



Distributions of Heavy Metals in Rice and Corn and Their Health Risk Assessment in Guizhou Province

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Abstract

Long-term exposure to heavy metals from high geological background area may lead to varieties of diseases. Therefore, risk assessment from agricultural products in these areas was crucial to ensure the health of customers. However, the effects of geological background on distributions of heavy metals and their accumulation in plant remain unclear. This study aimed to determine the distributions of mercury (Hg), cadmium (Cd), arsenic (As), lead (Pb), chromium (Cr) and copper (Cu) in 1036 corn and rice samples collected from 9 locations in Guizhou province and to evaluate their health risks. The concentrations of Hg, Cd, As, Pb, Cr and Cu in these two crops were analyzed by inductively coupled plasma-mass spectrometry (ICP-MS), and their health risk were estimated by the target risk quotient approaches. Results showed that the mean concentrations of Hg, Cd, As, Pb, Cr and Cu in corn and rice were 0.01, 0.07, 0.06, 0.27, 0.56 and 1.86 mg/kg which were lower than their respective maximum allowable concentrations (MAC) of 0.02, 0.20, 0.70, 0.20, 0.50 and 10.00 mg/kg except Pb and Cr. The concentrations of Cr and Cu in corn were higher than in rice while Cd, As and Pb in rice were higher than in corn. Moreover, the distributions of Hg, Cd and Cu in corn and rice samples were mainly observed from QDN located at southeast of Guizhou province while Pb, As and Cr were most detected at ZY, QXN and BJ areas, southwest zone. The hazard indices (HIs) values for corn and rice were 0.20 and 2.61. The high HIs (> 1) in rice indicated that the health risk of heavy metals in rice was relatively high and Pb was the major component that attributed to the risk, followed by Cd. These results could provide a reference for the distributions of heavy metals in agricultural products in Guizhou province under crop cultivation conditions, and to provide scientific basis for health risk assessment and ensure food safety.

Keywords Heavy metal · Corn · Rice · Hazard indices · Health risk assessment

Rice and corn, as the main components in our diet, provide us the sources of dietary fiber, sugar, protein, and vitamins. They are the substantial and highly consumed crops all

around world with the average consumption amount ranged from 35.2 to 340 g/day (Huang et al. 2013; Yu et al. 2017). However, the contamination of rice and corn by heavy metals are raising more and more public health concern. The amount of contaminated and production reduced agricultural products induced by heavy metals were 12 million and 10 million tons, respectively, with the total economic loss of 20 billion RMB every year (Ao et al. 2019). The sources of heavy metals in rice and corn might be from the polluted soil and irrigation water. Heavy metals were easily absorbed by rice and corn and accumulated in edible parts. For example, Cd, As and Pb was frequently enriched in rice (Norton et al. 2014; Perera et al. 2016). Heavy metals could be biologically accumulated in organisms even at low concentrations which may lead to various diseases including Itai-Itai disease, cancer, leukemia, gastrointestinal colitis, brain function disorders and genetic toxicity (Nejabat et al. 2017).

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Heavy metals are ubiquitous in environment due to natural occurrence as well as fossil fuels usage (Wu et al. 2018; Li et al. 2013). Meanwhile, the discharge of urban industrial production and household waste are also the main sources of heavy metal pollutants in farmland around cities (Zou et al. 2015). Heavy metals could be accumulated in crops through water, soil and air (Yang et al. 2018). It had been reported that heavy metal residues were determined in various plants. Naseri et al. (2015) and Shakerian et al. (2012) reported the average concentration of Pb in Iranian rice samples which were 980 ± 280 $\mu\text{g}/\text{kg}$ and 68 ± 19 $\mu\text{g}/\text{kg}$, while the concentration of Cd were 340 ± 70 and 62 ± 19 $\mu\text{g}/\text{kg}$, respectively. The concentrations of Cd and Pb in 9 vegetables (lettuce, spinach, cauliflower, pudina, celery, coriander, parsley, onion and radish) collected from wastewater irrigated suburban area in Titagarh, India were 17.79 mg/kg and 57.63 mg/kg, respectively (Gupta et al. 2008). The distributions of Cd in corn samples from Dongdagou and Xidagou stream valleys were determined, and the results showed that the levels of Cd were ranged from 0.06 to 0.09 mg/kg, which were higher than the maximum limits (0.05 mg/kg) (Zang et al. 2017). The average concentrations of Cd, Cr, Pb and Cu in rice samples were 19.99 ± 19.39 , 24.94 ± 24.80 , 70.47 ± 64.20 and 2.26 ± 0.55 mg/kg, respectively, which were higher than in wheat samples with the average concentrations of 12.77 ± 2.15 , 20.99 ± 6.4 , 64.69 ± 24.84 and 1.55 ± 0.24 mg/kg, respectively (Wei et al. 2020).

