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Contamination status and risk assessment of fifteen elements in peanuts: a case study of Shandong Province, China

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Abstract

Food safety has always attracted considerable attention in China. However, few studies have investigated the pollution, dietary exposure, and health risk assessment of fifteen elements in peanuts. Herein, we collected 620 samples from 16 districts in Shandong province from 2020 to 2021 and detected the concentrations of Mn, Cu, Li, Al, Ni, Ba, Cr, Cd, Se, Pb, Sn, V, As, Sb, and Hg. Further, we evaluated the contamination degree of elements in peanuts using the Single Factor Pollution Index, Nemerow Comprehensive Pollution Index methods, Pollutant Sharing Rate and Quality Grading Standards. Finally, we successful assessed the dietary exposure and health risk of thirteen elements using Target Hazard Quotient method based on the contents and peanut intake of Shandong residents. SPIM and NPIM results showed that the products were slightly polluted by Cu and Pb. THQ results showed that dietary exposure risk of children was two to three times than that of adults, suggesting that children were more susceptible to non-carcinogenic risk from metals. HI results indicated that excessive peanut intake would harm human health, especially to children. Higher dietary exposure risk existed in Cu, Ni and Mn.

Keywords: peanut; contamination status; risk assessment; multi-element analysis; food safety.

Practical Application: We applied the model provided by the project results to fit and calculate the content of heavy metals in peanuts, the pollution situation and the dietary exposure risk probability of peanut of Shandong Province, which provided a scientific basis for understanding the pollution situation of heavy metals in peanuts. According to the risk model of the project results, we have identified the hidden dangers of heavy metal pollution, and the linkage with the regulatory authorities and data sharing were actively carried out, which improved the pertinence and purpose of supervision, strengthened the health quality supervision of peanuts. It has made certain contributions to the construction of food safety in Shandong Province.

1 Introduction

Peanut, as a main oilseed crop in the world, is a rich source of dietary protein and culinary oil, playing an important role in animal and human nutrition (Settaluri et al., 2012; Zou et al., 2022c). Scientific research shows that peanuts have anti-cancer, antioxidant, anti-inflammatory and other biological properties (Zou et al., 2022a). Peanut is susceptible to contamination by heavy metals, which hurts the healthy development of the peanut industry in China. China is one of the top three peanut producing countries in the world (Zou et al., 2022d). In recent years, the output of peanuts in China has exceeded 17 million tons (Wang et al., 2021). The yield and unit yield of peanuts rank first in the world, and the overall consumption of peanuts has maintained a steady growth (Zou et al., 2022b). Export is the mainly form of China's peanut trade, which also plays an important role in the world peanut trade. Shandong produced 2.85 million tons of peanuts in 2019, which reached second place in China's peanut production (Zhang & Wang, 2020; Yang et al., 2020). Peanut is an underground bearing crop, in addition to the root system can absorb arsenic (As) and fourteen metal elements. There was a significant positive correlation between heavy metal pollution of peanuts and heavy metal content in soil (Yang et al., 2020).

Lead (Pb) is classified as "possibly carcinogenic to humans" (group 2B) by the International Agency for Research on Cancer (IARC) (Vogel et al., 2021). Pb can accumulate in multiple organs and then affect the function of the immune, endocrine, and other systems (Charkiewicz & Backstrand, 2020). Cd accumulation within the human body can induce kidney damage, probably through its potential hazard to the proximal tubular epithelial cells (Jin et al., 2020). Mercury (Hg) brings adverse effects to the environment and human beings and inorganic mercury (IHg) is a typical hepatic toxin. Besides the direct deposition in liver of Hg, the perturbance to gut microbiome and alteration of gut-liver axis related metabolites by IHg also contributed to its hepatoxicity, which provides new insights about the hepatoxicity of chemicals (Lin et al., 2021). Increased mercury levels were associated with infertility or subfertility status (Henriques et al., 2019). As is classified as a Group I human carcinogen by IARC. Arsenic exposure to human through various sources such as

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contaminated groundwater and other human activities have become a significant global concern. Major diseases that have been linked to arsenic poisoning are diabetes, hyperkeratosis, cancer, hypertension, and neurodegeneration (Fatoki & Badmus, 2022). Many adverse health effects including skin diseases, carcinogenesis, and neurological diseases have been reported due to arsenic exposure (Rahaman et al., 2021). Copper (Cu) and zinc (Zn) are essential elements for plants and humans, but their absence or excessive intake via food can cause anemia, human metabolic disorders, developmental retardation, and chronic toxicities (Wang & Liu, 2007; Kabata-Pendias & Mukherjee, 2007). Chromium (Cr) can cause cell muta-tion and carcinogenesis (Ognik et al., 2021), and Nickel (Ni) can lead to the decline of intelligence quotient (IQ) in children (Bi et al., 2018; Ghasemidehkordi et al., 2018). Trace elements pollution results in the decline in quality of peanut, seriously threatens people's life and health and ecological environment, and also affects the healthy development of peanut industry (Liu et al., 2020).

Peanut of different origins, affected by factors such as sunlight, air, soil, water, and the surrounding environment, the metabolism of the body shows different levels of enrichment or depletion of elements in the environment. Therefore, it is particularly important to investigate the enrichment conditions and enrichment rules in peanuts. In this study, 620 copies of peanuts were carried out in 16 prefecture-level cities of Shandong Province, China, to monitor the content of fifteen elements, and then the pollution status and degree were analyzed and evaluated by using the pollution index method and the target hazard factor method.

2 Materials and methods

2.1 Sample collection

From October 2020 to October 2021, 620 peanut samples were collected from 16 prefecture-level cities in Shandong Province (34°N-38°N, 114°E-122°E), which was one of the main peanuts producing areas in China. Each prefecture-level city set 20 sampling points per year (except Qingdao). To ensure representativeness and dispersity of samples, only one sample was allowed to collect at one sampling site. The peanuts sampling was collected during harvest time from the field or farmer's home. The locations of 16 prefecture-level cities were marked in Figure 1.

