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Heavy metals (mercury, arsenic, cadmium, plumbum) in selected marine fish and shellfish along the Straits of Malacca

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Abstract: Level of heavy metals in marine ecosystem has been intensively studied during recent years as these hazardous substances could be accumulated in the biota. Generally, the presence of contaminants in fish is a result of human activities such as industrial and agricultural wastes. In this study, the marine fish and shellfish from the Straits of Malacca were analyzed using Inductively Coupled Plasma Optical Emission Spectrophotometer (ICP-OES) and Flow Injection Mercury System (FIMS) for Cd, As and Pb and Hg, respectively. The Straits of Malacca is one of the busiest shipping routes in the world that make the level of heavy metals potentially high besides the various industrial activities along the west region of Peninsular Malaysia. The range of heavy metals in samples were 1.0-3-6.5-3 µg/ g wet sample for Hg, 0.5-2-47-2 µg/ g wet sample for Cd, 0.01-0.39 µg/ g wet sample for Pb and 0.14-6.57 µg/ g wet sample for As. Most part of the values was below the permitted limit set by FAO/WHO 2004 as well as Food Act 1983 and Food Regulations 1985. Therefore it can be generalized that fish and shellfish from the Straits of Malacca are safe to consume in terms of these heavy metals concentration and do not constitute a risk for human health.

Keywords: Fish, shellfish, Hg, As, Pb, Cd, ICP-OES

Introduction

Today, fish has become the main supply of protein besides meat and poultry products and contributed to a large percentage of dietary protein globally. In most Asian countries, especially in Southeast Asia like Thailand, Indonesia and Malaysia fish is taken as the main dish of their diet. It is particularly valuable for providing food with nutritional value in the form of protein, and minerals such as calcium, phosphorus, iron and copper. Besides protein, fish is also high in essential fatty acids (EFAs), known as eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA) that are important to the diet. In addition, consumption of fatty fish is recommended by nutritionist as a mean to prevent cardiovascular diseases (Siscovick *et al*., 1995; Daviglus *et al*., 1997; Domingo *et. al*., 2007).

However, fish also may contain mercury, arsenic, and cadmium that could give negative effects for health. Svensson *et al*. (1991) mentioned that diet and food of animals origin are the most predominant sources (>90%) of heavy metals and other chemical contaminants to human. Although, fish and shellfish only represent small percentage (10%) of these contaminants, these foods are one of the major routes of the contaminants in the body (Harisson *et.al*., 1998). Some of the metals are essential to the human body but some of them can cause harm to the body. The common heavy metals that can be found in fish and shellfish are potassium, copper, calcium, iron, iodine, mercury, lead and cadmium (Connel, 1984). The metals that cause toxic to human are mercury, lead, cadmium and arsenic. Basically, the marine organisms accumulate contaminants such as metals from the environment and have been extensively used in marine pollution monitoring programmes (Linde *et al*., 1998; Mora *et al*., 2004). These metals accumulate in fish from water, food, sediment and some suspended particulate materials (Agusa *et al*., 2005). In many countries, industrial wastes, geochemical structure and mining of metals create a potential source of heavy metals pollution in the aquatic environment due to their toxicity and accumulation behaviour. Under certain environmental conditions, these heavy metals might accumulate up to a toxic concentration and cause ecological damage (Sivaperumal *et al*., 2007). On the other hand, industrial and agricultural activities also were reported to be the largest contributor to the accumulation of pollutants in the aquatics including seawater (Jordao *et al*., 2002). Although heavy metal contamination exist at high concentrations in water

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or sediment, however, it does not involved direct toxicological risk to fish, especially in the absence of significant bioaccumulation (Fernandes *et al*., 2006). However, for certain fish and shellfish even in low concentration, the contaminant may lead to toxic level because of the bioaccumulation and magnification capability in their body. Furthermore, heavy metal pollution has been a serious matter to human health concern.

