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Trace Metal In Highly Commercial Fishes Caught Along Coastal Water Of Setiu, Terengganu, Malaysia

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Abstract

Fishes in Setiu, Terengganu, Malaysia are the main source of protein for people who live near to the water continent. However, the unknown composition and concentration of heavy metal in the fish in Setiu is still in controversy and people still eating without knowing the safety. Determination of heavy metal in organism especially wide-consume by human is really important useful to understand and realize the safety of consuming the marine fish that they have caught. 211 fish with 5 types of most species consumed were collected and analysed. From 5 species analysed (Thunnus sp., Megalaspis cordyla, Rastrelliger sp., Nemipterus sp., and Selaroides sp.), the concentration of Copper (Cu), Zinc (Zn), Lead (Pb), Cadmium (Cd) and Mercury (Hg) was determined in the muscle. Metal that have acute effect on human even at low concentration for these 5 species of fish such as Cd, Pb, and Hg which is did not exceed the maximum permissible limit provided by Malaysian Food Regulation (1985). Both metal Zn and Cu that toxic at high concentration did not exceed the permissible level in muscle. In conclusion, the fish in Setiu, Malaysia can be consume or for economical uses.

Keywords— heavy metal, safety human consumption, commercial fish, toxic, bioaccumulate

INTRODUCTION

Fish is known as one of top predator for most species and dominantly contribute to the marine food chain [2,3]. As a basic need human daily diet, fish contains high protein content together with the presence of omega-3 fatty acids, fats and minerals and several essentials vitamin [3]. The continuous consumption of contaminated fish with unknown concentration of heavy metal could poses vital health risks. This is because heavy metal in fish may give an acute affect to human body [4,5]. Tendency of heavy metal accumulation in a fish is influence by their level in the food chain with their feeding habits and their area inhabit. Fish in highest level of trophic level and species with high lipid and protein contents have been establish as the most suitable indicators of environmental contamination with lipophilic compounds [6,7]. Using fish as bio-indicators are really beneficial as they are long-lived which it can tolerate or survive to pollutants over time, making a continual observation in the place with possible presence of pollutants and the outcome result shows the pollution data besides they are easily to be collected [7].

Many research and observation have been made on concentration of heavy metal in fish for the last few decade [4,5,8]. However in East Coast of Peninsular Malaysia, just a few investigations and monitoring were carried out on commercial local marine fish especially for human diet. However according to Fuad *et al.* [9], just a few species has been directed towards local marine fish. People tend eat local marine fish as it is easy to find, where to find and fish, the type to eat and the quantity to eat. Focusing on limited information on local marine fish is very necessary in order to the public who consume local marine fish needs to know information on fish they ate and address concerns about health risks from consuming these types of fishes.

Setiu has become an area with major economic and domestic tourism as there are many stunning island need to visit by the regional and foreigner. Setiu in one the busiest area where fisherman capture marine organism for diet and economical use, bringing tourist into several significant area and many sport have been held due to large people visiting alongside of the Setiu’s seashore. Capturing fish is the most prevailed activity made by human due to large necessity by local people for economical purpose and food base industrial production. Setiu known as one of the biggest marine based food production in East Coast of Peninsular. As Setiu facing the urbanization and development of several industries, Setiu’s sea may receive heavy metal pollution. Moreover, Setiu’s sea is near to South China Sea, so variety kind of fish from different kind of place may render the diversity of the fish. Fish come near to the shore in order to rest from the strong wave after long-travelled movement.

METHODOLOGY

Sampling location

Setiu coastal water was selected for the collection of most consumed caught local marine species which obtained from the observation of selling activity, most species caught and question that have been asked to the local people. The most consumed local marine fish species in Setiu are *Thunnus* sp., *Megalaspis cordyla*, *Rastrelliger* sp., *Nemipterus* sp., and *Selaroides* sp. The fish species were bought directly from local fishermen as they returned from fishing activities from Setiu coastal water. Standard length and total weight for each individual marine local fish were measured before the fish is dissected and being analyzed. The fish were stored in freezer for -4˚C. Then, the fish is directly brought to the laboratory for further analysis.

