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Higher Bioavailability and Contamination of Copper in the Eastern Part of Johore Causeway: Will the Pattern Remain the Same Beyond 2020?

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ABSTRACT

The aim of this paper is to determine and discuss the Concentrations of Copper (Cu) in the different soft tissues of *Perna viridis* and surface sediments collected from western and eastern parts of Johore Singapore Causeway. In general, it is found that the different tissues of eastern mussel populations were found to have significant ($p < 0.05$) higher Cu levels than those in the western mussel populations. This indicated higher Cu bioavailability in the eastern part of causeway than that in the western part. The geochemical fractions (except for oxidizable-organic fraction) showed significant ($p < 0.05$) higher Cu levels in the eastern sediments than those in the western part of the causeway. This indicated higher Cu contamination in the eastern part of causeway than that in the western part. With consistent scientific reports of high metal levels in the eastern part of causeway between 2015-2018, it is predicted that there is a plausible constant source of anthropogenic metal contamination at the eastern part of the causeway beyond 2020 should there is no drastic effective control of the anthropogenic activities.

INTRODUCTION

The Straits of Johore (SOJ) is focused upon in this paper due to the significance from ecotoxicological point of view [1-3] such as monitoring of hydrochemistry changes in SOJ [4]. This is because of the fact that SOJ has been experiencing a fast development of different kinds of anthropogenic activities including an important cultivation site for mussels and fish [2,5]. Hamzah, et al. [6] concluded that both the western and eastern parts of the SOJ are extremely exposed to chemical pollution. According to Lim and Mohamed [7], the SOJ is one of the most polluted coastal areas in the southern part of Peninsular Malaysia. The water quality in the SOJ has long

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shown high level of metals [8]. The eutrophicated condition of the SOJ could be the reason for the occurrence of high biomass diatom blooms and formation of the hypoxic-anoxic zone in the SOJ [5].

Yap, et al. [2] reported the importance of the SOJ as a major cultivation site for the commercial green-lipped mussel *Perna viridis*. This mussel species has been suggested as a good biomonitor of heavy metal pollution since it has fulfilled many of the recommended criteria for the biomonitor [9]. The metal concentrations found in the tissues of mussels are indication of bioavailable metals in the coastal waters of the sampling sites [10]. The eastern part of the SOJ have been reported as having a lot of human activities such as fossil fuel fired electrical power plants, construction facilities, and shipping docks along [1,2,11].

The aim of this paper is to determine and discuss the Concentrations of Copper (Cu) in the different soft tissues of *P. viridis* and surface sediments collected from western and eastern parts of Johore Singapore Causeway in the SOJ, and

to discuss the possible trend of Cu contamination beyond 2020.

MATERIALS AND METHODS

The sampling sites for mussels and sediments in the Straits of Johore are shown in figure 1 and their descriptions are presented in table 1. The Cu data of all the parts of mussels collected in 2007 (KPPuteh-2, Senibong-2, and TgKupang) and 2008 (Senibong-3 and Pantai Lido) were unpublished. The Cu data for mantle, foot, muscle, and remaining soft tissues of KPPuteh-1 was cited from Yap, et al. [12] while other tissues were unpublished. The Cu data collected in 2004 (Senibong-1, Kg. Masai and Gelang Patah) for gill, mantle, foot, gonad, muscle and remaining soft tissue were cited from Yap, et al. [2] while Cu data for byssus and CS were unpublished. The Cu data collected in 2009 (KPPuteh-3 and KSMelayu) for byssus was cited from Eugene, et al. [13] while those Cu data for other tissues were unpublished.

