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RESEARCH ARTICLE

Accumulation of heavy metal in scalp hair of people exposed in Beijing sewage discharge channel sewage irrigation area in Tianjin, China

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Abstract Heavy metal concentrations in soil, wheat, and scalp hair exposed to Beijing sewage discharge channel sewage irrigation area (BSIA) in Tianjin were studied to evaluate the influence of sewage irrigation. Results showed that the continuous application of wastewater has led to an accumulation of heavy metals in the soil, with 55.2 and 8.62% of soil samples accumulating Cd and Zn, respectively, at concentrations exceeding the permissible limits in China. Concentrations of heavy metals in wheat grain from BSIA were higher than these from the clean water irrigation area by 63.2% for Cd, 3.8% for Cu, 100% for Pb, 6.6% for Zn, and 326.7% for Cr. The heavy metal bioaccumulation factor (BAF) of wheat/soil in BSIA showed the following order: Zn > Cd > Cu > Pb > Cr. Interestingly, these accumulation of heavy metals in soil after sewage irrigation could increase the migration ability of heavy metals (particularly Zn and Cd) from soil to wheat. Mean concentrations of heavy metals in the hair of residents followed the decreasing trend of Zn > Cu > Pb > Cr > Cd, which were higher than the control area by 110.0% for Cd, 20.0% for Cu, 55.9% for Zn, 36.6% for Pb, and 64.6% for Cr. Concentrations of heavy metals in male human hair in BSIA were higher than those of females. And the concentrations of heavy metals except for Pb in human hair increased with their increasing ages. The heavy metal BAF values of wheat/soil in BSIA showed the trend of Zn (98.0057) > Pb (7.0162) > Cr (5.5788) > Cu (5.4853) > Cd (3.5584); heavy metals had obvious biological amplification from wheat to human hair. These results indicated that local population health was potentially exposed to the heavy metal risk via wheat consumption.

Keywords Heavy metals accumulation · Soil-wheat-human hair system · Wastewater irrigation · Tianjin

Introduction

Heavy metals are the most important pollutants in the environment. Wastewater irrigation, solid waste disposal, sludge applications, vehicular exhaust, and industrial activities are the major sources of heavy metal contamination in soil (Rattan et al. 2005; Singh and Agrawal 2008, 2010; Qureshi et al. 2016). Furthermore, the continuous enrichment of heavy metals by food crops grown on such contaminated soils is frequently observed (Zeng et al. 2015).

Wastewater irrigation, which is a widespread practice in the world, can lead to the accumulation of heavy metals in agricultural soils and plants over time (Mapanda et al. 2005; Jamali et al. 2008; Marković et al. 2010; Khan et al. 2013; Yu et al. 2016). Although some of these heavy metals such as Zn and Cu act as micro-nutrients at lower concentrations, they can become toxic at higher concentrations. Several health risks due to heavy metal contamination in soil have been widely reported (Eriyamremu et al. 2005; Gebrekidan et al. 2013; Lu et al. 2015). Besides, crops and vegetables grown in soils contaminated with heavy metals have shown greater accumulation than those grown in uncontaminated soil, raising

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serious public health concern as food consumption is the primary path of heavy metal toxicity and uptake in human beings (Sharma et al. 2007). In previous studies, we demonstrated that continuous application of wastewater has led to significant accumulation of heavy metals in the soil and wheat crop in Tianjin, a very populated city in China that such contamination may pose serious health risks to adults and children (Wang et al. 2015; Zeng et al. 2015).

Scalp hair is one of the organs in the human body used to excrete trace elements including heavy metals. Hence, heavy metal concentration in scalp hair can reflect the physiological status and influence from environmental factors on the human body (Zhou et al. 2016). The concentration of heavy metals in hair is much more pronounced compared to that in blood and urine. Additionally, using scalp hair to study heavy metal content in human body has the added advantages of convenient material sampling, no damage to human body, and easy storage or transfer for pathological analysis, clinical diagnosis, and environmental pollution detection (Xue et al. 2015). Many scholars have conducted extensive research on heavy metal concentrations in hair (Ryabukhin 1976; Andrea et al. 2003; Mosaferi et al. 2005; Anwar 2005; Mohammed et al. 2007; Gonzalez-Reimers et al. 2014; Lamin et al. 2015; Massaquoi et al. 2015). These studies indicated that concentrations of heavy metals in hair are related to age, sex, environmental factors, ethnicity, local resident health differences, eating habit, and analytical method. Despite these numerous reports, the concentration of heavy metals derived from hair has varied greatly, sometimes across different regions (Zhou et al. 2016).

