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Assessment of Heavy Metal Contents and Pollution Risk in Reclaimed Soils of a Bauxite Mine

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Abstract: A study on the contents and pollution state of heavy metals in the soils at the Xiaoyi Bauxite Mine in Shanxi, China was conducted to provide a scientific basis for understanding and evaluating the risks of heavy metal pollution in reclaimed soils in mining areas. The contents of Cd, Cr, Cu, Pb, As, and Ni were analyzed by inductively coupled plasma mass spectrometry (ICP-MS), and evaluated with respect to the changes in their characteristics over different reclamation years. Using a single factor index and the Nemerow Pollution Index, the potential degree and risk of pollution were assessed. The mean concentration of Cd was 5.19 mg/kg, which exceeds the 0.3 mg/kg national standard in China by 17 times, while the concentrations of other elements did not exceed the national standard and there was no apparent pollution risk. With the extension of the reclamation time, the contents of Cd increased in the 0–15-cm layer and decreased in the 15–30-cm layer. Cadmium possessed the highest single pollution index, and exceeded the severe pollution level (Level 5), while the other five elements were at safe levels (i.e., lower than Level 1). The Nemerow Pollution Index ranged from 12.08–13.14, with an average of 12.43, and exceeded Level 5 pollution by 3–4 times, indicating a severe level of pollution. The soil being used to reclaim the mine was the main source of pollution, with the contents of six heavy metal elements exceeding the national standards. The soils in the reclamation area contain Cd pollution from the resource soil, manure, and dust. Therefore, for mine reclamation, the quality of the soil being used to reclaim the land should been stringently control.

Keywords: Reclamation, Cd, Heavy Metal, Single Factor Index, Nemerow Pollution Index

1. Introduction

With increasing social and economic development, the demand for mineral resources has also increased. Mineral exploitation also leads to a series of serious environmental problems. For example, the geological and geomorphological destruction of mining areas, water quality deterioration, loss of vegetation species diversity, soil pollution by heavy metals, soil erosion, and desertification are becoming increasingly serious [1–2]. This is especially true for heavy metal pollution of soils, which has attracted global research attention [3–7].

Heavy metals, as a persistent and potentially toxic pollutant, will remain for a long time once they enter the environment, as they cannot be biodegraded, and will accumulate continuously by enrichment, thus endangering biological and ecological security [8–13]. In order to reuse the lands of mining areas, they are usually covered with soil to reclaim the area. It is necessary to study the quality of these lands and to evaluate the states of heavy metals in the soils of reclaimed mines. This is especially important for determining the reclamation methods and land management measures employed. In this study, the soils from the Xiaoyi Bauxite Mine in the province of Shanxi was used to evaluate heavy metal pollution and the potential risk after different periods (years) of reclamation.

2. Material and Methods

2.1. Test Site Overview

The Xiaoyi Bauxite Mineis located in the town of Yangquanqu (37°07'44–37°05'12"N, 111°29'52'–111°27'12"E) in the city of Xiaoyi, the capital of the mountainous province of Shanxi in western China. The Xiaoyi Bauxite Mine is the largest open-air bauxite mine in

China. The production is 2.15 x 10^6 t/a, with an area of 1158.2 hm^2. After mining, the land is mainly reclaimed through soil coverage. By the end of 2015, 422 hm^2 of land had been reclaimed, and a total of 98 hm^2 had been greened. The mining area is located in a loess hilly region, and represents a typical continental semi-arid climate zone, with windy springs, hot summers, rainy autumns, and cold, dry winters. The highest temperature was 37°C and the lowest temperature was under-20°C. Rainfall is mainly concentrated from July to September, with an average rainfall of 529 mm. The reclamation area of the Xiaoyi Bauxite Mine is mainly distributed across five areas, and the reclamation time differs in each area. The natural geomorphological conditions and reclamation mode are similar and the same, respectively, and the reclaimed soil is a calcite loess transported from nearby. According to the period of land reclamation and the modelling of major vegetation allocations, 5 typical reclamation sample lands were selected, each of which had an area of~0.5hm^2. The soil profiles are described in Table 1.

