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A Review on Contamination of Heavy Metals, Linear Alkylbenzenes, Polycyclic Aromatic Hydrocarbons, Phenolic Endocrine Disrupting Chemicals and Organochlorine Compounds in *Perna viridis* from the Coastal Waters of Malaysia: A compilation of 1998 Data

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ABSTRACT

Since 1990s until today, the Asia-Pacific Mussel Watch approach has been widely used for biomonitoring purpose in Malaysia by using the green-lipped mussel *Perna viridis* in particular. This paper reviewed the concentrations of heavy metals (Cd, Cu, Pb and Zn), Linear Alkylbenzenes (LABs), Polycyclic Aromatic Hydrocarbons (PAHs), phenolic Endocrine Disrupting Chemicals (EDCs) [nonylphenol (NP), octylphenol (OP), and bisphenol A (BPA)] and organochlorine (OC) compounds (PCBs, DDTs, CHLs, HCHs and HCB) in nine mussel populations collected in 1998 from the coastal waters of Malaysia. In fact, all of these data were published separately in five different research journals in the literature. Since they discussed only based on the group of contaminants which they focused upon, this review paper aimed to see a holistic picture and understanding of the impacts of the different chemical contaminants in relation to the description of the sampling sites. Based on seven mussel populations with complete 11 chemicals (ranging from heavy metals, LABs, PAHs, phenolic EDCs and OCs), a dendrogram was established using single linkage cluster analysis. The clustering pattern showed two major subclusters. The first one comprising Tanjung Rhu, Trayong, Kuala Penyu and Pasir Panjang populations, indicating relatively uncontaminated conditions while the other subcluster consists of Penang, Kg. Pasir Puteh and Anjung Batu which indicated contaminated conditions as it is well supported by the elevated levels of some chemicals. The subcluster combining Penang and Kg. Pasir Puteh populations were mainly due to the elevated levels of LABs and PAHs in both sites while Kg. Pasir Puteh also had elevated levels of Cu, Pb, PCBs and CHLs. Anjung Batu, which is also clustering together with Penang and Kg. Pasir Puteh population can be explained by its elevated levels of three OC compounds namely DDTs, CHLs and HCHs. This comprehensive review is the first to report in the literature.

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1. INTRODUCTION

Nowadays, Mussel Watch approach has been widely employed for biomonitoring of chemical contaminants in the coastal waters. Until April 2010, a total 2,007 articles related to Mussel Watch program or approach can be easily found under the Sciencedirect website alone, indicating there is even more numbers of such studies can be found around the world with other scientific websites. According to Widdows and Donkin (1992), the main reasons for assessing the levels of different groups of chemical contaminants in coastal waters were 1) to protect human health by estimating exposure via dietary route back to man and 2) to protect valuable living natural resources [1]. Hence, the Mussel Watch approach initially proposed by Professor E. D. Goldberg in 1975 for the assessment of chemical contaminants in the coastal waters is of very much contributive to biomonitoring studies [2].

The most influential scientist on Asia-Pacific Mussel Watch is Professor Dr. S. Tanabe of Japan who have published a total of 168 publications specifically on two most leading journals in the field of pollution (Marine Pollution Bulletin= 94; Environmental Pollution= 74), as searched on 24 April 2010 in Sciencedirect website alone. Whereas the most recent article published (although still in press) on Mussel Watch is that by Apeti *et al.* (2010) on mussels collected from Birch Harbor, Maine [3].

Tracking back the history, in fact, the idea of using the green-lipped mussel *Perna viridis* as a biomonitor for Malaysian coastal waters is prompted by the many studies conducted using blue mussel *Mytilus edulis* [2],[4]. This is due to the ecological characteristics of marine mussels such as being wide geographical distribution, sedentary lifestyle, stable population, easy sampling, bioaccumulative of and correlative properties with the average pollutants of the environment, tolerance to salinity, resistance to stress due to high accumulation of wide range of pollutants and providing an assessment of bioavailability. Ever since, there is an increasing numbers of such biomonitoring studies utilizing mussels in Malaysia being reported in the literature [5]-[10].

The heavy metal data reviewed and cited in this paper was that by Yap *et al.* (2003a) [5] who reported the four popular heavy metal (Cd, Pb, Pb and Zn) concentrations in the total soft tissues of *P. viridis* from Peninsular Malaysian coastal waters while the data from East Malaysia (Sabah) was cited from Yap *et al.* (2003b) [6].

For Linear Alkylbenzenes (LABs), the data were both reported by Tsutsumi *et al.* (2002) and Isobe *et al.* (2007) [11],[12]. In particular, Tsutsumi *et al.* (2002) applied the LABs to be molecular markers for sewage input [11]. In addition, the data on Polycyclic Aromatic Hydrocarbons (PAHs), phenolic Endocrine Disrupting Chemicals (EDCs) were also reported from Isobe *et al*. (2007) [12]. The phenolic EDCs reported included Nonylphenol (NP) and Octylphenol (OP), and Bisphenol A (BPA) in which they are collectively termed as phenolic EDCs because of their phenolic structures in the molecules. The Alkylphenols such as nonylphenol (NP) and octylphenol (OP) are degradation products of alkylphenolpolyethoxylates (APnEO), which are commercially important nonionic surfactants with industrial, agricultural, and domestic applications.

Lastly, the most comprehensive data on monitoring Organochlorine Compounds (OCs) and PCBs pollution in the Asia-Pacific region including Malaysia, was that by Monirith *et al.* (2003) [13]. OCs represents ‘‘persistent organic pollutants (POPs)’’ and are of great concern due to their bioaccumulative nature and toxic biological effects on wildlife and humans [14]. The undesirable effects of some of these chemicals are linked to the occurrence of immunologic and teratogenic dysfunction, reproductive impairments and endocrine disruption in lower and higher trophic levels [15].

