Enrichment Condition and Security Risk Assessment of Heavy Metals in Soil-Crops System around the Gangue Dumps

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Abstract. This paper aims to make a study of the distribution patterns, the enrichment characteristics and the corresponding contamination conditions caused by the heavy metal contents in the soil-crops (soybean, corn and rice) around the coal gangue dump. Samples of soils and food crops were collected around Xinji coal gangue dump by the intercept method. The concentrations of Zn, Cu, Pb, Cd, and Cr in the soils and food crops were analyzed by inductively coupled plasma optical emission spectrometer (ICP-OES). With a contrastive analysis done with the enrichment features of the root and the transfer ability of the seeds among the different food crops. In order to evaluate the pollution condition of the three corps, the single factor pollution index and the Nemerow comprehensive index. The results showed that the soil is alkaline, and the max concentrations of Zn, Cu, Pb, Cd, Cr in the soils at the depth of 20~60 cm, which are below the background values of Huainan soil. In the top soil (0~20 cm), Zn, Cu, Pb, Cd, Cr each their peak values at a distance away from the coal gangue dump, and then, with increasing distance from the coal gangue dump gradually decreased; the remnant concentrations of Zn, Cu, Pb, Cd and Cr in the different plant organs are also in the degrading order of root > stem > seed, but that of Pb in the leaves turns to be significantly higher; The absorptive ability of the corn roots for the heavy metals proves to be higher than soybean and rice whereas the seed transferring ability turns to be reversed, for such differences account for the different root features and the stem transferring ability of the three food crops. The Nemerow comprehensive index of the three food crops has been found and illustrated also in the degrading order of corn > rice > soybean, with corn being affected by the light pollution, rice and soybean affected at the warning level.

1. Introduction
Gangue is a solid waste produced in the process of coal mining and processing, and its output accounts for about 10%~20% of the raw coal [1]. Due to natural factors such as weathering and erosion and leaching of rainfall, a large number of heavy metal elements in gangue are released during the process of long-term stacking, and they migrate into atmosphere, water and soil and eventually pollute the farmland ecosystem around [2]. On the one hand, heavy metal contaminants affect the soil microbial flora, and then affect the structure and function of the ecosystem. On the other hand, they cause human health risks through the human food chain, by affecting the quality of crops such as cereals and
vegetables [3-5]. Therefore, heavy metal pollution in soil and grain crops has become a hot issue in the related fields [6-7].

The content of Cd, Cu, Mn, Ni, Pb and Sn in gangue dump in Huainan area has been beyond the soil background value [8-10], which has resulted in the possible cumulative pollution of heavy metals in the soil around gangue dump [11]. In related studies, there are many studies of heavy metal pollution of soil in Huainan coal mining areas [12-15], but few sample analysis for the relevant environmental media (soil, plant, etc.) there. Taking the gangue in Xinji Huainan as the object, this research explored the distribution characteristics, enrichment and transfer regulations of heavy metal elements in the natural cropland soil and food crops around the gangue dump, and then evaluated the status of heavy metal pollution of crops, in order to provide theoretical basis for the environmental protection as well as the safety assessment and risk assessment of food crops around the gangue dump in mining areas.

2. Materials and methods

2.1. Sample collection
The experimental samples were collected from Xinji Coal Mine in Huainan City, which has been put into production since July 1, 1993 and the designed annual capacity was 900,000t. According to the dominant wind direction and topographic characteristics of the mining area, gangue dump was taken as the center, and 90° fan-shaped points were arranged in the direction of the downwind from west to south of the dominant wind. Sampling was conducted respectively in the areas 300m, 600m, 900m, 1200m and 1500m from gangue, at the interval of 300m; and 6 sampling points were uniformly arranged in each fan-shaped area in the distance. Soil samples were collected at the vertical distance of 0~20cm, 20~40cm and 40~60cm at each sampling point; According to the conditions of food crops around the sampling points, 1~2 same plants were collected in the range of 1m, and their roots, stems, leaves, hulls and seeds were respectively preserved in the zip lock bags.

