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RESEARCH ARTICLE

Trend analysis of anthropogenic activities affecting trace metals deposition in core sediments from the coastal and four rivers estuary of Sarawak, Malaysia

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Abstract
This study reports the concentrations of trace metals in core sediments profile from the coastal and four rivers estuary in the Kuching Division of Sarawak, Malaysia, and the controlling mechanisms influencing their availability in sediments of the studied area. The bonding of trace metals with non-mobile fractions was confirmed with the sequential extraction. Inductively coupled plasma–optical emission spectroscopy (ICP–OES) was used to measure the concentrations of the trace metals. Granulometric analyses were performed using normalized sieve apertures to determine the textural characteristics of the sediments. Enrichment factor was used to evaluate the level of metal enrichment. Heavy metals concentrations in sediment samples varied in the range: Pb (8.9–188.9 mg/kg d.w.), Zn (19.4–431.8 mg/kg d.w.), Cd (0.014–0.061 mg/kg d.w.), Ni (6.6–33.4 mg/kg d.w.), Mn (2.4–16.8 mg/kg d.w.), Cu (9.4–133.3 mg/kg d.w.), Ba (1.3–9.9 mg/kg d.w.), As (0.4–7.9 mg/kg d.w.), Co (0.9–5.1 mg/kg d.w.), Cr (1.4–7.8 mg/kg d.w.), Mg (68.8–499.3 mg/kg d.w.), Ca (11.3–64.9 mg/kg d.w.), Al (24.7–141.7 mg/kg d.w.), Na (8.8–29.4 mg/kg d.w.), and Fe (12,011–35,124.6 mg/kg d.w.). The estimated results of the enrichment factor suggested enrichments of Pb, Zn, and Cu in all the core sediment samples and depths at all sites. The other trace metals showed no enrichments in almost all the sampled stations. Continuous accumulation of Pb, Zn, and Cu metals over a period can be detrimental to living organisms and the ecology. The results obtained from the statistical analyses suggested that the deposition of trace metals in the studied sites is due to anthropogenic inputs from the adjacent land-based sources.

Keywords Trend analysis · Trace metal · Enrichment · Core sediments · Anthropogenic inputs · Sarawak

Introduction

Some of the most important places for human settlements are coastal and estuarine areas (Likuku et al. 2013; Weissmannova et al. 2015; Asare et al. 2021a, b); nevertheless, with heavy industrialization and speedy urbanization, trace metals are constantly transported to the estuarine and coastal sediments from up-stream of the creek (Kara et al. 2015; Weissmannova and Pavlovsky 2017). The contamination of trace metals in core sediment could harm water quality and bioaccumulation of trace elements in aquatic organisms could result in long-term adverse health effects on humans (Sekabira et al. 2010; Devesa-Rey et al. 2011; Alexakis 2011; Asare et al. 2019a, b, 2021a; Omorinoye et al. 2020; Nawrot et al. 2020). The presence of heavy metals in water, soils, and sediments cannot be regarded as non-aligned to the ecosystem or humans because of their mutagenicity and accumulation via the food chain (Ergonul and Altindag 2014; Maanan et al. 2015).

The differences in ecological conditions in and around storage reservoirs and streams are documented in the sediment profiles (Wang et al. 2017). Sediments contamination increases because they are an important indicator of

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environmental variation caused by anthropogenic activities (Alexakis and Gamvroula 2014; Guo et al. 2019). Trace metals gather in sediments in different forms are probable to be mobilized and become toxic in the environment in the path of utilization (Wen et al. 2016). Trace metals within the exchangeable or carbonate-bond portions emitted from sediments, approximately 1% is regarded safe to the ecosystem, and more than 50% of the total quantity may constitute an immense risk and perhaps through the food chain (Nayak 2015).

Quantification of environmental contamination with trace metals generally includes the various geochemical indices such as enrichment factor (*EF*), pollution load index (PLI), contamination factor (*CF*), and geoaccumulation index (*I*geo) (Hakanson 1980; Hakanson 1984; Likuku et al. 2013; Nazeer et al. 2016; Weissmannova and Pavlovsky 2017). The determination of trace metals distribution in core sediments and the levels of its enrichment provide the foundation for the understanding of sediments improvement techniques and assessment of the possible emission of metals into water and conveying downstream (Kowalska et al. 2018; Yang et al. 2018; Nawrot et al. 2020).

The Kuching Division of Sarawak, Malaysia underlines the South China Sea which forms part of the Pacific Ocean, surrounding an area from Kalimantan and Malacca Straits to the Taiwan Straits with about 3,500, 000 km^2 (Chen et al. 2015). The nature of the estuarine and coastal sediments in Kuching is dependent on the complex interaction of several factors that control sediment composition, transportation, deposition, and postdeposition of sediment (Morni et al. 2017). Furthermore, it is all based on the origin of material such as biological or geological sources or both to ascertain the constituents (Bale and Kenny 2008). Generally, the continental shelf content of sediments in Sarawak particularly the Kuching Division is biogenic, volcanic, and terrigenous substance (Liu et al. 2013) which is comprised of kaolinite, illite, smectite, chlorite, and a mixture of clay minerals (Chen et al. 2015). If interaction remains stable, the sedimentary environment will continue unchanged unless any of the factors change in the period to cause alteration of sediment (Holme and Mclntyre 1984). Transportation of sediment is a factor influenced by the mobility of particles and water current which is dependent on water velocity, roughness velocity, settling velocity, and threshold velocity (Gray 1981). Approximately 80% of the organic matter buried on the continental shelf was obtained from the rigorous sedimentary accumulation and high biological production (Kao et al. 2007).

The Kuching Division of Sarawak has experienced population growth and increased in development in the past 20 years, which is probably contributing to environmental contamination (Sim 2007). Sarawak's urbanization rate stood at 53.80% in 2010, compared with 36.00% in 1991 and 15.50% in 1970 as reported by the Department of Statistics Malaysia. In 2020, population and housing census conducted by the Department of Statistics Malaysia showed an increase in the human population in Kuching from the year 2010 with 617,877 inhabitants to 711,500 residents in the year 2020. The effect of urbanization on water quality and the macrobenthos community structure in the Fenhe River, Shanxi Provine, China reported by Wang et al. (2020) indicate that intensification of urbanization has strongly affected the water, sediment, and macrobenthos in the Fenhe River watershed. In addition, Sarawakians depend on gas fuels for heating and burning which produces numerous air contaminants such as PAHs, heavy metals, carbon (II) oxide, sulfide, and particulate matter into the environment. Floods gathered sediments within the nearby communities could be relocated inside the estuarine and finally transport to the coast of the South China Sea. The last major flood incident that occurred on January 17, 2015 has a massive influence on the quality of underneath sediments and transport trace metals downstream. Contaminants such as heavy metals trapped in the sediments can create environmental or human health risks which can be a problem of the greatest consequence in partially crowded areas like nearby towns of the study sites (Nawrot et al. 2020, 2018). However, pollutants deposition prevents their movement further downstream and accumulation in the ocean (Nawrot et al. 2020).

Most of the studies on heavy metals distribution in sediments within the estuarine and coastal sites of this region were mostly concentrated on quantitative data only without dealing with the anthropogenic activities affecting trace metals availability and implications of metals on the environment (Sim 2007; Sim et al. 2016; Omorinoye et al. 2019). Furthermore, information regarding sediment fractions and water flow velocity influence on heavy metals availability in the sediment of the estuarine and coastal sites of this region is lacking. Even if they exist, they have not been documented for easy retrieval. Hence, this study is a trend analysis of anthropogenic activities influencing trace metals deposition in core sediments from the coastal and four rivers estuary of Sarawak. This study is important because the results of the investigation would be of great importance to policymakers to identify the vulnerability site and take appropriate remedial effort to protect the studied coastal ecosystem. It can be deduced that there has been trace metals deposition in the core sediments of the coastal and four rivers estuary due to both natural and anthropogenic activities. Hence, the research is driven by the hypotheses that (a) there are significant strong relationships between trace metals concentrations and particle size fractions in core sediments collected from the coastal region and four rivers estuary, (b) there is a significant difference in trace metals concentrations in surface and bottom bed sediments between the coastal sites and the selected estuaries, (c) there is an enrichment of trace metals from the top layers to the bottom layers of the core sediments from the study area, and (d) the sources of trace metals deposition in the core sediment are greatly influenced by anthropogenic inputs. To test these hypotheses, core sediments were collected from the coastal and four rivers estuary in the Kuching Division of Sarawak, Malaysia to evaluate trace metals concentrations and predict the mechanisms influencing their availability. Also, multivariate analyses were used to predict the likely sources of trace metals contamination using the surface layer of the core sediments.

