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Mineral and heavy metal contents of marine fin fish in Langkawi island, Malaysia

[1]*Irwandi, J. and [2]Farida, O.

[1]*Biomolecular Engineering Research Unit (Biomeru), Department of Biotechnology Engineering, Faculty of Engineering, International Islamic University Malaysia, Jalan Gombak, 53100 Kuala Lumpur, Malaysia,*
[2]*Department of Biotechnology, Faculty of Science, International Islamic University Malaysia, Kuantan Campus, Jalan Istana, Bandar Indera Mahkota 25200, Kuantan, Pahang, Malaysia*

Abstract: A study was conducted to quantitate the concentrations of heavy metals, such as Hg, Pb and Cd in eight species of marine fin fish caught off the coast of Langkawi Island in Malaysia, as well as in its waters. The same fish were also used to determine the content of nutritional minerals, such as copper (Cu), zinc (Zn), calcium (Ca), and manganese (Mn). Fish and water samples were collected from four different areas, namely (1) Main Jetty Pulau Tuba (MJPT), (2) Teluk Cempedak Jetty (TCJ), (3) Simpang Tiga Chian Lian (STCL) and (4) Main Jetty Kuah (MJK) around Langkawi Island. Results showed that for the vital elements, all species had higher concentration of Zn compared to other elements. For the toxic elements, lead (Pb) and mercury (Hg) were found to have lower concentration of the mean values than the permissible limits set by FAO/WHO (1984). However, cadmium (Cd) level was slightly higher than the permissible limit but was still acceptable according to the Malaysian Food Regulation (1985). It can be concluded that all fish species studied are safe to be consumed.

Key words: Heavy metals, fin fish, toxic element, tolerable weekly intake, Malaysian waters

Introduction

Heavy metals are defined as metallic chemical element that has a relatively high density and are toxic or poisonous at low concentrations (Connel, 1984). Living organisms require trace amounts of some heavy metals, including calcium, copper, iron, manganese, molybdenum, vanadium, strontium, and zinc. However, heavy metals are also dangerous because they tend to bioaccumulate.

Fish, apart of being a good source of digestible protein vitamins, minerals and polyunsaturated fatty acids (PUFA), are also an important source of heavy metals. Some of the metals found in the fish might be essential as they play important role in biological system of the fish as well as in human being, some of them may also be toxic as might cause a serious damage in human health even in trace amount at a certain limit. The common heavy metals that are found in fish include potassium, copper, chlorine, phosphorus, calcium, iodine, iron, copper, zinc and manganese, mercury, lead and cadmium (Connel, 1984).

Potassium, chlorine, phosphorus, calcium, iodine, iron, copper, zinc and manganese are essential metals while, mercury, lead and cadmium are toxic metals. Besides, cadmium, chromium, mercury, lead, arsenic, cadmium and antimony are non-essential heavy metals of particular concern to the surface water systems (Kennish, 1992).

The seriousness of heavy metals leads the marine environmental pollution to be recognized as a serious matter to human health concern. Industrial and agricultural activities were reported to be the leading potential source of the accumulation of pollutants in the aquatic environment including the sea (Freedman, 1989; Gümgüm *et al.*, 1994; Nimmo *et al.*, 1998; Barlas, 1999; Tarra-Wahlberg *et al.*, 2001; Akif *et al.*, 2002; Jordao *et al.*, 2002). The noxious wastes in the sea are potentially accumulated in the sediments and marine organisms including fish which consequently transfer to human being through food chain (Tüzen, 2003). Since, fish are highly consumed by human being and may accumulate large amounts of some metals from the water, it is important to determine the concentration of heavy metals in commercial fish in

*Corresponding author
E-mail: *irwandi@iiu.edu.my*
Phone: +603 61964549

order to evaluate the possible risk of fish consumption for human health (Cid *et al.*, 2001).

