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Semimonthly ISSN 0944-1344

SPRINGER HEIDELBERG, TIERGARTENSTRASSE 17 HEIDELBERG, GERMANY, D-69121

Coverage

RESEARCH ARTICLE

Spatial distribution, enrichment, and source of environmentally important elements in Batticaloa lagoon, Sri Lanka

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Received: 1 June 2016 / Accepted: 24 October 2016
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Abstract The present paper is the first documentation of distribution and contamination status of environmentally important elements of superficial sediments in the Batticaloa lagoon that is connected to the largest bay of the world. Surface sediment samples were collected from 34 sites covering all over the lagoon. Concentrations of elements such as As, Cr, Cu, Fe, Nb, Ni, Pb, Sc, Sr, Th, V, Y, Zn, and Zr were measured by X-ray fluorescence analysis. Geochemically, the lagoon has three different zones that were influenced mainly by fresh water sources, marine fronts, and intermediate mixing zones. The marine sediment quality standards indicate that Zr and Th values are exceeded throughout the lagoon. According to the freshwater sediment quality standards, Cr levels of all sampling sites exceed the threshold effect level (TEL) and 17 % of them are even above the probable effect level (PEL). Most sampling sites of the channel discharging areas show minor enrichment of Cu, Ni, and Zn with respect to the TEL. Contamination indices show that the lagoon mouth area is enriched with As. Statistical analysis implies that discharges from agricultural channel and marine fluxes of the lagoon effects on the spatial distribution of measured elements.

Further research is required to understand the rate of contamination in the studied marine system.

Keywords Spatial distribution · Heavy metals · Enrichment · Sediment quality assessment · Batticaloa lagoon · Sri Lanka

Introduction

Spatial distribution and concentration of heavy metals and other environmentally important elements in large-scale coastal aquatic environments are influenced by natural and anthropogenic factors (Gao and Chen 2012; Dou et al. 2013; Maanan et al. 2015). Major natural environmental factors include weathering and erosion of source rocks, river, marine fluxes, and biogenic influences. Major anthropogenic factors include industrial, agricultural, and domestic wastewater discharges and shrimp and fish farming effluents (Selvaraj et al. 2004; Fujita et al. 2014; Syakti et al. 2015).

Heavy metals collected in aquatic bodies are mobilized by water fluxes and finally, they sink and accumulate in the bottom of coastal aquatic environments such as lagoons, estuaries, or continental shelf areas (Yeats and Bewers 1983). The low solubility, toxicity, wide sources, and bioaccumulation behavior of heavy metals and other toxic elements cause serious problems on the environment and aquatic biology and simultaneously on human health through the food chains (Munksgaard and Parry 2002; Yu et al. 2008). Therefore, it is necessary to investigate and monitor the temporal and spatial distribution and concentration of such elements in bottom sediments of coastal aquatic sediments (Bryan and Hummerstone 1977).

The coastal zone of eastern Sri Lanka faces the largest bay of the world, Bay of Bengal. The northern part of the eastern coast is world famous for its valuable heavy mineral deposits such as ilmenite, zircon, and rutile. However, so far, no studies

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have been conducted on contamination of heavy metals and other toxic metals in the eastern coastal aquatic bodies. Hence, the present study focuses on spatial distribution and concentrations of some environmentally important elements in the superficial sediments of a large semi-enclosed lagoon in eastern Sri Lanka. It also includes an interpretation of contamination status and probable sources of such elements. This work provides a baseline study for the status of contamination in a large-scale lagoon of southeastern Asia.

Material and methods

Study area

Batticaloa is a micro-tidal, semi-enclosed, and the largest lagoon of the eastern part of Sri Lanka (Fig. 1). Thin quaternary sediments cover the basement rocks, which comprises of inter-banded granitic and hornblende biotite gneisses (Cooray and Katupotha 1991). Batticaloa lagoon is shallow in the southern extent and elongated toward the north–south direction. The lagoon mouth is a narrow path located at the northern end. A manmade marine entrance operates only during heavy flood periods at Periyakallar of the southern region. Fresh water inputs inflow from two major channels of southern areas that passes long distances along the flat paddy cultivated lands and six subchannels at various landward points (Fig. 1). The eastern side of the lagoon is densely populated while the western side is cultivated. Therefore, the Batticaloa lagoon has industrial, urbanized, and agricultural catchment that dilutes with seasonal marine fluxes in northern areas (Kularatne 2014). Three decades before the year 2009, the Batticaloa lagoon area was leisurely developed due to political instability of the country. Yet, after 2009, development in surrounding areas increased in a rapid manner.

The Batticaloa lagoon has a mixed ecosystem, together with marine and freshwater habitats (Kularatne 2014). Widespread grass beds, salt marshes, and secondary scrubby mangroves are observed in the northern areas, while most mangroves of the western part of the lagoon have been cut and burned for various human purposes (Kularatne 2014). Fish and prawn farming is a basic income for the people living around the lagoon.

This area belongs to the dry zone of the country, with a temperature variation between 22.5 and 32.5 °C and annual rainfall of 1500 and 2000 mm (Department of Meteorology, Sri Lanka, 2015). High rainfall values record from November to February with northeast monsoon activations. The rest of the year has a dry climate.

Sample collection

Using a standard Ekman Grab sampler, 34 superficial sediment samples were collected from the Batticaloa lagoon

in September 2015 (Fig. 1). The Ekman Grab sampler has the capacity to penetrate to a depth of about 15 cm and can catch a volume of 3.5 L of sediment sample (Jayawardana et al. 2012). Based on the prevalent hydrodynamic stresses, the sampling locations were carefully selected. The collected total sediment samples were transferred to a plastic tray, and textural characteristics of sediments were recorded referring a texture chart (Jayawardana et al. 2012). Three to four kilograms of surface fraction of each sediment sample was sealed in polyethylene bags and transported to the laboratory.

