Spatial and seasonal variation of metal accumulation in brown seaweed, *Padina* spp. on the South China Sea coast of Terengganu, Peninsular Malaysia

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Abstract. Heavy metal (Cu, Zn, Cd, and Pb) concentrations in seawater and brown algae *Padina* spp. from three stations located along the coastline of northeast peninsular Malaysia were evaluated from April 2014 to May 2015. Metal concentrations of these samples were analysed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The average concentration of heavy metals in dry tissues of *Padina* samples were in the following order: Zn (11.41±2.91 μg g^{-1}) > Pb (2.17±2.05 μg g^{-1}) > Cu (1.25±0.59 μg g^{-1}) > Cd (0.19±0.05 μg g^{-1}); whereas the heavy metal concentration of seawater samples followed the order of Zn (2.96±1.57 μg L^{-1}) > Cu (0.37±0.12 μg L^{-1}) > Pb (0.16±0.07 μg L^{-1}) > Cd (0.008±0.01 μg L^{-1}). Zinc was the most abundant metal in both seawater and *Padina* samples. Strong correlation was observed between the concentrations of Cu and Pb in *Padina* spp. ($r = 0.843$) and between Pb and Cd ($r = 0.722$) in seawater samples. Based on the Malaysia Marine Water Quality Criteria and Standard (IMWQS), the heavy metal concentrations were in the range of acceptance limit, except Pb in seawater which may be considered slightly polluted. The Kruskal-Wallis variance test showed that the concentrations of metals in *Padina* spp. were not significantly different for almost all the studied metals among studied areas, although significant spatial variations ($p < 0.05$) occurred for Cd with comparable higher values in Bidara and Bari beaches. Comparably, the metals concentrations of Cu, Zn, Cd, and Pb in seawater samples showed no significant variation among studied areas. The strong correlation of Cd-Cu, and Cd-Pb pairs in *Padina* spp. and seawater samples indicates that these metal pollutants were of the same source, possibly from industrial activities and sewage. In addition to assessing the heavy metal concentrations along the coast of Terengganu, this study highlights the potential of *Padina* spp. as indigenous bioindicator for detecting anthropogenic impact on the coastal marine environment. Periodical assessment along the coast of Terengganu is highly recommended to ensure the level of heavy metals is always below national standards.

Key Words: heavy metals (Cu, Zn, Cd, Pb), *Padina* spp., bioaccumulation, spatial and temporal variations.

Introduction. Coastal waters are considered as endpoints for toxic chemicals including heavy metals (Ferreira et al 2004; Hudspith et al 2016). Although heavy metals are natural constituents of the hydrosphere, their high level of accumulation in the marine environment due to various anthropogenic actions such as shipping activities, industrial and urban effluents, agricultural fertilizer use, bedrock dredging, mining, and burning of fossil fuels is a global problem - damaging not only the regional ecosystem but also human health (Förstner & Wittmann 1979). Heavy metals that enter the aquatic environment generally bind to suspended matter and accumulate in sediments. When resuspended, sedimented heavy metals may be released back into the water column and become bioavailable. Sedimented heavy metals still pose a threat to various bottom dwellers such as bivalves and crustacean species including crabs and when resuspended, heavy metals could easily be ingested by aquatic organisms. Heavy metal pollution has gained international attention as they mobilize easily up the food chain and affect human health through consumption of seafood, often caught in the coastal areas (Bosch et al 2016; Zhang et al 2016).

More than half of the coastal zones throughout Malaysia have human settlements and are important areas for artisanal fisheries, tourism and recreation. Urbanization process along the coastal zones often results in release of unwanted pollutants, especially heavy metals into the marine environment (Sarimin & Mohamed 2012; Tan et al 2016). The accumulation of heavy metals is even more severe when water current is directed to certain location during certain seasons. Based on a study done by Utoomprurkporn & Snidvongs (1998), high concentration of Cr, Pb, Cd, Ni, and Fe were found at the offshore area of Sabah, Sarawak and Brunei during July-August period as a result of water current flowing northward as the wind blows from south to north (Utoomprurkporn & Snidvongs 1998).

Located at the north east coastline of peninsular Malaysia, the Dungun River is heavily influenced by northeast monsoon. Due to its strategic location, several petrochemical industries are situated at the coastal area and near the Dungun River. Studies have been conducted to evaluate the ecological impacts in this area (Shazili et al 2006), yet little research has been focused on the assessment of bioavailability of heavy metals for bioaccumulation by marine organisms. The assessment of environmental contamination by heavy metals directly by conventional water or sediment analysis is difficult and misleading because of the biogeochemistry complexity of these elements in aquatic environments (Topcuoglu et al 2003). Therefore, biomonitors such as seaweeds have been widely employed to monitor the bioavailable concentrations of the contaminants and to assess the status of environmental pollution (Rodríguez-Figueroa et al 2009; Karthick et al 2012; Chakraborty et al 2014). Aside from being a representative of the study area, seaweeds have long lifespan, sessile, abundantly found throughout most of the world's coastal zones and have the added advantage of being able to accumulate contaminants and not being killed or severely affected by it (Anastasakis et al 2011).

In line with this, the current study is conducted to monitor and assess the accumulation of Cu, Zn, Cd, and Pb in brown seaweed *Padina* spp. collected from three beaches of the northern coast of Terengganu State. The spatial and monthly variations of these heavy metal elements off the southern coast of Terengganu will serve as a baseline for future research in this coastal zone.

Material and Method

Padina spp. collection. Samples of the brown seaweed *Padina* spp. (Figure 1), were collected monthly (April 2014 to May 2015), over a year from three sites: Site 1 – Pantai Pandak (Chendering) 05° 16.135' N, 103°11.177' E; Site 2 – Pantai Teluk Bidara (Dungun) 04°46.962' N, 103° 26.333' E; Site 3 – Pantai Bari Kecil (Setiu) 05°34.014' N, 102°51.886' E (Figure 2), in order to assess the degree of heavy metal (Cu^{+2}, Zn^{+2}, Cd^{+2}, Pb^{+2}) contamination in the area. *Padina* spp. were hand-picked gently and washed three times in seawater to remove any unwanted particles adhered to the surfaces of the seaweed specimens. Samples were collected from three distinct subset of each site. Seaweeds from each station were pooled into one sample, stored in ziplock plastic bags containing seawater and kept in a cooler, then transported to the laboratory at Institute of Oceanography and Environment (INOS), Universiti Malaysia Terengganu. In the laboratory, all samples were washed under a flow of tap water and rinsed, then washed again three times with double deionized water to remove mineral particles, organisms and other external adherent. They were then kept frozen at -20°C in ziplock plastic bags until ready for further analysis.