Most interestingly, all the mussels samples from Malaysia from the above papers reviewed and reported, came from a series of sampling trips in which I involved in sampling field trip of Peninsular Malaysia when I was a training researcher. The sampling trip was in fact, one of the series for Asia-Pacific Mussel Watch program in 1998. As a research student, I focused on heavy metal analysis in the mussel samples collected in the same year in order to establish background data for Malaysian coastal waters. The aim of this paper was to review and compile all the reported data for heavy metals, linear alkylbenzenes, polycyclic aromatic hydrocarbons, phenolic endocrine disrupting chemicals and organochlorine compounds in *Perna viridis* collected in 1998, from the coastal waters of Malaysia, in order to discuss them in a more holistic understanding on the different groups of chemical contaminants focusing on Malaysia’s scenario.

2. MATERIALS AND METHODS

Nine sampling sites for mussels were conducted in 1998. Descriptions for each of the sampling sites are given in Table 1.

Table 1: Information of *Perna viridis* samples collected from Malaysian coastal waters in 1998.

No.	Sampling sites	Sampling dates	N	Moisture (%)	Lipid (mg/g dry)	Shell length (mm)	Site description
1.	Sangkar Ikan (SIkan), Langkawi	20 Sep 1998	25	83.8	44	69.8 (61.9-91.0)	A fish aquaculture site.
2.	Tanjung Rhu (TRhu), Langkawi	20 Sep 1998	25	84.9	73	70.6 (67.0-74.7)	A recreational beach and fish aquaculture site.
3.	Penang Bridge (Penang), Penang-1	21 Sep 1998	25	81.2	66	73.4 (47.4-100.0)	Shipping lane, industry and urban area.
4.	Pasir Panjang (PPjg), Negeri Sembilan	22 Sep 1998	25	81.3	86	88.6 (68.1-109.1)	A recreational beach and mussel aquaculture site.
5.	Anjung Batu (ABatu), Malacca	22 Sep 1998	25	83.7	60	92.2 (84.3-106.1)	An agriculture and fish aquaculture area.
6.	Pantai Lido (PLido), Johore	1) 30 May 1998 2) 23 Sep 1998	25	83.1	71	59.4 (46.8-72.2)	City area.
7.	Kg. Pasir Puteh (KPPuteh), Johore	23 Sep 1998	25	81.0	112	64.4 (41.9-92.9)	Shipping, near a port, industry and urban area.
8.	Trayong, Sabah.	29 Sep 1998	25	87.1	54	109.8 (89.8-120.6)	Agriculture and aquaculture area.
9.	Kuala Penyu (KPenyu), Sabah.	29 Sep 1998	21 - 25	88.1	59	66.9 (57.1-76.6)	Agriculture and aquaculture area.

Note: Data for moisture and lipids were cited from Isobe *et al.* (2007) [12]

Briefly, for metal analysis, the samples were digested in concentrated HNO_3 (AnalaR grade, BDH 69%). The digested samples were then diluted to a certain volume with double distilled water (DDW). After filtration, the prepared samples were determined for heavy metals by an air-acetylene flame atomic absorption spectrophotometer (AAS) Perkin-Elmer Model 4100 [5],[6].

For LABs analysis, approximately 15 g of homogenized wet samples were macerated with dichloromethane (DCM) and anhydrous sodium sulfate using a Polytron RT2000 (Kinematica, Switzerland). The extracts were then subjected to purification, fractionation, and instrumental analysis, as described by Hartmann *et al.* (2000) [16]. Briefly, the extracts were purified and fractionated using two-step silica gel column chromatography and the alkylbenzene fraction was determined by gas chromatography–mass spectrometry (GC–MS) in selected ion monitoring mode at m=z ¼ 91, 92 and 105 [11].

Polycyclic Aromatic Hydrocarbons (PAHs) and phenolic endocrine disrupting chemicals done by Isobe *et al.* (2007) covered a wide range of compounds from non-polar hydrocarbons (e.g. n-alkanes) to polar compounds (e.g., bisphenol A) [12]. They include various EDCs (i.e., PCBs, DDTs, PAHs, NP, OP, phthalates, BPA, and natural estrogens) and molecular markers (e.g., alkylbenzenes and hopanes). This method relied upon maceration/homogenization, extraction, gel permeation chromatography (GPC), two-step silica gel chromatography, and gas chromatography-mass spectrometry (GC-MS). Detailed analytical conditions and quantification procedures are described by Isobe *et al.* [12],[17].

Polychlorinated biphenyls (PCBs) and organochlrorine (OC) insecticides such as DDT and its metabolites (DDTs: p; p0-DDT, p; p0-DDD, and p; p0-DDE), chlordane compounds (CHLs: trans-chlordane, cischlordane, cis-nonachlor and oxychlordane), hexachlorohexane isomers (HCHs: a-HCH, b-HCH, and c-HCH) and hexachlorobenzene (HCB) were analyzed following the method described by Tanabe *et al.* (2000b) and Kanatireklap *et al.* (1997), as mentioned by Monirith *et al.* (2003) [18],[19],[13].

For the statistical analysis, the data were $\log_{10}$(mean + 1) transformed before cluster analyses. The cluster analysis was performed in the STATISTICA software, version 5.5A.

