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Seasonal variation of heavy metals (Cu, Mn, Ni and Zn) in farmed green mussel (*Perna viridis*) in Marudu Bay, Sabah, Malaysia

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Abstract: Concern over consumption safety of farmed green mussel (*Perna viridis*) in Marudu Bay rises due to the increasing agricultural activities taking place around the bay. This urges us to conduct a study to determine the status of heavy metals (Cu, Mn, Ni and Zn) content in the mussel and sediment samples from the aquaculture site and its relationship with the environmental parameters. Samples of 120 mussels and sediment were collected and analyzed monthly for one year period. Water parameters including temperature, pH, salinity, dissolved oxygen and current speed were collected *in-situ* during sampling. The mean content of Cu, Mn, Ni and Zn in the mussel was at 45.81±46.94, 48.55±22.21, 8.66±5.02 and 46.72±17.38, respectively. In sediment, the mean content of the metals was at 39.76±36.13, 276.24±189.32, 61.61±17.83 and 55.99±54.49, respectively. Current study also found that, the mean contents of Cu, Mn, Ni and Zn were generally higher during northeast monsoon and lower during southwest monsoon. In addition, the Mn content in green mussel was above the permissible limit. Hence an in-depth study is needed to ascertain the sources of Mn in the farmed green mussel in Marudu Bay.

Keywords: Heavy metal, *Perna viridis*, sediment, Marudu Bay

Introduction

Heavy metals can cause deleterious health impact to human being (Jaishankar, 2014). Industrialization, urbanization and non-stop human activities such as mining, can contribute to the deposition of more heavy metals in the environment (Tchounwou *et al*., 2012). Anthropogenic wastes including toxic metals from earth surface can enter the ocean through runoffs (Rzymski *et al*., 2014; Ferreira *et al*., 2004; Ponnusamy *et al*., 2014). Such condition raises a great concern worldwide (Jarup, 2003) as the introduction of more heavy metals into the ocean will alter the properties of marine environments and exposes marine organisms to heavy metals-related problems, and poses doubt on seafood (Zodape *et al*., 2011). Marine organisms especially filter feeders are known to accumulate toxic materials including heavy metals in their bodies (Yap *et al.*, 2004; Yusof *et al*., 2004; Kamaruzzaman *et al*., 2011; Abdullah *et al*., 2007; Ponnusamy *et al*., 2014) by means of bioaccumulation. The heavy metal bioaccumulation in bivalve has been extensively studied as the content of heavy metals in bivalve could reach at alarming level (Fang *et al*., 2001; Hossen *et al*., 2015; Dabwan and Taufiq, 2016). One of the main factors which greatly influence the heavy metals bioaccumulation in marine organisms is the environmental factor (Denton and Burdon-Jones, 1981; Barua *et al.,* 2011).

Bivalve such as green mussel (*Perna viridis*) is one of most sought after seafood restaurants in Sabah, Malaysia. The source of this bivalve is either from the wild or farmed at the natural marine environment. One of the popular bivalve aquaculture sites in Sabah is in Marudu Bay (Tan and Ransangan, 2016a). However, the escalation of palm oil plantation activity surrounding the bay may contribute to excessive heavy metals loading in the bay and feared to end up bioaccumulated into the farmed mussels. Such condition is not only detrimental to the health of consumers but also affect economics of the mussel aquaculture in the area. Hence in this paper, we report the status of heavy metals content in farmed green mussels and sediment of Tanjung Batu, Marudu Bay, Sabah, Malaysia and its relationship with the environmental parameters.

Materials and Methods

Sampling Site

Green mussel and sediment samples were collected from Tanjung Batu, Marudu Bay, Sabah, Malaysia (6° 35' to 7° N, 116° 45' to 117° E). The sampling site is

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close to fishermen village and also a prominent place for fishing activities. Raft culture oysters and green mussels are among the main economic activities in the area.

Environmental Parameters Measurement

Water

All water parameters were taken at 0.5 m below water surface. Multiparameter (YSI; Loveland, Co, USA) was used to measure water temperature, salinity, and dissolved oxygen *in-situ*. Water current speed was also measured *in-situ* by using current meter (Stanley, USA).

