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Heavy metal pollution and potential ecological risk in reclaimed soils in Huainan mining area *

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Abstract In order to determine the environmental quality condition of reclaimed soils in Huainan mining area, soil samples were collected from three representative mines, such as Panyi Mine, Xinzhuangzi Mine and Datong Mine. The total concentration of Cd, Hg, Cu, Pb and As in the samples were analyzed. The potential ecological risk was used to evaluate the heavy metals pollution. The investigation reveals that the reclaimed soils are contaminated to a certain degree and the trace elements in coal gangue transferred to the surface soil. The order of potential ecological risk is Cd>Hg>Cu>Pb>As; the pollution degree of each sampling site is arranged in the following order: Xinzhuangzi Mine> Datong Mine>Panyi Mine, and the multiform of heavy metals of potential ecological risk index is at 357.35~484.62.

Keywords heavy metal pollution, potential ecological risk, reclaimed soils

Introduction

Coal is the main energy resource in China. 96% of coal production comes from underground mining, which always lead to surface subsidence. The area of the subsidence is up to 130 km^2 every year. Meanwhile, the reclamation of subsidence land has become the focus of mining area reclamation. Coal gangue and other solid wastes were used to fill the subsidence land not only restoring the value of mining subsidence land, but also dealing with the solid waste and reducing the area of coal gangue. However, the toxic materials in coal gangue would contaminate the reclaimed soils. The status of reclaimed soil environmental quality is related to agricultural production and human health. Hence, it is necessary to investigate the characteristics of heavy metals and evaluate the potential ecological risk.

Huainan is located in the North-central of Anhui Province. It is the largest base of resource and industry in East China and it has nearly a hundred years of coal mining history. The exploitation of coal resources promoted the development of the economy and society of Huainan City, but it resulted in a large-scale coal mining area being subsided or to be subsided, and damaged the local ecological environment. According to incomplete statistics, the area of the whole subsidence land and the affected land is 132.82 km^2, accounting for 5.1% of the area of Huainan City, mainly distributed in Datong, Xiejiaji, Bagongshan, Panji and Fengtai County. According to the requirements of the Anhui and Huainan resources department, the stable subsidence land has been filled with coal gangue and other solid wastes. Because the range and the depth of the subsidence are large, groundwater is at a high level, and the type of the subsidence is complex, so harmful heavy metals in the coal gangue immersed in the groundwater will migrate to the surroundings, and then contaminate the reclaimed soil. According to the classification of meaningful environment elements by Wood (Wood, 1974) and Förstner (Förstner and Wittmann, 1982), the elements As, Cu, Pb, Hg and Cd are selected as evaluation object, and potential ecological risk index is used to evaluate the environmental quality of the reclaimed soils. The objectives of this study were: ① to assess the level of soil contamination with

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respect to average concentrations of toxic metals in the reclaimed soils, ② to make a systematic assessment and evaluation of soil contamination under the influence of a high influx of metal pollutants from the coal gangue, thus providing a scientific basis to the ecological reconstruction of mining areas.

1 Materials and methods

1.1 Study area

Huainan City located in 116°21′21″~117°11′59″E longitude and 32°32′45″~33°00′24″N latitude and its boundaries encompass a total area of 2 596.4 km^2, and a population of some 2 million. The city consists of four administrative districts and one county. The area of subsidence is 132.82 km^2 as shown in Fig.1. According to the date of mining and the status of reclaimed soils, three representative reclaimed soils are selected as research objects. Datong Mine is a scrap mine, the thickness of the covering soil is 60 cm; Xinzhuangzi Mine is an old mine, the thickness is 60 cm; Panyi Mine is a new mine, the thickness is 100 cm.

