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Migration and Accumulation of Heavy Metals in Soils and Crops around Coal Gangue Hill

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Abstract:

Concerns about soil heavy metal contamination have continued to rise in recent years, particularly concerning the surrounding soil heavy metal contamination caused by the prolonged stockpiling of coal gangue. In this study, we collected the surrounding soil and corn from a coal gangue hill in Qufu, Shandong Province, detected the content and occurrence forms of soil heavy metals, analyzed the correlation between coal gangue and soil heavy metals, and evaluated the impact of heavy metal pollution on environmental quality of the site. The results showed that Hg and Cd were major pollutants in coal gangue hill, which had reached serious pollution levels, and the content of the other was all lower than the risk control values in agricultural lands. With the distance from the coal gangue hill center from near to far (0-150 meters), the occurrence modes of soil heavy metals gradually increased to stable, and the potential risk decreased accordingly. The absorption and storage capacity of Pb in corn kernels was the strongest, which led to the content exceeding the standard by about 85.7 times. The study provided a reliable theoretical basis for the rational deposition of coal gangue and ecological environmental protection in the region.

Keywords: *Heavy metal, Coal gangue hill, Occurrence modes, Chemical and physical properties*

I. INTRODUCTION

With the rapid development of coal mine production, a series of environmental problems such as resource crisis, environmental pollution and ecological damage have become prominent, which have aroused widespread concern [1]. Gangue come from the underground reduction environment into the surface oxidation environment, and under the action of atmospheric precipitation and microorganisms, Cr, Hg, Pb and other heavy metals into the surrounding soil caused soil heavy metal pollution [2]. Heavy metal

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pollutants remained in the soil for a long time and were generally not easily transported and cannot be decomposed by microorganisms. When accumulated to a certain extent, they would have toxicity on soil-plant systems, leading not only to soil degradation, but also polluting surface and groundwater through runoff and scouring to degrade the hydrological environment, and endangering human life and health through direct contact and the food chain [3]. Li investigated the heavy metal pollution situation in a coal mine in Henan Province and found the Zn, Pb, Cu and Cd in the surface soil exceeded the standard, showing a decreasing trend from the mine to the surrounding [4]. Obasi assessed the potential ecological risk of Pb-Zn mines in south-eastern Nigeria, noting that the concentration of heavy metal ions in water resources is $Pb^{2+} > Hg^{2+} > As^{2+} > Cd^{2+} > Cr^{2+} > Cu^{2+}$. And the concentration of heavy metal ions around the mining area exceeds the standard concentration of drinking water stipulated by WTO except for Cu^{2+}, causing serious physical and mental harm to residents [5]. Therefore, the study of soil heavy metals in coal gangue mining areas was of great significance to the prevention and protection of similar areas polluted by heavy metals.

In recent years, the biogeochemical characteristics of the soil-plant system in mining areas have been paid more attention to environmental monitoring, pollution control and ecological restoration in mining areas [6, 7]. Halim found that the contents of As, Cr and other heavy metals in rice in the Apululia coal mine area of Bangladesh seriously exceeded the standard [8]. Liu exhibited that some crops in the Xiaoqinling gold belt have high background values of heavy metals and have a high risk of exposure to heavy metals [9]. Therefore, using the crops around the coal gangue area to monitor the degree of soil pollution was one of the important methods to measure soil health and evaluate the quality of crops from an ecological perspective.

The occurrence state of heavy metals determined the soil heavy metals ‘toxicity and effectiveness. Hua showed that the soil heavy metals content in the gangue area of Tai'an almost exceeded local background value in Shandong, and the carbonate and exchange states of Pb and Cr accounted for a large proportion [10]. Jia characterised the heavy metal pollution characteristics around coal gangue hills in Qufu City, Shandong Province, by measuring the content of heavy metal elements around coal gangue hills, and evaluated the heavy metal potential ecological risk [11], while occurrence modes, biological effectiveness and the form of transfer and storage between soil and crop have not been studied in depth [12, 13].

