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DISTRIBUTION CHARACTERISTICS OF SELENIUM AND HEAVY METAL ELEMENTS IN SOIL-CROP SYSTEM IN ANKANG, SHAANXI, CHINA

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Abstract

A total of 229 groups of root vegetables, rice, corn and their root soils were collected in Ankang, Shaanxi, to analyze the characteristics of element migration and transformation in crops and root soil, to compare the migration and enrichment capabilities of various elements in different crops, to explain the characteristics of root soil pH and element correlation, and provide a theoretical basis for agricultural development. Results showed that there were differences in the ability of crops to accumulate various elements in the soil. The absorption capacity of Se, Cd and Zn elements was strong, while the bioaccumulation ability of As, Pb and Cr elements was weak. The contents of Cr, As and Hg were the highest in rice, which were significantly higher than those in root vegetables and corn. The contents of Cd, Cu and Pb in root vegetables were the highest, significantly higher than those in rice and corn. The content of various elements in crops and root soil had a different intensity correlation. Generally speaking, the content of various elements in crops increased with the increase of root soil element content to a certain extent. There was a negative correlation between pH and soil elements, and there was a significant positive correlation between the elements in the root soil of different crops.

Introduction

The transfer and transformation of elements in cropland soil habitat refers to the process that crops absorb and accumulate various forms of elements from soil through root system, and then enrich into crops. The bioavailability, migration and transformation ability of elements in cropland soil habitat were related to the total content of elements, soil physical and chemical properties, elemental forms, crop varieties, coordination and antagonism of elements (Wang *et al.* 2001, Zhong *et al.* 2009, Zhang *et al.* 2011). The change of soil physical and chemical properties will have some effects on the migration and enrichment of elements, for example, the influence of soil pH is widely concerned (Yang *et al.* 2005, Wang *et al.* 2010). pH is an important physical and chemical index of crop root soil, which can affect the migration, transformation and effective activity of all elements in root soil (Lian 2015). Soil pH affects the solubility of soil minerals, which is closely related to the content of available nutrients in soil. It is important to analyze the correlation between pH and soil elements. In recent years, the Shaanxi Geological Survey Institute has carried out multi-scale Regional geochemistry surveys and assessments in the Ankang region at scales of 1:50,000 and 1:10,000, with the aim of ascertaining the soil environmental status of agricultural land, it provides a scientific theoretical basis for the optimization of agricultural planting structure, the promotion of land use value and the development of healthy geology.

Ankang is located in the southeast of Shaanxi province, between N 31°42′ ～ 33°50′and E 108°00′ ～ 110°12′. It is located in the junction of Shaanxi, Sichuan, Hubei and Chongqing. It is a subtropical continental monsoon with four distinct seasons and abundant rainfall. Soil types

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include yellow cinnamon soil, yellow brown soil, brown soil, and so on. The main crops are rice, corn, rape, and so on. The leading crops of agricultural development are konjac, potato, tea and so on (Zhang *et al.* 2021). Ankang is a typical selenium-rich area in Shaanxi. It is known as the "Selenium Valley of China" and is affected by secondary enrichment of elements during soil formation, the content of Se and Cd in soil is higher than that in other soil-forming parent materials, and has high background attributes of Se and Cd. The present study was undertaken to investigate the distribution characteristics of selenium and other heavy metal elements in soil-crop systems in Ankang, Shaanxi, China. Based on the study of the element content and enrichment characteristics of crops and their root soil, this paper evaluated the characteristics and safety of selenium-rich crops, rationally utilized the investigation results, explored the planting suitability of crops and the development of characteristic agricultural products, and provided basic geochemical data for land development and utilization and agricultural planting structure adjustment.

Materials and Methods

From June to August 2020, 229 edible parts of crops and their root soils, including 128 root vegetables, 55 rice and 46 corn, were collected from the main growing areas of Hanyin and Ziyang in Ankang, Shaanxi. The sampling sites were mainly distributed in the areas with thick soil layers, such as farmland and vegetable fields, the sampling points are more than 150 m away from the main roads such as railway and highway.

