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Heavy metals and associated health risk of wheat grain in a traditional cultivation area of Baoji, Shaanxi, China

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Abstract As a staple food for people worldwide, wheat is one of the major exposure pathways for heavy metals (HMs). Therefore, the safety of the wheat grain directly affects food security and human health. Long-term agricultural activities are sources of heavy metal pollution in farmland ecosystems. This study assessed the pollution situation of HMs in wheat grain from the major wheat-cultivation areas of Baoji, a typical agricultural area in Shaanxi, to assess the dietary health risks caused by consuming wheat grains and to prevent food pollution. The results showed that the mean grain concentration of Cr, Ni, Cu, Zn, Cd and Pb were 0.11, 0.09, 4.41, 26.79, 0.01 and 0.03 mg/kg, respectively. These values were all remarkably lower than the tolerance limits of the Chinese food hygiene standard (GB2762-2017). According to the metal pollution index (MPI) analysis, wheat grain consumption poses no direct threat to human health. The health risk assessment showed that there was a noncarcinogenic risk to adults and children for wheat consumed in the study area. In the study area, no carcinogenic risk was manifested. Principal component analysis (PCA) indicated that the source of Ni was different from that of the other tested HMs and was mainly from industry, where as the others were mainly derived from agricultural activities. Therefore, more attention should be paid to Cu and Zn input through agricultural activities in fields to further prevent the accumulation of these HMs in wheat grains and their related human health risks.

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Introduction

Heavy metals (HMs) are mainly from industrial and agricultural activities, which are the common pollutants in soil (Zhu et al. 2017). According to a national report of soil pollution in China from 2014, the total national contaminated soil occupied 16.1% of the total farmland, on the basis of environmental quality standards for soils (GB 15618-1995). Inorganic pollutants were the main type of pollutants, accounting for 82.8%, with Cd, Ni, Cu, Pb, Cr and Zn as the major contributors. Their accumulation in the soil deteriorates the soil ecological system and affects the quality of crops (Bermudez et al. 2011; Liu et al. 2014; Xiao et al. 2015), while also threatening human health through the food chain (Luo et al. 2015; Reboredo et al. 2018). As a staple food for people worldwide, wheat is one of the major exposure pathways for HMs, which can easily accumulate in the human body, causing potential health risks (Liu et al. 2014; Liu et al. 2017a, b). Therefore, the safety of wheat grain directly affects food security and human health.

Wheat is the 3rd largest crop as well as the staple food of northern residents in China. The cultivated land area accounts for 20% of cultivated lands in the world. It has a pivotal position in food production, circulation and consumption (Zhao 2013). Wheat absorbs some toxic elements during its uptake of essential elements from the soil; these toxic elements include Pb, Ni, Cd and Cr (Ran et al. 2016). Wheat showed different capabilities for HM accumulation in the different regions because of the variety, soil type, topography, climate and other natural conditions and the pattern of economic development. To date, numerous studies have been performed to investigate the HM content in wheat grains in China. Xing et al. (2016) observed that Cd in all of wheat grain samples exceeded China's national standards around the lead smelting factories in Henan Province, and the Pb in 64% of the wheat samples exceeded the upper limits of the national standard. Ru et al. (2010) found that Cd and Zn in the wheat grains from the wastewater irrigation areas in Luancheng District of Heibei Province were 3.96 times and 1.30 times higher than that of freshwater irrigation areas, respectively, and HMs in some samples exceeded the limits of the national standard. Even in the typical livestock manure application areas of Yucheng in Shandong Province, Ye et al. (2013) found that Cr, Ni, Cd and As in the wheat grains exceeded the limits of the national standard. All of the above results were gathered from either wheat-growing areas with known industrial pollution sources or wastewater input fields, but the condition of typical farmland, especially with mostly agricultural activity input, was still obscure.

From an economic point of view, the contamination occurred with the economic developed rapidly in the last few decades. Pollutants, such as heavy metals, were released into the environment and accumulated in soil finally (Liang et al. 2009). A typical agricultural area has a long history of farming which influenced farmland ecosystems. Irrigation, fertilization and pesticide spraying were essential to the growth of crops (Wang et al. 2005; Rahman et al. 2014), and these activities were also potential HM sources of pollution for farmland. In addition, some factories were located closer to the fields, and they discharged pollutants in many ways, such as through fossil fuel combustion, mining exploration, exhaust emissions from transportation, and especially animal husbandry (Zeng et al. 2015; Wang et al. 2017). Furthermore, agricultural and industrial activities will release HMs to cropland soil and even threaten grain security. Heavy metals were absorbed by wheat, which is the result of economic development (Liang et al. 2009). Consequently, exploring the HM contamination conditions of grain in a wheat producing area is necessary.