This study was conducted to determine the concentration of several types of metals in marine fin fish caught in Pulau Tuba, near Langkawi Island, the most famous tourism destination in Malaysia. Heavy metal contents of the water in the island were also studied to give a better picture of the environmental condition in Langkawi. Concentration of copper, calcium, zinc and manganese were determined as vital elements, while mercury, lead and cadmium as toxic elements.

Material and methods

Sampling location and procedure

Fish and water samples were collected at Pulau Tuba, Langkawi. A water sampler of 21cc capacity was used to collect surface (0-15 m depth) and bottom (20-34 m depth) water. Four main area namely, (1) Main Jetty Pulau Tuba (MJPT), (2) Teluk Cempedak Jetty (TCJ), (3) Simpang Tiga Chian Lian (STCL), and (4) Main Jetty Kuah (MJK) were selected. Representatives sub sample were transferred into a polypropylene bottle.

On collecting fish samples, the samples were washed with clean sea water at the point of collection, separated by species and location, packed in polyethylene plastic bags. The collected samples were transferred to the laboratory under ice boxed where they were kept under freezer at -27^{o}C until analyzed.

Eight fish species commonly found in the study area were analyzed. The species names and number of the fish species analyzed are presented in Table 1. Determinations of all metal concentrations were carried out by atomic absorption spectrophotometer (AAS). Calibration of the instrument was done using standard solutions that were prepared from commercially available materials. High purity argon was used as an inert gas for the AAS analysis. All reagents used during analysis were of analytical reagent grade. Deionized water was used throughout the study. All the plastics and glassware were washed in nitric acid solutions and rinsed with deionized water before used.

Analysis of heavy metals

Prior to the analysis, the fish were gutted, cleaned, and washed with distilled water. Bones were removed and fish fleshes with the skin were minced using a domestic blender. A sample (1 g) was placed in a high form porcelain crucible. The furnace temperature was slowly increased from room temperature to 450°C in 1 h. The samples were ashed for about 4 h until a white or grey ash residue was obtained. The residue was dissolved in 5 ml of HNO_3 (25% v/v) and the mixture, where necessary, was heated slowly to dissolve the residue. The solution was transferred to a 25 ml volumetric flask and made up to volume (Vaidya and Rantala, 1996). A blank digest was carried out in the same way. All metals were determined against aqueous standards. The elements analyzed were copper (Cu), calcium (Ca), manganese (Mn), zinc (Zn), cadmium (Cd), lead (Pb) and mercury

Table 1. Names and number of fish collected from Pulau Tuba analyzed

Local name	Common name	Scientific Name	No. of fish analyzed
"Jenahak"	Golden snapers	*Epinephelus sexfasciatus*	10
"Duri"	Marine catfishes	*Lutianus agentimaculatus*	10
"Kerapu"	Groupers	*Cynoglossus lingua*	10
"Tinggiri batang"	Spanish mackerels	*Scolidon sorrakowah*	10
"Kerisi"	Threadfin breams	*Scomberomorus commersoni*	10
"Malong"	Pike and conger	*Rastrelliger kanagurta*	8
"Kembong"	Indian mackerels	*Psettodes crumei*	10
"Kintan"	Pseudo rhombus	*Arius cumatranus*	8

Table 2. Wavelength and slit widths for determination of heavy metals in water and fin fish collected in Pulau Tuba, Langkawi using flame ionization mass spectrophotometer (FIS)

Metal	Wavelength (nm)	Slit width (nm)
Copper (Cu)	324.0	0.70
Manganese (Mn)	279.5	0.20
Zinc (Zn)	213.0	0.70
Calcium (Ca)	427.7	0.70
Cadmium (Cd)	228.0	0.70
Lead (Pb)	283.3	0.70
Mercury (Hg)	253.7	0.70

(Hg). For water analysis, the samples analyzed were prepared by the method of the Association of Official Analytical Chemists (AOAC, 1995)

Stock standard solutions (Sigma, SpectrosoL-1000 mg l^{-1}) of each element were used to prepare calibration solutions to obtain calibration curves. The metal analyses of samples (Cu, Mn, Ca, Zn, Pb, Cd and Hg) were carried out by using a flame atomic absorption spectrophotometer (Perkin-Elmer, UNICAM-929). The contents of heavy metals are expressed as µg/L of the sample. The maximum absorbance of each element was obtained by adjusting the cathode lamps at specific slits and wave lengths as shown in Table 2. The heavy metal analyses either in water or fish were recorded as means ± standard deviation (SD) of triplicate measurements (Steel and Torrie, 1980).