Up to this date, varied levels of heavy metals in fish samples in Malaysia were reported from various collection sites (Hajep *et al*., 2009; Irwandi *et al*., 2009; Kamaruzzaman *et al*., 2010). Findings of these studies are consistently important to indicate the safety of local fish products in terms of the heavy metals concentration. The sample from this study was collected from fish landing areas along the Straits of Malacca. The Straits of Malacca is one of the busiest shipping routes in the world, resulting the level of heavy metals be potentially high besides the various industrial activities along the West Region of Peninsular Malaysia. Therefore, this study is important to determine the level of mercury, arsenic, cadmium and plumbum in commonly consumed marine fish caught along the Straits. Additionally, the fishery industries along this coastal provide 70% of the total marine resources to the country (Annual Fisheries Statistics, 2004). Furthermore, data of this study could also be used to economically protect local and export market of the fish industries in Malaysia. In addition, it also gives important information on the safety aspect of local fish as consumer nowadays is aware on the beneficial intake of fish particularly for its high level of EFAs.

Materials and Methods

Chemical

The reagents used were pesticide residue grade; Nitric acid from Fisher Scientific, Leicestershire, UK.

Instrumentation

Inductively Couple Plasma Optical Emission Spectrophotometer (Optima 7300 DV, Perkin Elmer, Inc. Shelton, USA) and Flow Injection Mercury System (Perkin Elmer, Inc. Shelton, USA)

Sample collection

Stratified sampling method was used to collect fish and shellfish samples. Fresh fish and shellfish were collected from 10 identified fish landing areas along the Straits of Malacca (Figure 1). Collection of samples at each site was carried out twice (TI and T2) in August to November 2008. Two different sampling times was carried out to compare the effect of sampling time on bioaccumulation of heavy metals in fish and shellfish. Previously, Stow *et al*. (1994) and Bentzen *et al*. (1996) have reported abiotic factors such as proximity of fish to contaminated sediment, magnitude of contamination in their habitats and the trophic status may contribute to the degree and exposure of chemical contaminants in fish. Being one of the busiest shipping routes in the world, the Straits of Malacca is constantly exposed to various sources of contamination. All samples were immediately dipped in a mixture of water and ice to block any digestive and unfavourable changes. From collection site to laboratory, samples were transferred in polystyrene boxes containing ice and transported at refrigerated temperature (4°C).

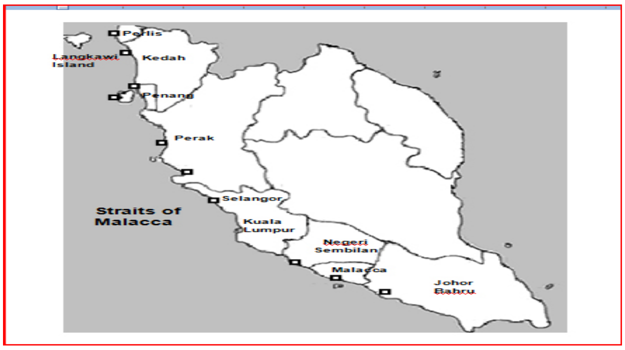

Figure 1. Location of sampling sites

Fish Samples

There were 12 species of fish; *Gymnura* spp. (Long-tailed butterfly ray), *Plotosus* spp. (Gray eel-catfish), *Nemipterus janonicus* (Japanese threadfin bream), *Epinephulus sexfasciatus* (Sixbar grouper), *Psettodes erumei* (Large-scale tongue sole), *Lutianus argentimaculatus* (Malabar red snapper), *Rastrelliger kanagurta* (Indian mackerel), *Scomberomorus guttatus* (Spanish mackerel), *Pampus argenteus* (Silver pompret), *Megalapsis cordyla* (Hardtail scad), *Eleutheronema tradactylum* (Fourfinger threadfin), *Chirocentrus dorab* (Dorab wolfherring), and three species of shellfish; *Sepia officinalis* (cuttle fish), *Anadara granosa* (Cockles), *Macrobrachium rosenbergi* (Prawn) collected. The selection of the fish samples was based on work by Osman *et al*. (2001), where most of the fish species are those very popular and most preferred by local consumers.

