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SELECTED HEAVY METALS CONTENT IN COMMERCIAL FISHES AT DIFFERENT SEASON LANDED AT FISHERIES DEVELOPMENT AUTHORITY OF MALAYSIA COMPLEX, KUALA TERENGGANU, MALAYSIA

ONG MENG CHUAN[1*], NORHAZIRAH ABD AZIZ[1], NOOR AZHAR MOHAMED SHAZILI[2] AND KAMARUZZAMAN YUNUS[3]

[1]*School of Marine and Environmental Sciences, Universiti Malaysia Terengganu, 21030 Kuala Nerus, Malaysia.*
[2]*Institute of Oceanography and Environment, Universiti Malaysia Terengganu, 21030 Kuala Nerus, Terengganu, Malaysia.*
[3]*Kulliyyah of Science, International Islamic University Malaysia, Bandar Indera Mahkota, 25200 Kuantan, Pahang, Malaysia.*

**Corresponding author: ong@umt.edu.my*

Abstract: Fish is a common table food consumed by people for protein nourishment in Kuala Terengganu state. Therefore, a study was carried out to determine the bioaccumulation level of selected heavy metals; copper (Cu), zinc (Zn), arsenic (As), cadmium (Cd) and lead (Pb) in muscle of four most popular and landed fish species which are *Nemipterus furcosus* (Fork-tailed threadfin bream), *Katsuwonus pelamis* (Skipjack tuna), *Decapterus macrosoma* (Shortfin scad) and *Atule mate* (Yellowtail scad) which were caught at different season and landed at Fisheries Development Authority of Malaysia Complex, Kuala Terengganu, Malaysia. A total of 30 individuals for each species were collected during monsoon (December 2015) and non-monsoon season (September 2016). Concentrations of these heavy metals (Cu, Zn, As, Cd and Pb were determined by using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) after 0.05 g of dried samples were digested with 1.5 mL of Suprapur nitric acid. The average concentrations of Cu, Zn, As, Cd and Pb for samples of non-monsoon season are 3.31±0.26 mg/kg dry wt., 14.9±2.68 mg/kg dry wt., 9.12±3.45 mg/kg dry wt., 0.05±0.01 mg/kg dry wt. and 0.23±0.08 mg/kg dry wt., respectively. On the other hand, the average concentration of the same metals in samples of monsoon season are 5.25±1.23 mg/kg dry wt. for Cu, 32.7±9.67 mg/kg dry wt. for Zn, 24.9±6.77 mg/kg dry wt. for As, 0.11±0.07 mg/kg dry wt. for Cd and 0.66±0.21 mg/kg dry wt. for Pb, respectively. Generally, the average metals concentrations in this study were below the permitted levels set by Malaysian Food Act 1983 and Food Regulation 1985 except for As and Cd. However, the provisional tolerable weekly intake (PTWI) have been calculated for each metals and found that the PTWI value for Cd in *K. pelamis* caught during monsoon season was exceeded the permitted level set by FAO/WHO. This study would be useful for creation of guidelines to protect the public from the harmful effects of the toxicant present in the fish that is consumed by Kuala Terengganu people.

KEYWORDS: Heavy metals, toxicology, fishes, muscle, season, permitted level

Introduction

Fisheries are one of the most important food production sectors in supplying protein to the human population (Kamaruzzaman *et al.,* 2011). It has always been seen as a food necessary for a good health (Ozuni *et al.,* 2010). Due to the increasing health consciousness of the consumers, seafood demand increases drastically during the past decade (Kamaruzzaman *et al.,* 2011; Alina *et al.,* 2012). Malaysian, being blessed with abundance fish supplies of various species and other seafood products, consumed about 60-70% of protein from such sources (Tukiman *et al.,* 2006; Zuraini *et al.,* 2006). However, the present of some heavy metals within the fish may change the good nutritional value to human into some chronic diseases. Marine organism such as fish can accumulate toxic trace metals both through the food chain and through water. They also can be considered as one of the most significant indicators of toxicity (Omar *et al.,* 2014; Authman *et al.,* 2015). Muscle is main edible part of fish by human and thus forms the most preferred tool for assessment of public health risk associated with metal pollution in fish (Reinfelder *et al.,* 1998; Kaneko & Ralston, 2007; Yi *et al.,* 2011).