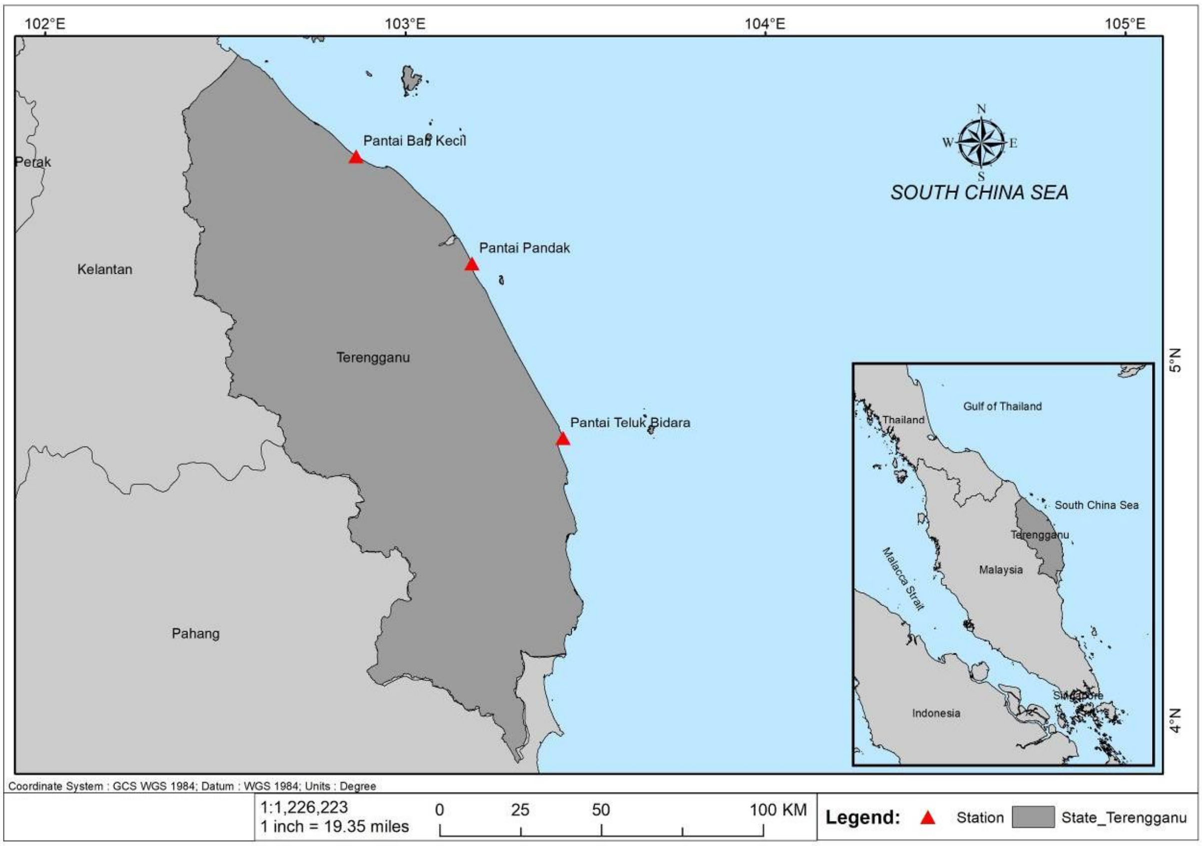

Figure 1. Map illustrating the Terengganu coast and the location of study sites.

Figure 2. *Padina* spp. collected from study area.

Seawater collection. Surface seawater samples were collected using acid-washed polyethylene bottles from each study site. Three replicates of surface seawater samples were collected from three different points of the same station. The collected seawater samples were labelled, kept on ice and transported to the laboratory within 3h. Upon reaching the laboratory, they were filtered using GF/C 0.45 µm (Whatmann), immediately preserved by the addition of a few drops of concentrated nitric acid (HNO_3) for stabilization and kept at room temperature until further analysis (Suttle 1992). The three replicate samples for each site were composited for all analytical procedures.

Determination of heavy metal in seawater. The analyses of Cd, Cu, Pb and Zn in seawater samples were conducted based on previously described solid extraction methods with slight modification (Freire & Santelli 2012; Søndergaard et al 2015). Briefly, Chelex-100 resin (5 gram of resin for every 200 mL of sample) was cleaned by soaking in 10% v/v nitric acid solution, further cleaned with ultrapure water until the solution reached a stable pH range of 6.5 to 7. Then, 50 mL of sample (seawater or standard) was added to the resin and pH was maintained between 5 and 7 using ammonium hydroxide or nitric acid. The mixtures were subjected to mechanical stirring overnight. On the following day, the mixtures were centrifuged at 2000 rpm for 30 minutes to separate the aqueous phase, and the supernatant was discarded. The trace metals were then extracted from the resin by the addition of 2.5 mL of 10% v/v nitric acid solution and 6.5 mL of ultrapure water. The mixtures were mixed for 30 minutes at 150 rpm and centrifuged (30 minutes at 2000 rpm). The supernatant (either standard solutions or seawater) was collected in a tube and introduced into the inductively coupled plasma mass spectrometry (ICP-MS) for analysis. Each sample was analysed in triplicate. One negative control (Blank) consisting of ultrapure water instead of seawater was analyzed for every 9 samples.

***Determination of heavy metals in Padina* spp**. Prior to digestion, samples were dried in oven at 80°C for 48 hr or until they reached a constant weight (Roméo & Gnassia-Barelli 1995). The dried samples were ground to a fine powder and sieved (pore size of 0.5 to 1 mm). They were then kept in a desiccator until further processing.