2.2. Treatment of samples
The collected soil samples were air-dried in the laboratory and divided into two parts after removing the plant residual and sundries. One soil sample was crushed and sieved, and then treated by the method of digestion with aqua regia reflux for content determination of heavy metal elements. The other soil sample was crushed and sifted to determine the physicochemical properties of the soil. After being washed in deionized water, the crop samples were dried, ground, sifted and treated by the method of sulfuric acid -- nitric acid -- perchloric acid. The content test of elements adopted ICP-OES. In the process of experimental testing and analysis, the national standard soil sample (GBW07403 (GSS-3)) and standard plant sample (GBW07603 (GSV-2)) were added respectively for quality control. The recovery of Zn, Cu, Pb, Cd and Cr in the standard soil sample was 95.10%, 97.09%, 92.58%, 92.70% and 97.14%, respectively. The recovery of Zn, Cu, Pb, Cd and Cr in the standard plant sample was respectively 96.08%, 94.35%, 93.59%, 93.68% and 98.45%; The relative standard deviation of heavy metals in all tests was less than 5%. Routine methods were used to measure the soil organic matter, pH, conductivity, total nitrogen, phosphorus and potassium [16].

2.3. Research methods
What were adopted in this study were the evaluation method of the single factor index and Nemerow comprehensive index [17].

According to China's agricultural industry standard NY 861-2004, The Limitation of Eight Elements (Pb, Cd, Cr, Hg, Se, As, Cu and Zn) in Foodstuff (Including Cereals, Beans and Potatoes), the limitation of elements and grading standards of pollution indexes are shown in Table 1 and Table 2, respectively.
Table 1. Limiting criterion of heavy metal elements in foodstuff [18]

| Crop type     | Zn (mg·kg⁻¹) | Cu (mg·kg⁻¹) | Pb (mg·kg⁻¹) | Cd (mg·kg⁻¹) | Cr (mg·kg⁻¹) |
|---------------|--------------|--------------|--------------|--------------|--------------|
| Cereal crops  | 50           | 10           | 0.4          | 0.05         | 1.0          |
| Pulse crops   | 100          | 20           | 0.8          | 0.2          | 1.0          |

Table 2. Grading standards of single factor index and Nemerow comprehensive index [18]

| Single factor index | Pollution level | Nemerow comprehensive index | Pollution level |
|---------------------|----------------|-----------------------------|----------------|
| P₁≤11               | Clean          | Pₙ<0.7                      | Safety         |
| 1<P₁<22             | Slight pollution| 0.7≤Pₙ<1                   | Alert level    |
| 2≤P₁<33             | Moderate pollution| 1≤Pₙ<2                    | Slight pollution|
| P₁≥33               | Heavy pollution| 2≤Pₙ<3                     | Moderate pollution|
| Pₙ≥3                | Heavy pollution|                            |                |

3. Study results and analysis

3.1. Analysis of soil physicochemical properties in the studied area

It can be seen from the Table 3 that, the soil pH in the studied area is more than 8.00, which indicates that the soil there is alkaline. The pH value and conductivity of soil increase with soil depth. The total nitrogen content of soil increases gradually with the increase of soil depth, but it is not significant (P>0.05). The total phosphorus content of soil decreases with soil depth increasing (P<0.01).The total potassium content reaches the lowest point at the depth of 0~20cm, and it increases with increase of soil depth (P<0.01).The soil organic matter in the studied area decreases gradually from 0 to 60cm, but has no significant relationship with soil depth (P>0.05).

