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Research Article

Heavy metal concentration and human risk assessment of rock oyster in the coastal area of Ha Long City, Quang Ninh Province

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Abstract: The accumulation of heavy metals in rock oyster is a significant concern, particularly because rock oysters are a common food source for local people. This study evaluated the concentrations of 10 heavy metals in oysters and assessed the associated health risks from their consumption. The target hazard quotient (THQ) methodology, as established by the United States Environmental Protection Agency (US EPA), was utilized to determine the level of potential health risks. The findings revealed that among the heavy metals studied, zinc (Zn) and copper (Cu) were found to accumulate at significantly high concentrations, with values of 402.9008 mg/kg and 100.7288 mg/kg, respectively. Other heavy metal values, ranged from 0.1166 to 1.897 mg/kg, which is a comparatively low level. The levels of cadmium (Cd) and lead (Pb) were within the allowable limits outlined in Ministry of Health QCVN 8-2:2011/BYT. However, the THQ values for arsenic (As) exceed 1 for both medium and high consumption levels, signaling a potential health risk for consumers. The THQ values for other heavy metals remained below 1, indicating minimal health concerns from these elements individually. In addition, the total THQ for all heavy metals combined exceeded 1 across all consumption scenarios, underscoring a cumulative risk to consumer health.

Keywords: Heavy metals; Human risk assessment; Rock oyster; Heavy metal in rock oyster, Ha Long City.

1. Introduction

Oysters, as bivalve mollusks, are widely consumed in coastal regions and are integral to local diets. These organisms attach to underwater substrates where they live and grow, obtaining nourishment by filtering plankton from the surrounding water. This unique feeding mechanism, while essential for their survival, also renders them particularly susceptible to bioaccumulation, especially of heavy metals [1]. The ability of oysters to bioaccumulate arises from their filtration process, during which they inadvertently concentrate contaminants present in their aquatic environment, including heavy metals. This characteristic makes them both an indicator species for environmental monitoring and a potential source of dietary exposure to harmful substances for humans [2]. Environmental pollution, resulting from anthropogenic activities such as industrial discharge [3], agricultural runoff [4], and urban waste [5–10], significantly contributes to the elevated levels of heavy metals in aquatic ecosystems [11, 12]. Consequently, oysters residing in polluted waters often exhibit increased concentrations of toxic metals, posing health risks to consumers [2, 13, 14]. There have been some studies about heavy metals accumulation in rock oyster [1, 11, 15, 16]. This highlights the need for effective environmental management and regular monitoring to ensure the safety of oyster consumption.

There have been some researches of heavy metals in oyster in Vietnam [17–19]. However, the research about heavy metal in natural rock oyster was limited. Rock oysters are often harvested and consumed by the local people in Ha Long City, Quang Ninh Province. However, the potential accumulation of heavy metals in rock oysters, driven by environmental pollution, raises concerns about food safety and health risks. Therefore, it is essential to investigate the levels of heavy metals present in oysters from this region to safeguard public health. This study focused on two primary objectives. The first was to evaluate the concentrations of heavy metals accumulated in oysters from Ha Long City, Quang Ninh Province, providing a comprehensive understanding of contamination levels. The second objective was to assess the associated health risks to consumers using oysters as a food source. By employing methods, the target hazard quotient (THQ) approach, the study aimed to quantify the potential risks posed by heavy metal exposure through rock oyster consumption.

2. Materials and Methods

2.1. Samples collection and analysis

Five samples were collected from different locations along the Ha Long City coastal (Figure 1, Table 1). The sample were then stored in a cold condition then brought to laboratory. In this study, 10 heavy metals were evaluated that included: Arsenic (As), Selenium (Se), Chromium (Cr), Cobalt (Co), Nickel (Ni), Copper (Cu), Zinc (Zn), Cadmium (Cd), Barium (Ba) and Lead (Pb). The samples were collected in August 2024.

After cleaning the rock oyster shell, the tissue was dissected from the shell, rinsed thoroughly with distilled water, and subsequently dried. The dried tissue was finely ground, and 0.2-0.5 g of the powdered sample was transferred into a Teflon digestion vessel. A total of 8 mL of concentrated nitric acid (HNO_3) was then added, and the vessel was hermetically sealed to maintain a closed system. Microwave-assisted digestion was conducted by ramping the temperature to 180°C over 20 minutes, holding at that temperature for 15 minutes, and

Figure 1. Location of the sampling points.

subsequently cooling for 20 minutes to approximately 70°C before returning to room temperature. The resulting digest was diluted to a final volume of 50 mL with deionized water and filtered through a 0.45 µm membrane filter. The filtrate was then analyzed using an ICP-MS NexION 2000 instrument (PerkinElmer, USA).

