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Contamination of Heavy Metal(Loid)S in Cereals, Vegetables, and Legumes Purchased from Local Markets of Jiaozuo, China and The Associated Health Risk Assessment

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ABSTRACT

The health implications associated with the consumption of food contaminated by heavy metals (HMs) and metalloids, specifically arsenic (As), remain a global concern. This study aimed to investigate the occurrence of HMs and As in various food categories, including cereals (rice, wheat, and corn), vegetables (leafy greens, edible mushrooms, kale, solanaceous fruits, and tubers), and legumes (peanuts, soybeans, and mung beans), commercially available in Jiaozuo city. Additionally, we sought to assess the potential health risks posed to human consumers by the ingestion of these three food categories. To evaluate the extent of contamination, both single-factor analysis and Nemerow's synthetical pollution index were employed. The assessment of health risks was conducted using the health risk model recommended by the United States Environmental Protection Agency. Specifically, cereals exhibited exceedance rates of 2.60% for Cr, 44.16% for Hg, 3.90% for Ni and 3.90% for Pb. Notably, vegetable samples exhibited exceedance rates of 0.90% for Hg and 17.12% for Zn. Among legume samples, exceedance rates were 16.98% for Cr, 75.47% for Ni and 13.21% for Pb, while other HMs and As remained within standard limits. The cumulative contamination level of HMs in each diet category ranked in the order of legumes > cereals > vegetables. Based on the daily intake of cereals, legumes and vegetables, the Total Target Hazard Quotient for HMs in both children and adults exceeded a value of 1, indicating the potential noncarcinogenic risks (non-CRs) associated with consumption of these three dietary categories. Conversely, the CR induced by HMs through the consumption of these foods remained within acceptable levels for both children (8.92×10^{-6}) and adults (2.65×10^{-6}). It is noteworthy that the health risk posed by HMs through food consumption was more pronounced in children than in adults.

KEYWORDS

Heavy metals; Arsenic; Cereals, Vegetables, Legumes; Health risk assessment.

1. INTRODUCTION

Recently, in light of population growth and economic development, environmental pollutants such as heavy metals (HMs), polycyclic aromatic hydrocarbons, and polychlorinated biphenyls have become pervasive. These substances exhibit long-term persistence in the environment due to the inherent stability of HMs and their resistance to degradation (Pei et al. 2020). The accumulation of HMs in living organisms through contaminated environments and the associated escalation in the risk of contaminating food sources have garnered significant attention among researchers (Naseri et al. 2015; Shaheen et al. 2016). The consumption of crops contaminated with HMs is responsible for more than 4.2 million fatalities worldwide annually and afflicts approximately 600 million individuals with diseases (Zheng et al. 2020). Notably, Cd, Cr, Cu, Hg, Ni, Pb, Zn, and arsenic (As, a metalloid) are designated priority control pollutants by the United States Environmental Protection Agency (USEPA)(USEPA 2014).

Among these toxic HMs, Cd, Pb, Cr, Hg, Ni, and As exhibit the capacity to induce various illnesses even at low exposure levels and can inflict harm upon the human body following prolonged exposure. For instance, As exposure may result in skin lesions or skin cancer, whereas protracted Cd intake can lead to conditions such as bone fractures, renal insufficiency, high blood pressure, and lung cancer (Liu et al. 2013; Jia et al. 2018; Bi et al. 2018). Elevated levels of Pb can adversely affect brain function in children, while chronic Hg exposure can impair the central nervous system (Huang et al. 2014, 2018). Ni can trigger contact dermatitis, and direct ingestion may provoke vomiting and diarrhea (Sall et al. 2020). Cu and Zn are essential metals for human physiological processes, serving as both catalytic and structural components of enzymes and proteins. However, when their concentrations exceed a certain threshold, they can become detrimental to human health (Chen et al. 2018). In addition to disrupting the body's endocrine system, Cu and Zn can also perturb other bodily functions, including respiration and the central nervous system (Doabi et al. 2018). The International Agency for Research on Cancer has classified As, Cr, and Cd as carcinogenic HMs, thereby indicating their potential to induce cancer in humans upon prolonged exposure (Zakaria et al. 2021).

Presently, numerous research endeavors focus on assessing the health risks associated with the dietary intake of metals. For instance, Pirsaheb et al. (2021) conducted a study on the human health risk assessment of HMs in grains in Kermanshah. Li et al. (2018b) investigated the exposure risks to HMs from edible vegetables in proximity to smelting facilities in central China. Rasheed et al. (2018) explored As exposure and its associated health effects in wheat, both in its raw and cooked forms. However, a substantial portion of these dietary studies tend to employ homogenous diets, which may result in an underestimation of HM health risks in food and, consequently, an incomplete comprehension of dietary exposures.

In our country, dietary patterns are characterized by an Oriental diet structure, primarily centered around the consumption of plant-based foods such as cereals and vegetables, complemented by animal products such as meat, eggs, and dairy (Yan 2014). Staple crops such as corn, wheat, and soybeans constitute the foundation of dietary habits in many regions and are extensively used in various food preparations (Corguinha et al. 2015). In accordance with the China Food and Nutrition Development Program (2014-2020), cereals, vegetables, and legumes collectively account for over 60% of the total dietary intake of individuals (General office the State Council 2014). Consequently, HMs ingested through these three fundamental food categories, namely, cereals, vegetables, and legumes, which are integral components of our daily sustenance, may entail heightened health risks for the human population.

This study was conducted in the region of Jiaozuo to examine the levels of HMs and As in commercially available cereal, vegetable, and legume samples. The objective was to assess the extent of HM contamination across different food categories. To evaluate the associated health risks of these three dietary components for both adults and children in the region, we employed the health risk assessment model recommended by the USEPA. The findings of this study aim to furnish a scientific foundation for comprehensive investigations into HM contamination in food and the development of strategies for its prevention and control within the Jiaozuo area.

2. MATERIAL AND METHODS

2.1. Study area

Jiaozuo City is situated in central China and falls within the administrative jurisdiction of Henan Province as a prefecture-level city. It is strategically positioned, nestled against the northern foothills of the Taihang Mountains, and shares its border with Jincheng City in Shanxi Province. This location makes it a pivotal regional center at the crossroads of Henan and Shanxi.

The topography of Jiaozuo city is characterized by a combination of mountainous and plain landscapes. The terrain gradually ascends from the southeast to the northwest, resulting in a wideranging elevation that spans from 80 to 1200 meters, contributing to significant elevation differentials within the region. The city covers a total area of 4,071 square kilometers and is administratively divided into 4 districts, 4 counties, and 2 county-level cities.

2.2. Sample collection

Sampling locations for this study were strategically chosen within the Shanyang and Jiefang districts of Jiaozuo City, since those two districts contain majority of the city population. These locations encompassed six supermarkets, six farmers markets, and one wholesale market. In total, 244 samples were meticulously collected. The sample collection comprised 78 samples of cereals, encompassing rice, corn, and wheat flour; 52 samples of legumes, including soybeans, mung beans, and peanuts; and 114 samples of vegetables, encompassing leafy greens, solanaceous fruits, tubers, edible mushrooms, and kale. Detailed information regarding the specific types and quantities of samples can be found in Table 1.

Table 1. Type and number of samples collected at each sampling point

Sampling point		Cereals			Legumes			Vegetables				
		Rice	Flour	Corn	Soybean	Peanut	Mung bean	Leaf greens	Tuber	Edible mushrooms	Solanaceous melons	Kale
	Xinhua Supermarket	3	2	1	1	1	1	2	1	1	1	1
	Dazhang Supermarket	3	2	1	2	2	1	2	2	2	2	1
	Yonghui Supermarket	2	1	1	1	1	1	1	1	1	1	1
Jiefang District	Wangfujing Supermarket	1	1	1	1	1	1	1	1	1	1	1
	Zhongzhou market	4	3	1	3	3	1	4	2	2	3	1
	Little Snail Supermarket	2	1	—	1	1	—	2	1	1	1	1
	Moon Season Farmers' market	6	3	1	3	3	1	5	2	2	3	1
	Jiaobei Marketplace	3	2	1	2	2	1	3	2	2	2	1
	Yutong Agricultural Trade	5	4	1	2	2	1	4	2	2	3	1
	Lubao Wholesale	2	2	—	1	1	—	4	2	2	2	1
Shan yang District	Pak Tai Ka Yuen	1	1	1	1	1	1	1	1	1	1	1
	Tanan farmers markets	3	2	1	2	2	1	3	2	2	2	1
	Baida supermarket	4	2	1	1	1	—	3	2	2	2	1
	Total	39	27	12	21	21	10	35	21	21	24	13

2.3. Sample preparation

In the initial step of our sample preparation, the vegetable samples underwent a meticulous cleaning process with deionized water. Subsequently, they were subjected to drying in an oven at 70°C for a duration of 30 minutes, followed by further drying at 105°C until a constant weight was achieved. For the cereal and bean samples, a similar cleaning process with deionized water was employed, followed by drying at 105°C to a constant weight. The dried samples were subjected to grinding and then passed through a 100-mesh sieve. The processed samples were stored in securely sealed plastic bags until testing.

