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Environmental Health Risk Assessment of Fe, Zn, As, Cd and Pb Concentration in Selected Asian Rice Grain (*Oryza sativa*)

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Abstract

This study was carried out to determine the concentration of chemical elements in food grains (*Oryza Sativa*). A total of eight brands of rice grains was selected which are commonly sold in major hypermarkets in Malaysia. The selection was based on the countries of origin (COO) namely Cambodia, Indonesia, Malaysia, Thailand, Vietnam, China, Japan and Taiwan. A total of 16 rice grain samples included both polished and unpolished rice grains in order to determine the concentration of Fe, Zn, As, Cd and Pb. From the laboratory result, it was found that the concentration of Fe, Zn, As, Cd and Pb was ranged from 0.067-14.446 $mgkg^{-1}$, 0.308-12.335 $mgkg^{-1}$, 0.000-0.048 $mgkg^{-1}$, 0.000-0.604 $mgkg^{-1}$ and 0.000-0.418 $mgkg^{-1}$ respectively. The rice grains samples from Vietnam, China and Cambodia showed that carcinogenic cancer risk for both adults and infants' group (Cumulative lifetime cancer risk, CLCR $>1x10^{-4}$).

Keywords: Environmental contamination, Heavy metals, Asian rice grain, *Oryza sativa*

1. Introduction

Rice grain is a staple food for a large portion of people in the world especially those living in

Asia and it has a very high potential to be affected by contamination through air, water and soil (Hoornweg et al., 2013). Heavy metals are the most common pollutants as they can be found in water, land and air pollution and therefore they have higher probability to influence the paddy rice (Viet and Yabe, 2013). Heavy metals contamination by Cd, Pb, Zn and Cr are serious environmental problem which are caused by numerous anthropogenic activities (Tan et al. 2018). These chemical elements capable to pose a danger to human health since they are non-biodegradable, can be accumulated and can enter the human body through the food chain (NCBI 2015; Kong et al., 2018).

Heavy metals are categorized into carcinogenic and non-carcinogenic (Liu at al., 2016) The International Agency for Research on Cancer grouped arsenic and lead as both carcinogenic and non-carcinogenic elements whereas cobalt, copper, iron, aluminium and zinc as non-carcinogenic (NCBI, 2014). Heavy metals which are classified as carcinogenic elements are those that have the potential to induce tumours when they get into the body through various pathways such as ingestion. Other non-carcinogenic heavy metals can cause other health problems even though they do not cause cancer if they present in large quantities. For example, highly concentrated copper may lead to chronic toxicity in body which cause heart problems, jaundice, coma and even death (NCR, 2000).

Paddy crops are cultivated in water flooded conditions which enhance the ease of pollutants uptake (GRiSP, 2013). As a result, heavy metals are more easily to be absorbed and stored in rice grain as compares with other food. Humans and animals have a higher chance to be influenced as rice acts as producer in the food chain.

2. Material and Methods

2.1 Collection and Preparation of Rice Grain Samples

Total eight brands of rice grain were selected which are being sold in major hypermarkets in Malaysia. The selection was based on the countries of origin (COO) namely Cambodia, Indonesia, Malaysia, Thailand, Vietnam, China, Japan and Taiwan. A total of sixteen samples for both polished and unpolished rice grain from each country were collected in order to carry out the laboratory test to identify heavy metals concentration which was used to conduct human health risk assessment. The samples preparation and processing were conducted according to Praveen and Omar (2017).

