Partitioning of heavy metals in different environmental and biotic components in the coastal waters of the Straits of Malacca, Peninsular Malaysia

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Abstract

Aim: Heavy metal contents in copepods, waters and sediments from an unpolluted Gula estuary, off a mangrove area in Perak (0455.185°N, 10027.761°E), were compared with those from a polluted Juru estuary, off an industrial area in Penang (05 19.906°N, 100 22.949°E), Peninsular Malaysia.

Methodology: Triplicate composite water, copepod and sediment samples were collected for a period of one year at one-month interval from three permanent stations in the coastal waters off a mangrove forest in Gula estuary, and near an industrial area in Juru estuary.

Results: Copepods and sediments from Juru estuary contained significantly higher ($p < 0.05$) concentrations of Zn, Cu, Pb, and Cd compared to those from Gula estuary. In fact, these heavy metal concentrations were also significantly higher ($p < 0.05$) in the water columns of Juru estuary than in Gula coastal waters. According to Pearson correlation coefficient analysis, significant correlations ($p < 0.05$) were observed between heavy metal contents in copepods and sediments (Zn, $r = 0.664$; Cu, $r = 0.603$; Pb, $r = 0.568$; Cd, $r = 0.501$), but no significant correlation ($p > 0.05$) was noted between heavy metal concentrations in water and copepods in both estuaries.

Interpretation: The concentration of heavy metals in sediments and copepods were relatively more stable than those in the water column, indicating that copepods and sediments could serve as more useful and reliable markers for environmental monitoring than seawater.

Keywords: Coastal waters, Heavy metals, Industrial area, Mangrove forest, Marine copepod, Sediment.

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Introduction

Rapid industrial developments without proper waste treatments have resulted in an increased amount of pollutants released into the environment. Heavy metals such as lead (Pb) and cadmium (Cd) are dangerous to many organisms in coastal waters as these harmful substances can cause negative impacts on organisms along the marine food chain, as well as other top consumers including humans. The threats are serious since these substances cannot be broken down by environmental processes and they are bioaccumulated and biomagnified to toxic levels (Harte *et al.*, 1991; Schüürmann and Market 1988; Shartooh *et al.*, 2013; Al-Imarah *et al.*, 2018; Hong *et al.*, 2020). With increased industrial, agricultural and other economic development activities in coastal areas, trace amount of heavy metals in the marine environment have been amplified to the levels that are unfavourable to aquatic life (Kalay and Canli, 2000; Looi *et al.*, 2013). Consequently, most heavy metals discharged into coastal waters are related to inputs from anthropogenic activities, atmospheric deposition and surface runoffs (Veena *et al.*, 1997; Bhattacharya *et al.*, 2015; Balogh *et al.*, 2016).

Heavy metal contamination in aquatic ecosystems have caused many negative consequences to environmental and human health mainly due to their accumulation in marine organisms, and consumption by top predators (such as fish, birds and marine mammals) and humans (Davies, 1978; Looi *et al.*, 2013; Zeitoun and Mehana, 2014; Fatima *et al.*, 2014). The uptake of these hazardous metals by plankton provides information and entry marine food chains (Gajbhiye *et al.*, 1985; Haris *et al.*, 2017). However, there is a considerable speculation about the partitioning of these substances in coastal waters and their transfer through various trophic levels along the marine food chain, especially in tropical areas, where food webs are complex (Wang, 2002; Zetina-Rejon *et al.*, 2003).

Studies on heavy metal contamination in the Straits of Malacca, especially in fish and shellfish, date back to more than 20 years ago (Babji *et al.*, 1979; Jothy *et al.*, 1983; Law and Singh, 1988; Yap *et al.*, 2004a, 2004b). Coastal waters bordering areas with active anthropogenic activities experience serious impacts from industrial, agricultural and urban waste discharges. Primary consumers in such areas are small copepods such as *Acartia*, *Paracalanus*, *Temora* and *Oithona* (Turner, 2004; Duggan *et al.*, 2008; Serranito *et al.*, 2016). However, information on heavy metal concentrations in lower trophic groups such as phytoplankton and zooplankton is limited (Rezai *et al.*, 1999, 2003), yet information regarding heavy metal contents in primary consumers, particularly copepods, is important to understand the impacts on higher trophic groups in the coastal food chain. Therefore, this study aimed to assess the partitioning of commonly found heavy metals including Cd, Cu, Pb and Zn in seawater, sediment and copepods in polluted and unpolluted estuaries in order to illustrate the relationship between heavy

metal contamination in different environmental (sediment and water) and biotic (copepods) compartments.

