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Ecological Risk Assessment of Heavy Metal Pollution in Surface Water and Sediment of Lahug River, Cebu, Philippines

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Abstract: The Lahug River, a significant urban waterway in Metro Cebu, was studied to assess its water quality and sediment characteristics. Physicochemical properties, such as pH and dissolved oxygen (DO), were measured in situ using a Multi-probe digital meter. At the same time, metal concentrations in both water and sediments were determined through Flame Atomic Absorption Spectroscopy (FAAS) with multiple standard addition techniques. In water, metal concentration showed a decreasing trend of Zn > Pb > Cu > Cr. Notably, copper (Cu) and chromium (Cr) exceeded the National Environmental Protection Agency (NEPA, 1989) threshold at the downstream station, while all stations exceeded the limit for zinc (Zn). In sediments, copper emerged as the most prevalent metal. Statistical analysis indicated significant correlations among Cu, Zn, Pb, and Cr, suggesting similar pollution sources or behaviors in the river environment. Ecological risk assessment revealed that the downstream area exhibited the highest risk, highlighting the urgency of rehabilitative measures to protect the river ecosystem. A comprehensive, large-scale environmental risk assessment is recommended to mitigate further degradation and ensure sustainable management of the Lahug River

Keywords: Kamputhaw river; ecological risk; water and sediment quality

1. Introduction

Metal contamination in the aquatic environment has attracted global attention due to its environmental toxicity, abundance, and persistence. Large quantities of hazardous chemicals, especially heavy metals, have been released into rivers worldwide due to rapid global population growth, intensive domestic activities, and expanding industrial and agricultural production (Srebotnjak et al. [1], Su et al. [2], Aziz et al. [3], Rizabal et al. [4]). Rivers in urban areas have also been associated with water quality problems because untreated domestic and industrial waste are discharged into the water bodies [5]. These problems of urban river pollution and ecological damage are becoming more and more critical, such that environmental monitoring and assessment are on the frontier to help environmentalists and government officials save and rehabilitate these river resources Rohde et al. [6], Richardson et al. [7]). This makes river pollution one of the most critical environmental problems of the 20th century, with agricultural, commercial, industrial, and anthropogenic activities as the main contributors Yi et al. [8], Bensig et al. [9], Maglangit et al. [10].

Barangay Lahug is in a densely populated area in Cebu City, Philippines. It has an 8.5 km long Lahug River with a basin area of 6.3 km². With the ever-increasing urbanization due to the rapid development of technology and the business process outsourcing (BPO) economy of Lahug, vast quantities of domestic and industrial wastes are disposed into its river, which leads to severe pollution and deterioration of its river ecosystem, as reported by Bensig et al. [9] and Maglangit et al. [10] that rapid oxygen depletion and high phosphorus content of the river water indicated that it is polluted. Dumping this untreated wastewater leads to an increase in pollutants present in the river. It causes dissolved oxygen (DO), biological oxygen demand (BOD), total phosphorus (TP), fecal coliform count (FC), and total coliform count (TC) in some parts of the Lahug River to exceed the Department of Environment and Natural Resources – DAO 34 (DENR-DAO 34) standards. In addition, the discharge of untreated waste into the water bodies also increases the level of metals in river water Khadse et al. [11], Venugopal et al. [12]

The introduction of metal pollutants in various forms in the environment can pose severe threats to the ecological system due to their negative impacts on most life forms (Yang et al. [13], Jaiswal et al. [14], [15]). Geolin et al. [16] reported that the univalve *Canarium labiatum* accumulated high concentrations of metals when there was a high concentration of metals in sediments. Although all life forms require some amounts of heavy metals, there is a threshold limit to this requirement (Cervantes et al. [17]). At high concentrations, heavy metal ions react to form toxic compounds in cells ([18], Choudhury et al. [19]). Another major problem with metals is their persistence, as they tend to persist indefinitely in the food chain (Gupta et al. [20], Aleem et al. [21]). Thus, assessing the concentrations and distribution of heavy metals in the Lahug riverine ecosystem is important.

2. Materials and Methods

2.1 Description of the study area

This study focused on an important urban river, the Lahug River, in the central part of Cebu City, Philippines. The river begins from the upper areas of Laguerta, passes into the midstream local communities of Kamputhaw, Capitol, and Lorega, and then courses into the downstream areas of T. Padilla, Day-as, and Tejero. This river is a natural flood drain for Cebu (Maglangit et al. [10]). The three sampling sites of Maglangit et al. [10] and Bensig et al. [9] were used as sampling sites in this study to represent the upstream, midstream, and downstream. The location of the sampling sites and their GPS coordinates are shown in Figure 1.

