

Original Article

Heavy metal concentration in the muscle tissues of selected commercially important fishes and health risk assessment in Tubay, Agusan del Norte, Philippines

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Abstract: Caraga Region is a hub for nickel-iron mining operations in Tubay, Agusan del Norte. Two notable mining companies are currently operating—San Roque Metal Inc. (SRMI) and Agata Mining Venture Inc. (AMVI). Three heavy metals, namely mercury (Hg), lead (Pb), and chromium (Cr), were analyzed from the muscle tissues of commercially important fishes, and a health risk assessment for human consumers was conducted. Heavy metal concentrations were determined using cold vapor atomic absorption spectrometry (AAS) for Hg and Flame AAS for Pb and Cr. Levels of heavy metals at various stations fall within FAO's approved maximum permitted limits, except for *Cheilinus trilobatus*, which exhibits elevated levels of Pb with a concentration of 6.21 mg/kg fresh weight in Lawigan. The highest fish consumption was observed in Binuangan (2.70 g/person/day), while the lowest was in La Fraternidad (1.15 g/person/day). Health risk indicators for environmental risk assessment revealed potential risk values surpassing the reference dose. The estimated daily intake (EWI) surpassed acceptable levels in *C. trilobatus*, while the Target Hazard Quotient (THQ) for Cr and the Total THQ exceeded one for most fish species. These findings underscore the critical need for ongoing research to thoroughly investigate and monitor fishery commodities, including water and sediments, ultimately safeguarding the people's welfare, considering the active operation of mining companies in the area.

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Introduction

Heavy metals include both essential and non-essential elements. Essential metals, such as copper, zinc, cobalt, chromium, manganese, and iron, play important biological roles, while non-essential metals like barium, aluminum, lithium, and zirconium have no known biological function. Some metals, such as tin and arsenic, are considered less toxic, while mercury, cadmium, and lead are highly toxic (Duffus, 2002). Despite advances in ecological waste management, heavy metal pollution, particularly in aquatic environments, remains a pressing issue for researchers, as these discharges continue to adversely affect aquatic organisms (Bakshi and Panagri, 2018). Gonzales and Armenta (2008) noted that mercury (Hg), cadmium (Cd), lead (Pb), and arsenic (As) are frequently used in various industrial and agricultural applications. However, their presence in the

environment poses significant health risks to humans due to their toxic properties, often leading to bioaccumulation. Ansari et al. (2004) identified lead, cadmium, zinc, copper, manganese, iron, mercury, arsenic, and barium as commonly studied heavy metals, as their concentrations are measurable in marine samples. Due to the relatively high concentrations of heavy metals in aquatic environments, fish tend to absorb these (Hao et al., 2013; Elkady et al., 2015). According to Tchounwou et al. (2012), environmental contamination and human exposure to these metals largely result from anthropogenic activities, including mining and smelting operations, industrial production and usage, and domestic and agricultural applications of metal-containing compounds.

The Caraga Region is rich in mineral deposits, notably copper, chromite, and coal, with coal

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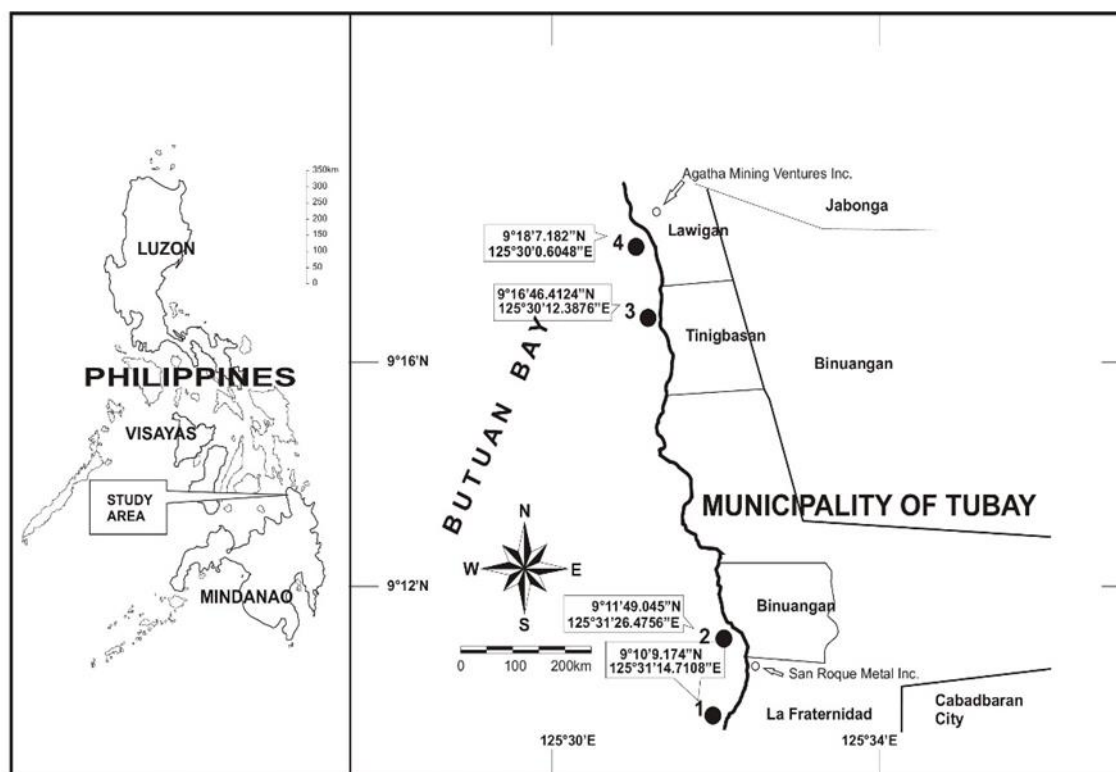


