

ACCUMULATION AND HEALTH RISK OF HEAVY METALS IN LOCAL AND IMPORTED FISH (*RASTRELLIGER KANAGURTA*) SOLD IN SHAH ALAM, SELANGOR

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ABSTRACT

Global aquatic ecosystems are becoming increasingly polluted with heavy metals posing health concerns associated with fish consumption. This study aims to determine the concentrations of cadmium (Cd) and lead (Pb) in local and imported fish (*Rastrelliger kanagurta*), and its potential human health risk. Total local and imported fish (60 samples) were randomly selected and purchased from ten different supermarkets in Shah Alam, Selangor. Cd and Pb concentrations were assessed by using dry ashing-acid digestion technique and analysed with atomic absorption spectrometry (AAS). The analytical results shows that the mean concentration of Cd in local and imported fish (*Rastrelliger kanagurta*) were 0.487 ± 0.25 mg/kg and 0.267 ± 0.11 mg/kg, respectively. The mean concentration of Pb in local fish was higher at 2.774 ± 1.46 mg/kg compared to the imported fish (1.482 ± 0.81 mg/kg). Further analysis indicated that there are significant differences ($p < 0.05$) for both Cd and Pb levels in local and imported fish (*Rastrelliger kanagurta*) sold within Selangor supermarkets. The concentration of Pb in local fish sold from supermarkets failed to comply with all the standards guidelines. The hazard quotient (HQ) used for health risk assessment of Pb and Cd was found to be < 1 ($HQ < 1$) for imported fish and potential risk for local fish ($HQ > 1$). The study reveals that the local fish are not totally safe for consumption and the pollution concentrations are on the borderline but can be treated.

Keywords: cadmium; lead; fish; *Rastrelliger kanagurta*; hazard quotient

1. Introduction

The global aquatic environment has been increasingly polluted with heavy metals from both natural and anthropogenic sources. This raised concerns about the level of exposure that can affect human health and the environment (Yu et al., 2020). The presence of heavy metals in aquatic ecosystems has been a source of concern for many decades. Agriculture, rapid urbanisation, and the discharge of untreated effluent by chemical industries and other human activities have been contributing to the severe deleterious effects on fish (Effah et al., 2021). The consequence of all these human activities is the accumulation of heavy metals in our natural rivers, lakes, and marine ecosystems, leading to the increase of heavy metal concentrations in fish (Yi et al., 2011). Therefore, fish is a commodity with potential health concerns as it can be contaminated with a wide range of heavy metals, such as cadmium (Cd), lead (Pb), mercury

(Hg), arsenic (As), and chromium (Cr), that may cause toxicity even in small amounts (Goyer & Clarksom, 2011). In current times, with climate change and unfettered industrialisation, the contamination of heavy metals in fish is a major environmental health concern, especially since the majority of Asia's freshwater and marine environments are threatened by contamination in various ways (Octavianti & Jaswir, 2017).

Generally, fish can absorb heavy metals directly from water and sediments and ingest heavy metals through the food chain (Agusa et al., 2007; Rahman et al., 2012). Commonly, heavy metals such as Pb, Hg, Cd, and Cr, have the ability to cause various severe health effects to the population of fish consumers due to its ability to accumulate in the body to levels above the acceptable limit. This process begins the moment heavy metals are introduced into water bodies. Once it enters the marine ecosystem, it would be readily available for marine life. The implications of consumed heavy metals would be stored in various organs and tissue muscles of the fish. Due to the metallic elements of the heavy metals, they would not be degraded, in fact, bioaccumulate (Rahman et al., 2012). This process would continue when the fish is consumed even at lower concentrations by consumers. Continuous heavy metal exposure can cause acute disease to consumers. For example, the high concentration of Cd may cause kidney failure, hypertension, neurological and digestive problems, skeletal weakness, and cancers (Cabral et al., 2015). Meanwhile, with respect to Pb, continuous Pb exposure can affect the human central nervous system, immunity and the cardiovascular system, cause digestive disorders, and many more (Núñez et al., 2018). Unfortunately, poisoning by heavy metals, such as Cd and Pb, is common. In Malaysia, numerous research has been done to determine the health risks associated with Cd and Pb exposure in foods (Ishak et al., 2020; Khandaker et al., 2015; Octavianti & Jaswir, 2017; Rajan & Ishak, 2017).

