ORIGINAL ARTICLE

Heavy Metals Contamination in Paddy Soil and Water and Associated Dermal Health Risk Among Farmers

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ABSTRACT

Introduction: This study aims to quantify the concentrations of Cu, Cr, Zn, Ni, Pb, As and Cd in paddy soil and water and assess their potential dermal health risk to the farmers at Kampung Sawah Sempadan, Malaysia. **Methods:** 72 water samples and 72 soil samples were collected and analysed using inductively coupled plasma mass spectrometry (ICP-MS). 117 respondents were interviewed using a questionnaire to obtain the exposure information for dermal health risk assessments. **Results:** All elements in water did not exceed the recommended concentration by Malaysia National Water Quality Standard (NWQS) and Food and Agriculture Organization (FAO). Nevertheless, the maximum concentration of As (31.49 mg/kg) in paddy soil exceeded the Dutch Target Value for soil protection (29 mg/kg). There was no significant chronic non-carcinogenic health risk for farmers working in the paddy soil and water (HQ<1, HI<1). The carcinogenic health risk of As was in the acceptable risk level (10-6 to 10-4). **Conclusion:** The contamination of the selected heavy metals in the paddy soil and water at Kampung Sawah Sempadan were lower than the available standards except for As in soil which exceeds the recommended value by Dutch target value. The health risk to these heavy metals were also minimal and within the acceptable level.

Keywords: Inductively coupled plasma mass spectrometry (ICP-MS), Water Quality, Soil, Risk assessment, Malaysia

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INTRODUCTION

Paddy is widely planted in Malaysia. Almost 97 percent of the total cultivated agricultural land in Malaysia is covered by paddy, together with other crops (1). Paddy plays a vital part in Malaysia's economy and diet as rice is a staple food for Malaysians. Rapid growing population has increased the demand for rice consumption. In order to fulfill this demand, Malaysia needs to sustain its production and increase the rice productivity. To sustain high productivity of crops, the application of large quantities of fertilizers became an important component. Contemporary farming with aimless utilization of agrochemicals such as pesticides and fertilizers, along with tractor development, for greater yield profitability has contaminated the environment with heavy metals (2-3).

Almost all types of water contain heavy metals from the natural occurrence or earth's surface (4). Large amount of water is supplied to the crops making this activity one of the concerns (5). The application of fertilizers by farmers, especially chemical fertilizers such as compound fertilizers and urea fertilizers, has contributed to higher contamination of the metals in the paddy water and soil (6). Heavy metals contamination in agriculture fields is commonly related to fertilizers used, as some fertilizers contain certain heavy metals (7).

Occupations such as paddy farmers are at risk since they are involved in wet work (8). Skin diseases among paddy farmers were found at a high prevalence in southern India through a study by Shrutakirthi, Suraj, Sugandhi, Girija and Sreekumaran (9). In addition to this, a study on skin diseases among farmers who use wastewater in paddy field in Nam Dinh, Vietnam found that agricultural work increased the risk of skin diseases, as farmers were required to have frequent contact with wastewater and fertilizers (8).

Farmers who do not wear appropriate and full personal protective equipment (PPE) while working may be exposed to heavy metals in paddy soil and water . Thus, it is important to assess the health risk that is imposed on the farmers by the exposure. This study was designed to quantify the concentration of selected heavy metals (Cu, Cr, Zn, Ni, Pb, As and Cd) in paddy water and soil, and assess their potential dermal health risk to the farmers at Tanjung Karang, Kuala Selangor.

MATERIALS AND METHODS

Sample population

117 farmers in Kampung Sawah Sempadan were randomly recruited as respondents according to the two following inclusion criteria: i) paddy farmers within the age group of 18 to 70 years old, ii) paddy farmers who work directly in the paddy water and soil as part of their job task. The specified age group was selected as to include the adult population (18 to 60 years old), however in order to achieve the desired sample size, the age limit was extended to 70 years old because there were farmers who were more than 60 years old and still working on paddy farming. The second inclusion criterion was set to ensure that the respondents were in contact with paddy water and soil to calculate health risk appropriately.