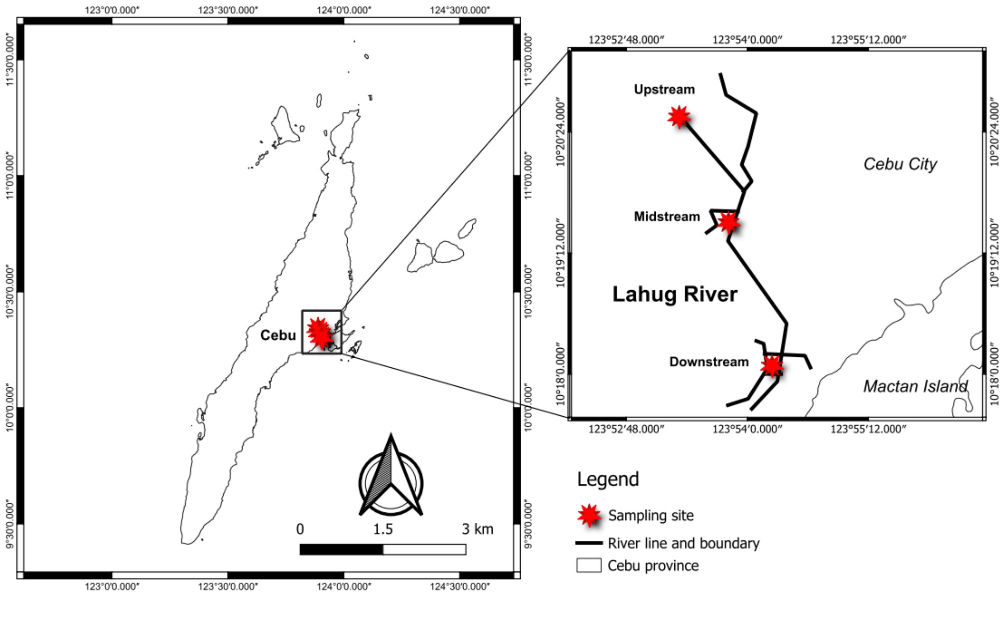

Figure 1. Location of sampling sites along Lahug River, Cebu City, Philippines.

2.2 Sampling design

As a preliminary study, the sampling for water and sediment was carried out in May, which is considered a dry season in the Philippines, with three sampling stations to represent the upstream, midstream, and downstream of the river, with a description of the stations and their coordinates tabulated in Table 1.

Table 1. Description of Sampling stations and their coordinates

Station	Coordinates	Description
Station 1 (Upstream) (5.8 km from river delta)	10° 20' 33.1" N, 123° 53' 19.4" E	Relatively clear river water. Fish and vegetation were seen; stones were covered with algae and moss; residents were seen bathing and doing laundry. A few meters up is a large construction site.
Station 2 (Midstream) (3.5 km from the river delta)	10° 19' 30.3" N, 123° 53' 48.8" E	The river water was turbid with an unpleasant smell due to the large volume of solid wastes such as feces, plastics, papers, and diapers dumped in the river. This part of the river transverses the residential and industrial areas. Concrete dikes modified the riverbanks.
Station 3 (Downstream) (900m from river delta)	10° 18' 4.90" N, 123° 54' 14.70" E	Floating solid wastes; stinky black water bubbling at the surface, plenty of households nearby; presence of fecal matter; stagnant water

2.3 Sample collection and preparation

2.3.1 Water sampling and analysis

The river water's pH and Dissolved oxygen (DO) were analyzed on-site. Approximately 30 L of river water was collected in each station and placed in a polyethylene bottle with 50% (v/v) HNO_3. The samples were then digested using the APHA method upon arrival at the laboratory. Exactly 500 mL of filtered river water was digested with 3 mL HNO_3 until the volume was about 10 mL, filtered, and diluted to the 50 mL mark of the volumetric flask. The sample was then analyzed using flame atomic absorption spectroscopy (FAAS) and the multiple standard addition technique. Five mL aliquots of the stock sample were transferred to 25-mL volumetric flasks, added with 0.00-, 0.10-, 0.20-, 0.30- and 0.40 ppm copper, chromium, lead, and zinc standards and diluted to the mark with distilled water. The samples were thoroughly mixed and analyzed in triplicates with AAS.