To continue the procedure, a precise amount of 300 mg (with an accuracy of 0.001 g) of each sample was carefully weighed into a 50 mL PTFE digestion jar. Following this, 8 mL of concentrated HNO_3 was added for predissolution. Subsequently, 2 mL of H_2O_2 was introduced, and the jar was sealed with a lid. The sealed jars were then placed within a microwave digestion instrument for digestion. The temperature within the instrument was incrementally raised to 150°C within a span of 10 minutes, after which it was further elevated to 180°C and maintained at this temperature for a duration of 20 minutes.

2.4. Determination of HMs in food

The analysis of Hg content in the samples was conducted using a Direct Mercury Analyzer (DMA-80, Milestone, Italy). Meanwhile, the determination of Pb, Cd, Cr, As, Ni, Cu, and Zn in the samples was carried out utilizing a inductively coupled plasma mass spectrometer (Varian 810-MS, Jena, Germany). The details of operating parameters are outlined in Table 2. It is noteworthy that the quantification of HMs and As in the samples was performed on a fresh weight basis.

Table 2. Instrument operating parameters

	Items	Parameter
ICP-MS	Power	1450w
	Plasma flow	18.0L/min
	Auxiliary gas flow	1.80L/min
	Nebulizer flow rate	0.85L/min
	Sheath gas flow	0.85L/min
	Peristaltic pump speed	2rpm
	Sampling depth	6.5mm
	Sampling cone aperture	1.0mm
	Interception cone bore diameter	0.4mm
	Analysis mode	Mass analysis
	Internal standard	72Ge

2.5. Quality control

To ensure the precision and accuracy of our analyses, each group of 24 samples was accompanied by two parallel samples: one constituted a standard reference material (SRM) sample (Hunan rice standard material, GBW10045a), and the other containing deionized water served as a blank sample. The results indicated that the response values of the blank samples fell below the detection limit. Moreover, the relative standard deviations of the parallel samples remained within the range of ±20%. Importantly, the determined values of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in the SRM all conformed to the prescribed error limits. Detailed outcomes of the standard substance measurements are provided in Table 3.

Table 3. Standard substance measurement results

Element	Standard reference value	Measured value						Mean ± standard deviation
		1	2	3	4	5	6	
As(mg/kg)	0.12±0.02	0.1	0.1	0.11	0.1	0.11	0.1	0.1±0.01
Cd(mg/kg)	0.32	0.35	0.35	0.34	0.31	0.3	0.32	0.32±0.02
Cr(mg/kg)	0.08	0.06	0.11	0.09	0.07	0.09	0.08	0.08±0.02
Cu(mg/kg)	2.4±0.2	2.32	2.22	2.20	2.09	2.03	2.51	2.23±0.16
Hg(μg/kg)	3.5±0.8	2.92	3.1	2.77	3.2	2.79	2.85	2.94±0.16
Ni(mg/kg)	0.27±0.03	0.24	0.31	0.23	0.26	0.19	0.22	0.244±0.04
Pb(mg/kg)	0.08±0.02	0.09	0.09	0.05	0.04	0.04	0.07	0.06±0.02
Zn(mg/kg)	12.4±1.2	11.1	10.9	10.4	10.0	10.3	11.1	10.63±0.42

Table 4. Standard Food Hygiene Limits for HMs (loid) in Diet

Element	Limit	Diet type	Sources of standards
Hg	≤0.02	Cereals	(National Health Commission of the People's Republic of China, State Administration for Market Regulation. 2022)
	≤0.01	Fresh vegetables	
	≤0.1	Edible mushrooms and their products	
Pb	≤0.2	Cereals	
	≤0.1	Vegetables (except Brassica vegetables, leafy vegetables, leguminous vegetables and potatoes)	
	≤0.3	Brassica vegetables, leafy vegetables	
	≤0.2	yams	
	≤0.5	Edible mushrooms and their products (except products of oyster mushrooms, mushrooms and their products)	
	≤0.3	oyster mushroom, mushroom and their products	
	≤0.2	Legumes	
As	≤0.5	Vegetables	
	≤0.5	Edible mushrooms and their products	
	≤0.5	Cereals (except paddy)	
	≤0.2	rice	
Cd	≤0.1	Cereals (except paddy)	
	≤0.2	Paddy, rice	
	≤0.05	Fresh vegetables	
	≤0.2	Leafy vegetables	
	≤0.1	Root, tuber and stem vegetables	
	≤0.2	Fresh Edible Mushrooms	
	≤0.5	Mushroom	
	≤0.2	Legumes	
	≤0.5	Peanut	
Cr	≤1.0	Cereals	
	≤0.5	Fresh vegetables	
	≤1.0	Legumes	
Cu	≤10	Cereals and products	(Ministry of Agriculture of the People's Republic of China. 2005)
	≤20	Beans and products	
	≤20	Vegetables	(FAO/WHO)
Zn	≤50	Cereals and products	(Ministry of Agriculture of the People 's Republic of China. 2005)
	≤100	Beans and products	
	≤1.0	Vegetables	(FAO/WHO.2001)
Ni	≤0.4	Cereals	(Yang et al. 2018)
	≤3.0	Legumes	
	≤0.3	Vegetables	

2.6. Methods used to evaluate the contamination levels and health risks of HMs

2.6.1. Assessment of HMs contamination in food

To assess the degree of HM contamination in the samples, we employed both single-factor analysis and Nemerow's synthetic pollution index (Huang et al. 2018). The single-factor contamination index (P_i) was calculated using equation (1) while the Nemerow's synthetic pollution index (P) was calculated using equation (2):

$$P_i = \frac{C_i}{S_i} \quad (1)$$

$$P = \sqrt{\frac{P_{ave}^2 + P_{imax}^2}{2}} \quad (2)$$

where C_i and S_i represent the content of a individual HM in the food and the standard limit set for the corresponding food type, respectively. The standard limits for HMs in the diet can be found in Table 4. P_{ave} denotes the mean of P_i for all HMs, and P_{imax} represents the maximum P_i. The extent of HM contamination can be classified into 5 distinct categories, as outlined in Table 5.

Table 5. Classification of heavy metal pollution levels(Huang et al. 2018)

Grade	Single factor and Nemerow' synthetical pollution index	Level of contamination
1	P≤0.7	safety
2	0.7<P≤1	Precaution
3	1<P≤2	Slightly polluted
4	2<P≤3	Moderately polluted
5	P>3	Severely polluted

2.6.2. Health risk assessment

In this paper, we utilized the health risk model introduced by the USEPA (FAO/WHO, 2001). This model encompasses two key components: the Target Hazard Quotient (THQ) and the Target Carcinogenic Risk (TCR), which collectively serve as tools for evaluating human health risks.

(1) Assessment of noncarcinogenic risk

The THQ is employed to evaluate the non-carcinogenic risk (non-CR) associated with the ingestion of a individual HM through food. The Total Target Hazard Quotient (TTHQ) provides insight into the cumulative impact of various HMs. An acceptable risk is indicated when the TTHQ is less than 1, while a TTHQ greater than 1 suggests a potential health risk. It is important to note that the larger the value of TTHQ is, the more significant the risk posed by contaminants to human health. The formula for calculating TTHQ is as follows:

$$THQ = \frac{EDI}{RfD} = \frac{ED \times EF \times IR \times C}{BW \times AT_n \times RfD} \quad (3)$$

$$TTHQ = \sum THQ \tag{4}$$

where EF is the exposure frequency of food, ED is the exposure duration of food, IR is the amount of daily food intake, C is the HM concentration in food, BW is the body weight of the exposed individual, AT_n is the duration of noncarcinogenic effects, and RfD is the corresponding reference dose.

