Pol. J. Environ. Stud. Vol. 24, No. 3 (2015), 1379-1386
DOI: *10.15244/pjoes/35282*

Original Research

Health Risks of Heavy Metals Uptake by Crops Grown in a Sewage Irrigation Area in China

Zuwei Wang[1], Xiangfeng Zeng[2, 4]*, Mingshuo Geng[1], Chunyi Chen[3], Jianchao Cai[5], Xiaoman Yu[2, 4], Yingying Hou[1], Hui Zhang[1]

[1]Tianjin Key Laboratory of Water Resource and Water Environment, Tianjin Normal University, Tianjin 300387, China
[2]Key Laboratory of Pollution Ecology and Environmental Engineering, Institute of Applied Ecology, Chinese Academy of Sciences, Shenyang 110016, P.R. China
[3]Center for Environmental Biotechnology, University of Tennessee, Knoxville 37996, USA
[4]University of Chinese Academy of Sciences, Beijing 100039, P.R. China
[5]Institute of Geophysics and Geomatics, China University of Geosciences, Wuhan 430074, P.R. China

Received: 30 November 2014
Accepted: 29 December 2014

Abstract

Ecological risks of heavy metal toxicity such as Cd, Cu, Pb, Zn, Cr, As, and Hg through crops (wheat and rice) grown in the Tianjin sewage irrigation area in northern China were studied in this paper. Wheat and rice samples as well as related soil samples from 77 select blocks were collected. The second grade of standards for Tianjin soil environmental quality was used for soil risk assessment. Chinese National Food Safe Criterion was used for health risk assessment of wheat and rice grains. Daily intake rate and Target hazard quotient were used for the potential health risk assessment of local population through the intake of wheat and rice grown in the sewage-irrigated site. The results showed that continuous application of wastewater has led to accumulation of heavy metals in the soil, and Cd, Zn, and Hg were the main pollutants. Zn and Cd were more mobile than other metals. Pb in wheat and rice had an ecological risk to human health. As and Hg in some rice samples as well as Cd, Zn, and As in some wheat samples had potential risk. Target hazard quotient (THQ) of individual metal was below 1.0, meaning the relative absence of health risks associated with intake of a single heavy metal through intake of either wheat or rice. THQs of As for wheat and rice would sum up to above 1.0, indicating As may pose a risk to the local population by intake of wheat and rice.

Keywords: heavy metals, health risks, crops, soil, Tianjin

Introduction

Wastewater irrigation is a widespread practice in the world and long-term wastewater irrigation may lead to the accumulation of heavy metals in agricultural soils and plants [1-9]. Although some of the heavy metals such as Zn, Mn, Ni, and Cu act as micronutrients at lower concentrations, they become toxic at higher concentrations. The health risks from heavy metal contamination of soil have been widely reported [10, 11]. Crops and vegetables grown in soils contaminated with heavy metals have greater accumulation of heavy metals than those grown in uncontaminated soil [12-14]. Intake of crops is an important path of heavy metal toxicity to humans.

Dietary intake of heavy metals through contaminated crops may lead to various chronic diseases. Duruibe et al. suggested that biotoxic effects of heavy metals depend upon the concentrations and oxidation states of heavy metals, the kinds of sources, and the modes of deposition [15]. Severe exposure to Cd may result in pulmonary effects

*e-mail: xf6.zeng@gmail.com

such as emphysema, bronchiolitis, and alveolitis. Renal effects may also result from subchronic inhalation of Cd [16]. Pb toxicity causes reduction in the haemoglobin synthesis; disturbance in the functioning of kidneys, joints, and reproductive and cardiovascular systems; and chronic damage to the central and peripheral nervous systems [17]. Higher concentrations of Zn can cause impairment of growth and reproduction [18].

In Tianjin region, wastewater irrigation is a choice to resolve the scarcity of agricultural water. But a large amount of matter enters the soils at the same time, polluting the soil environment and then threatening lives through the food chain. There has been research about heavy metals pollution of the Tianjin sewage-irrigated site. Wang et al. and Wu et al. studied heavy metals pollution in the sewage-irrigation area soil of Dagu sewage discharge channel [19, 20]. Shi et al., Zhai, and Dong discussed the risks of heavy metals on soil and vegetables of sewage-irrigated sites in Tianjin suburbs [21-23]. Li analyzed heavy metals pollution of soil and wheat at Beijing sewage discharge channel sewage-irrigated sites [24]. Ma et al. studied heavy metal concentrations of winter wheat at each stage in typical sewage-irrigated sites of Tianjin [25]. Although wheat and rice are important food crops in Tianjin, there is no published research on health risks of heavy metals uptake through wheat and rice grown in Tianjin sewage-irrigated sites simultaneously. In the present study, health risk was ascertained through the calculation of different hazard quotients. Health risk caused by the daily intake of heavy metals through contaminated wheat and rice was also assessed.

