การประเมินความเสี่ยงต่อสุขภาพจากการบริโภคปลาที่มีการปนเปื้อนโลหะหนักใกล้แหล่งฝังกลบมูลฝอยเทศบาล

HEALTH RISK ASSESSMENT OF HEAVY METALS INTAKE DUE TO FISH CONSUMPTION NEAR MUNICIPAL LANDFILLS

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บทคัดย่อ

การวิจัยครั้งนี้มีวัตถุประสงค์เพื่อประเมินความเสี่ยงด้านสุขภาพจากการบริโภคปลาที่ปนเปื้อนโลหะหนัก (สารหนู แคดเมียม โครเมียม ตะกั่ว นิกเกิล แมงกานีส สังกะสีและทองแดง) บริเวณรอบแหล่งฝังกลบมูลฝอยเทศบาล อำเภอธาตุพนม จังหวัดนครพนม และศึกษาปริมาณการปนเปื้อนของโลหะหนักในน้ำ และตัวอย่างปลา 5 ชนิด 30 ตัวอย่าง ได้แก่ ปลาช่อน ปลาซิว ปลากระสูบจุด ปลากด ปลากระดี่และปลาหมอ วิเคราะห์ปริมาณโลหะหนักในเนื้อปลาด้วยเครื่อง Inductively coupled plasma optical emission spectrometry (ICP-OES) ผลการศึกษาพบว่าปริมาณโลหะหนักทุกชนิดที่ปนเปื้อนในตัวอย่างน้ำมีค่าไม่เกินมาตรฐานยกเว้น สังกะสี และสารหนู ปริมาณโลหะหนักทุกชนิดที่ปนเปื้อนในปลาทุกชนิดค่าไม่เกินมาตรฐาน ยกเว้นโครเมียม ค่าประมาณการได้รับสัมผัส

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โลหะหนักสูงสุด (EDI) จากการบริโภคปลาได้แก่ สังกะสี>แมงกานีส>โครเมียม ซึ่งพบได้ในปลาทุกชนิด ค่าประมาณการได้รับสัมผัสโลหะหนักจากการบริโภคปลาทุกชนิดมีค่าไม่เกินปริมาณสูงสุดที่แนะนำให้บริโภคได้ต่อวัน การประเมินความเสี่ยงด้านสุขภาพจากการบริโภคปลาที่มีการปนเปื้อนโลหะหนักพบว่า โลหะหนักทุกชนิดค่า human risk index (HRI) ไม่เกิน 1 ในปลาทุกชนิด

คำสำคัญ: โลหะหนัก, ปลา, แหล่งฝังกลบมูลฝอยเทศบาล, การประเมินความเสี่ยง

Abstract

This study was conducted to assess heavy metal concentrations (arsenic, cadmium, chromium, lead, nickel, manganese, zinc and copper) in water and 6 species, 36 of fish (*Channa striata, Silver rasbora, Hampala barb, Hemibagrus nemurus, Toxotes microlepis and Anabas testudineus*) near municipal landfill in Thatphanom District, Nakhonphanom Province. The estimated daily intake (EDI) and health risk index (HRI) were measured. The samples were analyzed for heavy metals using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES). The results found that heavy metal concentrations in water samples were within the standard except Zn and As. The heavy metal concentrations in all fish samples were within the standard except Cr. The highest of the EDI value of heavy metals via fish consumption were Zn>Mn>Cr in all fish species. The EDI of heavy metals in all fish species was within the maximum tolerable intake (MTDI). The human risk index (HRI) of heavy metals via fish consumption was less than 1 in all fish species.