2.3.2 Sediment sampling and analysis

The pH of the soil was determined on-site. Composite sediment samples were collected at each sampling site using standard protocol [22]. The river bed sediment samples were taken at a 0 - 5cm depth using a core sampler. Three composite samples of mass approximately 200g were collected at each station. The samples were then chilled and contained in polyethylene bags. Upon arrival at the laboratory, the sediments were air-dried until parched, sieved at 180 µm, and homogenized using a ball mill. The homogenized samples were then oven-dried at 110°C to constant weight. The digestion method used to analyze metal concentrations in sediment was based on the USEPA Method 3050B. About 1 g of the sediment sample was placed in a 100-mL beaker and was digested with repeated additions of concentrated HNO_3 and H_2O_2 for 30 minutes to near boiling and then refluxed. The sediment digest was filtered and rinsed with hot HCl, followed by hot water. The filter paper and residue were returned to the digestion flask, refluxed with additional HCl, and then filtered again. The digest was diluted to a final volume of 100 mL and is now ready for analysis. Five mL aliquots of the sediment samples were transferred to 25-mL volumetric flasks, separately added with 0.00-, 0.10-, 0.20-, 0.30- and 0.40 ppm standards of copper, chromium, lead, and zinc and diluted to the mark with distilled water. The samples were thoroughly mixed and analyzed in triplicates with FAAS.

2.4 Instrumental analysis and quality assurance

The pH and dissolved oxygen of the river water were measured using an Orion pH meter and a Milwaukee DO meter, respectively. Metal concentrations were measured using the Shimadzu 6300 Flame Atomic Absorption Spectrophotometer (FAAS). FAAS grade MERCK Titrisol® solutions were used to prepare a calibration curve with R2 > 0.999 and were accepted for concentration calculation. All test batches were evaluated using an internal quality approach and validated if they satisfied the defined internal quality controls (IQCs). For each experiment, a run included a blank recovery test (RT), and samples were analyzed in triplicate with acceptable relative standard deviation (RSD, <5%) to eliminate any batch-specific error and provide good levels of accuracy and precision. The results were then reported in means and standard deviation of elemental values.

2.5 Statistical analysis

Means and standard deviations were calculated using Excel. A two-way Analysis of Variance (ANOVA) was performed in GraphPad Prism 6.0 to assess significant water quality differences between sampling sites. Pearson's r was used to compute the relationship between the heavy metals Cu, Pb, Cr, and Zn in the sediments.

2.6 Assessment of ecological risk

The following sediment ecological risk assessment guidelines were used to evaluate the degree to which the sediment-associated metals adversely affect aquatic organisms:

2.6.1 Pollution load index:

To assess the sediment quality, an integrated approach of the pollution load index of the four metals is calculated according to Suresh et al. [23]. The PLI is defined as the nth root of the multiplications of the contamination factor of metals (CF), as shown in the equation of

$$PLI = (CF_1 \, x \, CF_2 \, ... \, CF_n)^{1/n} \quad (1)$$

where the PLI value of 0 is interpreted as unpolluted, PLI of 1 means polluted, and PLI > 1 is highly polluted.

2.6.2 Contamination Factor (CF):

CF was calculated using the following equation or by dividing the content of each metal by the background values in sediment (Suresh et al. [23]):

$$CF_{metal} = C_{metal} / C_{background} \quad (2)$$

2.6.3 Geoaccumulation index (Igeo).

Geoaccumulation index values were calculated using the equation of

$$I_{geo} = log_2 \, (C_n / 1.5 \, B_n) \quad (3)$$

where Cn is the measured concentration of metal n in the sediment and B_n is the geochemical background value of element n in the background sample (Gao et al. [24], Islam et al. [25])

2.6.4 Potential ecological risk (PERI).

The potential ecological risk index (PERI) is also introduced to assess the degree of contamination of heavy metals in the present sediments. Equation of

$$PERI = \sum RI = \sum (T_{rf} \times CF)$$

It was used to calculate the PERI proposed by Gao et al. [24]). PERI is the comprehensive potential ecological index, the sum of RI. It represents the sensitivity of the biological community to the toxic substance and illustrates the potential ecological risk caused by overall contamination.

