# Influence of Soil Characteristics and Arsenic, Cadmium, and Lead Contamination on Their Accumulation Levels in Rice and Human Health Risk through Intake of Rice Grown nearby Abandoned Mines

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Transfer ability of arsenic (As), cadmium (Cd), and lead (Pb) from soil to polished rice, the influence of soil characteristics, and contamination on As, Cd, and Pb contents in rice and potential health risk were investigated through intake of polished rice grown in nearby abandoned mines. Transfer factors (TFs) of As, Cd, and Pb from soil to polished rice were evaluated based on the total and HCl-extractable concentration of As, Cd, and Pb in soil. Regression analysis was used to predict the relationship of soil properties, and total and HCl-extractable As, Cd, and Pb concentrations in rice and human health risk through the rice consumption. The results showed that As, Cd, and Pb contents in polished rice and human health risk through rice intake were more influenced by HCl-extractable concentrations of As, Cd, and Pb in soil than by total concentration. The contents of As, Cd, and Pb in polished rice showed the negative relationship with soil pH and positive relationship with soil organic matter and cation exchangeable capacity (CEC). As content in polished rice was significantly and positively correlated with total and HCl-extractable As concentration in soil. Same results were observed for the relationship between total and HClextractable As concentration in soil and cancer risk probability (R) as well as hazard quotient (HQ) from As exposure. This demonstrates that human health risk from As exposure could be well predicted by both of total and HCl-extractable As concentrations in soil.

Key words: arsenic, cadmium, human health risk, lead, polished rice, soil characteristics

Soil contaminations by toxic elements such as arsenic (As), cadmium (Cd), and lead (Pb) mainly originate from anthropogenic activities and cause a decrease in quantity and quality of agricultural products. Particularly, mine tailings and waste rocks contaminate the agricultural fields with toxic elements during weathering and heavy rainfall. High contents of toxic elements in soil lead to detrimental effects on the crop growth, development, and yields, and may pose severe toxic effects in animal as well as human beings through the food chain (WHO, 2007).

Among agricultural products, rice (*Oryza sativa* L.) is widely consumed as a staple food in Asian countries and is a significant dietary source of toxic elements due to

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high availability and accumulation compared to other crops; thus, consumption of contaminated rice may cause potential human health risk [Mondal and Polya, 2008; Su *et al.*, 2010].

The uptake of toxic elements by rice crop mainly depends on their solubility and mobility in soil [Zeng et al., 2011]. The available form of toxic elements from soil to rice crop is dissolvable and exchangeable [Zeng et al., 2011]. Moreover, it is associated with soil characteristics (i.e. pH, organic matter, cation exchange capacity (CEC), redox potential, mineral contents, and iron (Fe)/manganese (Mn) oxides) [Zeng et al., 2011]. Several studies have reported that low pH and high contents of the organic matter in soil increase the bioavailability, controlling speciation, solubility, and mobility of toxic elements [Kashem and Singh, 2001; Zhao et al., 2010]. Furthermore, the dramatic decreases in redox potential and Fe/Mn oxides also increase the availability of toxic elements [Marin et al., 1993]. Apart from soil characteristics on availability of toxic elements, their soil contents also

control the uptake and accumulation of toxic elements to rice grain [Huang et al., 2006]. As content in polished rice, impacted by As-enriched paddy soil, is estimated to be high at 7.5 mg/kg [Liao et al., 2005]. Cd and Pb can be easily absorbed from soil to rice roots, translocated and accumulated in rice grain, and thus are considered as toxic metals that cause health risk through rice intake [Liu et al., 2003; Qian et al., 2010]. In this respect, the soil contamination of toxic elements becomes a major source influencing their uptake and accumulation by the edible part of rice and health risk through rice consumption [Huang et al., 2006]. Several studies on the soil availability of toxic elements and their uptake by rice crop have reported [Liu et al., 2003; Bogdan and Schenk, 2009; Chen et al., 2009]; however, only few studies have been documented on the influence of soil characteristics and contamination of toxic elements in rice as well as possible health risks.

The objectives of the present study were to investigate the transfer ability of toxic elements, including As, Cd, and Pb from soil to polished rice, and the influence of soil characteristics and contamination on their contents in rice, and the potential human health risks from rice intake cultivated in paddy fields adjacent to the abandoned mines.