2.2 Trace Elements Determination

The elements namely iron (Fe), zinc (Zn), arsenic (As), cadmium (Cd) and lead (Pb) were selected in this study. The analysis method was adopted according to Ma et al. (2017). The stock solution that contained As, Pb, Cd, Zn and Fe was used to prepare solutions with different concentrations (1, 10, 20, 50, 100ugL^{-1}) by using ultrapure water and volumetric flasks and ready to be used in inductively coupled plasma mass spectrometry (ICP-MS) Perkin elmer nexion 2000. Three replications for each sample was tested by ICP-MS

2.3 Human Health Risk Assessment (HHRA) Procedure

Due to absence of cancer slope factor (CFS) by consumption data for Cd, only As and Pb were selected in the determination of carcinogenic health in this study. According to USEPA, Equation 1 was used to find the Average Daily Dose (ADD); Equation 2 was used to find the Hazard Quotient (HQ); Equation 3 was used to find the hazard index (HI); Equation 4 was used to find the Lifetime Cancer Risk (LCR); Equation 5 was used to find cumulative lifetime cancer risk (LCRT) (USEPA 2016). The summary of the constant value for the huma health risk assessment determination were shown in **Table 1-3.**

$$ADD(mg/kg.day) = (C \times IR \times ED \times EF) / (BW \times AT) \quad (1)$$

$$HQ = ADD / R_{fd} \quad (2)$$

$$HI = HQ_{As} + HQ_{Cd} + HQ_{Pb} + HQ_{Fe} + HQ_{Zn} \quad (3)$$

$$LCR = ADD \times CSF \quad (4)$$

$$LCRT = \text{Sum of } LCR \quad (5)$$

Where,

C = Containment Concentration

IR = Ingestion Rate

EF = Exposure Frequency

ED = Exposure Duration

BW =Body Weight

AT = Averaging time

LCR = Lifetime Cancer Risk

Cancer Slope Factor (CSF) for As was 1.5 and for Pb was 0.0085 (USEPA, 2011; OEHHA, 2011). The required information to determine the ADD was obtained by referring to Department of Statistics Malaysia (2014) and Praveen and Omar (2017). The reference dose, R_{fd} for selected chemical elements were used according to USEPA 2006 and WHO 2011 (USEPA, 2006; WHO, 2011).

Table 1. Parameter used in human health risk (HQ and LCR) calculation

HQ	**IR(kg day^{-1})**	**ED(years)**	**EF(days/year)**	**BW(kg)**	**AT(days)**
Adult	0.6	74	365	62.65	ED x 365
Children	0.1984	74	365	19.5	ED x 365
LCR	**IR(kg day^{-1})**	**ED(years)**	**EF(days/year)**	**BW(kg)**	**AT(days)**
Adult	0.6000	74	365	62.65	25550
Children	0.1984	74	365	19.50	25550

(Department of statistic, 2014; USEPA, 2012; Zheng et al., 2014)

Table 2. Refence dose for Fe, Zn, As, Cd and Pb (mg/kg.day)

Element	Reference dose	Reference
Fe	0.700	USEPA, 2006
Zn	0.300	WHO, 2011
As	0.002	WHO, 2011
Cd	0.001	WHO, 2011
Pb	0.004	WHO, 2011

Table 3. Cancer slope factor for As and Pb

Element	CSF	Reference
As	1.5000	(USEPA, 2011)
Pb	0.0085	(OEHHA, 2011)

3. Results and Discussion

The average for selected elements concentration found in polished and unpolished rice grain collected from eight countries (Vietnam, China, Cambodia, Indonesia, Malaysia, Thailand, Taiwan, Japan) are shown in **Table 4 - 5** respectively. Some of the elements in samples were not detected by the ICP-MS therefore were indicated as "n.d.". The highest concentration of Fe, Zn, As, Cd and Pb in both polished rice grain determined in this study were 14.45mgkg^{-1}, 12.34mgkg^{-1}, 0.048mgkg^{-1}, 0.156mgkg^{-1} and 0.418mgkg^{-1} respectively.

