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Penilaian Risiko Kesehatan Logam Berat pada Kerang Hijau (*Perna viridis*) dan Tiram (*Magallana bilineata*) yang Ditangkap di Luzon Utara, Filipina

Health Risk Assessment of Heavy Metals in Green Mussel (*Perna viridis*) and Oyster (*Magallana bilineata*) harvested in Northern Luzon, Philippines

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Abstrak

Bivalvia merupakan sumber makanan global yang penting karena harganya yang terjangkau, melimpah, dan nilai gizinya. Namun, urbanisasi dan pariwisata menimbulkan polutan seperti logam berat, khususnya kadmium (Cd), timbal (Pb), dan merkuri (Hg), yang meningkatkan masalah kesehatan masyarakat. Sampel bivalvia dikumpulkan menggunakan pengambilan sampel praktis selama musim kemarau dan musim hujan. Sampel air lingkungan juga dikumpulkan untuk penilaian perbandingan logam berat. Menggunakan Spektroskopi Serapan Atom Tungku Grafit, konsentrasi logam berat dalam jaringan bivalvia yang dapat dimakan dianalisis. Penilaian risiko kesehatan, termasuk Dosis Harian Rata-rata Seumur Hidup (LADD), Hazard Quotient (HQ), dan Risiko Karsinogenik (CR), dievaluasi mengikuti U.S. EPA dan FAO/WHO. Untuk kerang hijau, kadar Cd rata-rata 0,0071 mg/kg untuk kedua musim. Di sisi lain, kadar Cd tiram rata-rata 0,0298 mg/kg. Kedua bivalvia menunjukkan konsentrasi Pb dan Hg kurang dari batas deteksi metode masing-masing sebesar 0,0480 dan 0,0173 mg/kg. Konsentrasi ini berada dalam batas regulasi Cd, Pb, dan Hg yang ditetapkan oleh Peraturan UE 2023/915, yaitu masing-masing 1, 1,5, dan 0,5 mg/kg. LADD tidak melebihi Asupan Harian yang Dapat Ditoleransi Sementara, sementara nilai HQ dan CR berada di bawah nilai ambang batas satu, yang menunjukkan tidak ada risiko kesehatan terkait. Oleh karena itu, penelitian ini dapat digunakan untuk merumuskan langkah-langkah manajemen risiko dan nasihat kesehatan masyarakat. Lebih jauh, penelitian ini mengecualikan spesies bivalvia dan univalvia lain yang endemik di daerah pertumbuhan yang menyoroti perlunya pemantauan penilaian risiko untuk memastikan keamanan pangan.

Kata kunci: bivalvia, dosis harian rata-rata seumur hidup, rasio bahaya, risiko karsinogenik

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Abstract

Bivalves are a significant global food source due to their affordability, abundance, and nutritional value. However, urbanization and tourism introduce pollutants like heavy metals particularly cadmium (Cd), lead (Pb), and mercury (Hg), raising public health concerns. Bivalve samples are collected using convenience sampling during dry and wet seasons. Environmental water samples are also collected for comparative assessment of heavy metals. Using Graphite Furnace–Atomic Absorption Spectroscopy, heavy metal concentrations in edible tissues of bivalves were analyzed. Health risk assessments, including Lifetime Average Daily Dose (LADD), Hazard Quotient (HQ), and Carcinogenic Risk (CR), were evaluated following U.S. EPA and FAO/WHO. For green mussels the Cd levels averages to 0.0071 mg/kg for both seasons. On the other hand, Cd levels of oyster averages to 0.0298 mg/kg. Both bivalves show Pb and Hg concentrations less than the method detection limits of 0.0480 and 0.0173 mg/kg, respectively. These concentrations were within regulatory limits of Cd, Pb, and Hg set by EU Regulation 2023/915, i.e., 1, 1.5, and 0.5 mg/kg, respectively. LADD did not exceed the Provisional Tolerable Daily Intake, while HQ and CR values were below the threshold value of one, indicating no associated health risks. Hence the study can be used to formulate risk management measures and public health advisories. Furthermore, the study excluded other bivalve and univalve species endemic to the growing areas which highlights the need for risk assessment monitoring to ensure food safety.

Keywords: bivalves, lifetime average daily dose, hazard quotient, carcinogenic risk

1. Introduction

The Philippines boasts significant cultivation of two key fishery products, namely green mussel (*Perna viridis*) and oyster (*Magallana bilineata*). The Philippine Statistics Authority (PSA) – a government authority providing statistical records of agricultural supplies has verified that from 2013-2024, the country has consistently produced an average of 506,992.68 Metric Tons (MT) of these bivalves, with Northern Luzon (Region 1), Central Luzon (Region 3), CALABARZON (Region 4A), Western Visayas (Region 6), and Eastern Visayas (Region 8) emerging as the top producing areas. Bivalves play a crucial role as a primary food source nationwide due to their abundance, affordability, and nutritional benefits (Liu *et al*., 2018). However, their status as filter feeders makes them prone to accumulating toxic contaminants from their marine surroundings, such as heavy metals like Cadmium (Cd), Lead (Pb), and Mercury (Hg) (Liu *et al*., 2017).