3. Results and Discussion

3.1 pH and dissolved oxygen

Values for river water samples ranged from 6.71 to 6.82 ppm for dissolved oxygen. The dissolved oxygen values indicate sufficient oxygen supply to support aquatic life in the river. pH values also ranged between 6.68 and 6.77 for water samples and sediments and 6.71 and 6.82 for sediments.

3.2 Metal concentration in water

The results of heavy metal concentrations in the surface waters of Lahug are shown in Table 2, revealing a significant variation ($p<0.05$) among the sites. The average concentration of the studied metals in water followed a decreasing order of Zn>Pb>Cu>Cr. The levels of all metals exceeded the threshold values of the National Environmental Protection Agency [26] at station 3, while stations (1 and 2) failed for both Cu and Zn. This means the quality of water entering the Mactan Channel failed fishery water quality standards, making all fish susceptible to metal contamination. Copper contamination downstream is attributed to domestic sewage and runoff from the widespread flower farming in the area. On the other hand, zinc contamination mostly comes from everyday human activities—industrial waste, urban runoff, and even things like the wear and tear of tires and corrosion of galvanized metal ([27], Alburo et al. [28]). Together, these pollutants can build up in aquatic life and potentially affect the ecosystem's overall health, highlighting the need for regular monitoring and better waste management practices.

Table 2. Heavy metal concentrations in surface water (ug L^{-1}).

Sampling Stations	Mean Metal Concentrations in ug/L			
	Cu	**Pb**	**Cr**	**Zn**
1	0.97	nd	nd	23.2
2	2.30	nd	nd	40.2
3	10.2*	15.0*	6.0*	56.4*
NEPA	0.01	0.05	0.01	0.01

* Significantly different

3.3 Metal contamination in sediments

The mean concentration of heavy metals in the sediment samples from the Lahug River is presented in Table 3. Copper was found to be the most abundant element in the river, while lead, chromium, and zinc varied in the following sequence irrespective of sampling locations: Cu>Zn>Cr/Pb. Concentrations of the metals at Station 3 were much higher than at other sites because this site is located downstream of the river, where there is extensive discharging of urban waste (Islam et al. [25], Luo et al. [29]).

To predict the heavy metal pollution in Lahug sediments, a comparative study was made with both background World Surface Rock Average (WSA) and toxicological reference values (Effect Range Low (ERL), Effect Range Medium (ERM) and Toxicity Reference Value (TRV)). Comparative results are also presented in Table 3.

Table 3. Mean ± sd metal concentrations in Lahug River and comparison with background and toxicological reference values

Metals	Sampling Stations			WSA[a]	ERL[b]	ERM[b]	TRV[c]
	1	2	3				
Copper, mg/kg	74.40 ± 1.10	94.80 ± 0.75	1152 ± 52.2	32	70	390	16
Lead, mg/kg	6.22 ± 0.26	6.733 ± 0.12	39.33 ± 0.68	20	35	110	31
Chromium, mg/kg	8.927 ± 0.48	10.15 ± 0.23	26.73 ± 0.11	97	80	145	26
Zinc, mg/kg	65.63 ± 0.55	92.13 ± 0.55	416.3 ± 0.58	129	120	270	110

[a] World Surface Rock average (Martin et al. [30]).

[b] Effect range low and effect range medium for freshwater ecosystems (Bai et al., [31]).

[c] Toxicity Reference Value (Mohiuddin et al. [32]).

Only Cu exceeded the WSA in all stations of Lahug River sediments. Lead and zinc levels exceeded WSA only at Station 3. This indicates the anthropogenic introduction of the metals in the river. Based on toxicological reference thresholds, the average concentrations of Cu at all stations exceeded the Effects Range-Low (ERL) value, while only Station 3 surpassed the Effects Range-Median (ERM). Similarly, Zn concentrations exceeded the ERM at Station 3 alone. Lead at Station 3 is also beyond its corresponding ERL value and may cause occasional biological effects on aquatic organisms in the area. In contrast, no biological effect will be observed due to Cr (Harikumar et al. [33], Marin et al. [34]). All metals at station 3 are above the TRV values. Exposures above the TRV indicate that the dose or concentration may exceed the threshold where toxic effects are likely. Depending on the organism and the substance, this can lead to adverse health effects, such as biochemical, physiological, or reproductive harm (Allard et al. [35]). Table 4 shows that all metal contaminants in the sediments are significantly correlated. According to Suresh et al. [36], a high correlation coefficient between metals signifies that the metals have common sources, mutual dependence, and identical behavior during transport.