Human may get exposed to heavy metals through inhalation, skin contact, water or food intake (Liu et al. 2013). It was reported that heavy metal pollution caused more than 420,000 death and 600 million diseases every year around the world (Zheng et al. 2020). Therefore, there was growing demand for assessing the health risk posed by heavy metals in crops. The carcinogenic health risk was assessed by using the method of increased lifetime cancer risk (ILCR) and the non-carcinogenic risk was evaluated by the methods of estimated daily intake (EDI), target hazard quotient (THQ) and hazard indices (HI). In current study, these methods were used to evaluate the risk of heavy metals. The potential health risk in corn, rice and vegetables of Cd, Hg, Pb and Cr were assessed, and the results showed that the target hazard quotients (THQs) were 0.54, 0.75, 0.21 and 1.81 and the hazard indexes (HI) for those heavy metals was 2.18, suggesting its high likelihood of hazards to human (Wei et al. 2019). Pan et al. (2016) researched the potential health risks of As, Cd, Cr, Pb, Ni and Hg in marketed vegetables of Zhejiang, China which showed that the HI was less than 1 (0.32), while the HI of Hg and Cd at high exposure (97.5% levels) were 1.01 and 1.85. Zheng et al. (2020) reported the risks of Cd, Cr, Pb, Hg, As in rice, corn, vegetable, and fruit samples in the Pearl River Delta, and found that the highest risks were in rice and corn samples with the mean and maximum HI values (95% level) of 1.7 and 5.7, respectively. The average

THQs of Cd, Cu, Hg and Pb in corn were 0.068, 0.349, 0.018, 0.614, respectively, with the mean HI (1.61) higher than 1 which may pose a risk to human (Gu et al. 2019).

Guizhou Province, located at southwest of China, is the typical karst landform and its lithostratigraphy is dominated by carbonate rocks. The relatively high heavy metals background in Guizhou soil had been reported which was mainly attributed by the two sources: one was due to the effects of weathering and leaching, heavy metals are transferred and released into soil, which lead to the high geological background of heavy metals in Guizhou (Kong et al. 2018a). Another reason was the contamination from industry, agricultural production and urbanization including the irrigation with wastewater or polluted water, atmospheric sedimentation, production of electronic products, household appliances, Pb–Zn mining/smelting activities (Huang et al. 2020). Both natural factors and human activities may lead to high concentrations of heavy metals in corn and rice growing soil which may pose a high ecological risk to human health. Research showed that high Hg concentration was mainly distributed in GY, QDN, TR and QN, while Cd was mainly detected in GY and QN (Song et al. 2005). Bi et al. (2006) reported that high Cd and Pb were mainly distributed in BJ, Qianxi and Hezhang County, and high Cu and Cr were mainly distributed in Pu 'an county, QXN (Zhang et al. 2018). Rice and corn are the daily staple food and representative coarse diet in the dietary structure of residents in Guizhou province. Due to the relatively high heavy metals background in soils, and long-term historical of Pb–Zn mining/smelting activities, the potential risk of contamination of agricultural products had gained more and more attention (Luo et al. 2018). However, research on the distributions of heavy metals in corn and rice in Guizhou were scarce and their health risk were still unclear.

In this study, the distributions of 6 heavy metals including Hg, Cd, As, Pb, Cr, Cu in corn and rice samples collected from 9 districts of Guizhou province were detected and their potential risk to human were evaluated. The objectives of this study were to: (1) to determine the concentrations of 6 heavy metals in corn and rice samples; (2) to evaluate their distribution difference at 9 sampling locations; (3) to evaluate the health risk to human.

Materials and Methods

Sampling Locations

Guizhou province is located at southwest of China with the latitudes and longitudes ranges from $24^{\circ}37'$ to $29^{\circ}13'N$ and $103^{\circ}36'$ to $109^{\circ}35'E$, respectively. The annual average temperature is 14 – 16°C and the annual precipitation is generally 1100–1400 mm (Deng et al. 2004). As one of

the fastest growing province in China, Guizhou has a total population of 35.81 million and area of 176,167 square kilometers, which is consisted of 9 regions: Anshun (AS), Bijie (BJ), Guiyang (GY), Zunyi (ZY), Tongren (TR), Liupanshui (LPS), Qian dongnan (QDN), Qian xinan (QXN) and Qiannan (QN). In this study, 638 corn and 398 rice

samples were randomly collected from 56 counties (9 regions) during harvesting season (Fig. 1), and specific sample information was showed in Table.1. At each sampling site, a five-point sampling method was used to collect samples with 250 g for each sample. They were then stored in a polyethylene bag and transferred to the laboratory for analysis.

Fig. 1 Corn and rice sampling points distribution map in Guizhou, China (by ArcGIS 10.6)

