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SPATIAL-TEMPORAL VARIATION OF HEAVY METAL ELEMENTS CONTENT IN COVERING SOIL OF RECLAMATION AREA IN FUSHUN COAL MINE

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ABSTRACT: Grid method is employed for sampling covering soil at the test field, which is reclamation area filled by coal mining wastes for cropland in the Fushun coal mine, Liaoning Province, the Northeast China. The soil samples are taken at different locations, including three kinds of covering soil, three different depths of soil layers and four different covering ages of covering soil. The spatial-temporal variation of heavy metal element content in reclamation soil is studied. The results indicate that the content of heavy metal elements is decreasing year after year; the determinant reason why the content of heavy metal elements at 60cm depth layer is higher than that at 30cm depth layer and surface is fertilizer and manure application; the metal elements mainly come from external environment; there is no metal pollution coming from mother material (coal mining wastes) in plough layer of covering soil.

KEY WORDS: covering soil; heavy metal elements; spatial-temporal variation; reclamation; Fushun coal mine

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1 INTRODUCTION

Fushun coal mine is located in 123°04′48″ – 124°27′26″ E, 41°27′10″ – 42°04′01″ N. The climate belongs to typical East Asia continental monsoon climate in mid temperate zone. The average temperature is about 4 – 7℃ and the average annual precipitation is above 800mm in the past decade or so. The zonal strata are composed of Proterozoic, Mesozoic Cretaceous period purple shale and sand shale layer, Cenozoic Tertiary coal layer and Cenozoic Quaternary alluvial layer from bottom to top respectively. The zonal soil is brown earth, and the intrazonal soil is cultivated meadow soil. The characters of soil are looseness, good perviousness, high capacity of retaining the fertility and water. The main vegetation type is broadleaf-conifer mixture forest that mainly consists of the broadleaved tree.

Fushun coal mine with nearly 100 years of exploitation period is very famous in China. The whole diggings area is 45km^2 (15km long from east to west and 3km wide from south to north). Because of continuous expansion of underground mining range, a series of environmental problems have been appeared, such as the earth surface sinking, water accumulating, etc. The natural ecological environment has been destroyed, which leads to the contradiction between the population increasing and soil land decreasing being more and more acute (LIU *et al.*, 2000; YANG *et al.*, 1999).

In recent years, some Chinese scholars have done a series of studies about the reclamation of the abandoned mine land, especially in the field of vegetation planting technique (YANG *et al.*, 1999; ZHAO 1993; HU, 1996; LI and JI, 1995; ZHU, 1993; JIAO, 1999; ZHANG, 1992). The oversea and Chinese studies show that the major task of the reclamation is to make the covering soil suitable for vegetation growing. The primary factors affecting plant growing in reclamation area are nutrition, heavy metal pollution and acid pollution of the covering soil (DUAN *et. al.*, 1999; SHU *et al.*, 1999; HOSSNER, 1988; TU *et al.*, 2000; ZOOL,

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2001; CHEN, 2001; PAIN *et al*., 2000; MCBRIDE *et al*., 2000; SUN *e t al*., 2000). In the paper, we discussed the spatial-temporal variation laws of the heavy metal elements content in covering soil of coal mine reclamation area, taking Fushun coal mine area as a case.

2 MATERIAL AND METHODS

The Fushun Coal Mine Bureau was established in 1932. The coal storage is about 804.943 million tons, and the average thickness of coal layer is about 50m. After the centralized mining for more than 60 years, this region has appeared three sinks from east to west. Up to 1995, the sinking area has been beyond 22.5km^2. Among these sinks, the depth of Laohutai coal mine was about 25 – 28m, the accumulated water depth was about 5 – 6m and its volume was about 1.2 million m^3. The gangues refilled and reclamation experimental zone of Fushun coal mine was established in 1991. The refilled technique is called covering soil refilled method, a depth about 0.6 – 0.8m was covered after the coal gangues was refilled. Finally the altitude was 81.3m, surpassing the pre-mining altitude(80.5m) 0.8m. From June 1991 to October 1993, the reclamation project had been put in practice by Northeast Coal Company and Fushun Mine Bureau, and the pioneer plants had been selected and planted. During the period of June 1996 to September 1999, the Northeast critical coal mine ecological reconstruction experimental site was established here by Changchun Institute of Geography, the Chinese Academy of Sciences.

Mesh spot pattern is employed to choose 9 sampling sections in an area of 10ha, four samples sites in each section, and is repeated three times. The soil samples were taken with three different covering soil types(river bottom soil, loess-like soil, mixing garbage soil) and different depths(surface, 30cm, 60cm) in 1993 (covering for 2 years), 1995(covering for 4 years), 1997 (covering for 6 years) and 1999(covering for 6 years), respectively. The heavy metal elements including Cu, Zn, Cr, Mn and Cd were analyzed for each soil sample. The determination method is inductively coupled plasma analysis of emission spectroscopy (ICP-AES) method after extracted by different methods.

