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Eco-toxic risk assessment and source distribution of trace metals in surface sediments of the coastal and in four rivers estuary of Sarawak

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Abstract

Background: Trace metals pollution in the coastal and estuarine sediment could harm water quality and aquatic organisms, leading to potential long-term health risks on the environment and humans. Thus, the purpose of this study was to conduct an assessment of selected trace metals in surface sediments of the coastal and in four rivers estuary in the Sarawak State of Malaysia to investigate their distribution, environmental risk, and potential source distribution.

Results: Average concentrations of trace metals in sediment increased in the following order: Cd < As < Co < Cu < Ni < Cr < Zn < Mn < Mg < Fe. The enrichment, contamination, and geo-accumulation index results showed that surface sediments were polluted with Zn and Mg. In contrast, the other metals (i.e., As, Fe, Mn, Ni, Cr, Cu, Co, and Cd) indicated background concentration to minor contamination. Generally, the pollution load index values showed that almost all the sampling sites were unpolluted with the selected trace metals. Sediment quality guidelines (SQGs) and risk indexes were employed to assess the ecotoxicological risk of trace metal contamination in the sediments. The results proved that studied trace metals are not likely to have a deleterious impact on bottom-dwelling organisms. Still, a further accumulation of trace metals such as Zn, Ni, and Cr with time may adversely affect bottom-dwelling organisms. The risk index results showed a low ecological risk to the study sites. The correlation analysis and principal component analysis indicated that nine studied trace metals have strong interrelationships, suggesting common pollution sources or similar geochemical characteristics.

Conclusions: The study highlights the need to make tremendous efforts to monitor and control trace metal pollution in the coastal and estuarine areas.

Keywords: Trace metals, Eco-toxic risk, Surface sediments, Pollutants, Bottom-dwelling organisms

1 Background

Trace metals are a pressing concern regarding pollution in aquatic ecosystems because of their persistence, bioaccumulation, environmental toxicity, etc. [1–3]. Aquatic bodies, for example, reservoirs [4], rivers [5, 6],

estuaries and coastal [7, 8], wetlands [9], and lakes [10], receive trace metals in inadequately treated or untreated wastewater from agricultural, domestic, and industrial sources. As an essential constituent in riverine and estuarine environments, sediments are a source and a sink of trace metals [11, 12]. Trace metals entering rivers and estuaries rapidly deposit into the sediment and are much more concentrated than in the water body of riverine, estuarine, or coastal systems [13, 14]. When there is an alteration of the hydrological or the physicochemical

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conditions, trace metals in the sediment may resuspend or desorb to result in secondary pollution in the water body [15, 16]. Trace metals accumulation in the sediment directly influences benthic organisms. Trace metals in sediment also affect many other organisms via the food chain and threaten the comfort of the aquatic environment. Thus, it is necessary to evaluate and appreciate the accumulation and distribution of trace metals in sediment.

Rambungan, Sibul, Salak, and Santubong Rivers are key rivers in the Sarawak state of Malaysia. They flow through the Kuching city and support many less densely populated towns, offering services to both agriculture and industry. Over the past few decades, there have been trace metals pollution in riverine, estuarine, and coastal sediment of the Sarawak state of Malaysia because of the wastewater discharge from agricultural, metallurgical, and mining industries carrying trace metals. Even though many researchers have studied trace metals pollution in the Rambungan, Sibul, Salak, and Santubong Rivers and their associate estuary [17, 18]; a systematic study is still lacking to correlate the sediment with trace metals distribution, characteristics, risk assessment, likely sources, and their effect on the aquatic environment.

Geochemical indices used for ecological risk assessment of trace elements in surface sediments comprised of computation of enrichment factor (EF), contamination factor (CF), geo-accumulation index (I_{geo}), pollution load index (PLI) [19–21], potential ecological risk indices [22], and excess regression analysis [20, 23]. Analyses of trace metals in river sediments have been used considerably for the essence of pollution monitoring [19, 24, 25]. A handful of these studies focused on the paths of imparting trace metals content in sediments without evaluating their ecotoxicological risks [17, 18]. Therefore, this study aimed to determine ten trace metals (i.e., Zn, As, Fe, Mg, Mn, Ni, Cr, Cu, Co, and Cd) in surface sediments of the coastal and in four rivers estuary of the Kuching Division of Sarawak. Also, using the geochemical indices to evaluate the contribution of anthropogenic activities carrying trace metals into estuary and coastal sediment. Determining potential risks linked with trace metal toxicity using sediment quality guidelines and ecotoxicological risk index. Finally, ascertaining the source distribution of trace metals using statistical tools such as Pearson's coefficient correlation and principal component analysis.

2 Methods

2.1 Description of the study area

The field studies and sampling were conducted in the coastal and in four rivers (i.e., Santubong, Salak, Sibul, and Rambungan) estuary in the Kuching Division of Sarawak in the North-Western portion of Borneo Island,

Malaysia. The study area has a tropical rainforest climate, moderately hot yet very humid once in a while, and receives considerable rainfall. The mean annual precipitation is approximately 4200 mm. The study area receives an average of 247 rainy days per year with an average of 6 h of sunshine and a mean of 3.7 h per day during January. North-East Monsoon months of November to February is the wettest time while June to August is considered the driest months. The temperature of the study area range between 19 to 36 °C with a mean temperature of roughly 23 °C in the early hours of the morning and rises to approximately 33 °C in the course of mid-afternoon and can reach 42 °C in the dry season. Figure 1 shows the sampling locations of the coastal and four rivers estuary.

2.2 Sediment sampling and treatment

The field studies and sample collection were conducted according to the procedure outlined by Gao [26] from September to October 2020. The coordinates for each sampling site were determined using a portable Global Positioning System (GPS), Garmin etrex (Table 1). Sixteen surface sediments were collected from sampling sites. Samples were collected at each sampling site at approximately 5.0 cm depth using a Wildco grab sampler. To minimize contamination, the grab sampler was disinfected using biodegradable detergent and rinsed with deionized water before and after each use. Sediments were placed in cleaned polyethylene containers and kept in the cooler box at a temperature of 5 °C during transportation. In the laboratory, samples were air-dried in a well-ventilated area for a week. The bulky materials such as stones and unwanted materials were removed from the sediments using stainless-steel forceps and homogenized. The dried sediment samples were pulverized into fine particles using a mortar and a pestle. Pulverized sediments were then sieved using a 55 µm mesh size sieve to obtain powdered sediments which then placed in cleaned polyethylene containers. The samples were kept in a refrigerator for further analysis [27].