Materials and Methods

Sampling location : Three permanent stations approximately 1 km from the shore were randomly selected in the coastal waters off a mangrove forest along Gula estuary in Perak and the coastal waters off an industrial area along Juru estuary in Penang. The sampling sites were located in a relatively homogenous shallow area with a maximum depth of approximately 3 m (Fig. 1).

Sample collection : Triplicate composite water samples were collected at each sampling station using a Niskin water sampler on a monthly basis for a period of one year to cover annual seasonal changes. Samples were filter-sterilized through a 0.45 membrane filters, acidified with few drops of nitric acid (HNO₃) and kept in acid-washed sample bottles before further analyses. For the sediment analysis, samples from top 4-5 cm of sediment surface were collected using an Ekman grab and transferred to labelled acid-washed (acid bath 10% HCl) plastic bags and kept in a refrigerated box. Zooplankton samples were collected using a zooplankton net (100 µm mesh size) towed vertically from the deepest depth (approximately 20 cm from the bottom). Samples were then transferred to labelled sample bottles and kept cold in an ice-box to be carried off to the laboratory.

Analyses of metal contents

Seawater : Upon arrival at the laboratory, water samples were allowed to reach room temperature (approximately 27 °C), and 10 ml of each filtered sample was subjected to metal analyses (Cd, Cu, Pb and Zn) using an air-acetylene flame atomic absorption spectrophotometer (AAS: Perkin-Elmer Model 800).

Sediment analysis : Sediment samples were dried at 105 °C for at least 16 hr until a constant dry weight was achieved. Dried samples were then pounded and sieved through a 63 µm (mesh size) stainless steel sieve attached to an electronic sieve shaker (RETSCH 200 DIGIT USA). The sediments were shaken vigorously at 160 rpm on orbital shaker (Gymromax model 722) for three hours to produce homogenous samples. For the analysis of metal concentration in sediments, direct aqua regia method (ISO1955:Scancar *et al.*, 2000) was used. Sediment samples were then analysed using an air-acetylene flame AAS (Perkin-Elmer Model 800). Multiple-level calibration standards were used to generate calibration curves against which sample concentrations were calculated. Standard solutions were prepared from 1000 mg l⁻¹ stock solution (MERCK Spectrosol) of each metal and the data were expressed in µg g⁻¹ d.wt.

Copepods : After thawing to room temperature (approximately 27 °C), samples were sieved with a 63µm mesh size net and rinsed three times with distilled water to remove traces of salts. Copepods were sorted out from the zooplanktons before

Fig. 1 : Map of sampling location in Juru estuary, off an industrial area in Penang, and Gula estuary, off a mangrove area in Perak.

digestion. Approximately, 0.5 g - 1.0 g of copepods were digested for 3 hrs with 10 ml concentrated HNO_3 at 140 °C. The digests were then made up to 40 ml volume with deionized distilled water, after which heavy metal concentrations were analysed using AAS (Perkin-Elmer Model 800).

Quality control : To avoid possible contamination, all glassware and other materials used were acid washed. Procedural blanks and quality control samples made from standard solutions for Cd,

Cu, Pb and Zn were analysed every 5-10 samples to check for sample accuracy. Percentages of recovery for heavy metal analysis were acceptable at 80-100%. The analytical procedure for heavy metal contents in copepods was checked with the Certified Referenced Material (CRM) for dogfish liver (DOLT-3, National Research Council Canada). The recoveries of all metals in this study were considered satisfactory at 80-100%.

Statistical analyses : Mann-Whitney U-test and independent t-

test were employed to assess the concentrations of Cd, Cu, Pb and Zn in different compartments (seawater, sediments and copepods) at different sites (Juru and Gula estuaries) using statistical software SPSS version 15.0. Spearman Rank Correlation was applied to examine the correlation between heavy metal concentrations and their partitioning amongst different environmental and biotic compartments (seawater, sediments, and copepods). Euclidean distance was used to construct a similarity index as the basis for a 2-D ordination of non-metric multi dimensional scaling (nMDS) using PRIMER (version 6.1.9, PRIMER-E Ltd) software package (Clarke and Warwick 2001).