Sample Preparation and Analysis

The freeze local marine fish samples were defrosted in room temperature. Then the fish were lumped from center of the gill until the anus. Ceramic knife and sterilized (acid-washed) apparatus were used during dissection process. The targeted organ (liver and muscle) were removed and put on the small plate for drying purpose. Weight for each organ from each individual fish were measured and dried in the oven for 60˚C until constant weight was achieved. As the samples were completely dried, the samples were grinded and homogenize using porcelain mortar and pestle. Mortar and pestle were cleaned by using 70% ethanol after each organ. A total of 0.05 g of the grinded sample was digested with 1.5 mL of concentrated (suprapur) 75% nitric acid that derived from the method by Agusa *et al.* [10], Ahmad *et al.* [11] and Irwandi [12]. The sample was mixed with acid inside Teflon beaker covered with safety Teflon jacket in order to make sure the mixed sample was not vaporized during heating process. The mixture was heated at 100˚C for 7 hours. Then, the sample was transferred into centrifuge tube and added with filtered (milli-Q) water until 10mL. Analysis of metals (Cd, Cr, Pb, Cu, and Hg) in samples was performed by using induced coupled plasma mass spectrophotometer (ICP-MS).

Data analysis

\Descriptive statistics was performed to obtain mean, maximum and minimum values using SPSS for windows (version 20) software. Potential health risk assessment estimation was adapted from Wang *et al.* (2005). This method had been used by various studies in assessing health risks of heavy metals to the general public through food consumption via Hazard Quotient values [13,14]. Safe dietary intake and maximum allowable fish consumption rate estimations were adapted from Taweel *et al.* [3] using reference dose, body weight and measured concentration of heavy metal in edible portion of fish (fillet) values. One-way analysis of variance (ANOVA) was performed to test the effect of local marine species and heavy metal concentration.

Estimation of Potential Health Risk Assessment

Fish is an important food source for people who live along seashore of East Coast Peninsular Malaysia. Heavy metal intake via fish is a great concern as potential human health risk. In the estimation of potential health risk assessment, heavy metal data from fish samples were used. The safety for human consumption is using Malaysia Food Regulation 1985.

The actual concentration of heavy metal in fish (C_S) is calculated by using equation (1) [15]:

$$C_S\ (\text{ppb}) = \frac{C_E \times V_E \times D}{W} \qquad (1)$$

where C_E, is the analyzed concentration in final extract, µg/L, V_E is the final sample extract volume in milliliters, D is the dilution factor (diluted volume) and W is the sample weight in being used in grams

Another equation to determine potential health risk is the Hazard Quotients (HQ). HQ is a complex parameter used in estimation of potential health risks associated with long term exposure to chemical pollutants including heavy metal [14]. The following equation (2) was used in the estimation of potential health risk assessment adapted from Wang *et al.* [13]:

$$HQ = \frac{E_F E_D F_{IR} C}{R_{FI} W T_A} \times 10^{-3} \qquad (2)$$

where E_F is the exposure frequency (from 365 days/year for people who eat fish five times); E_D is the exposure duration (74.26 years, equivalent to average lifetime based on World Bank (2012); F_{IR} is the food ingestion rate (160 g/person/day) [16]; C is the metal concentration in sample (µg/day); R_{FC} is the oral reference dose (mg/kg/day); W is the estimation of body weight of a man (64 kg) and T_A is the averaging exposure time for non-carcinogens (365 days x E_D).

The oral reference intake (R_{FI}) for the heavy metals assessed in this study according to USEPA [15] were as follow: Cadmium (0.001 mg/kg/day); Copper (0.04 mg/kg/day); Lead (0.004 mg/kg/day); Zinc (0.3 mg/kg/day) and Hg (0.006 mg/kg/day).

Estimation of Maximum Allowable Fish Consumption Rate

Equation (2) was used and the results were expressed in kilogram of fish per day for estimating the safety daily intake. Assumption that there are no other sources of Cd, Zn, Hg, Pb and Cu exist in the local people diet, safe dietary was calculated using (3). The safe daily intake results indicated allowable fish diet rate of the studied commercial fishes. These safe dietary intake values were multiplied by seven to obtain a safety weekly intake and converted to maximum safe number of monthly fish meals using (4) applied by Moreau *et al.* [17].

$$CR_{lim} = (RfD \text{ x } W/C_m) \times 7 \qquad (3)$$

where CR_{lim} is the maximum safe daily intake rate of the fish samples (kg/day); RfD is the reference intake for each heavy metal (mg/kg/day); W is the body weight (kg) and C_m is the measured concentration of heavy metal in fillet fish (mg/kg).

$$CR_{mm} = (CR_{lim} \text{ x } T_{ap})/MS \qquad (4)$$

where CR_{mm} is the maximum permissible fish consumption rate (meals/month); CR_{lim} is the maximum safe daily intake rate of the fish samples (kg/week); T_{ap} is the typical time period in a month (4.3 week/month) and MS is the meal size, 227 g for a man [18].