2.3 Sediment extraction and analysis

The procedure used to analyze sediment absolute trace metal contents is acid digestion adapted by Hossner [28]. A powdered sample of 0.5 g was placed in a crucible. About 3 mL of H₂SO₄ (95%) and 4 mL of HCl (96%) were added to the crucible contents. The contents of the crucibles were then placed in an oven Memmert model 30 – 70 (UN 30) at 115 °C for 20 h to break down all organic materials and the weight was recorded. The sediment samples were reheated at 500 °C for 3 h 30 min in a muffle furnace (Model Ney Vulcan D – 550 series) and the weights were recorded. 2.0 mL of distilled water and

Table 1 Coordinates of sampling locations

Sample site code	Locality	Coordinates
CZ1(1),(2)	Rambungan River Estuary	N01°41'37.7" E110°08'24.5"
CZ2(1),(2)	Offshore of Batang Rambungan opposite small Satang Island	N01°44'46.8" E110°08'45.4"
CZ3(1),(2)	Offshore of Batang Rambungan adjacent big Satang Island	N01°46'22.6" E110°08'37.8"
CZ4(1),(2)	Sibu River Estuary	N01°44'46.8" E110°08'45.4"
CZ5	Offshore of Telaga Air opposite to small Satang Island	N01°45'50.4" E110°11'30.2"
CZ6	Offshore of Telaga Air adjacent big Satang Island	N01°47'23.5" E110°10'39.7"
CZ7(1),(2)	Salak River Estuary	N01°40'41.1" E110°16'59.2"
CZ8	Santubong Bay	N01°42'45.7" E110°27'63.1"
CZ9(1),(2)	Offshore of Santubong Resort	N01°44'49.6" E110°29'72.3"
CZ10	Santubong River Estuary	N01°42'32.6" E110°19'02.3"

2.0 mL concentrated HNO_3 were added to cool the samples and further heated to dryness on a hot plate (Model Favorit HS070V2 Serial 5434). 10 mL distilled water and 1.0 mL concentrated HCl were added to each crucible's content and stirred for 3 min for uniformity. The mixture was then filtered through Whatman No. 42 filter paper, and the content (filtrate) was top up with distilled water to 100 mL. Trace metals concentrations were analyzed using an inductively coupled plasma optical emission spectrophotometer (ICP-OES).

2.4 Quality assurance and control (QA/QC)

To ensure the quality and efficiency of the instrumental outcomes; QA and QC techniques were established. It includes cleaning laboratory materials and apparatus using 15% H_2SO_4 , applying standard operating methods, analyzing blanks, and standard calibration and recovery of actual additions. Instrumental validation is a crucial analytical precondition of the multi-elemental analysis process. The procedure lays out a system for describing the efficiency of the instrument. In addition, it proves the performance abilities of the procedure being examined by ensuring that it is coherent with the used method [6]. The limit of detection (LOD) and limit of quantification (LOQ) were calculated for the ten trace metals after establishing the calibration curves and equation of ICP-OES. The LOD and LOQ values were computed using the formulae adapted by [29]:

$$\text{LOD} = 3.3 \times (\text{S.d}_{\text{blank}}/y) \quad (1)$$

$$\text{LOQ} = 10 \times (\text{S.d}_{\text{blank}}/y) \quad (2)$$

where S.d refers to mean standard deviation of blanks, and y represents the sensitivity of the calibration curve [29]. Table 2 shows the properties' performance of the ICP-OES calibration equation.

Table 2 Characteristics performance of ICP-OES calibration equation

Trace metal	Parameters				
	R^2	Slope	Intercept	LODs (mg/kg)	LOQs (mg/kg)
Zn	0.9880	0.0114	0.0012	0.0404	0.1224
As	0.9910	0.0136	0.0014	0.0379	0.1149
Fe	0.9720	0.0371	0.0206	0.0106	0.0321
Mg	0.9830	0.0131	0.0017	0.0143	0.0143
Mn	0.9870	0.0211	0.0043	0.0348	0.1055
Ni	0.9910	0.0117	0.0013	0.0330	0.1000
Cr	0.9690	0.0431	0.0073	0.0429	0.1300
Cu	0.9950	0.0201	0.0134	0.0541	0.1639
Co	0.9990	0.0114	0.0036	0.0101	0.0306
Cd	0.9670	0.0633	0.0073	0.0132	0.0400

LOD limit of detection, LOQ limit of quantification, R^2 variance

The method efficiency for the study was determined using recovery tests. Analyte for individual trace metal was used to spike the digestion sample solution. The spiked samples were analyzed, and the average concentration of trace metal after spiking was compared with the concentration of analyte trace metal before spiking. The recoveries were calculated as follows [6]:

$$\text{Recovery(\%)} = (\text{Average Conc.}_{\text{spiked}} / \text{Analyte Conc.}) \times 100\% \quad (3)$$

The amount of trace metals concentrations was assessed according to the formula:

$$\text{Final Conc. (mg/L)} = \text{Conc.}_{\text{sediment}} \times \text{DF} \times \text{NV (mL)} \quad (4)$$

where, $\text{Conc.}_{\text{sediment}}$ represents trace metal concentration in sediment samples, DF denotes dilution factor, and NV is nominal volume.

The percentage recoveries of the spiked samples analyzed varied from 89 – 104% (Table 3). The analyte level of approximately 1 mg/kg has been reported, and the acceptable recovery range is 90.0–110% [6, 30, 31]. The obtained values in this current study were found to be within the acceptable recovery range. Hence, the analytical instrument is suitable for analysis of selected trace metals.

2.5 Environmental assessment of trace metals contamination

2.5.1 Enrichment factor (EF)

EF deals with estimating the anthropogenic impacts on the media, such as soil, sediments, and others. Iron (Fe) is used as a normalization metal because the collected sediments from the study area were abundant in Fe. The EF can be calculated by Eq. 5 proposed by Muller [32].

$$EF = \left(\frac{X_n}{Fe} \right)_{\text{sample}} \div \left(\frac{X_n}{Fe} \right)_{\text{crust}} \quad (5)$$

where, $(X_n/Fe)_{\text{sample}}$ represents ratios of arithmetic average concentrations (mg/kg, dry wt) of the target heavy metals; $(X_n/Fe)_{\text{crust}}$ denotes Fe in the investigated sediments and continental earth crust according to Muller [32]. The classifications of trace metals enrichment and their environmental risk grades in soil/sediment are shown in Table 4.

2.5.2 Contamination factor (CF)

CF is applied to assess pollution in an aquatic ecosystem by a given toxic substance. Thus, it serves as a vital indicator of sediment contamination [33]. The CF was computed using Eq. 6 formulated by Hakanson [34] as:

$$CF = (C_m)_{\text{sample}} / (C_m)_{\text{background}} \quad (6)$$

$C_{m_{\text{sample}}}$ denotes the concentration of a given trace metal in sediment, and $(C_m)_{\text{background}}$ is the values of reference metal, which are the average shale values of each study metal for sedimentary rock [35]. The CF values are grouped into four groups as listed in Table 4.

2.5.3 Geo-accumulation index (Igeo)

Igeo makes it possible to determine pollution by comparing the present and pre-industrial concentrations of trace metals in the earth's crust [32]. The geo-accumulation index (Igeo) can be calculated using Eq. 7 [32]:

$$I_{\text{geo}} = \log_2(C_n / 1.5 * B_n) \quad (7)$$

where, C_n refers to the measured concentration of the trace elements in the sediment samples and B_n represents the geochemical background value in the earth's crust. The factor of 1.5 is introduced to reduce the impact of possible variations in the background values due to lithogenic variations (i.e., alteration during early chemical reactions within freshly deposited sediment) [36]. The interpretation of the Igeo values is summarised in Table 4.

2.5.4 The pollution load index (PLI)

PLI for each sampling site is derived as the n th root of n number multiplied together by the values of the CF suggested by Tomilson et al., as shown by Eq. 8 [37].

$$PLI = (CF_1 * CF_2 * CF_3 * \dots * CF_n)^{1/n} \quad (8)$$

where, n represents the number of heavy metals. PLI index is ranked into several classes, as shown in Table 4.

