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RESEARCH ARTICLE

Preliminary determination of heavy metals in sediment, water, and some macroinvertebrates in Tawi-Tawi Bay, Philippines

Ainulyakin H. Imlani[1] • Yiğit Taştan[2*] • Albaris B. Tahiluddin[3,4] • Soner Bilen[5] • Yashier U. Jumah[1] • Adem Yavuz Sönmez[5]

[1] *Mindanao State University, Tawi-Tawi College of Technology and Oceanography, College of Fisheries, Department of Aquaculture, Sanga-Sanga, Bongao, Tawi-Tawi 7500 Philippines*
[2] *Kastamonu University, Faculty of Fisheries, Department of Aquaculture, 37150 Kastamonu, Turkey*
[3] *Mindanao State University, Tawi-Tawi College of Technology and Oceanography, College of Fisheries, Department of Marine Fisheries, Sanga-Sanga, Bongao, Tawi-Tawi, 7500 Philippines*
[4] *Kastamonu University, Institute of Science, Department of Aquaculture, 37150 Kastamonu, Turkey*
[5] *Kastamonu University, Faculty of Fisheries, Department of Basic Sciences, 37150 Kastamonu, Turkey*

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ABSTRACT

Determination of heavy metals is enormously important to determine the condition of the aquatic environment in terms of chemical pollution. In this study, a preliminary determination of heavy metal concentrations in sediment, water, and some macroinvertebrates in several sampling sites along Tawi-Tawi Bay, Philippines, was undertaken to have an initial status of heavy metal pollution in the area. Results revealed that the average concentration of heavy metals followed the order of Fe > Zn > Mn > Pb > Cu > Ni > Cd for sediment, Pb > Zn > Cu > Ni > Fe > Cd > Mn for seawater, Fe > Zn > Mn > Cu > Pb > Ni > Cd for spider conch (*Lambis lambis* Linnaeus, 1758), and Fe> Zn > Pb > Ni > Mn > Cu > Cd for sea cucumber (*Holothuria scabra,* Jaeger, 1833). However, all these determined heavy metals were within the safety limits set by WHO, US (EPA and FDA), and EMA. This study suggests that despite the anthropogenic activities in the coastal areas, heavy metal contamination in Tawi-Tawi Bay has not exceeded the safety limits.

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[*] Corresponding author
E-mail address: yigittastan@gmail.com (Y. Taştan)

Introduction

Potential sources for heavy metal pollution in the aquatic environment include but not limited to mining of metals, geochemical structure, and industrial wastes (Lee & Stuebing, 1990; Gümgüm et al., 1994; Sönmez et al., 2021). Moreover, heavy metals can accumulate to toxic concentrations under some environmental conditions (Güven et al., 1999) and cause ecological harm (Harms, 1975; Jefferies,1984; Freedman, 1989).

Tawi-Tawi is located at the southern tip of the Philippines, known to be at the center of the coral triangle, endowed with great marine biodiversity. Tawi-Tawi Bay serves as the major source of juvenile fish and macroinvertebrates recruits for much of the focal areas, perhaps even for areas farther east. The bay harbors different species of macroinvertebrates that include gastropods and haliotids (Campos et al., 2007).

This bay serves as the spawning ground of all aquatic organisms, including pelagic fishes such as tunas. However, this bay also serves as a sink for a series of anthropogenic activities coming from different coastal areas. Prominent activities undertaken in this bay are mariculture, urbanization, and nickel mining activities that were recently established within the mainland of Tawi-Tawi. Seaweed farming is the main livelihood of most people thriving in these areas. To increase their seaweed production, seaweed farmers immerse their seaweeds in agricultural fertilizers (Tahiluddin et al., 2021) that have a potential source of heavy metals. Mining activities in mainland Tawi-Tawi will find their way into the bay through sedimentation. Fine sediments are known to harbor heavy metals due to their size and adhesive properties (Hwang et al., 2016). In addition, heavy maritime traffic and deep-water movements in the region are also seen as important factors that can contribute to the chemical pollution.