Sample preparation

Upon arrival in the laboratory, the collected samples were measured for their length and weight individually. Fish samples were gutted, viscera removed, beheaded, washed and filleted before frozen. Shellfish samples were also prepared by removing inedible parts, washed and frozen. All samples were kept at -75°C without any prior treatment. Before analysis, composite sample of each species was prepared by mixing and grinding homogenously the prepared samples using food processor (National, Petaling Jaya, Malaysia). All composite samples were packed into polyethylene (PE) covered cup, stored in freezer at -20°C and analyzed within a week. Before digestion process, samples were dried for 72 hours at 60-70°C using air oven (Memmert GmbH and Co, KG) and grinded using mortar.

Analysis of fish sample for heavy metals

Dry fish sample was weighed for about 0.5 g. Then, 5 ml of 65% Nitric acid was added and the mix was stored at room temperature for 1 day. After that, the sample was introduced onto heat at 60-80°C for 3 hours for digestion. After digestion, the sample was allowed to cool and filtered. Then, elute was added with 25 ml deionizer water (Cascada AN MK2, ELGA Labwater, UK). Level of heavy metals were measured by Inductively Couple Plasma Optical Emission Spectrophotometer (ICP-OES) (Optima 7300 DV, Perkin Elmer, Inc. Shelton, USA) for cadmium (Cd), arsenic (As), plumbum (Pb) and Flow Injection Mercury System for mercury (Hg) (FIMS) (Perkin Elmer, Inc. Shelton, USA) (Falco *et al*., 2006). All samples were determined in triplicate reading.delivery vehicle.

Results and Discussion

Details of 12 species of fish and 3 species of shellfish species analyzed in this study are as shown in Table 1. The classification of the habitat was based on Abdul Majid, (2004) and the samples were analyzed according to their living habitat. Knowledge on the living habitat is very important as it may represent the basis for accumulation of mercury and others trace metals in fishes (Hajeb *et al*., 2009). Furthermore, many previous literatures stated that the occurrence of trace mineral including mercury and others were also related to length, weight and age of fish (Agusa *et al*., 2005; De marco *et al*., 2006).

Table 2 shows the levels of Hg, As, Cd and Pb that were expressed in µg/ g wet weight sample. The data clearly shows variation in the level of heavy metals in fish and shellfish species of the two sampling time (T1 and T2).

Generally, the levels in T1 were higher compared to T2 (65% of samples in T1). For example, Spanish mackerel had As at 1.10 ± 0.03 µg/ g wet weight of sample and 0.64 ± 0.03 µg/ g wet weight of sample for T1 and T2, respectively. This clearly indicates the influence of aquatic environments (Stow et al., 1994; Bentzen *et al*., 1996; Zhang and Wong, 2007) as well as surrounding human activities at specific point of time may contribute to accumulation of heavy metals in fishery products. Accumulation of heavy metals in the aquatic environments have been associated with urban runoff, sewage treatment plants, industrial effluents and wastes, mining operations, boating activities, domestic garbage dumps and agricultural fungicide runoff (Alemdaroglu *et al*., 2003).

Hg is one of the most toxic elements among the studied heavy metals and exposure to high level of this element could permanently damage the brain, kidneys and developing foetus (ATSDR, 2003a; Castro-González and Méndez-Armenta, 2008). The detected values of Hg in samples were in agreement with other studies in Malaysia such as Hajeb et al. (2009) and Irwandi *et al.* (2009). Results in Table 2 clearly show that the highest value of Hg was obtained in pelagic species of Dorab wolfherring at 0.0065 ± 0.00 µg/ g wet weight of sample while the lowest was in Indian mackerel at 0.0011 ± 0.00 µg/ g wet weight of sample.

