



Bioaccumulation of heavy metals and health risk assessment of the mangrove clam, *Pegophysema philippiana* (Reeve, 1850), in Davao region, Philippines

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Abstract. Davao region is the most urbanized and industrialized part of Mindanao, Philippines. However, there are limited investigations of heavy metal contamination in the region's marine environment, particularly in bivalves. One of the major shellfish commodities in the area is *Pegophysema philippiana*, locally known as imbao. Thus, this study was conducted to determine the cadmium, lead, and mercury concentrations in the flesh of mangrove clams in selected areas of the region with substantial harvest and evaluate the health risk associated with consumption. Heavy metal concentrations were assessed using cold vapor atomic absorption spectroscopy for mercury and atomic absorption spectroscopy for cadmium and lead. The estimated daily human consumption of mangrove clams was determined through a social survey. The results revealed that Cd, Pb, and Hg concentrations in mangrove clams in Malita, Mati, and Bangaga were below the tolerable limits established for bivalves, but Cd and Pb in Sta. Cruz slightly exceeded the permissible levels. The local consumption rate of mangrove clam in four sites showed a significant difference ($p < 0.05$), with the highest value recorded in Baganga ($12.40 \text{ g day}^{-1} \text{ ind}^{-1}$) and the lowest in Sta. Cruz ($1.31 \text{ g day}^{-1} \text{ ind}^{-1}$). The health risk indices, such as estimated daily intake (EDI) values, were far below the provisional tolerable daily intakes (PTDI) established by authorized agencies, while the target hazard quotient (THQ) and total target hazard quotient (TTHQ) were less than one. Thus, no health risks were associated with consuming mangrove clams in the four sampling areas. However, further investigation and monitoring of the mangrove clams and other fishery commodities are encouraged to secure consumer's safety.

Key Words: bivalve, cadmium, heavy metal contamination, lead, mercury.

Introduction. Heavy-metal pollution remains a global environmental problem, since heavy metals continuously enter our environmental systems. Heavy metals are considered serious pollutants due to their toxicity, persistence, and inability to degrade in the environment (Gao et al 2021). The nonbiodegradability and bioaccumulative properties of heavy metals are well recognized, and, when taken up by organisms through bioaccumulation, they are known to be nonreversible (Su et al 2015).

Natural and anthropogenic processes contribute to heavy metals released into the environment. Among the main contamination sources of heavy metals in natural water resources are: overpopulation, industrialization, rapid urbanization, overuse of pesticides and herbicides, detergents and agricultural chemicals, sewage treatment plants, garbage dumps, mining operations, boating and recreational use of water bodies, petroleum wastewater, and crude oil spills (Peters et al 1997; Kesavan et al 2013; Dar et al 2018). The concern heightens when these metals persist in our environment and end up in trophic relationships where they are being accumulated within the animal tissues.

Marine organisms can accumulate heavy metals, especially bivalves, because they have high capacities for bioaccumulating heavy metals from their aquatic environments (Liu et al 2017). Bivalves are widely distributed benthic animals that mostly live in coastal and estuarine environments. They are recognized bio-indicators for evaluating the contamination and transfer of pollutants between media in the aquatic environment due to their strong capacity to bioaccumulate aquatic contaminants (Sarkar et al 2008). This is particularly due to their sedentary nature or immobility, filter-feeding activity, low metabolism, contact with sediments, wide distribution in all environments, ability to bioaccumulate pollutants, and high tolerance to chemical exposure due to a remarkably active immune system (Waykar & Deshmukh 2012; Zuykov et al 2013). Heavy metals exist in the marine environment, particularly in sediments, which means that bivalves accumulate a variety of metals from sediment and seawater during feeding (Qin et al 2021). The concentration of heavy metals in bivalves is an indication of heavy metal concentration in the sediment (El Nemr et al 2016).

Heavy metal pollution in the marine environment has become a rising concern due to the growing industrialization and urbanization that contribute significantly to the release of these heavy metals into the environment, particularly in the Davao Region. Davao Region is touted as Mindanao's center for trade and investment with a developing industry, and among the subsectors, mining and quarrying had the fastest growth in 2021 with 21.9%, followed by construction with 12.5%, and manufacturing with 8.9% (PSA 2022). However, there are only limited studies on heavy metal assessment in the marine environment of the region, especially on bivalves.

In the Davao Region, one of the major shellfish commodities is *Pegophysema philippiana* locally known as "imbao", previously named *Anodontia philippiana* (Lumogdang et al 2022). *P. philippiana* belongs to the family Lucinidae, which is distributed abundantly in the Indo-West Pacific region and is well known to be a delicious seafood delicacy in the Philippines. The mangrove clam is considered one of the important commodities in the region due to its flavor, size, and demand as a locally favorite shellfish.

It is always important to determine the bioaccumulation capacity and monitor the metals in the animal's tissue, especially in the edible ones caught within the coastal areas. The relationships between the concentrations of pollutants in the sediments and bivalve mollusks within the area can be used as effective tools to evaluate contamination levels and risks to the human population (Su et al 2015). The current concern nowadays lies in assessing the potential risks that heavy metals may have to human health. In the present study, the heavy metal contamination in the coastal area of Davao Region using the mangrove clam, *P. philippiana* and health risks associated with human consumption were evaluated. Three heavy metal pollutants, such as Cd (cadmium), Pb (lead), and Hg (mercury), were measured in the study because these are the most common heavy metals that cause human poisoning.

Material and Method

Description of the study sites. Davao Region, officially designated Region XI, is located in the southeast portion of Mindanao, Philippines. The region lies at 7.3042°N, 126.0893°E. The region faces the Pacific Ocean in the east and the Celebes Sea in the south, surrounding the Davao Gulf. The sampling areas were identified based on the presence of *P. philippiana* and areas where the majority of mangrove clam supply originates. The study sites were located in the three provinces of the Davao region, namely Sta. Cruz, Davao del Sur (6°49'16"N, 125°23'27"E), Malita, Davao Occidental (6°49'16"N, 125°35'23"E), Mati, Davao Oriental (6°95'22"N, 126°21'73"E), and Baganga, Davao Oriental (7°34'29"N, 126°33'4138"E) (Figure 1).