Sample collection

This study was conducted at Kampung Sawah Sempadan,Tanjung Karang, Selangor, which is the third largest paddy field in Peninsular Malaysia and is widely known for its paddy cultivation activity. The study location was selected based on the following criteria: i) agriculture area with paddy cultivation as the main activity with no industrial activities near the study area ii) co-operation of respondents throughout the study, iii) short distance from the laboratory which permits effective transportation of samples.

72 water samples and 72 soil samples were randomly collected from all 24 paddy blocks at Kampung Sawah Sempadan (Block A to Block X) (Fig.1). High density polyethylene (HDPE) bottles were used to collect the water samples while topsoil samples were collected in plastic zip-locked bags. All samples were transported in ice to the laboratory immediately.

Water samples were acidified with nitric acid (HNO3) (Fisher Scientific, USA) to a pH of 2 and filtered through a 0.45-µm membrane filter paper (Millipore, MA). Water samples were kept at 4°C and analysed as soon as possible.

Soil samples were sieved (<2 mm) and homogenised. The aliquot was stored at -20 °C until they were analysed. The soil was extracted using Aqua-Regia digestion method according to Radojevic and Bashkin (2006) (10). Briefly, 130 mL of HCl (R&M Marketing, Essex, U.K) and 120 mL ultrapure water were added to prepare the Aqua-Regia. Then, 150 mL of the solution was added to 50 mL HNO3 (R&M Marketing, Essex, U.K) and mixed well. Kjeldahl flask was used to weigh one gram of soil sample and 5 mL of Aqua-Regia was added. The sample was left overnight. The flask was heated at 50 °C for 30 min on heating block. The temperature was increased to 120 °C and the heating process continued for 2 hours. 10 mL of 0.25 M HNO3 was added after the extract was cooled. The extract was filtered through 0.45-µm membrane filter paper.

Heavy metal analysis

Soil and water extracts were injected into ICP-MS (Perkin Elmer Sciex Elan 9000, USA) to quantify the concentration of Cu, Cr, Zn, Ni, Pb, As and Cd. ICP-MS was calibrated using heavy metal standards at concentrations 1, 5, 10, 25 and 50 ng/mL.

Quality control

Sample blanks were prepared for each batch of analysis throughout the whole sample extraction and analysis process (11). Ultrapure water (Milli-Q® Advantage A10 Water Purification System, France) and soil standard reference matrix (2711a Montana II soil) were used as sample blank. The blank was prepared using similar

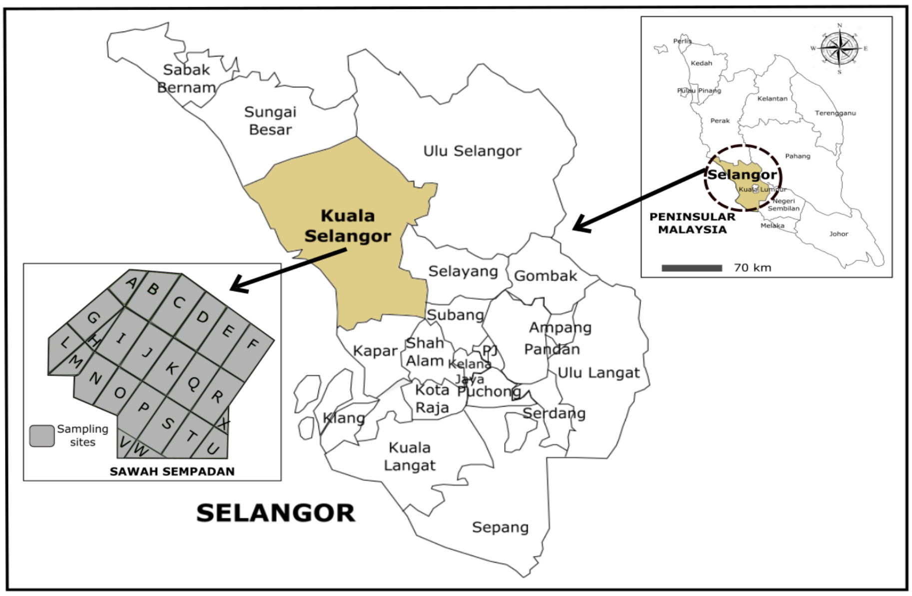

Figure 1: Sampling locations in the paddy field (Block A to X) at Kampung Sawah Sempadan, Kuala Selangor

method in the sample preparation.

