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Heavy Metal Concentrations in Mollusks and Crustaceans Harvested from Eastern Samar's Taft River in the Philippine and the Health Risks Posed to Consumers

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Heavy metal (HM) contamination from mining activities poses a health risk to humans who consume mollusks and crustaceans from mine-affected environments. One such example is the Taft River, which receives drainage water and sediments from the Bagacay mines in Samar, Philippines. To assess the accumulation of HM in mollusks and crustaceans and the associated risks to the humans, a comprehensive analysis was conducted on representative specimens, which were gathered from different sections of the river. The concentrations of Cu (97.67 mg kg^{-1}), Ni (29.30 mg kg^{-1}), Pb (0.59 mg kg^{-1}), and Sr (14.67 mg kg^{-1}) in the flesh of "bebe" (*Batissa violacea* Lamarck) were significantly higher than those in "tangili" (*Telescopium telescopium* Linn.), "tuway" (*Mercenaria mercenaria* Linn.), and "sihi" (*Nerita albicilla* Linn.); whereas for crustaceans, the flesh of mud crabs (*Scylla serrata*) contained higher concentrations of As (17.67 mg kg^{-1}), Cu (118.67 mg kg^{-1}), Ni (25.79 mg kg^{-1}), Rb (31.33 mg kg^{-1}), Sr (60.67 mg kg^{-1}), and Zn (331.00 mg kg^{-1}) compared with giant freshwater prawns (*Macrobrachium rosenbergii*). The consumption of *T. telescompium*, *N. albicilla*, and *B. violacea* may result in an estimated daily intake (EDI) that is greater than the provisional tolerable daily intake (PTDI) for As [2.03–4.48 μg kg^{-1} body weight (BW) d^{-1}], while all mollusks may have an EDI for Cr of between 124.59–136.23 48 μg kg^{-1} BW d^{-1}. This may subsequently lead to a high target hazard quotient (THQ), high hazard index (HI), and high target cancer risk (TCR) for adults. With respect to crustaceans, the consumption of *S. serrata* exceeds the PTDI index for As. Both *S. serrata* and *M. rosenbergii* exceed the PTDI for Cr by 20–21 times, as well as the elevated health risk values of 22.36 and 28.57 48 μg kg^{-1} d^{-1} for *M. rosenbergii* and *S. serrata*, respectively. Overall, current results indicate that human consumption of the Taft River mollusks and crustaceans may inadvertently lead to an increased intake of As and Cr that could negatively impact human health.

Keywords: freshwater clams, giant freshwater prawns, health risk assessment, mudcrabs

INTRODUCTION

Among aquatic organisms, mollusks and crustaceans – the filter and bottom feeders – appear to bioaccumulate large amounts of both essential and non-essential nutrients from their environment, making them the easiest species to analyze as bioindicators for chemical alterations in rivers and lakes (Liu *et al.* 2019). Mollusks – which contain large concentrations of calcium (Ca), potassium (K), zinc (Zn), iron (Fe), phosphorus (P), and copper (Cu) – are a rich source of minerals for humans. However, the consumption of mollusks – which are harvested from HM contaminated water bodies – could raise safety concerns because of the presence of a wide range of chemical pollutants, particularly HM (Kumar and Weerasooriyagedara

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2018). These species are good indicators of the presence of HM in their environments (Hembrom *et al.* 2020). It is also suggested that these species may be a point of entry for HM contamination into the food web and subsequently into human diets (Montojo *et al.* 2021). Similarly, crustaceans such as mud crabs and shrimp found in contaminated rivers and water bodies have exhibited elevated levels of HM. Examples include high accumulations of Zn, Cu, Cr, Pb, Cd, and Hg in the tissues of mud crabs from the Solo River estuary in Indonesia (Soegianto *et al.* 2022), and high concentrations of Cu, Fe, Pb, and Zn in shrimp tissues from the Iskenderun Gulf in Turkey (Aytekin *et al.* 2019). Mostafiz *et al.* (2020) reported high concentrations of Pb, Cr, Fe, Cu, Zn, and Mn in wild-caught prawn (*Macrobrachium rosenbergii*) and high Ni and Cd concentrations in farmed prawns in Bangladesh. The elevated concentrations of these HM can lead to high estimates of daily intake (EDI) above tolerable daily intake levels (Mostafiz *et al.* 2020).

The concentration of the different substances and the chemical constituents found in the soft tissues of mollusks and crustaceans has been assessed and reported as extensive (Liao *et al.* 2020). However, there is no available information about the HM concentrations in the mollusks and crustaceans from the Taft River (Dar *et al.* 2018). In addition, the information about the chemical constituents in many bivalves and crustaceans is significant since these species become prey to organisms on a higher trophic level (Mostafiz *et al*. 2020). The probable contribution of the mollusk and crustacean species to the biomagnification and bioaccumulation of contaminants – as evidenced in particular by the presence of HM in the food chain – could actually be higher than reported because most of these species, especially mollusks, can tolerate higher levels of HM and tend to accumulate more in their tissues (Yuan *et al.* 2020). The consumption of HM at high concentrations can have disastrous consequences for humans and other living species. For example, arsenic (As), mercury (Hg), cadmium (Cd), and lead (Pb) are naturally present in the environment, but their levels are rising due to anthropogenic activity, posing a concern to aquatic animals and humans (Cambia *et al.* 2019; Kortei *et al.* 2020). When HM and metalloids are present in large quantities, they become toxic to all living things, including those elements that are considered essential for bivalves and crustaceans. The result of humans consuming contaminated food sources is no different. A study by Keshavarzi *et al.* (2018) revealed that even if some metal concentrations in the tissues are within tolerable limits, the consumption of such products containing other metals will have a combined impact, resulting in a higher hazard index (HI). Since a single organism can accumulate multiple HM, the consumption of the whole will result in a higher potential health risk.

This study investigated the long-term impact of the Bagacay Mine operation on heavy accumulations in mollusks and crustaceans from the Taft River. This copper mine – which operated from 1956–1985 – is in the Municipality of Hinabangan, Western Samar, Philippines. The mineral extraction done in the Bagacay Mines over many years has deposited into the Taft River basin large quantities of sediment and water, which contain HM. The unplanned closure of the Bagacay Mines operation resulted in an abandoned area being left in an unrehabilitated state (Samaniego *et al.* 2020). Although government laws and regulations require mine operators to ensure pollution control and rehabilitation upon closure, these measures were not fully or properly implemented which worsened the pollution, especially in the off-site areas (DENR-EMB 2007). Taft municipality residents believe the Taft River has been dead since the 1970s because they have not been able to harvest aquafauna from this area. HM-laden sediments and drainage from the mine sites contaminate food sources and are hazards to animal and human lives. A preliminary report on the Taft River by a team from the Marine Geological Survey Division in June 2005 showed that acid mine drainage had manifested in the receiving bodies of water and the concentration of HM was higher than average (MGB-MESD 2006). The amount of HM contamination in the river system, particularly on the basin's aquafauna and flora, is little understood. Therefore, this study was carried out to determine the concentration of HM in mollusk and crustacean species that thrive along the Taft River, as well as the health risks to the humans who consume them. Specifically, the purpose of this study was to quantify the concentrations of As, Cu, Ni, Cr, Pb, Rb, Sr, Zr, Ti, Mn, and Zn in the flesh of different mollusk and crustacean species collected from the different sections of the Taft River, as well as to determine the associated health risks from the consumption of these mollusks and crustaceans.

