Potential Health Risk Assessment for the Occurrence of Heavy Metals in Rice field Influenced by Landfill Activity in Can Tho City, Vietnam

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Abstract— The study was conducted to assess potential risk of heavy metals in the soil and rice plants in the ricefield around the landfill in Dong Thang commune, Co Do district, Can Tho city, Vietnam. Four soil samples in which three samples were collected around the landfill and one sample was collected one km away from the landfill for the analysis of heavy metals including Mn, Zn, Cu, Cr, Ni, Pb and Cd. Rice samples were collected during ripening stage (few days before the harvest) at the same locations with the soil sampling, for the same heavy metal species analysis. The findings revealed that six out of seven heavy metals occurred in the soil. The decreasing order of theheavy metals concentrations in the soil samples was Mn > Zn > Ni > Cr > Cu > Pb. This study found that accumulation of heavy metals in parts of rice at S1-S3 was higher that those at S4 (except for Zn and Pb at rice roots) and decreased in the order Mn > Zn > Cu > Ni. Cr (except in rice grain, Cr> Cu> Ni). Heavy metalsgenerally in the rice partswere in the magnitude order of root > stem-leave> grain. The calculated hazard index (HI) indicated that the accumulation of heavy metals in soil and rice grain is not likely to pose a threat to public health (HI <1), however, potential health and ecological risk may still exist. Measures should be taken to prevent landfill leachate leaching into the agricultural areas to minimize potential environmental and health risks.

Keywords— Landfill; leachate; heavy metals; health risk; ricefield.

I. INTRODUCTION

Vietnam has recently been facing serious environmental pollution from solid wastes as the amounts of generated wasteshave been increasing in both quantity and toxic level. According to the National Environmental Report 2011-2015 (MONRE, 2015), the total amount of urban domestic solid waste generated in the country was 32,000 tons in 2014. The amount of solid waste generated in the Mekong Delta region accounted for 5% of the generation of the whole country. Can Tho city is generatingsolid wastes of approximate 893 tons day-¹(People's Committee of Can Tho City, 2015). Solid wastes have been collected and treated at landfills. However, landfills have also been identified as a cause of soil and groundwater pollution(Fatta et al., 1999). According to MONRE (2015), only 203 out of 660 landfills across the country are sanitary landfills, and the remaining were unsanitary. However, the majority of landfills have been overloaded, exacerbating the environmental impacts, which has led to increasingly serious and complex pollution problem in the landfilling areas.

The landfill at Dong Thang Commune, Co Do District, Can Tho City, Vietnam is in a state of serious overload due to receiving a fairly large amount of waste approximate 370 tons per day⁻¹ from several districts of Can Tho city. The untreated leachate has significantly affected water quality, soil and rice yield in the land adjacent to the landfill (**Nhien and Giao**, 2019). Leachate not only contains high levels of organic matter, nitrogen but also significant concentrations of heavy metals, so it may cause pollution of soil and surface water (**Nhien and Giao**, 2019). Several studies have also shown that heavy metals are often found in high concentrations in and around landfills all over the world (**Alam et al., 2012; Nava-Martinez et al., 2012; Ajah et al., 2015**). In addition, heavy metals could potentially present in paddy fields due to impurities of chemical fertilizers and pesticides (Liu et al., 2003; Kingsawat and Roachanakanan, 2011). Therefore, heavy metal contamination is always a major focus in several environmental studiessince it could bioaccumulate in microorganisms and then transfer into food chains, for example, from plant to animal and to human being(Munees and Abdul, 2012; Klinsawathom et al., 2017). The former study pointed out that heavy metals could move from soil and water to plants' tissues via uptakingby roots (Kingsawat and Roachanakanan, 2011), posingpotential risks for human health and ecosystems(Satachon et al., 2019).Currently, several studies reported on the quality of water and soil at the (Kanmani landfill and surrounding areas and Gandhimathi, 2013; Huang et al., 2013; Nhien and Giao, 2019) but very few studies have been carried out on assessment of potential risk resulting from exposure to heavy metals in rice grains and soil. This study was implemented to examine the potential risk for the presence of heavy metals in soil and rice parts around Dong Thang landfill, Co Do district, Can Tho city, Vietnam. The findings from this study could provide useful information for local authorities for managing risk resulting from heavy metal occurrence.