Keywords: heavy metals, fish, municipal landfill, risk assessment

Introduction

The expanding urban communities and life style changing from agricultural to urban society, population growth, promoting tourism and higher consumption are factors that cause higher amount of solid waste in various areas. The amount of municipal solid waste in Thailand generated in 2019 was 28.71 million tonnes which is a 3 percentages increase from 2018, 2 million tons is municipal plastic waste (PCD, 2019). The municipal solid waste (9.58 million tons or 34%) was separated at its source and re-utilized. For remainder of the municipal solid waste, 10.88 million tons (39%) was disposed of appropriately, while another 7.36 million tons (27%) was still disposed improperly (PCD, 2019). Thatphanom sub-district municipal located in Nakornphanom district of Thailand. The total municipal solid waste was generated 5 tons per day. However, household hazardous waste is not separated. The open dumpsite was used to dispose municipal solid waste. Moreover, leachate management is improper, resulting in heavy metal contamination in the environment. Leachate is formed due to the interaction between the waste in the landfill and water from soil moisture, precipitation, and other liquid waste disposed of in the landfill site. Leachate can also be formed as a result of chemical and biochemical processes within the landfill. One of the adverse effects caused by solid waste disposal onto landfills is the contamination of surface and groundwater by leachate. The quantity and quality heavily depends on the waste components and rain for leachate in landfills (El-Fadel et al., 1997; Capelo & de Castro, 2007). A wide variety of contaminants such as heavy metals was reported in various studies (Rikta et al., 2018; Baun & Christensen, 2004; Kanmani & Gandhimathi, 2013; Phoonaploy et al., 2016). The heavy metals present in municipal landfills include chromium (Cr), cadmium (Cd), mercury (Hg), lead (Pb), and a metalloid such as arsenic (As). Some metals such as Cu and Zn are essential for the normal physiological regulatory functions of organisms but can bioaccumulation to toxic levels, while others such as Pb, As and Cd are non-essential and lead to intoxication through bioaccumulation in tissues (decreased fertility, cellular and tissue damage, cell death and dysfunction in a variety of organs), depending on their potential toxic effects (Mishra & Mohanty, 2008; Paul et al., 2014; Pereira et al., 2016).

In last decades, studied on heavy metals in freshwater ecosystem have been major focuses (Plessl et al., 2014; Mutia et al., 2012; Ogamba et al., 2016; Arantes et al., 2016). Fish have been recognized as bioindicator for environmental contamination, providing an integrated insight into the status of their environment over longer periods of time. This is particularly valid for most metals, as they show very long biological half-lives. Therefore, elevated tissue concentrations can occur even if the exposure is not continuous (Hofer et al., 1995). Moreover, fish is an important food source for humans in many parts of the world, and monitoring their trace element levels is therefore also important to ensure food safety. Exposure to heavy metals can cause a variety of adverse health effects. Heavy metals are transported and compartmentalized into body cells and tissues binding to proteins, nucleic acids destroying these macromolecules and disrupting their cellular functions. As such, heavy metal toxicity can have several consequences in the human body. It can affect the central nervous function leading to mental disorder, damage the blood constituents and may damage the lungs, liver, kidneys and other vital organs promoting several disease conditions (Monisha et al., 2014; Jaishankar et al., 2014). The present study investigated heavy metal accumulation in the water, and fish near the municipal landfill area. Additionally, the estimated daily intake (EDI) and health risk index (HRI) were investigated.

Methods and Materials

Study area

The study site was the fishery pond located near a municipal landfill in the Um-mao sub-district, Thatphanom district, Nakornphanom province of Thailand with the latitude and longitude 16.8843799, 104.6795057. The distance between the affected reservoir and the municipal landfill was 50 m. A municipal landfill was located approximately 1 km from the nearest residential area. In the municipal landfill area, most of the untreated effluent flows into reservoir and a small stream, particularly in the rainy season, and then flows into the Mae Kong River. Major agricultural crops around this area are rice.

Water samples

Water samples were collected from fishery pond over a distance of 50 meters downstream of the municipal landfill with glass bottle samplers, which were previously washed and rinsed with deionized water. 6 water samples were collected at multiple locations in the water body. Water samples were taken directly with the bottle’s mouth facing the flow of the water. The samples were directly acidified in the field to analyze the metal content by adding concentrated nitric acid until the pH was less than 2. Bottles, which already contained water samples, were labeled according to the sampling station and time. The samples were then put in a cool box and taken to the laboratory for metal analysis.

Fish samples

Fish were caught with a bottom trap net from 6 sample sites. Fish samples were transferred to a properly aerated cage made of bamboo. All fish was identified to species onsite by project scientists using a range of taxonomic keys and brought to the laboratory for heavy metal analysis. Muscles were removed with stainless steel scalpels and forceps, washed in distilled water, and then stored in polyethylene bags and frozen for metal analysis.

Sample preparation and analysis

Heavy metals in water analysis

Each water sample (25 mL) was put in a beaker, 1.25 mL of nitric acid was added to the beaker, and the beaker was covered with a watch glass. The beaker was then placed in a water bath at 90 °C for 30 min. After cooling, the digested samples were adjusted to a final volume of 25 mL with deionized water. The final suspended mixture was filtered through an 11-mm membrane filter with standard quantitative cellulose filter paper. The samples were analyzed by inductively coupled plasma optical emission spectrometry (ICP-OES) (Chand & Prasad, 2013).