Figure 1. The sampling sites: 1. Brgy La Fraternidad, 2. Binuangan, 3. Tinigbasan, and 4. Lawigan Tubay Agusan del Norte; The locations of Agata Mining Ventures Inc and San Roque Metals Inc. are also shown on the map (with arrows).

representing most of the Philippines' lignite reserves located in three of the region's four provinces: Agusan del Sur, Surigao del Norte, and Surigao del Sur (Cuadrado et al., 2016). In Agusan del Norte, mining activities commonly focus on extracting nickel, iron, and other minerals (Cabuga et al., 2017; Sarmiento et al., 2023; Capilitan et al., 2023). Studies have documented heavy metal pollution in the Agusan River, Butuan City, Agusan del Norte, where fish muscle tissues in species like *Mesopristes cancellatus* (Cabuga et al., 2016) and *Mugil cephalus* (Cabuga et al., 2017) show levels of copper, zinc, lead, nickel, chromium, and mercury exceeding international safety standards. The decline in fish catch, likely resulting from mining operations and other harmful human activities, is also a notable concern (Presilda et al., 2016). Monitoring heavy metal exposure in fish muscle tissue is essential, as Cabuga et al. (2020) underscored, since fish, being at the top of the marine food chain, bioaccumulate metals from sources such as food, water, and sediment. This accumulation poses significant health risks to consumers, potentially causing renal failure, liver damage, cardiovascular

diseases, and even death (El-Moselhy et al., 2014).

While fish constitute a significant portion of our diet, there is currently a scarcity of available data determining heavy metal concentrations in the muscle tissues of commercially important fish species. Therefore, we determined the concentrations of mercury (Hg), lead (Pb), and chromium (Cr) in the muscle tissues of some commercially important fish species from four selected sampling stations in Tubay, Agusan del Norte. Additionally, we also evaluated the potential human health risks associated with heavy metal exposure through fish consumption.

Materials and Methods

Sampling area: The study focused on the evaluation of heavy metal concentrations in the muscle tissues of commercially important fish and the assessment of potential health risks in four selected barangays in Tubay, Agusan del Norte: La Fraternidad ($9^{\circ}10'9.174''\text{N}$, $125^{\circ}31'14.7108''\text{E}$): Binuangan ($9^{\circ}14'49.045''\text{N}$, $125^{\circ}31'26.4756''\text{E}$); Tinigbasan ($9^{\circ}16'46.4124''\text{N}$, $125^{\circ}30'12.3876''\text{E}$) and Lawigan ($9^{\circ}18'7.182''\text{N}$, $125^{\circ}30'0.6048''\text{E}$) (Fig. 1) located in

the Caraga Region, a part of the Mindanao group of islands and at the southern part of the Philippine archipelago. Near the sampling sites I, Tubay, Agusan del Norte are two mining companies: the Agata Mining Ventures Inc. (AMVI), and San Roque Metals Inc. (SRMI). AMVI is situated at Barangay Lawigan, a neighboring Barangay of Tinigbasan, while SRMI is situated in La Fraternidad, the neighboring Barangay of Binuangan.

Sample collection: A total of eleven fish samples, comprised of eight species, were collected, including *Upeneus vittatus* (2 sites), *Mugil cephalus*, *Katsuwonus pelamis*, *Lethrinus atkinsoni* (2 sites), *Siganus guttatus* (2 sites), *Carangoides bajad*, *Chlorurus bowersi*, and *Cheilinus trilobatus*, all of which hold significant commercial importance. However, their availability in the area is constrained due to lower catches by fishermen. At each sampling station, two to two-and-a-half kilograms (2-2.5 kg) of each fish species were procured directly from the fishermen at the landing center. Twenty-seven and a half kilograms of fish samples were collected, with two fish species obtained from La Fraternidad and three species each from Binuangan, Tinigbasan, and Lawigan. All samples were carefully packed in polyethylene bags and placed in a bucket with ice to preserve their freshness. Subsequently, the fish species were transported to the laboratory.

Fish morphological identification: Fish species samples were identified using the Field Guide to Coastal Fishes of Palawan by Gonzalez (2013) and FishBase. Each specimen was photographed, weighed in grams, and measured for total body length (TL) in millimeters.

Tissue preparation: Fish species were thoroughly cleaned in the laboratory with distilled water. The muscle tissues of fish species from four stations were analyzed. The pooled samples at each station were prepared by mixing the dissected muscle tissues. About 200 grams of homogenized samples were weighed using a digital scale, placed in labeled zip locks, and put in the bucket with ice. The tissue samples were sent to FAST Laboratories in Cagayan de Oro City for heavy metal analysis. Eleven fish

tissue samples were analyzed, with two replicates for each fish sample. The procedure used to analyze was 3112 B. Cold vapor - AAS to determine the heavy metal concentration in the samples for Mercury (Hg) (AOAC Official Method 971.21) and Flame AAS for Chromium and Lead (AOAC Official Method 999.11).

Survey on fish consumption rates: The fish consumption rates were estimated by surveying 100 households in each of the sampling areas of Tubay, Agusan del Norte (N = 400). The survey questionnaire was adapted from Molina (2014), Patricio and Alima (2010), and Elvira et al. (2021), with some modifications.

Human health risk assessment of fish consumption: To evaluate the possible health risks of heavy metal exposure, the concentrations of heavy metals in the fish flesh and the local consumption rate were utilized for the estimated daily intake of heavy metals (EDI), target hazard quotient (THQ), and total target hazard quotient (TTHQ). These indices were employed in numerous health studies to assess the risk of metal intake from fish consumption (Molina et al., 2014; Kortei et al., 2020).