2.2 Methods and standards

Detection methods and evaluation criteria

In accordance with the standard operating procedures specified in the *National Food Contaminants and Harmful Factors Risk Monitoring Workbook* (Yang & Li, 2013). Pb, Cd, As, aluminum (Al), chromium (Cr), nickel (Ni), Cu, selenium (Se), vanadium (V), manganese (Mn), lithium (Li), antimony (Sb), barium (Ba), and stannum (Sn) were detected by microwave ablation-inductively coupled plasma mass spectrometry, and Hg was detected by atomic fluorescence spectrophotometry. The detection limits of different elements are Pb: 0.004 $mg \cdot kg^{-1}$, Cd: 0.003 $mg \cdot kg^{-1}$, As: 0.004 $mg \cdot kg^{-1}$, Al: 0.3 $mg \cdot kg^{-1}$, Cr: 0.01 $mg \cdot kg^{-1}$, Ni: 0.01 $mg \cdot kg^{-1}$, Cu: 0.01 $mg \cdot kg^{-1}$, Se: 0.005 $mg \cdot kg^{-1}$, V: 0.004 $mg \cdot kg^{-1}$, Mn: 0.01 $mg \cdot kg^{-1}$, Li: 0.02 $mg \cdot kg^{-1}$, Sb: 0.004 $mg \cdot kg^{-1}$, Ba: 0.02 $mg \cdot kg^{-1}$, Sn: 0.004 $mg \cdot kg^{-1}$, Hg: 0.0005 $mg \cdot kg^{-1}$. In accordance with the *National Food Safety Standard Limits of Contaminants in Food (GB 2762-2017)* (National Health and Family Planning Commission of the People's Republic of China, 2017) and the *Eight Elemental Limits of Pb, Cd, Hg, Se, As, Cu, and Zn in Grain (including cereals, legumes, and potatoes) and Products (NY 861-2004)* (Ministry of Agriculture of the People's Republic of China, 2005) in peanuts and legumes elements (Peanut belongs to the legume family, so bean element standards have certain reference value) were finally determined as follows: Cd ≤ 0.5 $mg \cdot kg^{-1}$, Pb ≤ 0.2 $mg \cdot kg^{-1}$, As ≤ 0.5 $mg \cdot kg^{-1}$, Cr ≤ 1.0 $mg \cdot kg^{-1}$, Hg ≤ 0.02 $mg \cdot kg^{-1}$, Cu ≤ 20 $mg \cdot kg^{-1}$, Se ≤ 0.3 $mg \cdot kg^{-1}$.

Evaluation methods and criteria

The elements contamination in peanuts were evaluated according to the single factor pollution index method (SPIM), Nemerow composite pollution index method (NPIM), Pollutant sharing rate (PSR), and Quality grading standards (QGS) in

Figure 1. The locations of Shandong Province in China (a) and sixteen cities of Shandong Province (b).

NY/T 398-2000 (Ministry of Agriculture of the People's Republic of China, 2000). SPIM can only reflect the pollution degree of each heavy metal element, cannot fully reflect the contamination status of food. NPIM is one of the most commonly used methods for comprehensive pollution index calculation. It considers the average and the highest value of SPIM, can highlight the effect of pollutants with the largest pollution index on food quality (Equation 1).

$$SPIM : P_i = C_i / S_i \quad (1)$$

P_i: the single factor pollution index of i elements; C_i (mg·kg^{-1}): the measured concentration of i elements; S_i (mg/kg): the evaluation standard of i elements. QGS: $P_i \le 0.6$ is safe; $0.6 < P_i < 1.0$ is lightly polluted; $P_i \ge 1.0$ is heavily polluted.

Equation 2:

$$NPIM : P_{syn} = \sqrt{\left(P^2{}_{max} + P^2{}_{ave}\right)/2} \quad (2)$$

P_{syn}: the integrated pollution index of Nemerow; P_{max}: maximum value of a variety of single factor index in 7 elements (P_i); P_{ave}: average value of a variety of single factor index in 7 elements (P_i).

Equation 3:

$$PSR : K_i = \left(P_i / P\right) \times 100\% \quad (3)$$

K_i: i element occupies the sharing rate of all elements pollutant, %; P_i: single factor pollution index of i element; P: sum of single factor pollution index of each element.

Evaluation method and standard of edible safety

Target Hazard Quotient (*THQ*) method published by United States Environmental Protection Agency was used to conduct food safety evaluation. THQ method was a risk assessment method for assessing human exposure to heavy metals through food, which assumed that the absorbed dose of the contaminant was equal to the absorbed dose, using the ratio of the measured intake dose of pollutants to the reference dose as the evaluation criterion. $THQ \le 1$ indicated there was no significant health risk of exposed people; $THQ > 1$ indicated that health risk existed. The higher the *THQ* value, the more serious the risk to human health.

Single element risk calculation formula (Equation 4):

$$THQ = \frac{E_F \times E_D \times F_{IR} \times c}{R_{FD} \times W_{AB} \times AT_n} \times 10^{-3} \quad (4)$$

E_F(d/a): exposure frequency, value as 365 (United States Environmental Protection Agency, 2022).

E_D(a): exposure time, value as 70 (United States Environmental Protection Agency, 2022).

F_{IR}(g/d): peanut intake, general intake of 2~6 years old children was 1.66 g/d, general intake of 2~6 years old children was 24.9 g/d, general intake of standard person was 3.02 g/d, high intake of standard person was 35.7 g/d (Wang & Liu, 2007).

c(mg/kg): elements content in peanuts, from this study.

W_{AB} *(kg)*: the average weight data of 2~6 years old children were obtained from *The second monitoring report on the nutritional and health status of Chinese residents: the physical and nutritional status of residents from 2010 to 2013* (Piao & Huo, 2019), which was 17.3 kg; the standard human weight was taken as 60 kg (United States Environmental Protection Agency, 2022).

AT_n(d): non-carcinogenic mean exposure time, value as 25550 (United States Environmental Protection Agency, 2022).