Sediment

The organic carbon matter in sediment were determined by drying 5 g of wet sediment sample until constant weight obtained then heated in a muffle furnace at 550 °C for 6 hours and estimated according to Parsons *et al.* (1984). Clay-silt percentage was measured by using a laser diffraction particle size analyzer (Sequola, Canada) according to Agrawal and Pottsmith (2000).

Sample Collection

Green mussels

The specimens were collected from raft culture facility (6 m x 6 m). Ten individuals of green mussels (7.4 to 11.7 cm) were taken randomly at monthly interval for a period of 12 months (Feb 2014 to Jan 2015). The specimens were transported to the laboratory in polystyrene box containing ice cubes. The specimens were then dissected by using acid washed stainless steel knife. The whole soft tissues were kept in zip-lock bag then stored in –80 °C ultralow freezer (U570-86, New Brunswick). Prior to further analysis, the specimens were oven-dried at 105 °C for 24 hours (Carvalho *et al*., 2000). Then, the dried specimens were ground to powder using mortar and pestle and kept in zip-lock bags.

Sediment

Sediment samples were collected from seabed of the culturing site (Tanjung Batu) by using a bottom ponar grab sampler (Ponar 6''x6'' All SS, Wildco). Triplicate samples were taken monthly for a period of 12 months (Feb 2014 to Jan 2015). Sediment samples were stored in plastic bags and kept chilled in polystyrene box containing ice cubes throughout transportation to laboratory. The samples were air-dried for 7 days and grounded to powder using mortar and pestle. Individual sample was kept in zip-lock bags prior to heavy metal analysis.

Sample Digestion

The green mussel and sediment samples were acid digested using hot block digestor (Tecator Digestor Auto 8, FOSS) according to Yap *et al*. (2008). All glassware used in this study were rinsed with 1 N nitric acid. 0.5 g of powdered sample was weighed using electronic balance (Pecisa 404A) and placed in digesting tubes. 15ml of 65% nitric acid (Merck) and 2.5ml of 70-72% perchloric acid (Merck) were added in the tubes. The tubes were then placed on the hot block and left heated at 140 °C for 10min. Then, 5 ml of 37% hydrochloric acid (AR Grade, Qrec) were added in the tubes. The temperature was raised to 200 °C and digestion process proceeded for 3 hours. When digestion completed, the tubes were left to cool at room temperature before adding 1.25 ml of 65% nitric acid and 10ml of double distilled water. The solution was filtered using Whatman filter paper (No. 1001-110) and top-up to 35ml with double distilled water. Subsequently, the filtrates were kept in 20ml glass vials and stored at 4 °C until further analysis.

Heavy Metal Determination

Heavy metal content of the samples was determined by using Inductively Couples Plasma- Optical Emission Spectroscopy (ICP-OES), Perkin Elmer Optima 5300DV. Quality Control Standard 21 (Perkin Elmer Pure) was used as Standard Reference Material (SRM) for instrument recovery. Triplicate readings were obtained for every sample. Assessment of measurement quality and precision was achieved by running blank samples and certified standard materials Dogfish liver DOLT-5 and Marine sediment HISS-1 (National Research Council Canada) alongside with the samples.

Statistical Analysis

SPSS Windows Statistical Package (version 22.0) was used for all statistical analyses. All data were subjected to one-way ANOVA with all variables tested for normality and homogeneity of variances. Significance level was set at $p<0.05$. One-way ANOVA was performed and followed by the Tukey multiple comparison tests (Tukey HSD) to make specific contrast in temporal variation of environmental parameters (temperature, pH, salinity, DO, current speed, organic carbon matter and clay-silt composition), heavy metals (Cu, Mn, Ni and Zn) content in mussels and heavy metals content in sediment. Correlation coefficients were then calculated between main environmental variables and heavy metal content.