## **Materials and Methods**

Soil characteristic measurement. Soil pH value was measured at the ratio of 1:5 distilled water using pH meter (250A, Thermo Orion, Beverly, MA). Organic matter and available phosphate ( $P_2O_5$ ) were determined by wet oxidation and Lancaster method, respectively. The exchangeable cations, i.e. Calcium (Ca), Potassium (K), Magnesium (Mg), Sodium (N), were measured using 1 N ammonium acetate at pH 7.0, and analyzed by inductively coupled plasma mass spectroscopy (ICP-MS) (Agilent Technologies 7500a, Santa Clara, CA). Cation exchangeable capacity (CEC) was determined by summation of exchangeable cations including Ca, K, Mg, Na, and hydrogen (H).

**Sample pretreatment and chemical analysis.** Soil and rice samples were collected from paddy fields adjacent to eight abandoned mines (CY, DD, SD, SS, OS, TC, DS, HY) in November 2010. Based on the reports of the Ministry of Environment (2008), these abandoned mines have Au-Ag bearing quartz veins, abundant tailings, waste rocks of 10~200,000 m<sup>3</sup>, and mine water. Soil composite samples (n=34) comprised of five subsamples were taken within a depth of 15 cm from the soil surface in each location. All soil samples were packed in polyethylene bags, immediately transported to

laboratory, and then stored at room temperature. Soil samples were air-dried, crushed, and passed through a 20-mesh sieve. Standard reference material (SRM; Contaminated soil BAM-U112; BAM Federal Institute for Materials Research and Testing, Berlin, Germany) and 10 g soil samples were extracted with 1 N HCl for As and 0.1 N HCl for Cd and Pb compounds via soil standard method as described by the Ministry of Environment (2001) and the National Institute of Agricultural Science and Technology (2000). Soil extracts were continuously shaken at 30°C for 1 h, filtered with Whatman No. 5B filter, analyzed by ICP-AES inductively coupled plasma atomic emission spectroscopy (Integra XL Dual, GBC, Melbourne, Victoria), followed by the hydride generator for As. The accuracies of soil SRM As, Cd, and Pb were 8.41±0.52, 4.42±0.23, and 158.23±16.93 mg/kg, with certified values of  $10.4\pm0.4$ ,  $3.91\pm0.24$ , and  $195\pm8$  mg/ kg, respectively. The recovery values of As, Cd, and Pb were 81.27±4.98, 112.97±5.95, and 81.14±8.68%, respectively.

Harvested rice samples (n=34) were air-dried, polished, and then pulverized with a homogenizer (Ace Homogenizer, Nihonseiki Kaisha Ltd, Tokyo, Japan) three times for 1 min. SRM rice flour NIST 1568a, (National Institute Standards Technology, Gaithersburg, MD) and 0.5 g polished rice samples were transferred into a high pressured-polytetrafluoroethylene (PTFE) vessel and digested with 8 mL of 70% HNO<sub>3</sub> and 1 mL H<sub>2</sub>O<sub>2</sub> (Sigma-Aldrich, St. Louise, MO) using microwave digestion system (ETHOS, Milestone, Italy). After cooling to room temperature, the extracts were filtered through a 0.45-µm membrane filter, and adjusted to a final volume of 25 mL. As, Cd, and Pb contents in polished rice were determined by ICP-MS (Agilent technologies 7500a). The SRM accuracy values of As, Cd, and Pb were 0.29±0.05, 0.019±0.001, and 0.01±0.003 mg/kg, with certified values of 0.29±0.03, 0.022±0.002, and <0.01 mg/kg respectively. Extraction efficiencies (%), determined by dividing the extracted As content by total As content were 100.58±17.59 for As, 97.40±6.88 for Cd, and 106.15±1.88 for Pb.

Transfer factors (TFs) were based on total concentrations of As, Cd, and Pb (TF<sub>total</sub>)=total As, Pb, and Cd concentrations in polished rice (mg/kg)/total As, Pb, and Cd concentrations in soil (mg/kg). TFs were determined as follows: extractable concentrations of As, Cd, and Pb (TF<sub>total</sub>)= HCl-extractable concentrations of As, Pb, and Cd in polished rice (mg/kg)/HCl-extractable concentrations of As, Pb, and Cd in soil (mg/kg).