Table 3. Physicochemical properties of the soils around gangue dump from Xinji Coal mine (mean ± SD)

| Depth/cm | PH value | Conductivity/ (µs·cm⁻¹) | Total nitrogen/ (mg·kg⁻¹) | Total phosphorus/ (mg·kg⁻¹) | Total potassium/ (g·kg⁻¹) | Organic matter/ % |
|----------|----------|--------------------------|---------------------------|-----------------------------|----------------------------|-------------------|
| 0~20     | 8.04±0.61| 226.55±74.93             | 1943.43±1368.61           | 971.19±320.46              | 9.59±1.72                 | 2.59±1.34         |
| 20~40    | 8.29±0.51| 238.25±64.43             | 1983.67±932.46            | 919.30±419.77              | 10.03±2.94                | 2.22±1.14         |
| 40~60    | 8.49±0.29| 245.94±47.82             | 2439.74±968.10            | 842.14±432.90              | 9.68±2.59                 | 2.04±1.73         |

3.2. Heavy metal distribution in the soil of studied area

As can be seen from the Table 4 that, the contents of Zn, Cu, Pb, Cd and Cr in the soil from Xinji mine lots are below the soil background value in Huainan and do not meet the primary standard of the national soil environmental quality. In the soil profile, the concentrations of Zn, Pb and Cu in soil reach the highest point at the depth of 20~40 cm, which are 43.08 mg·kg⁻¹, 15.61 mg·kg⁻¹ and 24.27 mg·kg⁻¹ respectively. However, with the increase of soil depth, the concentrations of Zn, Pb, Cu and Cr in soil all decrease to some extent. Cd in the soil reaches highest at the depth of 0~20cm. With the increase of soil depth, Cd in soil decreases to some extent, but the concentration at 20~40cm is more than that at 40~60cm. Correlation analysis shows that there is no significant correlation between the content of Zn, Pb, Cd, Cu and Cr with the soil depth (P>0.05).

From the Table 4, it can also be seen that the content of Cd in soil is relatively high, ranging from 0.111 to 0.124 mg·kg⁻¹, which has been beyond the soil background value of Huainan, namely 83.00%~106.76%. In the range of soil depth of 20~60 cm, the content of Cu is 24.18~24.27 mg·kg⁻¹, exceeding the soil background value of 0.08%~0.46% in Huainan. It can be concluded from the above analysis that there is no obvious heavy metal pollution in the soil of Xinji mining lots.
Table 4. Total concentrations of Zn, Cu, Pb, Cd and Cr in soils (mg·kg⁻¹) (mean ± SD)

| Soil depth (cm) | Zn       | Cu       | Pb       | Cd       | Cr       |
|----------------|----------|----------|----------|----------|----------|
| 0–20           | 40.03±7.45 | 21.89±4.23 | 14.19±4.51 | 0.124±0.056 | 59.62±9.45 |
| 20–40          | 43.08±8.48 | 24.27±8.81 | 15.61±3.80 | 0.122±0.074 | 59.93±10.48 |
| 40–60          | 40.92±9.90 | 24.18±8.39 | 13.61±3.71 | 0.111±0.075 | 60.01±10.01 |

Soil background value in Huainan [19]  
National standard of soil environmental quality (Grade I) [19]

| Soil depth (cm) | Zn       | Cu       | Pb       | Cd       | Cr       |
|----------------|----------|----------|----------|----------|----------|
| 0–20           | 40.03±7.45 | 21.89±4.23 | 14.19±4.51 | 0.124±0.056 | 59.62±9.45 |
| 20–40          | 43.08±8.48 | 24.27±8.81 | 15.61±3.80 | 0.122±0.074 | 59.93±10.48 |
| 40–60          | 40.92±9.90 | 24.18±8.39 | 13.61±3.71 | 0.111±0.075 | 60.01±10.01 |

Under natural conditions, the gangue dump in coal areas has an impact on the heavy metal content in nearby soil. In this research (as can be seen in the Table 5), the highest contents of Zn, Cu, Pb and Cd are not in the soil closest to the gangue dump, and only the Cr content in the soil near the gangue dump reaches the highest point. The contents of Zn and Cu in the top soil (0–20cm) peak in the sample 600m far from the gangue dump; the contents of Pb and Cd reach the peak 300m from the gangue dump; the content of Cr peaks in the sample 50m from the gangue dump; and then, the content of heavy metals are reduced with the increase of distance from the gangue dump.

