Asian Journal of Chemical Sciences

Volume 14, Issue 2, Page 113-130, 2024; Article no.AJOCS.113303
ISSN: 2456-7795

Health Risks Assessment of Heavy Metal Concentration in Cultured *Chanos chanos* (Bangus) and *Scylla serrata* (Mudcrab) in Selected Municipalities in Northern Samar

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Authors' contributions

This work was carried out in collaboration among all authors. Authors JLE, TJLA, MLCA and KMCL conducted the preparation and analysis of heavy metals. Authors TJLA, KRLD and MCM have contributed on the analyses of heavy metals with its health risks implications as well as the statistical analyses. All authors read and approved the final manuscript.

Article Information

DOI: 10.9734/AJOCS/2024/v14i2298

Open Peer Review History:
This journal follows the Advanced Open Peer Review policy. Identity of the Reviewers, Editor(s) and additional Reviewers, peer review comments, different versions of the manuscript, comments of the editors, etc are available here: https://www.sdiarticle5.com/review-history/113303

Original Research Article

Received: 01/01/2024
Accepted: 05/03/2024
Published: 18/03/2024

ABSTRACT

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One of the common problem in fishponds is heavy metal contamination. Though there are some heavy metal elements that are naturally occurring, but due to human activities, their concentration goes beyond what is normal. In this study, health risks analysis using Estimated Daily Intake (EDI), Total Hazard Quotient (THQ), Target Cancer Risk (TCR) were done to assess if the quantities of the heavy metals, such as: Arsenic, Cadmium, Chromium, Lead, and Mercury, impose risks to consumer. Arsenic had the highest concentration among all other heavy metals in crab aligue, having 46.83 mg/kg. The consumption of bangus meat may result in an EDI that is greater than PTDI, especially for Arsenic [15.22731-18.10317 μg kg⁻1 BW d⁻1]. Similarly, consuming crab aligue may also result to a high EDI for Arsenic [2.48197-5.27841μg kg⁻1 BW d⁻1]. THQ was also evaluated as well as the sum of individual heavy metal values which is the Hazard Index (HI) that exceeded to 1 multiple times. In terms of TCR levels, all of the heavy metals exceeded the acceptable limit for cancer risks. Shapiro-Wilk Test had shown non-normal distribution of data for EDI, THQ, and TCR. Spearman's Correlation Test, meanwhile, suggested that there is a significant relationship between the quantities of heavy metals in bangus meat and crab aligue as well as EDI, THQ, and TCR. In general, based on the health risks assessments (EDI, THQ, and TCR), Arsenic, an established carcinogen, can be the greatest contributor in developing risks and disease, while the varying concentration of Chromium and Cadmium in the samples may also pose risks to consumers. This implies that strict management measures should be implemented to mitigate or lessen the discharge of these heavy metals in the aquatic systems.

Keywords: Heavy metals; health risks; THQ; TCR; EDI; hazard index; aquatic systems; developing risks; food sources; food safety.

1. INTRODUCTION

The presence of heavy metal concentration in different food sources such as fishes and crustaceans could alter the life processes of an organism that consumes it. The extent of absorption of these metals in the body is measured by bioaccumulation and bioconcentration. In the study done by Jakimska et al [1], elements such as Mercury (Hg), Lead (Pb), and Cadmium (Cd), are toxic and adversely affect the DNA and other enzymatic activity in the body. Increased levels of these heavy metals in the food may lead to renal failure, liver damage, coma, mental retardation, infertility, hypertension, tumours, and even death [2]. These heavy metals also attack proteins and membrane lipids, thereby disrupt cellular integrity and functions [3,4].

The consumption of fish and crustaceans had changed drastically. To combat the over-exhaustion and harvesting of such resources from the wild, aquaculture-like establishment of fishponds were considered [5-8]. Rapid urbanization, improper land use planning and pattern, fast industrial development, and human population explosion are the major activities that affect the aquatic ecosystems [9,10]. Meanwhile, in terms of the feeds used in the fishponds, tannery and poultry wastes are often used as a cheap source of fish feed globally. The use of such feed stocks may possibly, or theoretically, increase the accumulation of toxic contaminants such as heavy metals in cultured fish and may pose a food safety risk and health risks [11,12-17].

In aquatic systems, fish samples as well as invertebrate organisms (e.g. mollusks, crustaceans, and etc.,) are observed as one of the indicative organisms for the evaluation of metal pollution [18,19,20-24]. Fish accumulates substantial amounts of metals in its tissues, especially in their muscles, thus posing a major risk for humans as it is considered as one of their vital dietary sources [18,2,25]. In that way, the accumulated heavy metals from fish and crustaceans may enter the food chain [2]. This makes heavy metal contamination a problem requiring government intervention and global attention.

This paper aims to assess the associated health risks of five heavy metals As, Cd, Cr, Pb, and Hg detected from bangus meat and crab (aligue), in terms of estimated daily intake (EDI), total hazard quotient (THQ), and target cancer risk (TCR). In addition, the relationship of the levels of heavy metal concentration between bangus meat and crab aligue to health risks in the study site using Multivariate Analysis of Covariance (MANCOVA) and also provide recommendations on fishponds management to lower the risk on the consumption of heavy metals.

