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Article

Heavy Metal(loid) Levels in Paddy Soils and Brown Rice in Korea

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There is an increasing concern over heavy metal(loid) contamination of soil in agricultural areas including paddy soils. This study was conducted to monitor the background levels of heavy metal(loid)s, arsenic (As), cadmium (Cd), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb), and zinc (Zn) in major rice growing soils and its accumulation in brown rice in Korea. The samples were collected from 82 sites nationwide in the year 2012. The mean and range values of As, Cd, Cu, Hg, Ni, Pb, and Zn in paddy soils were 4.41 (0.16-18.9), 0.25 (0.04-0.82), 13.24 (3.46-27.8), 0.047 (0.01-0.20), 13.60 (3.78-35.0), 21.31 (8.47-36.7), and 54.10 (19.19-103.0) mg kg^{-1}, respectively. This result indicated that the heavy metal(loid) levels in all sampled paddy soils are within the permissible limits of the Korean Soil Environment Conservation Act. The mean and range values of As, Cd, Cu, Hg, Ni, Pb, and Zn in brown rice were 0.146 (0.04-0.38), 0.024 (0.003-0.141), 4.27 (1.26-16.98), 0.0024 (0.001-0.008), 0.345 (0.04-2.77), 0.113 (0.04-0.197), and 22.64 (14.1-35.1) mg kg^{-1}, respectively. The mean and range BCF (bioconcentration factor) values of As, Cd, Cu, Hg, Ni, Pb, and Zn in brown rice were 0.101 (0.01-0.91), 0.121 (0.01-0.70), 0.399 (0.05-2.60), 0.061 (0.016-0.180), 0.033 (0.004-0.44), 0.005 (0.003-0.013), and 0.473 (0.19-1.07), respectively, with Zn showing the highest. The results show that the levels of all metal(loid)s in all sampled brown rice are generally within the acceptable limit for human consumption.

Key words: Heavy metal(loid)s, Paddy soil, Brown rice, Bioconcentration factor

Mean and range of heavy metal(loid) concentrations in brown rice in Korea.

Heavy metal(loid)	Metal(loid) concentrations in brown rice (mg kg^{-1})
As	0.146 (0.04-0.38)
Cd	0.024 (0.003-0.141)
Cu	4.27 (1.26-16.98)
Hg	0.0024 (0.001-0.008)
Ni	0.345 (0.04-2.77)
Pb	0.113 (0.04-0.197)
Zn	22.64 (14.1-35.1)

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Introduction

Heavy metal(loid)s are released into the environment by both natural and anthropogenic sources. The concentrations of heavy metal(loid)s releasing into soil systems by pedogenic processes are low and depend largely on the origin and nature of the parent materials (Adriano, 2001). However, anthropogenic processes such as application of metal(loid)-containing pesticides, commercial fertilizers, wastewaters and sewage sludge have led to a widespread accumulation of metal(loid)s in agricultural soils (Bolan et al., 2014; Wuana and Okieiman, 2011). High levels of toxic elements in soil can have a negative impact on soil fertility, lead to detrimental effects on crop growth and yield, and may pose a severe toxic risk to animals as well as human beings through the food chain (Cardosa et al., 2013).

Heavy metal(loid) accumulation in crops is a function of complex interactions between the soil, the plant and environmental factors. It has been well documented that the content of heavy metal(loid)s in crop plants is closely related to the levels in the soil (Adriano, 2001; Cheng et al., 2006). Previous studies have investigated the effect of soil properties such as soil pH, organic matter (OM), redox potential, cation exchange capacity (CEC), and phosphorus content on the transfer of heavy metal(loid)s (Zhao et al., 2010). Heavy metal(loid) contamination in paddy soils is one of the most serious problems facing rice production and soil management in Asian countries. Among agricultural food products, rice (*Oryza sativa* L.) is one of the most widely consumed staple cereal foods in the world constituting about 89% of the diet of people in Asian countries. In many East and South Asian countries including Japan, Bangladesh, Indonesia, and Korea, the accumulation of metal(loid)s, particularly arsenic (As) and cadmium (Cd), in rice ecosystems and its subsequent transfer to the human food chain is a major environmental issue.