Materials and Methods

Study Sites

The study was conducted around Tianjin sewage-irrigated sites, which have a 20-year history of sewage irrigation. Tianjin sewage-irrigated sites include three main sites of sewage discharge channel irrigation, namely the Beijing sewage discharge channel sewage irrigation area (BSIA), the Dagu sewage discharge channel sewage irrigation area (DSIA), and North sewage discharge channel sewage irrigation area (NSIA) (Fig. 1).

BSIA is located mainly in Wuqing and Beichen districts, and part in Baodi and Ninghe districts, with an irrigation area of 8.35×10^4 hm^2. The wastewater for irrigation is industrial wastewater and domestic sewage coming from Beijing. DSIA is located in Xiqing District and Jinnan District with an irrigation area of 2.33×10^4 hm^2, and the wastewater for irrigation came from Tianjin urban and Xiqing District. NSIA is located in Dongli District with an irrigation area of 1.2×10^4 hm^2, and the wastewater for irrigation is industrial wastewater coming from Dongli District.

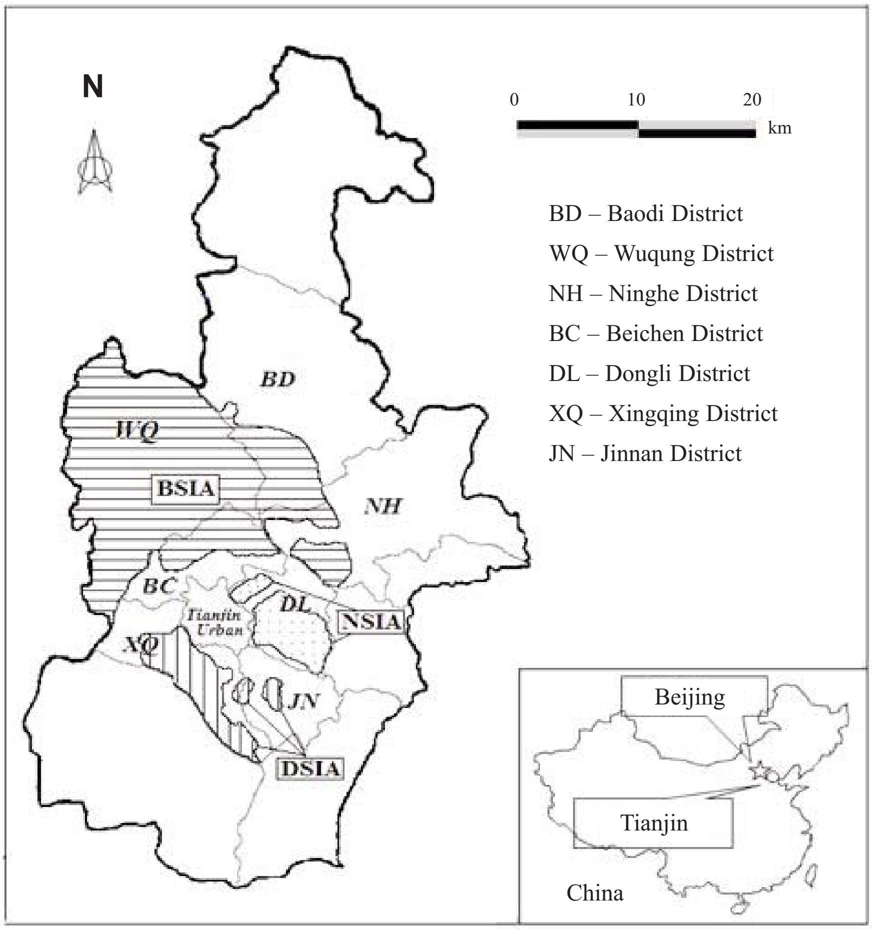

Fig. 1. A study site map showing the locations of TSIA and related districts.

In recent years, the sewage-irrigated sites were reduced due to the reduction of the wastewater amount from Beijing caused by Chinese national environmental policy and the advancement of wastewater treatment techniques.

Sampling and Analysis Methods

Representative 24 wheat samples and 29 rice samples grown in Tianjin wastewater irrigation area (WWI) and 10 wheat samples and 14 rice samples grown in clean water irrigation area (CWI) were collected randomly from a 5 m×5 m area of different fields in harvest time using the cinquefoil 5-point snakelike sampling method.

After washing with clean tap water to remove the soil particles, wheat and rice samples were oven dried at 80ºC to constant weight. The dried samples were ground, passed through a 2 mm sieve, and stored in plastic bags at room temperature before analysis.

At the same time, related soil samples were sampled by digging out a monolith of 10 cm×10 cm×15 cm size from the A layer soil and the C layer soil. After transport to the laboratory, samples were air dried, crushed, passed through a 2 mm mesh sieve, and stored at ambient temperature for analysis.