Results

Temporal Variation of Environmental Variables

The temporal variations of environmental variables in Tanjung Batu, Marudu Bay are illustrated in Table 1. Temperature ranged between 27.67 and 31.45°C, pH ranged between 7.80 and 8.30, salinity ranged between 25.60 and 34.23 ppt, current speed ranged between 6.50 and 63.13 m/min and Dissolved oxygen (DO) ranged between 2.87 and 6.06 mg/l (Tab. 1). Current speed was generally higher during northeast monsoon compared to southwest monsoon (Fig. 1). Other water parameters did not fluctuate throughout the study period (Fig. 1).

Tab. 1: Environmental variables recorded in Tanjung Batu throughout the study period.

Month	Tem. (°C)	pH	Salinity (ppt)	Current (m/s)	DO (mg/L)	Organic carbon matter (%)	Clay-silt composition (%)
Feb	27.67	8.03	30.03	15.60	2.87	7.80	47.20
Mar	28.51	7.80	30.39	25.80	4.39	7.89	51.85
Apr	29.38	8.10	34.23	36.50	6.06	7.83	58.03
May	27.96	8.03	33.54	36.13	4.17	6.75	43.04
June	31.45	8.30	33.47	39.61	5.63	7.91	51.92
July	30.70	8.18	31.67	63.11	5.93	8.39	59.14
Aug	31.05	8.04	25.60	25.20	5.12	8.20	56.61
Sept	29.90	8.20	31.54	40.40	4.28	8.55	58.86
Oct	29.20	8.04	30.74	16.19	4.49	8.82	55.66
Nov	30.10	8.06	31.10	9.60	3.32	9.17	52.86
Dec	29.65	7.88	29.69	*	*	8.15	66.10
Jan	27.80	8.12	32.94	6.63	3.75	7.68	69.25
Mean	29.44 (±1.23)	8.06 (±0.12)	31.24 (±2.24)	28.61 (±16.07)	4.54 (±1.01)	8.09 (±0.66)	55.88 (±7.18)
Min	27.67	7.80	25.60	6.50	2.87	6.14	43.00
Max	31.45	8.30	34.23	63.13	6.06	9.30	70.11

Note: * = missing value

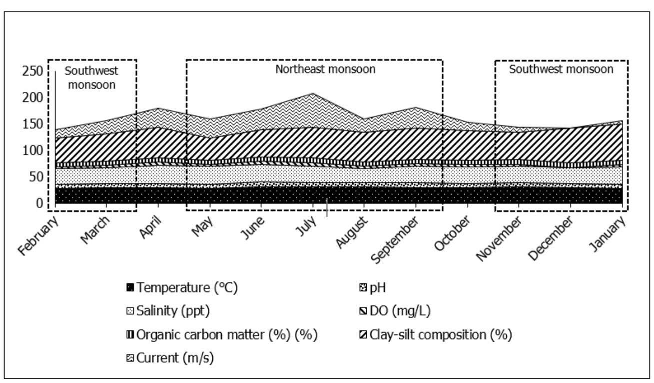

Fig 1: Environmental parameters recorded in Tanjung Batu, Marudu Bay throughout the study period

Organic Carbon Matter and Clay Silt Composition of the Aquaculture Site

The organic carbon matter in surface sediment of Tanjung Batu was recorded between 6.14 and 9.30%. The surface sediment of the site was mainly composed of clay silt at 43.0 to 70.11% (Tab. 1).

Heavy Metals in Green Mussel

The recovery of the certified reference material for heavy metals content in green mussel and sediment was satisfactory at 77% to 90%. Temporal variations of respective metal elements in green mussel tissue collected from Tanjung Batu are summarized in Table 2. The content of Cu ranged between 2.16 and 189.83 mg/kg, Mn ranged between 6.17 and 112.32 mg/kg, Ni ranged between -0.48 and 21.69 mg/kg and Zn ranged between 12.24 and 115.03 mg/kg (Tab. 2).

