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Assessment of Heavy Metal Content and Consumption Risks At Selected Paddy Field in Malaysia: A Review

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ABSTRACT

As the Malaysian population grows, there is a high demand for rice, the main staple food in this region. This has caused the overuse of agrochemicals that contain heavy metals and the utilization of contaminated groundwater to increase paddy yield, posing a risk to humans. This study reviewed the accumulated heavy metals in paddy fields of Malaysia's Selangor, Kedah and Sabah states and further calculated the consumption risks of rice grains from the selected areas. The study revealed that paddy soil in Ranau Valley (Sabah), Kota Marudu (Sabah) and Tanjung Karang (Selangor) showed presences of Cu and Cd in high concentration, respectively, creating higher potential to be uptake by paddy roots. These findings also revealed that Ranau Valley (Sabah) paddy grains contained high Cu and Cd concentrations, while Sabak Bernam (Selangor) contained high Pb concentrations. Further, a higher Cd concentration was reported from the Ranau Valley (Sabah), while the higher Pb concentrations were reported from the samples collected from Sabak Bernam (Selangor), Tanjung Karang (Selangor) and Kubang Pasu (Kedah). Based on the health risk indices calculation in this study, carcinogenic and noncarcinogenic health risks in all study areas except in Kubang Pasu (Kedah) and Langkawi (Kedah) are likely to occur due to Cu mining activities, ultrabasic soil contamination, utilization of contaminated groundwater and rock phosphate fertilizer and vehicular emission. Regular assessment of heavy metal content and consumption risks of paddy is essential to ensure the paddy field is free from contamination and will help protect the ecosystem and human health.

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1 Introduction

Apart from being the staple food for Malaysians, paddy serves as a primary income source for farmers and agricultural workers in Malaysia (Omar et al. 2019; Firdaus et al. 2020). An estimated agricultural land in Malaysia was 645,000 ha, used to cultivate the paddy (Akinbile et al. 2011). Since 2018, more and more lands have been developed for paddy farming in Malaysia to meet the country's high rice consumption demand (Malaysia Population 1950-2021 2021). This has led to farmers using excessive agrochemicals such as fertilizers, insecticides, and herbicides to manage weeds and pests while also ensuring higher yield of paddy cultivation (Rudzi et al. 2018; Engwa et al. 2019; Alengebawy et al. 2021).

Agrochemicals contain toxic heavy metals that bioaccumulate to concentrations higher than permissible (Ali et al. 2018). According to Rudzi et al. (2018), overuse of agrochemicals is one of the primary causes of limiting high rice production as it makes paddy unsafe for human consumption. Due to the heavy metals' nonbiodegradable characteristics, excessive use of agrochemicals, including lead (Pb), cadmium (Cd), and copper (Cu), causes environmental pollution and, more seriously, contamination of the paddy soil and grains themselves (Rudzi et al. 2018; Irshad et al. 2023). Using tainted irrigation water and soil for rice production is a problem since they contain heavy metals that can be transferred from the roots of the paddy into the grains (Akinbile et al. 2011).

Despite the grave consequences of heavy metals released from agrochemicals to humans and the environment, the health risk assessments of paddy in Malaysia are not well documented. Further studies on the heavy metals contained in paddy are crucial as rice production expands and intensifies. Therefore, this review aims to investigate the accumulation levels of Cu, Cd, and Pb in the soils of paddy fields and the risk of rice grains via ingestion in Selangor, Kedah, and Sabah.

2 Review of heavy metals and risk assessment in paddy fields at the selected states

Data obtained from the Selangor, Kedah, and Sabah state paddy fields were chosen for this study because they are Malaysia's leading rice-producing states. These sites are all situated in different parts of Malaysia: Selangor (central region) and Kedah (northern region) are both situated on Peninsular Malaysia's western coast, while Sabah is located in East Malaysia (Alias et al. 2014; Zulkafflee et al. 2019). This article is based on the compilation of data on the mean concentrations of Cu, Cd and Pb from soil and grains samples collected from Sabak Bernam (Selangor), Tanjung Karang (Selangor), Langkawi (Kedah), Kubang Pasu (Kedah), Kota Marudu (Sabah) and Ranau Valley (Sabah). Furthermore, Hazard Quotients (HQ) and Lifetime Cancer Risk (LCR) of rice grains calculated based on the data taken from reported studies in these areas were performed.

