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### ARTICLE

# The Heavy Metal Pollution Index in Seawater of the Coastal Aquaculture Zone in Quang Ninh, Vietnam

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### ABSTRACT

Heavy metal pollution in coastal waters is on the rise, presenting serious threats to both ecosystems and human health. Coastal aquaculture zones, such as those in Quang Ninh province, are especially vulnerable due to the accumulation of heavy metals from multiple sources, including industrial wastewater, land runoff, and maritime activities. This study applies the Heavy Metal Pollution Index (HPI) to assess pollution levels based on the concentrations of nine heavy metals (Cr, Mn, Fe, Cu, Zn, As, Cd, Hg, Pb) found in seawater samples collected from coastal aquaculture areas in Quang Ninh. According to the HPI method, values exceeding 100 indicate polluted water, serving as a benchmark for evaluating heavy metal contamination in the region. A total of 25 seawater samples were collected and analyzed; of these, 18 samples (72%) had HPI values above 100, signaling a concerning level of heavy metal pollution, while only 7 samples (28%) fell below the threshold, suggesting no significant contamination. These results underscore the urgent need for enhanced monitoring of water quality and stricter regulation of pollution sources to protect both environmental and public health.

*Keywords:* Heavy Metal Pollution; Seawater Quality; Aquaculture Zones; Heavy Metal Pollution Index (HPI); Quang Ninh Province

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### 1. Introduction

Heavy metal pollution in coastal marine environments has emerged as a critical environmental concern due to its negative impacts on ecosystems and human health. Coastal aquaculture areas, in particular, which are economically vital, are vulnerable to the accumulation of heavy metals from both natural and anthropogenic sources. Factors such as terrestrial runoff, industrial wastewater, and maritime traffic activities significantly exacerbate pollution levels. Numerous domestic and international studies have focused on assessing heavy metal pollution in coastal aquaculture zones, identifying emission sources, accumulation levels in aquatic organisms, and their impacts on human health through the food chain<sup>[1-4]</sup>. A study by Gupta et al. in India reported concerning accumulations of cadmium (Cd) and lead (Pb) in coastal aquaculture waters, adversely affecting fish growth and increasing the risk of contamination throughout the food chain<sup>[5]</sup>. Similarly, research conducted by Wang and Lu indicated that the accumulation of heavy metals in marine organisms could threaten ecosystem balance and pose risks to human health<sup>[6]</sup>. In addition, water and sediment quality play a crucial role in ensuring the quality of aquaculture products<sup>[7]</sup>. Therefore, collecting data on heavy metal concentrations in aquaculture zones is vital for ensuring seafood safety and supporting the sustainable growth of the aquaculture industry<sup>[8]</sup>.

Several recent studies have also demonstrated that heavy metals constitute major pollutants in marine environments, posing significant health risks to aquatic life<sup>[9]</sup>. They enter oceans through both natural processes like weathering and human activities such as industrial discharge and metal processing<sup>[10]</sup>. These contaminants can impair reproduction and reduce larval survival, leading to biodiversity loss<sup>[11]</sup>. Offshore waters are especially susceptible due to combined natural and anthropogenic pressures. Industrial waste releases metals like lead (Pb), cadmium (Cd), and mercury (Hg) into the sea<sup>[12]</sup>, while agricultural and urban runoff worsens the situation<sup>[13]</sup>. This pollution threatens fisheries and aquaculture, resulting in decreased fish stocks, food safety concerns, and financial losses for coastal communities<sup>[14]</sup>.

In Vietnam, several studies have been conducted to assess seawater quality in aquaculture areas, focusing specifically on physicochemical and nutritional parameters<sup>[15]</sup>, mi- 2. Site Description and Methods

crobiological contamination levels<sup>[16]</sup> and certain metals such as iron (Fe), manganese (Mn), and mercury  $(Hg)^{[17]}$ . Additionally, studies on coastal sediments in northern Vietnam have documented increased concentrations of certain metals such as copper (Cu), zinc (Zn), lead (Pb), arsenic (As), cadmium (Cd), and mercury (Hg)<sup>[18]</sup>. Nhu Da Le et al evaluated heavy metal pollution in coastal aquaculture areas of the Red River Delta, Vietnam<sup>[8]</sup>. Currently, although the Heavy Metal Pollution Index (HPI) has been applied in several studies, there are still limitations in providing a comprehensive assessment of pollution levels in aquaculture zones. Therefore, this study applies the HPI method to evaluate heavy metal pollution levels in cage aquaculture areas along the coast of Quang Ninh province. The results will provide important scientific data to support environmental management efforts and propose measures to mitigate pollution in the aquaculture sector.

Quang Ninh, one of the coastal provinces experiencing significant growth in aquaculture activities in Northern Vietnam, is particularly noted for its shrimp, fish, and mollusk farming. However, along with economic development, this region is also confronting environmental pollution risks arising from industrial activities, mineral extraction, and maritime transportation. Therefore, assessing heavy metal pollution levels in seawater within aquaculture zones is crucial to protecting the environment, ensuring product quality, and safeguarding public health.

In this study, the Heavy Metal Pollution Index (HPI) was applied to evaluate seawater pollution levels in coastal aquaculture areas of Quang Ninh province. Typically, an HPI value less than 100 suggests that the concentration levels of heavy metals are within acceptable limits, implying that the water is relatively unpolluted and safe for ecological and human use. Conversely, when the HPI value exceeds 100, it indicates a concerning level of heavy metal accumulation, classifying the water as polluted and potentially hazardous to both environmental and public health. This method not only synthesizes data on heavy metal concentrations but also provides an essential scientific basis to support environmental management efforts and propose more effective pollution mitigation measures.