Materials and methods

Study area and sample collection

The study was carried out in the coastal territory and four estuaries of Rambungan River, Sibu River, Salak River, and Santubong River in the Kuching Division of Sarawak state, Malaysia (Fig. 1). The collection of core sediment samples were carried out from 8 sampling points from September to October 2020. The sampling stations were located by employing geographical positioning system (GPS). The coordinates of the locations are shown in Table 1. The sample collection procedure was conducted according to Gao (2019). The core sediments were collected with the help of gravity corer. Approximately 30-cm-long core sediments were obtained from each site of the coastal and the four rivers estuary.

The sediment core samples were sliced at an interval of 5 cm. To avoid the samples undergoing oxidization due to the presence of air, the open ends of the pipes containing the samples were closed using corks (USEPA 2009). The sliced samples were placed in clean plastic bags and transported to the laboratory within 5 h.

Extraction of trace metals

The protocol used for sample treatment before the measurement was adapted by Asare et al. (2019b). Sediment samples were placed in crucibles and oven-drying at a temperature of 107 °C for 24 h. After oven-drying, the sediment samples were ground using mortar and pestle and passed through 2.0-mm sieves. To exclude external contamination, contact between the core sediment samples and metal materials was prevented throughout the process of preparation.

Total trace metal concentration: subsample of 1.0 g (0.001 g accuracy) from each sediment core sample was measured and placed into crucibles. HCl, HF, and $HClO_4$ (1:2:3; Suprapur) were added. The weighted samples in crucibles were placed in a muffle furnace for 6 h at a temperature of 250 °C. Subsequently, 10 mL of concentrated HNO_3 (Suprapur) was added after the solution was evaporated to dryness. The dried residue was dissolved in 15 mL of 0.1 M HNO_3 (Suprapur) and transferred into 50-mL test tubes.

Extractions of trace metals from sediments were done according to BCR sequential extraction procedure (Bodog et al. 1996). About 1.0 g (0.002 g accuracy) of a sediment subsample was subjected to Community of Bureau of Reference (BCR) sequential extraction with dilute 0.11 M CH_3COOH, an oxidizing agent (8.8 mol/L hydrogen peroxide), and 0.05 M hydroxyl ammonium chloride, pH = 2 as a reducing agent. The solutions used were the ones previously prepared except $NH_2OH.HCl$ solution. The next extraction phase was started immediately after stage one was completed. Room temperature condition was applied during the extraction of sediment core samples to obtain individual fractions and it was done for 20 h. The extracts obtained were transferred to 50-mL plastic tubes, then acidified with concentrated trioxonitrate (*v*) acid, and kept in a refrigerator before analysis. The following dilutions were prepared; × 10.0, × 100.0, and × 100.0, and the extracts were analyzed for Pb, Zn, Cd, Ni, Mn, Cu, Ba, As, Co, Cr, Mg, Ca, Al, Na, and Fe with inductively coupled plasma–optical emission spectroscopy (ICP-OES). The quantified results were expressed in mg/kg d.w. The quantifications were performed in four replications. The assurance of quality control was carried out by analyzing a prepared 0.5 mg/L, 1.0 mg/L, 3.0 mg/L, and "blanks" from 1000-mg/L-certified stock standard solutions. The certified stock standard solutions were purchased from Merck in Germany. Excellent agreement between analyzed and certified values were established because the correlation coefficient between the certificated and analyzed values was approximately 0.999984; recoveries were in the range of 98–101%, based on individual metals. Regarding precision, the relative standard deviation was in the range of 0.19–2.77%. The limit of detection (LOD) of every individual metal was computed as (3 × SD) + blank, where; *SD* stands for standard deviations of the blank ($n = 5$) (Asare et al. 2019a, b). The limit of detection were as follows: Pb = 0.009 mg/g, Zn = 0.020 mg/g, Cd = 0.044 mg/g, Ni = 0.038 mg/g, Mn = 0.013 mg/g, Cu = 0.031 mg/g, Ba = 0.007 mg/g, As = 0.003 mg/g, Co = 0.035 mg/g, Cr = 0.026 mg/g, Mg = 0.015 mg/g, Ca = 0.068 mg/g, Al = 0.099 mg/g, Na = 0.042 mg/g, and 0.051 mg/g.

The accuracy performance of quantifications depicts how close a value or an outcome appears to the certified value. The test of accuracy was assessed by using certified reference material (CRM), sewage sludge amended soil, standardized by the Community Bureau of Reference (BCR), the reason is that the results obtained from the certified reference material analysis are mostly suitable for the assessments of the accuracy of performance of any analytical technique because it is easy to compare measured values to certified values and draw a conclusion. The regression line was used to detect the values using SPSS statistics.

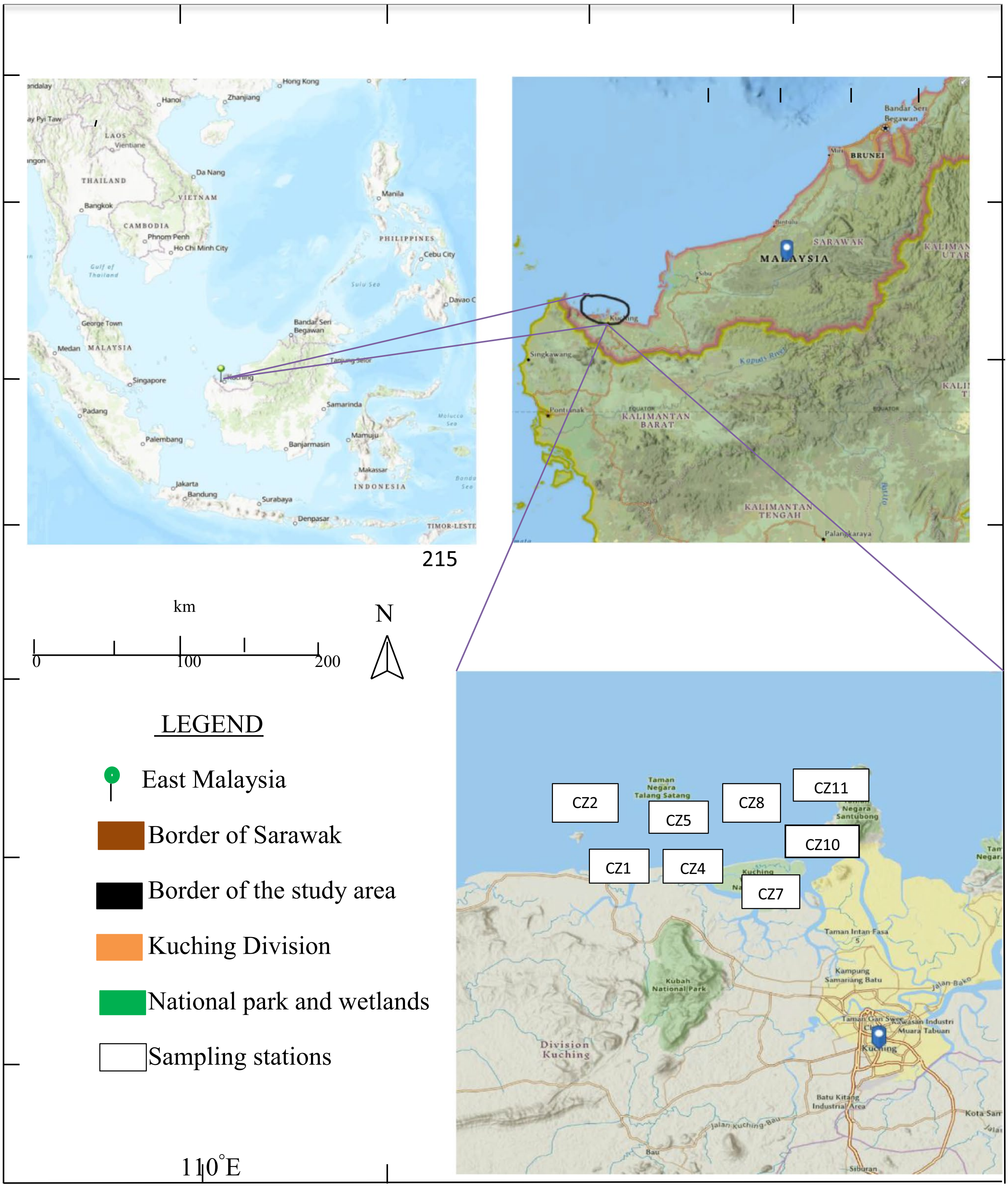

Fig. 1 Map of study area showing sampled sites (adapted from Asare et al. 2021a)

The certified values were plotted as the vertical axis and the measured values as the horizontal axis. It was observed that reference values at which the experimental values were derived are grouped around the perfect correlation line; this means that the measured values were in good association with the reference values. Also, the accuracy was evaluated

