

Original Research

Spatial Distribution and Influence Analysis of Soil Heavy Metals in a Hilly Region of Sichuan Basin

Hanhui Liu*, Guanghui Liu, Yunfan Zhou, Chao He

The Ministry of Land and Resources Key Laboratory of Geological Spatial Information Technology,
College of Earth Sciences, Chengdu University of Technology,
Chengdu, 610059, China

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Abstract

The hilly region of the Sichuan basin has unique natural topography, geomorphology, geology, and hydrology, with intensive human activities in the area. Therefore, it is of great significance to carry out research on heavy metal characteristics. In this study, taking five villages of Zigong city as a sample, the content of eight kinds of heavy metals in soil was determined (Cu, Cd, As, Pb, Cr, Zn, Ni, and Hg) based on ordinary Kriging. The results showed that Cd, Pb, Ni, and Zn content in the research area was higher than the background value and had a good concentration with all of the heavy metal contents lying within the national standards. Through soil parent material, soil environment, atmospheric environment, and water environment analysis of heavy metals, it was found that higher levels of heavy metals in the research area were mainly affected by agricultural and industrial activities (atmospheric dustfall). This was generally where there were intense areas of human activity – especially in and around transport routes and construction areas. It was also found that the soybean crop exhibited a strong Cd accumulation ability. This soybean planting should be reduced to control the distribution of Cd. Zn and Cu had a positive enrichment effect on rice, soybeans, and other crops, and had little enrichment effect on grapefruit. This study showed the effect human activities have on the quality of soil quality on the crop. This research has great significance for the ecologically sustainable development of society.

Keywords: heavy metals, soil environment, atmospheric dustfall, Sichuan, hilly region of Sichuan basin

Introduction

Soil is a key component of the terrestrial ecosystem and plays an essential role in food production, biochemical transformations, recycling of elements, and many

recreational activities [1]. With rapid industrialization and urbanization, as well as extensive application of agrochemicals, heavy metal pollution of soil has increasingly become a serious problem in China [2-3]. Agricultural land includes farmland, forestland, and grassland, and contamination of these types of soils with heavy metals is attracting considerable attention because of the potential threat to farmland, forestland, and grassland ecosystems [4-5]. Anthropogenic contamination

*e-mail: pjoes_mcse@hotmail.com

– including excessive use of fertilizers and pesticides, irrigation with sewage water, and elevated atmospheric deposition – is the main source of the existence of heavy metals in agricultural soils [6-8]. Heavy metal contamination in soil exhibits specific pollutant characteristics as they do not decay over time and can be toxic to plants when they exceed specific thresholds. At the same time, accumulation of heavy metals in soil has the potential to inhibit the proper functioning of soil, causing toxicity to plants and promote heavy metals to humans [9-10].

Agricultural production plays a key role in the development of China. However, with rapid industrialization, urbanization and increasing reliance on agrochemicals, heavy metal contamination in soil has become severe during the last two decades [11-12]. In the past decade, numerous studies have reported the concentration, distribution, and sources of heavy metals in Chinese soil [13-14]. However, most of these studies have focused on urban soil [13, 15] and only a few on rural, agricultural land – especially in the hilly region of the Sichuan basin.

Much food and other materials for daily life are produced in the hilly region of the Sichuan basin. Rapid economic development and increases in population have put an enormous pressure on the ecosystems and soil ecological environments in these regions. Combined with the development of various industrial activities, at the expense of the environment and the impact of rural-point source pollution, the land-bearing load is saturated in the hilly region of the Sichuan basin. This has brought about a series of consequences; for example, serious degradation of soil ecological environment, soil environmental quality is decreasing year by year and soil pollution problems have become increasingly prominent. In light of the above, this study has carried out heavy metal pollution and the distribution characteristics in soil of five villages in Zigong city and the control factors have been discussed.

Materials and Methods

Study Area

The study area is located in the central Sichuan basin in China. The size of the research area from east to west is 4.3 km long and 5.0 km wide from north to south.