**Human exposure estimation.** Average daily dose (ADD) values of As, Cd, and Pb were estimated for the mean and 95th percentile consumers; these values

represent the average and high-exposures to polished rice. The following equation modified based on US Environmental Protection Agency (EPA) Integrated Risk Information System (IRIS) was used in estimating the ADD values. The probability distribution of input parameters was used to predict the realistic ADD values using Crystal ball program 11.1.0 ver. (Denver, CO). The published input parameters used for estimating the ADD values through human intake of polished rice contaminated with As, Cd, and Pb were as follows:

ADD via rice intake (mg/kg-days)

$$=\frac{C_r \times IR_r \times FI_r \times EF \times ED}{W_{AB} \times AT}$$
(1)

where,  $C_r$  is total contents of As, Cd, and Pb in polished rice (mg/kg);  $IR_r$  is the rice ingestion rate (kg/day);  $FI_r$  is absorption factor for rice intake (unitless;1); EF is exposure frequency (365 days/year); ED is the exposure duration (70 years);  $W_{AB}$  is the average body weight of exposed person (kg); and AT is the average time of the exposed person (days).

The average body weight was provided by the fourth Korean National Health and Nutrition Examination Survey database in the Korea Centers for Disease Control and Prevention (2008). In the case of As, proportion of inorganic As (As<sub>i</sub>) to total As content was averagely considered as 57.4% [Paik et al., 2010]. Substitution of total As content with inorganic As content is more accurate in determining the health risk due to As exposure to humans through rice intake (WHO, 2007). The estimated ADD value was compared with a provisional tolerable daily intake (PTDI), recommended by Joint Food and Agriculture Organization/World Health Organization (FAO/WHO) Expert Committee on Food Additives (JECFA) which were 2.1 µg As<sub>i</sub>/kg body weight (bw)/day (1989), 1 µg Cd/kg bw/day (2003), and 3.6 µg Pb/kg bw/day (1993).

**Human Health risk assessment.** Health risk of humans from exposure to As, Cd, and Pb is associated with both cancer and non-cancer toxic effects [Mondal and Polya, 2008]. Based on ADD values, the probabilistic health risks at mean and 95th percentile values were assessed by both cancer risk probability (R) and hazard quotient (HQ). The R value represents the possibility of cancer due to lifetime exposure through rice intake, and is acceptable within  $10^{-6} \sim 10^{-4}$  for the regulatory purpose [Bartell, 1996]. This value was calculated using the following equation (2).

$$R=1-\exp(-CSF \times ADD) \tag{2}$$

where R is the cancer risk probability; CSF is oral cancer

slope factor (mg/kg)/day; and *ADD* is the estimated average daily dose through rice intake (mg/kg-day).

Non-cancer toxic effects estimated by HQ are determined by comparing estimated ADD values with a reference dose (RfD) using the following equation (3).

$$HQ = \frac{ADD}{RfD} \tag{3}$$

where *RfD* is the oral reference dose value (mg/kg-day).

The HQ values of As, Cd, and Pb in polished rice were summed to estimate the overall toxic risk, and the hazard index (HI) under the assumption of additive interaction among them using the following equation (4).

$$HI = \sum HQ_i; i=1...n \tag{4}$$

**Statistical analysis.** Regression analysis was performed to investigate the influence of soil characteristics, i.e. pH, organic matter, CEC, and contamination, on the contents of As, Cd, and Pb in rice using SPSS statistical program ver. 12.0 (SPSS Co., Chicago, IL). In regression analysis, the soil organic matter content, CEC, total and HCl-extractable soil concentrations, and contents of As, Cd, and Pb in rice were  $\log_{10}$ -transformed to make homogeneous variances.