Table 1 Coordinates of sampled locations

Sample ID	Locality	Coordinates
CZ1	Rambungan River Estuary	01° 41′3 7.7″ N 110° 08′ 24.5″ E
CZ2	Offshore of Rambungan River opposite to small Satang Island	01° 44′ 46.8″ N 110° 08′ 45.4′″″E
CZ4	Sibu River Estuary	01° 44′ 46.8″ N 110° 08′ 45.4″ E
CZ5	Offshore of Telega Air opposite to small Satang Island	01° 45′ 50.4″ N 110° 11′ 30.2″ E
CZ7	Salak River Estuary	01° 40′ 41.1″ N 110° 16′ 59.2″ E
CZ8	Santubong Bay	01° 42′ 45.7″ N 110° 27″ 63.1″ E
CZ10	Santubong River Estuary	01° 42′ 32.6″ N 110° 19′ 02.3″ E
CZ11	Offshore of Santubong Resort	01° 44′ 49.6″ N 110° 29′ 72.3″ E

from the *Z*-score value (Misa et al. 2014; Asare et al. 2019a). The *Z*-score is also known as a standard score, it is a score that generates an idea of how far from the mean a data point is. Scientifically, it's a measure of how many standard deviations below or above the variable mean a raw score is. The standard score (*Z*-score) of each metal was computed using Eq. 1.

$$Z = \frac{R_{Lab} - R_y}{U} \quad (1)$$

where R_{lab} signifies measured or laboratory value, R_y denotes certified value accepted as the true one, and *U* denotes the uncertainty of the certified value. Assessment of uncertainty of the certified values was quantified at the 95% confidence level by Eq. 2 (Eurachem, Middlesex 1998),

$$U = k \times RSD \quad (2)$$

where *U* denotes the uncertainty of the reference value and *k* represents the coverage factor ($k = 1.740$, for 95% and 18 points). If *Z*-score ˂ 2 then laboratory performance is reported as satisfactory and acceptable but if *Z*-score > 2 but ˂ 3 then the performance is questionable. The laboratory performance becomes unsatisfactory if the *Z*-score > 3 (Funk et al. 2007; Misa et al. 2014; Asare et al. 2019b). Table 2 highlights the measured values, the calculated *Z*-score values, and the ratio of difference relative to the certified values expressed in percentage. The accuracy test demonstrated that all the trace metals investigated their *Z*-score values were less than 2, indicating that laboratory performance is satisfactory and acceptable.

Granulometric evaluation

To evaluate the distribution of particle size in core sediment samples, granulometric analysis was carried out by using six normalizing sieve apertures (2.0 mm, 1.0 mm, 0.50 mm, 0.125, and ˂ 0.0625 mm) according to the procedure established by USEPA (2007). Approximately 300 g of dried surface layer sediment samples were sieved in 20 min with the help of a mechanical shaker. The residue of each sieve was measured (0.01 g accuracy). The fractions of each sediment texture for each sampled station in percentage content: gravel (> 2.0 mm), sand (0.5–2.00 mm), silt (0.125–0.0625), and clay (˂ 0.0625 mm) were evaluated.

Enrichment factor

To evaluate the enrichment of trace metals in core sediment, the enrichment factor formula developed by Muller (1979) is used. The enrichment factor allows removing the possible influence of variations in grain size of the sediments of a geochemical background (Muller 1969; Buat-Menard and Chesselet 1979; Hakanson 1980; Hakanson 1984; Likuku et al. 2013; Weissmannova and Pavlovsky 2017; Kowalska et al. 2018; Yang et al. 2018; Nawrot et al. 2020). The enrichment of metal was quantified based on the formula in Eq. 3;

$$EF = \left(\text{Conc}_n / \text{Conc}_{ref}\right) \div \left(M_n / M_{ref}\right) \quad (3)$$

Conc.$_n$ stands for the concentration of trace metals in the environment under study, Conc.$_{\text{ref}}$ denotes the concentration of trace metals in the reference environment, M_n represents the concentration of trace metals in the reference environment, and M_{ref} represents the reference element in the reference environment. The average continental shale abundance metal contents (Turekian and Wedepohl 1961) were used as the background metal concentrations. In this study, iron was used as a normalizing reference element owing to its conservative behavior (de Mora et al. 2004). Besides, the major constituent of clay minerals is aluminum and iron (Alkarkhi et al. 2009) and also, Fe has been used as a reference metal to determine the level of trace metals contamination for some areas in Malaysia because of its abundance in sediment (Lim and Kiu 1995; Sim et al. 2016). Table 3 shows the enrichment factor value and its classification.

Table 2 The results of CRM, sewage sludge amended soil, standardized by BCR (Community Bureau of Reference)

Element	Pb	Zn	Cd	Ni	Mn	Cu	Ba	As	Co	Cr	Mg	Ca	Al	Na	Fe
CV (ppm)	171.00 ± 6.00	1059.00 ± 22.00	76.00 ± 1.20	40.60 ± 3.50	873.00 ± 12.00	137.00 ± 9.00	53.40 ± 4.10	42.00 ± 1.30	23.50 ± 2.80	415.00 ± 9.60	307.00 ± 7.10	924.00 ± 11.00	219.00 ± 6.40	89.90 ± 5.20	1072.00 ± 14.00
[a]*MV* (ppm)	171.88 ± 0.83	1061.79 ± 3.08	77.08 ± 0.42	41.86*	873.90 ± 0.91	138.01*	55.07*	42.69 ± 0.41	24.03 ± 0.31	416.51 ± 0.40	309.11 ± 0.68	926.1 ± 0.73	221.03 ± 0.84	92.03 ± 0.76	1078.35 ± 0.96
R_{Lab}—R_y	1.01	2.79	1.08	1.26	0.90	1.01	1.67	0.69	0.53	1.51	2.11	2.10	2.03	2.13	6.35
U	10.44	38.28	2.09	6.09	20.88	15.66	7.13	2.26	4.87	16.70	12.35	19.14	11.14	9.05	24.36
|Z-score|	0.10	0.07	0.52	0.21	0.04	0.07	0.23	0.31	0.11	0.09	0.17	0.11	0.18	0.24	0.26
[b]Diff (%)	0.21	0.26	1.42	3.10	0.10	0.74	3.13	1.64	2.26	0.36	0.69	0.23	0.93	2.37	0.59

CV denotes the certified value.

MV represents the measured value.

[a]Mean measured values of three determinations.

[b]Diff. - Percentage of difference to true values (Z-scores) calculated based using Eq. 1

*Values without uncertainty due to insufficient CRM sample measurement repetitions.

Table 3 Enrichment factor and their environmental intensity (Muller 1979)

The value of the enrichment factor	Environmental risk grades
EF less than 1	No enrichment
EF less than 3 but greater than 1	Minor enrichment
EF 3 to 5	Moderate enrichment
EF 5 to 10	Moderately severe enrichment
EF 10 to 25	Severe enrichment
EF 25 to 50	Very severe enrichment
EF greater than 50	Extremely severe enrichment

Statistical analysis

The comparison between average concentrations of heavy metals and standard deviations were tested for significance ($P \leq 0.05$) applying ANOVA analysis. Pearson's correlation analysis was used to quantify the relationships of different metals in the top layers of core sediments. The correlations between trace metals and between a given trace metal and the percentage portion of granulometric fractions were examined. Cluster analysis (CA) and principal component analysis (PCA) for weighted trace metals were performed to examine the sources of the heavy metals. All statistical analyses were performed using the computer program STATISTICA software (StatSoft 1999).

Results

Relationships between sediment size fractions and trace metals

Sediment samples deposited in the coastal sites and four rivers estuary of Sarawak consisted of sand (53–71%), a reasonable amount of clay fraction (11–33%), and 9–21% silt fraction (Table 4).