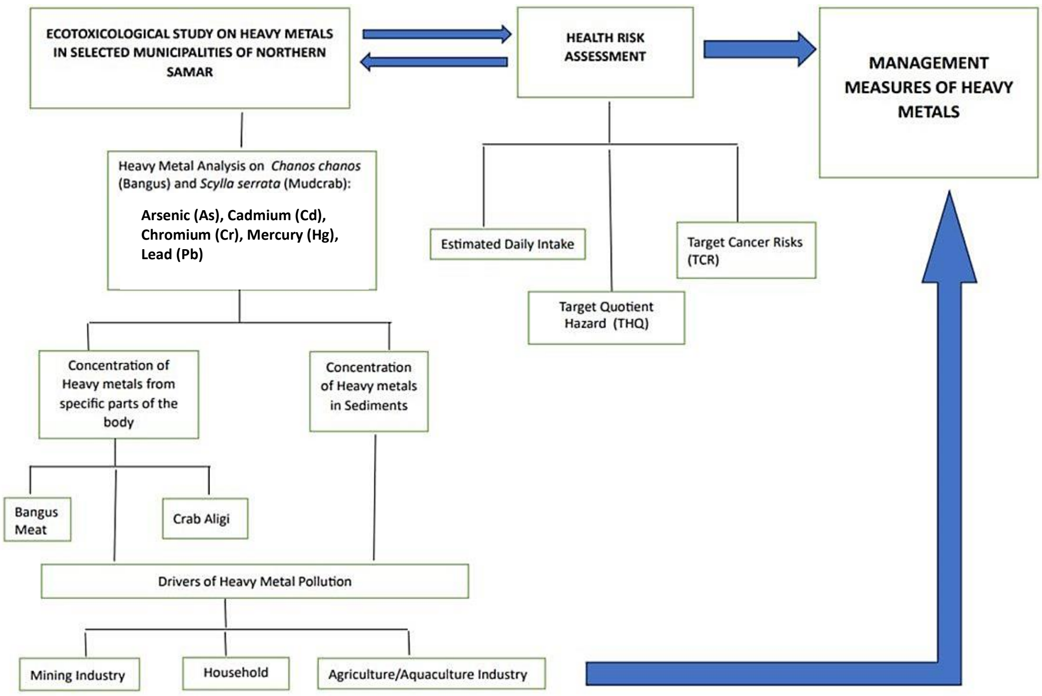

Fig. 1. Conceptual framework on heavy metals using DPSIR

1.1 Conceptual Framework

This study utilized a Driver-Pressure-State-Impact-Response (DPSIR) framework in analyzing the health risk indices on heavy metals concentration in cultured bangus (*Chanos chanos*) and mudcrab (*Scylla serrata*).

The DPSIR framework assesses the changes in environmental quality and its impact on the ecosystem, the society leading to political responses in terms of prioritization, and target setting in order to solve the specific problem in the environment [26,27]. In this study, it was found out that four municipalities of Northern Samar have cases of heavy metal concentration in bangus and crabs. Thus, this framework is used to examine further what are the drivers of these discharges and its possible impact on health. Management measures on heavy metals is shown to be a possible response of the local government units in order to mitigate or lessen the health risks to the surrounding communities (Fig. 1).

2. METHODOLOGY

2.1 Collection Site and Sampling

Samples were gathered from the four fishponds in four selected municipalities (Fig. 2) in the province of Northern Samar.

2.2 Preparation of Bangus and Crab Samples As Well As Tissue Preparation for Digestion

After gathering the samples from the sampling area, they were brought to the College of Science, Chemistry Laboratory, University of Eastern Philippines, for the removal of meat and meat near the stomach part which were done separately. The bangus meat and crab stomach/aligue were dried in an oven until it reached crispness and were pulverized, then, it was restrained separately in a clean vial for digestion.

2.3 Sample/ Tissue Digestion

Before the analysis, samples were subjected to microwave acid digestion. Briefly, dried bangus meat and crab stomach/aligue were weighed to approximately 0.5g and added with 7mL concentrated HNO_3 and 1 mL 30% H_2O_2. Digestion of each sample was done using Milestone Ethos Up Microwave Digestion System with the following digestion program:

2.4 Quantification of Heavy Metals

Digested samples were cooled to room temperature and filtered. Then, digested solutions were diluted to volume. Atomic emission spectrometric analyses were done in triplicates based on EPA 200.7 method using Shimadzu ICP-9000 spectrometer (ICP-OES). Various concentrations of certified reference standards for As, Cd, Cr, and Pb were also prepared to construct the calibration and determined the method LOD and LOQ. For Hg, the Standard Methods for Water and Wastewater serves as reference with the aid of Cold Vapor Atomic Absorption Spectrometer (CV-AAS). The digestion and quantification of As, Cd, Cr, and Pb were done at the UPLB Nanotechnology Laboratory, while digestion and quantification of Hg were done at Mach Union Laboratory. The results are reported in mg/kg.

2.5 Health Risk Assessment

Health risk assessments for the concentration of the five heavy metals were done to uncover health risks that are linked to the medical problems that can be developed over a longer period of exposure. In this study, the EDI, THQ, and TCR were assessed.

2.6 Estimated Daily Intake (EDI)

The calculation of Griboff et al*.* [11] was followed for the EDI of heavy metals [28,19]. The main guide for the calculation was based on the study of Cabahug et al*.* [29].

$$EDI = (C \times IRd)\ BW^{-1}$$

where C is the heavy metal concentration per species from each site (mg kg–1 DW), while IRd is the daily average ingestion rate (38.36 g d–1 or per day for fishes according to the Department of Science and Technology-Food and Nutrition Research Institute (DOST- FNRI) between 2018-2019 and 8.3 g d–1 or per day for crustaceans) (Massachusetts Department of Environmental Protection, 2008; Griboff et al*.*, 2017), meanwhile BW is the average body weight (BW) of Filipino adults (70 kg) [12,13]. EDI is measured by µg/kg BW/d.

2.7 Target Quotient Hazard (THQ)

The calculation of Cabahug et al*.* [29] and Tayone et al*.* [30] on THQ were adopted in this study. The THQ is the ratio of hazardous element exposure to the reference dose. It is the greatest amount at which no adverse health consequences are predicted. The reference dose is unique to the trace elements under investigation. The THQ identifies the non-carcinogenic health risk presented by the hazardous substance in question. A non-carcinogenic health impact is not predicted if the THQ is 1 or less. Otherwise, there is a possibility that negative health problems will occur. A THQ that is greater than 1, however, does not indicate a statistical likelihood of negative non-carcinogenic health consequences. The THQ can be estimated using the US-EPA formula, as demonstrated by Tayone et al*.* [30].