The climate in this region is subtropical, humid, and susceptible to monsoons, and has four distinct seasons. The annual average temperature is 17.8°C and the accumulated temperature is 6021.0°C. The year round sunshine is 1200-1280 hours, the average annual precipitation is 1031-1041mm, and average annual rainfall is 140 days. The characteristic of the terrain is high in the northwest, low in the southeast, with a creek widely distributed among small hills and a narrow mesa. There are some areas that have exposed bedrock in the research area, but most of the rest is in the quaternary. In the bedrock outcropping is Jurassic Shaximiao (J_2 s), and the

lithology is purple, brownish red mudstone, and unstable feldspar quartz siltstone interbedded. Quaternary alluvial material is divided into (Qpal) and alluvial material (Qdpl), scattered in the vicinity of the river terraces and Pingba, and the lithology is mainly packed clay and gravel with a gravel floodplain.

Collection of Samples

Soil samples were collected following the method in reference [16]. Sampling points were set in a 1:10,000 topographic map. The sampling density was 16 kg/km² and the sampling unit was 250m×250m. The research area was divided into 173 sampling cells (Fig. 1) and soil samples were collected for testing. As and Hg content in soil samples were estimated by the atomic fluorescence method. The content of Cr, Cu, Ni, Pb, and Zn was determined by x-ray fluorescence, and plasma mass

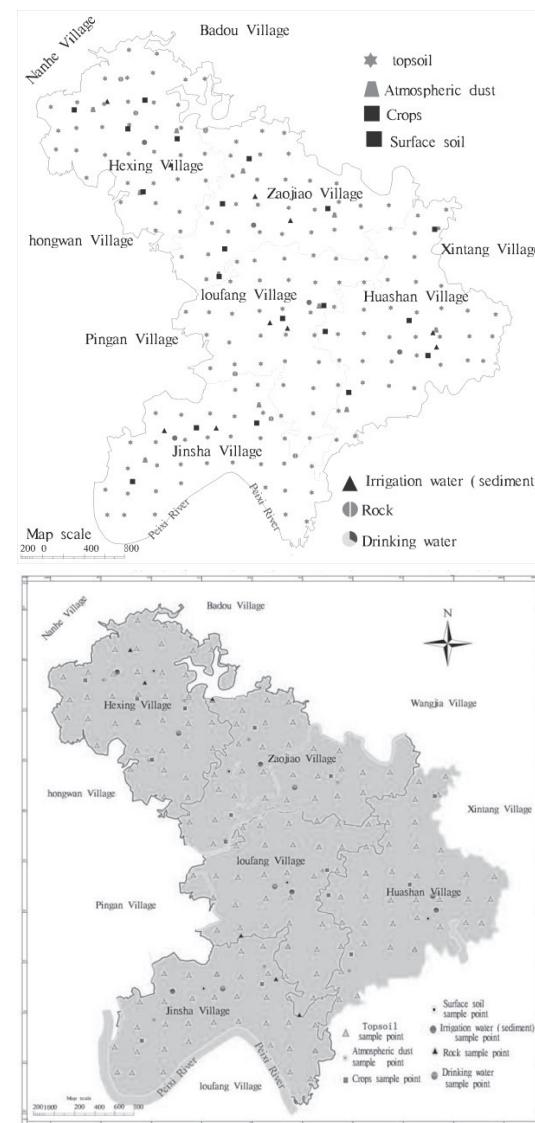


Fig. 1. Distribution of samples and sampling position in the study area.

spectrometry was used to determine the Cd content in soil samples [17].

Soil samples were collected by digging soil about 30 cm between locations, placed at different levels according to the level of soil excavated. Records were kept of profile number (field number), soil name (field naming), soil color, soil structure, mining land points, terrain (plus notes on parent material, vegetation, altitude, etc., if necessary), hunters, and acquisition date.

Atmospheric dustfall samples were collected from the windows of residential buildings and public facilities. While sampling, direct sources of pollution such as industrial pollution, civil coal, and peeling paint, etc., should be avoided. Each sample was made by mixing equal amounts of 3-4, collecting approximately 20 g.

Irrigation water samples were collected from the main irrigation water intake. The water intake section was generally laid on a larger main stream, and the control was less than 80% of the total runoff flow. Drinking water samples were collected at the typical model of the centralized water supply and untreated water sources with a sampling density of 1 kg/km². Well-water samples were collected from residential wells. While collecting irrigation water, sediment samples were also collected.