However, different pollutant sources lead to different HM pollution types and contents. Shaanxi is a major grain-producing province possessing several kinds of HM pollution. Serious moderate-to-severe Pb, Zn and Hg pollution was found around and in some Pb and Zn smelting areas in the western part of Guanzhong (Xu et al. 2014). Zn and Cd levels in the farmland close to the smelting area and the China National Highway 312 exceeded the limit of National Environmental Quality Standard of Soil level II (GB15618-1995) (Li and Zhang 2017). Because of the industrial wastewater irrigation in the past, As, Ni and Pb in the soil near Xi'an also exceeded the limits of the mentioned standard levels (Chen et al. 2016). Thus, the pollution source and types and concentrations of HM in the soil in typical wheat-cultivation areas must be investigated.

In addition, the pollution orders of different heavy metals in soil were not the same as that in the wheat grain. The farmland in Tongguan County was polluted by Cd, Pb and Hg, with over-limit rates of 57.1%, 12.2% and 89.8%, which resulted in over-limit levels of 39.1%, 44.4% and 39.1% in the wheat grain samples, respectively (Wang et al. 2014). Moreover, the average concentration of Hg, As, Cd, Cr, Pb, Cu, Zn and Ni in farmland soil of the Jinghui Canal irrigation area exceeded the base background values of the soil, but the average of the HMs in the corresponding wheat grains was significantly lower than the Chinese national upper limits (Lei et al. 2014). Therefore, proper assessment and analysis methods should be developed to fully understand HM contamination in wheat grains and the risk to human health.

In this study, we choose typical wheat-cultivation areas as the investigation subject, and the HMs (Cr, Ni, Cu, Zn, Cd and Pb) in wheat grains were determined. The specific objectives were as follows: (1) to clarify the HM pollution status in wheat grains in the typical agricultural area, (2) to further evaluate the potential risk to human health, and (3) to find out the potential sources of HMs in wheat by principal component analysis (PCA) and provide so that to lay for soil management in the similar area.

Materials and methods

Description of study area

Guanzhong Plain, located in Shaanxi Province, is one of the main origins of farming culture in China, with favourable climate, fertile soil and great natural conditions. Baoji is an important agricultural wheat-growing region in the Guanzhong Plain (106° 18′~108° 03′ N, 33° 35′~35° 06′ E). The topography of Baoji is complex, with mountains in the north, streams and plain in the central area, Qinling Mountains in the south and Wei River across the middle. Baoji has a temperate monsoon climate and a 183,000-ha wheat area with a total capacity of 811,700 tons (Baoji local chronicles office 2015).

Sampling and analysis

The main wheat cultivars in the study area were Xiaoyan 22 and Xiaoyan 6. The characterization of the soils was as follows: Soil organic matter content was 11.3–16.5 g/kg, CEC was 15.1–17.2 cmol/kg, pH was 7.3–7.7.

According to the distribution of farmland in Baoji, the study area was divided into 4.5 km × 4.5 km. The 81 pairs of surface soils (0–20 cm) and their corresponding wheat grains were collected in the study area during the wheat harvest season in 2016 from six counties (sampling number) (Fig. 1), including Chencang District (11), Fufeng County (18), Fengxiang County (15), Long County (13), Mei County (12) and Qishan County (12). To make the sample homogeneous and representative, we collected the samples by the quincunx point method.

Wheat grains were collected randomly in the same units with the soil samples and brought back to the laboratory. The latitude and the longitude of each sampling site were recorded. During the process of collection, preservation, and pre-processing, samples should be avoided any contact with metal materials to prevent pollution. The wheat grains were air-dried and the husk or glumes were removed by hand, and then the samples were oven-dried at 60 °C to achieve constant weight. Thereafter, grains were ground and stored in a sealed plastic bag for determination. The samples were digested by HNO_3 and $HClO_4$ (4:1) (GB/T17141-1997) and analysed by inductively coupled plasma mass spectrometer (ICP-MS). The total concentrations of Cr, Ni, Cu, Zn, Cd and Pb were determined. Soil samples were air-dried and sieved to pass through the 0.15-mm mesh of a nylon screen and then saved in a zip-lock plastic bag. Four acid digestion (HCl, HNO_3, HF, $HClO_4$) methods (GB 17171-97) were used to digest soil samples, and the abovementioned HMs were also analysed by ICP-MS (PerkinElmer, USA).

Data analysis

Metal pollution index

The metal pollution index (MPI) was applied to evaluate the pollution degree of the HMs in crops in different research areas. The formula follows (Garg et al. 2014):

$$\mathrm{MPI} = (\mathrm{Cf}_1 \times \mathrm{Cf}_2 \ldots\ldots \mathrm{Cf}_n)^{1/n} \quad (1)$$

where Cf_n is the concentration of n HMs in the wheat grain samples (mg/kg). Pollution does not exist when MPI is less than 1 and exists when MPI is greater than 1.

Bioconcentration factor

The bioconcentration factor (BCF) is a significant indicator of plant contamination. It has been commonly used to assess the transfer of heavy metals from soil to wheat grain (Ran et al. 2016). The formula is as follows (Lei et al. 2015):

$$\mathrm{BCF} = C_w / C_s \quad (2)$$

where C_w and C_s are the total concentration of heavy metal in wheat grain and soil, respectively (mg/kg).

