ORIGINAL ARTICLE

Assessment of heavy metals concentration in Gelama fish and water at Kuala Selangor and its potential health risk to human.

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Abstract:

Consumption of bioaccumulation heavy metals in fish had been found to cause severe health effect. This study was conducted in order to evaluate the concentration of selected heavy metals (Copper, Lead, Zinc, Cadmium) in the Gelama fish (*Tigertooth Croaker*) and water at Kuala Selangor estuary. This study also aimed to estimate the potential health risks through ingestion of this fish. The result showed the order of level concentration of metals studied found in Gelama fish was Zn > Cu > Cd > Pb. The study indicated that the concentration of these metals is below the permissible limit stated by Malaysian Food Regulation 1985. However, the concentration of Cd and Pb were slightly higher than Food and Agriculture Organization (FAO, 1984) but within the Malaysian government limits. Zn concentration was found to be significantly higher in the Gelama fish ($p<0.05$) in all sampling stations. For water samples, the result showed the concentration of metals studied was in the order of Zn>Cu>Pb>Cd. Target hazard quotient was calculated to determine the potential health risks associated with the heavy metals. The result showed that the THQ value was <1, which revealed that no adverse health effects expected from the consumption of fish at Kuala Selangor estuary. Hence, it can be concluded that Gelama fish caught at Kuala Selangor are safe to be consumed. However, a public awareness and continuous monitoring on heavy metals content in aquatic life should be done in order to reduce the potential health effect from the consumption of contaminated aquatic organisms.

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

1. INTRODUCTION

Pollution of heavy metals in water, especially at estuarine area has contribute to a major problem as it receives a large amount of pollutants input from both point and non-point sources and also from tourism activities and industries that are located along the estuarine [1]. Previous research conducted showed that accumulation of heavy metals in fish is an important issue to be studied as fishing activity one of the most important food production sectors in supplying protein to human. [2]

According to the Food and Agriculture Organization (FAO), during the past decade, demand for fish has drastically increase due to the increasing health awareness among the consumers [2]. This is because many fish species are consumed as a protein by a large section of population, especially those who live near the rivers [3]. Heavy metals found in fish tissue may be essential for their biological system and human but some of them may cause severe damage to human health above certain allowable limit [4].

Kuala Selangor estuary was selected as a study location by considering its nearby area such as Tanjung Karang, Sekinchan and Kapar, that tend to discharge their wastes along Selangor River which finally end up at Kuala Selangor estuary. Wide range of paddy and agricultural plantation at Tanjung Karang and Sekinchan contributed higher amount of fertilizers and pesticides residue into water body. Meanwhile, leachate from Kapar landfill may be leached and enter the Selangor river which then deposited at Kuala Selangor estuary. The numerous aquaculture farms and agriculture field located along the Selangor River is the reason why Kuala Selangor was selected as study location. Heavy metals from these activities may be introduced into adjacent area including Kuala Selangor estuary and accumulated in the sediments, water and aquatic organisms. This study was conducted in order to evaluate the accumulation of selected heavy metals (Cu, Zn, Pb, Cd) concentration in water and *Tigertooh Croaker* fish caught at Kuala Selangor estuary and their potential human health risk assessment. The concentration of these heavy metals in water also would give a clear picture of the environmental condition of Kuala Selangor estuary. The fish was selected

based on the frequently important of measuring the concentration of heavy metals in fish is because of its higher consumption by human [4].

2. MATERIALS AND METHODS

This study was conducted at Kuala Selangor estuary, which located at the mouth of the Selangor River in Kuala Selangor District. Seawater and fish samples (*Tigertooth croaker*) were collected at three different point along the Kuala Selangor estuary which were plotted as P1 (Upstream), P2 (Middle) and P3 (Downstream).