Results and discussion

Heavy metal contents in water samples

Table 3 shows the metal concentrations in the water collected from four main locations studied in Pulau Tuba, Langkawi. Results showed that in general, the concentrations of measured heavy metals of the water were comparable in the four locations. This might be caused by the fact that all four sampling location selected are jetties normally utilized by fishermen for their daily work or by local people for transportation. Although no supportive data available, this fact could be an indication that the intensity of use of the jetties is comparable, so that pollution mainly caused by water motor vehicles in the areas are more less the same. Means values of Zn, Cd and Hg were generally found at very low concentrations. For Zn, the values were below 0.02 ppm in all four locations, while for Cd, the contents were less than 0.02 ppm. For both types of metal, no significant differences were found between the surface and bottom of sampling areas in all locations. However, in the case of Hg, samples collected from the surface and bottom of sampling locations generally gave different values. In MPJT and TCJ locations, samples from the surface of sampling areas contained higher Hg (0.004 and 0.003 ppm, respectively) than those from bottom areas (0.001 ppm in both locations), while in MJK, Hg content of samples from the bottom area was much higher (0.007 ppm) than that of the surface area (0.002 ppm). In STCL, there were no significant differences between the Hg contents of samples taken from the surface and the bottom of sampling areas.

Cu contents ranged from 2.77 to 4.00 ppm. Samples collected from MJK location were relatively low in the contents. The values were 2.77 ppm at the bottom and 2.85 ppm on the surface. The values in STCL were significantly higher, i.e., 3.90 and 4.00 ppm for the bottom and surface samples, respectively. The values for the samples collected in both MJPT and TCJ locations were 3.05 (bottom) and 3.02 (surface), and 3.81 (bottom) and 3.65 (surface), respectively. It was also shown in Table 3 that for the Cu contents, there were significant differences between the surface and the bottom of sampling areas in all locations. The difference of certain metal contents in the different depths of the water might contribute to the difference in the metal concentration of different types of fish, as each type of fish lives in the certain water depth.

Pb contents of water samples in this study varied from one location to another. The lowest values were found in MPJT. In this location, the Pb contents were 1.58 and 2.08 ppm for the surface and the bottom, respectively. The values for other locations were 2.87 (surface) and 3.04 (bottom); 3.61 (surface) and 4.50 (bottom); and 4.73 (surface) and 3.74 (bottom), for TCJ, STCL, and MJK locations, respectively.

Mn contents of all samples in this study ranged from 0.41 to 0.44 ppm. Results showed that there was no significant difference between the four locations studied, nor between the surface and the bottom of sampling areas in each location.

Ca is the major metal evaluated in this study. The values were found to range from 25.85 to 27.89 ppm. In MJPT, samples collected from the surface part gave a significantly higher value of 27.89 ppm than the value from the bottom part (25.93). In other 3 locations, there were no significant differences between the surface values and the bottom ones.

Heavy metals contents in fish species

The heavy metal contents of selected fish species studied are presented in Table 4. For the vital elements, all species had higher concentration of Zn compared to other elements. In general, Zn contents of the fish caught in Pulau Tuba ranged from 34.33 ("kembong") to 49.39 ppm ("jenahak").

Results from Table 4 also shows that Cu and Mn contents of the fish varied, ranging from 11.48 to 13.95 ppm and 16.8 to 24.35 ppm, respectively. Unlike Zn content, for Cu, "kembong" was recorded to have the highest content, followed by "kintan" (12.75 ppm) and "malong" (12.68 ppm), while the lowest Cu content was found in "kerapu". As for Mn, the highest content was found in "malong" and the lowest one was in both "kembong" and "kintan".