Diet is considered as the main route of human exposure to heavy metals which can be toxic in larger amounts (Olmedo *et al.,* 2013). Hence, in the last few decades, the concentration of heavy metals in fish has been extensively studied in different parts of the world (El-Moselhy *et al.,* 2014).

In recent years, heavy metals accumulation in fish and other organism has been investigated along the Terengganu waters (Kamaruzzaman *et al.,* 2008; Kamaruzzaman *et al.,* 2009; Ong *et al.,* 2014; Ong *et al.,* 2016; Ong & Gan, 2017; Ong *et al.,* 2017a; b). Terengganu state is situated in north-eastern Peninsular Malaysia and is bordered in the northwest by Kelantan, the southwest by Pahang and the east by the South China Sea. Terengganu have the average temperature around 23 ºC to 32 ºC during non-monsoon season while it has been received heavy rain fall during monsoon season. North-east monsoon blows season which started from November until January every year. At this time, the rainfall quantities increase together with the strong winds and waves (Akhir *et al.,* 2017). Heavy rainfall during monsoon seasons will increased the amount of heavy metals loading into the ocean (Ong *et al.,* 2015). In addition, heavy metals discharged into the marine environment can damage both marine species diversity ad ecosystem due to their toxicity and accumulative behaviours (Gumgum *et al.,* 1994; Matta *et al.,* 1999).

This study was carried out to investigate the difference concentration of selected heavy metals within selected commercial fish in monsoon and non-monsoon seasons. Findings of this study revealed the influences of seasonal variation in South China Sea towards the concentration of heavy metals within the commercial fishes. In this study, the muscle tissues were analyzed to determine the level of heavy metal concentration as it is the major edible part to human as concerning on human health risk. There were five elements of heavy metals detected in four species of commercial fish landed at LKIM Complex, Kuala Terengganu. The average of copper (Cu), zinc (Zn), arsenic (As), cadmium (Cd) and lead (Pb) were detected in muscle of *N. furcosus* (Fork-tailed threadfin bream), *K. pelamis* (Skipjack tuna), *D. macrosoma* (Shortfin scad) and *A. mate* (Yellowtail scad). These four selected species of commercial fish are among the most popular species based on customer preferences in Kuala Terengganu (Aziz, 2016).

Materials and Methods

Fisheries Development Authority of Malaysia Complex or LKIM Complex located at Pulau Kambing, Kuala Terengganu was used to land all the fishes that caught at Terengganu including Pulau Kapas and Pulau Bidong area (Figure 1). Fishes that landed here then will be distributed to the other places in Kuala Terengganu region. In this study, *N. furcosus, K. pelamis, D. macsoma* and *A. mate* were purchased from the LKIM Complex at two difference seasons. The monsoon season samples were purchased in December 2016 while the non-monsoon season samples were purchased in September 2017. A total of 30 individual for each species and season have been purchased for this study.