(2) Carcinogenic risk assessment

The carcinogenic risk (CR) is utilized to assess the lifetime risk of cancer within the population resulting from the consumption of a individual HM through food. Based on USEPA standards, $CR < 1\times10^{-4}$ is acceptable, and $CR > 1\times10^{-4}$ indicates a lifetime cancer risk. The TCR is employed to depict the cumulative impact of multiple metals. CR and TCR is calculated using the following equation:

$$CR = \frac{C\times IR\times CSF\times ED}{BW\times AT_C} \tag{5}$$

$$TCR = \sum CR \tag{6}$$

where CSF is the cancer slope factor and AT_C is the duration of carcinogenic effects.

The values of parameters used in the above formula are listed in Table 6.

3. RESULTS AND DISCUSSION

3.1. HMs in food

The levels of HMs in food samples, reported on a fresh weight basis, are depicted in Figure 1. Specifically, the ranges of Cr, As, Cd, Pb, Hg, Ni, Cu, and Zn in cereals were as follows: not detected-1.500 mg/kg, 0.01-0.170 mg/kg, 0.010-0.040 mg/kg, 0.010-1.15 mg/kg, 0.002-0.444 mg/kg, not detected-0.700 mg/kg, 0.180-6.990 mg/kg, and 0.460-15.500 mg/kg, respectively. Meanwhile, in vegetables, the ranges were 0.003-0.278 mg/kg for Cr, 0.107 mg/kg for As, 0.104 mg/kg for Cd, 0.430 mg/kg for Pb, 0.014 mg/kg for Hg, 0.086 mg/kg for Ni, 0.002-0.599 mg/kg for Cu, and 3.991 mg/kg for Zn. For legumes, the ranges were 0.007-2.600 mg/kg for Cr, 0.001-0.003 mg/kg for As, 0.01-0.430 mg/kg for Cd, 0.010-0.380 mg/kg for Pb, 0.002-0.072 mg/kg for Hg, 0.500-20.100 mg/kg for Ni, 6.860-12.400 mg/kg for Cu, and 20.100-33.100 mg/kg for Zn.

Substantial variations were observed in the HM levels among different food categories, with legumes and cereals demonstrating higher HM levels compared to vegetables as a whole. Importantly, the disparity between the highest and lowest HM levels in various food categories spanned three orders of magnitude, underscoring the variability in the bio-accumulation of HMs by different crop types.

Table 6. Health risk assessment parameters

Index	Reference value		References
	Adult	Children	
BW(kg)	56.8	15.9	(Su et al. 2023)
ED(a)	30	10	
EF($d\cdot a^{-1}$)	365	365	
ATn (d)	ED*365	ED*365	
ATc(d)	365×70	365×70	
$IR_{vegetable}$($g\cdot d^{-1}$)	400	100	7([CSL STYLE ERROR: reference with no printed form.])
IR_{cereal}($g\cdot d^{-1}$)	300	55	
IR_{legume}($g\cdot d^{-1}$)	30	25	
RfD_{Pb}[mg(kg·d)$^{-1}$]	4.00E-03		(US EPA 2015) (Xiao et al. 2017)
RfD_{Cr}[mg(kg·d)$^{-1}$]	3.00E-03		
RfD_{Cd}[mg(kg·d)$^{-1}$]	1.00E-03		
RfD_{As}[mg(kg·d)$^{-1}$]	5.00E-02		
RfD_{Ni} [mg(kg·d)$^{-1}$]	2.00E-02		
RfD_{Zn}[mg(kg·d)$^{-1}$]	3.00E-01		
RfD_{Cu}[mg(kg·d)$^{-1}$]	4.00E-02		
RfD_{Hg}[mg(kg·d)$^{-1}$]	3.00E-04		
CSF_{Cd}[mg(kg·d)$^{-1}$]	6.1		
CSF_{As}[mg(kg·d)$^{-1}$]	1.5		
CSF_{Cr}[mg(kg·d)$^{-1}$]	5.00E-01		
CSF_{Pb}[mg(kg·d)$^{-1}$]	8.50E-03		
CSF_{Ni}[mg(kg·d)$^{-1}$]	8.40E-03		

The average contents of Zn, Cu, Cr, Ni, As, Cd, Pb, and Hg in cereals were 3.789 mg/kg, 1.087 mg/kg, 0.210 mg/kg, 0.126 mg/kg, 0.043 mg/kg, 0.011 mg/kg, 0.124 mg/kg, and 0.044 mg/kg, respectively (Table 7). In accordance with the standardized limits (Table 4), the exceedance rates for Cr, Pb, Hg and Ni in cereal samples were 2.60%, 3.90%, 44.16% and 3.90%, respectively. However, the contents of As, Cd, Cu and Zn remained within the specified standard range, indicating their safety. This suggests that Hg contamination has occurred in these cereals, posing potential health risks to consumers. Furthermore, varying crop capacities to absorb HMs may contribute to higher HM contamination in cereals compared to other crops. Notably, other studies have also reported high concentrations of Hg in cereals, such as a study by Horvat et al. (2003), where the Hg content in rice grains reached up to 569 mg/kg. The pronounced HM contamination observed may arise from several factors, including industrial waste discharge into the soil or naturally elevated HM content in the local area. An example can be found in the district of tungsten-polymetallic deposits in Chenzhou City, Hunan Province, and the lead-zinc deposits in Shaoguan City, Guangdong Province, where soil HM levels are notably high, consequently leading to elevated HM content in rice (Li et al. 2018a; Wei and Cen 2020).

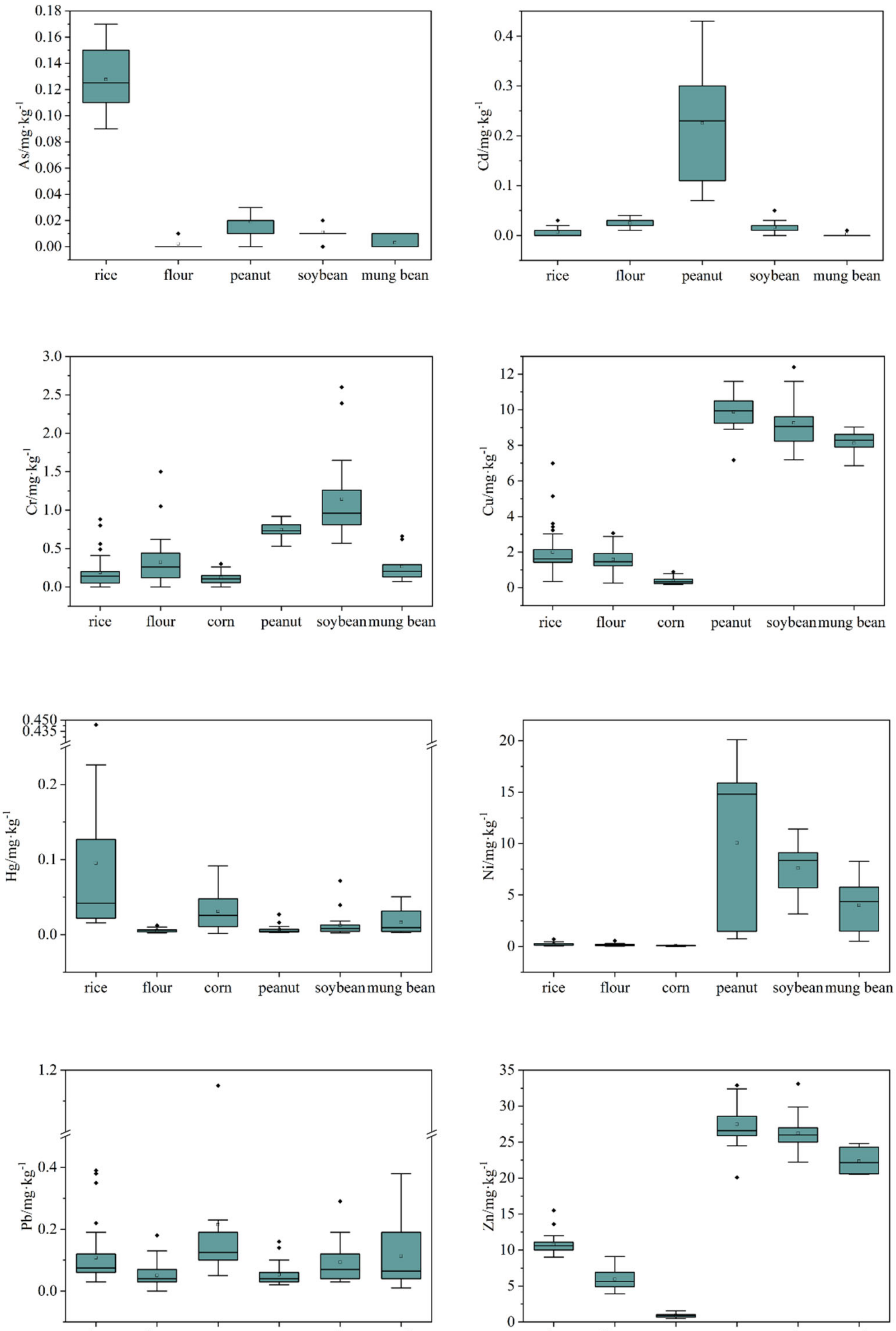
The average contents of Zn, Cu, Cr, Ni, Pb, Cd, As, and Hg in vegetables were 0.589 mg/kg, 0.064 mg/kg, 0.032 mg/kg, 0.016 mg/kg, 0.013 mg/kg, 0.008 mg/kg, 0.006 mg/kg, and 0.004 mg/kg, respectively (Table 7). These findings are generally consistent with the trend observed in HM concentrations in the study by Wang et al. (2012). With the exception of exceedance rates for Hg and Zn in vegetables samples were 0.90% and 17.12%, the levels of HMs and As in the remaining vegetables were below the corresponding standard limits. This indicates that these vegetables are safer overall for consumption within the food chain (Table 4). A comparison of the HM levels in food products in this study with those in other regions is provided in Table 7, revealing that the levels of HMs in vegetables were lower than those in Shanghai and Zhejiang.

