HEALTHRISKSASSESSMENTOFHEAVYMETALCONCENTRATIONIN CULTURED*Chanoschanos*(BANGUS)AND*Scyllaserrata*(MUDCRAB)IN SELECTED MUNICIPALITIES IN NORTHERN SAMAR

ABSTRACT

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 analysisusingEstimatedDailyIntake(EDI),TotalHazardQuotient(THQ),TargetCancer 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 aligi, having 46.83 mg/kg. The consumption of bangus meat may result in an EDI that is greater than PTDI, especiallyforArsenic[15.22731-18.10317μgkg^{-1}BWd^{-1}].Similarly,consumingcrabaligimayalsoresulttoahighEDIforArsenic[2.48197-5.27841μgkg^{-1}BWd^{-1}].THQwasalso evaluatedaswellasthesumofindividualheavymetalvalueswhichistheHazardIndex (HI) that exceeded to 1 multiple times. In terms of TCR levels, all of the heavy metals exceededtheacceptablelimitforcancerrisks.Shapiro-WilkTesthadshownnon-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 aligi as well as EDI, THQ, and TCR. In general, based on the healthrisksassessments(EDI,THQ,andTCR),Arsenic,anestablishedcarcinogen,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:*heavymetals,healthrisks,THQ,TCR,EDI,hazardindex*

INTRODUCTION

The presence of heavy metal concentration in different food sources such as fishes and crustaceanscouldalterthelifeprocessesofanorganismthatconsumesit.Theextentof absorption of these metals in the body is measured by bioaccumulation and bioconcentration.Inthe studydonebyJakimska*etal*(2011),elements suchasMercury (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 (Sarkar *et al.,* 2016). These heavy metals also attack proteins and membrane lipids, therebydisrupt cellular integrity and functions(Mattia *et al.,* 2004; Obrien *et al.,* 2003).

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. Rapid urbanization, improper land use planning and pattern, fast industrial development, and human population explosion are the majoractivities that affect the aquatic ecosystems(Rahman *et al.,* 2012; Rahman *et al.,* 2010). Meanwhile, in terms of the feeds used in the fishponds, tannery and poultry wastesareoftenusedasacheapsourceoffishfeedglobally.Theuseofsuchfeedstocks may possibly, or theoretically, increase the accumulation of toxic contaminants such as heavymetalsinculturedfishandmayposeafoodsafetyriskandhealthrisks(Shamshad *et al.,* 2009)

In aquatic systems, fish samples as well as invertebrate organisms (e.g. mollusks, crustaceans,andetc.,)areobservedasoneoftheindicativeorganismsfortheevaluation of metal pollution (Williams *et al.,* 2022; Erdogrul and Ateş, 2006). Fish accumulates substantial amounts of metals in its tissues, especially in their muscles, thus posing a majorriskforhumansasitisconsideredasoneoftheirvitaldietarysources(Williams *et al.,* 2022; Sarkar *et al*., 2016; Dural *et al*., 2006). In that way, the accumulated heavy metals from fish and crustaceans may enter the food chain (Sarkar *et al.,* 2016). 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 and crabs, in terms of estimated daily intake (EDI), totalhazardquotient(THQ), andtargetcancerrisk (TCR). Inaddition,therelationshipof the levels of heavy metal concentration between bangus and crab 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.

ConceptualFramework

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

The DPSIR framework assesses thechanges inenvironmental quality and itsimpacton 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. In this study, it was found out that four municipalities of Northern Samar have cases of heavy metal concentrationinbangusandcrabs.Thus,thisframeworkisusedtoexaminefurtherwhat 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 unitsinordertomitigateorlessenthehealthriskstothesurroundingcommunities(Figure 1).