To our knowledge, there has been no study investigating heavy metals in water, sediment, and macroinvertebrates (spider conch and sea cucumber) in Tawi Tawi Bay, Philippines, to date. To evaluate the metal pollution of the area, we aimed to determine the heavy metal contents (Cd, Cu, Fe, Mn, Ni, Zn, and Pb) in sediment, water, and macroinvertebrates originating from the coastal region of Tawi-Tawi bay. Further comparisons with different reported data were also conducted. Our results may help in evaluating the heavy metal pollution status of Tawi-Tawi bay, Philippines, regarding the quality of macroinvertebrates, water, and sediment.

Materials and Methods

Sampling Procedure

All samples were collected in four different coastal areas (Bongao, Panglima Sugala, Banaran, and Simunul) in October 2019 within Tawi-Tawi Bay, Philippines (Fig. 1). Although Tawi-Tawi is not much affected by season, October is the peak of the wet season. Samples were collected in triplicate at each sampling site. Three individuals of spider conch and sea cucumber were collected in each sampling area. Likewise, three samples of sediment and surface water were collected in areas where the macroinvertebrates were found.

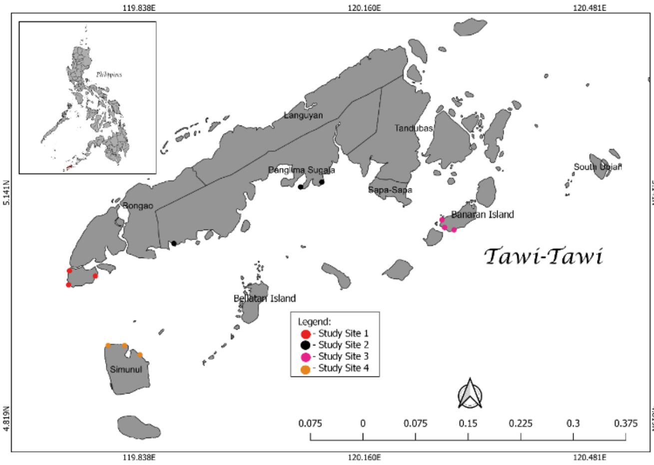

Figure 1. Map of the study sites

Water samples were collected from approximately 30 cm below the surface by immersing polyethylene bottles that were previously washed with 10% nitric acid (v/v) and rinsed thoroughly with distilled water. After collection, samples were treated with 10% nitric acid until obtaining the pH level of 2, filtered through 0.45 μm filter and stored at +4°C (Alam et al., 2001; Sönmez et al., 2012; Elderwish et al., 2019). Sediment samples were collected using a plastic shovel from the bottom (<5 cm) and placed into polyethylene ziplocked bags. The samples were then air dried in the laboratory. After drying, they were powdered, filtered through a 160-μm sieve and stored at -20°C (Öztürk et al., 2009). Macroinvertebrate samples, on the other hand, were collected by hand while freediving. Macroinvertebrate samples were washed with deionized water and stored in individual polyethylene bags at -20°C until transportation (Baharom & Ishak, 2015).

Heavy Metal Analyses

Water, sediment, and macroinvertebrate (spider conch and sea cucumber) samples were transported to Turkey by plane in a styrofoam box containing dry ice and analyzed in the Central

Research Laboratory, Kastamonu University, Turkey. Heavy metal (Cd, Cu, Fe, Mn, Ni, Zn, and Pb) concentrations in the water samples were measured directly. The heavy metal contents of the sediments were determined by digesting samples (0.5 g each) in a microwave oven for 1 h at 600 W using a high-pressure digestion vessel with 6 ml of HNO_3/HCl (v/v 2:1) as per Morillo et al. (2004). Macroinvertebrates were analyzed following the method of Sönmez et al. (2012). Briefly, samples were accurately homogenized, weighed (5 g, wet weight), and were digested with nitric acid-hydrogen peroxide (2:3) by exposing samples to 40 bar pressure microwave wet incineration units (Berghof speedwave MWS-2) in three different steps: (i) 75% microwave power for 5 min at 145°C, (ii) 90% microwave power for 10 min at 180°C, and (iii) 40% microwave power for 10 min at 100°C. The samples were then read in two parallel. All readings were conducted using inductively coupled plasma optical emission spectrometry (ICP-OES, PerkinElmer, Optima 2100 DV). Deionized water was used and all reagents used were of analytical grade. All glassware and plastic materials were washed using nitric acid for 15 minutes and were shaken using deionized water before use. The inert gas used was high purity argon. Standard solutions were prepared using stock solutions (Merck, multiple element standard). DORM-3 and DOLT-4 (National Research Council Canada, Ottawa, Canada) were used as certified reference materials.