# **Results and Discussion**

Soil characteristics. The results of characterized soil samples (n=34) are as follows: soil pH ranged from 5.26 to 6.85; soil organic matter contents ranged from 12.07 to 37.93 g/kg dry soil, with a mean value of 22.36 g/kg dry soil; available P<sub>2</sub>O<sub>5</sub> was 131.84 mg/kg on the average, ranged from 14.88 to 373.24 mg/kg; CEC, calculated from summation of exchangeable cation capacity, i.e. H, Ca, K, Mg, and Na varied from 7.00 to 43.10 cmol<sup>+</sup>/kg, in the range of  $1.42 \sim 6.21 \text{ cmol}^+/\text{kg}$  for Ca,  $0.07 \sim 1.16 \text{ cmol}^+/\text{kg}$ kg for K,  $0.26 \sim 2.37 \text{ cmol}^+/\text{kg}$  for Mg, and  $0.03 \sim 0.20$ cmol<sup>+</sup>/kg for Na, respectively (Table 1). The results showed the wide range of soil pH, organic matter, available P<sub>2</sub>O<sub>5</sub>, and CEC to examine the effect of characteristics of soil adjacent to abandoned mines on As, Cd, and Pb contents in polished rice harvested from nearby areas.

As, Cd, and Pb contents in soil/polished rice and availability to polished rice. Total and HCl-extractable As, Cd, and Pb contents in soil and polished rice were measured to evaluate their transfer abilities from soil to polished rice (Table 2). Total As and Cd concentrations in soil showed mean values of 22.99 and 3.48 mg/kg, respectively. These levels closely reached the soil guideline (Ministry of Environment, 2009) and maximum

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Mines	$\begin{array}{ccc} \text{Mines} & \text{pH} (\text{H}_2\text{O}) & \text{EC}^{\text{a}} & \text{Organic r} \\ \text{(n=34)} & 1:5 & (\text{dS/m}) & (\text{g/kg dry}) \end{array}$	EC <sup>a</sup>	Organic matter	Av-P <sub>2</sub> O <sub>5</sub>	Exchangeable cations (cmol <sup>+</sup> /kg)					
(n=34)		(g/kg dry soil)	y soil) (mg/kg)	Ca	K	Mg	Na	CEC <sup>b</sup>		
Mean	5.69	0.46	22.36	131.84	3.29	0.70	1.00	0.12	33.90	
SD	0.30	0.38	5.83	106.38	1.16	0.33	0.50	0.05	6.74	
Min	5.26	0.26	12.07	14.88	1.42	0.07	0.26	0.03	7.00	
Max	6.85	2.45	37.93	373.24	6.21	1.16	2.37	0.20	43.10	

#### **Table 1. Soil characteristics**

<sup>a</sup>Electrical conductivity.

<sup>b</sup>Cation exchangeable capacity.

Table 2. Total and HCl-extractable concentrations of A	s, Cd	, and Pb in	ı soil and	transfer	factor	(TF	) to j	polished	rice
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Toxic elements	No	Soil <sub>Total</sub> (mg/kg)	Soil <sub>Extractable</sub> (mg/kg)	Extractable fraction <sup>a</sup> (%)	Polished rice (mg/kg)	TF <sub>total</sub> <sup>b</sup>	$\mathrm{TF}_{\mathrm{extractable}}$ <sup>c</sup>
	110.	Mean (Min~Max)	Mean (Min~Max)	Mean (Min~Max)	Mean (Min~Max)	Mean (Min~Max)	Mean (Min~Max)
As	34	22.99 (2.51~84.90)	5.66 (0.44~24.26)	23.68 (2.62~50.80)	0.18 (0.08~0.44)	0.014 (0.002~0.056)	0.084 (0.008~0.334)
Cd	34	3.48 (2.07~4.95)	0.21 (0.004~2.08)	5.49 (0.09~43.15)	0.04 (0.001~0.27)	0.013 (0.0004~0.097)	0.722 (0.029~11.890)
Pb	34	46.57 (12.15~131.64)	10.16 (5.00~38.40)	23.35 (10.84~84.30)	0.07 (0.004~0.44)	0.002 (0.0001~0.017)	0.009 (0.0005~0.080)

<sup>a</sup>The proportion of HCl-extractable form in soil is equal to HCl-extractable concentrations of As, Cd, and Pb in soil/total concentrations of As, Cd, and Pb in soil.