The most significant anthropogenic source of these metal(loid)s including As, Cd, lead (Pb), and zinc (Zn), is the mining industry and the inappropriate management of associated mining wastes. In Korea, rice is the most common crop grown on agricultural land. The cultivated land in Korea is around 21%, and of that 61% is paddy fields. Large areas of agricultural land, including paddy fields, have been contaminated by As, Cd and Pb via effluent from mine tailings and other wastes generated by closed or abandoned mines (Kim et al., 2007; Kwon et al., 2013; Yang et al., 2006) and thus results to the uptake of heavy metal(loid)s by rice plants, posing a significant public health risk to the local community. Most previous studies on accumulation of heavy metal(loid)s in Korea were focused on special field areas such as industrial and mining regions. Only little information is available on the heavy metal(loid) accumulation in non-contaminated paddy fields throughout the country, and the relationship between heavy metal(loid)s in soil-rice system has been less investigated. Based on this fact, there was a need to survey the levels of heavy metal(loid)s in non-contaminated paddy soils and rice grains. The present study was undertaken in order to (1) assess the background levels of heavy metal(loid)s (As, Cd, copper (Cu), mercury (Hg), nickel (Ni), Pb, and Zn) in paddy soils and brown rice in Korea; (2) assess the degree of contamination by calculating bioconcentration factor (BCF) and (3) determine the effect of soil properties and total metal(loid) concentrations in influencing the availability of heavy metal(loid)s to rice.

Materials and Methods

Sample collection and analysis Eighty-two rice grain samples were collected in October 2012 from various rice cultivated fields throughout Korea (Fig. 1). Eighty-two soil samples were collected from the root zone (<15 cm depth), corresponding to rice samples during the early harvest period. Soil samples were air dried to constant weight, crushed and passed through 2 mm sieve. Paddy rice (rice grains with husks) was air dried to constant weight, husks were removed and pulverized using a homogenizer (Ace Homogenizer, Nihonseiki Kaisha Ltd, Tokyo, Japan) and stored until analysis.

The soil samples were analyzed for pH, electrical conductivity (EC), CEC, and total heavy metal(loid)s. The soil pH and EC were determined using end-over-end

Fig. 1. Sampling areas for paddy soils and brown rice in Korea.

equilibration of soil with water at a ratio of 1:5 for an hour and measuring the solution with calibrated pH and conductivity meters (250A, Thermo Orion, Beverly, MA). The exchangeable cations, i.e. calcium (Ca), potassium (K), magnesium (Mg), and sodium (Na), 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 was determined by the summation of exchangeable cations. For the analysis of total heavy metal(loid) contents, soil samples (3 g) were digested using aqua regia at 30 °C for 2 h, and then extracted at 90 °C for 150 min, by Kjeldatherm block digestion system (Gerhardt GmbH, Northants, UK). After cooling to ambient temperature, the digests were filtered through a 0.45-μm membrane filter, made up to a volume of 50 ml and analysed for total metal(loid) concentrations using hydride generation inductively coupled plasma atomic emission spectroscopy (HG-ICP-AES; Integra XL Dual, GBC, Melbourne, Australia). A standard reference material (SRM; contaminated soil BAM-U112a; certified by the BAM Federal Institute for Materials Research and Testing, Berlin, Germany) and a blank were included to validate the digestion operation. A direct Hg analyzer (DMA-80 Milestone, Italy) was used for total Hg concentration in soil and rice samples by following the EPA method 7473.

The brown rice samples (0.25 g) were transferred to a high pressured-polytetrafluoroethylene (PTFE) vessel and digested with 8 mL of 70% HNO_3 and 1 mL H_2O_2 using microwave digestion system (ETHOS, Milestone, Italy). After cooling to room temperature, the extracts were filtered through a 0.45-μm membrane filter, adjusted to a final volume of 25 mL and analysed for metal(loid) contents using HG-ICP-MS. A standard reference material (SRM rice flour NIST 1568a, certified by the National Institute Standards Technology, Gaithersburg, MD) and a blank were included to validate the digestion operation. All the analyses were carried out in triplicate.