According to NCBI (2014), the higher concentrations of heavy metals may pose higher human health risk since many adverse human health issues are associated with concentration of heavy metals adopted in human body. The increase of heavy metals concentration in the environment because of human activities such as mining and natural deformation such as weathering of the earth's crust has increased the human health risk since the heavy metals cannot be biodegradable and can enter human body through inhalation, dermal contact and ingestion. Therefore, it is essential to determine the concentration of heavy metals in rice grain and estimate its risk to human health. The toxicity of heavy metals is associated with the absorbed dose, exposure path and exposure duration. Hence, rice grain which act as a staple food in Asia should be demonstrated with low and acceptable heavy metals concentration.

Table 4. Average heavy metals concentration in polished rice grain (mgkg^{-1})

Sample (COO)	Fe	Zn	As	Cd	Pb
Vietnam	13.455 ±3.388	3.248 ±0.250	0.010 ±0.012	0.020 ±0.019	n.d.
China	6.980 ±2.203	6.808 ±0.615	0.048 ±0.020	0.156 ±0.100	0.418 ±0.725
Cambodia	14.446 ±6.428	9.165 ±3.198	0.009 ±0.009	n.d.	n.d.
Indonesia	10.072 ±0.328	0.661 ±0.611	0.002 ±0.002	n.d.	n.d.
Malaysia	0.067 ±0.115	12.34 ±0.492	n.d.	n.d.	n.d.
Thailand	7.004 ±1.880	0.402 ±0.697	n.d.	0.018 ±0.002	n.d.
Taiwan	8.123 ±2.858	0.308 ±0.534	n.d.	n.d.	n.d.
Japan	1.870 ±0.885	0.544 ±0.426	n.d.	n.d.	n.d.
Malaysia food regulations (1985)	-	100	1.0	1.0	2.0
FAO/WHO (1984)	-	150	1.4	0.4	0.2
FAO/WHO (1989)	100	-	-	-	-

- Non- Detectable – n.d.

Table 5. Average heavy metals concentration in unpolished rice grain (mgkg^{-1})

Sample	Fe	Zn	As	Cd	Pb
Vietnam	1.635 ±1.784	1.638 ±0.635	n.d.	n.d.	n.d.
China	1.279 ±1.185	1.964 ±0.801	n.d.	n.d.	n.d.
Cambodia	1.986 ±1.751	1.006 ±1.549	n.d.	n.d.	n.d.
Indonesia	4.909 ±2.417	1.701 ±0.398	n.d.	0.604 ±0.019	n.d.
Malaysia	0.655 ±1.135	2.315 ±0.353	n.d.	n.d.	n.d.
Thailand	5.941 ±1.066	5.454 ±0.496	n.d.	n.d.	n.d.
Taiwan	3.056 ±1.173	1.679 ±2.018	n.d.	n.d.	n.d.
Japan	3.766 ±0.972	1.997 ±0.815	n.d.	n.d.	n.d.
Malaysia food regulations (1985)	-	100	1.0	1.0	2.0
FAO/WHO (1984)	-	150	1.4	0.4	0.2
FAO/WHO (1989)	100	-	-	-	-

- Non- Detectable – n.d.

The As concentration in unpolished rice grains samples were not detected by ICP-MS. It may that the concentration was below the detection limit of ICP-MS (<0.001ppb). Among the polished rice grain samples, the As concentrations recorded for Vietnam, China, Cambodia and were 0.010 mgkg^{-1}, 0.048 mgkg^{-1}, 0.009 mgkg^{-1} and 0.002 mgkg^{-1}. The detected As concentration was lower than the permissible limit set by Malaysia Food Regulation (1985) (1.0 mgkg^{-1}) and FAO/WHO (1984) (1.4 mgkg^{-1}).