The main types of crops collected include root vegetables (sweet potatoes, garlic, konjac, potatoes), rice and corn. When collecting crops, for rice and corn, on the basis of field investigation of farmland area and yield, and combining the characteristics of crop planting density and grain size, the plots were divided into zones, and then the sampling points were selected, after air-drying, the samples were divided into four parts, and about 500 g of samples were obtained. For root vegetables, the number of samples was determined according to the area of plots and the growth of crops, and 5-10 representative plants were used to collect the roots of crops. At the point where the crops were collected, the corresponding root soil was collected. When collecting root soil, litter, animal remains, gravels and aggregates in the topsoil were discarded, and 0-20 cm depth soil was collected vertically, after air-drying, passed through 20 mesh size nylon screen, say to take 300 g samples submitted to the laboratory for analysis and testing. Both crop and root soil samples were tested by Shaanxi Experimental Institute of Geology and Mineral Resources. Arsenic, Hg, Se in soil samples were analyzed by atomic fluorescence spectrometry (AFS), Cd by Inductively coupled plasma mass spectrometry (ICP MS) and Cr, Cu, Ni, Pb, Zn by X-ray fluorescence spectrometry (XRF). As, Cd, Cr, Pb and Se of crop samples were determined by inductively coupled plasma mass spectrometry (ICP MS), and Hg of crop samples were determined by primordium fluorescence spectrometry (AFS). The report rate of each element is 100%, and the qualified rate of accuracy and precision of analysis element is 100%. The detection limit and analytical quality of the selected analytical methods meet the requirements of the standard of 'Code for evaluation of land quality geochemistry' (DZ/T0295-2016).

Data were analyzed by Excel 2016 and SPSS 17.0, and the differences of element contents in crops were compared by Duncan method, the plot of element content in crops and the plot of correlation analysis of pH and elements in root soil were drawn by R software.

Results and Discussion

The statistical parameters of the contents of various elements in root soil of different crops are presented in Table 1. The average contents of Se, Cd, Cr, Ni, Cu, Zn, As, Pb and Hg in different crop root soils were 1.56 ± 2.17, 1.19 ± 1.43, 101.01 ± 38.39, 66.72 ± 49.59, 57.50 ± 38.91, 163.99 ± 123.43, 17.03 ± 13.47, 25.47 ± 5.79 and 0.12 ± 0.11 mg/kg, respectively. The coefficient of variation (C.V) can reflect the dispersion degree of each element in the farmland soil, and can also quantify the variation degree of each element content in the soil in the spatial distribution. Coefficient of variation ≤ 10% belongs to weak variability, 10% < C.V < 100% belongs to medium variability, C.V ≥ 100% belongs to strong variability (Li *et al*. 2008). The coefficients of variation of each element content were Se > Cd > Hg > As > Zn > Ni > Cu > Cr > Pb. The coefficients of variation of Se and Cd in different crop root soils were relatively large, which were 139.10 and 120.05%, respectively, reaching strong spatial variability, indicating that Se and Cd contents in different sampling sites were highly dispersed and differentiated. The coefficient of variation of other elements ranged from 22.72 to 91.67%, and the element content dispersion was small, which belonged to the medium spatial variability.

Table 1. Contents of various elements in root soil of different crops.

Crop root soil		Se	Cd	Cr	Ni	Cu	Zn	As	Pb	Hg
					mg/kg					
Root vegetables	Mean value	1.66	1.28	104.63	69.86	59.80	167.74	17.56	26.44	0.12
	Standard deviation	2.48	1.54	41.99	55.14	40.58	99.65	14.63	6.31	0.10
	Coefficient of variation %	149.00	119.63	40.13	78.93	67.86	59.41	83.30	23.88	83.33
Rice	Mean value	1.28	0.87	90.16	51.65	45.26	126.59	12.70	24.15	0.11
	Standard deviation	1.60	1.04	24.65	24.90	24.58	58.93	6.82	3.73	0.05
	Coefficient of variation %	124.39	119.90	27.34	48.21	54.30	46.55	53.69	15.44	45.45
Corn	Mean value	1.62	1.32	103.92	76.00	65.75	198.27	20.72	24.37	0.13
	Standard deviation	1.80	1.46	39.00	51.53	44.42	202.90	14.68	5.77	0.16
	Coefficient of variation %	111.51	110.16	37.53	67.80	67.56	102.33	70.86	23.70	123.08
Total	Mean value	1.56	1.19	101.01	66.72	57.50	163.99	17.03	25.47	0.12
	Standard deviation	2.17	1.43	38.39	49.59	38.91	123.43	13.47	5.79	0.11
	Coefficient of variation %	139.10	120.05	38.01	74.33	67.68	75.27	79.11	22.72	91.67

