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Ecological Risk Assessment of Heavy Metal Pollution in Mangrove Sediments of the Sepang Besar River, West Coast Peninsular Malaysia

Kumar Krishnan[1,*], **Nadia AS**[1], **Chong MY**[2]

[1]Faculty of Health & Life Sciences, INTI International University, Negeri Sembilan, Malaysia
[2]Matrix Global School, Negeri Sembilan, Malaysia

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Abstract The impact of heavy metal contamination caused by human activities on mangroves, rivers, estuaries, and coastal wetlands has gained attention recently. In this study, the presence of heavy metals in the mangrove Sepang Besar River's surface sediments was examined. As, Mo, Pb, Cr, Co, Cd, Ni, and Hg concentrations in sand were measured using inductively coupled plasma mass spectrometry (ICP-MS). Across all sampling sites, Cr concentrations were the highest and Hg concentrations were the lowest. The concentration of As was found higher than shale's geochemical background value. The range of EFs for As, Mo, and Pb are 4.38 to 12.9, 1.71 to 3.67, and 0.94 to 2.68, respectively. The eight heavy metals and other environmental components were compared using the correlation coefficient. The ecological hazards posed by eight heavy metals (As, Mo, Pb, Cr, Co, Cd, Ni, and Hg) were assessed using the comprehensive enrichment factor (EF) and geo-accumulation index (Igeo). The Enrichment factor and geo-accumulation index, in particular, revealed that Arsenic is the most prevalent heavy metal pollutant in surface sediments in Sepang Besar River mangrove sediments, with some stations requiring specific attention due to high pollution levels.

Keywords Heavy Metal, Pollution, ICP-MS, Enrichment Factor, Geoaccumulation

1. Introduction

Heavy metals have long been regarded one of the most significant contaminants in aquatic environments, and heavy metal poisoning of rivers has become a common problem. The rising industrialization and urbanization of most Malaysian cities have resulted in considerable volumes of pollution entering rivers, raising concerns about the aquatic environment [1]. Although some trace elements and metals are biologically significant for aquatic life, live species are at risk when heavy metal concentrations exceed specified limits. The majority of heavy metals are also abundant in the environment and have the capacity to persist and bio-accumulate, which makes them a major worry [2].

Heavy metal pollution of bottom sediments is one of the most serious threats to the water environment. River bottom sediments account for about 97 percent of the bulk transfer of heavy metals to the seas [3-4]. Given that heavy metal poisoning of sediments is a common problem, research on their abundance, distribution and sources has gained momentum on a global scale [5]. Due to their well-documented detrimental effects on ecosystem health and human health, heavy metal pollutants' presence in the sediment environment has been a major cause for concern [6-7].

Heavy metal contamination is viewed as a serious threat to the water environment due to its chronic nature, stability, toxicity, tendency to enter the food chain, and bioaccumulation [8]. Following their release, heavy metals have the potential to spread throughout the river habitat, accumulating in the water, bottom sediments, and river fauna and plants.

Only a few of the mechanisms that mix heavy metals with the bottom sediments of the hyporheic zone are coprecipitation, particle surface adsorption, ionic exchange, hydrolysis, and organic matter deposition [9–10].

The most common sources of heavy metals are anthropogenic point and area sources linked to industry and agricultural activity [11]. Because heavy metals accumulate in plants and have negative effects on animals, they are released from bottom sediments and can harm river flora and agricultural crops [12]. They can also build up in plants with water from irrigation infrastructure, endangering agricultural crops and river vegetation. Several metals found in bottom sediments have been demonstrated to be dangerous even at low quantities, depending on their source [13].

The bottom sediment quality index must be built and analysed in light of the aforementioned information in order to identify the pollution and toxicity risks associated with the presence of heavy metals in the aquatic environment. The Geoaccumulation Index (Igeo), Enrichment Factor (EF), Pollution Load Index (PLI), and Metal Pollution Index (MPI), which have all been created, can all be used to assess the quality of bed sediment [1-2,14].

