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Assessment of heavy metals pollution and ecological risk in surface sediments in the Tam Giang - Cau Hai lagoon area, Thua Thien Hue Province

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ABSTRACT

The Tam Giang - Cau Hai lagoon system plays an important role in the economic development of the Thua Thien Hue area. In this project, the study and analysis of several heavy metals, such As, Cd, Pb, Zn, Hg, Cr, and Cu contained in the lagoon system were taken to assess pollution levels and ecological risk. Thirty-five sediment samples were collected in the research area in January 2024 (rainy season). Metals' concentration was analyzed by (Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The assessment method is based on comparisons with the national technical regulation on sediment quality (QCVN 43:2017). It includes the geo-accumulation index (I_{geo}), enrichment factor (*EF*), pollution load index (*PLI*), and ecological risk index (*RI*). Results indicate the concentration of heavy metals were As (4.65); Cd (0.08); Pb (11.93); Zn (33.66); Hg (0.07); Cr (12.88) and Cu (4.97) mg/kg that within the allowed limits for surface water and brackish water sediments in QCVN 43:2017/BTNMT (national technical regulation on sediment quality). The geo-accumulation index (Igeo) for the heavy metals (As, Cd, Pb, Zn, Hg, Cr and Cu) confirmed that the sediment was not polluted. The enrichment factor (*EF*) proved that human activities are the primary source of increasing concentration of As, Cd, Hg, and Pb in sediment. Based on *PLI*, the pollution level of Tam Giang - Cau Hai lagoon is considered good, and the ecological risk (*RI* = 20.75) in the study area is low.

Keywords: Sediment, heavy metals, Tam Giang - Cau Hai lagoon, ecological risk.

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INTRODUCTION

In the rapid industrialization and urbanization of coastal areas, many pollutants are produced through various activities. These pollutants are quickly released into coastal ecosystems [1, 2], especially enclosed and semi-enclosed bays such as lagoons [3, 4]. Metals are highly concerning because of their high toxicity, reduction, and biological accumulation, which happen when the absorption rate is higher than the metabolism rate. Sediment, less affected by hydrometeorological conditions and stable chemical properties, is the primary source of metal pollutants. After being released into the coastal system, almost all heavy metals will be adsorbed onto suspended particles and then precipitated in sediment [5], leading to rich heavy metals sediment formation. Moreover, sediment can contaminate seawater and significantly affect marine organisms [6]. Therefore, researching heavy metals in sediment and their effect on the ecosystem plays a vital scientific role.

Lagoons are considered vital ecosystems of the marine environment. Global biogeochemical cycles heavily depend on these brackish water ecosystems [7]. A lagoon environment is necessary for aquatic and terrestrial organisms, including humans.

Tam Giang - Cau Hai lagoon system is Vietnam's largest brackish water lagoon, stretching 68 kilometers along the coast in northwest to southeast direction. It is a dynamic ecosystem affected directly by tide and river flow. There are two entrances to exchange with seawater: Thuan An in the north and Tu Hien in the south. Lagoons are rich in plants and animals, considered the richest in Southeast Asia [8], providing many social and economic benefits such as fisheries resources, mangrove vegetation, and contributing to ecotourism. The economic activities surrounding the lagoon are mostly agriculture, fishing, marine exploitation, port transportation, and tourism services [9]. Besides, the environmental quality of the lagoon is affected by the upstream due to rivers flowing into it. Researching heavy metals in the lagoon sediment has not gained much attention. Hence, the primary purpose of this study is to assess the concentration of heavy metals and ecological risk to propose solutions to protect the lagoon.

METHODOLOGIES AND EXPERIMENTS

Sediment sampling

Thirty-five sampling areas were chosen based on distance from pollution sources such as fishing grounds, markets, ports, and marine traffic lines. The samples were collected in January 2024 (rainy season) from the top to 10 centimeters of sediment along the lagoon's left and right banks and center, then preserved in a cooler to transport to the laboratory. Sampling positions are shown in Figure 1.

Analysis method

Samples were prepared for analysis by air-drying and then grinding to powder with particle size less than 0.25 mm.

Heavy metal analysis procedure: approximately 0.5 g of prepared sediment was used for analysis. The samples were digested in a mixture of HCl, HNO_3, and H_2O_2 for 3 hours [10], then filtered and diluted to a specific volume. Metal concentrations were determined using an Agilent Technologies 7900 ICP-MS. The grain size distribution was determined according to the Vietnamese Standard TCVN 8567:2010, and the total organic carbon content was analyzed using the Walkley Black method (TCVN 8941:2011). The method's accuracy was verified using CRM016 from Sigma Aldrich; the recovery rates are 90–110%.

