Risk Assessment and Source Analysis of Heavy Metal in Agricultural Soil of a Township in Wuxi County

Hengchang Zhang, Chuan Fu, Tingzhen Li, Bin Yan, and Yan Wu

***Abstract*—According to the total amount and morphological results of heavy metals (Cd^{2+}, Cr^{3+}, Cu^{2+}, Pb^{2+}, Zn^{2+}) in cropland in Tianyuan Township, Wuxi County, Nemerow index, potential ecological risk index and geo-accumulation index method were utilized to assess the potential risks of heavy metals in soil. The consequences made clear that the average content of 5 heavy metals in the agricultural topsoil of Tianyuan Township, Wuxi County was: Zn^{2+} (85.92 mg/kg)>Cr^{3+} (73.00 mg/kg)>Pb^{2+} (31.17 mg/kg)>Cu^{2+} (31.16 mg/kg)>Cd^{2+} (1.61 mg/kg); Cr^{3+}, Cu^{2+} and Zn^{2+} were mainly residuals, the average proportion was 91.61%, 84.48% and 72.30%, respectively. Pb^{2+} was mainly Fe/Mn oxide bound (38.24%) and residuals (49.22%), and Cd^{2+} was mainly exchangeable (22.25%) and Fe/Mn oxide bound (47.57%). Nemerow index, potential ecological risk index method and geo-accumulation index method all indicated that Cd^{2+} pollution was relatively serious, and Cr^{3+}, Cu^{2+}, Pb^{2+} and Zn^{2+} were all in a relatively clean state. Correlation analysis and principal component analysis showed that Cr^{3+} was mainly affected by natural factor soil parent material. Human pollution mainly had an effect upon Cd^{2+}, Pb^{2+} and Zn^{2+}, including traffic emissions and agricultural activities, and Cu^{2+} was jointly affected by two factors.**

***Index Terms*—Ecological risk, heavy metals, pollution assessment, source apportionment.**

I. INTRODUCTION

Soil is an important component of the earth's eco-system, its elemental content affects the growth of surface organisms. Some of the heavy metal content in the soil exceeds the standard, which not only affects the normal growth of plants, but also enters the human body through food chain and other ways, causing physiological disfunction of the human body and inducing various special physiological diseases [1]-[3]. Chongqing lies on the upper stream of the Yangtze River and the hinterland of the Three Gorges Reservoir Area. It lies in the Yangtze River Economic Belt. Tianyuan Township lies on the upper reaches of Daning River in Wuxi County, Chongqing. It is the birthplace of the Daning River. Daning River runs through two counties of Chongqing, which are Wuxi and Wushan, and is injected into the Yangtze Rver at WuxiaKou in Wushan county, with a total length of more than 250 km. Wuxi County is located at the junction of the three provinces of Chongqing, Shanxi and Hubei, lying in the Qinba Mountains. The total population of the county is 540,000, including 60,000 poor people. It is a national-level poverty-stricken county with backward infrastructure construction and industrial development in rural areas. Tianyuan township, Wuxi County, 75 km from the administrative centre, covers an area of 213km^2 and has a total population of 8302 with steep mountains, gullies and scattered land, which all indicating it is in a typical three-dimensional alpine mountain climate zone [4]. Tianyuan Township has a high level of poverty, limited land resources while its land use intensity is kept in a high level, which causing a large amount of chemical fertilizers, organic fertilizers and pesticides enter the soil, as a result, its heavy metal pollution cannot be ignored. At present, the over-standard rate of soil pollution in China's arable land reaches 19.4%, among which 82.8% are heavy metals [5], which not only affects the sustainable development of modern agriculture and social economy, but also seriously threatens the agricultural ecological environment and the safety of agricultural products [6]. There is a lack of data about eco-agricultural environment in Tianyuan township and the long-term evaluation system of agricultural development and eco-environmental effect is not perfect [7], [8]. Simply pursuing the increase of output value by using extensive development may result in a blind layout of agricultural industry, even lead to the deterioration of the environment of local ecology. Therefore, it is significant and required to investigation and assess the heavy metal pollution and ecological risk of agricultural soil in Tianyuan township, Wuxi county.