#### 2.1. Site Description

Quang Ninh is a province located at the northeastern tip of Vietnam. The province's aquaculture zones extend across its coastal waters, primarily concentrated in the following localities: Ha Long City, Cam Pha City, Quang Yen Town, and the districts of Van Don, Co To, Hai Ha, Dam Ha, Tien Yen, and Mong Cai City<sup>[19]</sup>.

As of May 2023, the total potential marine aquaculture area in Quang Ninh province was 60,740 hectares. Among these, more than 40,000 hectares have been reviewed and identified by local authorities, with over 20,000 hectares located beyond the 6-nautical-mile boundary, classified as open sea areas. In 2023, the total area of estuarine and tidal flat zones, along with marine waters used for mollusk farming, reached 9,500 hectares, with a total output of 42,465.5 tons and an average yield of over 4.47 tons per hectare. The main cultured species included clams, cockles, estuarine oysters, and Pacific oysters, with cultivation methods primarily involving rafts, suspended cages, or tidal flat farming. These activities were mainly concentrated in Van Don, Dam Ha, Hai Ha, Quang Yen, and Mong Cai. Regarding marine fish farming, the total area in 2023 was 2,208 hectares, with approximately 15,000 fish cages distributed in localities such as Van Don, Dam Ha, Hai Ha, and Cam Pha. The main cultured species included grouper, seabass, and cobia, yielding a total production of 12,980.7 tons.

According to Decision No. 4209/QĐ-UBND dated December 15, 2016, approving the development plan for the fisheries sector of Quang Ninh province through 2020, with a vision to 2030, the province is projected to have 11,800 marine cage fish farming units by 2030, with an estimated production output of 7,420 tons (**Table 1**); The mollusk production is expected to reach 35,000 tons<sup>[19]</sup>.

<b>Table 1.</b> Cage Aduaculture Development Plan of Qualig Ninn Province by 2050. Source:	Table 1. Cage Aqua	culture Development	t Plan of Ouang	Ninh Province b	v 2030. Source: <sup>[20]</sup>
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No	Locality	Cage	Production (ton)
1	Van Don City	4500	2740
2	Tien Yen District	550	420
3	Hai Ha District	800	570
4	Dam Ha District	1500	1060
5	Mong Cai City	350	310
6	Ha Long City	700	450
7	Cam Pha City	3200	1750
8	Co To District	200	120
	Total	11,800	7,420

#### 2.2. Sample Collection

Water sampling in the intensive cage aquaculture areas along the coastal waters of Quang Ninh was conducted in May 2024, during low tide. The sampling process was carried out at multiple sites to ensure representativeness and accurately reflect water quality in the study area. Specifically, water samples were collected at 4 locations in Quang Yen Town, 4 in Cam Pha City, 5 in Van Don District, 4 in Tien Yen District, 4 in Dam Ha District, and 4 in Mong Cai City. A total of 25 sampling points were selected, covering key aquaculture areas throughout the province (**Figure 1**). The selection of sampling sites was based on geographical characteristics, aquaculture activities, and factors affecting seawater quality, with the aim of providing scientific data to support environmental assessment and management in

coastal zones. This study focused on analyzing water quality parameters, particularly the concentrations of the following heavy metals: chromium (Cr), manganese (Mn), iron (Fe), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), cadmium (Cd), mercury (Hg), lead (Pb), tin (Sn), antimony (Sb), cesium (Cs), barium (Ba), vanadium (V), selenium (Se), molybdenum (Mo), and silver (Ag).

Surface seawater samples were collected at a depth of approximately 50 cm to accurately reflect the environmental conditions at the sampling sites. The sampling process strictly adhered to technical guidelines based on the Vietnamese standard for seawater sampling (TCVN 5998:1995) and the international standard (ISO 5667-9:1992), ensuring the reliability and representativeness of the samples for heavy metal analysis. Immediately after collection, each water sample was treated on-site to maintain chemical stability and minimize compositional changes before laboratory analysis. Specifically, 2 mL of nitric acid (HNO<sub>3</sub>) in a 1:1 ratio was added to each liter of sample water. This acidification process helped maintain an appropriate pH level, prevent-

ing precipitation or adsorption of metal ions onto suspended particulate matter, thereby ensuring that the measured heavy metal concentrations accurately reflect the in situ conditions of the water at the time of sampling.



Figure 1. Map of the research area location

### 2.3. Sample Treatment and Analysis

Sample treatment: The treatment of seawater samples for heavy metal analysis followed the methodologies described by Brooks et al.<sup>[21]</sup>; ISO, 1986<sup>[22]</sup>; Satyanarayanan et al<sup>[23]</sup>. In the laboratory, the acidified water samples were allowed to reach room temperature and then filtered through Whatman cellulose acetate membranes (0.45 µm, Whatman, Merck) to collect the dissolved metal fraction. An acetic acid/ammonia buffer was used to adjust the pH of the solution to between 2.2 and 2.4. Subsequently, 5 mL of ammonium 1-pyrrolidine dithiocarbamate (APDC) at a concentration of 20 g/mL and 10.0 mL of methyl isobutyl ketone (MIBK) were added<sup>[21, 23]</sup>. The mixture was vigorously shaken for 2 minutes and then left to stand in a stoppered separating funnel for 1.5 hours at 30°C in the dark. The resulting inorganic phase was then used for heavy metal analysis. All glassware and polyethylene bottles were cleaned by soaking in a 10% v/v HNO<sub>3</sub> solution (Merck, Germany) for at least 24 hours, followed by rinsing with deionized water and drying in a sterile laminar flow cabinet prior to use<sup>[8]</sup>.