<sup>b</sup>Transfer factor based on total concentration in soil is equal to As, Cd, and Pb contents in polished rice/total As, Cd, and Pb concentrations in soil.

"Transfer factor based on HCl-extractable concentration in soil is equal to As, Cd, and Pb contents in polished rice/HClextractable As, Cd, and Pb concentrations in soil.

concentration (131.64 mg/kg) of Pb in soil was below the soil guideline. Soil HCl-extractable As, Cd, and Pb concentrations varied with mean values of 5.66, 0.21, and 10.16 mg/kg, respectively. Thus, extractable proportions of As, Cd, and Pb were estimated with mean values of 23.68, 5.49, and 23.35%, respectively. Soil extractable proportions of As and Pb were approximately four times greater than that of extractable proportion of Cd.

The average contents of As, Cd, and Pb in polished rice were 0.18, 0.04, and 0.07 mg/kg, respectively. The mean Cd and Pb concentrations in polished rice did not exceed the safety guideline (0.20 mg/kg), established by Korea Food and Drug Administration (2006), whereas the maximum contents of Cd and Pb were higher than the safety guideline. In addition, the As content in polished rice was as high as that in heavily As-contaminated rice of Bangladesh [Alam *et al*, 2002], and exceeded the global normal content of As level in the range of 0.082~0.202 mg/kg [Zavala and Duxbury, 2008]. High concentrations of As, Cd, and Pb in mine-impacted paddy soil resulted in high contents of As, Cd, and Pb in polished rice, ranging from 0.23 to 0.58, from 0.46 to 0.90, and from 0.08 to 0.33 mg/kg, respectively [Kim *et al.*, 2009].

TFs of As, Cd, and Pb to polished rice were evaluated based on the total and HCl-extractable soil concentrations (Table 2). The TF values of As, Cd, and Pb from total concentration in soil averaged at 0.014, 0.013, and 0.002, respectively. This TF value was consistent with the results reported by Huang et al. [2006], which showed that the median TF value of As from soil to polished rice was 0.02. For TF values of the Cd and Pb, similar ranges were observed with those reported by Chen et al. [2009], which were in the range of  $0.003 \sim 3.40$  and  $0.0001 \sim$ 0.007, respectively. The TF value of total As and Cd contents from soil to polished rice was five times higher than that of total Pb content in soil. Moreover, the TF values of As, Cd, and Pb, based on HCl-extractable soil concentration, were 0.084, 0.722, and 0.009 on the average, respectively. The average biological absorption coefficient (BAC) of As was 0.06 from HCl-extractable As content in soil to polished rice. Particularly, much higher TF value of 0.722 was observed in HClextractable Cd concentration in soil, even though TF

and the total and HCl-extractable concentrations in soil								
		Regression parameters						
Х	У	а	b	$\mathbb{R}^2$				
	Log <sub>10</sub> <sup>(As rice)</sup>	-0.37	-0.07	0.13				
Soil pH	Log <sub>10</sub> <sup>(Cd rice)</sup>	0.94	-0.47	0.25				
	Log <sub>10</sub> <sup>(Pb rice)</sup>	-1.50	0.005	0.002				
	Log <sub>10</sub> <sup>(As rice)</sup>	-0.87	0.08	0.06				
Log <sub>10</sub> <sup>(O.M.)a</sup>	Log <sub>10</sub> <sup>(Cd rice)</sup>	-2.55	0.64	0.14				
	Log <sub>10</sub> <sup>(Pb rice)</sup>	-1.60	0.10	0.02				
	Log <sub>10</sub> <sup>(As rice)</sup>	-0.89	0.08	0.07				
Log <sub>10</sub> (CEC)b	Log <sub>10</sub> <sup>(Cd rice)</sup>	-3.50	1.18	0.29				
-	Log10 (Pb rice)	-1.06	-0.27	0.07				
Log <sub>10</sub> <sup>(Tot. As soil)</sup>	Log <sub>10</sub> <sup>(As rice)</sup>	-0.98	0.17	0.42*°				
Log <sub>10</sub> <sup>(Ext. As soil)</sup>	Log <sub>10</sub> <sup>(As rice)</sup>	-0.84	0.15	0.46***				
Log <sub>10</sub> <sup>(Tot. Cd soil)</sup>	Log <sub>10</sub> <sup>(Cd rice)</sup>	-1.81	0.19	0.04				
Log <sub>10</sub> <sup>(Ext. Cd soil)</sup>	Log <sub>10</sub> <sup>(Cd rice)</sup>	-1.30	0.40	0.37*°				
Log <sub>10</sub> <sup>(Tot. Pb soil)</sup>	Log <sub>10</sub> <sup>(Pb rice)</sup>	-0.42	-0.64	0.20				
Log <sub>10</sub> <sup>(Ext. Pb soil)</sup>	Log <sub>10</sub> <sup>(Pb rice)</sup>	-2.08	0.64	0.24				