RfD(mg/(kg·d)): reference dose, $RfD_{(Pb)} = 0.0035$ (Ihugba et al., 2018), $RfD_{(Cd)} = 1 \times 10^{-3}$ (United States Environmental Protection Agency, 2022), $RfD_{(As)} = 3 \times 10^{-4}$ (United States Environmental Protection Agency, 2022), $RfD_{(Cr)} = 3 \times 10^{-3}$ (United States Environmental Protection Agency, 2022), $RfD_{(Se)} = 5 \times 10^{-3}$ (United States Environmental Protection Agency, 2022), $RfD_{(Mn)} = 1.4 \times 10^{-1}$ (United States Environmental Protection Agency, 2022), $RfD_{(Cu)} = 4 \times 10^{-2}$ (Ihugba et al., 2018), $RfD_{(Ni)} = 2 \times 10^{-2}$ (United States Environmental Protection Agency, 2022), $RfD_{(Ti)} = 4 \times 10^{-4}$ (United States Environmental Protection Agency, 2022), $RfD_{(Ba)} = 2 \times 10^{-1}$ (United States Environmental Protection Agency, 2022), $RfD_{(Hg)} = 4.54 \times 10^{-5}$ (Chinese Research Academy of Environmental Sciences, 2019), $RfD_{(V)} = 9 \times 10^{-2}$ (Pan et al., 2002), $RfD_{(Al)} = 1$ (Wang et al., 2019).

Hazard Index *(HI)* denotes the sum of *THQ*, Multiple elements risk calculation formula (Equation 5):

$$HI = \Sigma THQ \quad (5)$$

THQ_g denotes *THQ* general intake exposure, THQ_h denotes *THQ* high dietary exposure, HI_g denotes *HI* general intake exposure, HI_h denotes *HI* high dietary exposure.

Data processing

According to the WHO principles for handling undetected data, when the proportion of undetected data was higher than 60%, all undetected data were replaced by the limit of detection (LOD), while the proportion of undetected data was less than or equal to 60%, all undetected data were replaced by 1/2 LOD (Wang et al., 2002). Excel and PASW Statistics 18.0 trial version software were used for statistical analysis of the data.

3 Results and discussion

3.1 Concentrations of fifteen elements in samples

Fifteen elements were all checkout in 620 samples. The results (Table 1) showed that the elements with high detection rates were Cu, Ni, Mn, Ba, Al, and Li, all with 100% detection rates, followed by Cd, Se, Cr, As, and Pb, with detection rates of 95.65% (593/620), 89.68% (556/620), 85.81% (532/620), 78.71% (488/620), and 77.74% (482/620).The low detection rates were V, Sn, Hg, and Sb, with detection rates of 61.13% (379/620), 58.55% (363/620), 55.81% (346/620), and 14.19% (88/620), respectively. The average values of fifteen elements in descending

Table 1. Detection results of fifteen elements in peanut samples of Shandong province.

Element	detectable rate/%	pollution levels/(mg/kg)									
		range of checkout	average	standard deviation	P_{25}	P_{50}	P_{75}	P_{90}	P_{95}	$P_{97.5}$	P_{99}
Pb	77.74 (482/620)	ND~0.143	0.0290	0.0286	0.0122	0.0187	0.0402	0.0735	0.0919	0.1030	0.1281
Cd	95.65 (593/620)	ND~0.332	0.0628	0.0450	0.0306	0.0550	0.0849	0.1150	0.1438	0.1799	0.2129
Hg	55.81 (346/620)	ND~0.00958	0.0020	0.0020	0.0003	0.0017	0.0033	0.0049	0.0057	0.0062	0.0076
As	78.71 (488/620)	ND~0.0923	0.0220	0.0149	0.0131	0.0220	0.0300	0.0403	0.0484	0.0562	0.0674
Cu	100 (620/620)	0.487~18.969	7.9508	3.0098	5.7982	7.7875	9.7763	12.2535	13.6186	14.2908	14.6424
Cr	85.81 (532/620)	ND~0.359	0.1340	0.0914	0.0583	0.1290	0.2070	0.2629	0.2840	0.2985	0.3380
Se	89.68 (532/620)	ND~0.101	0.0447	0.0245	0.0264	0.0454	0.0625	0.0778	0.0850	0.0909	0.0958
Ni	100 (620/620)	0.035~16.8	2.9493	3.1021	0.5325	1.7000	4.9650	7.5790	9.1125	11.3475	13.0790
Mn	100 (620/620)	0.779~45.0	19.8726	9.9986	11.7000	18.6500	27.0000	33.9900	40.1750	43.0000	44.2790
Ba	100 (620/620)	0.0793~8.37	2.3597	1.6270	1.0100	2.0800	3.3200	4.6500	5.3085	6.2443	7.1817
Al	100 (620/620)	0.925~12.0	4.2674	2.2291	2.6925	3.70	5.4675	7.3680	8.8900	9.9390	11.2000
Li	100 (620/620)	0.182~23.2	6.7757	4.3403	3.7300	5.9700	8.8900	12.6000	15.9000	17.7475	21.479
V	61.13 (379/620)	ND~0.097	0.02429	0.0237	0.0020	0.0205	0.0354	0.0635	0.0708	0.0765	0.0879
Sn	58.55 (363/620)	ND~0.117	0.0278	0.0298	0.0020	0.0164	0.0493	0.0767	0.08790	0.0940	0.1028
Sb	14.19 (88/620)	ND~0.107	0.0061	0.0065	0.0040	0.0040	0.0040	0.0145	0.0176	0.0228	0.0258

Note: ND means not detected.