Rapid advancements in the socio-economic situation of many countries in Asia, including Malaysia, have resulted in significant changes in the lifestyles of communities, such as food habits, food purchasing, and consumption patterns. In a region where fish and fish products are part of the daily diet, expansion in the world's human population and economic development will increase future demand for fish. In most Asian regions, especially in Southeast Asian countries like Thailand, Indonesia, and Malaysia, fish is taken as the main dish of their diet (Yu et al., 2020). Marine fresh and fish products are considered a fundamental element of protein sources consisting of about 60 to 70% of protein. Apart from that, fish is notably a more valuable high quality protein source than protein derived from meat, milk, or eggs. Fish is also a good source of omega-3 fatty acids, calcium (Ca), phosphorus (P), iron (Fe), trace elements like copper (Cu), and a fair proportion of B-vitamins that will further contribute to healthy nutritional options for a balanced diet (Hajeb et al., 2009).

Malaysian facts showed that *Rastrelliger kanagurta* or Indian Mackerel is a common fish consumed by the Malaysian population (Osman et al., 2000). This situation arises due to its cheaper price when compared to other fish species in supermarkets, apart from being more easily available; thus, making it an easier and more convenient option (Alina et al., 2012; Nor Hasyimah et al., 2011). However, there is limited information on the heavy metal concentrations in local and imported selected fish species (*Rastrelliger kanagurta*). A study such as this is crucial to not only determine the level of heavy metals in fish, but also as an indicator of environmental pollution, both of which are important factors for the provision of healthy food for local communities. Therefore, this study aims to determine the concentrations of cadmium (Cd) and lead (Pb) in local and imported fish (*Rastrelliger kanagurta*) sold in Shah Alam, Selangor, in addition to assessing the human health risk assessment due to consumption of *Rastrelliger kanagurta*.

2. Materials and Methods

2.1. Questionnaire

A questionnaire was used to determine the socio-demographic data, fish consumption information, and health risks of exposure to heavy metals. The questionnaire was modified from the NHANES Food Frequency Questionnaire (FFQ) (US National Cancer Institute, 2008) in order to perform a heavy metal health risk assessment. The questionnaire was divided into three parts, and comprises socio-demographic profiles and the participants' fish consumption pattern. This includes information, such as gender, age, education level, fish as a food diet, and frequency of fish intake. Sixty (60) sets of questionnaires were randomly distributed to consumers who purchased fish, particularly *Rastrelliger kanagurta* species as their food diets, sold in supermarkets at Shah Alam, Selangor.

2.2. Samples and sampling procedure

Sixty (n = 60) fish samples (*Rastrelliger kanagurta* or Indian Mackerel) were randomly selected and purchased from ten different supermarkets located in Shah Alam area. The fish samples (*Rastrelliger kanagurta*) were from both local (n = 30) and imported (n = 30). All fish samples were packed in clean zipped polythene bags and transported to the laboratory in an ice box at a temperature of 0 °C to 4 °C. The fish samples were then transferred and stored in a chiller at -2 °C until further chemical analyses.

2.3. Determination of heavy metal content in fish

2.3.1. Sample preparation and analysis

The method for sample preparation was performed based on a method described by Nor Hasyimah et al. (2011) with slight modifications. Fish samples were rinsed thoroughly with deionised water to remove dirt, mucus, and fouling substances. Only stainless steel cutting equipment was used. All materials in this procedure were acid soaked (10% nitric acid) for at least 24 hours and rinsed thoroughly with deionised water prior to usage. After rinsing and cutting, the fish were skinned and filleted, and the samples were blended to homogenise.