The level of Hg in the pelagic samples can be considered low as the habitat of these species that are living near to the surface of water and believed to live on eating only plankton and water plants (Fisheries Research Institute, 2004).

Table 1. List of samples with narrow range of weight and length

Local name	Common name	Scientific name	n	Habitat	Weight (g) (min-max)	Length (cm) (min-max)
Bawal putih	Silver pomfret	*Pampus argentus*	8	Pelagic	100 – 200	15 - 25
Cencaru	Hardtail scad	*Megalapsis cordyla*	5	Pelagic	100 - 250	21 - 28
Parang	Dorab wolfherring	*Chirocentrus dorab*	2	Pelagic	200 - 900	40 - 71
Kembung	Indian mackerel	*Rastrelliger kanagurta*	7	Pelagic	50 - 100	14 - 20
Senangin	Fourfinger threadfin	*Eleutheronema tetradactylum*	6	Pelagic	150 - 300	27 - 32
Tenggiri papan	Spanish mackerel	*Scromberomorus guttatus*	2	Pelagic	200 – 450	30 - 42
Kerapu	Sixbar grouper	*Epinephulus sexfasciatus*	2	Demersal	480 - 750	33 - 36
Kerisi	Japanese threadfin bream	*Nemipterus japanicus*	8	Demersal	100 - 230	18 - 25
Merah	Malabar red snapper	*Lutjanus argentimeculatus*	2	Demersal	580 - 760	28 - 37
Pari	Long-tailed butterfly ray	*Gymnura* spp.	3	Demersal	1300 - 1700	32 - 36
Sebelah	Large-scale tongue sole	*Cynoglossus arel*	8	Demersal	50 - 100	24 - 32
Semilang	Gray eel-catfish	*Plotosus* spp.	7	Demersal	350 - 600	40 - 50
Sotong	Cuttlefish	*Sepia officinalis*	20	Shellfish	20 - 45	12 - 18
Udang	Prawn	*Metapenaeus affinis*	25	Shellfish	10 - 20	12 - 17
Kerang	Cockles	*Anadara granosa*	50	Shellfish	10 - 20	2 - 5

Table 2. Level of heavy metals (µg/ g wet sample) in fish samples along the Straits of Malacca