About 20 relatively similar sized mussels (4-6 cm) were

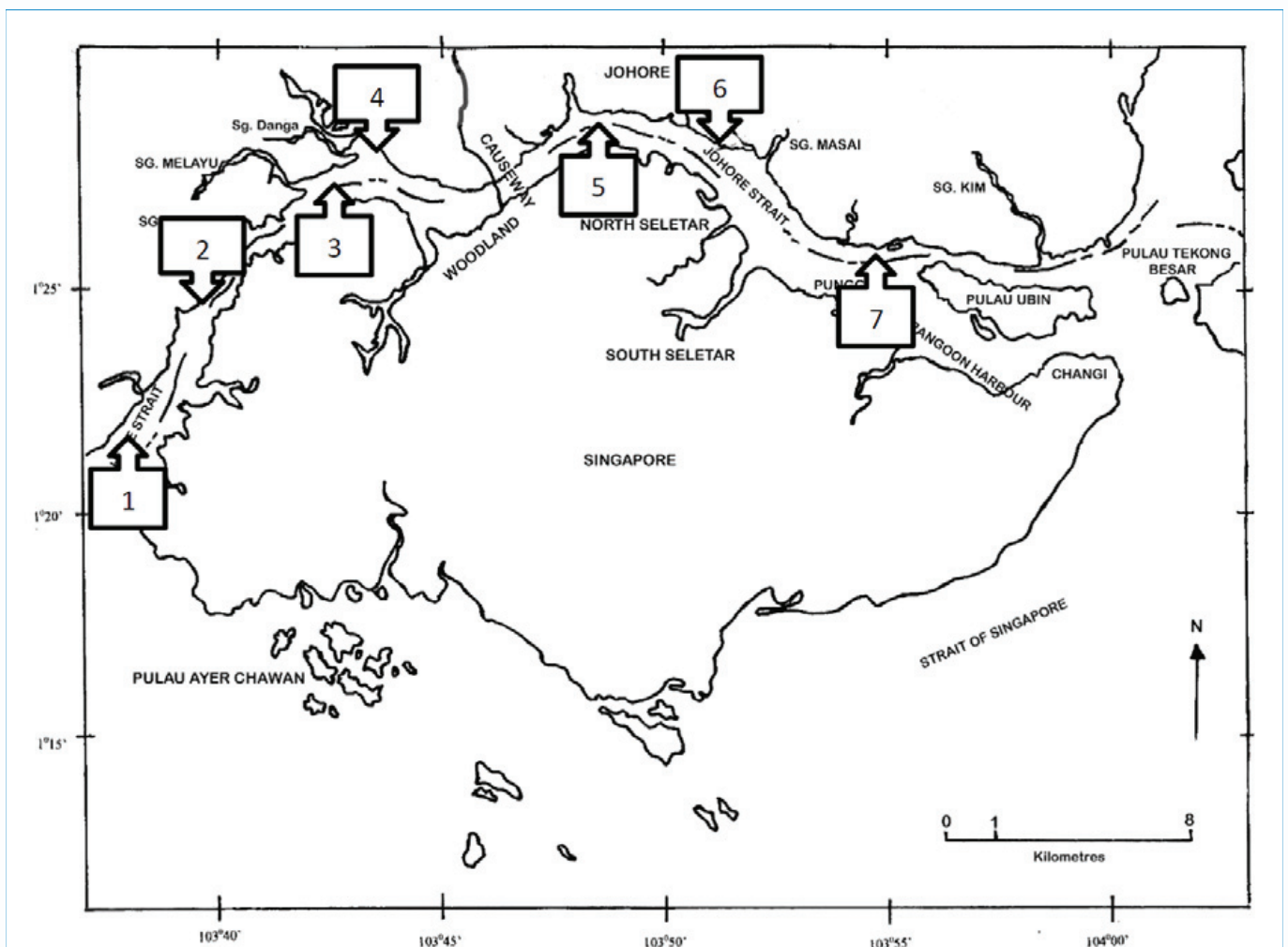


Figure 1 Map showing the sampling site of mussel *Perna viridis* and surface sediments in the Straits of Johore. The numberings of sampling sites follow those in table 1.

Table 1: The samplings sites, sampling dates and their geographical locations and site descriptions of mussels and sediments from the present study.