Three replicates of sample blank were spiked with 25 µg/L heavy metals standard. The concentration of heavy metals spiked before sample extraction and the concentration spiked after extraction in the blank matrix were compared to calculate the percentage of recovery (11). Blank solutions were spiked with heavy metal standards to quantify the instrumental limit of detection (LOD) and limit of quantification (LOQ). LOD and LOQ were calculated from the standard deviation (σ) of ten times analyses of these determinations (LOD = 3.σ and LOQ = 10.σ) (12-13).

The correlation coefficients (R^2), average extraction recovery for soil and water, LOD and LOQ for each heavy metal are shown in Table I.

Table I: Correlation coefficients (R^2), average extraction recovery for soil and water, LOD and LOQ for each heavy metal.

Compounds	R^2	Recovery (%)		LOD (ng/L)	LOQ (ng/L)
		Soil	Water		
Cu	0.9998	88.2	98.7	5	15
Cr	0.9991	89.1	94.9	4	15
Zn	0.9998	81.3	81.5	11	30
Ni	0.9999	89.9	98.5	10	30
Pb	0.9980	86.6	99.4	2	5
As	0.9999	88.7	93.8	8	20
Cd	0.9999	82.7	96.6	3	9

Health risk assessment

All respondents were interviewed using the questionnaire which was constructed according to Nordic Occupational Skin Questionnaire (NOSQ) (14) and the study by Taneepanichskul et al. (15) as references. The questionnaire comprised of the following three sections: section A (personal information of the respondents), section B (exposure frequency and exposure duration to the heavy metals), and section C (personal hygiene and the use of PPE).

The non-carcinogenic health risk was calculated using HQ using the formula provided by USEPA (16) (Eq. 1). Hazard index (HI) was calculated from the total of HQs to estimate the risk of mixed metal exposures (11).

$$HQ = \frac{DAD}{RfD} \quad \text{(Eq. 1)}$$

Where DAD is dermal absorbed dose (mg/kg/day) and RfD is reference dose (mg/kg/day). The values for dermal RfD were adapted from the Risk Assessment Information System (RAIS). The values of RfD are Cu: 1.2×10^{-2} mg/kg/day (17); Cr: 7.5×10^{-3} mg/kg/day (18); Zn: 6.0×10^{-2} mg/kg/day (19); Ni: 5.4×10^{-3} mg/kg/day (20); Pb: not available (21); As: 1.2×10^{-4} mg/kg/day (22); Cd: 5.0×10^{-6} mg/kg/day (23). The EPA has not developed an RfD for lead because it appears that lead is a non-threshold toxicant, and it is not appropriate to develop RfDs for these types of toxicants (21).

DAD is defined by the following equation (16):

$$DAD = \frac{DA_{event} \times EV \times ED \times EF \times SA}{BW \times AT} \quad \text{(Eq. 2)}$$

Where DA_{event} is absorbed dose per event (mg/cm^2-event), SA is skin surface area available for contact (cm^2), EV is event frequency (events/day), EF is exposure frequency (days/year, ED is exposure duration (years), BW is body weight (kg), and AT is averaging time (days) (Table II).

Table II: Information on farmer's exposure to paddy soil and water.