Core sediment collected from the Salak River estuary with sample ID CZ7 had the highest content of sand fraction (71%), 41% of the sand fraction in CZ7 sediment was finer, 23% was coarse sand, and the remained 7% was very coarse sand. The textural characteristics showed that sediment from the Salak River estuary (CZ7) is sandy. Sediments collected from the estuary of Rambungan River (CZ1) had the second-highest quantity of sand fraction (68%), 21% clay fraction; and 11% silt. It was observed that the percentage of fine sand (25%) is lower as compared to the percentage of coarse sand (i.e., 39%). The other percentage constitutes very coarse sand. The textural characteristics indicated that sediment collected from the Rambungan River estuary with sample code CZ1 is sandy loam. The percentage textural compositions of sediments collected from the remaining 2 estuaries

Table 4 Textural classification of core sediment at sampled stations

Sample ID	Locality	% Sand	% Silt	% Clay	Textural classification
CZ1	Rambungan River Estuary	68	21	11	Sandy loam
CZ2	Offshore of Rambungan River opposite to small Satang Island	53	18	29	Sandy clay loam
CZ4	Sibu River Estuary	64	25	11	Sandy loam
CZ5	Offshore of Telega Air opposite to small Satang Island	56	13	31	Sandy clay loam
CZ7	Salak River Estuary	71	09	20	Sandy
CZ8	Santubong Bay	58	16	26	Sandy clay loam
CZ10	Santubong River Estuary	61	28	10	Sandy loam
CZ11	Offshore of Santubong Resort	54	13	33	Sandy clay loam

(i.e., Sibu River estuary (CZ4) and Santubong River estuary (CZ10)) were similar to that of the Rambungan River estuary (CZ1), thus, they were also found to be sandy loam. Based on the percentages of textural compositions, it was observed that all the sediments collected from coastal sites were sandy clay loam. The order of very coarse sand is as follows: CZ7 > CZ1 > CZ4 > CZ10 > CZ8 > CZ5 > CZ11 > CZ2. During the separation of sediment samples into fractions, the larger and heavier portions of the suspended materials were found to be deposited in the sediments collected from the four Rivers estuary sites, whereas smaller and lighter materials were deposited in the sediments collected from the coastal sites. According to Farkas et al. (2007), with the decrease in flow velocity in the retention tank, the larger and heavier fractions of suspended solids are deposited near the inlet, whereas the smaller and lighter fraction settled towards the outlet. Based on the visual observation of the materials in sediment samples, granulometry confirmation, and Farkas et al. (2007) study, means that there may be a possibility of a decrease in flow velocity in the estuaries as water is moving from the rivers to join the sea. That is why larger and heavier fractions of suspended materials were assembled in the estuary sediments and smaller and lighter depositions of suspended materials were found in the coastal sediments.

Pearson correlation matrix between analyzed trace metals and granulometric fraction was performed to evaluate the inter-relationships between trace metals and sediment particle fractions (Table). Significant strong positive correlations occurred between Ba and < 0.0625 mm fraction ($r = 0.71$; $p < 0.01$), Pb and < 0.0625 mm fraction ($r = 0.74$; $p < 0.05$), Cu and < 0.0625 mm fraction ($r = 0.88$; $p < 0.05$), and Zn and < 0.0625–2.0 mm fraction ($r = 0.98$, $p < 0.05$). While significant strong negative correlations were observed between Zn and 0.0625–2.0 mm fraction ($r = -0.69$, $p < 0.01$) and Cd and 0.0625–2.0 mm fraction ($r = -0.71$, $p < 0.01$). Also, strong positive correlations were found between As and < 0.0625 mm fraction ($r = 0.61$) and Ca and < 0.0625 mm fraction ($r = 0.68$). Positive medium relationships were noticed between Al and < 0.0625 mm fraction ($r = 0.51$) and Co and < 0.0625 mm fraction ($r = 0.51$). Several studies have confirmed this relationship associated with sediment fraction > 0.0625 mm, the total trace metals in sediments decreases (Wang et al. 2016; Nawrot et al. 2020). No significant strong negative or positive inter-relationship occurred between Ni, Mn, Mg, As, Co, Ca, Al, Na, or Fe and the granulometric fractions. It has been generally noticed that trace metals are predominantly associated with the finer fraction of sediments (Silveira et al. 2016; Ji et al. 2018). This confirms the significant strong positive correlations between clay fractions and some trace metals (Pb, Zn, Cu, and Ba) and strong positive relationships between clay fractions and As or Ca as well as positive medium correlation between Al and clay fractions, unlike sand or gravel fractions.

Trend analysis of trace metal contents in core sediment of each sampled station

The average concentration of trace metals in core sediments of each sampled point is presented in Fig. 2. Because the mean levels of Fe element detected in sediment samples were extremely higher than the other trace metals, it was not included during the constructions of the mean concentration of trace metals chart in each sample in order to have a clear picture and the trend of the trace metals at each sampled point. Therefore, the average concentrations and standard deviations of Fe detected at each sediment depth/cm of each sample station of four determinations ($n = 4$) are outlined in Table 5. It was observed that the trace metal concentrations charts constructed at each sampled point were similar (Fig. 2) and thus, the content of each metal in each sediment sample are in the order: Cd < Co, As < Ba < Cr < Mn < Ni < Na < Ca < Pb < Zn < Cu < Al < Mg < Fe.

The average concentrations of trace metals detected in the coastal core sediments were higher as compared to the average concentrations of trace metals in the core sediments collected from the four rivers estuary. This could be the affirmation of the granulometry results obtained in this study and studies conducted by Silveira et al. (2016) and Ji et al.

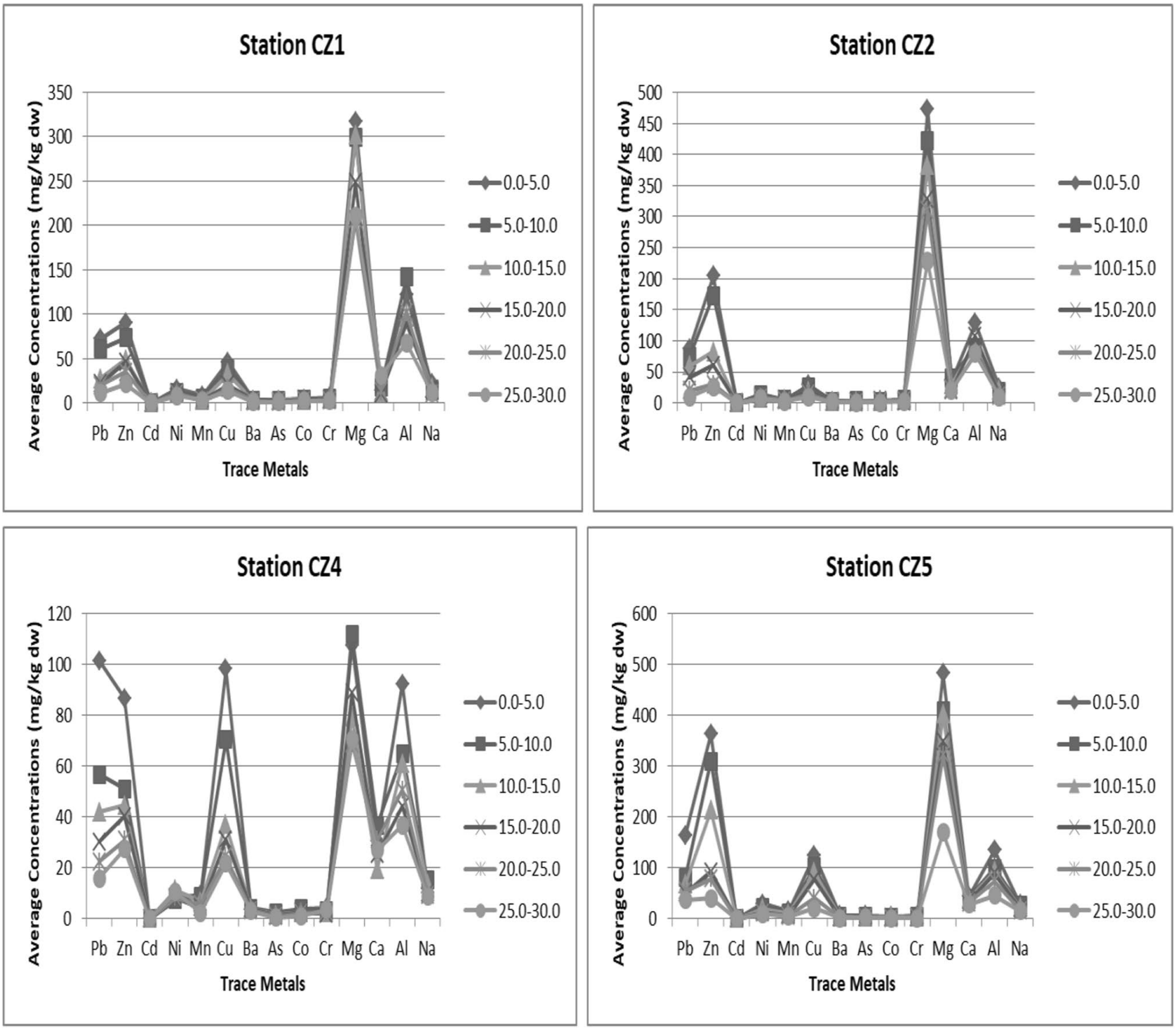

Fig. 2 Average concentration of trace metals (mg/kg d.w.) detected in core sediment samples in respective sampled stations

Table 5 Average concentration of Fe (mg/kg d.w) analyzed in core sediment samples ($n=4$)

