Journal of Environment Pollution and Human Health, 2013, Vol. 1, No. 1, 1-5
Available online at http://pubs.sciepub.com/jephh/1/1/1

DOI:10.12691/jephh-1-1-1

Human Health Risk Assessment of Metal Contamination through Consumption of Fish

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Received April 2, 2013; Revised April 24, 2013; Accepted June 20, 2013

Abstract The progress of industries has led to the increased of pollutants emission into the ecosystems. One of the most common pollutants is heavy metals. This research deals with human health risk assessment of metal contamination through the consumption of fish at selected river in Kuantan, Pahang. The objective of this research is to determine the concentration of Cu, Pb and Cd contaminant in fish. This research describes the heavy metal experiment analysis and health risk assessment. Inductively Coupled Plasma Membrane System (ICP-MS) was used to determine the concentration of heavy metals in fish. The average concentration of Cu, Pb and Cd in three locations are approximately 0.0205µg/g, 0.0145µg/g and 0.0004µg/g. Target Hazard Quotient (THQ) was used in the health risk assessment to determine carcinogenicity of the sample. The result shows that the concentration and THQ of all metal studied (Cu, Pb, Cd) are less than 1; signified that a daily exposure at this level is unlikely to cause any adverse effects during a person lifetime.

Keywords: *fish, health risk, heavy metal, human*

Cite This Article: Amirah, M.N., A.S. Afiza, W.I.W. Faizal, M.H. Nurliyana and S.Laili, "Human Health Risk Assessment of Metal Contamination through Consumption of Fish." *Journal of Environment Pollution and Human Health* 1, no. 1 (2013): 1-5. doi: 10.12691/jephh-1-1-1

1. Introduction

The rapid development of industrialization has resulted in heavy metals pollution, which is significant environmental hazard for invertebrates, fish, and humans [22]. Industrialization of Malaysia has led to polluted rivers where the heavy metals are inert in the environment and often considered to be conservative pollutants, hence causing potential threat to ecosystems. Thus, Pollutants may be directly or indirectly toxic to the aquatic flora and fauna [29]. Heavy metals are one of the pollutants that can spread in sediment components and react through ion exchange, absorption and precipitation [27]. These elements are discharge through numerous anthropogenic sources and collective into receiving systems such as sediment, soil and water. Heavy metals are non-degradable and very harmful to plants, aquatic organisms and human health at certain levels of exposure [14].

Heavy metals tends to accumulate in advanced organisms through bio-magnification effects in the food chain. Thus they can enter into human body, and accumulate in the human tissues to pose chronic toxicity. Chronic assimilation of heavy metals is known cause of cancer [15] and can damage vital organ functions. Accumulation of heavy metals in the food web can occur either by accumulation from the surrounding medium, such as water or sediment, or by bioaccumulation from the food source [20]. According to Syed Lal Shah and Ahmet Altindag [18] heavy metals may affect organisms directly or indirectly by transferring to the next tropic level of the food chain. The most serious results of their persistence are biological amplification through the food chain. In the aquatic environment, heavy metals in dissolved form are easily taken up by aquatic organisms where they are strongly bound with sulfhydryl groups of proteins and accumulate in their tissues. The accumulation of heavy metals in the tissues of organisms can result in chronic illness and cause potential damage to the population.

Mukesh K. Raikwar et al. [17] found that heavy metals such as Cd, Ni, As, Pb pose a number of hazards to humans. These metals are also potent carcinogenic and mutagenic. Heavy metal toxicity can result in damaged or reduced mental and central nervous system function, lower energy levels, and damage to blood composition, lungs, kidneys, liver, and other vital organs. Long-term exposure may result in slowly progressing physical, muscular, and Alzheimer's disease, Parkinson's disease, muscular dystrophy, and multiple sclerosis. Allergies are not uncommon and repeated long-term contact with some metals or their compounds may even cause cancer [12]. According to Ferner [7], heavy metal toxicity is a clinically significant condition when it does occur. If unrecognized or inappropriately treated, toxicity can result in significant illness and reduced quality of life.