For extraction of heavy metals such as Cd, Cu, Pb, Zn, and Cr, 1 g dried sample of plant or soil was digested in 15 ml of HNO_3, H_2SO_4, and $HClO_4$ mixture (5:1:1) at 80ºC until a transparent solution was obtained [26]. Water samples (50 ml) were digested with 10 ml of concentrated HNO_3 at 80ºC until the solution became transparent. These transparent solutions were then filtered through Whatman No. 42 filter papers and diluted to 50 ml with distilled water. The concentrations of Cd, Cu, Pb, Zn, and Cr in the filtrate were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES, Leeman Labs, USA), fitted with a specific lamp of particular metal using appropriate drift blanks. As and Hg concentrations were determined by atomic florescence (AFS-230).

Quality Control Analysis

The identification of pollutant sources is conducted with the aid of multivariate statistical analyses, such as correlation analysis, principal component analysis (PCA), and cluster analysis. Multivariate statistical analyses of the data in this work were performed with SPSS 12.0 for Windows (SPSS Inc., Chicago, IL).

Data Analyses

Transfer Factor (TF)

Transfer factor (TF) was calculated to understand the extent of risk and associated hazard due to wastewater irrigation and consequent heavy metal accumulation in an edible portion of test wheat and rice following Cui et al. [27]:

$$TF = \text{concentration of metal in edible part} / \text{concentration of metal in soil}$$

Daily Intake Rate (DIR)

Daily intake was calculated by the following equation:

$$DIR = (C_{metal} \times D_{food\ intake}) / B_{average\ weight}$$

...where C_{metal} (mg·kg^{-1}), $D_{food\ intake}$ (kg·person^{-1}), and $B_{average\ weight}$ (kg·person^{-1}) are the heavy metal concentrations in wheat grains and rice grains, daily intake of wheat grains and rice grains, and average body weight, respectively [28, 29]. In the paper, $D_{food\ intake}$ are 0.242 (kg·person^{-1}) for wheat and 0.235 (kg·person^{-1}) for rice [30], separately. $B_{average\ weight}$ is 55.9 kg [29].

Target Hazard Quotient (THQ)

For the assessment of health risks through consumption of wheat and rice grains by the local inhabitants, THQ was calculated following the methodology described by USEPA (USEPA 2000). THQ was determined based on formula [31]:

$$THQ = 10^{-3} (E_F E_D F_{IR} C / R_{FD} W_{AB} T_A)$$

...where E_F is exposure frequency (365 days·year^{-1}) [32]; E_D is the exposure duration (70 years), equivalent to the average lifetime [33]; F_{IR} is the food ingestion rate (kg·person^{-1}·day^{-1}); C is the metal concentration in food (mg·kg$^{-1)}$; R_{FD} is the oral reference dose (mg·kg^{-1}·day^{-1}) from USEPA (2000) [34]; W_{AB} is the average body weight (55.9 kg), and T_A is the average exposure time for noncarcinogens (assuming 70 years in this study).

Results and Discussion

Levels of Heavy Metals in Soil

Continuous application of treated and untreated sewage water to the soil led to higher concentrations of heavy metals in the soil at the WWI site as compared to the CWI site (Table 1). Li has also found higher concentrations of heavy metals in sewage effluents as compared to clean water irrigation [24].

Among all the heavy metals at Tianjin wastewater irrigated site, the concentration of Zn was found to be maximum and Cd was minimum in sewage-irrigated soils. Wang et al., Dong, and Li found similar trends of highest concentrations of Zn and lowest concentrations of Cd in soils [19, 23, 24].

The concentrations of Cd and Hg in Tianjin sewage-irrigated soils were higher than those reported by Wang et al. and Dong, and the concentrations of Cu, Pb, and Cr were lower [19, 23].

Heavy metals enriched in soils at the WWI sites compared to CWI sites (Fig. 2) shows that Cd and Hg in soils were enriched obviously, whereas Cu, Pb, Cr, As, and Zn were enriched slightly.

Table 1. Concentrations of heavy metals in soils and crops such as rice and wheat at the WWI sites.

Element		Cd	Cu	Pb	Zn	Cr	As	Hg
Soil	range	0.05-1.17	10.9-61.3	3.8-49.79	62.2-333	40.2-108	5.14-17.7	0.035-1.72
	mean	0.46	28.15	15.62	129.05	64.19	11.23	0.52
	SD	22.29	9.45	6.34	56.23	12.25	2.88	0.41
Wheat	range	0.025-0.176	2.15-4.16	0.06-0.24	16.21-53.0	0.28-0.62	0.096-0.20	0.001-0.015
	mean	0.062	2.99	0.14	27.67	0.49	0.128	0.013
	SD	0.020	0.51	0.05	7.33	0.10	0.059	0.006
Rice	range	0.013-0.215	2.62-6.67	0.26-1.73	14.0 -44.3	2.56-5.47	0.046-0.38	0.001-0.257
	mean	0.07	4.45	0.62	22.73	3.91	0.166	0.022
	SD	0.025	0.59	0.104	2.14	0.64	0.079	0.005

The concentrations of heavy metals in soils are not only related to human activity levels, but controlled by soil texture. The use of a ratio of concentration of heavy metals in the A and C layers can eliminate the influence of soil texture on concentrations of heavy metals. The A/C value of concentration of heavy metals is shown in Fig. 3.