Estimated daily intake (EDI): Estimated daily intake of heavy metals (EDI) of trace metal was done in mg/kg/day based on Molina (2014) and Kortei et al. (2020) as follows: $EDI = (C \times IR) / BW$, where: C is the heavy metal concentration in fish (mg/kg, wet weight), IR is the estimated daily ingestion rate of the fish (kg/person/day), and BW is the average body weight for Filipino people (65 kg).

Target hazard quotient (THQ): The THQ, the ratio of the exposure dose to the reference dose (RfD), is the risk of non-carcinogenic effects. If it is less than 1, the exposure level is less than the RfD. This points out that daily exposure at this level is not likely to cause conflicting effects during a person's lifetime and vice versa. Value indicator is provided by the target hazard quotient (THQ), used to assess non-carcinogenic health concerns resulting from exposure to hazardous substances based on Kortei et al. (2020) using the formula of $THQ = (EF \times ED \times CR \times C) / (RfD \times BW \times AT) \times 10^{-3}$. According to Liu et al. (2017), the









<p>A</p>  <p>182 g; 254 mm <i>Upeneus vittatus</i> (Forsskal 1775)</p>	<p>B</p>  <p>1017 g; 470 mm <i>Mugil cephalus</i> (Linnaeus, 1758)</p>
<p>C</p>  <p>2,370 g; 510 mm <i>Katsuwonus pelamis</i> (Linnaeus, 1758)</p>	<p>C</p>  <p>249 g; 240 mm <i>Lethrinus atkinsoni</i> (Seale, 1910)</p>
<p>E</p>  <p>445 g; 295 mm <i>Siganus guttatus</i> (Bloch, 1787)</p>	<p>F</p>  <p>292 g; 250 mm <i>Carangoides bajad</i> (Fabricius, 1775)</p>
<p>G</p>  <p>267 g; 246 mm <i>Chlorurus bowersi</i> (Snyder, 1909)</p>	<p>H</p>  <p>309 g; 254 mm <i>Cheilinus trilobatus</i> (Lacepede, 1801)</p>

Figure 2. Fish species collected in Tubay Agusan del Norte (www.fishbase.org). The weight (in grams) and length (in mm) of representative samples were also added in the table.

THQ is classified into the following five categories: no significant risk: $THQ \leq 1$; low risk: $1 < THQ < 9.9$; moderate risk: $10 < THQ < 19.9$; high risk: $20 < THQ < 99$; serious risk: ≥ 100 .

Statistical analysis: The differences in heavy metal concentration (expressed in mg/kg) in fish muscle tissue among different species and across various sampling sites and differences in the rate of human consumption between these sites were analyzed using ANOVA in RStudio v4.3.1. Post-hoc analysis was conducted using Tukey's HSD test. The differences in heavy metal concentration between pelagic and

demersal fishes and between piscivores and non-piscivores were examined using the Mann-Whitney test, conducted in GraphPad Prism 9 for MacOS Version 9.5.0.

Results and Discussions

Heavy metal concentration: Eleven fish samples were collected in the four coastal barangays in Tubay, Agusan del Norte, constituting a total of eight fish species (Fig. 2). Two demersal fish species were found in La Fraternidad: *U. vittatus* and *M. cephalus*; three fish species in Binuangan: one pelagic *K. pelamis* and

Table 1. Approved Maximum permitted limit/safety limit concentrations of heavy metals in fish. Maximum permitted limit/safety limit concentrations of heavy metals in fish.

Heavy metal	Approved Maximum Limit (mg/kg)	Reference
Mercury (Hg)	0.5	FAO, 1983
Lead (Pb)	0.5	FAO, 2001
Chromium (Cr)	12	FAO, 1983

Table 2. Heavy metal (HM) concentrations (mean \pm SD) in the muscle tissues of selected commercially important fishes in the four sampling stations in mg/kg).

Station	Species	Hg	Pb	Cr
La Fraternidad	<i>U. vittatus</i>	0.0947 \pm 0.001	0.0170 \pm 0.014	0.0929 \pm 0.024
	<i>M. cephalus</i>	0.0402 \pm 0.001	0.0238 \pm 0.014	0.0239 \pm 0.024
	<i>K. pelamis</i>	0.2941 \pm 0.001	0.0271 \pm 0.000	0.0653 \pm 0.015
Binuangan	<i>L. atkinsoni</i>	0.1854 \pm 0.001	0.0001 \pm 0.000	0.0446 \pm 0.015
	<i>S. guttatus</i>	0.0194 \pm 0.000	0.0103 \pm 0.005	0.1101 \pm 0.010
	<i>L. atkinsoni</i>	0.4063 \pm 0.001	0.0845 \pm 0.014	0.0377 \pm 0.024
Tinigbasan	<i>U. vittatus</i>	0.0820 \pm 0.001	0.0575 \pm 0.014	0.0722 \pm 0.024
	<i>S. guttatus</i>	0.2491 \pm 0.072	0.0474 \pm 0.029	0.0920 \pm 0.005
	<i>C. bajad</i>	0.3352 \pm 0.043	0.0575 \pm 0.005	0.0722 \pm 0.005
Lawigan	<i>C. bowersi</i>	ND	0.0069 \pm 0.010	0.0618 \pm 0.010
	<i>C. trilobatus</i>	0.0276 \pm 0.001	*6.2084 \pm 0.096	0.0173 \pm 0.014
Mean		0.1576 \pm 0.141	0.59646 \pm 1.817	0.0628 \pm 0.031

*Above the maximum safety limit by FAO

two were demersal: *L. atkinsoni*, and *S. guttatus*; three demersal species in Tinigbasan: *L. atkinsoni*, *U. vittatus*, and *S. guttatus*; and three species in Lawigan: one pelagic species *C. bajad* and two demersal species *C. bowersi* and *C. trilobatus*. Only a few common fish species were collected per sampling station due to the differences in the landed fish catch per barangay.