For the Ca element, fish species studied had various concentrations, ranging from 5.66 to 15.1 ppm. "Jenahak" was found to have the lowest Ca content, while the highest one was found in both "kembong" and "kintan". Different levels of metals were reported in different species or within the same species. The differences were explained due to the fact that the concentration of metals depends on species, sex biological cycle and on the portion of the fish analyzed (Tuzen, 2003) Moreover, ecological factors such as season, place of development, nutrient availability, temperature and salinity of the water may also cause the inconsistency of metal concentration in fish flesh.

Contamination levels of heavy metal in fish are normally compared to the permissible limits recommended by Food and Agriculture Organization and World Health Organization (FAO/WHO, 1984). However, the Ministry of Health Malaysia also has set standards called Malaysian Food Regulation (1985). Unlike for other elements, there are no limits are revealed for Cu and Mn in both FAO/WHO and Malaysian standards. Table 4 shows the mean concentrations of heavy metals in different kind of fish compared to the permissible limits set by FAO/WHO and Malaysian Government. Results from Table 4 indicates that all types of fish studied were found to contain Zn element much below the recommended limits set by FAO/WHO which is 150 ppm. The Malaysian standard for Zn is 100 ppm.

The concentration of Cu element was found slightly higher compared to the permissible limit set by FAO/WHO while Mn level was almost triple the concentration of the limit set by FAO/WHO. Though the concentration of Cu were found higher than the set limit by FAO/WHO when compared by the limit set by Malaysian Food and Regulation (1985) it is lower (Table 4).

For the toxic elements, Pb and Hg were found to have lower concentration of the mean values than the permissible limits set by FAO/WHO (1984). However, Cd level was slightly higher than the permissible limit but was still acceptable according to the Malaysian Food Regulation (1985). It can be concluded that all fish species studied are safe to be consumed.

When compared the metal concentration in water (Table 3) and fish (Table 4) it is seen that it varied. Ca and Pb were found much higher in water compared to those in the fish while Mn, Zn, Cd were less in water. Mercury (Hg) was found almost in equal concentrations. The difference can be explained due to the fact that heavy metals are not digested in the fish and they tend to accumulate (Tuzen, 2002).

The current study is the first study reporting the mineral and heavy metal contents in fish, as well as in the water samples from the Langkawi Island coastal areas. When compared to previous studies from different sampling areas in Malaysia, the metal levels in the fish evaluated in this study are comparable to the early reported studies (Table 5). However, from the results, it is also shown that the concentration of heavy metal is increasing since the concentration metal studied is almost two times compared to the early reports.

Nutritional significance of mineral content

As a consequence of heavy metal toxicity, and of the serious contamination of food that occurs from time to time during commercial handling and processing, most countries monitor the levels of toxic elements in foods. However, the potential hazards of metals transferred to humans are probably dependent on amount of muscles consumed by an individual.

Table 3. Metal concentration (ppm) recorded as means ± standard deviation (SD) in water samples collected from Langkawi coastal areas in Malaysia[1]