Analytical techniques

All samples were prepared for X-ray fluorescence spectrometry (XRF) (RIX 2000) at Shimane University, Japan. Oven-dried (105 °C for 24 h) samples were sieved to remove particles and materials above 2-mm size fraction and homogenized. Fifty grams (50 g) from each total sample was crushed using a RETSCH ball mill at the Department of Geology, University of Peradeniya. The homogenized powdered samples were packed in sealed polyethylene bags and transported to Shimane University in Japan for heavy metal analysis. The crushed sediment samples were pressed into a disk with a 200 kN force for 60 s. Next, the disk was analyzed for 14 heavy metals (As, Cr, Cu, Fe, Nb, Ni, Pb, Sc, Sr, Th, V, Y, Zn, and Zr) using a Rigaku RIX-2000 spectrometer equipped with an end window 4.KW Rh-anode X-ray tube. Instrumental calibrations, sample preparations, and concentrations of the elements were determined by the Press powder method (Ogasawara 1987). Average error for these elements was less than 10 % and the range of uncertainties for each element was based on the standards of Geological Survey of Japan (Ogasawara 1987).

Assessment of contaminations

Mathematical normalizations and statistical techniques help to differentiate the source of sediments in lagoon-estuarine systems (Woods et al. 2012). In this study, sediment contamination status was assessed for 14 heavy metals (As, Cr, Cu, Fe, Nb, Ni, Pb, Sc, Sr, Th, V, Y, Zn, and Zr) using the following sediment quality standards, contamination indices, and statistical techniques.

Sediment quality standards

Sediment quality inspections considered freshwater and marine sediment standards. Sediment Quality Guidelines (SQG) facilitated interpretation of the contamination of elements with respect to environmental concerns (MacDonald et al. 1996, MacDonald et al. 2000). The total chemical concentration of

six elements (As, Cr, Cu, Ni, Pb, and Zn) at each sampling location was compared with reference values of threshold effect concentration (TEC) and possible effect concentrations (PECs) of consensus-based freshwater ecosystem SQGs (MacDonald et al. 1996). Values in between TEC and PEC rarely or occasionally make adverse effects on organisms whereas values above PEC make adverse effects on a wide range of organisms (MacDonald et al. 1996).

Values of concentration of heavy metals in marine sediments recommended by the International Atomic Energy Agency (IAEA 2004) were employed to evaluate metal concentration standards, except for Nb and Y.

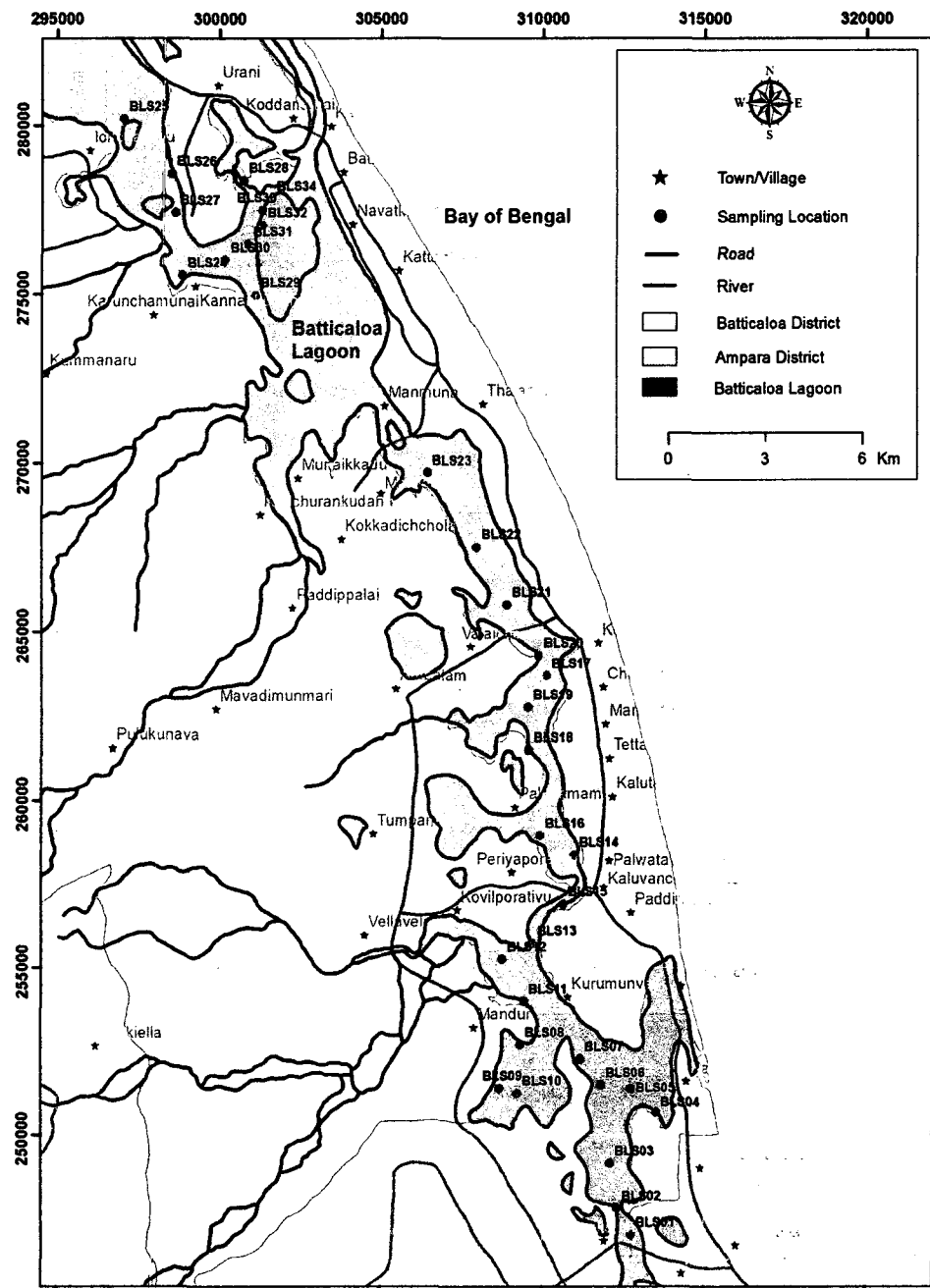
Sediment contamination indices

In this study, two different contamination indices facilitated to assess the degree of heavy metal contaminations of the Batticaloa lagoon.