Nine samples of *Tigertooh croaker* fish of about the same size (15-25cm) were purchased from local fisherman at Kuala Selangor. Three fish samples were collected at each plotted point which indicated as upstream, middle and downstream estuary. All collected samples were labeled and stored in a clean polyethylene plastic bag and immediately preserved in an ice box to reduce the activities and metabolism of the organism in the water [1]. For water samples, three sample of sea water were collected at each plotted point. *In-situ* measurement on its physical characteristics such as turbidity, dissolved oxygen, pH and temperature was conducted during the collection time.

Wet digestion method was used for sample preparation which is guided by Analytical Methods for Atomic Absorption Spectroscopy (AAS) with some modification [6]. The frozen fish samples were thawed and the length were recorded. One grams of boneless muscle tissue was removed and cut into small pieces. The samples were dried in oven at 105^0C until constant weight was reached. Next, the samples were homogenized and digested as described by [11]. 1g of sample with 10ml of HNO3 and 2ml of H2O2 was heated on the hot plate for about one hour. When the digestion is complete, the residue was dissolved and diluted with 0.2% of nitric acid to 20ml. The digested samples be stored in pre-cleaned polyethlene bottle for further analysis.

3. RESULTS AND DISCUSSION

Figure 1 shows the level concentration of heavy metals in seawater samples collected at three different point along Kuala Selangor estuary: P1 (Upstream), P2 (Middle stream) and P3 (Downstream). The order of heavy metals concentration in seawater samples are Zn > Cu > Pb > Cd. Downstream estuary accumulated the highest mean concentration of Zn (0.242 mg/L) and is significantly different ($p<0.05$) between sampling stations. Higher Zn level in the water samples might be due to the discharge of agricultural wastes along the riverbank, oil spills from local fisherman's boat and other activities that end up at downstream estuary. Meanwhile, the mean concentration of Cd was lowest among all metals assessed which was 0.048 mg/L (Upstream), 0.050 mg/L (Middle) and 0.096 mg/L (Downstream). Normally, heavy metals in seawater are compared with Malaysian Marine Water Quality Standard (MMWQS) Class E. Based on result obtained, all the assessed metals were complied with the limit stated by MMWQS.

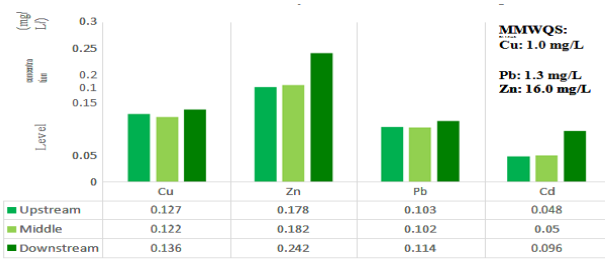

Figure 1: Heavy metals concentration in Seawater

Figure 2 below shows Zn concentration in fish samples collected at Kuala Selangor estuary were in the ranged of 7.20 mg/kg to 42.05 mg/kg. Zn concentration in fish samples is significantly different ($p<0.05$) among three sampling stations. The highest concentration of all metals studied was observed at downstream, while the lowest concentration was detected at middle stream. However, they were still below the allowable limit stated by Malaysian Food Regulations (1985) [5] and FAO (1983) which are 100 mg/kg and 150 mg/kg, respectively.

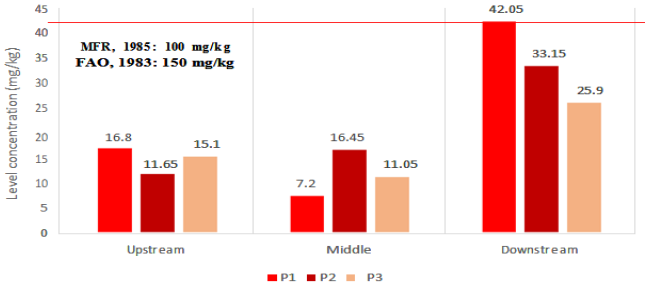

Figure 2: Zn concentration in *Tigertooth croaker* at Kuala Selangor estuary

Figure 3 shows the concentration of Cu in *Tigertooth croaker* fish was highest at middle stream which was 7.20 mg/kg. For downstream and middle stream, the average concentration of Cu was 6.87 mg/kg and 6.97 mg/kg, respectively. The maximum allowable Cu concentration for fish stated by Malaysian Food Regulations (1985) is 30 mg/kg while FAO is 10 mg/kg. This means the concentration of Cu in *Tigertooth croaker* fish at Kuala Selangor estuary were below the permissible limit.