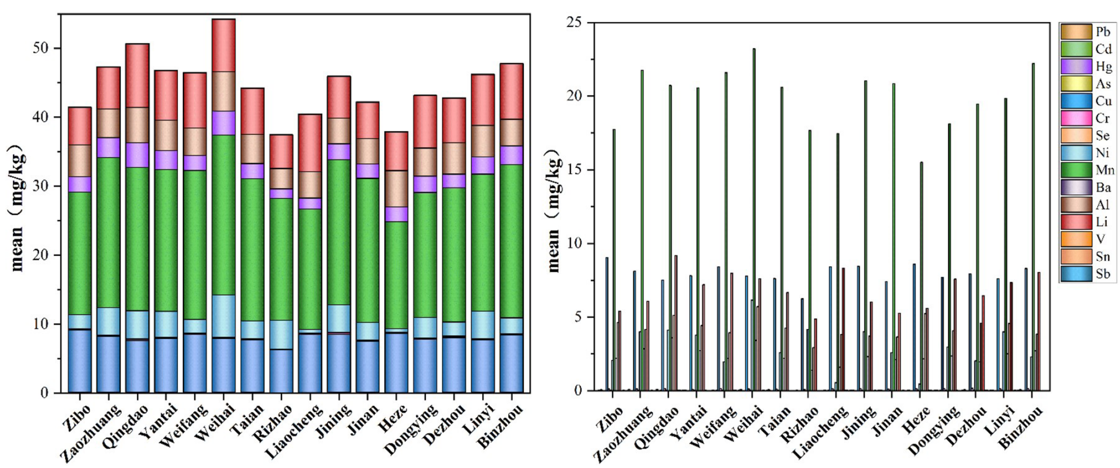

Figure 2. Detection of 15 elements in peanut in different regions.

order were Mn (19.8726 mg·kg^{-1}) > Cu (7.9508 mg·kg^{-1}) > Li (6.7757 mg·kg^{-1}) > Al (4.2674 mg·kg^{-1}) > Ni (2.9493 mg·kg^{-1}) > Ba (2.3597 mg·kg^{-1}) > Cr (0.1340 mg·kg^{-1}) > Cd (0.0628 mg·kg^{-1}) > Se (0.0447 mg·kg^{-1}) > Pb (0.0290 mg·kg^{-1}) > Sn (0.0278 mg·kg^{-1}) > V (0.0278 mg·kg^{-1}) > As (0.0220 mg·kg^{-1}) > Sb (0.0061 mg·kg^{-1}) > Hg (0.0020 mg·kg^{-1}). In general, Peanuts were polluted by many elements in Shandong Province.

3.2 Distribution of 15 elements in peanut in different regions

The contents of elements in peanut in different areas are shown in Figure 2 and Table 2, which showed that the total value was high in Weihai, Qingdao and Binzhou, while the total value was low in Rizhao and Heze. The average content of Hg (0.0027 mg·kg^{-1}), As (0.0354 mg·kg^{-1}), Ba (3.6089 mg·kg^{-1}), Li (9.1766 mg·kg^{-1}), and Sn (0.0589 mg·kg^{-1}) in Qingdao was the highest. The average content of Ni (6.1538 mg·kg^{-1}), Mn (23.2201 mg·kg^{-1}), and Al (5.7139 mg·kg^{-1}) in Weihai was the highest. The average content of Pb (0.0497 mg·kg^{-1}) and Cu (9.0404 mg·kg^{-1}) in Zibo was the highest. The average content of Cr (0.1727 mg·kg^{-1}) and Se (0.0548 mg·kg^{-1}) in Dezhou was the highest. The average content of V (0.0316 mg·kg^{-1}) in Heze and Sb (0.0093 mg·kg^{-1}) in Jinan was the highest.

Forty samples were randomly collected in each city (except Qingdao, where only 20 samples were collected due to the impact of COVID-19). The detection rates of 15 elements in peanuts in different cities were shown in Table 3. The detection rates of Pb (65%), Cd (80%), As (52.5%), Se (52.5%), Sn (45%), and Sb (2.5%) were the lowest in Rizhao City. The detection rates of Cr (62.5%), V (45%), Hg (45%) were the lowest in Weifang, Weihai, and Jinan City, respectively.

3.3 Pollution index of Pb, Cd, Hg, As, Cu, Cr, and Se

SPIM and NPIM were used to calculate the pollution index of Pb, Cd, Hg, As, Cu, Cr, and Se in peanut according to

Table 2. Average contents of 15 elements in peanut from different cities of Shandong province.

cities	Average Contents of 15 Elements in Peanut from different cities of Shandong province (mg/kg)														
	Pb	Cd	Hg	As	Cu	Cr	Se	Ni	Mn	Ba	Al	Li	V	Sn	Sb
Zibo	0.0497	0.0679	0.0020	0.0242	9.0404	0.1252	0.0345	2.0712	17.7358	2.2223	4.6287	5.3998	0.0241	0.0265	0.0065
Zaozhuang	0.0204	0.0961	0.0025	0.0196	8.1210	0.1271	0.0379	4.0108	21.7581	2.8593	4.1622	6.0711	0.0213	0.0314	0.0081
Qingdao	0.0241	0.0822	0.0027	0.0354	7.5139	0.1661	0.0370	4.1156	20.7307	3.6089	5.1258	9.1766	0.0289	0.0589	0.0048
Yantai	0.0414	0.0653	0.0018	0.0189	7.8112	0.1036	0.0459	3.7735	20.5573	2.7361	4.4339	7.1977	0.0249	0.0195	0.0045
Weifang	0.0194	0.0557	0.0019	0.0275	8.4175	0.1631	0.0448	1.9638	21.5906	2.1922	3.9448	7.9730	0.0252	0.0325	0.0054
Weihai	0.0251	0.0771	0.0023	0.0175	7.7865	0.1249	0.0435	6.1538	23.2201	3.4197	5.7139	7.6123	0.0197	0.0332	0.0083
Taian	0.0268	0.0676	0.0016	0.0226	7.6243	0.1119	0.0533	2.5924	20.5955	2.2076	4.2494	6.6716	0.0246	0.0219	0.0053
Rizhao	0.0167	0.0309	0.0024	0.0145	6.2509	0.0842	0.0165	4.1679	17.6658	1.3862	2.9317	4.8678	0.0227	0.0127	0.0043
Liaocheng	0.0199	0.0487	0.0018	0.0255	8.4154	0.1316	0.0506	0.5490	17.4655	1.5962	3.8178	8.3258	0.0262	0.0304	0.0056
Jining	0.0337	0.0598	0.0021	0.0228	8.4569	0.1581	0.0541	4.0078	21.0529	2.3163	3.7153	6.0255	0.0231	0.0286	0.0049
Jinan	0.0279	0.0596	0.0019	0.0209	7.4176	0.1184	0.0416	2.5995	20.8520	2.1186	3.6546	5.2743	0.0212	0.0314	0.0093
Heze	0.0166	0.0588	0.0022	0.0212	8.5891	0.1376	0.0473	0.4591	15.5119	2.1732	5.2392	5.5978	0.0316	0.0276	0.0063
Dongying	0.0264	0.0478	0.0018	0.0248	7.6997	0.1596	0.0543	2.9694	18.1231	2.3621	4.0792	7.5795	0.0251	0.0235	0.0053
Dezhou	0.0347	0.0718	0.0022	0.0214	7.9361	0.1727	0.0548	2.0384	19.4585	1.9569	4.5911	6.4456	0.0280	0.0262	0.0058
Linyi	0.0303	0.0549	0.0016	0.0233	7.6072	0.1347	0.0470	3.9954	19.8554	2.5079	4.5718	7.3503	0.0237	0.0354	0.0054
Binzhou	0.0487	0.0699	0.0023	0.0188	8.3068	0.1405	0.0485	2.3030	22.2114	2.7121	3.8428	8.0302	0.0205	0.0210	0.0055