Digestion methods. The *Padina* spp. samples were digested in triplicates using acid digestion following the method described by Vandecasteele & Block (1993) with minor modification. Briefly, 0.2 g of the samples were transferred directly into a Teflon digestion vessel. Then, 8.0 mL of mixture of concentrated nitric acid (65%) (Merck, Darmstadt, Germany), and hydrogen peroxide (30%) (Merck, Darmstadt, Germany) (with ratio 3:1) were added to the samples and digested using microwave system ETHOS One (Milestone, CT, USA).

***Analysis of Cu, Zn, Cd and Pb in Padina* spp**. Once the digestion was completed, each sample was transferred to a 15 mL centrifuge tube and kept at 4ºC. Concentrations of Cu, Zn, Cd and Pb were determined using an Elan 9000 inductively coupled plasma mass spectrometer (ICP-MS) (Perkin Elmer Sciex, Canada). The samples were analysed together with blanks and standards (ICP multi-element standard solution VIII (24 elements in dilute nitric acid) (Merck, Darmstadt, Germany).

The concentration of each metal was estimated based on the following equations:

$$\text{Metal concentration } (\mu g/g) = (F \times C \times V) / (M)$$

where: F = dilution factor;
C = ICP-MS reading ($\mu g\ L^{-1}$);
V = sample volume (mL);
M = dry weight sample (g).

The results were expressed as total concentrations ($\mu g\ g^{-1}$) dry weight (dw). Tomato leaves (NIST SRM 1573a) were used as reference material to determine the accuracy of the digestion and analytical methods. The calculation of recovery rate was based on the following equation (Kaewsarn 2002):

$$\text{Recovery } (\%) = (\text{C observed value})/(\text{C certified value}) \times 100$$

where: C observed value = mass fraction of analyte found, mg kg^{-1};
C certified value = certified value of analyte in the certificate, mg kg^{-1}.

Statistical analysis. All data analyses were performed using SPSS for Windows, Version 19. Kruskal-Wallis variance test was conducted in order to compare the average of metals concentration and to check for any spatial (between three groups of sampling area) or monthly significant differences between the average concentration of heavy metals in *Padina* spp. Mann-Whitney test was used to determine if significant difference occurred among groups. All tests assumed significant level at 5%.

Results

Heavy metal accumulation in seawater and Padina* spp*. along the coast of Terengganu. The overall mean and standard deviation of concentration of Cu, Zn, Cd and Pb (on dry weight basis) in seawater and *Padina* spp. are presented in Table 1. The concentration of the metals in the seawater of the study areas varied considerably following the order of Zn > Cu > Pb > Cd. It was observed that Zn was the most abundant metal in our study area with mean value of 2.96 μg L^{-1} while Cd and Pb were observed in trace amounts (mean value 0.008 and 0.16 μg L^{-1}, respectively). The mean concentration of Cu (0.37 μg L^{-1}) was approximately 10x lower than that of Zn. The average metal concentration in dry tissue of *Padina* spp. however, did not follow exactly the same concentration order as in seawater, with Zn (11.52±2.94 μg g^{-1}) > Pb (2.17±2.05 μg g^{-1}) > Cu (1.25±0.59 μg g^{-1}) > Cd (0.19±0.05 μg g^{-1}). Overall, the results of the Spearman correlation test showed that the most positive significant correlation were between Cu-Pb pair (r = 0.843, p < 0.05, n = 72), followed by Cu-Zn pair (0.416, p < 0.05, n = 72), Cd-Pb pair (0.39, p < 0.05, n = 72), and Zn-Cd pair (0.29, p < 0.05, n = 72). When narrowed down to a specific location, Zn-Cd pair (r = 0.624, p < 0.05, n = 24) showed positive significant correlation in Chendering while Cu-Pb pair (r = 0.99, p < 0.05, n = 21) were the most correlated pair in Teluk Bidara.

Table 1

Concentration of Cu, Zn, Cd, and Pb in seawater (μg L^{-1}) and *Padina* spp. (μg g^{-1} dry wt)

Sample		*Cu*	*Zn*	*Cd*	*Pb*
Seawater	Overall	0.37±0.12	2.96±1.57	0.008±0.008	0.16±0.07
	Range	0.003-0.73	0.05-7.39	0.0005-0.045	0.001-0.50
Padina	Overall	1.25±0.59	11.52±2.94	0.19±0.05	2.17±2.05
	Range	0.66-3.59	5.65-23.55	0.06-0.35	0.77-10.30

Spatial and temporal variation of heavy metal accumulation in seawater and Padina* spp*. The average concentration of Cu, Zn, Cd, and Pb in seawater samples did not vary significantly among the three sampled locations along the coast of Terengganu (p > 0.05) as shown in Figure 3. Significant differences were observed in the concentration of Cu, Cd and Pb retrieved from *Padina* spp. among locations, with Teluk Bidara having the highest concentration of all three heavy metals (Figure 4). Although the concentration of Zn did not vary significantly among locations, its concentration was 10-fold higher than the other metals.

When analysed temporally, the concentration of Zn showed increasing trend from June and peaked at November, followed by sharp decrease to its lowest recorded concentration during December. It then increased slightly to approximately 3 μg L^{-1} for several months before decreasing to a lower concentration in April (Figure 5) (p = 0.001). The other three heavy metals showed almost similar concentration patterns, with a high concentration during June 2014 and a decreasing trend towards May 2015 (all p ≤ 0.002).

Unlike in seawater samples, the concentration of Zn in *Padina* spp. increased from April and peaked at September 2014, then decreased through October and November before increasing again in December 2014. It then decreased gradually until May 2015 (p = 0.003) (Figure 6). Similarly, the concentration of Pb found in *Padina* spp. also varied significantly among different sampling periods, with two high peaks observed during May (4.43±4.26 μg g^{-1} dry wt) and December (5.4±0.70 μg g^{-1} dry wt) 2014 (p = 0.001). Although observed only in trace amount in the extract of *Padina* spp., the concentration of Cu and Cd varied temporally (p = 0.002 and 0.017, respectively), with the concentration of Cu always higher than that of Cd.