The approved maximum limit/safety limit concentration of heavy metals in fish set by the Food and Agriculture Organization (FAO) is shown in Table 1. The heavy metal concentrations (mg/kg wet weight) in the muscle tissues of selected commercially important fish species were compared to the approved maximum limits set by the FAO (Table 2). Based on the results, the concentrations of metals detected in fish muscle are ranked in the following order: Pb > Hg > Cr, with mean values of 0.59646 \pm 1.817, 0.1576 \pm 0.141, and 0.0628 \pm 0.031 mg/kg wet weight, respectively (Table 2).

Mercury concentration: The mercury levels observed in fish across the four sampling stations varied, ranging from 0.0194 \pm 0.000 (*S. guttatus*, Binuangan) to 0.4063 \pm 0.001 (*L. atkinsoni*, Tinigbasan) mg/kg. Notably, all recorded values fell within the approved maximum permissible limit for

Hg (0.5 mg/kg) set by the Fisheries Administrative Order (FAO, 1983). In the case of *C. bowersi* from Lawigan, Hg concentration was not detected (ND). In the study of Cabuga et al. (2020), Hg levels in marine fishes such as *Lutjanus malabaricus*, *Nemopterus japonicus*, and *Selar crumenophthalmus* were also below the detection level (BDL), similar to this study. In contrast, other investigations reported measurable Hg amounts, such as in *Mesopristes cancellatus* (0.888 ppm) (Cabuga et al., 2016) and *Johnius borneensis* (2.81 ppm) (Velasco et al., 2016).

Besides mining, potential anthropogenic sources of mercury encompass dental amalgams, fluorescent lights, thermometers, electric switches, batteries, insecticides, disinfectants, rat poisons, and even skin ointments (Perelonia et al., 2017). An investigation into the total mercury concentration in two marine fish species, mackerel (*Scomberomorus* sp.) and snapper (*Lutjanus* sp.), from various Mexican fishing ports highlighted the potential impact of local industrial activities as significant sources of Hg pollution (Ramírez-Islas et al., 2018). Despite prohibiting Hg use, Elvira et al. (2016) emphasized that operators often prefer it due to its affordability and widespread availability in the market. Between species, *U. vittatus* and *M. cephalus* exhibited Hg concentrations of 0.09

and 0.04 mg/kg, respectively, while *L. atkinsoni*, *S. guttatus*, and *U. vittatus* showed concentrations of 0.41, 0.24, and 0.08 mg/kg, respectively. The species of *K. pelamis*, *Auxis thazard*, and *Euthynnus affinis*, with concentrations of 0.500, 0.354, and 0.2444 µg/g, respectively, were within acceptable limits.

Lead concentration: For Pb, the values varied from 0.0001±0.000 (*L. atkinsoni*, Binuangan) to 6.2084±0.096 (*C. trilobatus*, Lawigan). Among the collected fish species, only *C. trilobatus* exceeded the permissible limit for Pb concentration, surpassing the limit of 0.5 mg/kg (or 0.5 ppm). This study aligns with the findings of Cabuga et al. (2017), who observed high lead concentrations in *M. cephalus*, exceeding the recommended safe limits set by FAO 2001 (≤0.5 ppm). Similarly, in the study of Solidum et al. (2013), most of the Pb concentrations in their fish samples were within the allowable limit of 0.5 ppm, except for the meat of *Selar crumenophthalmus*, which exceeded this limit (2.5029 mg/kg). Significantly higher concentrations of Pb were observed in Lawigan (2.0915 mg/kg wet weight) compared to Tinigbasan (0.06313 mg/kg wet weight), which may be attributed to the predominant mining activity in Lawigan (Fig. 4). This finding is consistent with the study by Mercado et al. (2021), where the average concentrations of Pb, Cd, and Cr were highest in fish samples obtained from Los Baños, while lowest in Bay, areas characterized by the presence of factories and manufacturing companies that dispose of high concentrations of heavy metals into the river, posing environmental concerns (De la Cruz et al., 2017). This aligns with the findings of Velasco et al. (2016), suggesting that the location of sampling sites can influence the levels of Hg, Ni, Pb, and Cd in fish muscles and sediments, with anthropogenic activities linked to industrialization being the source of heavy metal pollution based on the study of Dembitsky (2003). Pb naturally occurs in the environment due to anthropogenic activities (WHO, 1985), with mineral extraction and fossil fuel combustion as primary sources (Muzyed, 2011). Juberg (2000) notes that Pb is released into the environment through mining, smelting, and burning petroleum fuels emitted by

vehicles and engines.

Chromium concentration: For chromium, concentrations ranged from 0.0173±0.014 (*C. trilobatus*, Lawigan) to 0.1101±0.010 (*S. guttatus*, Binuangan). Like Hg, all recorded values for chromium fell within the approved maximum permissible limit for mercury (012 mg/kg) set by the Fisheries Administrative Order (FAO). Cabuga (2017) showed that *M. cephalus* chromium concentration was 0.05 ppm, exceeding the recommended safe limits in foods ≤0.01 set by the US EPA, FAO, and WHO. This implies that frequent consumption may result in health problems. Apart from being a necessary metal, chromium seriously threatens marine life. They degrade the ecosystem because of their harmful effects on the biota and the organisms' bioaccumulation (Aslam and Yousafzai, 2017). As a known carcinogen, excessive chromium exposure can cause acidosis, renal failure, acute tubular necrosis, and even death (USEPA, 2009). Naturally, through wastewater released by a variety of industries, including the textile, tannery, electroplating, mining, dyeing, printing photography, and medical sectors, chromium and its particles find their way into the aquatic medium as hexavalent chromium easily crosses cellular membranes and transforms into trivalent form, it is thought to be the most hazardous of these (Bashi and Panigrahi, 2018).