Heavy metal intakes by fish in polluted aquatic environment are varies depends on ecological requirements, metabolisms and other factors such as salinity, water pollution level, food and sediment. Fish accumulates metals in its tissues through absorption and human can be exposed to metals via food web. This will cause acute and chronic effect to human [8]. Several methods have been proposed for estimation of the potential risks to human health of heavy metals in fishes. The risks may be divided into carcinogenic and non-carcinogenic effects [29]. Risk assessment is one of fastest method which is need to evaluate the impact of the hazards on human health and also need to determine the level of treatment which are tend to solve the environmental problem that occur in daily life [28]. Current non-cancer risk assessment methods do not provide quantitative estimate of the probability of experiencing non-cancer effects from contaminant exposure. These method are typically are based on the Target Hazard Quotients (THQ).

Although the THQ-based risk assessment method does not provide a quantitative estimate of the probability of an exposed population experiencing an adverse health effect, it does provide an indication of the risk level associated with pollutant exposure. This method of risk estimation has recently been used by many researchers [3,26] and has been shown to be valid and useful. This non-cancer risk assessment method was also applied in this study.

Rivers are polluted by industrial and residential areas. Correlation between water quality and aquatic life gave an overall picture of biodiversity index and impact of industrial towards the river and the surrounding human population. Many studies have been done in Malaysia about heavy metal contamination in water. However, information on health risk associated with heavy metals is still limited. The purpose of this study is to determine the concentration of heavy metals in fish. By using the target quotient (THQ), health risk associated with heavy metals in fish was evaluated.

2. Materials and Methods

2.1. Study Sites

The research was carried out at a selected river in Kuantan. The river was situated 1008 kilometers west (281°) of the approximate center of Malaysia and 204 kilometers north east (65°) of the capital Kuala Lumpur. With a latitude of 3.93 (3° 55' 60 N) and a longitude of 103.37 (103° 22' 0 E), this river is a hydrographic (stream) located in the state of Pahang in Malaysia. In a 100km^2 area of this river has an approximate population of 480813 (0.004808 persons per square meter) and an average elevation of 38 meters above the sea. The river was classified as a stream (Pahang Drainage and Irrigation Department). This area was selected due to rapid growth of development which are mixed development area comprises residential, commercial, oil and gas industry, small and medium enterprises (SME) industry and lodging like resort and hotel.

2.2. Field Sampling

The samples of this study are fish tissue. Fish sample was collected from a single fisherman in order to assure regularity in fishing methods. The fish was collected from 3 selected points and transported to the laboratory on the same day in the pre-cleaned polyethylene bags. All samples were frozen and stored at -18ºC immediately upon returning from the field.