Statistical Analysis

The data obtained were analyzed using a one-way analysis of variance (ANOVA). Levene's test was used for homogeneity analysis. Significant differences among sampling sites were further interpreted using Duncan's Multiple Range Test (DMRT). Data were presented in mean ± SE. All significant levels were set at 0.05.

Results

The obtained levels for the Cd, Cu, Fe, Mn, Ni, Zn, and Pb in the sediment, water, spider conch, and sea cucumber in the coastal areas of Tawi-Tawi Bay, Philippines are summarized in Tables 1, 2, 3, and 4, respectively.

All heavy metals evaluated in sediments showed a statistically significant difference ($p<0.05$) in all experimental sites (Table 1). The Bongao area had the highest concentration of all heavy metals ($p<0.05$). However, the concentration of heavy metals (Mn, Cu, Zn, Pb, and Ni) found in sediment is considered a non-polluted category (Abbasi & Mirekhtiary, 2020). In general, the accumulated levels of heavy metals in sediment were in the following order: Fe > Zn > Mn > Pb > Cu > Ni > Cd. Fe was the most abundant metal, while the lowest abundance was observed for Cd.

The seawater samples collected from different experimental sites showed a statistically significant difference in the levels of Cd, Cu, Fe, Zn, Mn, Ni concentration ($p<0.05$) but not in Pb ($p>0.05$) as shown in Table 2. The highest level of cadmium, iron, manganese, and zinc content was recorded in seawater collected from Simunul. At the same time, the highest copper level was manifested by the seawater collected from the Panglima Sugala and Simunul areas ($p<0.05$). However, the highest Ni level was recorded at Bongao station. In general, the accumulated levels of heavy metals in seawater were in the following order: Pb > Zn > Cu > Ni > Fe > Cd > Mn. Pb was the most abundant metal, while the lowest abundance was observed for Mn.

The results showed that heavy metals (Cu, Fe, Mn, Ni, Zn, and Pb) in different coastal areas of Tawi-Tawi Bay differed significantly ($p>0.05$) in spider conch except for Mn (Table 3). Spider conch *L. lambis* collected in Panglima Sugala had the highest Cd level (1.48±0.18) compared to other coastal areas

Table 1. Heavy metal concentrations (ppm, dry weight) in sediment samples collected from different coastal areas of Tawi-Tawi bay

Sites	Cd	Cu	Fe	Mn	Ni	Zn	Pb
1	0.45 ± 0.01^{a}	20.17 ± 0.36^{a}	$9{,}948\pm113^{a}$	34.66 ± 0.45^{a}	6.40 ± 0.14^{a}	76.99 ± 2.21^{a}	23.79 ± 0.36^{a}
2	0.29 ± 0.00^{c}	10.41 ± 0.43^{b}	$4{,}257\pm177^{c}$	20.17 ± 0.08^{b}	4.53 ± 0.08^{b}	39.01 ± 1.90^{c}	14.99 ± 0.39^{b}
3	0.34 ± 0.02^{b}	10.37 ± 0.81^{b}	$4{,}443\pm535^{c}$	25.69 ± 3.85^{b}	4.24 ± 0.12^{b}	56.85 ± 6.03^{b}	14.98 ± 0.41^{b}
4	0.29 ± 0.01^{c}	11.84 ± 0.48^{b}	$6{,}709\pm55^{b}$	25.86 ± 0.57^{b}	4.32 ± 0.19^{b}	32.02 ± 3.32^{c}	16.04 ± 0.75^{b}

Note: Data are presented as mean±standard error (n:9). Different superscript letters in the same column indicate significant differences between sampling sites ($p<0.05$). 1: Bongao, 2: Panglima Sugala, 3: Banaran, 4: Simunul.