According to the actual situation in the research area, grain crops were chosen from rice, wheat, corn, sweet potato, and vegetables, with eggplant and pepper being the most commonly harvested. While collecting plant samples, plant roots and soil were collected at the same time. The plants were generally uprooted, shaking the soil under the plant roots and removing plant roots and other debris.

Quality Assurance and Quality Control

Soil standard reference material (GBW07401, GSS-1) obtained from the Center of National Standard Reference Material of China was used for quality assurance and quality control (QA/QC). Accepted recoveries ranged from 81.0-109%. Analysis methods were evaluated in blank ($n = 7$) and duplicate samples ($n = 13$) for each set of samples. Relative deviation of duplicate samples was <7% for all the batch treatments.

Data Analysis Methods

Ordinary Kriging is a moving weighted average method that uses variograms constructed to interpolate between samples by considering the shape, size, and relative positions of samples within the database [18-20]. These variograms are calculated by the following equation:

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i) - Z(x_i + h)]^2 \quad (1)$$

...where $\gamma(h)$ is the variogram, $Z(x_i)$ is the sample value at location I, h is lag distance, and $N(h)$ is the number of data pairs within lag distance [21].

Results and Discussion

This paper collected 173 samples of surface soil and, using various testing methods, obtained the content of eight heavy metals in the topsoil, and conducted a statistical analysis. Table 1 shows the maximum and minimum values of elements in the test sample. X_1 is the average of all elements in the sample, S_1 is the standard deviation of the measured values of deviation of the sample mean, and Cv is the coefficient of variation (the standard from the difference between the ratio of the arithmetic mean, $Cv=S/X_1$). The average concentrations of As, Cd, Cr, Cu, Pb, Ni, Zn, and Hg were 3.58, 0.49, 75.49, 27.95, 29.93, 33.30, 95.85, and 0.05 µg/g, respectively. Background concentrations of these heavy metals in the soils of Sichuan Province are 9.3, 0.07, 79, 29.7, 28.9, 32.6, 82.1, and 0.04 µg/g for As, Cd, Cr, Cu, Pb, Ni, Zn, and Hg, respectively [22]. The Chinese Environmental Quality Standard for Soils [23] provides critical values for heavy metal concentration in soil for the protection of agricultural products and human health, and these are 25, 1.0, 250, 200, 350, 300, and 1.0 µg/g for As, Cd, Cr, Cu, Pb, Zn, and Hg, respectively. In this study, Cd content was exceeded only in five samples. Although the heavy metal concentrations in the majority of the samples did not exceed the national guideline values, the obvious enrichment of these metals in the research area should be noted.

In the analysis of heavy metals in terms of distribution, the coefficient of variation can eliminate the influence of objective measurement scales and make it more dimensionless than the standard deviation. In Table 1, Cd and Hg coefficients of variation were significantly higher than those of the other elements, showing that aggregation was worse in these two elements.

Understanding the spatial distribution soil heavy metals in soil is a significant prerequisite for monitoring and evaluating the eco-environment in the research area. The spatial distribution of heavy metals in soil of the hilly region of the Sichuan basin was visualized by drawing distribution maps of eight heavy metals using ArcGIS.

Table 1. Total amount of statistical indicators of soil surface characteristics ($n = 173$; µg/g).

Element	Max	Min	X_1	S_1	Cv
As	5.83	2.03	3.58	0.66	0.18
Cd	4.03	0.16	0.49	0.39	0.86
Cr	124.00	59.90	75.49	7.13	0.09
Cu	36.10	18.20	27.95	2.97	0.11
Hg	0.19	0.02	0.05	0.02	0.42
Ni	57.80	18.40	33.30	4.34	0.13
Pb	38.20	22.70	29.93	2.73	0.09
Zn	140.00	47.20	95.85	12.73	0.13

According to the data, analysis of spatial interpolation was conducted based on 173 samples of the various elements, and this generated the corresponding grading evaluation map of Hg, Cd, As, Pb, Cr, Ni, Cu, and Zn elements (Fig. 2). The maps show some distinct geographical trends, especially for Cd, Pb, Ni, and Zn, which were found to be higher than the background values in areas of high concentrations. A previous study [24] found that enrichment of Cd, Pb, and Zn in agricultural soil was closely correlated to agricultural production.

