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Original Research

Geochemical Characteristics and Risk Assessment of Heavy Metals in Soil and Fruit of Major Blueberry Growing Areas in Guizhou Province

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Abstract

To investigate the health risk of heavy metals in soil and blueberry fruits in the blueberry growing area of Guizhou Province, the contents of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn were measured in nine blueberry parks. Using GB15618-2018, GB2762-2017, and GB2762-2012 as evaluation criteria, the Nemerow comprehensive pollution index method, potential ecological risk index method, and bio-concentration factors were used to evaluate the risk of planting soil and fruit in the producing areas. The results showed that the average values of As, Cr, Cu, Hg, Ni, and Zn in soil were higher than the background values in Guizhou Province, and 30.00%, 27.14%, 11.43%, 11.43%, 8.57%, and 7.14% of the As, Ni, Cr, Cd, Pb, and Zn values exceeded the control values, respectively. According to the Nemerow pollution index and potential ecological risk index, soil in the study area was mildly polluted, while the comprehensive potential ecological risk revealed a slight or medium pollution level. The heavy metal content in blueberry fruits in the study area did not exceed the standard and the enrichment coefficient was less than 1. The overall degree of soil pollution degree and risk was low, revealing blueberry fruit to be safe for consumption.

Keywords: blueberry, heavy metals, geochemical characteristics, risk assessment

Introduction

Blueberries are popular among consumers because of their high nutritional value and distinct flavor. In recent years, the blueberry planting area in various regions of China has expanded. Guizhou Province had the largest blueberry planting area and output in China at the end of 2020 [1], with blueberries becoming one of the main agricultural products driving the local economy. The soil environment is the foundation of agricultural development, and environmental health is inextricably linked to agricultural product quality

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and safety [2, 4]. Soil, as the growth environment of crops, has a considerable impact on the heavy metal content of crops [5, 6]. As the heavy metal content of crops directly affects human health, soil heavy metal content has become a focus of attention. Soil heavy metal content is influenced by various factors, such as elemental geological background content, element form, soil physical and chemical properties, land use type, root length, and human activities [7-13], while plant heavy metal content is influenced by the crop enrichment capacity and the major elements that make up plant metabolic processes [14-18]. Karst topography in Guizhou province is widely distributed, and studies have shown that the content of heavy metals in karst soil is higher than that in the average value across China [19]. Therefore, it is of great significance for the development of the Guizhou blueberry industry to explore the heavy metal content in Guizhou blueberries and their soil, as well as their potential ecological risks.

At present, there are many methods to evaluate the safety of soil and agricultural product pollution levels. Of these, the Nemerow comprehensive pollution index method, bio-concentration factor (BCF) method and potential ecological risk index (RI) method are the most suitable for studying the geochemical characteristics of heavy metals in basic agricultural ecosystems [20-21]. To date, the risk assessment of agricultural products has mainly involved apples, rice and other agricultural products, and aquatic products [22-26]. There have been few studies on the geochemical characteristics and risk assessment of heavy metals in blueberry soils, parent rocks, and fruits in China. Accordingly, in this study, the counties of Huangping, Ziyun, and Majiang in Guizhou Province – where blueberries are grown on a large scale – were selected as research objects. The geochemical characteristics of soil profiles and heavy metals in blueberry fruits were analyzed and the Nemerow comprehensive pollution index method, BCF method, and potential ecological RI method were adopted to comprehensively assess the risk of planting soil and blueberry fruits in the study area. This provided a reference for the large-scale planting of blueberries and the quality and safety of blueberry fruits.

Materials and Methods

Study Area

Ziyun County, located in the southwestern part of Guizhou Province, is geographically situated at 105°55'-106°29'E and 25°21'-26°3'N. It has a subtropical monsoon climate with an average annual temperature of 15.3ºC, an altitude of 623-1681 m, and average annual rainfall of 1337 mm. Majiang County is geographically located at 107°18'-107°54'E and 26°17'-26°37'N, with an elevation of 576-1862 m, a subtropical monsoon climate, an average annual temperature of 14-16ºC, and average annual rainfall of 1200-1600 mm. Huangping County is located in the southeastern part of Guizhou Province between 26º43'46''-27º14'30''N and 107º35'40''-108º12'48''E, with an elevation of 600-1200 m, a subtropical monsoon climate, an average annual temperature of 15.1ºC, and average annual rainfall of 1307.9 mm. The specific location of blueberry growing areas is shown in Fig. 1.

The dolomite of the Cambrian Loushanguan Group is mainly exposed in the Huangping growing area. The massive limestone (siliceous rock) of the Permian Wujiaping Formation, containing chert limestone, is mainly exposed in the Ziyun County plantation area. Dolomite of the Cambrian Gaotai Formation and Triassic limestone mixed with shale are mainly exposed in the planting area of Majang County.

Sampling and Testing

Through preliminary reconnaissance, nine typical soil profiles were selected in the study area, including three in each of Ziyun, Huangping, and Majjiang. After the fresh surface of each section was excavated, stratified samples were collected according to soil changes. One sample was collected at every 15 cm of each section, with each sample comprising ~500 g. The depth of each profile was determined according to the bedrock or the lowermost layer of the soil genetic horizon, with the depth of each profile varying. In total, 56 soil samples were collected from the profiles. The number of fruit and topsoil samples collected was subject to the strata and area of the study area. Topsoil samples were collected from a depth of 0-20 cm, with five topsoil samples randomly collected to form a mixed sample and 1.5 kg of each mixed sample collected in total. During collection, sundry items were removed, each sample was placed separately in a ziplock bag and transported back to the laboratory. Seventy topsoil samples were collected in total. Fruit samples from different plants of the same maturity were collected above the soil profile, with each sample being 300 g. The samples were loaded into the crisper, labeled and transported to the laboratory. A total of 90 fruits were collected.

Six pieces of Cambrian Loushanguan Group dolomite, Cambrian Gaotai Group dolomite products, and Permian Wujiaping Group massive limestone (siliceous rock) intercalated with flint limestone were taken below the profile.