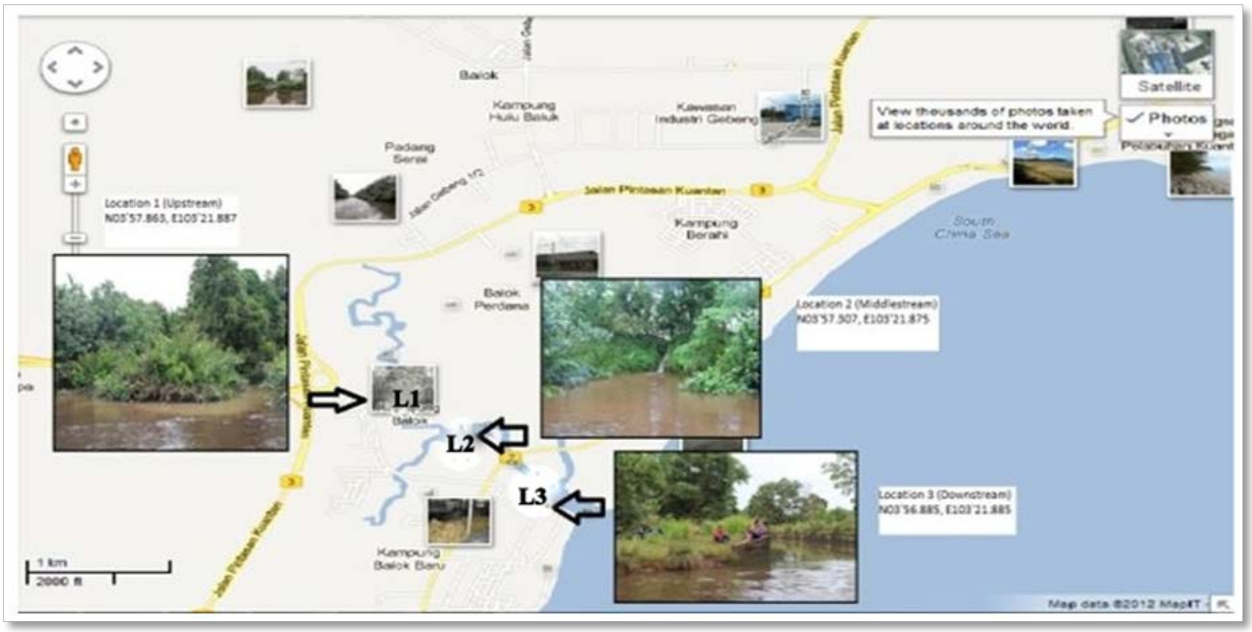

Figure 1. Location of sampling site

2.3. Sample Preparation

Sample of fish tissue weighted 0.5g was inserted directly into acid-washed Teflon digestion vessels. 10ml of ultra-pure nitric acid (HNO_3) was added into each vessel. The samples were heated to 100ºC using the XT-9800 pre-treatment heater to the point that almost all nitrogen dioxide was emitted. 4ml aliquot of concentrated HNO_3: HF (1:1 v/v) acid mixture was added before microwave digestion. One reagent blank for each digestion was included as a representative standard reference, homogeneity and process efficacy in sample replicated. The digested sample was transferred to a marked flask post-cooling.

2.4. Sample Test

All fish samples were analyzed for Cu, Pb and Cd using an Inductively Coupled Plasma Mass Spectrometer (ICP-MS). ICP-MS model used in this study was Perkin Elmer, Nexlon 300x ICP-MS from USA.

Sample digestates were typically diluted 10x with 1% HNO_3 for analysis. Greater dilutions (up to 10,000 xs) were necessary for some analytes presents at relatively high concentrations in fish digestates.

Results were quantified using an empirical calibration curve generated from the responses obtained from multiple dilutions of a multi-element calibration standard prepared from single-element standards (Alfa Aesar). Analytical quality control included analysis of a 1% ultra-pure HNO_3 blank and a sample duplicate from the microwave digestion [29].

3. Results and Discussion

3.1. Heavy Metal Concentration in Fishes

Heavy metal concentration of Cu, Pb and Cd in fish at downstream, middle stream and upstream of selected river in Pahang are summarized in Table 4.1. There are significant differences on the concentration presents in the water for the heavy metals total for Cu, Pb and Cd at all sampling points. The concentration of Cu is varied from 0.0193 to 0.0213µg/g. Pb concentration ranged from 0.0136 to 0.0152 µg/g and concentration of Cd is varied from 0.0004 to 0.0005 µg/g. This suggests the different bioavailability of metals to aquatic organisms. Among the heavy metals studied, Cu showed the highest level of accumulation. A similar situation was observed in studies by [29]. Reference [19] also reported high Cu concentration in fish which is consistent with its concentrations in surface soil, bottom sediment, and water in ponds. Cu also are one of the essential metal in fish. Thus, this study supported the fact by reference [21] which stated the levels of essential metals in the fish samples were higher than those of the non-essential metals. Pb had moderate mean concentration and the lowest mean concentration in fish is Cd at all location. Lower Cd in fish may be ascribed to its lower mean concentration in the water. Concentration of Cu and Pb are relatively lower in upstream. In contrast, concentration of Cd is highest in downstream.