These heavy metals, although naturally occurring in marine sediments, pose serious health risks when consumed in elevated concentrations. For instance, Cd is classified as a carcinogen by the World Health Organization (WHO) and can cause immediate kidney damage upon ingestion (Jafarzadeh *et al*., 2022). Similarly, Pb and Hg can lead to irreversible neurological damage, posing threats to both brain and kidney health (Agnihotri *et al*., 2018). Consequently, long exposure to these heavy metals through ingestion can lead to serious and irreversible health risks (Lin *et al*., 2024). Hence, conducting human health risk assessments regarding bivalve consumption is crucial for formulating preventive measures to safeguard public health and safety.

Various studies worldwide have focused on assessing the human health risks associated with bivalve consumption. For instance, Qin *et al*. (2021) investigated heavy metal concentrations in bivalve tissues from local markets in China's southern coastal region, concluding that there was no carcinogenic health risk associated with bivalve consumption in the area. However, they recommended continuous monitoring due to potential risks from prolonged and excessive consumption over a lifetime. In the Philippines, Montojo *et al*. (2021) examined green mussels and oysters from aquaculture farms around Manila Bay, indicating that these bivalves met safety standards based on local and international guidelines. Their health risk assessment suggested no risk associated with consumption, as the Estimated Daily Intake was below the Provisional Tolerable Daily Intake (PTDI), and hazard quotients were below one, according to U.S. EPA guidelines. Nonetheless, the study recommended further risk assessments on other fishery products or areas to ensure consumer safety.

With the rising concern in food safety and public health, we aimed to assess the levels of Cd, Pb, and Hg in green mussels and oysters collected from Bolinao, Pangasinan – one of the top producer of bivalves In Northern Luzon. This study area was limited to this place as part of the preliminary assessment of heavy metal contamination and health risks associated with it, from the top producing regions in the Philippines. In regards to this, some potential sources of heavy metals in Bolinao, Pangasinan include the increase of anthropogenic activities such as urbanization and aquaculture production. In 2014, researchers reported that the previous significant increase in milkfish culture had a negative impact on the seagrass ecosystems which increased nutrient loading and siltation (Tanaka *et al*., 2014). This change in composition can be linked to the accumulation of heavy metals in the sediments which were filtered by benthic organisms such as green mussels and oysters. Significant increase in tourism was also reported in the area by approximately 17% which can also be considered as possible source of heavy metal pollution (Austria, 2025).

Assessed levels of heavy metals were compared statistically to determine temporal variation and morphological correlation in heavy metal accumulation. Additionally we evaluated the levels of these heavy metals in water. Furthermore, we assessed the potential human health risks associated with the consumption of these bivalves. This assessment included assessment of lifetime averaging daily dose, hazard quotients, and carcinogenic risks for three age groups: children (1-10 years), adolescent (11-19 years), and adults (20 years and above). These age groups were selected to indicate varying vulnerabilities predominantly on the risk of the children to carry the health effects of long term heavy metal exposure. Risk assessment done from the consumption of bivalves in Bolinao, Pangasinan is crucial for formulating preventive measures in consumption, promote good aquaculture practices, and provide scientific evidences necessary for policy making aimed at protecting public health.

2. Material and Methods

Study Area

The samples are harvested in pre-determined growing areas of bivalves in Bolinao, Pangasinan (16.3846°, 119.9100°) which is a municipality under Northern Luzon (Region 1) as shown in Figure 1: mapped using Geographic Information System (QGIS version 1.8.0). Among the region stated as top-producers of bivalve in the country, Region 1 is selected as the focus of this study as the researchers plan to assess all the top producing regions in the country, thus limitations in the sample collection and area were present. Similarly, Pangasinan is identified as the sampling collection as this contributes to 84% and 100% production of green mussel and oyster, respectively. The heavy metals were assessed in the edible tissues of green mussels (*P. viridis*) and oysters (*M. bilineata*). Lastly, to compare the seasonal information of the commodities, the samples were collected twice during dry season (February-May) and wet season (June-August) of 2023 (PAGASA, 2025).

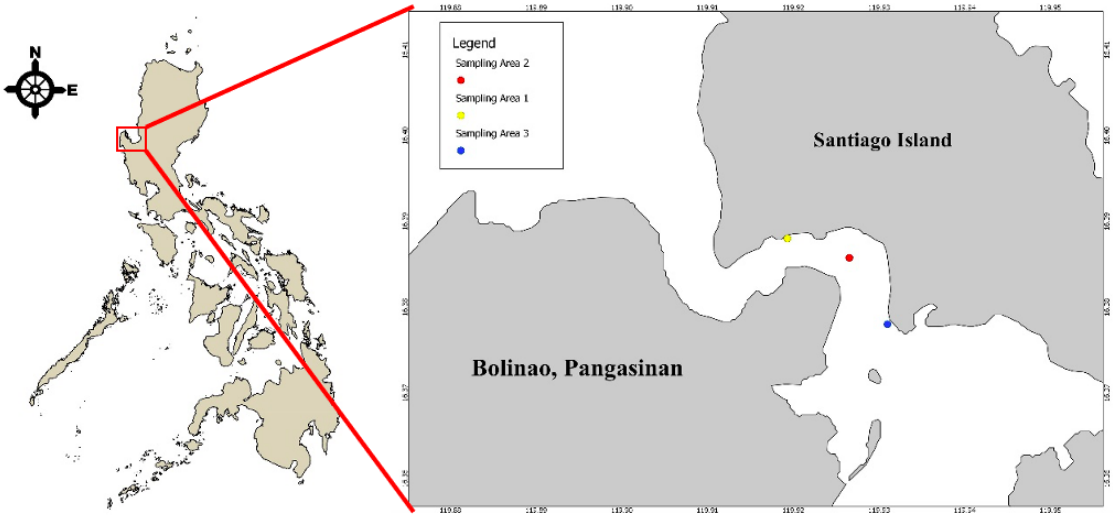

Figure 1. Simple map of the sampling areas of Bolinao, Pangasinan, Philippines.