Fig. 2 shows that the content of heavy metals in topsoil was unevenly distributed in the research area.

The present study has determined three main sources of heavy metal pollution. The first source was lithogenic components, and was associated with soil parent materials [25-26]. The second source was from industrial activity such as vehicular transport [24, 27-28]. The third source was controlled by the anthropogenic inputs from agricultural production, such as chemical fertilizers [29-30].

The original rock is the material basis for soil formation. In the corresponding biological climate condition, the surface of the original rock is gradually turned into soil through physical and chemical weathering,

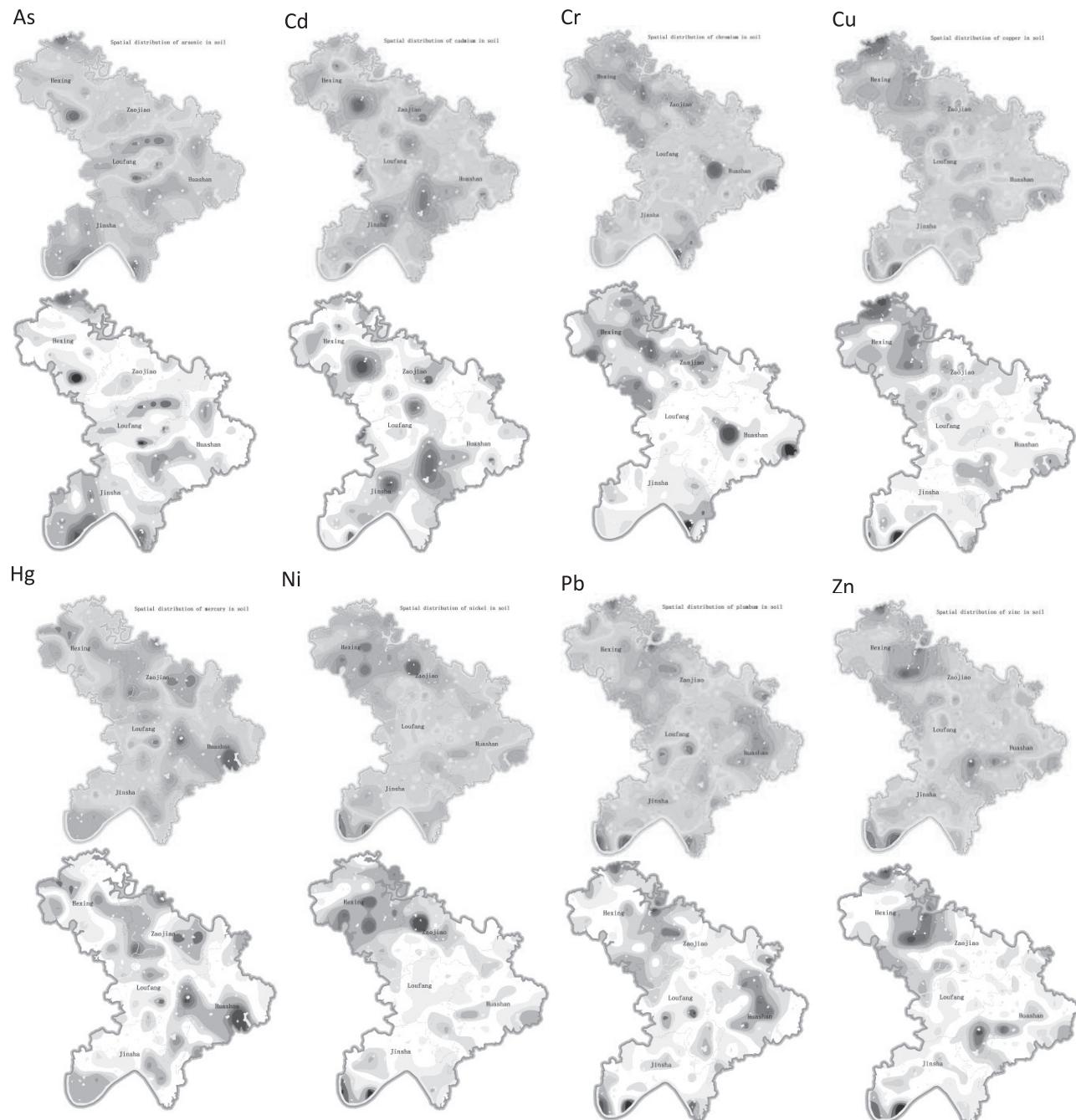


Fig. 2. Spatial distribution of different heavy metals on surface soil (dark representative element content is high, light representative element content is low).