Thus, this study focused on investigating the accumulation of heavy metals in soil, wheat, and human scalp hair from the BSIA, a typical long-term wastewater irrigation area in Northern China (Wang et al. 2015; Yu et al. 2016), and evaluating the relationship between concentrations of selected heavy metals in soil, wheat, and human scalp hair. Meanwhile, the Jixian District (JX) in North Tianjin was selected as the control area (CA) to assess the influence of heavy metals in hairs of people from the BSIA exposed to wastewater irrigation.

Materials and methods

Study sites

The study site is located in the Wuqing District of Tianjin, the main part of the BSIA. The BSIA includes Wuqing District (WQ), part of Baodi District (BD), Beichen District (BC) and Ninghe District (NH), with an irrigation area of 8.35 × 10^4 hm^2 (Fig. 1). The source of the wastewater for irrigation is the industrial wastewater and domestic sewage coming from Beijing.

This area has a continental monsoon climate (cold and dry winter with a hot and rainy summer), which is characterized by wide seasonal variation in annual rainfall (600 mm). The soils in this area are mainly cinnamon soil (Argosols) and fluvoaquic soil (Cambosols). The food crops are mainly wheat, corn, and vegetables (Liu et al. 2005). The wastewater irrigation commenced in the early 1960s, after which the reclaimed water has been used to irrigate agricultural soils since the 2000s. In recent years, the area of the sewage irrigation site decreased due to the reduced volume of wastewater from Beijing. This was a result of more stringent national environmental policies in China coupled with improved wastewater treatment technology.

Sampling and analysis methods

Nineteen representative wheat samples grown in BSIA and nine wheat samples grown in clean water irrigation areas around the BSIA were collected randomly at harvest time using the cinquefoil five-point snakelike sampling method. At the same time, related 68 and 28 soil samples from BSIA and clean water irrigation areas were taken with a 5-cm-diameter stainless steel auger at the 0–15 cm layer, homogenized in plastic bags, and stored at 4 °C. In the laboratory, wheat samples were thoroughly washed with running tap water to remove airborne dust and soil particles and then rinsed with deionized water. Then, samples were oven dried at 80 °C to a constant weight. The dried samples were grounded, passed through a 2-mm sieve, and stored in plastic bags at room temperature before analysis. Soil was air-dried and sieved through a 2-mm mesh, and then sealed in Kraft paper envelopes until analysis.

One hundred and four representative scalp hair samples were collected from barber shops from eight towns in BSIA and 50 representative hair samples were collected from barber shops from five towns in the control area. Scalp hair samples were thoroughly cleaned with detergent (washed 4 times with 15 ml acetone, followed by rinsing 4 times with ultra pure water, then washed once with 15 ml acetone) to remove the adhesion on the surface of the hair, the exogenous pollutants, and then rinsed with deionized water. Then, samples were oven dried at 80 °C to a constant weight. The dried samples were cut into pieces in 1–3 mm long and stored in plastic bags at room temperature before analysis.

To extract heavy metals, 1 g of dried wheat, soil, or hair was digested with 15 ml of mixture solution (HNO_3, H_2SO_4 and HClO4, 5:1:1) at 80 °C until a transparent solution was obtained (Allen et al. 1986). Water samples (50 ml) were digested with 10 ml of concentrated HNO_3 at 80 °C until the solution became transparent. These transparent solutions were then filtered through filter paper (0.22 lm, Whatman Filters, Maidstone, UK) and diluted to 50 ml with distilled water. The concentrations of heavy metals in the filtrate were determined by inductively coupled plasma atomic emission spectrometry