Fish samples

The fish muscle samples were thawed and then homogenized with a laboratory blender and digested by adding 7 mL of nitric acid to 1 g of the weighted sample. One milliliter of hydrogen peroxide was then added and the samples were heated in a water bath for 2 h at 90 °C. After cooling, the digested samples were adjusted with deionized water to a final volume of 25 mL. The final suspended mixture was filtered through an 11-mm membrane filter with standard quantitative cellulose filter paper. The fish tissues were analyzed by 5800 ICP-OES. The detection limits of each element analyzed were As: 0.006 mg/kg; Cd, Cr, Zn and Ni: 0.001 mg/kg; Pb: 0.005 mg/kg; Cu and Mn: 0.002 mg/kg. The ICP-OES wavelength analyses for As, Cd, Cr, Pb, Ni, Cu, Mn, and Zn were set to 188.979, 226.502, 267.716, 220.353, 231.604, 324.752, 259.327, and 213.857 nm, respectively. The accuracy of the metal concentrations was evaluated with certified reference material (CRM) via the standard 3111C method (APHA, 2005).

Estimated daily intakes (EDI) of heavy metals through fish consumption

The EDI of metals depends on both the metals concentration in food and the daily food consumption. In addition, the body weight of the human can influence the tolerance of pollutants. The EDI is a concept introduced to take into account these factors. The EDI was calculated as follows equation 1 (Fu et al., 2008):

$$EDI = (C \times Con)/Bw \qquad (1)$$

where C (mg/kg) is the heavy metal concentration in fish, Con (kg/day) is the average daily consumption of fish in the region (0.052 kg/day); and BW (kg/person) is the average body weight of people in the region (65.30 kg).

Health risk index (HRI) for heavy metals via fish consumption

Health risk index (HRI) for heavy metals via fish consumption In this study, the HRI associated with fish consumption was defined as the ratio of the estimated daily intake of metals to the oral reference dose (RfDo) for each metal. The oral reference doses were 0.003, 1.50, 0.001, 0.004, 0.20, 0.30, 0.14 and 0.040 mg/kg/day for As, Cr, Cd, Pb, Ni, Zn, Mn and Cu, respectively (USEPA, 1997). The HRI was calculated with the following formula (USEPA, 2000)

$$HRI = EDI/RfDo \qquad (2)$$

Statistical analysis

Descriptive statistic include mean and standard deviation were used in this study.

Results

Heavy metal concentrations in the water samples

Concentrations of heavy metals in water samples were shown in Table 1. The heavy metals concentration in water samples were within the standard except Zn and As (2.526±0.06 mg/L).

Table 1 Heavy metals concentration in water samples

Heavy metals	Water samples (mg/L) (N=6)	Standard[a]
As	0.011±0.01*	0.01
Cd	0.004±0.02	0.05
Cr	0.021±0.01	0.05
Cu	0.023±0.03	0.1
Mn	0.047±0.12	1.0
Ni	0.043±0.03	0.1
Pb	0.014±0.01	0.05
Zn	2.526±0.06*	1.00

[a] Water quality standards for surface water sources, *Exceed the standard

Heavy metal concentrations in fish samples

Concentrations of heavy metals in fish samples were shown in Table 2. The highest As (0.07±0.01 mg/kg), Ni (0.87±0.06 mg/kg), Pb (0.04±0.03 mg/kg) and Zn (47.06±1.33 mg/kg) were found in *S. Rasbora,* Cu (0.79±0.03 mg/kg) was found in *H. barb*, Cr (2.00±0.17 mg/kg) and Mn (8.29±0.24 mg/kg) were found in *T. microlepis*, and Cd (0.16±0.05 mg/kg) was found in *A. testudineus*. The Cr concentration in all fish species exceeded standard of Thailand's food quality standards.