Results showed that Se content of different crops has no significant difference, while Cd and As content had significant difference (Fig. 1). The contents of Cr, As and Hg were the highest in rice, which were significantly higher than those in root vegetables and corn. The contents of Cd, Cu and Pb in root vegetables were the highest, which were significantly higher than those in rice and corn. The contents of Cd, Cr, As and Hg in rice were higher than those in corn, while the contents of other elements in rice and corn were not significantly different. Results further showed that there were some differences in the enrichment ability of each element among different crops. In the process of transporting elements from roots to the upper part of crops, multiple factors such as migration and transformation ability of different crops, transport distance, root microecological environment, and hydrogeological conditions may affect the elements in farmland soil (Li *et al*. 2006, Li *et al*. 2018).

The selenium enrichment ability of crops was evaluated on the basis of selenium enrichment standard. The standard for selenium content of selenium-rich selenium-containing food and related products (DB 61/T 556-2018) stipulated that the selenium enrichment standard for grains was ≥ 0.05 mg/kg, and the selenium content standard was 0.010-0.045 mg/kg. The selenium enrichment standard of vegetables was ≥ 0.02 mg/kg, and the selenium content standard was 0.010 ~ 0.015

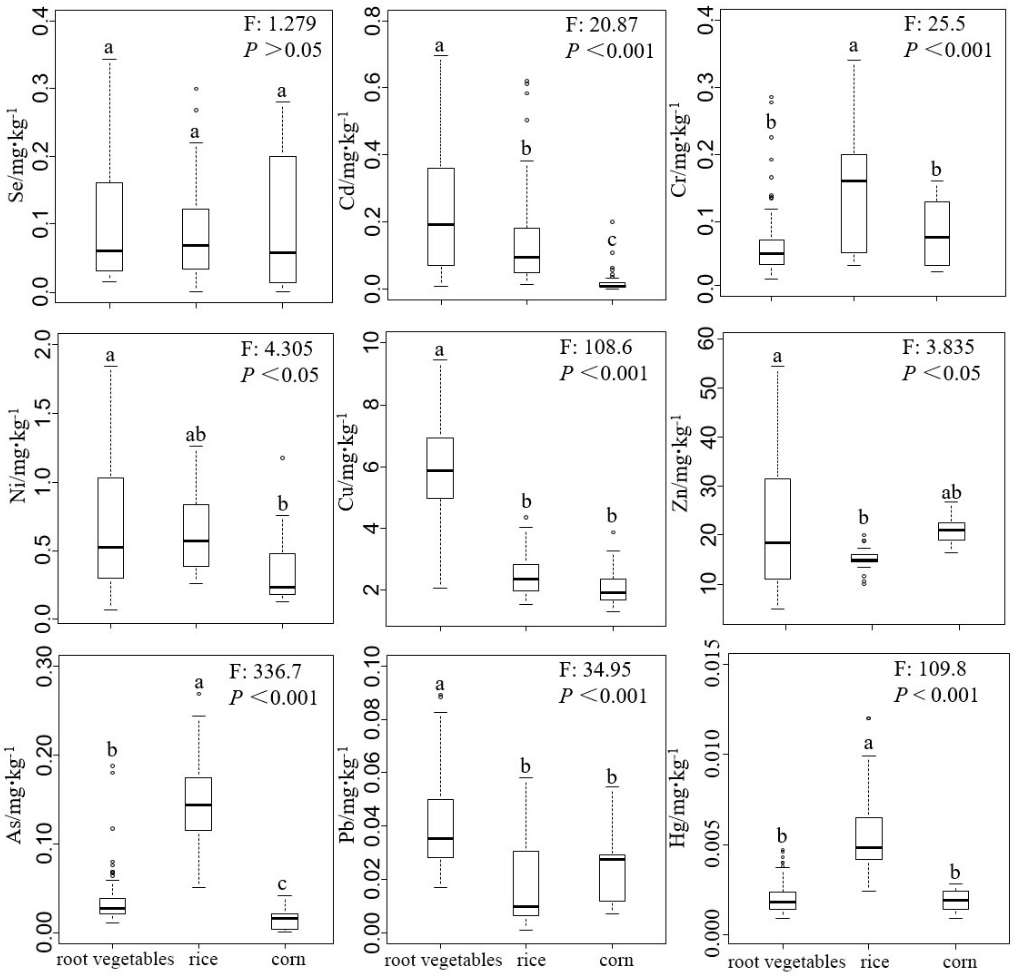

Fig. 1. Box plot of element content in different crops.