Table 2. Heavy metal concentrations (ppb) in water samples collected from different coastal areas of Tawi-Tawi bay

Sites	Cd	Cu	Fe	Mn	Ni	Zn	Pb
1	0.85±0.01^{b}	18.91±0.29^{b}	0.54±0.09^{b}	0.79±0.01^{b}	11.44±0.03^{a}	19.37±0.25^{b}	32.92±0.12
2	0.84±0.01^{b}	24.33±0.71^{a}	2.00±0.26^{b}	0.55±0.02^{b}	11.03±0.07^{b}	25.18±0.21^{b}	33.92±1.07
3	0.85±0.01^{b}	20.40±0.53^{b}	0.95±0.07^{b}	0.74±0.01^{b}	11.34±0.06^{ab}	21.57±0.62^{b}	38.86±2.56
4	0.90±0.01^{a}	22.70±0.60^{a}	10.40±2.71^{a}	1.31±0.23^{a}	11.32±0.14^{ab}	316.63±68.19^{a}	35.47±2.98

Note: Data are presented as mean±standard error (n:9). Different superscript letters in the same column indicate significant differences between sampling sites (p<0.05). 1: Bongao, 2: Panglima Sugala, 3: Banaran, 4: Simunul.

Table 3. Heavy metal concentrations (ppm, wet weight) in spider conch (*Lambis lambis*) samples collected from different coastal areas of Tawi-Tawi bay

Site	Cd	Cu	Fe	Mn	Ni	Zn	Pb
1	0.87±0.09^{b}	3.43±0.57^{ab}	42.19±1.26^{b}	9.85±0.26	1.00±0.06^{b}	21.42±0.62^{b}	1.21±0.02^{b}
2	1.48±0.05^{a}	6.06±1.13^{a}	138.08±19.78^{a}	9.01±0.43	1.74±0.18^{a}	36.16±2.43^{ab}	1.31±0.06^{ab}
3	0.83±0.05^{b}	2.13±0.24^{b}	49.87±8.18^{b}	7.87±0.48	1.02±0.01^{b}	24.30±0.80^{b}	1.52±0.13^{a}
4	0.43±0.10^{c}	4.92±1.11^{ab}	65.83±20.64^{b}	8.03±2.17	0.50±0.16^{c}	50.62±10.39^{a}	1.23±0.03^{b}

Note: Data are presented as mean±standard error (n:9). Different superscript letters in the same column indicate significant differences between sampling sites (p<0.05). 1: Bongao, 2: Panglima Sugala, 3: Banaran, 4: Simunul.

Table 4. Heavy metal concentrations (ppm, wet weight) in sea cucumber (*Holothuria scabra*) samples collected from different coastal areas of Tawi-Tawi bay

Sites	Cd	Cu	Fe	Mn	Ni	Zn	Pb
1	0.06±0.01^{b}	0.99±0.12^{ab}	132.66±16.73^{b}	0.78±0.06^{c}	1.06±0.06^{ab}	186.48±28.30^{b}	3.58±0.02^{a}
2	0.10±0.00^{a}	1.38±0.23^{a}	319.78±64.80^{ab}	1.57±0.21^{a}	0.89±0.03^{b}	88.70±4.65^{b}	3.58±0.15^{a}
3	0.08±0.00^{ab}	0.79±0.10^{b}	462.02±94.12^{a}	1.48±0.24^{ab}	0.96±0.02^{ab}	261.18±102.08^{ab}	3.40±0.07^{ab}
4	0.08±0.00^{ab}	0.68±0.02^{b}	159.54±10.61^{b}	0.84±0.09^{bc}	1.56±0.31^{a}	573.18±141.34^{a}	3.17±0.07^{b}

Note: Data are presented as mean±standard error (n:9). Different superscript letters in the same column indicate significant differences between sampling sites (p<0.05). 1: Bongao, 2: Panglima Sugala, 3: Banaran, 4: Simunul.

within Tawi-Tawi Bay (p<0.05). Cu, Fe, and Ni results were also higher at station 2, similar to Cd values. The Zn value was measured as highest at the 4th station, and the Pb value was the highest at the 3rd station. In general, the accumulated levels of heavy metals in spider conch were in the following order: Fe > Zn > Mn > Cu > Pb > Ni > Cd. Fe was the most abundant metal, while the lowest abundance was observed for Cd.