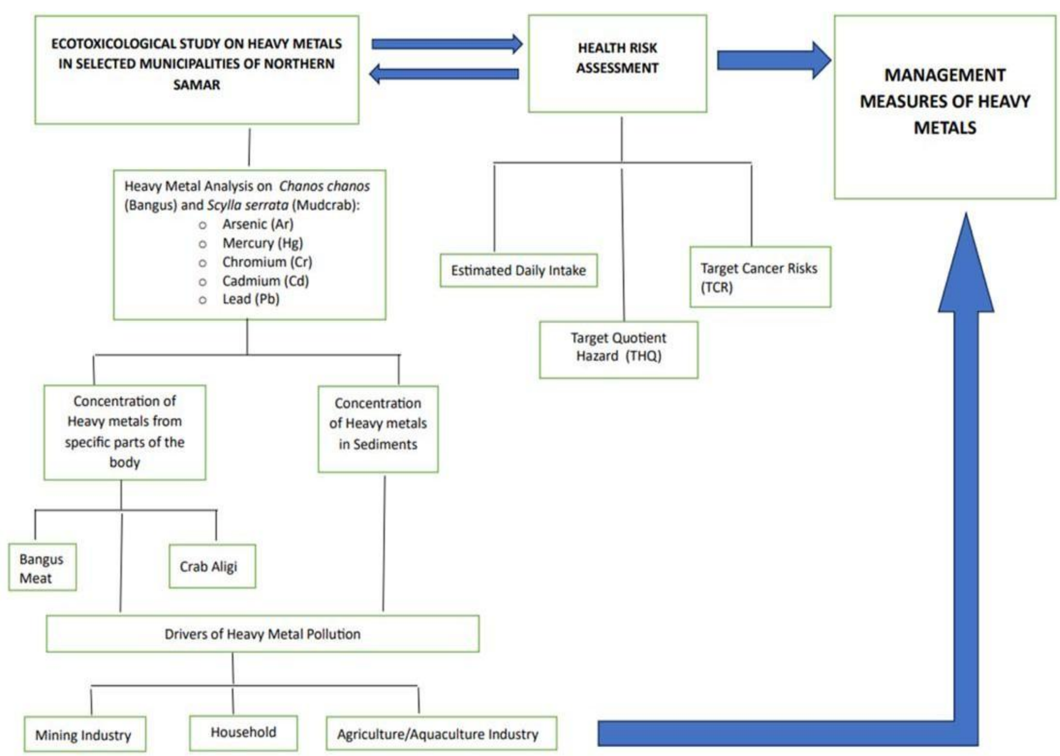

Figure1. ConceptualFrameworkonHeavyMetalsusingDPSIR

METHODOLOGY

CollectionSiteand Sampling

Samplesweregatheredfromthefourfishpondsinfourselectedmunicipalities(Figure2) in the province of Northern Samar.

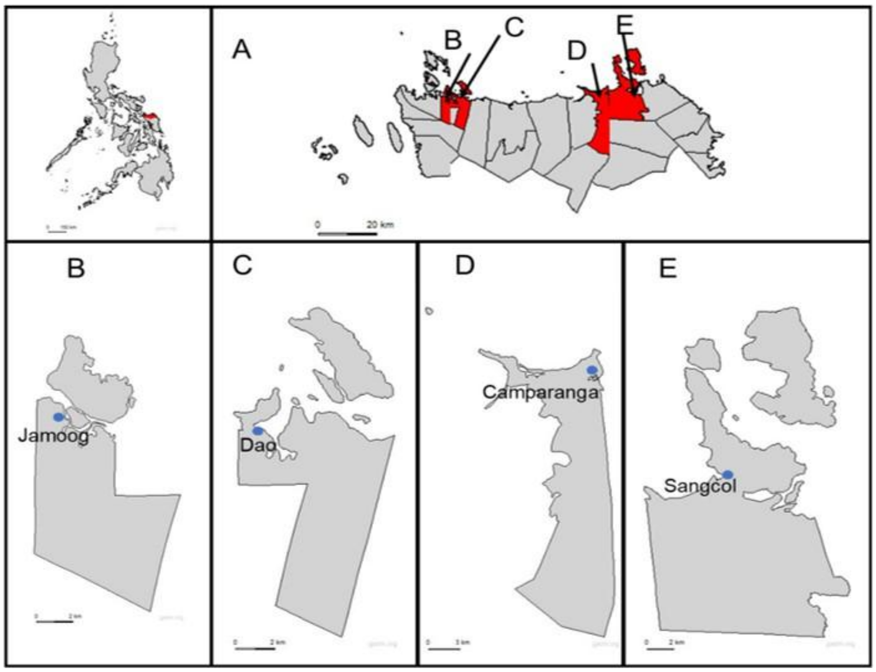

Figure2. Mapofthecollectionsitefortheculturedbangus andcrabs;A).ProvinceofNorthernSamar;B). Barangay Jamoog in the municipality of Rosario; C). Barangay Dao in the municipality of San Jose; D). Barangay Camparanga in the municipality of Pambujan; and E). Barangay Sangcol in the municipality of Laoang (Source: GADM.org)

PreparationofBangusandCrabsamplesaswellasTissuePreparationfor Digestion

Aftergathering the samplesfromthe sampling area, they were broughttothe Collegeof Science,ChemistryLaboratory,UniversityofEasternPhilippines,fortheremovalofmeat andmeatnearthestomachpartwhichweredoneseparately.Thebangusmeatandcrab stomach/aligiweredriedinanovenuntilitreachedcrispnessandwerepulverized,then, it was restrained separately in a clean vial for digestion.