II. MATERIALS AND METHODS

1) Soil sampling and analysis

Soil samples were collected at the depth of 0-25 cm at 4 locations, of which 3 locations in the rice field surrounding the landfill (namely S1, S2, S3) and 1 locationin the ricefield1km away from the landfill (namely S4) (Figure 1). After the collection, the soil samples were dried at room temperature, pulverized and sieved through mesh with the pore size of 0.5 mm for heavy metal analysis. The pulverized soil sample (0.5g) was digested using a microwave digester (Microwave digester, Milestone, Ethos) using the method of the United State Environmental Protection Agency (EPA3051) by adding 10 ml of 65% nitric acid and operated at 1,000 watts of power, temperature of 175°C for 15 minutes 30 seconds. Heavy metals including Cd, Cr, Cu, Fe, Ni, Mn, Pb and Zn were determined by atomic absorption spectrometry (AAS, Agilent, AA240). All glasswares used in heavy metal analysis were cleaned washed using 0.1 M nitric acid for 24 hours and then rinsed with distilled water. Analysis of heavy metals was performed in triplicates.



Fig.1: Soil sampling locations.

2) Rice sample collection and analysis for heavy metals

Rice samples were collected during ripening stage (few days before the harvest) at the same locations with the soil sampling (Figure 1). Five whole rice plants were carefully removed from soil at five positions in an area of 1 m^2 for every sampling location. The collected rice plants were divided into three parts including the root, stem and leave, and grain. The separated parts of the rice plants at three locations surrounding landfill (S1, S2, and S3) were pooled to reduce the analysis cost due to limited budget. The heavy metals including Cd, Cr, Cu, Ni, Mn, Pb and Zn were analyzed in the rice parts. The procedure for analyzing heavy metals in rice samples was performed in the similar manner to that for analyzing soil samples.

3) Risk assessment

Hazard index (HI) due to heavy metals in soil and rice (grain) was assessed according to **Hang** *et al.* (2009) and Ferreira- Baptista and de Miguel (2005). Heavy metal enters the body daily (D) through the three main routes including ingestion (D_{ing}), inhalation (D_{inh}), and dermal contact (D_{der}). Heavy metalsin rice grain only enter the body through ingestion (D_{ing}). Daily consumption levels through different contact routes were calculated based on Equation 2, 3, and 4.

- Direct ingestion rice grain (D_{ing}): $D_{ing} = \frac{C_s x Ing R x EF x ED x CF}{BW x AT}$ (Eq.2)
- Inhalation of suspended particles via mouth and noise (D_{inh}):

$$D_{inh} = \frac{C_s x InhR x EF x ED}{BW x AT x P EF}$$
(Eq.3)

• Dermal contact (D_{der}): $D_{der} = \frac{C_{sxSAxSLxABSxEFxEDxCF}}{BWxAT}$ (Eq.4)

Where: C_s is concentration of pollutants in soil or rice (mg/kg); IngR is the rate of ingestion of pollutants in soil or rice (mg day⁻¹); InhR is inhalation rate of suspended particles in soil (m³day⁻¹); EF and ED are frequency of exposure (dayyear⁻¹) and duration of exposure (years); CF is conversion factor = 1.00E-06 (kg mg⁻¹) and , BW is average body weight (kg); AT is average time of noncarcinogenic (days); PEF is soil-to-air particulate emission factor (m³ kg⁻¹), SL is soil-to-skin adherence factor (mgcm⁻²); SA is skin surface area available for exposure (cm²) and ABS is dermal absorption factor. Detail of these factorfor risk assessment wasindicated in Table 1.

The risk assessment for non-carcinogenic was calculated using Equation 5:

$$HI = \sum_{i=1}^{m} \sum_{j=1}^{n} HQ_{ij} = \sum_{i=1}^{m} \sum_{j=1}^{n} (\frac{D}{RfD})_{ij}$$
(Eq.5)

where m and n are type and number of pollutants; $R_f D$ is reference dose (mg kg day⁻¹) (Table 1); D is daily uptakedose (mg kg⁻¹ day⁻¹); HQ_{ij}is risk for the exposure path. HI <1 means there is no possibility of adverse human health effects, whereas HI> 1 means there is likely to have adverse effect on human health.