Table 1. *Soil sampling sites.*

Sample number	Sample name	Reclamation years (a)	Soil type	Soil texture	Vegetation types reclaimed
#1	Phase 2, #3	2	Lishi loess	Sandy loam	Walnuttree
#2	Phase 2, # 2	4	Lishi loess	Sandy loam	Walnuttree
#3	Phase 2, #1	6	Lishi loess	Light loam	Corn
#4	Phase 1, #2	8	Lishi loess	Light loam	Corn
#5	Phase 1, #1	10	Lishi loess	Light loam	Corn

2.2. Soil Sampling and Treatment

In April of 2015, samples were collected from the selected sites. Each sample was collected according to the "S" type at the level of 0–5 cm and 15–30 cm. After sample collection, the multi-point soil samples from the same depths were mixed, and the roots, rocks, and residues of plants in the soil samples were removed. Samples were stored in ice boxes and taken to the laboratory for natural drying. After the samples passed through a 100mesh screen, they were quartered and held for reserve.

2.3. Heavy Metal Determination

Arsenic content was assessed using silver diethyl dithiocarbamatespectrophotometry. Cadmium, Cr, Cu, and Pb were measured using an inductively coupled plasma mass spectrometer (Thermo 6300) after digestion by a strong acid. Heavy metal contents were recorded as the total amount, and the data used the mean value after 3 replications. Calculations were performed in Microsoft Excel v. ##.

2.4. Heavy Metal Evaluation

The single factor index is one of the most widely used methods for evaluating the degree of pollution of a certain pollutant in soils [14]. The calculation for this index is as follows:

$$P_i = {}^{C_i}\!/_{S_i}, \tag{1}$$

where P_i is the single factor pollution index of a given pollutant i, C_i is the measured concentration of pollutant i (mg/kg) and S_i is the criteria for assessment of the pollutant (mg/kg). The Level 2 soil environmental quality standard, Gb15618-1995, was selected as the evaluation standard for soil pollution (Table 2).

Compared with the singlefactor index, the Nemerow Pollution Indexcan comprehensively reflect the average pollution level of various pollutants in the soil, thereby highlighting the environmental damage caused by the most prominent pollutants [15]. The formula for this integrated index is as follows:

Table 2. *Soil environmental quality standard values.*

Heavy metal	Level 1	Level 2			Level 3
	pH natural background	pH<6.5	6.5≤pH≤7.5	pH>7.5	pH>6.5
Cd	0.2	0.3	0.3	0.6	1
Cr	90	150	200	250	300
Cu	35	50	100	100	400
Pb	35	250	300	350	500
As	15	40	30	25	40
Ni	40	40	50	60	200

$$P = \sqrt{{}^{(P_{jmax}^2 + P_{jave}^2)}\!/_{2}}, \tag{2}$$

where P is the Nemerow Pollution Index, P_{jmax} is the maximum from the single factor pollution index of the j monitoring point, and P_{jave} is the average value of the single factor pollution index of all pollutants at the monitoring point j. Soil heavy metal pollution can be divided into 5 levels according to the single factor index and the comprehensive index of Nemerow (Table 3).

Table 3. *Pollution classification standards for heavy metals.*

Level	Single factor pollution index	Nemerow Pollution Index	Pollution levels	Pollution level
1	≤0.7	≤0.7	Secure	Clean
2	0.7-≤1.0	0.7-≤1.0	Alert limit	Generally clean
3	1.0-≤2.0	1.0-≤2.0	Light pollution	Light pollution
4	2.0-≤3.0	2.0-≤3.0	Moderate pollution	Moderate pollution
5	>3.0	>3.0	Heavy pollution	Serious pollution

3. Results

3.1. Analysis of Heavy Metal Content in Reclaimed Soilsover Different Years

Figure 1 shows the statistical results of heavy metals in soils at 0–30 cm in depth after different periods (years) of reclamation in the Xiaoyi Bauxite Mine. It can be seen that the Cd concentrations at 0–15 cm were 5.04~5.27 mg/kg, with an mean of 5.11 mg/kg, while concentrations at depths of 15–30 cm were 5.05~5.49 mg/kg, with a mean of 5.27 mg/kg. The contents of Cd in the two layers were more than 10 times higher than the Level 2 national standard of 0.3 mg/kg for environmental soils. Therefore, the soils have been polluted by Cd. The Cd content at 0–15 cm has gradually decreased with reclamation time, while that at 15–30 cm has gradually increased over the same period of time.