A/C ratio of Cr and As in soil was close to 1, indicating they had been not affected by sewage irrigation. A/C ratios of Cu, Pb, Zn, and Cd in soils were between 1.0 and 2.0, meaning they had some enrichment in soil after sewage irrigation. A/C ratios of Hg were above 2.0, and it enriched obviously in soil after sewage irrigation.

Among heavy metals at BSIA, concentrations of Cu, Pb, Zn, and Cr were slightly lower than those reported by Wang [35], but Cd concentration was slightly higher. Compared with the heavy metal concentrations in soils at the CWI site, the concentrations of heavy metals in BSIA soil were higher by 64.6% for Cd, 18.7% for Cu, 57.5% for Pb, 37.6% for Zn, 24.6% for Cr, 46.6% for As, and 154.3% for Hg.

Among heavy metals concentrations in soils at the DSIA site, the concentrations of Cd, Zn, Cr, As, and Hg in soils were higher than those reported by Wu et al., but Pb was lower [20]. The concentrations of heavy metals in soils were higher by 243.5% for Cd, 28.8% for Cu, 36.5% for Pb, 85.4% for Zn, 44.4% for Cr, 7.0% for As, and 105% for Hg.

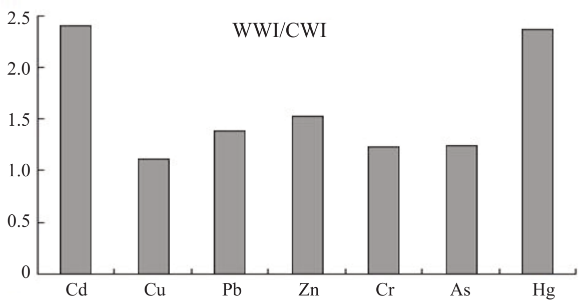

Fig. 2. Enrichment of heavy metals in soils of Tianjin sewage-irrigated sites.

Among heavy metals in soils at the NSIA site, the concentrations of Cu, Pb, Zn, Cr, and As were lower than those reported by Huang et al., but Cd was higher [36]. The concentrations of heavy metals in soils at the NSIA site were higher by 381.3% for Cd, 17.6% for Cu, 50.7% for Pb, 64.9% for Zn, 15.4% for Cr, 12.8% for As, and 133.3% for Hg.

The concentrations of heavy metals in soil at DSIA and NSIA sites were higher than the concentrations at BSIA site, and the reason may be that there are more rice fields in DSIA and NSIA, and the intensity of sewage irrigation for rice fields was higher (Fig. 4).

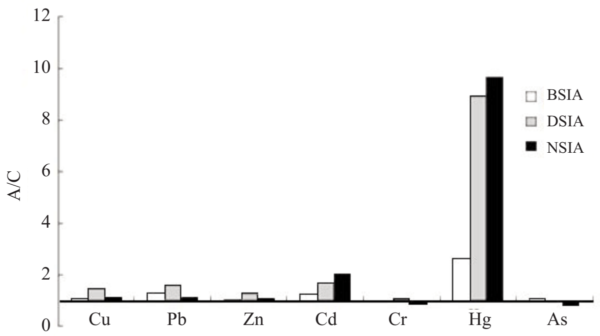

Fig. 3. A/C value of heavy metals concentrations in soils.

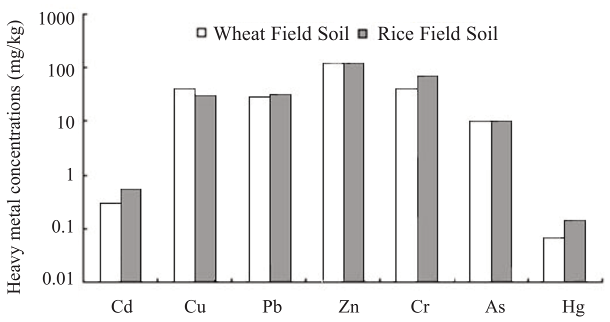

Fig. 4. Mean heavy metal concentrations in wheat and rice field soil at the WWI site.

Table 2. Guidelines for safe limits of heavy metals (mg·kg^{-1}).