Assessment method for sediment quality

To fully assess sediment quality and potential ecological risk, it is necessary to combine multiple standards and geochemical parameters. These include the Vietnamese sediment standards, the geo-accumulation index (Igeo), the enrichment factor (*EF*), the pollution load index (*PLI*), and the ecological risk index (*RI*).

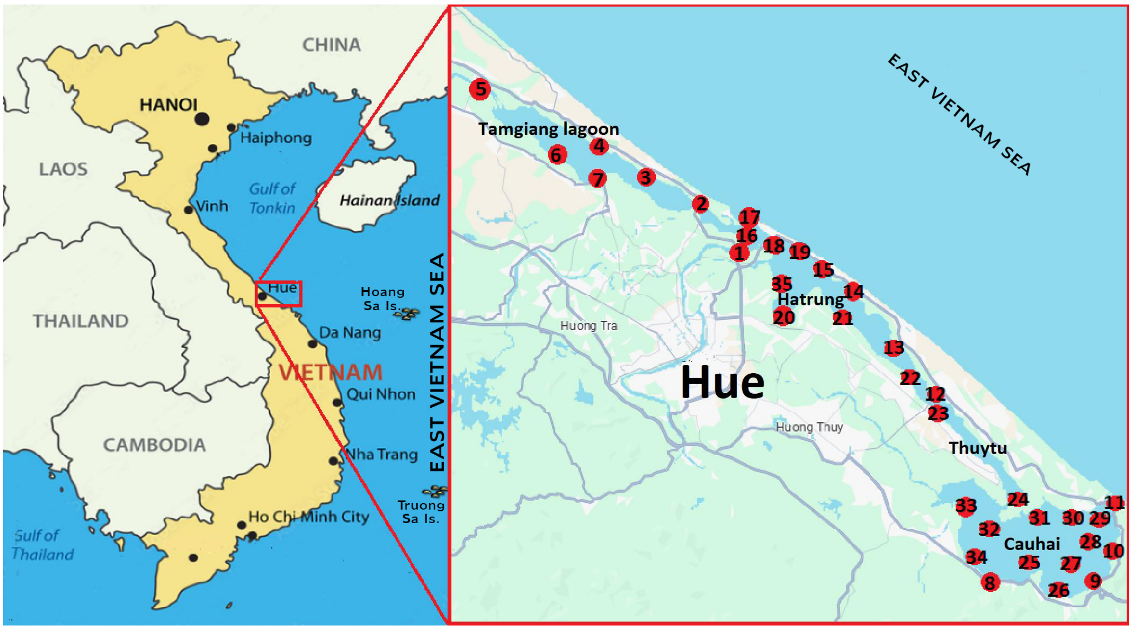

Figure 1. Diagram of sampling sites

Assessment comparing to the standard

The research results were compared with the National Technical Regulation on Sediment Quality of Vietnam (QCVN 43:2017/BTNMT). Threshold values were shown in Table 1.

Table 1. Threshold values of elements in saline and brackish water sediment

Element	As	Cd	Pb	Zn	Hg	Cr	Cu
Threshold (mg/kg)	41.6	4.2	112	271	0.7	160	108

The geo-accumulation index (I_{geo})

The geo-accumulation index (I_{geo}), a method proposed by Muller (1996) [11], is used to assess the pollution of heavy metals. The Igeo value of each metal is calculated by the following formula:

$$I_{geo} = \left(\frac{C_n}{1.5B_n} \right)$$

where: C_n is the concentration of metal in sample *n* (mg/kg); and B_n is the background value. However, the background value of this location is still undetermined, so the values according to K. Turekian and Hans Wedepohl (2014) [12] were used in this study. The B_n values of each metal, including As, Cd, Pb, Zn, Hg, Cr, and Cu, respectively, are 13, 0.3, 20, 95, 0.4, 90, and 45 ppm. The assessment is based on 7 levels shown in Table 2 [13].