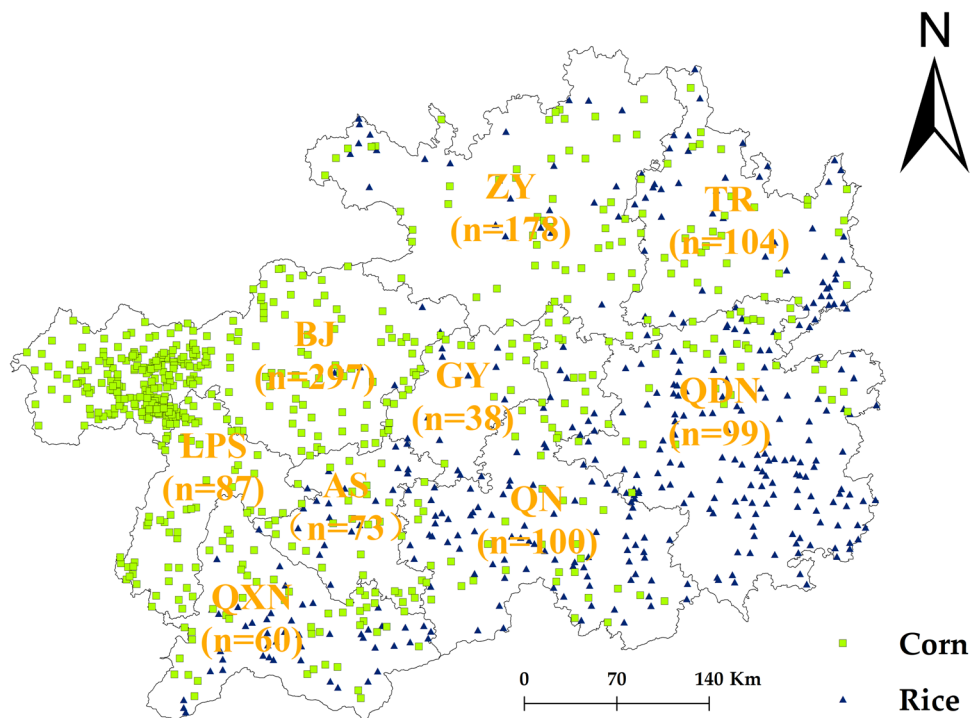


Table 1 Sampling locations and sample information in the study area

Region	Crops	Sample number	East longitude	North latitude	Altitude	The average area size of a crop plot (km ²)
AS	Corn	48	105.36–106.43	25.44–26.52	1211.95	1.40
	Rice	25				
BJ	Corn	237	103.71–106.57	26.44–27.73	1859.14	1.32
	Rice	60				
GY	Corn	21	106.16–107.19	26.30–27.34	1121.26	1.61
	Rice	17				
ZY	Corn	50	105.74–107.99	27.25–28.86	767.63	1.36
	Rice	128				
TR	Corn	35	107.81–109.42	27.26–29.07	633.30	1.12
	Rice	69				
LPS	Corn	87	104.35–105.67	25.38–26.86	1720.38	2.07
	Rice	69				
QDN	Corn	30	107.47–109.50	25.56–27.49	688.13	1.25
	Rice	69				
QXN	Corn	30	104.78–106.44	24.69–25.91	1121.03	1.06
	Rice	30				
QN	Corn	54	106.31–108.21	25.17–27.44	944.44	1.23
	Rice	46				

Sample Analysis

After shelled, corn and rice samples were grounded through a 0.15 mm nylon sieve and sealed in a new polyethylene bag. They were homogenized and powdered in a blender (7011S, K-corporation, USA). The dried samples (0.50 g) were added with 10 mL HNO₃ in sealed polytetrafluoroethylene (PTFE) digestion vessels, standing for 1 h. Samples were then placed in a microwave oven with the power at 1.50 kW and digested as following steps: First, samples were heated to 120°C within 8 min, and maintained at 120°C for 5 min, and then heated to 150°C within 5 min, and maintained at 150°C for 5 min. After that, samples were heated to 195°C within 10 min, and maintained at 195°C for 30 min (Lei et al 2015). After cooling to room temperature, the digest solution was degassed by an ultrasonic wave (KS-120AL, Shanghai, China) for 5 min, then the digest was diluted to 25 mL, and filtered through a 0.45 μm filter for analysis. Blank samples were carried out by following the above steps. Samples from the same collection point were conducted with 3 replicates.

Hg, Cd, As, Pb, Cr and Cu were determined by ICP-MS (NexION 2000, Perkin Elmer, USA). The multi-element standard solution was used as external standard solution. The operational conditions of ICP-MS were as follows: radio-frequency (RF) power 1.50 kW, plasma gas flow rate 15.00 L/min, auxiliary gas flow rate 1.00 L/min, carrier gas flow rate 1.07 L/min, make up gas flow rate 1.00 L/min, spray chamber 3°C, sampling depth was 8.2 mm and dwell time 20 ms. The mixed standard curves were prepared by diluting the multi-element standard solution (1000 μg/mL) into series concentrations of 0.0, 0.5, 1, 10, 50, 100, 200, 500 and 1000 μg/L. The correlation coefficients of Hg, Cd, As, Pb, Cr and Cu were 0.9992, 0.9999, 0.9996, 0.9992, 0.9998 and 0.9996, respectively. The calculation formula was as follows:

$$X = \frac{(\rho - \rho_0) \times v \times f}{1000 \times m}$$

where X (mg/kg) is the measured concentration of samples, ρ (μg/L) is the detected concentration of samples, ρ₀ (μg/L) is the detected concentration of blank samples, v (mL) is the volume of digestive solution, f is the dilution factor, m (g) is the sample weight.

Health Risk Assessment of Heavy Metal

The health risk of Hg, Cd, As, Pb, Cr and Cu in corn and rice with long-term exposure were assessed based on the estimated daily intake (EDI), target hazard quotient (THQ) and hazard indice (HI) (US EPA, 1989). The calculation formulas of EDI, THQ, and HI were as follows:

$$EDI = C \times C_{on} / BW \quad (1)$$

$$THQ = EDI / RfD \quad (2)$$

$$HI = \sum_{n=1}^i THQ_n \quad (3)$$

where C (mg/kg) is heavy metal concentrations in corn and rice, C_{on} (g/person/d) is the daily average consumption, BW (kg/person) represents body weight, RfD (mg/kg/d) is the oral reference dose. Based on the reports, the average corn and rice consumption are 35.2 and 420.0 g/d for adult, and the average body weight for adults is 65.7 kg. The RfD values for Hg, Cd, As, Pb, Cr and Cu are 0.0003, 0.001, 0.0003, 0.0015, 1.5, 0.04 mg/kg/day, respectively (Yu et al. 2017; Wu et al. 2013). Humans are considered to be safe if THQ or HI < 1.