RESULT AND DISCUSSION

Concentration of Heavy Metal in Commercial Marine Fish

The highest concentration of Cd was observed in *Selaroides* sp. (0.93 mg/kg) and lowest in *Nemipterus* sp. (0.03 mg/kg) as in Table 1. Meanwhile, Cu concentration shows highest in *Megalaspis cordyla* (3.04 mg/kg) and lowest in *Nemipterus* sp. (0.69 mg/kg). Highest Pb concentrations was detected in *Nemipterus* sp. (0.27 mg/kg) and lowest concentration can be found in *Thunnus* sp. (0.09 mg/kg).The level of Hg was highest in *Nemipterus* sp. (0.20 mg/kg), while *Rastrelliger* sp. (0.02 mg/kg) show the lowest. Lastly, the level of Zn was highest in *Thunnus* sp. (11.17 mg/kg) and lowest in *Nemipterus* sp. (5.86 mg/kg). Further details of the level of heavy metal in different fish species are in Table 1. The heavy metal in each studied fish species was compared with Permissible limits of Malaysian Food Regulations [1]. The levels of all metals were low and did not exceed the regulatory limit stated in Malaysian Food Regulations [1].

Table 1: Heavy metal level recorded as mean ± standard deviation (SD) in samples

Species (Common name)	Mean values for level of heavy metal (mg/kg)				
	Cu	Zn	Cd	Pb	Hg
Thunnus sp.	2.791 (± 1.21)	11.172 (± 6.92)	0.035 (± 0.03)	0.094 (± 0.06)	0.034 (± 2.34)
Megalaspis cordyla	3.043 (± 1.58)	8.969 (± 5.42)	0.304 (± 0.72)	0.119 (± 0.19)	0.055 (± 2.36)
Rastrelliger sp.	1.287 (± 1.29)	8.419 (± 5.05)	0.067 (± 0.28)	0.171 (± 0.24)	0.026 (± 2.45)
Nemipterus sp.	0.691 (± 1.11)	5.861 (± 4.99)	0.031 (± 16)	0.271 (± 0.50)	0.196 (± 6.58)
Selaroides sp.	1.362 (± 0.57)	10.411 (± 3.70)	0.927 (± 2.36)	0.236 (± 0.20)	0.126 (± 1.92)
Permissible limit Malaysia Food Regulation (1985)	30	100	1	2	0.5

ATSDR [19] reported that Cd level in marine ecosystem is only in trace quantity. Similarly, findings from local studies on coastal water fish reported lower range of Cd level in fish Fuad *et al.* [9] reported that the range of Cd mean concentration in Setiu Area using *Thais clavigera* was 5.30 mg/kg. Based on the findings of this study, it was observed that the level of Cd in fish decreased compared with the study done by Fuad *et al.* [9]. This could be an indicator of the pollution status in coastal water of Setiu. Cd was present in all of the analyzed fish samples. Cd level was the highest in *Selaroides* sp. with mean concentration value of 0.927 mg/kg, followed by *Megalaspis cordyla* (0.304 mg/kg), *Rastrelliger* sp. (0.067 mg/kg) and *Thunnus* sp. (0.035 mg/kg). Lowest Cd can be found in *Nemipterus* sp. (0.031 mg/kg). Cd levels were very low and not exceeded permissible limit stated in the Malaysian Food Regulations [1] which is 1 mg/kg. There are limited local studies performed to determine the Cu concentration in local marine fish in Malaysia especially in Setiu. A study by Fuad *et al.* [9] reported that Cu level in fish from coastal water of Setiu was in the mean of 71.46 mg/kg. The findings from present study also showed an incline in Cu levels in fish samples from coastal water of Setiu. The range of mean concentration of Cu in the samples is 0.69 to 3.04 mg/kg. The highest concentration of Cu is in *Megalaspis cordyla* (3.04 mg/kg) followed by *Thunnus sp* (2.791 mg/kg) and *Selaroides* sp. (1.36 mg/kg). Meanwhile in *Rastrelliger* sp. is 1.29 mg/kg and lastly is *Nemipterus* sp. (0.691 mg/kg). Cu levels in all the fish species assessed have not exceeded regulatory limit of the Malaysian Food Regulations (1985) which is 30 mg/kg.