In view of the above, we took the coal gangue hill in Qufu City, Shandong Province as the research object, by measuring the changes of the total amount of soil heavy metals at different distances from the gangue hill, the occurrence state and the physical and chemical properties of heavy metals, as well as heavy metals content in corn kernels. The pollution degree and spatial distribution of soil heavy metals around the gangue hill, as well as the bioconcentration and transfer, features characteristics of crops have also been investigated. The study provided a reference for the prevention and control of environmental pollution caused by coal gangue and the rational layout of agricultural land.

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II. MATERIALS AND METHODS

2.1 Collection and Preparation of Soil and Corn Samples

Soil samples pretreatment: This study selected the farmland soil around the coal gangue hill in Qufu as the research object. Centered on the gangue hill, samples were collected on the circumference of 0, 10, 50, 80, 100, 125, 150, respectively. There were 5 sampling points in each circle, which are collected into a complete sample. Soil samples were taken at a depth of 0-20 cm from the soil surface, then dried, crushed and passed through a 100 mesh sieve for subsequent analysis.

Corn samples pretreatment: Five corn of similar size were randomly collected from each sampling site to form a mixed sample, and the collected corn was hung in ventilated place to dry, then threshed and homogenized through a 100-mesh sieve. The cornflour of each sample is fully mixed and set aside.

2.2 Dissolution of Soil and Corn Samples

Accurately weight 0.5g soil samples or cornflour into digestion tube, added HNO_3 and H_2O_2 (volume ratio 3:1), mixed and soaked overnight. The digestion tube was placed in the digestion apparatus to raise the temperature to 90 ℃, and the heat preservation 30 min was used for pre-digestion treatment, and then the digestion was carried out until the sample solution became clear. At this time, the digestion was considered to be complete, and the tube cover was removed and heated to 160℃. When the solution in the tube has evaporated to 2 mL, the digester was switched off and cooled naturally. Finally, the solution was filtered into a 50 mL volumetric flask to fix the volume and stored it. Blank control was made in the same method.

2.3 Determination of Soil and Corn Samples

The contents of Cr, Pb, Cd, Hg, Cu, Zn, Ni in soil or corn were determined and compared with the soil background values and the control standard values in the soil Environmental quality-Agricultural Land soil pollution risk Control Standard (trial).

2.4 Occurrence Modes of Soil Heavy Metals

The Tessier five-step extraction method [14] using ICP-MS was used to analyze the exchangeable, carbon ate-bound, Fe-Mn oxide-bound, organic-bound and residue states of the soil heavy metals in sequence and were expressed as F1, F2, F3, F4 and F5 respectively.

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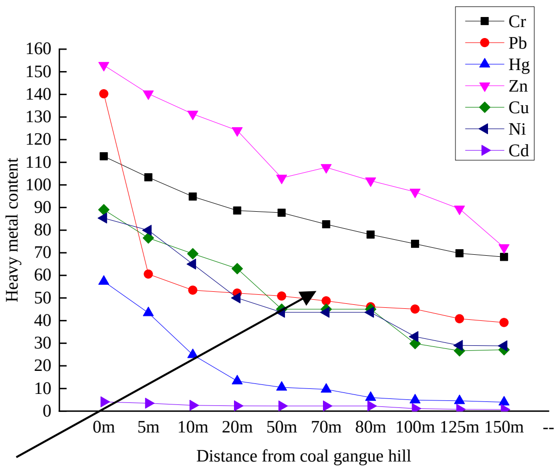
III. RESULTS AND ANALYSIS

3.1 Total Amount and Spatial Distribution of Soil Heavy Metals

As shown in Table I and Fig. 1, the total amount of Cr, Pb, Cd, Hg, Cu, Zn and Ni in agricultural soil around this gangue hill all surpassed the local soil background value content, with the total amount of Pb, Cd and Hg exceeding the control standard screening values of the national soil standards, and the highest exceeding times were 1.169, 13.587 and 23.876 times, respectively. This result demonstrated the risk of soil contamination in this region was extremely high and strict control measures should be taken. As the distance increased, the soil heavy metal content decreased significantly and eventually stabilises. As a whole, heavy metal contamination of agricultural soils around this gangue hill was prominent, which should be paid more attention to.