Health risk assessment

Health risk assessment can identify the possible risk source in the environment. The source, including gene toxic substances and nongene toxic substances, which can cause harmful effects on human health by exposure was quantitatively evaluated by HRI (Jin et al. 2014). The gene toxic substances refer to the radioactive material and chemical carcinogens such as Cd, Cr and Pb, whereas the nongene toxic substances refer to the non-carcinogenic substances such as Zn and Cu.

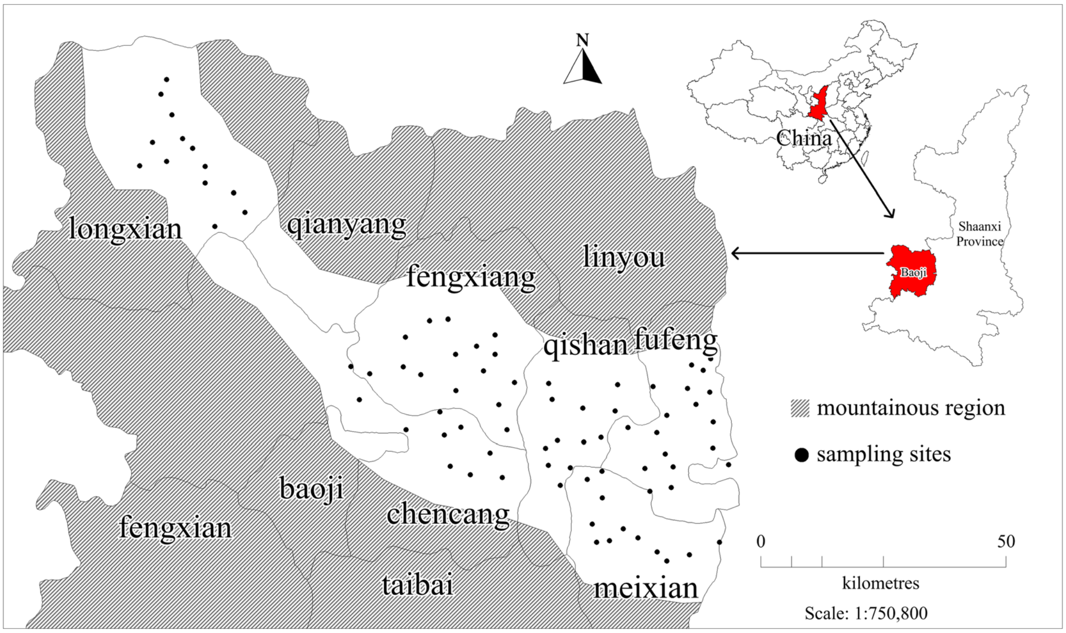

Fig. 1 Sampling sites distribution of wheat in Baoji

(1) Noncarcinogenic risk assessment

Humans could be exposed to HMs via three main pathways: dietary ingestion, dermal contact, and inhalation (Qing et al. 2015). The dietary ingestion accounted for the highest incidence, causing a direct effect (Liu et al. 2013). This study focuses on the human health assessment impacted through wheat dietary consumption, and the exposure assessment equations were as follows (US Environmental Protection Agency [USEPA] 1989):

$$\mathrm{CDI} = \frac{C \times \mathrm{IR} \times \mathrm{ED} \times \mathrm{EF}}{\mathrm{BW} \times \mathrm{AT}} \quad (3)$$

where CDI is the chronic daily intake of noncarcinogens (mg/(kg day)); *C* is the average concentration of HM in wheat grains (mg/kg); IR is the daily wheat intake (g/day); ED is the average human exposure time (years); EF is the exposure frequency (days/year); BW is the body weight (kg) and AT is the average exposure time (days).

Hazard quotient (HQ) is used to calculate the carcinogenic risk of residents consuming wheat in the study area. The formula (USEPA 1989) is as follows:

$$\mathrm{HQ} = \mathrm{CDI}/\mathrm{R_fD} \quad (4)$$

where R_fD is the HM chronic reference dose on human through daily exposure (mg/(kg day)), Values are listed in Table 1 (USEPA 2002).

The hazard index (HI) is added by the noncarcinogenic effects of various metals (Zeng et al. 2015), calculated as:

$$\mathrm{HI} = \mathrm{HQ_1} + \mathrm{HQ_2} + \ldots \mathrm{HQ_n} \quad (5)$$

Chronic risks do not occur if HI < 1 or HQ < 1, whereas potential risks can possibly occur if HQ > 1 or HI > 1.