The purpose of this study was to investigate variations in heavy metal concentrations in mangrove sediments in the Sepang Besar River on the west coast of peninsular Malaysia. The study examines the concentration of heavy metals in the surface sediments and to determine whether they are naturally occurring or artificially created based on the enrichment factor (EF), and uses Pearson correlation analysis to examine the main environmental factors that influence the metal deposition (CA). Furthermore, using sediment quality guidelines (SQG) and the Geoaccumulation Index (Igeo), it examines sediment quality and pollution stress of mangrove sediments.

2. Materials and Methods

2.1. Study Sites and Samples Collection

The mangrove sediments collected from Sepang Besar River (Sungai Besar Sepang) in 2019, which flows into the Straits of Malacca in Sepang District, Selangor, Malaysia, at GPS coordinates of 02 36'7. 41" N and 101 44' 8.62' E as shown in Figure 1, as well as nearby activities from sampling station stated in Table 1, were the subject of this study. The study area enjoys a tropical climate all year, with temperatures ranging from 27 to 34 degrees Celsius and a moderately humid atmosphere. The cloud cover is low, and the wettest month is November, albeit rain is uncommon. Because of its proximity to Malaysia's administrative capital, Putrajaya, the Sepang district witnessed rapid and large expansion from 1995 to 2015. This results in increased urbanisation, which has an impact on the ecosystems of Sungai Sepang Besar. The Sepang district's reserve forests and mangroves are small, with mangrove forests totaling around 550 hectares along the Sungai Sepang Kecil and Sungai Sepang Besar rivers. There are fish, crabs, and shrimp, all of which contribute to the mangrove ecosystem's richness. Mangroves are listed as Level 1 Environmentally Sensitive Areas in the Sepang 2025 Local Plan (Sepang 2017) [15-16].

Table 1. The description of sampling location

Location ID	Location and nearby activity	GPS Reading
Station S	Shrimp Farm	2◦ 36.3’23’’ N 101◦ 42.1’26’’ E
Station T	Pig Farm	2◦ 36.2’08’’ N 101◦ 42.4’4’’ E
Station U	Fish Farm	2◦ 36’03’’ N 101◦ 42.6’16’’ E
Station V	Tourist spot, raft house	2◦ 36.5’06’’ N 101◦ 42.2’7’’ E