Leafy vegetables and edible mushrooms displayed higher HM levels than vegetables such as solanaceous fruits, tubers, and kale. Edible mushrooms has been reported to be cable of accumulating HMs from the surrounding environment (Fu et al. 2020; Mleczek et al. 2021). Leafy vegetables exhibited higher levels of Pb and Hg than edible mushrooms, possibly because Pb and Hg can be absorbed by plants not only from the soil but also from the atmosphere (Bi et al. 2009; Ao 2017). In the case of leafy vegetables, those with larger leaves may be more exposed to the atmosphere and consequently enriched with higher HM levels (Sun et al. 2021).

The average contents of Zn, Cu, Ni, Cr, Pb, Cd, Hg, and As in legumes were measured at 25.988 mg/kg, 9.297 mg/kg, 7.914 mg/kg, 0.717 mg/kg, 0.086 mg/kg, 0.081 mg/kg, 0.012 mg/kg, and 0.011 mg/kg, respectively (Table 7). It is important to note that the contents of Cd, Cu and Zn in legumes met the requirements of food standard, indicating their safety as per established regulations. However, the exceedance rates for Cr, Ni and Pb in legumes were recorded at 16.98%, 75.47%, and 13.21%, respectively. In particular, the average concentration of Cr in soybeans reached 1.143 mg/kg, surpassing the standard by 1.143 times, with an exceedance rate of 42.86%. Since the national food safety standard (GB 2762-2022) does not specify limits for total Hg and As content in legumes, the contamination levels for those two elements were not evaluated in this study.

Table 7. Comparison of HM content in Jiaozuo food with the results of other regions (fresh weight)

Category	Source	Concentrations of HM ($mg \cdot kg^{-1}$)								References
		Cr	As	Cd	Pb	Hg	Ni	Cu	Zn	
Cereals	Jiaozuo	0.210±0.255	0.043±0.065	0.011±0.012	0.124±0.142	0.044±0.088	0.164±0.126	1.608±1.087	7.577±3.789	Present study
	Swat district, Pakistan	BDL-0.03	-	0.08-0.11	-	-	0.23-0.32	0.19-0.25	0.13-0.29	(Khan et al. 2013)
Legumes	Jiaozuo	0.717±0.476	0.011±0.009	0.081±0.128	0.086±0.072	0.086±0.0113	7.914±5.378	7.297±1.280	25.988±3.160	Present study
	Guangxi	0.03-1.05	0.01-0.2	0.01-1.02	0.11-0.85	0.15-24.22×10-3	-	-	-	(Huang et al. 2017)
Vegetables	Jiaozuo	0.032±0.054	0.006±0.006	0.008±0.017	0.0013±0.028	0.004±0.003	0.016±0.024	0.064±0.096	0.589±0.570	Present study
	Shanghai	-	0.035	0.028	0.039	0.002	-	0.67	3.96	(Bi et al. 2018)
	Zhejiang	0.057	0.013	0.017	0.022	0.094	0.002	-	-	(Pan et al. 2016)


As/mg·kg-1
Cd/mg·kg-1
Cr/mg·kg-1
Cu/mg·kg-1
Hg/mg·kg-1
Ni/mg·kg-1
Pb/mg·kg-1
Zn/mg·kg-1
rice
flour
corn
peanut
soybean
mung bean