Heavy metal results in sea cucumber (*H. scabra*) samples were statistically different at all stations (Table 4). Cd, Cu, Mn, and Pb levels were recorded as the highest in the second station. On the other hand, Fe was highest at the third station and Ni at the fourth station. In general, the accumulated levels of heavy metals in sea cucumber were in the following order: Fe> Zn > Pb > Ni > Mn > Cu > Cd. Fe was the most abundant metal, while the lowest abundance was observed for Cd.

Discussion

This study provides a preliminary determination of heavy metals in sediment, water, and macroinvertebrates in Tawi-Tawi Bay, Philippines. Our results revealed that the heavy metals (Cu, Fe, Mn, Ni, Zn & Pb) determined from different collected samples varied as presented in Tables 1, 2, 3, and 4. However, heavy metals in all samples were within the safety limits set by WHO, US (EPA and FDA), and EMA.

The average heavy metal content in the sediment samples decreased in the order of Fe > Zn > Mn > Pb > Cu > Ni > Cd. Fe content of Tawi-Tawi Bay's sediment ranged from 4,257-9,948 ppm, much greater than sediment determined in Quangzhou Bay (3.17 ppm, Yan et al., 2020) but considerably lower than the sediment sampled from Ryder Bay (14,900-26,300 ppm, Webb et al., 2020). The concentration of Zn in sediments of Tawi-Tawi Bay ranged from 32.02-76.99 ppm. In other studies, Zn concentrations vary according to the location; 7.6-145.9 in Monastir Bay (Amor et al., 2020), 48.6-538.2 ppm in Thessaloniki Bay (Christophoridis et al., 2019), 31.1-54.3 ppm in Ryder Bay (Webb et al., 2020), 95.2-443.4 ppm in Masan Bay (Lim et al., 2013), and 130.86 ppm in Quangzhou Bay (Yan et al., 2020). Mn content (20.17-34.66 ppm) in the sediment of Tawi-Tawi Bay is comparatively lower than other studies; 141.5-546.8 ppm in Thessaloniki Bay (Christophoridis et al., 2019), 211-404 ppm in Ryder Bay (Webb et al., 2020), and 612.6 ppm in Quangzhou Bay (Yan et al., 2020). Pb concentration (14.98-23.79 ppm) of the sediment obtained in the present study is relatively lesser than in Quangzhou Bay (42.18 ppm, Yan et al., 2020), Thessaloniki Bay (29.4-195.4 ppm, Christophoridis et al., 2019), and Masan Bay (29.0-82.5 ppm, Lim et al., 2013); however, it is higher than those obtained from Ryder Bay (4.8-5.0 ppm, Webb et al., 2020). Besides, the Pb concentration of Monastir Bay ranged from 0-47 ppm (Amor et al., 2020). Cu concentration sampled from the sediment of Tawi-Tawi Bay ranged from 10.37-20.17 ppm. In other Bays around the world, Cu concentrations in the sediments were 1.6-54.2 ppm in Monastir Bay (Amor et al., 2020), 21.3-180.1 ppm in Thessaloniki Bay (Christophoridis et al., 2019), 21.6-113.9 ppm in Masan Bay (Lim et al., 2013), 19.79 ppm in Quangzhou Bay (Yan et al., 2020), and 19.16-44.3 ppm in Ryder Bay (Webb et al., 2020). Ni concentrations in the sediment of the present study ranged from 4.24-6.4 ppm, which were relatively lower than those sampled from Thessaloniki Bay (41.8-171.3 ppm, Christophoridis et al., 2019), Masan Bay (15.5-46.6 ppm, Lim et al., 2013), and Quangzhou Bay (20.89 ppm, Yan et al., 2020). In addition, Ni concentrations in the sediments of Monastir Bay and Ryder Bay were 0.5-64.85 and 5.8-11.2 ppm, respectively (Webb et al., 2020; Amor et al., 2020). All heavy metals in the sediment were found to be higher in the first sampling site (Bongao) than any other site. Bongao is the most developed region among the sampling sites and it is inhabited by more people than others. The reason for high heavy metal concentration could be attributed to the high population and industrialization which may have led to increased anthropogenic activity.