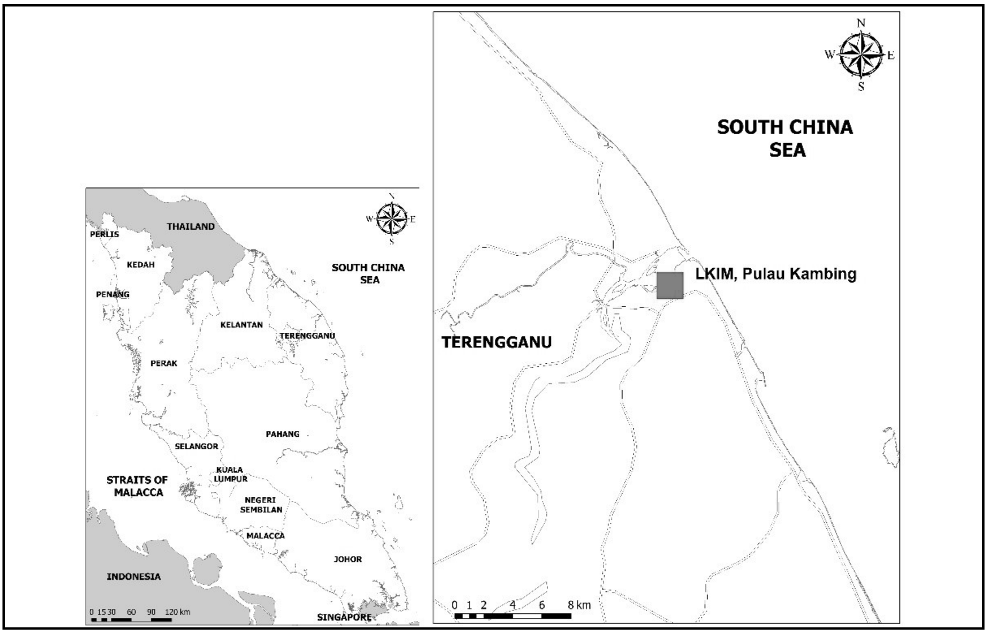

Figure 1: Map showing the location of LKIM Complex, Kuala Terengganu.

The samples were brought to the laboratory at the same day, weighed, measured and dissected. Pre-analysis process was done before the actual heavy metals analysis as to keep the samples from contamination with other impurities. The pre-analysis processes were including acid wash treatment, labeling and recording the details of samples, dissection process and samples homogenization.

For dissection process, the muscles of the fish were dissected and cleaned with deionized water before the drying process begins. Drying process was done at 60 ºC until they were reached to a constant weight. Next, the dried samples were grinded and the digestion process proceeded after 0.05 g of dried samples mixed with the 1.5 ml of Merck Suprapur nitric acid. The digestion process takes places in 8 hours with 100 °C. The digested samples were allowed to cool down to room temperature before it will be top up with Mili-Q water up to 10 ml. For the blank reagent, the solution was using the same suprapur nitirc acid (HNO_3) without addition of fish muscle samples. Meanwhile, for standard material, suprapur nitirc acid (HNO_3) was mixed up with 0.05 g of DOLT-4 fish liver. Both reagents were processes simultaneously with other samples each time the samples were run. Finally, the concentrations of these heavy metals were determined by using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (Ong & Gan, 2017; Ong *et al.,* 2017a).

In concerning on human health risk, the Provisional Tolerable Weekly Intake (PTWI) has been calculated for the four selected species (Chen *et al.,* 2018; Zaza *et al.,* 2015). The PTWI value is an estimation of the amount of the contamination that can be consumed and ingested over a lifetime without appreciable risk (Suhaimi *et al.,* 2005). The formula of PTWI calculation was shown as followed:

PTWI = Heavy metal conc. (mg/kg wet wt.) x 0.42 kg/person/week

The results calculated by using the formula were compared to the permitted values set by FAO/WHO (1984; 1988).

Results and Discussion

The average length and weight values of each species measured are tabled in Table 1. The levels of heavy metals (Cu, Zn, As, Cd and

Pb) measured in *N. furcosus, K. pelamis, D. macsoma* and *A. mate* for two different seasons are presented in Table 2 and Table 3, respectively. Each species has 30 fishes and the total samples are 240 samples for both sampling sessions. All species are chosen due to highly consumed by Kuala Terengganu people and available throughout the years.

Table 1: The species of fish, number of fish, mean total length and mean total weight.

Species	Season	N	Total Length	Total Weight
N. furcosus	Non-monsoon	30	19.5±1.42	107±18.3
	Monsoon	30	22.2±2.13	148±21.4
K. pelamis	Non-monsoon	30	32.5±3.25	433±46.2
	Monsoon	30	36.6±4.10	628±51.9
D. macrosoma	Non-monsoon	30	14.5±1.08	60.7±10.1
	Monsoon	30	25.0±2.15	152±23.6
A. mate	Non-monsoon	30	24.8±3.68	216±31.3
	Monsoon	30	21.0±4.13	109±13.7