Twenty-five grams of samples were placed in high-form porcelain crucibles. The furnace temperatures were slowly increased from room temperature to 450 °C. The samples were placed into a muffle furnace overnight until a white or grey ash residue was obtained. The residue was dissolved in 2 ml of Nitric Acid (HNO₃) and 20 ml of HCl. Then, the mixture was heated slowly on a hot plate to dissolve the residue. The samples were then filtered (0.45 µm pore size filter paper) into a 50 ml volumetric flask and distilled water was added. Concentrations of Cd and Pb in fish were analysed by using atomic absorption spectrometry (AAS) (Perkin Elmer Analyst 700). Detected metals were expressed in mg/kg wet weight. Each element was measured and validated in accordance with the limit of detection and standard deviation.

2.4. Human Health Risk Assessment

Humans may be exposed to heavy metals via three primary routes: direct ingestion, inhalation, and skin absorption. There are several methods for predicting the potential human health risk associated with the ingestion of heavy metals in fish. The level of human health risk of Cd and Pb exposure used hazard quotient (HQ) estimation to predict heavy metal for the consumer population of fish consumption (Praveena & Lin, 2015; Taweel et al., 2013). The equation for health quotient (HQ) is:

$$HQ = \frac{C \times IR \times ER \times ED}{RfD \times RW \times TA} \quad (1)$$

Where, C is the heavy metal concentration in fish (mg kg^{-1}), IR is the fish ingestion rate (160 g/person/day), EF is the exposure frequency (365 days/year for consumers who eat fish (*Rastrelliger kanagurta*) at least seven times a week, 52 days/year for consumers who eat fish once a week), ED is the exposure duration (74 years, according to lifetime based on World Bank (Praveena & Lin, 2015)), BW is the average body weight of each participants (49 kg obtained from questionnaire data), and TA is the average exposure time for non-carcinogens (365 days/year ED). RfD is the oral reference doses of Cd 0.001 mg/kg/day and Pb 0.005 mg/kg/day (US EPA 2007). If the HQ > 1.00, it means that there is a potential health risk to the exposed consumer.

2.5. Statistical Analysis

The data were statistically analysed using the Statistical Package for the Social Sciences (SPSS Inc., Chicago, IL, USA) version 21. Due to the data normality, the *t*-test parametric test was performed to assess the differences in the two independent samples. The independent *t*-test was used to compare the mean concentration of heavy metals (Cd and Pb) between local and imported fish (*Rastrelliger kanagurta*).

3. Results and Discussion

3.1. Demographic profile

Table 1 presents data on gender, age, education level, fish as a diet and frequency of fish intake. Sixty consumers participated in this study, with 55% female and 45% were male. The ages of the consumers ranged between > 20 and > 50 years old. Most consumers fall in the range of 21 to 30 years old (33%). Sixty per cent of the consumers were university graduates, and the lowest participant education background was primary school, at 3%. There are around 20 public and private universities in Selangor, which means that a large number of students are enrolled (Study Malaysia, 2020). Hence, the majority of the participants' population were students between 21 to 30 years old. Based on the data on fish consumption results, all consumers prefer fish as the main food in their diet (100%). Alina et al. (2012) stated that fish has been the main supply of cheap and healthy protein to a large percentage of the world's population. In most Asian countries, especially in Southeast Asian countries like Thailand, Indonesia, and Malaysia, fish is taken as the main dish in their diet. Fish is particularly valuable for providing food with nutrition in the form of protein, and minerals such as calcium (Ca), phosphorus (P), and iron (Fe), and copper (Cu). Previous study by Nor Hasyimah (2011) shows about 60 to 70% of Malaysians consume fish as a food diet compared to meat consumption because the price is cheaper. For the frequency of fish intake, the majority of the participants consumed fish 3-7 times per week (39%) and 3% of the participants consumed fish either once per week and 2 or more times per day. Generally, there is limited research related to the information on heavy metals in local and imported fish in Malaysia. According to Osman et al. (2000), most

Malaysian consumers (73%) prefer to eat Indian Mackerel (*Rastrelliger kanagurta*) as part of their daily diet due to its low cost and suitable size compared to ten other fish species.