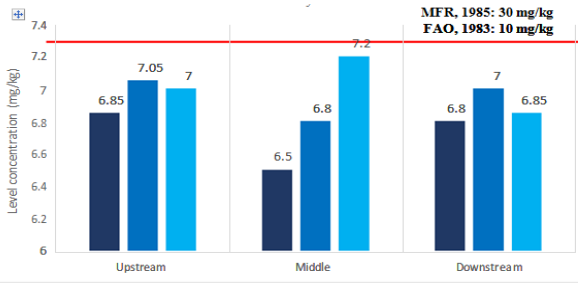

Figure 3: Cu concentration in *Tigertooth Croaker* at Kuala Selangor estuary

Figure 4 below shows that the concentration of Pb in *Tigertooth Croaker* fish were between 0.60 mg/kg to 1.15 mg/kg and is not significantly different among sampling stations ($p>0.05$). Malaysian Food Regulations (1985) and

FAO have stated that the maximum Pb level for fish is 2 mg/kg and 1.5 mg/kg, respectively. The graph shows that the Pb concentration in *Tigertooth croaker* collected at Kuala Selangor was below the Malaysian Food Regulations 1985 but slightly higher than FAO, 1983 allowable limits.

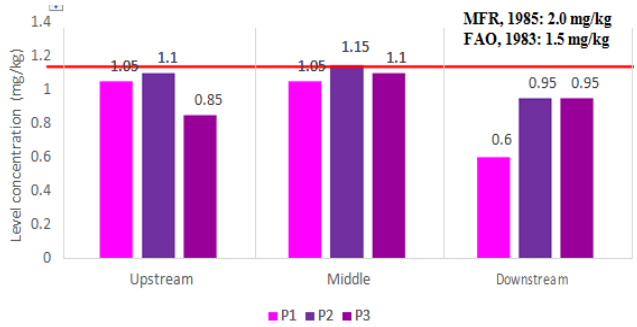

Figure 4: Pb concentration in *Tigertooth Croaker* fish at Kuala Selangor estuary

Figure 5 below shows the concentration of Cd in *Tigertooth Croaker* fish is highest at downstream with 0.90 mg/kg while the lowest concentration was reported at upstream with 0.55 mg/kg. Cd concentration in the *Tigertooth Croaker* fish was not significantly different ($p>0.05$) between different sampling stations. The maximum Cd level for fish set up by the Malaysian Food Regulations (1985) is 1.0 mg/kg while FAO limit is 0.2 mg/kg. However, the Cd concentration in fish sample exceeded the FAO but within Malaysian Food Regulations limit.

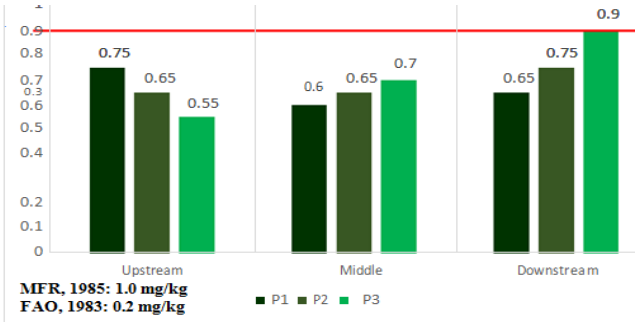

Figure 5: Cd concentration in *Tigertooth Croaker* at Kuala Selangor estuary

The results of *in-situ* water quality parameters measured at three sampling point along Kuala Selangor estuary were tabulated in Table 1. The pH values were 8.1 to 8.8 where the pH values of water were alkaline. Next, the results also show that the turbidity of the seawater was between 22 NTU to 56 NTU. The turbidity was highest at downstream followed with upstream and middle stream which were 56 NTU, 36 NTU and 22 NTU, respectively. Dissolve Oxygen (DO) was between 5.3-8.3. Usually, water temperature and salinity affect the reading of DO level. As the water's temperature and salinity increase, the solubility of oxygen, or its ability to dissolve in water decreases.