This study was based on the content and morphology of heavy metals (Cd^{2+}, Cr^{3+}, Cu^{2+}, Pb^{2+}, Zn^{2+}) in cropland of 9 villages in Tianyuan township, Wuxi county. Nemerow index, potential ecological risk index and geoaccumulation index method were utilized to assess the heavy metal pollution. Finally, Pearson correlation analysis and principal component analysis were utilized to dissect the pollution source for the purpose of understanding the quality status of local agricultural soil environment. It is expected to provide a theoretical basis for the healthy development of local agriculture and a scientific basis for the management of agricultural ecological environment in mountainous areas of the Three Gorges reservoir area.

II. MATERIALS AND METHODS

A. Sample Collection and Determination

There are villages in the study area, including Baoping(BP), Tianyuan(TY), Jilong(JL), Xinhua(XH), Zhenjiang(ZJ), Wanchun(WC), Xiangping(XP),

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Xiangyuan(XY) and Xintian(XT) village. A total of 77 samples were collected according to the grid method. Each sample was ground through a 100 mesh sieve after air drying and removing from sand and plant root, then they were conserved in a polyethylene plastic bag. The heavy metals content was digested by HF-HNO3 method and measured by inductively coupled plasma spectrometer (ICP-OES Optima 7000DV). The heavy metal form was determined by Tessier five-step continuous extraction method [9], including exchangeable state (F1), carbon. Acid salt bound state (F2), iron manganese oxide bound state (F3), organic bound state (F4) and residual state (F5).

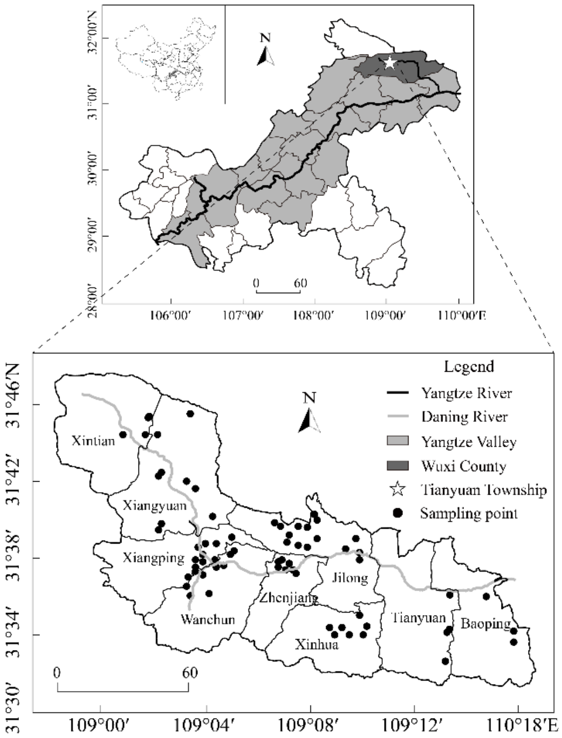

Fig. 1. Distribution of soil sampling point.

B. Evaluation Methods

1) Nemerow index

The Nemerow index can effectively estimate the pollution situation and contribution of various heavy metals [10].

(1) Single factor index

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

(2) Comprehensive factor index

$$P_i = \sqrt{\frac{\left(Max\frac{C_i}{S_i}\right)^2 + \frac{1}{n}\left(\sum_{i=1}^{n}\frac{C_i}{S_i}\right)^2}{2}} \quad (2)$$

where P_i is the pollution index of pollutant *i* in soil; C_i is the measured content of pollutant *i*, mg/kg; S_i is the control value of pollutants *i* in soil, mg/kg. In this paper, the background value of soil geochemistry in Chongqing [11] was used for calculation, as shown in Table I. *Max* is the maximum content value of heavy metals in soil; *n* is the total number of heavy metal species to be evaluated. The soil environmental quality was divided into five grades by the Nemerow comprehensive index method shown in Table II.