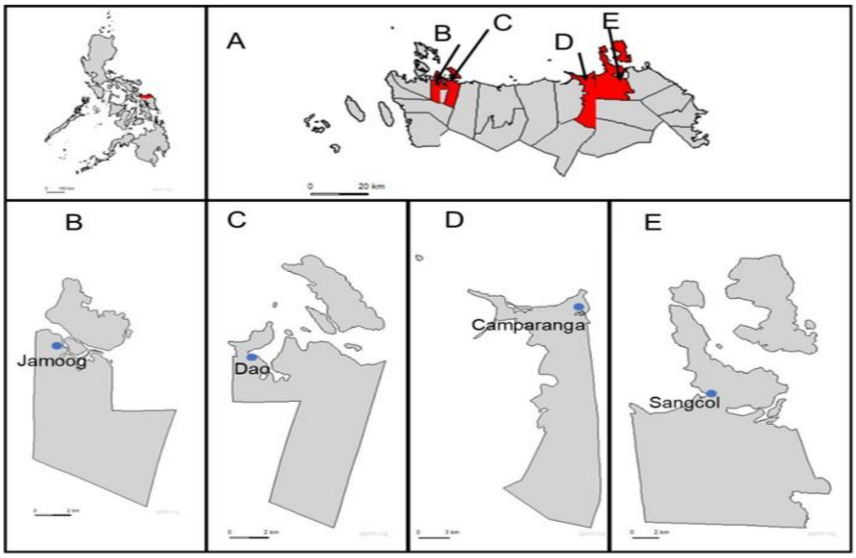

Fig. 2. Map of the collection site for the cultured bangus and crabs; A). Province of Northern Samar; B). Barangay Jamoog in the municipality of Rosario; C). Barangay Dao in the municipality of San Jose; D). Barangay Camparanga in the municipality of Pambujan; and E). Barangay Sangcol in the municipality of Laoang (Source: GADM.org)

Table 1. Digestion program

Step	Time	Temperature	Microwave Power
1	20 minutes	27°C-200°C	Up to 1800W
2	20 minutes	200°C	Up to 1800W

THQnon-carcinogenic = (EF × ED × Ird × C) / [(RfD × BW × AT)]

where THQnon-carcinogenic is the THQ for non-carcinogenic risk, EF is exposure frequency (104 d yr–1 assuming twice a week consumption), ED is the exposure duration (60 yr for adults), IRd is the ingestion rate (86.03 g per day for bangus and 7.89g per day for crustaceans) (Southeast Asian Fisheries Development Center [31], C is the heavy concentration in aquatic products (mg kg–1 DW), RfD is the oral reference dose values based from Liu et al. [32], BW is the average BW (70 kg), and AT is the average lifetime exposure (EF x ED).

2.8 Target Cancer Risks (TCR)

The TCR is a tool to determine the risk of cancer as a result of exposure to carcinogenic chemicals or materials. In this case, carcinogens are being eaten by the bangus and crabs which are the top cultured seafoods. An oral slope factor is used instead of an oral reference dosage to determine THQ. This component, combined with the carcinogen dosage, determines the likelihood of increased cancer risk over the lifespan of the exposed individual. The equation for TCR is adopted from Cabahug et al. [29]:

TCR = (EF × ED × Ird × C × CPSo)/ [(BW × AT)] × 10E^(–3)

where EF is the exposure frequency of 104 days (twice a week) exposure to the element, ED is the exposure duration average of 60 years for Filipinos (57 yr for males and 63 yr for females according to Banada and Andel [33], IRd is the food ingestion rate, C is the concentration in weight of the trace element from the representative composite samples (μg g–1), CPSO represents the oral cancer slope factor used in this study, wherein 1.5 for inorganic As, 0.5 for Cr, and 0.004 for Pb expressed as mg kg–1 d–1 [34] 0.38 for Cd, Hg was not included since it has no CPSO or is unable to cause cancer. BW is the estimated BW of 70 kg, AT is the average exposure time to the carcinogen (EF x ED or 104 d * 60 yr), and 10–3 is the unit conversion factor [34].

2.9 Data Analysis

The relationship of the heavy metals and health risks was analyzed using Multivariate Analysis of Covariance (MANCOVA). MANCOVA determines the relationship of two or more dependent variables and independent variables after controlling the effect of covariates. Shapiro-Wilk Test was used to determine the normality of the EDI, THQ, and TCR, while the Spearman's rank correlation coefficient was used to determine the correlation between the health risks and heavy metals present in crab aligue and bangus meat. Data analysis was done on R.

3. RESULTS

3.1 Concentration of Heavy Metals in Sediments

Table 2 shows the data heavy metal concentration in the sediments of selected fishponds. The quantification of heavy metals in the sediments of fishponds were also determined to trace if the crabs have possibly acquired the heavy metals in sediments due to their feeding habit. Based on Tables 2 and 3, results clearly showed that both bangus and mudcrabs contain the concerned heavy metals.

Table 2. Quantity of heavy metals in sediment in northern samar municipalities

	Heavy Metal Concentration in Sediment				
Heavy Metals	**Rosario**	**San Jose**	**Pambujan**	**Laoang**	**Standard Limit**
Arsenic	270.73*	373.53*	288.87*	342.09*	5.00
Cadmium	40.65*	41.69*	41.04*	39.90*	0.80
Chromium	88.52	90.79	89.37	86.91	100.00
Lead	131.40*	168.77*	137.91*	156.41*	85.00
Mercury	0.90	0.90	0.60	0.70	0.50-1.00

Notes. Standard limit based on the WHO (1993).
**Heavy metals that exceeded the limit.*

Table 3. Quantity of heavy metals accumulated in cultured *Chanos chanos* (Bangus meat) and *Scylla serrata* (Crab aligue)

Heavy Metals	Rosario	San Jose	Pambujan	Laoang	Standard Limit
Heavy Metal Concentration in Bangus Meat					
Arsenic	13.03	12.39	14.73	14.37	0.6-37
Cadmium	0.78	5.91*	5.95*	0.73	1.0
Chromium	11.08	12.97	13.06	12.33	50.0
Lead	1.47	2.19*	0.60	1.22	2.0
Mercury	0.35	0.35	0.32	0.35	0.5
Heavy Metal Concentration in Crab Stomach/Aligue Meat					
Arsenic	27.11	34.66	46.83*	22.02	0.6-37
Cadmium	0.97	12.84*	14.14*	0.93	1.0
Chromium	23.22	28.05	30.87	30.78	50.0
Lead	1.32	9.45*	7.79*	0.91	2.0
Mercury	0.50	0.50	0.48	0.48	0.5

Notes. Standard limit based on the WHO, the FAO, and the US-EPA.
**Heavy metals that exceeded the limit.*

3.2 Heavy Metals Concentration from the Specific Parts of Bangus and Mudcrab

Table 3 shows the quantity of heavy metals accumulated in the different parts of bangus and mudcrab samples. There are several variations observed in the concentrations of heavy metals in terms of As, Cd, Cr, and Pb both in bangus meat and aligue of crabs, whereas there is no significant variation observed in the concentration of Hg in bangus and crab. Among all heavy metals, As in crab aligue had the highest concentration with 46.83 mg/kg or ppm from Pambujan.