(2) Carcinogenic risk assessment

For carcinogens, the total exposure was evenly distributed to the whole life cycle in childhood and adulthood. The average daily exposure quantity model for life cycle is as follows:

$$\mathrm{LADD} = \frac{C \times \mathrm{EF}}{\mathrm{AT}} \times \left(\frac{\mathrm{IR_{child}} \times \mathrm{ED_{child}}}{\mathrm{BW_{child}}} + \frac{\mathrm{IR_{adult}} \times \mathrm{ED_{adult}}}{\mathrm{BW_{adult}}} \right) \quad (6)$$

$$\text{Cancer risk} = \mathrm{LADD} \times \mathrm{SF} \quad (7)$$

where LADD is the lifetime average daily dose (mg/(kg day)); AT is the average exposure time (days) and SF is the slop factor (mg/(kg day)). Because of the

Table 1 R_fD values of different heavy metals mg/(kg day)

Heavy metal	Cr	Ni	Cu	Zn	Cd	Pb
R_fD	1.5	0.02	0.04	0.3	0.001	0.0035

carcinogenicity of Cd, Cr, and Pb, this study calculates these metals to assess carcinogenic risk. The SF values were 6.1, 0.5 and 0.0085 (mg/(kg day)) for Cd, Cr and Pb, respectively. The meaning of C, EF, ED and BW was the same as above.

Principal component analysis

PCA was used for metal derivation analysis. It provided a basis for the source inquiry of the HMs in crops and correlated with HMs in farmland absorption characteristics of the plant.

Quality control and statistical analysis

The analysis used the national standard of wheat (GBW10011) as the material for determining quality control. The recovery of the standard samples ranged from 95 to 105% within the scope of permissible error.

Data were tested for normal and non-normal values before the statistical analysis by using the Kolmogorov-Smirnov test. Outliers are the data that are more or less than mean (A) ± 3 times the standard deviation (SD), which will be replaced by a maximum and minimum value (Lei et al. 2014). The following data were removed accordingly: Cr (2 samples), Ni (3 samples), Cu (1 sample), Zn (2 samples), Cd (1 sample) and Pb (1 sample).

Correlation analysis, normal distribution examination and PCA were conducted by SPSS 21. The graphs were charted by Origin 8.6, ArcGIS, Suffer and MapInfo.

Results

HM in wheat grains

The average HM concentrations in wheat grain from the study area were lower than the national standard limit for contaminants in Chinese food (GB 2762-2017). The highest HM concentration was Zn (26.79 mg/kg), followed by Cu (4.41 mg/kg), Cr (0.10 mg/kg), Ni (0.09 mg/kg), Pb (0.03 mg/kg) and Cd (0.01 mg/kg) (Table 2). A total of 3.7% of Zn in wheat grains exceeded the maximum levels (50 mg/kg). The Kolmogorov-Smirnov test showed that HM content in wheat grain followed a normal distribution, except for Ni and Cd, which followed a lognormal distribution.

The spatial distribution of wheat HMs in the study area is shown in Fig. 2, based on the principle of ordinary kriging. The spatial differentiation of Cd was small, and high Ni content samples were mainly located in the Chencang District. Other HMs with higher concentration were mainly found in Qishan, Mei and Fengxiang counties.

Pearson correlation coefficients of HM concentrations in the wheat grain and soil were low (Table S2), except for Cd (0.52**) and Pb (0.55**). Thus, HM content in the soil was not emphasized in this study (Table S1). A biological concentration factor (BCF) was applied to clarify the capability of the different HMs accumulated in wheat grains. BCF is the ratio of HM in wheat and HM in soil, reflecting different HM accumulation (Table 1). The BCF of HMs in wheat was in the order of Zn > Cu > Cd > Ni > Cr > Pb, indicating that wheat absorbed more Zn and Cu than other metals. In addition, the BCF of Cd was the largest, which was 27.9, 46.8 and 61.0 times higher than Ni, Cr and Pb, respectively.

MPI could unify the degree of HM pollution in wheat grains gathered from different regions. The overall MPI was less than 1, indicating that no pollution is noted at present. The MPI of different counties followed the order as Qishan (0.286) > Mei (0.286) > Chencang (0.263) > Fengxiang (0.263) > Long (0.234) > Fufeng (0.217) (Fig. 3). The difference of MPI was minimal, ranging from 0.2 to 0.3, demonstrating that HMs did not reach pollution level in the study area.

Human health risk assessment

The HQ and cancer risks were calculated by Eqs. 6 and 7. The annual intake of food for Chinese rural inhabitants was 147.10 kg in 2016. Residents in Shaanxi consumed cooked wheat-containing food, with wheat accounting for 78.23% of their food intake. Therefore, the daily intake of wheat for adults and children in Shaanxi was 315.28 and 105.09 g per day, respectively (Zhang et al. 2009; National Bureau of Statistics of China 2017; Ran et al. 2016). Other parameters are shown in Table 3 (National Bureau of Statistics of China 2012; USDOE 2011; USEPA 1989).