Contamination index 1: EF

Enrichment factor (EF) clearly defines heavy metal enrichments in sediments (Lin et al. 2012). In this study, Fe was selected as the standardized element to calculate the EF out of the many conservative elements used (Fe, Al, Mn) (Aprile

Fig. 1 Map showing the geography of the Batticaloa lagoon, Sri Lanka and sampling sites of the superficial sediments in this study



and Bouvy 2008; Martins et al. 2012; Uduma and Awagu 2013). Since primary observations indicated that the clay fraction of all samples of the present study is comparatively less, Fe is the ideal element for normalization. Furthermore, several studies have used Fe as the normalization metal to assess the contamination status of freshwater coastal sediments (Carvalho et al. 2005; Acevedo-Figueroa et al. 2006). The EF was calculated using the following expression:

$$EF = \frac{[X/Fe]_{\text{Sample}}}{[X/Fe]_{\text{UCC}}}$$

where X and Fe are the element concentration and iron concentration in each sample and upper continental crust (UCC), respectively. The average UCC values were used as described in Taylor and McLennan (1985). The EF values were interpreted as suggested by Birch (2003), where $EF < 1$ is no enrichment, $1 \leq EF < 3$ is minor enrichment, $3 \leq EF < 5$ is moderate enrichment, $5 \leq EF < 10$ is moderately severe enrichment, $10 \leq EF < 25$ is severe enrichment, $25 \leq EF < 50$ is very severe enrichment, and > 50 is extremely severe enrichment.

Contamination index 2: Igeo

Geo-accumulation index was used as the second contamination index to assess metal contamination. The expression is described as introduced by Müller (1979):

$$I_{\text{geo}} = \log_2 \left[\left(\frac{C_{\text{measured}}}{1.5 C_{\text{measured (reference)}}} \right) \right]$$

where C is the concentration of each element. The reference values were used as described in UCC. Interpretation and classification of geo-accumulation index (Igeo) was performed as described by Müller (1981); < 0 is class 0 (uncontaminated/unpolluted), $0 \leq I_{\text{geo}} < 1$ is class 1 (unpolluted to moderately polluted), $1 \leq I_{\text{geo}} < 2$ is class 2 (moderately polluted), $2 \leq I_{\text{geo}} < 3$ is class 3 (moderately to highly polluted), $3 \leq I_{\text{geo}} < 4$ is class 4 (highly polluted), $4 \leq I_{\text{geo}} < 5$ is class 5 (highly to extremely polluted), and $I_{\text{geo}} \leq 5$ is class 6 (extremely polluted).

Statistical techniques

Elemental correlations were examined using Spearman's rank correlation by SPSS 16 software. Spearman correlation matrix is a non-parametric correlation analysis method used to evaluate the relationship between two independent variables of the raw data (Corder and Foreman 2014). Trace element data of the study area are not normally distributed and failed to indicate a linear relationship between them. Therefore, Spearman correlation matrix was calculated and p values < 0.05 were considered significant. Principal component analysis (PCA) is the most

reliable statistical analysis method to interpret the source of heavy metals in estuarine sediments (Woods et al. 2012).

Therefore, PCA was conducted for concentrations of highly correlated elements in Spearman matrix by Minitab 16 statistical package. Factor loadings for original elemental concentrations were computed and then score plot and loading plot of the first two principle components were generated to display the relationship of sediment sampling sites and elemental variables.

Results and discussion

Spatial distribution of heavy metals in sediments

The analytical results of the 14 heavy metals are presented in Table 1. Concentrations were in the descending order of $Zr > V > Sr > Cr > Zn > Y > Cu > Th > Pb > Ni > Nb > Sc > Fe > As$, and the concentration ranges (ppm) of the samples were 3.4–9.6 for As, 52.3–145.9 for Cr, 5.6–216.4 for Cu, 8.1–38.1 for Nb, 7.0–41.1 for Ni, 14.5–26.4 for Pb, 7.8–22.0 for Sc, 86.5–329.7 for Sr, 8.5–44.1 for Th, 80.4–281.7 for V, 32.6–74.1 for Y, 20.9–155.1 for Zn, and 267.1–2243.1 for Zr. Similar to the variation of salinity (Silva et al. 2013), the spatial distribution maps of the measured elements demarcated three different areas of the lagoon, since the northern, middle, and southern areas have different compositions (Figs. 2 and 3). The distribution map of As and Zr indicates high concentrations in the northern areas and Cu, Ni, Sc, V, and Zn concentrations are high in the southern area. In contrast, the distribution of Cr, Nb, Pb, Sr, Th, and Y is relatively high in the middle area of the lagoon.

Table 1 presents the elemental concentrations of the sediments and compared standards. Concentrations of As and Pb of all the samples were almost below the values of marine and freshwater sediment quality standards. All samples had above TEC values of Cr, and six of them indicated exceeded values for PEC of freshwater sediment standards. These six locations belong to both the northern and southern parts of the lagoon.

Many sampling locations of the southern part indicate that the concentrations of Cu, Ni, and Zn were in between the TEC and PEC values of fresh water SQG and exceeded the values of IAEA. The spatial distribution of Cu-, Ni-, and Zn-contaminated sediments were distinct to the southern parts of the lagoon and associated with two main paddy channel inputs.

Zr indicates very high concentrations than IAEA values for all sampling sites. Similarly, Th indicates exceeded values for IAEA, except one site. Excluding the sampling sites near the lagoon mouth (nine sites), V also exceeded the standards of IAEA. Likewise, Sc indicated exceeded values for IAEA in irregularly distributed locations.

Table 1 Heavy metal concentrations of surface sediments in the Batticaloa lagoon, Sri Lanka