Table1 Parameters used for risk assessment (Hang et al., 2009)

Parameters	Adult	Children
IngR		
-Soil (mgday ⁻¹)	100	200
- Rice grain (g day ⁻¹)	389.2	198.4
InhR (m ³ day ⁻¹)	12.8	7.63
EF (day)	350	320
ED (year)	24	6
BW (kg)	59.95	23.9
AT (day)	8,760	2,190
SA (cm ²)	2,145	1,150
SL (mg cm ⁻²)	0.07	0.20
ABS	0.001	0.001
PEF (m ³ kg ⁻¹)	1.36E + 09	1.36E + 09

Table.2: Reference doses of some heavy metalsFerreira-Baptista and de Miguel (2005)

Heavy	RfD (mg kg ⁻¹ day ⁻¹)					
metals	RfDing	RfD _{inh}	RfD _{der}			
Cd	1.00E - 03		1.00E - 05			
Cr	3.00E - 03	2.86E - 05	6.00E - 05			
Cu	4.00E - 02	4.02E 02	1.20E - 02			
Mn	4.60E - 02	1.43E - 05	1.84E - 03			
Ni	2.00E - 02	2.06E - 02	5.40E - 03			
Pb	1.40E - 03	3.52E - 05	5.25E - 05			
Zn	3.00E - 01	3.00E - 01	6.00E - 02			

III. RESULTS AND DISCUSSION

1) Occurrence of heavy metals in soil

Table 3 presented the concentrations of heavy metals in the soil surrounding the landfill. Six out of seven heavy metals occurred in the two soil layers around the

landfill with average concentrations ranging from 12.3 \pm 2.14 - 291 \pm 38.85 mg kg⁻¹. The concentrations of Mn, Zn, Cu, Cr at the locations S1, S2 and S3 were all higher than those at S4 (1 km away from the landfill). Cd was the only metal not detected in all soil samples.

]	Heavy metal	OCVN 03				
Heavy metals	S1	S2 S3 S4		Average (S1, S2, S3)	MT:2015/BT NMT	CCME, 2007	
Mn	321ª±2	240 ^b ±0	315 ^a ±2	234 ^b ±8	291± 38.85	-	-
Zn	78.8 ^b ±0	$82.7^{a}\pm 0.70$	$\begin{array}{c} 65.8^{\rm d} \pm \\ 0.35 \end{array}$	74.7°±0	75.8 ± 7.70	200	200
Cu	20.4 ^a ±0	19.0 ^b ± 0.27	14.7 ^d ± 0.01	17.6°±0	18.1± 2.66	100	63
Cr	$9.66^{\circ} \pm 0.56$	27.5ª± 0.70	$28.3^{a}\pm 0.05$	$11.1^{b} \pm 0.4$	21.8± 9.12	150	64
Ni	34.9 ^b ±0	36.3ª± 0.50	30.5°± 0.25	35.6 ^b ± 0.45	33.9± 2.66	-	50
Pb	14.6ª± 0.03	12.6°± 0.02	$\begin{array}{c} 9.66^{d} \pm \\ 0.08 \end{array}$	13.1 ^b ± 0.50	12.3± 2.14	70	70
Cd	ND	ND	ND	ND	ND	-	3 ^a

Table.3: Heavy metals concentrations in the soilsurrounding the landfill

Notes: Data were presented as Mean \pm SD, n = 3. Different letters ^{a, b, c} indicated statistically significant at significance level 5% (p<0.05).^a **Error! Reference source not found.**; ND: Not detected.