Bioconcentration factor Biocencentration factor (BCF) is the ratio of the content of metal(loid)s in various parts of crop to that in the soil (Liu et al., 2009). In this study, the BCF was calculated for rice grains. If a BCF $\leq$ 1.00, it indicates that the plant can only absorb but not accumulate the metal(loid). On the contrary, if a BCF > 1.00, it means that the plant has the potential to accumulate metal(loid)s (Liu et al., 2009; Satpathy et al., 2014).

Statistical analysis All calculations and standard deviations between the replicates were done using the graphing software, SigmaPlot (version 10.0). Pearson correlations between metal(loid)s in grains and, soil properties and total metal(loid) concentrations were performed using SPSS software (version 18.0).

Results and Discussion

Heavy metal(loid) concentrations in paddy soils The mean and range values of As, Cd, Cu, Hg, Ni, Pb, and Zn in paddy soils were 4.41 (0.16-18.9), 0.25 (0.04-0.82), 13.24 (3.46-27.8), 0.047 (0.01-0.20), 13.60 (3.78-35.0), 21.31 (8.47-36.7), and 54.10 (19.19-103.0) mg kg^{-1}, respectively (Table 1). This result indicates that the heavy metal(loid) levels in all sampled paddy soils are within the permissible limits of the Korean Soil Environment Conservation Act. Satpathy et al. (2014) determined the metal(loid) (Cd, Cu, Pb and Zn) contamination in paddy soils and rice grains at the East coast of India and noted that the concentrations of Pb, Cd, Cu, and Zn ranged from 5.3-19.8, 0.2-0.6, 1.10-2.9, and 3.8-33.8 mg kg^{-1}, respectively. Zhao et al. (2009) observed that the soils in Zhejiang province, China were contaminated by Cd, Ni and Hg to some extent based on the permissible limits set by the Ministry of Environmental Protection in China. However, the pollution by Cu, Pb, and Zn were relatively low and were below the threshold values.

Table 1. Heavy metal(loid) concentrations in paddy soils and selected soil properties.

	Heavy metal(loid)s							pH	EC	CEC
	As	Cd	Cu	Hg	Ni	Pb	Zn		ds m^{-1}	cmol kg^{-1}
	mg kg^{-1}									
Mean	4.41 (0.16-18.9)	0.25 (0.04-0.82)	13.24 (3.46-27.8)	0.047 (0.01-0.20)	13.60 (3.78-35.0)	21.31 (8.47-36.7)	54.10 (19.19-103)	5.9 (4.9-7.0)	0.132 (0.043-0.923)	31.02 (9.36-55.39)
Med.	3.66	0.25	11.73	0.041	12.26	21.37	52.07	5.9	5.9	31.19
P95	10.31	0.54	25.38	0.124	27.79	30.61	85.47	6.4	0.258	45.16
Threshold value	25	4	150	4	100	200	300	-	-	-

EC: Electrical conductivity, CEC: Cation exchange capacity; *values in parentheses indicate range

Table 2. Heavy metal(loid) concentrations in brown rice and bioconcentration factor (BCF) values for heavy metal(loid)s.

Heavy metal(loid)	Metal(loid) concentrations in brown rice (mg kg^{-1})			Bioconcentration factor (BCF) values		
	Mean	Median	P95	Mean	Median	P95
As	0.146 (0.04-0.38)	0.141	0.246	0.101 (0.01-0.91)	0.032	0.345
Cd	0.024 (0.003-0.141)	0.013	0.077	0.121 (0.01-0.70)	0.069	0.395
Cu	4.27 (1.26-16.98)	3.30	11.59	0.399 (0.05-2.60)	0.288	0.984
Hg	0.0024 (0.001-0.008)	0.0021	0.0042	0.061 (0.016-0.18)	0.053	0.120
Ni	0.345 (0.04-2.77)	0.182	1.156	0.033 (0.004-0.44)	0.015	0.114
Pb	0.113 (0.04-0.197)	0.111	0.182	0.005 (0.003-0.013)	0.006	0.008
Zn	22.64 (14.1-35.1)	22.73	28.56	0.473 (0.19-1.07)	0.428	0.803