TABLE I: BACKGROUND VALUE OF SOIL GEOCHEMISTRY IN CHONGQING

Element	Pb	Cu	Cr	Cd	Zn
S_i (mg/kg)	26	26	80	0.11	253

TABLE II: NEMEROW COMPOSITE INDEX CLASSIFICATION

Grade	Safety	Prevention	Light pollution	Moderate pollution	Heavy pollution
Pi	*Pi*≤0.7	0.7≤*Pi*≤1.0	1.0≤*Pi*≤2.0	2.0≤*Pi*≤3.0	*Pi*>3.0

2) Potential ecological risk index

The index of potential ecological risk can integratedly take the potential impact of heavy metals into account on the ecosystem, and can be utilized to dissect soil in in large areas [12]. The relationship is as follows:

$$E_r^i = T_r^i \times P_i \quad (3)$$

$$RI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} \frac{T_r^i \times C_i}{S_i} \quad (4)$$

where *Pi* refer to (1); E_r^i is the single factor hazard coefficient; T_r^i is the toxic response coefficient of heavy metals; and *RI* is the multi-factor comprehensive potential ecological hazard index. According to the standardized toxic response coefficient of heavy metals formulated by Hakanson as the evaluation basis [13], the toxic response coefficients shown in Table III. The potential ecological risk assessment criteria is shown in Table IV.

TABLE III: TOXIC RESPONSE COEFFICIENT OF HEAVY METALS

Element	Cd	Cr	Cu	Pb	Zn
T_r^i	30	2	5	5	1

TABLE IV: POTENTIAL ECOLOGICAL RISK ASSESSMENT CRITERIA

Ecological risk coefficient	Degree of ecological risk				
	Slight	Moderate	Severe	Serious	Extremely serious
E_r^i	<40	40~80	80~160	160~320	>320
RI	<150	150~300	300~600	600~1200	>1200

TABLE V: CRITERIA FOR INDEX OF GEO-ACCUMULATION

Rank	I_{geo}	Degree of accumulation
0	I_{geo}≤0	Unaccumulated
1	0< I_{geo}≤1	Unaccumulated to moderately accumulated
2	1< I_{geo}≤2	Moderately accumulated
3	2< I_{geo}≤3	Moderately to heavily accumulated
4	3< I_{geo}≤4	Heavily accumulated
5	4< I_{geo}≤5	Heavily to extremely accumulated
6	I_{geo} >5	Extremely accumulated

3) Geoaccumulation index

The pollution degree of heavy metals in soil can be indicated by the geoaccumulation index method [14], [15], reflecting the impact of human activities on the environment. The calculation formula is as follows:

$$I_{geo} = log_2\left[\frac{C_i}{K \times S_i}\right] \quad (5)$$

where I_{geo} is the geological accumulation index of *i* pollutants; *K* is the correction coefficient that exhibits the characteristics of soil materials and other influences (generally $K = 1.5$) [16]. I_{geo} levels are shown in Table V.

III. Result and Analysis

A. Soil Heavy Metal Content and Morphological Distribution Characteristics

Table VI shows the contents of 5 heavy metal elements in agricultural soil of Tianyuan township, Wuxi county. The average content sequence of these five elements is: $Zn^{2+}>Cr^{3+}>Cu^{2+}>Pb^{2+}>Cd^{2+}$. Compared with the soil pollution risk screening values in the Soil Eenvironmental Quality Control Standard for Agricultural Land (Trial) (GB 15618-2018), except that 37 samples of Cd^{2+} exceeded the standard, Cr^{3+}, Cu^{2+}, Pb^{2+} and Zn^{2+} did not out of limits. At the same time, the average contents of these five heavy metal elements, except Cr^{3+} and Zn^{2+}, were all higher than the background values of soil geochemistry in Chongqing, the exceeding rates were 1049.91%, 20.81% and 20.23%, for Cd^{2+}, Cu^{2+} and Pb^{2+} respectively. This indicates that human activities began to lead to the accumulation of heavy metal elements in the soil in this area. The variation coefficients of the 5 elements were 68.17%, 22.52, 35.44%, 57.25 and 28.90%, for Cd^{2+}, Cr^{3+}, Cu^{2+}, Pb^{2+} and Zn^{2+} respectively. According to Wilding's classification of variation degree [17], Cr^{3+}, Cu^{2+} and Zn^{2+} belong to moderate variation (15%<CV<36%), while Cd and Pb^{2+} belong to high variation (CV>36%), indicating that these elements were unevenly distributied and may be affected by human activities. Meanwhile, it can be seen from the table that the heavy metals in the soil in this survey region are right skewed, and the skewness and kurtosis parameters of Cd^{2+}, Cr^{3+} and Pb^{2+} are larger; the P of KS test of Cd^{2+}, Cr^{3+} and Pb^{2+} reaches a significant level, indicating that non-normal distribution is more serious, and the P values of Cu^{2+} and Zn^{2+} are not significant and belong to a normal distribution.