3. RESULTS AND DISCUSSION

Heavy Metal Contamination

Table 2: Mean concentrations (μg/g dry weight) of Cd, Cu, Pb and Zn in the total soft tissues of *Perna viridis* collected from Malaysian coastal waters [5],[6]

	Location	Cd	Cu	Pb	Zn
1.	SIkan	0.37	6.80	4.02	100.28
2.	TRhu	0.25	9.83	4.38	94.52
3.	Penang	0.67	10.05	1.27	89.85
4.	PPjg	0.47	6.39	4.28	65.09
5.	ABatu	0.51	9.19	1.80	75.33
6.	PLido	0.65	9.80	3.02	80.49
7.	KPPuteh	0.53	14.92	5.23	97.16
8.	Trayong	1.18	10.74	2.51	121.79
9.	KPenyu	0.51	7.29	3.46	110.59

The mean concentrations (μg/g dry weight) of Cd, Cu, Pb and Zn in the total soft tissues of *P. viridis* are given in Table 2. The four metals ranged from 0.25-1.18 for Cd, 6.39-14.92 for Cu, 1.27-5.23 for Pb, 65.1-121.8 for Zn [20]. Most obviously, the highest concentrations of Cu and Pb were found in Kg. Pasir Puteh while the highest Cd and Zn level was found in Trayong. However, the Cd and Zn levels were lower than those at Kuala Linggi (1.25) for Cd and Kg. Pasir Puteh (128.90) for Zn, both collected in 2000 [5],[6]. The present ranges of Cd, Cu, Pb and Zn from the present study were also close to other mussel populations collected in the west coast of Peninsular Malaysia [5],[6]. In Asia, the highest levels of Cd (19.1 μg/g), Cu (279 μg/g), Pb (259 μg/g) and Zn (213 μg/g) in the soft tissue of *P. viridis* were reported from the Gulf of Thailand (Sukasem and Tabucanon, 1993), Hong Kong (Phillips, 1985), the Gulf of Thailand (Ruangwises and Ruangwises, 1998) and the Chao Phraya Estuary of Thailand (Menasveta and Cheeparanapiwat, 1981), respectively [21]-[24]. From the above comparison, the levels of Cd, Cu, Pb and Zn in *P. viridis* found in Peninsular Malaysia were therefore much lower than the highest values of these metals reported in the literature. Nevertheless, it should be noted that the higher levels of Cu and Pb at Kg. Pasir Puteh population were also found based on 2000 samples [5],[6]. The higher metals at Kg. Pasir Puteh could be related to the discharge of effluents from the nearby domestic and industrial inputs. Kg. Pasir Puteh is also a marina and some petro-chemical plants and port activity in the vicinity may also be suspected as anthropogenic sources in the area. Cu leachate from the antifouling paints of boats and the areas semi-enclosed topography may aggravate the pollution problem [5],[6],[7].

Linear alkylbenzenes Contamination

The concentrations of LABs (sewage input) are given in Table 3. Concentrations of LABs in mussel samples collected from the Malaysian coastal waters ranged from 11 to 807 ng/g-dry tissue [11]. High levels of LABs observed in Penang Bridge and Kg. Pasir Puteh and these levels were comparable to those in Tokyo Bay and Boston Harbor, which are among the most polluted coastal zones of the world [25]. This suggests the intensive input of sewage into coastal waters of these populated areas. However, relatively low concentrations of LABs were observed in other sampling sites may be due to lower human activities and/or less usage of synthetic detergents in the surrounding areas.

Table 3: Linear alkylbenzenes (LABs) in the total soft tissues of *Perna viridis* collected from Malaysian coastal waters [11]

No.	Site	Sampling dates	SL (MM)	LABs (ng/g-dry)[a]	$\sum C_{12}$-C_{14} (ng/g-dry)[b]
1.	SIkan	NA	NA	NA	NA
2.	TRhu	20 Sep 1998	85	29	22
3.	Penang	21 Sep 1998	91	764	560
4.	PPjg	21 Sep 1998	93	21	17
5.	ABatu	22 Sep 1998	85	198	147
6.	PLido	30 May 1998	69	430	315
7.	KPPuteh	23 Sep 1998	89	807	565
8.	Trayong	29 Aug 1998	70	11	8
9.	KPenyu	29 Aug 1998	79	11	8

Note:
NA= data not available.
[a]Sum of the 26 LAB congeners; n.c.: no reliable values calculated due to being overlapped by TAB peaks.
[b]Sum of 17 LABs congeners with alkyl carbons ranging from C_{12} to C_{14}.
[c]A ratio of (6-C_{12}AB + 5-C_{12}AB) relative to (4-C_{12}AB + 3-C_{12}AB + 2-C_{12}AB);

Table 4: Concentrations (ng/g dry tissues) of linear alkylbenzenes (LABs), octylphenol (OP), nonylphenol (NP) and bisphenol A (BPA) in the total soft tissues of *Perna viridis* collected from Malaysian coastal waters [12]

Site	Concentrations				
	LABs[a]	OP	NP	BPA	I/E ratio[b]
SIkan	NA	1	18	(0.56)	NA
TRhu	29	NA	NA	NA	NC
Penang	764	NA	NA	NA	1.79
PPjg	21	NA	NA	NA	NC
ABatu	198	9	290	(0.51)	2.83
PLido	430*	16	663	4.2	2.21*
KPPuteh	807	NA	NA	NA	1.99
Trayong	11	NA	NA	NA	NC
KPenyu	11	NA	NA	NA	NC

Note:
* data cited from Tsutsumi *et al*. (2002) [11]
[a]Sum of the concentrations of the 26 LAB congeners.
[b]A ratio of (6-C12AB+5-C12AB) relative to (4-C12AB+3-C12AB+2-C12AB).
NA= not analyzed
Values in parenthesis are not significant in comparison to the procedural blanks.
NC= no reliable values calculated due to low concentrations.