Mangrove clams in the region have an estimated price of 2.14 USD to 2.67 USD per kg, and they are one of the most exploited shellfish in Davao because of their taste, size, and claimed aphrodisiac properties (Primavera et al 2002; Bersaldo et al 2022). Mangrove clams are economically important for local gleaners' sustenance because most of them rely heavily on this commodity on a daily basis.

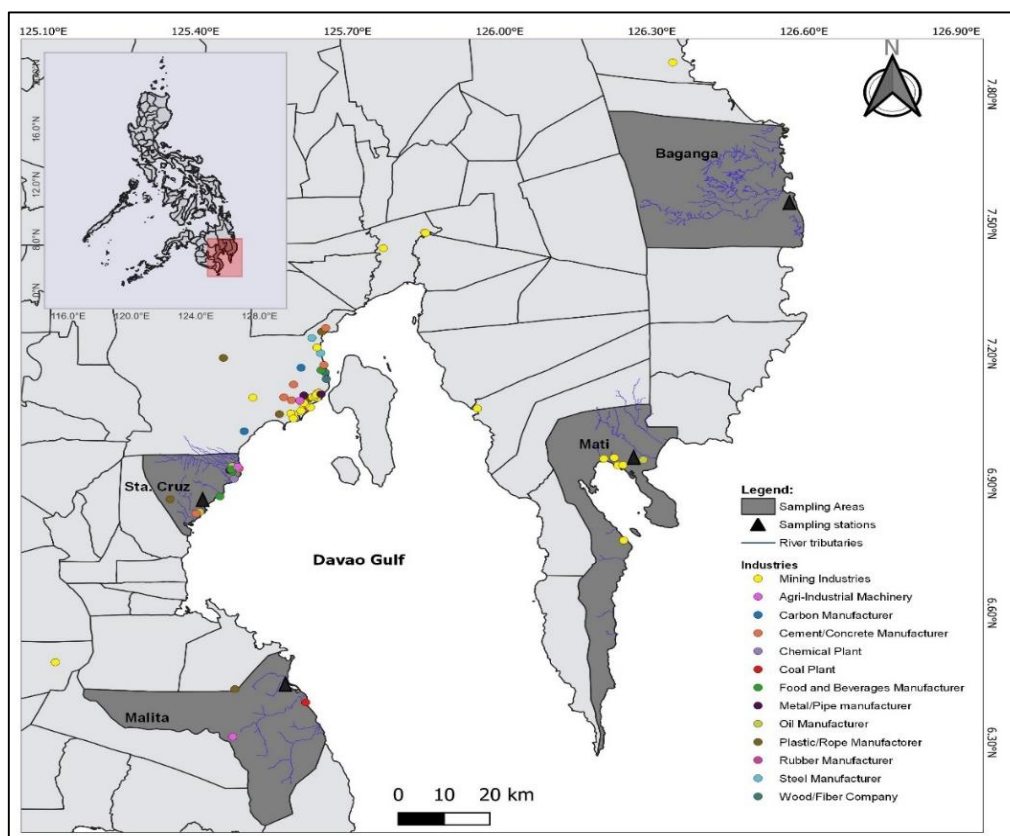


Figure 1. Map showing the four sampling areas in Davao Region (QGIS V3.26).

Based on geographic information displaying the different rivers in the Philippines (hydrosheds.org), it was observed that the Davao Region has a large number of river tributaries and is surrounded by a wide variety of industries, particularly mining and manufacturing industries, which are clear point sources for heavy metal pollution.

Sample collection and tissue preparation. In every sampling site, 60 individual samples of mangrove clam were collected. Only commercially sized individuals ranging from 5 to 7 cm in length were selected as shown in Figure 2, to avoid differences in heavy metal content because of size or reproductive stage (Saavedra et al 2004). The collection of individuals was randomly conducted by gleaning in the sampling area with the aid of local gleaners. The collected samples were immediately placed in an icebox and transferred to the laboratory for flesh extraction. In the laboratory, collected bivalve samples were properly cleaned with distilled water. Mangrove clam samples were dissected, and tissue was removed using a clean scalpel. The pooled samples at each station were prepared by mixing the dissected muscle tissue of the bivalves (Jahromi et al 2021). 500 g of homogenized muscle tissue were weighed using a digital scale and placed in a labeled plastic container and in a freezer until further analysis for the determination of Cd, Pb and Hg concentrations.

Sediment collection. The samples of sediment were acquired from the same point where bivalve samples were collected. Three random points were considered for the collection of sediment samples in every sampling station using a 2-foot-long PVC pipe with a minimum of 500 g of sediment per station. The collected sediment samples were secured in plastic containers and labeled with necessary data such as the date, time, and place where the sample was collected. Collected sediments in plastic containers were then placed in an ice box at 4°C for initial preservation.

Heavy metal analysis for bivalve tissue and sediments. The tissue and sediment samples were sent to Davao Analytical Laboratory, Inc., in Davao City for heavy metal

analysis. Three replicates for each sample were analyzed. Mangrove clam tissue and sediment samples were subjected to the flame AAS (atomic absorption spectrophotometry) method for detecting and quantifying heavy metals such as Pb and Cd, while cold vapor AAS was used to test the presence of Hg, following the standard procedures from AOAC (2012).



Figure 2. Mangrove clam, *Pegophysema philippiana* samples collected in the study.

Social survey. The survey questionnaire included information on the consumption of mangrove clams as well as a socio-demographic profile. The survey was conducted with the help of local residents in each of the sampling areas. The socio-demographic component of the survey contained inquiries regarding the respondents' demographics, including their age, sex, civil status, level of education, length of residency, occupation, family size, and number of gleaners in the household. The local consumption rate of mangrove clams in the sampling areas was assessed using the retrospective method adopted from the dietary assessment of FAO. A food frequency questionnaire (FFQ) was used to collect information on mangrove clams already consumed. The method greatly depends on the memory of the respondents and their ability to recall the amount of mangrove clam flesh consumed over a reference period of time. A random subsample of 100 households, representing each of the sampling areas, was interviewed (Zhuang et al 2013; FAO 2018).