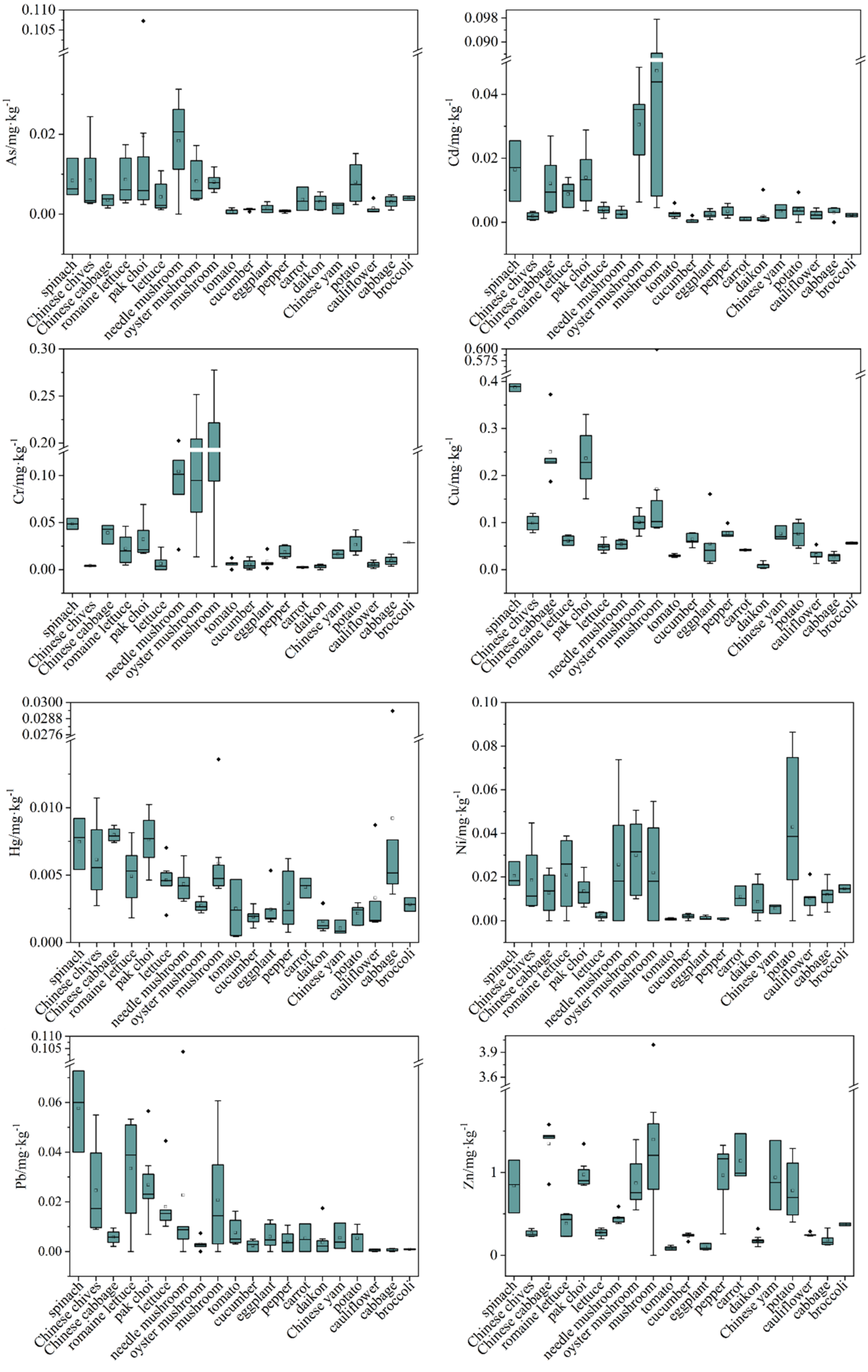


Figure 1. HM content in food

3.2. Evaluation of HM contamination in food

Results of HMs contamination levels assessed by the single-factor pollution index (Figure 2) indicated that As, Cd, Cu, and Zn were within safe limits; Cr in peanuts and Hg in spinach, Chinese cabbage, and pak choi were of precautionary concern; Cr in soybeans, Pb, Hg in corn, and Ni in mung beans exhibited slight pollution; Ni in peanuts reached a moderate pollution level; and both Hg in rice and Ni in peanuts were at a level of severe pollution.

Nemerow's synthetic pollution index provided an overall assessment of contamination levels by various HMs. As shown in Figure 3, corn, soybeans and mung beans were slightly polluted; peanuts were moderately polluted; rice showed severe pollution; and the remaining food products were deemed safe. In summary, the hierarchy of HM contamination in food products can be generalized as follows: legumes > cereals > leafy greens > kale > solanaceous fruits > edible mushrooms >tubers, with cereals and legumes being the primary contributors to contamination levels (Figure 2(a)).

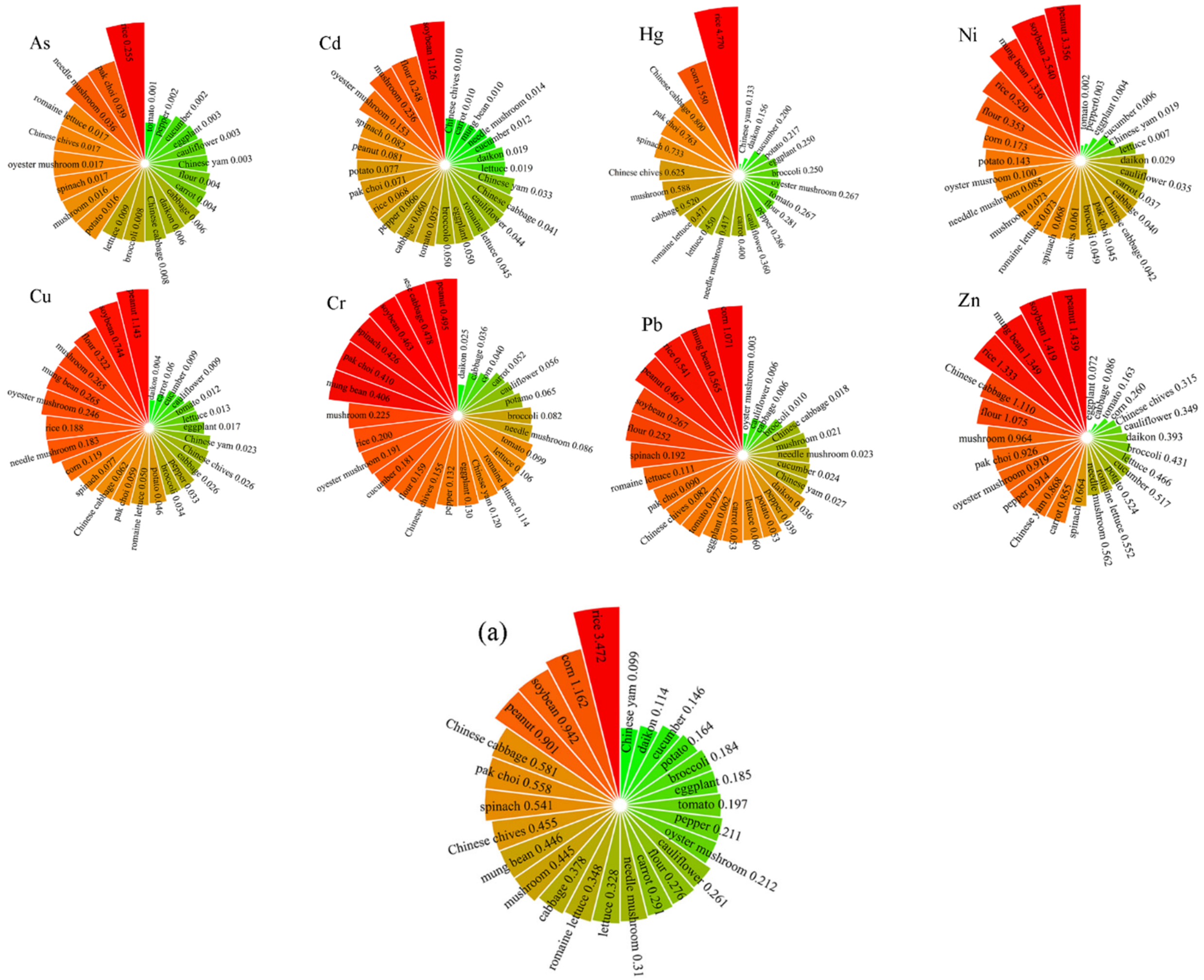


Figure 2. Single factor pollution index and (a) represents Nemerow's synthetical pollution index

3.3. Health risk assessment

3.3.1. Noncarcinogenic risk

As shown in Table 8, the THQ of individual HMs for children remained below 1, except for Hg in rice and corn, where it exceeded the threshold of 1. The health risk associated with exposure to an individual HM remains within acceptable limits. However, it is imperative to acknowledge that several HMs, when considered collectively, can elevate the risk associated with consumption (Zakaria et al. 2021).

As delineated in Figure 3(A) and Table 8, among cereals, rice exhibited the highest TTHQ (5.481), followed by corn (2.060) and wheat (0.853). The TTHQ of rice and corn were higher than 1, suggesting that long-term consumption of rice and corn may cause adverse health effects. Hg in rice exhibited a THQ of 3.983, contributing 72.7% to the TTHQ of rice, surpassing other HMs. Considering the indispensability of cereals in our diet, the heightened levels of HM contamination in rice represent a direct health risk to human consumers. Consequently, it is imperative to emphasize and regulate the presence of HMs, especially Hg, in cereals.

In the realm of vegetables, the hierarchy of TTHQ, in descending order, was as follows: edible mushrooms (0.554, mean value of different varities, the same bellow), leafy vegetables (0.333), kale (0.132), tubers (0.111), solanaceous fruits (0.104) (Figure 3(A) and Table 8). The TTHQ of all vegetables was lower than 1, suggesting that the health effects of prolonged consuming these vegetables were acceptable. Cd in mushrooms showed the highest THQ values among all the THQ of HMs in vegetables.

Likewise, among legumes, peanuts presented the highest TTHQ at 2.035, followed by soybeans (1.741) and mung beans (0.949) (figure 3(A)). Prolonged consumption of peanuts and soybeans elevates health risks beyond acceptable levels. Ni contributed the most to TTHQ of peanuts.

For adults, as listed in the Figure 3(B) and Table 9, the THQ of individual HMs was consistently below 1, except for the Hg in rice, which is 1.673, exceeding the 1 threshold.

In the context of cereals, the descending order of TTHQ values was as follows: rice (2.704), wheat (1.213), and corn (1.117). TTHQ greater than 1 indicates a potential health risk posed by long term consumption of cereals by adults.