Metal	Location[2]							
	MJPT		TCJ		STCL		MJK	
	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom
Zn	0.02 ± 0.01^{aA}	0.02 ± 0.00^{xA}	0.02 ± 0.01^{aA}	0.02 ± 0.01^{xA}	0.02 ± 0.01^{aA}	0.03 ± 0.01^{xA}	0.02 ± 0.00^{aA}	0.02 ± 0.00^{xA}
Cu	3.02 ± 0.31^{cA}	3.05 ± 0.33^{bA}	3.65 ± 0.32^{bA}	3.81 ± 0.40^{xA}	4.00 ± 0.36^{aA}	3.90 ± 0.42^{xA}	2.85 ± 0.28^{cA}	2.77 ± 0.30^{cA}
Mn	0.44 ± 0.04^{aA}	0.43 ± 0.03^{xA}	0.42 ± 0.04^{aA}	0.42 ± 0.05^{xA}	0.44 ± 0.03^{aA}	0.43 ± 0.04^{xA}	0.41 ± 0.04^{aA}	0.41 ± 0.05^{xA}
Ca	27.89 ± 3.64^{aA}	25.93 ± 2.98^{yB}	25.85 ± 3.00^{bA}	26.37 ± 4.20^{yA}	26.87 ± 4.11^{aA}	26.91 ± 5.28^{xA}	26.81 ± 3.98^{aA}	26.97 ± 4.88^{xA}
Cd	0.01 ± 0.00^{aA}	0.01 ± 0.00^{xA}	0.02 ± 0.00^{aA}	0.02 ± 0.01^{xA}	0.01 ± 0.00^{aA}	0.02 ± 0.00^{xA}	0.02 ± 0.00^{aA}	0.01 ± 0.00^{xA}
Pb	1.58 ± 0.31^{cB}	2.08 ± 0.44^{zA}	2.87 ± 0.62^{bA}	3.04 ± 0.52^{yA}	3.61 ± 0.88^{bB}	4.50 ± 1.01^{xA}	4.73 ± 0.89^{aA}	3.74 ± 0.66^{yB}
Hg	0.004 ± 0.00^{aA}	0.001 ± 0.00^{zB}	0.003 ± 0.00^{bA}	0.001 ± 0.00^{zB}	0.005 ± 0.00^{aA}	0.003 ± 0.00^{yA}	0.002 ± 0.00^{bB}	0.007 ± 0.00^{xA}

[1] Mean of three replicates; $^{a\text{-}c}$ Means within row for surface position with different letters significantly differ ($P < 0.05$); $^{x\text{-}z}$ Within row for bottom position with different letters significantly differ ($P < 0.05$); $^{A\text{-}B}$, within a row for each location with different letters significantly differ ($P < 0.05$)
[2]Locations: MJPT, Main Jetty Pulau Tuba; TCJ, Teluk Cempedak Jetty ; STCL, Simpang Tiga Chian Lian; MJK, Main Jetty Kuah

Table 4. Heavy metal contents recorded as means ± standard deviation (SD) of selected fin fish in Pulau Tuba, Langkawi

Type of Fish	Heavy Metal Concentration (ppm)						
	Vital Element				Toxic Element		
	Cu	Mn	Ca	Zn	Cd	Pb	Hg
"Jenahak"	11.55 ± 2.10^{ab}	19.95 ± 2.67^{bc}	5.66 ± 0.88^{d}	49.39 ± 8.21^{a}	0.30 ± 0.08^{b}	1.00 ± 0.18^{b}	0.08 ± 0.02^{b}
"Duri"	12.07 ± 1.98^{a}	21.81 ± 3.20^{b}	9.66±1.02c	38.63 ± 6.44^{b}	0.90 ± 0.08^{a}	1.00 ± 0.22^{b}	0.04 ± 0.01^{c}
"Kerapu"	11.48 ± 1.90^{b}	17.85 ± 1.89^{c}	30.25 ± 4.98^{a}	38.70 ± 7.20^{b}	0.20 ± 0.05^{b}	1.10 ± 0.20^{b}	NDd
"Tinggiri"	11.74 ± 1.88^{ab}	20.13 ± 3.08^{b}	10.21 ± 1.80^{c}	38.81 ± 5.98^{b}	0.30 ± 0.06^{b}	1.00 ± 0.25^{b}	NDd
"Kerisi"	12.60 ± 2.50^{a}	17.75 ± 2.10^{c}	10.42 ± 1.90^{c}	37.23 ± 5.60^{b}	0.20 ± 0.08^{b}	0.90 ± 0.11^{b}	0.03 ± 0.01^{c}
"Malong"	12.68 ± 2.64^{a}	24.35 ± 4.21^{a}	11.86 ± 2.10^{c}	38.95 ± 7.34^{b}	0.30 ± 0.10^{b}	0.80 ± 0.10^{c}	0.02 ± 0.01^{c}
"Kembong"	13.95 ± 2.70^{a}	16.8 ± 1.80^{c}	15.10 ± 2.55^{b}	34.33 ± 5.90^{b}	0.30 ± 0.09^{b}	0.90 ± 0.10^{b}	0.02 ± 0.00^{c}
"Kintan"	12.75 ± 1.80^{a}	16.8±1.77c	15.10 ± 3.07^{b}	38.70 ± 6.80^{b}	0.20 ± 0.04^{b}	0.90 ± 0.13^{b}	0.02 ± 0.01^{c}
Permissible limits (FAO/WHO)*	10	5.4	-	150	0.2	1.5	0.14
Permissible limit Malaysia)**	30	-	-	100	1	2	0.5