MATERIALS AND METHODS

Study Area

The Taft River is a stream located in Province of Eastern Samar, Eastern Visayas, Philippines (latitude: 11°54'11.02"; longitude: 125°25'22.02"). The estimated terrain elevation is 1 m above sea level, and it receives surface water from Hinabangan, Samar where the Bagacay mine is located (Patindol 2016). The principal study area examined is the segment from the lower of the river, which extends roughly 30.59 km to the Guila-guila Creek (Figure 1).

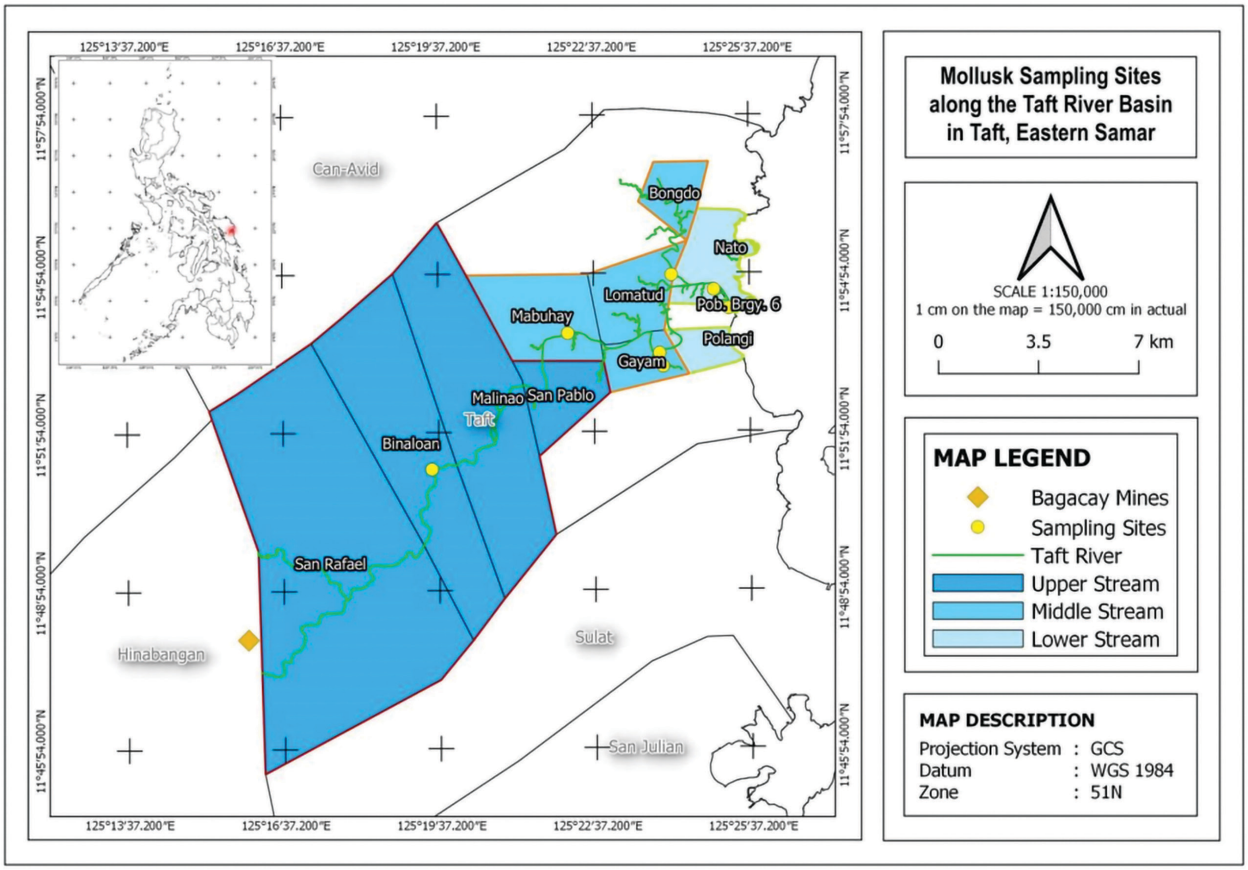

Figure 1. Sample sites along Taft River, Philippines.

Collection and Treatment of Samples

River segments along the Taft River were divided into upper, middle, and lower sections. In each section, three sites were established to collect the samples from 27 Oct to 03 Nov 2020. Indigenous methods including the use of nets and manual hand-picking were employed to reduce habitat disturbances and preserve most of the species (Appendix Figures I and II). During sample collection, no mollusk species were found in the upper section and only four mollusk species were collected in the middle and lower sections of the river, whereas crustacean species were distributed throughout all sections of the river. Identification of the specimens was done *in situ* utilizing their local names. Field guides, other related material, and the marine species identification portal (http://species-identification.org/) were used to confirm the identification using genuine samples and high-resolution images. For each site, 10 samples of mollusks and five samples of crustaceans were randomly collected as representative composite samples per species, per site. Collected samples were rinsed in river water and placed in pre-cleaned zip lock bags, which were then stored in an ice chest filled with ice to preserve the specimens before being transferred to the Northwest Samar State University (NwSSU) laboratory for further processing.

Water and sediment samples were collected simultaneously at each site where the specimens were chosen for laboratory analyses. Pre-cleaned 1000-mL polythene bottles were used to collect grab samples of river water from a depth of not more than 20 cm from the surface. At each location, triplicate samples were collected, preserved in an ice chest, and then transferred to the laboratory for chemical analysis. Similarly, the surface sediments on the riverbed were collected at each site along the river using a grab sampler. A total of three composite samples from each site was collected and taken to the lab for chemical analysis. The sediment samples were air-dried for 24–48 h, sieved through a 2-mm stainless steel wire mesh, and kept at 4 °C until analysis.

Processing of Mollusk and Crustacean Samples

The collected samples were washed and cleaned to remove external debris and attached materials. The flesh was separated from the exoskeleton and washed with distilled water. A total of 12 representative composite samples of mollusks derived from 10 individuals and 15 representative composite samples of crustaceans derived from five individuals were obtained and labeled with the species name and location. The representative composite flesh samples were oven-dried for about 72 h at 60 °C in a convection oven and then stored in sealed vials inside the refrigerator for further chemical processing (Habte *et al.* (2015).

Analyses of HM

Mollusks and crustaceans. The oven-dried flesh samples were finely crushed into powder using a blender in preparation for analysis. The method used was adapted from Habte *et al.* (2015) with modifications. Briefly, approximately 0.50 g of the tissues were digested in a mixture of 8-mL nitric acid (HNO_3) and 2 mL hydrogen peroxide (H_2O_2) in a microwave digester (Ethos Easy) at 200 °C for 1 h. The digests were then diluted to 25 mL with ultrapure water and filtered through Whatman No. 42. Using an inductively coupled plasma–mass spectroscopy (ICP-MS, Thermo Scientific TM iCAP Q ICP-MS, Illinois 60060 USA), the digests were examined for total concentrations of As, Cu, Ni, Cr, Pb, Rb, Sr, Zr, Mn, and Zn. In this study, the concentrations of these elements in the flesh are expressed on a dry-weight basis. Based on an X-ray fluorescence analyses, these metals were selected due to their high abundance in the sediment samples.