Statistical Analysis

One way analysis of variance (ANOVA) was used to evaluate the significant differences of heavy metal concentrations collected from different locations and crops. Data analysis was performed with SPSS software, version 13.0 (IBM Corporation, Armonk, New York, USA).

Results

Concentrations of Heavy Metals in Corn and Rice

The concentrations of Hg, Cd, As, Pb, Cr and Cu in corn and rice were ranged at 0.002–0.11 (mean 0.007), 0.001–6.35 (mean 0.07), 0.002–1.12 (mean 0.06), 0.001–14.30 (mean 0.27), 0.01–9.27 (mean 0.56), 0.10–27.00 (mean 1.86) mg/kg, respectively. The percentages of rice and corn samples that exceeding the maximum allowable concentration (MAC) value was 14.14% for Hg (MAC value 0.02 mg/kg), 23.17% for Cd (MAC value 0.2 mg/kg), 0.26% for As (MAC value 0.7 mg/kg), 26.89% for Pb (MAC value 0.2 mg/kg), 32.03% for Cr (MAC value 0.5 mg/kg) and 1.16% for Cu (MAC value 10.0 mg/kg). The maximum concentrations of Cd (6.35 mg/kg), Cr (9.27 mg/kg) and Pb (14.30 mg/kg) were nearly 32 to 72-fold higher than their corresponding MAC levels.

As shown in Fig. 2, the detected rates for Hg, Cd, As, Pb, Cr and Cu in rice were 71.39%, 96.20%, 93.42%, 53.42%, 79.24% and 100.00%, respectively. For corn samples, the detection rates were 65.52%, 94.67%, 61.44%, 68.34%, 87.46% and 100.00%, respectively. In addition, the total metal concentration in rice was 0.46 mg/kg which was higher than in corn (0.16 mg/kg). For rice, the concentrations of

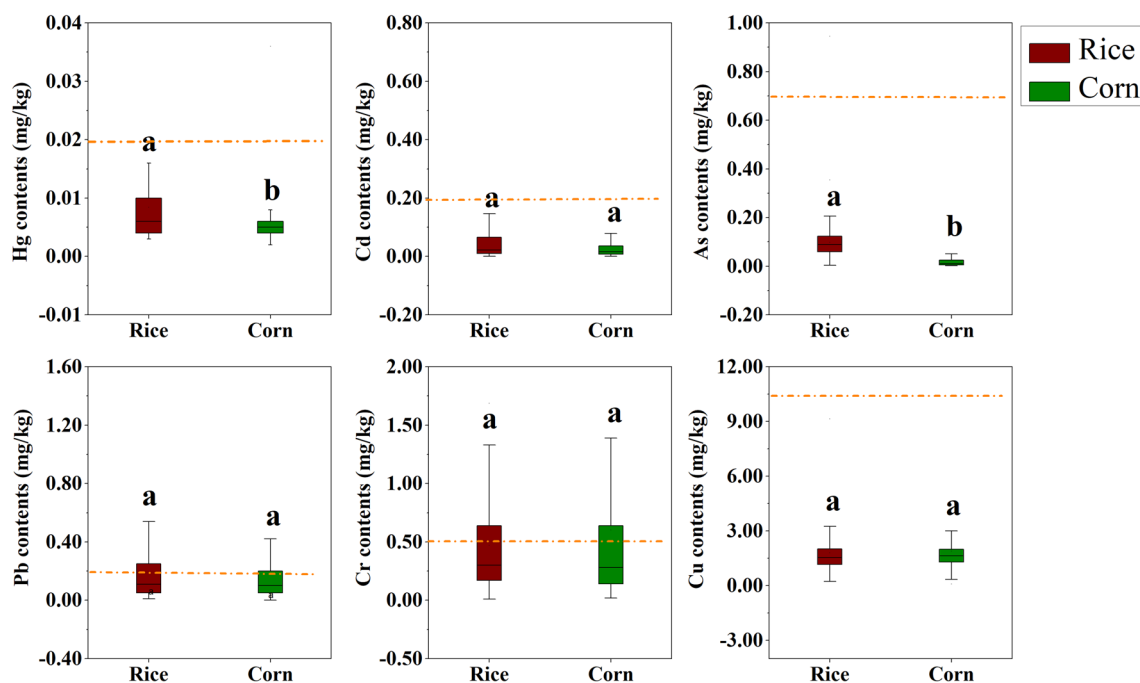


Fig. 2 Concentrations of heavy metal in corn and rice samples in Guizhou province. The orange dotted line indicated the permissible level of each heavy metal, and the concentrations of heavy metals

significantly differed between corn and rice when different lowercases existed. The significance level (α) was set as 0.05

Hg, Cd, As, Pb, Cr and Cu were ranged from 0.003–0.06 (mean 0.01), 0.001–6.35 (mean 0.09), 0.03–0.95 (mean 0.10), 0.01–7.12 (mean 0.27), 0.01–2.93 (mean 0.47), 0.24–27.00 (mean 1.81) mg/kg ($n=398$), and for corn were 0.02–0.11 (mean 0.01), 0.001–4.78 (mean 0.06), 0.02–1.12 (mean 0.02), 0.001–14.30 (mean 0.26), 0.02–24.40 (mean 0.60), 0.10–21.80 (mean 1.90) mg/kg ($n=638$), respectively. The concentrations of Cr and Cu were higher in corn than in rice while Cd and Pb were higher in rice than in corn.