After natural air drying, sundries such as weeds, roots, and gravel were removed from the soil samples, which were ground through a 200-mesh screen. The blueberries were washed and pounded. Rocks were washed with non-ionic water, dried, and ground through a 200-mesh screen. The organic matter content was determined by the potassium dichromate volumetric method with oil bath heating. Here, organic matter in the soil was disintegrated with potassium dichromate and then titrated back with ferrous sulfate. The content of clay was determined by the hydrometer

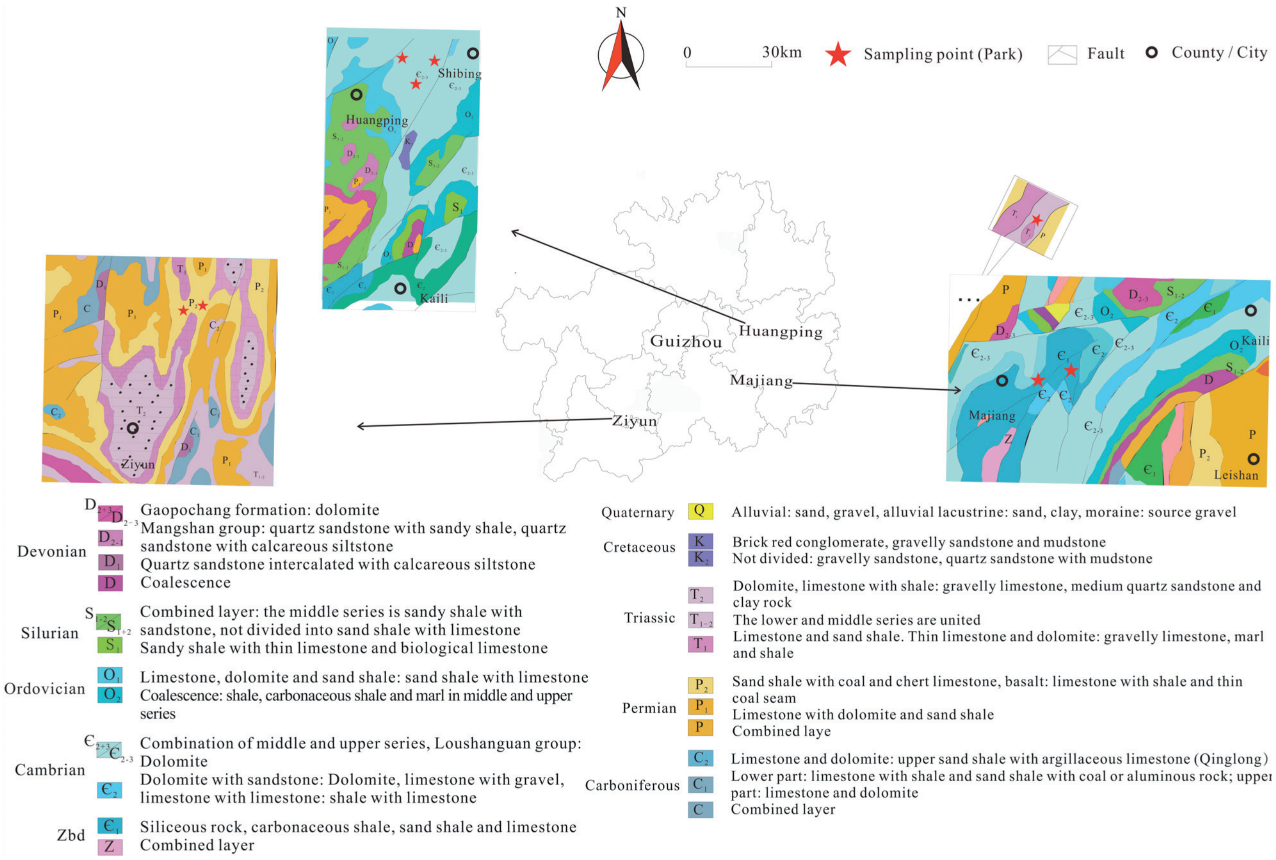

Fig. 1. Map of Guizhou blueberry production areas (Modified from Li et al.[25]).

method. Here, sodium hydroxide was selected as the dispersant according to the pH value of the soil. The soil was soaked with sodium hydroxide, passed through a 0.01 mm iron sieve and the volume was adjusted to 1000 mL for hydrometer analysis. The soil, rock, and blueberry samples were digested by aqua regia and the total content of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn was determined by inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma emission spectrometry (ICP-AES). Quality control was performed by inserting standard substances and blank samples simultaneously. The elemental analysis of all samples was performed by Aoshi Analytical Testing Co., Ltd. (Guangzhou, China) and the relative deviations (RDs) and relative errors (REs) were controlled to be less than 10%. The recoveries of the standard samples ranged from 90% to 110%, showing the methods to be reliable. Excel and SPSS 25.0 were used for statistical analysis and processing, and Origin 2019 and CorelDRAW 2018 were used for plotting.

Single-Factor Index Method and Nemerow Comprehensive Pollution Index

Based on the draft Environmental Quality-Soil Pollution Risk Control Standard for Agricultural Land (GB15618-2018), the total content of heavy metals in soil were assessed by the single-factor index method and the Nemerow comprehensive index method [28, 29].

The single-factor index was calculated as follows:

$$P_i = C_i / C_s \tag{1}$$

Here, P_i is the superstandard index of element *i*, C_i is the total amount of element i, and C_s is the soil pollution risk control value of element *i* in agricultural land. The Nemerow comprehensive index formula is as follows:

$$PN = \sqrt{\frac{{P_{imax}}^2 + {P_{iave}}^2}{2}} \tag{2}$$

Here, *PN* is the Nemerow pollution index, P_{iave} is the mean value of the single-factor index and P_{imax} is the maximum value of the single-factor index.

Potential Ecological Risk Index Method

The evaluation of heavy metals in a study area using the Nemerow comprehensive pollution index method is influenced by the heavy metal content in the research area. Accordingly, Hakanson proposed the potential ecological risk index method [30], which is calculated as follows [21, 31]:

$$Er = T_r \times C_s / C_{ref} \tag{3}$$

$$RI = \Sigma_f^m E_r \quad (4)$$

Here, E_r represents the potential ecological risk index of a single metal, T_r represents the toxicity coefficient of a heavy metal, RI is the potential ecological risk index of comprehensive heavy metal pollution, C_s represents the measured total concentration of heavy metals, and *Cref* represents the background values of soil elements in Guizhou Province. The toxicity coefficients of heavy metals As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn were 10, 30, 2, 5, 40, 5, 5, and 1, respectively [32]. The ecological risk classification of these heavy metals is shown in Table 1 [20, 21].