The range of mean concentration of Zn in the samples is 5.86 to 11.17 mg/kg. The highest concentration of Zn is in *Thunnus sp* (11.17 mg/kg) followed by *Selaroides sp* (10.411 mg/kg) and *Megalaspis cordyla* (8.97 mg/kg) followed by *Rastrelliger* sp. (8.42 mg/kg) and lastly in *Nemipterus* sp. (5.861 mg/kg). Zn levels in all the fish species assessed still not have exceeded regulatory limit of the Malaysian Food Regulations [1] which is 100 mg/kg. Study by Fuad *et al.* [9] on fish from Setiu reported mean Zn of 80.76 mg/kg. There are a few records that have been established regarding on the concentration of Zn in Setiu besides Fuad *et al.* [9]. Concentration of Pb is in the range of mean between 0.09 to 0.27 mg/kg. The highest concentration of Pb can be found in *Nemipterus* sp. (0.27 mg/kg) followed *Selaroides* sp. (0.236 mg/kg), *Rastrelliger* sp. (0.17 mg/kg) and *Megalaspis cordyla* (0.12 mg/kg). The lowest concentration of Pb is shown in *Thunnus* sp. All species contain a level of Pb concentration not exceed to the level provided by Malaysia Food Regulation [1] which is 2 mg/kg.

The level of Hg was highest in *Nemipterus* sp. (0.20 mg/kg) followed by *Selaroides* sp. (0.13 mg/kg), *Megalaspis* sp. (0.06 mg/kg), *Thunnus* sp. (0.034 mg/kg) and lowest in *Rastrelliger* sp. (0.026 mg/kg). Hg levels in all the fish samples also did not exceed regulatory limit stated in the Malaysian Food Regulations [1] and the value provided is 0.5 mg/kg. Less study have been observed with concentration of Hg in the fish. From research by Fuad *et al*. [9], the mean concentration of Hg in *Thais clavigera* is 0.15 mg/kg. The average concentration of Hg in current study show the data is close to the Fuad *et al.* [9].

Among all the heavy metal assessed, Zn was present in the highest level in all of the fish species followed by Cu, Cd, Pb and Hg. This is due to the high transfer factor of Zn compared with other heavy metal. This indicated that bioaccumulation of Zn from water to muscle of fish is greater compared to other metals. Zinc is really useful and needed for metabolic activities and basic requirement for growth and development if taken at proper dose. Besides transfer factor, there are quite a few elements that could affect the concentration of heavy metal in fish. The other influences include seasonal variation, temperature of water and the size (length and weight) of marine organism can affect the content of heavy metal in the sample [20]. Increasing of heavy metal accumulation in fish is also associated with water temperature [21]. The example for effect of water temperature towards concentration of metals is research by Ray [20] told that Cd intensively accumulate with gradual rise of water temperature meanwhile Carvalho *et al.* [22] revealed that accumulation of Cu increases with decreasing of temperature. This research told the difference of heavy metal level in each different fish species. Seasonal variant time consist of changes in temperature, also has a role in accumulation of heavy metal by contribute to the changes affecting the temperature, pH value of water and also the body size of the marine organism. Other characteristic of water that influence the heavy metal accumulation in fish are water viscosity or density, salinity and pH of water [23]. Besides, concentration of heavy metal in commercial marine fish is dependent on size, weight and age of the marine organisms [24]. Several studies conducted on fish from local coastal water found positive correlations between level of heavy metal in fish and the weight and length of fish [7,25,26]. Other factors that possibly that can affect the concentration of heavy metal in fish are sex of fish, sex biological cycle, behavior and feeding habits, nutrient availability and habitats [21].

Potential Health Risk Assessment Output

If Hazard Quotation is more than 1 is gained, potential health risk is associated to the studied heavy metal [3]. The mean values of HQ calculated for Cd, Cr, Zn, Pb and Cu were below 1 for all fish species analyzed, thus diet on fish from study area have no significant effects of for Cd, Cr, Zn, Pb and Cu (Table 2). However, exception for Cd in *Selaroides* sp. which is more the 1.