Sample Collection of Bivalves and Environmental Water

The protocol for the collection, handling, and preservation of samples were followed from American Public Health Association (APHA, 2025, Method 1060). Convenience sampling was done in three bivalve farms and approximately one kilogram each of green mussel and oyster samples were collected. The samples were placed in a polyethylene (PE) plastic bag, labeled, and placed in a container with a maintaining temperature of less than 4ºC during transport to the laboratory for sample preparation and analysis. To ensure the repeatability of the sampling method, three (N = 3) replicates were collected per farm. Each sampling replicates contains ten (10) pieces of bivalves with similar lengths subject for morphometric and heavy metal analyses. Morphometric analyses were done every after sampling to prevent loss of sample integrity. Tissues were collected for the heavy metal analyses and stored in zip locks prior to analyses. These tissues were transported to the laboratory and analyzed within 24 hours to mitigate sample deterioration and decrease in analyte recovery.

Moreover, one liter of environmental water samples were also collected per sampling area. The water samples were collected in the surface of the rearing areas. Water samples for the analysis of Cd and Pb were placed in a PE bottle and preserved using 1 mL of concentrated nitric acid per liter of sample while samples for the analysis of Hg were placed in a borosilicate glass and preserved using 5 mL of concentrated hydrochloric acid was used for samples used for Hg determination. These samples were transported to the laboratory and analyzed within 48 hours to avoid poor analyte recovery.

Analysis of Heavy Metals

The sample preparation and digestion of Cd, Pb, and Hg in bivalve tissues were modified from AOAC Official Method (AOAC, 1976, Method 973). For the digestion of heavy metals in bivalve tissue, 0.3±0.0010 g was weighed in a digestion vessel. 7 mL of concentrated nitiric acid and 1 mL of 30% hydrogen peroxide was added to the tissue. The samples were then placed under microwave digestion system at 140±10 ºC for 50 minutes.

On the other hand, the digestion of Cd and Pb of water samples were followed from SMEWW 25th Edition (SMEWW, 2025, Method 3030) utilizing concentrated nitric acid and microwave digestion. digestion of Hg in water was adopted from Wahidah *et al*., (2019). Shimadzu AA-17000 was utilized for the detection of heavy metals. Analyses of Cd and Pb were conducted using Graphite Furnace Atomizer – Atomic Absorption Spectrophotometer (GF-AAS) while Cold

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Vapor Method was used for Hg. The evaluated method detection limit for the bivalve tissue are 0.0033, 0.048, and 0.0291 mg/kg for Cd, Pb, and Hg, respectively. While the method detection limit for water samples are 0.0020, 0.0004, and 0.0010 mg/L for Cd, Pb, and Hg, respectively. To ensure analytical replicability, three measurement were conducted per sample and limited the percent relative standard deviation to 7%. Meanwhile, spike samples were also used for the determination of method reproducibility as Certified Reference Material (CRM) was not available at the time of validation. Spike samples' percentage recoveries ranged from 84 – 92% for the bivalve tissue and 85 – 110% for water samples. Acceptable limits of heavy metal levels in bivalve tissues sourced from various regulatory bodies and institute were presented in Table 1.

Bivalve Morphometric Analyses

To assess the morphology of each bivalve, the selected shells with identical lengths were selected, cleaned with distilled water, and air-dried. Bivalve length (in cm) was identified as the longest distance from front to edge in lateral view. Meanwhile, bivalve weight (in g) was measured using an electrical balance with a 0.0100 g accuracy. Bivalve weight was recorded as the total weight of sample – the weight of the shell and organism. The morphometric data were used to assess the relationship of size to the levels of heavy metal detected. Furthermore, the correlation of morphometric data to the concentration of heavy metal was done using Pearson Correlation with probability level of rejecting the null hypothesis set at 5%.

Calculation of Associated Health Risk

a. Data collection

The data required for the calculation of the associated human health risk upon the consumption of bivalves harvest in the area were gathered from the population of Bolinao, Pangasinan. A stratified sampling due to their proximity to the bivalve rearing area was done to collect data for the calculation of health risks. Samples and data were collected according to the National Fisheries Research and Development Institute (NFRDI), Republic of Philippines, protocols. Samples and data were collected voluntarily from all participants.

A risk assessment interview were conducted to the target population to obtain their consumption data: approximate weight of bivalve per consumption in grams, and frequency of weekly consumption of the current year, age, weight, and years of residency. A data privacy consent form outlining exclusive usage of collected data for the study was included in the questionnaire. The consent was discussed with the participants prior to the collection of data. As the bivalve consumption rate and frequency differ per age group, the data were organized by three age groups namely: children (0-12 years old), adolescent (13-19 years old), and adults (20-65 years old).

b. Evaluation of associated health risk

Heath Risk Assessment (HRA) was evaluated using guidelines from the World Health Organization (WHO) and the US Environmental Protection Agency. In this study, the PTDI values of for Cd (0.83 μg/kg/day) (JECFA, 1999), Pb (3.57 μg/kg/day) (JECFA, 2010), and Hg (0.57 μg/kg/day) (JECFA, 2011) were used for adults with 70 kg body weight. All data gathered were calculated using the decision limit of 99^{th} percentile rank score, which also refers to the top 1% of the data calculated.

