Bull Environ Contam Toxicol (2007) 79:209–213
DOI 10.1007/s00128-007-9164-0

Uptake of Toxic Heavy Metals by Rice (*Oryza sativa* L.) Cultivated in the Agricultural Soil near Zhengzhou City, People's Republic of China

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Received: 12 April 2007 / Accepted: 1 June 2007 / Published online: 17 July 2007

Abstract Higher accumulation of toxic heavy metals in rice grown in agricultural soil may lead to health disorder. A field experiment was carried out to investigate uptake and translocation of Cd, Cr, Pb, As, and Hg by different parts of rice plant in various irrigation regions. The results showed the rice grain contained significantly lower amounts of five metals than straw and root in all sampling sites. Rice root accumulated Cd, As, and Hg from the paddy soil. Moreover, the rice plant transported As very weakly, whereas Hg was transported most easily into the straw and grain among studied heavy metals.

Keywords Heavy metal uptake · Translocation · Bioconcentration factor · Rice

The potential public health risk associated with dietary intake of heavy metals has become of increasing concern. Intake of heavy metals via the soil–crop system has been considered as the predominant pathway of human exposure to environmental heavy metals. Among the heavy metals, cadmium (Cd), chromium (Cr), lead (Pb), mercury (Hg) and arsenic (As) are commonly considered as toxic to both plants and humans (Rahman 1999; Gibb and Chen 1989; Trichopoulos 1997). Therefore, some regulations to restrict the emission of heavy metals and the tolerance limit in food have been set up in many countries or areas. It is necessary to decrease toxic heavy metal accumulation in cereals for food production, particularly in rice, which is one of the most frequently consumed cereals worldwide.

Bioavailability of heavy metals is mainly affected by total content of soil heavy metal, soil chemical and physical properties and plant species (Cheng et al. 2006; Rubio et al. 1994). In recent years, researchers have focused their attention on the significance of soil types and genotype on the uptake and accumulation of heavy metal in the pot experiment or hydroponic solution (Li et al. 2005; Lee et al. 1998; Du et al. 2005). However, the pot experimental measurement and hydroponic solution result may not predict uptake of heavy metal by crop under actual field conditions. Accordingly, it is important to study heavy metals accumulation in natural environment. Single heavy metal pollution occurs in nature occasionally, but compound pollution of multiple metals is a more common phenomenon. Interactions between different elements create a different toxic effect on an ecosystem compared to that of single pollutant. Therefore, in the present research, we performed a comprehensive study of toxic heavy metals in rice plant from the agricultural soil under natural condition. The objective of this study was to illuminate heavy metal uptake by the paddy crops growing naturally, and various heavy metal translocation in different parts (root, straw, and grain) of rice plant under real field conditions.

Materials and Methods

The growing period of rice plant (*Oryza sativa* L.) is about 155 days (May–November) in the suburb of Zhengzhou city, Henan Province, China (34°42′N; 113°45′E; altitude 110.4 m). The area has a warm spring continental weather

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with an annual average temperature and rainfall of 14.3°C and 640.5 mm, respectively. Rice Plant is broadly cultivated, and has a large abundance yield in the area. Moreover, most of the rice product enters the local market, supplying for a population of approximately 2,000,000 inhabitants.

Rice plant was sampled during maturity from eight sites in November 1998, which were divided into three districts: municipal sewage irrigation region (MSIR) (Chengang, Xuzhuang, and Jiagang), Yellow River irrigation region (YRIR) (Zhaolanzhuang, and Jingshuicun), and groundwater irrigation region (GWIR) (Xincun, Ershilipu, and Huayuankouxiliuhuangcun). Additionally, the corresponding soils (at 0–20 cm in depth) were collected. Sampled plants were separated into roots, straws and grains, and then washed three times with distilled water and finally rinsed with deionized water and dried in an oven at 65°C. Dry weights were determined and the plant parts ground with a tissue grinder. The soil samples were air-dried at room temperature, finely powdered and sieved through 2 mm nylon mesh to remove large debris, stones and pebbles. Soil samples (500 g) were dried at 105°C for 2 h and ground to pass through 60 mesh sieve and homogenized for analysis.