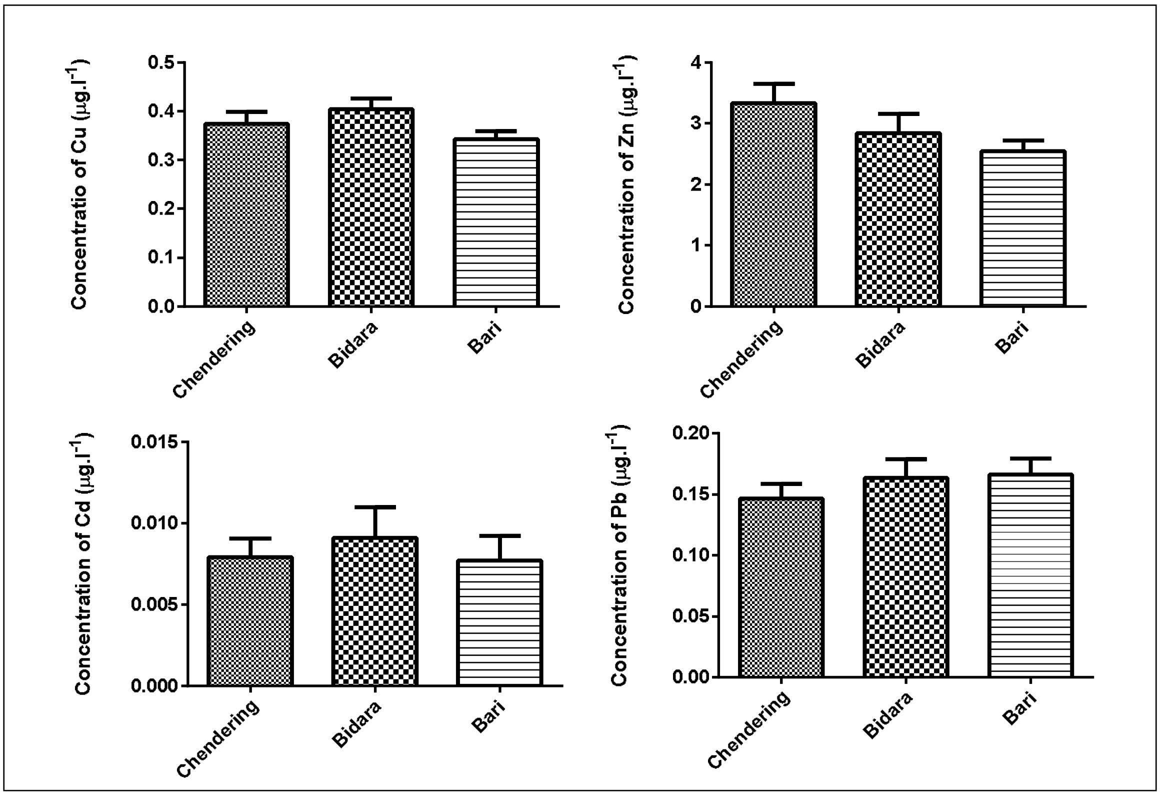

Figure 3. Heavy metal concentrations in seawater (µg L^{-1}) samples collected from the three study areas.

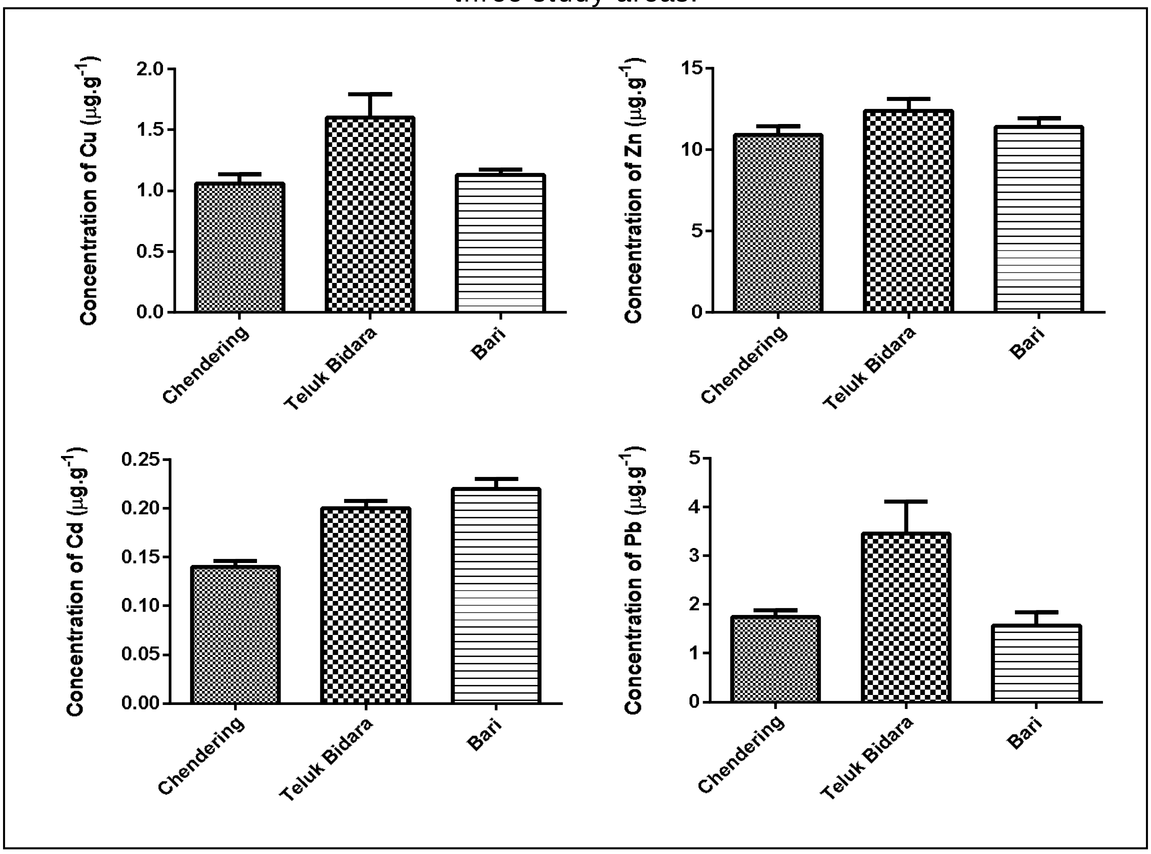

Figure 4. Heavy metal concentration (mean±SD), in *Padina* spp. (µg g^{-1} dry wt) samples collected from Chendering, Teluk Bidara, and Pantai Bari.