The use of mean values in determining the local consumption rate was derived from a similar study conducted by Zhuang et al (2013). The average consumption per day was computed by dividing the total household consumption per day by the frequency of consumption (on a weekly, monthly, or yearly basis, depending on the rate of consumption per household). To get the mean ingestion rate of individuals per day, the average household consumption per meal per day was divided by the number of household members consuming the mangrove clam.

Human health risk assessment of bivalve consumption. The concentrations of heavy metals in the flesh of *P. philippiana* and the local consumption rate were used to compute the estimated daily intake of heavy metals (EDI), target hazard quotient (THQ), and total target hazard quotient (TTHQ) to assess the potential health hazards of heavy metal exposure. These indices were used in many studies for the evaluation of the ingestion of metals through the consumption of bivalves (Wang et al 2018; Jahromi et al 2021; Elvira et al 2021; Ding et al 2022). The equations considered are the established methods used to estimate the potential risk of toxic metals on human health.

The EDI takes into consideration the human population's daily exposure to amounts of heavy metals from consuming edible bivalve tissues (Yap et al 2016). The equation was adopted from the study of Jahromi et al (2021).

$$EDI=(C \times CR)/BW$$

A risk value indicator is provided by the target hazard quotient (THQ), which is used to assess non-carcinogenic health concerns resulting from exposure to hazardous substances (Yap et al 2016). The formula was adopted from the study of Wang et al (2018) and Elvira et al (2021).

$$THQ = (EF \times ED \times CR \times C) / (RfD \times BW \times AT) \times 10^{-3}$$

According to Liu et al (2017), a THQ value below 1 has no substantial risk, and a THQ above 1 poses a health risk to consumers.

By adding up all the individual metal THQ values, the total THQ (TTHQ) is calculated to determine the cumulative and overall health hazards (Chien et al 2002).

$$TTHQ = THQ(Hg) + THQ(Pb) + THQ(Cd)$$

If the value of TTHQ is below 1, there is no significant risk and if it is higher than 1, there is a chance that adverse effects may occur, at a probability tending to increase with the value. The greater the value of TTHQ, the greater the level of concern. The values of exposure factors used in the calculation of health indices and their respective units are shown in Table 1.

Table 1
Exposure factors, their corresponding unit and values gathered from literature

<i>Exposure factor</i>	<i>Symbol</i>	<i>Unit</i>	<i>Values</i>	<i>References</i>
Exposure frequency	EF	days year ⁻¹	365	Elvira et al (2021)
Exposure duration	ED	year	70	Elvira et al (2021)
Local consumption rate in mangrove clam/ Average daily dose	CR	g person ⁻¹ day ⁻¹		This study
Concentration of Hg, Pb, and Cd in bivalve	C	mg kg ⁻¹		This study
Oral reference dose				
Hg			0.0057	Ding et al (2022)
Pb	RfD	mg kg ⁻¹ day ⁻¹	0.0035	(USEPA, IRIS) adopted from Elvira et al (2021)
Cd			0.001	
Average body weight of Filipino	BW	Kg	65	Molina (2014)
Averaging time (EF x ED)	AT	Days	25.55	Wang et al (2018)

Statistical analysis. Means and standard deviations were used to express the heavy metal concentrations in the edible tissues of the mangrove clams and sediments. Microsoft Excel 2010 and IBM SPSS V.20 were utilized for the analyses. The difference in the means of the levels of Hg, Cd, and Pb in the tissue of *P. philippiana* and the rate of human consumption between the sample sites were tested using one-way ANOVA. The significance of differences between sampling sites was further determined using Tukey's test. Also, the data on metal concentrations were examined for homogeneity of variance. For further statistical analysis, the data were logarithmically transformed if the homogeneity requirements were not met. At $p < 0.05$, differences were deemed significant.

Results and Discussion

Levels of heavy metals in mangrove clam tissue. The levels of Cd, Pb, and Hg of mangrove clams in four selected sites in the Davao Region are presented in Table 2. The three metals detected in mangrove clams in the region in descending order were

Pb>Cd>Hg, with mean values of 0.341 mg kg⁻¹, 0.072 mg kg⁻¹, and 0.003 mg kg⁻¹, wet weight, respectively. All values of heavy metals across all stations were not significantly different (p>0.05) based on one-way ANOVA analysis.

Table 2

Heavy metal concentrations (mean ± SD) in tissue of mangrove clam (*Pegophysema philippiana*) collected from four selected sites in the Davao Region

Metal (mg kg ⁻¹)	Sta. Cruz	Malita	Mati	Baganga	Safe limit (mg kg ⁻¹)
Cd	0.210±0.072	0.017±0.006	0.033±0.006	0.027±0.006	0.05 ^a
Pb	0.553±0.093	nd	0.457±0.065	0.353±0.100	0.5 ^a
Hg	0.005±0.001	nd	0.005±0.000	nd	0.5 ^a

Note: ^a - safe limit of heavy metals based on FAO Legal Limits for Hazardous Substances in bivalves FAO, 1987).