Sample	Sediment properties	Metal concentration (mg/kg)													(g/kg)
		As	Cr	Cu	Nb	Ni	Pb	Sc	Sr	Th	V	Y	Zn	Zr	
BLS 01	CS	3.4	122.8	51.7	17.1	41.1	22.8	21.8	86.5	21.4	280.0	61.4	155.1	458.3	97.9
BLS 02	CS	3.6	113.3	175.0	16.2	33.7	20.5	22.0	98.2	19.6	279.1	55.7	150.8	267.1	103.0
BLS 03	CS	4.4	103.5	64.9	14.6	25.1	18.4	18.4	112.5	19.1	273.0	56.3	126.5	574.4	122.7
BLS 04	CS	3.8	66.3	19.6	15.7	13.1	22.1	12.6	165.8	14.6	172.3	41.3	76.6	933.9	63.1
BLS 05	CS	4.3	87.0	56.5	14.5	22.2	19.8	17.2	146.5	17.5	216.4	47.0	106.1	548.1	88.1
BLS 06	FS	4.9	77.2	11.1	17.2	7.3	16.8	12.1	126.1	21.0	171.4	46.0	55.0	1857.7	73.4
BLS 07	FS	6.1	78.2	14.6	15.0	13.2	19.0	14.0	161.5	28.7	204.5	65.9	78.7	1844.8	96.2
BLS 08	FS	4.5	92.3	50.1	18.4	24.4	20.2	19.2	193.3	24.0	231.2	55.6	107.9	896.5	82.2
BLS 09	FS	5.3	93.4	9.5	13.4	11.3	14.5	19.8	271.0	17.8	206.7	37.5	70.4	1511.3	67.7
BLS 10	CS	3.5	93.8	216.4	17.0	25.5	22.4	18.2	135.5	22.4	230.3	54.5	129.4	527.3	84.5
BLS 11	CS	5.1	71.3	20.2	18.6	17.5	21.1	13.0	136.3	22.7	185.8	54.4	83.3	1275.3	61.1
BLS 12	CS	3.7	88.1	35.3	19.2	25.5	24.4	17.8	114.1	22.0	234.7	70.5	127.1	448.2	86.0
BLS 13	FS	3.8	99.1	31.0	18.2	24.2	19.5	18.5	161.1	20.8	247.1	62.3	130.0	383.3	97.4
BLS 14	SC	6.5	98.5	18.9	20.1	18.4	24.8	13.5	176.1	37.2	221.3	74.1	93.6	1921.2	73.4
BLS 15	FS	4.6	121.8	33.3	22.2	29.5	21.8	18.6	100.8	28.3	281.7	66.3	129.9	972.3	98.9
BLS 16	CS	4.6	105.7	25.8	22.0	23.5	26.4	14.6	144.0	27.0	245.7	60.9	108.5	1196.6	82.2
BLS 17	FS	6.6	104.3	16.1	38.1	12.4	24.1	12.1	207.9	44.1	273.6	52.6	77.6	2064.3	53.4
BLS 18	SC	5.1	97.2	19.1	20.5	17.3	22.6	13.9	224.4	21.0	202.7	49.1	80.1	1054.5	59.1
BLS 19	FS	4.7	92.0	22.8	17.9	20.6	22.7	14.6	192.1	20.8	211.9	54.0	95.5	642.5	68.9
BLS 20	FS	4.7	102.1	10.5	27.3	7.2	20.3	12.3	329.7	23.3	194.6	37.4	53.4	1207.4	38.2
BLS 21	FS	4.6	99.1	27.0	17.6	24.2	22.9	16.1	180.7	19.6	219.5	54.5	102.5	506.5	75.7
BLS 22	FS	3.8	110.5	17.2	22.1	22.7	21.7	17.0	228.3	21.0	229.4	47.5	94.8	857.0	67.9
BLS 23	FS	5.7	91.3	11.5	31.9	13.2	23.0	11.0	251.7	24.5	215.0	38.8	63.9	1591.4	42.8
BLS 24	SC	4.5	79.0	5.6	10.4	8.0	17.7	7.8	152.2	18.9	80.4	34.7	20.9	1889.1	16.0
BLS 25	FS	5.2	65.3	26.9	12.4	13.1	15.5	16.2	220.1	13.0	168.0	40.4	68.9	1094.9	53.7
BLS 26	FS	5.8	52.2	12.3	8.1	10.1	19.5	9.2	201.4	8.5	93.3	35.6	44.7	1481.5	28.8
BLS 27	FS	7.0	69.2	18.4	13.5	18.4	17.0	12.8	159.7	15.3	166.9	43.9	63.7	1800.3	49.7
BLS 28	CS	7.8	145.9	22.8	13.0	19.2	21.3	12.4	143.2	16.9	157.6	45.3	89.5	1053.5	60.3
BLS 29	CS	7.7	82.3	18.2	13.5	16.7	20.0	11.7	153.0	20.8	145.5	47.8	77.9	1372.5	53.2
BLS 30	FS	9.4	63.7	13.0	13.6	11.2	19.6	13.4	178.4	14.6	126.2	35.0	47.3	2124.6	34.9
BLS 31	SC	7.6	73.7	9.0	13.9	7.0	19.1	8.9	163.8	15.3	106.3	32.6	36.8	2243.1	27.8
BLS 32	CS	5.9	81.7	14.8	11.6	14.6	20.7	13.0	198.0	13.6	127.3	37.7	59.0	1457.2	43.4
BLS 33	CS	7.4	124.8	19.6	11.7	15.0	18.8	17.8	200.6	15.6	146.8	40.8	70.1	1357.2	50.1
BLS 34	CS	9.6	120.0	19.7	12.6	16.0	17.7	17.2	200.3	14.7	151.3	41.1	79.3	1370.1	53.7
Min		3.4	52.2	5.6	8.1	7.0	14.5	7.8	86.5	8.5	80.4	32.6	20.9	267.1	16.0
Max		9.6	145.9	216.4	38.1	41.1	26.4	22.0	329.7	44.1	281.7	74.1	155.1	2243.1	122.7
Average		5.4	93.1	33.5	17.3	18.3	20.6	15.0	174.0	20.8	197.0	49.4	86.9	1199.5	65.4
SQG values	TEC	9.8	43.4	31.6	...	22.7	35.8	121.0
	PEC	33.0	111.0	149.0	...	48.6	128.0	459.0
IAEA		18.9	136.0	30.8	...	39.4	26.0	14.6	302.0	9.8	160.0	101.0	148.0	...	40.8

CS coarse sand, FS fine sand, SC silty clay

Light blue shaded numbers are in between TEC and PEC. Dark blue shaded numbers are above PEC. Bold numbers indicate exceeded values for IAEA

Comparing the selected sediment heavy metal data (Cr, Cu, Ni, Pb, and Zn) of the Batticaloa lagoon with available published data of other coastal aquatic environments around Bay of Bengal reveal that sediments of the Sri Lankan lagoon are in a status of less contamination (Table 2). Published data indicate that the east and west coastal areas of Bay of Bengal are contaminated with the discussed heavy metals, but the northern areas have less contamination (Table 2). In contrast to the west coastal areas of Bay of Bengal, the main lagoon of eastern Sri Lanka has fewer contaminations.