TABLE VI: Statistical Result of Soil Heavy Metal Content in Tianyuan Township, Wuxi County

Heavy metal element	Maximum/ (mg·kg^{-1})	Minimum/ (mg·kg^{-1})	Average value/ (mg·kg^{-1})	Standard deviation	Variable coefficient CV/%	Skewness	Kurtosis	K-s *P*	Background values	Exceeding standard rate
Cd	0.01	3.29	1.26	0.86	68.17%	0.81	0.10	0.01	0.11	1049.91%
Cr	48.31	147.15	73.33	16.51	22.52%	1.51	4.61	0.01	80.00	-8.34%
Cu	10.57	74.31	31.41	11.13	35.44%	0.62	1.63	0.20	26.00	20.81%
Pb	14.86	117.24	31.26	17.89	57.25%	3.12	10.81	0.00	26.00	20.23%
Zn	31.38	160.94	86.69	25.05	28.90%	0.22	0.23	0.20	253.00	-65.74%

The proportional distribution of various forms of heavy metals is shown in Fig. 2.Cr^{3+}, Cu^{2+} and Zn^{2+} were mainly in residual state, and the average F5 proportions of Cr^{3+}, Cu^{2+} and Zn^{2+} were 91.61%, 84.48% and 72.30%, respectively, which were at a high level. Due to the stable nature of residual heavy metals, which are bound in mineral lattice, it is generally difficult to release, indicating that Cr^{3+} and Cu^{2+} have low bioavailability and low ecological risk. Pb^{2+} mainly exists in F3 and F5, with an average value of 38.24% and 49.22%, respectively. F3 is mainly formed by iconicity union, its strong binding ability with the hydroxide of iron and manganese in the soil causes it difficult to release and is mainly affected by redox potential. Cd^{2+} is dominated by F1 and F3, which are 22.25% and 47.57%, respectively. A high proportion of F1 indicates that Cd^{2+} has strong mobility and high ecological risk.

B. Evaluation Results of Heavy Metal Pollution in Soil

1) Evaluation results of Nemerow index method

It can be seen from Table VII that from the whole range, the mean value of soil pollution coefficient is $Cd^{2+}>Cu^{2+}>Pb^{2+}>Cr^{3+}>Zn^{2+}$. The average pollution coefficient of single factor Cd^{2+} is greater than 3, and the cumulative effect is the most significant, which is at the level of severe pollution. Single factor pollution coefficients of Cr^{3+}, Cu^{2+} and Pb^{2+} are all less than 2, which belong to the level of prevention and mild pollution. The average pollution coefficient of single factor Zn^{2+} is less than 0.7, which belongs to a safe range.

2) Potential ecological risk assessment results

The evaluation results of potential ecological risks are shown in Table VIII. The potential ecological risk index is shown as $Cd^{2+}> Cu^{2+}> Pb^{2+}> Cr^{3+}> Zn^{2+}$. The average value of E_r^i of Cd^{2+} is 227.79, which is the main contributing factor, and its risk reaches the level of serious pollution. The average of E_r^i of Cr^{3+}, Cu^{2+}, Pb^{2+} and Zn^{2+} were all much below than 40, indicating mild pollution. According to the heavy metal *RI* value of Table VIII, it can be determined that the survey region is moderately polluted.