Cd concentration. The concentrations of Cd in mangrove clams were found to be in the range of 0.017–0.21 mg kg⁻¹. The Cd content in mangrove clams from Malita, Mati, and Baganga was below the permissible limit (0.05 mg kg⁻¹), as set by the Fisheries Administrative Order (FAO 1983). However, the value of Cd in Sta. Cruz exceeded the safety level. There were multiple studies carried out on the bioaccumulation of Cd in marine bivalves in the Philippines (Elvira et al 2016; Montojo et al 2021; Benitez et al 2021). In Davao Region, only one study has been conducted on levels of Cd in bivalves using *Anadara maculosa* and *Antigona puerpera*, specifically in Pujada Bay, Davao Oriental (Tayone et al 2020). The study discovered similar results of Cd (0.03 mg kg⁻¹) as in this current finding in Mati, Davao Oriental. Moreover, the present findings were comparable to those of a study in Manila Bay on green mussels, *Perna viridis*, and oysters, *Crassostrea iredalei*, with Cd concentrations ranging from 0.0043 to 0.3326 mg kg⁻¹ (Montojo et al 2021). The Cd content found in this study was significantly lower compared to levels in scallop, *Bractechlamys vexillum*, in the Visayan Sea, which contained high concentrations ranging from 0.54 to 4.68 mg kg⁻¹ due to the increase in anthropogenic activities (Benitez et al 2021). Compared to other metals, Cd is relatively water-soluble, which can make it bioavailable to marine organisms. It is efficiently assimilated by marine bivalves due to their sedentary and filter-feeding behavior. Aside from natural sources, the occurrence of Cd in marine waters is due to industrial wastes and agricultural and mining activities that have caused an increase in its level (Ghani 2015). Plastic stabilizers, solar panels, motor oil, pigments, batteries, fungicides, polyvinyl chloride plastic, rubber, and textile manufacture are other sources of Cd (Khan et al 2017). It can also be found in phosphate fertilizers, detergents, and refined petroleum products (Mustapha & Lawal 2014). Elevated concentrations of Cd in mangrove clams in Sta. Cruz, Davao del Sur, compared to the rest of the sampling areas, can be associated with a higher state of industrialization. Among the four sites in Davao Region, Sta. Cruz is surrounded by extensive industries since it is bounded by Davao City and Digos City in the province of Davao del Sur. It should be emphasized that Cd is highly toxic to most marine and freshwater animals, even at low concentrations in natural waters (Lovett et al 1972).

Pb concentration. The highest concentration of Pb in mangrove clams was found in Sta. Cruz (0.55 mg kg⁻¹), followed by Mati (0.46 mg kg⁻¹), Baganga (0.35 mg kg⁻¹), and was not detected (nd) in Malita. Only Pb in Sta. Cruz was slightly above the safety threshold set by FAO, which is 0.5 mg kg⁻¹. The rest of the sampling areas were below the acceptable limit. Two studies have been conducted in the Davao Region on the accumulation of Pb in bivalves (Tayone et al 2020; Marquez & Avenido 2022). In comparison with the present findings, Pb levels in mangrove clams in the four sampling areas were lower than in the recent study of Marquez & Avenido (2022), with high concentrations in some bivalve species (*Anadara* sp., *Gafrarium* sp., and *Lioconcha* sp.) in Malita, Davao Occidental, with values ranging from 0.77 to 1.19 mg kg⁻¹. However, the levels of Pb in this study are higher compared to those in a study in Pujada Bay, Davao Oriental, using bivalve *A. maculosa* and *A. puerpera*, with an average 0.1 mg kg⁻¹ (Tayone et al 2020), and those in Cagayan

Valley in *Mytilus edulis*, with an average 0.1 mg kg^{-1} (Raju et al 2021). However, the levels were significantly lower compared to those in the study of Elvira et al (2016) in Butuan Bay in *Polymesoda erosa* (3.44 to 4.5 mg kg^{-1}). Pb is a heavy metal of environmental concern, just like Cd and Hg. It is the one of the abundant, widely used, and widely dispersed metals. It is mainly used in gasoline as tetraethyl lead, batteries, corrosive liquid containers, glassware, ammunition, paint, ceramics, and even cosmetics (US EPA 2015). Considering Pb is not biodegradable, the accumulation is insurmountable if Pb waste from industrial activities is continuously disposed of over time. Pb is a typical example of anthropogenic metal pollution. Anthropogenic activities are responsible for Pb pollution.

Hg concentration. The Hg contents detected in mangrove clams in four sites in Davao Region ranged from not detected to 0.005 mg kg^{-1} . These values were far below the allowable limit for Hg (0.5 mg kg^{-1}) imposed by the Fisheries Administrative Order (FAO). Similar findings were revealed in the study of Marquez & Avenido (2022), where bivalves had Hg ranging from 0.003 to 0.005 mg kg^{-1} in Malita, Davao Occidental. The Hg concentrations detected in mangrove clams were much lower than those found in bivalves from Butuan Bay, which were reported to have Hg values ranging from 1.67 to 6.72 mg kg^{-1} (Elvira et al 2016). Hg is also a persistent and toxic pollutant. One of the largest sources of Hg in marine waters is artisanal and small-scale mining. In Eastern Mindanao, it was reported that Hg contamination occurred in water and sediment in the 1900s in the Naboc River due to extensive small-scale mining and mineral processing affecting minor tributaries of the river flowing north to Agusan channel and west to Davao channel (Appleton et al 1999). Aside from mining, potential anthropogenic sources of Hg are dental amalgams, fluorescent lights, thermometers, electric switches, batteries, insecticides, disinfectants, rat poisons, and skin ointments (Perelonia et al 2017).

Levels of heavy metals in sediment. Several studies show significantly higher concentrations of heavy metals in sediments than in bivalve tissues (Sarkar et al 2008; Elvira et al 2016; Swaleh et al 2016; Diwa et al 2022). Hu & Cheng (2013) explained that sediments act as a pollution sink and show an indication of the severity and effects of heavy metal pollution in aquatic environments. However, the results of this study did not detect Cd, Pb, and Hg in sediments in the four sampling areas. This coincides with the findings of Lias (2013), where no significant correlation was determined between heavy metal concentrations in sediments and bivalves. This suggests that *P. philippiana* regulates the concentrations of heavy metals in its tissue, which may differ from the concentrations in the sediment to which it is exposed. Bivalves exhibit greater spatial sensitivity than sediments, making them the most effective method for detecting sources of heavy metal contamination (Szefer 1986). Bivalves are filter feeders and can bioconcentrate contaminants, which would normally be found at concentrations too low for detection in sediments (Phuong 2014). Variation in the metal content of sediments can be attributed to several factors, including metal deposition, particle sedimentation rates, particle size, and organic content (Ghabbour et al 2006; Chakraborty & Owens 2014).

Socio-demographic profile of the respondents. Most of the respondents were female (59%) and married (49%). The average ages of the respondents were 45, 43, 41, and 36 years old in Sta. Cruz, Mati, Malita, and Banganga, respectively. Almost half of the respondents (49%) have attained a high school education, while 42% have an elementary education, and only 9% have a college education. The households have an average of five members, and only one member is engaged in fishing or gleaning. Many of the respondents have lived near the coastal areas for more than three decades, an average of 39 years in Sta. Cruz, 33 years in Malita, 32 years in Mati, and 30 years in Baganga (Table 3).