and biological degradation [31-34]. We found that parent materials and pedogenic processes were major factors contributing to high concentrations of heavy metals in

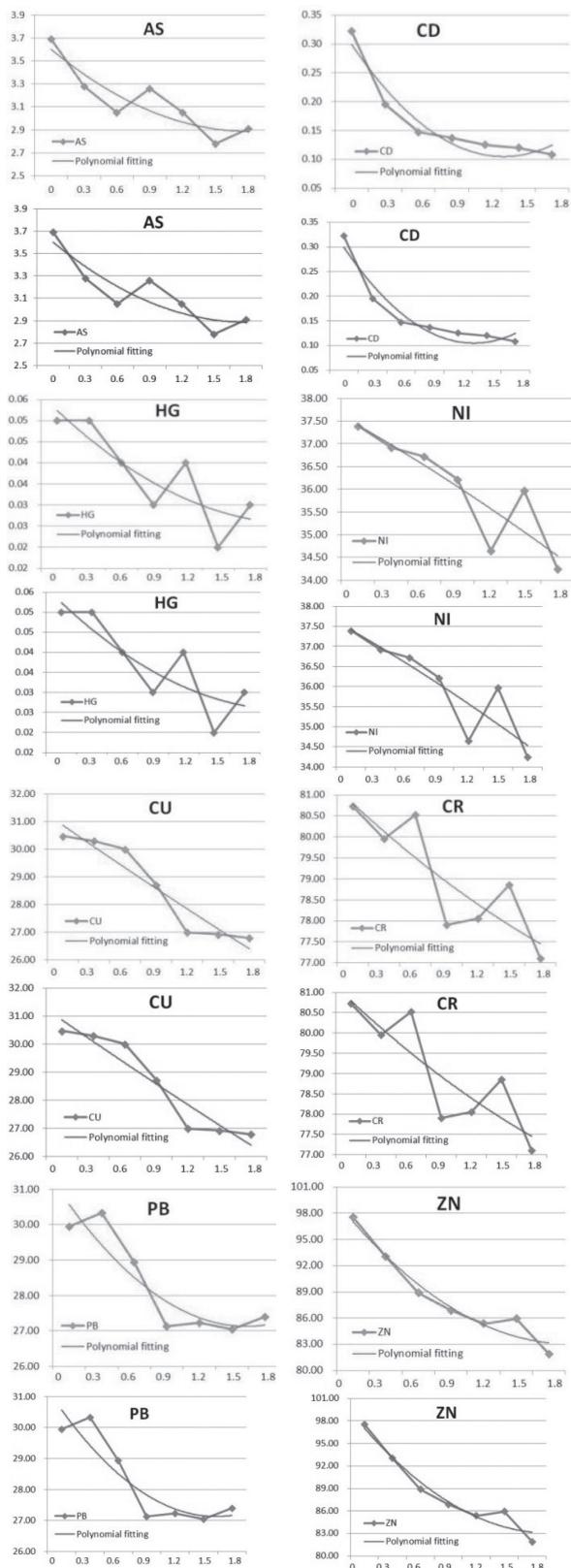


Fig. 3. Vertical change of heavy metal content in soil (Y unit for ug/g, X unit for meter).

soil [26]. In the research area, soil mainly originated from quaternary eluvial, alluvial, and diluvial deposits. The exposed bedrock was a Jurassic Shaxi Miao formation (the lithology was purple, brown-red mudstone, and siltstone unstable feldspar quartz interbedded), and all the soil originated from the same parent material. There was no significant difference in space between soil and parent material. Heavy metals in the soil background values were consistent. According to the analysis of heavy metals content in rock (Table 2), the concentration of heavy metals in rock was lower than that of the surface soil.