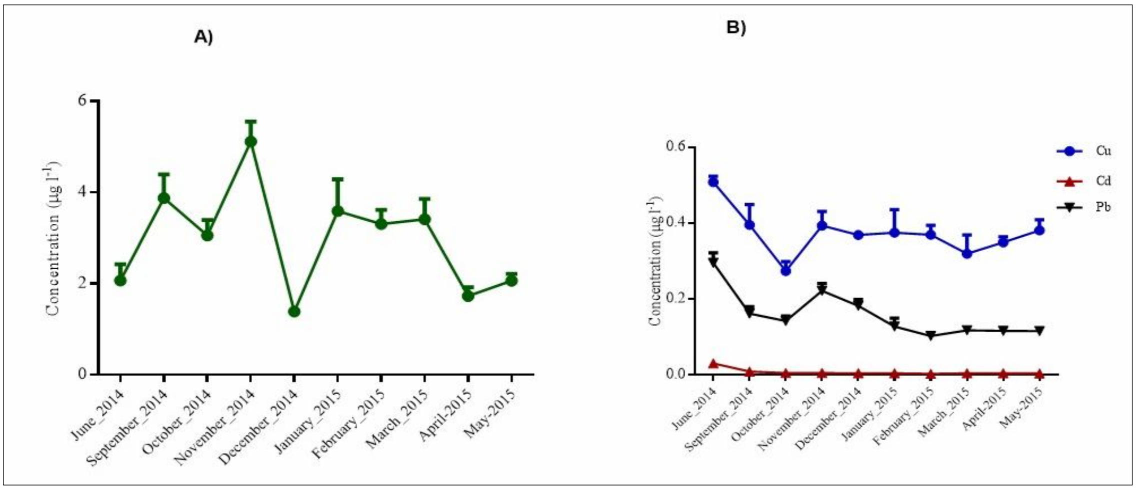

Figure 5. The overall temporal concentration of (A) Zn and (B) Cu, Cd, and Pb in seawater samples.

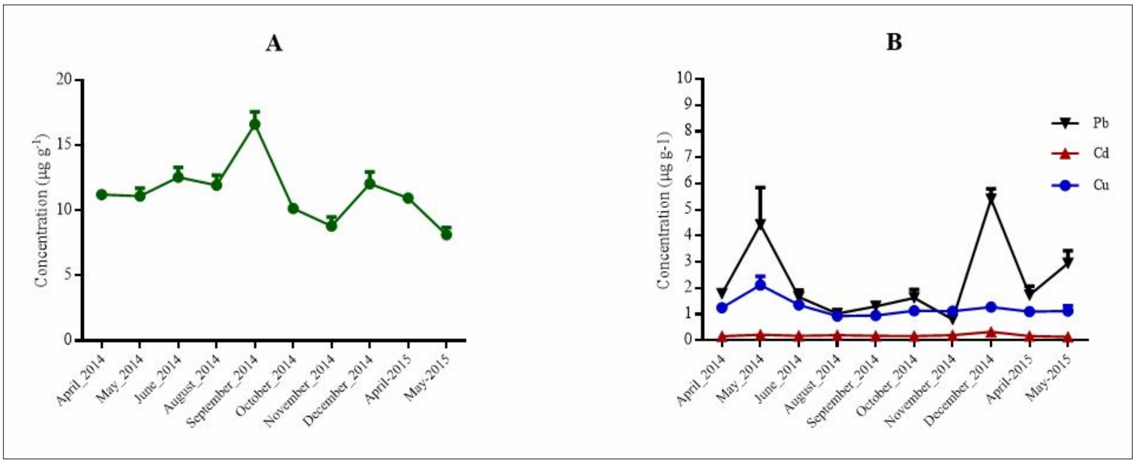

Figure 6. The overall temporal concentration of (A) Zn and (B) Cu, Cd, and Pb in *Padina* spp. samples.

Temporal variation of heavy metal concentration in seawater and Padina spp. according to location

Chendering. From seawater samples, the maximum concentration of Zn in Chendering was observed in September with an average of 5.49 µg L^{-1}, followed by November, while the lowest concentration was detected in December 2014, followed by April and May 2015, with value 1.69, 1.19, and 1.64 µg L^{-1}, respectively. The maximum Cd levels were found in June with an average of 0.03 µg L^{-1}, followed by September, November, and December with same value of 0.01 µg L^{-1}. However, the lowest concentration of Cd was detected in all the remaining months with range between 0.005 and 0.006 µg L^{-1}. In regards to the concentration of Pb, the highest concentration was observed in June with 0.27 µg L^{-1}, followed by September 0.22 µg L^{-1} (Figure 7).

There were significant differences recorded in the concentration of heavy metals in *Padina* spp. ($p < 0.05$) except for Cd ($p > 0.05$) in Chendering (Figure 6). Cu, Pb showed higher concentration in June and May 2014, respectively. Zn presented higher concentration in September, and June 2014, while Cd ranged from 0.13-0.17 µg g^{-1} from April 2014 to April 2015 and dropped to 0.09 µg g^{-1} in May 2015 (Figure 8).

Teluk Bidara. The concentration of Cu in seawater from Teluk Bidara was highest in January 2015 (Figure 7). The maximum concentration of Zn was detected in November 2014 with 6.71 µg L^{-1}, while the lowest Zn concentration was observed in December 2014. Pb was detected in trace amounts in the seawater, with maximum and minimum concentration of 0.34 $\mu g\ L^{-1}$ in June and 0.096 $\mu g\ L^{-1}$ in April 2015, respectively. Similarly, the highest level of Cd was recorded in June 2014 with 0.37 $\mu g\ L^{-1}$ (Figure 7).