c. Lifetime average daily dose

Lifetime Average Daily Dose (LADD) is a parameter for the determination of lifetime dose of heavy metals from the consumption of foodstuff. Calculation of LADD (Benitez *et al*., 2021) is shown in Equation 1 (Eq 1.), where C is the concentration of metal in mg/kg, IR is the ingestion rate (kg consumed bivalve/day), EF is the Exposure Frequency which shows the days of frequency of consumption over the year

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consumed (e.g., a person who consumes bivalves seven (7) times a week has an EF of 365), ED is the Exposure Duration which is the period of duration in years wherein a consumer was exposed to carcinogenic risk which is similar to the duration of a consumer to the area, BW is the current body weight of the participant in kilograms, and AT is the average time for the healthy life expectancy which is 70 years old derived from the Projected Life Expectancy at Birth calculated for 2020-2025 from PSA (PSA, 2025) multiplied by 365 days/year.

$$LADD\left(\frac{\frac{mg}{kg}}{day}\right) = \frac{C\left(\frac{mg}{kg}\right) x\ IR\left(\frac{kg}{day}\right) x\ EF\left(\frac{day}{year}\right) x\ ED(years)}{BW\ (kg)\ x\ AT\ (day)} \quad \text{(Eq 1.)}$$

d. Hazard quotient

The Hazard Quotient (HQ) was calculated using an equation derived from Benitez *et al*. (2021) as presented in Equation 2 (Eq 2.). The HQ was used to determine the ratio of the population's daily intake to the reference doses by FAO/WHO. RfD is the reference dose in mg/kg of the studied metal, which was referenced from the U.S. EPA updated in 2010 for Cd, Pb, and Hg (US EPA, 2024). If the HQ of the heavy metal is less than one, the chemical exposure is less than the benchmark which exhibits low chance of adverse effect on consumption. On the contrary, the sampling population in which data exceeded the HQ benchmark of one concludes possible adverse effects in the consumption of the bivalves. This method is under the assumption that the intake dose is equal to the absorbed dose.

$$HQ = \frac{LADD\ \left(\frac{mg}{kg}/day\right)}{RfD\ \left(\frac{mg}{kg}/day\right)} \quad \text{(Eq 2.)}$$

e. Carcinogenic risk

The Carcinogenic Risk (CR) is a valuation tool used to quantify the risk associated with exposure to metals that may cause cancer. The carcinogenic risk was calculated using Equation 3 (Eq 3.), where CSF is the Cancer Slope Factor established by the California Office of Environmental Health Hazard Assessment for Cd, i.e., 15 mg/kg/day (OEHHA, 2025). CR of less than 10^{-6} is considered negligible for carcinogenic risk while CR of less than 10^{-4} or greater than 1 in ten thousand is considered as benchmark for gathering additional information or relaxing conservative assumptions that may be used in the screening analysis.

$$CR\left(\frac{\frac{mg}{kg}}{day}\right) = LADD\ \left(\frac{mg}{kg}/day\right) x\ CSF\left(\frac{mg}{kg}/day\right) \quad \text{(Eq 3.)}$$

Statistical Analysis

Statistical difference in the variation of heavy metals concentration during dry and wet season was done using Independent Samples t-test with $p < 0.05$ using R (version 4.3.3).

3. Results and Discussions

Levels of Heavy Metals in Bivalves and Water

The concentrations of Cd, Pb, and Hg in green mussels, oysters, and environmental water samples are investigated during dry and wet seasons as presented in Table 1. Concentration of heavy metals in bivalves showed that Pb and Hg in the edible tissues of bivalves are below the method detection limit of 0.0480 and 0.0173 mg/kg, respectively which indicates values below the detectable quantity of the method. These low concentrations of Pb and Hg can be extracted from the characteristics of these heavy metals as toxic pollutants that cause death to the tissues of benthic organisms upon exposure to high concentrations (Zaynab *et al*., 2022). Hence, Pb and Hg tend to accumulate and detected in low concentrations only. On the other hand, Cd concentrations for oyster are observed to be higher than the levels of green mussels. While Cd is also toxic to organisms, it naturally exists as salt and organic Cd which is significantly soluble in water and is easily digested by the bivalve (Benitez *et al*., 2021).

This characteristic of Cd allows its accumulation in higher concentrations than Pb and Hg (Haeruddin *et al*., 2021). Moreover, Cd accumulation of oyster is higher than green mussel due to the differences in their physiological mechanism such as accumulation and excretion (Liu *et al*., 2019). These data are in good agreement with a published paper on the levels of heavy metals in collected bivalves from Manila Bay, Philippines' aquaculture farms (Perelonia *et al*., 2017). The paper also observed low concentrations of Pb and Hg in bivalves and oyster accumulates more Cd concentration than green mussels. Moreover, this study was validated by the published data on levels of heavy metals of bivalves collected from the aquaculture farms of Manila Bay, which are determined to be below the regulatory limits (Montojo *et al*., 2021).