Meanwhile, the vertical cross-section of heavy-metal soil showed a decreasing trend with increasing depth (Fig. 3), and with the increase in depth, the amount of variation decreased. This shows that heavy metal content tends to be stable at the depths of soil and it also indicates that heavy metals on the surface soil have super-imposed external sources.

Therefore, the original rock is not the reason for heavy metal accumulation in the surface soil.

Industrial activities cause heavy metal pollution in two ways. One is direct pollution, such as wastewater and solid waste [35-36]. The second is indirect pollution, such as atmospheric dust. Samples were collected from agricultural land where there was no direct wastewater or waste discharge. Therefore, industrial pollution was caused by atmospheric dust. In order to investigate if the heavy metals in the atmospheric dustfall had an impact on the topsoil, the content of heavy metals in the dustfall and the topsoil were compared. The results are shown in Table 2, including the average heavy metal content of all the samples.

Table 2 shows the heavy metal content in atmospheric dustfall. In addition to Hg and NI, the remaining elements' content was much higher than the background value of the topsoil. The Zn content in the atmospheric dustfall was 4.33 times greater than that of the topsoil, and the As, Cd, and Pb content in the atmospheric dustfall was 3.5 times higher than in the topsoil. This suggests that atmospheric particles settling into the soil was one of the external sources of heavy metals in the topsoil. By comparing the spatial distribution of heavy metals between the topsoil elements and the atmospheric dustfall, the spatial distribution of elements had a better consistency [37-38].

There was a road to the town in the west, north of Hexing village. The road was frequented by a large number of diesel vehicles and farm trucks in both directions. Heavy metals concentrations in the topsoil and the atmospheric dustfall had high values, which suggests that the atmospheric dust has an impact on the heavy metals in the soil. It is worth noting that the district had just built a concrete road to a school called the Hope School. Although currently there is no contamination, the near-surface portion of the heavy metal content in the atmospheric dustfall was relatively high, with the accumulation of atmospheric dustfall ultimately affecting soil quality. Similarly, in a place not far from Huashan village, a highway will be built which will have a great impact on the future of the soil and will require attention and monitoring.

Table 2. Heavy metal content in atmospheric dustfall.

Topsoil and rock Index	Heavy metal content ($\mu\text{g/g}$)				
	Atmospheric dustfall (AD)	Topsoil (TS)	Rock (R)	AD / TS	TS/R
As	13. 16	3. 54	2. 02	3. 72	1. 75
Cd	1. 59	0. 40	0. 23	3. 98	1. 74
Cr 93. 6		75. 21	56. 28	1. 24	1. 34
Cu	54. 73	28. 01	17. 37	1. 95	1. 61
Hg	0. 25	0. 49	0. 01	0. 50	49
NI	30. 3	33. 24	27. 57	0. 91	1. 21
Pb	104. 72	29. 88	23. 54	3. 50	1. 27
Zn	413. 52	95. 392	60. 78	4. 33	1. 57

In the present study, soil samples were collected from agricultural soil. A previous study [24] found that enrichment of Cd, Cu, and Zn in agricultural soil was closely correlated to agricultural production. For example, cadmium is usually considered a marker element for agronomic practices, which includes the use of chemical fertilizers such as phosphate fertilizers, livestock manure, etc. [28]. Annual Cd input to agricultural soil in China has been reported as 113, 778, and 30t for total fertilizers, livestock manures, and irrigation water, respectively [30]. Additionally, Zn and Zn compounds are widely used

as agricultural fertilizers, and cattle slurry contains a significant amount of Zn and Cu since these elements are fed to cattle as a trace metal supplement [29, 39]. In the research area, enrichment of heavy metals (Cd, Cu, and Zn) had a good correlation with the production of rice, and in turn rice production has a direct relationship with fertilizer use. So, agricultural production is a main source of heavy metal pollution.