Normally, factors such as higher DO and total suspended solid (TSS) presented in the water can influenced the turbidity value. During the monitoring, the water at downstream area appeared to be cloudiest due to higher concentration of DO and TSS. This may be due to decaying plants and animals suspended in the water column. The results also showed temperature between 30.5-31.4ºC. The temperature of waters was varied at the three sampling points depending on sampling time collection and also factors such as surrounding vegetation. Overall, all locations of sampling station have high salinity in estuaries, salinity levels are generally high near the river mouth.

Table 1: Water quality parameter at Kuala Selangor estuary

Sampling Location	Temp (ºC)	Turbidity (NTU)	Dissolved Oxygen (%)	Total Suspended Solid (TSS)	Salinity (mg/L)	pH
P1	30.5	36	6.3	31.7	31.3	8.8
P2	31.4	22	5.9	31.1	31.9	8.4
P3	30.9	56	8.3	32.5	32.9	8.1

Table 2 shows the heavy metals content in water samples. The mean concentration of all heavy metals studied in seawater samples were lower than the maximum allowable values stated by Malaysian Marine Water Quality Standard (MMWQS) Class E.

Wastes discharged along the riverbank from mainland activities increase the accumulation of metals in water body. From the study, mean value for Cu and Zn were found to be higher at all sampling stations and did not significantly differ between upstream and middle stream. Higher Zn concentrations in water samples collected possibly might be because of Zn are not digested or excreted efficiently by fish and tend to dissolved and accumulate in the sediment, soil and surface water [6].

The downstream point has the highest concentration of all metals assessed. The fact that downstream area is point of waste deposition from both point and non-point upstream sources conducted along Selangor River and it is one of factors that increase the accumulation of heavy metal at the area. Kuala Selangor is located near the largest coal power station, Kapar Energy Venture, where high amount of heavy metals in cooling water may be discharged and dissolved in the nearby water column. Besides, residues from agricultural activities at nearby district also released higher amount of metals into water body. Upstream area showed the lowest heavy metals in water samples. Strong water current at upstream area may be a factor that decrease the chemical reaction in the water, thus cause lower accumulation of such metals in the water body.

Table 2: Heavy metals contents in water samples

Sampling location	Heavy metals concentration (mg/L)			
	Cu	Zn	Pb	Cd
Upstream	0.127±0.0173	0.178±0.0027	0.103±0.0990	0.048±0.0022
Middle stream	0.122±0.0165	0.182±0.0040	0.102±0.0193	0.050±0.0011
Downstream	0.136±0.0019	0.242±0.0018	0.114±0.0303	0.096±0.0364
p-value	0.05	0.01	0.03	0.03
Malaysian Marine Water Quality Standard	1.0	16.0	1.3	1.0

Source: Department Of Environment Malaysia

Table 3 shows the contents of heavy metals in fish samples collected along Kuala Selangor estuary. The result showed that different heavy metals accumulated differently at different sampling stations.