2.6 Ecological risk assessment

2.6.1 Sediment quality guidelines

Sediment-quality guidelines (SQGs) were developed in Australia and New Zealand in 2000 to predict the adverse biological impacts caused by contaminated sediments [38]. The technique has been employed to determine the potential risk to aquatic organisms due to trace metal pollution in aquatic bodies [39]. The assessment is established by comparing the measured trace metal contents in sediment samples with the consensus-based threshold effect concentration (TEC), probable effect concentration (PEC) values, and midway values between the TEC and PEC (i.e., MEC) [40].

2.6.2 Threshold effect concentration (TEC)

TEC is a sediment contamination concentration at which a toxic response has begun to be observed in benthic organisms. Florida Department of Environmental Protection developed Eq. 9 to determine TEC based on the

Table 3 Method efficiency data for analysis of ten trace metals in spiked surface sediment samples

Trace metal	Analyte Conc. (mg/kg)	Mean Conc. of a spiked sample (mg/kg)	% Recovery
Zn	1.00	0.98	98
As	3.00	2.75	92
Fe	2.00	2.08	104
Mg	1.00	1.01	101
Mn	2.00	1.95	98
Ni	2.00	1.78	89
Cr	2.00	1.98	99
Cu	1.00	0.97	97
Co	2.00	1.94	97
Cd	2.00	1.96	98

Table 4 Enriched factor (EF), contamination factor (CF), geo-accumulation index (Igeo) and pollution load index (PLI) range values and their environmental risk grades

Index type	Value	Environmental risk grade	References
Single Indices			
EF			[32]
	0 – 1	Background concentration or no enrichment	
	1 – 3	Minor enrichment	
	3 – 5	Moderate enrichment	
	5 – 10	Moderately severe enrichment	
	10 – 25	Severe enrichment	
	25 – 50	Very severe enrichment	
	> 50	Extremely serve enrichment	
CF			[34]
	< 1	Minimum contamination	
	1 to 3	Moderate contamination	
	3 to 6	Considerable contamination	
Igeo	> 6	Very high contamination	[32]
	0	Uncontaminated or background concentration	
	0–1	Uncontaminated	
	1–2	Moderately contaminated to uncontaminated	
	2–3	Moderately contaminated	
	3–4	Moderately to highly contaminated	
	4–5	Highly contaminated	
	> 5	Very highly contaminated	
Integrated Index			
PLI			[37]
	0	Perfection	
	1	Baseline	
	> 1	Increasing contamination	

concentrations at which benthic organisms from aquatic ecosystems exhibited toxic responses in the laboratory [41].

$$TEC = \sqrt{(EDS - L * NEDS - M)} \quad (9)$$

EDS-L represents the concentration at which 15% of benthos showed effects, and NEDS-L denotes concentration at which 50% of benthos showed no impact.

2.6.3 Probable effect concentration (PEC)

PEC is the concentration at which a large percentage of the benthic population shows a toxic response. PEC can be calculated using Eq. 10 as proposed by the Florida Department of Environmental Protection [41].

$$PEC = \sqrt{(EDS - M * NEDS - H)} \quad (10)$$

EDS-M denotes concentration at which 50% of benthos showed effects, and NEDS-H indicates concentration at which 85% of benthos showed no impact.

2.6.4 Median effect concentration (MEC)

It is assumed that sediment contamination concentrations below TEC are acceptable and concentrations above the PEC are unacceptable. The region in between the TEC and PEC is called median effect concentration (MEC). The MEC values require further study and judgment to ascertain the likelihood of environmental consequences.

2.6.5 Potential ecological risk index (RI)

RI can be calculated using Eqs. 11–13 developed by Hakanson [34]. RI is widely used in evaluating the ecological risk of trace metals contamination in sediments [42]. Thus, RI is calculated using the formula:

$$RI = \sum E_i \quad (11)$$

$$E_i = TiFi \quad (12)$$

$$Fi = Ci/Cb \quad (13)$$

where, RI is the sum of all risk factors in the sediment samples; E_i is the monomial potential ecological risk factor for individual factors; T_i is the metal toxic response factor and F_i is the metal contamination factor, C_i is the calculated concentration of trace metal in the sediment sample, and C_b refers to the value of the reference element [35]. Metal contamination factor (F_i), risk index classification, and their environmental risk intensity are highlighted in Table 5. The metal contamination factor data of eight out of the ten detected trace metals in the samples were available in literature [34]. Therefore, the potential ecological risk assessment of 8 trace metals was evaluated in this work.

2.7 Statistical analysis

SPSS 24.0 software (IBM Corp., Armonk, NY, USA) was used for basic descriptive statistical analysis of the sample data [43]. This provided the mean and standard deviation for the sampled trace metal concentrations. Bivariate analysis such as Pearson's correlation method was used to assess the correlations between trace metals and multivariate analysis such as principal component analysis (PCA) was applied to study the source distribution of the selected trace metals.

3 Results

3.1 Distribution of trace metals in surface sediments

Trace metal concentrations in surface sediments sampled from the coastal and in four rivers estuary are shown in Table 6. The concentration of detected zinc (Zn) in the surface sediments varied from 57.02 to 155.05 mg/kg with an average concentration of 133.45 mg/kg. It has been reported that Zn has high mobility, and dissolved Zn can potentially increase its biological availability in an aquatic ecosystem [19, 44–46]. Osullivan et al. suggested that Zn can readily adsorb and be scavenged by the hydroxides and oxides [47]. The high level of Zn concentration in the surface sediments could be attributed to the fuel station, vehicle emissions, and commercial discharges [19, 48].

The detected arsenic (As) concentration in the samples detected ranged from 3.02 to 6.88 mg/kg with a mean value of 5.13 mg/kg (Table 6). Arsenic primarily exists in the ecosystem because of natural processes (i.e., volcanic processes and weathering of rocks, etc.) and anthropogenic activities such as mining, industrial pollution, fertilizer, pesticides, and insecticides [27, 49]. The elevated concentration of As in the study area could be attributed to the excessive use of inorganic fertilizer and domestic sewage discharges [50].

The iron (Fe) concentrations in the analysed samples ranged from 10,142.25 to 18,483.22 mg/kg, with a mean value of 16,415.17 mg/kg. In the current study, it was observed that Fe concentrations detected in all samples were higher compared to other trace metals. Apart from erosion, weathering, and some natural sources; the dominance of Fe in the surface sediments can be caused by large-scale anthropogenic processes including agricultural activities, solid waste, and urban-industrial discharges [51].

The occurrence of magnesium (Mg) in sediments is either a solute in pore-fluids or an essential constituent in the formation of late-stage diagenetic chlorite and dolomite [52]. Mg concentration ranged from 7183.17 to 9561.75 mg/kg with an average value of 8597.46 mg/kg (Table 6). Mg is the second highest concentration following Fe in the collected surface sediments from the study area. The relatively high content of Mg may be attributed to CaCO_3 sweating out from the sedimentary column or human activities such as industrial pollution, waste discharges, excessive pesticides, and fertilizer application [53].