2.1 Concentrations of Cu, Cd, and Pb in Soil Samples

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The mean levels of Cu, Cd, and Pb (mg/kg) in paddy soil collected from Sabak Bernam, Tanjung Karang, Langkawi, Kubang Pasu, Kota Marudu, and Ranau Valley are displayed in Table 1. Based on compiled data (Table 1), the highest levels of Cu and Cd (154.83 and 0.776 mg/kg, respectively) were found in paddy soil

Table 1 The mean concentration data of Cu, Cd, and Pb (mg/kg) in paddy soil samples collected from the selected study are	a
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Study Areas	Mean concentration (mg/kg)		(mg/kg)	References	
	Cu	Cd	Pb		
SabakBernam (Selangor)	0.451	0.012	0.984	Zulkafflee et al. 2019	
Tanjung Karang (Salangor)	0.538	0.014	1.088	Zulkafflee et al. 2019	
Tanjung Karang (Selangor)	Tanjung Karang (Selangor)3.87		6.64	Rudzi et al. 2018	
Langkawi (Kedah)	0.178	0.014	0.380	Jusoh et al. 2013; Zakaria et al. 2021	
Kubang Pasu (Kedah)	0.331	0.117	0.571	Zulkafflee et al. 2021	
Rubang Fasu (Redail)	-	0.20	3.72	Looi et al. 2014; Zakaria et al. 2021	
Kota Marudu (Sabah)	ND	0.776	ND	Yap et al. 2009; Zakaria et al. 2021	
Ranau Valley (Sabah)	154.83	0.45	-	Aziz et al. 2015; Zakaria et al. 2021	
Comparison with standard data					
GB15618-2018	50	0.30	80	Soil Environmental Quality Standards (GB 15618-2018), 2018)	
European Standards Agriculture Soils	140	3	300	EU Soil Policy 2018	

*ND: Not Detected; *GB15618-2018: Soil environmental quality: Risk control standard for soil contamination on agricultural land. *Significant level for one sample T-test at p<0.05.

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samples taken from Ranau Valley in Sabah and Kota Marudu in Sabah (Yap et al. 2009; Aziz et al. 2015; Rudzi et al. 2018; Zakaria et al. 2021). Pb was reported to have the highest concentration of 6.64 mg/kg in paddy soil collected from Tanjung Karang, Selangor (Rudzi et al. 2018).

Soil analysis from the paddy field of Ranau Valley in Sabah demonstrates a high concentration of Cu of 154.83 mg/kg, which exceeded the permissible limit, and this can be associated with the ultra-basic soil pollution and absorption by organic matter and clay minerals in the soil (Aziz et al. 2015). It was reported that some metals were enriched through precipitation in ultrabasic soil and positively correlated with Cu (Aziz et al. 2015).

Precipitation immobilizes the Cu in the soil, raising the concentration of Cu present (Yoon et al. 2019). Furthermore, the sampling site is close to the ex-Mamut Cu Mine (MCM), which also releases Cu into the environment during smelting, possibly flowing through neighbouring paddy soil (Lo and Saibeh 2013; Kiprop 2021). MCM mining activities generate large amounts of wastewater contaminating the groundwater in Ranau Valley (Wuana and Okieimen 2011; Masindi and Meudi 2018). Runoff and using contaminated groundwater for paddy cultivation can result in high Cu concentrations in the sediment (Lo and Saibeh 2013). At minute levels, Cu promotes plant growth; therefore, it is necessary to maintain crop yield. On the contrary, excessive Cu buildup and leaching from agricultural soils might lead to groundwater pollution, causing harm to all organisms via the food chain (Stanislawska and Korzeniowska 2018; Aziz et al., 2023). Heavy metals contaminated soils are likely to be directly absorbed by paddy roots and eventually translocated to edible plant parts, endangering rice consumers (Aziz et al. 2015).