Cs represents the total amount of heavy metals measured.

Bio-Enrichment Factor (BCF)

The BCF indicates the ratio of the content of an element absorbed by a plant from the ground to the content of that element in the ground. The BCF of heavy metal elements in soil for blueberries is calculated as follows [30, 33]:

$$\text{BCF}=\frac{C_B}{C_S} \quad (5)$$

Here, C_B is the total concentration of an element in blueberry fruit and C_S is the total concentration of an element in soil.

Leaching Coefficients

The degree of weathering and leaching of soil elements is measured by leaching coefficients. Since Al is inert in elemental migration, it is often assumed that it does not leach. The leaching coefficient formula is as follows [34]:

$$t = \frac{(t_1 - t_2)}{2} \times 100\% \quad (6)$$

$$t_2 = \frac{t^1 \times Al_2O_{3\,rock}}{Al_2O_{3\,soil}} \times 100 \quad (7)$$

Here, t is the leaching coefficient, t_1 is the elemental content in rock, t_2 is the elemental content in soil under the assumption that aluminum does not move, and t^1 is the elemental content in soil.

Chemical Index of Alteration (CIA)

The CIA can provide a good indicator of the weathering of a soil profile and is often used to evaluate the degree of soil weathering. The higher the CIA value, the more intense and the higher the degree of soil weathering. The CIA is calculated as follows [35]:

$$I_{CIA} = W_{Al_2O_3} / (W_{Al_2O_3} + W_{K_2O} + W_{CaO^*} + W_{Na_2O}) \quad (8)$$

In this formula, the mole fraction of each component is used, with CaO* representing the mole content of silicate minerals. A CIA value of between 0.75 and 1 indicates that the soil has experienced strong weathering. A CIA value of between 0.65 and 0.75 indicates that the soil has experienced moderate weathering. A CIA value of between 0.50 and 0.65 indicates that the soil has undergone low-intensity weathering.

Health Risk Assessment

To assess the potential risk from the regular consumption of blueberry fruit, the health risk of blueberry fruit was assessed using the temporary tolerable weekly intake (%PTWI) and target hazard quotient (THQ) based on Arvay et al. [36]. The %PTWI was calculated as follows.

$$\%\text{PTWI} = \frac{C_b \times \text{intake}}{\text{PTWI}} \quad (9)$$

The intake was calculated by referring to the annual per capita consumption of fresh melons and fruits in the China Statistical Yearbook 2021 [37], which was 60.1 kg. Accordingly, the intake was calculated as 1.17 kg/week, with an average daily consumption (*ADC*) of 164.66 g/day and PTWI of 0.28 mg/adult.

The THQ predicts the ratio of toxic element exposure to its maximum threshold of no adverse health effects and is calculated as follows.

Table 1. Ecological risk of heavy metals.

Er	Single ecological risk level	RI	Potential ecological risk level
Er<40	Low	RI≤150	Low
40≤Er<80	Moderate	150≤RI<300	Moderate
80≤Er<160	Considerable	300≤RI<600	Considerable
160≤Er<320	High	600≤RI<1200	High
320≤Er	Very high	1200≤RI	Very high

$$\mathrm{THQ} = \frac{EFr \times ED \times ADC \times C_E}{RfDo \times BW \times ATn} \times 100\% \quad (10)$$

Here, *EFr* is the frequency of exposure (365 days), *ED* is the duration of exposure (70 years), *ADC* is the average daily consumption (0.17 g/d), *CE* the average content of Hg in blueberry fruit, *RfDo* is the oral reference dose of Hg (0.0003 mg/kg/day), *BW* is the average adult body weight (70 kg), and ATn is the average duration of exposure (365 days × 70 years = 25550 days).

Results and Discussion

Physical and Chemical Properties of the Soil Profile

SOM, pH, clay content, and other basic physical and chemical properties are the main factors affecting elemental geochemical properties [38]. In the present study, the pH range of blueberry plantation soil in Majang, Ziyun and Huangping counties was 3.95-5.86, which was acidic and therefore suitable for the growth and development of blueberries. The clay content ranged from 58.25% to 91.07% according to Kaczynski's soil classification criteria [39], revealing that most of the soil in the study are was clay and that its content increased with depth. The SOM content of the blueberry plantations in the three counties ranged from 1.82 to 57.34 g/kg and decreased gradually from the topsoil downward. In combination with the soil color and root distribution records from the field survey, a soil profile was created (Fig. 2). The root distribution was concentrated from 0-45 cm.

Analysis of Heavy Metals in Soil

The total content of heavy metals in the topsoil of the blueberry gardens in Huangping, Ziyun, and Majjiang counties (Table 2) was measured and evaluated using GB15618-2018, with soil element background values from Guizhou Province as the standards. The variation coefficients of the heavy metals were indicative of the degree of variation in the heavy metal content. Generally speaking, CV≤20% indicates low variability, 20%<CV<50% indicates moderate variability, and CV≥50 indicates high variability [40]. As can be seen from Table 2, Cr, Zn, and Ni showed moderate variability, while As, Cd, Hg, Pb, and Cu showed high variability, which may have been related to the physical and chemical properties of the soil parent rock, soil pH, organic matter and texture, and root density. The content of the heavy metals was in the order Zn>Cr>Ni>Cu>As>Pb>Hg>Cd, and the average content of As, Cu, Hg, Ni, and Zn exceeded the corresponding background values [41]. The mean values of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn were 1.78, 0.22, 0.88, 1.2, 2.63, 1.17, 0.98, and 1.23 times of the background values in Guizhou, respectively.