Table 1: Demographic characteristics of consumers from selected supermarket at Shah Alam, Selangor (n = 60)

Variable	Items	Frequency (n)	Percentage (%)
Gender	Male	27	45
	Female	33	55
Age	< 20	5	8
	21- 30	20	33
	31 – 40	15	25
	41 – 50	15	25
	> 50	5	8
Education level (EF)	Primary school	2	3
	Secondary school	14	23
	University	36	60
	Others	8	13
Fish taken as a food diet	Yes	60	100
	No	0	0
Frequency of fish intake	1 time/month	2	3
	2-3 times/month	7	12
	1 time/week	13	22
	2 times/week	11	18
	3-7 times/week	23	39
	1 times/week	2	3
	2 or more times/day	2	3

3.2. Concentration of Cadmium (Cd) and Lead (Pb) in Fish

The concentrations of Cd and Pb in local and imported fish (*Rastrelliger kanagurta*) were detected in both samples (Table 2). The mean concentrations of Cd and Pb in local and imported fish ranged between 0.267 mg/kg to 0.481 mg/kg and 1.482 mg/kg to 2.774 mg/kg, respectively. The mean concentration of Cd was detected lower in imported fish (0.267 mg/kg) compared to local fish (0.481 mg/kg). Previous study by Nor Hasyimah et al. (2011) stated that a higher mean concentration of Cd (1.62 ± 0.13 mg/kg to 1.82 ± 0.14 mg/kg) was obtained in commercial *Rastrelliger kanagurta* that was sold in Klang Valley supermarkets. However, another study conducted in the coastal area of Port Dickson observed that the range of Cd in local fish (*Rastrelliger kanagurta*) was lower at 0.09 mg/kg (Praveena & Lin, 2015). Meanwhile, the levels of Cd reported in this study, for both local and imported fish, were higher than the permissible limit set by the Joint FAO/WHO (2004) Expert Committee on Food Additives (JECFA, 2003). According to the FAO/WHO (2004), the recommended concentration of Cd in fish for human consumption should be around 0.05 mg/kg (JECFA, 2003). The present finding may be due to the textile, chemical pollution from industries, municipal sewage, and agricultural waste located near coastal or upstream rivers (Ishak et al., 2020).

The mean concentration of Pb was also lower in imported fish (1.482 ± 0.81 mg/kg) compared to local fish (2.774 ± 1.46 mg/kg). The mean concentration of Pb in this study was greater than those reported by Mziray & Kimirei (2016). Additionally, a higher mean concentration of Pb (12.44 ± 1.66 mg/kg) in the fish species *Rastrelliger kanagurta* along the coast of Port Dickson was reported by Praveena & Lin (2015). It was observed that the concentration of Pb in local fish exceeded the maximum permissible limit (about 2 mg/g) as stated in the Malaysian Food Regulations (1985) (Act, 1983). This indicates that the pollution

status in the Malaysian coastal environment may be moderate to high and needs to be monitored. In India, the concentration of Pb is on the borderline due to *Rastrelliger kanagurta* that was harvested along the coast of Visakhapatnam has been recognised as a high industrial area (Mangalagiri et al., 2020).

Table 2: The mean concentrations (mg/kg) of cadmium (Cd) and lead (Pb) in local and imported fish (*Rastrelliger kanagurta*) sold in supermarkets at Shah Alam, Selangor

Fish source	Heavy metals	Mean \pm SD
Local (n = 30)	Cadmium	0.481 \pm 0.25
	Lead	2.774 \pm 1.46
Imported (n = 30)	Cadmium	0.267 \pm 0.11
	Lead	1.482 \pm 0.81