West Part	Sampling Site	Sampling Date	Description of Sampling Site
1	Tanjung Kupang (TgKupang)	10 May 2007	Near a mangrove Area, an aquaculture Site
2	Gelang Patah (GPatah)	11 August 2004	Fish cultivation (floating cages); mussel cultivation (floating cages)
3	Pantai Lido (PLido)	05 March 2008	Urban and agricultural area
4	Kg. Sungai Melayu (KSMelayu)	28 November 2009	Fish cultivation (floating cages); mussel cultivation (floating cages)
East part	Sampling site	Sampling date	Description of sampling site
5	Senibong-1	11 August 2004	Mussel cultivation (floating cages and buoyant); a power-generating facility was seen in the vicinity; construction activities; restaurants.
	Senibong-2	10 May 2007	
	Senibong-3	05 March 2008	
6	Kg. Masai	11 August 2004	Mussel cultivation (buoyant); a seaport.
7	Kg. Pasir Puteh (KPPuteh)-1	19 January 2005	Receiving industrial wates; municipal wastes; shipping activity in the surrounding and a marina site.
	KPPuteh-2	10 May 2007	
	KPPuteh-3	28 November 2009	

selected and dissected and pooled into byssus, Crystalline Style (CS), gill, mantle, gonad, muscle, foots and remaining soft tissues. All the samples were oven-dried at 105° C until constant dry weights and later they were digested in concentrated HNO₃ (AnalaR grade, BDH 69%), following the method used by Yap, et al. [14]. The prepared samples were determined for Cu by an air-acetylene flame Atomic Absorption Spectrophotometer (AAS) Perkin-Elmer Model AAnalyst 800.

For sediment data of geochemical fractions, only samples collected in 2004 (Senibong-1 and Kg. Masai) were cited from Yap, et al. [15] while other sediment data were unpublished. The sediment samples were analysed for Cu followed the method used by Yap, et al. [15]. Geochemical fractions of Cu in the sediment were obtained by using the sequential extraction technique which was described by Badri and Aston [16]. The four fractions used in this study were Easily, Freely, Leachable or Exchangeable fraction (EFLE; F1), Acid-Reducible fraction (AR; F2), Oxidizable-Organic fraction (OO; F3), and Resistant fraction (Res; F4).

Besides all glassware and equipment used were acid-washed, procedural blanks and quality control samples made from the standard solutions for Cu were analysed in every five samples to confirm the Cu analysis in this study. Percentage recoveries for heavy metal analyses were between 80 - 110 %. The analytical procedures for the mussels were checked with the Certified Reference Material (CRM) for dogfish liver (DOLT-3, National Research Council Canada), with 85% Cu recovery (certified Cu measured (certified value, 31.2 mg/kg; measured value, 26.5 mg/kg). For sediment analysis, the quality of the method used was checked with a CRM for Soil (International Atomic Energy Agency, Soil-5, Vienna, Austria), with 88.3% Cu recovery (certified value, 77.1 mg/kg; measured value, 68.1 mg/kg).

For the statistical analysis, *t*-test analysis was performed for the Cu levels in the different parts of mussels and

geochemical fractions of sediments, between the eastern and western parts of causeway are significantly different (*p* < 0.05). The *t*-test analysis was conducted by using STATISTICA StatSoft Inc. version 8.0 for Windows.

RESULTS

The concentration of Cu in the different tissues of *P. viridis* collected from the SOJ are presented in figure 2. Obviously, the Cu levels in the eastern sampling sites (7 populations from 3 sampling sites) of Johore Causeway are higher than those in western sampling sites, especially for remaining soft tissues, muscle, foot, gonad, gill, mantle and byssus. The concentration of Cu in geochemical fractions of the surface sediments collected from the SOJ are presented in figure 3. Obviously, the Cu levels in the eastern sampling sites (7 populations from 3 sampling sites) of Johore Causeway are higher than those in western sampling sites, especially for F1, F3, F4 and summation of all geochemical fractions.