Parameters	Average
Event frequency (events/day)	1[a]
Exposure duration (years)	26[a]
Exposure frequency (days/year)	220[a]
Body weight (kg)	70.3[a]
Averaging time (days)	5659.5[a]
Skin surface area (hands) for adult male (cm^2)	990[b]
Skin surface area (feet) for adult male (cm^2)	1310[b]
Skin surface area (hands and feet) for adult male (cm^2)	2300[b]

[a]Information obtained from questionnaires administered to the respondents
[b]Adapted from USEPA (49)

DA_{event} for water samples is defined by the following equation (16):

$$DA_{event} = K_p \times C_w \times t_{event} \quad \text{(Eq. 3)}$$

Where K_p is dermal permeability coefficient of compound (Cu:1.0×10^{-3} cm/hr; Cr: 1.14×10^{-3} cm/hr; Zn: 3.19×10^{-4} cm/hr; Ni: 3.06×10^{-4} cm/hr; Pb: 1.0×10^{-4} cm/hr; As: 1.62×10^{-3} cm/hr; Cd: 3.29×10^{-4} cm/hr) (16)(49), C_w is concentration of heavy metals in water (mg/cm^3), t_{event} is hour of contact to the heavy metals in paddy water per event (average of 2 hours obtained from questionnaire).

DA_{event} for soil samples was calculated by following equation (16):

$$DA_{event} = C_s \times CF \times AF \times ABS_d \quad \text{(Eq. 4)}$$

Where DA_{event} is absorbed dose per event (mg/cm^3-event), C_s is concentration of heavy metals in soil (mg/kg), CF is conversion factor (10^{-6} kg/mg), AF is adherence factor of soil to skin (mg/cm2-event)(farmers: 0.1) (16), and ABS_d is dermal absorption fraction (As: 0.03; Cd: 0.001) (16) (other metals: 0.1) (24).

Dermal cancer risks for soil and water were estimated using lifetime cancer risk (LCR) as follows:

$$LCR = DAD \times SF \quad \text{(Eq. 5)}$$

Where DAD is dermal absorbed dose (mg/kg/day) and SF is absorbed cancer slope factor (mg/kg/day) (As: 3.66 mg/kg/day) (22).

RESULTS

Information of respondents

Socio-demographic information of respondents is summarised in Table III.

Personal hygiene and personal protective equipment (PPE)

The result on the personal hygiene and the use of PPE among respondents is presented in Table IV.

81.2% of the farmers took a shower after farming as a routine of personal hygiene. 95.7% of the respondents changed their clothes after farming. Out of 117 respondents, 43 respondents (36.8%) used full set of PPE which consisted of boots and gloves, while the other 74 respondents (63.2%) did not wear proper PPE.

Table III: Socio-demographic Information of Respondents (n=117)

Variables	Mean±SD	Minimum	Maximum
Age (years)	49.6±12.02	24	70
Weight (kg)	70.37±12.79	45	120
Height (m)	1.63±0.05	1.52	1.74
BMI (kg/m^2)	26.31±4.78	17.01	39.64
Variables	**Category**	**Frequency**	**Percentage (%)**
Gender	Male	117	100
Race	Malay	117	100

Table IV: Personal hygiene and use of personal protective equipment (PPE) among respondents

Variables	Frequency (n=117)	Percentage (%)
Wash up/Shower after farming	95	81.2
Change clothes after farming	112	95.7
Use full PPE (boots and gloves)	43	36.8

Concentration of heavy metals in paddy water

The selected heavy metal concentrations in paddy water samples are summarised in Table V. Arsenic (As) has the highest mean concentration ($1.00x10^{-2}$ mg/L), followed by Zn ($8.40x10^{-3}$ mg/L), Cr ($4.50x10^{-3}$ mg/L), Ni ($2.60x10^{-3}$ mg/L), Cu ($2.40x10^{-3}$ mg/L), Pb ($1.60x10^{-3}$ mg/L) and Cd ($2.20x10^{-5}$ mg/L). None of the heavy metal elements exceeded the maximum recommended value for irrigation water by Malaysia is National Water Quality Standard (NWQS) (25) and by Food and Agriculture Organization (FAO) (26).

Concentration of heavy metals in paddy soil

The concentrations of heavy metals in soil samples are summarised in Table V. As has the highest mean concentration ($1.71x10^{1}$mg/kg), following the list are Cr (1.48×10^{1}mg/kg), Zn (7.21 mg/kg), Pb (6.64 mg/k), Cu (3.87 mg/kg), Ni (1.82 mg/kg), and Cd (6.00×10^{-2}mg/kg). The concentration of heavy metals in this study did not exceed Dutch target value for soil protection (27) except for the maximum concentration of As (Table V).