Water. Water samples were analyzed for HM content based on the procedure stated in USEPA Method 352.1 (USEPA 1983). Briefly, As was determined using AAS with a manual hydride generation system, Hg by a cold vapor in AAS, the Ni, Pb, Zn, and Cu by direct air-acetylene flame-AAS.

Sediments. The sediments were analyzed for the total As, Cu, Ni, Cr, Pb, Ti, Mn, and Zn were quantified after the digestion of 1.0 g samples with concentrated nitric acid and hydrogen chloride in a microwave digester (Ethos Easy) at 200 ºC for 1 h. ICP-MS (Thermo Scientific TM iCAP Q ICP-MS, Illinois 60060, USA) was used to determine the concentration of HM in the digest (Manyiwa *et al.* 2021). Concentration of these elements in sediments are expressed on a dry-weight basis.

Quality Control/ Method Validation

Precision and accuracy were ensured for all chemical analyses by batch inclusion of an internal reference sample whose contents were cross verified by three analytical laboratories in Botswana and repeat analysis was made for samples where the coefficient of variation of duplicate analysis (n = 2) was greater than 10%. For all elements examined, the internal standard sample had a recovery rate of > 95%. The detection limit for ICP-MS is 0.005–0.0005 ng mL^{-1} for mid-elements. The BIUST (Botswana International University of Science and Technology) Environmental Remediation and Management Laboratory, Department of Earth and Environment Sciences, BIUST, Botswana, completed all chemical analyses for the flesh of mollusks and crustaceans and the sediments, whereas the water samples were analyzed at OMLI (Ostrea Mineral Laboratory Inc.), Mandaue City, Philippines.

Health risk assessment. Health risk assessment was done to uncover health risks that are directly linked to medical problems which develop over a long period. In this study, the target hazard quotient (THQ) and target cancer risks (TCRs) were examined for various HM detected in samples to determine the non-carcinogenic health risk of consuming mollusks within the area.

Estimated daily intake (EDI). The preceding equation was used to calculate the EDI of HM (Griboff *et al.* 2017):

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

where C is the average HM concentration for each species from each site (mg kg^{-1} DW), IRd is the daily average ingestion rate (17.1 g d^{-1} for mollusks and 8.3 g d^{-1} for crustaceans), BW is the average body weight (BW) of Filipino adults (70 kg) (Tayone *et al.* 2020). EDI is measured in µg/kg BW/d. There is no published reference data for Rb, Sr, Zr, and Mn in the EDI computation. Hence, they are excluded from the discussion.

Target hazard quotient (THQ). The THQ is the ratio of hazardous element exposure to the reference dose (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. If the THQ is more than 1; however, there is a possibility that negative health problems will occur. A THQ greater than 1, however, does not indicate a statistical likelihood of negative non-carcinogenic health consequences. The THQ was estimated using the US EPA formula, as demonstrated by Tayone *et al.* (2020):

$$\text{THQ}_{\text{non-carcinogenic}} = (\text{EF} \times \text{ED} \times \text{Ird} \times \text{C}) \\ [(\text{RfD} \times \text{BW} \times \text{AT})]\ \hat{}(-1) \qquad (2)$$

where $\text{THQ}_{\text{non-carcinogenic}}$ 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, C is the HM concentration in aquatic products (mg kg^{-1} DW), RfD is the oral reference dose values based from Liu *et al.* (2019), BW is the average BW (70 kg), and AT is the average lifetime exposure (EF x ED).

The HI was also computed since it aids in determining the total noncarcinogenic harm to human health from many HM. The hazardous quotient of all HM is added together to form the HI. Toxins with a HI of 1 or below are unlikely to induce noncancerous health consequences over a lifetime of exposure (Tayone *et al.* 2020).

Target cancer risk (TCR). The TCR is a tool for determining the risk of cancer from exposure to carcinogenic chemicals over the course of a person's lifetime. An oral slope factor is used instead of an oral reference dosage, which is used 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:

$$TCr = (EF \times ED \times Ird \times C \times CPSo)\ [(BW \times AT)]\ {}^\wedge(-1) \times 10E{}^\wedge(-3) \qquad (3)$$

where EF is the exposure frequency of 104 d (twice a week) exposure to the element, ED is the exposure duration average of 60 yr 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: 1.5 for inorganic As, 1.7 for Ni, 0.5 for Cr, and 0.004 for Pb expressed as mg kg^{-1} d^{-1} (Liu *et al*. 2019). 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 (Antoine *et al*. 2017). Only the TCR for As, Ni, Cr, and Pb was computed while other elements have no established oral cancer slope data.

Statistical Analysis

A one-way analysis of variance based on species was used to compare the HM levels of the various species for mollusks and crustaceans separately. When significant differences existed, Tukey's test was used to compare means at a 5% level of significance. SPSS 12 was used to conduct the analysis.

RESULTS

Concentrations of HM in Mollusk, Crustaceans, Water, and Sediments

The concentration of the ten HM in the flesh of four mollusk species are presented in Table 1. Significant variation was observed between species on the concentration of As, Cu, Ni, Pb, Rb, Sr, Zr, and Zn, whereas nonsignificant variation was observed for Cr and Mn. Of these metals, the average concentration for four mollusk species was found to be in the following order: Mn (729–660 mg kg^{-1}) > Cr (510–557.67) > Zn (483.33–43.33) > Cu (97.67–35) > Rb (37.67–3.33) > Ni (29.30–10.50) > As (18.33–0.00) > Sr (14.67–0.00) > Zr (3.00–0.00). The As concentration in *T. telescompium*, *N. albicilla*, and *B. violacea* was 4,000 time higher than the allowable limit for seafood of 0.002 (JECFA 2003). The Cu concentration was significantly higher in *T. telescompium* and *B. violacea* as

Table 1. Heavy metal concentrations (mg kg^{-1})[1] in the tissues of different mollusks species collected from the Taft River in March 2021.