3.3 Sources of Heavy Metal Pollution

Based on the ocular observation of study sites, there are some buildings/houses, small- scale piggeries, dumpsites in the vicinity or just a few meters from the fishponds to where the samples are collected. Cheng et al*.* [35] mentioned that much of the As is concentrated in the sediments of fishponds. Although As is naturally occurring in the environment, its inorganic form is considered as harmful and its presence in an aquatic environment can be due to anthropogenic activities such as electronics, agriculture, and metallurgy [35]. However, the nearest cause of contamination in fishponds could be from agriculture since McNelly (2022) recounted that a wide range of fertilizers contains elevated amounts of As and other heavy metals like Cd and Pb. It was also observed during sample collection that fishponds, where the bangus and crab samples were collected, have farms around its vicinity. Cd's presence in the aquatic environment can be due to both natural and anthropogenic activities [18]. Moreover, the Centers for Disease Control and Prevention [36] had suggested that the entry of Cd in the bodies of water can be due to action of wind and rain (e.g. surface runoff).

Contrary to As, Cd was found concentrated on the surface waters [37]. Fish and crabs accumulate Cd in the waters [38] coming from agriculture, feeds, and water sources (e.g. river and sea). As observed during the sampling, one fishpond in San Jose sourced out its water from a river. As noted by Mannzhi et al. [39], rivers are contaminated with hazardous chemicals such as: pesticides, trace metals, and effluents from houses. Mannzhi et al*.* [40] also recounts that quality of water as well as the feeds have an impact on the cultured organisms in the fishponds. Among all heavy metals included in the study, Cr is the one that did not exceed to the tolerable levels set by FAO and WHO. Main source of Cr in sediments could be suspected from leaching from chromite mining sites [40] in other towns in Samar Island or there could be a possibility that sediments in selected sampling sites have high chromite and chromium reserves.

Pb is a nonessential element that is not needed by most organisms. Pb are introduced through discharge of waste water from industries and anthropogenic activities [41]. The fishpond in San Jose has houses nearby and also some rear livestocks, which might contribute to the fluctuation of Pb in the sampled fishpond. According to the University of Toledo [42], main sources of Pb contamination in bodies of water can be found in household materials which includes: lead-containing waste products (e.g. batteries), lead-based paints, lead dust, water pipes, home remedies, and cosmetics, among others.

In addition to Cr, Hg is another heavy metal that did not exceed the tolerable limit set by WHO, US-EPA, and FAO. However, although Hg did not exceed the tolerance limit, its presence in fishes, crabs, and sediments is alarming. Mercury is a common pollutant of aquatic ecosystems and has a substantial impact on both human and wildlife health. Contamination may be attributed to the improper disposal of house materials containing the said element (e.g. light bulbs). Moreover, mercury can be converted through microorganisms into methylmercury, a highly toxic chemical that builds up in fish, shellfish and animals that eat fish [43].

3.4 Health Risk Assessment

The Philippines is composed of island provinces, having direct access to the seas and ocean, Filipinos have included fishes and crustaceans, as two of the staple animal food products, in their diet and serves as their source of protein. In an article by Lagniton [44], a report by DOST-FNRI stated that individual Filipinos in the year 1993 consumed 36 kg (which accounts for 98.63g/day) of fish. However, in a recent study by SEAFDEC [31], there is a gradual change in the consumption of fish by Filipino at only 31.4kg (86.03g/d). This slight decrease in fish consumption can be attributed to a variety of food choices nowadays. However, the decrease in consumption of fish does not necessarily mean the health risks posed by dangerous chemicals that are included in the water system or either in sediments to where fishes are exposed when they are in fishponds is lesser. As such, possible health risks will be assessed through EDI, THQ, and TCR.

3.4.1 Estimated Daily Intake (EDI)

Tables 4 and 5 shows the PTDI (μg $kg^{-}1BW$ $d^{-}1$) and the EDI of heavy metals from consuming bangus and crabs. Results show that consumption of bangus meat brought the EDI level of As and Cr above the PTDI at 18.10317 μg $kg^{-}1$ BW $d^{-}1$ and 16.05074 μg $kg^{-}1$ BW $d^{-}1$, respectively. The rest of the heavy metals, meanwhile, are below the PTDI.

Table 4. Estimated daily intake (adult) in mg kg–1 body weight d–1 (Bangus)

Heavy Metals		Estimated Daily Intake			
		Bangus Meat			
	Rosario	San Jose	Pambujan	Laoang	PTDI
Arsenic	16.01387*	15.22731*	18.10317*	17.66073*	0.30
Cadmium	0.95862	7.26339*	7.31255*	0.89717	1.00
Chromium	13.61732*	15.94013*	16.05074*	15.15357*	3.00
Lead	1.80663	2.69151	0.73740	1.49938	3.57
Mercury	0.43015*	0.43015*	0.39328*	0.43015*	0.1

Notes. [PTDI] provisional tolerable daily intake [32]
**Heavy metals that exceeded PTDI*

Table 5. Estimated daily intake (adult) in μg kg–1 body weight d–1 (Crabs)

Heavy Metals		Estimated Daily Intake			
		Crab Stomach/Aligue			
	Rosario	San Jose	Pambujan	Laoang	PTDI
Arsenic	3.05568*	3.90668*	5.27841*	2.48197*	0.30
Cadmium	0.10993	1.44725*	1.59378*	0.10482	1.00
Chromium	2.61723	3.16164*	3.47949*	3.46935*	3.00
Lead	0.14878	1.06515	0.87804	0.10257	3.57
Mercury	0.05636	0.05636	0.05410	0.05410	0.1

Notes. [PTDI] provisional tolerable daily intake [32]
**Heavy metals that exceeded PTDI*

Table 5, on the other hand, shows the EDI of the crab aligue. Similar to the results of the crab, As and CR also exceeded the PTDI at 5.27841 μg kg^{-1} BW d^{-1} and 3.47949 μg kg^{-1} BW d^{-1}. The other elements also have lower EDI than the PTDI.