Health Risk Assessment

The health risks were categorised into non-carcinogenic and carcinogenic by calculating HQ and LCR respectively. The hazard index (HI) was calculated from HQs to estimate the risk of mix metal exposures (11). The HQ and HI and LCR are listed in the Table VI.

The HQs calculated for feet exposure, hands exposure and for both hands and feet exposures were less than 1 for each heavy metal when exposed in paddy soil and water. For carcinogenic health risk, only LCR of As was reported in this study due to the availability of its dermal cancer slope factor. The LCRs were in the range of $1.67x10^{-6}$ to $1.00x10^{-5}$.

Table V: Heavy metal concentrations in paddy water and soil and comparison with available standards (n=72)

	Cu		Cr		Zn		Ni		Pb		As		Cd	
	Water (mg/L)	Soil (mg/kg)	Water (mg/L)	Soil (mg/kg)	Water (mg/L)	Soil (mg/kg)	Water (mg/L)	Soil (mg/kg)	Water (mg/L)	Soil (mg/kg)	Water (mg/L)	Soil (mg/kg)	Water (mg/L)	Soil (mg/kg)
Mean	$2.40x10^{-3}$	3.87	$4.50x10^{-3}$	$1.48x10^{1}$	$8.40x10^{-3}$	7.21	$2.60x10^{-3}$	1.82	$1.60x10^{-3}$	6.64	$1.00x10^{-2}$	$1.71x10^{1}$	$2.20x10^{-5}$	$6.00x10^{-2}$
SD	$1.20x10^{-3}$	1.92	$1.80x10^{-3}$	6.61	$4.40x10^{-3}$	3.17	$1.20x10^{-3}$	$7.40x10^{-1}$	$9.00x10^{-4}$	2.70	$7.50x10^{-3}$	7.11	$1.40x10^{-5}$	$4.00x10^{-2}$
Median	$2.10x10^{-3}$	3.51	$4.30x10^{-3}$	$1.45x10^{1}$	$8.10x10^{-3}$	6.72	$2.40x10^{-3}$	1.65	$1.50x10^{-3}$	6.04	$8.50x10^{-3}$	$1.79x10^{1}$	$1.90x10^{-5}$	$5.00x10^{-2}$
Minimum	$1.10x10^{-3}$	$7.70x10^{-1}$	$1.10x10^{-3}$	3.90	$1.10x10^{-3}$	1.91	$9.00x10^{-4}$	$8.80x10^{-1}$	$7.00x10^{-4}$	2.31	$1.30x10^{-3}$	4.42	ND	$2.00x10^{-2}$
Maximum	$6.30x10^{-3}$	7.98	$9.50x10^{-3}$	$3.67x10^{1}$	$2.21x10^{-3}$	$2.09x10^{1}$	$7.50x10^{-3}$	4.63	$6.40x10^{-3}$	$1.60x10^{1}$	$3.34x10^{-3}$	$3.14x10^{1}$	$1.03x10^{-4}$	$2.90x10^{-1}$
Water standards	$2.00x10^{-1}$		$1.00x10^{-1ab}$		2.0^{ab}		$2.00x10^{-1ab}$		5.0^{b}		$1.00x10^{-1ab}$		$1.00x10^{-2ab}$	
Soil standard		$3.60x10^{1c}$		$1.00x10^{2c}$		$1.40x10^{2c}$		$3.50x10^{1c}$		$8.50x10^{1c}$		$2.90x10^{1c}$		$8.00x10^{-1c}$

ND = Not detected

[a] Recommended concentration of heavy metals in irrigation water (Class IV) by Malaysia National Water Quality Standard (25)

[b] Recommended maximum concentration of heavy metals in irrigation water by Food and Agriculture Organization (FAO) (26)

[c] Recommended concentration of heavy metals in soil by Netherlands for soil protection - Dutch target value (27)

Table VI: Hazard quotient (HQ), hazard index (HI) and lifetime cancer risk (LCR) of heavy metals for farmers.