In order to intuitively show the variation trend of element content in a figure, the mean value standardization method of datum is applied. That method means that the datum used in the figure is the ratio of measured value to mean value.

3 RESULTS AND DISCUSSION

3.1 Temporal Variation of Heavy Metal Elements Content in Covering Soil

Before covering the soil in experimentation area, the background value of typical elements in gangues and covering soil were monitored(Table 1). After covering the soil(July 1991), several pioneer plants species such as radish(*Raphanus*), Chinese cabbage (*Brassica pekinensis*), potherb onion(*Allium oleraceum*), potherb mustard(*Brassica juncea* var. *crispifolia*), etc. were planted for experiment.

Table 1 The background content of heavy metal elements in different covering soils and coal mining wastes (mg/kg)

Soils and gangues		Cu	Zn	Cd	Mn	Cr
Soils	River bottom soil	20.81	62.79	0.023	453.61	72.41
	Loess-like soil	23.05	62.48	0.021	435.53	70.92
	Mixing garbage soil	59.12	71.52	0.011	413.22	74.13
Gangues	Chlorite shale	32.61	93.58	2.000	357.36	82.00
	Refused shale	35.01	86.97	2.212	306.65	100.03
	Kerogen	31.69	101.10	1.531	368.40	87.31

During 1991 to 1999, different covering soils were sampled in September of 1993, 1995, 1997 and 1999 and analyzed, respectively. The variation of heavy metal content in different kinds of covering soil with the covering age of soil is shown in Fig. 1 to Fig. 3. The results show that the decreasing trend of each metal element(except Cd) content with the increasing of covering age of soil appears in different covering soils. The reason for the phenomenon is that the heavy metal elements content in covering soil is high, and with cultivation period increasing, heavy metal leached down continuously from topsoil layer. The variation of Cd is fluctuating violently, but they show a similar regulation in three different covering soils. The main reason is yearly change of fertilizer sort and quantity. For example, Cd content in phosphate fertilizer is obviously higher than that in other fertilizers (BIAN and ZHANG, 1999).

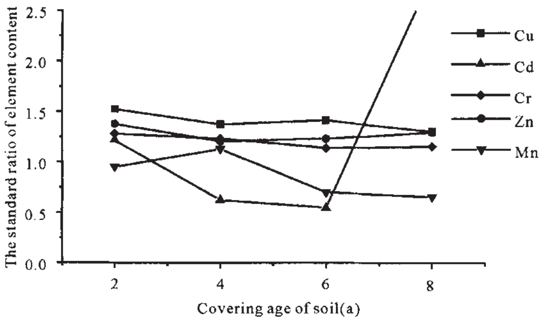

Fig. 1 The content variation of heavy metal elements with covering age of river bottom soil

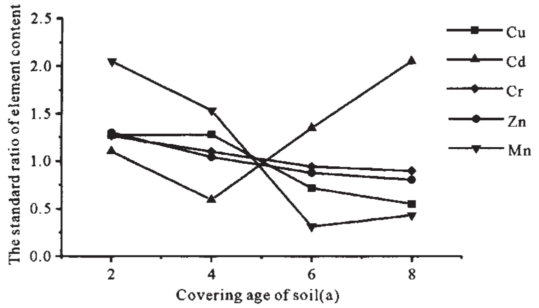

Fig. 2 The content variation of heavy metal elements with covering age of loess-like soil

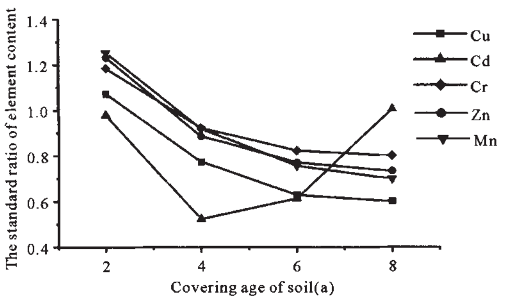

Fig. 3 The content variation of heavy metal elements with covering age of mixing garbage soil

3. 2 Spatial Variation of Heavy Metal Elements Content in Covering Soil

3. 2. 1 Horizontal variation of heavy metal elements content

According to the analytical results of 0 – 30cm depth topsoil samples which were sampled in 1999, the differences of heavy metal elements content of three kinds of covering soil were showed in Table 2. Obviously, the heavy metal elements contents are sharp different in three kinds of covering soil after cultivation for 8 years. The contents of all heavy metal elements in mixing garbage soil are lower than that in river bottom soil and loess-like soil except for Mn.