The comparison of Cu concentrations between east part (7 populations from 3 sites) and west part (4 populations from 4 sites) of Johore Causeway in the different parts of *P. viridis* collected from the SOJ are given in table 2. It is found that overall Cu levels the eastern populations are significantly (*p* < 0.05) higher than those in the western populations for byssus, gills, mantle, foot, gonad, muscle and remaining soft tissues. The comparison of Cu concentrations between east part (7 sampling periods from 3 sites) and west part (4 sampling periods from 4 sites) of Johore Causeway in geochemical fractions of the surface sediments collected from the SOJ are given in table 3. It is found that overall Cu levels the eastern sediments are significantly (*p* < 0.05) higher than those in the western populations for F1, F3, F4 and summation of all geochemical fractions.

DISCUSSION

Generally, most of the different parts of soft tissues

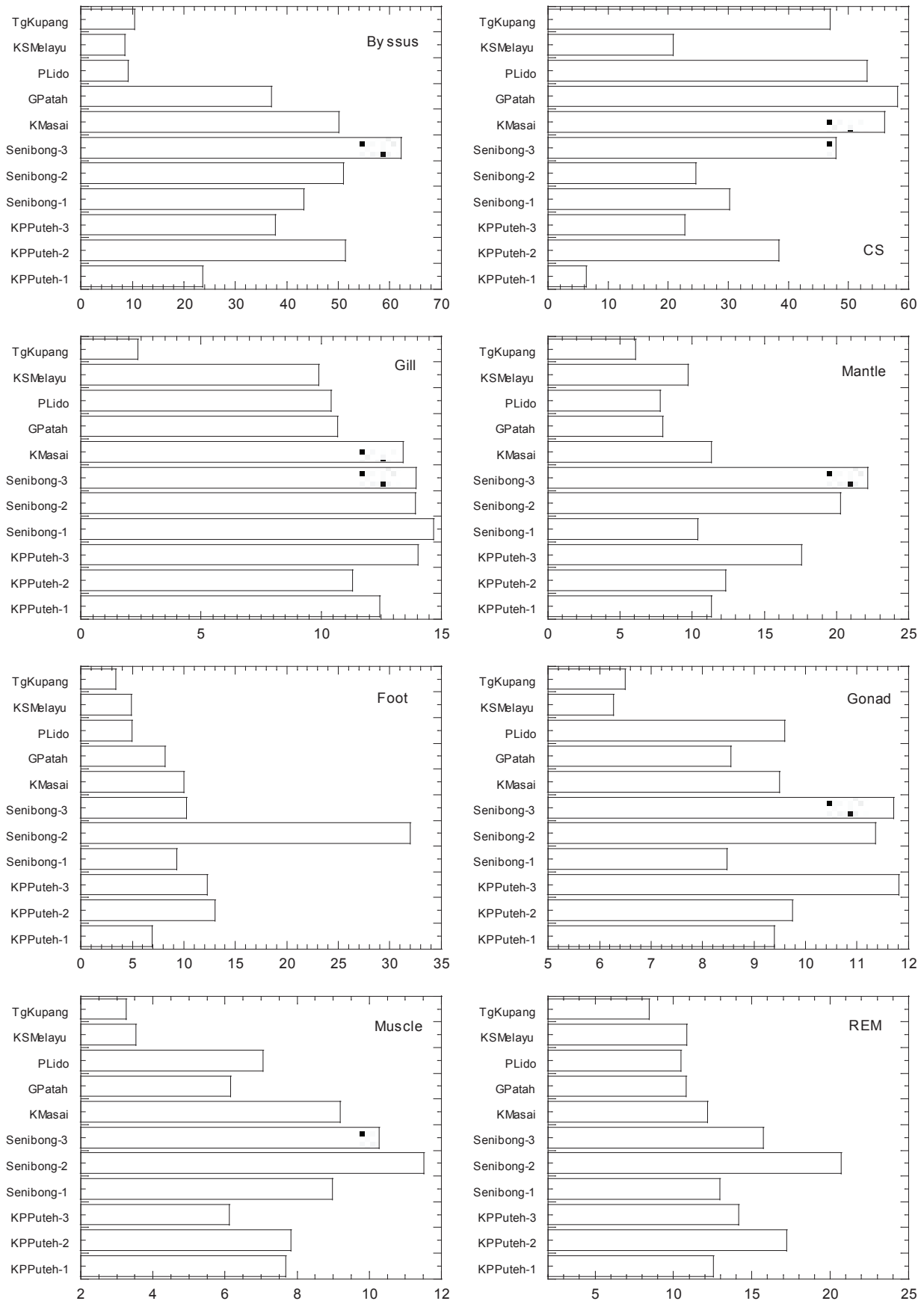


Figure 2 Concentrations (mean, mg/kg dry weight) of Cu in the different tissues of green-lipped mussel *Perna viridis* collected from the Straits of Johore.