Sample/Tissue Digestion

Before the analysis, samples were subjected to microwave acid digestion. Briefly, dried bangusmeatandcrabstomach/aligiwereweighedtoapproximately0.5gandaddedwith

7mLconcentratedHNO_3and1mL30%H_2O_2. Digestionofeachsamplewasdoneusing Milestone Ethos Up Microwave Digestion System with the following digestion program:

Table1.Digestionprogram

Step	Time	Temperature	MicrowavePower
1	20minutes	27°C-200°C	Upto1800W
2	20minutes	200°C	Upto1800W

QuantificationofHeavyMetals

Digestedsampleswerecooledtoroomtemperatureandfiltered.Then,digestedsolutions were diluted to volume. Atomicemission spectrometricanalyses weredone intriplicates based on EPA 200.7 method using Shimadzu ICP-9000 spectrometer (ICP-OES). Various concentrationsof certified reference standards forAs, Cd, Cr, andPb werealso preparedtoconstructthecalibrationanddeterminedthemethodLODandLOQ.ForHg, theStandardMethodsforWaterandWastewaterservesasreferencewiththeaidofCold VaporAtomicAbsorptionSpectrometer(CV-AAS).ThedigestionandquantificationofAs, Cd, Cr, andPb were doneat the UPLB NanotechnologyLaboratory, while digestionand quantification of Hg were done at Mach Union Laboratory. The results are reported in mg/kg.

HealthRiskAssessment

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.

EstimatedDailyIntake(EDI)

ThecalculationofGriboff*etal*(2017)wasfollowedfortheEDIofheavymetals(Cabahug *et al.,* 2023; Griboff*et al.,* 2017). The main guide for the calculation was based on the study of Cabahug *et al.* (2023).

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

where**C**istheheavymetalconcentrationperspeciesfromeachsite(mgkg^{-1}DW),while **IRd** is the daily average ingestion rate (38.36g d^{-1}orperday forfishes accordingto 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 theaveragebodyweight(BW)ofFilipinoadults(70kg)(Cabahug *etal,*2023;Tayone*et a*l. 2020). EDI is measured by µg/kg BW/d.

TargetQuotientHazard (THQ)

The calculation of Cabahug *et al* (2023) and Tayone*et al* (2020) on THQ were adopted inthisstudy.TheTHQistheratioofhazardouselementexposuretothereferencedose. 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.TheTHQcanbeestimatedusingtheUS-EPAformula,asdemonstrated by Tayone*et al* (2020):

$$THQ_{non\text{-}carcinogenic}=(EF\times ED\times Ird\times C)/[(RfD\times BW\times AT)]$$

where $THQ_{non\text{-}carcinogenic}$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 (SEAFDEC), 2023), **C** is theheavyconcentrationinaquaticproducts(mgkg^{-1}DW),**RfD**istheoralreferencedose valuesbasedfromLiu*etal*(2019),**BW**istheaverageBW(70kg),and**AT**istheaverage lifetime exposure (EF x ED).

TargetCancerRisks(TCR)

The TCR isatooltodeterminethe riskofcancerasa resultofexposuretocarcinogenic chemicals or materials. In this case, carcinogens are being eaten by the bangus and crabswhicharethetopculturedseafoods.Anoralslopefactorisusedinsteadofanoral 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* (2023):

$$TCR=(EF\times ED\times Ird\times C\times CPSo)/[(BW\times AT)]\times 10E\text{^}(-3)$$

where**EF**istheexposurefrequencyof104days(twiceaweek)exposuretotheelement, **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 (2018), **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 $kg^{-1}d^{-1}$(Liu *et al* 2019)0.38 forCd, Hg was not includedsince it has no CPSOoris unable to cause cancer. **BW** is the estimated BW of 70 kg, **AT** is the average exposure time to the carcinogen(EFxEDor104d*60yr),and10–3istheunitconversionfactor(Antoine *et al* 2017).

DataAnalysis

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-WilkTestwasusedtodeterminethenormalityoftheEDI,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 aligi and bangus meat. Data analysis was done on R.

RESULTS

ConcentrationofHeavyMetalsinSediments

Table2showsthedataheavymetalconcentrationinthesedimentsofselectedfishponds. Thequantificationofheavymetalsinthesedimentsoffishpondswerealsodeterminedto trace if the crabs have possibly acquired the heavy metals in sediments due to their feeding habit. Based on Table 2 and 3, results clearly showed that both bangus and mudcrabs contain the concerned heavy metals.