Noncarcinogenic risk assessment

Table 4 lists the mean and the range of each HM in the wheat grain of the HI according to Eqs. 3 to 5. The

Table 2 Heavy metal concentration of wheat grain and soil in Baoji wheat-cultivation area

Heavy metals	Distribution	Heavy metal concentration in wheat (mg/kg)				Tolerance limits of Chinese standards (mg/kg)	Heavy metal in soil (mg/kg)	BCF	Percent exceeded standards%
		Range	Median	Mean	SD				
Cr	Normal	0.001~0.58	0.09	0.10	0.10	1.00	72.20	0.002	0
Ni	Logarithmic	0.012~0.35	0.07	0.09	0.07	1.00	35.80	0.002	0
Cu	Normal	2.68~9.84	4.41	4.61	1.26	10.00	27.56	0.173	0
Zn	Normal	9.92~60.74	24.19	26.79	9.83	50.00	86.61	0.325	3.70
Cd	Logarithmic	0.005~0.035	0.012	0.013	0.007	0.10	0.19	0.070	0
Pb	Normal	0.013~0.068	0.03	0.03	0.01	0.20	28.85	0.001	0

Reference from Chinese Hygiene Standard for wheat (GB2726-2017) and Agricultural standard in China (NY861-2004)

results of the HQs and HI for individual metals in adults and children are demonstrated in Fig. 4. The USEPA acceptance criterion is 1. Except for Fufeng County, the total HIs of the other counties were larger than 1, indicating that potential noncarcinogenic risks existed for consumed wheat in the study area. The order of HI was Fufeng (0.98) < Long (1.05) < Qishan (1.11) < Chencang (1.12) < Fengxiang (1.23) < Mei (1.37) in adults. The same order of risk for children was also observed. The highest risk county was Mei. In Mei, the potential risk of HMs followed the order Cu > Zn > Cd > Pb > Ni > Cr, and the HQ of Cu and Zn accounted for 49.77% and 38.5%, respectively, of the total HI.

Carcinogenic risk assessment

Figure 5 shows that the ranking of carcinogenic risk factors in different counties was Mei (1.21×10^{-6}) > Qishan (1.11×10^{-6}) > Long (8.89×10^{-7}) > Chencang (8.67×10^{-7}) > Fengxiang (7.60×10^{-7}) > Fufeng (6.75×10^{-7}), which was lower than the maximum acceptable risk level of the International Commission of Radiological Protection (5.0×10^{-5}). Thus, wheat ingested in this study area will not cause cancer risk to humans. The carcinogenic risks of Cd and Cr were considerably higher than those of Pb and were the major HMs with carcinogenic risk in this study, with an average ratio of 60.60% and 39.19%, respectively.

PCA of HMs in wheat grains

In this study, the Bartlett's test of sphericity result, the Kaiser–Meyer–Olkin (KMO) value gained from PCA was 0.637 (KMO value higher than 0.5 is the evaluation basis), and the results were gathered after the orthogonal rotation (Table 5). Two principal components were partitioned from the variables with eigenvalues exceeding 1. They reflected 51.68% of the total characterization of six HMs. PC1 was dominated by Cr, Cu, Zn, Cd and Pb, accounting for 37.72% of the total variance. The higher load of Ni was found in PC2, with an 18.96% contribution rate. These results indicated that the source of Ni was different from other HMs.

Discussion

Characteristic of heavy metal in wheat grains from study area

Previous studies found that agricultural activity in farmland was one of the reasons that heavy metals accumulated in soil and absorbed by crop. In the typical livestock manure application areas of Yucheng in Shandong Province, Cr, Ni and Cd in the wheat grains exceeded the limits of the national standard (Ye et al. 2013). In the wastewater irrigation area of Abbottabad district in Pakistan, Cr and Ni in the wheat grains were beyond recommended dietary limits (Hassan et al. 2013). In the cultivated land of Ropar wetland in India, Pb and Zn in wheat grain had above the safe limit (Sharma et al. 2018). In this study, a total of 3.7% of Zn in wheat grains exceeded the maximum levels. The averages of HM in the study area were lower than the Chinese national standard limit (GB 2762-2017); there was no direct risk.

Soil is not only the direct source of nutrient elements for wheat but also the source of toxic HMs (Khan et al. 2017). Therefore, clarification of the HM pollution in soil, identification of the pollutant sources and provision of basic management to reduce HM contents in wheat