Table 1. Coordinates of the sampling points.

Ord.	Code	Location	Longitude	Latitude
1	P1	Bridge to Tuan Chau	106.986384	20.945149
2	P2	Port to Reu Island	107.024434	20.946818
3	P3	Bai Chay old ferry port	107.060792	20.958578
4	P4	Port behind Halong market	107.082872	20.948116
5	P5	Port near Hongai beach	107.120696	20.936422

2.2. Statistical method

After analyzing samples, the resultant data were subjected to both basic and advanced statistical analysis using the R statistical software.

2.3. Human health risk assessment

In this study, estimated daily intakes (EDI) and target hazard quotient (THQ) were used to assess human health risk (HHR). The calculation of EDI is as followed:

$$EDI = \frac{C_w \times C_r}{B_w} \quad (1)$$

where C_w is the heavy metal concentration (mg/kg) in rock oyster tissue (wet weight), C_r is rock oyster consumption rate 17.86 g/day for average level and 35.7 g/day for high level [20–22]. B_w is body weight for adults was 60 kg.

In this study, target hazard quotient (THQ) equation of the US EPA was used to calculate the non-carcinogenic risk assessment of consuming oyster that contained heavy metals:

$$THQ = \frac{E_f \times E_d \times C_w \times C_r}{RfD \times B_w \times AET} \times 10^{-3} \quad (2)$$

where E_f is exposure frequency (365 d/year), E_d is the exposure duration for this study is 75 years (average life expectancy), C_w, C_r, and B_w are heavy metal concentration, rock oyster consumption rate, and body weight for adults as described in EDI equation. The average exposure time (AET) is 365 days/year × E_d for non-carcinogens, 10^{-3} is for unit conversion. The oral reference dose (RfD - mg/kg) values for this study were suggested by

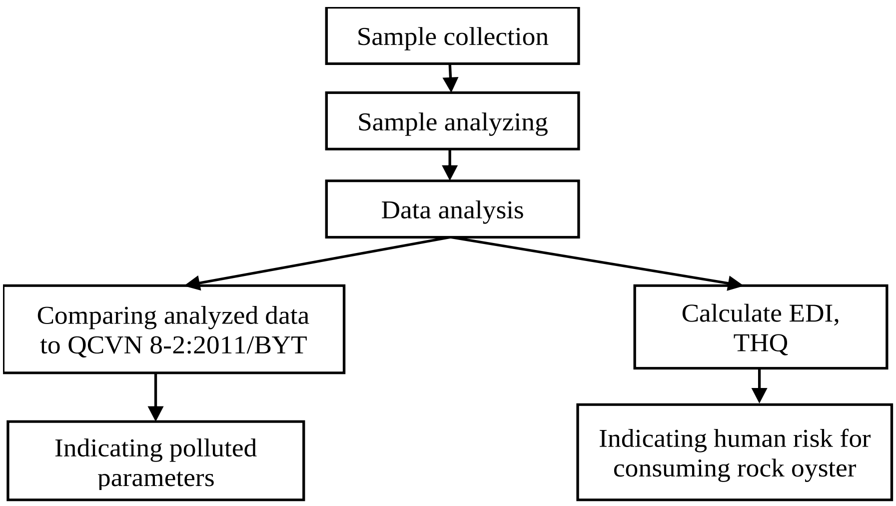

Figure 2. Research flowchart.

the EPA's Integrated Risk Information System (IRIS) [23]: As: 0.0003, Se: 0.005, Cr: 0.003, Ni: 0.02, Cu: 0.04, Zn: 0.3, Cd: 0.001, Ba: 0.2. Since RfD of Co and Pb are not listed in the database of the EPA's IRIS, Co and Pb values referenced from the studies of Finley et al. for Co: 0.03 [22, 24] and for Pb: 0.0035 [22]. If the THQ > 1 is high risk and THQ < 1 is low risk. The research flowchart was shown in the Figure 2.