Table2.QuantityofHeavyMetalsinSedimentinNorthernSamarMunicipalities

HeavyMetals	H avyMetalC ncentration in Sedimen				Standard Limit
	Rosario	SanJose	Pambujan	Laoang	
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.*StandardlimitbasedontheWHO(1993).
*Heavymetalsthatexceededthelimit.

HeavyMetalsConcentrationfromtheSpecificPartsofBangusandMudcrab

Table3 showsthequantityofheavymetals accumulated inthedifferentpartsofbangus 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 aligi of crabs, whereas there is no significant variation observed in the concentration of Hg in bangus and crab.

Table 3. Quantity of Heavy Metals Accumulated in Cultured *Chanoschanos*(Bangus meat) and *Scylla serrata* (Crab aligi).

HeavyMetals	He vyMetalCo ncentrationir BangusMe at				Standard Limit
	Rosario	SanJose	Pambujan	Laoang	
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
	HeavyMetalConcentrationinCrabStomach/AligiMeat				
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.*StandardlimitbasedontheWHO,theFAO,andtheUS-EPA.

*Heavymetalsthatexceededthelimit.

Amongallheavymetals,Asin crabaligi hadthehighest concentration with46.83mg/kg or ppm from Pambujan.

SourcesofHeavyMetalPollution

Based on the ocular observation of study sites, there are some buildings/houses, small-scalepiggeries,dumpsitesinthevicinityorjustafewmetersfromthefishpondstowhere the samples are collected. Cheng *et al* (2013) 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 anthropogenicactivities such aselectronics, agriculture, and metallurgy (Cheng *et al.,* 2013). However, the nearest cause of contamination in fishpondscouldbefromagriculturesinceMcNelly(2022)recountedthatawiderangeof fertilizerscontainselevatedamountsof**As**andotherheavymetalslike**Cd**and**Pb**.Itwas 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 environmentcanbeduetobothnaturalandanthropogenicactivities(Cheng*etal.,*2013). Moreover,theCentersforDiseaseControlandPrevention(2017)hadsuggestedthatthe entry of **Cd** in the bodies of water can be due to action of wind and rain (e.g. surface runoff).

Contraryto**As**,**Cd**wasfoundconcentratedonthesurfacewaters(daSilvaandMartinez, 2014). Fish and crabs accumulate **Cd** in the waters (Luo *et al.,* 2020) 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* (2021), rivers are contaminated with hazardous chemicals such as: pesticides, trace metals, and effluents from houses. Mannzhi*et al* (2021) also recounts thatqualityofwateraswellasthefeedshaveanimpactontheculturedorganismsinthe fishponds.Amongallheavymetalsincludedinthestudy,**Cr**istheonethatdidnotexceed to the tolerable levels set by FAO and WHO. Main source of **Cr** in sediments could be suspected from leaching from chromite mining sites (Koleli and Demir, 2016) in other townsinSamarIslandortherecouldbeapossibilitythatsedimentsinselectedsampling sites have high chromite and chromium reserves.

Pb is a nonessential element that is not needed by most organisms. **Pb** are introduced throughdischargeofwastewaterfromindustriesandanthropogenicactivities(Wiriawan*etal.,*2017).ThefishpondinSanJosehashousesnearbyandalsosomerearlivestocks, which might contributeto the fluctuationof **Pb**in thesampled fishpond.According to the UniversityofToledo(2023),mainsourcesof**Pb**contaminationinbodiesofwatercanbe 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** isanotherheavymetal thatdidnotexceedthetolerablelimit setby 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 microorganismsintomethylmercury,ahighlytoxicchemicalthatbuildsupinfish,shellfish and animals that eat fish (US-EPA, 2023).

HealthRiskAssessment

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 (2022),areportbyDOST-FNRIstatedthatindividualFilipinosintheyear1993consumed 36 kg (which accounts for 98.63g/day) of fish. However, in a recent study by SEAFDEC (2023), there is a gradual change in the consumption of fish by Filipino at only 31.4kg (86.03g/d).Thisslightdecreaseinfishconsumptioncanbeattributedtoavarietyoffood 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 systemoreitherinsedimentstowherefishesareexposedwhentheyareinfishpondsis lesser. As such, possible health risks will be assessed through EDI, THQ, and TCR.

a. EstimatedDailyIntake(EDI)

Table 4 and 5 shows the PTDI (μg kg^{-1}BW d^{-1}) and the EDI of heavy metals from consuming bangus and crabs.Results show that consumption of bangus meat brought theEDIlevelofAsandCrabovethePTDIat18.10317μgkg^{-1}BWd^{-1}and16.05074μg kg^{-1} BW d^{-1}, respectively. The rest of the heavy metals, meanwhile, are below the PTDI.