LABs consist of isomers with different phenyl substitutional positions. External isomers are preferentially biodegraded as compared to internal isomers [26]. To quantitatively express the isomer composition, a ratio of internal to external isomers (I/E ratio: a ratio of sum of 6-C12 AB and 5-C12 AB relative to sum of 4-C12 AB, 3-C12 AB and 2-C12 AB) has been proposed as an index of the degree of LAB degradation [27]. In synthetic detergents and untreated sewage I/E ratios are around 0.7, whereas secondary (biological) sewage treatment increases the ratio to as much as 7 [28]. A higher value of this ratio means a greater depletion of external isomers and, hence, greater degradation, whereas a lower I/E ratio means less degradation. I/E ratios for mussels collected from Malaysian coastal waters are given in Table 4 and they ranged from NC (no reliable values calculated due to low concentrations) to 2.83. These values are clearly much lower than those from Tokyo Bay (3.00–8.43), and indicate that LABs in the Malaysian coastal zones are less degraded. This can be explained by the difference in the type of wastewater being discharged. Primary effluent as well as raw sewage has a low I/E ratio (0.5–0.9) because it is essentially a physical settlement of sewage particles that allows for the limited opportunity of aerobic degradation [28]. On the other hand, due to facilitated microbial degradation, secondary effluent shows a much higher I/E ratio, ranging from 2 to 7 [28]. The considerably lower I/E ratios (NC-2.83) found in all the sampling sites indicated that the Malaysian coastal zones have been receiving sewage effluents with limited (primary effluent) or no (raw) treatment, and is probably due to a lack of sewage treatment facilities or experience frequent heavy rainfall with subsequent overflow of sewage treatment plants, thereby flushing the untreated sewage into the coastal zones. In summary, LABs determination demonstrated extensive inputs of poorly treated wastewater to aquatic environments in Malaysia.

Phenolic endocrine disrupting chemicals

For Phenolic EDCs in mussels, the concentrations of alkylphenols (NP) in mussels from Malaysian coastal waters are given in Table 4 [12]. The NP concentrations ranged from 18 to 663 ng/g dry tissue. Elevated concentrations of NP were observed in Pantai Lido (663 ng/g dry tissues as shown in Table 4) which was were comparable to although lower than those in Tokyo Bay (47-1347 ng/g dry tissues). Monitoring results of the present study suggest that the status of NP pollution in some locations in Malaysia had already as severe as those in industrialized countries such as Japan. OP concentrations in the Malaysian mussels were one to two orders of magnitude lower than those of NP. This is probably due to a combination of less production and usage of octylphenol ethoxylate surfactants and lower hydrophobicity of OP than NP [12].

Relatively high concentrations of BPA was found in the mussels from Pantai Lido (4.2 ng/g dry tissues) which was comparable to and within those in Tokyo Bay (0.54-13.4 ng/g dry tissues) and India (1.1-

13.7 ng/g dry tissue). The fact that BPA was significantly detected in marine biological samples in the present study warrants the necessity for a wider range of monitoring of BPA in marine biological tissues.

For source estimation of the phenolic EDCs, LABs concentrations in the mussels were compared with those of phenolic EDCs. Elevated NP concentrations were detected for locations where low LABs were found at Pantai Lido. This result indicates that there are some other sources of phenolic EDCs other than from urban sewage, although urban sewage is one of the major sources of phenolic EDCs.

Polycyclic Aromatic Hydrocarbons (PAHs)

Based on Isobe *et al.* (2007), they quantified PAHs with three to six rings [12]. Total concentrations of PAHs in mussels collected from all the nine populations of Malaysian coastal waters ranged from 11 to 309 ng/g, comparing to South and Southeast Asia which ranged from 11 to 1,494 ng/g dry (Table 5). Based on categorization of PAH concentrations in mussels proposed by Baumard *et al*. (1998), PAHs concentrations in Malaysia can be classified as "low (0–100 ng/g dry) to moderate (100–1,000 ng/g dry)" [29].

A comprehensive review on the source identification ratios were also done by Isobe *et al*. (2007) [12]. The mussel populations collected from Penang Bridge and Kg. Pasir Puteh showed a considerably higher methylphenanthrenes/phenathrene MP/P ratio (5.15-5.69) than those observed in mussels from Tokyo Bay (0.8–3.1), suggesting a higher contribution of petrogenic inputs to coastal waters in these areas.

A ratio of CPP to the sum of methylphenanthrenes (CPP/MP ratio) has been proposed as another index to distinguish petrogenic and pyrogenic PAHs [30]. The CPP/MP ratios from this study ranged from 0.012 to 0.115 and therefore they were of between petrogenic sources (<0.07) to petrogenic plus minor contributions from pyrogenic sources (0.07–0.2). The CPP/MP ratios were also less than the mean values observed in Tokyo Bay mussels (0.15). The other ratios of PAH species (pyrene to fluoranthene: Py/Flu ratio, phenanthrene to anthrancene: Phe/An) were also examined for source-identification. All populations except for Pasir Panjang and Tanjung Rhu, their Py/Flu ratios were >1, supporting that these samples had petrogenic contribution more from pyrogenic inputs. For some populations, the pyrogenic signature was not consistent with the petrogenic signature suggested from the MP/P ratio and CPP/MP ratio. This can be explained by the fact that the contribution from petrogenic PAHs varies among PAH species. Actually lower Py/Flu ratios have been observed in some crude oil samples [31]. Phe/An ratios higher than 10 at Penang Bridge and Kuala Penyu supported petrogenic contributions to these sites whereas if conflicted with the estimation from the alkylated PAHs for some other locations (such Kg. Pasir Puteh). Lower Phe/An ratios ("pyrogenic" signature) were again observed in some crude oil and petroleum products [32]. In general, sampling sites at Penang Bridge and Kuala Penyu, all compositional parameters (MP/P, CPP/MP, Py/Fluo, Phe/An ratios) consistently exhibited petrogenic signatures. In conclusion, these sites in Malaysian coastal waters were heavily affected by petrogenic PAHs, although inputs from pyrogenic sources were detected.