Sample analysis: The seawater samples described above were analyzed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS), specifically the Agilent 7700x model equipped with an integrated autosampler operating in helium (He) mode. For all determinations, the performance of the ICP-MS instrument was tested, and the instrument was continuously and properly calibrated using Tune solution (Agilent, code 5188-6564) at a concentration of 1 µg/L. The instrument sensitivity, oxide formation ratio, doubly charged element formation ratio, and background were checked daily. A standard solution (Agilent, code 8500-6940) and an internal standard solution (Agilent, code 5188-6525) were used. The standard curve ranged from 0 to 220  $\mu$ g/L for Zn, Fe; from 0 to 20 µg/L for Cu, Pb, As, Cd, Co, Mo, Sb, Ba, Mn, and Ni The correlation coefficients of the calibration curves for each element must be  $\geq 0.995$ . The percentage recovery of the spike sample should be 90 - 110%. The percent recovery of the continuous calibration standards (at the beginning and end of the analysis) should be between 85 and 115% (U.S. EPA, 1994)<sup>[24]</sup>.

#### 2.4. Statistical Analysis

In this study, statistical methods were applied to assess the distribution and relationships of heavy metal concentrations in seawater from coastal aquaculture areas in Quang Ninh province, Vietnam. Principal Component Analysis (PCA) was employed to identify the key factors influencing the variation of heavy metals while reducing data dimensionality, thereby aiding in the identification of major pollution sources. Cluster Analysis was used to group sampling sites with similar pollution characteristics, helping to pinpoint areas with high contamination risk. Additionally, Pearson correlation analysis was conducted to evaluate the relationships among heavy metals, providing insights into their common sources and geochemical characteristics. These statistical approaches contribute to a comprehensive understanding of the dynamics of heavy metal pollution in the study area.

All statistical analyses were performed using Origin 2024 software. This software facilitates graphing and the visual representation of PCA results, clustering, and correlation matrices, enabling effective evaluation of data trends. The use of Origin 2024 ensured both the accuracy and clarity of the data analysis, thereby enhancing the reliability of the research findings.

#### 2.5. Heavy Metal Pollution Index (HPI)

The Heavy Metal Pollution Index (HPI) evaluates the combined impact of individual heavy metals on water quality, serving as a ranking technique<sup>[25]</sup>. Each selected parameter (from 1 to *n*) is assigned a weight (Wi) or ranked in order to calculate the HPI using the following formula<sup>[26]</sup>:

$$HPI = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i}$$

In this formula,  $W_i$  and  $Q_i$  represent the weight and the sub-index of the *i*-th parameter, respectively.

#### Sub-index ( $Q_i$ )

The sub-index  $(Q_i)$  is calculated using the following equation sau<sup>[26]</sup>:

$$Q_i = \sum_{i=1}^{n} \frac{M_i - I_i}{S_i - I_i}.100$$

Where  $M_i$ ,  $I_i$ , and  $S_i$  represent the observed concentration, the ideal value, and the standard permissible value of the i-th heavy metal, respectively, according to QCVN 10:2023/BTNMT – the National Technical Regulation on Seawater Quality.

#### Calculation of the Weight (Wi)

Entropy is applied as a method to quantify the amount of information contained within a given indicator. The greater the amount of information an indicator holds, the more significant its role becomes in the decision-making process. Therefore, entropy is also used to assign weights to environmental indicators<sup>[27, 28]</sup>.

The main steps for determining entropy-based weights are as follows<sup>[27–31]</sup>.

#### Step 1: Normalize the original data.

Assuming there are m monitoring points and n evaluation parameters, the original data matrix X is represented as follows:

$$\mathbf{X} = \begin{bmatrix} \mathbf{x}_{11} & \mathbf{x}_{12} & \cdots & \mathbf{x}_{1n} \\ \mathbf{x}_{21} & \mathbf{x}_{22} & \cdots & \mathbf{x}_{2n} \\ & & \ddots & \\ \mathbf{x}_{m1} & \mathbf{x}_{m2} & \cdots & \mathbf{x}_{mn} \end{bmatrix}$$

After normalization, the matrix becomes  $P = (y_{ij})$  m x n (i = 1,2,...,m; j = 1,2,...,n) where  $P_{ij}$  is the normalized value of the i-th moniotring point for the j-th parameter and  $P_{ij}$  [0,1].