Most of heavy metal concentrations in soil were in compliance with QCVN 03-MT: 2015/BTNMT, CCME (2007). Concentration of Mn was the metal with the highest concentrations in soil ranging from 240 \pm 0 - 321 \pm 2 mg kg⁻¹ (Table 3). Mn concentrations at S1 and S3 were always higher than that at S4 showing the negative impact of the landfill leachate on soil environment. Similar to Mn, Cr concentration at S4 was lower than those at S2 and S3 and this could be because Cr is not directly affected by the landfill leachate. The presence of Cr in soil is a major threat to plants and humans because under appropriate environmental conditions, Cr (III) is easily converted to Cr (VI) - a form always toxic to plants (Ba, 2008). At locations around the landfill sites (except S3), Ni concentration ranging from 30.5 ± 0.25 - 36.3 ± 0.50 mg kg⁻¹. The results of Zn concentration ranged from 65.8 \pm $0.35 - 82.7 \pm 0.70 \text{ mg kg}^{-1}$. The distribution of Ni and Zn concentration at the locations and the soil were mainly influenced by the impact of leachate, mobility of the metals and soil properties. Cu and Pb were presented in soil with relatively low concentration at $16.3 \pm 2.20 - 18.1 \pm 2.66$ mg kg⁻¹ and $11.2 \pm 0.46 - 12.3 \pm 2.14$ mg kg⁻¹, respectively (Table 3). Pb concentration in the soilat S4 (13.1 ± 0.50 mg kg⁻¹) was higher than those at S1-S3 ($9.66 \pm 0.08 - 12.6 \pm 0.02$ mg kg⁻¹). Six out of seven heavy metals occurred in the soil samples collected at the surrounding landfill and 1km away from landfill. The presence of heavy metals not only affects the quality of the soil but also threatens the groundwater and rice production.

2) Heavy metals in rice plant

It was found that six out of seven heavy metals occurred in the rice plant parts including root, stem-leave, and rice grain (Table 4). The Cd concentration was below the detection limit, and below the FAO/WHO regulatory standard (0.2 mg kg⁻¹).Heavy metals were found highly accumulated in the rice roots in this study (Table 4). The concentrations of Mn, Zn, Cu, Pb, Ni and Cr in the rice root at S1-S3 were 674 ± 12.53 mg kg⁻¹, 87.6 ± 0.93 mg

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kg⁻¹, 29.3 \pm 0.20 mg kg⁻¹, 11.7 \pm 0.07 mg kg⁻¹, 16.9 \pm 0.68 mg kg⁻¹ and 10.4 \pm 0.06 mg kg⁻¹, respectively, while these heavy metals at S4 were 403 \pm 6.66 mg kg⁻¹, 104 \pm 2.08 mg kg⁻¹, 28.0 \pm 1.85 mg kg⁻¹, 14.5 \pm 0.80 mg kg⁻¹, 7.95 \pm 0.34 mg kg⁻¹ and 5.04 \pm 0.09 mg kg⁻¹, respectively (Table 4). The results indicated that heavy metal concentrations in rice roots in the area influenced by the landfill leachate were higher than those without influenced by the landfill activity.

The heavy metals including Mn, Zn, Cu, Ni, and Cr at the locations surrounding the landfills (S1-S3) were detected in the stem and leave of the rice plants at the concentrations of $645 \pm 8.72 \text{ mg kg}^{-1}$, $47.6 \pm 1.08 \text{ mg kg}^{-1}$, $5.42 \pm 0.34 \text{ mg kg}^{-1}$, $4.37 \pm 0.16 \text{ mg kg}^{-1}$, $2.30 \pm 0.05 \text{ mg kg}^{-1}$, respectively. There were only Mn, Zn, Ni and Cu found at S4 at the concentrations of $544 \pm 15.87 \text{ mg kg}^{-1}$, $61.5 \pm 0.55 \text{ mg kg}^{-1}$, $2.78 \pm 0.09 \text{ mg kg}^{-1}$ and $1.96 \pm 0.82 \text{ mg kg}^{-1}$, respectively (Table 4).

Sampling	Heerry metals	Concentration of heavy metals (mg kg ⁻¹)					
sites	Heavy metals	Root	Stem - Leave	Grains			
	Mn	403 ^b ±6.66	544 ^a ±15.87	129°±11.59			
S4	Zn	$104^{a}\pm 2.08$	61.5 ^b ±0.55	17.7°±0.82			
	Cu	$28.0^{a} \pm 1.85$	$1.96^{b} \pm 0.82$	1.45 ^b ±0.13			
	Cr	5.04±0.09	ND	0.57 ± 0.01			
	Ni	$7.95^a\pm0.34$	2.78 ^b ±0.09	1.68°±0.30			
	Pb	14.5±0.80	ND	ND			
	Cd	ND	ND	ND			
	Mn	674 ^a ±12.53	645 ^a ±8.72	237 ^b ±21.79			
	Zn	$87.6^{a}\pm0.93$	$47.6^{b} \pm 1.08$	35.8°±0.17			
	Cu	29.3ª±0.20	5.42 ^b ±0.34	4.27°±0.07			
S1-S3	Cr	10.4 ^a ±0.06	2.30°±0.05	5.67 ^b ±0.25			
	Ni	16.9 ^a ±0.68	4.37 ^b ±0.16	4.25 ^b ±0.13			
	Pb	11.7±0.07	ND	ND			
	Cd	ND	ND	ND			

Table.4: Concentrations of heavy metals in rice plants

Notes: Data were presented as Mean \pm SD, n = 3. Different letters ^{*a*, *b*, *c*} indicated statistically significant at significance level 5% (p<0.05). ND: not detected.