Table4. Estimateddailyintake(adult)inmgkg–1bodyweightd–1(Bangus)

HeavyMetals	EstimatedDailyIntake BangusMeat				
	Rosario	SanJose	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]provisionaltolerabledailyintake(Liuetal.2019)
**HeavymetalsthatexceededPTDI*

Table 5, on the other hand, shows the EDI of the crab aligi. Similar to the results of the crab,AsandCRalsoexceededthePTDIat5.27841μgkg^{-1}BWd^{-1}and3.47949μgkg^{-1} BW d^{-1}. The other elements also have lower EDI than the PTDI.

Table5. Estimateddailyintake(adult)inμgkg–1bodyweightd–1(Crabs)

HeavyMetals	EstimatedDailyIntake CrabStomach/Aligi				
	Rosario	**SanJose**	**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]provisionaltolerabledailyintake(Liuetal.2019)
**HeavymetalsthatexceededPTDI*

b. **TargetQuotientHazard(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 from60.34180-50.75593,5.35006-4.53895,7.31230-0.89714,and4.30135-3.93266, respectively.ItshouldbenotedthattheTHQofPbisnotconsideredsincetheamountof bangusmeatconsumedislessthan1.Thehazardindex(HI)ofheavymetalperbangus ranged from 77.14750-63.69276.

Table6. EstimatedTHQandHIduetotwiceadayconsumptionofbangusmeatfromthe selected municipalities of Northern Samar.

HeavyMetals	TotalQuotientHazard(THQ) BangusMeat				
	Rosario	**SanJose**	**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
HazardIndex	63.69276*	68.40259*	77.14750*	69.54493*	>1

Notes.[PTDI]provisionaltolerabledailyintake(Liuetal.2019) Total HI Index should be > 1 (Sarkar et al., 2016)
**HeavymetalsthatexceededPTDI*

On the other hand, the THQ (Table 7) of As and Cd from consuming crab aligi ranged from17.59470-8.27323and1.59378-1.44725,respectively.WhereastheTHQofCr,Pb, and Hg are below its PTDI with some being less than 1. The HI of heavy metal per crab aligirangedfrom21.14021-16.39129whichislowerascomparedtoHIofbangusmeat.

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

HeavyMetals	TotalQuotientHazard(THQ) CrabStomach/Aligi				
	Rosario	SanJose	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
HazardIndex	11.77343*	16.39129*	21.14021*	10.10484*	>1

Notes.[PTDI]provisionaltolerabledailyintake(Liuetal.2019) Total HI Index should be > 1 (Sarkar et al., 2016)
**HeavymetalsthatexceededPTDI*

c. ***TargetCancerRisks*(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,duetoitsinabilitytocausecancer.Bethatasitmay,mercuryintheenvironment has hazardous and toxic effects once it is converted into methylmercury.

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

HeavyMetals	TotalCancerRisk BangusMeat			
	Rosario	SanJose	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]referencevalues(Antoineetal.2017):“unacceptable”ifgreaterthan0.0001aftershortperiod of exposure; “acceptable” if lesser than 0.000001; “acceptable for lifetime” if 0.0001–0.000001*

The Total Cancer Risk (TCR) due to consumption of crab aligi by adults from the four selectedmunicipalitiesinNorthernSamarwaspresentedinTable9.Theconsumptionof crabaligishowedthattheTCRforAs,Cd,Cr,andPbranged0.02375-0.01117,0.00182-0.00012, 0.00522-0.00393, and 0.00067-0.00000, respectively.

Table9.EstimatedTargetCancerRiskforadultsduetotwiceadayconsumptionofcrab stomach/aligi from selected municipalities of Northern Samar.

HeavyMetals	TotalCancerRisk CrabStomach/Aligi			
	Rosario	SanJose	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]referencevalues(Antoineetal.2017):“unacceptable”ifgreaterthan0.0001aftershortperiod of exposure; “acceptable” if lesser than 0.000001; “acceptable for lifetime” if 0.0001–0.000001*

StatisticalAnalysis

Shapiro-Wilk Test was used to check for the normality of EDI, THQ, and TCR. The Shapiro-Wilk test statistic for the variable EDI (Figure 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.