In the study area, the proportion of the soil profiles that exceeded the heavy metal threshold for As, Ni, Cr, Cd, Pb, and Zn was 30.00%, 27.14%, 11.43%, 11.43%, 8.57%, and 7.14%, respectively. The content of Hg, Cu, and Pb was lower than the soil pollution control standard. The CIA values of topsoil in the study area ranged from 0.77 to 0.92, which were strongly weathered. The leaching coefficients of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn were –138.10 to –12.68, –0.37 to 0.00, –219.35 to 544.72, –126.07 to –7.47, –1.94 to –0.03, –178.05 to 317.70, –203.54 to 5.78, and –439.10 to –48.71, respectively. There were negative values of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn, and the order of leaching indexes was Cd>Zn>Cu>As>Hg>Cr>Pb>Ni. Ma et al. [42] studied heavy metals in agricultural soils in carbonate rock areas and found that residual heavy metals were enriched by strong weathering and leaching under alternating redox conditions. Hence, it was presumed that chemical weathering and leaching were

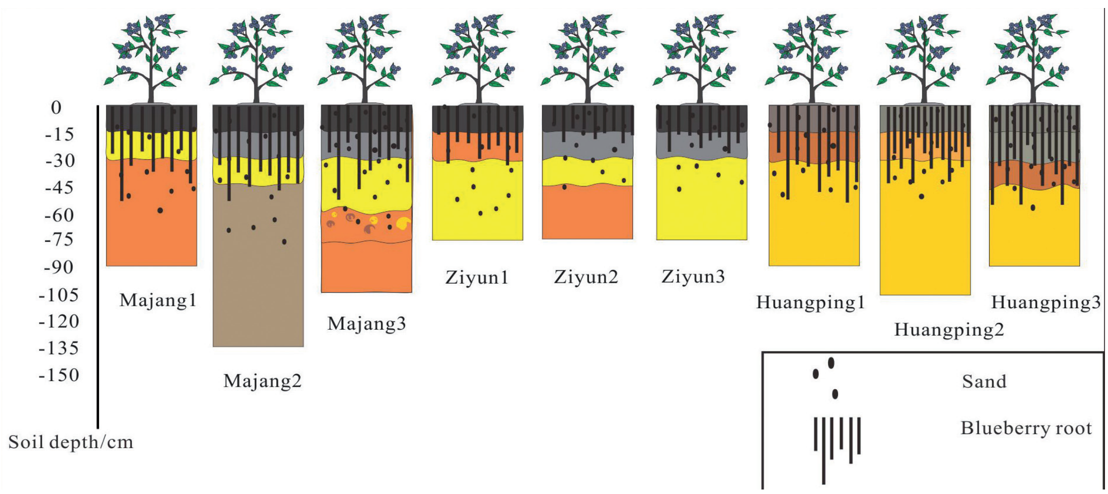

Fig. 2. Soil profile in the study area.

Table 2. Statistical results of heavy metal content in topsoil in the study area (mg/kg).

		As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
The study area (n = 56)	Ave	35.60	0.15	83.93	38.45	0.29	45.61	34.59	122.31
	Min	16.20	0.04	48.00	6.90	0.08	15.70	10.90	54.00
	Max	108.00	0.57	190.00	98.20	1.03	146.00	146.00	263.00
	C.V/%	0.44	0.84	0.44	0.59	0.65	0.59	0.69	0.40
Super control value/%		55.36	12.50	26.79	0.00	0.00	48.21	0.00	30.36
Soil elements in Guizhou The background value[41]		20	0.659	95.9	32	0.11	39.1	35.2	99.5
GB15618-2018		40	0.3	150	150	1.3	60	70	200

Note: „C.V." stands for coefficient of variation.

strong in the study area and affected the enrichment of heavy metals Cd, Zn, Cu , As , Hg, and Cr in the soil.

To further study the concentration, distribution characteristics, and characteristics of factors influencing the soil in the study area, a vertical distribution map of heavy metals in the soil profile was prepared. As shown in Fig. 3, the heavy metal content in the vertical direction sharply decreased from 0 to 30 cm, representing a deficit state, slowly increased from 30 to 90 cm, and sharply increased when the soil depth was greater than 90 cm. That is, with the increase in soil depth, there was an overall increasing trend in heavy metal content. At 0 to 30 cm, the soil in the blueberry park was acidic and most of the metal ions in the environment were hydrated. This layer contained a large amount of organic matter, and the duel effects of organic matter chelation and leaching caused a sharp loss of heavy metal content. At 30 to 90 cm, the organic matter content decreased sharply, the clay content gradually increased, and the leaching effect was stronger than in the 0 to 30 cm section; these factors resulted in a slow increase in heavy metal content with depth. Below 90 cm, where the SOM content was low, the clay content was high, and heavy metals were enriched due to leaching from the upper layers, there was a sharp increase in heavy metal content.

The results showed that there were differences in the heavy metal concentrations, elements and profiles in the blueberry orchard soils in the three regions. Ziyun had lower concentrations of As, Pb, Hg, Zn, and Ni but higher concentrations of Cr, Cu, and Cd than Huangping and Majjiang. The developing parent rocks