Table 3. Detection rate of 15 elements in peanut from different cities of Shandong Province (%).

cities	Detection rate of 15 elements in peanut from different cities of Shandong Province (%)														
	Pb	Cd	Hg	As	Cu	Cr	Se	Ni	Mn	Ba	Al	Li	V	Sn	Sb
Zibo	77.5	97.5	60	90	100	92.5	95	100	100	100	100	100	67.5	62.5	15
Zaozhuang	72.5	97.5	65	77.5	100	90	87.5	100	100	100	100	100	52.5	70	42.5
Qingdao	75	100	70	95	100	100	90	100	100	100	100	100	75	100	5
Yantai	82.5	95	50	77.5	100	70	92.5	100	100	100	100	100	57.5	52.5	5
Weifang	77.5	97.5	50	95	100	62.5	97.5	100	100	100	100	100	65	60	10
Weihai	77.5	95	62.5	67.5	100	80	77.5	100	100	100	100	100	45	55	12.5
Taian	82.5	97.5	47.5	82.5	100	87.5	95	100	100	100	100	100	62.5	57.5	10
Rizhao	65	80	57.5	52.5	100	72.5	52.5	100	100	100	100	100	57.5	45	2.5
Liaocheng	75	95	50	80	100	87.5	92.5	100	100	100	100	100	67.5	57.5	12.5
Jining	82.5	95	57.5	87.5	100	92.5	95	100	100	100	100	100	55	55	10
Jinan	77.5	97.5	45	80	100	87.5	87.5	100	100	100	100	100	65	65	37.5
Heze	75	100	50	80	100	77.5	92.5	100	100	100	100	100	70	57.5	15
Dongying	72.5	95	55	80	100	97.5	95	100	100	100	100	100	52.5	47.5	7.5
Dezhou	77.5	100	62.5	70	100	92.5	95	100	100	100	100	100	77.5	57.5	12.5
Linyi	77.5	92.5	47.5	85	100	82.5	95	100	100	100	100	100	62.5	62.5	10
Binzhou	95	97.5	70	67.5	100	82.5	95	100	100	100	100	100	52.5	52.5	15

Equations 1-2. The results (Figure 3, Table 4) showed that the highest single factor pollution index was Cu, while the lowest single factor pollution indices was As and Hg. $P_{Cu(P90)}$, $P_{Cu(P95)}$, $P_{Cu(P97.5)}$, $P_{Cu(P99)}$ and $P_{Pb(P99)}$ were all higher than 0.6, which showed that the product was in a lightly polluted state on Cu and Pb. However, due to the extremely high detection rate of Cu and Pb, attention should be paid to health effects. The value of $P_{syn(mean)}$, $P_{syn(P25)}$, $P_{syn(P50)}$, $P_{syn(P75)}$, $P_{syn(P90)}$, $P_{syn(P95)}$, $P_{syn(P97.5)}$ and $P_{syn(P99)}$ was 0.3021, 0.2138, 0.2934, 0.3774, 0.4805, 0.5380, 0.5701 and 0.5984, respectively, and the $P_{syn(P99)}$ is close to 0.6, indicating a higher combined pollution rate at the higher dietary intake percentile.

3.4 PSR of Pb, Cd, Hg, As, Cu, Cr, and Se

PSR of *Pb, Cd, Hg, As, Cu, Cr, and Se* was calculated according to Equation 3, and the results showed that K_{Cu} was the highest among the 7 elements (Figure 4). Cu and Pb were the most

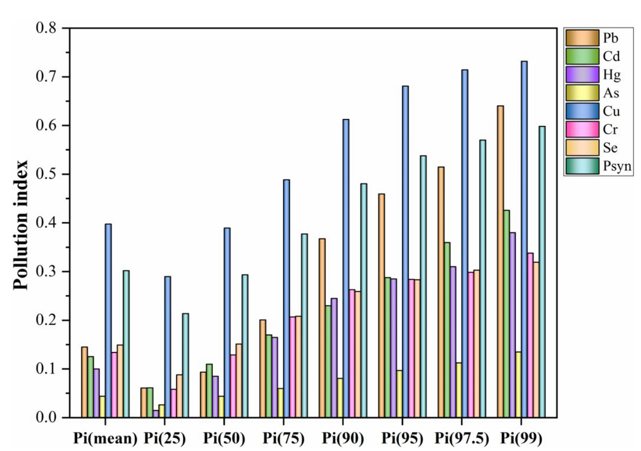

Figure 3. Single factor pollution index and comprehensive pollution index of Pb, Cd, Hg, As, Cu, Cr, and Se in peanut. P_{syn}: the integrated pollution index of Nemerow.

Table 4. Single factor pollution index and comprehensive pollution index of Pb, Cd, Hg, As, Cu, Cr and Se in peanut.