*values in parentheses indicate range

Heavy metal(loid) concentrations in brown rice In this study, the mean and range values of As, Cd, Cu, Hg, Ni, Pb, and Zn in brown rice were 0.146 (0.04-0.38), 0.024 (0.003-0.141), 4.27 (1.26-16.98), 0.0024 (0.001-0.008), 0.345 (0.04-2.77), 0.113 (0.04-0.197), and 22.64 (14.1-35.1) mg kg^{-1}, respectively (Table 2). The results showed that the metal(loid) concentrations in rice samples were within the permissible levels as recommended by the Korean Food and Drug Administration (Pb and Cd in rice: 0.2 mg kg^{-1}), the Joint FAO/WHO Expert Committee on Food Additives (As: 1 mg kg^{-1}; Zn: 60 mg kg^{-1}; and Cu: 40 mg kg^{-1}), and the Ministry of Health of the People's Republic of China (Hg: 0.02 mg kg^{-1}).

According to (Jung, 1995), the Cd content in rice grown in various countries ranged from 0.01 to 0.05 mg kg^{-1}. Machiwa (2010) surveyed the concentrations of heavy metal(loid)s in paddy soils and grains at a wider scale in different locations including closer to mining and within urbanized areas in Tanzania. The results showed that the levels of Cd, Cu, Hg, Pb, and Zn in brown rice were generally within the acceptable levels for human consumption, with Hg content in polished rice ranging from <0.0001 to 0.0062 mg kg^{-1}. Park et al. (2008) investigated the Hg content in paddy soils and its uptake to rice plant in Korea and noticed that the Hg levels in brown and polished rice were 0.00166 and 0.00145 mg kg^{-1}, respectively. Al-Saleh and Shinwari (2001) reported that the average Hg in polished rice grains was 0.003 mg kg^{-1} and Cd and Pb values ranged from <0.001-0.178 and 0.023-1.529 mg kg^{-1}, respectively. Zhao et al. (2015) observed that concentrations of Cd in rice ranged from 0.003 mg kg^{-1} to 0.103 mg kg^{-1}, with an average of 0.011 mg kg^{-1}. The concentrations of Cu and Zn ranged from 1.06-4.23 and 9.45-21.97 mg kg^{-1}, respectively.

Huang et al. (2006) investigated the health risks of As in soils collected from paddy rice fields in Fujian Province, China and noted that the As concentration in brown rice ranged from 0.041-0.201 mg kg^{-1}. Rogan et al. (2007) showed that the As in unpolished rice was in the range 0.11−0.52 mg kg^{-1}, with a median value of 0.18 mg kg^{-1}. While Payus and Talip (2014) noticed that concentrations of Cu, Cd and Zn, except Pb, in rice grains were within FAO/WHO (2002) permissible limits for human consumption. Halim et al. (2015) observed that As and Pb uptake in rice grains were 6.87- and 5.26-fold higher than the maximum permissible concentration.

Bioconcentration factor Toxic metal(loid)s are mostly geochemically mobile where they are readily taken up by plant roots and translocated to aerial parts (Satarug et al., 2003). In order to understand the relationship of heavy metal(loid)s in soil−rice system, the BCF value was applied in this study. The BCF value provides a useful indication of the metal(loid) availability from soil to plants. In this study, BCF values of less than 1 were obtained for all the metal(loid)s in the rice grains (Table 2). The mean BCF values for As, Cd, Cu, Hg, Ni, Pb, and Zn were 0.101, 0.121, 0.399, 0.061, 0.033, 0.005, and 0.473, respectively, indicating the availability of heavy metal(loid)s to rice was generally in the order of Zn>Cu>Cd>As>Hg>Ni>Pb. The results showed that the bioavailability of metal(loid)s was low in the investigated paddy fields.

Zhao et al. (2015) reported that the average BCF values for Cd, Cu, Ni, and Zn were 0.0829, 0.0567,

Table 3. Correlation matrix [Pearson correlation coefficients (r)] between metal(loid) concentrations in rice grains and, soil properties and total metal(loid)s in soil.