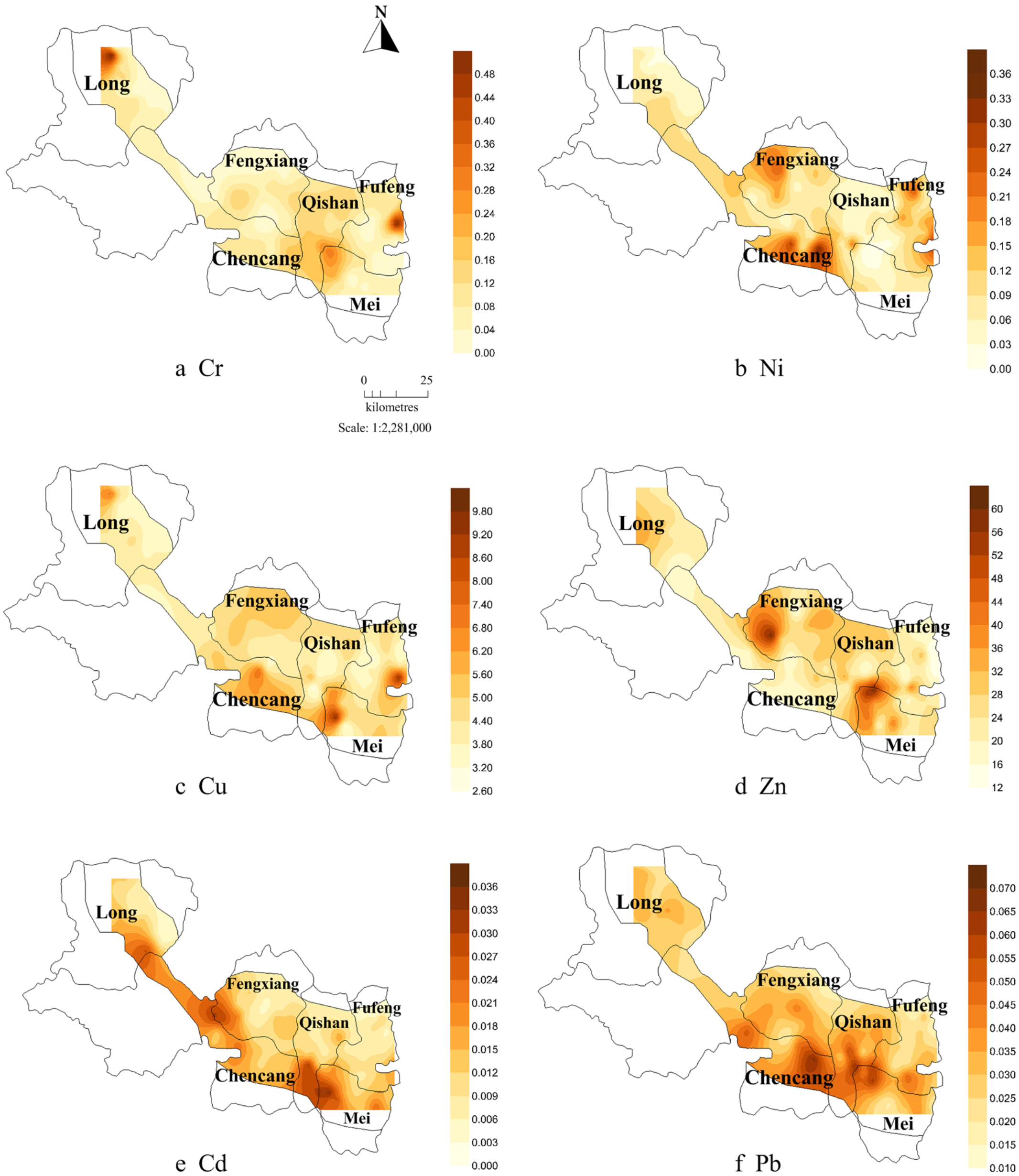

Fig. 2 Spatial distribution maps of heavy metal contents with chromium (a Cr), nickel (b Ni), copper (c Cu), zinc (d Zn), cadmium (e Cd), and lead (f Pb) in wheat grain of Baoji (mg/kg)

grain are necessary. From the results of Tables 1 and S1, six HMs found in soil samples were all lower than the National Environment Quality Standard of soil (GB15618-1995GB), but numerous samples showed HMs higher than the background values in Shaanxi. Thus, the food safety of wheat in these areas is not guaranteed. However, a significant correlation between wheat grain and soil was found only for the Cd and Pb concentrations (Table S2). Consequently, a more appropriate parameter—the biological enrichment coefficient (BCF)—was selected to reflect the migration and transformation of the HMs from soil to wheat. The BCF was calculated by Eq. 2.

Because zinc is an essential micronutrient for plants, and the mobility of Zn from soils to crops

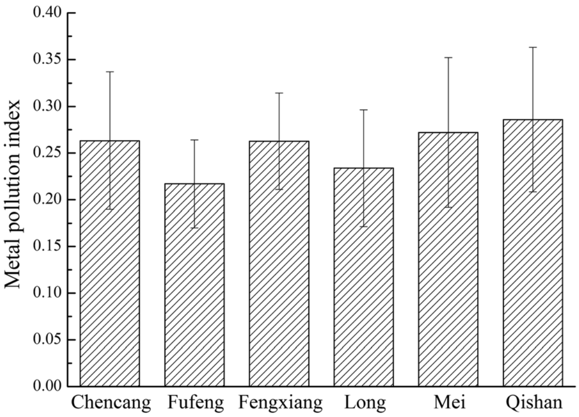

Fig. 3 Metal pollution index of wheat grain in study areas

is higher than that of other metals, Zn can easily transfer from the soil to the wheat grains (Boussen et al. 2013). This phenomenon could explain the BCF of Zn (0.32) in wheat grains, and Zn was the highest among all tested HMs in agricultural soils. In contrast, Pb mainly existed in a carbonate combination (24–55%) and ferric-manganese oxidation state in soil, and the mobility and bioavailability of Pb was low under natural conditions (Iavazzo et al. 2012). Therefore, the lowest BCF was found in Pb (0.001), even though the element existed at a high value and exceeded the background values (76.97%). Previously conducted investigative reports from Henan (Xing et al. 2016), Hebei (Ru et al. 2010) and Tianjin (Zeng et al. 2015) confirmed that the order of the BCF of the metals contained in the Baoji wheat grains was as follows: Zn > Cu > Cd > Ni > Cr > Pb.