Table 5: Compositional parameters of PAHs in the total soft tissues of *Perna viridis* collected from Malaysian coastal waters [12]

Code	PAH[a] (ng/g dry)	Compositional parameters				
		H/L[b]	MP/P[c]	CPP/MP[d]	Py/Flu[e]	Phe/An[f]
SIkan	NA	NA	NA	NA	NA	NA
TRhu	18	0.37	1.13	0.065	0.74	8.92
Penang	265	0.20	5.15	0.012	1.06	12.21
PPjg	41	1.19	2.67	0.115	0.70	5.55
ABatu	57	0.58	1.78	0.067	1.01	7.36
PLido*	157	0.16	2.39	0.047	5.12	7.46
KPPuteh	309	0.44	5.69	0.035	2.50	2.70
Trayong	38	0.16	1.79	0.066	1.04	8.43
KPenyu	11	0.11	1.90	0.061	1.05	14.74

Note:
Data for PLido* were based on samples collected on 30 May 1998.

[a]Sum of concentrations of phenanthrene, anthracene, 1-methylphenanthrene, 2-methylphenanthrene, 3-methylphenanthrene, 9-methylphenanthrene , fluoranthene, pyrene, benz [a] anthracene, chrysene, benzo [b] fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, benzo [e] pyrene, benzo[a] pyrene, perylene, indeno [1,2,3-cd] pyrene, benzo [ghi] perylene, and coronene.

[b] H/L ratio: a ratio of the sum of benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, benzo[e]pyrene, benzo[a]pyrene, perylene, indeno[1,2,3-cd]pyrene, benzo[ghi]perylene and coronene to sum of phenanthrene, anthracene, methylphenanthrenes, fluoranthene, pyrene.

[c] MP/P ratio: a ratio of the sum of 1-methylphenanthrene, 2-methylphenanthrene, 3-

methylphenanthrene, 9-methylphenanthrene to phenanthrene
[d] CCP/MP ratio: 4H-cyclopenta [def] phenanthrene to sum of 1-methylphenanthrene, 2-methylphenanthrene, 3-methylphenanthrene, and 9-methylphenanthrene.
[e] Py/Flu ratio: a ratio of pyrene to fluoranthene.
f Phe/An ratio: a ratio of phenanthrene to anthracene.

In Kuala Penyu (Sabah), which is close to an oil platform, where the contribution of crude oil may be occurring. A comprehensive study reported by Zakaria *et al.* (2002) identified the used crankcase oil as the major source of petrogenic PAHs in Malaysian aquatic environments [33]. Of course, there are some other potential sources including accidental oil spills, routine tanker operations, spillage from oil fields and oil refineries, spillage from fishing boats, leakage from land-based storage, gasoline and diesel fuel from automobiles, street dust, and others.

To conclude, at several populations collected from Penang Bridge, Kuala Penyu, all compositional parameters (MP/P, CPP/MP, Py/Fluo, Phe/An ratios) consistently exhibited petrogenic signatures.

Organochlorine Contamination

Six compounds of OC in *P. viridis* collected Malaysian coastal waters are given in Table 6. In general, lower DDTs levels were observed in mussels from Malaysia, which indicates less usage of DDTs in these countries when compared to mussels from China (830–54,000 ng/g lipid wt.), Hong Kong (640–61,000 ng/g lipid wt.) and Vietnam (220–34,000 ng/g lipid wt.) [13]. Mussels from Singapore and Malaysia contained higher percentages of c-HCH (up to 90%) (Monirith *et al.*, 2003). Previous reports indicated the use of lindane (purified c-HCH) in Malaysia (Tan and Vijayaletchumy, 1994; Kannan *et al.*, 1995), supporting the result of the present study. In general, low concentrations PCBs were found in mussels from Malaysia, which indicate fewer local sources [34],[35]. Our PCBs were much lower than some urban/industrialized cities in Asian countries which showed relatively higher concentrations, such as Bombay and Cochin in India, Fuzhou Lian Jian in China and Manila in the Philippines [13].

Table 6: Concentrations of organochlorines (ng/g lipid wt.) in the total soft tissues of *Perna viridis* collected from Malaysian coastal water [13]

Location of sample	Lipid (%)	PCBs	DDTs	CHLs	HCHs	HCB
SIkan	0.92	6.0	95	41	9.4	<1.1
TRhu	1.1	5.1	16	2.5	4.9	<0.90
Penang	1.0	60	71	180	<0.10	2.4
PPjg	1.6	11	93	60	3.5	<0.60
ABatu	1.3	22	100	610	12	<0.80
PLido	NA	NA	NA	NA	NA	NA
KPPuteh	2.1	250	130	470	5.2	<0.50
Trayong	.65	.3	2	.1	.1	1.5
KPenyu	.73	.5	00	.7	1.4	1.4

Note:
NA: No data available.
DDTs: *p, p'*-DDE + *p, p'*-DDD + *p, p*'-DDT.
CHLs: *trans*-chlordane + *cis*-chlordane + *trans*-nonachlor + *cis*-nonachlor + oxychlordane.
HCHs: α-HCH + β-HCH + γ-HCH.