Significant temporal variations ($p < 0.05$) were observed for all heavy metals in Teluk Bidara except for Cd ($p > 0.05$) (Figure 6). Cu and Pb concentrations were higher (3.57 $\mu g\ g^{-1}$, 10.09 $\mu g\ g^{-1}$ respectively) during May 2014, while Zn concentration was highest (19.1 $\mu g\ g^{-1}$) in September 2014 (Figure 8).

Pantai Bari. Highest concentration of Zn was recorded in seawater samples from Pantai Bari in March 2015. The concentrations of Cu, Pb and Cd were highest in June 2014, with an average concentration of 0.56 $\mu g\ L^{-1}$, 0.27 $\mu g\ L^{-1}$ and 0.031 $\mu g\ L^{-1}$, respectively (Figure 7).

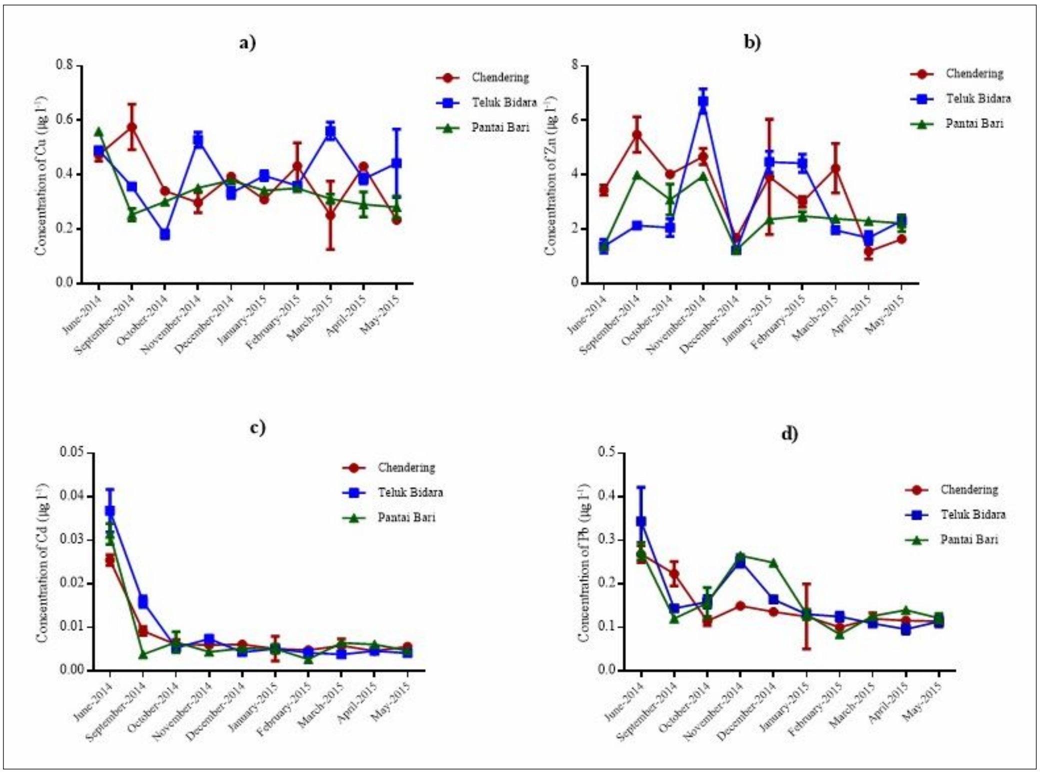

Figure 7. Monthly variation in heavy metals (Cu, Zn, Cd and Pb) concentration of seawater from Chendering, Teluk Bidara, and Pantai Bari.

Overall, there were significant temporal variations in all metal concentrations extracted from *Padina* spp. in Pantai Bari beach (Figure 6). Different heavy metals, however, showed different accumulated concentrations at different times. The peak concentrations for Cu, Zn, Cd and Pb retrieved from *Padina* extracts obtained in Pantai Bari were observed in May, September and December (for both Cd and Pb) 2014, with 1.49 $\mu g\ g^{-1}$, 15.95 $\mu g\ g^{-1}$, 033 $\mu g\ g^{-1}$ and 5.37 $\mu g\ g^{-1}$ respectively (Figure 8).

The bivariate correlation analysis as depicted in Table 2 showed significant correlation between Pb_{Padina} - Cu_{Padina} ($r = 0.843$). Weak correlations were also demonstrated between Cd_{Padina} - Cu_{Padina} ($r = 0.416$), Pb_{Padina} - Cd_{Padina} ($r = 0.392$), and Zn_{Padina} - Cd_{Padina} ($r = 0.288$). While there was moderate correlation between $Pb_{Seawater}$ - $Cd_{Seawater}$ ($r = 0.722$), weak correlations were observed between $Cd_{Seawater}$ - $Cu_{Seawater}$ ($r = 0.424$), $Pb_{Seawater}$ - $Cu_{Seawater}$ ($r = 0.464$), $Zn_{Seawater}$ – $Cu_{Seawater}$ ($r = 0.226$).

When the heavy metal data of *Padina* spp. and seawater were analysed together, significant positive correlation ($p < 0.01$) was found between $Cu_{Seawater}$ – Cu_{Padina} ($r = 0.447$), $Cd_{Seawater}$ – Cu_{Padina} ($r = 0.541$), $Pb_{Seawater}$ – Cu_{Padina} ($r = 0.447$) and $Zn_{Seawater}$ – Pb_{Padina} ($r = 0.44$); while significant negative correlation ($p < 0.05$) was found between $Zn_{Seawater}$ – Cu_{Padina} ($r = -0.250$) and $Zn_{Seawater}$ – Pb_{Padina} ($r = -0.364$). Weak correlation between $Pb_{Seawater}$ – Cu_{Padina} ($r = 0.291$) and $Pb_{Seawater}$ – Zn_{Padina} ($r = 0.281$) were also observed.