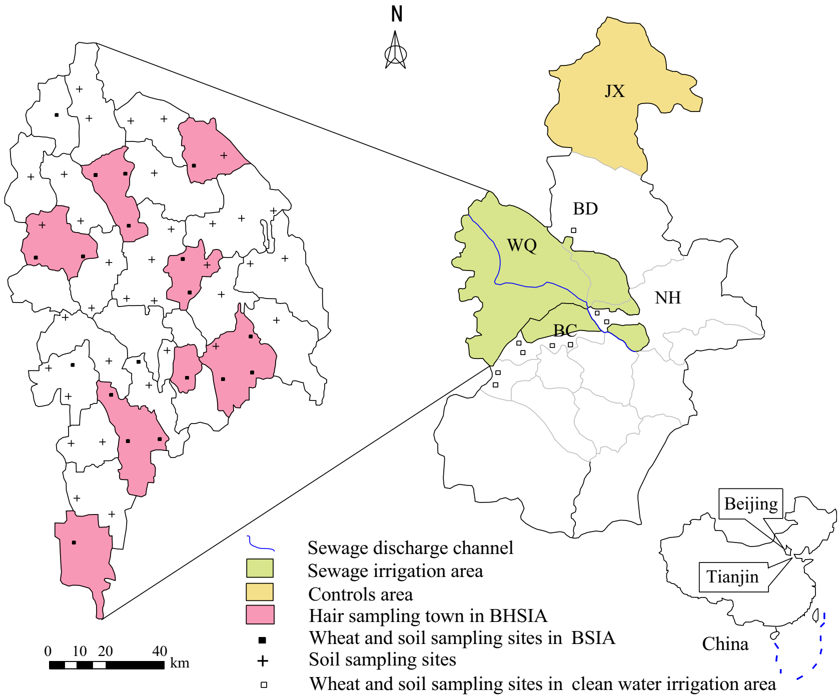

Fig. 1 Sampling locations for hair, wheat grain, and soil

(ICP-AES, Leeman Labs, USA) fitted with a specific lamp for specific metals using appropriate drift blanks.

Quality control and data analysis

The accuracy of analyses was checked on samples of wheat and soil with certified concentrations (GBW10011, GBW07401, respectively, China National Center for Standard Materials) of selected heavy metals. In each analytical batch, 10% of the samples were analyzed twice to ensure the precision and accuracy of the analysis. Internal reference standard materials and reagent blanks were also used in the analysis to ensure high precision.

Bioaccumulation factor (BAF) is the ratio of heavy metal concentration to wheat grain and soil. BCF is used to evaluate the extent of risk and associated hazard due to wastewater irrigation and consequent heavy metal accumulation in wheat, and is calculated following Zeng et al. (2015)

$$\mathrm{BAF} = \mathrm{Cp}/\mathrm{Cs}$$

where Cs is the concentration of heavy metals in soil, and Cp is the concentration of heavy metals in wheat grain.

Results and discussion

Levels of heavy metals in soil and wheat grain at BSIA

Continuous application of treated and untreated sewage water led to higher concentrations of heavy metals in the soil and wheat grain at BSIA compared to those at the control area. The range and mean values of five heavy metals in soils and wheat grain are shown in Table 1.

More than half of soil samples (55.2%) had mean concentrations of Cd above the maximum permitted levels in GB15618-1995, a standard set by the Chinese Environmental Protecting Administration (CEPA 1995) for soils in China. Soil samples (8.62%) accumulated Zn in concentrations exceeding the permissible limits in China. The minimum concentrations of all five heavy metals in soils were below the maximum permitted levels, partly due to lower background values in the soil (Liu et al. 2005). Zinc had the highest mean concentration (118.59 ± 55.79 mg/kg), followed by Cu, Pb, As, and Cd. The mean concentration of Cd was the lowest among the five heavy metals, but it still exceeded the permissible limits of China's standard. The average concentration of heavy metals in this study was comparable to previously reported values in this area (Ma et al. 2010). The average concentrations of heavy metals in the soil at the BSIA were lower than those at Zhangshi Sewage Irrigation Area of Shenyang, which has a similar 30-year wastewater irrigation history (Li et al. 2009a, b), and the Mexico Irrigation Area, which has an 80-year wastewater irrigation history (Siebe 1995). Different water sources, irrigation times, and background values at the three locations could be the reasons for the differences in soil heavy metal accumulation. ZSIA received mostly industrial wastewater and had a higher background value, while BSIA mainly received domestic wastewater with a lower background value. The Mexico Irrigation Area has had a longer irrigation time (80 years) than the Tianjin sewage irrigation area.