Depth (cm)	CZ1	CZ2	CZ4	CZ5	CZ7	CZ8	CZ10	CZ11
0–5	19,142.0 ± 6.7	20,186.2 ± 6.4	16,322.2 ± 5.4	33,381.4 ± 6.3	19,244.1 ± 5.2	29,131.4 ± 6.2	24,170.3 ± 5.5	35,124.6 ± 8.9
5–10	16,033.2 ± 4.2	18,463.7 ± 4.1	14,275.9 ± 3.7	31,684.9 ± 5.9	17,168.7 ± 4.4	27,426.6 ± 3.8	23,672.7 ± 6.1	33,644.3 ± 3.6
10–15	15,188.7 ± 5.8	16,335.5 ± 10.3	14,381.4 ± 9.8	30,926.1 ± 9.7	16,301.4 ± 5.1	25,871.6 ± 5.1	23,088.1 ± 8.9	30,911.1 ± 7.9
15–20	13,946.1 ± 4.6	14,898.3 ± 9.5	13,913.7 ± 5.6	29,118.4 ± 9.6	15,397.8 ± 2.9	24,812.0 ± 3.7	22,814. ± 6.8	29,349.6 ± 4.3
20–25	13,529.4 ± 7.7	14,146.2 ± 8.8	12,636.4 ± 3.9	26,763.1 ± 9.1	15,004.4 ± 6.4	23,089.6 ± 8.9	22,077.3 ± 6.1	27,081.2 ± 5.1
25–30	12,086.1 ± 8.5	13,861.6 ± 7.9	12,011.5 ± 2.9	20,431.3 ± 5.8	14,117.5 ± 6.6	19,338.8 ± 4.1	21,462.5 ± 7.4	24,653.8 ± 6.8

(2018). The higher average concentrations of trace metals in the coastal core sediment samples and low average concentrations of trace metals in the four rivers estuary may also uphold the study conducted by Farkas et al. (2007). It was noted that the contents of trace metals detected in the top layers were higher as compared to the levels of trace metals detected in the middle and in the bottom layers sediments in almost all the studied sites. There were significant differences between the levels of trace metals detected in the top layers and in the bottom layers sediment samples and this may be due to the deposition of new trace metals because of the influence of anthropogenic activities within the vicinity and natural sources such as flood which took place 16 years ago or changes in geochemical characteristics.

Lead (Pb) concentrations ranged from 8.9 mg/kg d.w. in the bottom layer of the Salak estuary (CZ7) to 188.9 mg/kg d.w. in the top layer of the coastal site CZ8. The level of Pb in sediment collected from the Sibu River estuary is three times higher than the values reported by Omorinoye et al. (2019). The Pb concentrations detected in all locations were below the average shale value of Pb for sedimentary rock. Several natural processes may be attributed to higher Pb concentrations and these include forest fires and various erosional processes. Also, anthropogenic influence for instance mining of metallic ores results in higher concentration and dispersion of Pb (Kao et al. 2007). The highest concentration of Zn was recorded in the top layer sediment of Santubong Bay (i.e., coastal site CZ8), while the lowest concentration of Zn was detected in the bottom layer of Santubong River estuary (CZ10). Most of the Zn concentrations detected along the depth of each location were below the natural background level (100 mg/kg d.w.) except in the top to the middle layer sediments of the coastal sites and the top layer sediment of the Salak River estuary (109.2 mg/kg d.w.). This means that some extent of anthropogenic activity has taken place within the locations which recorded higher content of Zn. The potential sources of Zn are sewage sludge, motor oil and grease, transmission fluid, phosphate fertilizers, and undercoating and concrete (Monaci and Bargagli 1997; Shaari et al. 2015). The contents of Cd detected were similar in all studied sites and also along with the sediment depth (see Fig. 2). The highest concentrations of Cd were observed in the top layer of the sample collected from the offshore of Rambungan River estuary opposite to small Satang Island (i.e., coastal site CZ2). The highest concentration of Ni was detected in the top layer sediment collected from the offshore of Santubong resort (i.e., coastal site CZ11). It was observed that the concentrations of Ni detected in the sediment samples from the coastal sites CZ5, CZ8, and CZ11 were twice higher as compared to their corresponding estuaries (i.e., CZ4, CZ7, and CZ10). The concentrations of Fe varied from 12,011 mg/kg d.w. in the bottom layer of the estuary of Salak River (CZ4) to 35,124.6 mg/kg d.w. in the top layer of the coastal site (CZ11) (Table 5). Low Fe concentrations were observed in all the top layers when compared to the average shale value (47,200 mg/kg d.w.) (Turekian and Wedepohl 1961). This may be attributed to the leaching of Fe compounds from the surface layer due to rain and high salt content build-up (salinity control). In addition, when the Fe concentration detected in the top layer sediment of Estuary of Sibu river (16,322.2 mg/kg d.w.) is compared with the Fe content obtained by Omorinoye et al. (2019) in the sediment of Sibu River (4325 mg/kg d. w.), the difference in value may be attributed to anthropogenic inputs. Iron hydroxides are capable of absorbing a massive amount of trace metals by the process of cation exchange (Salomons and Forstner 1984) and iron oxides also help in holding trace metals in aquatic sediments (Horowitz and Elrick 1987; Shaari et al. 2015).

Trace metals charts shown in Fig. 2 revealed a decrease in the content of Cr with an increase in depth except station CZ2 (0–0.5 cm (4.6 mg/kg d.w.) and 5.0–10.0 cm (4.9 mg/kg d.w.)). Low concentrations of Co and Ba elements were detected in the core sediment samples in all stations. The contents of Mg ranged from 68.8 mg/kg d.w. in the bottom layer sediment of station CZ4 (20.0–25.0 cm) to 499.3 mg/kg d.w. in the top layer sediment of Santubong Bay (i.e., location CZ8). The low content of Mg in the estuarine sites could be due to the influence of river inflow on sediment transport because as the water current increases, the particle of the sediment is lifted into the water column and permits it to move from the estuarine towards the sea coast. Hence, the majority of the sediment may assemble in the coastal lane responsible for a higher level of trace metals compared to the estuary. It was noted that the concentrations of Ca in the coastal sites were higher than their counterparty estuaries, while the contents of Na detected in the coastal sites were similar to their corresponding estuaries. Inverse relationships between the average concentrations of Ca or Na and depth were observed. The higher concentration of Al was observed in the top layer sediment of the coastal site CZ11, and this may be influenced by both natural and anthropogenic origins. The movement of aluminum-rich sediment by the river flow may be the reason behind the higher levels of Al in the coastal stations. Aluminum metal is predominant in soil, rocks (mostly igneous rocks), minerals (turquoise, rubies, and sapphires), and clays (ATSDR 2008; Shaari et al. 2015). A study carried out in estuary sediments of the east coast of Peninsular Malaysia by Shaari et al. (2015), the contents for Mn (207.58–491.33 μg/g d.w.), Co (2.00–11.12 μg/g d.w.), Cu (14.49–22.33 μg/g d.w.), Zn (36.13–125.93 μg/g d.w.), Fe (6.20–8.95 μg/g d.w.), and Al (0.94–5.59 μg/g d.w.) were lower than the values obtained in this study. Regarding this study, the four Rivers Estuary are connected and joined to the coast of the South China Sea and are close to Telega Air and Sibu communities; therefore, water drainage system discharge into the rivers maybe be the reason for higher levels of trace metals as compared to the study conducted by Shaari et al. (2015).

The trend of trace metal enrichments in core sediments

Enrichments were evaluated for each of the six layers to study the variation of trace metals along with the depth/cm to help appraise the biological and environmental risk that may be associated with the surface layer. Therefore, the trends of three trace metals that are enriched along the depth/cm of the core sediments and also in all sampled stations are tabulated in Table 6. In general, higher enrichments were detected in the top layer sediments in almost all the samples, and also, sediments collected from the coastal sites were somehow enriched as compared to sediments collected from the four rivers estuary (Table 6 and see Supplementary Table S1). Though there were no notifications of significant differences of enrichment between the sediments collected from the coastal sites and four rivers estuary and along with the sediment depths. The estimated enrichment factor values obtained for trace metals in sediments occurred in the order: Pb > Zn > Cu > As > Co > Mg > Ni > Cd > Cr > Ba > Ca > Mn > Na > Al (Table 6 and see Supplementary Table S1). Out of the 15 trace metals analyzed in the sediment samples, only 3 (i.e., Pb, Zn, and Cu) were enriched in all sampled sites and along all depths (Table 6 and 7).