Heavy metal concentration in pelagic vs. demersal and in piscivore vs. non-piscivore fishes: Several factors influence heavy metal concentrations in fish, including feeding habits, species, age, size structure, trophic levels, and the ingestion of organic matter present in the water (WHO, 1990; Kamaruzzaman, 2008). Demersal and pelagic fishes also exhibit different bioaccumulation potentials (George et al., 2022). This study categorized species based on feeding guild (piscivore vs. non-piscivore) and habitat (pelagic vs. demersal). Our results indicate that Hg and Pb concentrations do not differ significantly between non-piscivorous and piscivorous fishes (Fig. 3A-B). However, the levels of chromium in non-piscivores are significantly different compared to piscivorous fishes (Fig. 3C).

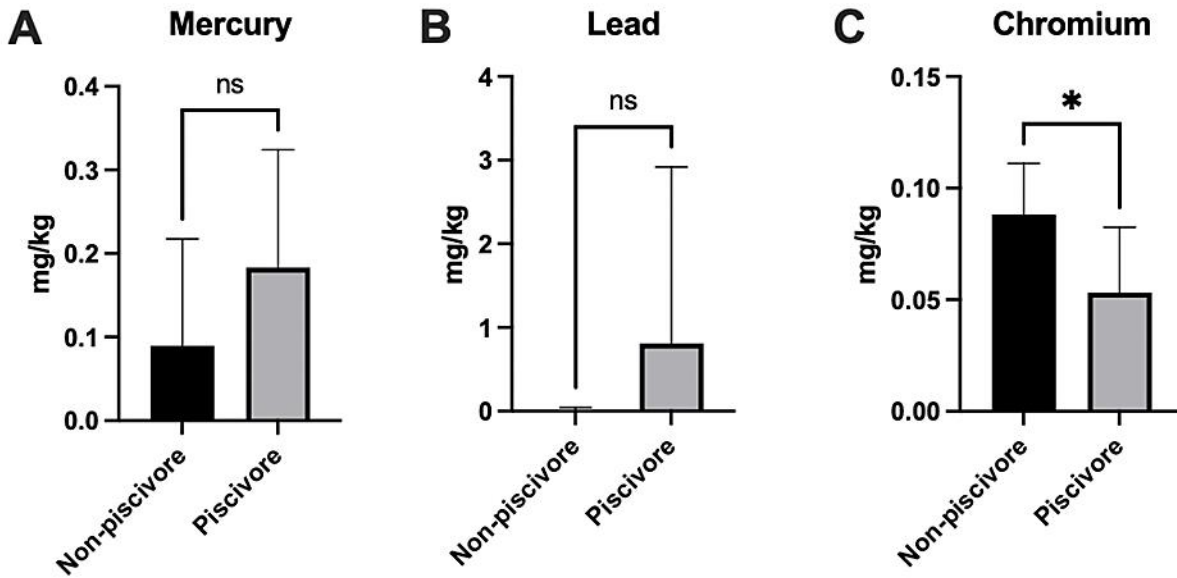


Figure 3. Heavy metal concentration of piscivore and non-piscivores species in four sampling stations of commercially important fishes in Tubay, Agusan del Norte.

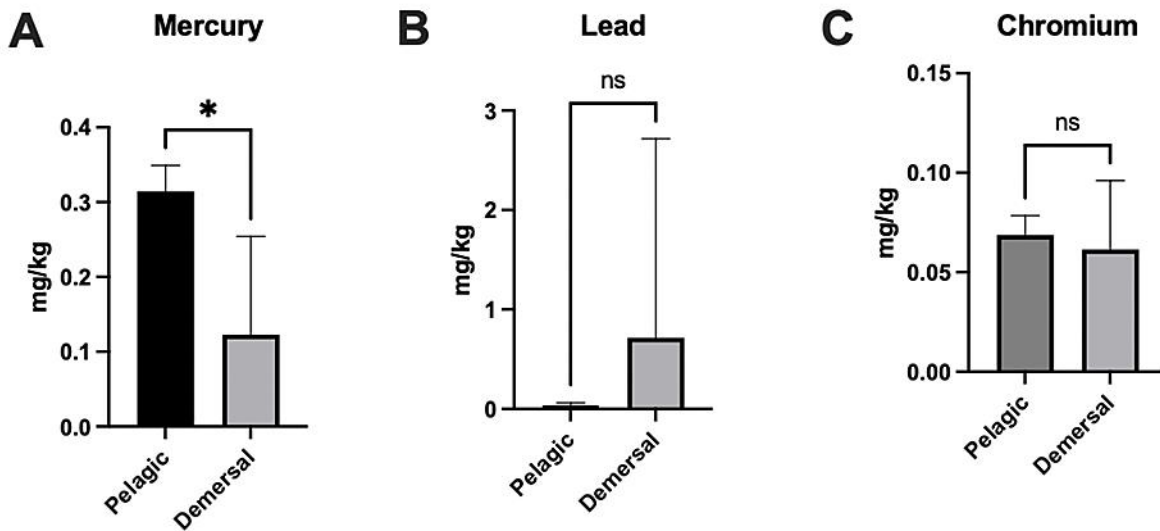


Figure 4. Heavy metal concentrations of pelagic and demersal fish in the four sampling stations of commercially important fishes in Tubay Agusan del Norte.