Cu content in green mussel did not show significant difference ($p>0.05$) throughout the study period. Mn was significantly higher ($p<0.05$) in December 2014 (66.02 mg/kg) than in March (23.86 mg/kg), May (22.63 mg/kg), July (19.95 mg/kg) and

August 2014 (24.54 mg/kg) but did not significantly different with that in April (54.81 mg/kg), September (44.57 mg/kg), November 2014 (54.99 mg/kg) and January 2015 (62.09 mg/kg). Ni was significantly higher ($p<0.05$) in February (8.57 mg/kg), April (8.31 mg/kg), May (8.18 mg/kg) and December 2014 (7.83 mg/kg) than in July 2014 (1.38 mg/kg) but did not significantly different with that in March (7.02 mg/kg), June (3.15 mg/kg), September (6.12 mg/kg) to November 2014 (5.60 mg/kg) and January 2015 (7.49 mg/kg). Zn was significantly higher ($p<0.05$) in April 2014 (49.71 mg/kg) and January 2015 (58.02 mg/kg) than in October 2014 (26.47 mg/kg) but did not significantly different than those in the other months (Tab. 2). The study found that the mean contents of Cu, Mn, Ni and Zn in farmed green mussels was generally higher during the northeast monsoon and lower during southwest monsoon

Tab 2: Heavy metals content in green mussels collected from Tanjung Batu throughout the study period.

Month	Cu (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Feb	35.52±56.78 ^{a}	40.14±16.89 ^{ab}	8.57±4.38 ^{c}	45.75±21.07 ^{ab}
Mar	14.44±20.60 ^{a}	23.86±18.08 ^{a}	7.02±3.49 ^{bc}	40.08±32.36 ^{ab}
Apr	15.73±18.33 ^{a}	54.81±39.12 ^{bcd}	8.31±5.03 ^{c}	49.71±18.02 ^{b}
May	10.09±12.64 ^{a}	22.63±8.23 ^{a}	8.18±6.25 ^{c}	40.93±15.54 ^{ab}
June	10.52±8.26 ^{a}	37.85±5.66 ^{ab}	3.15±1.63 ^{abc}	39.32±9.33 ^{ab}
July	27.95±28.21 ^{a}	19.95±7.07 ^{a}	1.38±1.11 ^{a}	40.40±7.77 ^{ab}
Aug	28.58±38.05 ^{a}	24.54±7.78 ^{a}	2.01±1.01 ^{ab}	39.92±11.56 ^{ab}
Sept	27.04±30.12 ^{a}	44.57±12.91 ^{abcd}	6.12±5.05 ^{abc}	38.49±6.46 ^{ab}
Oct	16.29±26.62 ^{a}	34.53±18.37 ^{ab}	4.97±2.38 ^{abc}	26.47±6.96 ^{a}
Nov	7.76±3.67 ^{a}	54.99±20.52 ^{bcd}	5.60±3.73 ^{abc}	39.71±11.20 ^{ab}
Dec	46.95±60.97 ^{a}	66.02±14.64 ^{d}	7.83±3.84 ^{c}	48.21± 7.61 ^{ab}
Jan	9.48±1.39 ^{a}	62.09±6.52 ^{cd}	7.49±2.26 ^{bc}	58.02±8.09 ^{b}
Mean	20.81±32.13	40.47±22.60	5.88±4.28	42.28±16.07
Min	2.16	6.17	-0.48	12.24
Max	189.83	112.32	21.69	115.03

Heavy Metals in Sediment

Temporal variation of heavy metals in surface sediment of Tanjung Batu is illustrated in Table 3. Cu content in surface sediment was ranged between 10.56 and 163.63 mg/kg, Mn ranged between 11.80 and 650.27 mg/kg, Ni ranged between 26.84 and 96.74 mg/kg and Zn ranged between 17.57 and 285.16 mg/kg. Cu, Mn, Ni and Zn content in sediment did not significantly fluctuate ($p>0.05$) throughout the study period (Tab. 3).

Correlation between Heavy Metals in Sediment and Environmental Variables

Elements such as Cu and Ni in sediment showed no significant correlation ($p>0.05$) with the environmental variables recorded throughout the study period. Mn was positively correlated with clay silt composition ($r^2=0.39$, $p<0.05$). Zn was positively correlated with DO ($r^2=0.43$, $p<0.05$) and clay silt composition ($r^2=0.44$, $p<0.05$) (Tabs. 4 and 5).