Table 1. Heavy metal concentrations (mean±SD) in bivalves and environmental water samples

Commodity	Levels of heavy metals (Bivalve: mg/kg, Water: mg/L)					
	Cd		Pb		Hg	
	Bivalve	Water	Bivalve	Water	Bivalve	Water
Green Mussel	0.0071±0.0014	<0.0020	<0.0480	<0.0004	<0.0173	<0.0010
Oyster	0.0298±0.0053	<0.0020	<0.0480	<0.0004	<0.0173	<0.0010
Limit for bivalve, mg/kg (EU 2023/915	1	--	1.5	--	0.5	--
Limit for bivalve, mg/kg (BFAR FAO 210 s2001)	0.5	--	0.5	--	0.5	--
Limit for water, mg/L (DAO 2016-08, Class SB)	--	0.003	--	0.01	--	0.001

Note: Double dash line indicate that data is not applicable

Additionally, the environmental water samples in the bivalve rearing area exhibited levels of Cd, Pb, and Hg detected below the method detection limits of 0.0020, 0.0004, and 0.0010 mg/L, respectively. Although Cd was detected in the bivalve tissue, it is undetectable in water samples. This phenomenon could be explained as heavy metals tend to naturally partition into sediments and is bioaccumulated in the tissues of filter feeders such as bivalves rather than remain in the water column. A study conducted in Haizhou Bay, China reported that heavy metals were detected in bivalves even when water concentrations are low (Zhang *et al*., 2023).

Table 2. Comparison of assessed levels of heavy metals from Green mussel and Oyster collected from various locations globally

Study area	Green mussel			Oyster			Study
	Cd	Pb	Hg	Cd	Pb	Hg	
Manila Bay, Philippines (aquaculture farms)	0.0269 – 0.0384	0.1781 – 0.2079	0.0179 – 0.0844	0.0804 – 0.2044	0.1203 – 0.1515	0.2249 – 0.5590	Perelonia *et al*., (2017)
Manila Bay, Philippines	< 0.50	< 0.50	< 0.50	< 0.50	< 0.50	< 0.50	Cambia *et al*., (2019)

Study area	Green mussel			Oyster			Study
	Cd	Pb	Hg	Cd	Pb	Hg	
(landing sites)							
Manila Bay, Philippines (aquaculture farms)	0.0028 – 0.0338	<0.00480	<0.0173	0.0455 – 0.3326	0.0042 – 0.0099	0.0173 – 0.0386	Montojo *et al*., (2021)
South China Sea (Daya Bay)	0.31	0.48	0.02	--	--	--	Yuan *et al*., (2020)
Java, Indonesia (Jakarta Bay)	0.195	1.27	--	1.92	5.76	--	Yulianto *et al*., (2019)
Bolinao, Pangasinan (Anda Bay)	0.0045 – 0.0085	< 0.0480	< 0.0173	0.0178 – 0.0417	< 0.0480	< 0.0173	Present study (2025)
Acceptable limit for HM in bivalve (mg/kg) (EU 2023/915)	1.00	1.50	0.50	1.00	1.50	0.50	

Note: Double dash line indicate that data is not applicable

Comparison of the studies presenting heavy metal contamination in bivalves locally and globally was shown in Table 2. Studies where bivalves are collected from growing areas with brackish water as a water source was focused on this section. It was noticeable that only the collected oyster from the study of (Yulianto *et al*., 2019) exceeded the acceptable heavy metal concentration in bivalve tissue with EU 2023/915 as basis. In the Oyster sample collected from Jakarta Bay, the levels of Cd and Pb which were 1.92 and 5.76 mg/kg, respectively exceeded the acceptable limit of 1.00 and 1.50 mg/kg, respectively. The authors suggested that this level of Cd and Pb from the collected oyster samples were sourced from the high number of anthropogenic activities in the area such as plywood industry and garment manufacturing. Therefore it was then concluded that these levels indicated strong correlation to pollution and that bivalves in the area were not safe for consumption. Albeit of this, the researches gathered did not exceed the regulatory limit from the EU 2023/915 and was often below the laboratory's detection limits. Although few researches published a significant contamination of these bivalves, constant monitoring of these pollutants in a growing area should be of utmost importance considering that there was a strong correlation of the increase in anthropogenic activities and heavy metal accumulation in bivalve tissues.

Over-all, measured levels of Cd, Pb, and Hg in both bivalves and water remain below the regulatory limits established by Commission Regulation (EU) 2023/915 (European Commission, 2023) and Bureau of Fisheries Aquatic Resources No. 210 s2001 (BFAR, 2001) for bivalves, and Department of Environment and Natural Resources Administrative Order 2016-08 (DENR, 2016) for Class SB environmental water samples. Class SB water is categorized as marine waters suitable for commercial propagation of shellfish. These findings affirm that the bivalves harvested from Bolinao, Pangasinan are safe for human consumption under current conditions. To safeguard food safety, continuous monitoring and implementation of guidelines is recommended and future studies should investigate potential sources and control of Cd contamination and monitoring in other bivalves and univalves endemic in the area.

Temporal Variation of Cd Concentration

On account of Pb and Hg concentrations being below the method detection limit, only Cd levels are acceptable for the statistical comparison of its temporal variation as presented in Table 3. The concentrations of Cd in green mussels and oysters exhibited significant temporal variation between dry and wet seasons. Cd concentration in green mussels is significantly higher in dry season than in wet season. Similarly, Cd concentration of oysters followed a similar trend to green mussel of higher values in dry season than in wet season.