Table 4. Pearson's correlation coefficient between the heavy metals in river sediments ($p<0.05$).

Metals	Sampling Stations			
	Cu	**Pb**	**Cr**	**Zn**
Cu	1			
Pb	0.9993	1		
Cr	0.9975	0.9981	1	
Zn	0.9974	0.9982	0.9995	1

3.4 Ecological Risk Assessment of metal pollution in Lahug River sediments

3.4.1 Pollution Load Index

The calculated pollution load index (PLI) provides a comparative assessment of the status of sediment quality in the Lahug River, as summarized in Figure 2. Based on this information, station 3 of the river is the most polluted due mainly to the effects of urban activities.

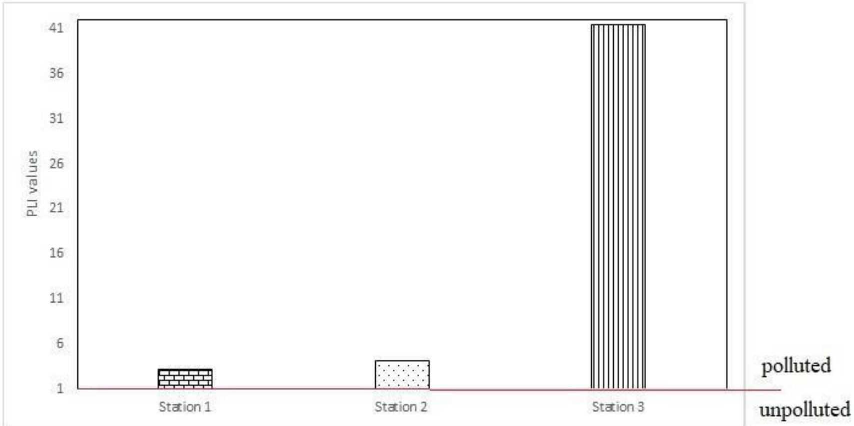

Figure 2. Pollution load index (PLI) value of heavy metals in Lahug River, Cebu, Philippines, sediment.

3.4.2 Contamination Factor

Figure 3 illustrates that the contamination factor for all metals followed a descending order of Cu > Zn > Pb > Cr. The CF values indicated a low to moderate degree of contamination (CF > 1) across all metals, except for Cu at station 3, which exhibited a considerably higher contamination level. This elevated CF for Cu suggests a significant localized source, likely linked to anthropogenic inputs such as domestic sewage, urban runoff, or corrosion of copper-containing materials. Considerable contamination of Zn levels, particularly at station 3, may be attributed to the widespread use of zinc-containing fertilizers or galvanization processes in nearby areas. The relatively lower CF values for Pb and Cr, with detections only at station 3, further support the presence of point-source pollution in the downstream portion of the Lahug River. The contamination factor (CF) of 36.01 for Cu at station 3 indicates a very high level of contamination, far exceeding the threshold for significant ecological concern. Such an elevated value suggests the presence of a major localized source, potentially stemming from continuous inputs of untreated domestic wastewater, improper disposal of copper-containing industrial effluents, or runoff from urban infrastructure and agricultural areas using copper-based agrochemicals (Oquiñena-Paler et al. [37], Cañete et al. [38]).

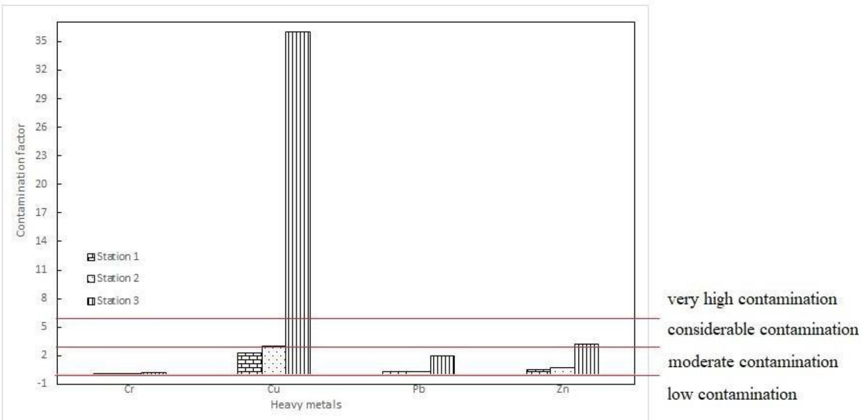

Figure 3. Contamination factor (CF) values of heavy metals in river sediments.