Heavy Metals	Hand exposure			Feet exposure			Hand and feet exposure		
	Soil	Water	Soil & water	Soil	Water	Soil & water	Soil	Water	Soil & water
					HQ				
Cr	2.78x10^{-4}	1.93x10^{-5}	2.97x10^{-4}	3.68x10^{-4}	2.55x10^{-5}	3.93x10^{-4}	6.46x10^{-4}	4.48x10^{-5}	6.90x10^{-4}
Ni	4.75x10^{-5}	4.15x10^{-6}	5.16x10^{-5}	6.28x10^{-5}	5.49x10^{-6}	6.83x10^{-5}	1.10x10^{-4}	9.64x10^{-6}	1.20x10^{-4}
Cu	4.54x10^{-5}	5.63x10^{-6}	5.10x10^{-5}	6.01x10^{-5}	7.45x10^{-6}	6.75x10^{-5}	1.06x10^{-4}	1.31x10^{-5}	1.19x10^{-4}
Zn	1.69x10^{-5}	1.26x10^{-6}	1.82x10^{-5}	2.24x10^{-5}	1.66x10^{-6}	2.41x10^{-5}	3.93x10^{-5}	2.92x10^{-6}	4.22x10^{-5}
As	6.03x10^{-3}	3.80x10^{-3}	9.83x10^{-3}	7.98x10^{-3}	5.03x10^{-3}	1.30x10^{-2}	3.93x10^{-5}	8.83x10^{-3}	8.87x10^{-3}
Cd	1.69x10^{-5}	4.08x10^{-5}	5.77x10^{-5}	2.24x10^{-5}	5.40x10^{-5}	7.64Ч10^{-5}	1.40x10^{-2}	9.47x10^{-5}	1.41x10^{-2}
					HI				
	6.43x10^{-3}	3.87x10^{-3}	1.03x10^{-2}	8.52x10^{-3}	5.12x10^{-3}	1.36x10^{-2}	1.49x10^{-2}	9.00x10^{-3}	2.39x10^{-2}
					LCR				
As	2.65x10^{-6}	1.67x10^{-6}	4.32x10^{-6}	3.50x10^{-6}	2.21x10^{-6}	5.71x10^{-6}	6.15x10^{-6}	3.88x10^{-6}	1.00x10^{-5}

DISCUSSION

There are few studies which reported higher heavy metals concentrations than this study. Reddy et al. (6) found that Cd in paddy water ranged from 1.4 to 5.8 mg/L, followed by Zn (1.00x10^{-1} – 2.00x10^{-1} mg/L), Cu (4.00x10^{-2} mg/L) and Pb (1.00x10^{-1} –2.00x10^{-1} mg/L). Assubaie (28) reported the heavy metal concentrations in mixed irrigation at Oasis farms, Saudi Arabia were 1.10x10^{-2} mg/L (Cu), 1.00x10^{-2} mg/L (Zn), 9.00x10^{-3} mg/L (Cd) and 5.00x10^{-3} mg/L (Pb), respectively. Another study from Tanzania testified the concentrations of Cr, Pb and Cu ranged from 1.00x10^{-2} to 1.414 mg/L, 1.13x10^{-1} to 8.30x10^{-2} mg/L and 1.30x10^{-2} to 1.60x10^{-2} mg/L, respectively, while the concentration of Cd was below the AAS detection limit (1.00x10^{-2} mg/L) at all sampling points (29).

Nevertheless, our results were comparable with few similar studies, where the heavy metals concentration in agricultural water did not exceed the permissible level. For instance, Bambara et al. (30) reported that the average concentrations of As, Ni, Cr, Zn and Pb in Goudrin irrigation water were less than the FAO recommended limit. Assubaie (28) quantified that the levels of Zn and Pb in the irrigation water were in acceptable range and suitable for irrigation use in Saudi Arabia. Reddy et al. (6) also stated that the concentrations of Cd, Pb, Zn and Cu in paddy water below the permissible limit of Indian standard.