Metals As, Hg, Cd, Pb and Cr were determined according to previously described methods (Schuhmacher et al. 1993). A microwave assisted digestion procedure was used. About 0.5–3 g of homogenized samples was digested under pressure in Teflon vessels with 4 mL of nitric acid and 1.5 mL of hydrogen peroxide. On completion of the digestion and after adequate cooling, solutions were filtered and made up to 50 mL with 1% nitric acid.

Metals Cr, Cd, and Pb contents were analyzed by flame atomic absorption spectrometry (FAAS, Hitachi Z-8000, Hitachi Ltd., Tokyo, Japan), whereas concentrations of Hg and As were determined using cold-vapor atomic absorption spectrometry (CV-AAS) with a hydride generation VA-90 model (TongJi University, China) and sodium borohydride as the reductant. All reagents were supra-pure and high-purity water was employed throughout. A sample of standard reference material (NIST SRM 2709), a blank, and a determination in duplicate were included for assurance of analytical accuracy. The analytical results showed no signs of contamination and that the precision and bias of the analysis were generally <10% for metals. The recovery rates for heavy metals in SMR were around 85%–105%.

Results and Discussion

Heavy metals concentrations in soils and different parts of rice plants are presented in Table 1. The average concentrations in agricultural soils ranged 0.11–0.14 mg kg^{-1} for Cd, 57.54–65.67 mg kg^{-1} for Cr, 14.42–19.28 mg kg^{-1} for Pb, 0.041–0.10 mg kg^{-1} for Hg, and 6.10–6.97 mg kg^{-1} for As, respectively, which were below the threshold levels in natural background soil as defined by China (National Environmental Protection Agency of China, GB15618, 1995). Therefore, the paddy soils were uncontaminated with heavy metals. Concentrations of Cd, Cr, Pb and As in paddy soils did not vary significantly among the different irrigation regions, while content of Hg was higher in MSIR and YRIR than GWIR. Irrigation of municipal sewage did not result in the heavy metal pollution in the soil. This was attributed to municipal sewage with low heavy metal content and high organic matter (BOD 53.8–74.8 mg L^{-1}). Compared with vegetable soils, concentration of Cr was higher in the paddy soils, whereas content of Hg was very much lower (Liu et al. 2006).

Average Cd values in rice roots were 6.20, 2.10, and 0.11 mg kg^{-1} for MSIR, YRIR, and GWIR, respectively. Cadmium concentration of root showed significant difference between various irrigation regions. Average Cd concentration in rice grains was 0.016 mg kg^{-1} for MSIR, 0.038 mg kg^{-1} for YRIR, and 0.0079 mg kg^{-1} for GWIR. Among the different analyzed parts of the paddy crop (root, straw and grain), the lowest value of Cr was observed for the grain (0.22 mg kg^{-1} for MSIR, 0.18 mg kg^{-1} for YRIR, and 0.37 mg kg^{-1} for GWIR). Average values of Pb in rice roots were 7.14, 8.34, and 3.90 mg kg^{-1} for MSIR, YRIR, and GWIR, respectively. Among root, straw and grain, the lowest value of Pb was also observed for the grain (0.53 mg kg^{-1} for MSIR, 0.33 mg kg^{-1} for YRIR, and 0.16 mg kg^{-1} for GWIR).

The arsenic average level varied from 20.87 to 23.89 mg kg^{-1} in roots, from 0.40 to 0.93 mg kg^{-1} in straws, and from 0.062 to 0.10 mg kg^{-1} in rice grains. Arsenic concentrations in the grains were remarkably less than that in the roots and straws of the paddy crops in various irrigation regions. Root of paddy from uncontaminated agricultural soil accumulated the highest arsenic followed by straw and rice-grain, which was in agreement with the results of Alam et al. (2003). Alam et al. found the quantity of accumulation arsenic concentration was in the order: grain < husk < leaf < stem in rice plant from arsenic contaminated irrigation water. The concentrations of Cd, Pb, Cr and As in the edible grain were well below the Chinese national food guideline limit. Thus, the rice grain was uncontaminated with Cd, Pb, Cr and As.