The concentration of Cd found in Indonesia unpolished rice grain sample was 0.604mgkg^{-1}. Polished rice grain samples collected from Vietnam, China and Thailand, on the other hand, were detected to contain Cd element. The China polished rice grain sample recorded the highest value (0.156 mgkg^{-1}). In comparison with the permissible concentration in FAO/WHO (1984) (0.4 mgkg^{-1}) and Malaysia Food Regulation (1985) (1.0 mgkg^{-1}), the highest Cd concentration recorded in Indonesia unpolished rice grain sample was between these two standards. The Pb concentration in brown rice and white rice from all the selected countries were not detected except the polished rice grain sample from China exceeded the permissible concentration (0.2mgkg^{-1}) set by FAO/WHO (1984). However, it is under the permissible limit (2.0mgkg^{-1}) set by Malaysia Food Regulation (1985).

According to Hironori (2012), small paddy farms are practiced in China, Indonesian Java, and Red River Delta in Vietnam which have average size less than 0.5 ha; the size of paddy farms in Bangladesh, Eastern of India, and the Mekong River Delta in Vietnam have average size of less than 1 ha.; the paddy farms in Japan have average size of about 1 ha.; the rice farms in most other Asian countries have average size of 1–2 ha.; the rice farms in Thailand, Myanmar, Cambodia, and Punjab, India are average farm size larger than 2 ha. (Yagi, 2012; Ji et al. 2016). Hence, it cannot be concluded that the amount of heavy metals assimilated by the rice grain is constant concentration. The concentration of heavy metals in soil may not be the same as uptake concentration. There may be factors that influence the growth of rice grains that affect the heavy metal assimilation. Those paddy crops that are cultivated under the optimal conditions where sunlight exposure, water provision and nutrients provision are optimum, may contain more heavy metals. In a large paddy field, it is impossible to ensure that the sunlight, water and nutrients are distributed evenly to every area since the composition of soil may slightly be different and many other unknown factors may disturb the distribution such as invertebrates which are living in soil which may change the characteristic of soil.

3.1 Human Health Risk Assessment

Human health risk assessment was conducted based on the result obtained from laboratory analysis**.** The calculated hazard quotient and hazard index are tabulated in **Table 6** and **7**. For adult receptor group, the hazard quotient and hazard index for all polished rice grain samples are determined as lower than the risk level limit (1.0) except China's sample (HQ_{cd} = 1.579, HQ_{Pb} = 1.058, HI = 3.182). On the other hand, the hazard quotient and hazard index for all unpolished rice grain samples were below the risk limit except Indonesia samples (HQ_{cd} = 6.115, HI =6.237).

In the children receptor group, the hazard index of China polished rice samples was determined as exceeded the risk level limit (HQ_{cd} = 5.074, HQ_{Pb} = 3.399, HI= 10.222). Also, the unpolished rice grain sample from Indonesia was found to have unacceptable hazard index >1.0 (HQ_{cd} = 19.647, HI = 20.037). The calculated hazard quotient is based on the reference dose which is safe to human health. The concentrations of Cd, Zn and Pb in some rice grain samples exceeded the permissible level for safe consumption and also can pose severe health risk to human. High cadmium concentration can cause human health problems. For examples, it can pose significant damage to kidney and has the potential to induce tumor effects and thus its risk to human health is high. An incident in Japan, many victims in Japan who were living near to the contaminated area had developed Itai-itai disease because of cumulative cadmium exposure through ingestion (Nishijo et al. 2017).

Hence, the amount of daily intake should be reduced so that it will not develop significant effects to human health (USEPA, 2011). Although the hazard values of the selected trace elements may not be in excess of 1.0. it doesn't mean that the rice is safe for consumption daily because full health risk description is not enough (Praveen and Omar 2017) Therefore, the hazard index which is the cumulative hazard values from selected trace elements are calculated since cumulative exposure can increase the risk. Thus, the hazard index which is more than 1 will also pose potential health risk although the individual hazard value is less than 1.