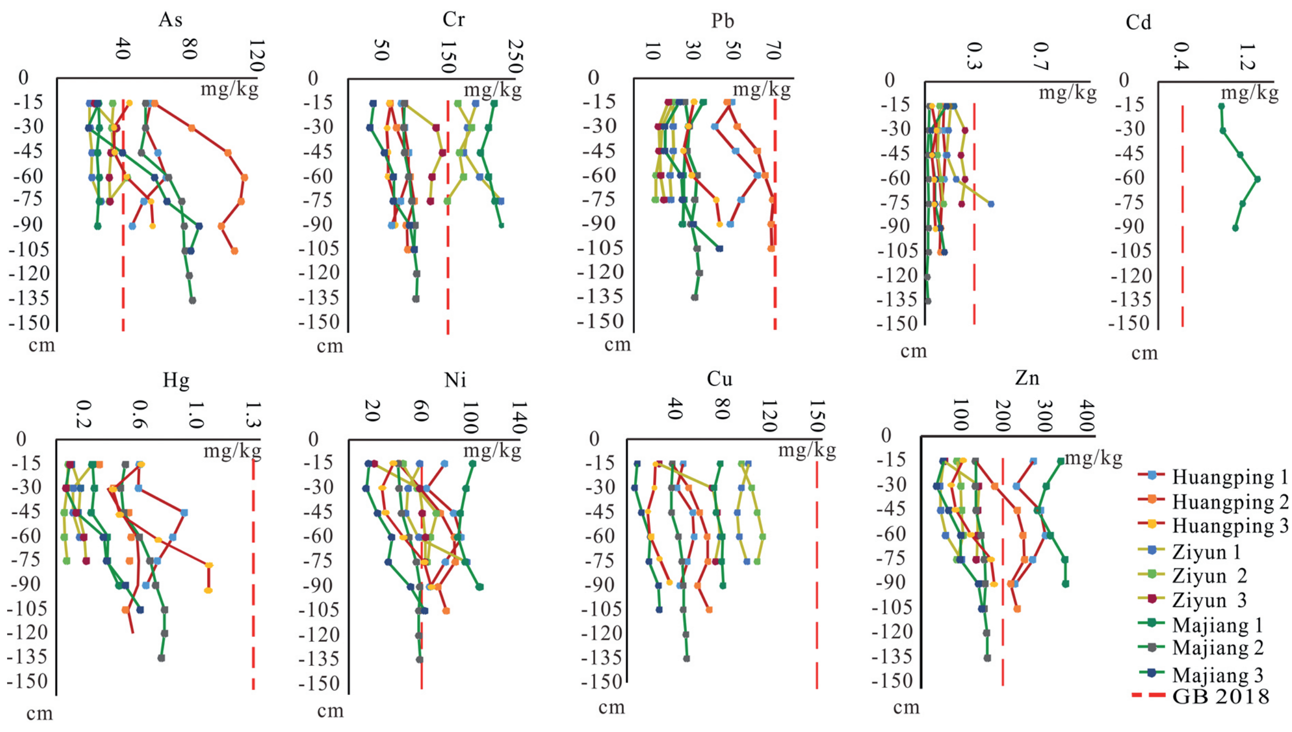

Fig. 3. Vertical distribution of heavy metal in soil profile.

of Ziyun are limestone, and the developing parent rocks of Huangping and Majiang2 and Majiang3 are dolomite on different strata. Overall, limestone contains lower concentrations of As, Pb, Hg, Zn, and Ni than dolomite, while limestone contains more Cr, Cu, and Cd than dolomite. The different stratigraphy of the three study areas was also one of the main factors for the elemental differences. In addition to Hg, Cu, and Pb, the levels of As, Cd, Cr, Ni, and Zn all exceeded the national control standards to varying degrees in the soils of the study area, with the greatest exceedance being for As. The concentration of all heavy metals in the soil of the study area, except for Cd and Hg, was higher than the background value of soil elements in Guizhou (Table 2).

The Pearson correlation coefficient is often used to measure the degree of correlation between different variables [43]. In agricultural land, the heavy metal content in the plowed layer (0-45 cm) and non-plowed layer (>45 cm) are affected by physical and chemical properties such as organic matter and clay content [44]. The root length of blueberry is generally less than 45 cm, so the soil depth of less than 45 cm and greater than 45 cm in a blueberry park can be divided into the plowed layer and the non-plowed layer. Eleven indexes – pH, SOM, clay content, and concentration of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn – in the profile soil were tested for normality using the kurtosis skewness method (J-B test). The indexes with absolute values of skewness and kurtosis less than one were selected. The correlation between the two depth segments is shown in Table 6. There was a negative correlation between clay content and Cr, Ni, Zn, SOM, and As ($P<0.05$). There was no correlation between pH and heavy metal concentrations. In the plowed layer, the correlation between physicochemical properties and heavy metals, and between heavy metals, was enhanced. The clay content was significantly positively correlated with As, Cu, and Hg concentration ($P<0.05$), and highly significantly positively correlated with Ni, Pb, and Zn concentration ($P<0.01$). There was no correlation between heavy metal concentrations and pH, and organic matter was significantly positively

Table 3. Pearson correlation analysis of heavy metal content and physicochemical properties in the arable layer.

Depth	Indicators	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
The plowed layer (<45cm)	As	1							
	Cd	0.37	1						
	Cr	0.474 *	679 * *	1					
	Cu	0.238	0.355	0.890 * *	1				
	Hg	0.682 * *	0.197	0.392	0.307	1			
	Ni	0.169	0.687 * *	0.620 * *	0.553 * *	0.303	1		
	Pb	0.800 * *	0.057	0.304	0.17	0.724 * *	0.423 *	1	
	Zn	0.337	0.626 * *	0.278	0.152	0.492 *	0.896 * *	0.597 * *	1
	SOM	0.463 *	0.084	0.217	0.158	0.326	0.2	0.231	0.325
	pH	0.061	0.009	0.186	0.206	0.271	0.126	0.111	0.076
	Clay content	0.479 *	0.06	0.281	0.493 *	0.472 *	0.638 * *	0.570 * *	0.516 * *
The non-plowed layer (>45cm)	As	1							
	Cd	0.496 *	1						
	Cr	0.629 * *	0.744 * *	1					
	Cu	0.453 *	0.298	0.753 * *	1				
	Hg	0.404	0.247	0.624 * *	0.722 * *	1			
	Ni	0.035	0.597 * *	0.537 * *	0.429 *	0.069	1		
	Pb	0.821 * *	0.274	0.570 * *	0.459 *	0.536 * *	0.27	1	
	Zn	0.162	0.589 * *	0.111	0.129	0.308	0.730 * *	0.507 *	1
	SOM	0.508 *	0.076	0.083	0.087	0.227	0.023	0.245	0.177
	pH	0.206	0.147	0.119	0.218	0.335	0.048	0.05	0.024
	Clay content	0.613 * *	0.376	0.433 *	0.217	0.540 * *	0.322	0.805 * *	0.305

Note: „**" means at the 0.01 level (double-tailed) and the correlation is significant; „*" means at the 0.05 level (double-tailed) and the correlation is significant.

correlated with As (P<0.05). In the non-plowed layer, there were both positive and negative significant correlations between heavy metals, indicating that the heavy metal content of this soil was not only influenced by the parent rock, but also by leaching and weathering. The negative correlation between some heavy metals in the plowed layer was weakened and the positive correlation was enhanced. The clay content and soil physicochemical properties had a greater influence on the content of most heavy metals, whereas the correlation between soil heavy metals was stronger than that between heavy metals and physicochemical properties, indicating that the geochemical effect of soil physicochemical properties on heavy metals in the carbonate rock areas where blueberries are grown may be less than the effect of soil-forming parent rock and the interaction between heavy metal elements, which was consistent with the results of previous studies [45-46]. Furthermore, altitude, slope, soil thickness, and topography also have a considerable influence on soil heavy metal content [47, 48]; these factors should be considered in future rese research.