The concentrations of heavy metals in wheat grain were in the order of Zn > Cu > Cr > Pb > Cd (as shown in Table 1),

Table 1 Concentrations of heavy metals in soils and wheat grain from BSIA

Heavy metal		Cd	Cu	Pb	Zn	Cr
Soil	Range	0.05–0.77	10.9–43.6	3.8–21.42	62.2–333.6	40.2–108
	Mean	0.37	24.75	13.41	118.59	64.19
	Stdev.	0.18	7.41	3.51	55.79	12.25
	GB 15618-1995	0.3	100	250	200	150
Wheat grain	Range	0.025–0.08	2.15–4.16	0.06–0.24	16.21–53.0	0.28–0.62
	Mean	0.062	2.99	0.14	27.67	0.49
	Stdev.	0.020	0.51	0.05	7.33	0.10
	Chinese Standard (GB 2762-2005)	0.1	10	0.2	50	20
	E.U. Standard (EC: no. 629/2008)	0.2	–	0.2	–	–
	WHO/FAO	0.2	40	5	60	–

similar to the result reported by Yang et al. (2005) for wheat grain grown in the Liangfeng sewage irrigation area in Beijing. Compared to permissible limits of the Chinese Standard (GB 2762-2005), the mean concentrations of heavy metals in wheat grains were actually lower. Some wheat samples contained Pb (two samples) and Zn (one sample) exceeding the permissible limits in China, suggesting that they may have potential health risk to human. The mean concentrations of heavy metals were also lower than the safe limits provided by WHO/FAO (WHO/FAO 2007) and the EU commission regulation (EU 2008), with the exception of As. Compared to the concentrations of heavy metals in wheat grain grown in clean water irrigation area, the concentrations of heavy metals in wheat grain from BSIA were higher by 63.2% for Cd, 3.8% for Cu, 100% for Pb, 6.6% for Zn, and 326.7% for Cr, suggesting that heavy metals had enriched in wheat grains due to sewage irrigation.

Levels of heavy metals in scalp hair in BSIA residents

The range and mean values of five heavy metals in the hair of people in BSIA are shown in Table 2, along with the concentrations of heavy metals in hair from Jixian as the control area. The concentrations of heavy metals in scalp hair in BSIA were in the order of Zn > Cu > Pb > Cr > Cd, same as the results obtained in the control area.

The concentration range of Cd in BSIA residents' hair was lower than the world average value (0.4–1.0 mg/kg) (Iyenger 1987). The mean value of Cd in BSIA hair samples was 0.21 mg/kg, which was lower than both the that of Chinese residents (0.25 mg/kg) and the upper limit of the normal Cd value (0.8 mg/kg or 1.0 mg/kg) determined in many clinical laboratories around the world (Miekeley et al. 1998). The concentration ranges of Cu and Zn in BSIA residents' hair were consistent with the results from the International Atomic Energy Agency (IAEA 1978) (Cu 9.6–20.6 mg/kg, Zn 138–308 mg/kg), while similar study results were obtained both from Ward et al. (1987) for Oxford city (Cu 4.6–19.4 mg/kg, Zn 141.9–259.6 mg/kg) and Tavakkoli et al. (2000) for the Esfahan area of Iran (Cu 4.8–66.7 mg/kg, Zn 36–329 mg/kg). The mean concentrations of Cu and Zn in BSIA were 16.82 and 197 mg/kg, which were higher than the typical values 12.4 and 168 mg/kg in Tianjin (Sun et al. 1987), as well as 11.3 and 165 mg/kg in China. Our results also indicated that the concentration range of Pb in hair is very large. The concentration range of Pb in BSIA residents' hair was 1.13–86.8 mg/kg, which was higher than that reported by Wang (2001), and Petering et al. (1973). However, the mean value was higher than the normal values of 7.1 mg/kg in China, 0.93 mg/kg in Tianjin, 6 mg/kg in France, and 12 mg/kg in England (Qin 2004; Sun et al. 1987). The mean concentration of Cr was 2.65 mg/kg, which was higher than the normal hair Cr value of 0.98 mg/kg in Tianjin, 0.262 mg/kg in Beijing, 1.21 mg/kg in China, 0.82 mg/kg in Japan, and 1.42 mg/kg in Pakistan (Qureshi et al. 1982; Sun et al. 1987; Chen et al. 1994).