According to Bawuro et al. (2018), metal buildup differs according to species. In a previous study, metal concentrations were higher in herbivorous fish *Heterotis niloticus* than in carnivorous fish like *Clarias anguillaris*. Malakootian et al. (2016) showed that *Thunnus tonggol* is higher than the other two studied fish species (*Liza klunzingeri* and *Pleuronectiformes*) in average Ni and Cr contents. This is due to feeding patterns, which feed on shrimp, crabs' small fish, and a wider food chain. Furthermore,

non-piscivore species may have higher chromium levels due to environmental exposure, and the quantities would be determined by habitat, food, and local contaminants (Eisler, 1996). Consequently, agricultural and industrial activities in the area of the Helman River may be the reason for the higher Cr (Miri et al., 2017) (Fig. 4).

In the case of habitat (i.e. pelagic vs demersal), Hg concentrations in pelagic fishes were significantly different than in the demersal fishes (Fig. 4). This

Table 3. Socio-demographic survey of fish consumption from four sampling station of Tubay, Agusan del Norte.

Frequency Variable		La Fraternidad	Binuangan	Tinigbasan	Lawigan
		N=100	N=100	N=100	N=100
Age	15-30	28	21	7	14
	31-45	20	36	34	37
	46-60	31	28	41	38
	61 above	21	15	18	11
Gender	Male	69	79	77	72
	Female	31	21	23	28
Civil Status	Single	9	3	4	16
	Married	90	93	96	80
	Widow	1	3	0	4
	Separated	0	1	0	0
Educational Attainment	Elementary Level	23	11	14	38
	Elementary Graduate	14	20	19	26
	High School level	30	38	38	22
	High School graduate	28	27	18	14
	Vocational	1	1	0	0
	College Graduate	4	3	4	0
Years of Stay	1-20	6	2	9	23
	21-40	39	45	37	28
	41-60	35	38	42	44
	61 above	20	15	12	5
Occupation	Farmer	3	3	10	9
	Fisherman	33	57	44	37
	Government employee	3	0	4	22
	Housewife	15	8	15	12
	Laborer	17	18	16	5
	Panday	1	0	0	0
	Vendor	3	10	9	6
	Private worker	11	3	2	9
Income	7k and below	35	47	40	35
	8k and above	65	53	60	65

Table 4. Estimated daily intake (EDI; mg/kg per day) of Hg, Pb, and Cr in commercially important fish in four sampling stations of Tubay Agusan del Norte.

Station	Average No. of Consumers	Consumption per week (g/week)	Average Consumption per day (g/day)	Consumption rate (g/person/day)
La Fraternidad	4.63±1.57	96.48±38.89	19980±7.38	1.15±0.59
Binuangan	4.78±2.10	85.44± 36.47	21100±8087.28	2.70±0.84
Tinigbasan	4.66±1.85	67.2±0.32	18330±7183.11	1.53±0.79
Lawigan	4.48±1.75	85.92±36.61	20130±7390.49	1.439±1.16

study was similar to the findings of Choy et al. (2009), who indicated higher Hg concentrations in the predatory fish and their prey rose with a median depth of occurrence in the water column and mimic concentrations of dissolved organic mercury in seawater for the species *Thunnus obesus*, *T. albacares*, *Katsuwonus pelamis*, *Xiphias gladius*, *Lampris guttatus*, *Coryphaena hippurus*, *Taractichthys steindachneri*, *Tetrapturus audax*, and *Lepidocybium flavobrunneum*, and suggested that the main source for a pathway of Hg into marine food webs was a mesopelagic habitat. Rocha et al. (2015) noticed that

Hg concentrations were the same between pelagic and demersal fish foraging habits, whereas Lavoie et al. (2010) observed stronger biomagnification in the pelagic food web. Azevedo et al. (2019) emphasized that understanding the impact of the foraging environment on Hg levels is especially important for human health and safety, as commercial and subsistence fishing may depend on a specific food web.

Socio-demographic profile of survey respondents on fish consumption rates: The fish consumption rates were estimated by surveying 100 households in

Table 5. Estimated daily intake (EDI; mg/kg per day) of Hg, Pb, and Cr in commercially important fish in four sampling stations of Tubay Agusan del Norte.

Stations	Species	mg/kg per day		
		Hg	Pb	Cr
La Fraternidad	<i>U. vittatus</i>	0.060634	0.013474	0.06063
	<i>M. cephalus</i>	0.026949	0.013474	0.01347
Binuangan	<i>K. pelamis</i>	0.121723	7.60768E-06	0.03043
	<i>L. atkinsoni</i>	0.015215	0.007608	0.08368
	<i>S. guttatus</i>	0.220623	0.022823	0.05325
Tinigbasan	<i>L. atkinsoni</i>	0.396577	0.077381	0.03869
	<i>U. vittatus</i>	0.077381	0.058036	0.06771
	<i>S. guttatus</i>	0.241815	0.048363	0.08705
Lawigan	<i>C. bajad</i>	0.257216	0.04539	0.05296
	<i>C. bowersi</i>	ND	0.00756	0.04539
	<i>C. trilobatus</i>	0.022696	4.69797	0.01513

each sampling area of Tubay, Agusan del Norte (Table 4). The majority of respondents were aged 46-60 in Barangay La Fraternidad (31%), Tinigbasan (41%), and Lawigan (38%), while those aged 31-45 predominated in Barangay Binuangan (36%). Males comprised the majority of respondents in all four sampling stations: Binuangan, Tinigbasan, La Fraternidad, and Lawigan, accounting for 79, 77, 69, and 72% respectively. Most respondents were married. Regarding educational attainment, 38% of respondents from Binuangan and Tinigbasan had completed high school, while those in Lawigan were primarily elementary graduates, and 28% in La Fraternidad had graduated from high school. La Fraternidad and Binuangan had residency durations of 21-40 years (39 and 45%, respectively), while Tinigbasan and Lawigan had residency 347 durations of 41-60 years (42% each).