The residue levels of CHLs in mussels collected from Malaysia coastal waters are given in Table 6. Our levels (17–630 ng/g lipid wt.) of CHLs from Malaysia were considered 'higher' and comparable to those observed in mussels from Japan (150–1800 ng/g lipid wt.), China (40–870 ng/g lipid wt.), Hong Kong (18–750 ng/g lipid wt.) and Singapore (520 ng/g lipid wt.) [13]. Malaysia showed higher concentrations of CHLs at sites in proximity to harbor, aquaculture, urban and densely populated areas such as ABatu and KKPuteh. Higher percentages of trans-nonachlor were found in mussels from Malaysia and again it is comparable to those found from Japan, Singapore, India, Philippines and China. The presence of trans-nonachlor in the Malaysian mussels might suggest the recent usage of technical CHLs. The present concentrations and compositions of CHLs found in the recent study in mussels from Malaysia might reflect the recent input of CHLs into coastal environment. In general, Malaysia had higher concentrations of CHLs.

General Discussion

Nevertheless, cautions should be exercised when interpreting all the above data since they can be influenced by biological and environmental factors. Biological factors such as gender (Yap *et al.*, 2006b), sizes (Yap *et al.*, 2003c) and spawning conditions (Yap *et al.*, 2002) can mask all major anthropogenic impacts while environmental factors such as salinity, temperature, tidal levels and stress due to desiccation can also cause the data interpretation misleading [36],[37],[20]. We can argue that since the present samplings were conducted between 20-29 September 1998 (which was a relatively short interval) except for the first sampling at Pantai Lido conducted in May 1998, the spawning factor thus is greatly reduced to a minimal. Other biological factor which is the mussel sizes investigated indicated by shell lengths could influence the data reviewed in this paper. For example, the mean shell lengths for Pantai Lido population was 64.4 cm, comparing to 109.8 cm for Trayong population. This indicates Trayong population is bigger or older when compared to Pantai Lido population. However, it should be noted that the growth rate of mussels are subjected to food availability and population structures. For the environmental factors such as temperature and salinity, since those water parameters varied within a range of tolerance for the mussel mussels, it can be assumed that these factors can little influence the present data reviewed. Certainly, this is still debatable whether the present data reviewed can accurately reflect the anthropogenic impact on the accumulation of the different contaminant groups in *P. viridis*. Nevertheless, it is argued that since 1) the clustering pattern are very much related to the site description (although it is not always true), and 2) there is not other background data reported in the literature such as the data reviewed in this paper, the present data reviewed can serve as important future reference which had covered the important contaminant groups such as heavy metals, LABs, PAHs, EDCs and OCs.

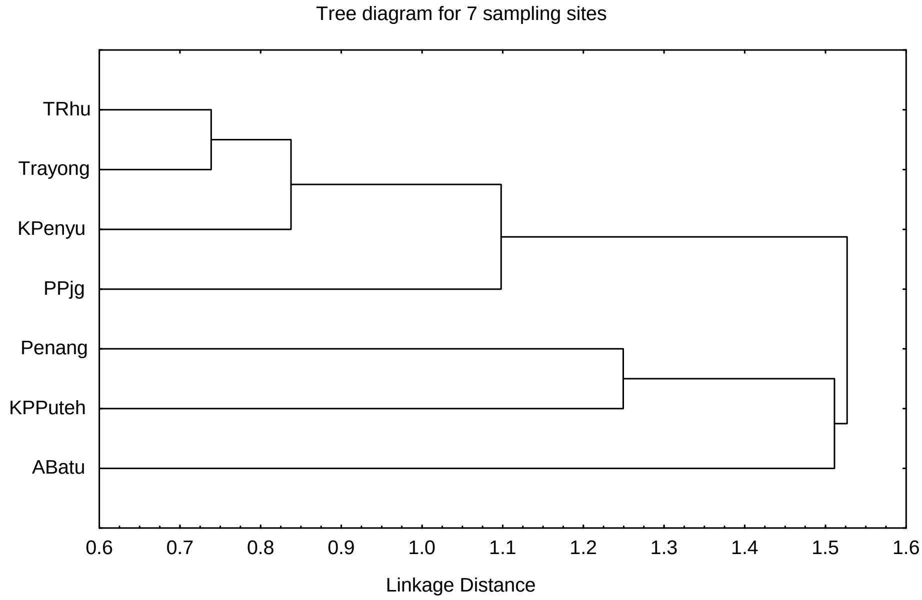

Figure 1: Cluster analysis based on Single Linkage Euclidean distances, on the eleven chemicals (PCB, DDT, PAH, LAB, CHL, HCH, HCB, Cd, Cu, Pb and Zn) in the seven mussel populations, based on $\log_{10}$(mean + 1) transformed data.

To conclude the overall chemical contamination, a dendrogram established using single linkage cluster analysis based on seven sampling sites covering eleven chemicals namely Cd, Cu, Pb, Zn, LABs, PAHs, PCBs, DDTs, CHLs, HCHs and HCB, is shown in Figure 1, since only seven sampling sites had a complete data for the eleven chemicals. The clustering pattern shows two major subclusters. The first one comprising Tg. Rhu, Trayong, KPenyu and PPjg population, indicating relatively uncontaminated conditions while the other subcluster consists of Penang, Kg. Pasir Puteh and ABatu which indicated contaminated conditions as it is well supported by the absolute values of some chemicals. The subcluster combining Penang and Kg. Pasir Puteh populations could be mainly due to the elevated levels of LABs and PAHs in both sites (Table 3) while ABatu is within the major clusters with Penang and Kg. Pasir Puteh is due to the elevated levels of DDTs, CHLs and HCHs. Kg. Pasir Puteh also had elevated levels of Cu, Pb, PCBs and CHLs.