The other 12 trace metals (i.e., Cd, Ni, Mn, Ba, As, Co, Cr, Mg, Ca, Al, and Na) were not enriched in all sampled stations and all depths (see Supplementary Table S1). The values of EF estimated for Pb were higher in all sediments at all sampled stations. In regards to the Rambungan River estuary (CZ1), the enrichment values for Pb at depth: 5.0–10 cm, 10.0–15.0 cm, 15–20 cm, and 20–25 cm were > 3 which suggest moderate enrichment while at the depth of 0–5.0 cm showed minor enrichment of Pb. Concerning sediment collected from the offshore of Rambungan River estuary opposite to small Satang Island (CZ2), the estimated EF values at a depth of 0–5 cm, 5–10 cm, 10–15 cm, and 15–20 cm

Table 6 Enriched trace metals in the core sediment samples at all sampled stations

Station/depth (cm)	CZ1	CZ2	CZ4	CZ5	CZ7	CZ8	CZ10	CZ11
Pb								
0–5.0	2.22	10.14	11.22	14.66	5.76	14.94	7.56	6.19
5.0–10.0	8.93	9.36	6.00	9.32	4.15	9.01	6.10	5.25
10.0–15.0	4.21	9.25	4.98	6.88	3.14	5.40	5.62	4.03
15.0–20.0	3.67	6.53	3.96	5.13	2.84	2.54	2.35	3.84
20.0–25.0	3.38	3.24	4.18	4.66	2.64	3.14	2.04	2.87
25.0–30.0	2.21	1.92	3.10	4.09	1.49	3.86	1.47	2.75
Zn								
0–5.0	2.35	5.30	2.64	5.39	2.82	7.37	1.82	5.40
5.0–10.0	2.28	5.34	1.77	4.86	2.34	7.33	1.41	4.76
10.0–15.0	1.60	2.68	1.54	3.41	1.93	6.75	0.92	3.34
15.0–20.0	1.61	2.20	1.43	1.56	1.68	5.44	0.72	2.73
20.0–25.0	1.28	1.09	1.21	1.45	1.33	4.43	0.60	2.29
25.0–30.0	0.88	1.09	1.13	0.98	0.83	2.60	0.45	2.07
Cu								
0–5.0	2.54	1.49	6.31	3.91	2.21	3.22	4.39	3.98
5.0–10.0	2.53	1.48	5.17	3.35	2.00	2.71	2.85	3.42
10.0–15.0	2.25	1.18	2.68	3.08	2.12	2.48	2.15	3.20
15.0–20.0	1.53	1.35	2.35	2.79	1.53	2.25	1.67	1.42
20.0–25.0	1.29	1.09	2.02	1.56	1.41	1.91	1.35	1.17
25.0–30.0	1.21	0.71	1.90	1.03	1.06	1.97	1.07	1.12

Table 7 Pearson correlation matrix between analyzed trace metals and sediment granulometric fraction expressed in percentage of the top layer of the core sediment samples

Particle fraction	Pb	Zn	Cd	Ni	Mn	Cu	Ba	As	Co	Cr	Mg	Ca	Al	Na	Fe
˂ 0.0625 mm	0.74*	0.98*	0.36	0.16	0.29	0.88*	0.71**	0.61	0.51	0.41	0.41	0.68	0.51	0.24	0.28
0.0625–2.0 mm	− 0.39	− 0.69*	− 0.71*	− 0.21	0.26	− 0.17	− 0.21	− 0.44	− 0.51	0.52	− 0.41	− 0.24	0.25	− 0.56	0.06

*Correlation is significant at the 0.5 level (2-tailed).

**Correlation is significant at the 0.1 level (2-tailed).

were above 5 indicating moderately severe enrichments of Pb. The bottom layer sediments (20–25 cm and 25–30 cm) at station CZ2 showed a decrease in values of enrichment for Pb as compared to the top layer and the middle layer sediments, although observation of minor enrichment was noticed in the bottom layer sediments. In a comparison of the Sibu River estuary (i.e., location CZ4 which serve as an immediate inlet) to the offshore of Telega Air opposite to small Satang Island (i.e., location CZ5 which serve as the outlet of the Sibu River estuary), it was noticed that there were higher enrichments of Pb in all layers of sediment collected from the coastal site CZ5 than their corresponding layers sediment collected from the Sibu River estuary. It was also observed that the calculated values of EF for Pb in the topmost most layer sediments of both sites (i.e., CZ4 and CZ5) were > 10, indicating severe enrichment. Also, at a depth of 5–10 cm, it was found that the enrichment values for Pb of both sites (i.e., CZ4 and CZ5) were > 5, suggesting moderately severe enrichments. There was a notification of an inverse relationship between the enrichments and depth in almost all the sites (i.e., enrichment value decreases as the depth increase and vice versa). Considering sediment from the Santubong River estuary (CZ7) and sediment from Santubong Bay (i.e., coastal site CZ8), it was observed that the enrichments for Pb decrease along with the depth. The EF results obtained in the topmost layer sediment from the coastal site CZ8 > 10, indicating severe enrichment while the calculated *EF* value of Pb in the topmost layer sediment of Salak River estuary > 5, suggesting moderately severe enrichment. At a depth of 5–15 cm, there was an observation of moderately severe enrichment of Pb in the sediment at the coastal site CZ8 and minor enrichment of Pb in the sediment collected from the Salak River estuary. Minor enrichments of Pb were noticed at the depth of 15–30 cm of a sediment sample from the coastal site CZ8, whereas no enrichment of Pb was observed at a depth of 15–30 cm of a sediment sample from the Salak River estuary. In regards to the sediment sample from Santubong River estuary (CZ10), and the sediment sample from the offshore of Santubong resort (i.e., coastal site CZ11), moderately severe enrichment of Pb was observed at a depth of 0–15 cm. Minor enrichments of Pb were noticed in the bottom layer samples collected from both stations (i.e., CZ10 and CZ11).

The enrichments of Zn decrease with an increase in depth at all sampled sites. The calculated *EF* values for Zn in the top layers sediment collected from four rivers estuary (i.e., CZ1, CZ4, CZ7, and CZ10) were in the range of 1 to 3, indicating moderate enrichments, whereas the estimated *EF* values for Zn in the top layer of sediments from the coastal sites (i.e., CZ2, CZ5, CZ8, and CZ11) were in the range of 5 to 10, suggesting moderately severe enrichments. Minor enrichments to no enrichment of Zn were recorded in the bottom layer sediments at all sampled sites. Surprisingly, the calculated values of *EF* for Cu in the top layer sediments from the four Rivers estuary (i.e., CZ1, CZ4, CZ7, and CZ10) were higher than their corresponding coastal sites (i.e., CZ2, CZ5, CZ8, and CZ11). Moderate enrichments of Cu were recorded in the top layer sediments at all sampled sites except the Sibu River estuary site (CZ4) which showed moderate-severe enrichment. Minor enrichments to no enrichments of Cu were recorded in the bottom layer sediments at all sampled sites. Generally, no enrichment of Cd, Ni, Mn, Ba, As, Co, Cr, Mg, Ca, Al, and Na were observed in all sediment samples at all sampled stations (see Supplementary Table S1). Enrichments of Pb, Zn, and Cu in the sediment samples showed that these metals may exhibit toxic impacts in the sediment biota by negatively influencing key microbial processes and decrease the number of sediment microorganisms in the studied area (Yang et al. 2018; Xu et al. 2019).

Possible sources of trace metals contamination

Inferences concerning the likely sources of trace metals in sediments of the coastal and four rivers estuary were developed using the Pearson correlation coefficients, dendrogram from the cluster analysis, and plot loading components from principal component analysis within and between trace metals detected at all sampled sites.

Pearson's correlation analysis

Pearson's correlation analysis was used to assess the interrelationship between measured metals in the surface layer (0–5 cm) sediments. Significant strong negative and positive correlations between some trace metals detected in the surface layer sediments are illustrated in Fig. 3.