Table 2. levels of pollution based on I_{geo}

I_{geo}	Scale	Level
$I_{geo} \le 0$	0	Unpolluted
$0 < I_{geo} \le 1$	1	Low Pollution
$1 < I_{geo} \le 2$	2	Moderate pollution
$2 < I_{geo} \le 3$	3	Moderate to heavy pollution
$3 < I_{geo} \le 4$	4	Heavy pollution
$4 < I_{geo} \le 5$	5	Serious pollution
$I_{geo} > 5$	6	Very serious pollution

The enrichment factor (EF)

Enrichment Factor (*EF*) is a geochemical index used to assess the degree of anthropogenic enrichment of a specific element in a sample compared to the background level [14]. It is used to discriminate the origin of elements between natural and anthropogenic sources of contamination. In this study, Fe is chosen to represent a reference element, and the *EF* was calculated using the following formula:

$$EF = \frac{\frac{C_i}{C_{Fe}} sample}{\frac{C_i}{C_{Fe}} background}$$

where the (C_i/C_{Fe}) × *sample* and (C_i/C_{Fe}) × *background* are ratios between the concentration of a particular metal (C_i) and Fe (C_{Fe}) of the lagoon sediment and the corresponding background sample, for this research, the background iron content of shale was used as a reference value which is 47,200 ppm [12]. An *EF* higher than 1.5 indicates the anthropogenic origin of metals, while other values show the natural origin. The degree of enrichment can be classified into five levels shown in Table 3 [15]:

Table 3. The levels of metals enrichment

Enrichment Factor (*EF*)	Enrichment level
EF ≤ 2	Low
2< *EF* ≤ 5	Moderate
5 ≤ *EF* ≤ 20	Significant
20 < *EF* ≤ 40	High
EF > 40	Very high

Pollution Load Index (PLI)

The pollution index (*PLI*) proposed by Tomlinson et al., (1980) [16] is used to assess the rate of pollution. The *PLI* was calculated using the following formula:

$$CF_i = \frac{C_{m\ sample}}{C_{m\ background}}$$

$$PLI = \sqrt[n]{CF_1 * CF_2 * \ldots CF_n}$$

where: CF_i is the pollution index for a specific element; $C_{m\ sample}$, and $C_{m\ background}$ is the element's concentration in the sample and background; according to K. Turekian and Hans Wedepohl (2014) [12], *n* is the number of metals tested. The *PLI* is classified into three categories: *PLI* < 1 indicates good environmental quality and no pollution, *PLI* = 1 indicates a basic pollution level, and *PLI* > 1 suggests declining environmental quality and rapidly increasing pollution.

Risk Index

The risk index is a method for assessing the potential risk of metals [17]. The RI was calculated using the following formula:

$$RI = \sum_i^n \frac{T_r^i C_i}{B_i}$$

where: C_i indicates the concentration of heavy metals in sediment; B_i is the value for the background concentration of the corresponding metal. Tri represents toxicity and biological sensitivity applied according to the values proposed by Hakanson in 1980 [17]. The *RI* value is classified into four categories (Table 4).

Table 4. Risk Index

RI value	Classification
< 150	Low risk
150–300	Moderate risk
300–600	High risk
> 600	Very high risk

Statistical data analysis

The data collected was processed and converted to the chartboard with Microsoft Excel software.

RESULTS AND DISCUSSION

The concentration of heavy metals in sediment of researched area

The mean concentration and range of data in studied area were as follow: Zn: 33.66 (0.28–92.14); Cr: 12.88 (0.06–41.7), As: 4.65 (0.08–13.46), Pb: 11.93 (0.09–31,52), Cu: 4.97 (0.05–32.71); Cd: 0.08 (0.02–0.21); Hg: 0.07 (0.04–0.09) ppm. Therefore, the highest concentration was found in Zn, while the lowest was in Hg (Fig. 2). The concentration of heavy metal in sediment was unevenly distributed among samples. However, there is no evidence of pollution in the studied area because the concentration of all analyzed metals was under the permitted threshold according to The research results were compared with the National Technical Regulation on Sediment Quality of Vietnam [18] applied for saline and brackish water.