Table I. Heavy metal content in farmland, local soil background value and control standard screening value.

element	Maximum (mg/kg)	Minimum (mg/kg)	Average (mg/kg)	Standard deviation	Variable coefficient	Background value (mg/kg)	Control standard screening value (mg/kg)
Cr	112.611	68.119	85.951	2.04	1.29	65.106	200
Pb	140.247	39.135	57.714	5.48	1.61	30.798	120
Cd	4.076	0.695	1.748	0.74	2.09	0.233	0.3
Hg	57.303	3.953	17.847	1.55	1.36	3.641	2.4
Cu	89.045	27.11	51.699	1.92	2.27	25.875	100
Zn	152.877	72.329	111.986	3.75	3.52	64.973	250
Ni	85.31	28.865	50.197	2.81	2.66	26.938	100



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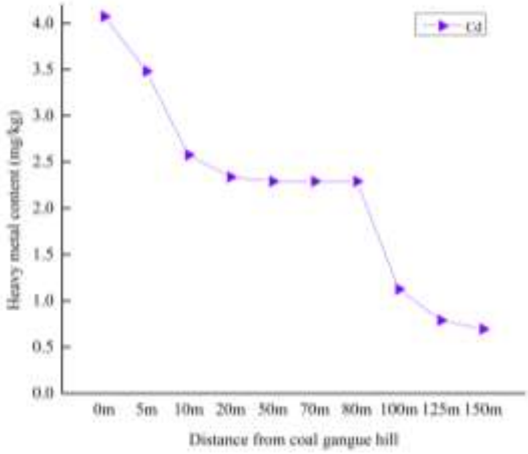

Fig 1: Folding line graph of the variation of various metals with distance

3.2 Occurrence Modes of Soil Heavy Metals

When exploring the pollution status of soil heavy metal, the gross of heavy metal were generally analyzed, but their migration ability in soil was mainly affected by occurrence modes [10, 15]. Furthermore, the occurrence modes of heavy metals had an influential role in the uptake and enrichment of heavy metals by plants.

The distribution of the occurrence modes of h soil heavy metals around the coal gangue hill with distance was shown in fig. 2. Cr, Ni and Cd were mainly in the residual state, which was not easily released or enriched by plant uptake under natural conditions and was relatively the most stable. Notably, the exchangeable state of Cd was between 11.48% and 17.42%. Exchangeable heavy metals can be dissolved through water flow and migration in soil and directly enter the ecological environment and food chain accumulation, thus affecting human health [10]. Cd with a higher exchangeable state in this area should be highly valued. Pb was predominantly in the residue and Fe-Mn oxide-bound states. Fe-Mn oxides were generally present as mineral epilimnion and finely powdered loose particles and were mainly influenced by pH and redox properties [16]. Hg was predominantly in the residue and organically bound state. Cu and Zn were predominantly in the residue, Fe-Mn oxide bound and organic-bound states. Organic bound heavy metals chelated with organic matter to form more stable substances [17]. The content of carbonate-bound in each metal was less than 9%. As the distance from the gangue hill increases, there was a decreasing trend in carbonate-bound and exchangeable heavy metals, with decreasing biological effectiveness and diminishing potential for harm.