Table 3. Temporal variation of Cd concentration in bivalves

Commodity	Season	Concentration (mean±SD)	p-value (α = 0.05)
Green Mussel	Dry	0.0091±0.0027	0.002
	Wet	0.0051±0.0017	
Oyster	Dry	0.0417±0.0113	0.006
	Wet	0.0178±0.0053	

Variation of Cd concentration during the seasonality of sample collection exhibited significant temporal effects. The seasonal influence of the bioaccumulation of Cd which is observed to be higher in dry season than in wet season may be attributed to rise in surface water temperature during dry season. During dry season, water dilution is reduced due to lower precipitation and water inflow thus increasing concentration of pollutants in the aquatic ecosystem. The high temperature of water enhances the activity of free Cd metal ion which becomes the most bioavailable form of Cd easily accumulated in bivalve tissue (Mubiana and Blust, 2007). Another cause for the significant increase in the levels of Cd were anthropogenic activities in the area primarily fishing and tourism. Activities such as aquaculture production was reported to be higher in dry season (i.e. 99 MT of green mussels, and 321 MT of oyster) in contrast to the wet season (i.e. 42 MT of green mussels, and 173 MT of oyster) (PSA, 2024). This increase in the aquaculture production can also lead to use of materials possibly coated with antifouling paints. Antifouling paints used in cages and structures submerged in water contains Cadmium and other metals that can leach off to the water (Satarug, 2024).

Similarly, tourism activities in 2024, recorded a 17% increase in the tourist arrivals relative to 2023 i.e. 635,935 tourists in 2023 and 744,430 tourists in 2024. Peak season of arrivals was reported to be during the dry season particularly in the month of April (Austria, 2025). These activities could elucidate the notable increase in Cd uptake during seasons which is characterized by elevated temperatures, as both a consequence of alterations in chemical reactions in the growing areas and the diffusion of bioavailable Cd ions into the bivalve. Comparatively, the temporal variation of Cd concentration to these bivalves is corroborated by some studies. For instance, Perelonia *et al*., (2017) reported levels of heavy metals in bivalves collected from aquaculture farms around Manila Bay, Philippines were higher in dry season as compared to the wet season. Similarly, (Milazzo *et al*., 2014) stated that the seasonality (dry and wet) carries a significant influence to the bioavailability and bioconcentration of metals in estuarine zone. Furthermore, despite displayed temporal variation, Cd levels in both bivalves remain acceptable for human consumption. In conclusion, the observed seasonal variation outlines the importance of periodic and continuous monitoring of these pollutants primarily during dry season.

Morphological Correlation of Cd Levels

Pearson correlation coefficients (r) of the correlation of Cd levels to the morphological parameters: length and weight of the bivalves are presented in Table 4. Both bivalve exhibited a negative correlation of morphological data with Cd levels. For the correlation of green mussel morphological data and Cd levels, moderate negative correlation in length and a strong negative correlation in weight are observed. Similarly, the correlation of the length and weight to the levels of Cd measured in oysters presented moderate negative correlation with length and weight.

Table 4. Correlation of bivalve length and weight to the Cd levels

Levels of Cd	**Morphological data**		**Pearson correlation coefficient (r)**	
	Length	**Weight**	**Length**	**Weight**
Present study (2023)				
Green mussel	7.77 – 9.04	37.00 – 46.07	-0.5067	-0.7089
Oyster	9.00 – 11.44	63.06 – 140.22	-0.4824	-0.1616

Observed negative correlations between Cd levels and morphological parameters of both bivalves indicated that the larger and heavier bivalves tend to accumulate lower Cd levels. These observations were also comparable to published studies Click or tap here to enter text.to which in similar manner exhibited negative correlation and concluded that this relationship may be influenced by the size and age of the bivalve. In 2019, Cambia *et al*., (2019) reported that the correlation coefficient (r) between green mussel and its length and weight were negatively and moderately correlated r = -0.4944 and r = -0.3305, while oyster's weight was negatively and significantly correlated with r = -0.8591. The negative and moderate correlation of their study was in agreement with the present analysis. Another study by (Sami *et al*., 2020) presented correlation analysis of some bivalves collected from Lake Timsah, Egypt in relation to their sizes and levels of heavy metals to which resulted significantly negative correlation to most of the heavy metals studied. These studies discussed that the inverse relationship of bivalve size and heavy metal accumulation might come from the age of the bivalve.

This phenomenon may be explained by the dilution effects in these organisms. Mature bivalves tend to pump less water hence the uptake of pollutants from the water and sediments were reduced. Smaller bivalves have the ability to consume more than the mature ones which lead to larger uptake and accumulation of heavy metals. With the increase in bivalve size, the surface area to volume ratio of exposure intensify which results to decrease in the relative spread of absorbed metal in bivalve tissue. Thus the smaller the bivalve size, the higher its pollutant intake and has more localized metal accumulation as compared to larger bivalves. Overall, uptake and excretion of heavy metals depend on the size of bivalves thus assuming that mature ones are safer for consumption (El-Moselhy and Yassie, 2005).

Health Risk Assessments based on LADD, HQ, and CR

a. Lifetime averaging daily dose

The LADD values for Cd, Pb, and Hg in green mussels and oysters are assessed for characterized age groups of their consumption during dry and wet season as summarized in Table 5. The LADD values are consistently highest in the adult group and their consumption rates appear to be higher during wet season while LADD values of children group exhibited lowest LADD values for both seasons (Figures 2a and 2b). None of the LADD values exceeded the PTDI values for Cd, Pb, and Hg based on the

Provisional Tolerable Weekly Intake (PTWI) from the joint FAO/WHO Expert Committee on Food Additives which are 0.83, 3.57, 0.57 μg/kg/day, respectively (JECFA, 2011).