Contamination indices

Enrichment factor

The enrichment factor for six heavy metals for all stations is presented in Fig. 4. Cu, Ni, Pb, and Zn indicated no or minor enrichments for the Batticaloa lagoon, except for one sampling site for Cu. Minor enrichments for Cr are observed throughout the lagoon. A moderate to moderately severe enrichment of As is recorded for the surface sediments of the Northern part of the lagoon, while the sample obtained from the BLS24 location of the northern part reveals moderate to moderately severe enrichments for As and Cr. Moderate EF values indicate that the element concentration has an effect from non-natural processes (Zhang and Liu 2002).

Geo-accumulation index

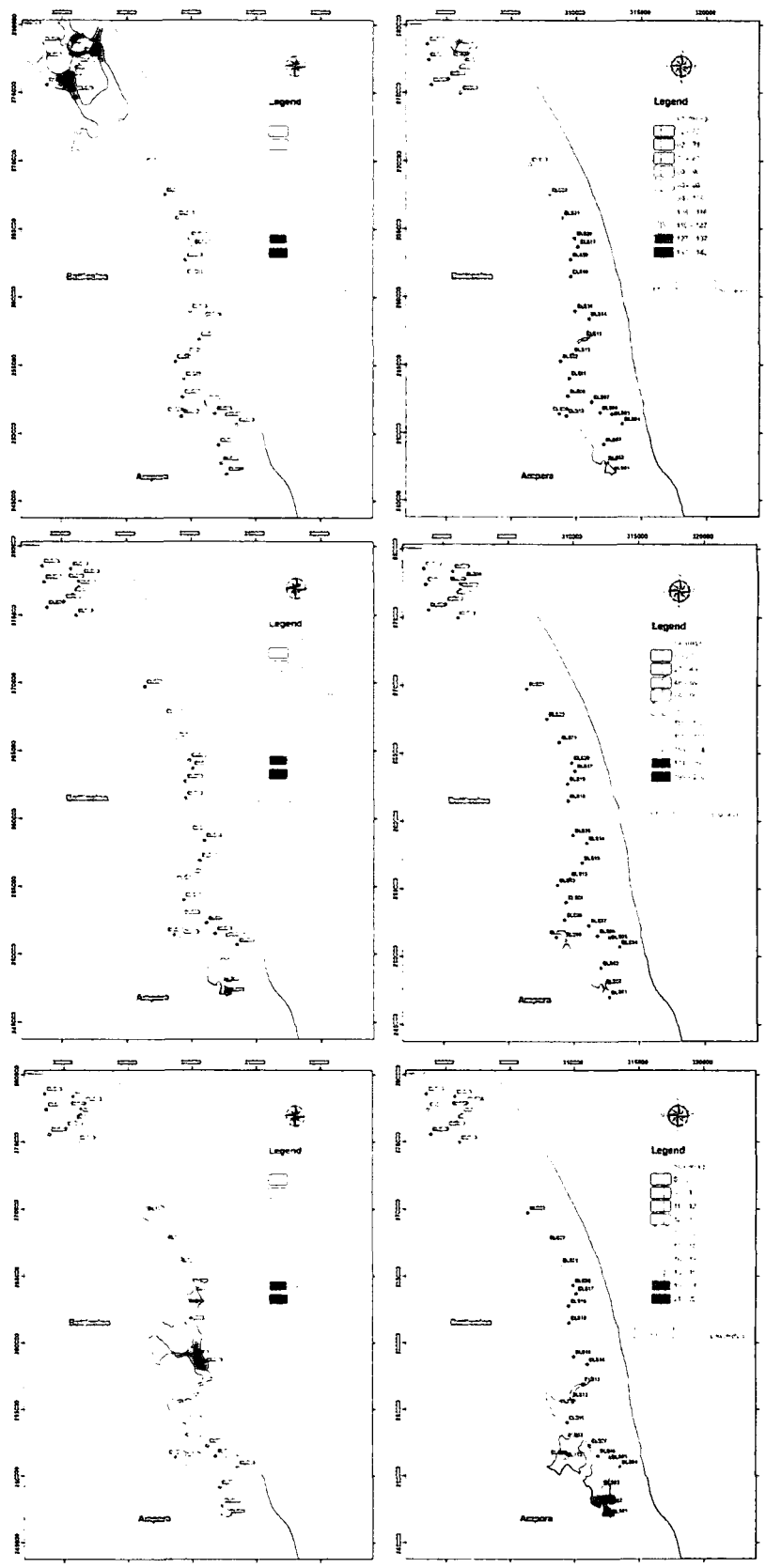
Pb in all the sampling sites of the Batticaloa lagoon indicated Igeo values below zero, and hence it can be categorized into the “unpolluted” class (Fig. 5). Cu, Ni, and Zn indicate unpolluted to moderately polluted sampling sites in most of the southern areas. Igeo values of Cr denote unpolluted to moderately polluted status for almost all sampling sites and that of As indicated moderate pollution for the northern part of the lagoon.

Statistical analysis for source

Table 3 explains the Spearman correlation matrix for all sediment samples. Very strong positive correlations were obtained in two metal clusters. Cluster 1 includes Cu, Ni, and Zn while cluster 2 includes Fe, V, Y, and Zn. Moreover, Cu and Ni also reveal strong correlations with Fe, Sc, V, and Y. Cluster 1 metals display a very strong negative correlation with Zr while that of cluster 2 indicate moderate to strong negative correlation.