Health risk assessment of Baoji residents

The average HM concentrations in Baoji wheat grain were lower than the national standard limits for contaminants in Chinese food, indicating no direct risk at present. However, HMs can bioaccumulate in the human body as the feeding time and intake increase. Therefore, the potential health risks of these pollutants need to be assessed (Qaswar et al. 2017; Ahmed et al. 2015). The noncarcinogenic HI greater than 1 in this study can be explained for three reasons. (1) The total risk of Zn and Cu accounted for 88.30% in HI, which was considerably higher than observed for the other metals. Cakmak (2008 and 2010) suggested that the average of Zn in global ordinary wheat grain should be 35 mg/kg or within 40–60 mg/kg to reach nutritional requirements. From the perspective of nutrition, the Zn concentration in the wheat grain of Baoji is acceptable, as is true for the Cu. However, in calculating health risk assessment, both the Zn and Cu contents inevitably increased the risk coefficient concurrently. (2) The USEPA evaluation system is used mostly to assess health risk of human. However, the standards of different country are different (Reboredo et al. 2018; Sharma et al. 2018; Xing et al. 2016), due to their specific soil types (Krami et al. 2013), plant species (Pajević et al. 2018) and even dietary habits of people are different. The priority monitoring list of pollutants made by the Chinese Ministry of Environmental Protection does not include Zn (Jiang et al. 2016). Thus, the health risk assessment system for Chinese residents should be

Table 3 Parameters for human health assessment model

Factor	Definition	Unit	Value	
			Children	Adult
IR	Concentration of the heavy metals	g/day	105.09	315.28
ED	Exposure duration	Years	6	30
BW	Average body weight	kg	16.20	61.80
EF	Exposure frequency	Days/year	350	
AT	Average time	Days	365×ED[a] 365 × 74.8[b]	

[a] Noncarcinogenic risk

[b] Carcinogenic risk

Table 4 Noncarcinogenic risk for adults and children for consuming wheat grains

Heavy metal	Adult		Children	
	Range	Mean	Range	Mean
Cr	2.72E−06~1.88E−03	3.43E−04	3.46E−06~2.39E−03	4.36E−04
Ni	3.03E−03~8.66E−02	2.20E−02	3.86E−03~1.10E−01	2.79E−02
Cu	3.27E−01~1.20E+00	5.64E−01	4.16E−01~1.53E+00	7.18E−01
Zn	1.62E−01~9.90E−01	4.37E−01	2.06E−01~1.26E+00	5.56E−01
Cd	2.24E−02~1.70E−01	6.41E−02	2.86E−02~2.16E−01	8.15E−02
Pb	1.85E−02~9.44E−02	4.62E−02	2.35E−02~1.20E−01	5.88E−02
HI	5.33E−01~2.54E+00	1.13E+00	6.78E−01~3.33E+00	1.44E+00

established based on the trace elements in the human body, diet habits, and capability of the crop to absorb HMs. (3) This study used the HMs of pulverized grain to evaluate risk. Most of the HMs exist in the outermost part of wheat grains (testa and the aleurone layers), which is lost in the flour processing, with the HM contents being reduced (Xing et al. 2016; Ajiboye et al. 2015). Humans consume flour directly rather than whole meal. Therefore, the potential risk was overestimated by the HM content of the whole wheat flour. This difference should be addressed in future studies.

Herein, we evaluated the risk of HMs such as Cr, Ni, Cu, Zn, Cd and Pb and Cd, Cr and Pb for carcinogenic risk. The carcinogenic risks include children and adults because such risks can be involved in the whole life cycle of the human body (Lei et al. 2015). The carcinogenic risk of the Cd and Cr was considerably higher than that of the Pb (Fig. 5); this result is consistent with the health risk assessment of HMs in wheat grains around Zhengbian road in Zhengzhou, Henan Province (Li et al. 2009) and in the outskirts of Beijing (Zhu et al. 2011). All those studies showed that Cd had the highest cancer risk, which is related to the stronger ability of Cd to be absorbed by the wheat grain. In conclusion, the carcinogenic risk of the HMs was low and did not exceed the risk range in wheat grains in the study area.