mg/kg. The selenium enrichment rate of root vegetables was the highest, and the selenium enrichment rate of rice and corn was similar. The sum of selenium enrichment rate and selenium content rate of root vegetables, rice and corn are 94.53, 96.37 and 73.92%, respectively (Table 2).

The correlation analysis of element contents in root soil-crop system presented in Table 3 showed that the content of each element in different crops and root soil has the correlation of different strength. There was no correlation between Cu and Hg in root soil of different crops, and no correlation between Cr, Cu, As and Hg in root soil-crop systems of rice and corn. The reason for this result might be that the sensitivity of crops to the above elements is low, the absorption and enrichment ability is weak, or the above elements are stable in the soil, and the crops absorb less of them. As a whole, the content of elements in corn has a low correlation with the root soil, which might be due to the weak enrichment of soil elements by corn itself. The contents of Cr, As and Pb in the root soil-root vegetable system were positively correlated (0.295, 0.292, 0.383, respectively) ($P < 0.001$). The contents of Se, Cd and Zn in rice and its root soil were significantly

Table 2. Selenium-enriched ratio characteristics of crops.

Crop type	Total sample count	Number of samples enriched with selenium	Selenium enrichment rate (%)	Number of samples containing selenium	Selenium content rate%
Root vegetables	128	119	92.97	2	1.56
Rice	55	30	54.55	23	41.82
Corn	46	25	54.35	9	19.57

Table 3. Correlation analysis of various elements in root soil-crop system.