Figures 6 and 7 illustrate the results obtained from PCA analysis. Two PCs were selected having cumulative variance of 74 % with eigenvalues greater than 1.6. Sediment samples associated with the two main channel inputs were grouped to the positive side of both PCs and mainly consist of Cu, Ni, Sc, and Zn. Sediment samples

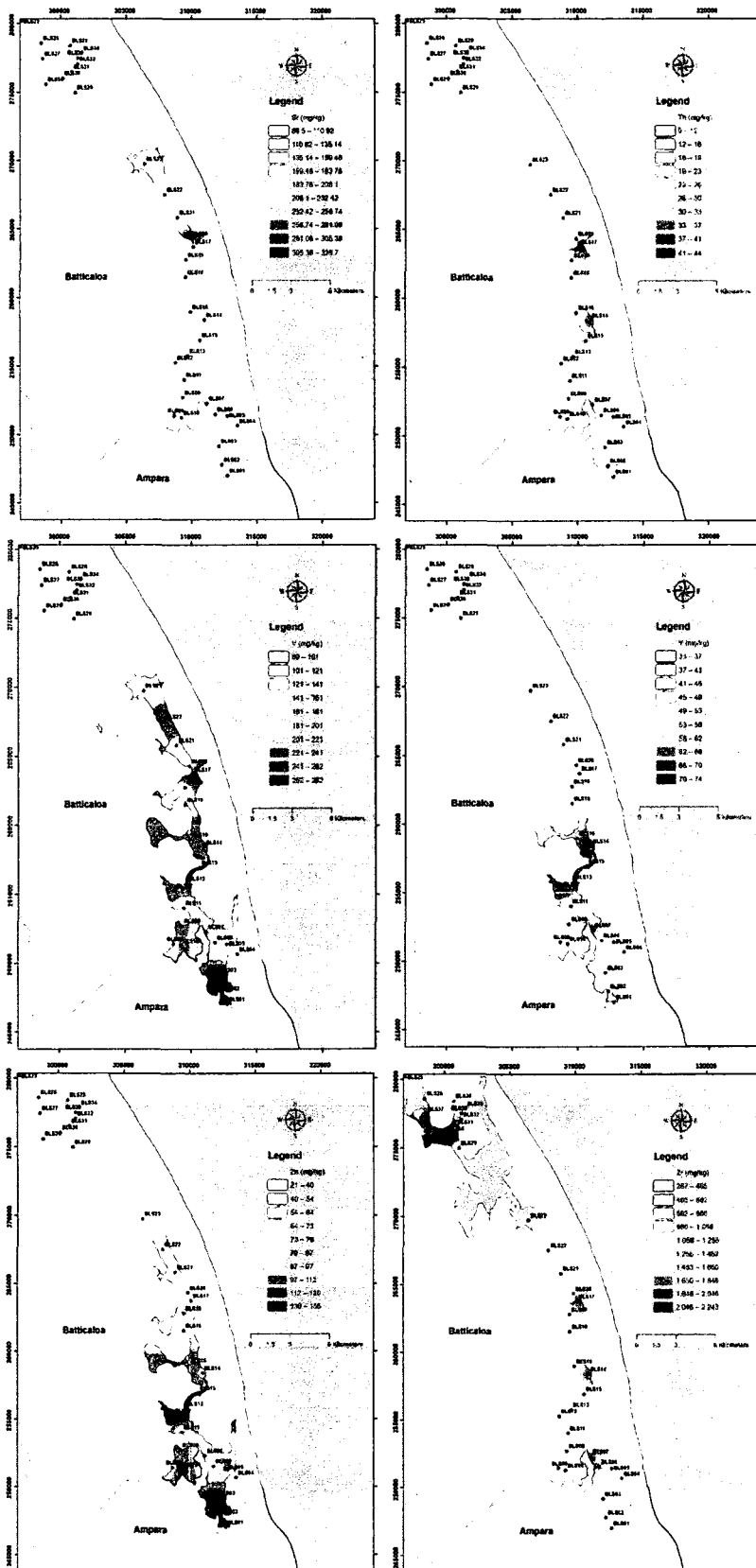
Fig. 2 Spatial distribution of As, Cr, Ni, Cu, Pb, and Sc in surface sediments of Batticaloa lagoon, Sri Lanka



of the northern area associated with the lagoon mouth have the lowest contents of heavy metals. They are

grouped in the negative side of the PC1, indicating their reduced influence for heavy metal concentrations. PCA

Fig. 3 Spatial distribution of Sr, Th, V, Y, Zn, and Zr in surface sediments of Batticaloa lagoon, Sri Lanka



clearly implies that the enrichment of Zr and As is free from marine sediments.

The center part of the Batticaloa lagoon is rich in natural metal enrichments. However, BLS04, BLS06, and BLS09

Table 2 Concentration of selected heavy metals of the Batticaloa lagoon, Sri Lanka and coastal aquatic environments of Bay of Bengal

Location		Concentration (ppm)					Reference
		Cr	Cu	Ni	Pb	Zn	
Batticaloa lagoon, Sri Lanka	Min	52.20	5.60	7.00	14.50	20.90	Present study
	Max	145.90	216.40	41.10	26.40	155.08	
	Average	93.14	33.48	18.31	20.55	86.91	
Gulf of Mannar		177.00	57.00	24.00	16.00	73.00	Jonathan et al. (2004)
Kalpakkam, Southeast India		118.00	–	53.00	21.70	119.00	Selvaraj et al. (2004)
Pondicherry, Southeast Coast, India		333.55	47.74	19.72	32.91	52.04	Sotali et al. (2013)
Gange's estuary, India		67.00	26.00	32.00	29.00	71.00	Subramanian et al. (1988)
Krishna Estuary		174.00	59.00	149.00	4.00	1482.00	Ramesh and Subramanian (1988)
West Bengal, Northeast India		36.50	35.70	33.50	17.20	74.10	Sarkar et al. (2004)
Chittagong coast, Bangladesh		658.45	189.18	32.64	18.09	355.00	Hasan et al. (2013)

locations are close to the inlet opening at the southern area, opened only during heavy rains. Hence, these sampling sites have marine influences.

Very strong positive correlations of metals among each other indicated a common source of input to the lagoon. Generally, Fe and Al are used as reference parameters to distinguish natural and anthropogenic sources (Woods et al. 2012). Hence, very strong positive correlations and uniform distribution of Fe, V, and Y specified their common source and crustal contribution of the area. In the case of Cu, Ni, and Zn, their distribution and correlation specify that their source is greatly influenced by channel inputs that follow long distances through the paddy fields over crustal contribution. These channels receive paddy discharges from Malwaththa, Sammanthurai, Nithavur, and Karthive at Kalumunei input and Vellaveli, Navithanveli, and Annamalei at Mandurinput.

Here, paddy lands are cultivated during dry and wet seasons. Therefore, the source of Cu, Ni, and Zn can be highly affected from agricultural effluents and can be concluded as anthropogenic inputs (Xue et al. 2000; Fonseca et al. 2013; Liu et al. 2014; Maanan et al. 2015).