Table 3

Socio-demographic profile of the respondents from Sta. Cruz, Malita, Mati and Baganga, Davao Region

Variable	Sta. Cruz		Malita		Mati		Baganga	
	%	Mean	%	Mean	%	Mean	%	Mean
Age		45		41		43		36
15-20			4		5		5	
21-40	33		45		41		74	
41-60	59		48		47		20	
≥61	8		3		7		1	
Sex								
Male	24		35		57		49	
Female	76		65		43		51	
Civil status								
Single	4				7		18	
Married	95		85		93		82	
Separated			11					
Widowed	1		4					
Educational attainment								
No formal education			1		1			
Elementary level	29		50		51		37	
Highschool level	59		45		37		53	
College level	12		4		11		10	
Duration of residency		39		33		32		30
Household size		5		4		5		5
Members involved in gleaning		1		1		1		1

Estimated daily consumption of mangrove clams. The consumption rates of mangrove clams were estimated by surveying 100 households in each of the sampling areas of Davao Region (Table 4). Mangrove clam was consumed by residents in corresponding coastal areas only one to two times per month. Among the four sampling stations, Baganga had the highest daily consumption of mangrove clams (12.4 g person⁻¹ day⁻¹), while Sta. Cruz had the lowest, at 1.31 g person⁻¹ day⁻¹. Significant differences were observed between the daily consumption rate across the stations ($p < 0.005$). As described by Bersaldo et al (2022), around 50 kg of mangrove clams are harvested daily in Baganga, and gleaning and fishing are the primary sources of income in the area. High consumption rates in Baganga compared to the other sampling areas can be attributed to the frequency of the gleaning of mangrove clams. As observed during the study, the residents of Baganga gleaned every day, while the residents in other sampling areas gleaned only once a week or twice a week. Furthermore, Tayone et al (2020) determined that the ingestion rate of mollusks (gastropods and bivalves) in Pujada Bay, Davao Oriental, was 82 g person⁻¹ day⁻¹, which is considerably higher than the daily consumption values in this study.

Table 4

Average consumption rates of mangrove clams (*Pegophysema philippiana*) per day across sampling areas

Station	Average no. of consumers	Times of consumption per month	Average consumption per day (g day ⁻¹)	Consumption rate (g person ⁻¹ day ⁻¹)
Sta. Cruz	5±2	1	136±88	1.31±1.55 ^a
Malita	4±2	1	282±136	7.87±11.17 ^b
Mati	5±2	1	280±158	4.69±4.54 ^{b, c}
Baganga	5±2	2	241±102	12.4±27.29 ^b

Note: different superscripts denote significant differences at $p < 0.05$.

Human health risk assessment of bivalve consumption. The estimated daily intake values of heavy metals through the consumption of mangrove clam are depicted in Table 5. To determine the health risk associated with Cd, Pb, and Hg contamination in humans, the EDI values of heavy metals in mangrove clams were compared to the provisional tolerable daily intakes (PTDI) recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA 2003; JECFA 2011) and the European Food Safety Authority (EFSA 2010). The comparison of EDI and PTDI values has been widely used to evaluate the adverse health effect of bivalve consumption (Mahat et al 2017; Qin et al 2021). The EDI values for Cd were 0.00223, 0.0003, 0.0001, and 0.00003 mg kg⁻¹ day⁻¹; for Pb, they were 0.006, 0, 0.0014, and 0.0004 mg kg⁻¹ day⁻¹; and for Hg, they were 0.0001, 0, 0.00002, and 0 mg kg⁻¹ day⁻¹ in Sta. Cruz, Malita, Mati, and Baganga, respectively. The EDI values for Cd (0.00223 mg kg⁻¹ day⁻¹) and Pb (0.0060 mg kg⁻¹ day⁻¹) in Sta. Cruz were relatively higher compared to the rest of the values in all the sampling stations, but they represented only 0.29 and 0.4% of the PTDI values for Cd (0.80 mg kg⁻¹ day⁻¹) and Pb (1.50 mg kg⁻¹ day⁻¹). Despite the mangrove clams' Cd and Pb levels in Sta. Cruz exceeding the safety limit imposed by FAO, the EDI values represent no health risk to humans considering the low consumption rate (1.31 g person⁻¹ day⁻¹) of mangrove clams in the area. Similarly, a study by Elvira et al (2021) on *Corbicula fluminea* in Laguna de Bay shows no health concern, revealing estimated EDIs for Cd (2.4×10^{-6} to 7.2×10^{-6} mg kg⁻¹ day⁻¹) and Pb (3.4×10^{-6} to 9.7×10^{-6} mg kg⁻¹ day⁻¹) in moderate to low consumption rates (0.4 to 2.2 g person⁻¹ day⁻¹). The current study indicates that ingestion of Cd, Pb, and Hg in mangrove clams does not present a health risk for the average consumer. Nevertheless, it should be considered that regular or excessive consumption of mangrove clams, particularly in Sta. Cruz, could exceed the recommended intake for Cd and Pb.

The provisional tolerable weekly intake (PTWI) and permissible weekly consumption of the mangrove clam, calculated based on the metal concentrations in the flesh of the mangrove clam, are summarized in Table 6. According to Alipour et al (2015), a PTWI value is an estimate of the quantity of a contaminant that humans can ingest over a lifetime without incurring risks on their health. JECFA established PTWIs for Cd, Pb, and Hg, which are 0.007, 0.025, and 0.0007 mg kg⁻¹ body weight for Cd, Pb, and Hg, respectively. Thus, PTWI for Filipino adults with an average weight of 65 kg is 0.46, 1.63, and 0.05 mg week⁻¹ for Cd, Pb, and Hg, respectively. Based on PTWI values and concentrations of heavy metals in mangrove clams, the permissible weekly consumption of mangrove clams was determined. This would serve as a basis for weekly human-safe consumption of mangrove clams in four sampling areas. Ideally, permissible consumption was calculated in mangrove clams for Cd, which ranged from 2.2 to 27.3 kg week⁻¹, and Pb, which ranged from 2.9 to 4.6 kg week⁻¹ for a 65 kg adult across the four sampling areas. Similarly, when we refer to the concentration of Hg, consuming 8.4 to 9.8 kg week⁻¹ is not a serious problem for health. Referring to the different metal concentrations in the four sampling areas, it is recommended to consume according to the lowest limit of 2.2 kg week⁻¹ for mangrove clams.