Significant strong negative correlations occurred between Pb and Ni (r= -0.89; $p < 0.05$) and Ba and Al ($r = -0.96$; $p < 0.01$), whereas significant strong positive correlations were observed between As and Fe ($r = 0.96$; $p < 0.05$) and Mg and Na ($r = 0.97$; $p < 0.01$) (Fig. 3). It was noted that there were strong positive inter-relationships between the following metals: Zn and Mg ($r = 0.65$), Na and Zn (r = 0.62), Ni and Cd ($r = 0.85$), Cr and Ni ($r = 0.79$), Mg and As ($r = 0.61$), and Fe and Na ($r = 0.61$) but the correlations were not significance. Medium positive correlations were documented between Al and Pb ($r = 0.50$), Al and Zn (r = 0.53), and Na and As ($r = 0.57$), but these relationships were not significance. These results suggest that the sediments of the coastal and four rivers estuary could be receiving multiple contaminants from the same emission sources or similar sources (Liang et al. 2017). On contrarily, Zn, Cd, Mn, Cu, and Ca showed no strong positive or negative correlation with the other trace metals (see Supplementary Table S2). The significant strong positive inter-relationships between As and

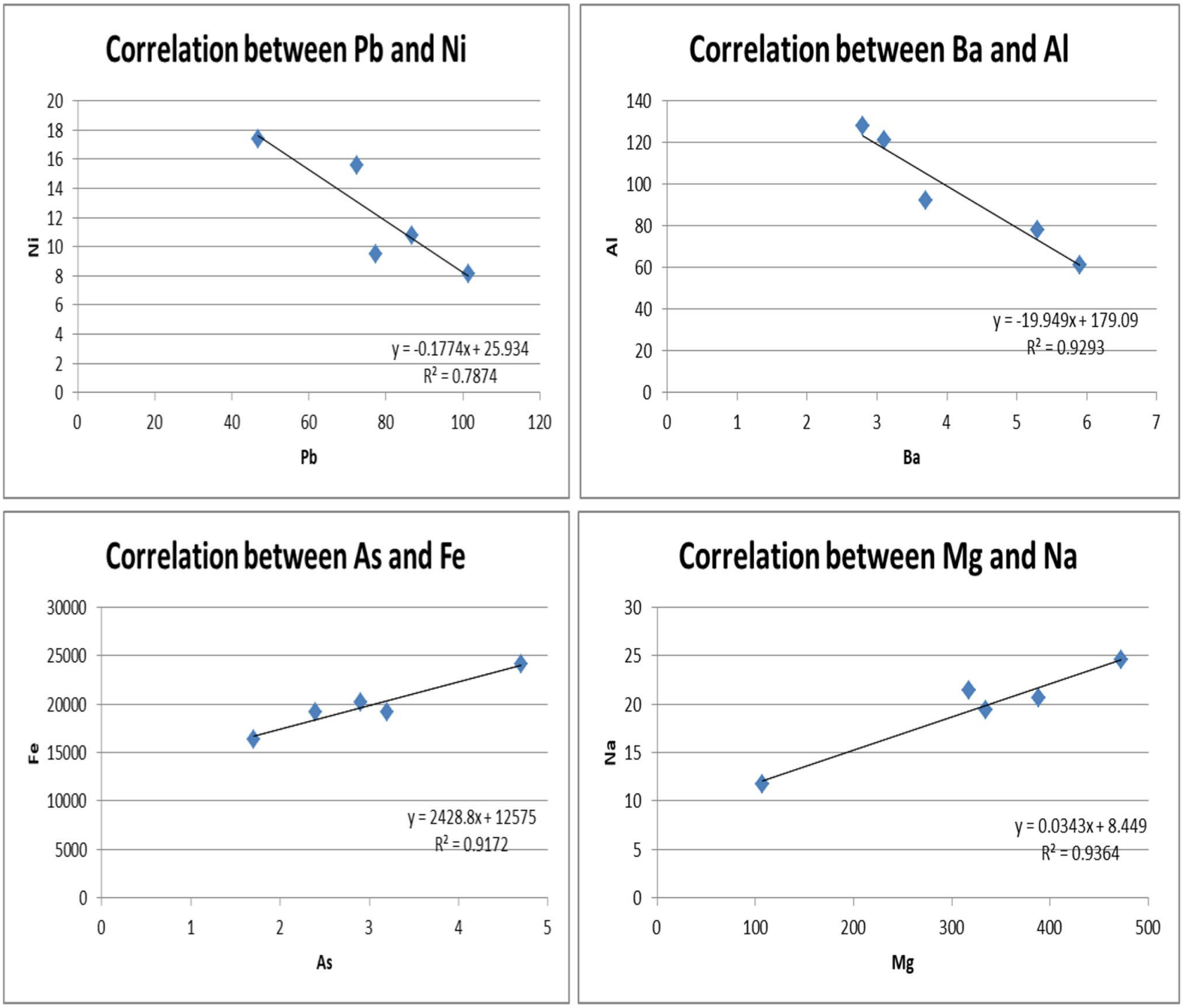

Fig. 3 Significant strong negative and positive correlations between Pb and Ni, Ba and Al, As and Fe, and Mg and Na in the surface layer of the core sediments

Fe and Mg and Na suggests common contaminants sources (Yang et al. 2012, 2018; Gupta et al. 2014) or similarity in geochemical features (Pandey and Singh 2015; Tian et al. 2017; Tiana et al. 2020). The significant inverse correlations between Pb and Ni and Ba and Al indicate different external inputs (Zarei et al. 2014; Kanda et al. 2018; Saleem et al. 2020). The coexistence of As and Fe and Mg and Na within an agricultural, tourism, and industrial catchment indicates agronomic inputs for example the excessive application of pesticides and phosphate fertilizer, tourism sources such as oil spillovers from boat activities and other human activities, and industrial inputs including effluents and waste discharges, which can enter the river to the coast through soil runoff (Ke et al. 2015; Wang et al. 2015; Gholizadeh et al. 2019; Fakhradini et al. 2019).

Cluster analysis

The dendrogram obtained using Ward Linkage of cluster analysis for the average concentrations of trace metals in the surface layers of the core sediments at each sampled point is shown in Fig. 4.

Based on the dendrogram, two clusters were displayed. Cluster 1 consists of As, Co, Ba, Cr, Mn, Cd, Ni, Na, Pb, Cu, Ca, Zn, Al, and Mg while cluster 2 is a cluster of Ca Zn, Al, Mg, and Fe metals. The appearance of Ca, Zn, Al, Mg, and Fe metals in cluster 2 may be due to their abundance in the sediment samples at the sampled sites when compared to the other trace metals (Yang 2012; Xu et al. 2019). Furthermore, because Ca, Zn, Al, Mg, and Fe metals were in the same cluster also indicate their common source. Trace

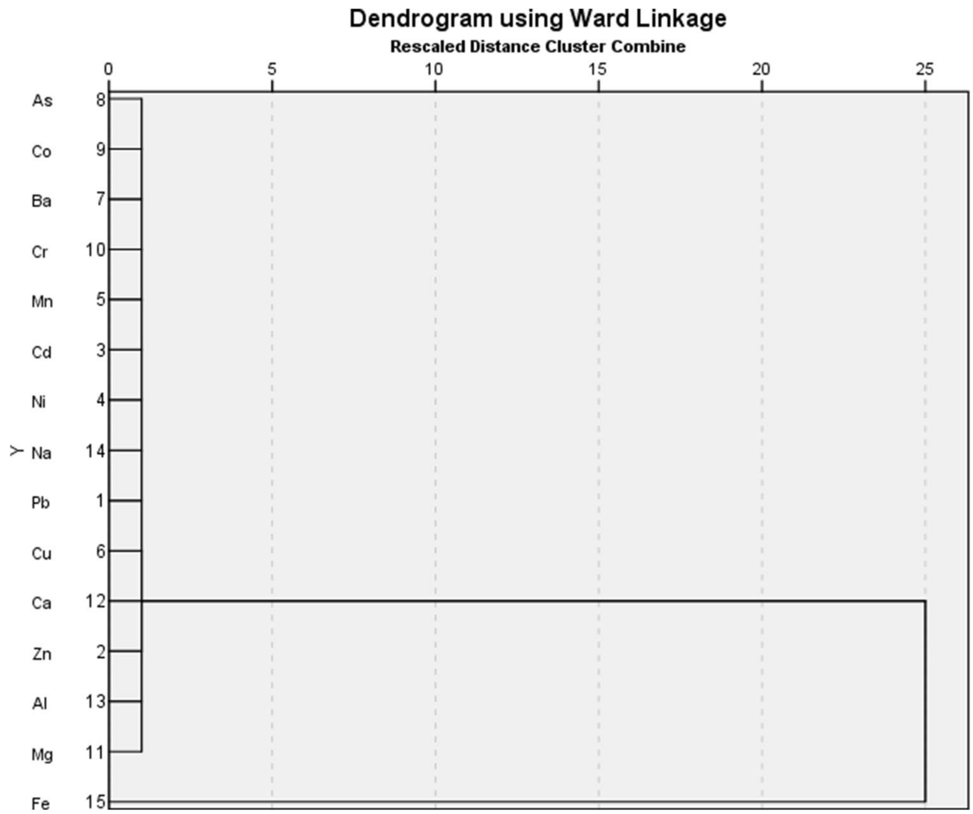

Fig. 4 Cluster analysis of average total trace metals concentrations in the core sediments at all sampled stations

metals in cluster 1 also suggest mutual inter-relationships between them and maybe originated from similar sources (Chien et al. 2002; Dias-Ferreira et al. 2016; Bhuyan et al. 2019).