Ranges of Hg concentration were 0.078–0.16 mg kg^{-1} for root, 0.045–0.062 mg kg^{-1} for straw, and 0.028–0.034 mg kg^{-1} for grain among the various irrigation regions. It is worth noticing that Hg content of the grain exceeded the Chinese food guideline limit of 0.02 mg kg^{-1}. This showed the rice grain was slightly contaminated by Hg. Nevertheless, the contents of the studied heavy metal

Table 1 Concentrations of heavy metals in the soils and rice plants (mg kg^{-1} dw)

	MSIR (n = 15)		YRIR (n = 10)		GWIR (n = 15)	
	Range	Mean	Range	Mean	Range	Mean
Cd						
Soil	0.11–0.17	0.14	0.10–0.13	0.12	0.11–0.11	0.11
Root	6.08–6.31	6.20	1.08–3.12	2.10	0.10–0.11	0.11
Straw	0.051–1.05	0.71	0.84–1.03	0.94	0.043–0.060	0.052
Grain	0.014–0.22	0.016	0.011–0.065	0.038	0.0029–0.013	0.0079
Cr						
Soil	50.23–67.52	60.47	59.55–71.80	65.67	51.49–62.10	57.54
Root	2.73–3.28	3.01	2.04–3.10	2.57	0.70–2.28	1.99
Straw	0.61–0.91	0.77	0.22–0.30	0.48	0.33–0.71	0.52
Grain	0.036–0.38	0.21	0.066–0.30	0.18	0.30–0.44	0.37
Pb						
Soil	14.57–21.43	18.34	14.66–23.99	19.28	12.38–16.46	14.42
Root	6.56–7.98	7.14	7.24–9.45	8.34	2.73–5.07	3.90
Straw	1.62–2.53	2.05	1.93–2.01	1.97	1.71–2.31	2.01
Grain	0.47–0.56	0.53	0.046–0.62	0.33	0.047–0.27	0.16
Hg						
Soil	0.057–0.14	0.10	0.069–0.13	0.090	0.035–0.046	0.041
Root	0.11–0.19	0.14	0.069–0.088	0.078	0.15–0.19	0.16
Straw	0.030–0.054	0.045	0.041–0.051	0.046	0.044–0.080	0.062
Grain	0.022–0.046	0.031	0.034–0.035	0.034	0.025–0.032	0.028
As						
Soil	5.34–7.64	6.81	6.08–6.12	6.10	4.76–8.74	6.97
Root	19.07–30.42	23.89	22.58–23.35	22.97	20.07–21.68	20.87
Straw	0.66–1.09	0.93	0.52–0.53	0.52	0.16–0.65	0.40
Grain	0.069–0.12	0.099	0.091–0.11	0.10	0.049–0.074	0.062

in the edible rice grain were lower than that in the edible vegetable cultivated in agricultural soil (Liu et al. 2006).

The bioconcentration factor (BCF) was calculated as the ratio of content of heavy metal in the plant or the part of plant to that in the soil. Figures 1 and 2 show the BCF values for Pb, Cd, Cr, Hg, and As in paddy plant. When a BCF ≤ 1, it shows that the plant can only absorb but not accumulate heavy metals; when a BCF > 1, it shows that plant can accumulate metals. BCF range of Pb in the paddy roots was 0.27–0.43, with average BCF of 0.36. BCF range of Pb was 0.10–0.14 for straw, and 0.011–0.029 for grain. The results indicated that Pb bioavailability was low. In general, the percent of exchangeable Pb in the soils was very low. Heavy metals in the exchangeable and carbonated-bound fractions are considered readily and potentially bio-available, while the Fe–Mn oxide and organic/sulphide fractions are relatively stable under normal soil conditions (Wong et al. 2002). Nevertheless, lead in the crop soils was largely associated with the Fe–Mn oxide phase, followed by the organic/sulphide and residual fractions, therefore Pb bioavailability was not high.