Results and Discussion

Assessment of harmful metal levels in plankton (mainly copepods) is important because planktonic organisms are often the main diet for many aquatic predators. Thus, copepods play an important role in transferring these harmful elements from water and sediments to higher trophic levels, such as fish and other commercially important invertebrates that form important marine food product for human populations. This study showed that metal concentrations in copepods from Juru estuary were significantly higher ($p < 0.05$) than those in copepods from Gula estuary throughout the study period, indicating that the industrial area in Juru contributed higher levels of metals to the estuary compared to the mangrove area in Gula. In Juru estuary, Cd, Cu, Pb and Zn concentrations in copepods ranged from 0.31 to 2.55

$\mu\text{g g}^{-1}$, 47.26 to 66.19 $\mu\text{g g}^{-1}$, 47.96 to 117.97 $\mu\text{g g}^{-1}$ and 177.32 to 256.17 $\mu\text{g g}^{-1}$, respectively (Fig. 2). However, in Gula estuary, the metal concentrations were significantly lower, ranging from 0.00 to 1.57 $\mu\text{g g}^{-1}$, 17.02 to 37.3 $\mu\text{g g}^{-1}$, 11.45 to 24.25 $\mu\text{g g}^{-1}$ and 43.69 to 73.02 $\mu\text{g g}^{-1}$ for Cd, Cu, Pb and Zn, respectively (Fig. 2). Significantly lower ($p < 0.05$) metal concentrations in copepods from Gula compared to those in Juru ($p < 0.05$) were perhaps due to the presence of undisturbed mangrove forests and absence of metal-based industries in the former. According to Tam and Wong (1999) and Yunus *et al.* (2011), mangrove ecosystems (including vegetation and sediments) have the capacity to sequester chemical compounds from river and sea waters, and hence are known as natural sinks for heavy metals. Similarly, Machado *et al.* (2002) noted that the sediment metal load in the mangrove forest, which are predominantly in trace metal forms, have strong affinity to substrates and low possibility of remobilization and uptake by organisms. In addition, Zhou *et al.* (2010) encouraged constant renewal of mangrove forest cover, because this approach would help to increase the absorption of heavy metals by the mangrove forest floor and thereby decrease their availability in the estuarine and coastal waters. In fact, studies from Balakrishnan *et al.* (2017) and Sarkar (2017) have shown that mangroves can sequester trace metals from tidal waters. In addition, Lacerda (1998) noted that mangrove roots and above ground biomass could induce changes in sediment chemistry, which would decrease heavy metal availability and remobilization into estuarine and coastal waters.

Fig. 2: Comparison of Cd, Cu, Pb, and Zn concentrations in copepods throughout the study period in Juru and Gula estuaries.

Fig. 3 Comparison of Cd, Cu, Pb, and Zn concentrations ($\mu\text{g g}^{-1}$) in sediment throughout the study period in Juru and Gula estuaries. Solid lines with an arrow indicate the International Sediment Quality Guidelines Canada (ISQ Guidelines, 1997).

Similar to copepods, Juru estuarine bottom sediments contained significantly higher ($p < 0.05$) concentration of heavy metals compared to Gula sediments throughout the year (Fig. 3). High concentration of metallic substances in Juru sediments could be due to prolonged exposure and accumulation of industrial waste discharges from the surrounding industrial area. Heavy metal levels in Juru estuarine sediment ranged from 0.24 to $1.40 \mu\text{g g}^{-1}$, 9.56 to $64.93 \mu\text{g g}^{-1}$, 15.11 to $59.46 \mu\text{g g}^{-1}$ and 72.08 to $303.59 \mu\text{g g}^{-1}$ for Cd, Cu, Pb and Zn, respectively, which were higher than their respective permissible limits (Fig. 3). These concentrations, however, were lower than those reported by Mat and Maah (1994) in the intertidal mudflat of Juru and Muda estuaries, because their study location was nearer to the pollutant source (factory outlets) compared to the present study in the estuarine area. However, the results of study was comparable to those reported by Yap *et al.* (2002, 2003) who also noted that sediments in Gula estuary had significantly lower metal concentrations compared to those in Juru area. Accumulation of non-degradable pollutions such as heavy metals in sediments over a long period of time can exceed the concentrations in water columns (Lyons and Fitzgerald, 1980; Barron, 1995; Yi *et al.*, 2008; Khan *et al.*, 2018). Under this condition, sediments can serve as a sink as well as a source of pollutants such as heavy metals. Invertebrates and fish feeding at sea bottom can transfer these heavy metals to other components along the marine food chain.