HM	***T. telescompium*** **(n = 3)**	***M. mercenaria*** **(n = 3)**	***N. albicilla*** **(n = 3)**	***B. violacea*** **(n = 3)**	***p*-value**	**Regulatory limits (mg kg^{-1})**	**Reference**
Stream location	Lower	Lower	Lower	Middle			
As	18.33 ± 2.25a	0.00 ± 0.00a	8.33 ± 3.67a	14.33 ± 3.18a	0.106ns	0.002	JECFA (2003)
Cu	56.67 ± 8.32b	52.00 ± 12.36b	35.00 ± 16.21b	97.67 ± 7.24a	0.004**	0.5	FAO/WHO (2005)
Ni	18.13 ± 2.56b	16.83 ± 1.85b	10.50 ± 6.22b	29.30 ± 3.21a	0.001**	3.7	FAO/WHO (2005)
Cr	510.00 ± 12.32a	556.00 ± 27.39a	557.67 ± 22.73a	532.00 ± 18.73a	0.293ns	2.0	FAO/WHO (2005)
Pb	0.34 ± 0.08b	0.31 ± 0.16b	0.21 ± 0.19b	0.59 ± 0.04a	0.004**	0.12–2.0	FAO/WHO (2005)
Rb	26.33 ± 1.26b	37.67 ± 2.20a	9.00 ± 3.40c	3.33 ± 2.38c	0.000***	NR	
Sr	4.67 ± 0.26b	0.00 ± 0.00c	2.33b ± 0.12c	14.67 ± 1.16a	0.000***	NR	
Zr	0.00 ± 0.00b	0.00 ± 0.00b	3.00 ± 0.54a	2.00 ± 0.83ab	0.006**	NR	
Mn	660.33 ± 12.44a	698.67 ± 18.23a	729.00 ± 20.24a	706.67 ± 21.55a	0.554ns	0.01	WHO (1985)
Zn	162.33 ± 6.23b	43.33 ± 10.28d	77.00 ± 14.22c	483.33 ± 22.38a	0.050*	5.0	FAO/WHO (2005)

[1] Means within the metal across the different species under the same group of organisms, followed by the same letter(s) are not significantly different from each other based on Tukey's test at 5% level of significance. [*] significant; [**] highly significant; [***] very highly significant; [NR] no reference available for allowable critical values safe for human consumption of mollusk species.

compared to the other two mollusk species and the highest concentration was about 195 times higher than the 0.5 critical limit (FAO/WHO 2005). The Ni concentration was significantly higher in *B. violacea* than the other three species, but all their Ni contents are at least 2.8 times higher than the critical limit. The Cr and Mn concentrations between mollusk species did not vary significantly, but their concentrations are about 255–278 times the critical value of 2.0 for Cr and for Mn, respectively. The highest Zn concertation was recorded in *B. violacea*, whereas the lowest was in *M. mercenaria*, and these values are more than 8.6–96 times higher than the critical limit of 5.0. *M. mercenaria* had the highest Rb, whereas *N. albicilla* had the highest Zr, and *B. violacea* had the highest Sr. The Pb concentration differed significantly between species, but all the values are below the critical limits of 0.12–2.0 (FAO/WHO 2005).

The concentration of the ten HM in the flesh of two crustacean species from different sections of the river were presented in Table 2. Significant variations in the concentrations were observed between species collected from different location for all metals under consideration. Based on the average concentration of the two species from the different locations, Mn was the highest ranging from 750.00–669 mg kg^{-1}), followed by Cr (574.00–507.67) > Zn (331.00–40.67) > Cu (118.67–22.33) > Sr (60.67–0.00) > Rb (31.33–8.00) > Ni (25.79–6.70) > As (17.67–0.00) > Zr(5.67–0.00) > Pb (0.71–0.25). The As concentration in *S. serrata* collected from the middle section was higher than those from the lower section of the river, whereas As was not detected in *M. rosenbergii* from all sections. The As concentration in *S. serrata* was 7000–8835 times higher than the allowable limit for seafood of 0.002 (JECFA 2003). The Cu and Ni concentrations were significantly higher in *S. serrata* from the lower section of the river compared to other samples, and these concentrations were about 236 times higher than the 0.5 critical limit for Cu and about 6.9 times higher than the 3.7 critical limit for Ni, respectively (FAO/WHO 2005). The lowest concentration of Cu and Ni was obtained in *M. rosenbergii* collected from the middle section of the river. The Cr and Mn concentrations were significantly higher in *M. rosenbergii* from the upper section and *S. serrata* collected from the middle section of the river, and the concentration is up to 287 times the critical value of 2.0 for Cr and 750 times for Mn, respectively. The highest Zn concertation was recorded in *S. serrata* from the lower section of the river, whereas the lowest was in *S. serrata* from the middle section, and these values are more than 8–66 times higher than the critical limit of 5.0. *S. serrata* collected from the lower section of the river had the highest Rb, Sr, and Zr concentrations compared to other samples. The Pb concentration was significantly higher also in *S. serrata*, but all the values are below the critical limits of 0.12–2.0 (FAO/WHO 2005).

The HM concentrations in water samples obtained from the lower and the middle sections of the river were lower than the DENR-EMB limit set for class C (Appendix Table I) (DENR-DAO 2016-08). In contrast, the water samples from the upper section of the river have higher values of As, Pb, and Cu than those samples from the lower and middle section of the river and these values are above the DENR-DAO 2016-08 limit for class C, which is 0.02 mg L^{-1} for As, 0.5 mg L^{-1} for Pb, and 0.02 mg L^{-1} for

Table 2. Heavy metal concentrations (mg kg^{-1}) in the tissues of different crustacean species collected from the Taft River in March 2021.

HM	***M. rosenbergii* (n = 3)**	***S. serrata* (n = 3)**	***S. serrata* (n = 3)**	***M. rosenbergii* (n = 3)**	***M. rosenbergii* (n = 3)**	***p*-value**	**Regulatory limits (mg kg^{-1})**	**Reference**
Stream location	Lower	Lower	Middle	Middle	Upper			
As	0.00 ± 0.00c	14.00 ± 0.27b	17.67 ± 0.52a	0.00 ± 0.00c	0.00 ± 0.00c	0.024*	0.002	JECFA (2003)
Cu	41.00 ± 3.26b	118.67 ± 10.28a	35.00 ± 2.58bc	22.33 ± 5.82c	49.33 ± 5.98b	0.000***	0.5	FAO/WHO (2005)
Ni	12.30 ± 1.48b	25.79 ± 2.74a	10.50 ± 1.74b	6.70 ± 1.86c	14.80 ± 0.88b	0.046*	3.7	FAO/WHO (2005)
Cr	554.00 ± 6.28a	508.00 ± 2.44b	569.33 ± 6.29a	507.67 ± 3.22b	574.00 ± 5.40a	0.048*	2.0	FAO/WHO (2005)
Pb	0.25 ± 0.08b	0.71 ± 0.14a	0.21 ± 0.06b	0.13 ± 0.02c	0.30 ± 0.08b	0.018*	0.12–2.0	FAO/WHO (2005)
Rb	26.67 ± 2.14ab	31.33 ± 2.20a	21.67 ± 1.84b	28.67 ± 1.20a	8.00 ± 2.79c	0.047*	NR	
Sr	22.33 ± 5.65b	60.67 ± 7.24a	3.33 ± 1.52c	0.00 ± 0.00c	2.33 ± 0.86c	0.008**	NR	
Zr	0.00 ± 0.00c	5.67 ± 1.28a	0.00 ± 0.00c	0.00 ± 0.00c	2.33 ± 0.63b	0.046*	NR	
Mn	669.33 ± 10.25b	688.00 ± 6.72b	724.00 ± 4.88a	685.67 ± 6.26b	750.67 ± 22.30a	0.028*	0.01	WHO (1985)
Zn	111.67 ± 8.36b	331.00 ± 10.22a	40.67 ± 6.74c	56.33 ± 4.88c	44.00 ± 5.30c	0.003**	5.0	FAO/WHO (2005)

[1] Means within the metal across the different species under the same group of organisms, followed by the same letter(s) are not significantly different from each other based on Tukey's test at 5% level of significance. [*] significant; [**] highly significant; [***] very highly significant; [NR] no reference available for allowable critical values safe for human consumption of mollusk species.