The concentrations of Mn, Zn, Cr, Cu and Ni in the rice grains surrounded the landfill were 237 ± 21.79 mg kg⁻¹, 35.8 \pm 0.17 mg kg⁻¹, 5.67 \pm 0.25 mg kg⁻¹, 4.27 \pm 0.07 mg kg⁻¹, 4.25 \pm 0.13 mg kg⁻¹, respectively. At the S4 location, the concentrations of Mn, Zn, Ni, Cu and Cr in the rice grains were $129 \pm 11.59 \text{ mg kg}^{-1}$, $17.7 \pm 0.82 \text{ mg}$ kg⁻¹, 1.68 \pm 0.30 mg kg⁻¹, 1.45 \pm 0.13 mg kg⁻¹ and 0.57 \pm 0.01 mg kg⁻¹, respectively. The average concentration of heavy metals in the rice grains at the locations S1-S3 were significantly higher than those at S4 from 1.49 - 2.94 times. The concentration of Cr in rice grainat S1-S3 (near the landfill)was 10 time higher than that at S4 (1km away from the landfill) could indicate serious impact of landfill leachate on the rice production and pose a threat to rice consumption since Cr is considered carcinogenic metal (Ba, 2008).

Among the heavy metals, Mn was highly accumulated in rice plants that could be due to its higher mobility compared to the others (**Prechthai et al., 2008**). This study found that accumulation of heavy metals in parts of rice at S1-S3 was higher that those at S4 (except

for Zn and Pb at rice roots) and decreased in the order Mn> Zn> Cu> Ni> Cr (except in rice grain, Cr> Cu> Ni). Heavy metals accumulated in rice parts with decreasing order root> stem - leave>grain (except for Mn at S4 and Cr at S1 - S3).

3) Health risk assessment

The mean concentration of heavy metals found in soils and rice grainswere used to calculate health risk and the results were showed in Table 5.

Health risk assessment was performed for heavy metals contaminated in soil and rice grains. The result indicated that there is no health risk for children and adults since all the hazard indexes (HI) were less than 1. Children were likely to suffer more risk than adults because the HI values for children were higher than adults (Table 5). Previous studies also indicated that there was no possible risk for human when exposed to soil and rice grains contaminated with heavy metals surrounding the landfill [13, 20]. It was clearly showed that HI values calculated for the heavy metals in the area surrounding the landfill (S1-S3) were higher than those calculated for heavy metals at the location S4 (Table 5). This could mean that higher health risk was expected for the area around the landfill. In the soil sample, the level of health risk (for adult) gradually decreases via $HQ_{ing}>HQ_{inh}>HQ_{der}$ routes; However, the potential health risk for children via ingestion (HQ_{der}) was higher that that via inhalation (HQ_{inh}). Among the heavy metals, Pb was the metal could pose the highest health risk, although this is the metal present with the lowest concentration in the soil. The health risk levels of the examined heavy metals were arranged as decreasing order Pb> Mn> Cr> Ni> Cu > Zn. In the rice grains, the

estimated potential health risk of the heavy metals were in the order of Mn> Cr> Ni> Zn> Cu. Comparing the values of HI between soil and rice grains in both child and adult, it could be seen that the risk for rice grain consumption was higher than the human exposed to the soil contaminated heavy metals. This study suggested that the agricultural activity, especially rice cultivation in the area surrounding the landfill is no longer suitable because the soil was contaminated with heavy metals and the heavy metals started to be accumulated in the rice parts. Long-term consumption of agricultural products produced in the study area could lead to potential health risk.