Metal concentrations in estuarine waters fluctuated inconsistently throughout the year with no distinct patterns (Fig. 4), perhaps due to water dynamics related to current and tidal changes, which in turn influenced the concentration of heavy metals at both locations. Moreover, water dynamics are naturally influenced by many factors, such as rainfall and wastewater runoffs. In fact, according to Etim *et al.* (1991), it was difficult to estimate the amount of biologically available concentration based marine water analyses alone, and this problem necessitates the analyses of chemicals in the organism itself. In Juru estuarine waters, the concentration of heavy metals ranged from 0.01 to 0.12 mg l^{-1} , 0.01 to 0.16 mg l^{-1} , 0.11 to 1.31 mg l^{-1} and 0.01 to 0.12 mg l^{-1} for Cd, Cu, Pb and Zn, respectively. However, in Gula estuary, heavy metal concentrations were significantly lower ($p < 0.05$) than those in Juru, ranging from 0.01 to 0.09 mg l^{-1} (Cd), 0.01 to 0.86 mg l^{-1} (Cu), 0.02 to 0.85 mg l^{-1} (Pb) and 0.01 to 0.07 mg l^{-1} (Zn).

Heavy metal contents in copepods and sediments were more consistent, with lower variability (maximum of eight order of magnitude) and distinct patterns, compared to those in water column, which experienced drastic fluctuation up to 16 times the lowest concentration (Fig. 2, 3, 4). A significantly higher ($p < 0.05$) concentration of heavy metals in Juru waters, copepods and sediments compared to those in Gula estuary might be attributed to industrialization and urbanization along the Juru coastal area. Wang and Fisher (1998) and Kadiene *et al.* (2019) found that copepods can completely assimilate ingested trace elements

Fig. 4 : Comparison of Cd, Cu, Pb and Zn concentrations (mg l^{-1}) in seawater throughout the study period in Juru and Gula estuaries. Solid lines with an arrow indicate the International Marine Water Quality Standard Malaysia (IMWQS Malaysia, 2005).

Fig. 5 Correlations of heavy metal concentrations (Cd and Cu) between sediments and copepods in Gula (a, c) and Juru (b, d) estuaries.

Fig. 6 : Correlations of heavy metal concentrations (PB and Zn) between sediments and copepods in Gula (a, c) and Juru (b, d) estuaries.

within 4 -15 hrs. This ability explains why the concentrations and bioavailability of heavy metals in copepods in Juru estuary were consistently detectable. The primary source of heavy metal contamination along the Juru estuary is due to factories discharging wastewaters directly into the coastal area. According to IMPAK (1998), the coastal waters along the Straits of Malacca are mainly polluted due to high input of organic and inorganic pollutants contributed mainly by human activities which could be land-based (agricultural and industrial wastes, sewage and domestic wastes, deforestation and land reclamation). In addition, sea-based activities can also be important source of pollution as the Straits of Malacca is the second busiest shipping lane in the world.

In this study, the concentration of heavy metal (Cd, Cu, Pb and Zn) in copepods showed a significant correlation ($p < 0.05$) with those at the bottom sediment of Juru estuary (Fig. 5 and 6) indicating that there was an uptake of sediment particles containing heavy metals by copepods. Kontas (2008) and Khan *et al.* (2018) suggested that the mobilization of particulates from

the bottom layer provided food particles to planktonic organisms, including copepods, although they were not in actual contact with the sediment. In fact, increased phytoplankton and chlorophyll *a* in the water column formed an important food source for the zooplankton partly due to mobilization of nutrients, especially inorganic nitrogen and phosphorus together with heavy metals from the enriched bottom sediment (Kontas 2008; Khan *et al.*, 2018). In a polluted area, the bottom sediment plays a significant role in accumulation, storage and transport of metals among different environment and biotic compartments (Solomon *et al.*, 1987). Dauvalter (1998) reported that metals in aquatic ecosystems tend to be adsorbed by suspended particulate matter that would finally sink to the bottom and become trapped in the sediment. In addition, Dawson and Macklin (1998) also noted that metals are transported either in solution or on the surface of suspended sediments as these metals have strong affinity to be adsorbed to particles (Lourna 1990; Robin *et al.*, 2012; Rauf *et al.*, 2018). In unpolluted Gula estuary, no significant correlation ($p > 0.05$) was observed between copepods and sediments, probably due to a significant lower ($p < 0.05$) concentration of