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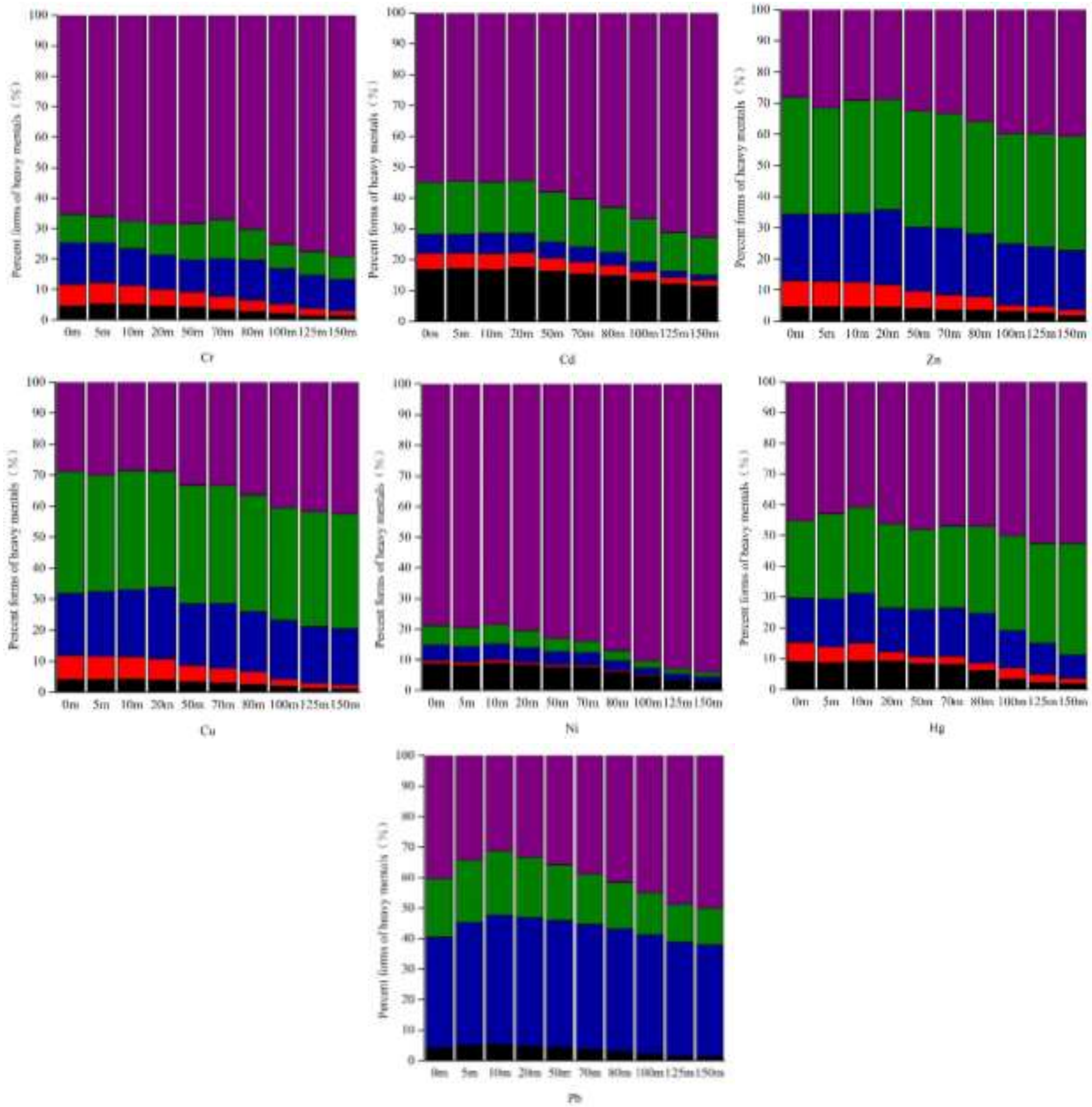

Fig 2: Percentage distribution of soil heavy metal occurrence modes with distance

3.3 Correlation Analysis between the Spatial Distribution of Soil Heavy Metals and Physical and Chemical Properties

The results of the correlation analysis between soil heavy metals and physico-chemical properties surrounding this coal gangue hill were shown in Table II. The analysis showed that Cr, Pb, Hg, Zn, Cu, Cd and Ni were positively correlated with pH. Many scholars argued that when pH decreased, the competition for H^+ sorption was reduced, which may lead to stronger binding of carbonates, organic matter and Fe-Mn

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oxides to heavy metals. Hence reducing their effectiveness, which to some extent hinders the uptake of heavy metals by plants.

Table II. Correlation between heavy metal content and physicochemical properties

	Cr	Pb	Hg	Zn	Cu	Cd	Ni
pH	0.783**	0.708*	0.885**	0.701*	0.709*	0.724*	0.803**
Cation exchange capacity	0.840**	0.817**	0.924**	0.755*	0.758*	0.822*	0.847**
organic matter	0.636*	0.488	0.793**	0.591	0.646*	0.613	0.758*

Note: **. Correlation significant at 0.01 level (two-tailed). *. Correlation significant at 0.05 level (two-tailed).