The average heavy metal concentration in seawater samples was observed in the order of Pb > Zn > Cu > Ni > Fe > Cd > Mn. Pb concentrations determined from the coastal waters of Tawi-Tawi Bay ranged from 32.92-38.86 ppb, which are comparable to those sampled from Sheyang Estuary (0.15-1.57 ppb, Zhao et al., 2018), East GD coastal regions (0.0-7.7 ppb, Zhang et al., 2015), and West GD coastal regions (0.1-6.5 ppb, Zhang et al., 2015). Also, the Pb concentrations in the water of the Gulf of Cambay ranged from 27-2,203 ppb (Reddy et al., 2005). Cu levels (18.91-24.33 ppb) obtained from the water in the present study are relatively higher than those in Sheyang Estuary (1.94-7.39 ppb, Zhao et al., 2018), East GD coastal regions (0.2-9.3 ppb, Zhang et al., 2015), and West GD coastal regions (0.3-18.1 ppb, Zhang et al., 2015) but much lower compared to Gulf of Cambay (27-4,062 ppb, Reddy et al., 2005). Ni and Fe concentrations determined in the coastal waters of Tawi-Tawi Bay ranged from 11.03-11.44 and 0.54-10.4 ppb, relatively lower than those in the Gulf of Cambay with a concentration 23-1,076 and 24-3,785 ppb, respectively (Reddy et al., 2005). Cd concentrations in seawater samples in this study ranged from 0.84-0.9 ppb, which are greater than other studies such as 0.04-0.30 ppb in Sheyang Estuary (Zhao et al., 2018), 0.01-0.66 ppb in East GD coastal regions (Zhang et al., 2015), and 0.01-0.89 ppb in West GD coastal regions (Zhang et al., 2015), but comparably lesser than those in Gulf of Cambay with a concentration range from 27-4,062 ppb (Reddy et al., 2005). Mn concentrations in the water samples of Tawi-Tawi Bay ranged from 0.55-1.31 ppb, which are considerably lesser than those in the Gulf of Cambay (Reddy et al., 2005) with a concentration range from 25-5,152 ppb. Surprisingly, contrary to the sediment results, the heavy metals in the water except Ni and Pb were found to be higher in the fourth sampling site (Simunul) than the other sites. This could, somehow, be the result of a spontaneous pollution event that emerged in the region during the sampling period. However, it is inconclusive whether such event occurred. This emphasizes the need for a periodical study that determines the heavy metal concentrations at certain intervals in the region.

The heavy metals content in spider conch showed a trend in an order of Fe > Zn > Mn > Cu > Pb > Ni > Cd. Fe content (42.19-138.08 ppm) in spider conch was found to be considerably lesser than in limpet with a 1048-2866 ppm concentration (Webb et al., 2020). Zn content in spider conch in the present study ranged from 21.42-50.62 ppm. Gastropods normally contain Zn ranging from 2.2-60.5 ppm (Cubadda et al., 2001). Limpets, in particular, contain varying Zn contents such as 40.3-158 ppm (Pérez et al., 2019) and 61.2-82.6 ppm (Webb et al., 2020). Mn content in spider conch obtained in this study ranged from 7.87-9.85 ppm, while in limpets, it ranged from 8.8-41.9 ppm (Webb et al., 2020). Cu content in spider conch in this study ranged from 2.13-6.06 ppm. Cu content in gastropods ranges from 0.47-34.7 ppm (Cubadda et al., 2001).

Shellfish and limpets have been reported to contain Cu content ranging from 0.9-137 ppm (El Nemr et al., 2016) and 1.49-25.2 ppm, respectively (Pérez et al., 2019; Webb et al., 2020). Pb content in gastropods ranges from 0.06-2.18 ppm (Cubadda et al., 2001). In this study, Pb concentration in spider conch ranged from 1.21-1.52 ppm. Pb content in limpets was previously determined, ranging from 0.5-2.2 ppm (Webb et al., 2020) to 0.68-2.75 (Pérez et al., 2019). In shellfish, Pb concentration ranged from 0.2-17 ppm (El Nemr et al., 2016). Ni content (0.5-1.74 ppm) in spider conch was found to be relatively lower compared to other studies such as in shellfish (7.8-41 ppm, El Nemr et al., 2016) and limpets (0.82-8.4, Pérez et al., 2019; Webb et al., 2020). Cd content in spider conch determined in this study ranged from 0.43-1.48 ppm. These values are within the values of Cd content in gastropods (0.1-6.6 ppm, Cubadda et al., 2001); however, they are higher than those reported in limpets with 1.49-25.2 ppm concentrations (Pérez et al., 2019; Webb et al., 2020). In addition, Cd content in shellfish ranged from 0.04-1.7 ppm (El Nemr et al., 2016).