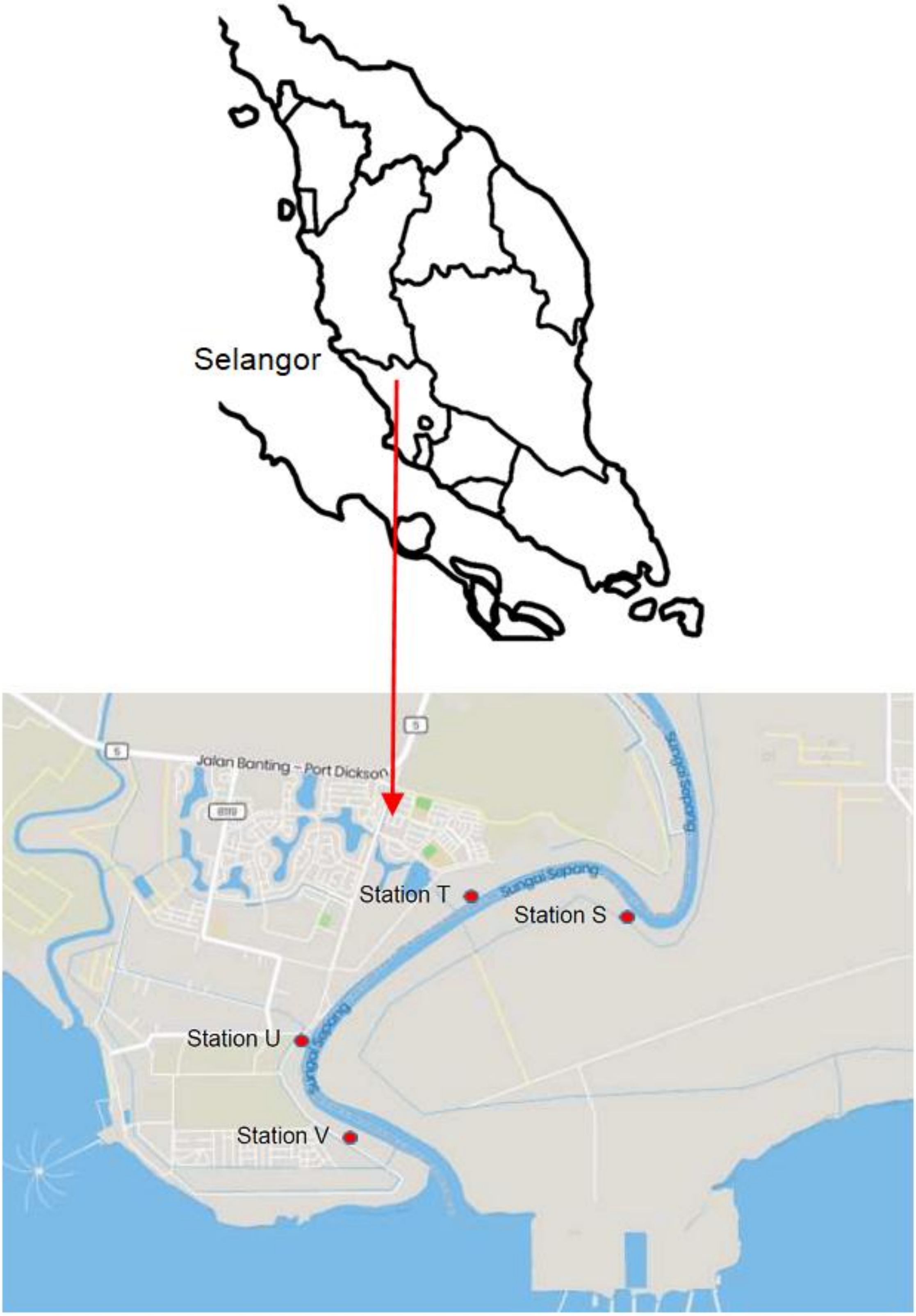

Figure 1. Map of the sampling site mangrove sediments Sepang Besar River, Selangor, Malaysia

2.2. Instrumentation

By scraping the top layer with a plastic spoon, the mangrove sediments, which weighted about 500 g, were collected in a clean plastic bag with a label at a depth of about 5.0 cm. To obtain a dry weight in the lab, sediments were dried in an oven at 80 ℃ for at least 72 hours. Using a glass mortar, each sediment sample was ground into powder. A 63-meter stainless steel hole is used to filter the powder after that. The samples were thoroughly stirred before being placed in plastic pill containers and prepared for further examination [2]. Plastic containers and all glassware used for collection, transit, and laboratory analysis were acid-washed with nitric acid, HNO_3 (10% concentrated AnalaR grade; BDH 69 percent) to avoid and restrict contamination.

Perchloric Acid, $HClO_4$ (AnalaR grade; BDH 60%), and HNO_3 were used in a 4:1 ratio to digest a total of 0.50 g of each dried and powdery sample (AnalaR grade; BDH 69 percent). The digestion of the sediment, standard, and blank samples took place for at least 3 hours at a low temperature (40 ℃) in a hot block digester and digestion tube [17]. After the digestion process, the resulting solution was diluted with 40 mL of double-distilled water and filtered using WhatmanTM No. 1 filter paper. The solution was then filtered into a polyethylene container to provide a volume of 50 mL for ICP-MS analysis using filter paper (Whatmann brand, diameter 125 mm) and Milli-Q water [18]. Three replicates of each sample were examined [19].

3. Results and Discussion

The SRM (IAEA Soil-7) was used as a quality control and assurance measure throughout the analysis of the analytical procedure. The calculation of SRM followed the same steps as a sample analysis [1]. The z-score and recovery analysis were used to evaluate the precision and accuracy of the analytical procedures that were applied. In order to demonstrate the accuracy of the analytical procedure, the standardised difference z is determined by taking into account the uncertainty in both the certified value and the measured values from the SRM. All of the elements' recovery rates and z-scores ranged from 78 to 109 percent and - 0.4 to +1.2, respectively. The ICP-MS method's recoveries and z-score between measured and certified samples are within acceptable bounds.