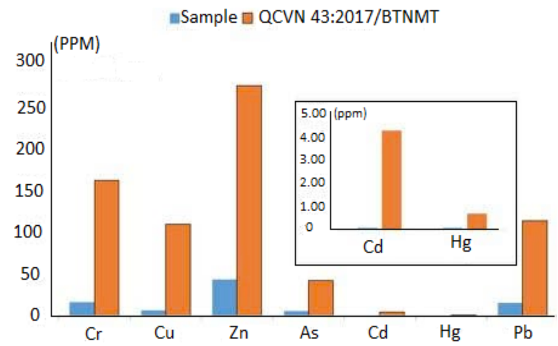

Figure 2. Concentration of metals in the lagoon sediment compared to the threshod

Variations in metal concentration among the studied samples can be attributed to differences in sampling locations, area characteristic, human activities, and difference in particle sizes. The results show that sediment samples dominated by sand with over 90% particle sizes collected along two sides of the lagoon are low in metal content. However, the samples in the lagoon's central area, which have higher proportion from 30% to 40% in silt and clay, possess higher metal concentration, suggesting that the waste sedimentation mainly progresses in the middle of the lagoon. Correlation analyses illustrate a strong positive correlation between heavy metal concentration and silt and clay content in the lagoon (Fig. 3), indicating that fine-grained sediments are more efficient at retaining metals, except for Hg.

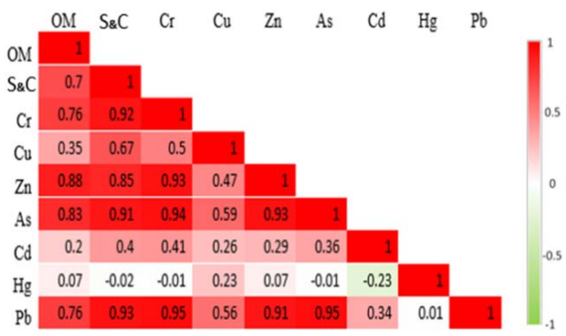

Figure 3. Correlation of heavy metals, organic matter and particle sizes

Assessment of heavy metal accumulation

The geo-accumulation index shown in Figure 4 suggests that sediments in the studied area are unpolluted according to the reference [12]. The order of the mean geo-accumulation index of seven heavy metals was Pb (-2.57) > Cd (-2.76) > Hg (-3.20) > As (-3.22) > Zn (-3.81) > Cr (-5.41) > Cu (-7.11). The Cd and Hg contamination levels varied insignificantly among sampling sites across the lagoon and had the lowest coefficients of variation. The water exchange with open sea heavily influences the possibility of metal accumulation in coastal lagoons. Especially semi-enclosed lagoons such as Tam Giang - Cau Hai, which contain a huge amount of freshwater input during rainy seasons, play a vital role in distributing and diluting contaminants. The rapid water exchange rate in the study area contributes to lower pollution levels than other areas with lower flow rate. This factor helps reduce pollution and accumulation of heavy metal in the lagoon.

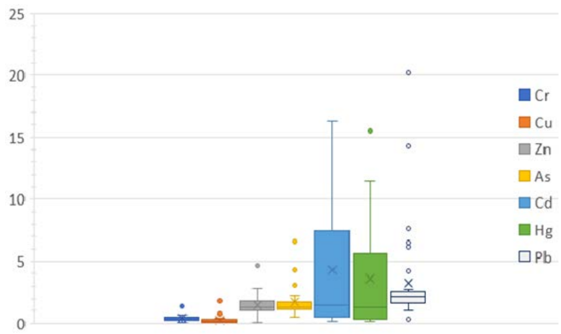

Figure 4. I_{geo} values of metals in the lagoon sediment

The Enrichment Factor was used to identify the source of contamination in the study area. The assessment based on calculated *EF* (Fig. 5) shows several metals, including Cu, Cr, and Zn in the lagoon sediment have the corresponding *EF* respectively 0.4, 0.21, and 1.44, which were lower than 1.5, proving that those metals are not enriched, and they have natural origin from geological processes. The remaining elements, including As (1.70), Cd (4.25), Hg (3.60), and Pb (3.20), have the *EF* higher than 1.5 indicating their anthropogenic origin. Cadmium was mainly released from industrial and recreational activities; Lead may be generated through industrial production or transportation [19, 20]. The degree of enrichment of each element varied from low (As) to medium (Cd, Hg, Pb). It was noticed that the EF values of Cd, Hg, and Pb in TT21 and TT22 sites are in high levels.