Table 5. Relationship between the contents of Zn, Cu, Pb, Cd, Cr (mg·kg⁻¹) in the top soils (0–20 cm) and the distance

| Distance (m) | Zn   | Cu   | Pb   | Cd   | Cr   |
|--------------|------|------|------|------|------|
| 50           | 42.65 | 21.18 | 14.67 | 0.138 | 62.52 |
| 100          | 38.53 | 22.31 | 15.01 | 0.058 | 61.21 |
| 300          | 46.49 | 25.2  | 17.48 | 0.176 | 60.58 |
| 600          | 50.32 | 27.69 | 16.04 | 0.166 | 57.89 |
| 900          | 34.78 | 25.03 | 11.26 | 0.110 | 54.68 |
| 1200         | 46.36 | 21.06 | 16.03 | 0.168 | 49.37 |

3.3. The distribution of Zn, Cu, Pb, Cd and Cr in three kinds of food crops

The same heavy metal element distributes differently in different plant organs, and different heavy metal elements distribute differently in different organs of the same plant. As can be seen from the Table 6, the mass ratio of each heavy metal element among the three kinds of food crops, namely soybean, corn and rice, is significantly different. Specifically, among the three kinds of food crops studied, the mass ratio of each heavy metal element in corn is the highest; The mass ratio of Pb, Cd and Cr in the root, stem, leaf, hull and seed is also the highest in corn; for the soybean, Cd is mainly distributed in the root; Zn, Pb and Cu are more likely to migrate to above-ground parts of soybean, especially Zn and Cu are easy to migrate to the seed of soybean; to rice, Pb, Cd and Cu are most abundant in the root, and only Zn is more distributed in the stem.

In general, the heavy metal elements that crops absorb from soil first accumulate at the root, and then some of the heavy metals are transported to other parts of the crop body through various transport routes, and thus the content of heavy metal elements in crop roots is often higher than that in other organs.[20] But the mass ratio of some heavy metal elements in the leaf obviously exceeds that in the root; for example, the mass ratio of Pb in corn and soybean leaves is higher than that their roots. On the one hand, this anomaly may be due to the selective absorption of different elements by leaves during the transport of heavy metals from the root of crops, thereby leading to the over-accumulation of some heavy metal elements in the leaves.[21] On the other hand, it is also possible that the pores on the surface of crop leaves can also absorb harmful heavy metal elements such as Pb and Hg in the polluted air; [22] however, other factors such as chemical fertilizer, pesticides and dust-fall might lead to the mass ratio increase of heavy metal elements in leaves.
Table 6. The concentrations of Zn, Cu, Pb, Cd and Cr in the root, stem, leaf, hull and grain of soybean, corn and rice (Dry weight, mean±SD)