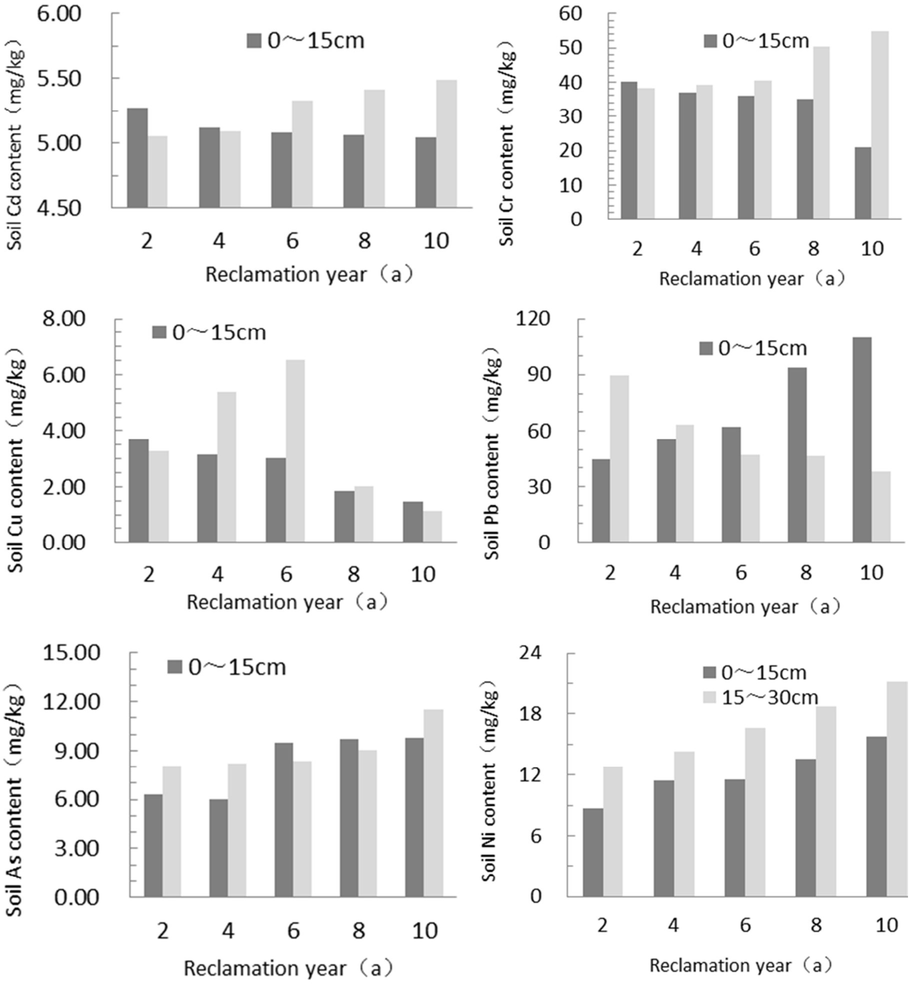

Figure 1. *Contents of soil heavy metals in the reclamation area: (a) description, (b) description, (c) description, (d) description, (e) description, and (f) description.*

Chromium contents in the 0–15-cm layer were 20.89~40 mg/kg, with a mean of 33.78 mg/kg, while at 15–30 cm, they were 38.08~54.88 mg/kg, with a mean of 44.56 mg/kg. Concentrations of Cr in the two soillayers did not exceed the Level 2 national standard of 200 mg/kg for environmental soils, and there was no pollution risk to the soils. Chromium contents in the 0–15-cm layer declined with reclamation time, but the rate of decline was slow, while in the 15–30-cm layer, concentrations gradually increased with reclamation time.