Copper concentrations in muscle of *N. furcosus, K. pelamis, D. macsoma* and *A. mate* were lower during non-monsoon season. The mean Cu concentration during non-monsoon season ranged from 2.51±0.32 to 4.52±0.12 mg/kg dry wt. for all the four fish species collected during that season. Meanwhile, highest average concentration of Zn for non-monsoon samples were found in *K. pelamis* with 28.1±6.54 mg/kg dry wt. and the lowest one was *N. furcosus* with 12.5±1.67 mg/kg dry wt. On the other hand, the As level in non-monsoon samples were ranged from 6.54±1.69 to 12.3±1.90 mg/kg dry wt. However, Cd level were considered as the lowest among other detected elements which they were ranged from 0.02±0.01 to 0.08±0.01 mg/kg dry wt. Pb concentration do not varied significantly among four species ranged were from 0.13±0.01 to 0.20±0.02 mg/kg dry wt. with the p value > 0.05 calculated from t-test statistical analysis.

Table 2: The concentration of heavy metals (Cu, Zn, As, Cd and Pb) in muscle samples of four commercial fish caught during monsoon and non-monsoon seasons.

Species	Season	Concentration (mg/kg dry wt.)				
		Cu	Zn	As	Cd	Pb
N. furcosus	Non-monsoon	2.51±0.32	12.5±1.67	7.05±0.89	0.02±0.01	0.15±0.07
	Monsoon	2.82±0.54	22.7±1.34	56.4±15.3	0.05±0.01	0.46±0.11
K. pelamis	Non-monsoon	4.52±0.12	28.1±6.54	7.92±2.35	0.08±0.01	0.20±0.02
	Monsoon	5.51±0.78	36.6±8.70	15.1±4.83	0.20±0.12	0.54±0.19
D. macrosoma	Non-monsoon	2.82±0.51	10.4±2.31	6.54±1.69	0.02±0.01	0.19±0.05
	Monsoon	5.99±1.63	42.3±12.3	14.9±5.81	0.14±0.01	0.43±0.15
A. mate	Non-monsoon	3.32±0.42	14.0±1.68	12.3±1.90	0.06±0.01	0.13±0.01
	Monsoon	6.67±0.22	29.2±0.21	13.3±0.12	0.05±0.01	1.23±0.01

For monsoon season, all metals detected were drastically increased in compare to non-monsoon season samples. The highest concentration of Cu in monsoon samples were found in *A. mate* with 6.67±0.22 mg/kg dry wt. while the lowest one was in *N. furcosus* with 2.82±0.54 mg/kg dry wt. Meanwhile, the ranges of Zn levels for monsoon season samples were from 22.7±1.34 to 42.3±12.3 mg/kg dry wt. Concentration of As and Cd in muscle of monsoon season fish samples were detected from 13.3±0.12 to 56.4±15.3 mg/kg dry wt. and from 0.05±0.01 to 0.20±0.12 mg/kg dry wt., respectively. Besides, the highest concentration of Pb can be found in *A. mate* with 1.23±0.01 mg/kg dry wt. followed by *K. pelamis* and *N. furcosus* with 0.54±0.19 mg/kg dry wt. and 0.46±0.11 mg/kg dry wt., respectively.

Table 3 showed the mean concentration of heavy metals detected in two different seasons. For non-monsoon seasons, the highest mean concentration of heavy metals was Zn which is 14.9±2.68 mg/kg dry wt. followed by As and Cu with 9.12±3.45 and 3.31±0.26 mg/kg dry wt., respectively. Meanwhile, mean concentration of Pb and Cd were almost the same with 0.23±0.08 and 0.05±0.01 mg/kg dry wt. respectively. However, the concentrations of selected metals were dramatically increased in the samples caught during monsoon seasons. The highest concentration can be found in Zn with 32.7±9.67 mg/kg dry wt. followed by As and Cu with 24.9±6.77 and 5.25±1.23 mg/kg dry wt., respectively. Lead concentration also showed significantly increasing value in compare to the non-monsoon season with 0.66±0.21 mg/kg dry wt. Cd concentration was the lowest one with 0.11±0.07 mg/kg dry wt. It can be concluded that the levels of heavy metals detected during monsoon seasons with the following sequence, Zn>As>Cu>Pb>Cd.