**Beneficial parameters** (e.g., DO, where higher values are better) are normalized using the following formula:

$$y_{ij} = \frac{x_{ij} - (x_{ij})_{min}}{(x_{ij})_{max} - (x_{ij})_{min}}$$

Adverse parameters (i.e, parameters where higher values indicate greater pollution) are normalized using the following formula:

$$y_{ij} = \frac{(x_{ij})_{max} - x_{ij}}{(x_{ij})_{max} - (x_{ij})_{min}}$$

#### Step 2: Entropy Determination

After transformation, the standardized matrix Y is obtained as follows:

$$\mathbf{Y} = \begin{bmatrix} y_{11} & y_{12} & \cdots & y_{1n} \\ y_{21} & y_{22} & \cdots & y_{2n} \\ & & \ddots & \\ y_{m1} & y_{m2} & \cdots & y_{mn} \end{bmatrix}$$

The formula to calculate the proportion of the *j*-th parameter in the *i*-th sample is:

$$P_{ij} = \frac{y_{ij}}{\sum_{i=1}^{m} y_{ij}}$$

The entropy parameter is represented by the formula:

$$e_{j} = -\frac{1}{\ln\left(m\right)} \sum_{i=1}^{m} P_{ij} . \ln\left(P_{ij}\right)$$

Step 3: The entropy value is calculated using the following formula:

$$W_{j} = \frac{1 - e_{j}}{\sum_{j=1}^{n} (1 - e_{j})}$$

The parameters applied in calculating the Heavy Metal Pollution Index (HPI) include: Cr, Mn, Fe, Cu, Zn, As, Cd, Hg, and Pb. These nine metals were selected based on the National Technical Regulation on Seawater Quality (QCVN 10:2023/BTNMT), which specifies threshold values for these substances due to their ecological toxicity and relevance to marine environmental monitoring in Vietnam.

*Assessment criteria:* An HPI value below 100 indicates that the water is not polluted, whereas a value above 100 signifies that the water is polluted by heavy metals<sup>[25, 32]</sup>.

### 3. Results and Discussion

### 3.1. Assessment of Heavy Metal Concentrations in Seawater of Coastal Aquaculture Areas in Quang Ninh Province

The analysis results of heavy metal concentrations in seawater from cage aquaculture areas along the coast of Quang Ninh revealed significant variations among different metals, reflecting the influence of various sources. Among them, zinc (Zn) exhibited the highest concentrations, ranging from 102.7 to 4365  $\mu$ g/L, with an average value of 844.4  $\mu$ g/L. This abnormally high concentration of Zn may be associated with oyster farming activities in the area, as oysters have the ability to accumulate and excrete zinc into the aquatic environment. Additionally, the decomposition of biomass from oyster shells may also contribute to the elevated levels of this metal (**Table 2**).

Iron (Fe) was also recorded at a considerable level, with an average concentration of 187.6  $\mu$ g/L (ranging from 61.2 to 400.4  $\mu$ g/L), indicating potential influence from natural mineral sources or industrial activities. Other metals such as manganese (Mn) with an average of 16.2  $\mu$ g/L (3.4–69.3  $\mu$ g/L), copper (Cu) at 37.8  $\mu$ g/L (9.1–126.7  $\mu$ g/L), and nickel (Ni) at 8.1  $\mu$ g/L (2.2–18.0  $\mu$ g/L) also showed variable distributions across the sampling sites (**Table 2**).

Notably, some toxic metals, despite their low average concentrations, still pose potential pollution risks. These include arsenic (As) at 2.1  $\mu$ g/L (0.8–3.9  $\mu$ g/L), cadmium (Cd) at 0.1  $\mu$ g/L (0–0.3  $\mu$ g/L), and mercury (Hg) at 1.0  $\mu$ g/L (0.2–3.9  $\mu$ g/L). Lead (Pb) showed an average of 4.8  $\mu$ g/L (1.5–16.3  $\mu$ g/L), suggesting possible influence from maritime traffic or coastal industrial activities. In addition, elements such as tin (Sn), antimony (Sb), cesium (Cs), barium (Ba), vanadium (V), selenium (Se), molybdenum (Mo), and silver (Ag) were also detected with considerable variation. Among them, Ba reached the highest level at 109.3  $\mu$ g/L, Se up to 11.7  $\mu$ g/L, and Ag as high as 26.9  $\mu$ g/L (**Table 2**).

The observed high standard deviations for certain metals - particularly Zn (836.9  $\mu$ g/L), Fe (78.1  $\mu$ g/L), and Cu (26.0  $\mu$ g/L) - indicate uneven spatial distribution across the study area. This may be related to differences in pollution sources or specific environmental characteristics of each sampling location. As shown in the boxplot, Zn exhibits the widest concentration range with extreme outliers, indicating significant spatial variability and possible point-source pollution. Cu and Fe also show relatively high variability. In contrast, most other metals display narrow ranges and limited variability, suggesting more uniform distribution (**Figure 2**).

Notably, zinc (Zn) exceeded the permissible limit set by QCVN 10:2023/BTNMT at all 25 out of 25 surveyed sites, with site VD3 recording an abnormally high value of 4364.5  $\mu$ g/L - 43.6 times higher than the regulatory standard. Copper (Cu) also surpassed the standard at 17 out of 25 locations, particularly at CP2 (126.7  $\mu$ g/L) and VD4 (77.5  $\mu$ g/L). Mercury (Hg) exceeded the limit at 19 out of 25 sites, mainly concentrated in the Hoang Tan area. Meanwhile, metals such as Cr, Mn, Fe, As, and Pb remained within permissible limits but still exhibited considerable variation among sampling sites. Notably, iron (Fe) reached its highest value at CP2 (400.4 µg/L), approaching the QCVN 10:2023/BTNMT threshold of 500  $\mu$ g/L. The uneven spatial distribution of heavy metals highlights a clear association with socio-economic activities in the region, emphasizing the need for stricter management and monitoring measures to safeguard water quality.