The heavy metal concentrations in carbonate areas are affected by the heritability of the parent rock. Exploring the elemental content of parent rock is an important aspect of studying the heavy metal content characteristics and geochemical laws of soil. The concentration range of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in bedrock collected in the Huangping area was 1.3-3.7, 0.02-0.06, 1-15, 1.9-2.8, 0.005-0.019, 0.3-9.0, 1.4-1.7, and 2-4 mg/kg, respectively. The concentration range of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in bedrock from the Majiang area was 1.9-3.0, 0.07-0.09, 2-14, 2.0-1.9, 0.006-0.011, 4.0-8.6, 4.6-6.0, and 4-6 mg/kg, respectively. The content of bedrock As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn collected in Ziyun area was 1.0, 0.03, 1225, 12.0, 0.015, 690, 0.5, and 3 mg/kg, respectively. The heavy metal content of bedrock from the Huangping and Majiang areas was generally higher than that in the upper crust [49]: 0.65-1.85 times higher in Huangping and 0.95-1.52 times higher in Majiang. However, the Cr and Ni content in the Ziyun area was high, at 35 and 37 times higher than that in the upper crust, respectively. The high geological background of As, Cr, and Ni was the main factor underpinning the high levels of As, Cr, and Ni in soil. The Cd, Cu, Hg, Pb, and Zn content of the bedrock was low and varied between regions, as well as being lower than in the upper crust. This geologically low background was indicative of low elemental enrichment.

The content of Cd in agricultural soils in carbonate areas is generally higher than the background value [50]. The content of Cd in the present study was lower than the background value for Guizhou, and the soil Cd in most parks did not exceed the standard. The low geological background of Cd in the study area was the main reason for the low soil Cd in the study area. It was found that in the study areas of Majiang1 and Ziyun, where the parent rock was the same limestone, the Cd content in Majiang was greater than in Ziyun, and the difference in Cd content was significant (Fig. 3). Han et al. [51] found that the Cd content in Triassic Daye Group strata was 0.35 mg/kg, which is higher than other strata in this study. Therefore, it is speculated that the Cd anomaly in the Majang1 area was related to different strata. Compared with the other two Majjiang areas, there was a great difference in the Cd content in Majjiang1. In an assessment of blueberry garden soil pollution for the Majiang region, Gou et al. [52] determined that heavy metals in the region were mainly inherited from parent rocks. Zhang et al. [53] showed that agricultural activities were second only to natural factors as a source of heavy metals in agricultural land. In this study, Ziyun had less As, Pb, Hg, Zn, and Ni overall than Huangping and Majiang, and more Cr, Cu, and Cd overall than Huangping and Majiang (Fig. 3). Field investigations showed that, except for the annual application of organic fertilizer in winter, there was no application of other fertilizers in the study area, there is no industrial zone around the park, and there is relatively low agricultural activity. Therefore, based on the existing research speculation, the origin of parent rock may be the main factor underpinning the differences in the heavy metal content between the different areas.

Table 4. Heavy metal content in blueberry fruit (n = 90 mg/kg).

Element	Min	Max	Ave	GB2762-2017/2012	Super control value
As	0.0020	0.0176	0.0058	0.5	0.00%
Cd	0.0006	0.0030	0.0012	0.05	0.00%
Cr	0.0135	0.4155	0.0741	0.5	0.00%
Cu	0.3390	2.3475	0.7665	10	0.00%
Hg	0.0003	0.0026	0.0009	0.01	0.00%
Ni	0.0615	0.4035	0.2025	1	0.00%
Pb	0.0080	0.1190	0.0238	0.2	0.00%
Zn	0.6900	3.4950	1.5742	5	0.00%

Analysis of Heavy Metal Content in Blueberry Fruit

The heavy metal content in blueberry fruits in the study area is shown in Table 4. The range of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn concentrations in blueberries in the study area was 0.0020-0.0176, 0.0006-0.0030, 0.0135-0.4155, 0.3390-2.3475, 0.0003-0.0026, 0.0615-0.4035, 0.0080-0.1190, and 0.6900-3.4950 mg/kg, respectively. Thus, the average content of heavy metals in blueberries in the study area followed the order Zn>Cu>Ni>Cr>Pb>As>Cd>Hg. The content of heavy metals in the blueberry fruit was less than the contamination limit, which showed that the blueberry fruit from the study area was safe for consumption.

The BCF reflects the ability of plants to enrich nutrients from the soil and indicates the degree of anthropogenic disturbance [53]. With heavy metals as the horizontal coordinate and the logarithm of the enrichment coefficient of blueberries relative to the plowing layer soil as the horizontal coordinate, the BCF in Huangping, Ziyun, and Majjiang is shown in Figure 4. The BCF of Cd, Zn, Cu, Ni, Cr, Hg, Pb, and As was 0.302-7.105%, 0.429-4.474%, 0.735-4.872%, 0.213-2.129%, 0.038-1.953%, 0.062-1.416%, 0.049-0.338%, and 0.005-0.052%, respectively, all of which were less than 1. The BCF values were small, indicating low bioavailability and a low degree of anthropogenic interference. The BCF values in decreasing order were as follows: Cd>Zn>Cu>Ni>Cr>Hg>Pb>As. The BCF values for Hg, Pb, and As in blueberries were small and had little variation. The BCF values of Cd, Zn, and Cu in blueberries were large, and the variation in the BCF values of Cd, Cr, and Hg was large. The variation in BCF values in Ziyun was smaller than that in Huangping and Majjiang. Past studies have shown that the bioconcentration capacity of carbonate areas is low and that vegetables and fruits are more enriched in heavy metals Cu, Zn, and Cd [54]. Additionally, the enrichment capacity of Cu, Zn, and Cd is related to the soil heavy metal content and morphology, the fruit and vegetable species, and different plant tissues [55, 56].

Nemerow Comprehensive Pollution Index Results

The assessment of topsoil heavy metals in the park plays an important guiding role in the planning of agricultural planting. The analysis and assessment of soil heavy metals help with understanding the potential threat posed by soil heavy metals in an agricultural environment. In the present study, the Nemerow comprehensive pollution index method was used to comprehensively evaluate the surface soil (0-20 cm) metals in the study area. The single-factor index, Nemerow comprehensive pollution index and evaluation results are shown in Fig. 5 , below. As shown in Fig. 5a), a single-factor index of less than 1 indicates that the soil is not polluted, which was the case for most of the blueberry gardens. The maximum and minimum values of the single-factor index were for As and Cu in Majang, respectively. The single-factor index for Cu and Hg was less than 1. The single pollution index for As, Cd, Cr, Ni, Pb, and Zn in blueberry gardens was greater than 1. As shown in Fig. 5b), the Nemerow comprehensive pollution index of soil in the study area mostly showed a pattern of $PN_{\text{Majjiang}}>PN_{\text{Ziyun}}>PN_{\text{Huangping}}$. The PN in the three areas was < 2 and most of the soil was at the light pollution level. The evaluation results of the three areas showed that the degree of pollution was in the order Majjiang>Ziyun>Huangping, indicating a low degree of pollution in the study area.