Cation exchange and soil heavy metal content all showed significant positive correlations [18, 19]. Cr, Hg, Cu and Ni were positively correlated with soil organic matter content, while Pb, Zn and Cd were not significantly correlated. This indicates that soil physicochemical properties influenced soil heavy metal content.

3.4 Analysis of Heavy Metal Content in Corn

Table III showed the content and enrichment level of heavy metals in corn kernels. Cr, Pb, Cd, Cu and Zn all exceeded the limit standard by a factor of 25.08, 85.7, 5.6, 2.289 and 1.4 respectively, except for Hg. The most serious exceeding was Pb, whose average content was 17.14mg/kg, about 85.7times higher than the limit standard, followed by Cr, with an average of 25.08 mg/kg, exceeding about 25 times. Noteworthily, owing to the strong mobility of Cd, it was easy to enter the human body under the action of the food chain when the gross of Cd was excessively large [20]. The average of Cd was 5.6 times of the limit standard, which should be paid more attention to. Hg and Ni were at high levels in corn kernels, although the limits were not exceeded. The coefficient of variation (CV) reflected the average change extent of content among all samples [21]. Among them, the coefficients of variation of Cr, Pb, Ni, Hg, Zn, Cu and Cd were all more than 36%, which were in a high variation, indicating that the spacial distribution of heavy metals around the coal gangue area varied considerably. The bioconcentration coefficients of these seven heavy metals in corn were Cd > Zn > Cu > Pb > Cr > Ni > Hg. To sum up, the chemical properties of heavy metals were essential elements in determining their transport and enrichment in soil-crop systems when identifying crop species. Cd, Zn and Cu were relatively active and easy to migrate and enrich from the soil, followed by Pb, Cr and Ni. Hg was mostly trapped in the soil, which was difficult to enrich into corn.

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Table III. Heavy metal content in maize kernels

Element	Maximum (mg/kg)	Minimum (mg/kg)	Average (mg/kg)	Standard deviation	Variable coefficient	Limitations	BCF	Standard
Cr	48.46	12.61	25.08	1.12	1.17	1	0.292	GB 2762-2017
Pb	28.53	15.84	17.14	2.07	1.22	0.2	0.297	GB 2762-2017
Cd	2.86	0.49	1.12	0.11	2.07	0.2	0.641	GB 2762-2017
Hg	0.043	0.006	0.011	0.0001	1.06	0.02	0.0006	GB 2762-2017
Cu	53.14	16.25	28.29	1.27	2.08	10	0.547	NY 861-2004
Zn	102.57	58.33	70.36	2.84	2.17	50	0.628	NY 861-2004
Ni	21.63	9.15	12.19	1.35	1.12	/	0.243	/

IV. DISCUSSION

In this study, the heavy metal contamination of farmland soil around this coal gangue area was more serious, Cr, Pb, Cu, Zn, Ni, Hg, Cd all exceed the local soil background value, which was consistent with Li's study [22]. Toxic heavy metals for instance Cd and Hg even exceed the screening value of control standards, so the remediation of heavy metal pollution has become an urgent problem to be addressed. And the levels of Cr, Cu, Zn, Ni, Hg and Cd all showed a slow downward trend with the increase of distance and gradually approached the soil background value. Pb decreased step by step in the initial stage and finally tended to be stable, indicating that the gangue hill had an impact on the contents of heavy metals in the neighboring farmland, and the effect gradually decreased with the increase of distance. In the natural environment, exchangeable carbonate-bound states could be transformed into organic-bound, Fe-Mn oxide-bound and residual states [23]. The morphological changes of seven heavy metals were consistent with this, in which Cu, Zn, Pb, Cr, Ni and Cd were gradually transformed to residual state, and Hg was gradually transformed into organic-bound state and residual state. Cr in the soil around the coal gangue was highly transferable [24]. Cr was mainly combined with carbonates, organic matter and oxides, and a large part of it existed in the soil solution, which would change the morphological distribution of Cr in the original soil [25]. The occurrence modes of heavy metals were mainly affected by the physicochemical properties, including cation exchange capacity, pH and organic matter content [26]. Carbonate-bound heavy metals dissolved and released metal ions under the condition of low pH [27]. The pH in this area was mainly weakly alkaline soil, but the closer to the coal gangue hill, pH was relatively rised, the proportion of carbonate states was less than 10%, and as the distance increases, the content of carbonate metals decreased gradually, demonstrating that pH of the area affected the content of carbonate heavy metals. The further away from the gangue area, the lower the pH and cation exchange, while the overall content in the Fe-Mn oxide-bound state of Cd, Hg and Ni showed a decreasing trend and the obvious correlation between these three heavy metals and pH and cation exchange capacity, indicating that with the decrease of pH and cation exchange capacity, the heavy metals were partially dissolved and converted to soluble state for plant uptake. Elevated organic matter in soils increased the solubility of some heavy