3.4.2 Target Quotient Hazard (THQ)

To further assess the health risks, THQ was calculated. The THQ of the samples were presented in Table 6. The THQ of As, Cr, Cd, and Hg when consuming bangus ranged from 60.34180-50.75593, 5.35006-4.53895, 7.31230-0.89714, and 4.30135-3.93266, respectively. It should be noted that the THQ of Pb is not considered since the amount of bangus meat consumed is less than 1. The hazard index (HI) of heavy metal per bangus ranged from 77.14750-63.69276.

On the other hand, the THQ (Table 7) of As and Cd from consuming crab aligue ranged from 17.59470-8.27323 and 1.59378-1.44725, respectively. Whereas the THQ of Cr, Pb, and Hg are below its PTDI with some being less than 1. The HI of heavy metal per crab aligue ranged from 21.14021- 16.39129 which is lower as compared to HI of bangus meat.

3.4.3 Target Cancer Risks (TCR)

The TCR due to consumption of bangus meat by adults from the four selected municipalities in Northern Samar was presented in Table 8. The consumption of bangus meat showed that the TCR for As, Cd, Cr, and Pb ranged 0.6852-0.07206, 0.00834- 0.00102, 0.02408-0.02043, and 0.00003-0.00001, respectively. Hg has no TCR value, however, due to its inability to cause cancer. Be that as it may, mercury in the environment has hazardous and toxic effects once it is converted into methylmercury.

The Total Cancer Risk (TCR) due to consumption of crab aligue by adults from the four selected municipalities in Northern Samar was presented in Table 9. The consumption of crab aligue showed that the TCR for As, Cd, Cr, and Pb ranged 0.02375-0.01117, 0.00182-0.00012, 0.00522-0.00393, and 0.00067-0.00000, respectively.

Table 6. Estimated THQ and HI due to twice a day consumption of bangus meat from the selected municipalities of Northern Samar

Heavy Metals	Total Quotient Hazard (THQ)				
	Bangus Meat				
	Rosario	San Jose	Pambujan	Laoang	PTDI
Arsenic	53.37771*	50.75593*	60.34180*	58.86705*	0.30
Cadmium	0.95859	7.26314*	7.31230*	0.89714	1.00
Chromium	4.53895*	5.31319*	5.35006*	5.05101*	3.00
Lead	0.51616	0.76898	0.21068	0.42838	3.57
Mercury	4.30135*	4.30135*	3.93266*	4.30135*	0.1
Hazard Index	63.69276*	68.40259*	77.14750*	69.54493*	>1

Notes. [PTDI] provisional tolerable daily intake [32] Total HI Index should be > 1 [2]
**Heavy metals that exceeded PTDI*

Table 7. Estimated THQ and HI due to twice a day consumption of crab stomach/aligue meat from selected municipalities of Northern Samar

Heavy Metals	Total Quotient Hazard (THQ)				
	Crab Stomach/Aligue				
	Rosario	San Jose	Pambujan	Laoang	PTDI
Arsenic	10.18561*	13.02226*	17.59470*	8.27323*	0.30
Cadmium	0.10933	1.44725*	1.59378*	0.10482	1.00
Chromium	0.87241	1.05388	1.15983	1.15645	3.00
Lead	0.04251	0.30433	0.25087	0.02931	3.57
Mercury	0.56357*	0.56357*	0.54103*	0.54103*	0.1
Hazard Index	11.77343*	16.39129*	21.14021*	10.10484*	>1

Notes.[PTDI] provisional tolerable daily intake [32] Total HI Index should be > 1 [2]
**Heavy metals that exceeded PTDI*

Table 8. Estimated Target Cancer Risk for adults due to twice a week consumption of bangus meat from selected municipalities of Northern Samar

Heavy Metals	Total Cancer Risk			
	Bangus Meat			
	Rosario	San Jose	Pambujan	Laoang
Arsenic	0.07206	0.6852	0.08146	0.07947
Cadmium	0.00109	0.00828	0.00834	0.00102
Chromium	0.02043	0.02391	0.02408	0.02273
Lead	0.00002	0.00003	0.00001	0.00002
Mercury	-	-	-	-

Notes.[TCR] reference values [34]: "unacceptable" if greater than 0.0001 after short period of exposure; "acceptable" if lesser than 0.000001; "acceptable for lifetime" if 0.0001–0.000001*

Table 9. Estimated Target Cancer Risk for adults due to twice a week consumption of crab stomach/aligue from selected municipalities of Northern Samar

Heavy Metals	Total Cancer Risk			
	Crab Stomach/ Aligue			
	Rosario	San Jose	Pambujan	Laoang
Arsenic	0.01375	0.01758	0.02375	0.01117
Cadmium	0.00012	0.00165	0.00182	0.00012
Chromium	0.00393	0.00474	0.00522	0.00520
Lead	0.00067	0.00001	0.00001	0.00000
Mercury	-	-	-	-

Notes.[TCR] reference values [34]: "unacceptable" if greater than 0.0001 after short period of exposure; "acceptable" if lesser than 0.000001; "acceptable for lifetime" if 0.0001–0.000001*

3.5 Statistical Analysis

Shapiro-Wilk Test was used to check for the normality of EDI, THQ, and TCR. The Shapiro-Wilk test statistic for the variable EDI (Fig. 3) is 0.7726 with a p-value equals 2.334e-07 thus, rejecting the null hypothesis that EDI follows a normal distribution. Below is a histogram of EDI that supports the non-normal distribution of the data.