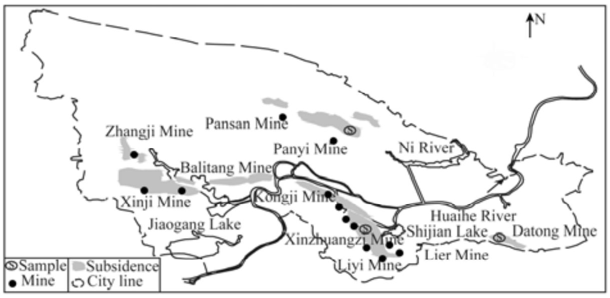

Fig.1 Distribution of coal-mining subsidence lands and sampling points in Huainan Coal Mine district

1.2 Sampling and analysis

Soil samples were collected from these reclaimed soils in May 2009. All soil samples were taken between the soil surface and the coal gangue, and had 20 cm thickness and about 1 kg in weight. Based on the principles of Global Position System (GPS), the exact longitude and latitude of the sampling sites was determined. The location of the sampling sites is shown in Fig.1. All the soil samples were collected in self-locking polythene bags, and air-dried to remove stones and coarse plant roots or residues. The dry soil samples were finely powdered by swing-grinding mill, then thoroughly mixed and ground to pass a 0.149 mm nylon sieve. Sample pellets were stored in brown glass bottles and prepared for chemical analyses. Mixed acid HF+$HClO_4$+HNO_3 was used to digest soil heavy metal elements Cd, Pb and Cu, and tested by atomic absorption spectrophotometry. Aqua regia was used to digest Hg and As, and tested by atomic fluorescence spectrometry.

1.3 Quality control

To monitor the quality of the chemical analysis data and examine the accuracy of the data, soil reference materials (GBW-07403) issued by the center of Chinese Standard Material were analyzed along with the soil samples during the course of analysis as unknown samples. The analytical concentrations of the metals in reference samples and the recovery rate are presented in Table 1. The results show that the measured values are within the range of certified values of the reference materials, and all elements obtained better recoveries; the quality of analysis was effectively controlled.

Table 1 Comparison between measured values of the soil samples and certified values

Elements	As	Hg	Cd	Cu	Pb	Cr	Zn
Measured values	3.9	536	0.055	11.2	23.7	29.5	28.5
Certified values	4.4	590	0.059	11.4	26.0	32.0	31.0
Recovery values (%)	89	90	93	98	91	92	92

1.4 Potential ecological risk index

The method of potential ecological risk index presented by Hakanson was used to assess the ecological risk of heavy metals (Hakanson, 1980). It is based on the nature and environmental characteristics of heavy metals, put forward by the view of sedimentation, and evaluate heavy metals in soil or sediment. This method not only takes into account the heavy metal contents, but also associates with ecological effects, environmental effects and toxicology, and adopts a comparable and equivalent attribute index classification to evaluate (Guo, 2005). Its calculation formula is as follows:

$$C_{\mathrm{f}}^{i}=C_{\mathrm{s}}^{i}/C_{\mathrm{n}}^{i}, E_{\mathrm{r}}^{i}=T_{\mathrm{r}}^{i}/C_{\mathrm{f}}^{i}, R_{\mathrm{I}}=\sum_{i=1}^{n}E_{\mathrm{r}}^{i}$$

where, C_{f}^{i} is the single element index, C_{s}^{i} is the measured value of heavy metals contents, C_{n}^{i} is the pre-industrial concentration of individual metal (the background values of soil heavy metals in Huainan are used as the reference values in this study), T_{r}^{i} is the toxic response factor (Hg: 40, Cd: 30, As: 10, Pb: 5, Cu: 5). E_{r}^{i} is the potential ecological risk factor of a single heavy metal; R_{I} is the potential ecological risk index of multiform heavy metals in soil.

The potential ecological risk coefficients were classified into 5 grades and the potential ecological risk index was classified into 4 grades. The relationship between the potential ecological risk coefficients, the potential ecological risk index of heavy metals and the degree of pollution is shown in Table 2.

Table 2 Relationship between potential ecological risk coefficients, risk indices of heavy metals and pollution level

E_r^i	Pollution levels	R_I	Pollution levels
E_r^i <40	Low	R_I<150	Low
40≤ E_r^i <80	Moderate	150≤R_I<300	Moderate
80≤ E_r^i <160	Considerable	300≤R_I<600	High
160≤ E_r^i <320	High	R_I ≥600	Very high
E_r^i ≥320	Very high		

1.5 Background data

In order to better reflect the status of the soil heavy metal pollution, this paper chose the background values of Huainan soil as the reference. The concentrations of As, Cd, Hg, Pb, Cu were 16.86, 0.06, 0.041, 30.47 and 24.20 mg/kg respectively.