The concentrations of Cu in the 0–15-cm layer were 1.45~3.70 mg/kg, with a mean of 2.64 mg/kg, while in the 15–30-cm layer, they were 1.12~6.54 mg/kg, with a mean of 3.67 mg/kg. The Cu content in the two layers did not exceed the 100 mg/kg Level 2 national second for environmental soil quality, and did not cause pollution. The concentration of Cu in the 0–15-cm layer decreased gradually and uniformly with the extension of reclamation time, while in the 15–30-cm layer, concentrations fluctuated and increased gradually between years 2 and 6, before gradually decreasing between years 6 and 10.

The concentrations of Pb at 0–15 cm were 44.86–110 mg/kg, with a mean of 73.23 mg/kg. The Pb contents in the 15–30-cm layer were 38.23~89.66 mg/kg, with a mean of 57.04 mg/kg. Lead concentrations in the two layers did not exceed the Level 2 national standard value of 300 mg/kg for environmental soils, and did not pose a pollution hazard to the soil. The Pb content in the 0–15-cm layer exhibited a gradually decreasing trend with reclamation time, while the 15–30-cm layer had a gradually increasing trend.

The contents of As in the 0–15-cm layer were 6.33~9.82 mg/kg, with a mean value of 8.26 mg/kg, while in the 15–30-cm layer, they were 8.04~11.49 mg/kg, with a mean of 9.01 mg/kg. In the two layers, the contents of As did not exceed the Level 2 national standard of 30 mg/kg for environmental soils, and did not pose a pollution hazard to the soils. The contents of As in the 0–15- and 15–30-cm layers gradually increased with reclamation time, and their rates of increase were similar.

The concentrations of Ni at 0–15 cm were 8.68~15.78 mg/kg, with a mean value of 12.19 mg/kg, while at 15–30, they were 12.85~21.17 mg/kg, with a mean of 16.72 mg/kg. In the two layers, the contents of Ni did not exceed the 50 mg/kg Level 2 national standard for environmental soils, and did not pose a pollution hazard to the soils. The concentrations of As in both the 0–15- and 15–30-cm layers gradually increased with reclamation time, and their rates of increase were similar. Based on the results of these analyses, after 2~10 years of reclamation, only Cd exceeded the Level 2 national standard in both layers, resulting in the pollution of the soil, while the other elements did not pollute the soil.

3.2. Assessment Results Using a Single Factor Index

Table 4 shows the contamination assessment results for heavy metals in the soils of the Xiaoyi Bauxite Mine in different reclamation years, based on the single factor index. According to these results, only the Cd pollution index (P_i) was greater than 3.0, reaching Level 5, the level of serious contamination. This is consistent with the results of heavy metal contents in the soil. The pollution indices of the other five heavy metals were all less than the 0.7 value of the standard (Level 1), and at a safe cleaning level. The pollution index (P_i) values, were ordered as Cd > Cr > As >Pb> Ni > Cu.

As shown in Table 4, the pollution index (P_i) of Cd ranged from 16.81~18.30, with an average of 17.32, both of which were higher than the Level 5 value, indicating a serious pollution level. The P_i of Cr ranged from 0.10~0.24, with an average of 0.20, which was less than that required for Level 1 classification, indicating a safe and clean level. The P_i of Cu ranged from 0.01–0.07, with an average of 0.03, which was less than that required for Level 1 classification, indicating a safe and clean level. The P_i of Pb ranged from 0.37~0.13, with an average of 0.223, which was less than that required for Level 1 classification, indicating a safe and clean level. The P_i of As ranged from 0.20–0.38, with an average of 0.29, which was less than the pollution index required for Level 1 classification. The pollution level was therefore considered safe and clean. The P_i of Ni was between 0.17 and 0.42, with an average of 0.29, which was less than that required for Level 1 classification, indicating a safe and clean level.