Dissanayaka and Chandrajith (2009) reported that the phosphate fertilizers used in the agricultural zones of Sri Lanka contain high amounts of heavy metals including Ni, Cr, and Pb. Further, Zn, Cu, Cd, Pb, and As are widely used in agricultural fertilizers, and pesticides–fungicides finally accumulate in aquatic environments (Gimeno-Garci'a et al. 1996; Kelepertzis 2014).

Cattle farming is a common commercial practice in eastern Sri Lanka, where Zn and other trace metals are fed as trace metal supplements for health improvements of cattle. These metals finally accumulate in agricultural

Fig. 4 Calculated enrichment factor for selected heavy metals and other toxic elements for all sampling sites of Batticaloa lagoon, Sri Lanka

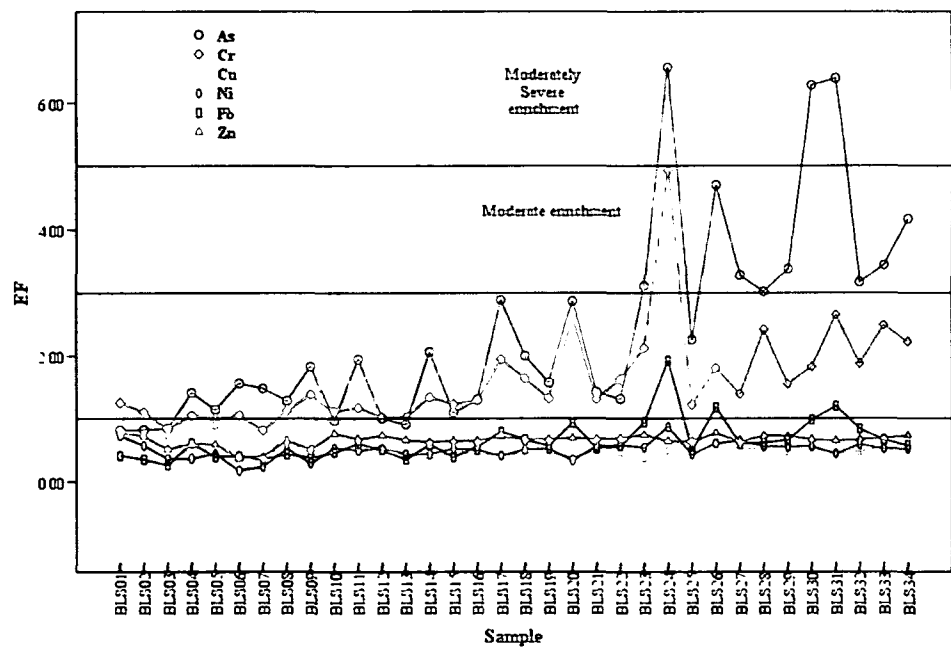
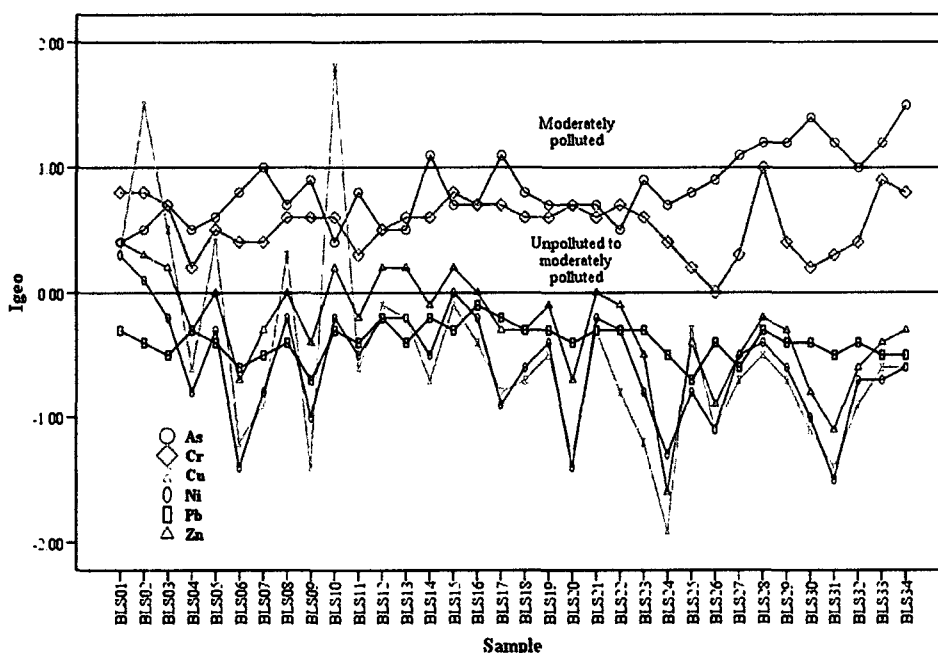


Fig. 5 The results of Igeo values for selected heavy metals and other toxic elements were plotted against the sampling location of Batticaloa lagoon, Sri Lanka



lands as urine and feces (Gimeno-García et al. 1996; Nicholson et al. 2003; Franco-Urriá et al. 2009).

In contrast, the negative correlations of Zr with the above-discussed two groups (channel inputs and natural geology) imply their different influences. Enrichment of Zr can be attributed solely as natural accumulations. The northeastern part of Sri Lanka is rich in mineral sand deposits including ilmenite, rutile, and zircon (Jinadasa and Wijayadeva 2013), and obviously, the northeastern coastal sediments are rich in Zr. The sea currents generated due to northeast monsoonal winds in January period are directed to the south, with sediment movement accompanying the currents (Suwannathatsa et al. 2012).

Correspondingly, Chandramohan et al. (1990) identified Batticaloa in Sri Lanka as a nodal drift point of long-shore transports, indicating that the volume of sediments arrive from northerly and southerly is equal. Therefore, high concentrations and spatial distributions of Zr are perhaps due to the seasonal marine sediment flux that is rich in mineral sands, which enters through the lagoon mouth.