	Standards		Cd	Cu	Pb	Zn	Cr	As	Hg
Soil	Chinese Standards (second grade) (GB 15618-1995)		0.6	100	350	300	250_D 350_P	25_D 20_P	1.0
	Tianjin Standard (second grade)		0.159	43.71	32.83	115	107_D 124_P	16.64_D 14.64_P	0.258
	European Union Standard (EU2002)		3.0	140	300	300	150	-	-
	WHO/FAO		-	-	-	-	-	-	-
Plant	Chinese Standard (GB 2762-2005)	wheat	0.1	10	0.2	50	20	0.15	0.02
		rice	0.2	10	0.2	50	-	0.15	0.02
	European Union Standards (EC: No. 629/2008)		0.2	-	0.2	-	-	0.1	0.02
	WHO/FAO		0.2	40	5	60	-	0.15	0.02

D – upland soils, P – paddy soil

The concentrations of heavy metals such as Cd, Cu, Pb, Zn, Cr, As, and Hg in soils at the WWI site were below the permissible limits of Chinese and EU standards. The concentrations of Cd and Hg clearly exceeded the permissible limits of level of the second grade of the Tianjin Standard (Table 2) and the concentration of Zn was slightly higher, suggesting that Cd and Hg were important pollutants in the soils. They had potential ecological risks, and Zn had, to some extent, ecological risks.

Levels of Heavy Metals in Wheat Grains and Rice Grains

Levels of Heavy Metals in Wheat Grains

All the heavy metal concentrations in wheat grains at the WWI site were much higher compared to the CWI site (Fig. 5). Li and Ma et al. also found higher concentrations of heavy metals in wheat grown under wastewater irrigation as compared to those at the CWI site [24, 25]. The concentrations of heavy metals in wheat grain grown in the CWI site were higher by 63.2% for Cd, 3.8% for Cu, 100% for Pb, 6.6% for Zn, 11.4% for Cr, 326.7% for As, and 18.2% for Hg.

Among heavy metals in wheat grains at the WWI site, concentrations of Zn were highest and Hg lowest. Similar results were reported by Yang for wheat grain grown in Liangfeng sewage-irrigated sites (Beijing) [37]. The concentration of Zn of wheat grain was higher than that reported by Zhai [22]. The concentrations of other heavy metals such as Cd, Cu, Pb, and Cr were lower than those reported by Zhai [22].

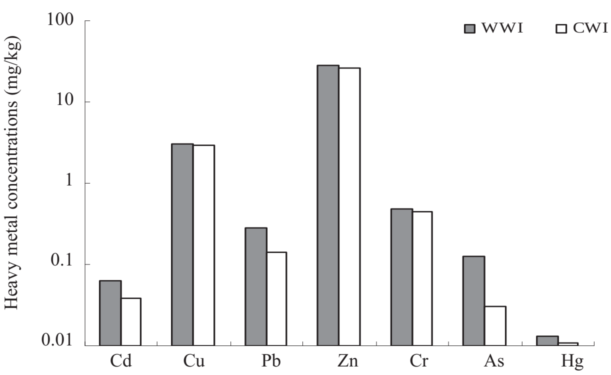

Fig. 5. Mean concentrations of heavy metals in wheat grains.

Compared with permissible limits of Chinese Standards (GB 15618-1995), the mean concentrations of Cd, Cu, Zn, Cr, As, and Hg in wheat grains were lower at the WWI sites, whereas Pb concentration was higher by 40%, indicating Pb in wheat grains has a potential ecological risk to human health. Maximum concentrations of heavy metals, such as Cd, Zn, and As in some wheat samples exceeded permissible limits of Chinese Standards, suggesting they may pose a potential ecological risk to human health.

The mean concentrations of heavy metals were also lower as compared to the safe limits given by WHO/FAO (WHO/FAO, 2007) and the safe limits given by commission regulation (EU, 2008) except for Pb and As (Table 2) [38].

Levels of Heavy Metals in Rice Grains

Among heavy metals in rice grains at the WWI site, concentration of Zn was highest and Hg was lowest, followed by Cd, As, Pb, Cr and Cu. The mean concentrations of Zn and Cu were higher than that reported by Zhai but Cr was lower [22].

All the concentrations of heavy metals in wheat grains at the WWI site were higher as compared to the CWI site (Fig. 6), and the concentrations of heavy metals in rice grains were higher by 27.3% for Cd, 18.7% for Cu, 65.4% for Pb, 21.6% for Zn, 46.4% for Cr, 54.9% for As, and 40% for Hg.

The concentrations of Cd, Cu, Zn, and Cr in rice were lower as compared to the safe limits given by commission regulations (EU, 2008) and Chinese Standards (GB 15618-1995), except for Pb, As, and Hg. The concentration of Pb was higher by 115%, indicating that Pb in wheat grains may have an ecological risk to human health (Table 2).

Table 3. TF, DIR, and THQ values of crops.

Elements		Cd	Cu	Pb	Zn	Cr	As	Hg
TF	Wheat	0.111	0.099	0.008	0.214	0.007	0.012	0.087
	Rice	0.242	0.105	0.015	0.181	0.095	0.011	0.100
DIR	Wheat	0.0002	0.0115	0.0011	0.1068	0.0019	0.0005	0.00005
	Rice	0.0003	0.0187	0.0018	0.0956	0.0164	0.0005	0.00003
	PTDI*	60	300	214	60	–	–	–
THR	Wheat	0.0980	0.1181	0.0111	0.1457	0.0005	0.6742	0.0411
	Rice	0.1074	0.1707	0.1650	0.1163	0.0040	0.5626	0.0215
	∑	0.2054	0.2888	0.1761	0.2620	0.0045	1.2368	0.0627

*Joint FAO/WHO Expert Committee on Food Additives, 1999; PTDI – Potential tolerable daily intake

The mean concentrations of As and Hg in rice were slightly higher than the safe limits given by commission regulations (EU, 2008) and Chinese Standards. The ecological risk to human health can be a big concern. All heavy metal concentrations were lower as compared to safe limits given by WHO/FAO (WHO/FAO, 2007) except for As [38].