3) Geoaccumulation index results

Fig. 3 shows the results of the accumulation index of heavy metals in the soil. Zn^{2+} and Cr^{3+} in the soil were 97.40% and 96.10%. They are in a state of no accumulation, indicating that the study area was basically free from Zn^{2+} and Cr^{3+} pollution. 76.62% of Cu^{2+} and 80.52% of Pb^{2+} are in a state of no accumulation, which is clean on the whole. Cd^{2+} is distributed from no accumulation to extremely strong accumulation with 3.90%, 48.05%, 7.79%, 18.18%, 12.99%, 7.79%, 7.79% and 1.30%, respectively, indicating the management standards and governance need to be strengthened.

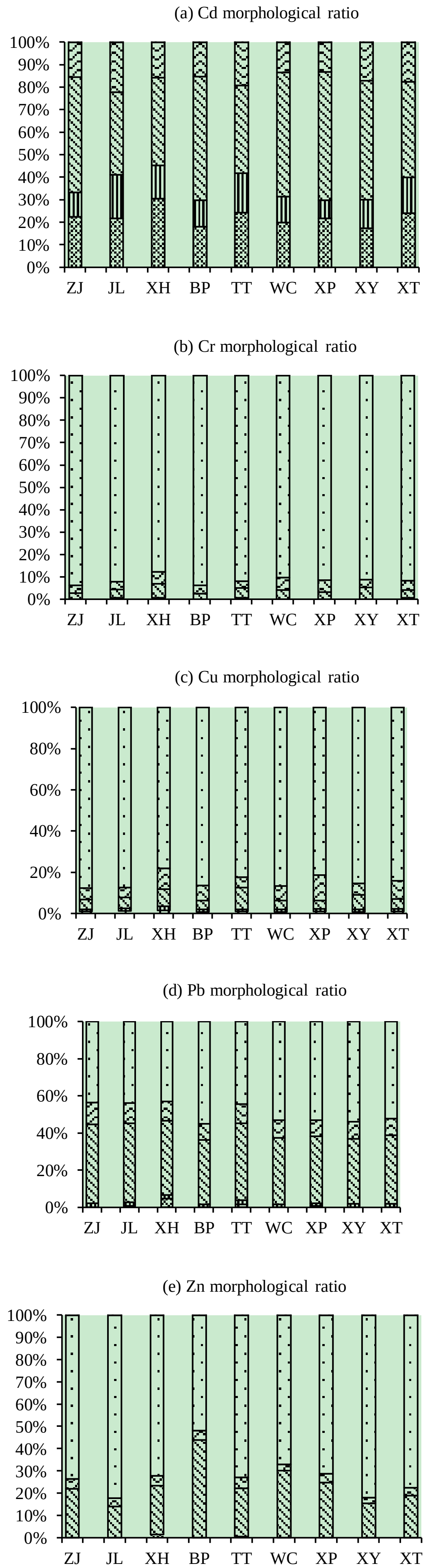

Fig. 2. Distribution ratio of heavy metal fractions.

TABLE VII: RESULTS OF NEMEROW INDEX ON SOIL HEAVY METAL

Project	Single factor pollution factorp_i					Comprehensive pollution coefficientp_i
	Cd	Cr	Cu	Pb	Zn	
Average	8.00	0.89	1.17	1.12	0.33	1.53

TABLE VIII: EVALUATION ON POTENTIAL RISK OF SOIL HEAVY METAL

Sampling point	E_r^i					Risk index	Risk level
	Cd	Cr	Cu	Pb	Zn	RI	
ZJ	52.82	1.94	7.13	4.62	0.29	66.79	Slight
JL	13.40	1.62	4.88	4.45	0.27	24.62	Slight
XH	979.65	1.50	5.09	9.37	0.35	1138.22	Serious
BP	512.75	1.86	7.09	8.98	0.45	531.12	Severe
TY	78.87	2.12	6.69	6.04	0.37	75.27	Slight
WC	103.15	1.74	6.08	5.26	0.34	128.23	Slight
XP	405.54	2.01	6.23	5.88	0.40	420.05	Severe
XY	165.50	1.94	6.57	4.75	0.36	179.11	Moderate
XT	195.72	1.89	5.34	4.78	0.34	208.08	Moderate
Average	227.79	1.88	6.26	6.36	0.36	242.65	Moderate