Cu. The Ni concentration is less than 0.02 in all sampling points, whereas the Pb and Zn concentration in the lower and middle sections of the river were below the detection limit of 0.02 and 0.003 for Pb and Zn, respectively.

The concentration of different metals in the sediment varied significantly between sampling locations (Appendix Table I). For As, Cu, and Zn, their concentrations were higher in the lower section than in the upper and middle sections but the values in the upper section were significantly higher than the middle section. For Ni and Cr, the concentration in the middle section was significantly higher than those from the lower and upper section, whereas for Pb and Zr, the lower and the upper section had comparable values but were higher than the middle section. For Mn, Rb, and Sr, significantly higher concentrations were obtained from samples in the upper section of the river. Furthermore, the concentration of HM and metalloids in the flesh of crustaceans and mollusks were not correlated with the concentration in these metals in sediments or in water.

Human Health Risk Assessment of Mollusks and Crustaceans

The PTDI (µg kg^{-1}BW d^{-1}) and the EDI of HM from the consumption of several mollusks and crustaceans found in the Taft River by the local community is presented in Table 3. Consumption of *T. telescompium* will result in an EDI level, which is higher than the PTDI for As equivalent to 4.48 µg kg^{-1} BW d^{-1}and Cr at 124.59 µg kg^{-1} BW d^{-1}; all other elements are below the PTDI or no-reference value is available. For *M. mercenaria*, only the EDI$_{Cr}$ equivalent to 135 µg kg^{-1} BW d^{-1} was above the PTDI$_{Cr}$, for *N. albicilla*, the EDI for As is about 2.03 µg kg^{-1} BW d^{-1}, and Cr at 136.23 µg kg^{-1} BW d^{-1} are above their respective PTDIs, whereas for *B. violacea*, EDI for As at 3.50 µg kg^{-1} BW d^{-1} and Cr at 129.96 µg kg^{-1} BW d^{-1} are above their respective PTDIs. The EDI for Cu among the different mollusk species ranged from 8.55–23.86 µg kg^{-1} BW d^{-1}, 7.16–2.57 µg kg^{-1} BW d^{-1}for Ni, 0.14–0.05 µg kg^{-1} BW d^{-1} for Pb, 9.20–0.81 µg kg^{-1} BW d^{-1}for Rb, 3.58–0.00 µg kg^{-1} BW d^{-1} for Zr, 178.08–161.31 µg kg^{-1} BW d^{-1} for Mn, and 118.07–10.58 µg kg^{-1} BW d^{-1} for Zn. All these values are below the PTDI for their respective elements (Table 3).

For crustaceans, the EDIs for As and Cr were higher than their respective PTDIs. The EDI for Cr in *M. rosenbergii* collected form lower, middle, and upper sections of the river ranged from 60.20–68.06 µg kg^{-1} BW d^{-1}, and these values are higher than the 3.0 µg kg^{-1} BW d^{-1} PTDI for Cr (Table 3). Consuming *S. serrata* will result in an EDI for As and Cr above their respective PTDIs with the EDI$_{As}$ of 1.66 µg kg^{-1} BW d^{-1} and EDI$_{Cr}$ of 20.23 µg kg^{-1} BW d^{-1}, respectively, for samples collected from the lower section of the river. Those that were collected from the middle section have EDI$_{As}$ of 2.10 µg kg^{-1} BW d^{-1}and EDI$_{Cr}$ of 67.51 µg kg^{-1} BW d^{-1}. For both crustacean species collected from different sections of the river, the EDI for Cu ranged from 14.05–2.65 µg kg^{-1} BW d^{-1},

Table 3. Estimated daily intake (adult) in mg kg^{-1} body weight d^{-1}.

HM	Estimated daily intake (EDI)									PTDI (µg/kg BW/d)
	Mollusks				Crustaceans					
	T. telescompium	*M. mercenaria*	*N. albicilla*	*B. violacea*	*M. rosenbergii*	*S. serrata*	*S. serrata*	*M. rosenbergii*	*M. rosenbergii*	
Stream location	Lower	Lower	Lower	Middle	Lower	Lower	Middle	Middle	Upper	
As	4.48	0.00	2.03	3.50	0.00	1.66	2.10	0.00	0.00	0.30
Cu	13.84	12.70	8.55	23.86	4.86	14.07	4.15	2.65	5.85	40.00
Ni	4.43	4.11	2.57	7.16	1.46	3.06	1.25	0.79	1.75	20.00
Cr	124.59	135.82	136.23	129.96	65.69	60.23	67.51	60.20	68.06	3.00
Pb	0.08	0.08	0.05	0.14	0.03	0.08	0.02	0.02	0.04	3.57
Rb	6.43	9.20	2.20	0.81	3.16	3.71	2.57	3.40	0.95	ND
Sr	1.14	0.00	0.57	3.58	2.65	7.19	0.39	0.00	0.28	ND
Zr	0.00	0.00	0.73	0.49	0.00	0.67	0.00	0.00	0.28	ND
Ti	1555.29	0.00	3029.14	4238.36	1458.43	193.67	458.48	1158.05	1011.81	ND
Mn	161.31	170.68	178.08	172.63	79.36	81.58	85.85	81.30	89.01	ND
Zn	39.65	10.58	18.81	118.07	13.24	39.25	4.82	6.68	5.22	300.00

[ND] no available data
[PTD] provisional tolerable daily intake (Liu *et al.* 2019)

3.06–0.79 μg kg^{-1} BW d^{-1} for Ni, 0.08–0.02 μg kg^{-1} BW d^{-1} for Pb, 3.71–0.95 μg kg^{-1} BW d^{-1}for Rb, 7.19–0.00 μg kg^{-1} BW d^{-1}for Sr, 0.67–0.00 μg kg^{-1} BW d^{-1} for Zr, 89.01–79.36 μg kg^{-1} BW d^{-1} for Mn, and 39.25–4.82 μg kg^{-1}BW day^{-1} for Zn. All these EDI measurements are below their respective PTDIs. On average, the EDIs for most of the elements under consideration for mollusks are higher than those of the crustaceans.

To further assess the risk associated with the consumption of these mollusk and crustacean species from the Taft River, the THQ was estimated and presented in Table 4. Following the same trend as the EDIs, the THQ_{As}, THQ_{Cr} and THQ_{Mn} of consuming the different mollusk species ranged from 14.93–0.00, 45.27–41.53, and 1.27–1.15, respectively. The THQ for other elements under consideration due to consumption of mollusks were less than 1. The overall HI for the different mollusk species ranged from 58.33–47.07. For crustaceans, the THQ_{Cr} for the two species harvested from different sections of the river ranged from 22.69–20.08, whereas the THQ_{As} of consuming *S. serrata* from the lower and middle sections of the river were 5.53 and 6.98, respectively. The THQ for Cu, Ni, Pb, Mn, and Zn due to consumption of these two crustacean species collected from different sections of the river were less than 1. The HI for the *S. serrata* from lower and middle section were 26.85 and 30.29, respectively. These values are higher than *M. rosenbergii* with HI ranging from 23.58–20.78.