4. CONCLUSION

From this review, Kg. Pasir Puteh received higher concentrations of Cu, Pb, LABs, PAHs, PCBs and CHLs while Penang population received higher concentrations of LABs and PAHs. ABatu population particularly received anthropogenic OCs including DDTs, CHLs and HCHs. There is always need and concern as we may ask ‘What are these chemical pollutant status in the Malaysian coastal waters in 2010 ?’ This benchmark review based on 1998 data covering heavy metals, LABs, PAHs, phenolic EDCs and OCs can always be used for future comparison and reference since more extensive and systematic monitoring of all these chemical pollution in Malaysian coastal waters using mussels or other molluscs as biomonitors are always encouraged and welcome in future.

REFERENCES

[1] Widdows, J. and Donkin, P. “Mussels and environmental contaminants: bioaccumulation and physiological aspects”, in Gosling, E, editor. *The mussel Mytilus: ecology physiology, genetic and culture,* Amsterdam: Elsevier. Pp. 383-424, 1992.
[2] Goldberg, E.D. “The Mussel Watch - A first step in global marine monitoring”, *Mar. Pollut. Bull.*, Vol. 6. Pp. 111, 1975.
[3] Apeti, D.A., Lauenstein, G.G., Christensen, J.D., Kimbrough, K., Johnson, W.E., Kennedy, M. and Grant. K.G. “A historical assessment of coastal contamination in Birch Harbor, Maine based on the analysis of mussels collected in the 1940s and the Mussel Watch Program”, *Mar. Pollut. Bull*. In Press, 2010.
[4] Phillips, D.J.H. and Rainbow, P.S. *Biomonitoring of trace aquatic contaminants*, Elsevier Science Publishers Limited, London. Pp. 371, 1993.
[5] Yap, C.K., Ismail, A. and Tan, S.G. “Background concentrations of Cd, Cu, Pb and Zn in the green-lipped mussel Perna viridis (Linnaeus) from Peninsular Malaysia”, *Mar. Pollut. Bull.*, Vol. 46. Pp. 1043-1048, 2003a..
[6] Yap, C.K., Rahim Ismail, A., Ismail, A. and Tan, S.G. “Studies on heavy metal accumulations in green-lipped mussel Perna viridis by using multiple linear stepwise regression analysis”, *Pertanika J. Sci. Tech.*, Vol/Issue: 11(1). Pp. 43-55, 2003b.
[7] Yap, C.K., Ismail, A, Tan, S.G. and Rahim Ismail, A. “The impact of anthropogenic activities on heavy metal (Cd, Cu, Pb and Zn) pollution: Comparison of the metal levels in green-lipped mussel Perna viridis (Linnaeus) and in the sediment from a high activity site at Kg. Pasir Puteh and a relatively low activity site at Pasir Panjang”, *Pertanika J. Trop. Agric. Sci.*, Vol/Issue: 27(1). Pp. 73-78, 2004.
[8] Yap, C.K., Ismail, A., Edward, F.B., Tan, S.G. and Siraj, S.S. “Use of different soft tissues of Perna viridis as biomonitors of bioavailability and contamination by heavy metals (Cd, Cu, Fe, Pb, Ni and Zn) in a semi-enclosed intertidal water, the Johore Straits”, *Toxicol. Environ. Chem.*, Vol/Issue: 88(4). Pp. 683 – 695, 2006a.
[9] Yap, C.K., Che Mohd Zaidi, C.B., Edward, F.B., Ismail, A. and Tan, S.G. “Heavy metal concentrations (Ni, Pb, and Zn) in the green-lipped mussel, Perna viridis collected from the northern coastal waters of Peninsular Malaysia”, *J. Sustain. Sci. Manage*, Vol/Issue: 4(1). Pp. 10-19, 2009a.
[10] Yap, C.K., Yeow, K.L., Edward, F.B. and Tan, S.G. “Revealing Cu-contaminated mussels collected from the coastal water near Penang Industrial Area by using Malaysian Mussel Approach”, *Asian J. Microbiol. Biotech. Environ. Sci.*, Vol/Issue: 11(4). Pp. 683-689, 2009b.
[11]
[12] Isobe, T., Takada, H., Kanai, M., Tsutsumi, S., Isobe, K. O., Boonyatumanond, R. and Zakaria, M.P. “Distribution of Polycylic Aromatic Hydrocarbons (PAHs) and phenolic endocrine disrupting chemicals in South and Southeast Asian mussels”, *Environ. Monitor. Assess.*, Vol. 135. Pp. 423-440, 2007.
[13] Monirith, I., Ueno, D., Takahashi, S., Nakata, H., Sudaryanto, A., Subramanian, A., Karuppiah, S., Ismail, A., Muchtar, M., Zheng, J., Richardson, B.J., Prudente, M., Hue, N.D., Tana, T.S., Tkalin, A.V. and Tanabe, S. “Review: Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries”, *Mar. Pollut. Bull.*, Vol. 46. Pp. 281–300, 2003.
[14] Tanabe, S., Prudente, M.S., Kan-atireklap, S. and Subramanian, A. “Mussel watch: marine pollution monitoring of butyltins and organochlorines in coastal waters of Thailand, Philippines and India”, *Ocean Coast. Manage*, Vol. 43. Pp. 819–839, 2000.
[15] Colborn, T. and Smolen, M.J. “Epidemiological analysis of persistent organochlorine contaminants in cetaceans”, *Rev. Environ. Contam. Toxicol*, Vol. 146. Pp. 91–172, 1996.
[16]
[17] Isobe, T., Nishiyama, H., Nakashima, A. and Takada, H. “Distribution and behavior of nonylphenol, octylphenol, and nonylphenol monoethoxylate in Tokyo metropolitan area – their association with aquatic particles and sedimentary distributions”, *Environ. Sci. Tech.*, Vol. 35. Pp. 1041–1049, 2001.
[18] Tanabe, S. “Asian developing regions: persistent organic pollutants in seas”, In: Sheppard, C.R.C. (Ed.), *Sea at the Millennium: an Environmental Evaluation*, Elsevier Science, Amsterdam, Pp. 447–462, 2000b.
[19]
[20] Yap, C.K., Ismail, A. and Tan, S.G. “Condition index of green-lipped mussel Perna viridis (Linnaeus) as a potential physiological indicator of ecotoxicological effects of heavy metals (Cd and Pb)”, *Malays. Appl. Biol.*, Vol/Issue: 31(2). Pp. 37-45, 2002.
[21]
[22] Phillips, D.J.H. “Organochlorines and trace metals in green-lipped mussels Perna viridis from Hong Kong waters: A test of indicator ability”, *Mar. Ecol. Prog. Ser.*, Vol. 21. Pp. 251-258, 1985.
[23] Ruangwises, N. and Ruangwises, S. “Heavy metals in green mussels (Perna viridis) from the Gulf of Thailand”, *J. Food Protect*, Vol. 61. Pp. 94-97, 1998.