Element	$P_{i(mean)}$	$P_{i\,(25)}$	$P_{i\,(50)}$	$P_{i\,(75)}$	$P_{i\,(90)}$	$P_{i\,(95)}$	$P_{i\,(97.5)}$	$P_{i\,(99)}$
Pb	0.1450	0.0610	0.0935	0.2010	0.3675	0.4595	0.5150	0.6405
Cd	0.1256	0.0612	0.1100	0.1698	0.2300	0.2876	0.3598	0.4258
Hg	0.1000	0.0150	0.0850	0.1650	0.2450	0.2850	0.3100	0.3800
As	0.0440	0.0262	0.0440	0.0600	0.0806	0.0968	0.1124	0.1348
Cu	0.3975	0.2899	0.3894	0.4888	0.6127	0.6809	0.7145	0.7321
Cr	0.1340	0.0583	0.1290	0.2070	0.2629	0.2840	0.2985	0.3380
Se	0.1490	0.0880	0.1513	0.2083	0.2593	0.2833	0.3030	0.3193
Psyn	0.3021	0.2138	0.2934	0.3774	0.4805	0.5380	0.5701	0.5984

Table 5. PSR of Pb, Cd, Hg, As, Cu, Cr and Se in peanut (%).

Type of element	$K_{i\,(mean)}$	$K_{i\,(25)}$	$K_{i\,(50)}$	$K_{i\,(75)}$	$K_{i\,(90)}$	$K_{i\,(95)}$	$K_{i\,(97.5)}$	$K_{i\,(99)}$
Pb	13.24%	10.17%	9.33%	13.40%	17.86%	19.33%	19.71%	21.56%
Cd	11.47%	10.21%	10.98%	11.32%	11.18%	12.10%	13.77%	14.33%
Hg	9.13%	2.50%	8.48%	11.00%	11.90%	11.99%	11.86%	12.79%
As	4.02%	4.37%	4.39%	4.00%	3.92%	4.07%	4.30%	4.54%
Cu	36.30%	48.35%	38.85%	32.59%	29.77%	28.64%	27.34%	24.65%
Cr	12.24%	9.72%	12.87%	13.80%	12.77%	11.95%	11.42%	11.38%
Se	13.61%	14.68%	15.10%	13.89%	12.60%	11.92%	11.59%	10.75%

important pollutant in peanuts. The sum of $K_{Cu+Pb\,(mean)}$, $K_{Cu+Pb\,(P50)}$, $K_{Cu+Pb\,(P75)}$, $K_{Cu+Pb\,(P90)}$, $K_{Cu+Pb\,(P95)}$, $K_{Cu+Pb\,(P97.5)}$, and $K_{Cu+Pb(P99)}$ reached 49.54%, 58.52%, 48.18%, 45.99%, 47.63%, 47.97%, 47.05%, and 46.21%, respectively (Table 5).

3.5 Dietary exposure and risk index results

THQ method was used to evaluate dietary exposure risk of thirteen elements in peanuts. Equations 4-5 were used to evaluate the risk index of dietary exposure. The $HI_{h(mean)}$, $HI_{h(50)}$, $HI_{h(75)}$, $HI_{h(90)}$, $HI_{h(95)}$, $HI_{h(97.5)}$, and $HI_{h(99)}$ of children and $HI_{h(95)}$, $HI_{h(97.5)}$, and $HI_{h(99)}$ of adults were all greater than 1, which proved that high peanut intake would cause harm to human health, especially to children (Table 6).

Figure 5 showed that dietary exposure risk of children was two to three times than that of adults, suggesting that children were more susceptible to non-carcinogenic risk from metals. This result is in agreement with the results reported in Australia (Saha et al., 2017) and the Hong Kong Environmental Protection Department (Rajeshkumar et al., 2018). This may be related to the low body weight of children and the small difference in peanut intake compared with that of adults.

The proportion of dietary exposure (Table 7) of thirteen elements showed that dietary exposure risk of Cu accounts for the largest proportion, with the highest THQ value in each percentile, followed by Ni and Mn. The dietary exposure risk of Pb, Al, and V was low. The THQ were all smaller than 1, which proved that there was no significant health risk of exposed people. The risks associated with dietary intake of metals was $THQ_{Cu} > THQ_{Ni} > THQ_{Mn} > THQ_{As} > THQ_{Cd} > THQ_{Cr} > THQ_{Hg} > THQ_{Sb} > THQ_{Ba} > THQ_{Se} > THQ_{Pb} > THQ_{Al} > THQ_{V}$ for same population with same exposure levels.

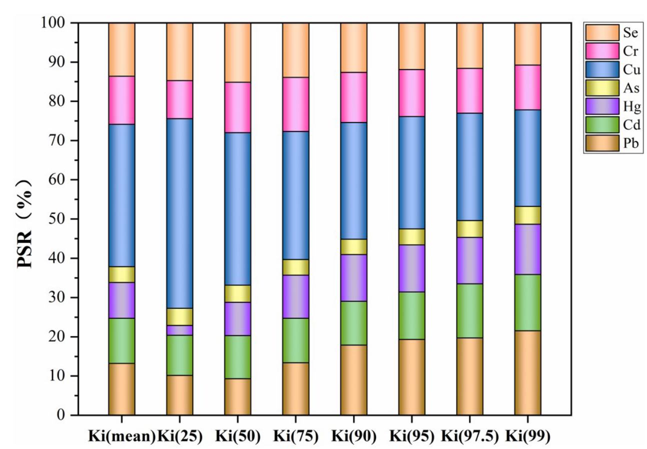

Figure 4. PSR of Pb, Cd, Hg, As, Cu, Cr and Se in peanut (%).

Table 6. Target Hazard Quotient (THQ) and Hazard Index (HI) of thirteen elements for adults and children through dietary intake of peanut.