Distributions of Heavy Metals in Rice and Corn Collected From Different Sampling Locations

The mean concentrations of Hg, Cd, As, Pb, Cr and Cu in 9 locations (AS, BJ, GY, ZY, TR, LPS, QDN, QXN, QN) were 0.01, 0.07, 0.06, 0.27, 0.56 and 1.86 mg/kg ($n=1036$), respectively. The highest levels of Hg (0.11 mg/kg), Cd (6.35 mg/kg) and Cu (27.00 mg/kg) in rice and corn samples were detected at QDN locations and the lowest concentrations of Hg (0.002 mg/kg), Cd (0.02 mg/kg) and Cu (1.57 mg/kg) was determined at BJ, LPS and ZY. Besides, the highest levels of As (1.12 mg/kg), Pb (14.30 mg/kg), Cr (24.40 mg/kg) were detected at QXN, ZY and BJ locations, while the lowest concentrations of As (0.02 mg/kg), Pb (0.15 mg/kg) and Cr (0.40 mg/kg) were determined at BJ, GY and ZY. In a whole, the highest concentrations of Hg, Cd and Cu were mainly detected from QDN located at southeast

of Guizhou province while the highest concentrations Pb, As and Cr were detected in ZY, QXN and BJ samples, southwest zone. The distributions of Hg, Cd, As and Cu in Guizhou were decreased by a rank of QXN > GY > BJ. Similar distributions of Pb and Cd in rice and corn were observed with ZY and TR higher than AS and LPS areas (Fig. 3). To be specific, the average concentrations of Cr in agricultural products from those 9 areas showed a decreased order of GY (0.82 mg/kg) > QDN (0.64 mg/kg) > LPS (0.61 mg/kg) > AS (0.60 mg/kg) > BJ (0.59 mg/kg) > TR (0.54 mg/kg) > QXN (0.44 mg/kg) > QN (0.42 mg/kg) > ZY (0.39 mg/kg).

Health Risk Assessment of Heavy Metals in Corn and Rice

The EDIs, THQs and accumulative HI values of heavy metals in corn and rice samples were presented in Table 2 and Fig. 4. For corn consumption, the THQs of Hg, Cd, As, Pb, Cr and Cu were 0.01, 0.07, 0.004, 0.09, 0.0002 and 0.03, respectively, with the accumulative HI of 0.20, which were all less than 1 indicating low health risk. The relative contributions to HIs were 5%, 35%, 2%, 45%, 0.1% and 15% for Hg, Cd, As, Pb, Cr and Cu in corn. The THQs of Hg, Cd, As, Pb, Cr and Cu for rice consumption were Pb (1.02) > Cd (0.91) > Cu (0.29) > As (0.21) > Hg (0.18) > Cr (0.0002). The accumulative HI of 6 heavy metals was 2.61, which was beyond 1, indicating higher risk in rice than in corn.