2 Results and discussion

2.1 Heavy metal concentrations

The mean concentrations of some heavy metals in the reclaimed soil are given in Table 3. Compared with the background values of Huainan soil, except As, the mean concentration of Cd, Hg, Pb and Cu in those reclaimed soil were higher than the background values. The Cd concentrations ranged from 0.31 to 0.58 mg/kg, and the Hg concentrations ranged from 0.113 to 0.237 mg/kg, which greatly exceeded the natural background values. From the results we can see that reclaimed soils were subject to a certain degree of heavy metals pollution.

Table 3 Concentration of some heavy metals in the reclaimed soil

Reclaimed land	Depth(cm)	Concentration of heavy mentals(mg/kg)				
		As	Pb	Cu	Cd	Hg
Panyi Mine (n=30)	0~20	6.44±2.29	52.93±17.72	40.28±7.66	0.43±0.16	0.126±0.093
	20~40	6.71±2.99	53.78±15.60	41.83±5.97	0.46±0.22	0.113±0.064
	40~60	6.33±2.53	52.97±15.45	42.20±5.57	0.50±0.25	0.121±0.058
	60~80	6.40±2.23	52.58±15.37	42.16±5.87	0.45±0.12	0.114±0.061
	80~100	6.45±2.91	58.24±18.67	44.21±8.16	0.53±0.31	0.128±0.071
Xinzhuangzi Mine (n=17)	0~20	5.86±1.85	42.58±13.81	43.52±9.87	0.51±0.14	0.177±0.091
	20~40	6.29±2.97	45.94±10.97	42.85±8.10	0.57±0.19	0.171±0.084
	40~60	6.92±2.52	43.60±12.31	42.22±6.44	0.58±0.27	0.179±0.094
Datong Mine (n=7)	0~20	3.82±1.74	44.75±7.83	32.06±5.13	0.31±0.12	0.205±0.070
	20~40	3.85±1.89	45.17±7.20	31.70±5.17	0.31±0.15	0.236±0.087
	40~60	3.83±1.38	47.97±7.68	32.31±5.06	0.32±0.10	0.237±0.109

Note: n is the number of sample points; mean±standard deviation

2.2 Vertical distribution and migration

In order to study the vertical changes of heavy metals in soil, we analyze the reasons for the distribution of heavy metals. In this paper, we make a comparison of the mean concentrations of heavy metals in each sampling thickness of the reclaimed soil. The results were shown in Fig.2.

Fig.2 shows that the concentrations of heavy metals were different in the different sample layer of the reclaimed soils. The soil heavy metals content that contact with the coal gangue were higher than the surface layer soil. This indicated that the coal gangue heavy metals had migrated to the lower soil layer. Moreover, the mean concentration of one metal is different in three reclaimed soils.

Recently, highly toxic element pollutants (such as As, Cd, Hg, Pb) have received much concern. In the study of soil heavy metal pollution in the reclaimed land in Xuzhou, the soil was subject to a certain degree of pollution, Cd and Cu pollution was the most serious, followed by Hg and Pb (Wang and Dong, 2009). In the evaluation of the Antaibao opencast coal mine reclaimed land heavy metal pollution, the order was Cd>As>Pb>Cr>Hg, in which Cd exceeded the first standard of GB15618−1995 (Ge et al., 2008). Overall, the process of using coal gangue to fill in the subsidence land will cause a certain degree of pollution to soil. The environmental quality conditions of the reclaimed soils should be monitored and serious pollution of soil should be repaired.

2.3 Evaluation of potential ecological risk index

Table 4 presents the potential ecological risk coefficients and risk indices of heavy metals in reclaimed soils. From Table 4, we can see that in the reclaimed areas the E_r^i of As, Pb, and Cu values were less than 40, which is classified as low ecological hazard. However, the E_r^i of Cd in the soil layer 0~20 cm and 20~40 cm in Datong Mine were between 80 and 160,