Type of element	Population classification	THQ (mean)		THQ (25)		THQ (50)		THQ (75)		THQ (90)		THQ (95)		THQ (97.5)		THQ (99)	
		a	b	a	b	a	b	a	b	a	b	a	b	a	b	a	b
Pb	**Child**	0.0008	0.0119	0.0003	0.0050	0.0005	0.0077	0.0011	0.0165	0.0020	0.0302	0.0025	0.0378	0.0028	0.0424	0.0035	0.0527
	Adult	0.0004	0.0049	0.0002	0.0021	0.0003	0.0032	0.0006	0.0068	0.0011	0.0125	0.0013	0.0156	0.0015	0.0175	0.0018	0.0218
Cd	**Child**	0.0060	0.0904	0.0029	0.0440	0.0053	0.0792	0.0081	0.1222	0.0110	0.1655	0.0138	0.2070	0.0173	0.2589	0.0204	0.3064
	Adult	0.0032	0.0374	0.0015	0.0182	0.0028	0.0327	0.0043	0.0505	0.0058	0.0684	0.0072	0.0856	0.0091	0.1070	0.0107	0.1267
Hg	**Child**	0.0042	0.0634	0.0006	0.0095	0.0036	0.0539	0.0070	0.1046	0.0104	0.1553	0.0120	0.1807	0.0131	0.1966	0.0161	0.2409
	Adult	0.0022	0.0262	0.0003	0.0039	0.0019	0.0223	0.0037	0.0432	0.0054	0.0642	0.0063	0.0747	0.0069	0.0813	0.0084	0.0996
As	**Child**	0.0070	0.1055	0.0042	0.0628	0.0070	0.1055	0.0096	0.1439	0.0129	0.1933	0.0155	0.2322	0.0180	0.2696	0.0216	0.3234
	Adult	0.0037	0.0436	0.0022	0.0260	0.0037	0.0436	0.0050	0.0595	0.0068	0.0799	0.0081	0.0960	0.0094	0.1115	0.0113	0.1337
Cu	**Child**	0.0191	0.2861	0.0139	0.2086	0.0187	0.2802	0.0235	0.3518	0.0294	0.4409	0.0327	0.4900	0.0343	0.5142	0.0351	0.5269
	Adult	0.0100	0.1183	0.0073	0.0862	0.0098	0.1158	0.0123	0.1454	0.0154	0.1823	0.0171	0.2026	0.0180	0.2126	0.0184	0.2178
Cr	**Child**	0.0043	0.0643	0.0019	0.0280	0.0041	0.0619	0.0066	0.0993	0.0084	0.1261	0.0091	0.1363	0.0095	0.1432	0.0108	0.1622
	Adult	0.0022	0.0266	0.0010	0.0116	0.0022	0.0256	0.0035	0.0411	0.0044	0.0521	0.0048	0.0563	0.0050	0.0592	0.0057	0.0670
Se	**Child**	0.0009	0.0129	0.0005	0.0076	0.0009	0.0131	0.0012	0.0180	0.0015	0.0224	0.0016	0.0245	0.0017	0.0262	0.0018	0.0276
	Adult	0.0004	0.0053	0.0003	0.0031	0.0005	0.0054	0.0006	0.0074	0.0008	0.0093	0.0009	0.0101	0.0009	0.0108	0.0010	0.0114
Ni	**Child**	0.0141	0.2122	0.0026	0.0383	0.0082	0.1223	0.0238	0.3573	0.0364	0.5454	0.0437	0.6558	0.0544	0.8166	0.0627	0.9412
	Adult	0.0074	0.0877	0.0013	0.0158	0.0043	0.0506	0.0125	0.1477	0.0191	0.2255	0.0229	0.2711	0.0286	0.3376	0.0329	0.3891
Mn	**Child**	0.0136	0.2043	0.0080	0.1203	0.0128	0.1917	0.0185	0.2776	0.0233	0.3494	0.0275	0.4130	0.0295	0.4421	0.0303	0.4552
	Adult	0.0071	0.0845	0.0042	0.0497	0.0067	0.0793	0.0097	0.1148	0.0122	0.1445	0.0144	0.1707	0.0155	0.1828	0.0159	0.1882
Ba	**Child**	0.0011	0.0170	0.0005	0.0073	0.0010	0.0150	0.0016	0.0239	0.0022	0.0335	0.0025	0.0382	0.0030	0.0449	0.0034	0.0517
	Adult	0.0006	0.0070	0.0003	0.0030	0.0005	0.0062	0.0008	0.0099	0.0012	0.0138	0.0013	0.0158	0.0016	0.0186	0.0018	0.0214
Al	**Child**	0.0004	0.0061	0.0003	0.0039	0.0004	0.0053	0.0005	0.0079	0.0007	0.0106	0.0009	0.0128	0.0010	0.0143	0.0011	0.0161
	Adult	0.0002	0.0025	0.0001	0.0016	0.0002	0.0022	0.0003	0.0033	0.0004	0.0044	0.0004	0.0053	0.0005	0.0059	0.0006	0.0067
V	**Child**	0.0000	0.0004	0.0000	0.0000	0.0000	0.0003	0.0000	0.0006	0.0001	0.0010	0.0001	0.0011	0.0001	0.0012	0.0001	0.0014
	Adult	0.0000	0.0002	0.0000	0.0000	0.0000	0.0001	0.0000	0.0002	0.0000	0.0004	0.0000	0.0005	0.0000	0.0005	0.0000	0.0006
Sb	**Child**	0.0015	0.0874	0.0010	0.0072	0.0010	0.0738	0.0010	0.1274	0.0035	0.2285	0.0042	0.2548	0.0055	0.2753	0.0062	0.3163
	Adult	0.0008	0.0091	0.0005	0.0060	0.0005	0.0060	0.0005	0.0060	0.0018	0.0216	0.0022	0.0262	0.0029	0.0339	0.0032	0.0384
HI	**Child**	0.0731	1.1620	0.0367	0.5426	0.0634	1.0099	0.1025	1.6510	0.1417	2.3023	0.1662	2.6841	0.1902	3.0455	0.2132	3.4220
	Adult	0.0383	0.4533	0.0192	0.2273	0.0332	0.3930	0.0538	0.6358	0.0743	0.8789	0.0872	1.0305	0.0997	1.1791	0.1119	1.3222

Note: a = general intake population; b = high intake population.