Chromium BCF value range in different irrigation regions was 0.035–0.050 (root), 0.007–0.013 (straw), and 0.0028–0.0065 (grain). Chromium present in the forms Cr(VI) or Cr(III) in the soils, and the toxicity and mobility of Cr(VI) are higher than those of Cr(III). Soil redox potential can influence the distribution of Cr between Cr(VI) and Cr(III) forms. Cr(VI) reduction occurred in the flooded soils, thus Cr(VI) can be transformed to low-solubility cationic forms via reduction Lee et al. 2005. Therefore, Cr bioavailability was very much low in the paddy soil. Additionally, Cr uptake by rice plant was not significantly different among the various irrigation regions.

BCF value for As ranged from 3.00 to 3.76 (root), from 0.058 to 0.14 (straw), and from 0.009 to 0.017 (grain) in different irrigation regions. Average BCF of As for rice root was 3.46, which was above 1. This indicated that rice roots enriched in As, as compared with the soil. It is known that rice maintain relatively high redox potentials in the rhizosphere by a continuous flux of O_2 from the shoots toward the roots. The release of O_2 enables the accumulation of Fe-oxyhydroxide in the rhizosphere of living plants (Bigham et al. 2002; Wang et al. 1993). Consequently, rice roots seem to adsorb and concentrate As on the rich mineral coating. A similar situation was reported by Hansel et al. (2002) who demonstrated a spatial

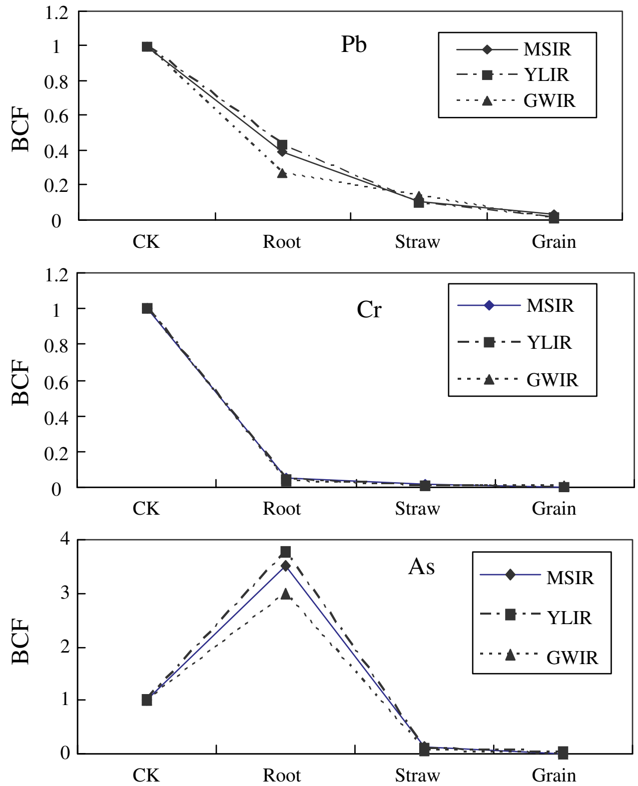

Fig. 1 BCF values of Pb, Cr and As in rice plants from the suburb of Zhengzhou, China

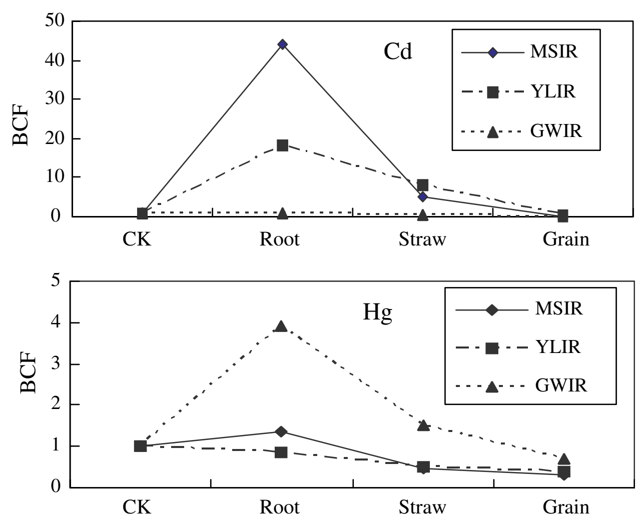

Fig. 2 BCF values of Cd and Hg in rice plants

correlation between Fe and As in roots of aquatic plant by means of XRF microtomography.