Metal(loid) in rice	pH	EC	CEC	Total metal(loid) in soil
As	-0.175	-0.151	0.099	-0.040
Cd	0.186	-0.283*	-0.280*	0.031
Cu	0.061	-0.123	-0.075	-0.029
Hg	0.036	-0.002	-0.121	0.567**
Ni	-0.009	-0.047	-0.014	-0.019
Pb	0.056	-0.114	-0.115	0.090
Zn	0.116	-0.172	-0.124	0.058

EC: Electrical conductivity, CEC: Cation exchange capacity
* Correlation is significant at p<0.05; ** Correlation is significant at p<0.0001

0.0041, and 0.1429, respectively, indicating the availability of heavy metal(loid)s to rice was generally in the order of Zn>Cd>Cu>Ni. Kashem and Singh (2001) reported that the accumulation of Cd and Zn was higher than that of Ni in rice. Jung and Thornton (1997) noticed that Cd and Zn concentrations in rice increased with increasing metal(loid) concentrations in paddy soil than that of Cu and Pb. Rice genotypes also influence the transfer and bioavailability of heavy metal(loid)s in the soil-rice system. For instance, Zeng et al. (2008) have reported that hybrid rice accumulated more heavy metal(loid)s than *Japonica* rice under the same soil conditions.

Influence of soil properties and total soil metal(loid)s on metal(loid) availability to rice grains The Pearson correlation coefficient, r, was used to establish the relationship between the concentrations of metal(loid)s in rice grains and, soil properties and total metal(loid)s in soil. The influence of soil properties on the availability of heavy metal(loid)s to brown rice varied among the metal(loid)s. Only Cd and Hg showed some significant correlations amongst the metal(loid)s. Rice grain Cd showed negative and significant correlations with soil EC and CEC (p<0.05) (Table 3). In a recent study, Zhao et al. (2015) also observed negative correlations between EC and metal(loid)s, Cu, Ni, Pb, and Zn in rice grains. The concentration of Hg in rice grains showed a positive and significant correlation only with the total Hg in soil (p<0.0001). Kosolsaksakul et al. (2014) observed a linear relationship between soil total Cd and rice grain Cd. Correlation analysis by Zhang et al. (2013) showed that As concentration in rice had a significant positive correlation with soil As concentration (R^2= 0.759, p <0.01), a negative correlation with soil pH, K, Ca, and Mg contents, and a positive correlation with soil OM and iron content.

Zhao et al. (2010) used cross-correlograms and illustrated that BCF values (availability of metal(loid)s, Cu, Cd, Pb, and Zn to rice grains) were significantly correlated with most soil properties, among which soil pH and OM had the strongest correlations with BCFs. However, BCF value of Cu showed relative weak correlations with soil properties; particularly soil pH and OM had no correlations with BCF of Cu, indicating that the availability of Cu may be influenced by other factors. Feng et al. (2012) found the spatial variation of the heavy metal(loid)s contents in rice grains was mainly affected by the soil OM or soil pH. However, in this study, the metal(loid) concentrations in rice grains showed weak correlations with the investigated soil properties and total metal(loid)s in soil.

Conclusion

Heavy metal(loid) contamination in agriculture soils and their transfer in soil–rice system have been of increasing concern. This study provides useful information on the heavy metal(loid) concentrations in major rice growing soils and brown rice in Korea. The results indicate that the heavy metal(loid) levels in all sampled paddy soils are within the permissible limits as per the Korean Soil Environment Conservation Act. The levels of metal(loid)s in brown rice are also generally within the acceptable limit for human consumption. The BCF values in this study indicate that the rice grains only absorb and does not accumulate the metal(loid)s. While the statistical analysis provided useful information about the relationship between metal(loid)s in grains as affected by the soil properties and total metal(loid)s in soil, the bioavailability may also be influenced by other factors such as organic matter in soil. Further study on soil properties is required to understand its influence on the bioavailability of metal(loid)s in rice grains.

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