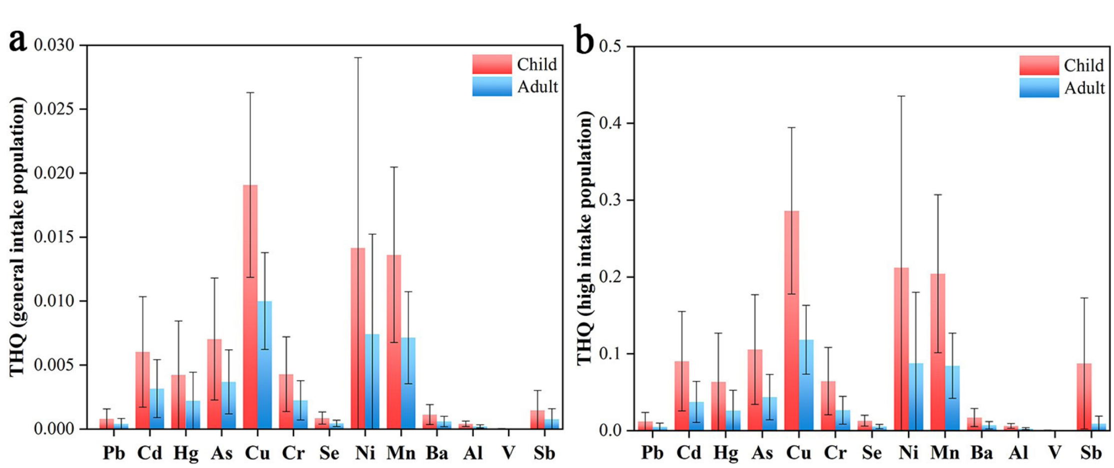

Figure 5. Estimated THQ for dietary intake of peanuts by adults and children. Note: a = general intake population; b = high intake population.

Table 7. Proportion of dietary exposure of thirteen elements.

Element	$THQ_{(mean)}$	$THQ_{(25)}$	$THQ_{(50)}$	$THQ_{(75)}$	$THQ_{(90)}$	$THQ_{(95)}$	$THQ_{(97.5)}$	$THQ_{(99)}$
Pb	1.09%	0.91%	0.81%	1.07%	1.42%	1.52%	1.49%	1.65%
Cd	8.24%	8.01%	8.33%	7.95%	7.79%	8.30%	9.08%	9.58%
Hg	5.78%	1.73%	5.67%	6.80%	7.31%	7.25%	6.89%	7.53%
As	9.63%	11.43%	11.10%	9.36%	9.09%	9.32%	9.45%	10.11%
Cu	26.09%	37.95%	29.48%	22.87%	20.74%	19.66%	18.03%	16.47%
Cr	5.86%	5.09%	6.51%	6.46%	5.93%	5.47%	5.02%	5.07%
Se	1.17%	1.38%	1.37%	1.17%	1.05%	0.98%	0.92%	0.86%
Ni	19.36%	6.97%	12.87%	23.23%	25.65%	26.31%	28.63%	29.43%
Mn	18.63%	21.88%	20.17%	18.05%	16.44%	16.57%	15.50%	14.23%
Ba	1.55%	1.32%	1.57%	1.55%	1.57%	1.53%	1.58%	1.62%
Al	0.56%	0.70%	0.56%	0.51%	0.50%	0.51%	0.50%	0.50%
V	0.04%	0.01%	0.03%	0.04%	0.05%	0.05%	0.04%	0.04%
Sb	2.00%	2.62%	1.51%	0.94%	2.45%	2.54%	2.88%	2.90%

4 Conclusion

In this study, we conducted quantitative risk assessment of fifteen elements in peanuts. This study is one of the few studies involving risk assessment of multiple element pollution in peanuts in Shandong, China. Fifteen elements were detected on the 620 samples, and the average values of fifteen elements in descending order were Mn (19.8726 mg·kg^{-1}) > Cu (7.9508 mg·kg^{-1}) > Li (6.7757 mg·kg^{-1}) > Al (4.2674 mg·kg^{-1}) > Ni (2.9493 mg·kg^{-1}) > Ba (2.3597 mg·kg^{-1}) > Cr (0.1340 mg·kg^{-1}) > Cd (0.0628 mg·kg^{-1}) > Se (0.0447 mg·kg^{-1}) > Pb (0.0290 mg·kg^{-1}) > Sn (0.0278 mg·kg^{-1}) > V (0.0278 mg·kg^{-1}) > As (0.0220 mg·kg^{-1}) > Sb (0.0061 mg·kg^{-1}) > Hg (0.0020 mg·kg^{-1}). The results of SPIM and NPIM noted that $P_{Cu(90)}$, $P_{Cu(95)}$, $P_{Cu(97.5)}$, $P_{Cu(99)}$, and $P_{Pb(99)}$ were all higher than 0.6, which showed that the product was in a lightly polluted state on Cu and Pb. Whether adults or children, the order of risk exposure caused by eating peanuts was $THQ_{Cu} > THQ_{Ni} > THQ_{Mn} > THQ_{As} > THQ_{Cd} > THQ_{Cr} > THQ_{Hg} > THQ_{Sb} > THQ_{Ba} > THQ_{Se} > THQ_{Pb} > THQ_{Al} > THQ_{V}$ for same population with same exposure levels. It also shows that children were more vulnerable to elemental pollution, as dietary exposure risk of children was higher than adults. $HI_{h(mean)}$, $HI_{h(50)}$, $HI_{h(75)}$, $HI_{h(90)}$, $HI_{h(95)}$, $HI_{h(97.5)}$, *and* $HI_{h(99)}$ of children and $HI_{h(95)}$, $HI_{h(97.5)}$, *and* $HI_{h(99)}$ of adults were all greater than 1, which proved that high peanut intake would cause harm to human health, especially to children. Therefore, strategies and interventions to reduce elements contamination should be subject of the future studies. We believe that our manuscript could provide effective preventive and control measures for production, storage, transportation, and marketing, and then provided a scientific basis for the revision of local peanut standards and production, distribution regulation and risk warning. Our study also pinpointed the importance for continuously monitoring and conduct a food consumption survey.

Conflict of interest

No conflict of interest associated with this work.

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