Spearman matrix values of As with Zr indicate a strong positive correlation (0.76). Hence, it can be suggested that the enrichment of As and Zr in lagoon mouth area is due to marine fluxes (Wang et al. 2012). It indicates strong negative correlation with Fe, V, and Zn and very strong negative correlations with Cu and Ni. The largest application of As is

Table 3 Spearman correlation matrix of heavy metals and other toxic elements from sediments of the Batticaloa lagoon, Sri Lanka

	As	Cr	Cu	Nb	Ni	Pb	Sc	Sr	Th	V	Y	Zn	Zr	Fe
As	1.00													
Cr	-0.18	1.00												
Cu	-0.57	0.41	1.00											
Nb	-0.36	0.32	0.16	1.00										
Ni	-0.59	0.54	0.89	0.31	1.00									
Pb	-0.29	0.33	0.30	0.68	0.44	1.00								
Sc	-0.52	0.50	0.73	0.11	0.74	0.02	1.00							
Sr	0.40	-0.11	-0.54	0.02	-0.55	-0.09	-0.25	1.00						
Th	-0.26	0.36	0.13	0.83	0.32	0.56	0.13	-0.15	1.00					
V	-0.65	0.56	0.65	0.69	0.74	0.51	0.66	-0.33	0.66	1.00				
Y	-0.46	0.41	0.68	0.53	0.79	0.49	0.56	-0.53	0.66	0.80	1.00			
Zn	-0.63	0.59	0.88	0.42	0.94	0.46	0.77	-0.52	0.43	0.84	0.86	1.00		
Zr	0.76	-0.42	-0.84	-0.21	-0.80	-0.30	-0.69	0.35	-0.06	-0.60	-0.53	-0.81	1.00	
Fe	-0.65	0.42	0.77	0.35	0.79	0.24	0.76	-0.58	0.42	0.82	0.86	0.89	-0.70	1.00

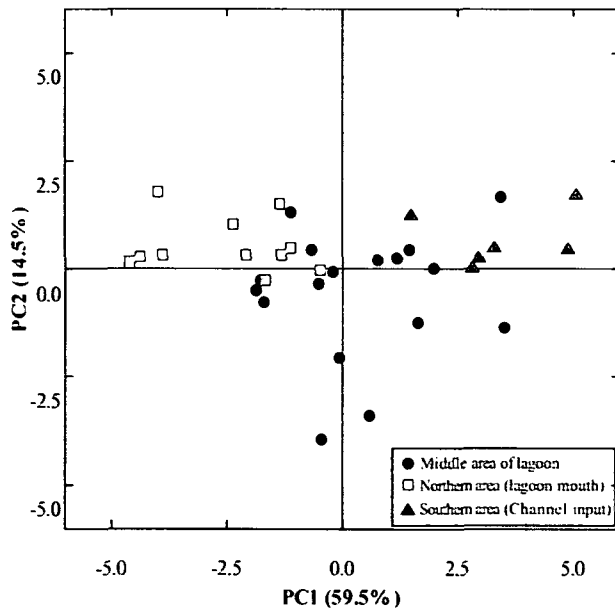


Fig. 6 Principal component score plot of first two PCs for all sampling sites

in agricultural products such as herbicides and insecticides, and this is one possible source of As in Batticaloa lagoon due to regional agricultural base. Moreover, it is reported that most marine algae produce organoarsenic compounds (Penrose 1974; Andrae and Klumpp 1979). Distribution of As in northern areas is probably due to marine biological processes of coastal waters.

The Cr enrichment throughout the lagoon can be explained as a natural process as well as an anthropogenic source. PCA of Cr indicates its relation to cluster 2 (Fig. 7). Moderate spearman correlations between both clusters evidenced the probable sources. Since the agricultural

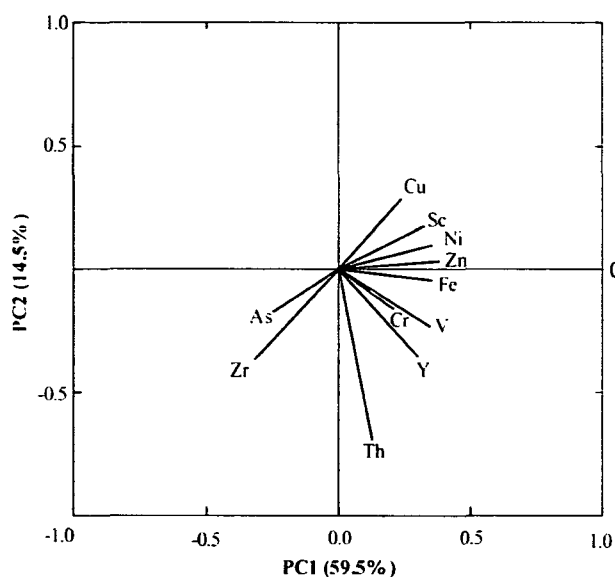


Fig. 7 Principal component loading plot of first two PCs for highly correlated variables from sediments of the Batticaloa lagoon, Sri Lanka

effluents are higher than industrial effluents, the probable source of Cr may have been affected widely from phosphate fertilizers (Dissanayaka and Chandrajith 2009).

Conclusions

For the first time, the present study demonstrates about the metal contamination status of the Batticaloa lagoon in Sri Lanka. Heavy metal enrichments described here reveal major areas affected by marine fluxes and agricultural channel inputs. Northern areas closer to the lagoon mouth have high enrichment of Zr, indicating that natural ocean fluxes that are rich in mineral sands influence the spatial distribution. Southern areas are nourished by paddy channel inputs, and Cu, Ni, and Zn are the enriched metals with exceeded values for sediment quality standards.

In addition, Cr, Fe, Th, V, and Y are randomly distributed throughout the lagoon, pointing out their natural lithogenic origin. Hence, it is possible to suggest that heavy metal distribution and nourishment of Batticaloa lagoon are from both natural processes, including marine fluxes and weathering products, and anthropogenic processes.

Acknowledgments The authors highly appreciate Professor Yoshihiro Sawada of Shimane University, Japan, for access to the XRF facility. Head of the Department of Geology, University of Peradeniya and Head of the Department of Physical Sciences, South Eastern University of Sri Lanka are also acknowledged for providing the laboratory facilities. The research was financially supported by University Grant Commission, Sri Lanka (Grant No. UGC/DRIC/PG/2014AUG/SEUSL/01).

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