Habitat	Common name	Moisture (%)		Mercury (Hg)		Arsenic (As)		Cadmium (Cd)		Plumbum (Pb)	
		T1	T2	T1[a]	T2[a]	T1	T2	T1[b]	T2[b]	T1	T2
Demersal	Large-scale tongue sole	84.4	77.9	1.7 ± 0.00	3.7 ± 0.00	0.59 ± 0.02	1.06 ± 0.03	1.3 ± 0.00	1.1 ± 0.00	0.19 ± 0.01	0.08 ± 0.03
	Japanese threadfin bream	76.4	74.7	4.2 ± 0.00	6.5 ± 0.00	0.77 ± 0.06	1.56 ± 0.07	0.9 ± 0.00	0.7 ± 0.00	0.20 ± 0.01	0.39 ± 0.01
	Gray eel-catfish	82.3	76.7	2.1 ± 0.00	2.1 ± 0.00	1.36 ± 0.03	0.60 ± 0.06	2.6 ± 0.00	0.5 ± 0.00	0.13 ± 0.01	0.02 ± 0.00
	Long-tailed butterfly ray	77.1	75.7	3.6 ± 0.00	5.8 ± 0.00	6.57 ± 0.05	2.13 ± 0.06	3.3 ± 0.00	1.4 ± 0.00	0.11 ± 0.02	0.08 ± 0.03
	Sixbar Grouper	75.7	78.6	3.7 ± 0.00	2.9 ± 0.00	0.14 ± 0.03	0.70 ± 0.02	0.6 ± 0.00	0.5 ± 0.00	0.01 ± 0.02	0.02 ± 0.01
	Malabar red snapper	75.6	71.0	5.4 ± 0.00	2.1 ± 0.00	0.24 ± 0.01	0.45 ± 0.02	6.0 ± 0.00	1.1 ± 0.00	0.24 ± 0.01	0.28 ± 0.01
Pelagic	Hardtail scad	76.6	76.3	2.8 ± 0.00	3.6 ± 0.00	0.54 ± 0.03	0.98 ± 0.07	1.4 ± 0.02	2.59 ± 0.00	0.18 ± 0.02	0.19 ± 0.01
	Silver pompret	74.9	75.9	1.6 ± 0.00	1.4 ± 0.00	0.58 ± 0.06	0.57 ± 0.03	3.5 ± 0.16	1.38 ± 0.00	0.31 ± 0.01	0.08 ± 0.03
	Dorab wolfherring	75.4	74.6	6.5 ± 0.00	4.3 ± 0.00	1.00 ± 0.03	0.65 ± 0.04	0.9 ± 0.38	0.68 ± 0.00	0.08 ± 0.01	0.06 ± 0.02
	Spanish mackeral	73.8	74.9	2.2 ± 0.00	2.0 ± 0.00	1.10 ± 0.03	0.64 ± 0.03	1.4 ± 0.05	0.89 ± 0.00	0.04 ± 0.01	0.01 ± 0.00
	Fourfinger threadfin	76.0	75.1	2.4 ± 0.00	1.8 ± 0.00	0.67 ± 0.03	0.78 ± 0.03	2.7 ± 0.29	0.61 ± 0.00	0.08 ± 0.02	0.01 ± 0.02
	Indian mackeral	78.5	76.3	1.1 ± 0.00	1.0 ± 0.00	0.25 ± 0.01	0.24 ± 0.03	0.7 ± 0.07	1.05 ± 0.00	0.03 ± 0.01	0.15 ± 0.02
Shellfish	Cuttlefish	84.6	82.6	2.2 ± 0.00	1.1 ± 0.00	0.57 ± 0.01	0.75 ± 0.01	6.2 ± 0.00	2.9 ± 0.00	0.33 ± 0.00	0.06 ± 0.01
	Prawn	76.7	76.3	2.8 ± 0.00	1.4 ± 0.00	1.13 ± 0.08	0.90 ± 0.01	3.2 ± 0.00	0.9 ± 0.00	0.12 ± 0.02	0.03 ± 0.01
	Cockles	75.7	76.9	3.6 ± 0.00	6.0 ± 0.00	0.88 ± 0.02	0.75 ± 0.01	47.0 ± 0.00	25.1 ± 0.00	0.19 ± 0.01	0.28 ± 0.01
Permitted Level (FAO/WHO, 2004)[c] (µg/ g)				0.5		0.1-5.0		0.05		1.5	
Permitted Level (Malaysia)[d] (mg/ g)				0.5		1		1		2	

a Level are to times with 10-3
b Level are to times with 10-2
c Joint FAO/WHO Expert Committee on Food Additives (FAO/WHO, 2004)
d Food Act 1983 and Food Regulations 1985
TI is Trip 1 (12 August 2008 until 9 September 2008)
T2 is Trip 2 (15 October 2008 until 12 November 2008)

Cd also was detected in each sample (0.6^{-2} to 3.5^{-2} µg/ g wet sample in fish and 0.9^{-2} to 47^{-2} µg/ g wet sample in shellfish) and the levels were considered low. It has been well established that the occurrence of Cd in marine aquatic environment is only in trace concentrations (ATSDR, 2003b). Cd toxicity in human may affect some organs such as kidney, lung, bones, brain as well as central nervous system (Castro-González and Méndez-Armenta, 2008). In this study, Cd was highest in cockles at both sampling time in the amount of 0.470 ± 0.00 µg/ g and 0.251 ± 0.00 µg/ g of wet weight of samples, respectively. This could be due to the fact that the habitat of Anadara granosa (cockles) that are living at the bottom of sea, nearly to the sediment where various kinds of hazardous and toxic substances are accumulated (Kamaruzzaman *et al*., 2011). According to Boscolo *et al*. (2007), bivalve mollusc species had a high capacity and propensity to concentrate pollutants. Furthermore, the environmental conditions and seasonal fluctuations of lipids may influence the heavy metals to accumulate.