Most respondents identified as fishermen, with proportions highest in Binuangan, Tinigbasan, Lawigan, and La Fraternidad at 57, 44, 37, and 33% respectively. Other occupations reported included farmers, government employees, housewives, laborers, carpenters, vendors, and private employees. The majority of respondents in all four sampling stations reported an income of P8,000.00 and above (Table 3).

Estimated daily consumption of fish: The estimated daily intake of fish varied significantly across the four sampling stations. Binuangan reported the highest

daily consumption rate of fish flesh at 2.70 g/person/day, whereas La Fraternidad recorded a comparatively lower rate of 1.15 g/person/day. These differences in daily consumption rates were statistically significant ($P < 0.05$).

Survey results indicated that the respondents' moderate consumption rate of each fish species correlated with their preferences for certain species. Some residents opted for alternative fish species due to their higher market value, while others were less available in the area due to overfishing (Patricio and Alima, 2010). Similarly, Laudino et al. (2023) found a connection between the accumulation of heavy metals in the muscles of striped snakehead murrel (*Channa striata*) and the selection of fish species, as some individuals preferred marine fish over freshwater fish for their taste preferences.

Estimated daily intake (EDI): The calculated EDI for three heavy metal concentrations is presented in Table 5. The EDI values of Hg for *S. guttatus* (Binuangan), *L. atkinsoni* (Tinigbasan), *C. bajad* (Lawigan), *K. pelamis* (Binuangan), *S. guttatus* (Tinigbasan), *U. vittatus* (Tinigbasan), *U. vittatus* (La Fraternidad), *L. atkinsoni* (Binuangan), *M. cephalus* (La Fraternidad), *C. trilobatus* (Lawigan), and *C. bowersi* (Lawigan) were ranged 0.01204–ND, respectively. For Pb, EDI for *C. trilobatus* (Lawigan), *L. atkinsoni* (Tinigbasan), *U. vittatus* (Tinigbasan), *C. bajad* (Lawigan), *S. guttatus* (Binuangan), *S. guttatus* (Tinigbasan), *L. atkinsoni* (Binuangan),

Table 6. Estimated weekly intake (EWI) of fish (kg) by a 65-kg adult relative to PTWI criteria (mg kg⁻¹ body weight week⁻¹).

Stations	Species	Hg	Pb	Cr	Hg (1.6)	Pb (0.025)	Cr (0.0233)
La Fraternidad	<i>U. vittatus</i>	0.019084	0.004240923	0.019084	↓	↓	↓
	<i>M. cephalus</i>	0.008482	0.004240923	0.004241	↓	↓	↓
Binuangan	<i>K. pelamis</i>	0.030046	1.87785E-06	0.007511	↓	↓	↓
	<i>L. atkinsoni</i>	0.003756	0.001877846	0.020656	↓	↓	↓
	<i>S. guttatus</i>	0.054458	0.005633538	0.013145	↓	↓	↓
Tinigbasan	<i>L. atkinsoni</i>	0.060554	0.011815385	0.005908	↓	↓	↓
	<i>U. vittatus</i>	0.011815	0.008861538	0.010338	↓	↓	↓
	<i>S. guttatus</i>	0.036923	0.007384615	0.013292	↓	↓	↓
Lawigan	<i>C. bajad</i>	0.064208	0.011330769	0.013219	↓	↓	↓
	<i>C. bowersi</i>	ND	0.001888462	0.011331	ND	↓	↓
	<i>C. trilobatus</i>	0.005665	1.172734615	0.003777	↓	↑	↓

↓ indicates below PTWI; ↑ indicates above PTWI

U. vittatus (La Fraternidad), *Mugil cephalus* (La Fraternidad), *Katsuwonus pelamis* (Binuangan), and *Chlorurus bowersi* (Lawigan) were ranged 0.13662–0.00022 mg/kg per day, respectively. For Cr, EDI for *L. atkinsoni* (Binuangan), *S. guttatus* (Binuangan), *Siganus guttatus* (Tinigbasan), *K. pelamis* (Binuangan), *U. vittatus* (Tinigbasan), *U. vittatus* (La Fraternidad), *C. bajad* (Lawigan), *C. bowersi* (Lawigan), *L. atkinsoni* (Tinigbasan), *C. trilobatus* (Lawigan), and *M. cephalus* (La Fraternidad) were ranged 0.00456–0.00035 mg/kg per day, respectively.

According to Laudiño et al. (2023), the results of EDI of heavy metals will be compared to the value proposed by the United States Environmental Protection Agency (USEPA, 1989) RfD: Hg = 0.0001; Pb = 0.0000001, and Cr = 0.003 mg/kg per day. The EDI value of the three elements ranged from 0–0.0120, 0.0002–0.1366, and 0.0003–0.0045 mg/kg per day means that Hg, Pb, and Cr were above the USEPA Oral Reference Dose (RfD). Molina (2014) emphasized a high risk in some fish species and all sampling stations.