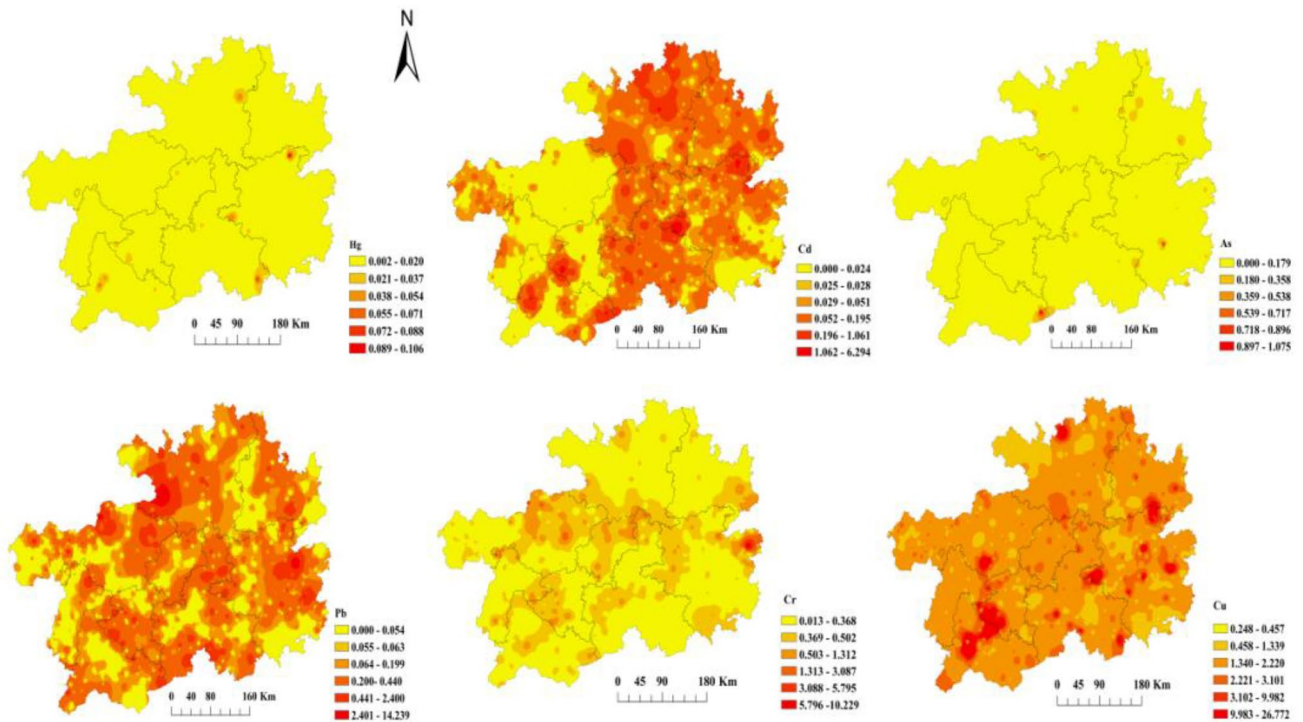


Fig. 3 The geographical spatial distributions of the Hg, Cd, As, Pb, Cr and Cu in corn and rice from Guizhou province. (drawn by software of ArcGIS 10.6)

Table 2 EDI and THQ of heavy metals from corn and rice

Element	Crops	Estimated daily intake		The calculated target hazard quotients	
		mg/kg/ d		(THQ)	
		Mean	P95	Mean	P95
Hg	Corn	0.003	0.010	0.011	0.032
	Rice	0.054	0.121	0.179	0.402
Cd	Corn	0.034	0.115	0.068	0.229
	Rice	0.469	2.058	0.911	4.114
As	Corn	0.013	0.044	0.004	0.015
	Rice	0.632	1.374	0.211	0.458
Pb	Corn	0.142	0.443	0.094	0.295
	Rice	1.511	7.460	1.018	4.961
Cr	Corn	0.322	0.981	0.000	0.001
	Rice	3.016	8.458	0.002	0.006
Cu	Corn	1.017	1.611	0.025	0.040
	Rice	10.726	19.226	0.289	0.518

The relative contributions were 6.9%, 34.9%, 8.1%, 39.1%, 0.01% and 11.1% for Hg, Cd, As, Pb, Cr and Cu in rice. The average and P95 levels of heavy metals were used to present the common and high exposure levels, respectively. The average and high consumption (THQs) of Pb for rice

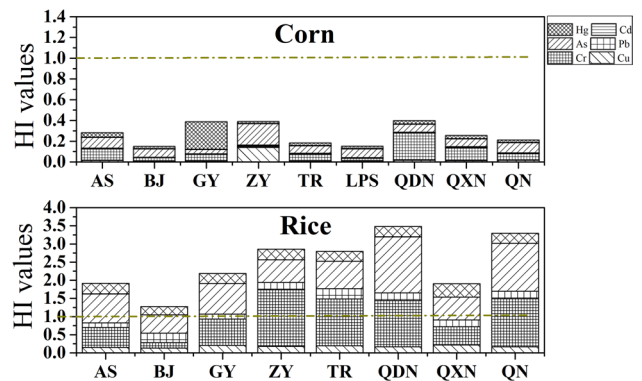


Fig. 4 The calculated target hazard quotients (THQs) of Hg, Cd, As, Pb, Cr and Cu and accumulative HI values for adults from corn and rice consumption. The dark yellow dotted line indicated the standard value of THQ and HI (1.0) corresponding to each element from different regions

were 1.02 and 4.96, respectively, followed by Cd, which were 0.91 to 4.11. The relative high values of THQs for Cd and Pb from rice indicated that rice intake may lead to high adverse health risk. The HI value in rice other than in corn for 6 heavy metals was beyond 1 which may pose a potential risk and Pb was the major component attributed to potential health risk, followed by Cd. Moreover, HI values for the corn were ranked as QDN (0.38) > GY (0.37) > As

(0.27) > ZY (0.25) > QXN (0.24) > QN (0.20) > TR (0.17) > BJ = LPS (0.14), and for rice displayed in the following orders: QDN (3.48) > QN (3.30) > ZY (2.85) > TR (2.80) > GY (2.19) > As (1.92) > QXN (1.90) > BJ (1.28). The high HIs (> 1) in rice indicated the high health risk, and the greatest risk of 6 heavy metals from corn and rice were detected at QDN area.

Discussion

Heavy Metals in Corn and Rice

Based on our obtained results, the average concentrations of Hg, Cd, As and Cu in corn and rice were all below the acceptable levels (0.01 mg/kg for Hg, 0.07 mg/kg for Cd, 0.06 mg/kg for As and 1.86 mg/kg for Cu) except Pb and Cr which were higher than their permissible levels (0.2 and 0.5 mg/kg) (GB 2762-2017) (CHNC 2017). Moreover, the concentrations of heavy metals in rice and corn samples followed the orders: Hg < As < Cd < Pb < Cr < Cu. The concentrations of Cu in corn and rice samples were relatively high but did not exceed the limit (10 mg/kg), while Cd, Cr and Pb were exceeded the limit which may pose risks to human health (Zheng et al. 2020). Cao et al (2020) reported that the mean concentrations of As, Cd, Cr and Pb in corn were 0.04, 0.04, 0.30 and 0.06 mg/kg, respectively, indicated that the mean concentrations of As, Cd and Cr in our current work were higher than this study except Pb. In previous study, the maximum values of Pb and Cd in rice were reported as 0.72 and 1.32 mg/kg, which were 10 and 5 times lower than our study (Lei et al. 2015). Compared to the heavy metal concentrations in crops that collected from South and Northeast China, relatively high concentrations were detected in our study. This might due to the maldistributions of heavy metals in environment. Study had been reported that the mean heavy metal background in Guizhou province is higher than the average value of China (Kong et al. 2018b). This might be the reason that higher concentrations of heavy metal in plant were observed at Guizhou province than other places.