Table 4. Estimated target hazard quotient (THQ) and hazard index (HI) due to twice a day consumption of different mollusk and crustacean species harvested from Taft River, Philippines.

HM	**Target hazard quotient (THQ)**									**RfD[1] (μg kg^{-1})**
	Mollusks				Crustaceans					
	T. telescompium	*M. mercenaria*	*N. albicilla*	*B. violacea*	*M. rosenbergii*	*S. serrata*	*S. serrata*	*M. rosenbergii*	*M. rosenbergii*	
Stream location	Lower	Lower	Lower	Middle	Lower	Lower	Middle	Middle	Upper	
As	14.93	0.00	6.78	11.67	0.00	5.53	6.98	0.00	0.00	0.30
Cu	0.35	0.32	0.21	0.60	0.12	0.35	0.10	0.07	0.15	40.00
Ni	0.22	0.21	0.13	0.36	0.07	0.15	0.06	0.04	0.09	20.00
Cr	41.53	45.27	45.41	43.32	21.90	20.08	22.50	20.07	22.69	3.00
Pb	0.02	0.02	0.01	0.04	0.01	0.02	0.01	0.00	0.01	3.50
Mn	1.15	1.22	1.27	1.23	0.57	0.58	0.61	0.58	0.64	140.00
Zn	0.13	0.04	0.06	0.39	0.04	0.13	0.02	0.02	0.02	300.00
Hazard index	58.33	47.07	53.88	57.61	22.71	26.85	30.29	20.78	23.58	

[1] Liu *et al.* (2019)

Table 5. Estimated target cancer risk for adult due to twice a day consumption of different mollusk species harvested from Taft River, Philippines.

HM	**Target cancer risk (TCR)**								
	Mollusks				Crustaceans				
	T. telescompium	*M. mercenaria*	*N. albicilla*	*B. violacea*	*M. rosenbergii*	*S. serrata*	*S. serrata*	*M. rosenbergii*	*M. rosenbergii*
Stream location	Lower	Lower	Lower	Middle	Lower	Lower	Middle	Middle	Upper
As	0.0067166	0.0000000	0.0030524	0.0052509	0.0000000	0.0051300	0.0064748	0.0000000	0.0000000
Ni	0.0075291	0.0069893	0.0043605	0.0121679	0.0051080	0.0107102	0.0043605	0.0027824	0.0061462
Cr	0.0622929	0.0679114	0.0681154	0.0649800	0.0676671	0.0620486	0.0695396	0.0620083	0.0701100
Pb	0.0000007	0.0000006	0.0000004	0.0000012	0.0000005	0.0000015	0.0000004	0.0000003	0.0000006

TCR reference values (Antoine *et al.* 2017): "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

The TRC for adults due to consumption of mollusks and crustaceans from the different sections of the river was presented in Table 5. The consumption of *T. telescompium* will result in TRC_{As} of 0.0067166, TRC_{Ni} of 0.0075291, TRC_{Cr} of 0.0622929, and TRC_{Pb} of 0.0000007. For *M. mercenaria* from the lower section of the river has TRC_{Ni} of 0.0069893, TRC_{Cr} of 0.0679114, and TRC_{Pb} of 0.0000006. For *N. albicilla*, TRC_{As} is 0.0030524, TRC_{Ni} of 0.0043605, TRC_{Cr} of 0.0681154, and TRC_{Pb} of 0.0000004; whereas for *B. violacea*, the TRC_{As} is 0.0052509, TRC_{Ni} of 0.0121679, TRC_{Cr} of 0.06498, and TRC_{Pb} of 0.0000012. For *M. rosenbergii* from different sections of the river, the TRC_{Ni} ranged from 0.0027–0.0061, TRC_{Cr} ranged from 0.062–0.70 and 0.0000003–0.0000006 for TRC_{Pb}. In contrast, *S. serrata* has a TRCAs of between 0.0051–0.0065, TRC_{Ni} of 0.0043–0.0107, TRC_{Cr} between 0.06294–0.0695, and TRC_{Pb} of between 0.0000004–0.0000015.

DISCUSSION

The off-site impacts of Bagacay mines on the Taft River, Philippines, especially on the HM accumulation in the flesh of mollusks and crustaceans harvested from the different sections of the river, were presented in this study. The concentrations of these HM of interest in the flesh of mollusks and crustaceans is a function of the concentrations in their habitat (sediments and water) and the contents and accumulation of these metals in other organisms serving as their food sources. With respect to the concentration of these metals in the flesh of mollusks and crustaceans, the results showed significant variation in the concentration between species and on the sampling site; and the average concentration of four mollusk species was in the following order: Mn > Cr > Zn > Cu >Rb > Ni > As > Sr > Zr > Pb. The significant variation on the amount of HM accumulated on the flesh in different species are inherent to the species characteristics such as feeding habits including ingestion rate, maturity of the individual species, concentration and availability in the growing substrate, and other environmental factors (Bonsignore *et al.* 2018). Similarly, differences on the degree of accumulation between these elements depends on the chemical characteristics of the elements, its role and function in the metabolism for each species (species specific), metal assimilation efficiency in the dietary phase, and the efflux rate of these elements inherent to species characteristics (Bonsignore *et al.* 2018). The results indicated that Mn and Cr are extremely high compared to their concentration reported in previous studies, wherein the concentration for Zn and Fe are much higher than those of Cr and Mn (Swaleh *et al.* 2016). The Mn concentration in mollusks collected in Magellan Straight, Chile was only about 1.46 mg kg^{-1} (Astorga España *et al.* 2004), compared to about 19.31 ± 1.71 µg g^{-1} for samples from Tudor Creek in Mombasa, Kenya (Swaleh *et al.* 2016). Similarly, the Cr concentrations in the clams were < 4 mg g^{-1} but again can be somewhat higher (9.6 mg g^{-1}) in some regions of the Moroccan Atlantic coast (Maanan 2008). Both Cu and Zn are essential trace elements for the bivalves and mussels, and our results are slightly above the concentrations from non-polluted environments (< 20 and < 200 mg g^{-1}, respectively) but much lower than those reported in *P. nobilis* samples (up to 200 and 4000 mg g^{-1}) from Spain (Vazquez-Luis *et al.* 2016) and in *M. edulis* (Cu up to 1250 mg g^{-1} and Zn up to 2755 mg g^{-1}) collected from Gulf of Maine and Narragansett Bay, United States (Chen *et al.* 2016). In contrast, Ni concentration in the samples from Taft River are within the reported Ni concentrations for mussels from < 5 mg g^{-1} to as high as 33 mg g^{-1} in *M. galloprovincialis* from the Moroccan coastal region (Maanan 2008). The Pb concentrations in the samples were below 1 mg kg^{-1}, which was much lower than the values reported in *M. galloprovincialis*, which were less than 5 mg g^{-1} to as high as 59–64 mg g^{-1} from the Moroccan coastal region and Gulf of Maine and Narragansett Bay, United States (Maanan 2008).