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Region	Sample Point	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Cd	Hg	Pb	Sn	Sb	Cs	Ba	v	Se	Мо	Ag
Hoang Tan	HT 1	2.2	12.5	237.2	1	7.8	35.7	345.4	1.9	0.1	3.9	1.5	0.4	1.5	0.4	19.9	1.4	11	0.9	10
HT	HT 2	4.2	19.7	196.6	1.3	11.5	63.3	659.2	2.4	0.2	3.1	7.1	0.8	1.9	0.5	34.8	1.4	4.4	1.1	19.1
	HT 3	0.6	3.4	118.5	0.6	4.8	18.7	1482	1.5	0.1	1.5	3.8	0.3	1.3	0.3	16.8	0.5	7.3	0.9	2.2
	HT 4	2.9	3.5	61.2	1.1	8.5	37.1	863.4	2	0.2	1.2	1.6	0.8	1.1	0.4	17.4	1.1	9	0.9	0.4
Tra Co	TC 1	1.3	9	101.3	0.4	3.7	12.6	1250.7	1.6	0	0.3	2.7	0.1	1.2	0.3	22.9	1	2.7	0.5	1.2
	TC 2	0	69.3	88.2	0.7	2.2	9.9	102.7	0.8	0	0.4	2.3	0.1	1.4	0.3	15.7	0.5	2.3	0.3	3.9
	TC 3	7.1	4.5	114.9	0.8	10.9	16.7	878.7	2.7	0.1	0.9	2	1	1.9	0.4	109.3	2.8	6.4	5.6	2.8
	TC 4	0	19.9	267.8	1.1	6.9	46.6	631.5	1.6	0.1	1	6.5	0.5	1.4	0.3	38.4	0.6	7.2	0.8	21.7
Dam Ha	DH 1	0.8	16.4	236.6	1.5	8.4	50.9	470.6	1.8	0	0.4	4.6	0.4	1.1	0.4	32.1	0.8	4.2	0.9	7.1
	DH 2	4.8	15.6	111.9	0.9	10	23.1	452	1.2	0.2	0.8	1.8	1.3	1.3	0.4	23.3	0.5	4.1	1	3.5
	DH 3	0.8	16.3	134.7	1.4	5.6	59.4	594.3	1.3	0.2	0.6	6.5	0.4	1.7	0.5	21.9	1.4	5.2	1	2.5
	DH 4	1.6	13.9	141.2	1.2	8.2	36	567.4	1.5	0.1	0.7	4	0.7	1.6	0.3	30.4	1.1	4.5	0.9	15
Tien Yen	TY 1	14.6	10.2	252.4	1.3	9.3	36.5	593.1	2.8	0	1.7	4.2	0.7	2.5	0.5	64.8	1.9	6.7	7.5	4.5
	TY 2	1.1	21.4	238.1	1.7	18	45.2	423.6	3.3	0.2	1.2	4.4	0.3	1.9	0.5	61.6	2.5	10.4	1.3	17.2
	TY 3	0.8	15	262.9	1.2	6.5	29.3	1078.2	1.9	0	0.7	4.7	0.5	1	0.3	32.2	0.4	6.6	1	8.1
	TY 4	11.4	17.2	168.8	1.4	16.4	37.1	1236.9	1.8	0.1	1.1	5.7	2.2	1.2	0.5	19.3	0.9	5.2	0.9	1.5
Van Don	VD 1	1.7	5.2	131.5	0.9	4.8	19.4	1456.2	1.1	0	0.6	3.6	0.5	1.5	0.5	21	0.8	9.7	1.1	2.9
	VD 2	0.6	5.2	174.2	1.1	2.7	27.1	289.4	1.5	0	0.6	1.9	0.4	0.7	0.3	21.4	1.3	8.2	1.1	5
	VD 3	2.8	8.7	189.2	1.3	11.9	43.2	4364.5	3.9	0.1	0.2	9	0.8	3.7	0.5	45.1	5.8	8.8	1.5	0.8
	VD 4	2.2	27.8	272.4	1.3	7.9	77.5	426.8	2.2	0	0.6	6.7	0.5	1.4	0.4	21.9	1.3	9.3	1.2	26.2
	VD 5	1.4	8	225	1.3	3.8	12.2	227.6	2.5	0.2	1.7	3.1	0.6	0.8	0.5	22.6	1.4	11.7	1.3	11.4
Cam Pha	CP 1	3	15.4	273.4	1.4	10.5	55.8	836.5	2.7	0.2	0.3	5.4	0.4	1.7	0.4	30.6	2	9.4	1.3	17.4
	CP 2	3.6	50.3	400.4	1.8	11.4	126.7	1299	3.5	0	0.4	16.3	0.4	1.8	0.4	33.9	1.5	11.3	1.3	26.9
	CP 3	1.1	9.8	142.3	0.9	6.5	9.1	177.6	2.6	0.3	0.6	9.2	0.5	1.7	0.6	17.7	2.7	11.7	1.1	4.7
	CP 4	1.3	6.3	150	0.8	4.8	16.2	401.6	1.9	0	1	2.2	0.3	1.5	0.6	24.2	0.6	7.6	1.2	10.4
QCVN 10:2023/BTNMT	100	500	500	-	-	20	100	20	5	0.5	50	-	-	-	-	-	-	-	-	

Table 2. Heavy Metal Concentrations (µg/L) in Seawater from Intensive Cage Aquaculture Areas in the Coastal Region of Quang Ninh Province.