3. Results and Discussion

3.1. Heavy metals concentration in rock oyster

The average concentration of heavy metals ranged from 0.1166 mg/kg (Co) to 402.9 mg/kg (Zn) (Table 2). The total concentration of the studied heavy metals was 507.78 mg/kg. Of which, Zn accounted for the highest proportion with 79% of the mass, followed by Cu with 20% of the mass. The remaining metals accounted for only 1%.

Zinc showed the highest concentrations in rocky oysters, with a mean of 402.9008 mg/kg and a wide range between 229.937 mg/kg and 559.522 mg/kg. Zn alone occupied 79% of heavy metal mass in rock oyster. Copper had the second highest concentration among the heavy metals in this study, with a mean value of 100.7288 mg/kg. The range of values was from 67.833 mg/kg to 186.533 mg/kg, with a notably high standard deviation of 48.662 mg/kg. This wide range suggests significant variability, likely influenced by localized sources. Arsenic mean concentration value was 1.897 mg/kg. The values ranged from a minimum of 1.174 mg/kg to a maximum of 2.791 mg/kg, indicating significant variability (standard deviation of 0.582 mg/kg). Selenium had a lower mean concentration of 0.406 mg/kg, with a narrow range from 0.366 to 0.489 mg/kg. The small standard deviation (0.0497 mg/kg) implies relatively stable levels in the environment. Chromium and cobalt, both trace elements, exhibited similar patterns of low concentrations. Chromium's mean concentration was 0.258 mg/kg, with a broader variability (standard deviation of 0.058 mg/kg) compared to cobalt, which had a mean of 0.1166 mg/kg and a standard deviation of 0.0196 mg/kg. Nickel had a mean value of 0.2812 mg/kg and a relatively higher variability (standard deviation of 0.0776 mg/kg). The range was from 0.183 to 0.391 mg/kg. Cadmium, a highly toxic heavy metal, had a mean concentration of 0.6758 mg/kg. Its values ranged from 0.626 mg/kg to 0.775 mg/kg, with a low standard deviation of 0.0656 mg/kg, indicating consistent levels across samples. Barium concentrations in rocky oysters are relatively low, with a mean of 0.2714 mg/kg. The range was from 0.143 mg/kg to 0.463 mg/kg, and the standard deviation of 0.1382 mg/kg indicates moderate variability. Lead exhibits low concentrations with a mean of 0.2414 mg/kg. The values ranged narrowly from 0.224 mg/kg to 0.258 mg/kg, and the standard deviation is minimal at 0.0158 mg/kg, suggesting consistent exposure levels. Lead contamination is often associated with industrial emissions, old paint residues, and urban runoff. Even at low concentrations, lead can pose significant ecological and health risks due to its toxicity and tendency to bio-accumulate.

Table 2. Heavy metals concentration in rock oyster (mg/kg).

Heavy metal	Mean	Minimum	Median	Maximum	SD	QCVN 8-2:2011/BYT
As	1.897	1.174	1.832	2.791	0.58205	-
Se	0.4064	0.366	0.387	0.489	0.04973	-
Cr	0.258	0.206	0.231	0.349	0.05803	-
Co	0.1166	0.084	0.122	0.135	0.01963	-
Ni	0.2812	0.183	0.272	0.391	0.07762	-
Cu	100.7288	67.833	84.365	186.533	48.66245	-
Zn	402.9008	229.937	413.825	559.522	117.0836	-
Cd	0.6758	0.626	0.639	0.775	0.06557	2
Ba	0.2714	0.143	0.215	0.463	0.1382	-
Pb	0.2414	0.224	0.24	0.258	0.01584	1.5

The concentrations of heavy metals in oysters differed across study locations, indicating that oysters' heavy metal accumulation is strongly influenced by local environmental conditions and pollution sources. Among the investigated sites, P3 exhibited the highest concentrations of Cu and Zn, while As was notably elevated at P1 (Figure 3). A comparison of the heavy metal concentrations in rock oysters with the QCVN 8-2/2011/BYT standards indicates that most measured parameters in the study area fall below the regulatted limits (Table 2).