Within the domain of vegetables, the hierarchy of TTHQ, in descending order, unfolded as follows: edible mushrooms (0.672, mean value of different varities, the same bellow), leafy greens (0.384), kale (0.153), tubers (0.140), and solanaceous fruits (0.123) (Figure 3(B) and Table 9). The TTHQ of all the vegetables was lower than 1, suggesting that the health effects of consuming these vegetables were acceptable.

In contrast, the TTHQ values of legumes for adults, in descending order, were peanuts (0.667), soybeans (0.617), and mung beans (0.351) (Figure 3(B)). An acceptable level of noncarcinogenic health risk associated with legume consumption was thus suggested.

Numerous studies in the field of food research have consistently underscored that children face greater health risks than adults. In a study conducted by Su et al. (2023) examining health risks associated with vegetable consumption in South China, it was found that the HMs TTHQ in vegetables consumed by children was approximately 2.4 times higher than that of adults. Similarly, Wang et al. (2021) investigated the effects of steel-making activities and isotopic source distribution on individuals of varying age groups, including children, adults, and elderly individuals, who ingested vegetables. The results of this study concurred with those previous ones, emphasizing the heightened susceptibility of children. This heightened risk faced by children can be attributed to their developmental stage and the incomplete development of their bodily functions.

Table 8. Target hazard quotient for children

Category			Cr	As	Cd	Pb	Hg	Ni	Cu	Zn	TTHQ
	Legumes	Peanut	0.390	0.001	0.354	0.021	0.038	0.791	0.389	0.052	2.035
		Soybean	0.599	0.000	0.025	0.037	0.068	0.599	0.364	0.049	1.741
		Mung bean	0.139	0.000	0.003	0.044	0.085	0.315	0.319	0.043	0.949
	Cereals	Rice	0.002	0.032	0.029	0.339	3.983	0.693	0.173	0.231	5.481
		Wheat	0.003	0.001	0.105	0.157	0.231	0.036	0.138	0.183	0.853
		Corn	0.001	0.000	0.000	0.673	1.280	0.024	0.035	0.047	2.060
Vegetables	Leaf greens	Spinach	0.081	0.001	0.103	0.091	0.154	0.006	0.061	0.008	0.505
		Chinese chives	0.027	0.001	0.013	0.039	0.131	0.006	0.016	0.002	0.234
		Chinese cabbage	0.064	0.000	0.052	0.009	0.168	0.004	0.039	0.005	0.342
		Romaine lettuce	0.064	0.001	0.057	0.053	0.099	0.007	0.010	0.001	0.291
		Pak choi	0.064	0.002	0.089	0.042	0.160	0.004	0.037	0.005	0.404
		Lettuce	0.064	0.001	0.024	0.028	0.094	0.001	0.008	0.001	0.222
	Edible mushrooms	Needle mushroom	0.192	0.002	0.018	0.036	0.087	0.008	0.008	0.001	0.353
		Oyster mushroom	0.258	0.001	0.192	0.004	0.056	0.009	0.016	0.002	0.539
		Mushroom	0.278	0.001	0.297	0.033	0.123	0.007	0.027	0.004	0.769
	Solanaceous fruits	Tomato	0.013	0.000	0.018	0.012	0.056	0.000	0.005	0.001	0.104
		Cucumber	0.009	0.000	0.004	0.004	0.042	0.001	0.010	0.001	0.071
		Eggplant	0.017	0.000	0.016	0.010	0.052	0.000	0.009	0.001	0.106
		Pepper	0.035	0.000	0.021	0.006	0.060	0.000	0.012	0.002	0.136
	Tubers	Carrot	0.006	0.000	0.006	0.008	0.069	0.003	0.007	0.001	0.101
		Daikon	0.007	0.000	0.012	0.006	0.033	0.003	0.001	0.000	0.063
		Chinese yam	0.052	0.000	0.021	0.008	0.028	0.002	0.012	0.002	0.125
		Potato	0.048	0.001	0.024	0.008	0.045	0.013	0.012	0.002	0.154
	Kale	Cauliflower	0.010	0.000	0.014	0.001	0.075	0.003	0.005	0.001	0.109
		Cabbage	0.027	0.000	0.019	0.001	0.109	0.004	0.004	0.001	0.165
		Broccoli	0.036	0.001	0.016	0.002	0.052	0.005	0.009	0.001	0.121
TTHQ			2.488	0.048	1.529	1.670	7.380	2.546	1.725	0.644	18.031

Table 9. Target hazard quotient for adults

Category			Cr	As	Cd	Pb	Hg	Ni	Cu	Zn	TTHQ
	Legumes	Peanut	0.131	0.000	0.119	0.007	0.013	0.266	0.131	0.001	0.667
		Soybean	0.201	0.000	0.009	0.012	0.023	0.201	0.122	0.048	0.617
		Mung bean	0.047	0.000	0.001	0.015	0.029	0.106	0.107	0.046	0.351
	Cereals	Rice	0.331	0.014	0.036	0.142	1.673	0.055	0.264	0.189	2.704
		Wheat	0.567	0.000	0.132	0.066	0.097	0.037	0.210	0.105	1.213
		Corn	0.210	0.000	0.000	0.283	0.538	0.018	0.053	0.016	1.117
Vegetables	Leaf greens	Spinach	0.091	0.001	0.115	0.102	0.172	0.007	0.068	0.020	0.576
		Chinese chives	0.031	0.001	0.014	0.043	0.147	0.006	0.017	0.006	0.266
		Chinese cabbage	0.072	0.001	0.058	0.010	0.188	0.004	0.044	0.032	0.408
		Romaine lettuce	0.072	0.001	0.063	0.059	0.111	0.007	0.011	0.009	0.333
		Pak choi	0.072	0.003	0.099	0.048	0.179	0.005	0.042	0.023	0.470
		Lettuce	0.072	0.001	0.027	0.032	0.106	0.001	0.009	0.006	0.253
	Edible mushrooms	Needle mushroom	0.215	0.003	0.020	0.040	0.042	0.098	0.009	0.011	0.437
		Oyster mushroom	0.289	0.001	0.215	0.005	0.027	0.063	0.018	0.020	0.638
		Mushroom	0.311	0.001	0.333	0.037	0.059	0.138	0.030	0.033	0.942
	solanaceous fruits	Tomato	0.014	0.000	0.020	0.013	0.063	0.000	0.005	0.002	0.118
		Cucumber	0.010	0.000	0.004	0.004	0.047	0.001	0.011	0.005	0.083
		Eggplant	0.020	0.000	0.018	0.011	0.059	0.000	0.010	0.002	0.119
		Pepper	0.039	0.000	0.023	0.007	0.067	0.000	0.014	0.023	0.173
	Tubers	Carrot	0.007	0.001	0.006	0.009	0.078	0.004	0.007	0.027	0.138
		Daikon	0.008	0.000	0.013	0.006	0.037	0.003	0.002	0.004	0.074
		Chinese yam	0.059	0.000	0.023	0.009	0.031	0.002	0.013	0.022	0.161
		Potato	0.054	0.001	0.027	0.009	0.051	0.015	0.013	0.018	0.189
	Kale	Cauliflower	0.011	0.000	0.015	0.001	0.085	0.004	0.006	0.006	0.127
		Cabbage	0.031	0.000	0.021	0.001	0.122	0.004	0.005	0.004	0.188
		Broccoli	0.040	0.001	0.018	0.002	0.059	0.005	0.010	0.009	0.142
TTHQ			3.003	0.030	1.431	0.973	4.098	1.052	1.231	0.688	12.506