Note: QCVN 10:2023/BTNMT - National Technical Regulation on Seawater Quality; (--) not specified / not regulated.



Figure 2. Heavy Metal Concentrations in Seawater from Cage Aquaculture Areas along the Coast of Quang Ninh Province

The results of the Spearman correlation analysis of heavy metal concentrations in seawater from cage aquaculture areas along the coast of Quang Ninh revealed several statistically significant relationships. Chromium (Cr) exhibited a strong positive correlation with nickel (Ni) (r = 0.68)and tin (Sn) (r = 0.67), suggesting a common origin possibly high correlation with vanadium (V) (r = 0.77), likely due to

related to industrial activities or natural weathering processes. Iron (Fe) and cobalt (Co) also showed a close correlation (r = 0.67), while copper (Cu) had a very strong correlation with Co (r = 0.79), reflecting the typical co-occurrence of these metals in aquatic environments. Arsenic (As) displayed a

shared sources from industrial pollution or similar geochemical behaviors. Meanwhile, molybdenum (Mo) and silver (Ag) showed a positive correlation (r = 0.74), potentially associated with electronic industry waste or agricultural activities. In contrast, zinc (Zn) showed negative correlations with mercury (Hg) (r = -0.31) and silver (Ag) (r = -0.36), possibly due to competition in adsorption processes or dif-

ferences in emission sources. Environmental factors also demonstrated significant influence: salinity and pH were highly correlated (r = 0.83), reflecting a close relationship between ionic balance and alkalinity. Additionally, selenium (Se) and molybdenum (Mo) showed strong correlations with pH (r = 0.50 and 0.77, respectively), indicating their sensitivity to the chemical conditions of the water (**Figure 3**).



Figure 3. Correlation Among Heavy Metal Parameters in Seawater from Cage Aquaculture Areas along the Coast of Quang Ninh Province.

The results show that the first two principal components (PC1 and PC2) explain 46.86% of the total variance in the dataset, with PC1 accounting for 29.91% and PC2 for 16.95%. PC1 has high loadings for As (0.35), Pb (0.27), Fe (0.26), Co (0.31), Ni (0.28), Cu (0.25), V (0.26), Sb (0.25), Cs (0.20), salinity (0.27), and pH (0.27). The strong association between these metals and salinity suggests a potential origin from the intrusion of polluted seawater or runoff from terrestrial sources. Arsenic (As) shows the highest loading (0.35), indicating a possible origin from mineral weathering processes or coal and metal ore mining activities in the region. Pb, Ni, and Cu may be related to industrial wastewater and maritime traffic, while Fe and Co could originate from mining operations or rock and soil erosion. PC2 shows high positive loadings for Mo (0.30), Cr (0.29), Sn (0.24), Ba (0.24), and Sb (0.21), and strong negative loadings for Cu (-0.31), Fe (-0.31), Mn (-0.31), Pb (-0.23), and Ag (-0.39). The separation of these metals along PC2 suggests the presence of at least two distinct sources of contamination (**Figure 4**):

*Industrial and transportation sources:* Cr, Sn, Ba, and Mo may originate from industrial wastewater, particularly from metallurgical processes, construction material production, and transportation activities. Sb is commonly used in alloys, anti-corrosion paints, and batteries, indicating potential contributions from industrial or domestic waste sources. *Natural sources and aquaculture activities:* The contrasting behavior of Cu, Fe, Mn, Pb, and Ag compared to the aforementioned metals suggests that these elements may derive from sediment deposition, dispersion from underlying soil layers, or interactions between the water column and the seabed.

The spatial distribution of sampling points on the PCA biplot (**Figure 4**) indicates that certain sites exhibit higher

levels of heavy metal pollution than others - particularly samples TC1, TC2, and TC3, which may be located near areas affected by industrial wastewater discharge and mining activities. Samples located near the center of the plot display more stable metal compositions, reflecting less pronounced environmental impacts.

Mercury (Hg) contributes minimally to both principal components, suggesting that its emission source is independent from those of the other metals.



Figure 4. Principal Component Analysis (PCA) of Heavy Metal Parameters in Seawater from Cage Aquaculture Areas along the Coast of Quang Ninh Province.

Based on the results of the cluster analysis, several groups of parameters were identified and optimized according to their relative proximity. Variables such as Cr, Sn, Ni, Ba, Mo, Zn, As, Sb, and V were grouped into the first cluster, indicating a high degree of similarity among them. This cluster includes metals commonly associated with pollution sources from coastal industrial and agricultural activities such as mining, metallurgy, the use of fertilizers and pesticides. Some of these metals may also be present in industrial feed used in aquaculture. The second cluster consists of Mn, Fe, Ag, Co, Cu, and Pb, reflecting a close relationship among metals that may originate from both natural sources (e.g., weathering, redox reactions in aquatic environments) and anthropogenic sources (e.g., domestic waste, marine traffic, aquaculture operations). The third cluster includes Cs and Se, along with environmental parameters such as salinity and pH. This grouping suggests that in addition to salinity and pH acting as baseline factors regulating the dissolution, adsorption, and transformation of metals, Cs and Se may be trace elements that are sensitive to environmental conditions or share similar distribution characteristics influenced by these factors. Additionally, Cd and Hg are found in separate, tics that may be linked to specific emission sources or unique

isolated positions, indicating distinct distribution characteris- transport mechanisms in the coastal aquaculture environment of Ouang Ninh province (Figure 5).