			Root soil								
			Se	Cd	Cr	Ni	Cu	Zn	As	Pb	Hg
Crop	Root vegeta-bles	Se	0.098	0.174*	0.248**	0.245**	0.261**	0.242**	0.253**	0.131	0.116
		Cd	0.067	0.125	0.157	0.081	0.101	0.104	0.124	0.131	-0.025
		Cr	0.144	0.205*	0.295***	0.151	0.235**	0.187*	0.300**	0.186*	0.128
		Ni	0.057	0.052	0.040	0.106	0.062	0.072	0.076	-0.035	0.109
		Cu	0.131	-0.072	-0.100	0.029	-0.025	0.017	-0.020	-0.123	-0.011
		Zn	-0.001	0.089	0.057	0.087	0.078	0.076	0.037	0.046	-0.022
		As	0.097	0.088	0.251**	0.206*	0.314***	0.205*	0.292***	0.105	0.174*
		Pb	0.194*	0.454	0.465***	0.318***	0.348***	0.413***	0.429***	0.383***	0.328***
		Hg	-0.056	-0.013	-0.009	0.106	0.044	0.053	0.005	-0.060	0.080
	Rice	Se	0.665***	-0.121	0.383	0.437*	0.425*	0.385	0.372	0.566**	0.526**
		Cd	0.391	0.866***	0.666***	0.545**	0.497*	0.540**	0.561**	0.614**	-0.164
		Cr	0.145	0.386	0.258	0.315	0.217	0.277	0.238	0.191	0.311
		Ni	-0.108	0.168	0.042	0.047	-0.001	0.042	-0.041	0.052	0.088
		Cu	-0.032	0.189	0.015	-0.006	-0.033	0.068	0.008	0.028	-0.074
		Zn	0.306	0.702***	0.666***	0.576**	0.558**	0.643***	0.556**	0.641***	0.088
		As	0.191	0.356	0.270	0.078	0.161	0.184	0.248	0.300	-0.168
		Pb	0.532**	0.249	0.267	0.186	0.139	0.121	0.196	0.479*	0.040
		Hg	0.077	0.329	0.089	0.021	-0.063	-0.010	0.059	0.101	-0.321
	Corn	Se	0.100	0.051	0.036	0.022	0.064	0.061	0.154	0.285	-0.202
		Cd	0.189	0.490*	0.215	-0.087	0.265	-0.037	0.539**	0.616***	-0.144
		Cr	0.256	0.347	0.269	0.005	0.002	0.074	0.128	0.114	0.288
		Ni	0.490*	0.247	0.416*	0.397*	0.261	0.441*	0.288	0.127	0.013
		Cu	0.134	-0.074	-0.032	0.222	0.124	0.223	0.089	-0.138	0.345
		Zn	-0.016	0.120	0.133	-0.162	-0.235	-0.176	-0.128	-0.062	0.262
		As	0.139	0.182	0.212	0.056	0.039	-0.059	0.138	0.236	0.335
		Pb	0.020	0.043	0.067	-0.055	0.065	-0.016	0.015	0.132	-0.161
		Hg	-0.221	-0.373	-0.333	-0.310	-0.301	-0.262	-0.332	-0.193	0.253

*,**,***Significatly at 5%, 1% and 0.01% level, respectively.

positively correlated (0.665, 0.866, 0.643, respectively) ($P < 0.001$), and the contents of Pb was correlated ($P < 0.05$). The contents of Cd and Ni in corn and its root soil were correlated ($P < 0.05$). The elements Cr, Cu, Zn and As in root soil of root vegetables are positively correlated with

Se, Cr, As and Pb in root vegetables, and the elements Cd, Cr, Ni, Cu, Zn, As and Pb in root soil of rice are positively correlated with the elements Cd and Zn in rice, which may indicate that the chemical properties of these elements are similar or homologous. In general, the content of elements in crops increased with the content of elements in root soil to a certain extent, there is element inheritance, but it is mainly determined by the total content and the effective state proportion of each element in the root soil, which shows that the element content in the crop is restricted by the background soil condition (Li *et al.* 2020, Wei *et al.* 2020). In the present study, a relatively small number of crop species were collected. In later studies, more crop species, such as leafy vegetables, should be added to objectively reflect the characteristics of element migration in the root soil-crop system in Ankang.

Correlation analysis between pH and elements was conducted on the collected crop root soil samples (Fig. 2). Meanwhile, it can be seen from Table 4 that for root vegetables, pH in root soil was negatively correlated with As ($P < 0.001$), Cd ($P < 0.01$), and Se, Ni, Cu and Zn ($P < 0.05$). There was a negative correlation between pH and Cd in rice root soil ($P < 0.05$), while there was no correlation between pH and other elements in corn root soil. It shows that with the increase of soil pH, the elements in soil migrate and transform into the content of crops decreases, which is consistent with the research results of previous studies (Chen *et al.* 2021). With the decrease of soil pH, H^+ in the soil migrating to farmland increases, which accelerates the migration of soil elements from roots to crops and promotes the accumulation of elements in crops (Zeng *et al.* 2011, Chen *et al.* 2016). On the other hand, the dissolution rate of various elements adsorbed by farmland soil increases and the solubility increases, which accelerates the migration and transformation of elements in the root-soil-crop system, while the increase of pH leads to the precipitation and retention of elements (Yu *et al.* 2012). There was a significant positive correlation between heavy metal elements in the root soil of different crops, especially in the root soil of root vegetables. In the correlation between Se and heavy metal elements, there was a very significant positive correlation between Se and heavy metal elements in root soil of root vegetables and rice ($P < 0.001$). There was no correlation between Se and Pb elements in corn root soil, and there were significant correlations between Se and Hg, Se and Cr ($P < 0.05$), and extremely significant correlations between Se and other elements ($P < 0.001$).