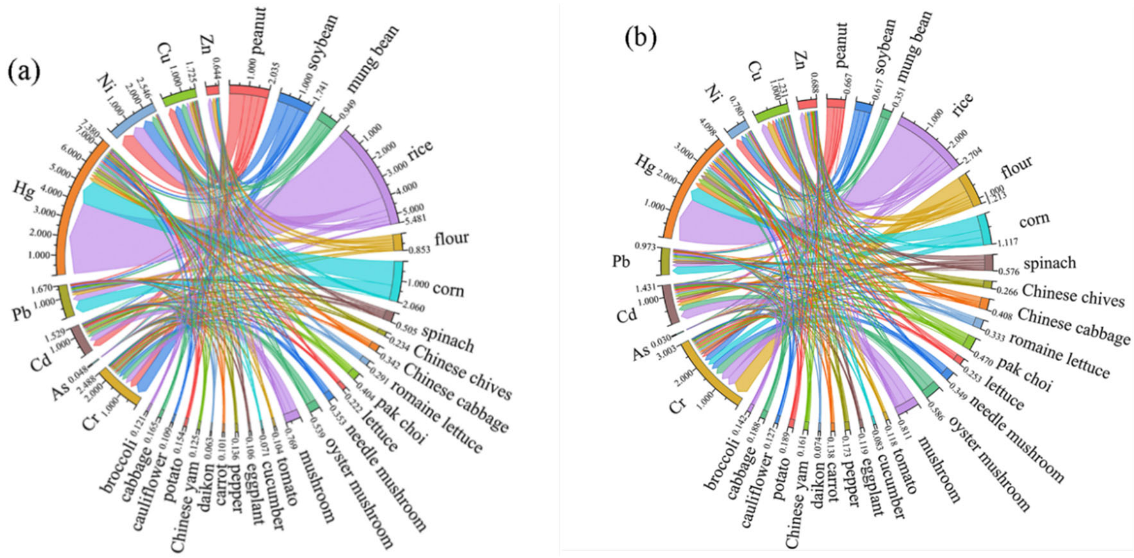


Figure 3. (a) represents THQ for children and (b) represents THQ for adults

3.3.2. Carcinogenic risk assessment

Figure 4(a) and Table 10 portrays the CR for children associated with HMs in different food types. The CR value of each individual HM was smaller than 1.00×10^{-4}, indicating that children are not subjected to significant carcinogenic health risks from the dietary intake of any single HM.

When examining cereals in more detail, the TCR values in descending order were as follows: wheat (8.65×10^{-7}) > rice (6.98×10^{-7}) > corn (2.42×10^{-7}). Meanwhile, within the vegetable category, the hierarchy of TCR, descendingly, was observed as follows: edible mushrooms (6.63×10^{-6}) > leafy greens (4.64×10^{-6}) > tubers (1.04×10^{-6}) > solanaceous fruits (7.43×10^{-7}) > kale (7.34×10^{-7}). For legumes, the TCR sequence was peanuts (1.55×10^{-6}) > soybeans (1.13×10^{-6}) > mung beans (2.56×10^{-7}). In general, the carcinogenic risk associated with each food group remains at acceptable levels.

As shown in Figure 4(b) and Table S11, adults exhibit a descending order of TCR for cereals as follows: rice (2.03×10^{-6}) > wheat ($1.96E\times10^{-6}$) > corn (3.69×10^{-7}). The TCR for vegetables, in descending order, unfolded as follows: edible mushrooms (1.98×10^{-6}) > leafy greens (6.80×10^{-6}) > tubers (2.32×10^{-7}) > kale (6.38×10^{-7}) > solanaceous fruits (6.60×10^{-7}). Meanwhile, among legumes, the TCR hierarchy in descending order was peanuts (1.10×10^{-6}) > soybeans (4.26×10^{-7}) > mung beans (9.34×10^{-8}). The carcinogenic risk associated with each food group was deemed within acceptable limits.

Based on the daily intake of cereals, legumes and vegetables, the health risks associated with the daily intake of the three food groups are shown in Table 1, the TCR for children linked to daily consumption of three food groups was below threshold of 1.00E-04. Similarly in adults, the daily intake of cereals, legumes and vegetables resulted in TCR values, the TCR values of legumes and vegetables less than 1.00E-04.

Table 10. Carcinogenic risks for children

Category			Cd	As	Cr	TCR
Legumes		Peanut	8.45E-07	1.71E-08	6.87E-07	1.55E-06
		Soybean	6.08E-08	1.012E-08	1.06E-06	1.13E-06
		Mung bean	8.19E-09	2.94E-09	2.45E-07	2.56E-07
Cereals		Rice	5.68E-08	2.60E-07	3.82E-07	6.98E-07
		Wheat	2.06E-07	4.64E-09	6.54E-07	8.65E-07
		Corn	0.00E+00	0.00E+00	2.42E-07	2.42E-07
Vegetables	Leaf greens	Spinach	9.81E-07	1.23E-07	1.43E-07	1.25E-06
		Chinese chives	1.20E-07	1.26E-07	4.80E-08	2.94E-07
		Chinese cabbage	4.96E-07	5.54E-08	1.14E-07	6.64E-07
		Romaine lettuce	5.41E-07	1.29E-07	1.14E-07	7.83E-07
		Pak choi	8.48E-07	2.86E-07	1.14E-07	1.25E-06
		Lettuce	2.33E-07	6.28E-08	1.14E-07	4.09E-07
	Edible mushrooms	Needle mushroom	1.70E-07	2.68E-07	3.38E-07	7.76E-07
		Oyster mushroom	1.83E-06	1.23E-07	4.55E-07	2.41E-06
		Mushroom	2.84E-06	1.18E-07	4.90E-07	3.45E-06
	Solanaceous fruits	Tomato	1.70E-07	7.38E-09	2.28E-08	2.00E-07
		Cucumber	3.60E-08	1.77E-08	1.62E-08	7.00E-08
		Eggplant	1.50E-07	1.97E-08	3.08E-08	2.01E-07
		Pepper	1.97E-07	1.27E-08	6.17E-08	2.72E-07
	Tubers	Carrot	5.42E-08	5.43E-08	1.04E-08	1.19E-07
		Daikon	1.14E-07	4.58E-08	1.27E-08	1.73E-07
		Chinese yam	2.00E-07	2.46E-08	9.23E-08	3.17E-07
		Potato	2.30E-07	1.16E-07	8.43E-08	4.30E-07
	Kale	Cauliflower	1.32E-07	2.36E-08	1.70E-08	1.73E-07
		Cabbage	1.80E-07	4.43E-08	4.80E-08	2.72E-07
		Broccoli	1.50E-07	5.91E-08	6.28E-08	2.72E-07
TCR			1.09E-05	2.01E-06	1.14E-07	1.85E-05

Table 11. Carcinogenic risks for adults

Category			Cd	As	Cr	TCR
Legumes		Peanut	8.51E-07	1.73E-08	2.31E-07	1.10E-06
		Soybean	6.12E-08	1.02E-08	3.54E-07	4.26E-07
		Mung bean	8.25E-09	2.96E-09	8.22E-08	9.34E-08
Cereals		Rice	2.60E-07	1.19E-06	5.83E-07	2.03E-06
		Wheat	9.46E-07	2.13E-08	9.98E-07	1.96E-06
		Corn	0.00E+00	0.00E+00	3.69E-07	3.69E-07
Vegetables	Leaf greens	Spinach	8.24E-07	1.03E-07	1.60E-07	1.09E-06
		Chinese chives	1.01E-07	1.05E-07	5.37E-08	2.60E-07
		Chinese cabbage	4.16E-07	4.65E-08	1.27E-07	5.90E-07
		Romaine lettuce	4.54E-07	1.08E-07	1.27E-07	6.89E-07
		Pak choi	7.12E-07	2.40E-07	1.27E-07	1.08E-06
		Lettuce	1.95E-07	5.27E-08	1.27E-07	3.75E-07
	Edible mushrooms	Needle mushroom	1.43E-07	2.25E-07	3.78E-07	7.46E-07
		Oyster mushroom	1.54E-06	1.03E-07	5.09E-07	2.15E-06
		Mushroom	2.38E-06	9.92E-08	5.48E-07	3.03E-06
	Solanaceous fruits	Tomato	1.43E-07	6.20E-09	2.55E-08	1.75E-07
		Cucumber	3.03E-08	1.49E-08	1.82E-08	6.34E-08
		Eggplant	1.26E-07	1.65E-08	3.45E-08	1.77E-07
		Pepper	1.66E-07	1.06E-08	6.91E-08	2.45E-07
	Tubers	Carrot	4.55E-08	4.56E-08	1.17E-08	1.03E-07
		Daikon	9.60E-08	3.85E-08	1.42E-08	1.49E-07
		Chinese yam	1.68E-07	2.07E-08	1.03E-07	2.92E-07
		Potato	1.93E-07	9.72E-08	9.44E-08	3.85E-07
	Kale	Cauliflower	1.11E-07	1.98E-08	1.90E-08	1.50E-07
		Cabbage	1.51E-07	3.72E-08	5.37E-08	2.42E-07
		Broccoli	1.26E-07	4.96E-08	7.03E-08	2.46E-07
TCR			1.03E-05	2.68E-06	5.29E-06	1.82E-05