Manganese (Mn) concentration varied from 98.93 to 194.90 mg/kg with an average of 142.92 mg/kg (Table 6). The behavior of Mn in sediment is different compared to other trace metals and can be influenced by the existence of MnO_2 in oxic surface sediments [19, 54]. Loska and Wienchula suggested that Mn pollution results from deposition in the atmosphere and organic material emissions [55]. The nickel (Ni) concentration in the surface sediments ranged from 10.54 to 27.28 mg/kg with an average value of 20.18 mg/kg (Table 6). Loska and Wienchula

Table 5 Metal contamination factor (F_i) of the selected trace metals, classification and environmental risk intensity

Trace metal	Co	Cu	Ni	Cr	Zn	Mn	Cd	As
Metal contamination factor	5.0	5.0	5.0	2.0	1.0	1.0	30.0	10.0
	Risk Index Value				Environmental Risk Categories			
1	< 150				Low ecological risk			
2	$\geq 150 < 300$				Moderate ecological risk			
3	$\geq 300 < 600$				Considerable ecological risk			
4	≥ 600				Very high ecological risk			

Table 6 Trace metals concentrations (mg/kg) in surface sediments from the coastal and in four rivers estuary of Sarawak

Sample site code	Zn	As	Fe	Mg	Mn	Ni	Cr	Cu	Co	Cd
CZ1(1)	57.02 ± 0.09	3.02 ± 0.08	13,572.17 ± 5.21	7261.00 ± 2.02	102.61 ± 0.94	18.63 ± 0.71	21.64 ± 1.13	11.02 ± 1.05	5.08 ± 0.31	0.01 ± 0.003
CZ1(2)	101.34 ± 0.21	3.73 ± 0.11	14,356.00 ± 8.61	7183.17 ± 5.06	116.43 ± 1.52	11.30 ± 1.01	34.56 ± 1.10	10.82 ± 0.84	3.98 ± 0.16	0.01 ± 0.001
CZ2(1)	104.09 ± 0.17	2.98 ± 0.04	14,895.02 ± 9.87	7929.05 ± 2.71	99.42 ± 0.83	16.42 ± 0.81	33.81 ± 2.21	10.13 ± 0.95	4.33 ± 0.41	0.02 ± 0.003
CZ2(2)	107.14 ± 0.26	5.41 ± 0.10	10,142.25 ± 6.41	8018.33 ± 1.81	98.93 ± 0.74	19.95 ± 1.21	33.53 ± 1.41	10.01 ± 0.46	2.99 ± 0.11	0.03 ± 0.001
CZ3(1)	134.03 ± 0.11	4.19 ± 0.07	15,524.00 ± 9.01	8099.57 ± 2.68	108.67 ± 2.11	20.06 ± 0.09	34.66 ± 0.89	9.69 ± 0.89	4.73 ± 0.31	0.01 ± 0.001
CZ3(2)	137.11 ± 0.42	4.88 ± 0.13	15,872.31 ± 5.81	8131.08 ± 1.41	117.59 ± 2.48	24.01 ± 0.16	36.61 ± 2.22	9.89 ± 0.41	5.93 ± 0.24	-
CZ4(1)	141.26 ± 0.39	5.43 ± 0.09	16,324.19 ± 6.91	9561.75 ± 0.96	142.07 ± 1.79	24.98 ± 0.71	38.11 ± 1.51	10.98 ± 1.21	5.98 ± 0.31	0.06 ± 0.001
CZ4(2)	144.09 ± 0.41	6.29 ± 0.10	18,483.22 ± 10.73	8938.60 ± 2.31	149.62 ± 1.69	22.00 ± 0.11	40.21 ± 2.61	12.06 ± 1.13	6.07 ± 0.43	0.02 ± 0.002
CZ5	147.03 ± 0.72	6.67 ± 0.09	17,327.71 ± 9.19	9183.01 ± 4.86	152.88 ± 2.51	25.57 ± 0.94	41.85 ± 1.31	12.77 ± 0.91	7.01 ± 0.68	-
CZ6	148.17 ± 0.18	5.99 ± 0.13	17,114.81 ± 12.61	9172.66 ± 3.14	183.36 ± 3.10	26.11 ± 0.69	41.28 ± 1.88	13.61 ± 1.01	6.99 ± 0.77	0.01 ± 0.003
CZ7(1)	150.06 ± 0.21	6.88 ± 0.23	17,210.64 ± 6.43	9317.09 ± 2.51	183.94 ± 1.42	26.91 ± 0.76	44.69 ± 2.10	14.93 ± 1.66	7.61 ± 0.39	0.03 ± 0.005
CZ7(2)	154.19 ± 0.66	5.96 ± 0.11	18,221.73 ± 12.11	8989.73 ± 5.19	194.90 ± 3.51	27.28 ± 0.82	43.74 ± 1.21	14.96 ± 1.44	7.92 ± 0.94	-
CZ8	151.14 ± 0.28	4.85 ± 0.27	17,982.43 ± 7.17	9077.48 ± 2.38	164.27 ± 2.62	20.32 ± 0.91	47.99 ± 0.99	15.05 ± 0.89	6.28 ± 0.61	0.03 ± 0.002
CZ9(1)	155.05 ± 0.53	5.12 ± 0.11	14,942.99 ± 6.46	9141.12 ± 4.92	154.99 ± 1.11	17.32 ± 0.94	38.28 ± 1.03	14.91 ± 0.64	6.81 ± 0.95	0.04 ± 0.001
CZ9(2)	150.18 ± 0.81	5.11 ± 0.15	15,337.01 ± 8.04	8552.71 ± 3.41	158.26 ± 2.75	10.54 ± 0.61	36.18 ± 1.71	10.64 ± 0.48	5.01 ± 0.91	-
CZ10	153.22 ± 0.95	5.59 ± 0.21	15,194.05 ± 8.71	9002.93 ± 3.21	158.84 ± 1.46	11.45 ± 0.94	33.60 ± 2.04	10.71 ± 0.71	5.13 ± 0.17	-
Mean	133.45 ± 0.01	5.13 ± 0.021	16,415.17 ± 6.36	8597.46 ± 2.49	142.92 ± 1.25	20.18 ± 0.72	37.55 ± 0.61	12.01 ± 0.89	5.74 ± 0.01	0.02 ± 0.001
Average shale	95.00	13.00	47,200.00	15,000	850.00	68.00	90.00	45.00	19.00	0.300

Details of sample site codes are mentioned in the materials and methods section

World average shale values (ASV) for trace metals as reported by Turekian and Wedepohl, 1961 were used as background values for calculating the geochemical indices because there were no geochemical background concentrations for the selected trace metals for the study area

suggested that Ni is primarily available in the organically bound form in the soil, which under certain pH (acidic or neutral) conditions accelerate its movement and biological availability [55]. According to Anderson et al., the primary anthropogenic sources of Ni pollution are fuel combustion and agricultural wastes [56].

The concentration level of chromium (Cr) varied from 21.64 to 47.99 mg/kg with an average concentration of 37.55 mg/kg (Table 6). Cr is regarded as low mobility trace metal, usually under moderately oxidizing and reducing conditions and nearly neutral pH. Zarei et al. reported that Cr and its associate compounds are used in synthesizing steel and some alloys, pigment production, and chrome plating [57]. Thus, it can be deduced that the steel and iron industry and chrome plating are the primary sources of Cr in the study area. Copper (Cu) concentration levels detected in all sampling sites varied from 9.69 to 15.05 mg/kg with a mean concentration of 12.01 mg/kg (Table 6). Cu as a mineral is of great importance for the proper growth and development of plants due to its constituent of different enzymes and proteins [40, 55]. Cu is extensively applied in roofing, electrical wiring, and manufacturing pigments, piping, alloys, and cooking utensils [49]. Therefore, manufacturing industries of electrical appliances, alloys, roofing materials, etc., near the study area are the primary source of Cu. It has been reported that the pollution of the aquatic system with Cu is linked with agrochemicals [19, 58]. Cobalt (Co) concentration levels in the surface sediment varied from 2.99 to 7.92 mg/kg with an average concentration of 5.74 mg/kg (Table 6). The concentration levels of the detected cadmium (Cd) in the samples ranged from nil (zero) to 0.06 mg/kg with a mean concentration of 0.02 mg/kg (Table 6). It was noticed that the estuarine sediments exhibited the highest concentration of Zn, Fe, Mg, and Mn compared to the coastal sediments.