In Malaysia, the concentration of heavy metals increases may be due to marine pollution and activities, such as cross-channel shipping, transit traffic, and fishing activities (Khandaker et al., 2015). In addition to marine accidents, marine pollution is also caused by ships. For example, any leakage of fuel oil or lubricating oil from machinery, installations, pipes, tanks, and other spillage on board ships together with washing residue or sea water accumulates in the ship through the propulsion system. Apart from that, some shippers have a habit of filling cargo tanks with ballast water when oil tankers sail without cargo. The washing of cargo tanks when there is a change of cargo or the cleaning of dirty tanks also generates oil waste, which is then discharged into the sea (Wisaksono & Bilal, 2006). This is clearly a problem in Malaysia, where higher Cr and Pb levels have been observed around the Malaysian coastal waters (Agusa et al., 2007). Apart from that, Hong-fei et al. (2010) and Asraf et al. (2012) stated that leachate from landfills, shipping and boating activities, fossil fuel combustion, waste incineration, and agricultural utilisation of pesticides and herbicides also contributed to the anthropogenic input of heavy metals in the environment. The Pb content can be increased with the addition of inorganic lead compounds from batteries found in landfills, whereas the Cd concentration can come from the nickel-cadmium battery used in mobile phones, which is frequently discarded with household waste and may also originate from fertilisers.

Table 3 shows the comparison of heavy metals between the local and imported fish. A two-sample *t*-test shows that there were significant differences ($p < 0.05$) in the mean concentrations of Cd and Pb in local and imported fish (*Rastrelliger kanagurta*) sold in Shah Alam supermarkets. This finding supports the previous study by Nor Hasyimah et al. (2011) and Praveena & Lin (2015), where the concentrations of Cd and Pb detected in their local fish samples purchased from supermarkets around Klang Valley were higher than the acceptable limit. Therefore, this finding concluded that the concentrations of Cd and Pb in local fish were higher than the imported fish.

Table 3: The comparison of mean concentration of heavy metals in local and imported fish (*Rastrelliger kanagurta*)

Variables	Local fish (Mean± SD)	Imported fish (Mean± SD)	Mean diff (95% CI)	t-test (df)	p-value
Cadmium (mg/kg)	0.481 ± 0.25	0.267 ± 0.11	-0.1983 (-0.3087, -0.08793)	-3.596 (-36.459)	0.01 ^a
Lead (mg/kg)	1.482 ± 0.81	2.774 ± 1.46	-1.292 (-1.9056, 0.6784)	-4.240 (45.277)	0.0001 ^a

^aMean values are significantly different (p < 0.05).

Several countries such as Indonesia, Thailand, China, and Taiwan are involved in supplying Indian Mackerel fish (*Rastrelliger kanagurta*) to Shah Alam supermarkets. In Malaysia, the sources of fish caught came from various aquaculture sites. One of the sources is from the Straits of Malacca. The Straits of Malacca was claimed as the leading fishing ground (nearly 70%) in Malaysia (Agusa et al., 2007). Not only that, the Straits of Malacca is one of the busiest shipping routes in the world, with more than 100,000 oil tankers, container vessels, and cargo vessels transiting there each year. Moreover, Bashir et al. (2011) reported high levels of heavy metals in the waters, principally on the West Coast of Peninsular Malaysia. Lead (Pb), copper (Cu), zinc (Zn), and cadmium (Cd) levels in several water samples were also found to be higher than the interim guidelines. The most common ship pollution is oil spill, which can poison Malaysian waterways. It is evident that the shipping sector is one of the primary suppliers of heavy metals in Malaysian coastal waters.

The concentrations of Cd and Pb in local and imported fish (*Rastrelliger kanagurta*) were evaluated by using the national and international standard guidelines: the Fourteenth Schedule (Regulation 38) of the Malaysian Food Regulation (1985), the Food and Agriculture Organization/World Health Organization (FAO/WHO) (2004), and the United States Food and Drug Administration (USFDA) 1990. Based on Table 4, the concentration of Cd in imported fish (0.267 mg/kg) and local fish (0.481 mg/kg) are still under the permissible limit of the Malaysian Food Regulation 1985, FAO/WHO (2004), and USFDA (1990) standard guidelines. However, the concentration of Pb in local fish (2.774 mg/kg) contravened the national and international standard guidelines. Nevertheless, the mean concentration of Pb in imported fish (1.482 mg/kg) was below the permissible limit, thus complying with the Malaysian Food Regulation (1985) and USFDA (1990) standards. However, it contravened the FAO/WHO (2004) standard (JECFA, 2003). Therefore, this research provides regional baseline data for Malaysian consumers who consume fish with regards to the concentrations of Cd and Pb between local and imported fish (*Rastrelliger kanagurta* or Indian Mackerel).