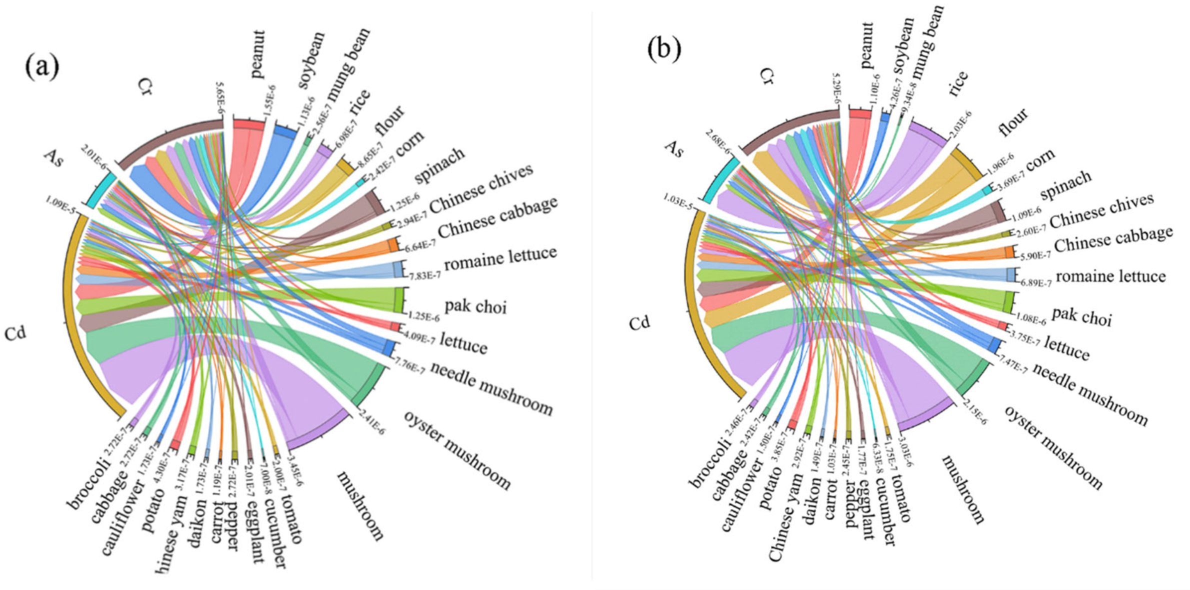


Figure 4. (a) represents TCR for children and (b) represents TCR for adults

3.4. Cumulative health risks due to simultaneous consumption of the three types of food

People's diet is always composed of various types of food, such as cereals, vegetables, as well as some other supplementary foods. Thus, the health risks posed by HMs in any individual type of food would not represent the "real" risks encountered by people. As mentioned above, cereals, vegetables, and legumes collectively account for over 60% of the total dietary intake of individuals. It is thus interesting to calculated the accumulative health risks induced by HMs when eating all the three types of food. It is important to recognize that humans are exposed to HMs through various pathways, including inhalation from the air and dermal absorption from soil, particularly through the consumption of plants grown in HM-contaminated soils (Sawut et al. 2018). This multifaceted exposure may further elevate overall health risks within the population.

Table 12 presented the mean values of TTHQ and TCR of each group of foods, as well as the sums of all the three groups of foods. It was suggested that prolonged consumption of those three types of food by both children and adults may pose noncarcinogenic health risks, since the cumulative TTHQs were higher than 1. It is noteworthy that the cumulative TCR values for both children and adults remained below 1.00×10^{-4}, indicating that the CR stemming from food intake was within acceptable limits for both age groups.

Yang et al. (2017) elucidated that different foods contribute unequally to noncarcinogenic health risks, with vegetables exerting the most significant impact, followed by cereals, while meat and eggs played comparatively smaller roles, accounting for 71%, 23%, and 6% of the contribution, respectively. Chen et al. (2023), in their study of dietary lead exposure in Chongqing, found that vegetables and their derivatives accounted for 39.1%-45.3% of the contribution to health risk, grains and grain products accounted for 37.4%-39.5%, whereas meat and meat products made up only 7.5%-8.9%. Other foods examined contributed even less. Notably, the Fifth Chinese Whole Diet Study (Liu et al. 2019) pinpointed that the most substantial sources of lead intake were cereals and vegetables. However, our research found a quite different result. The data in Table 12 clearly showed that cereal contributed the most to both TTHQ and TCR for children and adults. This discrepancy might be attributed to the difference in sampling sites, since contents of HMs in plants are mainly affected by the growing environment.

Although other types of food, such as meat, egg and dairy products, were not tested in the present study, their contribution to health risk may not be significant, as revealed by previous studies (Yang et al. 2017; Liu et al. 2019; Chen et al. 2023). Results obtained in this study could reasonably approximate the overall health risks posed by HMs in the commercially available foods to local population.

Table 12. Cumulative health risks caused by HMs in the three types of food

	TTHQ		TCR	
people	Children	Adults	Children	Adults
Vegetables	0.246	0.292	7.34×10-6	6.53×10-7
Cereals	2.798	1.678	6.02×10-7	1.46×10-6
Legumes	1.575	0.545	9.77×10-7	5.39×10-7
Cumulative TTHQ/TCR	4.619	2.518	8.92×10-6	2.65×10-6

4. CONCLUSION

In this research endeavor, cereals, vegetables, and legumes were purposefully selected as subjects of investigation to scrutinize the extent of HMs contamination within these dietary constituents and the consequential health hazards they may impose upon human consumers. The findings revealed that cereals and legumes exhibited elevated levels of HMs in comparison to the entirety of vegetables. Analysis revealed that the proportions of Cr, Pb, and Hg and Ni exceeding the standards in cereal samples were 2.60%, 3.90%, 44.16% and 3.90%, respectively, while the contents of As, Cd, Cu and Zn remained within the specified standard range. With the exception of exceedance rates for Hg and Zn in vegetables samples were 0.90% and 17.12%, the levels of HMs in the remaining vegetables were below the corresponding standard limits. Additionally, the legume category showed a 16.98% exceedance for Cr, a 13.21% exceedance for Pb and a 75.47% exceedance for Ni. The dietary constituents exhibiting more pronounced deviations from the prescribed standards included rice (Hg), corn (Hg), soybeans (Cr, Ni), peanut (Ni) and mung bean (Ni). Comparing these results, it becomes evident that the contamination scenario is considerably more favorable within the realm of vegetables.

A comprehensive assessment of contamination levels across the sampled foods indicated the following hierarchy: legumes > cereals > vegetables. This hierarchy underscores that cereals and legumes bear the brunt of contamination within this context.

In the context of noncarcinogenic risk assessment, the THQ for individual metals generally remained below the stipulated standards in the study area, except for Hg in rice and wheat consumed by children, where THQ exceeded the threshold of 1. Nevertheless, when considering the combined effect of multiple HMs, the TTHQ surpassed 1 for rice and corn in cereals, as well as peanuts and soybeans in legumes, indicating potential adverse health effects associated with the consumption of these foods by children. For adults, the THQ of single HMs were mostly below 1, with the exception of Hg in rice. The TTHQ for rice, wheat, and corn in the cereal group were greater than 1, suggesting that the consumption of these three types of foods by adults would result in adverse health effects. The CR values for individual HMs are all less than 1.00×10^{-4} for children and adults. The TCR values for various food types were 1.85×10^{-5} for children and 1.82×10^{-5} for adults, both of which remained below the 1.00×10^{-4} threshold, thereby placing the TCR associated with the prolonged consumption of these three dietary groups within an acceptable range.

Based on the daily intake of cereals, legumes and vegetables, the cumulative TTHQ exceeded a threshold of 1. Prolonged consumption of these three categories of food elevates health risks beyond acceptable levels. Conversely, the carcinogenic risks associated with the daily intake of the three food groups were within an acceptable range. It is imperative to highlight that children are potentially more susceptible than adults to health risks arising from the consumption of contaminated foods.

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