THQ (Fig. 4), meanwhile, has a Shapiro-Wilk test statistic equals to 0.52741 with a p- value equals 3.622e-10 thus, also rejecting the null hypothesis that THQ follows a normal distribution. Below is a histogram of THQ that supports the non-normal distribution of the data.

Lastly, the Shapiro-Wilk test (Fig. 5) statistic for the variable TCR has a value equal to 0.65996 with a p-value of 2.307e-07. This also means that we will reject the null hypothesis that the data follows a normal distribution. Below is a histogram of TCR that supports the non-normal distribution of the data.

3.6 MANCOVA Results

MANCOVA (Table 10) was done with EDI, THQ, and TCR as dependent variables while heavy metals in sediment, heavy metals in crab aligue and bangus meat, and the dummy variable for heavy metals as independent variables. Moreover, we included the dummy variables for the type of resource and their location as covariates. Results show that the independent variables and the type of resource is significant at 99% confidence level.

3.7 Correlation Result (Spearman's Rank Correlation Coefficient)

Spearman's correlation is a test to measure the strength of relationship of data. It is used when the data does not follow normal distribution.

Results on THQ and the heavy metals found in the resources showed a coefficient of 0.496 indicating a positive correlation. The correlation is also deemed significant due to its p-value of 0.001.

Similar to THQ, TCR is also positively correlated to heavy metals found in the resources by 0.638. It is also deemed significant with a p-value of 8.5e-05

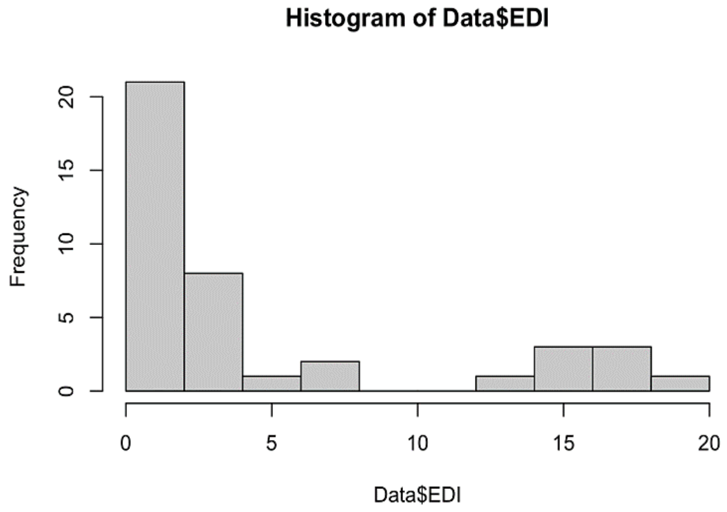

Fig. 3. Shapiro- Wilk test for normality of EDI

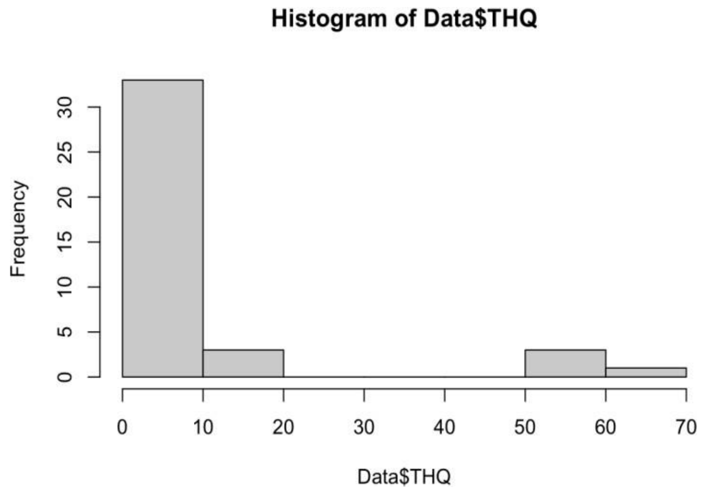

Fig. 4. Shapiro- Wilk Test for normality of THQ

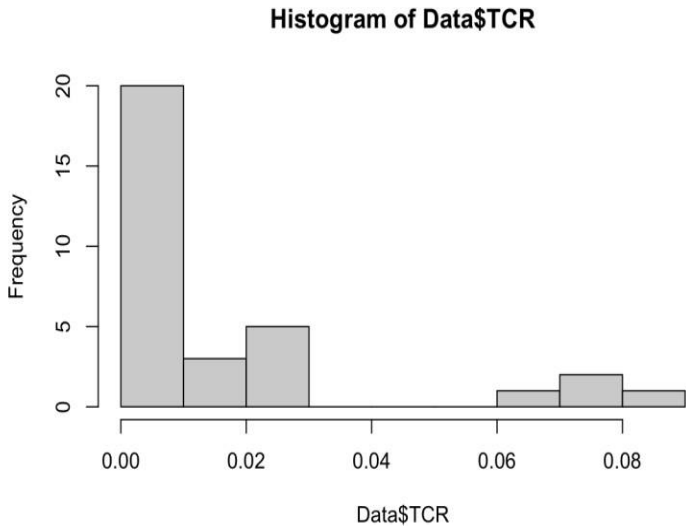

Fig. 5. Shapiro- Wilk test for normality of TCR

Table 10. MANCOVA Results

Degrees of Freedom		Pillai's Trace	Approx F	Num Df	Den Df	Pr(>F)
Independent Variables	3	0.90469	3.7420	9	78	0.0005997***
Location_Dummy	1	0.00151	0.0121	3	24	0.9981286
Resource_Dummy	1	0.61683	12.8785	3	24	3.24e-05***
Residuals	26					