Irrigation water was collected in the research area and was the corresponding total of six sediment samples from reservoirs, ponds, and rivers [40-41]. Table 3 shows the

Table 3. Heavy metal content in water environment ($\mu\text{g/g}^{-1}$).

Index	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Water	1.34	0.23	<0.01	2.87	0.02	6. 86	9. 85	24.59
Sediment	2.51	0.34	69.55	26.72	0.03	35.02	27.06	75.64
Topsoil	3.54	0.40	75.21	28.01	0.49	33.24	29.88	95.39

Table 4. Heavy metal content in plant and root soil ($\mu\text{g/g}^{-1}$).

Type	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Rice	0.118	0.010	0.173	5.719	0.003	0.067	0.038	13.570
Root soil	2.939	0.711	72.091	27.454	0.056	0.858	28.236	95.100
Root soil /rice	24.9	71.1	416.7	4.8	18.7	12.8	743.0	7.0
Peanut	0.016	0.091	0.122	12.910	0.002	0.041	0.095	45.413
Root soil	3.36	0.441	73.5	26.8	0.03	1.06	27.7	96.8
Root soil /peanut	210.0	4.8	602.4	2.0	15.0	25.8	291.5	2.1
Soybean	0.030	0.080	0.121	16.420	0.001	0.070	0.034	53.201
Root soil	3.25	0.461	74.8	27.6	0.034	0.852	26.8	95.5
Root soil /soybean	108.3	5.8	618.2	1.7	34	12.2	788.2	1.8
Grapefruit	0.028	0.001	0.110	3.370	0.001	0.015	0.015	1.370
Root soil	2.49	0.490	68.9	32.8	0.034	0.693	26.7	101
Root soil / grapefruit	88.9	490.0	626.4	9.7	34	46.2	1780	73.7
Sweet potato	0.017	0.012	0.164	15.424	0.002	0.092	0.096	28.012
Root soil	5.33	0.880	74.6	32.3	0.035	1.10	37.5	116
Root soil / sweet potato	313.5	73.3	454.8	2.1	17.5	11.9	390.6	4.1

average heavy metal content of all the samples. As can be seen, the heavy metal content in the water was below that of the topsoil. The heavy metal content in the sediment and soil background value was basically the same, indicating that the influence of soil heavy metals on water is limited.

The research area has collected crops (rice, peanuts, sweet potatoes, and grapefruit) and corresponding root soil for a total of 15 sets. Table 4 shows the various types of plants and their corresponding roots with heavy metal content in the soil. All the samples were from the fruit of plants. As the table shows, most of the heavy metal content of root soil was dozens of times or even 100 times that found in the plant. Only individual heavy metals content was not very different between plants and root soil and this only occurred a few times. High Cd content of the research area can be seen in Table 4. The soybean has a strong ability to accumulate Cd, therefore its planting should be reduced to reduce the distribution of Cd. It can also be seen from Table 4 that Cu has a beneficial effect on various types of plants gathered, but Cu content in the research area did not reach hazardous levels, and therefore the impact of Cu could not be determined. The Zn content was higher in the research area and the Zn found on rice, soybeans, and other crops had a good enriching effect on the crop while it had little effect on the grapefruit. Therefore, the type of vegetation should be adjusted according to Zn distribution.

Conclusions

This paper collected a large number of samples, including 173 topsoil, 11 atmospheric dustfall, six water and sediment, and 15 core samples. Eight heavy metals were identified and their content was determined for all of the samples. This paper presents the concept of using the ordinary Kriging method to generate the spatial distribution of heavy metals in the research area. By analyzing the spatial distribution of heavy metals in the soil, it was found that the distribution of heavy metals is various. By analyzing the content of heavy metals in original rock, atmospheric dustfall, and agricultural production, it can be found that the differences of heavy metals in soil are derived from atmospheric dustfall and agricultural production, which differs depending on the specific environments. This suggests that attention should be paid to environmental protection while developing the economy. In this paper, the effects of heavy metals on the water environment and crops were also analyzed. Analysis showed that the influence of various elements was different. For the water environment, the influence of Zn, Pb, and Ni was significant. For the core samples, the influence of Zn and Cu was significant. This study can indicate the choice of crops in order to ensure the low adsorption rate of heavy metals.

Acknowledgments

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