Maximum concentrations of heavy metals in some rice samples, such as Cu and Cd, exceeded permissible limits of Chinese Standards, suggesting they may have potential ecological risk to human health.

Transfer Factor

Among different metals in wheat grains, Zn showed highest Transfer Factor (TF) value, followed by Cd, Cu, Hg, and As (Table 3). The transfer factor of Cr is minimum followed by Pb. The result showed that Zn and Cd are more mobile than other metals. Lokeshwari and Chandrappa have reported that Cd and Zn are retained less strongly by soils and hence they are more mobile than other metals [39].

Among different metals in rice grains, Cd showed highest TF values followed by Zn, Cu, Hg, and Cr. TF values of Pb and As were lower. The result showed that Cd and Zn are more mobile than other metals, which is the same as wheat grains. TF values of metals in rice were higher than that in wheat grains except for Zn.

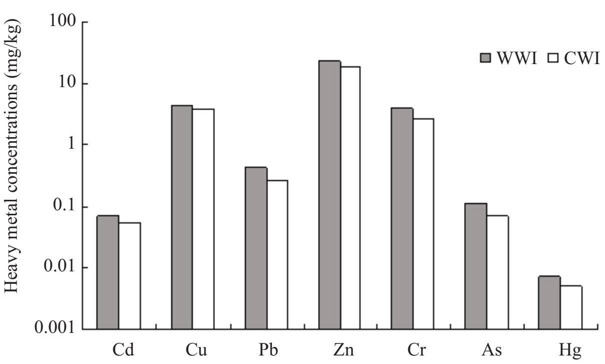

Fig. 6. Mean concentrations of heavy metals in rice grains.

Daily Intake Rate

The degree of toxicity of heavy metals to humans depends on their daily intake. Heavy metals intake through wheat and rice grown in the Beijing sewage discharge channel sewage irrigation area showed slight variations (Table 3). The standard of FAO/WHO (1999) has established a reference value for the tolerable daily intake [38].

Our estimated daily intake rate for all the metals were below the tolerable daily intake rates. Radwan and Salama and Khan et al. have also observed no risk due to consumption of common foodstuffs grown in wastewater irrigated sites [40].

Target Hazard Quotient (THQ)

The results of estimated target hazard quotient calculations showed that all heavy metals in plants had no potential health risk to the local population (Table 3). Although individual metal THQ (<1.0) values indicated the relative absence of health risks associated with the intake of a single heavy metal through consumption of either wheat or rice only, the summation of individual THQs of both wheat and rice was almost 1.0. If individual THQs resulting from crop consumption are considered, the health risks would be greater when the THQ values are greater than 1.0. THQs of As was above 1.0, and consumption of such wheat and rice may pose a risk to the local population (Table 3). Wang et al. drew a consistent conclusion when they discussed the health risks of heavy metals to the general public in Tianjin regarding consumption of vegetables and fish [29]. Other metals are less responsible for causing risks to the local population since the THQs were below 1.0 plants.

Conclusions

Heavy metals were enriched in soils at the WWI sites compared with the CWI sites. Cd and Hg in soils were clearly enriched, whereas Cu, Pb, Cr, As, and Zn were slightly enriched.

The concentrations of Cd and Hg clearly exceeded the permissible limits of the second grade of Tianjin Standards. The concentration of Zn was slightly higher, suggesting that Cd and Hg were important pollutants in the soil and posed potential ecological risks. Zn had some ecological risks as well.

The concentrations of Pb in wheat and rice had crossed the safe limits for human consumption, indicating that Pb posed ecological risks to human health. The mean concentrations of As and Hg in rice were slightly higher than the safe limits of commission regulation (EU, 2008) and Chinese Standards, and they had some ecological risks to human health. The concentrations of heavy metals such as Cd, Zn, and As in part of the wheat samples, and Cu and Cd in part of the rice samples exceeded safe limits of Chinese National Standards, suggesting that they pose a potential ecological risk to human health.

Heavy metal concentrations varied in wheat and rice samples, which reflect the differences in their uptake capabilities and their further translocation to edible portion of the plants. Zn and Cd were more mobile than other metals.

The target hazard quotient of individual metal was below 1.0, indicating the relative lower health risks associated with intake of a single heavy metal through consumption of either wheat or rice only. The summation of THQs of both wheat and rice was above 1.0, indicating that the consumption of wheat and rice may pose a risk to the local population.