Table 2 Heavy metal concentration in fish samples

Heavy metals	*C. striata*	*S. rasbora*	*H. barb*	*H. nemurus*	*T. microlepis*	*A. testudineus*	Standard[a]
As	0.04±0.01	0.07±0.01	0.04±0.01	0.04±0.01	0.04±0.01	0.06±0.01	0.1
Cd	0.40±0.03	0.06±0.01	0.05±0.01	0.05±0.02	0.07±0.01	0.16±0.05	0.5
Cr	1.41±0.03**	1.90±0.06**	1.57±0.08**	1.37±0.13**	2.00±0.17**	1.59±0.11**	1.0
Cu	0.49±0.02	0.66±0.04	0.79±0.03	0.51±0.11	0.71±0.04	0.59±0.06	-
Mn	4.42±0.19	4.06±0.44	2.88±0.14	2.56±0.36	8.29±0.24	3.74±0.17	-
Ni	0.61±0.01	0.87±0.06	0.86±0.04	0.70±0.12	0.80±0.11	0.79±0.07	-
Pb	0.03±0.01	0.04±0.03	ND*	0.03±0.01	ND*	ND*	0.3
Zn	38.87±0.22	47.06±1.33	43.09±0.84	45.06±1.53	45.80±0.35	47.72±1.52	200-600

[a]Thailand's food quality standards, * ND =Not Detected, ** Exceed the standard

Potential health risks from heavy metal exposure via fish consumption

The EDI and HRI values for heavy metal exposure via fish consumption are shown in Table 3 and Table 4. The highest EDI of Cd was found in *C. striata*, As, Pb and Zn were found in *S. Rasbora,* Cu and Ni were found in *H. barb*, Cr and Mn were found in *T. microlepis*. The HRI of heavy metals via fish consumption were less than 1 all fish species.

Table 3 The EDI of heavy metals via fish consumption

Heavy metals	EDI (µg/kg/day)						
	C. striata	*S. rasbora*	*H. barb*	*H. nemurus*	*T. microlepis*	*A. testudineus*	MTDI
As	0.032	0.056	0.032	0.032	0.032	0.048	1.00[c]
Cd	0.319	0.048	0.040	0.040	0.056	0.127	2.00[c]
Cr	1.123	1.513	1.250	1.091	1.593	1.266	1.00[a]
Cu	0.390	0.526	0.631	0.406	0.565	0.470	4.50[b]
Mn	3.520	3.233	2.293	2.039	6.602	2.978	11[d]
Ni	0.486	0.693	0.696	0.557	0.637	0.629	0.90[c]
Pb	0.024	0.034	ND	0.024	ND	ND	0.30[c]
Zn	30.953	35.438	34.314	35.882	34.435	38.001	40[d]

MTDI Maximum tolerable intake [a] FAO/WHO (2002), [b] FAO/WHO (2004), [c] JECFA (2005), [d] Institute of Medicine (US) Panel on Micronutrients (2001)

Table 4 The HRI of heavy metals via fish consumption

Heavy metals	*C. striata*	*S. rasbora*	*H. barb*	*H. nemurus*	*T. microlepis*	*A. testudineus*	RfDo (mg/kg/day)
As	0.106	0.186	0.106	0.106	0.106	0.159	0.0003
Cd	0.319	0.048	0.040	0.040	0.056	0.127	0.001
Cr	0.001	0.001	0.001	0.001	0.001	0.001	1.50
Cu	0.010	0.013	0.016	0.010	0.014	0.032	0.04
Mn	0.003	0.002	0.002	0.001	0.005	0.002	1.40
Ni	0.024	0.035	0.035	0.028	0.032	0.031	0.02
Pb	0.007	0.010	ND*	0.007	ND*	ND*	0.0035
Zn	0.103	0.151	0.114	0.120	0.148	0.127	0.30

*ND= not detected

Discussion

Heavy metal concentrations in the water samples

The Heavy metal concentrations in the water sample were within the water quality standards for surface water sources recommended by the Pollution Control Department in the Ministry of Natural Resources and Environment of Thailand except Zn (PCD, 1994). The Heavy metal concentrations in municipal landfill depend on various factors such as the composition of the waste, biochemical processes that occur in the degradation stages of the waste, amount of moisture, and the local parameters (Ma et al., 2018). The results of this study found that Zn is the highest concentration in water samples. Most of the Zn concentrations in the water from the municipal landfill are likely to be from spent battery. Since the solid waste in this site is not sorted. Portable batteries have become essential in supplying energy to various electronic devices such as remote controls. A considerable amount of battery wastes is generated due to the short lifespan. In particular, primary cells such as alkaline and zinc-carbon batteries are non-rechargeable and disposed of after one-off discharge. Hence, this can create serious environmental problems during disposal process as there are hazardous components such as heavy metals (Nyame et al., 2012; Intamat et al., 2017; Phoonaploy et al., 2016; Sriuttha et al., 2017). Heavy metals contaminated in water samples were from leachate. Leachate causes the pollution of the surface water, groundwater and would finally be enriched in the sediments by adsorption, complexation, flocculation, and sedimentation (Wang et al., 2016). The improper leachate management in this area resulting in leachate spread to environment and organism. Landfill leachate is a concern since it is a complex mixture composed of several pollutants, such as heavy metals, soluble inorganic and organic compounds (Mavakala et al., 2016; Naveen et al., 2017).