Correlation between Heavy Metals in Green Mussel and in Sediment

Nickel in green mussel showed significant positive but weak correlation with Cu in the sediment ($r^2=0.37$, $p<0.05$). Zn in both green mussel and sediment showed significant weak positive correlation ($r^2=0.39$, $p<0.05$) (Tab. 6).

Discussion

Temporal Variation of Heavy Metals in Green Mussel and Sediment

It was found that the mean content of Cu, Mn, Ni and Zn in the farmed green mussel was generally higher during the northeast monsoon and lower during southwest monsoon (Fig. 2). The finding is in agreement with previous studies (e.g Otchere, 2003; Ferreira *et al*., 2004; Barua *et al*., 2011). Rainy season during the northeast monsoon carries surface soil and runoff from land into the bay (Talwar *et al*.,

2014) which could explain the high heavy metals contents observed in the present study. Domestic and anthropogenic wastes from the land which washed off into the bay during heavy rain contributed heavy metals load into the water body (e.g Ferreira *et al*., 2004; Rzymski *et al*., 2014; Ponnusamy *et al*., 2014) and increased their bioavailability

Tab. 3: Heavy metals content in sediment of Tanjung Batu throughout the study period.

Month	Cu (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Feb	55.04±41.73	90.06±109.29	43.04±11.50	78.04±11.20
Mar	23.21±9.15	101.05±124.43	59.02±29.29	139.29±86.30
Apr	33.66±19.21	87.43±125.52	45.25±8.13	24.98±2.11
May	79.93±51.85	367.71±43.94	59.05±8.36	39.32±0.60
June	42.66±28.75	144.95±111.83	58.46±26.26	108.64 ±152.87
July	23.65±3.66	472.21±155.25	66.52±14.77	45.37±5.39
Aug	23.36±2.62	426.62±116.75	72.36±14.34	44.98±4.55
Sept	99.02±68.93	430.87±85.06	73.99±14.71	40.01±11.73
Oct	16.32±4.49	256.27±208.19	70.93±5.18	30.91±11.56
Nov	20.88±8.95	106.44±77.10	57.70±22.62	25.87±8.08
Dec	21.10±4.27	409.56±128.87	64.95±18.03	46.95±11.42
Jan	38.26±32.41	421.77±197.97	68.04±24.85	47.58±9.46
Mean	39.76±36.13	276.24±189.32	61.61±17.83	55.99±54.49
Min	10.56	26.84	26.84	17.57
Max	163.63	96.74	96.74	285.16

Tab. 4: Pearson's correlation between heavy metals in green mussels and environmental variables.

	Tem.	pH	Salinity	Current	DO	Organic Content	Clay Silt Composition
Cu	-	-	-0.41*	-	-	-	-
Mn	-	-	-	-	-	-	0.38*
Ni	-0.53**	-0.35*	-	-	-	-0.44**	-
Zn	-0.46**	-0.52*	-	-	-	-	-

Note: *= p<0.05; **= p<0.01; - = p>0.05

Tab. 5: Pearson's correlation between heavy metal in sediment and environment variables.

	Tem.	pH	Salinity	Current	DO	Organic Content	Clay Silt Composition
Cu	-	-	-	-	-	-	-
Mn	-	-	-	-	-	-	0.39*
Ni	-	-	-	-	-	-	-
Zn	-	-	-	-	0.43*	-	0.44**

Note: *= p<0.05; **= p<0.01; - = p>0.05

Tab. 6: Pearson's correlation between heavy metal in both green mussels and sediment.