Bioaccumulation coefficient (BCF) can be used to quantify and compare the ability of different crops to transfer and enrich various elements from farmland soil. C*crop* and C*root* soil represent the contents of each element in crop and root soil respectively, the formula is as follows:

$$\text{Bioaccumulation coefficient (BCF)} = C_{crop} / C_{root\ soil}$$

Crop varieties will have a certain impact on the absorption process of different elements. Simply focusing on the content characteristics of elements in soil and crops is likely to cause errors in the evaluation of element transfer ability. In order to more comprehensively measure the enrichment and migration ability of each element in the root soil-crop system and analyze the degree of absorption and accumulation of elements by different crops, the biological enrichment coefficients of each element were calculated (Table 4). In the present study, the bioenrichment coefficients of total crops were Se > Cd > Zn > Cu > Hg > Ni > As > Pb > Cr. Results showed that Se, Cd and Zn had strong migration ability, and their bioenrichment coefficients were 0.674, 0.199 and 0.135, respectively. Cadmium showed strong migration and transformation ability in the root soil-crop system. On the one hand, it might be because Cd element mainly exists in the form of easy migration in farmland soil, which is easy to be absorbed by crops (Cong *et al.* 2009) and the synergistic effect of various elements in the root soil-crop system may promote the migration and transformation of Cd to crops (Nan *et al.* 2002, Zhao *et al.* 2005).

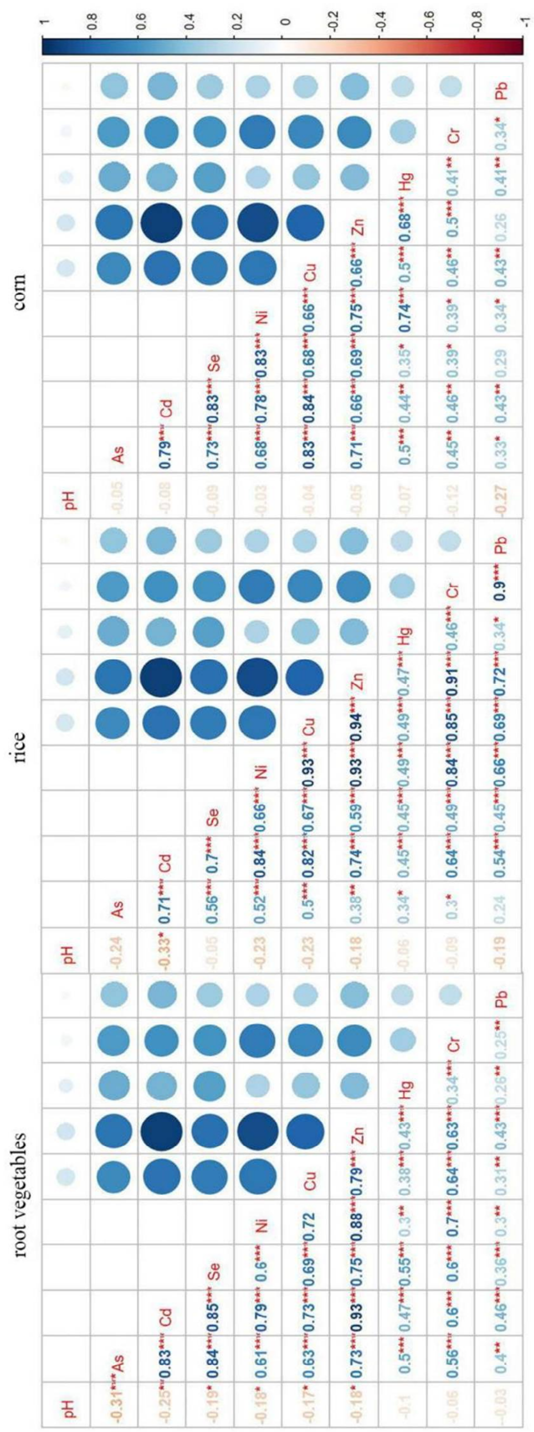

Fig. 2. Correlation analysis between pH and elements of root soil.
Figures without * indicate P > 0.05; * : P < 0.05; ** : P < 0.01; *** : P < 0.001.