Thus regular monitoring of heavy metal contamination in crops grown in wastewater-irrigated sites is necessary. Consumption of contaminated crops should be avoided in order to reduce the health risks caused by intake of the contaminated crops. The wastewater treatment technology should be perfected in terms of improving the removal of heavy metals.

Acknowledgements

This research was funded by the National Nature Science Foundation of China (No. 40973078), the Geping Green Action Environmental Research and Education "123 Project" of Liaoning Province, China (CEPF2011-123-1-1), and the State Scholarship Fund organized by the China Scholarship Council (CSC2013). We thank reviewers for their valuable and constructive comments.

References

1. SINGH R. P., AGRAWAL M. Variations in heavy metal accumulation, growth and yield of rice plants grown at different sewage sludge amendment rates. Ecotox. Environ. Safe., **73**, 632, **2010**.
2. SINGH R. P., AGRAWAL M. Potential benefits and risks of land application of sewage sludge. Waste Manage., **28**, 347, **2008**.
3. MARKOVIĆ M., CUPAĆ S., ĐUROVIĆ R., MILINOVIĆ, J., KLJAJIĆ P. Assessment of heavy metal and pesticide levels in soil and plant products from agricultural area of belgrade, Serbia. Arch. Environ. Con. Tox., **58**, 341, **2010**.
4. LIU W. H., ZHAO J. Z., OUYANG Z. Y., SODERLUND L., LIU G. H. Impacts of sewage irrigation on heavy metals distribution and contamination in Beijing, China. Environ. Int., **31**, 805, **2005**.
5. MAPANDA F., MANGWAYANA E. N., NYAMANGARA J., GILLER K. E. The effect of long-term irrigation using wastewater on heavy metal contents of soils under vegetables in Harare, Zimbabwe. Agric. Ecosyst. Environ., **107**, 151, **2005**.
6. RATTAN R. K., DATTA S. P., CHONKAR P. K., SURIBABU K., SINGH A. K. Long term impact of irrigation with sewage effluents on heavy metal content in soils, crops and groundwater – a case study. Agric. Ecosyst. Environ., **109**, 310, **2005**.
7. JAMALI M. K., KAZI T. G., ARAIN M. B., AFRIDI H. I., JALBANI N., MEMON A. R., ANSARI R., SHAH A. The feasibility of using an industrial sewage sludge produce in Pakistan as agricultural fertilizer used for cultivation of Sorghum bicolor L. Arch. Agron. Soil Sci., **53**, 659, **2007**.
8. JAMALI M. K., KAZI T. G., ARAIN M. B., AFRIDI H. I., JALBANI N., MEMON A. R., SHAH A. Heavy metals from soil and domestic sewage sludge and their transfer to Sorghum plants. Environ. Chem. Letter, **5**, 209, **2008**.
9. MORERA M. T., ECHEVERRÍA J., GARRIDO J. Bioavailability of heavy metals in soils amended with sewage sludge. Can. J. Soil Sci., **82**, 433, **2002**.
10. ERIYAMREMU G. E., ASAGBA S.O., AKPOBORIE I. A., OJEABURU S. I. Evaluation of lead and cadmium levels in some commonly consumed vegetables in the Niger-Delta oil area of Nigeria. B. Environ. Contam. Tox., **75**, 278, **2005**.
11. NAWAG U. H., QAISAR M., AMIR W., MUHAMMAD I., FARIDULLAH, ARSHAD P. Assessment of heavy metals in wheat plants irrigated with contaminated wastewater. Pol. J. Environ. Stud., **22**, 115, **2013**.
12. SHARMA R. K., AGRAWAL M., MARSHALL F. M. Heavy metals contamination of soil and vegetables in suburban areas of Varanasi, India. Ecotox. Environ. Safe., **66**, 258, **2007**.
13. SHARMA R. K., AGRAWAL M., MARSHALL F. M. Heavy metals contamination in vegetables grown in wastewater irrigation areas of Varanasi, India. B. Environ. Contam. Tox., **77**, 311, **2006**.
14. CHANG A. C., PAGE A. L., HYUN H. Cadmium uptake Swiss for Chard Grown on composted sewage sludge treated plots: Plateau or time bomb. J. Environ. Qual., **26**, 11, **1997**.
15. DURUIBE J. O., OGWUEGBU M. D. C., EGWURUQWU J. N. Heavy metal pollution and human biotoxic effects. Int. J. Phys. Sci., **2**, 112, **2007**.
16. YOUNG R. A. Toxicity Profiles: Toxicity Summary for Cadmium, Risk Assessment Information System. University of Tennessee (rais. ornl. Gov/ tox/ profiles/ cadmium.html), **2005**.
17. OGWUEGBU M. O. C., MUHANGA W. Investigation of lead concentration in the blood of people in the copper belt province of Zambia. J. Environ., **1**, 66, **2005**.
18. NOLAN K. R. Copper toxicity syndrome. J. Orthomol Physiol., **12**, 270, **2003**.
19. WANG B., MENG H. T., ZHANG Z., GONG H.Y., JIANG W. Heavy metals content and potential Ecological risk assessment in field soil in Tianjin suburbs. Environ. Res. Test, **4**, 11, **2010**.