BCF range of Cd was 0.97–44.13 for root, 0.47–8.05 for straw, and 0.071–0.32 for grain. Cd uptake by rice root was significantly different between irrigation regions, whereas no significant difference for Pb was found. The Cd uptake by paddy roots has the relation: MSIR > YRIR > GWIR. It

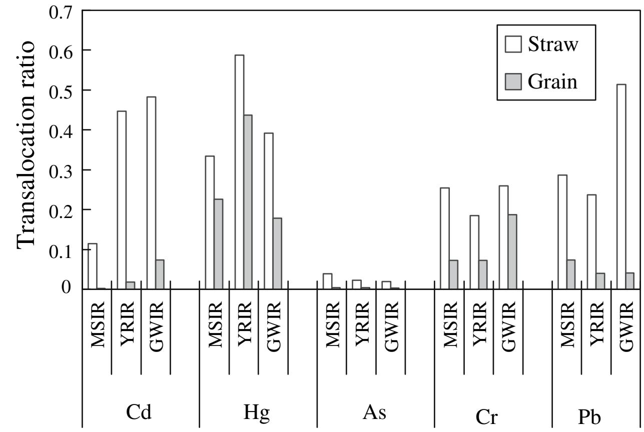

Fig. 3 Translocation ratios of metals in the straws and grains of paddy plant

is possible that the organic contaminant in municipal sewage enhanced Cd uptake. Zarcinas et al. (2004) also reported that Cd uptake was strongly correlated with organic matter. Rice root accumulated high quantities of Cd^{2+} when grown in non-pollution areas as in a medium containing this metal (Rubio et al.1994). This metal, as divalent cation, may compete with other cation (including Ca^{2+}, Mg^{2+}, Fe^{2+}) in their transport across membranes. In addition, Cadmium was more easily taken up and accumulated than Pb by rice plants through the root systems from soil. Other authors have also indicated that Cd^{2+} is more bio-available to plant than other heavy metal, such as Zn, Cu, and Pb, having a higher biological absorption coefficient (Lee et al. 1998).

BCF range of Hg was 0.86–3.9 for root, 0.45–1.53 for straw, and 0.31–0.70 for grain. Significant difference for Hg uptake was observed in various irrigation regions. However, the exact mechanisms are largely unknown. In conclusion, the heavy metals uptake by paddy plants showed the greatest accumulation of Cd, As, and Hg in the roots, although there are different in various irrigation regions. However, Pb and Cr were hardly taken up by the rice root. The heavy metal uptake by paddy roots was in the following order: Cd > As > Hg > Pb > Cr. On the other hand, BCF values of Pb, Cr, As and Cd in edible rice grain were lower than that in edible vegetable from the suburb of Zhengzhou city (Liu et al. 2006). This showed that rice grain could take up less heavy metal than edible vegetable, with the exception of Hg.

Further, translocation ratios ($HM_{straw\ or\ grain}/HM_{root}$), from root to straw or grain, were calculated for each heavy metal. Figure 3 showed translocation ratios of Cd, Hg, As, Cr, and Pb in the rice plant in various irrigation regions, and all values were below 1. This is because most of the heavy metals are often confined in the roots after paddy plant uptake. Other than the roots, the aerial organs such as straws and grains also display competence in absorbing heavy metals. Translocation ratio of straw was more than that of grain for each heavy metal in the different irrigation regions. For five toxic heavy metals, absorption of rice plant had the relation: root > straw > grain. Additionally, the results revealed that the rice plant transported As very weakly into the straw and grain, whereas Hg was transported most easily into the straw and grain of rice among studied heavy metals.

Acknowledgments This research was supported by Technology Program of Henan province of China (0424007, 981190704).

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