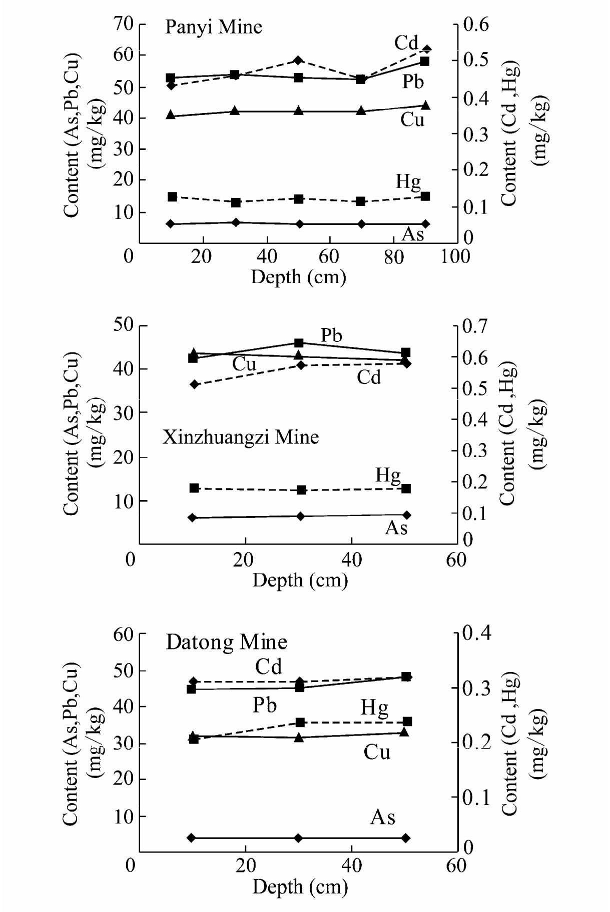

Fig.2 The vertical variation of heavy metal elements content in different reclaimed areas

Table 4 Potential ecological risk coefficients and risk indices of heavy metals in the reclaimed area

Reclaimed land	Depth (cm)	E_r^i					R_I
		As	Pb	Cu	Cd	Hg	
Panyi Mine	0~20	3.82	8.69	8.32	215	122.93	358.75
	20~40	3.98	8.83	8.64	230	110.24	361.69
	40~60	3.75	8.69	8.72	250	118.05	389.21
	60~80	3.80	8.63	8.71	225	111.22	357.35
	80~100	3.83	9.56	9.13	265	124.88	412.39
Xinzhuangzi Mine	0~20	3.48	6.99	8.99	255	172.68	447.14
	20~40	3.73	7.54	8.85	285	166.83	471.95
	40~60	4.10	7.15	8.72	290	174.63	484.62
Datong Mine	0~20	2.27	7.34	6.62	155	200.00	371.23
	20~40	2.28	7.41	6.55	155	230.24	401.49
	40~60	2.27	7.87	6.68	160	231.22	408.04

which is classified as considerable ecological hazard. The other sample points were between 160 and 320, or high ecological hazard. The E_r^i of Hg between 160 and 320 in Xinzhuangzi Mine and Datong Mine, belonged to high ecological hazard. The E_r^i of Hg between 80 and 160 in Panyi Mine, belonged to considerable ecological hazard. From the analysis of single heavy metal pollution, Cd showed significant ecological hazard effects. The order of ecological hazard of various metals is Cd>Hg>Cu>Pb>As. A multiform of heavy metals of potential ecological risk index was at 357.35~484.62, the reclaimed soil has reached a high degree of ecological hazard. The order of potential ecological risk index is Xinzhuangzi Mine>Datong Mine>Panyi Mine. It is necessary to analyze the environmental problems in the reclaimed soils caused by the coal gangue to avoid secondary pollutions.

3 Conclusion and suggestion

(1) The reclaimed soils have been subjected to a certain degree of heavy metals pollution. It is contaminated due to the use of coal gangue to fill the subsided land.

(2) The high content of toxic heavy metals in coal gangue induces an increase in their content in ground water as a result of leaching, then heavy metals migrate to the lower soil layer. The concentrations of Cd, Hg, Pb and Cu exceeded the background values of Huainan soil.

(3) The order of ecological hazard of various metals is Cd>Hg>Cu>Pb>As. The order of potential ecological risk index is: Xinzhuangzi Mine>Datong Mine>Panyi Mine.

(4) The reclaimed soil has reached the high degree of ecological hazard. Therefore, in the process of remediation, it is necessary to monitor the heavy metals that leach from coal gangue, and take measures to reduce the chance of migration of harmful pollutants.

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