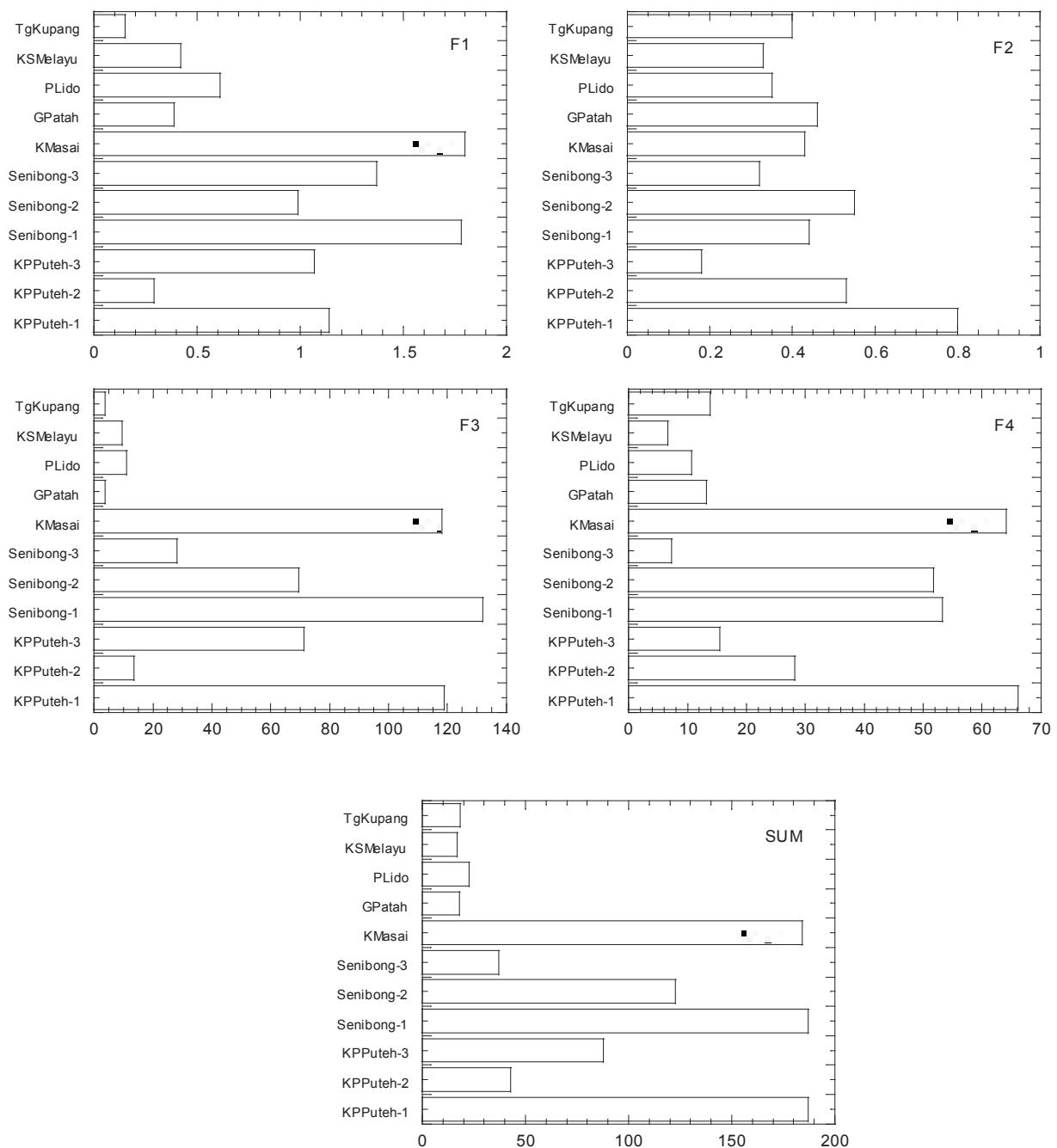


Figure 3 Concentrations (mean, mg/kg dry weight) of Cu in geochemical fractions of the surface sediments collected from the Straits of Johore.

Note: F1 = Easily, freely, leachable or exchangeable fraction; F2 = Acid-reducible fraction; F3 = Oxidizable-organic fraction; F4= Resistant fraction; SUM= Summation fractions of F1, F2, F3 and F4.

recorded higher levels of Cu in the eastern mussel populations than those in the western populations. Different parts of the soft tissues accumulated different Cu levels were found. Wong, et al. [17] reported that the variation of metal concentrations could be explained by the variation in water salinity and temperature, besides season and physiology of the mussels [18].

The soft tissues of *P. viridis* from the eastern part of the causeway recorded higher levels of Cu indicated higher bioavailability of Cu in the eastern part than those in the western part of the causeway [19]. This is most likely related to the many anthropogenic activities such as petro-chemical plants, land reclamation, urbanization, shipping, and other industrial activities [20].

Table 2: Comparison of Cu concentrations (mg/kg dry weight) between east part (7 populations from 3 sites) and west part (4 populations from 4 sites) of Johore Causeway in the different parts of green-lipped mussel *Perna viridis* collected from the Straits of Johore.

	East	West	East	West	East	West	East	West