Table 2 shows the concentration of heavy metal in mangrove sediments collected at the four sampling locations (Station S.T.U and V). The concentration of As, Cd, Co, Cr, Hg, Mo, Ni, and Pb (in mg/kg) from all stations were ranging 3.31 – 9.827, 0.015 – 0.024, 1.434 – 3.122, 5.34 – 15.81, 0.004 – 0.009, 0.860 – 1.851, 2.09 – 6.931 and 5.54 – 15.76, respectively. In general, heavy metal accumulation in sediments at Station S, Station T, Station U and Station V were found to be in order of Pb > Cr > As > Ni > Co > Mo, Pb > Cr > As > Ni > Co > Mo > Cd, Cr > Pb > As > Ni > Co > Mo > Hg and Cr > Pb > As > Ni > Co > Mo > Cd > Hg respectively. The concentration of Cr was the highest and Hg was the lowest at all sampling sites. The mean concentrations of all heavy metal were found to be greater at stations V, U and S than at station T. Heavy metal concentration is higher in the sediment because these elements are enriched directly from the weathering profile and are kept by clay particles in the soil, which may also be a result of anthropogenic activity close to the study area [33]. Sediment particles, which are made up of both biogenic and non-biogenic (lithogenic) components, are regarded final sinks for heavy metals that have been transported to the mangrove environment [20]. Agriculturally produced animal wastes including chicken, cattle, and pig manures are frequently spread on crops and pastures as solids or slurries. High levels of As, Cu, and Zn are present in the manures produced by animals fed such diets, and if these manures are frequently spread over confined areas of land, a significant amount of these metals may eventually accumulate in the soil [31-32]. Given that the location of the study is near the shrimp, and pig farm, a high concentration of heavy metals may be attributable to the above reasons.

Table 2. Level of heavy metals concentration in mangrove sediments from Sungai Besar River (mg/kg)

	Station S				**Station T**				**Station U**				**Station V**			
	min	max	ave	std	min	max	ave	std	min	max	ave	std	min	max	ave	std
As	2.857	3.753	3.316	0.448	5.945	7.545	6.762	0.800	8.716	9.562	9.014	0.475	8.456	11.302	9.827	1.426
Cd	nd	nd	nd	nd	0.021	0.028	0.024	0.004	nd	nd	nd	nd	0.008	0.020	0.015	0.006
Co	1.241	1.604	1.434	0.183	2.179	2.640	2.424	0.232	3.025	3.234	3.122	0.106	2.490	3.059	2.771	0.285
Cr	4.664	5.966	5.340	0.652	8.379	10.809	9.752	1.245	14.641	17.929	15.815	1.834	13.440	18.127	15.563	2.375
Hg	nd	nd	nd	nd	nd	nd	nd	nd	0.003	0.017	0.009	0.007	0.002	0.006	0.004	0.002
Mo	0.729	0.996	0.884	0.138	1.449	1.740	1.615	0.150	0.808	0.934	0.860	0.065	1.099	2.324	1.851	0.659
Ni	1.825	2.367	2.090	0.271	3.673	4.848	4.250	0.588	6.610	7.472	6.931	0.471	5.248	6.948	6.027	0.859
Pb	4.638	6.429	5.543	0.896	8.401	11.232	9.975	1.442	14.871	17.330	15.762	1.362	13.180	17.729	15.176	2.325

nd – not detected

Table 3. Comparison of heavy metal in this study with other studies and SQG sediment quality guidelines (mg/kg)