Figure 5. Cluster Analysis of Heavy Metal Parameters in Seawater from Cage Aquaculture Areas along the Coast of Quang Ninh Province.

### 3.2. Assessment of the Heavy Metal Pollution Index (HPI) in Seawater from **Coastal Aquaculture Areas in Quang Ninh** Province

Table 3 resents the calculated Heavy Metal Pollution Index (HPI) values for seawater samples collected from coastal aquaculture areas in Quang Ninh province, with a total of 25 water samples analyzed. The results show that 18 samples (accounting for 72%) had HPI values exceeding the threshold of 100 - commonly used as a critical limit to assess the risk of heavy metal pollution in water - thus categorized as "polluted." Only 7 samples (28%) recorded HPI values below this threshold and were classified as "non-polluted." The high proportion of samples exceeding the limit reflects a concerning level of heavy metal pollution in the study area, highlighting the urgent need for enhanced water quality monitoring and management in aquaculture operations (Figure 6).

Further analysis revealed that the metals contributing most significantly to the HPI values were mercury (Hg) and zinc (Zn). Notably, sample HT1 recorded an HPI (Hg) of 93.6 - a remarkably high value compared to typical background levels in aquatic environments. Meanwhile, sample VD3 had an HPI (Zn) of 523.7, the highest value among all samples. The exceptionally high concentration of Zn in this sample led to a total HPI of 556, far exceeding the threshold and identifying VD3 as the most severely polluted site in the study. Additionally, copper (Cu) was also detected at significant concentrations at several sampling points, indicating the widespread presence of heavy metals originating from both natural and anthropogenic sources (Figure 6).

When compared to international studies, the HPI values in Quang Ninh reflect a moderate to high level of heavy metal pollution. For example, in the Xuwen offshore area (China), HPI values in 2021 ranged from 28.46 to 65.79 and decreased to 7.24-10.16 in 2022<sup>[33]</sup>. Conversely, coastal areas of the Gulf of Mannar and Palk Strait (India) recorded

average HPI values of 233.2 and 136.4, significantly above the critical limit<sup>[34]</sup>. In the Al-Uqair coastline of Saudi Arabia, HPI values remained low, ranging from 9.55 to 13.04<sup>[35]</sup>. Based on comparative analysis with other coastal regions, the findings of this study indicate that the coastal aquaculture areas of Quang Ninh province are subject to a relatively high

level of environmental pressure from heavy metal contamination. The substantial proportion of sampling sites exceeding the established HPI threshold reflects a widespread and significant degree of pollution, thereby positioning this area among the more critically affected coastal zones in terms of heavy metal impact within the regional context.

<b>Fable 3.</b> Heavy Metal Pollution I	ndex (HPI)	) in Seawater from Coastal A	quaculture Areas in Q	Juang Ninh Province
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Sample Point	Cr	Mn	Fe	Cu	Zn	As	Cd	Hg	Pb	HPI	Assessment	
HT 1	0.3	0.4	2.4	17.9	41.4	0.5	0.3	93.6	0.3	157	Polluted	
HT 2	0.6	0.6	2.0	31.7	79.1	0.6	0.6	74.4	1.6	191	Polluted	
HT 3	0.1	0.1	1.2	9.4	177.8	0.4	0.3	36.0	0.8	226	Polluted	
HT 4	0.4	0.1	0.6	18.6	103.6	0.5	0.6	28.8	0.4	154	Polluted	
TC 1	0.2	0.3	1.0	6.3	150.1	0.4	0.0	7.2	0.6	166	Polluted	
TC 2	0.0	1.9	0.9	5.0	12.3	0.2	0.0	9.6	0.5	30	Not polluted	
TC 3	1.1	0.1	1.1	8.4	105.4	0.7	0.3	21.6	0.4	139	Polluted	
TC 4	0.0	0.6	2.7	23.3	75.8	0.4	0.3	24.0	1.4	128	Polluted	
DH 1	0.1	0.5	2.4	25.5	56.5	0.5	0.0	9.6	1.0	96	Not polluted	
DH 2	0.7	0.4	1.1	11.6	54.2	0.3	0.6	19.2	0.4	89	Not polluted	
DH 3	0.1	0.5	1.3	29.7	71.3	0.3	0.6	14.4	1.4	120	Polluted	
DH 4	0.2	0.4	1.4	18.0	68.1	0.4	0.3	16.8	0.9	107	Polluted	
TY 1	2.2	0.3	2.5	18.3	71.2	0.7	0.0	40.8	0.9	137	Polluted	
TY 2	0.2	0.6	2.4	22.6	50.8	0.8	0.6	28.8	1.0	108	Polluted	
TY 3	0.1	0.4	2.6	14.7	129.4	0.5	0.0	16.8	1.0	166	Polluted	
TY 4	1.7	0.5	1.7	18.6	148.4	0.5	0.3	26.4	1.3	199	Polluted	
VD 1	0.3	0.1	1.3	9.7	174.7	0.3	0.0	14.4	0.8	202	Polluted	
VD 2	0.1	0.1	1.7	13.6	34.7	0.4	0.0	14.4	0.4	65	Not polluted	
VD 3	0.4	0.2	1.9	21.6	523.7	1.0	0.3	4.8	2.0	556	Polluted	
VD 4	0.3	0.8	2.7	38.8	51.2	0.6	0.0	14.4	1.5	110	Polluted	
VD 5	0.2	0.2	2.3	6.1	27.3	0.6	0.6	40.8	0.7	79	Not polluted	
CP 1	0.5	0.4	2.7	27.9	100.4	0.7	0.6	7.2	1.2	142	Polluted	
CP 2	0.5	1.4	4.0	63.4	155.9	0.9	0.0	9.6	3.6	239	Polluted	
CP 3	0.2	0.3	1.4	4.6	21.3	0.7	1.0	14.4	2.0	46	Not polluted	
CP 4	0.2	0.2	1.5	8.1	48.2	0.5	0.0	24.0	0.5	83	Not polluted	

Note: The weights used in the HPI calculation for each metal are as follows: Cr (0.15), Mn (0.14), Fe (0.05), Cu (0.10), Zn (0.12), As (0.05), Cd (0.16), Hg (0.12), and Pb (0.11).