Table 1. Average concentration of Cu, Pb and Cd in fish (µg/g) based on location

Element	Location 1 Upstream	Location 2 Middle stream	Location 3 Down stream	Average
Cu	0.0193	0.0213	0.0210	0.0205
Pb	0.0136	0.0147	0.0152	0.0145
Cd	0.0004	0.0004	0.0005	0.0004

Table 2. Heavy metals concentration (µg g^{-1} DW) in muscle of tilapia fish [1]

Element	Cd	Zn	Pb	Ni	Co
Serdang night market	0.02±0.000^{a}	11.36±01.28^{a}	0.10±0.020^{a}	3.95±0.290^{a}	0.25±0.220^{a}
Bangi night market	0.03±0.010^{a}	16.72±03.39^{a}	0.17±0.040^{b}	03.97±0.170^{a}	0.26±0.020^{a}
Kajang wet market	0.03±0.010^{a}	16.42±04.11^{a}	0.22±0.060^{b}	03.84±0.410^{a}	0.26±0.020^{a}
Bangi wet market	0.03±	14.35±0.72^{a}	0.21±0.03^{b}	03.97±0.25^{a}	0.26±0.020^{a}
FAO and WHO (1984)*	1 µg g^{-1}	100 µg g^{-1}	2 µg g^{-1}	-	-
MFR (1985)**	1 µg g^{-1}	100 µg g^{-1}	2 µg g^{-1}	-	-
Other study	0.021	16[1]	0.17[1]	03.60[2]	0.27[3]

Values are Mean±SD, Values in each row with the same superscript letter are not significantly different at p<0.05. *World Health Organization, **Malaysian Food Regulations (values are in µg g^{-1})

From the result, concentration of Cu, Cd and Pb is consistent for every location. At all location, Cu is the highest concentration in fish with 0.0205µg/g followed by Pb with 0.0145µg/g and the least concentrated is Cd, 0.0004µg/g. The concentration of Cu, Pb and Cd decreased in the following sequence: Cu > Pb > Cd. The concentration of Cu was higher than Pb and Cd in the water of the river. Therefore, fish inhabiting the waters accumulated and concentrated elevated levels of Cu in their body tissues from the river water. Comparing to the other studies that study on heavy metals concentrations (Cd, Zn, Pb, Ni,Co) in Tilapia fish (*Oreochromis niloticus*) from four selected markets in Selangor, Peninsular Malaysia, they found that the mean concentrations of heavy metals in the fish muscle obtained from all markets was increasing in order of scales; Zn > Ni > Co > Pb > Cd. [1].

Metal concentrations of the muscle tissues of fish caught from downstream were higher compared to that of upstream of the river because the upstream was relatively less contaminated than downstream due to the inflow of industrial effluents and municipal sewage from urban system. In consistence to the present findings, samples collected from that river showed high concentration of heavy metals due to the increasing pollution levels that resulted from input of industrial waste waters and anthropogenic sources, and municipal sewage inflow into the river. Concentrations of heavy metals in tissues of fishes from different sampling sites also showed discernible variations [2], which also corroborated the present findings. Bio concentration factor of Cu and Cd in the fish muscle tissues from the water were high. It can be concluded that the highest metal concentration is Cu.

3.2. Health Risk Assessment

The THQ which is the ratio between the exposure and the reference dose (a reference dose or RfD), is used to express the risk of non-carcinogenic effects. Ratio of less than 1 signifies non-obvious risk. Conversely, an exposed population of concern will experience health risk if the dose is equal to or greater than the RfD. The method for the determination of THQ was provided in the United States EPA Region III risk-based concentration table [24]. The dose calculations were carried out using standard assumptions from an integrated United States EPA risk analysis. Assumptions for the health risk calculation in this study are listed in Table 3.

A THQ below than 1 implies that the level of exposure is smaller than the reference dose; a daily exposure at this level is believed to cause any adverse effects during a person's lifetime.

The models for estimating THQ are [2]:

$$THQ = \frac{Efr \times EDtot \times FIR \times C}{RfDo \times BWa \times ATn} \times 10^{-3} \quad (1)$$

Where EFr is exposure frequency (365 days/year); EDtot is the exposure duration 70 years, average lifetime); FIR is the food ingestion rate (g/day); C is the heavy metal concentration in fish (μg/g); RfD0 is the oral reference dose (mg/kg/day, Table 2); BWa is the average adult body weight (55.9 kg); and ATn is the averaging exposure time for non-carcinogens (365 days/year x number of exposure years, assuming 70 years). Estimates of fish consumption in coastal cities of China [12,29], indicate that the general population eats 105 g/day of fish.