		As	**Cd**	**Co**	**Cr**	**Hg**	**Mo**	**Ni**	**Pb**	**Ref**
Sepang River Present study		3.316 -9.827 (7.230)	0.015-0.024 (0.020)	1.434 -3.122 (2.438)	5.340-15.815 (11.618)	0.004-0.009 (0.007)	0.860-1.851 (1.303)	2.090-6.931 (4.825)	5.543-15.762 (11.614)	
Kuala Gula, Perak, Malaysia		na	0.9–1.7	na	na	na	na	na	28.0–47.0	[28]
Sungai Puloh mangrove estuary, Malaysia		na	0.94	na	na	na	na	35.54	78.8	[29]
Juru River		9.15 – 11.47 (10.5)	0.572 – 0.696 (0.639)	7.70 – 8.89 (8.23)	78.85 – 89.04 (82.21)	na	na	na	27.64 – 45.25 (34.52)	[1]
Gulf of Khambhat region, West Coast of India		2.798±0.171	0.086±0.024	0.246±0.079	48.189±6.659	0.119±0.109	-	34.655±2.791	7.135 ±0.858	[30]
SQG sediment quality guidelines	USEPA	6	1	na	8.1	0.1	na	20.9	21	[1]
	average shale values	13	0.3	na	90	0.08	na	68	20	[25]

na – not available

Table 3 compares the heavy metal in this study to the Sediment Quality Guidelines (SGQ) and other river studies. SGQ values are used as a general measure to help ensure that some physical and chemical stresses in rivers do not exceed dangerous levels. A guideline value is a quantifiable amount (threshold) or condition of an indicator for a particular community value that is below or, for some stresses, over which we consider there to be a minimal likelihood of unacceptable impacts arising. The majority of heavy metals were discovered below the SGQ value, however arsenic (As) was located above the SGQ value of typical shale. On the other hand, it was found that most of studied elements in this research were lower than metal concentration in other studies except for Pb, which was slightly higher than the Gulf of Khambhat region, West Coast of India.

The enrichment factor (EF) was derived based to assess possible anthropogenic sources of hazardous elements as shown in Equation (1):

$$EF = [(M_X/M_Y)_{\text{sample}}]/[(M_X/M_Y)_{\text{shale}}] \qquad (1)$$

where $(M_X)_{\text{sample}}$ is referring to concentration of element in the experiment sample, $(M_Y)_{\text{sample}}$ refers the concentration of Fe in the sample, $(M_X)_{\text{shale}}$ refers to the concentration of abundant and common element in the average shale and $(M_Y)_{\text{shale}}$ refers to the concentration of Fe in the average shale. The EF values were interpreted as described in other study [22]. Classification of EF is listed in Table 4 [22].

Table 4. Enrichment Factor (EF) Classification

Classification	Sediment enrichment status
≤ 2	Low enrichment
2–5	Moderate enrichment
5–20	High enrichment
20–40	Very high enrichment
> 40	Extremely enrichment

EFs can be used as an indirect indicator of sediment pollution or toxicity but just assessing on the sediment toxicity at a given location merely on the basis of the enrichment factor is insufficient. Enrichment factor (EF) of Sepang Besar River mangrove is presented in Table 5. The EF analysis demonstrated an anthropogenic source in metal buildup in Sepang river mangrove sediments. The EF of As, Cd, Co, Cr, Hg, Mo, Ni, and Pb showed enrichment from a variety of sources, including industrial discharges, aquaculture activity, use of fertilisers, agricultural inputs, and mangrove exploitation [1-2]. In all sampling locations, the EFs of As, Mo and Pb exhibit enrichment (EF values 4.38 – 12.9, 1.71 – 3.67 and 0.94 – 2.68, respectively). Heavy metals, such as Cd, Cr, Co, Hg, and Ni, have low concentration at all test sites. Severe to excessive enrichment of As contamination exists. On the other hand, Mo and Pb enrichment ranges from low to moderate. These findings show that heavy metals (As, Mo, and Pb) have contaminated the sediments in these locations, with the main source being anthropogenic inputs from agricultural activities near the sampling area [23]. It is most likely that agriculturally produced animal waste such as pig manure is a higher enrichment of As, Mo and Pb in the study location. To evaluate sediment toxicity at a specific site, the enrichment factor is insufficient on its own. Assessing sediment toxicity for a specific location involves taking into account the degree of sediment contamination using the geo-accumulation index and comparison to sediment standards.