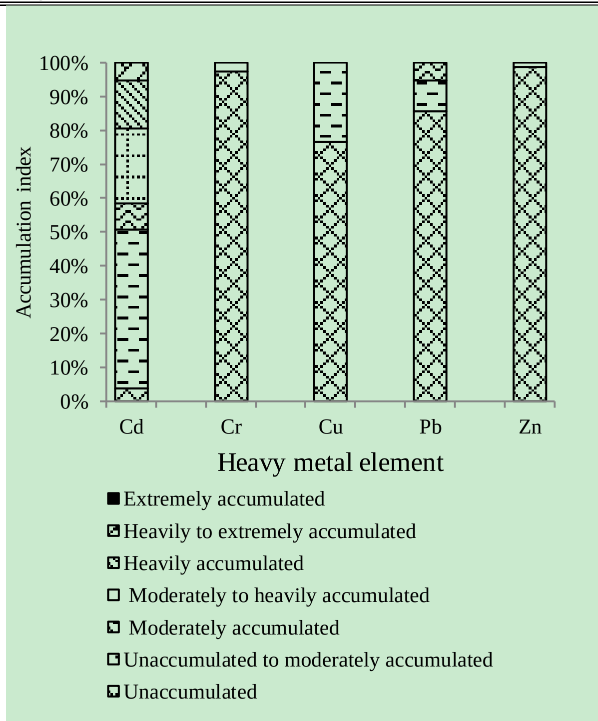

Fig. 3. The result chart of accumulation index of each metal element.

IV. SOURCE ANALYSIS OF HEAVY METAL ELEMENTS IN SOIL

In order to have a more comprehensive understanding of the geochemical properties of heavy metals in the soil in this survey region, Pearson correlation analysis was conducted on Cd^{2+}, Cr^{3+}, Cu^{2+}, Pb^{2+} and Zn^{2+} to determine the source of heavy metals. The results are shown in Table Ⅸ. According to the table, they are remarkably correlative between Cu, Cr and Zn^{2+} ($P< 0.01$), and Pb^{2+}, Cd^{2+} and Zn^{2+} ($P< 0.01$). It indicates that the sources of Cu^{2+}, Cr^{3+} and Zn^{2+}, and Pb^{2+}, Cd^{2+} and Zn^{2+} are similar or controlled by the same factor.

The heavy metals in the soil are mainly derived from natural factors and anthropogenic factors, the principal component analysis method can be used to judge the pollution sources of heavy metals. Through the Table X and Fig. 4, the eigenvalues of the first two principal components are all greater than 1, and the contribution rate is up to 71.182%, which can represent the data inclusion information. Cd^{2+}, Pb^{2+} and Zn^{2+} have a high load in the first principal component factor, Cr^{3+} has a high load in the second principal component factor, and Cu has a similar load in the two component factors, indicating the joint influence of the two factors.

TABLE IX: PERSON CORRELATION COEFFICIENTS OF HEAVY METAL CONTENT

Heavy metal element	Cd	Cr	Cu	Pb	Zn
Cd	1				
Cr	0.022	1			
Cu	-0.048	0.602**	1		
Pb	0.593**	0.050	0.175	1	
Zn	0.169	0.149	0.470**	0.464**	1

Heavy metals come from a variety of sources, which are influenced by different regions, different approaches, industrial and agricultural distribution and other factors [18]. Soil samples are mainly collected from concentrated points of villagers, and due to the steep terrain, almost all the cultivated land is close to the traffic roads. Zn^{2+} is an important additive in the production of automobile tires [19]. Cu^{2+} has high corrosion resistance and high thermal conductivity, and is often used to prepare vehicle braking systems and automobile radiators. Therefore, Cu^{2+} and Zn^{2+} can be used as identification elements of traffic pollution sources [20]. Some pesticides and agricultural plastic films contain Cd^{2+} and Pb^{2+} [21], which also cause soil pollution. However, Cr^{3+} content depends on soil parent material, which is generally the least polluted heavy metal in China [22], [23]. Therefore, the first principal component factor can be regarded as "human factor" by combining the sampling point position and correlation analysis with the results of principal component analysis. The second principal component factor can be regarded as a "natural factor" as shown in Table X.