		Sediment			
		Cu	Mn	Ni	Zn
Green Mussel	Cu	-	-	-	-
	Mn	-	-	-	-
	Ni	0.37*	-	-	-
	Zn	-	-	-	0.39*

Note: *= p<0.05; **= p<0.01; - = p>0.05

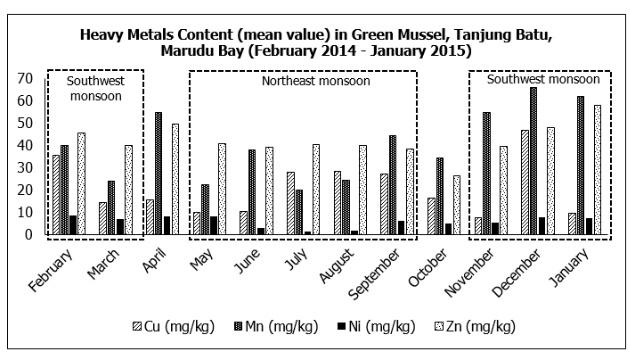

Fig. 2: Mean content of heavy metals in green mussels farmed in Tanjung Batu, Marudu Bay (February 2014- January 2015).

Temporal variation of heavy metals content in sediment of Tanjung Batu, Marudu Bay was not significantly different throughout the study period. However, higher heavy metals content in sediment was observed during the southwest monsoon but contradicted with the heavy metals content in the green mussels. This can be explained by the nature of heavy metals that tend to associate with sediment during low water volume input (Rajan *et al*., 2012). Generally, the southwest monsoon in Malaysia brings less rain making the season to be drier and hotter (Tan and Ransangan, 2016b.). In contrast, northeast monsoon brings much rain which increases volume of runoff from the land and causes water turbulences in the bay. Such condition brings more heavy metals inputs from the land and causes heavy metals deposit to redissolve into the water column.

Heavy Metals Content in Green Mussel and Sediment

Green mussels farmed in Tanjung Batu, Marudu Bay contained Zn that comparable to the finding of studies conducted in the west-coast of Peninsular Malaysia (75.1- 129.0 mg/kg) (Yap *et al*., 2004) and in Pekan, Pahang (45.54 mg/kg) (Kamaruzzaman *et al*., 2011). Meanwhile, Cu content in green mussel of the present study was found higher than that in Mengkabong, west-coast of Peninsular Malaysia and Pekan, Pahang (2.26, 7.76- 20.1 and 19.05 respectively) (Abdullah *et al*., 2014; Yap *et al*., 2004; Kamaruz-zaman *et al*., 2011). However, the Cu content of present study are comparable with some studies done on other bivalve species (Tab. 7). Ni and Mn content in green mussel are, nonetheless, less studied in Malaysia but finding of the present study showed their contents are comparable with other studies conducted on other bivalve species (Tab. 7). The abundance, in decreasing order, of heavy metals content in green mussel in the current study is Mn>Zn>Cu>Ni. Unlike in green mussels, the metal elements (Cu, Mn, Ni and Zn) in sediment of Marudu bay are comparable and fallen within the records of other studies (Tab. 8). The abundance, in decreasing order, of heavy metals in sediment of Tanjung Batu, Marudu bay is Mn>Ni>Zn>Cu.

By comparing the results of the current study with the permissible limits of heavy metals consumption stipulated by national and international standards (Tab. 9), Mn content in farmed green mussels seems exceeded the permissible limit of 5.4 mg/kg set by FAO/WHO (1984).

Tab. 7: Comparison of heavy metals content of bivalve soft tissue in Malaysia.