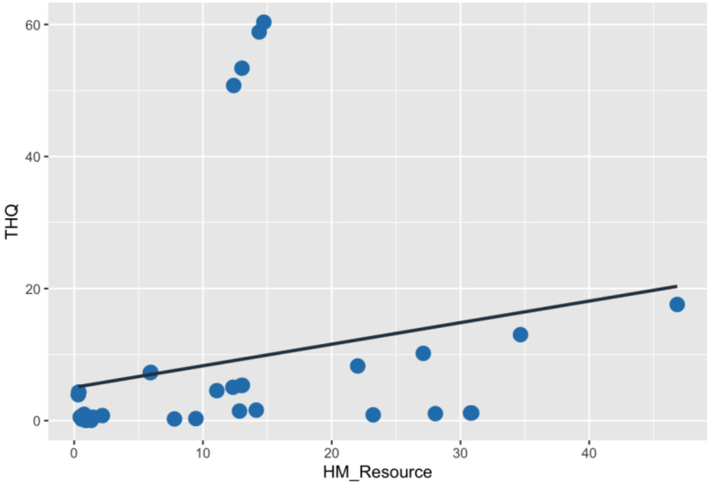

Fig. 6. Spearman Correlations test for THQ

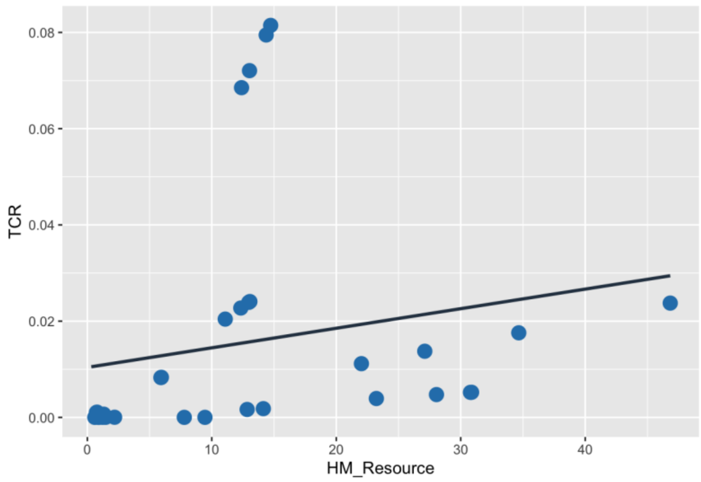

Fig. 7. Spearman Correlations test for TCR

4. DISCUSSION

The presence of heavy metals in aquatic systems originates from the natural interactions between the water, sediments, and atmosphere [28]. However, the impacts of activities around the fishponds in selected municipalities of Northern Samar had contributed and altered the aquatic systems and added to the existing natural interactions which resulted in high concentrations of heavy metals in sediments, cultured bangus, and crabs. The varying concentrations of heavy metals in bangus meat and crab aligue/stomach can be attributed to their feeding habits. Bangus, being a pelagic fish, was assumed to have a lower heavy metal in its meat but results showed that it has high amounts of heavy metals instead. This entails that in a fishpond set-up, bangus is also considered a substratum/bottom feeder and an iliophagous since fishpond provides a shallow environment to the bangus [46]. On the other hand, crab is a bottom feeder where they depend on the organic materials that sink down in the sediments and, in other way, they also filter microparticles in the bottom of the bodies of water. Compared with bangus meat, the heavy metal such as As and Cr in crab aligue is higher. There is no doubt that there is a positive correlation on the higher heavy metals in sediments and the higher heavy metal concentration in crabs as presented in Table 2. Heavy metal such as As is much concentrated in sediments [18]. In addition, high concentrations of Fe, Cu, Mn, Cr, Zn and Pb are also recorded in bottom sediments as reported by Aledesanmi et al. [45].

Furthermore, Hg levels in sediments were the lowest but is considered alarming as compared to other concerned heavy metals since mercury is present in household products (e.g. thermometers, gas appliances, and fluorescent lamps/ lights) (Vermont DEC, nd). There are regulations provided for the proper disposal of mercury-containing products through the enactment of Republic Act (RA) No. 6969, otherwise known as the “Toxic Substance, Hazardous, and Nuclear Wastes Control Act”, and the issuance of the DENR Administrative Order No. 2013-22, which provides the procedure for the disposal of hazardous substances in pursuant to the said Act. As such, Hg should be recycled, managed, and disposed of as hazardous waste. Mercury is a naturally occurring element, its existence in the aquatic ecosystem is common and is actually avoidable. However, in the Philippines, its presence in the environment and bodies of water could be due to poor implementation of RA 6969. US-EPA [46] mentioned that once mercury enters the environment it can be converted by microorganisms in the sediments into methylmercury, a highly toxic chemical that builds up in fish, shellfish and animals that consume fish.

The health risk assessment of contaminants, specifically the heavy metals, in humans is based on a mechanistic assumption that such chemicals may either be carcinogenic or non-carcinogenic [47,48]. EDI and PTDI of heavy metal from the consumption of bangus meat and crab aligue by the community showed that there is an increase of heavy metal in the human body when consumed. For instance, in bangus meat from the four municipalities, As had exceeded 50-60 times against its PTDI. As is a known carcinogen. Several studies have shown that the inorganic form of arsenic can cause lung, bladder, liver, kidney, prostate, and skin cancer. There is also evidence that inorganic arsenic may harm pregnant women and their fetuses [49]. In addition, Cr and Cd had also exceeded the normal EDI and PTDI. This means that, in consuming bangus meat, there is a high possibility that risks and possible diseases can be developed overtime where As, Cr, and Cd are the main contributors. But based on the number of times that As had exceeded its PTDI, this makes As the highest contributor for the associated risks [50-52]. On the other hand, in terms of consuming the aligue of crab from the four municipalities, as shown in Table 5, As was also found to have exceeded PTDI by 8-17 times the normal amount. Cr and Cd were also observed to exceed PTDI. As shown in Table 4 and 5, the value of EDI of the concerned heavy metals in crab aligue is lower as compared to the bangus meat. This can be attributed to the fact that, on average, a Filipino consumes 31.4 kg of bangus annually while only 2.89 kg of crab annually, as reported by SEAFDEC [31]. However, the lower consumption of crab aligue compared to bangus meat does not mean that the danger of developing health problems is low. Risk is always there and is determined by the concentration of all HM present in the samples as well as the amount that an individual human body can tolerate [29,53].