Table.5: Health risk assessmen	t for	exposure to	heavy metals	contaminated	soil	and	rice	grain
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Heavy metals	Cr	Cu	Mn	Ni	Pb	Zn	Total		
For soil sample at S4									
Adult									
HQ_{ing}	5.92E-03	7.05E-04	8.14E-03	2.84E-03	1.50E-02	3.98E-04	3.30E-02		
HQ_{inh}	5.84E-05	6.60E-08	2.46E-03	2.60E-07	5.62E-07	3.75E-08	2.52E-03		
HQ _{der}	4.44E-04	3.53E-06	3.05E-04	1.58E-05	6.01E-04	2.99E-06	1.37E-03		
HI	6.42E-03	7.09E-04	1.09E-02	2.86E-03	1.56E-02	4.01E-04	3.69E-02		
			Chil	dren					
HQ _{ing}	2.71E-02	3.23E-03	3.73E-02	1.30E-02	6.89E-02	1.83E-03	1.51E-01		
HQ_{inh}	8.79E-05	9.93E-08	3.70E-03	3.91E-07	8.45E-07	5.64E-08	3.79E-03		
HQ _{der}	1.56E-03	1.24E-05	1.07E-03	5.56E-05	2.11E-03	1.05E-05	4.82E-03		
HI	2.88E-02	3.24E-03	4.21E-02	1.31E-02	7.08E-02	1.84E-03	1.60E-01		
			For soil sam	ples at S1-S3					
			Ad	lult					
HQ_{ing}	1.16E-02	7.22E-04	1.01E-02	2.71E-03	1.40E-02	4.04E-04	3.96E-02		
HQ_{inh}	1.15E-04	6.76E-08	3.07E-03	2.48E-07	5.25E-07	3.80E-08	3.19E-03		
HQ_{der}	8.73E-04	3.61E-06	3.81E-04	1.51E-05	5.61E-04	3.03E-06	1.84E-03		
HI	1.26E-02	7.25E-04	1.36E-02	2.73E-03	1.46E-02	4.07E-04	4.46E-02		
			Chil	dren					
HQ_{ing}	5.33E-02	3.31E-03	4.65E-02	1.24E-02	6.43E-02	1.85E-03	1.82E-01		
HQ_{inh}	1.73E-04	1.02E-07	4.62E-03	3.72E-07	7.89E-07	5.72E-08	4.79E-03		
HQ _{der}	3.07E-03	1.27E-05	1.33E-03	5.30E-05	1.98E-03	1.07E-05	6.45E-03		
HI	5.65E-02	3.33E-03	5.24E-02	1.25E-02	6.64E-02	1.86E-03	1.93E-01		
			For rice g	rain at S4					
			Ad	lult					
HI	1.18E-03	2.26E-04	1.75E-02	5.23E-04	-	3.67E-04	1.98E-01		

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Heavy metals	Cr	Cu	Mn	Ni	Pb	Zn	Total	
Children								
HI	1.38E-03	2.64E-04	2.04E-02	6.11E-04	-	4.29E-04	2.31E-01	
For rice grain at S1-S3								
Adult								
HI	1.18E-02	6.65E-04	3.21E-02	1.32E-03	-	7.43E-04	4.66E-01	
Children								
HI	1.38E-02	7.77E-04	3.75E-02	1.55E-03	-	8.68E-04	5.44E-01	

IV. CONCLUSION

Six out of seven heavy metals including Mn, Zn, Ni, Cr, Cu and Pd were detected and were lower than the permitted limits of QCVN 03-MT: 2015/BTNMT and CCME. The concentration of the detected heavy metals in the topsoil (0-25cm) around the landfill (S1-S3) were higher than those at the location 1 km from the landfill (S4) with the exception for Ni, Pb. The concentration of the heavy metals in the rice parts in the surrounding landfill sites decreased from Mn> Zn> Cu> Ni> Cr (except for the heavy metals in the rice grains with the order of Cr> Cu> Ni). Cd was not detected in the rice and Pb only appeared in the rice roots. The calculation of the hazard index (HI) shows that the health risk due to heavy metals contamination in soil and rice grain for children was higher than for adult, however, all HI values fell into safe level. Health risk for rice consumption was higher than that for exposure to soil contaminating heavy metals. In addition, health risk for due to exposure to heavy metals by all routes in the area surrounding the landfill were higher than that at 1km away from the landfill. Measures should be taken to minimize the leakage of leachate into rice fields.

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International Journal of Environment, Agriculture and Biotechnology, 5(4) Jul-Aug, 2020 | Available: <u>https://ijeab.com/</u>

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