Table 5

The estimated daily intake (EDI) of Cd, Pb, and Hg in mangrove clams (*Pegophysema philippiana*) in four sampling areas of Davao Region

Metal	EDI (mg kg ⁻¹ day ⁻¹)				PTDI (mg kg ⁻¹ day ⁻¹)
	Sta. Cruz	Malita	Mati	Baganga	
Cd	0.0023	0.00003	0.0001	0.00003	0.80 ^a
Pb	0.0060	0	0.0014	0.0004	1.50 ^b
Hg	0.0001	0	0.00002	0	0.14 ^c

Note: EDI - estimated daily intake; PTDI - provisional tolerable daily intake; ^a - the PTDI value of Cd imposed by JECFA (2011); ^b - the PTDI value of Pb imposed by EPSA (2010); ^c - the PRDI value of methyl mercury was adopted as that of total Hg based on JECFA (2003).

Table 6

Permissible weekly consumption of mangrove clams (*Pegophysema philippiana*) (kg) by a 65-kg adult relative to PTWI criteria

Heavy metal	PTWI (mg 65 kg ⁻¹ weight wet)	*Permissible weekly consumption (kg)			
		Sta. Cruz	Malita	Mati	Baganga
Cd	0.46	2.2	27.3	13.7	17.1
Pb	1.63	2.9	-	3.6	4.6
Hg	0.05	9.8	-	8.4	-

Note: PTWI - provisional tolerable weekly intake; * - permissible weekly consumption = PTWI/concentrations of mangrove clams in wet weight.

Table 7 shows the target hazard quotient (THQ) values of heavy metals calculated according to the consumption of mangrove clams by residents in four selected areas in the Davao Region. The THQ value is considered to be a reliable parameter to assess the health risk of consuming bivalves contaminated with heavy metals (Mahat et al 2017; Wang et al 2018). If the THQ is equal to or greater than 1, consumers are potentially in danger of developing health problems. Conversely, if the THQ is less than 1, it poses no health-destructive impacts (Elvira et al 2021). To determine the overall danger to consumers, the cumulative risk expressed as the arithmetic sum of the THQ value (TTHQ) of each metal was also determined. The THQs of Cd, Pb, and Hg across the sampling areas were found to be less than 1 and the TTHQ values in all four sampling areas were far lower than 1, which signifies that it is unlikely to experience any deleterious effects from the consumption of mangrove clams in the Davao Region.

Table 7

Target hazard quotient (THQ) and the cumulative health risk (TTHQ) of heavy metals via consumption of mangrove clam (*Pegophysema philippiana*) by local residents in four selected areas in Davao Region

Metal	Sta. Cruz	Malita	Mati	Baganga
Cd	0.0042	0.00202	0.00241	0.00509
Pb	0.0032	0	0.00942	0.01926
Hg	0.00002	0	0.00007	0
TTHQ	0.0074	0.0020	0.0119	0.0243

The results of this study are comparable with the findings of the investigation of Tayone et al (2020) on metal exposure through the consumption of bivalves *A. maculosa* and *A. puerpera* in Pujada Bay, Davao Oriental. The computed THQ for Cd and Pb in their study was 0.035 and 0.029, respectively, neither of which exceeded the allowable value of 1. However, a study conducted by Mahat et al (2017) revealed that the consumption of mussel *P. viridis* in the coastal areas of Malaysia posed detrimental health impacts on the consumers, which recorded THQ values for Pb (16.72 to 25.71) and Cd (2.60 to 7.42) higher than the permissible limit of 1.

Conclusions. This study evaluated the levels of heavy metals (Cd, Pb, and Hg) in mangrove clams harvested in substantial quantities from coastal areas of the Davao Region. It was revealed that only Cd and Pb levels in Sta. Cruz exceeded the standard limits imposed by FAO. The assessment of human health risk was driven by both the concentration of heavy metals in mangrove clams and the estimated ingestion rate of the residents. The estimated daily intake (EDI) for all the heavy metals was found to be far below the provisional tolerable daily intake (PTDI). Based on the provisional tolerable weekly intake (PTWI), the recommended permissible weekly consumption of mangrove clams in four sampling areas was 2.2 kg per week for an adult with an average weight of 65 kg. The target hazard quotient (THQ) and total target hazard quotient (TTHQ) were less than 1, thus justifying no health concern with the consumption of mangrove clams. Overall, all the health risk indices were below the acceptable levels set by authorized agencies. Hence, Cd, Pb, and Hg posed no potential risk to residents in four selected areas of the

Davao Region. The study cannot establish a clear comparison of heavy metal concentrations in the edible tissue of mangrove clams in the environment. Mollusks can directly absorb heavy metals through direct exposure to water through their gills and the ingestion of sediments. Aside from mangrove clam tissue and sediments, it is also suggested to investigate the levels of heavy metals in the water. The study further recommends the risk assessment of hazardous chemicals and contaminants in other fishery commodities present in the area. This is to guarantee that seafood eaten by consumers is risk-free.

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Conflict of Interest. The authors declare that there is no conflict of interest.