Table 4: Comparison of heavy metal permissible limits (mg/kg) in local and imported fish (*Rastrelliger kanagurta*) with legal compliance

Parameters	Local	Imported	Malaysian Food Regulation (1985)	FAO/WHO (2004)	USFDA (1990)
Cd	0.481	0.267	1.0	0.5	3.7
Pb	2.774 ^a	1.482 ^b	2.0	0.5	1.7

^a not comply with the Malaysian Food regulation (1985) and FAO/WHO (2004) and USFDA (1990)

^b not comply with FAO/WHO (2004)

3.3. Risk of Cd and Pb Exposure to Human

A health risk assessment was conducted to assess the exposure of the concentrations of Cd and Pb from the ingestion of local and imported fish purchased from local supermarkets at Shah Alam, Selangor. Most researchers believed that Pb could cause neurotoxicity, nephrotoxicity, and other health effects (Rahman et al., 2012; Goyer & Clarksom, 2011). Apart from that, studies reviewed by Rahman et al. (2012) on the health effects of the concentrations of Cd stated that Cd might directly affect human health organs and tissues, mainly the kidney, lung, heart, brain, and the central nervous system. It was also discovered that Cd might contribute to poor learning abilities and trigger hyperactivity in children (Castro-González & Méndez-Armenta, 2008).

In this study, Table 5 presents the consumer health risk for Cd and Pb exposure via fish consumption. The assessment of fish consumption was based on the frequency of fish intake once a week and seven times a week. The hazard quotient (HQ) applied to both Pb and Cd for local fish (*Rastrelliger kanagurta*) indicates to be more than 1 ($HQ > 1$) considering all parameters examined (Table 5). The HQ value has revealed that the exposure to both Pb and Cd from the Indian Mackerel fish (*Rastrelliger kanagurta*) may pose a health effect on a local population who consumed the fish. However, the health risk estimates for imported fish consumption were considered safe with HQ value less than 1 ($HQ < 1$). This indicates there is a low risk to human health from the consumption of imported fish. However, this needs to be consistently monitored as the HQ value is nearly 1. Moreover, Mangalagiri et al. (2020) reported that the accumulation of Pb in *Rastrelliger kanagurta* from India was higher than the limit set by the Joint FAO/WHO (2004) Expert Committee on Food Additives (JECFA, 2003).

Table 5: Health risk estimates for cadmium (Cd) and lead (Pb) ingestion from *Rastrelliger kanagurta* based on the fish consumption pattern

	Hazard Quotient (HQ)			
	Cd (once a week)	Cd (7 times a week)	Pb (once a week)	Pb (7 times a week)
Local	1.29	1.30	1.85	1.84
Import	0.71	0.71	0.99	0.98

*HQ < 1 indicates no potential risk of fish consumption

4. Conclusion

The present study shows that local or imported fish species, *Rastrelliger kanagurta*, sold in supermarkets in Shah Alam, Selangor, do accumulate Cd and Pb. The concentration of Cd in both local and imported fish complied with the national and international standard guidelines. Meanwhile, the concentration of Pb in the local fish was above the permissible limits for all the standards guidelines. Furthermore, the mean concentration of Pb in the imported fish complied with the Malaysian Food Regulation (1985) and USFDA (1990) standards but contravened the Joint FAO/WHO (2004) Expert Committee on Food Additives standard. Further analysis for human health risk assessment related to daily or seven times a week imported fish consumption of *Rastrelliger kanagurta* may not pose a health risk (Pb and Cd intoxication) to the consumers in Shah Alam, Selangor since the value was less than 1 ($HQ < 1$). However, the hazard quotient for the local fish intake was more than 1 ($HQ > 1$), which suggests that the daily consumption

or a week of *Rastrelliger kanagurta* fish sold in Shah Alam, Selangor may pose a threat to human health related to both Pb and Cd.

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