Table 2 The average content of heavy metal elements in 0 – 30cm depth of different covering soils (mg/kg)

Soil	Cu	Zn	Cd	Mn	Cr
River bottom soil	26. 57	63. 47	0. 613	156. 69	51. 93
Loess-like soil	11. 25	39. 40	0. 442	103. 43	40. 32
Mixing garbage soil	10. 86	36. 02	0. 217	167. 57	36. 07

The primary reason of such result is the difference of physical properties and chemical properties of covering soils. The water permeability of loess-like soil with heavy texture is weak. The soil particle size of river bottom soil is fine, the diameter of about 15. 53% of soil particular is less than 0. 001mm, about 45. 25% of soil particular is between 0. 01mm to 0. 001mm. So the water permeability of river bottom soil is weak than that of loess – like soil. The majority of soil particular diameter of mixing garbage soil is between 0. 25mm to 0. 01mm, about 66. 68%. So the loss of element in mixing garbage soil is fast relatively.

3. 2. 2 Vertical variation of heavy metal elements content

Because of the sustaining fertilization, plough and other effected factors such as rainfall, evaporation and leaching etc., the heavy metal content in topsoil is different obviously to below soil. But with the similar cultivation, field management and other outer conditions, the vertical variation trend of heavy metal elements contents in every soil type shows a similar law(Fig. 4 to Fig. 6).

The results show that the content of all heavy metal elements but Mn is gradually decreasing from topsoil to bottom layer soil. The potential sources of heavy metal in the covering soil include: 1) heavy metal elements in gangues parent material are released during the cultivation by the soil microorganism and the plants; 2) heavy metal elements in the covering soil; 3) input from outside, such as the exhaust gas from automobile, waste gas and waste solid material from power station, irrigation; 4) input during fertilization. For example, phosphate fertilizer contains high concentration of Cd, Cr and V; pig dung contains high concentration of Cu; sewage and sludge contain high concentration of Pb, Zn, Cr, Ni and Cu. About 66% – 84% heavy metal exist in bio-flocculate, about 14% – 27% exist in particular constituent and 0. 15% – 13. 4% exist in water-soluble constituent (BIAN, 1993; ZHOU *et al*., 2000). Ac-

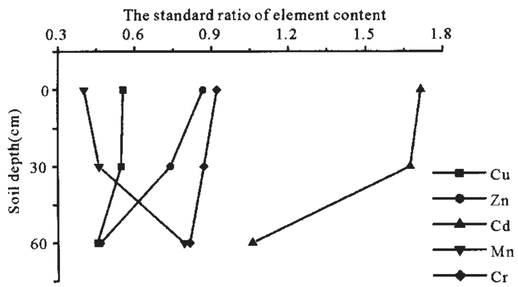

Fig. 4 The vertical variation of heavy metal elements content in covering river bottom soil

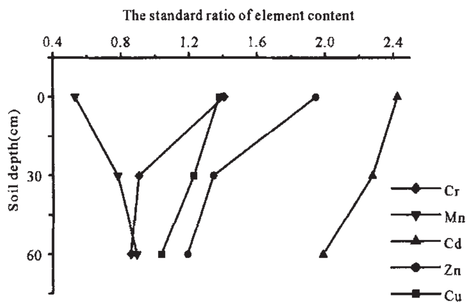

Fig. 5 The vertical variation of heavy metal elements content in covering loess-like soil

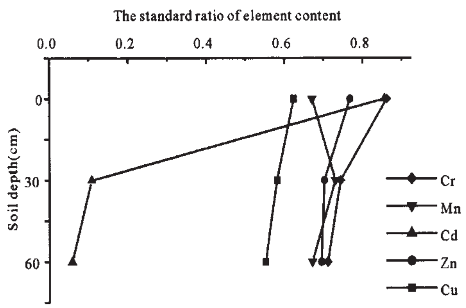

Fig. 6 The vertical variation of heavy metal elements content in covering mixing garbage soil

cording to the contrast of heavy metal contents in different depth soils to the background concentration in each covering soil, we can see that the content of all heavy metals except individual elements in 60cm depth soil is lower than that in 30cm depth soil. The difference of heavy metal content between topsoil and bottom layer soil is quite obvious. The content of Cu, Zn in topsoil of river bottom soil are 28.09mg/kg and 95.40mg/kg, respectively, 7.29mg/kg and 32.60 mg/kg higher than their background concentration, respectively. The results indicate that the heavy metal elements didn't come from the gangues parent material, but from outside input. It is related to fertilization and topsoil pollution by atmosphere and dust. The eluviation is main reason of heavy metal content difference between topsoil and below layer soil.

4 CONCLUSIONS

(1) The content of heavy metal elements in covering soil is decreasing with the covering soil age increasing.

(2) Compared with the heavy metal elements content in different covering soils, the mixing garbage soil is fairly to be covering soil in coal mine reclamation.

(3) The heavy metal elements content in gangues doesn't impact soil quality for cultivation, the outside input is the main source of heavy metal.

(4) The reason of similar vertical variation law of heavy metal content in different covering soils is similar field management and outer environment.

(5) Eluviation is the major influence factor on the vertical transference of heavy metal elements.

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