TABLE X: RESULTS OF PRINCIPAL COMPONENT ANALYSIS OF HEAVY METAL ELEMENTS IN SEDIMENTS

Project	The first principal component factor	The second principal component factor
Cd	0.748	-0.157
Cr	0.259	0.897
Cu	0.577	0.690
Pb	0.752	-0.473
Zn	0.672	-0.233
eigenvalue	1.976	1.583
variance contribution rate/%	39.522	31.660
accumulating contribution rate/%	39.522	71.182

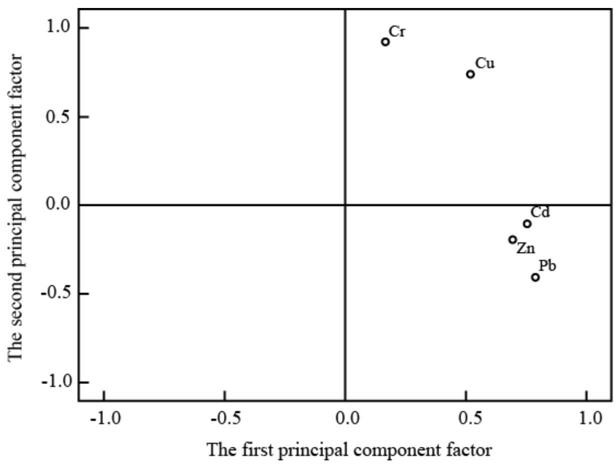

Fig. 4. Multidimensional scaling analysis of heavy metal in soils.

V. CONCLUSION

1) The mean values of Cd^{2+}, Cu^{2+} and Pb^{2+} were higher than the background values of soil geochemistry in Chongqing. Compared with the soil pollution risk screening values in the Soil Eenvironmental Quality Control Standard for Agricultural Land (Trial) (GB 15618-2018), except that 37 samples of Cd^{2+} exceeded the standard, Cr^{3+}, Cu^{2+}, Pb^{2+} and Zn^{2+} did not exceed the standard.
2) Through the analysis of heavy metal form proportion, except Cd^{2+}, Cr^{3+}, Cu^{2+}, Pb^{2+} and Zn^{2+} are all based on residue state, indicating that these four heavy metal elements are of low bioavailability and pollution risk.
3) The pollution levels obtained by Nemerow index method and potential ecological risk index method were both $Cd^{2+}>Cu^{2+}>Pb^{2+}>Cr^{3+}>Zn^{2+}$. According to the evaluation results of geoaccumulation index, the four heavy metals Cr^{3+}, Cu^{2+}, Pb^{2+} and Zn^{2+} in the soil in the survey region are mainly non-accumulation and relatively clean, which is consistent with the conclusions of the previous two evaluation methods.
4) Correlation analysis and principal component analysis make clear that Cr^{3+} was mainly affected by natural factors, and its content was related to soil parent material. Cd^{2+}, Pb^{2+} and Zn^{2+} are mainly affected by man-made pollution. As the sampling sites are mainly located near the concentration points of villagers and traffic roads, they are mostly affected by agricultural activities and traffic emissions. Cu^{2+} is affected by two factors.

CONFLICT OF INTEREST

The authors declare no conflict of interest

AUTHOR CONTRIBUTIONS

Hengchang Zhang conducted the research. Chuan Fu, Tingzhen Li, Bin Yan, Yan Wu calculated and analyzed the data. All authors wrote this paper together and had approved the final version.

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