References

- Alipour H., Pourkhabbaz A., Hassanpour M., 2015 Estimation of potential health risks for some metallic elements by consumption of fish. *Water Quality, Exposure and Health* 7(2):179-185.
- Appleton J. D., Williams T. M., Breward N., Apostol A., Miguel J., Miranda C., 1999 Mercury contamination associated with artisanal gold mining on the island of Mindanao, the Philippines. *Science of The Total Environment* 228(2-3):95-109.
- Benitez K. C. D., Cambia F. D., Banicod R. J. S., Perelonia K. B. S., Tadifa G. C., Tanyag B. E., Rivera A. T. F., Rivera A. T. F., Montojo U. M., 2021 Concentrations, seasonality, and risk assessment of cadmium in scallop, *Bractechlamys vexillum* (Reeve 1853) in the Visayan Sea, Philippines. *Food Control* 126:108021.
- Bersaldo M. J., Lacuna M. L., Macusi E., 2022 Status of mangrove clam (*Anodonta philippiana* Reeve, 1850) in Baganga, Davao Oriental, Philippines. *ABAH Bioflux* 14(2):55-66.
- Chakraborty S., Owens G., 2014 Metal distributions in seawater, sediment and marine benthic macroalgae from the South Australian coastline. *International Journal of Environmental Science and Technology* 11:1259-1270.
- Chien L., Hung T., Choang K., Yeh C., Meng P., Shieh P., Han B., 2002 Daily intake of TBT, Cu, Zn, Cd and As for fishermen in Taiwan. *Science of The Total Environment* 285(1-3):177-185.
- Dar M. A., Belal A. A., Madkour A. G., 2018 The differential abilities of some molluscs to accumulate heavy metals within their shells in the Timsah and the Great Bitter lakes, Suez Canal, Egypt. *The Egyptian Journal of Aquatic Research* 44(4):291-298.
- Ding H., Zhao Y., Sheng X., Kang X., Ning J., Zhong X., Shang D., 2022 Heavy metal bioaccumulation in bivalves from coastal areas of yellow sea and Bohai Sea, China: Evaluation of contamination and human health risk. Preprint. Available at: <https://doi.org/10.21203/rs.3.rs-1512790/v1>
- Diwa R. R., Elvira M. V., Deocarís C. C., Fukuyama M., Belo L. P., 2022 Transport of toxic metals in the bottom sediments and health risk assessment of *Corbicula fluminea* (Asiatic clam) collected from Laguna de Bay, Philippines. *Science of The Total Environment* 838:156522.
- EFSA Panel of Contaminants in the Food Chain (CONTAM), 2010 Scientific opinion on lead in food. *EFSA Journal* 8(4):1570.
- El Nemr A., El-Said G. F., Ragab S., Khaled A., El-Sikaily A., 2016 The distribution, contamination and risk assessment of heavy metals in sediment and shellfish from the Red Sea coast, Egypt. *Chemosphere* 165:369-380.
- Elvira M. V., Garcia C. M., Calomot N. H., Seronay R. A., Jumawan J. C., 2016 Heavy metal concentration in sediments and muscles of mud clam, *Polymesoda erosa* in Butuan Bay, Philippines. *Journal of Biodiversity and Environmental Science* 47(3):47-56.

- Elvira M. V., Faustino-Eslava D. V., de Chavez E. R., Lososo J. A., Fukuyama M., 2021 Human health risk associated with heavy metals from consumption of Asiatic Clam, *Corbicula fluminea*, from Laguna de Bay, Philippines. *Environmental Science and Pollution Research* 28(27):36626-36639.
- Gao Y., Wang R., Li Y., Ding X., Jiang Y., Feng J., Zhu L., 2021 Trophic transfer of heavy metals in the marine food web based on tissue residuals. *Science of The Total Environment* 772:145064.
- Ghabbour E. A., Shaker M., El-Toukhy A., Abid I. M., Davies G., 2006 Thermodynamics of metal cation binding by a solid soil-derived humic acid: binding of Fe(III), Pb(II), and Cu(II). *Chemosphere* 63(3):477-483.
- Ghani S. A., 2015 Trace metals in seawater, sediments and some fish species from Marsa Matrouh Beaches in north-western Mediterranean coast, Egypt. *The Egyptian Journal of Aquatic Research* 41(2):145-154.
- Hu Y., Cheng H., 2013 Application of stochastic models in identification and apportionment of heavy metal pollution sources in the surface soils of a large-scale region. *Environmental Science & Technology* 47(8):3752-3760.
- Jahromi F. A., Keshavarzi B., Moore F., Abbasi S., Busquets R., Hooda P. S., Jaafarzadeh N., 2021 Source and risk assessment of heavy metals and microplastics in bivalves and coastal sediments of the Northern Persian Gulf, Hormogzan Province. *Environmental Research* 196:110963.
- Kesavan K., Murugan A., Venkatesan V., Kumar B. S., 2013 Heavy metal accumulation in molluscs and sediment from Uppanar Estuary, southeast coast of India. *Thalassas* 29(2):15-21.
- Khan M. A., Khan S., Khan A., Alam M., 2017 Soil contamination with cadmium, consequences and remediation using organic amendments. *Science of The Total Environment* 601-602:1591-1605.
- Lias L., 2013 A preliminary study on heavy metal concentration in the marine bivalves *Marcia marmorata* species and sediments collected from the coastal area of Kuala Perlis, north of Malaysia. *IOSR Journal of Applied Chemistry* 4(1):48-54.
- Liu J., Cao L., Dou S., 2017 Bioaccumulation of heavy metals and health risk assessment in three benthic bivalves along the coast of Laizhou Bay, China. *Marine Pollution Bulletin* 117(1-2):98-110.
- Lovett R. J., Gutenmann W. H., Pakkala I. S., Youngs W. D., Lisk D. J., Burdick G. E., Harris E. J., 1972 A survey of the total cadmium content of 406 fish from 49 New York State fresh waters. *Journal of the Fisheries Research Board of Canada* 29(9):1283-1290.
- Lumogdang L. P., Llamag M. B., Antonio E. S., Bersaldo M. J. I., 2022 DNA barcoding based on 16S mitochondrial DNA (mtDNA) molecular marker of mangrove clams from the selected sites of Davao Region, Philippines. *Asian Journal of Fisheries and Aquatic Research* 16(6):40-47.
- Mahat N. A., Muktar N. K., Ismail R., 2017 Toxic metals in *Perna viridis* mussel and surface seawater in Pasir Gudang coastal area, Malaysia, and its health implications. *Environmental Science and Pollution Research* 25:30224-30235.
- Marquez E. T., Avenido P. M., 2022 Bioaccumulation potential of marine bivalve seashells on heavy metals along the intertidal zone of selected coastal barangays in Malita Davao occidental, Philippines. *South Florida Journal of Development* 3(2):2915-2929.
- Molina V. B., 2014 Health risk assessment of heavy metals in Manila catfish (*Arius dispar*) from Laguna Lake. *Acta Medica Philippina* 48(1):22-27.
- Montejo U. M., Baldoza B. J. S., Cambia F. D., Benitez K. C. D., Perelonia K. B. S., Rivera A. T., 2021 Levels and health risk assessment of mercury, cadmium, and lead in green mussel (*Perna viridis*) and oyster (*Crassostrea iredalei*) harvested around Manila Bay, Philippines. *Food Control* 124:107890.
- Mustapha O. M., Lawal O. S., 2014 Comparative study of heavy metal pollution of sediments in Odo-Owa and Yemoji Streams, Ijebu-Ode local government area, SW Nigeria. *IOSR Journal of Applied Chemistry* 7:12-17.
- Perelonia K. B., Abendanio C., Rana J. P., Opinion A. G., Villeza J., Cambia F. D., 2017 Heavy metal contamination in water and fishery resources in Manila Bay aquaculture farms. *The Philippine Journal of Fisheries* 24(2):74-97.