Values are means of three replicate analyses of n for each species as depicted in Table 3
* FAO/WHO (1984)
**Malaysian Food regulation (1985)
ND means non detected

Table 5. Heavy metals contents in fin fish from Pulau Tuba, Langkawi, compared to previous studies in Malaysia

	Heavy Metal Concentration on selected Fish (ppm)						
	Vital Element				Toxic Element		
Studies	Cu	Mn	Ca	Zn	Cd	Pb	Hg
Present study	0.01	24.35	15.1	49.39	0.9	1.1	0.08
Ahmad *et al*, (1994)	0.84	12.06	-	23.32	0.26	1.44	-
Yap *et al.* (2004)	0.64	-	-	19.66	0.08	0.66	-

Table 6. Heavy metals concentration of fin fish in Pulau Tuba, Langkawi in human based on 2.5g for seven days consumption

Fish Common Name	Heavy Metal Concentration in selected Fish (µg)						
	Vital Element				Toxic Element		
	Cu	Mn	Ca	Zn	Cd	Pb	Hg
“Jenahak”	4.85	8.37	2.38	20.74	0.22	0.42	0.03
“Duri”	5.07	9.16	4.06	16.22	0.38	0.42	0.02
“Kerapu”	4.82	7.50	12.71	16.25	0.08	0.46	ND
“Tinggiri”	4.93	8.45	4.29	16.30	0.13	0.42	ND
“Kerisi”	5.30	7.46	4.38	15.64	0.08	0.38	0.01
“Malong”	5.33	10.22	4.98	16.36	0.13	0.34	0.08
“Kembong”	5.86	7.06	8.43	14.42	0.13	0.38	0.08
“Kintan”	5.36	7.06	8.43	16.25	0.08	0.38	0.08
Tolerable weekly intake (µg) by WHO/FAO (1984)	5.00	-	-	25	6.70-8.30	50	0.043

In Malaysia, the consumption of fish is estimated to be 21 kg/person/year or 0.06 kg/day (21 kg/265 days). The Joint Food and Agriculture Organisation/ World Health Organization Expert Committee on Food Additives (FAO/WHO, 1984) has suggested a provisional tolerable intake of Cd, Hg and PB, as well as for Ca, Mn, Cu, and Zn weekly. Table 6 shows the heavy metals that are approximately taken per week (average element concentration (ppm) × 0.06 kg × 7 days) by a Malaysian compared to tolerable weekly intake set by the FAO/WHO.

However, a precaution should be considered for the Hg element which has slightly higher and almost equal to the recommended limit for the provision tolerable weekly intake. Similarly if an adult consumes approximately 0.25 kg of fish muscles per day, then a person who consumes the fish studied will consume lower limit recommended by FAO/WHO for the provisional tolerable weekly intake for Cu, Mn as well as for Ca and Zn (Table 6).

CONCLUSION

The heavy metal concentration of muscles fish in Pulau Tuba Langkawi could be attributed to natural of anthropogenic metal sources affecting their habitats. Though the results reveal the safety consumption of fish from the human health point of hview, it is important to examine the metal concentration in fish time by time.

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