In this study, Cd showed the lowest concentration compared to other metals which between 7.00x10^{-6} and 1.03x10^{-4} mg/L. According to Kahn et al. (31), concentration of Cd could be contributed by various factors such as industrial activity, atmospheric release and organic sediments' deposition. Cd reported in the water near the electroplating plant in China varied from 1.50x10^{-3} mg/L to 2.30x10^{-3} mg/L (32). Therefore, absence of industrial or mining activities near the paddy field at Kampung Sawah Sempadan may suggest the minimum presence of Cd in the paddy water.

Elevation of heavy metal pollution in agriculture is often reported with wastewater to irrigate the crops (33). However, the use of municipal wastewater to irrigate the agricultural field is not practiced commonly in Malaysia. According to Amin et al. (34), the water used for irrigation in the paddy field in Kampung Sawah Sempadan is from Tengi River, and the upstream is Bernam Head River. It is the only source for irrigation supply in this area. The irrigation scheme is a run of the river without reservoir and the paddy plots receive water directly from eight tertiary canals. This may suggest the possible reason of the low concentration of heavy metals is because the water used to irrigate the paddy field is from the upstream of Bernam Head River, where minimal anthropogenic activities were expected. Currently, Kampung Sawah Sempadan is mainly active in paddy cultivation activity and there are no industrial or mining activities around the area.

For soil contamination, there were a few studies reported higher readings of heavy metals compared to our results. The mean concentrations of heavy metals found in paddy soil of China were 3.40x10^{-1} mg/kg (Cd), 5.71x 10^{1} mg/kg (Cr), 2.07x10^{1} mg/kg (Cu), 1.70x10^{1} mg/kg (Ni), 3.51x10^{1} mg/kg (Pb) and 6.11x10^{1} mg/kg (Zn), respectively (35). Agriculture soil in Iran was found to be contaminated with Pb (2.99x10^{1} mg/kg), Zn (1.98 x10^{1} mg/kg), Ni (1.85x10^{1} mg/kg) and Cu (7.26 mg/kg) (36). Contrarily, the heavy metal contamination in paddy soils of India were lower compared to this study except for Cd where the concentrations of Cd, Zn and Cu were ranged between 8.0 to 9.3 mg/kg, 1.4 to 5.0 mg/kg and $4.00x10^{-1}$ to $5.00x10^{-1}$ mg/kg, respectively (6).

The mean concentration of As in soil for this study ($1.71x10^{1}$ mg/kg) has exceeded the recommended value by Dutch Target Value for soil protection while concentration of As in water sample ($1.00x10^{-2}$ mg/L) was within the permissible limit by NWQS and FAO. The result is comparable with other studies which measured the concentration of As in both soil and water samples. Islam et al. (39) reported As concentrations in 100 irrigation water with mean of $7.50x10^{-2}$ mg/L and soil samples contained mean concentration of As at $1.12x10^{1}$ mg/kg in Chapai Nawabganj (Bangladesh). Similarly, Otero et al. (40) found a higher concentration of As in soil samples (4.48 mg/kg) compared to water (generally < $1.00x10^{-2}$ mg/L).

Previous study in Beijing (China) reported the contents of fertilizers and pesticides could accumulate heavy metals in soil (37). Malidareh et al. (38) also showed that fertilization is responsible for the increase of average concentration of As. The average As concentrations of soil samples in Ghaemshahr, Iran elevated from $3.00x10^{-3}$ mg/kg to $2.25x10^{-1}$ mg/kg after fertilization (38). This shows that the fertilizers introduced to soils could be a possible source of As pollution. Since the soils in this study were collected from agriculture area which also exposed to fertilizers, hence explaining the high readings of As in soil samples.

Cd has the lowest concentration in this study among the selected heavy metals which was between $2.00x10^{-2}$ and $2.90x10^{-1}$ mg/kg. Similarly, Aziz et al. (41) reported that Cd has the lowest concentrations in paddy soil in Ranau Valley, Sabah, Malaysia, as compared to As, Co, Cr, Cu Ni and Zn. Khairiah et al. (42) indicated in their study the low contamination of Cd in paddy soil was originated from natural soil deposits.