| Plant   | Organ | Zn    | Cu    | Pb    | Cd    | Cr    |
|---------|-------|-------|-------|-------|-------|-------|
| Soybean | Root  | 9.22±2.53 | 7.89±1.94 | 0.78±0.55 | 0.048±0.032 | 3.31±1.36 |
|         | Stem  | 11.16±5.12 | 5.91±2.62 | 0.64±0.32 | 0.039±0.033 | 1.21±0.08 |
|         | Leaf  | 21.88±3.24 | 5.88±1.44 | 0.92±0.57 | 0.040±0.037 | 1.15±0.32 |
|         | Hull  | 14.52±2.89 | 4.82±1.30 | 0.47±0.43 | 0.018±0.015 | 0.41±0.10 |
|         | Seed  | 30.38±8.09 | 8.38±1.09 | 0.48±0.41 | 0.018±0.041 | 0.38±0.09 |
| Corn    | Root  | 30.08±4.29 | 26.82±3.85 | 1.42±0.75 | 0.34±0.15 | 6.62±2.36 |
|         | Stem  | 21.48±1.24 | 17.63±3.21 | 0.91±0.11 | 0.11±0.08 | 1.51±0.63 |
|         | Leaf  | 27.82±2.85 | 19.72±7.12 | 1.72±0.63 | 0.24±0.07 | 1.84±0.97 |
|         | Hull  | 11.16±0.92 | 3.44±0.84 | 0.81±0.22 | 0.07±0.01 | 0.69±0.18 |
|         | Seed  | 9.22±0.53  | 3.23±0.77 | 0.78±0.17 | 0.07±0.01 | 0.62±0.13 |
| Rice    | Root  | 20.12±0.24 | 11.69±3.16 | 1.22±0.79 | 0.20±0.20 | 2.53±1.05 |
|         | Stem  | 21.09±2.12 | 5.85±2.01 | 0.18±0.15 | 0.011±0.010 | 0.93±0.15 |
|         | Leaf  | 19.88±3.14 | 4.17±1.06 | 0.19±0.20 | 0.018±0.017 | 0.53±0.05 |
|         | Hull  | 18.62±4.12 | 3.87±0.97 | 0.22±0.10 | 0.008±0.007 | 0.23±0.04 |
|         | Seed  | 17.38±2.09 | 3.89±1.18 | 0.50±0.41 | 0.011±0.003 | 0.20±0.02 |

3.4. Heavy metal enrichment and transport conditions of three kinds of food: crops soybean, corn and rice

Enrichment coefficient is one of the indicators to evaluate the ability of plants to enrich heavy metals. The enrichment coefficients of Zn, Cu, Pb, Cd and Cr in the root of three kinds of food crops, namely soybean, corn and rice, were calculated respectively in this research (As shown in the Table 7).

It can be seen from the Table 7 that, the enrichment coefficients of Zn, Cu, Pb, Cd and Cr were different in the root of the soybean, corn and rice. The enrichment coefficients in the root can be presented as Cd> Zn> Cu> Pb> Cr, indicating that Cd is more easily absorbed by crop roots. The enrichment coefficient in corn is the highest among the three kinds of food crops. This may be due to the fact that the heavy metals in the studied area are mainly concentrated in the top soil, and the root system of corn can absorb the water and trace elements more fully because its roots mainly distributed in the top soil.

Table 7. Enrichment coefficient of heavy metals in soybean, corn and rice

| Enrichment coefficient in the root | Zn   | Cu   | Pb   | Cd   | Cr   |
|-----------------------------------|------|------|------|------|------|
| Soybean                           | 0.24 | 0.375| 0.013| 0.806| 0.023|
| Corn                              | 0.891| 0.646| 0.106| 0.943| 0.062|
| Rice                              | 0.5  | 0.53 | 0.07 | 1.31 | 0.084|

The transfer coefficient in crop seed is to evaluate the ability of crops to transport heavy metal elements from the underground to the seed. The transfer coefficients of Zn, Cu, Pb, Cd and Cr in the three kinds of food crops, namely soybean, corn and rice, were calculated respectively in this research (as shown in the Table 7). As can be seen from the Table 8, the seed shows relatively high transfer coefficients of Cu and Pb, indicating that Cu and Pb are more likely to accumulate in crop seed. Although the enrichment coefficient in the corn root is high, the transfer coefficient in seed is lower than that of soybean. This is because the stalks of corn are relatively high, and thus it is more difficult for heavy metals to migrate upward during the transport of elements from root to the seed.
Table 8. Transfer coefficient of heavy metals in soybean, corn and rice

|          | Zn   | Cu   | Pb   | Cd   | Cr   |
|----------|------|------|------|------|------|
| Soybean  | 0.57 | 0.898| 0.568| 0.542| 0.113|
| Corn     | 0.212| 0.119| 0.515| 0.200| 0.084|
| Rice     | 0.102| 0.107| 0.014| 0.265| 0.017|

3.5. Assessment of heavy metal pollution of soybean, corn and rice

The table 9 shows the single factor pollution index of heavy metals and the Nemerow comprehensive pollution index of seed of soybean, corn and rice.