- Peters E. C., Gassman N. J., Firman J. C., Richmond R. H., Power E. A., 1997 Ecotoxicology of tropical marine ecosystems. *Environmental Toxicology and Chemistry* 16:12-40.
- Primavera J. H., Leбата M. J., Gustilo L. F., Altamirano J. P., 2002 Collection of the clam *Anodontia edentula* in mangrove habitats in Panay and Guimaras, Central Philippines. *Wetlands Ecology and Management* 10(5):363-370.
- Qin L. Y., Zhang R. C., Liang Y. D., Wu L. C., Zhang Y. J., Mu Z. L., Deng P., Yang L. L., Zhou Z., Yu Z. P., 2021 Concentrations and health risks of heavy metals in five major marketed marine bivalves from three coastal cities in Guangxi, China. *Ecotoxicology and Environmental Safety* 223:112562.
- Raju J., Johnson J., Jeevan A., Capili J. T., 2021 Heavy metal determination of bivalves in Cagayan Valley, Philippines. *Scholars Academic Journal of Biosciences* 9(10):256-258.
- Saavedra Y., Gonzalez A., Fernandez P., Blanco J., 2004 Interspecific variation of metal concentrations in three bivalve molluscs from Galicia. *Archives of Environmental Contamination and Toxicology* 47:341-351.
- Sarkar S. K., Cabral H., Chatterjee M., Cardoso I., Bhattacharya A. K., Satpathy K. K., Alam M. A., 2008 Biomonitoring of heavy metals using the bivalve molluscs in Sunderban mangrove wetland, northeast coast of Bay of Bengal (India): Possible risks to human health. *CLEAN - Soil, Air, Water* 36(2):187-194.
- Su G. L. S., Basilio J. E. S., Villanueva E. L. C., Su M. L. L. S., Rraggio E. M., Ramos G., Ajero M., 2015 Assessing copper bioaccumulation in edible bivalves sold in Lucena market, Quezon province, Philippines. *International Journal of Current Science* 15:E36-41.
- Swaleh M. M., Ruwa R. K., Wainaina M. N., Ojwang L. M., Shikuku S. L., Maghanga J. K., 2016 Heavy metals bioaccumulation in edible marine bivalve mollusks of Tudor Creek Mombasa Kenya. *IOSR Journal of Environmental Science, Toxicology and Food Technology* 10:43-52.
- Szefer P., 1986 Some metals in benthic invertebrates in Gdansk Bay. *Marine Pollution Bulletin* 17(11):503-507.
- Tayone J. C., Camille C. O., Tayone W., 2020 Selected mollusks from Pujada Bay, Philippines: Heavy metal health risk assessment and antibacterial activities. *Asian Journal of Biological and Life Sciences* 9(2):177-184.
- Wang X. N., Gu Y. G., Wang Z. H., Ke C. L., Mo M. S., 2018 Biological risk assessment of heavy metals in sediments and health risk assessment in bivalve mollusks from Kaozhouyang Bay, South China. *Marine Pollution Bulletin* 133:312-319.
- Waykar B., Deshmukh G., 2012 Evaluation of bivalves as bioindicators of metal pollution in freshwater. *Bulletin of Environmental Contamination and Toxicology* 88:48-53.
- Yap C. K., Cheng W. H., Karami A., Ismail A., 2016 Health risk assessments of heavy metal exposure via consumption of marine mussels collected from anthropogenic sites. *Science of The Total Environment* 553:285-296.
- Zhuang P., Li Z., McBride M., Zou B., Wang G., 2013 Health risk assessment for consumption of fish originating from ponds near Dabaoshan mine, South China. *Environmental Science and Pollution Research* 20(8):5844-5854.
- Zuykov M., Pelletier E., Harper D., 2013 Bivalve mollusks in metal pollution studies: From bioaccumulation to biomonitoring. *Chemosphere* 93(2):201-208.
- *** AOAC, 2012 Official method of analysis. 19th Edition, Washington DC, pp. 121-130.
- *** FAO (Food and Agriculture Organization), 1983 Compilation of legal limits for hazardous substances in fish and fishery products. Fisheries Circular No. 464:5-100.
- *** FAO (Food and Agriculture Organization), 2018 Dietary assessment: A resource guide to method selection and application in low resource settings. Rome, 172 p.
- *** hydrosheds.org
- *** JECFA, 2003 Summary and conclusions of the 61st meeting of the Joint FAO/WHO Expert Committee on Food Additives. Joint Food and Agriculture Organization/World Health Organization Expert Committee on Food Additives.
- *** JECFA, 2011 Evaluation of certain food additives and contaminants. Seventy third report of the Joint FAO/ WHO Expert Committee on Food Additives. WHO Technical Report Series, No. 960. Joint FAO/WHO Expert Committee on Food Additives.

- *** PSA (Philippine statistic Authority), 2022 2021 Economic performance of Davao Region. Reference Number SR-22RSS011-010.
- *** US EPA (United State Environmental Protection Agency), 2015 Learn about lead. Available at: <http://epa.gov>

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