The results of Sharafi et al. (2019) and Pirsaeheb et al. (2020) reported that relatively high concentrations of essential metals (Cu and Cr) were observed in rice and corn compared with toxic metals (As, Pb, Cd, Hg). However, our study revealed that the concentrations of Cr and Cu were higher in corn than in rice while Cd and Pb were higher in rice than in corn. Similar findings were reported in Iran and Japan (Akiko et al. 2020; Mohajer et al. 2019). Besides, research had shown that the concentration of Cd in corn and rice at karst area were 0.004–0.26 (mean 0.04 mg/kg) and 0.003–0.48 mg/kg (mean 0.07 mg/kg), respectively. The average concentration of Cd in rice was higher than in corn ($p < 0.05$), which was consistent with our paper (Wang

et al. 2021). Contrast to our result, the highest concentration of Pb (329.0 ± 383.0 µg/kg) and Cr (35.7 ± 25.0 µg/kg) were detected in corn and the highest concentration of As (1.4 ± 2.3 µg/kg) and Hg (4.3 ± 0.1 µg/kg) were observed in rice in Iran (Pirsaeheb et al. 2020). These results indicated that although corn and rice both belonged to gramineous plants, the heavy metal absorption and accumulation abilities are significantly different due to their biological characteristics and growing environment. First, the heavy metal enrichment capacities of edible part for rice and corn were different. Moreover, enrichment capacities for one crop to different heavy metal were different. For example, the adsorb abilities to Cu and Cr were higher than to Pb and Cd for rice and corn. Besides, the interactions of heavy metals, the existence form, the soil pH, organic content, and cation exchange capacity were also the important reasons that influence the accumulation of heavy metal in crops.

The sources of heavy metals in different regions are diverse. Anthropogenic activities, mining, sewage irrigation, and fertilization could cause heavy metals accumulation and pollution in soil (Atafar et al. 2010). Soil forms including carbonate bound, iron manganese oxide, organic bound, residual state, etc., and geological background are the two main heavy metal sources in corn and rice at karst areas of Guizhou province. Heavy metals in soil are absorbed by crops through the process of release, migration and membrane permeation. The exchangeable heavy metals could be attached to soil colloid, humus and other components through non-specific adsorption and then utilized by plants (Arias et al. 2002). The proportions of exchangeable forms of Pb, Cu and Hg in Guizhou soil ranged from 0.7% to 2.3% while the proportion of Cd is relative high which was 12.5% (Chu et al. 2007). Moreover, the background levels of Hg, Cd, As, Pb, Cr and Cu in Guizhou carbonate rocks soil were 0.11, 0.66, 20.00, 35.20, 95.90 and 32.00 mg/kg which were significantly higher than the average background levels in China with Hg, Cd, As, Pb, Cr and Cu of 0.07, 0.10, 11.2, 26.00, 61.00 and 27.10 mg/kg, respectively (Kong et al. 2018b; Qu et al. 2018). In addition, previous studies (Jia et al. 2020; Tu et al. 2011) have shown that soils formed by carbonate rocks has a high content of soil organic matter (SOM), and the adsorption capacity of SOM on heavy metals can be further improved by the complexation and integration with organic functional groups. Zhang et al. (2014) reported that the organic bound forms ratios of Pb and Cd in Guizhou soil were 26.1% and 15.2%, respectively. In the Fe–Mn oxidation state clay soil, the ratios of Pb and Cu were 18.4% and 3.5%, respectively. Those might be the reason that high concentrations of Pb and Cd were detected in rice and corn samples. Furthermore, soil in Guizhou is mainly acidic acid (pH = 3.90–6.29), and the active state is relatively high which increased the chances

of their accumulation in plant. Therefore, the high natural background levels of Pb and Cd in carbonate and limestone soil leads to the high levels of Pb and Cd in plants at 9 sampling locations. Besides, atmospheric deposition induced by coal combustion and traffic activities are the main sources of Pb and Hg. In general, excluding human activities, geological background played a dominant role on the distributions of heavy metals in crops.