For THQ, the estimation of the total potential non-carcinogenic health impacts caused by exposure to a mixture of heavy metals from bangus was calculated using HI, the HI is the sum of THQ for each heavy metals analyzed [30,34]. Based on Table 6, among all the heavy metals, the mean THQ of As from the bangus meat collected from the four municipalities of Northern Samar was 55.84. Consumption of bangus meat will also lead to mean THQ value for Cd, Cr, and Hg of 4.11, 5.06, and 4.21, respectively. The THQ value of Pb is less than 1 indicating that it does not pose risks and that the level of exposure is below the reference dose as well as the daily consumption has a low probability of causing adverse effects during a person’s lifetime [29,53]. Meanwhile, for the THQ values in consuming the crab aligue, only As had a mean THQ value of 12.3, the remaining heavy metals have a THQ value of less than 1 which indicates that As is the highest contributor in the risks in consuming crab aligue. Based also on Table 6 and 7, the HI of bangus meat and crab aligue ranges from 77.14750-63.69276 and 21.14021-10.10484, respectively. Therefore, consuming both bangus meat and crab aligue from the selected fishponds is considered hazardous since the HI had exceeded 1. As mentioned by Sarkar et al. [2], the safe level of HI should be less than 1. The study, however, showed that the HI values exceeded the normal HI values multiple times. As such, continuous consumption of bangus and crabs from the sample sites at a rate of two or more per week in a person's lifetime of 60 years will actually impose risks of developing diseases and adverse health effects. Once a person reaches adulthood, with the values presented from EDI and THQ, we can clearly determine the heavy metal that contributes the most to the development of diseases is As [54-57].

Liu et al*.,* [32] had suggested that TCR greater than 0.00001 is considered unacceptable for a short period of exposure or ingestion of the samples. Moreover, it is noted that the acceptable level of cancer risk for lifetime exposure/ingestion of the samples ranges 0.0001-0 is considered as acceptable [29,32]. As shown in Table 8, all of the value of TCR from heavy metals in bangus meat had exceeded the acceptable limit both for short period of exposure and lifetime exposure, excluding Hg. While TCR presented in Table 9 for crab aligue showed the same, it also exceeded the acceptable limit for short and long periods of exposure or consumption.

For the data analysis, Shapiro-Wilk Test was used to determine the distribution of EDI, THQ, and TCR. Presented in Figs 3, 4, and 5, EDI, THQ, TCR have lower than 0.05 p-values at 2.334e-07, 3.622e-10, and 2.307e-07, respectively. Thus, rejecting the null hypothesis that EDI, THQ, and TCR follows a normal distribution. Since the data did not follow the normal distribution, Spearman's Correlation Test was used. Based on Figs 6 and 7, THQ and TCR have p-values less than 0.05 at 0.001 and 8.5e-05, respectively, which suggests that there is a significant and positive correlation between TCR and THQ and the concentrations/values of heavy metals in bangus meat and crab aligue. For the MANCOVA test (Table 10), the result indicates a significant relationship between the independent variables (heavy metals in sediments, heavy metals, and heavy metals in the samples) as well as the dummy variable for the type of resource and the dependent variables (EDI, THQ, and TCR) at 99% confidence level.

5. CONCLUSION

Arsenic being an established carcinogen has been found to exceed the identified health risks parameters, EDI, THQ, and TCR. This entails that continuous consumption of bangus and crab at a frequency of twice or more a week over the span of 60 years, suggests that it would cause adverse health effects with Arsenic as the highest contributor, with the addition of varying concentrations of Chromium and Cadmium. The high HI for crabs could be attributed to the presence of elevated multi-metals in the sediments that eventually transferred to the crabs through bioaccumulation since crabs are filter feeder/bottom feeder. For the bangus, the high concentration of heavy metals in its body is attributed to the fact that since the sampling site is a fishpond, they are considered as an iliophagous wherein they typically feed on mud present in the shallow waters. In terms of concentrations of Mercury both in bangus, crabs, and sediment, the computed rates are alarming. Mercury, in its methylmercury form, is toxic in the environment system even though the concentrations of Hg did not exceed to the set standards. There should be no Mercury in sediments and in fishes and crustaceans, otherwise, this could be attributed to the poor implementation of laws and issuances.

6. RECOMMENDATIONS

Based on the result of the study we were able to draw recommendations that can help the locality.

1. Mitigating measures should be done by the authorities to protect the health of communities, especially the children, in consuming bangus and crab that is cultured from the identified fishponds.
2. Regular monitoring of heavy metals in cultured fishes and crustaceans should be done.
3. Issuance of regular health and environmental advisories regarding quantitative health and environmental risks associated with bangus and crab consumption coming from the fishponds in the province of Northern Samar that had exceeded the tolerable limit of heavy metals in the cultured fishes and crustaceans. The Department of Health, DENR, Department of Agriculture, local government units, and barangay local government units should be tapped on this.
4. Create a river management committee since most of the fishponds included in the study receives water from a river/freshwater source.

5. Develop and implement an appropriate risk communication program for all stakeholders including, but not limited to fishermen, fish pond owners, farmers, industries, if any, and the general public.
6. Conduct further research on other possible contamination of heavy metals in aquatic organisms aside from crab and bangus.
7. Conduct a follow-up study on quantification of heavy metals in water from fishponds in the selected municipalities of Northern Samar.
8. Conduct a follow-up study on quantification of heavy metals in feeds in feeding the cultured fishes and crustaceans.

ACKNOWLEDGMENT

The authors would like to acknowledge the participation of fishpond owners during the collection of samples. Also, the current head of Integrated Research Laboratory, University of Eastern Philippines (UEP-IRL), Ms. Perth Kennesa P. Ambida. Lastly, the two laboratories who accommodated the quantification of heavy metals, the Mach Union Water Laboratory, Inc. and the UPLB-Nanotech Laboratory specially its Research Staff, Ms. Maris Asuncion L. Bayhon and Mr. Ronaniel A. Almeda.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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