Table 4. Biological enrichment coefficient of each element in root-soil-crop system.

	Se	Cd	Cr	Ni	Cu	Zn	As	Pb	Hg
Total	0.674	0.199	0.001	0.011	0.086	0.135	0.003	0.002	0.023
Root vegetables	0.831	0.222	0.001	0.012	0.100	0.142	0.002	0.002	0.017
Rice	0.145	0.210	0.001	0.013	0.056	0.120	0.012	0.001	0.059
Corn	0.276	0.041	0.001	0.005	0.032	0.106	0.001	0.001	0.018

The bioenrichment coefficients of elements in root vegetables were Se > Cd > Zn > Cu > Hg > Ni > As ≈ Pb > Cr, indicating that root vegetables have strong absorption capacity of Se, Cd, Zn and Cu. The bioenrichment coefficients of each element in rice are Cd > Se > Zn > Hg > Cu > Ni > As > Pb ≈ Cr, indicating that rice has strong absorption capacity of Se, Cd and Zn. In addition, the absorption of Cd in root vegetables and rice was larger, and the bioenrichment coefficients were as high as 0.222 and 0.210, which are hundred times higher than those of As, Pb and Cr. Therefore, it is suggested that root vegetables and rice should be avoided for the safety of agricultural products in farmland soil with high Cd content. The bioenrichment coefficients of each element in corn (maize) were Se > Zn > Cd > Cu > Hg > Ni > As ≈ Pb ≈ Cr, indicating that the absorption capacity of Se and Zn in corn was strong. The selenium enrichment capacity of the above crops was in the order of root vegetables > corn > rice. The biological enrichment ability of root crops was stronger than that of seed crops. In conclusion, considering the distribution characteristics of soil elements in crop root system, it is suggested that priority should be given to planting corn with low cadmium uptake and high selenium enrichment to produce safe selenium-enriched agricultural products.

The bioenrichment coefficients of Se in root vegetables, rice and corn were 0.831, 0145 and 0.276, and the corresponding bioenrichment coefficients of Zn were 0.142, 0.120 and 0.106. The bioenrichment coefficients of As, Pb and Cr were significantly lower than those of other elements. The bioenrichment coefficients of As in different crops were 0.002, 0.012 and 0.001, and the corresponding bioenrichment coefficients of Pb were 0.002, 0.001 and 0.001, respectively. The bioenrichment coefficients of Cr in the root soil-crop system were 0.001, 0.001 and 0.001, respectively. The low bioenrichment coefficients of As, Pb and Cr elements might be due to the synergistic antagonism between elements, which makes their forms stable and is not conducive to crop enrichment. It might also be noted that they exist mainly in stable form in soil, which hinders the absorption of crop roots. It might also be due to the transformation of the above elements from the active form to the stable form caused by long-term fertilization in the tillage process, which reduces the availability of the elements and makes it difficult to migrate to crops (Li 2014).

In the present study, there were differences in the ability of crops to absorb and enrich elements in root soil. There was no significant difference in selenium content in root vegetables, rice and corn, and all reached the standard of selenium enrichment to different degrees. The contents of Cr, As and Hg were the highest in rice, which were significantly higher than those in root vegetables and corn. The contents of Cd, Cu and Pb in root vegetables were the highest, significantly higher than those in rice and corn. The bioenrichment capacity of Se, Cd and Zn was strong, but the absorption and accumulation capacity of As, Pb and Cr was weak. The content of elements in crops and root soil was correlated with each other at different intensity, and the content of elements in crops increased with the increase of element content in root soil to a certain extent. There was a significant positive correlation between various elements in the root soil of different crops, and a significant negative correlation between pH of root soil of root vegetables and a variety of soil elements.

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