Site	Species	Weight basis	Year	Cu (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)	Ref.
Tg. Batu, Marudu bay, Sabah	*Perna viridis*	d.w.	2014-2015	45.81	48.55	8.66	46.72	1
Mengkabong, Sabah	*Perna viridis*	d.w.	-	2.36	-	-	16.52	2
Kota Kinabalu markets, Sabah	*5 species of commercial bivalves	w.w.	-	3.21-36.22	-	-	28.62-1771.2	3
Likas estuary, Sabah	*Meretrix meretrix*	d.w.	-	4.26-10.30	-	-	50.3-223.5	4
Likas estuary, Sabah	*Crassostrea iredalei*	d.w.	-	4.08-47.17	-	-	77.2-1445.6	4
Kota Belud Estuary, Sabah	*Meretrix meretrix*	d.w.	-	0.82-3.93	-	-	42.6-149.8	4
Moyan, Sarawak	*Solen regularis*	d.w.	2007	5.2	67.3	-	-	5
Serpan, Sarawak	*Solen regularis*	d.w.	2007	6.5	48.9	-	-	5
Peninsular Malaysia (West Coast)	*Perna viridis*	d.w.	1999-2001	7.76-20.1	-	-	75.1-129.0	6
Peninsular Malaysia (Jeram, Kuala Kuru & Kuala Kurau)	*Anadara granosa*	d.w.	2005	5.41-7.39	-	0.80-16.15	91.89-203.47	7
Kuala Perlis, Perlis	*Marcia marmorata*	d.w.	-	21.58	-	-	-	8
Kuala Kemaman, Terengganu	*Polymesoda expansa*	d.w.	2014	15.9	-	-	12.8	9
Kuala Kemaman, Terengganu	*Anadara granosa*	d.w.	2014	56.5	-	-	22.9	9
Bandar Baharu, Kedah	*Anadara granosa*	d.w.	-	2.01	-	-	57.20	3
Parit Jawa, Johor	*Polymesoda erosa*	d.w.	2006	6.50	-	7.80	222.0	10
Kg Pasir Putih, Johor	*Polymesoda expansa*	d.w.	2006	36.0	-	6.04	368.0	10
Pantai Remis, Perak	*Schapharca broughtonii*	d.w.	2006	4.54	-	5.26	67.80	10
Pantai Remis, Perak	*Trisidos kiyonoi*	d.w.	2006	5.84	-	4.93	61.50	10
Pasir Panjang, Negeri Sembilan	*Donax faba*	d.w.	2006	7.23	-	3.65	51.20	10
Merlimau, Melaka	*Anadara granosa*	d.w.	-	9.10	-	-	98.72	11

Tab. 7: Continued.

Site	Species	Weight basis	Year	Cu (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)	Ref.
Telok Mas 1, Melaka	*Polymesoda erosa*	d.w.	2006	5.91	-	4.29	249.0	10
Tanjung Lumpur, Pahang	*Solen brevis*	d.w.	2009	8.64	-	-	87.74	12
Pekan, Pahang	*Perna viridis*	d.w.	2009	19.05	-	-	45.54	12

Note: * *Meretrix meretrix, Anadara granosa, Tridacna squamosal, Polymesoda erosa* and *Crassostrea gigas*; d.w.= dry weight; - = not mentioned
Ref.: 1 (This study); 2 (Abdullah *et al*., 2014); 3 (Budin *et al*., 2014); 4 (Abdullah *et al*., 2010); 5 (Kanaraju *et al*., 2008); 6 (Yap *et al*., 2004); 7 (Yap *et al*., 2008); 8 (Lias *et al*., 2013); 9 (Dabwan and Taufiq, 2016); 10 (Edward *et al*., 2009); 11 (Yusof *et al*., 2004); 12 (Kamaruzzaman *et al*., 2011);

Tab. 8: Comparison of heavy metals content of sediment in Malaysia.

Site	Weight basis	Year	Cu (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)	Ref.
Tg. Batu, Marudu bay, Sabah	d.w.	2014-2015	39.76	276.24	61.61	55.99	1
Likas Estuary, Sabah	d.w.	-	45.4-142.5	-	-	286.0-420.0	2
Kota Belud Estuary, Sabah	d.w.	-	68.6-82.7	-	-	47.9-57.6	2
Moyan, Sarawak	d.w.	2007	180.6-210.4	252.8-420.1	-	-	3
Serpan, Sarawak	d.w.	2007	206.7-274.0	316.2-512.2	-	-	3
Kuala Kurau, Peninsular Malaysia	d.w.	2005	12.91	-	114.1	74.64	4
Kuala Juru, Peninsular Malaysia	d.w.	2005	32.91	-	510.93	317.4	4
Jeram, Peninsular Malaysia	d.w.	2005	17.81	-	24.01	98.97	4
Peninsular Malaysia (East Coast)	nm	2008	17.48	-	273.42	63.01	5
Peninsular Malaysia (West Port)	d.w.	2009-2010	7.4-27.6	-	7.2-22.2	23.4-98.3	6
Kuala Perlis, Perlis	d.w.	-	41.85	252.8- 420.1	-	-	7
Port Klang Coast, Selangor	nm	2009-2010	11.35-40.6	-	6.2-17.83	32.8-126.9	8