Table 3. Fitted linear regressions in the form of y=a+bx

for As, Cd, and Pb contents in rice to soil characteristics

Soil organic matter, cation exchangeable capacity, total and HCl-extractable concentrations of As, Cd, and Pb in soil, and their contents in polished rice were Log<sub>10</sub>-transformed to give homogeneous variances.

<sup>a</sup>Organic matter

<sup>b</sup>Cation exchangeable capacity

<sup>°</sup>Asterisk (\* and \*\*) indicates the significant difference at the 0.05 and 0.01 probability levels, respectively.

value of Cd to polished rice was similar to that of total concentration of As in soil. HCl-extractable Cd concentration in soil showed a dramatic increase in transfer ability to polished rice than those of As and Pb. This indicated that soil HCl-extractable Cd concentration was strongly absorbed and accumulated by polished rice,

and intake of Cd-contaminated rice could cause health risk. The transfer ability of Pb to polished rice was much lower, regardless of total and HCl-extractable soil concentration. Overall, higher transfer ability was observed from soil HCl-extractable contents than from the total As, Cd, and Pb contents.

Effects of soil characteristics and contamination of As, Cd, and Pb on rice contents. Simple linear regression analysis was performed to identify the influence of soil characteristics (i.e. pH, organic matter, CEC), as well as total and HCl-extractable concentrations of As, Cd, and Pb on thier contents in polished rice (Table 3). The soil availability of toxic metals to rice plants is mainly controlled by soil properties, i.e. pH, soil organic matter, and CEC [Zeng *et al.*, 2011].

The As and Cd contents in polished rice were negatively correlated with soil pH, but with no significant difference (p > 0.05). This was consistent with the result that uptake of As and Cd in rice plants is typically reduced due to the decreased availablility in soil with higher pH [Römkens et al., 2009]. Morever, there was no obvious relationship between Pb contents in polished rice and soil pH. Soil organic matter positively influenced the As, Cd, and Pb contents in polished rice. The soil organic matter bound with toxic elements in soil increases the availability to rice plant, and enhances the mobility and solubility in soil. The influence of CEC in soil was positive for As and Cd contents, but negative for Pb content in polished rice. Particularly, the CEC in soil strongly showed a positive relationship with Cd content in polished rice as compared to As and Pb contents. However, the soil organic matter and CEC did not show a significant relationship with the As, Cd, and Pb contents in polished rice (p>0.05). Cd content in polished rice was more influenced by soil pH, organic matter, and CEC properties than by As and Pb contents. Apart from soil

Table 4. Average daily dose (ADD) values and human health risk posed by environmental exposure to As, Cd, and Pb through rice intake

Toxic - Elements	ADD (mg/kg-day)			Cancer risk p	robability (R) <sup>c</sup>	Hazard Quotient (HQ) <sup>d</sup>	
	Mean	P95 <sup>a</sup>	% of PTDI <sup>b</sup> (Mean/P95 <sup>a</sup> )	Mean	P95 <sup>a</sup>	Mean	P95ª
As	4.6×10 <sup>-4</sup>	9.7×10 <sup>-4</sup>	21.7/46.2	6.8×10 <sup>-4</sup>	1.5×10 <sup>-3</sup>	1.45	3.33
Cd	$1.1 \times 10^{-4}$	$4.1 \times 10^{-4}$	10.7/40.7	-	-	0.12	0.40
Pb	$3.3 \times 10^{-4}$	$1.2 \times 10^{-3}$	9.3/33.3	$2.7 \times 10^{-6}$	$1.0 \times 10^{-5}$	0.63	2.38

<sup>a</sup>95th percentile

<sup>b</sup>The proportion of provisional tolerable daily intake (PTDI) was calculated by dividing the ADD values of As, Cd, and Pb into their respective PTDIs of 2.1 µg As<sub>i</sub>/kg bw/day (FAO/WHO, 1989), 1 µg Cd/kg bw/day (FAO/WHO, 2003), and 3.6 µg Pb/kg bw/day (FAO/WHO, 1993).