Table 4. *Evaluation results of soil heavy metal using the single factor index.*

Serial number	Level/cm	Single factor pollution index (P_i)						Rating	Degree of pollution
		Cd	Cr	Cu	Pb	As	Ni		
1	0–15	17.56	0.20	0.04	0.15	0.21	0.17	5	Serious pollution
2	15–30	16.85	0.19	0.03	0.30	0.27	0.26	5	Serious pollution
3	0–15	17.05	0.18	0.03	0.19	0.20	0.23	5	Serious pollution
4	15–30	16.98	0.20	0.05	0.21	0.27	0.29	5	Serious pollution
5	0–15	16.94	0.18	0.03	0.21	0.32	0.23	5	Serious pollution
6	15–30	17.75	0.20	0.07	0.16	0.28	0.33	5	Serious pollution
7	0–15	16.88	0.17	0.02	0.31	0.32	0.27	5	Serious pollution
8	15–30	18.02	0.25	0.02	0.16	0.30	0.37	5	Serious pollution
9	0–15	16.81	0.10	0.01	0.37	0.33	0.32	5	Serious pollution
10	15–30	18.30	0.27	0.01	0.13	0.38	0.42	5	Serious pollution

Based on the results presented in sections 3.1 and 3.2, the concentrations of Cd in the reclaimed soils of the bauxite mine over different periods (years) of reclamation remain a serious source of contamination. Conversely, the concentrations of Cr, Cu, Pb, As, and Ni five were all atsafe and clean levels, and did not cause environmental damage. Table 5 shows the assessment results of soil heavy metal contamination in the Xiaoyi Bauxite Mine across different years based on the Nemerow Pollution Index. It can be seen that for this more comprehensive index (P), values at 0–15 cm and 15–30 cm ranged between 12.08 and 13.14, with an average of 12.42, which is four times greater than the single factor pollution index of 3.0. The value is therefore classified at a level of 5, indicating a serious level of contamination.

Table 5. *Degree of pollution of heavy metals in soil using the Nemerow Pollution Index.*

Serial number	Reclamation year (a)	Level/cm	Nemerow pollution index (*P*)	Rating	Degree of pollution
1	2	0–15	12.61	5	Serious pollution
2	2	15–30	12.10	5	Serious pollution
3	4	0–15	12.24	5	Serious pollution
4	4	15–30	12.19	5	Serious pollution
5	6	0–15	12.16	5	Serious pollution
6	6	15–30	12.75	5	Serious pollution
7	8	0–15	12.12	5	Serious pollution
8	8	15–30	12.94	5	Serious pollution
9	10	0–15	12.08	5	Serious pollution
10	10	15–30	13.14	5	Serious pollution

3.3. Source Analysisof Heavy Metal Pollutants

According to the analyses presented in sections 3.1 and 3.2, there is a risk of heavy metal contamination in the reclamation area of the Xiaoyi Bauxite Mine, and the main contaminating element is Cd. Therefore, the contents of heavy metals in the reclaimed and surrounding soils used to reclaim the mining area were analyzed (Table 6). It can be seen from Table 6, that the concentrations of Cd were 4.91 mg/kg (reclaimed soils) and 5.04 mg/kg (surrounding soils), which exceeded the national Level 2 standard, and must therefore be classified as contaminated. It can also be seen that all of the soils and surrounding Malan loess in the mine reclamation sites were already in the state of Cd pollution, while the contents of Cr, Cu, Pb, As, and Ni were all below the Level 2 national soil standard, which is a safe and clean state.

The average content of Cd in the soil after different reclamation periods was 5.19 mg/kg, which is higher than the concentrations in either the reclaimed soilsor the surrounding Malan loess. The average Cr content was 39.17 mg/kg, which fell between the concentrations in the reclaimed soils and the surrounding Malan loess. The average Cu content was 3.15, which is higher than the concentrations in the reclaimed soils and surrounding Malan loess. The average Pb content was 65.13 mg/kg, which is also higher than the concentrations in the reclaimed soils and surrounding Malan loess. Finally, the average contents of both As (8.64 mg/kg) and Ni (14.15 mg/kg) fell between the irrespective concentrations in the reclaimed soils and the surrounding Malan loess. Based on these findings, the Cd in the reclaimed soil is mainly derived from the soil used for reclamation, which is the source of soil contamination. The existence of other external sources of pollution warrants further investigation.