Note: - = not mentioned, d.w.= dry weight
Ref: 1 (This study); 2 (Abdullah *et al*., 2007); 3 (Kanaraju *et al*., 2008); 4 (Yap *et al*., 2008); 5 (Shaari *et al*., 2005); 6 (Sany *et al*., 2011); 7 (Lias *et al*., 2013); 8 (Sany *et al*., 2013)

Tab. 9: Guidelines on heavy metal for food set by different countries or organisations.

Guidelines provider	Cu (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Malaysian Food Regulation (MFR, 1985)	30.0	-	-	100
Food and Drug Administration of the United States (USFDA, 1993b)	-	-	80	-
Australian Legal Requirements for Food Safety (NHMRC, 1987)	350	-	-	750
Ministry of Public Health, Thailand (MPHT, 1986)	133	-	-	677
Brazilian Ministry of Health (ABIA, 1991)	150	-	-	250
FAO/WHO (1984)	-	5.4	1.0	-

Note: - = not mentioned

Correlation between Heavy Metals in Green Mussel and in Sediment

Heavy metals in marine organisms can be originated from various sources including foods, water and sediment. However, metal uptake by organisms is affected by multiple factors of both physical and chemical properties of the habitat. Studies show that sediment provides better indicator for contamination (for aquatic environment) than that of water as it subjects to water current and wind effect (Tekin-Ozan, 2008). Positive correlation of certain heavy metals in bivalves and sediment has been reported by a number of studies (e.g Astudillo *et al*., 2005; Usero *et al*., 2005; Rzymski *et al*., 2014). In this study, significant positive correlation was observed on Zn (r^2= 0.39, $p<0.05$) in sediment and green mussels. Similar positive correlation was also observed between Cu in sediment and Ni in green mussel (r^2= 0.37, $p<0.05$). This shows that Zn content in green mussel is most likely attributed to the sediment. Similar observation had been noted for Mn in other bivalve species (*Anodonta anatina*, *Anodonta cygnea* and *Unio tumidus)* from Maltanski reservoir, mid-western Poland was positively correlated with the amount of metal in sediment (Rzymski *et al*., 2014).

Impact of Heavy Metals to Green Mussel Aquaculture in Marudu Bay

Green mussel culture in Marudu Bay, which was started in 2000 experiencing mass mortality in the year 2010 (Tan and Ransangan, 2015). Tan *et al.* (2016) have found that failure of mussel culture in Marudu Bay is due to the altered phytoplankton composition following the introduction of green mussel culture in the bay. However, one cannot deny the existence of negative impact of heavy metal contamination to bivalve culture. Inflammation of gill, mucus production and significant avoidance of the sediment containing high level of heavy metals are among the symptoms of heavy metal contamination reported in bivalves (Hietanen *et al*., 1988; Burdon *et al*., 2014; Sonawe, 2015). Although no sign of acute heavy metal contamination, efforts to monitor heavy metals in Marudu Bay should be intensified to help restore the stock of green mussels in the bay as persistent pollution can cause stress reactions on bivalves which can possibly wipe out the entire stock due to mass mortality (Burdon *et al*., 2014). Apart from the effect of heavy metals on the health of the green mussel itself, consumption safety of the mussel farmed in the bay could become a health issue to consumers and lead to market rejection.

Conclusion

Heavy metals content in the farmed green mussel and sediment in Marudu Bay is most likely influenced by a synergistic effect of combined factors (physical or chemical). Manganese (Mn) in green mussel was found above the permissible limit. Prolonged heavy metals contamination might put the mussel stock in the bay at risk of mass mortality and market rejection. Thus, an in-depth study needs to be carried out to ascertain the origin of the manganese contamination in Marudu Bay.

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