Table 3: Mean concentration of heavy metals during non-monsoon and monsoon seasons.

Season	Concentration (mg/kg dry wt.)				
	Cu	**Zn**	**As**	**Cd**	**Pb**
Non-monsoon	3.31±0.26	14.9±2.68	9.12±3.45	0.05±0.01	0.23±0.08
Monsoon	5.25±1.23	32.7±9.67	24.9±6.77	0.11±0.07	0.66±0.21

Target organs such as liver, gonads, kidney and gills have the tendency to accumulate metals in high values. However, muscle part was used in this study as it was important because it is the chief edible part of fish. According to Yilmaz (2005) stated that, muscle is not an active tissue in accumulating metals. However, this study was concerned on public human health due to the highly intake of fish of Malaysian communities. The study of the metals concentration in this part can be a guideline to the consumer to estimate the intake of metals once they consumed the fish.

The affinity for metal absorption from contaminated water may be differ from one species to others due to some factors such as ecological needs, metabolism and the contamination level of water, feeding habits as well as other factors such as salinity, temperature and increasing agents (Romeoa *et al.,* 1999; Al-Yousuf *et al.,* 2000). In this study, samples were collected in two different seasons which were monsoon and non-monsoon seasons. There were some differences between these two different seasons including the temperature and rate of rainfall received. Monsoon season in Terengganu started on November until January every year with heavy rainfall (400 mm) and the temperature around 19-22ºC (Terengganu Tourism, 2015). However, during non-monsoon season, the temperature was slightly increased up to 23-32ºC with decreasing in rainfall (190mm) (Akhir *et al.,* 2015).

It was clearly shown that the level of metals detected were higher in samples during monsoon season in compare to non-monsoon. It can be due to the changes of temperature and the rainfall received in these two different seasons. The increasing the rainfall quantities might lead to the dilation factors in Terengganu waters which then resulted in the decreasing of pH and salinity. According to Prabal *et al.* (2011), the lowering pH might facilitate the dissolution of the precipitated forms of metals and increase the amount of metallic ions in solution. Other than that, the variation in metals concentration might be due to the sizes, length of fish and the habitat (Nor Hasyimah *et al.,* 2011; Canli & Atli, 2013). In this study, there were three species of pelagic fish were selected namely, *A. mate, D. macrosoma* and *K. pelamis,* and one demersal fish, *N. furcosus*.

Table 4 shows that As, Zn and Pb level in samples of monsoon season were exceeded the permitted level while in non-monsoon season samples, only As was exceeded the permitted level. It can be concluded that As level during both seasons were in higher concentration and aquatic organism in this study area were exposed to its pollution. Hence, further assessment on As pollution in Terengganu waters should be focused and treated.

Table 4: The permitted level set by varies regulation and the level of heavy metals in this study.

Regulation /Act	Weight Basis	mg/kg dry wt.				
		Cu	Zn	As	Cd	Pb
MFR (1985) FAO/WHO (1984)	Wet	30	100	1	2	1
	Dry	10	150	0.2	1.5	0.1-5.0
This Study (monsoon)	Wet	21.1±3.98	132±39.7	0.39±0.18	2.68±0.79	103±23.4
	Dry	5.25±1.23	32.7±9.67	0.11±0.07	0.66±0.21	24.9±6.77
This Study (non-monsoon)	Wet	8.43±1.06	37.9±6.82	0.13±0.02	0.58±0.23	23.2±7.92
	Dry	3.31±0.26	14.9±2.68	0.05±0.01	0.23±0.08	9.12±3.45

On the other hands, there were few other factors that control the accumulation of heavy metals in fish such as biological factors as well as environmental factors (e.g: age, body size, feeding habits and living habitat). In this study, the highest value of average heavy metal concentration during monsoon seasons was found in *N. furcosus* with 16.5±2.46 mg/kg dry wt. According to Ambak (2010) stated that, *N. furcosus* was mainly feed on crustaceans and small fishes. Heavy metals are bioaccumulative in ecosystem, and they can biomagnified in predatory animals such as fish that are also used as human food (Ganbi, 2010; Guerin *et al.,* 2011). Other than that, *N. furcosus* is a demersal fish that living near to the bottom of the sea. It was suggested that the bottom fish were likely to have a tendency to accumulate higher heavy metal concentrations than fish inhabiting the upper part of water column as they were in direct contact with the sea floor (El-Moselhy *et al.,* 2014). Moreover, Irwandi and Farida (2009) have suggested that the wastes in the sea are potentially accumulated at the bottom of the sea and other marine organism which then consequently transfer to human via food chain.