The fertilizer utilized in the paddy fields in Kota Marudu and Tanjung Karang is mainly in the form of rock phosphate, which contains Cd and has a solubility of 65% (Yap et al. 2009; Rudzi et al. 2018; Teles 2020; Zakaria et al. 2021). It shows that only 65% of the total rock phosphate is accessible for paddy, while the remaining 35% remains in the soil, resulting in elevated levels of Cd (Teles 2020; Suciu et al. 2022). The Cd concentrations in paddy soil were higher in Kota Marudu (Sabah) and Ranau Valley (Sabah), which suggests that the potential for Cd to be absorbed by rice roots in these regions is higher than the suggested one (Payus et al. 2015).

Further, Cd concentrations in Ranau Valley and Kota Marudu, Sabah are 0.45 mg/kg and 0.776 mg/kg, respectively and found to be exceeding the maximum allowable concentration suggested by Soil Environmental Quality Standards and European Standards Agriculture Soils (EU Soil Policy 2018; Soil Environmental Quality Standards (GB 15618-2018) 2018). This suggests that the soil samples from Kota Marudu and Ranau Valley may not be

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suited for paddy agriculture since excessive levels of Cd tend to bio-accumulate in paddy plant roots and translocate into paddy grains, thereby reducing the overall paddy yield and detrimental to human health (Satpathy et al. 2014). Various causes could affect Cd concentrations, such as industrial activities, air emissions, and organic sediment deposition (Kahn et al. 1992). It is also evident that, based on in-depth analyses of numerous fertilizer products, some micronutrient and phosphate fertilizers contain elevated Cd compared to others. The public's main concern is the persistent nature of Cd and its bioaccumulation in the food chain (Gao et al. 2022). The effect of Cd on the human body is kidney disease and can potentially affect the pulmonary, cardiovascular, and musculoskeletal systems (Munir et al. 2023). The 'itai- itai' disease is also associated with Cd toxicity in rice grown on industrial waste-contaminated soils (Nishijo 2017; Zulkafflee et al. 2019). Cd uptake and bioaccumulation in crops are influenced by soil factors such as pH, salinity, fertilizers, level of organic matter, and species of crops (Tang et al. 2023). The application of fertilizer, therefore, could increase the risk of Cd being transferred to the food chain (Robert 2014).

2.2 Concentrations of Cu, Cd and Pb in paddy grain samples

The mean concentrations of Cu, Cd and Pb (mg/kg) in the paddy grain from Sabak Bernam, Tanjung Karang, Langkawi, Kubang Pasu, Kota Marudu and RanauValley paddy field are tabulated in Table 2.

According to Aziz et al. (2015) and Zakaria et al. (2021), paddy grain samples taken from Ranau Valley (Sabah) has the highest concentrations of Cu (2.61 mg/kg) and Cd (0.54 mg/kg), which surpassed the permissible limit of Malaysian Food Regulations 1985 and FAO/WHO CAC 1984. This is due to a greater tendency for Cu and Cd in the soil to be taken up by the paddy roots and translocated into the grains (Chibuike and Obiora 2014; Yan et al. 2020). It is also evident from Table 1 that the soil samples analysis of Ranau Valley indicated a high concentration of Cu. This shows strong Cu translocation from the paddy soil to the grain sample. The ability of different parts of crop plants to bioaccumulate and translocate heavy metals among plants varies according to the species (Zulkafflee et al. 2021). Genotypic variation and organic soil material are other factors that may affect the metals in the crops (Liu et al. 2015). This is something to be concerned about, whereby the ingestion of metal-contaminated crops could cause detrimental effects to consumers.

According to Zulkafflee et al. (2019), paddy grain samples from Sabak Bernam (Selangor) had the highest Pb concentration (2.245 mg/kg). This might be due to the proximity of sampling sites to Route 5, considered a busy route in Malaysia (InventoriRangkaian Jalan Utama Persekutuan Semanjung Malaysia, 2021). The increased Pb concentration in paddy grains is a result of the Assessment of Heavy Metal Content and Consumption Risks At Selected Paddy Field in Malaysia: A Review