Potential Ecological Risk Assessment Results

Compared to other evaluation methods, the potential ecological risk index method can eliminate the differences caused by the background values of elements. Accordingly, the integrated potential ecological risk assessment of soil samples in the study area enabled the ecological risk of the area to be determined.

As shown in Fig. 6a), in the three study areas, the potential ecological risk of most individual metals in the soils of the study area was low (Er<40). Only Hg had a low (Er<40), moderate (40≤Er<80),

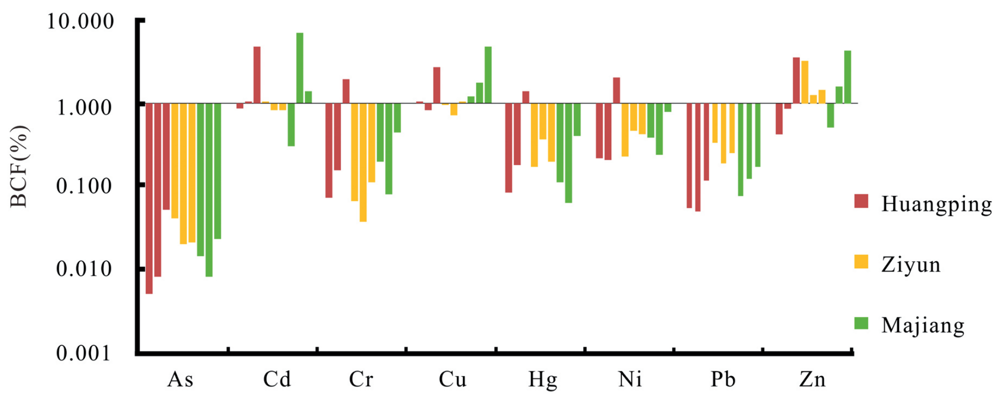

Fig. 4. Enrichment coefficient of heavy metal elements of blueberry fruits in plowed soil.

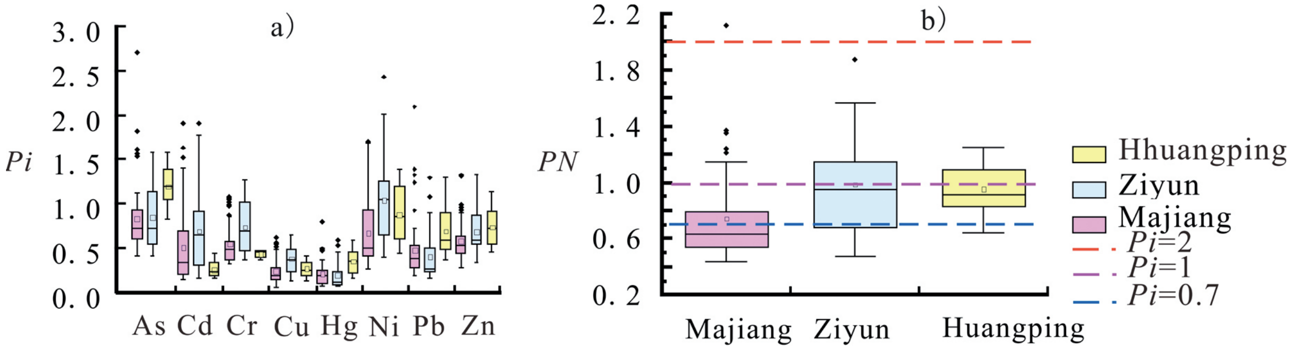

Fig. 5. Box diagram of topsoil single-factor index and Nemerow comprehensive pollution index.

considerable (80≤*Er*<160), high (160≤*Er*<320), and very high (320≤*Er*) ecological risk, and Hg as a whole was dominated by moderate, considerable, and high individual potential ecological risk hazards. As shown in Fig. 6b), in terms of mean values, the potential ecological risk of soil in Huangping and Majjiang was a moderate potential ecological risk (150<RI<300), and the potential ecological risk of Ziyun was a low potential ecological risk (RI<150). Overall, the potential ecological risk assessment results were in the order Huangping>Majjiang>Ziyun.

It was found that the degree of soil pollution and potential ecological risk level of the blueberry parks was low. In terms of the potential ecological risk of soil in the study area, Hg was the characteristic pollution factor, which was related to the large toxicity coefficient of Hg. To further clarify the Hg risk profile of fruits grown on soils with higher Hg risk, the health risk assessment of fruit Hg showed that the %PTWI and THQ of fruit Hg were 0.37% and 0.007, respectively. The %PTWI and THQ values of was <100% and <1, respectively, indicated that the Hg content of blueberry fruits in the study area would not pose a potential risk to human health based on long-term consumption and that there was no non-carcinogenic health risk.

In addition to Hg, As also had a considerable impact on the potential ecological risk index. Li et al. [50] found that high background values and low enrichment of plants occurred in soil, which was similar to the results of the present study. Consequently, the ecological benefits of heavy metals should be considered to adjust the local soil safety threshold.

The evaluation using the Nemero comprehensive pollution index method and the potential ecological risk index method both showed that the pollution level and ecological pollution risk in the study area were low. However, the evaluation results of the two methods were not the same. The restriction factors of the former were the high soil content of As, Cd, Cr, Ni, Pb, and Zn, while that of the latter was Hg. The former evaluates the pollution caused by the content of heavy metals in the soil and its results are influenced by the content of heavy metals and national control standards, while the latter also considers the toxic effects of each heavy metal on plants. Liu et al. [57] showed that Hg, Cd, Pb, Zn, and As are commonly associated with a high-risk assessment of agricultural soils in typical karst areas. Additionally, Li et al. [48] showed that there was low bio-availability of some elements to crops in karst areas, which was similar to the results of this study.