This may be due to the new deposition of sediments in the estuarine sediments. Average concentrations of ten trace metals were compared to the average shale value for sedimentary rock [35]. It was observed that only Zn contents in all sampled sites exceeded the average shale concentration for sedimentary rock. The other trace metals concentrations in all sampled sites were below the average shale values for sedimentary rock. Thus, it can be concluded that the fundamental source of trace metals at the studied sites is due to natural activities and little influence of anthropogenic activities (Table 7).

3.2 Assessment of trace metals contamination

The average shale concentrations for sedimentary rock were used as background concentrations for trace metals in evaluating EF [35]. Zn is more enriched in sampled sites CZ2 (i.e., offshore of Rambungan River opposite to small Satang Island), CZ9 (i.e., offshore of Santubong resort), and CZ10 (i.e., Santubong River estuary). The other sampled sites showed moderate enrichments of Zn (Table 8).

High enrichment of Zn was detected in a sample collected from the offshore of Batang Rambungan opposite small Satang Island i.e., CZ2(2), which could be attributed to industrial discharges and domestic sewage. The EF values for As in collected samples at sampled sites CZ1 (i.e., Rambungan River estuary), CZ2 (i.e., Batang Rambungan opposite small Satang Island), and CZ3(1) (i.e., offshore of Batang Rambungan adjacent big Satang Island) were below 1, indicating no enrichment. The EF values for As for other sampled sites were between 1 and 3, showing minor enrichments. The highest EF value for Mg is observed in a sample collected from Rambungan River estuary, CZ2(1), with a value of 2.49, indicating minor enrichment (Table 8). Except for CZ1(1), the other sampled sites have EF values for Mg between 1 and 3,

Table 7 Detected trace metal concentrations in surface sediments in the study sites and in some selected world rivers

Location	Unit	Zn	As	Fe	Mg	Mn	Ni	Cr	Cu	Co	Cd	References
Coastal and selected estuaries, Malaysia	mg/kg	133.45	5.13	16,415.17	8597.46	142.92	20.18	37.55	12.01	5.74	0.02	This study
Langat River, Malaysia	µg/g	–	–	28,300.00	–	–	7.84	21.03	–	–	–	[33]
River Subin, Ghana	mg/kg	49.70	4.82	–	–	–	–	55.80	6.66	–	1.16	[59]
River Ganga, India	µg/g	67.76	–	31,988.60	–	372.04	26.70	69.94	29.75	–	–	[51]
River Tigris, Turkey	mg/kg	509.84	–	–	–	–	284.00	135.81	1257.76	–	–	[60]
River Jialu, China	mg/kg	107.58	–	–	–	–	42.44	60.80	39.22	–	–	[61]
Mangonbangan River, Philippines	mg/kg	213.45	–	22,006.14	–	261.97	61.14	89.45	116.36	15.31	–	[40]
Shur River, Iran	Ppm	522.00	–	–	–	–	–	–	9174.00	–	–	[62]
Korotoa River, Bangladesh	mg/kg	–	–	–	–	–	95.00	109.00	76.00	–	–	[63]
River Gomti, India	mg/kg	76.34	–	–	–	–	23.92	16.19	23.23	–	–	[64]
River Huaihe, China	mg/kg	–	–	28,300.00	–	–	7.84	21.03	–	–	–	[39]

Table 8 Enrichment of detected trace metals in surface sediments of the selected study sites

Sample site code	Zn	As	Mg	Mn	Ni	Cr	Cu	Co	Cd
CZ1(1)	2.09	0.81	0.17	0.42	0.95	0.84	0.85	0.93	0.12
CZ1(2)	3.51	0.94	1.58	0.45	0.55	1.26	0.79	0.67	0.11
CZ2(1)	3.47	0.73	2.49	0.37	0.77	1.19	0.71	0.72	0.21
CZ2(2)	5.25	1.94	1.64	0.39	1.37	1.73	1.04	0.73	0.47
CZ3(1)	4.29	0.98	1.61	0.75	0.90	1.17	0.66	0.76	0.10
CZ3(2)	4.29	1.12	1.61	0.80	1.05	1.21	0.65	0.93	–
CZ4(1)	4.30	1.21	1.84	0.83	1.06	1.22	0.71	0.91	0.58
CZ4(2)	3.87	1.24	1.52	0.93	0.83	1.14	0.68	0.82	0.17
CZ5	4.22	1.40	1.67	0.92	1.02	1.27	0.77	1.00	–
CZ6	4.30	1.27	1.69	1.11	1.06	1.27	0.83	1.02	0.09
CZ7(1)	4.33	1.45	1.70	1.10	1.09	1.36	0.91	1.10	0.27
CZ7(2)	4.20	1.19	1.55	1.20	1.04	1.26	0.86	1.08	–
CZ8	4.18	1.00	1.59	1.00	0.78	1.40	0.88	0.87	0.26
CZ9(1)	5.15	1.24	1.93	0.94	0.80	1.34	1.05	1.13	0.42
CZ9(2)	4.88	1.21	1.76	1.03	0.48	1.24	0.73	0.81	–
CZ10	5.01	1.34	1.86	0.98	0.52	1.16	0.74	0.84	–
Mean	4.21	1.19	1.64	0.83	0.89	1.25	0.80	0.90	0.18

Details of sample site codes are mentioned in the materials and methods section

suggesting minor enrichment. Most of the EF values for Ni and Cr in the samples were between 1 and 3, indicating minor enrichment while, most of the estimated EFs for Co and Cd were below 1, suggesting background concentration. The high EF values obtained for some trace metals in some sampled sites may be ascribed to anthropogenic sources such as urbanization, industrial wastes deposition, etc. Trace metals bioavailability and toxicity in sediments are determined by their concentrations and chemical form [19, 65]. Thus, trace metals in sediments with high EF values associated with labile fractions have the potential for mobility and bioavailability in aquatic environments [63].

The list of CFs of ten trace metals in surface sediments is highlighted in Table 9. The highest CF value for Zn (i.e., CF = 1.63) was recorded in a sample collected from the offshore of Santubong Resort i.e., CZ9(1), suggesting moderately contamination. This may be attributed to commercial activities and vehicular effluence. Also, the CFs values obtained for As, Fe, Mg, Mn, Ni, Cr, Cu, Co, and Cd were below 1, which could be ascribed to lithogenic influences. Anthropogenic activities such as residential discharges, chemical control of surrounding weeds, etc., may also play a minor role.