20. WU G. H., SU R. X., LI W. Q., ZHENG H. Q. Source and enrichment of heavy metals in sewage-irrigation area soil of Dagu sewage discharge channel. Environ. Sci., **29**, 1693, **2008**.
21. SHI R. G., ZHOU Q. X., LIU F. Z., ZHAO Y. J., ZHENG X. Q., ZHANG H. Cadmium accumulation and pollution risks to human health based on Monto-Carlo model of soil and vegetable-using vegetable field in Tianjin Suburbs as example. China Environmental Science, **28**, 634, **2008**.
22. ZHAI H. Q. Investigation and assessment on heavy metals in soil and vegetables in sewage irrigation areas. Tianjin Normal University Master's Thesis, **2010**.
23. DONG W. H. Environmental geochemistry of heavy metals in irrigation water and soil of key areas in Tianjin, China. China University of Geosciences Master's Thesis, **2007**.
24. LI Z. M. Assessment to heavy metals pollution in Tianjin sewage irrigation area soil and wheat and relative analysis. Tianjin Normal University Master's Thesis, **2006**.
25. MA C. X., WANG Z. R., GAO Q., JIN X. L., HE S. Heavy metal analysis of winter wheat at each stage in typical sewage irrigation areas of Tianjin. Environ. Chem., **29**, 44, **2010**.
26. ALLEN S. E., GRIMSHAW H. M., ROWLAND A. P. Chemical analysis. In: Moore P. D., & Chapman S. B., (Eds.) Methods in Plant Ecology. Blackwell Scientific Publication, Oxford, London, 285, **1986**.
27. CUI Y. J., ZHU Y. G., ZHAI R. H., CHEN D.Y., HUANG Y. Z., QUI Y., LIANG J. Z. Transfer of metals from near a smelter in Nanjing, China. Environ. Int., **30**, 785, **2004**.
28. GE K. Y. The status of and Nutrient and meal of Chinese in 1990s. Beijing People’s Hygiene Press, **1992**.
29. WANG X., SATO T., XING B., TAO S. Health risks of heavy metals to the general public in Tianjin, China via consumption of vegetables and fish. Sci. Total Environ., **350**, 28, **2005**.
30. WANG X., HUAN G. W., TIAN H. G., DONG S. R., YIN H. G., CAO X. H. Investigation on dietary of urban residents in Tianjin from 2000-2004, China. J. Publ. Health, **23**, 1245, **2007**.
31. CHIEN L. C., HUNG T. C., CHAOANG K. Y., YEH C. Y., MENG P. J., SHIEH M. J. Daily intake of TBT, Cu, Zn, Cd and As for fishermen in Taiwan. Sci. Total Environ., **285**, 177, **2002**.
32. SINGH A., SHARMA R. K., ETAL M. A., MARSHALL F. M. Risk assessment of heavy metal toxicity through contaminated vegetables from wastewater irrigation area of Varanasi, India. Trop. Eco., **51**, 375, **2010**.
33. BENNETT D. H., KASTENBERG W. E., MCKONE T. E. A multimedia multiple pathway risk assessment of atrazine: the impact of age differentiated exposure including joint uncertainty and variability. Reliab. Eng. Sys. Saf., **63**, 185, **1999**.
34. USEPA. Risk-based concentration table. Philadelphia PA: United States of Environmental Protection Agency, Washington D.C., **2000**.
35. WANG X. X. Spatial distribution of heavy metals contamination fractions and influence factors in Beijing sewage discharge channel sewage irrigation area of Tianjin. Tianjin Normal University Master's Thesis, **2012**.
36. HUANG J. Y., XU H., LIU S. Y. Comprehensive evolution on heavy metal pollution in soil of Dabizhuang in Dongli District of Tianjin. J. AnH. Agri. Sci., **39**, (22), 13442, **2011**.
37. YANG J., CHEN T. B., ZHENG Y. M., LUO J. F., LIU H. L., WU W. Y., CHEN Y. C. Dynamic of heavy metals in wheat grains collected from Liangfeng Irrigation Area, Beijing and a discussion of availability and human health risks. Act. Sci. Circum., **25**, 1661, **2005**.
38. WHO/FAO. Joint FAO/WHO Food Standard Programme Codex Alimentarius Commission 13th Session. Report of the Thirty Eight Session of the Codex Committee on Food Hygiene. Houston, United States of America, ALINORM 07/30/13, **2007**.
39. LOKESHWARI H., CHANDRAPPA G. T. Impact of heavy metal contamination of Bellandur Lake on soil and cultivated vegetation. Current Science, **91**, 620, **2006**.
40. RADWAN M. A., SALAMA A. K. Market based survey for some heavy metals in Egyptian fruits and vegetables. Food Chem. Toxicol., **44**, 1273, **2006**.