Table 3: Heavy metals concentration in fish samples

Sampling point	Heavy metals concentration (mg/kg)			
	Cu	Pb	Zn	Cd
Upstream	0.137±0.0019	0.021±0.0025	0.336±0.01102	0.015±0.0025
	0.130±0.0021	0.022±0.0031	0.233±0.00667	0.013±0.0020
	0.136±0.0015	0.017±0.003	0.302±0.01457	0.011±0.0017
Middle	0.141±0.0037	0.021±0.0040	0.144±0.00306	0.012±0.0023
	0.136±0.0021	0.023±0.0042	0.329±0.00665	0.013±0.0021
	0.140±0.0015	0.022±0.0044	0.221±0.01082	0.014±0.0015
Downstream	0.140±0.0035	0.012±0.0025	0.841±0.00702	0.013±0.0042
	0.144±0.0050	0.019±0.0040	0.663±0.00500	0.015±0.0040
	0.137±0.0032	0.019±0.0030	0.518±0.00451	0.018±0.0030
p-value	0.002	0.14	0.005	0.012
MFR 1985*	30 mg/kg	2 mg/kg	100 mg/kg	1 mg/kg
FAO 1983**	10 mg/kg	1.5 mg/kg	150 mg/kg	0.2 mg/kg

*Malaysian Food Regulation (MFR, 1985)
**Food and Agriculture Organization (FAO, 1983)

Kuala Selangor is rich in mineral resources as it was once known as centre of tin mining industry that help defined the early history of Malaysia. Furthermore, Kuala Selangor is located adjacent to the Sekinchan, Tanjung Karang and Jeram district where all these areas are popular in releasing their pollutants that comes from their main activities such as fisheries, paddy plantation, industry as well as wastes disposal site. Based on the result obtained, Zn concentration was found to be significantly higher in the *Tigertooth Croaker* fish ($p<0.05$) between the different sampling station.

Normally, concentration of heavy metal in fish were compared to the permissible limits recommended by Food and Agriculture Organization (FAO,1983) and Malaysian Food Regulation (1985) [5]. The total mean heavy metals concentration of Tigertooth Croaker fish in this study was an order of Zn > Cu > Cd > Pb. The fact that different heavy metals tend to accumulate differently in fish may be one of different bioaccumulation factor. Although Zn was showed the highest in *Tigertooth Croaker* compared to other metals assessed, but the level is still below the permissible limit stated by Malaysian government. The highest level of Zn was found at downstream, followed by middle and upstream with mean concentration of 0.841 mg/kg, 0.663 mg/kg and 0.512 mg/kg, respectively.

Generally, Zn enter the water through several pathways such as burning of waste materials, steel production, coal-fired power station, and also leaching of fertilizer used in plantation. The higher Zn content in *Tigertooth Croaker* at downstream (0.841 mg/kg) must be due to metals released from nearby largest coal-fired power plants in Malaysia, Kapar Energy Venture is located off the coastal road between Port Klang and Kuala Selangor. Kapar Energy Venture is one of the largest power plant in Malaysia, where higher amount of heavy metals in cooling water might be discharged into the nearby water column, thus negatively cause accumulation of metals in the water, sediments and aquatic organisms. Besides, fertilizers containing Zn from nearby paddy plantation, Tanjung Karang and Sekinchan that are carried by surface runoff is normally end up at downstream point, thus cause higher accumulation of Zn at the area.

According to the previous study, all fish species were claimed to have higher concentration of Zn compared with other metals which ranging from 34.33 mg/kg to 49.39 mg/kg [4]. So, it is not surprising when Zn contribute the highest concentration in *Tigertooth Croaker* for this study. Generally, Zn is important element for embryo development and reproductive organs but a constant accumulation of Zn in fish over a period of time tend to cause toxicity that eventually harm the consumers through their consumption. [2]

The concentration of Cu in *Tigertooth Croaker* fish was 0.130 to 0.144 mg/kg and significantly differ among the three sampling stations with ($p<0.05$). Copper is highly detected at downstream with mean concentration of 0.144 mg/kg while the lowest concentration of Cu was detected at upstream (0.130 mg/kg). Usually, Cu enters the water body and settled in the water, sediments or soils through the releasing of copper compounds from agricultural activities. Kuala Selangor is located adjacent to Tanjung Karang, Sekinchan and Sungai Besar district where these areas contribute a wide range of paddy field and agricultural activities, where excessive fertilizers and pesticides residues used in controlling the quality of the crops might be discharged and carried along the riverbanks and finally end up at the downstream estuary.