3.4.3 Geo-accumulation Index, I_{geo}

According to the Muller scale, the calculated results of Igeo values shown in Figure 4 for Cu sediment quality are considered as heavily to significantly contaminated (4≤Igeo≤5) for station 3, moderately contaminated at station 2, and uncontaminated to moderately contaminated at station 1. Zinc is also uncontaminated to moderately contaminated at station 2, with no contamination for Cr and Zn in all stations.

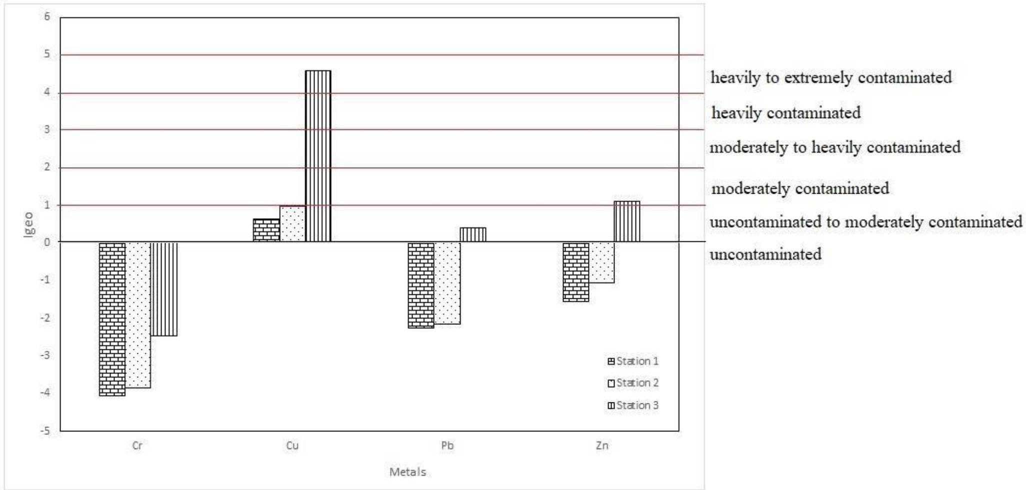

Figure 4. Geo-accumulation index (Igeo) values of heavy metals in river sediments.

3.4.4 Potential ecological risk index

The potential ecological risk index (RI) of surface sediments from the Lahug River was further calculated to confirm the other evaluations, with the results summarized in Table 6. In this study, the metals tested were copper (Cu), lead (Pb), zinc (Zn), and chromium (Cr), with the calculated Ei_r values indicating varying levels of ecological risk. Copper exhibited the highest risk, with the ecological risk coefficients following the order Cu > Pb > Zn > Cr, suggesting that copper posed the most significant potential threat to the aquatic ecosystem in the Lahug River, consistent with its elevated toxicity factor compared to the other metals. The mean Ei_r values for Pb, Cr, and Zn were all below 40, categorizing these metals under the low ecological risk classification, indicating that their concentrations in the sediments were not high enough to impact the ecosystem significantly. However, copper showed a notably higher Ei_r value, particularly at station 3, where the risk index (RI) exceeded 150, indicating a high ecological risk due to the significantly elevated copper concentrations at this site. This pronounced ecological threat highlights the need for further investigation and targeted mitigation strategies. In comparison, Villacarlos et al. [39] reported a low ecological risk classification in the sediments of the Balamban coastline, an area exposed to a diffused source of metal input from a nearby shipyard facility, suggesting that the localized and possibly point-source contamination in the Lahug River may pose a more acute environmental concern.

Table 5. Evaluation of the potential ecological risk of heavy metals pollution in sediments from Lahug River.