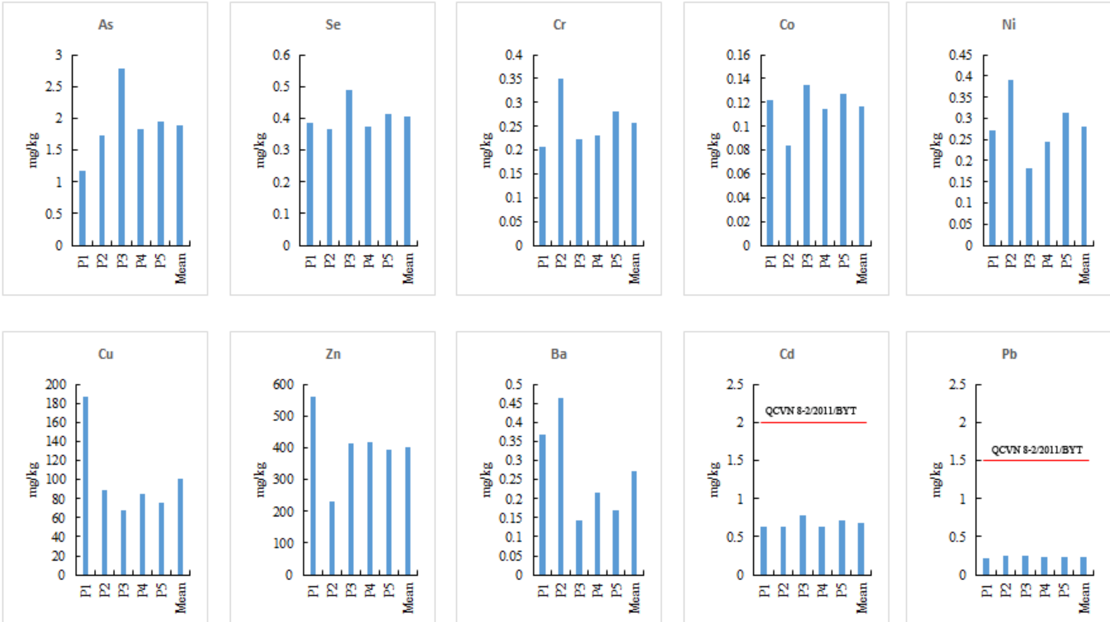

Figure 3. Concentration of heavy metals at sampling points in the study area.

In comparison with previous studies, the findings of this research demonstrated that rock oysters exhibited a significant capacity for bioaccumulating As, Cu, and Zn (Table 3). In these studies showed that oyster accumulated Zn with the highest concentration, 402.9 mg/kg (Halong Bay, Vietnam), 80-665 mg/kg (New South Wales, Australia), 100-2200 mg/kg (Sydney, Australia), and 774.44 mg/kg (Hab River Delta, Balochistan, Pakistan). These results aligned closely with those observed in other investigations focusing on the bioaccumulation of heavy metals in rocky oysters. It is important to note that the extent of metal accumulation is influenced by the concentration of heavy metals in the surrounding environment where the oysters reside. Nonetheless, the bioaccumulation patterns of As, Cu, and Zn in the oysters were clearly evident in the present study.

Table 3. The comparison heavy metals concentration in oyster in different countries.

Location	As	Se	Cr	Co	Ni	Cu	Zn	Cd	Ba	Pb	Ref.
Ha Long, Quang Ninh, Viet Nam	1.9	0.41	0.26	0.12	0.28	100.73	402.90	0.68	0.27	0.24	This study
Iranian coasts of the Oman Sea	-	-	-	-	6.45	208.86	-	-	-	4.55	[2]
New South Wales, Australia	1.2	-	-	-	-	3-48	80-665	0.2	-	0.8	[25]
Sydney, Australia	-	-	-	-	-	20-400	100-2200	2-10	-	50-1080	[11]

Location	As	Se	Cr	Co	Ni	Cu	Zn	Cd	Ba	Pb	Ref.
Hab River Delta, Balochistan, Pakistan	2.059	-	0.471	-	1.867	0.791	774.44	0.027	-	1.322	[26]

3.2. Human risk assessment

The THQ values were shown in the Table 4 and Figure 4 indicated that at medium CR level the THQ values for most metals remained below 1, indicating a low potential for health risks of consumers. Such as Se and Cr exhibited values of 0.011907 and 0.025798, respectively, signifying negligible risks (Table 4). However, As with a THQ value of 1.061678, which was higher tan the safety threshold of 1, indicating a significant health risk even at medium consumption levels.

At high CR level, the THQ values increased substantially. Arsenic (As) again posed a high risk with a THQ of 2.122167, further emphasizing its potential danger. Other metals like Se and Cr maintained low risk levels with THQ values of 0.023800 and 0.051567, respectively, suggesting that they remained relatively safe even with higher consumption. This pattern highlighted the variability in risk depending on both the metal type and consumption rate.