In addition, Cu found at Kuala Selangor estuary may also come from the existing activities such as cleaning and maintenance of boats, loading and offloading of fishes and fuelling the craft. These kind of activities cause heavy metals such as Cu to settle and bind in the sediment and soil, which then will be ingested by the aquatic organism such as fish. A constant ingestion of such metals by fish can cause high rate of copper compound in the fish tissue. Cu is essential for mammalian nutrition but consumption of fish polluted with Cu may cause several health effects including liver toxicity [7].

Pb concentration in *Tigertooth Croaker* fish shows the average mean concentration of Pb is in order of 0.012 < 0.019 < 0.023 mg/kg. The highest concentration of Pb was detected at middle stream which (0.023 mg/kg) while the lowest is at downstream (0.012 mg/kg). Pollutants originally from the mainland activities such as agriculture field and nearby factory generally flow through the Selangor River which then would deposited at Kuala Selangor estuary. Agricultural pollution such as contamination of Pb from fertilizers is a common issue because Kuala Selangor are surrounded with agricultural field. The pollutants may be carried into the coastal area through surface runoff, thus accumulate in the sediments, soil and surface water. Ingestion of polluted sediments and water by *Tigertooth Croaker* fish tend to accumulate them in their body.

In general, Pb is a cumulative toxin and can cause serious effect to human health such as damaging blood circulation, central nervous system, liver and kidneys [2]. In this study,

Pb level in *Tigertooth Croaker* fish at downstream point is slightly higher than other station. In fish, Pb causes decrease in survival, growth, development, behaviour and metabolism, and increase in the formation of mucus [3].

Cadmium concentration was found to be significantly lowest in the *Tigertooth Croaker* fish ($p<0.05$) between the different sampling stations. The highest Cd concentration is reported at downstream and lowest at middle estuary with 0.018 mg/kg and 0.011 mg/kg, respectively. The result showed Cd concentration at downstream is slightly higher than FAO permissible limit but within Malaysian Food Regulation limit. In this study, common components of commercial fertilizer used for wide range of paddy field at nearby district such as Tanjung Karang and Sekinchan increase Cd in the water as the metals are carried along the Selangor river and end up at downstream estuary [8].

Downstream estuary usually received more agricultural wastes that flow along the riverbank, and also oil spillage from boat used for regular transportation along Kuala Selangor river. Besides, released of wastes and sewage from mainland to the river may be an additional factor that increase the accumulation of Cd in downstream estuary.

Table 4 shows the result of estimated daily intake (EDI) of Tigertooth Croaker fish collected at Kuala Selangor estuary. The EDI of four heavy metals studied is determined based on assumption of 62.65-kg body weight per person and exposure duration of 70 year [9]. The average EDI value for consumption of Tigertooth Croaker fish collected at Kuala Selangor is in order of Zn > Cu > Pb > Cd. At downstream estuary, Zn shows the highest value with 0.00656 mg/kg/person, followed by Cu (0.00136 mg/day/person), Pb (0.00066 mg/day/person), and Cd (0.00058 mg/day/person). In middle estuary, the decreasing order of EDI value is Zn > Cu > Pb > Cd with 0.0022, 0.00135, 0.00086 and 0.00051 mg/kg/person, respectively. For upstream estuary, Zn contribute the highest EDI value while Cd indicates the lowest with value of 0.00283 mg/kg/person and 0.00051 mg/kg/person, respectively. Basically, the purpose of conducting THQ was to calculate the risk of non-carcinogenic effects. According to Mohamad et al [10] potential health risk is presented if THQ obtained is more than 1.