Distributions of Heavy Metals in Rice and Corn Collected From Different Sampling Locations

The parent materials of soil formation (sand shale, limestone, diluvial and alluvial), the production of electronics, home appliances and lighting appliances and the exploitation of manganese ore and silicon ore might be led to the high concentrations of Hg, Cd and Cu in rice and corn samples at QDN samples. Affected by limestone soil of Liping County at QDN area, the concentrations of Cr, Cd, As and Hg were relatively high with the values of 97.07, 0.37, 8.50 and 0.23 mg/kg, respectively (Chen et al. 2019). QDN area owns a lot of electrical appliances manufacture industries. The discharge of these electronic waste induced the high concentrations levels of Cd, Cr, Pb, Hg, As and Cu in soil which might be related to the relatively high concentrations of Hg, Cd, Cu in our paper. Moreover, research reported that the concentration of Hg in rice was mainly distributed in TR with the range of ND ~ 0.023 mg/kg while in our study Hg was mainly distributed in QDN (0.003–0.06 mg/kg) (Yang et al 2015). The human activities such as zinc-iron mining and smelting in BJ area may induce the high concentrations of heavy metal in soil and crops. Previous report showed the average concentrations of Cr, Pb and Cd in soil of Hezhang smelting region at BJ area were 157.50, 48.71 and 1.62 mg/kg, respectively, indicating high background heavy metals in soil. This might be the reasons that the highest concentration of Cr was detected in BJ area in our study (Briki et al. 2015). The concentrations of Pb in rice and corn from ZY area were ranged at 2.31–2.46 mg/kg which were higher than their permissible level in accordance with the highest concentration of Pb detected in ZY area in this paper (Zhang et al. 2021). Pb particles in air are easily settled down and accumulated in surface soil with the insoluble forms of Pb(OH)₂, Pb₃(SO₄)₂, Pb(CO₃)₃, which caused the increasing of Pb in crops (Gu et al 2018). Furthermore, due to the mining and utilization of arsenic coal in QXN (Xingren County) area, a large amount of acid wastewater was generated and used as irrigation water. This may induce a relative high acidity in soil (pH=5) and activation of heavy metal states (Tao et al. 2017). It might be

the reason that high concentration of Cr was detected in crops of QXN area.

Health Risk Assessment

The average THQ values of heavy metals in rice were higher than in corn with exception of Cr. Similarly, other report found that the mean THQ values of Cd, Pb and Cu for rice were 1.39, 5.27 and 0.59 which were higher than for corn with the values of 0.11, 0.56 and 0.05 (Zhang et al. 2021). Yu et al. (2017) reported Cd and Cu were key components in corn contributing to the potential health risk with the relative contributions of 13% and 30%. Huang et al. (2013) showed that the health risks in rice for local citizens were primarily due to As and Pb which contributed 44% and 33% of THQ, respectively. In this study, Pb and Cd were the major contributors to THQ in corn and rice samples with the relative contributions ranged at 35%–45% which were higher than the results of Yu. et al. and Huang et al. Furthermore, the average HIs of heavy metals in corn (0.20) was lower than 1, while in rice (2.61) was beyond 1 indicating that heavy metals in rice posed a higher risk than in corn. It may be caused by two factors: one was the amount of daily intake of crops. The average daily intake of rice (420 g/d) is 12 times higher than corn. Another reason was that the higher heavy metal concentrations were detected in rice than in corn. A research associated with rice reported the mean concentrations of Cd, As and Pb were 0.34, 2.33 and 0.04 mg/kg, respectively, with the THQ levels of 0.34, 2.33 and 0.22 (Mao et al. 2019). In Tian's study (Tian et al. 2020), the concentrations of Cu, Pb and Cd in rice ranged at 2.36–5.64 (mean 4.08), 0.11–0.50 (mean 0.19) and 0.10–1.70 (mean 0.93) mg/kg, respectively, and the THQ values of Cu, Pb and Cd in rice were 0.49, 0.11 and 1.79. Those results indicated that the concentrations of Cd and Pb were above the standard limit (0.2 mg/kg for Cd, 0.5 mg/kg for Pb) which was associated with higher health risk. In this study, 26.89% for Pb and 23.17% for Cd in rice and corn samples exceeded the MAC values and Pb and Cd were the major components contributing to HIs. The mean THQ values of Pb and Cd in corn and rice were 1.02 and 0.91, respectively, which were inconsistent with the findings of Adel et al. (2020) and Kong et al. (2018a, b) who demonstrated that the THQ values of Pb and Cd in corn and rice were < 1. The relatively high health risk of Pb and Cd in our study might be due to the high concentrations that detected in rice and corn samples collected from Guizhou province. Expect the difference enrichment capabilities for different crops, the high background of Pb and Cd in environment might be the main reason that causing high health risk for corn and rice samples. In addition, the highest HI values of corn and rice were detected in QDN area which was consistent with the high concentrations of heavy metals in this area. The finding of this study highlighted the human health risk

of Pb and Cd in corn and rice and more attention should be forced on the health risk of 6 heavy metals from QDN area given its high detected risk.

The concentrations of Hg, Cd, As, Pb, Cr and Cu in 398 rice and 638 corn samples were detected in this study. The percentages of Hg, Cd, As, Pb, Cr and Cu in corn and rice samples that higher than their permissible level were 14.14%, 23.17%, 0.26%, 26.89%, 32.03% and 1.16%, respectively. Attention should be paid to high concentrations of Hg, Cd and Cu which were detected in the QDN sampling area. The health risks of 6 heavy metals in rice were higher than in corn, and Pb was the major component attributed to the potential health risk, followed by Cd. Based on our study, further specific and detailed exposure estimates need to be performed in targeted area. In addition, the regular monitoring of heavy metals in corn and rice is still necessary in this area to ensure the dietary safety.

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Declarations

Conflict of interest The authors declare no conflict of interest.

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