It has been reported that exposure to two or more pollutant may result in additive and/or interactive effects [11,29]. In this study the total THQ is treated as the arithemetic sum of the individual metal THQ values, derived by the method of Chien et al. [3]:

$$\text{Total THQ }(\text{TTHQ}) = \text{THQ }(\text{toxicant 1}) + \text{THQ}(\text{toxicant 2}) + ... + \text{THQ}(\text{toxicant n}) \quad (2)$$

Table 4 shows the estimated THQ for individual metals and the total THQ from consumption of fish by general population in that area.

Table 3. Oral reference doses of heavy metals [25]

Heavy metal	Cd	Pb	Cu	Zn
Rfdo (mg/kg-day)	1 x 10^{-3}	4 x 10^{-3}	4 x 10^{-2}	3 x 10^{-1}

Table 4. THQ of Cu, Pb and Cd in fish

Element	Concentration	RfDo	THQ
Cu	0.0205	0.04	0.0010
Pb	0.0145	0.004	0.0068
Cd	0.0004	0.001	0.0008
		TTHQ	0.0085

The target hazard quotients (THQs) of studied metals through consumption of fish for residents were derived and listed in Table 3. Highest THQ value belongs to Pb and it was higher than comparable values for Cu and Cd. This research found that Pb was a major risk contributor for general population in that river, accounted for 79% of the total THQ. The tolerable weekly Pb intake limit recommended by the FAO/WHO for adults is 25μg/kg body weights [5], which corresponds to 3.57μg Pb/kg body weight/day. Taking into account the average body weight of 55.9kg for adults in these areas, the tolerable daily intake of Pb will be 200μg. The dietary intake of Pb (14μg) estimated for the inhabitants in the study area is far below the tolerable limit. The next higher risk contributor element was Cu, contributing about 11% of the total THQ. Recommended daily intake of Cu for adults is 6.5mg [16]. Daily intake of 0.20mg for Cu is less than the recommended values, suggesting a dietary intake deficiency for the inhabitants the study areas. The lowest risk contributor was Cd, accounted for 9% of the total THQ. The Cd intake of 5μg/day through the consumption of fish is also less than the tolerable daily dietary intake limt (57-71μg/day) of FAO/WHO [5]. This demonstrated the relatively minor risk from Cu and Cd and dominant contribution from Pb for the inhabitants study area. The estimated THQ for individual metal decreased in following sequence: Pb > Cu > Cd. Compared with the sequence of heavy metal concentration, the THQ of Pb has been promoted to first from second. This may due to the fact that Pb is the more toxic than Cu [5]. The maximum permissible doses for an adult are 3 mg Pb and 0.5mg Cd per week, but the recommended doses are only one-fifth of those quantities. The THQ of each metal studied from this research is generally less than 1, shows that people would not experience significant health risk from the consumption of individual metals through contaminated fish from the selected river.

Table 5. Assumption for THQ calculation

Assumption for THQ calculation	Reference
Ingested dose is equal to the absorbed	
Pollutant dose	[24]
Cooking has no effect on the pollutants	[4]
The average body wweight of the chinese	
Assumed to be 55.9kg	[9]
Average lifeline of Chinese is 70 years	

4. Conclusions

The concentration of Cu is highest in all location at average 0.0205μg/g. In contrast, the concentration of Cd was the least in all location. Downstream have the highest concentration of Cu. The factors that cause it is bioavailability of metals to aquatic organisms, Cu concentration in surface soil, bottom sediments and waters in ponds. As Cu is one of the essential metals in fish then it higher than non-essential metals concentration. This shows that downstream was the most polluted river area. The concentration of Cu, Pb and Cd decreased in the following sequence: Cu > Pb > Cd. The total average concentration of Cu, Cd and Pb in all location is 0.0354μg/g.

THQ value of Cd was lowest compared with Cu and Pb. The health risks posed by exposure to heavy metals Cu, Cd and Pb through consumption of contaminated fish that were investigated based on THQ. The result show that THQ of each metal investigated in this study were generally less than 1. Thus this study supported study of Yujun Yi et al. [28] that suggest people would not experience significant health risks from intake of individual metals through fish consumption.

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