The concentrations of heavy metal in BSIA residents' hair were higher than those of the control area (Fig. 2). Specifically, the concentrations of heavy metals were higher by 110% for Cd, 20.0% for Cu, 55.9% for Zn, 36.6% for Pb, and 64.6% for Cr. The results showed that heavy metals had significant enrichment in the hair of residents living along the sewage irrigation area. Such enrichment is likely due to long-term exposure to heavy metals by food ingestion, as wheat is the staple food for people residing in BSIA. Concentrations of heavy metal in wheat grown in the sewage irrigation areas were higher than that grown in clean water irrigation areas, meaning that heavy metals could be entering the human body by food.

Levels of heavy metals in human hair of different genders and age stages in BSIA

Concentrations of heavy metals in human hair of genders in BSIA are shown in Table 3. Concentrations of heavy metals in

Table 2 Concentrations of heavy metals in scalp hair (mg/kg)

Heavy metal		Cd	Cu	Zn	Pb	Cr
BSIA ($n = 199$)	Range	0.01–0.58	7.91–26.3	79.83–315.6	1.13–86.8	1.3–9.08
	Mean	0.21	16.82	197.29	13.69	2.65
	Stdev.	0.17	3.76	51.15	8.75	1.54
CA ($n = 93$)	Range	0.01–0.43	6.79–19.5	55.71–195.2	1.21–38.6	1.10–6.55
	Mean	0.10	14.01	126.56	10.02	1.61
	Stdev.	0.11	2.93	46.72	6.58	1.32
Chinese resident normal hair content[a]	Range	<0.5	8–16	90–170	<10	0.1–2.0
	Mean	0.25 ± 0.14	11.0 ± 3.5	130 ± 30	7.1 ± 3.2	1.21 ± 0.63

[a] Standards from Chinese trace elements scientific society (H/ZWY03-2005, H/ZWY01-2007)

male hair in BSIA were higher than those of female; the same results were also obtained in the control area. The difference of Pb content in scalp hair between males and females had statistical significance ($P < 0.01$), whereas other heavy metals had no significant difference ($P > 0.05$). The same result was obtained for the concentrations of heavy metals in human hair of different genders in the control area. Previous reports in the literature on heavy metal content in scalp hair of different genders also showed some differences. Most study results showed that concentrations of heavy metals in male hair were higher than that of females, and the differences were possibly due to the higher frequency and intensity of outdoor work among males than females (Trojanowski et al. 2010; Abdulrahman et al. 2012). Some study results suggested that there are no consistent correlations for concentrations of heavy metals in hair of different sexes, while other studies suggest that different heavy metal contents in male hair may be higher but other kinds of heavy metal content in female may be higher (Georgescu et al. 1997; Khalique et al. 2005; Perumal and Thangamani 2011). The results of this study are similar to most other study results.

Concentrations of Cd, Cu, Zn, and Cr in human hair increased with age, with the exception of Pb (Fig. 3). The difference of Cd content of age groups 40–49 and 50–59 years had statistical significance ($P < 0.01$), while other age groups had no significant difference ($P > 0.05$). There was no statistical difference for Cu content between different age groups, similar to the results of a previous study (Tavakkoli et al. 2000). Cd content gradually increased from 0.14 mg/kg in teenagers (<19 years of age) to 0.33 mg/kg in older men (>59 years of age). Cu content gradually increased from 10.03 mg/kg in teenagers (<19 years of age) to 20.25 mg/kg of older men (>59 years of age). Pb content declined as age increased, which was similar to the results from other studies around the world (Petering et al. 1973; Tavakkoli et al. 2000; Andrea et al. 2003; Mosaferi et al. 2005; Mohammed et al. 2007; Gonzalez-Reimers et al. 2014; Lamin et al. 2015). Zn content increased by age, which is in agreement with previous reports from China (Dong and Cai 1999; Zhang and Meng 2001; Lei et al. 2007), but different or even contradictory to the findings outside China. Bales found that Zn content decreased as age increased (Bales et al. 1990). Khalique found that female Zn content increased with age, while male Zn content decreased with age, and the total Zn content in human hair was not statistically different between age groups (Khalique et al. 2005).