Sampling Stations	Risk Index (E^i_r)				Potential Ecological Risk Index (Peri)	Risk Grade [40[
	Cu $C^i_f \; x \; T^i_f$	Pb $C^i_f \; x \; T^i_f$	Cr $C^i_f \; x \; T^i_f$	Zn $C^i_f \; x \; T^i_f$		
1	11.6	1.94	0.251	0.517	14.3	Low ecological risk
2	14.8	2.10	0.286	3.63	20.9	Low ecological risk
3	180.1	12.3	0.753	16.3	209	High ecological risk

3.4.5 Sediment Quality Guideline (SWQ) by USEPA

The chemical contaminations in the sediments were evaluated by comparison with the sediment quality guideline proposed by USEPA. These criteria are shown in Table 5. The present study indicates that all sites are heavily polluted with Cu, while Pb, Cr, and Zn exhibit varying contamination levels depending on the location. Specifically, station 2 is moderately polluted with Pb, Cr, and Zn, whereas station 1 shows no significant contamination concerning these metals. The Sediment Quality Guidelines (SQGs) established by the United States Environmental Protection Agency (USEPA) further assessed the chemical contamination levels in sediments. These guidelines benchmark for interpreting sediment contamination and potential ecological risks by providing specific contaminant concentration thresholds based on dry sediment weight. This approach directly categorizes pollution levels into not polluted, moderately polluted, or heavily polluted. In contrast, other commonly used indices such as the Contamination Factor, Geoaccumulation Index, Pollution Load Index, and Potential Ecological Risk Index rely on comparisons to background or reference values to assess anthropogenic input, making them more indicative of pollution sources rather than ecotoxicological effects.

In the present study, Cu concentrations across all three sampling stations exceeded 60 mg/kg, with values ranging from 74.40 ± 1.10 mg/kg at station 1 to 1152 ± 52.2 mg/kg at station 3, classifying all sites as heavily polluted and indicating significant anthropogenic input, particularly at station 3. Pb levels surpassed the 6 mg/kg mark in all stations, suggesting ecological risk despite the absence of a defined pollution category in the USEPA guidelines. Cr levels at station 1 (8.927 ± 0.48 mg/kg) were within the not polluted range, whereas station 3 (26.73 ± 0.11 mg/kg) slightly exceeded the 25 mg/kg threshold, indicating moderate contamination. Zn concentrations showed a gradient from not polluted at station 1 (65.63 ± 0.55 mg/kg) to heavily polluted at station 3 (416.3 ± 0.58 mg/kg), further supporting the presence of localized contamination sources.

Table 6. Environmental Protection Agency (EPA) guidelines for sediments, mg/kg dry weight

Metal	Not Polluted	Moderately Polluted	Heavily Polluted	Present Study		
				Station 1	Station 2	Station 3
Cu	<40	40-60	>60	74.40 ± 1.10	94.80 ± 0.75	1152 ± 52.2
Pb	-	-	>6	6.22 ± 0.26	6.733 ± 0.12	39.33 ± 0.68
Cr	<25	25-75	>75	8.927 ± 0.48	10.15 ± 0.23	26.73 ± 0.11
Zn	<90	90-200	>200	65.63 ± 0.55	92.13 ± 0.55	416.3± 0.58

4. Conclusion

The study assessed heavy metal contamination in the surface sediments of the Lahug River using multiple indices-CF, PLI, Igeo, and RI/PERI. Results showed that copper posed the highest contamination and ecological risk, particularly at station 3, where the RI exceeded 150, indicating a high ecological threat. CF and Igeo values indicated moderate to considerable Cu enrichment, while Pb, Zn, and Cr generally fell within low to moderate contamination and low-risk classifications. PLI values suggested cumulative pollution, emphasizing the impact of anthropogenic activities. When compared against the Sediment Quality Guidelines established by the U.S. Environmental Protection Agency (USEPA), Cu concentrations at all stations and Zn in station 3 exceeded the guidelines, indicating possible adverse biological effects. In contrast, Pb, Zn, and Cr were mostly below these thresholds. These results confirm that the Lahug River is experiencing localized but significant heavy metal pollution, predominantly from copper, likely due to point-source discharges. The study highlights the need for targeted pollution source identification, strengthened monitoring, and the implementation of appropriate remediation strategies to protect the river's ecological integrity.

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Validation: Alburo, RP; Formal analysis: Alburo, RP and Villegas, LMG; Investigation: Alburo, RP and Villegas, LMG; Resources: Alburo, RP; Data curation: Alburo, RP and Villegas, LMG; Writing—original draft preparation: Alburo, RP and Villegas, LMG; Writing—review and editing: Alburo, RP and Villegas, LMG; Visualization: Villegas, LMG; Supervision: Alburo, RP; Project administration: Alburo, RP; and Funding acquisition: Alburo, RP. Additionally, all authors have read and agreed to the published version of the manuscript.

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