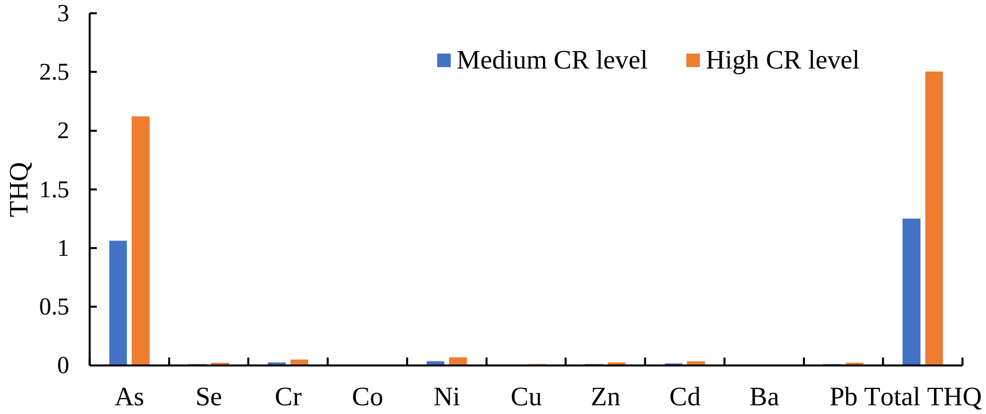

Figure 4. THQ values of heavy metals.

The data highlighted arsenic as a high-risk metal at both medium and high consumption levels, necessitating caution in dietary exposure. Given its widespread presence in the environment, especially in areas with contaminated water sources, efforts to reduce arsenic exposure are crucial for public health. On the other hand, metals such as Se, Co, and Ni consistently showed low risk, which indicated that, under the studied levels, these metals did not pose significant health hazards. This suggested that their intake levels, even at higher consumption rates, remain within safe limits for most individuals.

Table 4. THQ values for medium and high CR levels.

Heavy metals	Medium CR level	High CR level	Risk
As	1.0617	2.1222	High risk
Se	0.0119	0.0238	Low risk
Cr	0.0258	0.0516	Low risk
Co	0.0037	0.0073	Low risk
Ni	0.0353	0.0705	Low risk
Cu	0.0061	0.0122	Low risk
Zn	0.0127	0.0254	Low risk
Cd	0.0179	0.0357	Low risk
Ba	0.0017	0.0034	Low risk
Pb	0.0111	0.0221	Low risk
Total THQ	1.2518	2.5023	High risk

4. Conclusion

Among the studied heavy metals, zinc (Zn) and copper (Cu) were found to accumulate in rock oyster at notably high concentrations of 402.9008 mg/kg and 100.7288 mg/kg, respectively. In contrast, the concentrations of other heavy metals were relatively low, ranging from 0.1166 to 1.897 mg/kg. The concentrations of lead (Pb) and cadmium (Cd) were below the permissible thresholds outlined in QCVN 8-2:2011/BYT by the Ministry of Health of Viet Nam, which regulates heavy metal limits in bivalve mollusks.

The target hazard quotient (THQ) values for arsenic (As) indicated high-risk levels at both medium and high consumption rates, with values exceeding 1. Specifically, the THQ values for medium and high levels were 1.0617 and 2.1222, respectively. These findings suggested that regular oyster consumption at medium (17.86 g/day) and high (35.7 g/day) levels could pose long-term health risks to consumers. In contrast, the THQ values for other heavy metals remained below 1, signifying a low health risk. However, the cumulative total THQ for both medium and high consumption levels surpassed the critical threshold of 1, indicating a potential risk to consumer health.

The number of samples in the study was limited and collected from coastal areas. To provide a more comprehensive assessment of health risks, further research is required, including the analysis of rock oyster from various regions of Ha Long Bay. This would enable a more accurate evaluation of the potential health risks associated with rock oyster consumption.

Author contribution statement: Constructing research idea: H.T.D.; Select research methods: H.T.D.; Data processing: H.T.D.; Sample analysis: H.T.D.; Take samples: H.T.D.; Writing original draft preparation: H.T.D.; Writing review and editing: H.T.D.

Conflicts of Interest: The authors declare no conflict of interest.

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