The mean heavy metal content in the sea cucumber samples (*H. Scabra*) followed the order of Fe> Zn > Pb > Ni > Mn > Cu > Cd. Fe content (132.66-462.02 ppm) in *H. scabra* obtained from the present study was determined to be relatively higher than those obtained from other sea cucumber species such as in *Stichopus herrmanni* with a concentration of 14.6 ppm (de Fretes et al., 2020), in *H. edulis* with a concentration of 39.82 ppm (Jinadasa et al., 2014) and in *H. atra* with a concentration of 11.72 ppm (Jinadasa et al., 2014). Zn content (88.7-573.18 ppm) in *H. scabra* was also found to be comparably greater than those other sea cucumber species; 2.634 ppm in *Apostichopus japonicas* (Jiang et al., 2015), 20.95 ppm in *H. edulis* (Jinadasa et al., 2014), 20.30-36.21 ppm in *A. japonicas* (Mohsen et al., 2019), and 24.38 ppm in *H. atra* (Jinadasa et al., 2014). Pb content (3.17-3.58 ppm) in *H. scabra* was found to be much higher than in *A. japonicas* (Jiang et al., 2015), *H. edulis* (Jinadasa et al., 2014), and *H. atra* (Jinadasa et al., 2014) with concentrations of 0.065, 0.03, 0.1 ppm, respectively. Ni content (0.89-1.56) in *H. scabra* was determined to be relatively similar to *A. japonicas* with a concentration of 1.18-1.77 ppm (Mohsen et al., 2019). Mn content (0.78-1.57 ppm) in *H. scabra* was observed to be comparably lower than *A. japonicas* with 16.37-58.91 ppm concentrations (Mohsen et al., 2019). Cu content (0.68-1.38 ppm) in *H. scabra* was found to be much lower compared to *H. atra* (3.18 ppm, Jinadasa et al., 2014), *A. japonicas* (1.55-8.21 ppm, Mohsen et al., 2019), *H. edulis* (1.84 ppm, Jinadasa et al., 2014), however, it was determined to be higher than in *A. japonicas* (0.179 ppm, Jiang et al., 2015). Cd content (0.06-0.1 ppm) in *H. scabra* was observed to be much lesser than in *A. japonicas* (0.161-0.85 ppm, Jiang et al., 2015; Mohsen et al., 2019), and relatively similar with *H. edulis* (0.11 pm, Jinadasa et al., 2014) and *H. atra* (0.07 ppm, Jinadasa et al., 2014). In general, these results suggest that contents of heavy metals in sea cucumber from the coastal area of Tawi-Tawi are similar, lower, or higher than previously reported contents. It should be noted that the sea cucumbers inhabiting different areas were compared and the heavy metal contents among sampling sites varied. However, the chemical contents between different species as well as individuals of the same species are under the influence of many factors including but not limited to collection site and time, gender, habitat, age, feeding habits, etc. (Morgano et al., 2011). Nonetheless, to our knowledge, there has been no study regarding the heavy metal contents in Tawitawi Bay, Philippines, and the reliable data concerning the heavy metals in sea cucumbers and spider conch are also limited.