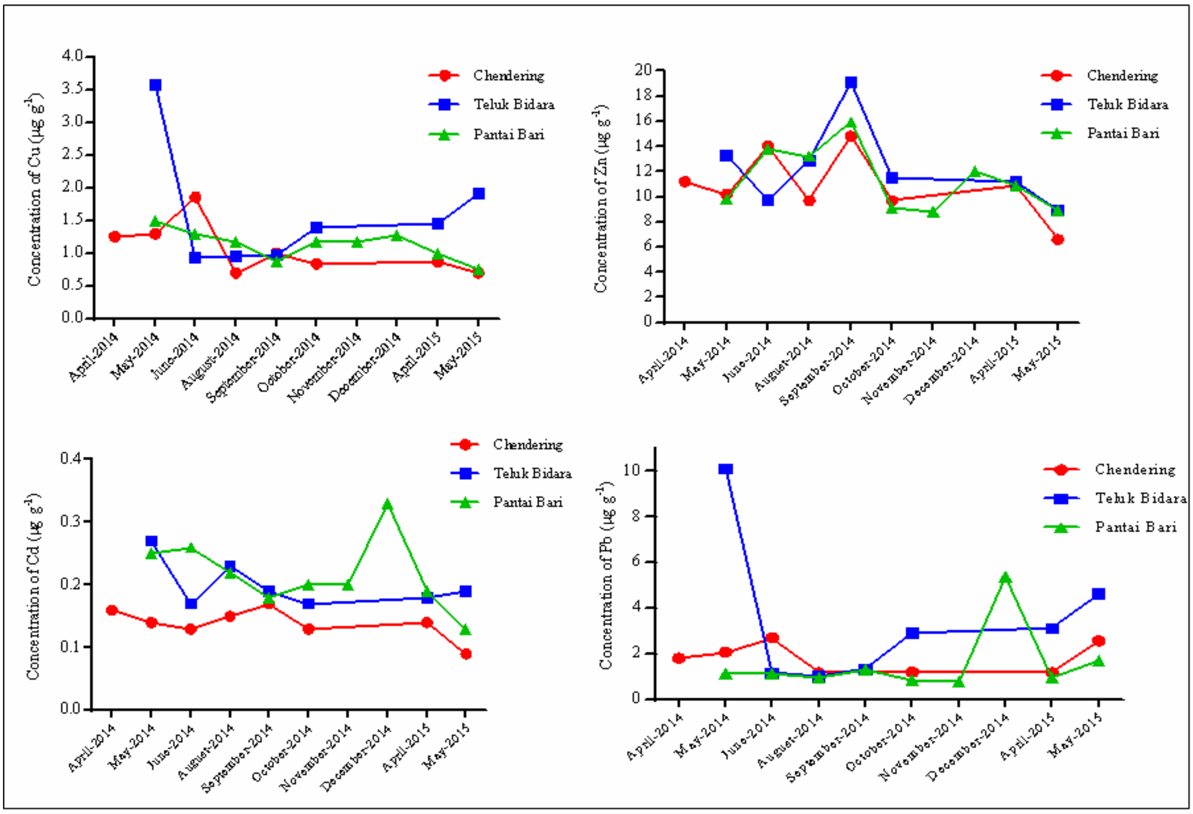

Figure 8. Temporal concentrations of Cu, Zn, Cd and Pb μg g^{-1} (in dry wt.) in *Padina* spp. at Chendering, Teluk Bidara and Pantai Bari.

Table 2

Correlation between elements of heavy metals in seawater and *Padina* spp.

	Cu_{Padina}	Zn_{Padina}	Cd_{adina}	Pb_{Padina}	$Cu_{Seawater}$	$Zn_{Seawater}$	$Cd_{Seawater}$	$Pb_{Seawater}$
Cu_{Padina}	1							
Zn_{Padina}	0.142	1						
Cd_{Padina}	0.416**	0.288**	1					
Pb_{Padina}	0.843**	0.054	0.392**	1				
$Cu_{Seawater}$	0.447**	0.096	0.179	0.252*	1			
$Zn_{Seawater}$	-0.250*	0.004	0.055	-0.36**	0.226*	1		
$Cd_{Seawater}$	0.541**	0.070	-0.015	0.44**	0.424**	-0.128	1	
$Pb_{Seawater}$	0.291*	0.281*	0.018	0.210	0.464**	0.169	0.722**	1

*. Correlation is significant at the 0.05 level (2-tailed).
**. Correlation is significant at the 0.01 level (2-tailed).

Of the four measured heavy metals, Zn had the highest recovery rate (99.55%), followed by Cd (96.71%) and Cu (90.64%) (Table 3). Due to the unavailability of Pb certified value, its recovery rate was not estimated.

Table 3

Recovery Rate of Cu^{+2}, Zn^{+2}, Cd^{+2}, and Pb^{+2} in reference material INST SRM 1573a (tomato leaves) mg g^{-1}

Element	*Certified value*	*Obtained value*	*Recovery rate (%)*
Cu	4.70±0.14	4.26±0.38	90.64
Zn	30.9±0.7	30.76±0.77	99.55
Cd	1.52±0.04	1.47±0.14	96.71
Pb	-	0.84±0.08	-

Discussion. Seaweeds play an important role in monitoring heavy metal accumulation in the marine environment and have been used as effective tool to develop policy and strategy for the environmental remediation. Their ability to bind only to free metal ions, of which the concentrations depend on the nature of the suspended particulate matter, allow assessment of the biologically available levels of contaminants in a given ecosystem (Akcali & Kucuksezgin 2011). Thus, the change in heavy metal contents in seaweeds directly reflects the temporal variation of the studied heavy metal concentrations (El-Din et al 2014; Bat et al 2019). In this study, *Padina* spp. was selected due to its suitability as bioindicator of heavy metals compared to other macroalgae and its availability in our study locations (Vandecasteele & Block 1993; Ferletta et al 1996). The effect of heavy metal contamination in the marine environment was evaluated in the South China Sea, because these areas are close to various anthropogenic inputs such as industrial waste and domestic sewage drainage (Shazili et al 2006; Adiana et al 2014).