For health risk assessment, three different situations were considered to estimate the health risk of farmers. The first situation was that the farmers wore gloves but did not wear boots, hence exposing the feet to the paddy soil and water. Second situation was that the farmers wore boots but did not wear gloves and thus exposing the hands. Meanwhile, the third situation was the farmers did not wear both gloves and boots and consequently exposing their hands and feet in soil and water.

Among the three situations, farmers with both hands and feet exposure to heavy metals recorded the highest HI, followed by feet exposure and hand exposure respectively (Table IV). The reason for different HI calculated for different area of exposure can be relate to the surface area of exposure, for example combination of hands and feet have larger surface area than feet only and hands only. However, overall there was no significant chronic non-carcinogenic health risk to the farmers working in the paddy soil and water for the 26 years while being exposed to a mixture of heavy metals (HI<1) for all situations. The health risk of Pb was not assessed in this study due to the unavailability of RfD in RAIS.

The carcinogenic health risk was only assessed for exposure to As because USEPA do not list the dermal cancer slope factors for Cd, Cr, Cu Ni and Pb although they are identified as potential carcinogens. Thus, the carcinogenic dermal health risk of farmers due to the exposure of these heavy metals was not assessed. The LCRs calculated for As were $1.67x10^{-6}$ to $1.00x10^{-5}$ which was in the acceptable risk level of 10-4 to 10-6, indicating the carcinogenic health risk was clearly acceptable. Another study reported the dermal LCR of As was $4.27x10^{-6}$ for the exposure of As in the water from natural environments as well as mining areas in Ghana while HQ for Cr and Cd from the mining and the pristine sites were less than 1 (43).

To the best of the authors' knowledge, there were no studies reported on the chronic non-carcinogenic and carcinogenic health risk of farmers related to occupational exposure in the agricultural field. Recent studies reported the health risk of heavy metals in water samples (44-45), soil samples (45), and vegetable samples (46-48) among the general population. Hence, the result in this study was not compared with previous studies.

CONCLUSIONS

Overall, the contamination of the selected heavy metals in the paddy soil and water at Kampung Sawah Sempadan were lower than the available standards in spite of As exceeding the recommended value by Dutch target value. The mean concentrations of heavy metals in soil samples from highest to lowest were $1.71x10^{1}$ mg/kg (As) > $1.48x10^{1}$ mg/kg (Cr) > 7.21 mg/kg (Zn) > 6.64 mg/kg (Pb) > 3.37 mg/kg (Cu) > 1.82 mg/kg (Ni) > 6.00 $x10^{-2}$ mg/kg (Cd). Meanwhile, the mean concentrations of heavy metals in water samples in decreasing order were $1.00x10^{-2}$ mg/L (As) > $8.4x10^{-3}$ mg/L (Zn) > $4.5x10^{-3}$ mg/L (Cr) > $2.60x10^{-3}$ mg/L (Ni) > $2.40x10^{-3}$ mg/L (Cu) > $1.60x10^{-3}$ mg/L (Pb) > $2.20x10^{-5}$ mg/L (Cd). The HQ and HI for all selected heavy metals were below 1, indicating that non-carcinogenic dermal health risk of farmers working in paddy soil and water for 26 years was not significant. The LCR for As was $1.00x10^{-5}$ for the exposure of both hands and feet in the paddy soil and water, indicating the risk level for the farmers was within the acceptable range. There were several limitations of this study that should be considered to improve on further research. The sources of heavy metals contamination in the paddy water and soil were not well considered and scientifically tested thus it cannot be certain that the heavy metals pollution are caused by the use of agrochemicals such as fertilizers or pesticides. The concentrations of heavy metals detected were from the environmental samples (water and soils) and not from human samples, thus the biomarker of exposure was not measured. The health risks calculated were only the possibility of exposure and not the actual representation of exposure. In addition, some of the compounds studied do not have cancer slope factor and reference dose, so their respective cancer risk and hazard quotient cannot be determined.

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