The *Igeo* values for trace metals in surface sediments from the coastal area and in four rivers estuary of Kuching Division were shown in Table 10. The *Igeo* values for each trace metals are as follows: -0.49 to 0.11 for Zn, -2.74 to -1.50 for As, -2.81 to -1.94 for Fe, -1.62 to -1.22 for Mg, -3.69 to -2.71 for Mn, -3.28 to -1.91

for Ni, -2.64 to -1.49 for Cr, -2.80 to -2.18 for Cu, -3.25 to -1.85 for Cd, and -5.49 to -2.91 for Co. The *Igeo* values for As, Fe, Mg, Mn, Ni, Cr, Cu, Cd, and Co in the sediments from the study area were below class 0, indicating unpolluted site. The high *Igeo* value for Zn was recorded in surface sediment collected from the offshore of Santubong resort i.e., CZ9(1), suggesting unpolluted to moderately polluted. The positive *Igeo* values for Zn at the sampled sites from CZ4(2) to CZ10 may be attributed to sewages discharges and/or effluents.

The PLI values for trace metals of the studied site were summarised in Table 10. The PLI values varied from 0.22 to 0.52 and with a mean value of 0.31. This indicates that there has been no occurrence of contamination in the studied site. High PLI value was found in sediment from the offshore of Telaga Air opposite to small Satang Island. In contrast, low PLI value was observed in sediment from the Rambungan River estuary and the offshore of Batang Rambungan opposite small Satang Island. Based on the PLI values, no significant disturbances of the aquatic environment due to heavy metals pollution were observed.

3.3 Ecotoxicological risk assessment

To determine the risks associated with trace metals toxicity on organisms living at or near the bottom of the aquatic bodies (i.e., bodies of water forming a physiological feature for example a river, sea, etc.); trace metals concentrations were compared with consensus-based

Table 9 Contamination levels of detected trace metals in surface sediments of the selected study sites

Sample site code	Zn	As	Fe	Mg	Mn	Ni	Cr	Cu	Co	Cd
CZ1(1)	0.60	0.23	0.29	0.48	0.12	0.27	0.24	0.25	0.27	0.03
CZ1(2)	1.07	0.29	0.30	0.48	0.14	0.17	0.38	0.24	0.21	0.03
CZ2(1)	1.10	0.23	0.32	0.53	0.12	0.24	0.38	0.22	0.23	0.07
CZ2(2)	1.13	0.42	0.22	0.54	0.12	0.29	0.37	0.22	0.25	0.10
CZ3(1)	1.41	0.32	0.33	0.54	0.13	0.30	0.39	0.22	0.31	0.03
CZ3(2)	1.44	0.38	0.34	0.54	0.14	0.35	0.41	0.22	0.32	–
CZ4(1)	1.47	0.42	0.35	0.64	0.17	0.37	0.42	0.24	0.32	0.07
CZ4(2)	1.52	0.48	0.39	0.60	0.18	0.32	0.45	0.27	0.37	0.07
CZ5	1.55	0.51	0.37	0.61	0.18	0.38	0.47	0.28	0.37	–
CZ6	1.56	0.46	0.36	0.61	0.22	0.38	0.46	0.30	0.40	0.03
CZ7(1)	1.58	0.53	0.37	0.62	0.22	0.38	0.50	0.33	0.42	0.10
CZ7(2)	1.62	0.46	0.37	0.60	0.23	0.40	0.47	0.33	0.33	–
CZ8(1)	1.59	0.37	0.38	0.61	0.19	0.30	0.53	0.33	0.36	0.10
CZ9(1)	1.63	0.39	0.32	0.61	0.18	0.25	0.43	0.33	0.26	0.13
CZ9(2)	1.58	0.39	0.33	0.57	0.19	0.16	0.40	0.24	0.26	–
CZ10	1.61	0.43	0.32	0.60	0.19	0.17	0.37	0.24	0.26	–
Mean	1.40	0.33	0.34	0.57	0.17	0.30	0.42	0.27	0.31	0.05

Details of sample site codes are mentioned in the materials and methods section

Table 10 Geo-accumulation indices (*I_{geo}*) and pollution load indices (PLI) values of detected trace metals in surface sediments of the selected study sites

Sample site code	Zn	As	Fe	Mg	Mn	Ni	Cr	Cu	Co	Cd	PLI
CZ1(1)	−1.32	−2.69	−2.38	−1.62	−3.64	−2.45	−2.64	−2.62	−2.49	−5.49	0.22
CZ1(2)	−0.49	−2.38	−2.30	−1.58	−3.46	−3.17	−1.97	−2.64	−2.84	−5.49	0.24
CZ2(1)	−0.45	−2.74	−2.25	−1.49	−3.68	−2.64	−2.00	−2.74	−2.72	−4.49	0.22
CZ2(2)	−0.41	−1.85	−2.81	−1.47	−3.69	−2.35	−2.01	−2.76	−3.25	−3.91	0.29
CZ3(1)	−0.09	−2.22	−2.19	−1.46	−3.56	−2.35	−1.96	−2.80	−2.59	−5.91	0.28
CZ3(2)	−0.06	−2.00	−2.16	−1.45	−3.44	−2.09	−1.88	−2.77	−2.27	–	0.34
CZ4(1)	−0.01	−1.84	−2.12	−1.22	−3.17	−2.03	−1.82	−2.62	−2.25	−2.91	0.34
CZ4(2)	0.02	−1.63	−1.94	−1.32	−3.10	−2.21	−1.75	−2.48	−2.23	−4.49	0.35
CZ5	0.05	−1.55	−2.03	−1.28	−3.06	−2.00	−1.69	−2.40	−2.02	–	0.52
CZ6	0.06	−1.70	−2.05	−1.28	−2.80	−1.97	−1.71	−2.31	−1.91	−5.49	0.34
CZ7(1)	0.08	−1.50	−2.04	−1.26	−2.79	−1.92	−1.60	−2.18	−1.85	−3.91	0.40
CZ7(2)	0.11	−1.71	−1.96	−1.31	−2.71	−1.91	−1.63	−2.18	−1.85	–	0.49
CZ8	0.08	−2.01	−1.98	−1.29	−2.96	−2.33	−1.49	−2.19	−2.18	−3.91	0.37
CZ9(1)	0.12	−1.93	−2.45	−1.28	−3.04	−2.56	−1.82	−2.18	−2.06	−3.49	0.35
CZ9(2)	0.08	−1.93	−2.21	−1.38	−3.01	−3.28	−1.90	−2.65	−2.45	–	0.35
CZ10	0.11	−1.80	−2.22	−1.31	−3.69	−3.16	−1.79	−2.64	−2.47	–	0.36
Mean	−0.11	−1.97	−1.62	−1.38	−3.24	−2.40	−1.85	−2.07	−2.34	−3.09	0.31

Details of sample site codes are mentioned in the materials and methods section

threshold effect concentration (TEC), probable effect concentration (PEC), and the midway concentration between TEC and PEC (i.e., MEC) values fetched from the sediments quality guidelines developed by ANZECC/ARMCANZ, 2000 [38]. The TEC, MEC, and PEC data of eight out of the ten detected trace metals in the samples

were available in the sediments quality guidelines developed by ANZECC/ARMCANZ, 2000. Thus, risks associated with 8 trace metals toxicity on bottom-dwelling organisms in the study area were appraised. The comparisons of consensus-based sediment-quality guidelines (SQGs) with detected trace metals levels in the surface

sediments of the selected study sites were presented in Table 11. Twenty-five percent of the samples contained Zn concentrations less than the TEC value for Zn, while 62.50 % of the sampled sites contained Ni concentrations lower than the TEC value for Ni (Table 11). In addition, 81.25 % of the samples had Cr concentrations below the TEC value for Cr. All the sampled sites contained As, Fe, Mn, Cu, and Cd concentrations below TEC values for As, Fe, Mn, Cu, and Cd, respectively. None of the samples contained trace metals exceeding the PEC values. Trace metals concentrations in surface sediments below TEC values were unlikely to negatively impact bottom-dwelling organisms [60]. Seventy-five percent of the sampled sites contained Zn concentrations exceeding TEC but

equal or less than MEC, whereas 37.50 % of the samples had Ni concentrations surpassing TEC but equal or less than MEC. Furthermore, 18.75 % of the sampled sites contained Cr concentrations above TEC but equal or less than MEC.