<sup>c</sup>R=1-exp[-cancer slope factor (CSF)×ADD]

<sup>d</sup>HQ=Average daily dose (ADD)/reference dose (RfD)

characteristics, the total and HCl-extractable As concentration in soil significantly showed positive influence on the As contents in polished rice, with regression coefficients ( $\mathbb{R}^2$ ) of 0.42 and 0.46 ( $p \le 0.01$ ). Liao et al. [2005] reported that the polished rice impacted by As-enriched soil was estimated to be high at 7.5 mg/ kg. Zhu et al. [2008] reported that the As level accumulated in rice is also high in heavily As-contaminated soil, due to the increment of As bioavailability to rice grain from soil. Significantly positive relationship was found between Cd contents in polished rice and HCl-extractable Cd concentration in soil, with  $R^2$  of 0.37 (p<0.05). This was demonstrated by the higher TF of Cd from HClextractable concentration in soil to polished rice (Table 2). Both total and HCl-extractable Pb concentrations in soil showed no significant effect on the Pb content in polished rice. Overall, HCl-extractable concentrations of As, Cd, and Pb in soil had the greater effect on polished rice than their total concentrations. This suggests that HCl-extractable contents of toxic elements in soil could be the dominant factor in predicting their contents in rice.

Influence of soil As, Cd, and Pb contamination on the health risk through rice intake. Human exposure to As, Cd, and Pb and the corresponding human health risk through rice consumption was assessed to identify the influence of As, Cd, and Pb concentrations in soil (Table 4). Estimated ADD values at mean and 95th percentile were  $4.6 \times 10^{-4}$  and  $9.7 \times 10^{-4}$  for As,  $1.1 \times 10^{-4}$  and  $4.1 \times 10^{-4}$ for Cd, and  $3.3 \times 10^{-4}$ , and  $1.2 \times 10^{-3}$  for Pb. When ADD values were compared with PTDIs of As, Cd, and Pb, the exposure amount accounted for 21.7 and 46.2% for As, 10.7 and 40.7% for Cd, and 9.3 and 33.3% for Pb at mean and 95th percentile. Furthermore, cancer risk probabilities (R) for both average and excessive consumption of rice contaminated with As and Pb were estimated to be  $6.8 \times 10^{-4} \sim 1.5 \times 10^{-3}$  and  $2.7 \times 10^{-6} \sim 1.0 \times 10^{-5}$ , respectively, which exceeded the limit set at  $10^{-6} \sim 10^{-4}$  for regulatory purpose, implying that cancer risk may be caused through intake of rice contaminated with As and Pb in mineaffected paddy fields. Furthermore, HQ for population exposed to As was respectively estimated to be 1.45 and 3.33 at mean and 95th percentile. This result indicates that the chronic toxic of As effects on the consumer of As-contaminated rice based on HQ beyond 1.0.

The chronic toxic effects of As-contaminated polished rice on consumer may be proposed based on HQ beyond 1.0.