Muller (1969) developed the Geo-accumulation index (Igeo), which is one of the most dependable indexes for calculating a system's contamination state. The equation (4) can be used to compute it [24]:

$$I_{\text{geo}} = \log_2(M_n/1.5B_n) \qquad (2)$$

where M_n is referred to concentration of element in the experiment sample, B_n refers to the concentration of abundant and common element in the average [25]. The background matrix correction factor of 1.5 is used to reduce variation caused by lithogenic effects. Classification of Igeo is listed in Table 6 [26].

Table 5. Enrichment Factor (EF) of heavy metals in mangrove sediments from Sungai Besar River

	Station S				**Station T**				**Station U**				**Station V**			
	min	max	ave	std dev	min	max	ave	std dev	min	max	ave	std dev	min	max	ave	std dev
As	3.78	4.96	4.38	0.59	7.86	9.97	8.94	1.06	11.52	12.64	11.92	0.57	11.18	14.94	12.99	1.88
Cd	nd	nd	nd	nd	0.33	0.44	0.38	0.06	nd	nd	nd	nd	0.13	0.32	0.23	0.09
Co	0.12	0.15	0.14	0.02	0.21	0.25	0.23	0.02	0.29	0.31	0.30	0.01	0.24	0.29	0.26	0.03
Cr	0.11	0.14	0.12	0.02	0.20	0.25	0.23	0.03	0.34	0.42	0.37	0.04	0.31	0.42	0.36	0.05
Hg	nd	nd	nd	nd	nd	nd	nd	nd	0.10	0.48	0.26	0.19	0.05	0.16	0.12	0.05
Mo	1.45	1.98	1.75	0.27	2.87	3.45	3.20	0.29	1.60	1.85	1.71	0.12	2.18	4.61	3.67	1.23
Ni	0.05	0.07	0.06	0.01	0.10	0.14	0.12	0.02	0.19	0.21	0.20	0.01	0.15	0.20	0.17	0.02
Pb	0.79	1.09	0.94	0.15	1.43	1.91	1.70	0.24	2.53	2.95	2.68	0.21	2.24	3.01	2.58	0.39

nd – not detected

Table 6. Geo-accumuation (I_{geo}) classification

Classification	**Sediment pollution status**
< 0	Unpolluted
0–1	Unpolluted to moderately polluted
1–2	moderately polluted
2–3	moderately to strongly
3–4	Strongly
4–5	strongly to extremely strongly
> 5	extremely polluted

Table 7. Geo-accumulation index (I_{geo}) of heavy metals in mangrove sediments from Sungai Besar River

	Station S				**Station T**				**Station U**				**Station V**			
	min	max	ave	std dev	min	max	ave	std dev	min	max	ave	std dev	min	max	ave	std dev
As	0.082	0.475	0.296	0.197	1.139	1.483	1.325	0.172	1.691	1.824	1.739	0.068	1.647	2.066	1.864	0.209
Cd	nd	nd	nd	nd	-3.438	-3.008	-3.246	0.215	nd	nd	nd	nd	-4.757	-3.485	-3.940	0.644
Co	-4.918	-4.547	-4.709	0.186	-4.105	-3.828	-3.951	0.139	-3.632	-3.535	-3.587	0.048	-3.913	-3.616	-3.758	0.149
Cr	-5.036	-4.681	-4.841	0.178	-4.191	-3.823	-3.972	0.185	-3.385	-3.093	-3.274	0.148	-3.509	-3.077	-3.297	0.216
Hg	nd	nd	nd	nd	nd	nd	nd	nd	-5.212	-2.888	-3.782	1.172	-6.115	-4.514	-4.891	0.837
Mo	-1.303	-0.854	-1.026	0.227	-0.313	-0.049	-0.156	0.133	-1.155	-0.947	-1.065	0.104	-0.712	0.369	0.041	0.554
Ni	-6.109	-5.734	-5.914	0.188	-5.100	-4.700	-4.890	0.200	-4.253	-4.076	-4.184	0.089	-4.586	-4.181	-4.386	0.202
Pb	-2.179	-1.708	-1.922	0.236	-1.322	-0.903	-1.074	0.211	-0.498	-0.277	-0.414	0.111	-0.672	-0.244	-0.469	0.214