The maximum allowable fish consumption rate (CR_{mm}) and estimation of safe dietary intake (CR_{lim}) are shown in Table 3. The Hg concentrations in all studied fish samples resulted in the highest maximum allowable fish consumption rates compared with other studied heavy metal. On the other hand, Cd concentrations were the lowest in all the studied fish samples

Table 2. Estimation of potential health risk and HQ values for commercial marine fish species.

Fish Species	Mean values for HQ of heavy metal				
	Cu	Zn	Cd	Pb	Hg
Thunnus sp.	2.39×10^{-3} (±0.001)	1.28×10^{-3} (±0.06)	1.18×10^{-3} (±0.07)	0.80×10^{-3} (±0.04)	0.19×10^{-3} (±0.01)
Megalaspis cordyla	2.60×10^{-3} (±0.001)	1.02×10^{-3} (±0.05)	10.42×10^{-3} (±1.83)	1.02×10^{-3} (±0.12)	0.32×10^{-3} (±0.01)
Rastrelliger sp.	1.10×10^{-3} (±0.001)	0.96×10^{-3} (±0.04)	2.30×10^{-3} (±0.72)	1.46×10^{-3} (±0.15)	0.15×10^{-3} (±0.01)
Nemipterus sp.	0.59×10^{-3} (±0.001)	0.67×10^{-3} (±0.04)	1.07×10^{-3} (±0.40)	2.32×10^{-3} (±0.32)	1.12×10^{-3} (±0.10)
Selaroides sp.	1.17×10^{-3} (±0.001)	1.19×10^{-3} (±0.03)	31.74×10^{-3} (±5.99)	2.02×10^{-3} (±0.13)	0.72×10^{-3} (±0.04)

It is really important to note that Hazard Quotation must be used with risk avoidance. USEPA [27] has intentionally focus that HQ value greater than 1 will not inevitably mean that acute effects will occur. Apart from local marine fish and concentration from the fish itself, there are many other sources of heavy metal exposures such as pollution and contamination from crops grown in contaminated land, metal mining, contaminated drinking water and cigarette smoke [28,29]. Even the trace amount of heavy metal could contribute to significant health risks of heavy metal in our daily life from all of these sources. Due to the existence of heavy metal in different kind of fish species, human may ingest heavy metal with potential to cause significant health effects. The maximum allowable fish consumption and specific consumption limit safe dietary intake indicated that it would be proper to minimize the weekly meals of the analyzed fish species, thus it will reduce the threat to fish consumers and the most important is it can avoid the chronic systemic effects due to effective affect from heavy metal pollution. Therefore, the maximum allowable fish consumption and estimation limits of safe dietary intake will provide to local people with specific information regarding the specific species of fish that can safely be consumed.

Table 3. Estimation of safe dietary intake (CR_{lim}) and maximum allowable fish consumption rate (Cr_{mm}) for studied heavy metal in fish sample

Fish Species	Type of Metal	Mean Concentration	Estimation of safe dietary intake, CR_{lim} (kg/week)	Maximum allowable fish consumption rate , CR_{mm} (meal/month)
Thunnus	Cu	2.791	6.36	0.12
sp.	Zn	11.172	11.92	0.23
	Cd	0.035	12.86	0.24
	Pb	0.094	18.93	0.36
	Hg	0.034	79.13	1.50
Megalaspis	Cu	3.043	5.84	0.11
cordyla	Zn	8.969	14.85	0.28
	Cd	0.304	1.46	0.03
	Pb	0.119	14.93	0.28
	Hg	0.055	48.25	0.91
Rastrelliger	Cu	1.287	13.80	0.26
sp.	Zn	8.419	15.82	0.30
	Cd	0.067	6.60	0.13
	Pb	0.171	10.41	0.20
	Hg	0.026	100.99	1.91
Nemipterus	Cu	0.691	25.70	0.49
sp.	Zn	5.861	22.73	0.43
	Cd	0.031	14.25	0.27
	Pb	0.271	6.55	0.12
	Hg	0.196	13.56	0.26
Selaroides	Cu	1.362	13.04	0.25
sp.	Zn	10.411	12.79	0.24
	Cd	0.927	0.48	0.01
	Pb	0.236	7.51	0.14
	Hg	0.126	21.21	0.40

In conclusion, all five heavy metal that have been analyzed shows below the limit enforced by the Malaysian Food Regulations [1]. The HQ values calculated for all metal are less than 1, so the targeted fish is safe to consume in long period of time.

The maximum allowable fish consumption rate based on Hg was the highest while Cd concentrations were the lowest in all the studied fish samples.

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