The excessive intake of rice exposed to Pb may also cause adverse toxic effects, with an HQ estimation of 2.38. The HI values for As, Cd, and Pb exceeding HQ value of 1.0, implies the possibility of non-carcinogenic toxic risks. The uptake of toxic metals in rice grain from contaminated soil contributes to health risk [Brus et al., 2009; Qian et al., 2010]. The relationship of cancer risk and HQ value with total and HCl-extractable soil contamination was also inveatigated. (Tables 5 and 6). Cancer risk from exposure to As was positively influenced by both total and HCl-extractable As concentrations in soil, with high  $R^2$  of 0.91 (p<0.01) and 0.71 (p<0.05). This result indicated that cancer risk by As exposure may be well predicted by soil contamination of both total and extractable As. Unlike As, Pb concentration in soil did not affect the cancer risk, which can be attributed to the very low transfer ability of Pb from soil to polished rice (Table 2). Moreover, the total and HCl-extractable Pb concentrations in soil did not show significantly positive relationship with Pb content in polished rice (Table 3), indicating that Pb availability to polished rice and Pb content in polished rice were not affected by the soil characteristics and contamination. Zeng et al. [2011]. suggested that the Pb uptake and transportation in plants are mainly controlled by the physiological feature of crops. Likewise, the total and HCl-extractable As concentrations in soil showed a significantly positive influence on the HQ estimation, with high  $R^2$  of 0.92 (p <0.01) and 0.72 (p<0.05). In Cd and Pb, HCl-extractable concentrations were positively related with HQ value with R<sup>2</sup> values of 0.41 and 0.37; however, no significant

Table 5. Fitted linear regressions in the form of y=a+bx for the cancer risk probability (R) through rice intake to the total and extractable concentrations of As, Cd, and Pb in soil

	Cancer risk probability (R)						
Parameters	Log	(R As) 0	Log <sub>10</sub> <sup>(R Pb)</sup>				
	$\mathrm{Log_{10}}^{(\mathrm{Tot.}\ \mathrm{As\ soil})}$	$\mathrm{Log}_{10}^{(\mathrm{Ext. As soil})}$	$Log_{10}{}^{(\text{Tot. Pb soil})}$	$\mathrm{Log}_{10}^{(\mathrm{Ext. \ Pb \ soil})}$			
а	-3.48	-3.27	-5.79	-5.33			
b	0.25	0.16	-0.03	-0.81			
$\mathbb{R}^2$	0.91**a	0.71*a	0.03	0.22			

Total and HCl-extractable concentrations of As and Pb in soil, and the cancer risk probability of As and Pb from rice consumption were Log<sub>10</sub>-transformed to give homogeneous variances.

<sup>a</sup>Asterisk (\* and \*\*) indicates the significant difference at the 0.05 and 0.01 probability levels, respectively.

	Hazard Quotient (HQ)							
Regression parameters	Log <sub>10</sub> <sup>(HQ As)</sup>		Log <sub>10</sub>	(HQ Cd) 0	Log <sub>10</sub> <sup>(HQ Pb)</sup>			
	$Log_{10}^{(\text{Tot. As soil})}$	$Log_{10}^{(Ext. As soil)}$	$Log_{10}^{(Tot. Cd soil)}$	$Log_{10}^{(Ext. Cd soil)}$	$Log_{10}^{(Tot. Pb soil)}$	Log <sub>10</sub> <sup>(Ext. Pb soil)</sup>		
а	-0.14	0.08	-0.36	-0.32	1.09	-1.09		
b	0.25	0.16	-0.33	0.26	-0.89	0.73		
$\mathbb{R}^2$	0.92** <sup>a</sup>	0.72* <sup>a</sup>	0.12	0.41	0.21	0.37		

Table 6. Fitted linear regressions in the form of y=a+bx for the hazard quotient (HQ) via rice intake to the total or extractable concentrations of As, Cd, and Pb in soil

Total and HCl-extractable concentrations of As and Cd in soil, and HQs of As and Cd from rice consumption were  $Log_{10}$ -transformed to give homogeneous variances.

<sup>a</sup>Asterisk (\* and \*\*) indicates the significant difference at the 0.05 and 0.01 probability levels, respectively.

relationship was observed between them. Particularly, the total concentrations of Cd and Pb in soil did not significantly influence the non-cancer toxic risks. Soil contamination of Cd and Pb did not show the obvious correlation with their contents in polished rice. (Table 3).

In the case of Pb, no significant relationship was observed between the total and HCl-extractable concentrations in soil and cancer or non-cancer risk. Thus, the absorption mechanism, except for soil chracteristics and contamination, can be attributed to Pb uptake by polished rice, and potential human health risk through rice intake. Therefore, further study on the uptake and accumulation mechanism of toxic metals in the rice plants may be nessasary to predict the risk induced by toxic metals for human health.

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