As can be seen from the Table 9, single factor pollution index of heavy metal elements tends to be the same in these three kinds of food crops. Pb is the highest, followed by Cd, Zn, Cu and Cr which were all lower than 1, indicating that the seed of the three kinds of food crops had not been polluted by Zn, Cu and Cr. The pollution index of Cd in corn seed is 1.062, belonging to the slight pollution; but its pollution indexes in the seeds of soybean and rice are 0.832 and 0.936 respectively, which shows that it was at the alert level although it was not polluted. The pollution indexes of Pb in the seeds of corn, soybean and rice respectively are 1.635, 1.012 and 1.214, all belonging to the slight pollution. The comprehensive pollution indexes of rice and soybean are 0.891 and 0.841 respectively, which are at the alert level. The comprehensive pollution index of corn is 1.311, which belongs to the slight pollution.

Table 9. Assessment results of heavy metal pollution in soybean, corn and rice near gangue dump

| Crop type | Soybean | Corn | Rice |
|-----------|---------|------|------|
| Number of samples | 12 | 8 | 10 |
| Pi | | | |
| Zn Range | 0.231–0.356 | 0.682–1.012 | 0.398–0.872 |
| Average value | 0.254 | 0.887 | 0.488 |
| Cu Range | 0.749–0.832 | 0.189–0.321 | 0.876–0.983 |
| Average value | 0.811 | 0.249 | 0.891 |
| Pb Range | 0.869–1.213 | 1.056–2.213 | 0.909–1.413 |
| Average value | 1.012 | 1.635 | 1.214 |
| Cd Range | 0.752–0.934 | 0.735–1.286 | 0.857–1.134 |
| Average value | 0.832 | 1.062 | 0.936 |
| Cr Range | 0.271–0.386 | 0.388–0.912 | 0.373–0.586 |
| Average value | 0.318 | 0.534 | 0.414 |
| PN | 0.841 | 1.311 | 0.891 |
| Pollution level | Alert level | Slight pollution | Alert level |

4. Conclusion

The research result shows that the content of Zn, Pb and Cr in the soil around the gangue dump in the studied area is lower than the background value in Huainan City. In the soil depth of 20-60cm, the contents of Cd and Cu are higher than the background value of soil of Huainan City, but the contents of Zn, Cu, Pb, Cd and Cr in soil are lower than the primary standard of national soil environmental quality. The contents of Zn, Cu, Pb, CD and Cr in the surface layer of soil reaches its peak value after a certain distance from the gangue dump. They first increase with increasing distance. After reaching a certain distance, they all tend to decrease with the increase of the distance between the sampling location and the gangue dump. It is speculated that the situation may be caused by the dust particles which are produced by the coal gangue hill with aerial migration.

The mass ratio of heavy metal elements in the organs of soybean, corn and rice presents the degrading order as root> stem> shell> seed. This is because the heavy metal elements first accumulate in the root system when they migrate from soil to crops, and then gradually transfer to the leaves and
seeds through conducting tissue in stem. However, the mass ratio of some heavy metal elements is obviously higher in leaves, which may be related to other factors such as chemical fertilizer, pesticides and dust reduction. The exact reasons remain to be confirmed.

The single factor pollution index and the Nemerow comprehensive pollution index were used to evaluate the pollution of heavy metals in the seed of corn, rice and soybean. As for the three kinds of corps around gangue dump in the Huainan mine lots, the mass ratio of Pb and Cd in corn exceeds the prescribed limits; and soybeans and rice are at the alert level.

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