Table 6. Hazard Quotient (HQ) and Hazard Index (HI) for adult in relation to test results of samples from different countries

Sample COO	Fe	Zn	As	Cd	Pb	HI
Vietnam (w)	0.184	0.104	0.048	0.202	0.000	0.538
China (w)	0.095	0.217	0.231	***1.579***	***1.058***	***3.182***
Cambodia (w)	0.198	0.293	0.043	n.d.	n.d.	0.534
Indonesia (w)	0.138	0.021	0.010	n.d.	n.d.	0.169
Malaysia (w)	0.001	0.394	n.d.	n.d.	n.d.	0.395
Thailand (w)	0.096	0.013	n.d.	0.182	n.d.	0.291
Taiwan (w)	0.111	0.010	n.d.	n.d.	n.d.	0.121
japan (w)	0.026	0.017	n.d.	n.d.	n.d.	0.043
Vietnam (b)	0.022	0.052	n.d.	n.d.	n.d.	0.075
China (b)	0.017	0.063	n.d.	n.d.	n.d.	0.080
Cambodia (b)	0.027	0.032	n.d.	n.d.	n.d.	0.059
Indonesia (b)	0.067	0.054	n.d.	***6.115***	n.d.	***6.237***
Malaysia (b)	0.009	0.074	n.d.	n.d.	n.d.	0.083
Thailand (b)	0.081	0.174	n.d.	n.d.	n.d.	0.255
Taiwan (b)	0.042	0.054	n.d.	n.d.	n.d.	0.095
Japan (b)	0.052	0.064	n.d.	n.d.	n.d.	0.115

*Non- Detectable – n.d.

- (w) Polished rice grain

- (b) Unpolished rice grain

Table 7. Hazard Quotient (HQ) and Hazard Index (HI) for children in relation to test results of samples from different countries

Sample COO	Fe	Zn	As	Cd	Pb	HI
Vietnam (w)	0.591	0.333	0.155	0.651	n.d.	1.730
China (w)	0.307	0.698	0.743	***5.074***	***3.399***	***10.222***
Cambodia (w)	0.635	0.940	0.139	n.d.	n.d.	***1.714***
Indonesia (w)	0.443	0.068	0.031	n.d.	n.d.	0.541
Malaysia (w)	0.003	***1.265***	n.d.	n.d.	n.d.	***1.268***
Thailand (w)	0.308	0.041	n.d.	0.585	n.d.	0.935
Taiwan (w)	0.357	0.032	n.d.	n.d.	n.d.	0.389
japan (w)	0.082	0.056	n.d.	n.d.	n.d.	0.138
Vietnam (b)	0.072	0.168	n.d.	n.d.	n.d.	0.240
China (b)	0.056	0.201	n.d.	n.d.	n.d.	0.258
Cambodia (b)	0.087	0.103	n.d.	n.d.	n.d.	0.190
Indonesia (b)	0.216	0.174	n.d.	***19.647***	n.d.	***20.037***
Malaysia (b)	0.029	0.237	n.d.	n.d.	n.d.	0.266
Thailand (b)	0.261	0.559	n.d.	n.d.	n.d.	0.821
Taiwan (b)	0.134	0.172	n.d.	n.d.	n.d.	0.307
Japan (b)	0.166	0.205	n.d.	n.d.	n.d.	0.370

*Non- Detectable – n.d.

- (w) Polished rice grain

- (b) Unpolished rice grain

The hazard index (HI) and hazard quotient (HQ) are used to describe the non-carcinogenic health risk, whereas the carcinogenic health risk is described by the lifetime cancer risk. The acceptable range of lifetime cancer risk according to the standard in Malaysia is ranging from $1x10^{-6}$ to $1x10^{-4}$. For carcinogenic elements Arsenic (As) and Lead (Pb), the life time cancer risk values were determined based on Equation 4 and 5. As shown in **Table 8**, the polished rice samples from Vietnam, China and Cambodia showed LCR_{As} for both children and adult exceeding LCR limit, especially for China's polished rice grain $LCR_{As,\ children}$. This study found that the polished rice sample from China exceeded acceptable level (2.34×10^{-3}).