For Pb in T1, the highest level was observed in cuttlefish at 0.33 ± 0.00 µg/ g wet weight of sample while in T2, the highest level was in Japanese threadfin bream at 0.39 ± 0.01 µg/ g wet weight of sample. Compared with previous study, Sixbar grouper, Spanish mackerel and Indian mackerel of this study had lower Pb levels than those samples from Langkawi Island (0.11 µg/ g, 0.1 µg/ g and 0.09 µg/ g, respectively) (Irwandi *et al*., 2009). Level of Pb in Spanish mackerel of this study ranged between 0.01 to 0.04 µg/ g wet weight of sample which was lower that other mackerel species from Aegean and Mediterranean seas (0.21-0.54 mg/ kg ww for *Scomber japonicas*) (Türkmen *et al*., 2009) and 0.01-0.02 mg/ kg ww for *Scomber scombrus* L., (Falcó *et al*., 2006). Some of the known symptoms of Pb poisoning are headache, irritability, abnormal pain and various symptoms related to nervous system (Jarup, 2003)

As is the element found in the highest concentration of the edible tissue of the species studied. Inorganic As is most toxic and considered as a Group A human carcinogenic and can effects mainly to lung, kidney, and skin disorder (ATSDR, 2003a). In this study, Long-tailed butterfly ray had the highest amount of As in both trips which were at 6.57 ± 0.05 µg/ g and 2.13 ± 0.06 µg/ g of wet weight samples in T1 and T2, respectively. These however, were still lower than the permitted level prescribed in Malaysian Food Act 1983 and Regulation 1985. The range of As in Spanish mackerel (*Scromberomorus guttatus*) of this study was 0.64 to 1.10 µg/ g of wet weight samples in the muscles tissue, which is undoubtedly lower than values in other *Scomber* sp. (*Scomber scombrus* L) with concentration range of 1.73 – 7.47 mg/kg ww (Falcó *et al*., 2006). This discrepancy could be due to the different environmental condition as well as the water and weather changes.

From this study, it can be clearly seen that the concentrations were varied among the metals and trips of sample collection. The differences in concentration in each sample were depending on species, sex biological cycle and the portion of sample being analyzed (Tuzen, 2003). Furthermore, ecological factors such as season, place of development, nutrient availability, temperature and salinity of the water also may contribute to the inconsistency of metals concentration in fish tissue (Clearwater *et al*., 2002; Tuzen, 2003). Moreover, some marine organisms have the ability to concentrate heavy metals in their tissue in several orders of magnitude higher than those in water and sediment (Law and Singh, 1991).

Contamination of heavy metals in fish and shellfish are usually compared with the permitted level recommended by the Food and Agriculture Organization and World Health Organization (FAO/WHO, 2004). Locally, the Ministry of Health Malaysia also has set the standard as stated in the Food Act 1983 and Food Regulations 1985 (Legal Research Board, 2010). From Table 2, the levels of Hg and Pb in all samples were lower than the permitted level set by FAO/WHO (2004) except Cd and As in some samples. However, the Malaysian Food Act 1983 and Regulations 1985 had higher permitted levels for Hg, As, Cd and Pb thus making the studied fish and shellfish are safe in terms of these heavy metals concentration.

Conclusion

This study has identified the level of heavy metals such as Hg, As, Cd and Pb in fish and shellfish caught along the Straits of Malacca. All metals were present in all samples but values obtained were below permitted limits set by FAO/WHO, (2004) and Food Act 1983 and Food Regulations 1985. Therefore, the local fish and shellfish from the Straits of Malacca are safe to consume in terms of these selected heavy metal concentrations.

Acknowledgements

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