Fig. 7 : Correlations of heavy metal concentrations (Cd, Cu, Pb and Zn) between water and copepods in Juru (a,b,c,d) and Gula (e,f,g,h) estuaries.

heavy metals in this estuarine bottom sediment compared to those in the Juru area (Fig. 5, 6). Weak and/or absence of correlation between heavy metals in copepods and sediments in Gula estuary suggested that heavy metal concentrations in Gula sediments was too low to be mobilized to the water column and was not effectively transferred to the marine food chain.

No significant correlations ($p > 0.05$) were observed between heavy metals in seawater and those in copepods in both Gula and Juru estuaries (Fig. 7). According to Davis (1978), Robin *et al.* (2012), and Rauf *et al.* (2018), there are two pathways through which zooplankton accumulate metals in their bodies either by direct uptake from water or assimilation from ingested food (phytoplankton/suspended organic particles). In this study, absence of significant correlation between heavy metal contents in seawater and copepods indicated that ingestion of suspended particles would be the main pathway of heavy metal uptake by the

copepods. On the other hand, Fang *et al.* (2006) reported that the concentration of heavy metals in copepods were significantly correlated to the contaminated seawater in an outfall area, indicating that direct uptake can occur in the water column.

Results based on non-metric multidimensional scaling (nMDS) revealed two major groups with a Euclidean distance of 3.2. The first group was characterized by low metal concentrations, which mainly consisted of samples from coastal waters off Gula estuary. The second group was characterized by high heavy metal concentrations from the coastal waters off the Juru industrial area (Fig. 8). Similarly, metal concentration in copepods from Gula and Juru estuaries were separated into two groups with a Euclidean distance of 5.1 (Fig. 8). The nMDS showed distinct separation of two study sites in terms of metals contents in copepods and sediments, indicating that high metal contents in sediments would be available to copepods and would

Fig. 8 Non-metric multidimensional scaling (NMDS) plot of heavy metal concentrations in copepods, seawater and sediment components of Juru and Gula estuaries throughout one-year study period.

affect the health of marine food chain. Fang *et al.* (2006) and Kadiene *et al.* (2019) reported that copepods have high capacity of accumulating and bioconcentrating trace metals and can be used as an effective biological indicator for assessing and monitoring metal contamination in marine environment. In addition, Wang and Fisher (1998) also noted higher fraction of metals found in copepods' soft tissues compared to those obtained from water indicating higher uptake rate of metals through ingestion of food particles compared to direct uptake from dissolved phase. Nriagu (1978), Rainbow and White (1989), Wang (2002) and Robin *et al.* (2012) noted that heavy metals were highly persistent in sediment and biological compartments, such as copepods, making them suitable markers for environmental health monitoring.

Partitioning of heavy metals (Cd, Cu, Pb and Zn) in various environmental and biotic compartments (seawater, copepods, and bottom sediments) was studied in unpolluted (Gula estuary) and polluted waters (Juru estuary). The polluted Juru estuary showed

significantly higher heavy metal concentration in all compartments compared to those in unpolluted Gula estuary. This observation was supported by multidimensional scaling, which showed two distinct groups (locations) with different heavy metal contents. A positive correlation ($p < 0.05$) of heavy metal contents between copepods and sediments was found in the Juru estuary, but no significant correlation was noted in the unpolluted Gula area. Heavy metal concentrations in copepods showed in significant correlation with those in seawater in both estuaries, indicating that uptake of heavy metals by copepods was mainly through ingestion of heavy metal contaminated particulate matter rather than direct uptake from seawater. This study illustrates that copepods and sediments can serve as useful markers for environmental health monitoring due to persistent heavy metal contents in these compartments.

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