From the study, results for cumulative lifetime cancer risk ($LCR_{As,\ Pb}$) for both adults and children receptors were $7.65x10^{-4}$ and $2.46x10^{-3}$ respectively, both had exceeded the Malaysia's LCR standard. The cumulative lifetime cancer risk is increased due to the presence of individual lifetime cancer risk of Arsenic (As) in rice grain sample from China. The increase in cumulative lifetime cancer risk shows that daily consumption of rice have higher risk to get cancer. Overall, the polished rice grains and its products imported from Vietnam, China and Cambodia didn't pose potential carcinogenic health risk to adult and children in Malaysia.

Table 8. Lifetime cancer risk for adult ($LCR_{As,\ adult}$; $LCR_{Pb,\ adult}$) for different polished rice samples from different countries

Sample COO	$LCR_{As,\ adult}$	$LCR_{Pb,\ adult}$	$LCR_{As,\ Pb,\ adult}$	$LCR_{As,\ children}$	$LCR_{Pb,\ children}$	$LCR_{As,Pb,\ children}$
Vietnam	$1.52x10^{-4}$	n.d.	$1.52x10^{-4}$	$4.88x10^{-4}$	n.d.	$4.88x10^{-4}$
China	$7.29x10^{-4}$	$3.60x10^{-5}$	$7.65x10^{-4}$	$2.34x10^{-3}$	$1.16x10^{-4}$	$2.46x10^{-3}$
Cambodia	$1.37x10^{-4}$	n.d.	$1.37x10^{-4}$	$4.39x10^{-4}$	n.d.	$4.39x10^{-4}$
Indonesia	$3.04x10^{-5}$	n.d.	$3.04x10^{-5}$	$9.76x10^{-5}$	n.d.	$9.76x10^{-5}$
Malaysia	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Thailand	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Taiwan	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Japan	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

*Non- Detectable – n.d.

From the result of lifetime cancer risk, cumulative lifetime cancer risk, hazard quotient and hazard index, the polished rice COO from China, Vietnam and Cambodia may have higher risk as compared to other countries and it is recommended to reduce ingestion rate of the rice grain where adults should consume less than 600$gday^{-1}$ and less than 198.4$gday^{-1\ +}$ for children. The polished rice grain from Malaysia has low health risk for both children and adult consumption. However, the hazard index for children was slightly over the permissible limit (1.0), the ingestion rate may need to be reduced to to less than 198.4$gday^{-1}$ to ensure that the children will not develop complicated health effect for both carcinogenic and non-carcinogenic in the future.

Beside the local supplier, majority of the polished and unpolished rice in the market are imported from Thailand. The results showed that both rice samples collected would not bring significant health risk to both adult and children except for the white rice from Malaysia which slightly exceeded the permissible limit for children due to the zinc content. Ingestion rate of 198.4g per day and 600g per day was used in calculating the average daily dose for both children and adults. If the ingestion rate for children is reduced, the hazard index for Malaysia polished rice grain may not exceed the safety limit.

4. Conclusion

The heavy metals concentration in all rice grain samples collected from the selected eight countries namely Cambodia, China, Indonesia, Malaysia, Thailand, Vietnam and Taiwan comply Malaysia food regulations (1985), FAO/WHO standard. The increase in cumulative lifetime cancer risk shows that the daily consumption of the rice grain will have higher risk to get cancer. Overall, the polished rice grains and its products imported from Vietnam, China and Cambodia doesn’t pose potential carcinogenic health risk to adults and children in Malaysia. In order to reduce the rate of potential heavy metals exposure, the residents in Malaysia are required to be aware the country of origin (COO) for the rice product.

Since rice is a staple food for most population in Malaysia and many products are made from rice, it is difficult to reduce the ingestion rate by replacing with other foods. Therefore, innovative green technologies may be required to process the rice to reduce the heavy metals in rice and more efforts from industries which produce waste that contains heavy metals, agricultural sectors and government are needed to counter the problems.

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