Table 4: EDI of Tigertooth Croaker fish at Kuala Selangor Estuary

Heavy metals	Estimated Daily Intake (mg/day/person)		
	P1 (Upstream)	P2 (Middle)	P3 (Downstream)
Cd	0.00051	0.00051	0.00058
Cu	0.00130	0.00135	0.00136
Zn	0.00283	0.00225	0.00656
Pb	0.00078	0.00086	0.00066

The values of THQ calculated for all studied metals were below 1 for all fish samples and is significantly showed differ ($p<0.05$) among three different sampling station (Table 5). The result showed Cd has higher THQ value at all sampling stations which ranging from 0.12651 to 0.14597 mg/kg/person. The THQ value of Cd is highest at downstream followed by upstream and middle stream. Besides, the result also shows that Zn, Cu and Pb were found higher at downstream estuary compare to middle and upstream point. So, it can be concluded that downstream estuary polluted where Tigertooth Croaker fish tend to highly accumulate heavy metals at the area. However, the THQ values among location of estuaries were below 1 which indicated no adverse health effect expected from the result of exposure.

Table 5: Target Hazard Quotient of non-carcinogenic risk for the consumption of Tigertooth Croaker fish at Kuala Selangor Estuary

Heavy metals	RfD (mg/kg/day)	THQ		
		P1 (Upstream)	P2 (Middle)	P3 (Downstream)
Cd	0.001	0.12651	0.12651	0.14597
Cu	0.04	0.03260	0.03381	0.03406
Zn	0.3	0.01732	0.01852	0.02287
Pb	0.004	0.04866	0.05353	0.04136

Based on Table 6, HI values that have been calculated for every sampling stations showed that downstream estuary indicated the highest value compared to upstream and middle stream which is 0.24126. Overall, HI values obtained is below than 1 which means there is no health effects expected from the result of exposure.

Table 6: Hazard Index for consumption of Tigertooth Croaker fish at Kuala Selangor Estuary

Sampling station	Hazard Index, HI
Upstream (P1)	0.22509
Middle (P2)	0.23237
Downstream (P3)	0.24126

Carcinogenic risk assessment refers to the probability that the individual will develop cancer over a lifetime due to exposure of particular contaminant or to a mixture of contaminants [12]. Based on Table 7 below, the result shows the highest carcinogenic risk of Pb recorded at middle stream which is 1.8198×10^{-6}, followed by upstream and downstream area with value of 1.6544×10^{-6} and 1.4062×10^{-6} respectively. The result obtained is within 10^{-6} which means there is potential effect expected from the result of exposure. Lead exposure can promote serious human health by damaging blood circulation, central nervous system, liver and kidneys [2] According to World Health Organization (WHO), children less than 5 years old and pregnant women are most susceptible to the adverse effects of lead where children absorb 4-5 times as much Pb as adults.

Table 7: Carcinogenic Risk level for Pb Concentration in *Tigertooth Croaker* fish

Metal	CSFo $(mg/kg/day)^{-1}$	P1 (Upstream)	P2 (Middle)	P3 (Downstream)
Pb	8.5×10^{-3}	1.6544×10^{-6}	1.8198×10^{-6}	1.4062×10^{-6}

4. CONCLUSION

Zn concentration was found to be significantly higher in the Tigertooth Croaker fish ($p<0.05$) between different sampling stations. Overall, all assessed metals accumulates highly at downstream, followed by upstream and middle stream. Downstream estuary or known as river mouth is the collection point where it receives wastes from mainland activities along the Selangor River. This situation causes high accumulation of such metals in the sediments, soils and surface water which finally negatively affect the fish. This finding is supported by the fact that water samples collected at downstream estuary also measured the highest metals concentration compared to other sampling station.

However, the present study indicated that Tigertooth Croaker fish collected at Kuala Selangor estuary are safe for human consumption as it selected heavy metals were below the allowable limit specified by Malaysian Food Regulations 1985. Meanwhile, for health risk assessment, THQ values calculated for Zn, Cu, Pb and Cd were below 1, thus indicated there is no adverse health effects resulting from the consumption of Tigertooth Croaker fish from Kuala Selangor. Public awareness and continuous monitoring on heavy metals in aquatic environment should be conducted as it provides useful knowledge and information for the consumer.

ACKNOWLEDGEMENTS

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