The preliminary source analysis of HMs in wheat grains

PCA analysis helps to infer the preliminary source of the HMs in the wheat grains into two parts. The study area was the typical agricultural cultivation region, and industry was developed in the surrounding area during past decades. Therefore, PC1 was inferred as agricultural activities (including fertilization, wastewater irrigation and pesticides spraying), which is a main contributor of the Cd, Cr, Cu, Zn and Pb. PC2 was closely related

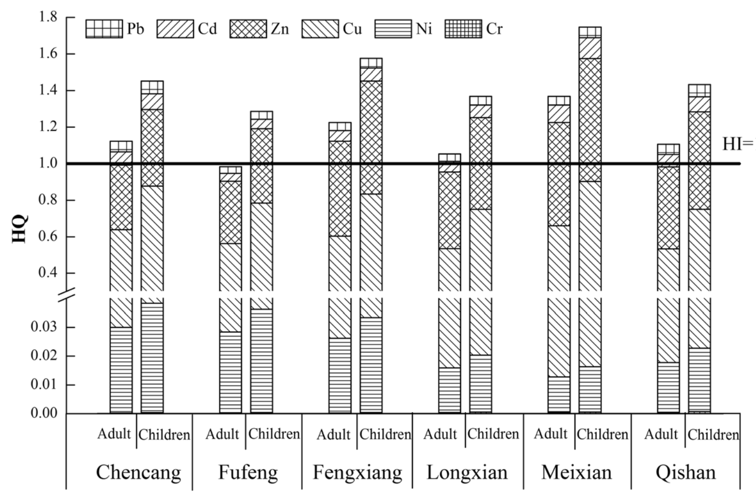

Fig. 4 Noncarcinogenic risks through wheat consumption in study areas

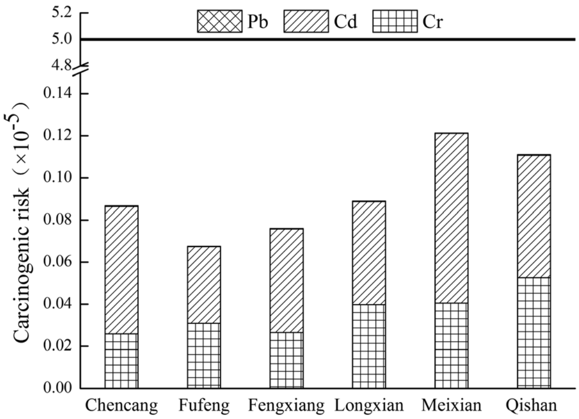

Fig. 5 Carcinogenic risks through wheat consumption in study areas

with industrial activity, referring to the Ni and Ti manufacture, which causes emissions of Ni to the local environment during the long-term production activities. Baoji is a city with well-developed industries of Ni and Ti. In some plants, mainly located in the Chencang District and Weibin District, the exhaust gas of the Ni smelters discharged some HMs to the atmosphere (He et al. 2014). The emission of smoke exhibited a high concentration from the drying haze which is a process of production technology and monitoring showed that the Ni emission (34.95 kg) was dozens or even hundreds of times higher than that of other metals (Zhang 2013). Therefore, the Chencang District mainly showed higher values for Ni in the wheat grain.

Mei County had the highest risk of carcinogens and noncarcinogens, whereas Fufeng County had the lowest risk (Figs. 4 and 5). With the rapid industrial development, a large-scale science and technology industrial park has been established in Mei County, becoming the leading industry in the whole province. Meanwhile, Fufeng County focuses on the development of agriculture and tourism and has a standardized demonstration area for the breeding of improved wheat varieties at the national level (Baoji local chronicles office 2015). Therefore, the noncarcinogenic and carcinogenic risk factors of Fufeng were the lowest among the six counties in this study. In addition, irrigation is necessary in the wheat-growing season in the study area. Local farmers usually use river water from nearby farmland for irrigation. Irrigation water may also serve as a pollution route (Zeng et al. 2015; Wang et al. 2017). Especially, cement, pitch and electricity production plants in Fengxiang County; pig farms; power plants; and other factories in the Chencang District and paper mills in Qishan County all release HMs to nearby rivers. As a typical agricultural area, Baoji has a long history of farming. Long-term fertilization and pesticide spraying are also sources of pollution in farmland ecosystems that cannot be ignored (Wang et al. 2005; Rahman et al. 2014).

Table 5 components matrix with varimax rotation of heavy metals in wheat grains

Heavy metal	PC1	PC2
Cr	0.53	0.13
Ni	0.05	0.89
Cu	0.70	0.29
Zn	0.63	− 0.44
Cd	0.57	− 0.26
Pb	0.67	− 0.09
Eigenvalue	1.96	1.14
Variance proportion	37.72	18.96
Accumulation contribution	32.72	51.68

Conclusion

The mean grain concentration levels of HMs in the Baoji wheat-cultivation areas are lower than the Chinese food hygiene standard. The BCF of six HMs was in the order as follows: Zn > Cu > Cd > Ni > Cr > Pb. The sources of Cr, Cu, Zn, Cd and Pb were different from Ni, with the former being derived from the agricultural activities of farmland. Conversely, the Ni in this wheat grain originated from the emissions of Ni smoke and dust discharged into the environment from Ni and Ti smelting industries.

The result of the human health risk assessment showed that the wheat grain (whole wheat flour) in Baoji wheat-cultivation areas has a potential noncarcinogenic risk, and Cu and Zn were the main contributors. The carcinogenic risk was lower than the acceptable coefficient (security coefficient) regulated by the International Commission on Radiation Protection and the USEPA. More attention should be paid on how to prevent and control the input of Cu, Zn, Cr and Cd through the agricultural activities in the fields of the study area and to further mitigate their related human health risks.

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