Heavy metal concentrations in fish samples

This study investigated the heavy metal concentrations in local fish that were consumed by local people who live around municipal landfill. Six fish species, 30 samples were investigated. The Cr concentration in all fish species exceeded standard of Thailand’s food quality standards. Whereas the other heavy metal concentrations were within the standard. This result indicates that to consume fish from this area may expose the Cr. The range of Cr concentrations in fish samples was 1.37-2.00 mg/kg. This result concurs with a previous study in the Khon Kaen municipal landfill (Sriuttha et al., 2017), in that the range of Cr concentration in four species of fish was 0.57-1.90 mg/kg. Hexavalent chromium (Cr (VI)) is the toxic form of chromium whereas trivalent form is relatively non-toxic (Velma et al., 2009). Cr (VI) has higher solubility or higher mobility rate in aquatic medium (Vinodini & Narayanan, 2009a). It can readily penetrate the gill membrane by the process of passive diffusion which is mediated by pH of the system; as a result, they are accumulated in fish which live in the surface water. Cr (VI) is allowed to enter easily into the cytoplasm of aquatic organisms (Vinodini & Narayanan, 2009b). Thus, it can be said chromium enters the body of the fishes mainly through the gills (Velma & Tchounwou, 2009). The bioaccumulation of Cr depends upon size and organs. With subsequent increase in size and dimension, the concentration of Cr in soft tissues and shell is reduced substantially. Cr accumulation occurs differently in various sorts of tissues. Its concentration have found to be highest in gills, kidneys and liver of fish while hardly any tendency for Cr accumulation in muscular tissues (Sadiq, 1992). The high Cr concentrations were found in fish samples in this study. In addition, both physical and chemical properties of water and seasonal changes are the main factors accountable for the intensification of heavy metals in various types of fish tissues Cr accumulated in fish may affect the physical, behavioral, biochemical, histological alterations, genetic and immunological (Arunkumar et al., 2006). The aquatic toxicology of Cr depends on both biotic and abiotic factors. The biotic factors include the type of species, age and developmental stage. The temperature, concentration of Cr, oxidation state of Cr, pH, alkalinity, salinity, and hardness of water constitute the abiotic factors. Moreover, lethal and sub-lethal concentrations of the metal and its speciation also determine the sensitivity of the individual organism (Velma et al., 2009). Fish provides essential nutrients especially proteins. Local people eat fish regularly, thus they are at a risk of Cr exposure.

Potential health risks from heavy metal exposure via fish consumption

Fish consumption has been identified as one of the major pathway of human exposure to the toxic heavy metals. The EDI of heavy metals in all fish species were within the maximum tolerable intake (MTDI). This indicates that these fish might low risk of heavy metals to the consumers in the study area. Though the EDI of the studied metals were lower than the MTDI, but periodic surveillance will be necessary to set up regulatory norms for dietary intake of those fish species.

HRI are parameters for risk assessment which compare the ingestion amount of a pollutant with a standard reference dose and have been widely used in the risk assessment of metals in contaminated foods. In the present study, the average HRI values for all the six fish species were below 1, which indicates that the intakes of metals by consuming these fish do not result in an appreciable hazard risk for the human body. HRI exceeding 1 indicates that the metals are toxic and present a hazard to human health (Li et al., 2013). Toxic heavy metals (Pb, Cd, As) are generally considered the most toxic to humans and animals; the adverse human health effects associated with exposure to them, even at low concentrations, are diverse and include, but are not limited to, neurotoxic and carcinogenic actions (Castro-González & Méndez-Armenta, 2008).

Suggestion

The public should be in control of the leachate, and increased public awareness is needed to reduce the risk to human health.

Conclusion

This is the first study that revealed heavy metals concentration in water and fish samples near Thatphanom municipal landfill. All of the samples contained heavy metals contamination, although the heavy metal concentrations were within the standard ranges allowed by Thailand food quality standard except Cr concentration in all fish species. It may be a potential source of human health risks for those living around the municipal landfill.

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