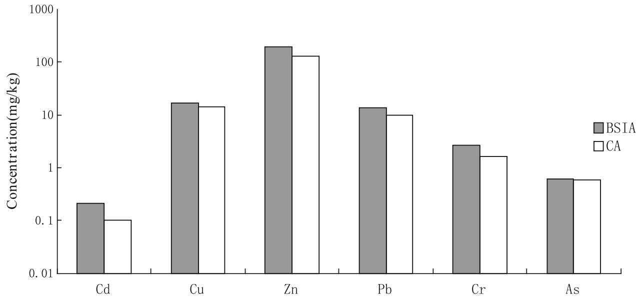

Fig. 2 Mean concentrations of heavy metals in human hair

BAF

Bioaccumulation factor (BAF) is a biological enrichment factor defined as the ratio of heavy metal content between organisms and their environment or food medium (Zeng et al. 2015). BAF was calculated to understand the extent of the risk and associated hazards due to wastewater irrigation and consequent heavy metal accumulation in crops portion. Many scholars of the world use BAF as a evaluation metric to study the transport of heavy metal from soil to plants (crop) (Ivanciuc et al. 2006; Arnot and Gobas 2006; Li et al. 2009a, b; Zhang et al. 2010; Rezvani et al. 2011).

The heavy metal BAF values of wheat/soil in BSIA were in the order of Zn (0.2241 ± 0.0886) > Cd (0.1800 ± 0.1030) > Cu (0.1439 ± 0.089) > Pb (0.01136 ± 0.0040) > Cr (0.0087 ± 0.0020). In comparison, the BAF values in clean water irrigation areas were much lower (Fig. 4), where the wheat/soil BAF values were ordered as Zn (0.0280) > Cd (0.0255) > Cu (0.0180) > Pb (0.0014) > Cr(0.0011). These results suggest that accumulation of heavy metals in contaminated soil after sewage irrigation could increase the capacity of heavy metal uptake by

Table 3 Concentrations of heavy metals in human hair of difference genders

Heavy metal			Cd	Cu	Zn	Pb	Cr
BSIA	Male	Range	0.01–0.52	8.64–25.7	88.6–315.6	1.13–86.8	1.3–9.08
	n = 104	Mean	0.22	17.53	198.38	15.03	2.84
		Stdev.	0.18	3.35	58.92	9.34	1.72
	Female	Range	0.01–0.47	7.91–26.3	79.8–295.4	1.21–77.1	1.37–7.27
	n = 95	Mean	0.2	15.98	196	12.1	2.42
		Stdev.	0.16	4.19	43.23	8.21	1.36
CA	Male	Range	0.01–0.43	7.57–18.53	55.71–190.07	1.83–38.63	1.10–6.55
	n = 50	Mean	0.10	15.53	135.34	11.47	1.65
		Stdev.	0.14	3.31	50.18	7.75	1.16
	Female	Range	0.01–0.41	6.79–19.52	60.24–195.22	1.21–31.22	1.15–5.52
	n = 43	Mean	0.10	12.24	116.35	8.33	1.56
		Stdev.	0.09	2.78	44.21	5.38	1.51

crops. Zn and Cd had higher wheat/soil BAF values, indicating their higher migration ability. Because Zn and Cd are the main pollutants in the soil at BSIA, it is necessary to further investigate the potential health risks caused by these heavy metals.