Table 6. *Contents of heavy metals in reclaimed and surrounding soils.*

Soil source	Heavy metal content/(mg.kg^{-1})					
	Cd	Cr	Cu	Pb	As	Ni
Reclaimed soil	4.91	45.33	2.30	49.80	9.27	15.42
Malan loess	5.04	34.53	1.15	51.56	8.41	9.34
4.	5.19	39.17	3.15	65.13	8.64	14.45

4. Discussion

Mineral resources in Chinaare widely distributed, especially the coal, iron ore, aluminum ore, and other minerals found in Shanxi. Therefore, it is essential to understand how best to recover and control land quality after the exploitation of mineral resources for the protection and continued utilization of land resources. Land quality has a direct and important impact on ecosystems, the environment, and agro-forestry products. Therefore, not only the reclamation of mining areas, but also the quality of the land *after* reclamation should be improved and maintained.

The results of this study revealed that the soils of the Xiaoyi Bauxite Mine were significantly over the national limit and therefore heavily polluted by Cd. Moreover, the main source of pollution is derived from the surrounding Lishi loess used for reclamation. The sources of excessive heavy metals in soils, such As, Cd, Cr, Pb, Cu, and As, are found in existing organic and inorganic fertilizers in China [16–18]. Therefore, the organic and inorganic fertilizers and pesticides applied during agricultural activities after reclamation, as well as the deposition and transportation of dust into mining areas, can introduce pollutants [19–21].

Heavy metals from different sources contribute to soil pollution through enrichment. Moreover, the slag lamp debris after mining also contains high concentrations of heavy metals, and the migration of materials between the reclaimed soil and waste is another reason for the difference of heavy metal contents in different soil layers. Additionally, the absorption of heavy metals into the soil by plants also leads to a certain degree of difference and transport.

Excessive amounts of fertilizer can be used to improve soil fertility after reclamation, but the content of heavy metals in the soils will be gradually increased through enrichment, which aggravates the degree of soil pollution. Furthermore, heavy metals in soils can be transported into the soils of reclamation areas by means of atmospheric subsidence and traffic, leading to changes in the concentrations of heavy metals in the reclamation area. Therefore, the selection and control of soil quality during mine reclamation is key to the success of reclamation efforts. Further study of the movement and circulation of materials and energy between mining slag, reclaimed soils, and plants is of great importance to the redevelopment of a healthy soil ecology in mining areas.

5. Conclusions

The only heavy metal that exceeds the Level 2 national quality standard in the reclamation soils of the Xiaoyi Bauxite Mine is Cd. The mean concentration of Cdin the 0–15-cm layer is 5.11 mg/kg, while in the 15–30-cm layer, the mean value is 5.27 mg/kg. The contents of the other five elements do not exceed the national standard. The contents of Cd, Cr, and Cu gradually decrease, and Pb, As, and Ni gradually increase at 0–15 cm. At 15–30 cm, concentrations of Cd, Cr, As, and Ni gradually increase, while those of Pb gradually decrease, and Cu contents first increase and then decrease.

The assessment results based on the single factor index reveal that the main contaminating element is Cd, which reaches Level 5 heavy pollution (i.e., there is serious contamination by Cd), while the other elements do not act as contaminants. The Nemerow Pollution Index of the reclamation soils is between 12.08 and 13.14, with an average of 12.42, which is four times as high as the Level 5 pollution index of 3.0, indicating a very serious level of contamination. The content of Cd in the overburden and surrounding soil in the reclamation area exceeds the Level 2 national standard. Therefore, the soil used for reclamation is the main source of heavy metal pollution in the reclamation area.

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Biography

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