LADD values for Cd, Pb, and Hg remain below the PTWI established by regulatory authorities which suggests that consumption of both bivalves during dry and wet seasons does not pose significant health risks. LADD values were notably higher during wet season compared to dry season this suggests that bivalve consumption of the sample population are dependent on the seasonality of harvest. As Philippines is a tropical country, the growth of a bivalve is faster during or after the rainy periods due to the influx of nutrients and other minerals into the ocean. This process boosts the production of phytoplankton which serves as the primary food source of bivalves, hence the increase in availability of these food stuff in the market (Sami *et al*., 2020). Consequently, adults were consistently exposed to highest LADD values due to their consumption rates thus suggests that monitoring programs should be targeted at this demographic area. Despite of Cd, Pb, and Hg levels being below the PTDI, accumulation of this heavy metal can lead to serious irreversible health risks thus younger age group can also be considered as one of the focus of this monitoring as their consumption duration and accumulation rate is higher than that of adults. Moreover, higher LADD values during wet season highlights the importance of continuous monitoring as Cd levels are higher during this season. Lastly, public health advisories should be issued to recommend safe consumption levels as ensure continued food safety and public health.

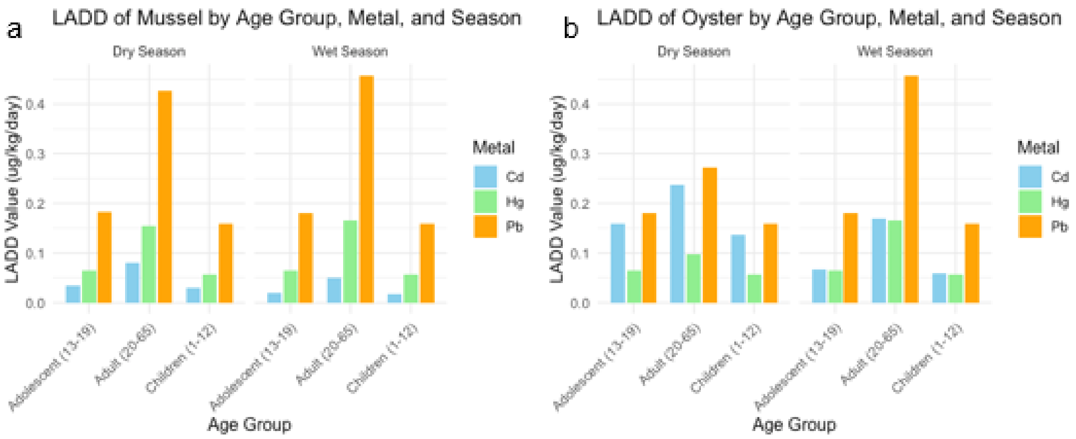

Figure 2. Lifetime Averaging Daily Dose (LADD) by Age Group, Metal, and Seasonality of collection of a) Green Mussel b) Oyster

b. Hazard quotient and carcinogenic risk

The summarized HQ values (mean and P99) of Cd, Pb, and Hg in green mussels and oysters are presented in Table 5 and illustrated in Figures 3a and 3b. Despite the HQ values of oysters are higher than that of green mussels, HQ values of all age groups did not exceed a value of 1.0 expressing no immediate concern in the consumption of these bivalves (US EPA, 2024).

Table 5. Estimated exposures to Cd, Pb, and Hg through consumption of bivalves from Northern, Pangasinan

Heavy metal	LADD (μg/kg/day)		HQ		CR (Cd)	
	Mean	**P99**	**Mean**	**P99**	**Mean**	**P99**
Green mussel						
Cd	0.1423	0.0751	0.0138	0.0086	0.0002	0.0011
Pb	0.1790	0.3960	0.0208	0.0129		

Heavy metal	**LADD (µg/kg/day)**		**HQ**		**CR (Cd)**	
	Mean	**P99**	**Mean**	**P99**	**Mean**	**P99**
Hg	0.4124	0.1427	0.0461	0.0286		
Oyster						
Cd	0.0498	0.1646	0.0498	0.1646		
Pb	0.0573	0.1895	0.0164	0.0541	0.0007	0.1198
Hg	0.0207	0.0683	0.0362	0.1198		

On the other hand, the mean and P99 carcinogenic risk used to quantify the associated risk with Cd exposure through consumption of bivalves is shown in Table 5 and illustrated in Figures 4a and 4b. In parallel to the HQ values, CR values of oysters are higher than that of green mussels. Furthermore, assessed CR values using the carcinogenicity factor of Cd (15 mg/kg/day) (OEHHA, 2025) did not exceed the threshold values (CR < 10^{-4}) (US EPA, 2024).