In this study, the geological background of As was relatively high, while the geological background of Cd, Cr, Cu, Hg, Ni, Pb, and Zn was relatively low. Some studies have shown that the soil elemental content of a park is influenced by the development of the parent rock, the stratigraphy, the soil formation process, and the daily management of the park [58, 59]. Apart from the application of organic fertilizer in winter, there were no other daily management measures

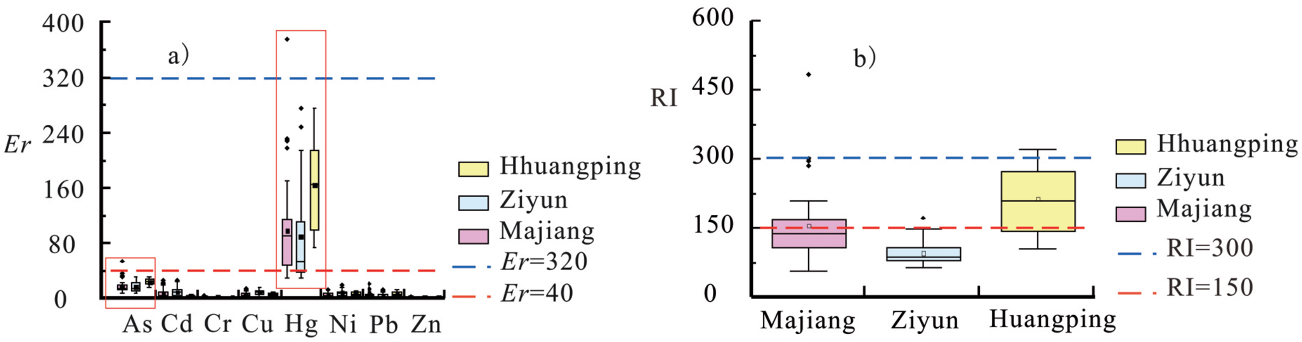

Fig. 6. Evaluation results of topsoil potential ecological risk.

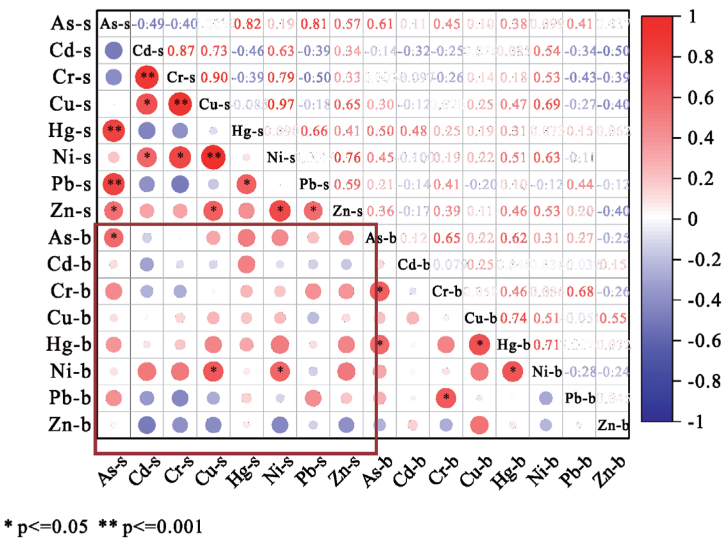

Fig. 7. Topsoil -fruit Pearson correlation diagram for heavy metals.

in the study area. As a result, the enrichment coefficient, pollution evaluation, and risk evaluation of blueberries were all low, which was speculated to be related to the low content of heavy metals in the parent rock of the study area and the few external inputs of heavy metals in the park. Gou et al. [52] showed that the potential ecological risk posed by heavy metals in the soil in the Majjiang area was relatively high. However, the results of the present study showed that the potential ecological risk in the Majjiang area was moderate. The differences in soil parent rock, soil weathering intensity, element leaching characteristics, and clay content at the sampling sites resulted in the differences in heavy metal content in the developed soils.

Additionally, it is not only high levels of heavy metals that can lead to toxic phenomena in plants. The deficiency of essential nutrients such as Zn, Cu, and Ni, which are components of numerous proteins and enzymes in plants, so deficiencies of these elements in plants can also be toxic to plants. Zn in plants is a cofactor for catalysis by many enzymes. Zn in humans is essential for protein and collagen synthesis, such that insufficient supply of the element via daily food intake can lead to diseases such as cancer, endocrine disorders, and neurological and behavioral changes in humans. Zn in plants is also involved in the Zn-Fe permeation process of Fe, so when insufficient Zn is absorbed by plants, this also affects the Fe content absorbed by plants. In calcareous and alkaline soils, the low solubility and bioavailability of Zn leads to reduced Fe uptake by plants and Fe deficiency chlorosis [18]. Ni is an important component of enzymes in the nitrogen cycle and Ni deficiency can also retard nitrogen metabolism [60]. In this study area, the content of essential heavy metals Zn, Cu, and Ni were 1.23, 1.2 and 1.17 times higher than the background values in Guizhou, respectively, but did not produce toxic effects and were within the safe range.

To further understand the relationship between fruit and soil heavy metal content, a Pearson correlation analysis was conducted for heavy metals in topsoil (0-20 cm) and fruit in the study area for each park. As shown in Fig. 7, blueberry was positively correlated with soil heavy metal content in general; fruit Pb and Zn was negatively correlated with most heavy metals in soil. Soil As was positively correlated with fruit As, soil Cu with fruit Ni, and soil Ni with fruit Ni at the 0.05 level, indicating that soil As and Ni content in the blueberry parks had some influence on the uptake of As and Ni by fruit. However, in general, there was no significant correlation between soil and fruit content of most heavy metals.

Conclusion

(1) Soil levels of As, Cu, Hg, Ni, and Zn were higher than background values, while soil levels of As, Ni, Cr, Cd, Pb, and Zn were higher than control values. In rock samples, levels of As in the Majjiang and Huangping areas were generally higher than the background values, while Cr and Ni levels in Ziyun were higher than the background values. The high geological background of As, Cr, and Ni was the main reason for the high content of As, Cr, and Ni in soil.

(2) The heavy metal levels in fruits of major blueberry parks in Guizhou did not exceed the standard, and the heavy metal enrichment coefficient of blueberry fruits was less than 1. The bioavailability of heavy metals in soil was low, indicating the fruits to be safe for consumption.

(3) The Nemerow comprehensive pollution index and potential ecological risk assessment showed that the degree of soil pollution and the potential pollution levels were low in the study area. The potential ecological hazard was medium in Majiang and Huangping and

was slight in Ziyun. The former two were limited by As, Cd, Cr, Ni, Pb, and Zn, and the latter was limited by Hg.

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Conflict of Interest

The authors declare no conflict of interest.

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