nd – not detected

According to the information in Table 7, the examined mangrove sediment in the Sepang mangrove forest is unpolluted since the geo-accumulation index (Igeo) of Cd, Co, Cr, Pb, Hg, Mo, and Ni is less than zero for all stations. The mangrove zone is considerably contaminated, according to the positive value of the As and geoaccumulation index (Igeo) at stations S and T, which is between 0 and 1. However, the Igeo index of 1 to 2 at stations T, U, and V indicates that the Sepang mangrove forest's analysed mangrove silt is contaminated with As. On the other hand, agricultural pesticides, lead-arsenate insecticides, and phosphate fertilisers are possible sources of As [1,27]. At Sungai Besar Sepang, a significant shift in mangrove vegetation was found, indicating a transformation from primary mangroves to mixed mangroves and inferior mangroves. Another noticeable alteration was the removal and degradation of mangroves at the mouth of the Sungai Sepang Besar river. Changes in land use and land cover in Sepang District have altered river features and resulted in a marked shift in mangroves [15,35].

In order to assess the interrelationships between the metal concentrations studied, correlation matrices were created using the dataset and statistical analysis. In Table 8, the values of the Pearson correlation coefficients (r) between metal concentrations are provided. The literature states that Al, Fe, and Mn are variables often connected to the geochemical matrix of sediments, while trace metals would provide a general pattern of the sediment contamination [1]. The correlation coefficients between Co and Cr (r=0.953), Co and Ni (r=0.985), Co and Pb (r=0.964), Cr and Ni (r=0.986), Cr and Pb (r=0.999), Ni and Pb (r=0.990), As and Co (r=0.951), As and Cr (r=0.977), and As and Ni (r=0.959) are all significantly positive. This relationship was also evident between Co and Ni (r=0.959). These considerable correlations between the concentrations of the elements in sediments suggested that the same pollution sources in the coastal environment were responsible for the sediments' strong positive heavy metal correlations [2,34]. The negative correlations between cadmium (Cd) and Co (r=0.034), Cr (r=0.417), Ni (r=0.310), Pb (r=0.374), as well as Hg (r=0.243), As (r=0.343), and As (r=0.343) are all present. This demonstrates that they compete for sediment binding locations, have a diversity of input sources, or utilise various sediment interaction methods.

4. Conclusions

Heavy metal pollution from the sediments of the Sepang Besar River mangrove was investigated by measuring the amount of various metals in the sediments at four sampling points. The results of this study showed the presence of As, Cd, Pb, Cr, Ni, Co, Mo and Hg in the mangrove sediments of the Sepang Besar River. We were able to define the relevant amounts of heavy metals in the designated section of the Sepang Besar River using pollution assessment methods. At all sampling places, the concentrations of the examined metals in sediments were lower than the resulting average shale values and the USEPA, indicating that adverse impacts do not occur frequently in all locations. Based on the results of the enrichment factor and the geo-accumulation index, concentrations of As, Mo and Pb must be monitored in the future to avoid any potential pollution hazards. Elemental profiling of the adjacent biotic community may be affected by increased sediment contamination from a variety of point and nonpoint.

Table 8. Pearson correlation coefficient matrix among significant metals within the sediments from Sungai Besar River

	As	**Cd**	**Co**	**Cr**	**Hg**	**Mo**	**Ni**	**Pb**
As	1							
Cd	-0.343	1						
Co	.951**	-0.034	1					
Cr	.977**	-0.417	.953**	1				
Hg	0.182	0.995	0.740	0.710	1			
Mo	0.514	0.423	0.345	0.355	-0.243	1		
Ni	.959**	-0.310	.985**	.986**	0.792	0.286	1	
Pb	.979**	-0.374	.964**	.999**	0.682	0.357	.990**	1

Pearson's correlation between Trace elements (N = 12)

** Correlation is significant at the 0.01 level (2-tailed); * Correlation is significant at the 0.05 level (2-tailed)

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