Table 2 Mean concentrations data of the Cu, Cd and Pb (mg/kg) of grains collected from the selected study area					
Study Areas	Mean concentration (mg/kg)			References	
	Cu	Cd	Pb		
SabakBernam (Selangor)	1.166	0.026	2.245	Zulkafflee et al. 2019	
Tanjung Karang (Selangor)	1.477	0.324	0.406	Zulkafflee et al. 2019	
Langkawi (Kedah)	0.058	0.029	0.069	Jusoh et al. 2013; Zakaria et al. 2021	
Kuhang Dagu (Kadah)	0.125	0.003	0.043	Zulkafflee et al. 2021	
Kubang Pasu (Kedah)	-	0.060	1.05	Looi et al. 2014; Zakaria et al. 2021	
Kota Marudu (Sabah)	0.312	0.180	ND	Yap et al. 2009; Zakaria et al. 2021	
Ranau Valley (Sabah)	2.61	0.54	-	Aziz et al. 2015; Zakaria et al. 2021	
Comparison with the standard value					
Malaysian Food Regulations 1985	30	1.0	2.0	Malaysia Food Regulations 1985, 1985	
FAO/WHO CAC 1984	10	0.4	0.2	FAO/WHO 2011	

*ND: Not Detected; *Significant level for one sample T-testat p<0.05.

disclosure to exhaust fumes released by motor vehicles due to numerous inhabitants and visitor's heavy use of Route 5 (França et al. 2017; Rai et al. 2019; Inventori Rangkaian Jalan Utama Persekutuan Semanjung Malaysia 2021).

Looi et al. (2014) and Zakaria et al. (2021) found elevated levels of Cd and Pb (0.01 and 0.21 mg/kg, respectively) in paddy grain samples in Kubang Pasu (Kedah) than the value (0.003 and 0.043 mg/kg, respectively) reported by Zulkafflee et al. (2021), it denoting that high concentrations of Cd and Pb in sediment are more likely to be uptaken by paddy roots and translocated into the grains. This will increase the risk that consumers may experience health concerns like bone malformation, liver damage, and pancreatic cancer (Eske 2020; Guo et al. 2020; TatahMentan et al. 2020; Rai et al. 2019).

Malaysian Food Regulations 1985 and FAO/WHO CAC 1984 reported that the paddy grains' Cd concentration in Ranau Valley, Sabah (0.54 mg/kg) and Pb concentration in Sabak Bernam, Selangor (2.245 mg/kg), Tanjung Karang, Selangor (0.406 mg/kg) and Kubang Pasu, Kedah (1.05 mg/kg) surpassed the maximum permissible level. Paddy grains containing Cd and Pb concentrations over the acceptable limits are not safe for ingestion by humans since they may cause kidney dysfunction, bone pain, and lung cancer when consumed in large quantities (Eske 2020; Genchi et al. 2020; TatahMentan et al. 2020; Hasan et al. 2022).

2.3 Hazard Quotient Values of Cu, Cd and Pb in Adults

The paddy grain consumption risks can be accessed through hazard quotients (HQ)(Zheng et al. 2020). Here, the assessment can be divided into different degrees of risk. Table 3 shows the adult's Hazard Quotient (HQ) values for Cu, Cd and Pb calculated based

Journal of Experimental Biology and Agricultural Sciences http://www.jebas.org on reported data in Table 2 by using the formula listed below (Calculating Exposure Doses 2005; IRIS 2011; Fan et al. 2017; Zulkafflee et al. 2019):

 $HQ = D (mg/kg \cdot day)/RfD (mg/kg \cdot day)$

$$HQ = A x \frac{I x E x D}{W x T} / RfD$$

Where

D = Average daily intake

RfD = Reference dose

A = Exposure point concentration

I = Intake rate of contaminated medium, 0.425

- E = Exposure factor, 74 days per year
- D = Exposure duration, 1 years

W=Body Weight, 70 kg

T=Averaging time, 74 x 1 year

RfD used are Cd = 0.001, Cu = 0.040, and Pb = 0.004(IRIS 2011).