[24] Menasveta, P. and Cheevaparanapiwat, V. "Heavy metals, organochlorines, pesticides and PCBs in green mussels, mullets and sediments of river mouth in Thailand", *Mar. Pollut. Bull*, Vol. 12. Pp. 19-25, 1981.
[25] Sherblom, P.M. and Eganhouse, R.P. "Bioaccumulation of molecular markers for municipal wastes by Mytilus edulis", In R. A. Baker (Ed.), *Organic substances and sediments in water*, Washington, DC: American Chemical Society, Vol. 3. Pp. 139-158, 1991.
[26] Gustafsson, O., Long, C.M., MacFarlane, J. and Gschwend, P.M. "Fate of linear alkylbenzenes released to the coastal environment near Boston Harbor", *Environ. Sci. Tech.*, Vol. 35. Pp. 2040–2048, 2001.
[27] Takada, H. and Ishiwatari, R. "Biodegradation experiments of linear alkylbenzenes (LABs): Isomeric composition of C12 LABs as an indicator of the degree of LAB degradation in the aquatic environment", *Environ. Sci. Tech.*, Vol. 24. Pp. 86–91, 1990.
[28] Takada, H. and Eganhouse, R. "Molecular markers of anthropogenic waste: Their use in determining sources, transport pathways and fate of wastes in the environment", In R. Mayers (Ed.), *The Encyclopedia of environmental analysis and remediation*, New York: Wiley. Pp. 2883-2940, 1998.
[29] Baumard, P., Budzinski, H., Garrigues, P., Sorbe, J. C., Burgeot, T. and Bellocq, J. "Concentrations of PAHs (polycyclic aromatic hydrocarbons) in various marine organisms in relation to those in sediments and to trophic level", *Mar. Pollut. Bull.*, Vol. 36. Pp. 951–960, 1998.
[30] Garrigues, P., Budzinski, H., Manitz, M.P. and Wise, S.A. "Pyrolytic and petrogenic inputs in recent sediments: A definitive signature through phenanthrene and chrysene compounds distribution", *Polycyc. Aromatic Comp.*, Vol. 7. Pp. 275–284, 1995.
[31] Yunker, M.B., Macdonald, R.W., Vingarzan, R., Mitchell, R.H., Goyette, D.S. and Sylvestre, S. "PAHs in the Fraser River basin: A critical appraisal of PAH ratios as indicators of PAH source and composition", *Organic Geochem*, Vol. 33. Pp. 489–515, 2002.
[32] Zakaria, M. P., Okuda, T. and H. Takada. "Polycyclic aromatic hydrocarbon (PAHs) and hopanes in stranded tarballs on the coasts of Peninsular Malaysia: Applications of biomarkers for identifying sources of oil pollution", *Mar. Pollut. Bull.*, Vol. 42. Pp. 1357–1366, 2001.
[33] Zakaria, M.P., Takada, H., Tsutsumi, S., Ohno, K., Yamada, J. and Kouno, E. "Distribution of polycyclic aromatic hydrocarbons (PAHs) in rivers and estuaries in Malaysia: A widespread input of petrogenic PAHs", *Environ. Sci. Tech.*, Vol. 36. Pp. 1907–1918, 2002.
[34] Tan, G.H. and Vijayaletchumy, K. "Organochlorine pesticide residue levels in Peninsular Malaysian Rivers", *Bull. Environ. Contam. Toxicol*, Vol. 53. Pp. 351–356, 1994.
[35] Kannan, K., Tanabe, S. and Tatsukawa, R. "Geographical distribution and accumulation features of organochlorine residues in fish in Tropical Asia and Oceania", *Environ. Sci. Tech.*, Vol. 29. Pp. 2673–2683, 1995.
[36] Yap, C.K., Ismail, A, Tan, S.G. and Rahim Ismail, A. "Is gender a factor contributing to the natural variations in the accumulation of heavy metals (Cd, Cu, Pb and Zn) by the green-lipped mussel Perna viridis?", *Indian J. Mar. Sci.*, Vol/Issue: 35(1). Pp. 29-35, 2006b.
[37] Yap, C.K., Ismail, A. and Tan, S.G. "Effects of total soft tissue and shell thickness on the accumulation of heavy metals (Cd, Cu, Pb and Zn) in the green-lipped mussel Perna viridis (Linnaeus)", *Russian J. Mar. Biol.*, Vol/Issue: 29(5). Pp. 323-327, 2003c.