The concentrations of As, Cu, Ni, Cr Mn and Zn in *T. telescompium*, *N. albicilla*, *M. mercenaria*, and *B. violacea* are above the allowable limits indicating high risk of exposure to these metals when these species are consumed by humans. Among these metals, As, Cr, and Mn would contribute more to potential risk than Cu and Ni. The highest Zn concertation was recorded in *B. violacea* while the lowest was in *M. mercenaria*, and these values are more than 8.6–96 times higher than the critical limit of 5.0. *M. mercenaria* had the highest Rb, whereas *N. albicilla* had the highest Zr, and *B. violacea* had the highest Sr. The Pb concentration differed significantly between species, but all the values are below the critical limits of 0.12–2.0 (FAO/WHO 2005).

In comparison with other species, Cu, Ni, Cr, Pb, Pb, Mn, and Zn were detected in the flesh of *M. mercenaria*. Moreover, all the 10 metals under consideration in this study were detected in other mollusks that were examined. The feeding behavior of *M. mercenaria* may explain the low content for the majority of HM examined. *M. mercenaria* is a benthic bivalve that is considered a filter feeder that obtains its food from suspended matter in the surrounding water, which – incidentally – had rather low levels of HM levels (Liu *et al.* 2019). In contrast, *B. violacea* has a high concentration of HM as compared to *M. mercenaria*. This could be attributed to its food source and feeding habit, which mainly comes from sediments. It should be emphasized that the HM level of the water samples in the study area was low, but the HM content of the sediment was extremely high, contributing to the high concentration of these metals in the flesh of *B. violacea*. On the other hand, the other two species – *T. telescompium* and the natural scavenger, *N. albicilla* – eat mostly organic materials and collect HM through trophic transfer (Liu *et al.* 2019).

Specifically, the highest concentrations of Cu and Ni were found in *B. violacea*, and the lowest value was from *N. albicilla* samples, respectively (Table 1). High concentrations of Cu may be traced back to the mining activities in the Bagacay mines, which mainly extracted copper minerals. Cu concentrations above a certain threshold, along with a low pH, are thought to be lethal to fish; but at low concentrations, Cu is a constituent of oxidases – which is important for the regulation of redox reactions, respiration, and cartilage formation (Khayatzadeh and Abbasi 2010). In addition, high levels of Cu result in an increase in the unsaturation of fatty acids in the phospholipids of the gills of mussels which may cause a loss of membrane integrity and high susceptibility to DNA damage (Fokina *et al.* 2013), Ni is composed of a variety of proteins and enzymes and stimulates growth, but elevated levels of Ni can inhibit multiple enzymes and interfere with detoxification processes in the liver, often by decreasing its filtration capacity (Fokina *et al.* 2013). A high concentration of Zn is also present in all samples with values of 43.33 μg g^{-1} for *M. mercenaria*, to as high as 483.33 μg g^{-1} found in *B. violacea*. Although zinc is an essential element for animals and a component of many enzymes, excessive concentrations of zinc can injure mollusks by reducing filtration rates and by causing gill injury, but little is known about the zinc absorption mechanism in fish and other aquafauna (Khayatzadeh and Abbasi 2010). Low levels of Pb concentrations were found in the flesh of all four species, whereas many HM are essential nutrients in low amounts, Pb is non-essential and a well-known industrial hazard, causing severe toxic consequences in higher animals when exposed acutely or chronically (Khayatzadeh and Abbasi 2010).

Other traces of elements were also detected in the samples. Rb had the highest value of 37.67 μg g^{-1} in *M. mercenaria* and the lowest value of 3.33 μg g^{-1} in *B. violacea*. The element Sr was not detected in *M. mercenaria* but had the values of 4.67 μg g^{-1} in *T. telescompium*, 2.33 μg g^{-1} in *N. albicilla*, and 14.67 μg g^{-1} μg g^{-1} for *B. violacea*, respectively. The Zr was not detected in *T. telescompium* and *M. mercenaria* samples but had the values of 3 μg g^{-1} in *N. albicilla* and 2 μg g^{-1} in *B. violacea*. Although there is no established critical acceptable daily intake of other metals such as Sr, Zr, and Rb, there is a need to monitor their concentrations in the edible parts of the different aquatic species that will be harvested from the Taft River. For example, as indicated that excessive intake of Sr may produce insoluble Sr phosphates leading to phosphorus deficiency and may cause diseases such as rickets, and together with high Cr concentrations in the samples, there is a possibility that strontium chromate may be formed, which is harmful to humans in small amounts (Nardone *et al*. 2015). On the other hand, rubidium is nontoxic, but depending on the counter anion, it may cause serious health problems. For example, rubidium fluoride causes renal and hepatic toxicities in rats (Usuda *et al*. 2014). Similarly, Zr has not yet been associated with any specific metabolic function, but it is being monitored as it can easily penetrate soft tissues and the bone, and it can cross the blood-brain barrier to be deposited in the brain and the level of toxicity has been found to be moderately low, both histologically and cytologically (Lee *et al*. 2010).

Between the two species of crustaceans, As was only found in the flesh of *S. serrata* with a higher concentration in samples collected from the middle section and the lower section of the river (Table 2). This may be traced to the movement of the water, where elements tend to concentrate or stay in river sections, where salt and fresh water meet. *S. serrata*, which was collected in the lower section of the river, had all 11 HM detected; all other crustacean samples have at least one undetected element (Table 2). For Cu, *S. serrata* in the lower section of the river has the highest concentration, followed by *M. rosenbergii* in the upper section of the river, *M. rosenbergii* in the lower section, *S. serrata* in the middle section, and *M. rosenbergii* in the middle section (Table 2). Ni was detected in all crustacean samples tested with the highest concentration found in *S. serrata* from the lower section of the river, and the lowest Ni concentration was detected in *M. rosenbergii* collected from the middle section of the river. A similar trend was noted for Cr concentrations in all samples examined with the highest value in *M. rosenbergii* from the upper section of the river and the lowest value of the same species found in the middle section of the river. The highest concentration of Zn was found in *S. serrata* in the lower river section and the lowest concentration was detected in the same species from the middle section of the river. These concentrations are above the critical levels that are intended for human consumption in seafood (UNEP 1986). Similar to the mollusk species, the concentration of the 10 HM in the flesh of two crustacean species from different sections of the river showed that Mn and Cr have the highest concentration, and the consumption of these species could contribute to dietary intake by humans.