Based on Table 3, all HQ values of Cu exposure for adults were below 1 (HQ <1) and did not show any non-carcinogenic health risk caused by Cu, such as kidney failure, liver damage and hemolysis in all study areas (Eske 2020; Moussiegt et al. 2020). HQ values of Cd exposure for adults were below 1 (HQ <1) except for the HQ values in Tanjung Karang, Selangor (1.967), Kota Marudu, Sabah (1.093) and Ranau Valley, Sabah (3.279). These Cd levels could cause potential non-carcinogenic health risks, for

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Table 3 Hazard Quotient (HQ) values of Cu, Cd and Pb in adults calculated based on the data taken from the reported studies
and using the formula listed above

Areas of Study	HQ values			
	Cu	Cd	Pb	
SabakBernam (Selangor)	0.177	0.158	3.407	
Tanjung Karang (Selangor)	0.224	1.967	0.616	
Langkawi (Kedah)	0.009	0.176	0.105	
Kubang Pasu (Kedah)	0.019	0.018	0.065	
Kota Marudu (Sabah)	0.047	1.093	ND	
Ranau Valley (Sabah)	0.396	3.279	-	

*ND: Not Detected

instance, cardiovascular disease, kidney dysfunction and severe bone pain (Nishijo 2017; Rahimzadeh et al. 2017; Zulkafflee et al. 2019) for inhabitants in these areas. All the HQ values of Pb exposure for adults were below 1 (HQ <1) except for the HQ value for Sabak Bernam, Selangor (3.407). This indicates noncarcinogenic health effects caused by Pb, for instance, bone malformation, nervous system damage and high blood pressure (Chari 2016; Health Problems Caused by Lead 2018), which consumers would likely experience in Sabak Bernam, Selangor.

2.4 Lifetime Cancer Risk Values of Cadmium and Lead in Adults

The adult's Lifetime Cancer Risk (LCR) values for Cd and Pb are calculated based on the data taken from the reported studies in Table 2 by using the formula listed below, and the value is tabulated in Table 4 (Calculating Exposure Doses 2005; RAIS 2011; Fan et al. 2017; Zulkafflee et al. 2019):

$$LCR = DS (mg/kg \cdot day) \times CSF (mg/kg \cdot day)^{-1}$$

LCR =
$$(A \times \frac{I \times E \times D}{W \times AT}) \times CSF$$

LCR = $(0.026 \times \frac{0.425 \times 74 \times 1}{70 \times 74 \times 1}) \times 0.38$
LCR = 6.00×10^{-5}

Where

DS =Average Daily Dose (mg/kg/day)

CSF = Cancer Slope Factor

This study did not determine the LCR values of Cu as, according to RAIS (2011), Cu's cancer slope factor (CSF) values were inaccessible, and Cu was regarded as non a cancer-causing element.

Based on Table 4, all LCR values of Cd exposure for adults were less than 1 x 10^{-4} (LCR <1 x 10^{-4}) except for the LCR values of Cd

Journal of Experimental Biology and Agricultural Sciences http://www.jebas.org in Tanjung Karang, Selangor (7.48×10^{-4}) , Kota Marudu, Sabah (4.15×10^{-4}) and Ranau Valley, Sabah (1.25×10^{-3}) . This indicates that among every 10,000 adult individuals in Tanjung Karang (Selangor), Kota Marudu (Sabah) and Ranau Valley (Sabah), there is a probability of 4 to 13 individuals who may develop carcinogenic-related health risks caused by Cd exposure such as lung, pancreas and breast cancers over a period 74 years (Rahimzadeh et al. 2017; Genchi et al. 2020; Tatah Mentan et al. 2020). Intake of Cd-contaminated rice may pose a risk of contracting the itai-itai disease. During the 1950s, ingesting cadmium-contaminated rice (itai-itai disease) impacted humans' well-being (Nishijo 2017). Itai-itai disease complications are associated with osteomalaecia and severe bone pain, as well as renal tubular failure (Shi et al. 2020).

All adult's LCR values of Pb exposure were below 1×10^{-4} (LCR <1 x 10^{-4}) except for the LCR value of Pb in Sabak Bernam, Selangor (1.16 x 10^{-4}). This indicates that among every 10,000 adult individuals in SabakBernam, Selangor, there is a probability of 1 individual who may develop carcinogenic-related health risks caused by Pb exposure, such as brain, stomach and lung cancers over 74 years (Lead Poisoning and Health 2019; Rai et al. 2019).