In aquatic system, heavy metal pollutants can be entered the fish body system through several pathways such as absorption from the water surrounding, or by the uptake of contaminated food, thus biomagnified via food chains to eventually pose a risk to the health of human that consumed the fish (Phillips, 1995). Generally, the increasing of metal concentration in aquatic environment will give the adverse effects to the marine organism and disturbed the cycle of food chain and food web. Heavy metal could reach food chain through various biochemical processes such as bioconcentration, bioaccumulation and ultimately biomagnified in various trophic levels and eventually threaten the health of human by seafood consumption (Kamaruzzaman *et al.,* 2011). In this study, *K. pelamis* showed the highest concentration of heavy metals during non-monsoon seasons. This species is one of the tuna species that can reach up to 1 m in length. Basically, the sizes of fish are often related to the age of the species or the fish life stage, the life phases of fish might give a suggestion on the amount of heavy metals that accumulated within the fish body.

Table 5 shows the provisional tolerable weekly intake (PTWI) (wet wt.) which calculated in four species of marine fish. The calculated values will be compared with the permissible level of PTWI set by FAO/WHO and USEPA. Based on the result, only the value of PTWI for Cd in *K. pelamis* during monsoon season has exceeded the permitted level set by FAO/WHO. The permissible limit for Cd is 0.441 mg kg^{-1} bw/week for 63 kg adult. In this study, a reference adult which is 63 kg of body weights was used to determine the amount of heavy metals consumed by an average Malaysian. This average body weights data of Malaysian adults was referred from the Malaysian Adults Nutritional Survey (MANS) conducted by Malaysian Ministry of Health in 2003. Other than that, the calculated level of PTWI in the samples of marine fishes collected for these elements were lies in the normal ranges and they were assumed as safe to be consumed.

Table 5: The Provisional Tolerable Weekly Intake (PTWI) (wet wt.) calculated in selected marine fish species for both monsoon and non-monsoon seasons.

Species	Season	Cu	Zn	As	Cd	Pb
N. furcosus	Non-monsoon	0.105	0.105	0.29	0.001	0.006
	Monsoon	1.18	9.53	23.7	0.021	0.193
K. pelamis	Non-monsoon	0.19	1.18	0.33	0.003	0.008
	Monsoon	2.31	15.36	6.32	0.840	0.227
D. macrosoma	Non-monsoon	0.12	0.437	0.27	0.001	0.008
	Monsoon	2.52	17.77	6.15	0.057	0.181
A. mate	Non-monsoon	0.14	0.59	0.52	0.003	0.005
	Monsoon	2.80	12.26	5.586	0.021	0.517
PTWI (mg/kg bw/ week)		0.35- 3.5[a]	0.21-7[a]	40.0[b]	0.007[a]	0.025[a]
Average PTWI (mg/kg bw/week) for 63 kg adult		22.1-221	13.2-441	2520	0.441	1.58
Remarks		Normal for consumption			Exceed in *K. pelamis* (monsoon)	Normal for consumption

[a]: FAO/WHO, 1984; [b]1988

Conclusion

As the conclusion, the findings of this study showed that these four fish species were tend to accumulate higher concentration of metals during monsoon season in compare to non-monsoon season in Terengganu waters. This phenomenon might be due to the changes of the temperature, the tide and also the strong wind that occur during monsoon season. Despite of seasonal variation, there were few controlling factors that contribute to the amount of heavy metal accumulated within the fish such as the size of fish body, the feeding habit as well as the living habitat of the fish. However, this study was emphasized on the level of As that considered as very high in compare to its permissible level which is should be focused and treated. More frequent sampling activities should be done in order to monitor the level of heavy metal especially As in Terengganu waters.

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