Figure 6. Heavy Metal Pollution Index (HPI) in Seawater from Coastal Aquaculture Areas in Quang Ninh Province.

Additionally, the analysis results indicate that the distribution of heavy metal pollution varies significantly across different areas, reflecting the influence of multiple emission sources, including both natural factors and socio-economic activities specific to each region. In particular, the abnormally high concentration of Zn in sample VD3 may be associated with large-scale oyster farming in the area. Oyster shells contain substantial amounts of Zn, and during the farming process, the decomposition of oyster shells after harvest or the accumulation of waste from cultured organisms can lead to elevated Zn levels in the water. This explains why oyster farming areas tend to exhibit higher Zn concentrations compared to other regions. Similarly, the high Hg concentration observed at Hoang Tan (HT1) may originate from the discharge of organic waste, industrial residues, or maritime traffic. Previous studies have also shown that Hg can stem from sources such as seafood processing, the use of chemical preservatives, or mining activities near coastal areas. Furthermore, coal and mineral mining activities in Quang Ninh contribute to increased heavy metal pollution, particularly Hg, Cu, and Pb. Quang Ninh is home to several large coal mines, and during the mining process, heavy metals can be transported by rainwater runoff into rivers and streams, eventually reaching the sea. This increases the risk of pollution in coastal zones located near inland water flows. Notably, mine tailings can be a significant source of Hg emissions, as mercury is often released into the environment during coal extraction and combustion. In addition to the impact of mining, the rapid development of coastal urban areas and cargo ports also contributes to marine water pollution. Domestic and industrial wastewater, as well as oil and grease from waterway transport, may contain metals such as Cu and Pb, increasing contamination levels in port areas and densely populated regions. In this study, Cu was also recorded at significant concentrations at several sampling sites, highlighting the widespread presence of this metal in coastal seawater.

This study provides a valuable snapshot of heavy metal contamination in seawater within coastal aquaculture zones of Quang Ninh; however, several limitations should be acknowledged. First, the entropy weighting method, while widely used for index construction, is inherently dependent on data variability and does not account for expert judgment or site-specific ecological sensitivity. This may influence the relative weight assigned to each metal in the HPI calculation. Second, the sample collection period was limited to May 2024, representing a single time point. As such, the findings may not fully capture seasonal variations in heavy metal concentrations driven by fluctuations in aquaculture activities, hydrodynamics, or weather conditions (e.g., rainfall, tidal influence). Third, although correlation analyses were conducted with salinity and pH, other potentially influential environmental parameters such as dissolved organic matter, current velocity, or sediment resuspension were not measured. The absence of these factors may limit the interpretation of heavy metal distribution patterns and potential sources. Future research should aim to incorporate multiseasonal monitoring and integrate a more comprehensive suite of environmental variables to improve the robustness and ecological relevance of heavy metal pollution assessments in coastal waters.

### 4. Conclusions

This study provides a comprehensive assessment of the current status of heavy metal contamination in seawater from cage aquaculture areas along the coast of Quang Ninh province. The analysis reveals that concentrations of certain metals - most notably zinc (Zn), copper (Cu), and mercury (Hg) - exceed the permissible limits established by national water quality standards at several monitoring sites. The spatial heterogeneity in heavy metal concentrations highlights the influence of multiple pollution sources, including natural inputs and anthropogenic activities such as industrial discharge, maritime transport, mineral exploitation, and aquaculture operations.

Although the sources of contamination vary, the cumulative presence of heavy metals significantly compromises water quality, potentially disrupting marine ecosystems and posing risks to human health. Multivariate statistical analyses, combined with the Heavy Metal Pollution Index (HPI), further demonstrate that certain regions - particularly those near unregulated industrial discharge zones and poorly managed aquaculture facilities - exhibit elevated pollution levels, with some sites classified as severely impacted.

These findings underscore the critical need to strengthen environmental management in coastal aquaculture areas. We recommend the implementation of systematic and long-term water quality monitoring programs, the enforcement of stricter national regulations on pollution sources, and the promotion of integrated coastal zone management. Such measures are essential not only to mitigate ecological and public health risks but also to ensure the sustainable development of the aquaculture industry in Quang Ninh province.

## **Author Contributions**

L.V.N.: Conceptualization, methodology, validation, data curation, writing—original draft preparation, writing—review and editing, supervision. N.T.M.L.: data curation, writing—original draft preparation, writing—review and editing. N.V.B.: writing—review and editing. L.X.S.: writing—review and editing; D.M.H.: writing— review and editing. N.T.T.H. writing—review and editing.

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### **Data Availability Statement**

Data will be made available on request.

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## **Conflicts of Interest**

All authors declare no conflict of interest.

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