Principal component analysis

While a Pearson correlation analysis (see Supplementary Table S2) can be used to make references about input sources for metals, it is a comparatively facile analysis given the complexity of the estuarine and coastal environment. Thus, PCA was employed in the data from the coastal and four rivers estuary sites, because its flow patterns, and consequently its dispersion of trace metals, are known to be especially complex. Plot loading of three principal components in PCA results which explains the relationships between the selected trace metals is depicted in Fig. 5.

The principal component analysis (PCA) extracted three components with eigenvalues describing 83.28% of the total variance. The results of the PCA confirmed with the results obtained from the cluster analysis with component 1 that accounted for 32.13% of the total variance with high positive loadings for As (0.69), Mg (0.99), Na (0.95), and Fe (0.79), and high negative loading for Mn (−0.88). The combinations of these trace metals in component 1 indicate similar anthropogenic sources because Fe in this component had values exhibiting enrichment. Also, the inverse correlation of the high positive loading metals and Mn is an indication of different external inputs (Li and Zhang, 2010; Decena et al. 2018). Component 2 accounted for 28.41% of the total variance with high positive and negative loadings for Cd (0.874), Ni (0.873), and Pb (−0.886). Component 2 could originate from industrial activities because Cd is likely to be from printing, electronics, and dyeing, chemical, and electroplating industry sources (Wu 2014). Additionally, mineral weathering and atmospheric deposition from coal-combustion dust may be attributed to the accumulation of Ni in sediments (Liang et al. 2017). The inverse correlation of Pb with Cd and Ni is evidence of different external inputs sources (Decena et al. 2018). The external inputs could be originating from agricultural, industrial, pharmaceutical, geogenic, domestic effluents, and atmospheric sources. Component 3 accounted for 22.73% of the total variance with high positive loading for Al (0.808), indicating that Al is derived from natural sources such as weathering of parent rocks and minerals. The results obtained from the analysis of PCA, which showed small positive or negative loadings, suggest weak relationships between metals, whereas large loadings are indications of strong relationships between trace metals (Attia and Ghrefat 2013). The results of multivariate analyses that exhibited strong positive inter-relationships between studied metals are indications of common pollutants sources or similarity in geochemical characteristics.

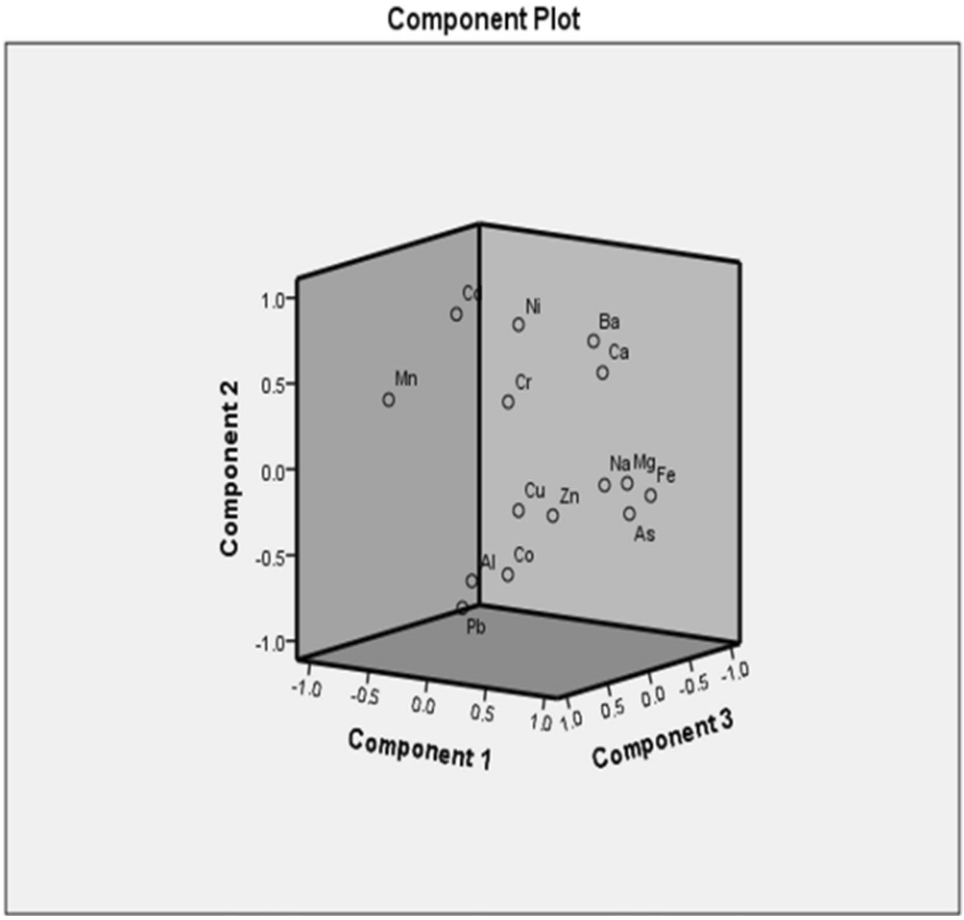

Fig. 5 Plot of loading of three principal components in PCA results

Discussion

Nevertheless, the single factor pollution index (enrichment factor) method has been extensively used; it is only useful to a single pollutant and does not necessarily consider the mixture of trace metals usually available in the contamination conditions but it has helped to determine how much the presence of a metal in sampling sites has increased relative to average natural abundance due to human activity. In general, all sampled sites are enriched with Pb, Zn, and Cu. The enrichment of Pb and Zn may be due to ore smelting centers and waste dumps from non-ferrous smelters close to the water bodies or sewage discharge from the processing of Zn-containing minerals, chemical enterprise, the wear and tear of automobile tires, and the manufacture of metal machinery (Guan et al. 2017). The enrichment of Pb may be attributed to the industrial utilization of minerals containing Pb and the burning of fossil fuels. The enrichment of Cu could be due to the urbanization of the area that leads to the gradual growth of anthropogenic processes resulting in the high concentrations of Cu in the sediments until the surface which is the latest deposition occurs. Urban runoff and vessel maintenance activities may also be the reasons for Cu enrichment. The source distribution by Pearson correlation connected with cluster analysis and PCA suggested diverse sources in sediments of the coastal and four rivers estuary, with As, Fe, Mg, Cd, and Na being thought to originate from industrial activities and Cu thought to be mainly from the emissions of traffic due to urbanization. Thus, it must be noted that this is not formal source distribution and is dependent on deduction drawn from the available information.

The coastal and four rivers estuary in Sarawak are important aquatic areas and therefore their sediment environment quality will affect the security of water for drinking and irrigation for inhabitants and agricultural production, micro, and macrobenthos activities, fishes, and other aquatic organisms along the river's estuary to the coast. Therefore, dissimilar regulatory actions should be paid to the environment treatment in the future for the coastal and selected estuaries based on the correspondent contamination features. Upgrading and control in agricultural management might be the adapted plan for lessening the input of contamination sources in sediments from the coastal and four rivers estuary, where the most essential input sources appear to mainly be from farming activities. However, the industrial sites, located in the upper and middle reaches of the rivers, seem to be the preference to control and management for reducing the inputs of contaminants from the wastewater discharge and atmospheric deposition plus avoiding the negative impact on population density zones in the lower reach.

Conclusion

The study evaluated the concentrations of trace metals predominantly associated with human activities: Pb, Cd, Ni, Ba, As, Co, Mg, Zn, Mn, Cu, Cr, Ca, Al, Na, and Fe in core sediments from the coastal and four rivers estuary sites in Sarawak. The study also looked at other factors such as sediment particle fractions and inflow velocity influencing the availability of metals in sediment. The assessment of the mobility of trace metals in the studied environment was established by the sequential extraction technique. Enrichment factors were calculated to assess the trend of enrichment of metals in each sampled station and also along with the sediment depth. To predict the sources of trace metals contamination in the studied sites, multivariate statistical analyses were used. This research work contributes to the evaluation of urbanization's and other anthropogenic activities influence on coastal and rivers estuary bodies and could be needed in establishing quality limits and planning alleviation programs. In this instance, the coastal and four rivers estuary (i.e., Rambungan, Sibu, Salak, and Santubong Rivers) in Sarawak can be considered as an ideal urban rivers and coastal site. In addition, the results of this research are not only for the interest of the local but as well have extensive possible future impacts.

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Author contributions EAA, ZA, and RW conceived of the study and carried out the design of the experiment. EAA and ZA carried out the sample preparation and analysis; EAA, TB, and SSD assessed the data; and EAA, ZA, and RW helped to draft and edited the manuscript. The author(s) read and approved the final manuscript.

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Data availability All data generated or analyzed during this study are included in this paper.

Declarations

Competing interests The authors declare no competing interests.

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