The monomial potential ecological risks (E_i) for each trace metal and possible environmental risk index (RI) in all collected samples from the coastal and in four rivers estuary were detailed in Table 12. The RI index was computed based on the eight heavy metals (i.e., Zn, As, Ni, Cr, Cu, Co, and Cd). The mean potential ecological risk for studied trace metals follow the order of $Mn < Cr < Ni < Cu < Zn < Cd < Co < As$. Among the analysed trace metals; Mn, Cr, and Ni had relatively lower RI values due

Table 11 Comparisons of consensus-based sediment-quality guidelines (SQGs) with detected trace metals levels in the surface sediments of the selected study sites

	Zn	As	Fe	Mn	Ni	Cr	Cu	Cd
SQGs								
TEC	120	9.8	20,000	460	23	43	32	0.99
MEC	290	21.4	30,000	780	36	76.5	91	3.00
PEC	460	33	40,000	1100	49	110	150	5.00
% sample sites of detected trace metal less than TEC	25.00	100.00	100.00	100.00	62.50	81.25	100.00	100.00
% sample sites of detected trace metal greater than TEC but equal or less than MEC	75.00	0	0	0	37.50	18.75	0	0
% sample sites of detected trace metal greater than PEC	0	0	0	0	0	0	0	0

Details of detected trace metals value in each sample site are mentioned in Table 6 in the results section

Table 12 The potential ecological risk index values (RI) of detected trace elements in surface sediments of the selected study sites

Sample site code	E _i								RI
	Zn	As	Mn	Ni	Cr	Cu	Co	Cd	
CZ1(1)	0.60	2.30	0.12	1.35	0.48	1.25	1.35	0.90	8.05
CZ1(2)	1.07	2.90	0.14	0.85	0.76	1.20	1.05	0.90	8.87
CZ2(1)	1.10	2.30	0.12	1.20	0.76	1.10	1.15	2.10	9.83
CZ2(2)	1.13	4.20	0.12	1.45	0.74	1.10	1.25	3.00	12.99
CZ3(1)	1.41	3.20	0.13	1.50	0.78	1.10	1.55	0.90	10.57
CZ3(2)	1.44	3.80	0.14	1.75	0.82	1.10	1.60	–	10.65
CZ4(1)	1.47	4.20	0.17	1.85	0.84	1.20	1.60	2.10	13.43
CZ4(2)	1.52	4.80	0.18	1.60	0.90	1.35	1.85	2.10	14.30
CZ5	1.55	5.10	0.18	1.90	0.94	1.40	1.85	–	12.92
CZ6	1.56	4.60	0.22	1.90	0.92	1.50	2.00	0.90	13.60
CZ7(1)	1.58	5.30	0.22	1.90	1.00	1.65	2.10	3.00	16.75
CZ7(2)	1.62	4.60	0.23	2.00	0.94	1.65	1.65	–	12.69
CZ8	1.59	3.70	0.19	1.50	1.06	1.65	1.80	3.00	14.49
CZ9(1)	1.63	3.90	0.18	1.25	0.86	1.65	1.30	3.90	14.67
CZ9(2)	1.58	3.90	0.19	0.80	0.80	1.20	1.30	–	9.77
CZ10	1.61	4.30	0.19	0.85	0.74	1.20	1.30	–	10.19
Mean	1.40	3.30	0.17	0.85	0.84	1.35	1.55	1.50	10.96

Details of sample site codes are mentioned in the materials and methods section

to their low toxicity response factors. Furthermore, RI results in the surface sediment varied from 8.05 (CZ1) to 16.75 (CZ7). The obtained RI values for all sampled sites were lower than 150, indicating that the sediments in the studied site posed a minimum risk. The most significant Ei values were recorded for As, because, based on Hakanson's approach, the toxic response of this metal is the highest. Although, it does not show a high ecological risk in the coastal and in four rivers estuary sediment, owing to the fact that the quantified As values are positioned below the acceptable limit. The other metals that made an important contribution to the final result of the RI index include Cu, Ni and Co.

3.4 Trace metal pollution source

Evaluating the sources of trace metals can help comprehend their distribution. Thus, Pearson's correlation analysis and principal component analysis (PCA) were employed to analyze the relationship and source of the trace metals [66, 67].

The correlation coefficient matrix recording the Pearson's product-moment coefficients were shown in Table 13. Positive correlations were recorded between (Zn and As), (Zn and Fe), (Zn and Mg), (Zn and Mn), (Zn and Cr), (As and Mg), (As and Cr), (As and Co), (Fe and Mg), (Fe and Mn), (Fe and Cr), (Fe and Co), (Mg and Mn), (Mg and Cr), (Mg and Co), (Mn and Cr), (Mn and Cu), (Mn and Co), (Ni and Co), (Cr and Cu), (Cr and Co), and (Cu and Co) at 0.01 significant level. Positive correlations were also noticed between (Zn and Cu), (Zn and Co), (As and Ni), (As and Cu), (Fe and Cu), (Mg and Cu), (Mg and Cd), (Mn and Cu), and (Mn and Cd) at 0.05 significant level. The high positive interrelationships between studied trace metals are indication of a common source.

In this current study, PCA was conducted based on the evaluated concentrations of trace metals with varimax rotation. The Kaiser–Meyer–Olkin (KMO) test gives 0.49, and the Barlett test gives 99.82 ($df=45$), showing strong interrelationships among variables and substantiating that PCA can be used to reduce the dimensionality of variables. Based on PCA results (Fig. 2 and Additional file 1: Table S1), PCA1 accounts for 67.33% of the variation, and its representative congeners include Mn, Mg, Zn, Co, Cr, As, Cu, Fe, and Ni, suggesting a common source that is probably exogenous discharge [68, 69]. PCA2 accounts for 11.80% of the variation, and its representative congeners are Cd and Mg, implying a similar source that may be attributed to industrial and domestic discharge. The weak correlation between Cd and other trace metals is an indication of different external sources.

Fig. 2 Principal component profile of the ten trace metals collected from the sediments of the selected sampling sites

Table 13 Correlation coefficients between different detected trace metals in surface sediments of the selected study sites

	Zn	As	Fe	Mg	Mn	Ni	Cr	Cu	Co	Cd
Zn	1									
As	0.747**	1								
Fe	0.630**	0.48	1							
Mg	0.849**	0.801**	0.625**	1						
Mn	0.768**	0.741**	0.715**	0.790**	1					
Ni	0.268	0.543*	0.477	0.4790	0.375	1				
Cr	0.807**	0.686**	0.711**	0.739**	0.740**	0.496	1			
Cu	0.499*	0.511*	0.581*	0.612*	0.799*	0.452	0.707**	1		
Co	0.619*	0.630**	0.808**	0.726**	0.832**	0.667**	0.667**	0.812**	1	
Cd	0.435	0.377	0.065	0.649*	0.252	0.331	0.342	0.241	0.267	1

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

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

Computed values were for all the selected study sites

4 Discussion

The coastal and four rivers estuary (i.e., Santubong, Salak, Sibul, and Rambungan) is a vital agricultural and transportation water resources for the Kuching Division of Sarawak. Therefore, it is necessary to evaluate the pollution status, ecotoxicological risks, and likely sources of trace metals in the surface sediments of the coastal and four selected rivers estuary. In aquatic ecosystems, sediment is both a source and a sink of trace metals. Thus, this study investigated ten trace metals in surface sediments from the coastal and four rivers estuary.