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metals and increase their uptake by plants [28], while soil organic matter had a significant positive correlation with Cr, Hg, Cu and Ni, so it was possible that the amount of these in soil was also influenced by organic matter leading to their reduction or were the result of a combination of both soil organic matter and plant uptake. It was noteworthy that soil organic matter has a significant complexation of mercury. When the organic matter content was high, the ability of the soil to fix mercury increases. Moreover, so far it had not been possible to eliminate the accumulation of mercury in living organisms, but only to reduce and control the transport and transformation of mercury in environmental systems [29]. Cd, Zn and Cu had strong migration ability and caught be absorbed and enriched by corn, which was mainly because the exchangeable and carbonate-bound states of Cd in soil were about 20%, both of which were unstable and easily absorbed by plants.

The carbonate states and exchangeable of Zn and Cu were nearly 10%. These two heavy metals were essential trace elements for plant growth, which might be the reason for the migration to plants and the high enrichment factor of plants. The excessive content in corn kernels of Pb might be caused by the high background value of soil. Cr was positively corroborated with cation exchange capacity, organic matter and pH, and the unstable state was about 10%. Cr in the soil around the coal gangue was highly transferable. Cr was mainly combined with carbonates, organic matter and oxides after entering the soil, at the same time a large part of which was present in the soil solution and would change the morphological distribution of Cr in the original soil, a large part of resulting in the easy migration of Cr into the plant, which was the cause for the accumulation of Cr within corn kernels. This results of this experiment provided a brief analysis of the soil heavy metal content and morphology in the gangue hill, as well as the enrichment of heavy metal migration by crops, which was of reference significance on the protection and management of heavy metal pollution contamination in the area.

V. CONCLUSION

(1) The total amount of heavy metals Cr, Pb, Cd, Hg, Cu, Zn and Ni around the farmland in the gangue hill exceeded the local soil background value, and average Hg content of was 17.847 $mg \cdot kg^{-1}$, which the risk screening value was 7.43 times and the risk control level was 4.9 times, and the mean amount of Cd was 1.748 $mg \cdot kg^{-1}$, which was 5.83 times of the risk screening value and 5.3 times of the risk control level, both of which seriously exceed the standard.

(2) Correlation analysis of soil heavy metals with physical and chemical properties showed that Cr, Pb, Hg, Zn, Cu, Cd and Ni were positively correlated with cation exchange, organic matter and pH showed significant correlations for Cr, Hg, Cu and Ni.

(3) The occurrence modes of the soil heavy metals revealed all seven heavy metals were predominantly in the residue state, and the proportion of residue state gradually increased to a stable level with increasing distance.

(4) The average levels of Cr, Pb, Cd, Cu and Zn in corn seeds all seriously exceeded the limits. The average value of Cr is 25.08 times the limit standard, and the average value of Pb is 85.7 times the limit standard.

VI. PROSPECT

This study investigated the correlation between total heavy metals in this gangue area, the occurrence modes patterns and the physicochemical properties of soils in the area, and the analysis of total heavy metals in corn. Although this study was comprehensive, the migration and enrichment mechanism of heavy metals was only generalized, as well as only corn seeds were studied, but not crop tissues, which can be further studied. In addition, studies on the transport and enrichment of heavy metals can be carried out on a variety of plants in the area, with a view to obtaining methods and approaches for the management of heavy metals in the area.

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