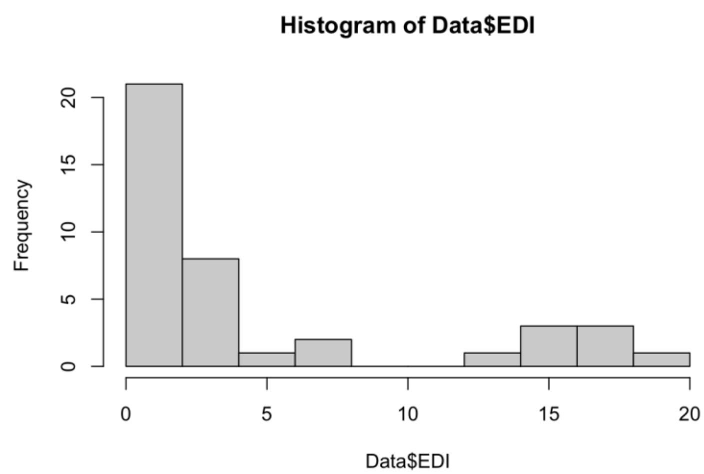

Figure3.Shapiro-WilkTestfornormalityof EDI

THQ (Figure 4), meanwhile, has a Shapiro-Wilk test statistic equals to 0.52741 with a p-value equals $3.622e^{-10}$thus, also rejecting thenullhypothesis that THQ follows a normal distribution.BelowisahistogramofTHQthatsupportsthenon-normaldistributionofthe data.

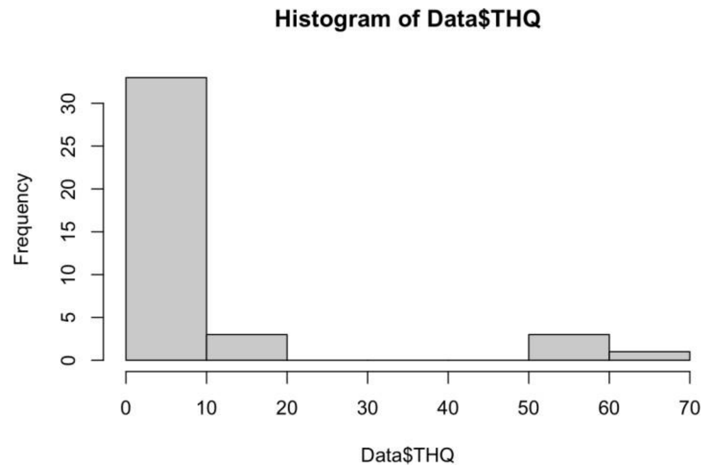

Figure4.Shapiro-WilkTestfornormalityofTHQ

Lastly,theShapiro-Wilktest(Figure5)statisticforthevariableTCRhasa valueequalto 0.65996withap-valueof2.307e^{-07}.Thisalsomeansthatwewillrejectthenullhypothesis thatthedatafollowsanormaldistribution.BelowisahistogramofTCRthatsupportsthe non-normal distribution of the data.

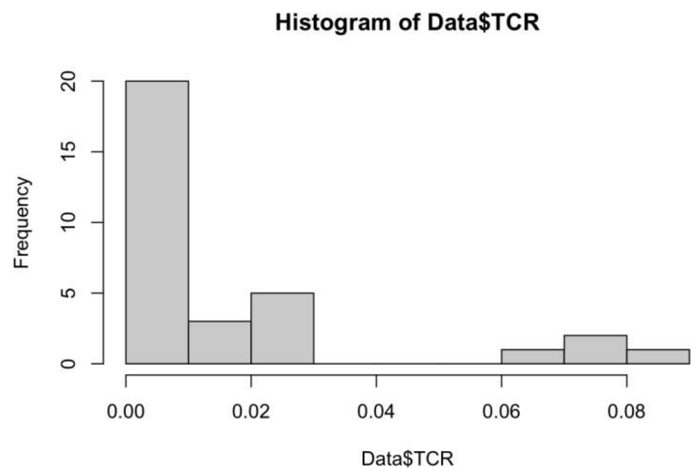

Figure5.Shapiro-WilkTestfornormalityofTCR

MANCOVAResults

MANCOVA (Table 10)wasdonewithEDI, THQ, and TCRasdependent variables while heavy metals in sedimentl, heavymetals in crab aligi and bangus meat, and the dummy variable for heavy metals as independent variables. Moreover, we included the dummy variables for the type of resourceand their location as covariates. Results show that the independent variables and the type of resource is significant at 99% confidence level.

Table10.MANCOVAResults

	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					

CorrelationResult(Spearman'srankcorrelationcoefficient)

Spearman'scorrelationisatesttomeasurethestrengthofrelationshipofdata.Itisused when the data does not follow normal distribution.