	Bys	Bys	CS	CS	Gill	Gill	Mantle	Mantle
Minimum	23.7	8.55	6.35	20.8	11.3	2.36	10.4	6.08
Maximum	62.2	37.1	56.0	58.2	14.7	10.7	22.2	9.69
Mean	45.7*	16.3	32.3	44.8	13.4*	8.34	15.1*	7.87
SD	12.3	13.9	16.7	16.6	1.15	4.00	4.86	1.48
	East	West	East	West	East	West	East	West
	Foot	Foot	Gonad	Gonad	Muscle	Muscle	REM	REM
Minimum	6.95	3.37	8.47	6.27	6.12	3.25	12.19	8.47
Maximum	31.9	8.19	11.81	9.60	11.5	7.05	20.7	10.9
Mean	13.4*	5.35	10.3*	7.73	8.80*	4.99	15.1*	10.2
SD	8.42	2.03	1.32	1.61	1.79	1.90	3.07	1.14

Note: Bys = Byssus; CS = Crystalline Style; REM = Remaining Soft Tissues; SD = Standard Deviation. * = Significantly higher ($p < 0.05$).

Table 3: Comparison of Cu concentrations (mg/kg dry weight) between east part (7 sampling periods from 3 sites) and west part (4 sampling periods from 4 sites) of Johore Causeway in in geochemical fractions of the surface sediments collected from the Straits of Johore.

	East	West	East	West	East	West	East	West	East	West
SED	F1	F1	F2	F2	F3	F3	F4	F4	SUM	SUM
Minimum	0.29	0.15	0.18	0.33	13.7	3.66	7.24	6.62	37.13	17.1
Maximum	1.80	0.61	0.80	0.46	132	11.1	66.1	13.8	187	22.7
Mean	1.21*	0.39	0.46	0.39	78.8*	7.08	40.9*	11.1	121*	18.9
SD	0.52	0.19	0.19	0.06	46.4	3.87	23.8	3.26	66.9	2.57

Note: F1 = Easily, freely, leachable or exchangeable fraction; F2 = Acid-reducible fraction; F3 = Oxidizable-organic fraction; F4 = Resistant fraction; SUM = Summation fractions of F1, F2, F3 and F4. SD = Standard Deviation. * = significantly higher ($p < 0.05$).