In sediment samples, all ten trace metals were detected, and their average concentration followed the order of $Cd < As < Co < Cu < Ni < Cr < Zn < Mn < Mg < Fe$. The assessment showed moderate As, Mg, Cr, and Ni pollution. The average concentrations of trace metals in surface sediment were compared with other studies of trace metals in rivers in the world (Table 7). The Zn concentrations detected in all sampled sites were lower than Zn content in Tigris River sediment, Mangobangon River sediment [40], and Shur River sediment [62]. Furthermore, the average concentrations of Cu in the studied area were below Cu contents reported values from surface sediments from Tigris River [60], Mangobangon River sediment [40], Jialu River sediment [61], Krotoa River sediment [63], and Gomti River sediment [64] (Table 7). The mean concentrations of Ni in all sediment samples were lower than values reported in surface sediments from Tigris River [60], Jialu River [61], Mangobangon River [40], Korotoa River [63], and Gomti River [64]. The average concentration of Cr in this current study was below the detected level of Cr in surface sediment's from the Subin River [59], Tigris River [60], Mangobangon River [40], and Korotoa River [63]. The Mn in the sediments of Mangobangon River [40] exceeded the values of Mn concentration in this study. Furthermore, the detected level of Cd was higher than Cd levels detected in surface sediments from the Subin River [59]. In contrast, the average concentrations of detected Fe in this study were below the concentrations of detected Fe in surface sediments from Mangobangon River [40] and Huaihe River [39].

Although the single factor pollution indices (i.e., EF, CF, and I_{geo}) method has been widely used, it is functional to a single pollutant. Thus, it does not consider a mixture of trace metals primarily available in the pollution conditions. However, it has helped to ascertain how much the available metal in sampled sites has elevated relative to average natural abundance due to human activity [7]. Nevertheless, an integrated pollution index (i.e., PLI) was employed to help considering the mixture of trace metals present in the contamination conditions. Based on EF results, the trend of trace metal enrichments in all

sampled sites followed the order: $Cd < Cu < Mn < Ni < Co < As < Mg < Cr < Zn$ (Table 8). Generally, the evaluation showed minor enrichments of almost all the trace metals in all sampled sites. The enrichments of trace metals may be attributed to both natural processes and anthropogenic sources, including industrial wastes deposition, sewage discharges, and urbanization. The trend of obtained CFs values for trace metals in all samples followed the order: $Cd < Mn < Cu < Ni < Co < As < Fe < Cr < Mg < Zn$. Almost all the trace metals CF values were below 1 in all sampled sites, suggesting minimum contamination conditions except for Zn, which showed moderate contamination in some sample sites (Table 10). All the trace metals except Zn have I_{geo} values lower than 0, indicating background concentration. Generally, the I_{geo} values obtained for Zn indicate a minor role played by anthropogenic activities. From the PLI results; there is no significant disturbance of the aquatic environment due to trace metals pollution. The PLI values can be used as baseline contamination levels in the future for pollution monitoring in a selected site.

The ecological risk index has been demonstrated as a highly productive tool to evaluate the total pollution of sediments of an aquatic ecosystem [54]. Protano et al. narrated that the inadequacy of updated reference metal values for a specified ecological site or geographical zone can lead to underestimating or overestimating the actual pollution load in sediments and the environmental risk index [70]. Decena et al. reported that to get an accurate estimation of the ecological risk of metals, regular updates of reference concentrations after a certain period are required, especially in geological zones with sensitive ecological environments [40]. Average concentrations of trace metals were compared with sediment-quality guidelines (SQGs). It was observed that almost all the trace metals in all sampled sites were below TEC values for respective trace metals except Zn, Ni, and Cr, in which their concentrations exceeded TEC values for Zn i.e., from sampled site CZ3(1) to CZ10; Ni i.e., from sampled site CZ3(2) to CZ7(2); and Cr i.e., from sampled site CZ7(1) to CZ8. It can be deduced that trace metals concentrations in sampled sites above TEC but equal or less than MEC may probably affect bottom-dwelling organisms. According to the IR results, a low potential ecological risk from all the trace metals in all sampled sites was noticed.

Since the studied trace metals in sediments have a moderate adverse health impact on the biome, it is necessary to assess and control the pollution source. Trace elements in sediments often show complex interrelationships. Many factors influence their relative abundance, for instance, parent materials and rocks, anthropogenic activities, and soil formation processes [42, 71]. Pearson's correlation analysis was used to assess the relationship between the trace metals. Principal component analysis (PCA)

was performed to evaluate the most common pollution sources. Correlation analysis and PCA results showed strong positive interrelationships between trace metals, suggesting a common source or similar geochemical characteristics except for Cd, which showed a weak correlation with other trace metals except for Mg (Fig. 2 and Additional file 1: Table S1). The inverse relationships between Cd and other trace metals are indication of different external sources.

Despite a low level of absolute content, the As, Cd, and Ni in sediment already render a moderate monomial ecological risk and therefore calls urgent attention. The primary source of trace metals in sediment is natural processes and sediment properties. Anthropogenic activities may also influence trace metals distribution of the studied area.

5 Conclusions

The concentrations of ten trace metals in surface sediments collected from the coastal and in four rivers estuary in Sarawak, Malaysia, were examined. All ten trace metals were detected at all sampled sites, with a concentration lower than the average shale value for sedimentary rock except Zn. Pollution appeared more severe in the coastal of the studied area, probably due to point source contamination nearby. Among the trace metals of interest, Zn, Ni, and Cr concentrations surpassed the TEC values and should be carefully monitored and remediated because they may cause unfavorable impacts on bottom-dwelling organisms. Potential ecotoxicological risk analysis of trace metals concentrations in surface sediments indicated that most sampling sites posed a minor or moderate ecological risk. Correlation analysis and principal component analysis revealed that nine trace metals (Zn, As, Fe, Mg, Ni, Cr, Cu, and Co) were derived from common sources. The results will guide controlling trace metal contamination and protecting agricultural and transportation water sources in the coastal and the four rivers estuary in the Kuching Division of Sarawak.

Abbreviations

EF: Enrichment factor; CF: Contamination factor; Igeo: Geo-accumulation index; PLI: Pollution load index; SQGs: Sediment quality guidelines; TEC: Threshold effect concentration; PEC: Probable effect concentration; RI: Risk index; PCA: Principal component analysis; ICP-OES: Inductively coupled plasma-optical emission spectroscopy.

Supplementary Information

The online version contains supplementary material available at <https://doi.org/10.1186/s43088-022-00199-y>.

Additional file 1. Total variance explained and component matrices for trace metals in the surface sediments of the coastal and in four rivers estuary of Sarawak.

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Authors' contributions

EAA, ZA, and RW conceived of the study and carried out the design of the experiment. EAA carried out the sample preparation and analysis, EAA, ZA, and JRF assessed the data, and EAA, ZA, and RW helped to draft and edited the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials

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

Declarations

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Competing interests

The authors declare that they have no competing interests.

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