Estimated weekly intake with provisional tolerable weekly intake (PTWI): The summary of the Provisional Tolerable Weekly Intake (PTWI) and permissible weekly fish consumption were calculated based on metal concentrations in fish muscle (Table 7). The PTWI, as defined by the joint FAO/WHO Expert Committee on Food Additives (JECFA), serves as a reference dose indicating a safe weekly

intake of pollutants, according to Miri et al. (2017). Specifically, the PTWI values for mercury (Hg), lead (Pb), and chromium (Cr) were 1.6, 0.025, and 0.233 mg/kg body weight, respectively, as stated in USEPA 2011 guidelines. These values determine the safe weekly fish consumption across the four sampling stations. In this study, the PTWI for Hg, Pb, and Cr ranged from Not Detected (ND) to 0.396577, 7.60768E-06 to 4.697975, and 0.013474 to 0.083684 mg/kg body weight week⁻¹ respectively. The intake of Hg and Cr per kg body weight per week varies depending on the species of fish consumed, all of which fall well below the safety limit of 1.6 mg kg⁻¹ body weight week⁻¹. However, for lead, certain species such as *Lethrinus atkinsoni*, *Upeneus vittatus*, *Siganus guttatus*, *Carangoides bajad*, and *Cheilinus trilobatus* from Lawigan exceeded the PTWI value set by USEPA 2011 (0.025 mg/kg body weight weekly), with concentrations ranging from 0.077381 to 4.697975 mg/kg body weight per week. This indicates a potential health hazard associated with lead intake from these species, 0.058036, 0.048363, and 4.697975 mg/kg body weight week⁻¹, which may pose a health hazard. Also, a comparison between PTWI recommended by USEPA (2011), Murao et al. (2017), and (JECFA/WHO, 2003) and the result of this study to evaluate ingestion-related weekly metal exposure in human results with the mentioned PTWI were compared.

This study was comparable to Zhang et al. (2018),

Table 7. Target hazard quotient (THQ) and the cumulative health risk (TTHQ) on heavy metals consumption of commercially important fish by residents in four Tubay Agusan del Norte sampling stations.

Stations	Species	THQ			Total THQ
		Hg	Pb	Cr	
La Fraternidad	<i>U. vittatus</i>	6.5286E-07	2.2E-05	3.02619	3.02621
	<i>M. cephalus</i>	2.9016E-07	2.2E-05	0.67249	0.67251
Binuangan	<i>K. pelamis</i>	2.808E-06	2.7E-08	3.25397	3.25397
	<i>L. atkinsoni</i>	3.51E-07	2.7E-05	8.94842	8.94844
	<i>S. guttatus</i>	5.0895E-06	8.1E-05	5.69445	5.69453
Tinigbasan	<i>L. atkinsoni</i>	4.0775E-06	0.00012	1.84392	1.84404
	<i>U. vittatus</i>	7.956E-07	9.2E-05	3.22685	3.22695
	<i>S. guttatus</i>	2.4863E-06	7.7E-05	4.14881	4.14889
Lawigan	<i>C. bajad</i>	3.1603E-06	8.6E-05	3.01595	3.01604
	<i>C. bowersi</i>	0	1.4E-05	2.5851	2.58511
	<i>C. trilobatus</i>	2.7885E-07	0.00892	0.8617	0.87062

and overall, for both cultured and wild fish, Pb was recognized as the major contributor to non-carcinogenic risk based on the calculation results of EWI. Therefore, Pb and Cr were selected as the representative trace elements to compare the potential health risk between cultured fish and wild fish and between muscle consumption and mixed edible tissue consumption according to Malakootian et al. (2016) in the study of heavy metals bioaccumulation in fish of southern Iran and risk assessment of fish consumption for EWI, Ni and Pb are higher based on the recommended PTWI. In the case of Hg, however, it is lower.

Target hazard quotient (THQ) of heavy metals in the fish species: The target hazard quotient (THQ) values of heavy metals calculated based on the consumption of fish flesh by residents of Tubay Agusan del Norte in four sampling stations are summarized in Table 7. Exposure doses should identify the toxic potency of these metals. In this regard, this study compares the concentrations of the metals with the estimated daily intake limits as an active tool to evaluate the balance between benefits and risks. According to Adebola et al. (2021), daily intake of heavy metals was estimated based on the concentrations in the fish's muscle samples. THQ values for Hg and Pb were < 1 , suggesting no health risk through fish consumption. On the other hand, both Cr and Total THQ of fishes *U. vittatus* of La Fraternidad, *K. pelamis*, *L. atkinsoni* and *S. guttatus* of Binuangan, *L. atkinsoni*, *U. vittatus*, *S. guttatus* of

Tinigbasan and *C. bajad*, *C. trilobatus* of Lawigan were greater than > 1 indicate there is a potential health risk. It shows that the $THQ > 1$ may pose some health problems. Therefore, this study advises routine heavy metal monitoring of fish to implement the regulatory standard by the government environmental health management agencies. Kortei et al. (2020) pointed out that consumption of fish for a longer period from Ogun River might pose some health problems, suggesting that consumers from the sites experience significant noncarcinogenic health risks and, therefore, must take some caution from the toxic potentials of As, Cd, Ni, and Cr.

Conclusion

The analysis of heavy metal concentrations in the muscle tissues of commercially important fishes from Tubay, Agusan del Norte revealed that levels of mercury (Hg), lead (Pb), and chromium (Cr) were generally within approved permissible limits. However, notable exceptions were observed, particularly in the case of Pb concentrations in *C. trilobatus* from Lawigan, which exceeded limits observed in other fish species. Assessment of health risks involved evaluating both heavy metal concentrations in fish and estimated consumption rates of individuals in the sampling stations. The estimated daily intake exceeded the oral reference dose, indicating potential health risks associated with fish consumption. Moreover, the estimated daily intake (EWI) surpassed acceptable levels in

C. trilobatus, while the Target Hazard Quotient (THQ) for Cr and the Total THQ exceeded one for nine fish species. These findings highlight the potential health risks of consuming fish muscle tissues across the four sampling stations. This underscores the critical need for ongoing research to thoroughly investigate and monitor fishery commodities, including water and sediments, ultimately safeguarding the people's welfare considering mining activities are operating.

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