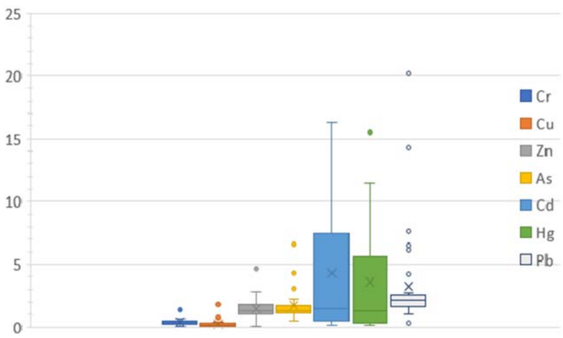

Figure 5. The *EF* values of metals in the lagoon

Assessment of ecological risk

The *PLI* was used to assess the lagoon's total pollution based on the concentration of the seven elements. The result show that the average *PLI* of the elements are less than 1, which indicates that the sediment environment is unpolluted. The figure for the *PLI* value of Pb was the highest; the opposite was seen in Cu. The RI values for heavy metals in sediment was shown in Table 5.

Table 5. RI and *PLI* values of metals in the lagoon sediment

Heavy metals	I_{geo}	*CF*	T_r	E_r (C_i/B_i)
Cr	-5.41	0.13	2	0.26
Cu	-7.11	0.08	5	0.40
Zn	-3.81	0.34	1	0.34
As	-3.22	0.35	10	3.5
Cd	-2.76	0.22	30	6.6
Hg	-3.20	0.17	40	6.8
Pb	-2.57	0.57	5	2.85
			RI	20.75
			Cdeg	1.86
			PLI	0.32

The RI at 20.75 is considered low risk, showing no threat to the local ecosystem. The order of ecological risk degree for each element was E_r (Hg) > E_r (Cd) > E_r (As) > E_r (Pb) > E_r (Cu) > E_r (Zn) > E_r (Cr). The concentration of Mercury is low at 0.07 mg kg^{-1}; however, due to the high toxicity (T^i_r = 40), Mercury poses the highest risk among studied elements.

Factors affecting heavy metal concentrations in sediments

The correlation analyses indicate a relatively strong correlation among six heavy metals. The most noticeable activity, aquaculture, which over 41,000 citizens rely on [8], may significantly contribute to the heavy metal pollution. Previous research has proven that discarding animal manure, feed supplementation, and fertilizer discharge in aquaculture increases heavy metal pollution [21–24]. Analysis of fish feed illustrates that the concentrations of Cu, Pb, Zn, and Cr, respectively, were 8.11, 0.97, 34.00, 0.31, and 3.22 mg kg^{-1} [25]. Over time, it may accumulate, increasing the heavy metal content in the sediment. In addition, anti-fouling paint and wastewater discharged by fishermen from boats are also significant sources of pollutants [21, 26]. In the past, aquaculture and fishery in the lagoon area used various chemicals (around 130 kg were released daily [27] for cleaning fishing gear and environmental improvement before culturing shrimp and fish. The overuse of chemicals will destroy the lagoon environment.

Furthermore, sediment characteristics are also significant factors contributing to the concentration and distribution of heavy metals. The sediments in the Tam Giang - Cau Hai lagoon mainly comprise mud, silt, sandy mud, and silty sand [28], creating good conditions for accumulating waste from the mainland and human activities in the lagoon. Smaller size particles have larger specific surface area and higher adsorbility, demonstrating higher capacity in accumulating heavy metals [29, 30]. This study also confirms that the middle lagoon areas with high silt and clay content (30–40%) accumulate higher levels of metals compared to the sediment areas on both sides of the lagoon with over 90% sand.

CONCLUSION

This study investigated the presence of seven heavy metals (As, Cu, Pb, Zn, Cd, Hg, and Cr) in the Tam Giang - Cau Hai lagoon sediments. The results of 35 sampling sites all fell within the limits in QCVN 43-2017/BTNMT applied for brackish and saline water sediments, with average concentration for metals as follows: Zn (33.66); Cr (12.88), As (4.65), Pb (11.93), Cu (4.97); Cd (0.08), and Hg (0.07) ppm. The geo-accumulation index (Igeo) indicates that the metals were at unpolluted levels. The enrichment factor (*EF*) showed that Cr Cu, and Zn were from natural processes (geological activities), while As, Cd, Pb, and Hg were released through anthropogenic activities. The pollution load index (*PLI*) indicates that all sampling sites had a low pollution level at a slow pace. The ecological risk in the studied area was low, with an *RI* value at 20.75. Although having lower concentrations, Hg posed the highest potential pollution risk and required special attention.

This is the result of the rainy season. To fully assess the ecological risk, monitoring the area during the dry season is required. Additionally, the study used background values of shale from global publications, so to accurately assess the pollution status in the lagoon, it is required to determine the area's background values.

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