2.5 Determination of Safety Consumption of Paddy

According to the findings, paddy from Langkawi and Kubang Pasu in Kedah is safe to eat because the HQ values of Cu (0.009 and 0.019), Cd (0.176 and 0.018) and Pb (0.105 and 0.065) are less than 1. The LCR values for Cd (6.69×10^{-5} and 6.92×10^{-6}) and Pb (3.56×10^{-6} and 2.22×10^{-6}) are less than 1×10^{-4} , indicating that serious health risks like cardiac failure, kidneys dysfunction and pancreas cancer would not occur due to exposure to these chemicals or lack thereof (Chari 2016; Genchi et al. 2020; Moussiegt et al. 2020). These data are in line with the studies reported in Iranshahr, Iran (Djahed et al. 2018); Zhejiang Province, China (Huang et al. 2013); Enugu, Nigeria and Omar (Ezeofor et al. 2019); and Yan and Pendang, Kedah (Omar et al., 2015). Table 4 The Lifetime Cancer Risk (LCR) values of Cd and Pb in adults were calculated based on the data taken from the reported studies by using the formula listed above

Areas of Study	as of Study LCR values	
	Cadmium (Cd)	Lead (Pb)
Sabak Bernam (Selangor)	6.00 x 10 ⁻⁵	1.16 x 10 ⁻⁴
Tanjung Karang (Selangor)	7.48 x 10 ⁻⁴	2.10x 10 ⁻⁵
Langkawi (Kedah)	6.69 x 10 ⁻⁵	3.56 x 10 ⁻⁶
Kubang Pasu (Kedah)	6.92 x 10 ⁻⁶	2.22 x 10 ⁻⁶
Kota Marudu (Sabah)	4.15 x 10 ⁻⁴	ND
Ranau Valley (Sabah)	1.25 x 10 ⁻³	-

*ND: Not Detected

It appears to be unsafe to consume paddy from SabakBernam (Selangor), Tanjung Karang (Selangor), Kota Marudu (Sabah), and Ranau Valley (Sabah) as the HQ values of Cd (1.093 to 3.279) and Pb (3.407) are more than 1. The LCR values of Cd (4.15×10^{-4} to 1.25 x 10⁻³) and Pb (1.16×10^{-4}) are more than 1 x 10⁻⁴, indicating that severe health risks such as liver damage, bone malformation and lung cancer might occur (Rai et al. 2019; Eske 2020; TatahMentan et al. 2020). These data are in line with those reported in Hunan Province, China (Zeng et al. 2015), the Pearl River Delta of China (Zheng et al. 2020), Enugu, Nigeria (Ihedioha et al. 2016; Ezeofor et al. 2019), and Iran (Fakhri et al. 2018).

Conclusion

Based on information from published studies, elevated concentrations of Cu and Cd in the paddy soil in Ranau Valley and Kota Marudu increased the possibility of paddy roots absorbing these metals. Cu and Cd levels in Ranau Valley and Cd levels in Kota Marudu exceeded the maximum allowable concentration suggested by Soil Environmental Quality Standards and European Agricultural Standards Soils. The paddy grains' Cd concentration in Ranau Valley and the Pb concentrations in Sabak Bernam, Tanjung Karang, and Kubang Pasu exceeded the maximum allowable level recommended by the Malaysian Food Regulations of 1985 and the FAO/WHO CAC 1984.

Based on the calculated HQ and LCR values of Cu, Cd and Pb, paddy from Langkawi and Kubang Pasu were safe for consumption. This study also indicated that the paddy grain taken from Sabak Bernam, Tanjung Karang, Kota Marudu and Ranau Valley was not safe for consumption based on the HQ and LCR value analysis. From this review, we found that the paddy is not safe for consumption, and it is important to carry out a monitoring program on the presence of Cu, Cd and Pb in paddy soils and grains. These findings also provide valuable insights for evaluating future impacts of environmental pollution towards rice production and potential health risks from Cu, Cd and Pb. Regular assessment of paddy consumption risks should be done to protect the ecosystem and human health.

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