Several reports designating anthropogenic activities as the main source of heavy metals contamination have been documented in different parts of the globe. Like in the case of metal concentrations in Korean coastal sediment, which are significantly influenced by human activities associated with urbanization (Hwang et al., 2016). The same report on heavy metals in the nearshore sediment of Daya Bay was all closely related to the import of anthropogenic activities (Qu et al., 2018). Anthropogenic influences on heavy metals accumulation were also observed along the Karachi coast, Pakistan (Saher & Siddique, 2019). Likewise, anthropogenic sources of contamination of heavy metals such as Zn, Cd, Cu, and Pb were also evident in the coastal sediments from the coastal areas of the Bohai Sea and the Yellow Sea (Tian et al., 2020). Sediments of Edku at the Deltaic coast of Egypt receiving sewage and untreated wastewater were also heavily polluted with heavy metals (Keshta et al., 2020). Influence by anthropogenic activities was also suggested as contamination and risk of heavy metals found in the northwest coastal area of the Bohai Sea (Wang et al., 2020). Domestic effluents and urban runoff were suggested as the sources of Zn contamination in New York Bight. Corrosion within the urban water supply network may contribute to the Zn levels in domestic effluents significantly. Contamination of the urban stormwater as well as zinc release from the roofs as a result of the corrosion could also elevate the Zn levels in near water sources where the runoff waters are discharged into (Amiard & Amiard-Triquet, 1993).

The correlation of anthropogenic activities with heavy metals (Cu, Ni, Pb, and Zn) was evident in marine surface sediments of the Thessaloniki Bay, Northern Greece (Christophoridis et al., 2019). Enrichment of heavy metals such as Cd, Cu, Ni, and Zn in coastal sediments of the Dammam, Al-Jubail area, Saudi Arabian Gulf was reported to be originating from anthropogenic sources (El-Sorogy et al., 2018). Wastewater discharges were also seen as one of the major contributors to heavy metal pollution in the surface sediments of Monastir Bay (Eastern Tunisia, Mediterranean Sea) (Amor et al., 2020). Sediment contamination associated with Cu, Zn, Pb, Mn, and Cd has occurred in Quanzhou Bay, southeast China since the 19th century caused by domestic emission (Yan et al., 2020). Jiangsu coastal environment in China was also affected by heavy metals pollution of Cu, Pb, Cd, and Zn, and these were strongly influenced by anthropogenic activities (Chen et al., 2020). The significant anthropogenic sources of heavy metal contamination were reported in Sabratha coastal sediments. High levels of presence in the sediment of heavy metals were defined as an anthropogenic component, as Karthikeyan et al. (2020) observed in the surface sediment of Emerald Lake, India. They found that almost all the sites were polluted by heavy metals. The anthropogenic origin of Pb contamination was also reported in the coastal sediments of the Egyptian Mediterranean coast (El Baz & Khalil, 2018). Various anthropogenic activities along the Daya Bay, South China Sea, were reported to contribute mainly to the heavy metal contamination (Liu et al., 2018).

Conclusion

In conclusion, this study revealed that the heavy metals (Cu, Fe, Mn, Ni, Zn & Pb) in the sediment, water, and macroinvertebrate samples collected from different sites along Tawi-Tawi Bay, Philippines, varied greatly. The mean concentration of heavy metals decreased in the order of Zn > Mn > Pb > Cu > Fe > Ni > Cd for sediment, Pb > Zn > Cu > Ni > Fe > Cd > Mn for seawater, Fe > Zn > Mn > Cu > Pb > Ni > Cd for spider conch, and Fe> Zn > Pb > Ni > Mn > Cu > Cd for sea cucumber. However, heavy metals in all samples in the present study were determined to be within the safety limits set by WHO, US (EPA and FDA), and EMA. This implies that despite urbanization and industrialization in the coastal areas of Tawi-Tawi province, Philippines, heavy metal levels, especially in the commercially important macroinvertebrates such as spider conch and sea cucumber, do not exceed the safety limits; hence, they cannot be considered as polluted. Moreover, it is also important to have frequent and periodical heavy metal monitoring as the other sites constantly receive heavy metal pollution pressures.

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Compliance With Ethical Standards

Authors' Contributions

AHI: Sampling, transportation of the samples, article writing
YT: Statistical analyses, article reviewing & editing
AT: Article reviewing & editing
SB: Article reviewing & editing
YUJ: Article writing
AYS: Study design, data evaluation, article reviewing & editing

Conflict of Interest

The authors declare that there is no conflict of interest.

Ethical Approval

For this type of study, formal consent is not required.

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