ResultsonTHQandtheheavymetalsfoundintheresourcesshowedacoefficientof 0.496 indicating a positive correlation. The correlation is also deemed significant due to its p-value of 0.001.

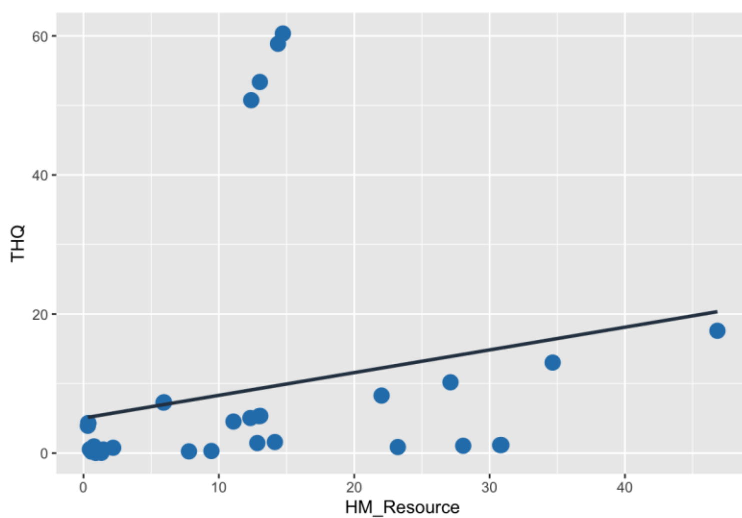

Figure6.SpearmanCorrelationstestforTHQ

SimilarlytoTHQ,TCRisalsopositivelycorrelatedtoheavymetalsfoundintheresources by 0.638. It is also deemed significant with a p-value of $8.5e^{-05}$.

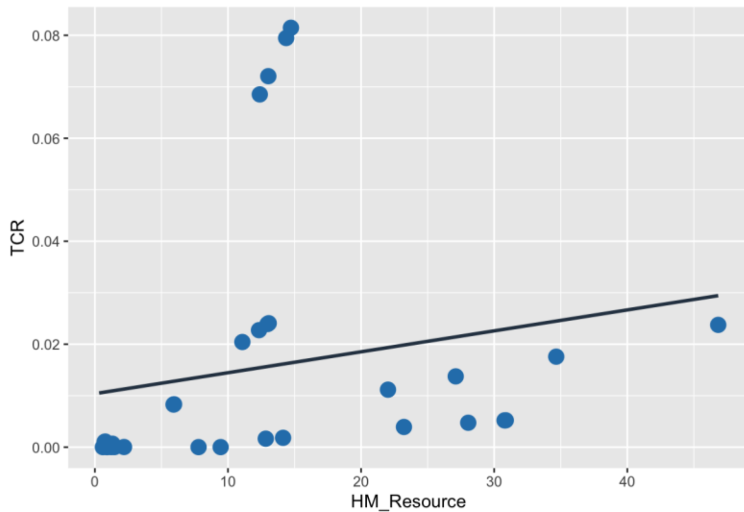

Figure7.SpearmanCorrelationstestforTCR

DISCUSSION

Thepresenceofheavymetalsinaquaticsystemsoriginatesfromthenaturalinteractions betweenthewater,sediments,and atmosphere(Aledesanmi*etal.,*2016).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 interactionswhichresultedinhighconcentrationsofheavymetals insediments,cultured bangus,andcrabs.Thevaryingconcentrationsofheavymetalsinbangusmeatandcrab aligi/stomach can be attributed to their feeding habits. Bangus, being apelagicfish, 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 (FAO, 2024). On the other hand, crab is a bottom feederwheretheydependontheorganicmaterialsthatsinkdowninthesedimentsand, inotherway,theyalsofiltermicroparticlesinthebottomofthebodiesofwater.Compared with bangusmeat, theheavymetal suchasAs and Crin crabaligi is higher. There isno doubtthatthereisapositivecorrelationonthehigherheavymetalsinsedimentsandthe higherheavymetalconcentrationincrabsaspresentedinTable2.Heavymetalsuchas As is much concentrated in sediments (Cheng *et al.,* 2013). In addition, high concentrations of Fe, Cu, Mn, Cr, Zn and Pb are also recorded in bottom sediments as reported by Aledesanmi*et al.,* 2016.