Overall, *Padina* spp. recorded higher average concentration of measured heavy metals (i.e. Cu, Zn, Cd and Pb) compared to direct seawater analysis. Similar higher average concentrations of these four heavy metals were also observed in molluscs in comparison to seawater analysis at the coastal waters of Guangdong province, China (Zhang et al 2016). Additionally, the significant differences in Cu, Cd and Pb among locations retrieved from *Padina* spp. but not in seawater analysis also highlight the sensitivity of seaweed to heavy metal accumulation compared to direct seawater analysis. This could be attributed to the ability of heavy metal accumulation in aquatic organisms, thus enabling a more accurate assessment of the heavy metal availability in marine environment. Direct seawater analysis is more subjective and is influenced by various factors, including the sedimentation of heavy metals and the frequency of the tidal fluxes (low contaminant concentration during frequent tidal fluxes and vice versa) (Chaudhuri et al 2007; Mantiri et al 2019).

Further, the metal accumulation levels in *Padina* spp. followed the order of Zn > Pb > Cu > Cd along the coast of Terengganu. The concentrations of Cd, Cu and Pb in *Padina* spp. in this study is similar to a previous study (Mashitah et al 2012) in the same region while Zn is lower. Although a similar order has been reported in eastern coastal area of Guangdong, the concentration reported in our study is much lower as eastern Guangdong coastal waters, an area that is well-known for its industrial activities, including chemical engineering, toy making, clothing and mariculture (Zhang et al 2016) whereas the coast of Terengganu is less populated and has minimal industrial activities. Although the Zn concentration in *Padina* spp. was much higher than the other three metals and not significantly different among three locations, its concentration (10-12 μg g^{-1}) placed the coast of Terengganu in the range of uncontaminated zone as proposed by Say et al (1990) (< 50 μg g^{-1} for uncontaminated area, 50-150 μg g^{-1} for moderate contamination, > 150 μg g^{-1} area of high contamination). The slightly higher concentration of Zn compared to other heavy metals is reasonable as it is an essential element for biological activity including cell division and synthesis of DNA and protein of most organisms, including algae (Bhowmik et al 2010).

Additionally, the average concentration of Cu and Cd were also well within the range of areas with low pollution impact and together with Zn, these values were lower than the maximum concentration for human consumption stipulated by Malaysian Food Regulation 1985, FAO max limits for prawn, and WHO 1989 standards (Table 4). The concentration of Pb^{+2} however, was slightly higher than the maximum concentration for human consumption. The detection of Zn, Pb, Cu and Cd at this study area could be linked to increase in anthropogenic inputs such as shipping, biofouling wastes from fishing boats, river runoffs, urban as well as industrial activities, that have been previously reported by Carrillo Domínguez et al (2002); Lagerstrom et al (2016), and Bighiu et al (2017). Our findings coincide with results found by Mamboya (2007) which would suggest that the high urban development, agricultural activities and petrochemical industries along the coastline from Dungun to the Kemaman are the most likely contributing factors to the relatively high metal content.

Table 4

Summary of international standard

Standard	*Heavy metal concentration (μg g⁻¹)*				*Reference*
	Cu	*Zn*	*Cd*	*Pb*	
Malaysian Food Regulation (1985)	30.0	100	1.00	2.00	Abdel-Aty et al (2013)
WHO (1989)	30	100	0.05-5.50	2	Ferreira et al (2004)
FAO max limits for prawn	10	1000	-	-	Ferreira et al (2004)

Specifically, the highest level of Pb, Zn and Cu were found at Teluk Bidara. This is expected as several petrochemical-based factories are situated nearby along the Paka and Kerteh coastline. Thus, the higher Pb concentration might be attributed to effluent from the petroleum refinery and domestic sewage (Adiana et al 2014; Tan et al 2016). In addition, transportation activities, shipping and export terminal activities at the Kemaman estuary also serve as a major source of heavy metals (Ahmad 1996; Carrillo Domínguez et al 2002; Chiu et al 2000; Kamaruzzaman et al 2012). Similar slightly higher concentration of Pb than the standard was observed in the neighbouring state of Kelantan, of which effluent from petroleum production plants and commercial factories such as battery and paint production processes were postulated as the main contributors (Wannahari et al 2013). Other study conducted on the distribution of heavy metals in seawater and surface sediment samples in the coastal area of Sabah, Malaysia also reported that the levels of heavy metals were in the acceptable range of the limit of Malaysia Marine Water Quality Criteria except for Pb in seawater which was considered slightly polluted (Tan et al 2016).

The strong correlation of Cd-Cu, and Cd-Pb pairs in *Padina* spp. and seawater samples indicates that these metal pollutants were of the same source, possibly from industrial activities and sewage. It was shown that the correlation between Zn and other heavy metals were very low (except for Zn-Cd pair in *Padina* spp.), both in *Padina* spp. and seawater samples, suggesting that the pollution source of Zn may be different to those of other heavy metals. The significant correlation in Zn-Cd pair only existed in *Padina* spp. and suggests that the metal-metal correlation in seaweeds were different to that of seawater, possibly due to the distinct distribution mechanisms of these metal pairs in seaweeds (Zhang et al 2016; Liu et al 2018).

Although the concentrations of heavy metals, except Pb were below the recommended maximum levels allowed in the study area, environmental monitoring in the Malaysian coastline of the South China Sea is essential due to rapid urban and industrial development. The current data on metals in the *Padina* spp. are important and serve as baseline information for future monitoring and comparison purposes.

Conclusions. This study shows that the total Pb, Zn, and Cu contents in *Padina* spp. along Teluk Bidara recorded their highest levels, while the Cd was higher in Bari. The current study indicates that *Padina* spp. could be useful biomonitoring tools for heavy metal pollution in the marine environments. As seaweed is fed upon by marine organisms it is most likely the main source of anthropogenic metals for many animals such as fish and invertebrate that nourishes on them. Thus, measurements of heavy metal concentrations in the algae species may offer useful information on the transfer of potentially toxic elements from abiotic compartments (water and sediments) to higher consumers, including human beings. Consequently, constant monitoring of heavy metals concentration in these areas is recommended with the purpose of examining the strength of the routine use in marine biomonitoring, in order to fully clarify their actual temporal accumulation pattern.

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