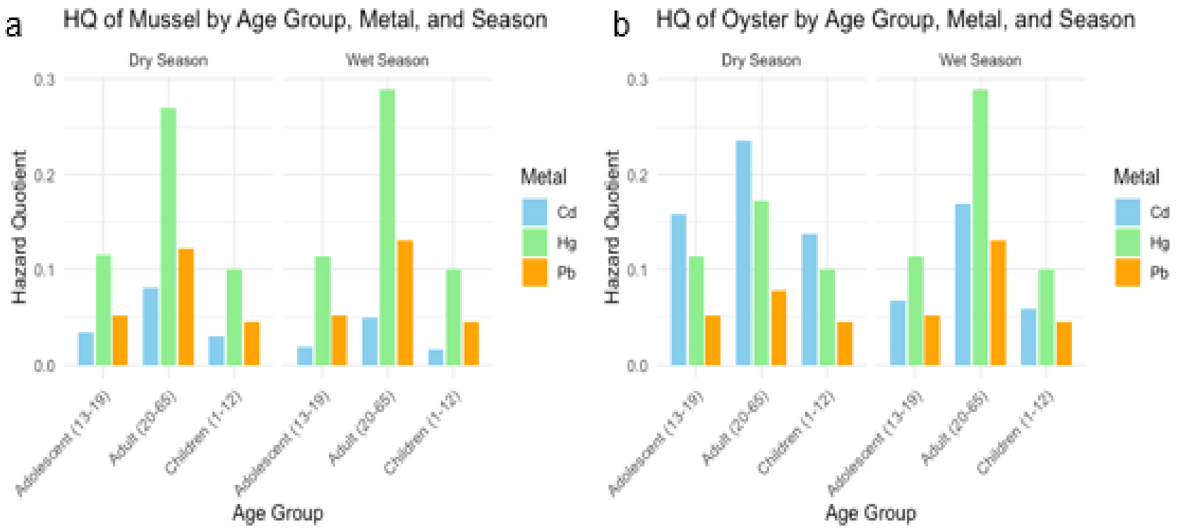

Figure 3. Hazard Quotient (HQ) by Age Group, Metal, and Seasonality of collection of a) Green Mussel b) Oyster.

The HQ values evaluated indicate that the consumption of both bivalves during dry and wet seasons poses minimal health risks to all age groups. However, significant variations were observed between seasons and age groups. Assessed HQ values for both values exhibited results which are less than one which implies that hazard exposure through consumption of these foodstuff is less than the benchmark of one (US EPA, 2024) and is unlikely to result in adverse effect during the time of assessment. Meanwhile, CR value for Cd revealed risk values of less than 10^{-4} inferring that excess lifetime cancer risk through long periods of exposure is less than one in a ten thousand indicating that additional information should be gathered (Benitez *et al.*, 2021). Despite of acceptable HQ values for the consumption of green mussels and oysters, risk management due to prolonged exposures should be implemented. Meanwhile, although CR values are below risk threshold, impact due to prolonged exposures necessitate further investigation.

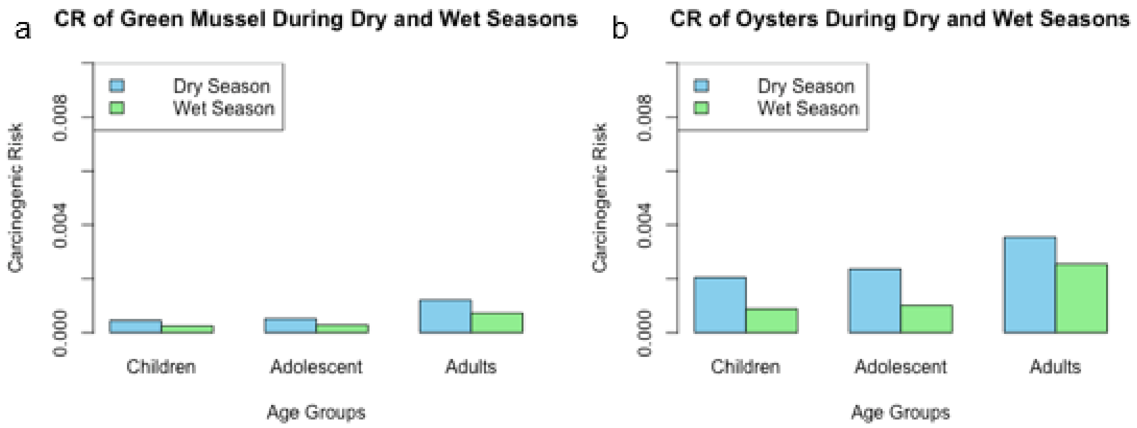

Figure 4. Carcinogenic Risk (CR) by Age Group, Metal, and Seasonality of collection of a) Green Mussel b) Oyster

4. Conclusion

The study assessed the levels of heavy metals particularly Cd, Pb, and Hg from edible tissues of green mussels and oysters harvested from growing areas of Bolinao, Pangasinan. Along with the evaluated levels, risk associated with its consumption were also assessed and evaluated with the risk thresholds from regulatory bodies. The results showed that the concentration of Cd in both bivalves remain below the limit and is safe for food consumption while Pb and Hg were observed to be below detection limit, a significant correlation was observed with the seasonality of sample collection and the Cd levels of both bivalves, a negative correlation between size and Cd levels, and there were no calculated health risk with the consumption of both bivalves. Despite of this, long term exposure and consumption of these implicates possibility of adverse health effects. Thus to ensure food safety and public health continuous monitoring of the levels of these contaminants in the studied area, implementation of environmental laws and policies related to wastewater discharge, and announcement of consumption advisories should be enforced. Moreover, other types of bivalves and univalves endemic in the area should be studied for possible heavy metal contamination and health risk associated.

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Author Contributions

All authors contributed to the final manuscript. Contributions of all authors: Athlene Cleah D. Benitez, Allysa Ross N. Almagro, Bernajocele Jalyn S. Baldoza, Karl Bryan S. Perelonia, Bryan E. Tanyag, Jerick Jann M. Quiambao, and Ulysses M. Montojo include conceptualization, methodology, format analysis, original draft preparation, writing-review and editing. All authors have read and approved the published version of the manuscript.

Conflict of Interest

The authors declare no financial or commercial conflict of interest.

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