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 asthe “ToxicSubstance,Hazardous,andNuclearWastesControlAct”,andtheissuanceofthe 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,anddisposedofashazardouswaste.Mercuryisanaturallyoccurringelement, itsexistencein theaquaticecosystem iscommonandisactuallyavoidable. However,in thePhilippines,itspresenceintheenvironmentandbodiesofwatercouldbeduetopoor implementation of RA 6969. US-EPA (2023) mentioned that once mercury enters the environmentitcanbeconvertedbymicroorganismsinthesedimentsintomethylmercury, a highly toxic chemical that builds up in fish, shellfish and animals that consume fish.

The health riskassessment of contaminants,specificallytheheavymetals,inhumansis based on a mechanistic assumption that such chemicals may either be carcinogenic or non-carcinogenic (Gnonsoro*et al.,* 2022; Dorne *et al.,* 2011). EDI and PTDI of heavy metalfromtheconsumptionofbangusmeatandcrabaligibythecommunityshowedthat thereisanincreaseofheavymetalinthehumanbodywhenconsumed.Forinstance,in bangusmeatfromthefourmunicipalities,Ashadexceeded50-60timesagainstitsPTDI. Asisaknowncarcinogen.Severalstudieshaveshownthattheinorganicformofarsenic can cause lung, bladder, liver, kidney, prostate, and skin cancer. There is also evidence that inorganic arsenic may harm pregnant women and their fetuses (Fondriest Environmental,Inc.,2019).Inaddition,CrandCdhadalsoexceededthenormalEDIand 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.ButbasedonthenumberoftimesthatAshadexceededitsPTDI,thismakes As the highest contributor for the associated risks. On the other hand, in terms of consumingthealigiofcrabfromthefourmunicipalities,asshowninTable5,Aswasalso found to have exceeded PTDI by 8-17 times the normal amount. Cr and Cd were also observedtoexceedPTDI.Asshownin Table4and5,thevalueofEDIoftheconcerned heavymetalsincrabaligiislowerascomparedtothebangusmeat.Thiscanbeattributed to the fact that, on average, aFilipino consumes 31.4 kg of bangus annually while only 2.89 kg of crab annually, as reported by SEAFDEC (2023). However, the lower consumption of crab aligi 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 (Cabahug *et al.,* 2023; Keshavarzi *et al.,* 2018).

ForTHQ,theestimationofthetotalpotentialnon-carcinogenichealthimpactscausedby exposuretoamixtureofheavymetalsfrombanguswascalculatedusingHI,theHIisthe sum of THQ for each heavy metals analyzed (Gnonsoro*et al.,* 2022; US-EPA, 1986). 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 (Cabahug *et al*., 2023; Keshavarzi *et al*., 2018). Meanwhile, for the THQ values in consuming the crab aligi, only As had a mean THQ value of 12.3, the remaining heavy metalshaveaTHQvalueoflessthan1whichindicatesthatAsisthehighestcontributor intherisksinconsumingcrabaligi.Basedalsoon Table6 and 7,theHIofbangusmeat and crab aligi ranges from 77.14750-63.69276 and 21.14021-10.10484, respectively. Therefore, consuming both bangus meat and crab aligi from the selected fishponds is consideredhazardoussincetheHIhadexceeded1.AsmentionedbySarkar*etal*(2016), thesafelevelofHIshouldbelessthan1.Thestudy,however,showedthattheHIvalues exceeded the normal HI values multiple times. As such, continuous consumption of bangus and crabs from the sample sites at a rate of two ormore per week in a person's lifetime of60 yearswill actually impose risks of developingdiseases and adverse health effects.Onceapersonreachesadulthood,withthevaluespresentedfromEDIandTHQ, we can clearly determine the heavy metal that contributes the most to the development of diseases is As.

Liu *et al.,* (2019) 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 (Cabahug *et al*., 2023; Liu *et al.,* 2019). 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,excludingHg.WhileTCRpresentedinTable9forcrabaligishowedthesame, 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 Figures 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 followthe normaldistribution, Spearman'sCorrelation Test was used. Based on Figures 6 and 7, THQ and TCR have p-values less than 0.05 at 0.001 and $8.5e^{-05}$, respectively, whichsuggeststhatthereisasignificantandpositivecorrelationbetweenTCRandTHQ and the concentrations/values of heavy metals in bangus meat and crab aligi. 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.

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 arefilterfeeder/bottomfeeder.Forthebangus,thehighconcentrationofheavymetalsin 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.

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 heavymetals in cultured fishesand 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. Createarivermanagementcommitteesincemostofthefishpondsincludedinthe 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. Conductafollow-upstudyonquantificationofheavymetalsinfeedsinfeedingthe cultured fishes and crustaceans.

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