Lim and Mohamed [7] reported that the green mussels from the wild contained the highest mean activity of ^{210}Po in the stomach and tissues of mussels due to contamination of industrial and domestic inputs in the SOJ. They assumed that the mussels might be a radiation risk to seafood consumers from the SOJ since the radiation dose of ^{210}Po exceeded permitted levels by the USEPA. Based on sediments collected in May 2013 from the SOJ, Keshavarzifard, et al. [21] concluded that Polycyclic Aromatic Hydrocarbons (PAHs) can be classified as moderate level pollution. Earlier, Sakari et al. [22] analysed two sediment cores in the SOJ for PAH and concluded that ocean-going ships and Singapore International Airport as the main sources of petroleum pollution in recent decades.

Zulkifli, et al. [23] reported Cu (57.8 mg/kg) in the surface sediments of the SOJ, in which the total Cu levels are dominated by non-resistant geochemical fraction (> 50%). According to Wood, et al. [1], the not elevated total Cu levels in the sediments collected from SOJ in 1990s was due to increased solubility of Cu during intense chemical weathering in the hot, humid tropical climate of the drainage basin. In addition, the Cu levels were significantly different ($p < 0.05$) between the eastern and western part of the SOJ. This could be attributed to the impacts of reclamation and dredging works, municipal or industrial discharge, marine aquaculture and shipping activities in the SOJ [4]. This

contrasted with the finding by Othman, et al. [24] who reported that the water quality in the coastline of Johor was less affected by the coastal reclamation activities.

Many recent reports did indicate that the eastern part of the Johore Causeway will be receiving continuous anthropogenic inputs. Yap, et al. [25] reported contamination in Pasir Gudang Area as the Kim Kim River chemical waste contamination was published in the local newspaper. Yap, et al. [26] reported higher bioavailability and contamination by Cu in three molluscs including *P. viridis* at Kg. Pasir Puteh, located at the eastern part of Johore Singapore Causeway.

Based on samples collected in 2015, Mahat, et al. [11] reported Cu levels ranging from 11.2-13.8 mg/kg dry weight in the total soft tissues of *P. viridis* collected from Kg. Pasir Puteh, comparing to 20.1 mg/kg dry weight for the mussels collected from the same site in 2000 [27]. This shows a lower level of Cu in the mussels for the 2015 samples when compared to those in 2000. However, Mohamat-Yusuff, et al. [28] reported a moderate level of contamination of Cu in *P. viridis* at Kong Kong Laut. Previously, high levels of Cu both in soft tissues of *P. viridis* and in the sediments were found in the eastern part of the causeway [1-3,13-15,29-31].

Since the eastern part of Johore Causeway such as Kg. Pasir Puteh and Kg. Masai are situated close to an active seaport and industrial areas near Pasir Gudang, the

possibility of constant sources of anthropogenic heavy metal contaminations at this area is expectedly high beyond 2020. Therefore, future effective control management of metal pollution in the eastern part of the SOJ should go in line with the Goal #12 under United Nation's Sustainable Development Goals (UNSDGs), in which economy, environmental and social are being mentioned [32].

CONCLUSION

In general, higher levels of Cu found in most of the soft tissues of *P. viridis* from east coast than the west coast of the SOJ indicated that the eastern part has higher bioavailability of Cu than the western part of the causeway. With some reports of high metal levels in the eastern part of Causeway in 2015–2018, it is predicted that there will be a constant source of anthropogenic metal contamination at the eastern part of the causeway in the SOJ beyond 2020 without a drastic, effective control of the human activities.

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