Analysis of the water and sediment obtained from the sampling site for these specimens revealed that the surface water could be classified under class C (Appendix Table I; DENR-DAO 2016-08). The concentration of As, Ni, Pb, Cu, and Zn in the lower and middle sections of the river are below the DENR-EMB limit, whereas in the upper section, the concentration of As, Pb, and Cu are higher than the limits of 0.02, 0.05, and 0.2 for Class C. This indicates that the water quality of the lower and the middle section of the river is considered safe for aquaculture, whereas the upper section could have limitations due to the elevated levels of As, Pb, and Cu. The elevated levels of As, Pb, and Cu could contribute to higher bioaccumulation of these metals in these aquatic organisms. In a similar manner, the concentration of As, Cu, Ni, Cr, and Zn in sediment samples from the lower and upper sections of the river greatly exceed the severe effect of levels established in NOAA 1999 (Buchman 1999), whereas Cr was excessively high in the middle section (Appendix Table I). Although there was poor correlation between concentration in sediments with that found in the flesh of mollusks and crustaceans, these high concentrations in the sediment could serve as the point of origination for the HM that accumulated in these aquafauna. The poor correlation could be due to the limited samples that were analyzed as there was high variability in the concentration of these metals in the flesh of the specimens. In addition, the poor correlation indicates that the bioaccumulation of these elements in the flesh is not only a function of the concentration in the surrounding environment but also relates to other factors such as inherent characteristics of the different species, feeding habits, and even maturity of the specimen (Dietrich and Ayers 2021; Tanhan *et al*. 2022). Moreover, the sediments with high concentrations of metals are the result of the long-term deposition from the Bagacay mine, especially during high intensity precipitation that has carried the tailings and mine waste down the slope to the river and floodplains of the Taft River basin.

The PTDI (µg kg^{-1} d^{-1}) and the EDI of HM from the consumption of several mollusk species from the Taft River by the local community showed that these species can lead to an increase in HM in the humans when consumed. The consumption of this species introduces HM, including As and Cr in particular, into the body in higher amounts than PTDI. With the exception of *M. mercenaria* for the mollusk species, the amount of As entering the human body due to the consumption of these species is more than 11 times higher than the PTDI. In terms of Cr, the consumption of all four species of mollusks can be up to 44 times higher than the PTDI. For crustaceans, only *S. serrata* has a detectable amount of As but it likewise exceeds the PTDI by up to 6.3 times. The EDI for both *S. serrata* and *M. rosenbergii* exceeds the PTDI for Cr by 20–21 times. Other metals detected in the flesh of the species had EDI that did not exceed the PTDI. These results indicate that the consumption of these mollusks and crustaceans from the Taft River by local consumers will increase their intake of As and Cr, which may possibly negatively impact their health. Furthermore, among the four species of mollusks, *M. mercenaria* had the least number of HM detected, which translates to a smaller amount of HM entering the human body upon consumption. However, this does not mean that the danger of developing health problems is low; risk is always determined by the concentration of all HM in the samples, as well as the amount that an individual human body can tolerate (Keshavarzi *et al*. 2018).

To further assess the risks associated with the consumption of these mollusk and crustacean species from the Taft River, the THQ was estimated. It showed that, among the HM, chromium appeared to have the highest potential health risk with a mean value of 43.88 in mollusks and 21.45 in crustaceans. Consumption of *T. telescompium*, *N. albicilla*, and *B. violacea* will also lead to THQ equivalents of 14.93, 6.73, and 11.67, respectively, whereas the consumption of *S. serrata* from the lower and middle sections of the river will have a THQ equivalent to 6.25. In the case of Mn, high potential health risk was calculated for all mollusk species with THQ values ranging from 1.15–1.27 but none for the crustacean species. Copper, Ni, Pb, and Zn had THQ values of less than 1, indicating that the level of exposure is below the reference dose and that daily consumption has a low probability of causing any adverse effects during a person's lifetime (Keshavarzi *et al*. 2018). Continuous consumption of all sample species at a frequency of twice a week over the span of 60 yr, suggests that it would cause adverse health effects with Cr making the highest contribution. A higher HI was estimated for the consumption of the mollusk species with values of 58.33 for *T. telescompium*, 57.61 for *B. violacea*, 53.88 for *N. albicilla*, and 47.07 in *M. mercenaria*. Although lower in magnitude, consumption of the crustacean species also poses health risks to consumers with average values of 22.36 for *M. rosenbergii* and 28.57 for *S. serrata*. The high HI for all species that were studied could be attributed to the presence of elevated multi-metals in the sediments that eventually transferred to the mollusks and crustaceans through bioaccumulation. This indicates that mollusks and crustaceans could be a major source of HM transfer from sediments to a higher form of organisms, including humans, *via* biomagnification and they may, therefore, harm human health and other aquafauna in the area (Keshavarzi *et al.* 2018).

The estimates of the TCR greater than 0.0001 are considered unacceptable for the short period of exposure (Liu *et al.* 2019). The acceptable level of cancer risk for lifetime exposure has a range of 0.0001–0.000001, whereas value of lower than 0.000001 is considered acceptable (Liu *et al.* 2019). The result of this study suggests that a high cancer risk could be a consequence of consumption or direct ingestion of all sampled species.

The result also implies that the Ni and Cr levels exceeded the acceptable limit in all four species of mollusks. Therefore, even consuming these four species over a short period of time could cause cancer in local consumers. The same result is found for As with the values of 0.030524 for *N. albicilla*, 0.0052509 for *B. violacea*, 0.0067166 for *T. telescompium*, and an average of 0.0058024 for *S. serrata*. In the case of Pb, there were no associated risks for both the short period and a lifetime period of consumption of the species under consideration.

CONCLUSION

The meat of the four mollusk and two crustacean species harvested from the Taft River contained high levels of As, Cu, Ni, Cr, Pb, and Zn – all of which were beyond the acceptable limits for human consumption set by authorities. The concentration of these metals varied between mollusk species with "bebe" (*B. violacea*) having significantly higher Cu, Ni, Pb, and Sr compared to those in "tangili" (*T. telescopium*), "tuway" *(M. mercenaria*), and "sihi" (*N. albicilla*); for crustaceans, the flesh of *S. serrata* contained higher As, Cu, Ni, Rb, Sr, and Zn than *M. rosenbergii*, which could be related to their feeding habit. Consumption of *T. telescompium*, *N albicilla*, and *B. violacea* could result in an EDI for As at 4.48, 2.03, and 3.5 $\mu g\ kg^{-1}$ BW d^{-1}, respectively – whereas the same species, including *M. mercenaria*, will result in an EDI for Cr of between 124.59–136.23 $\mu g\ kg^{-1}$ BW d^{-1}. These values are higher than the PTDI for As and Cr and, consequently, resulted in very high THQ for As and Cr. All these mollusk species have HI values between 47.07–58.33, which are considered to be hazardous to human health. The consumption of *T. telescompium* and *S. serrata* for crustaceans will result in the highest TCR for adults and HI as compared to other species, which is attributed to a relatively high THQ due to high concentration of As in its flesh. Overall, results indicate that consumption of mollusks and crustaceans – especially *T. telescompium*, *B. violacea*, and *S. serrata* – from the Taft River poses some human health risks due to high HM concentrations, especially As and Cr, found in the flesh of these aquatic animals. Our initial results on the HM accumulation in the mollusks and crustaceans that were examined warrant further investigation with respect to their different growth stages and seasonal variations in the area. Given the high concentration of HM in these species, legislation for harvest restriction is recommended.

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STATEMENT ON CONFLICT OF INTEREST

The authors have no competing interests to declare that are relevant to the content of this article.

NOTES ON APPENDICES

The complete appendices section of the study is accessible at https://philjournsci.dost.gov.ph

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