There are many challenges residing in the calculation of human hair heavy metal BAF on the environmental medium, mainly because of the diverse exposure pathways and the lack of effective methods to identify the sources of heavy metals in human hair. Considering that residents in BSIA use locally grown wheat as their main food source, and ingestion is the main way to bioaccumulate heavy metals in human hair, the mean concentrations of heavy metals in soil and wheat samples in eight hair sampling towns in BSIA were used to calculate the BAF of scalp hair. Although the BAF values of hair/wheat may not be completely consistent with the actual situation, they still can generally reflect migration of heavy metals from environment medium to human hair (Xue et al. 2015). The heavy metal BAF values of hair/wheat in BSIA showed the trend of Zn (98.0057) > Pb (7.0162) > Cr (5.5788) > Cu (5.4853) > Cd (3.5584). The BAF values of hair/wheat were higher than 1, suggesting that evident biological amplification occurred and that heavy metals in wheat grown in contaminated soil could be accumulated in the human body through ingestion.

Conclusions

Continuous application of wastewater has led to accumulation of heavy metals in the soil in Tianjin-Beijing sewage discharge channel sewage irrigation area. The maximum concentrations of Cd and Zn in the soil were above the maximum permitted levels according to GB15618-1995. In particular, 55.2 and 8.62% of soil samples accumulated Cd and Zn in concentrations exceeding the permissible limits in China.

Concentrations of heavy metals in wheat grain in BSIA were higher than the clean water irrigation area by 63.2% for Cd, 3.8% for Cu, 100% for Pb, 6.6% for Zn, and 326.7% for Cr. BAF of the heavy metals from soil to wheat in BSIA showed the trend of Zn > Cd > Cu > Pb > Cr. Accumulation of heavy metals in soil after sewage irrigation could increase heavy metal, especially Zn and Cd migration ability.

Mean concentrations of Cd, Cu, Zn, Pb, and As in the hair of BSIA residents were 0.21, 16.82, 197.29, 13.69, and 0.62 mg/kg, respectively. These values were higher than the control area by 110% for Cd, 20.0% for Cu, 55.9% for Zn, 36.6% for Pb, and 64.6% for Cr, respectively. Concentrations of heavy metals in male hair in BSIA were higher than those of female, and the concentrations of heavy metals with the exception of Pb in

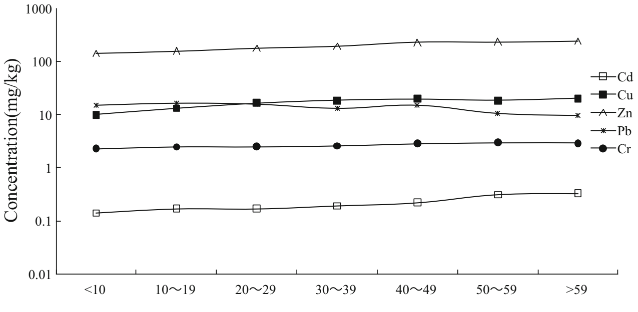

Fig. 3 Trend of concentrations of heavy metals in human hair with increasing age

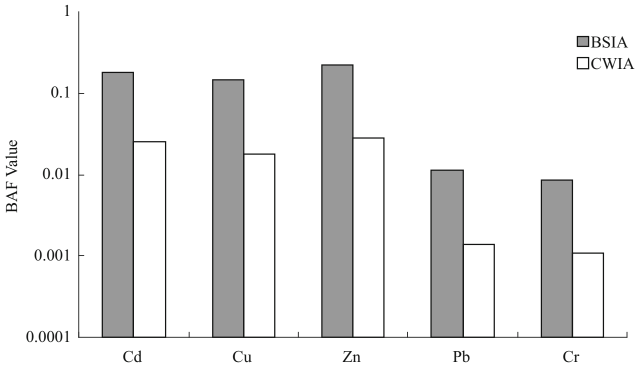

Fig. 4 Mean BAF values of wheat/soil for five heavy metals

human hair increased with ages. The BAF values of the heavy metals from wheat to human hair in BSIA showed the trend of Zn (98.0057) > Pb(7.0162) > Cr(5.5788) > Cu (5.4853) > Cd (3.5584), with heavy metals showing obvious biological amplification from wheat to human hair.

More attention should be paid to the continuing threat from heavy metals to the health of people through wheat consumption in Tianjin.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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