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Full Length Research Paper

The evaluation of reclamation effects for the methods of covering soils in a coalmine reclamation area

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An experimental area has been established for studying the ecological reconstruction and reclamation in China's Fushun coalmine where coal resource in this mine is almost exhausted now and parts of the surrounding land have undergone subsidence. The experimental area is a large subsided area that has been filled in with gangues (matrix) wastes and then covered with ~80 cm of topsoil. The resulting land was used to grow crops. A grid method was employed to guide sampling of different types of topsoil, covering different depths and reconstruction ages (2, 4, 6 and 8 years). The spatial and temporal variations in nutrient elements and heavy metals (Cu, Zn, Mn, Cr and Cd) in the soil were studied. Nutrients (N, P and K) in the topsoil recovered quickly after reclamation, whereas the heavy metal content decreased with time (except Cd). Nutrient levels were the highest in the top 30 cm of the topsoil and decreased with depth, as a result of the usage of fertilizer and manure. In contrast, the heavy metal contents (except Mn) of the topsoil (0 to 30 cm) were higher than those at depths of 30 to 80 cm, suggesting the high heavy metals were not from the gangues.

Key words: Heavy metals, soil nutrients, reclamation, coalmine, pollution, subsidence.

INTRODUCTION

The main factors that constrain plant growth in reclamation areas are the soil's nutrient status, heavy metal contents, and acid pollution (Hobbs et al., 2008; Hu, 1996; Jiao, 1999; Querol et al., 2006; Yang, 1999; Zhao, 1993; Zhu, 1993). Factors that affect the characteristics of the topsoil, such as fertilization, nutrient status, and pollution by heavy metals and acids, were controlled by the composition of the gangues (coal matrix) and garbage used as filling material, as well as by the local geochemistry, topography, and climatic conditions (Bian et al., 2009; Hossner, 1998; Loczy et al., 2007; McBride and Martinez, 2000; Shu and Huang, 1999; Sun, 2000).

Restoration of mine wasteland in China began in the late 1970s, but the restoration process was sluggish. The overall restoration rate of mine wasteland was some 10 - 12% with a higher rate for coal mine spoils but a lower rate for metal-mined derelict land (Li, 2006). Since the 1980s, the disposal technique for coal mining wastes has been changing and in many instances the wastes are now transported directly to subsided land as a fill to enable the reuse of the land. Today, both coal mining waste dumps from the past and filled subsided lands are in existence. But, the comparative impacts of these different disposal techniques on the environment and farmland productivity have not been studied in detail (Bian et al., 2009). The Fushun coalmine in China's Liaoning province is one of the oldest and largest coal mines in northeastern China, and has been exploited industrially since 1932, with some evidence of exploitation as early as the 12th century (Li, 2006; Liu et al., 2000; Yu et al., 2002). The annual output of coal ranged between 100×10^4 and 200×10^4 t from 1949 to 1990. Currently, the resource is nearly exhausted, and there is very little coal production. The mine covers an area of 45 km^2 (15 km from east to west and 3 km from

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north to south), bounded by two branches of the Hunhe River. As the range of the underground mining expanded, a series of environmental problems appeared, such as subsidence, pooling of mine water in low-lying land, and contamination of agricultural land by heavy metals, acidic water, and other mine wastes. In essence, the ecology of the land above and around the coalmine has been badly damaged (Liu et al., 2000; Yang, 1999; Yu et al., 2002). To investigate the possibility of rehabilitating the area, we chose a subsidence area of ~10 ha as an experimental site. This area had first been refilled with gangues and other solid wastes in 1990 - 1991, followed by a reclamation project from 1991 to 1993, by the Northeast Coal Company and the Fushun Mine Bureau, and was then maintained by the Changchun Institute of Geography, the Chinese Academy of Sciences from 1996 and 1999. Our purposes are: (a) to study the spatial and temporal variations of nutrient elements (N, P, K) and heavy metals (Cu, Zn, Cd, Mn, Cr) in the topsoil of this reclamation area; (b) to estimate the reclamation effects of different covering soils of urban garbage, stream sediment and loess soils, and (c) to evaluate the heavy metal pollution status of covering soils in filled subsided lands of reclamation area.

MATERIALS AND METHODS

Site description

The city of Fushun is located in the middle temperate zone, with typical eastern Asian continental monsoon climate. The annual air temperature averages between 4 and 7°C, and the average annual rainfall is about 800 mm based on data from 1970 to 2000 provided by the Fushun Meteorological Station. Part of the Fushun coalmine lies in the southern part of the city, between 41°27'10" and 42°04'01" N and between 123°04'48" and 124°27'26" E. The landform is higher in the eastern part of Fushun, and lower in the west. The Hunhe River runs from east to west through the southern part of the city. The regional soil is a brown earth and meadow soil with high capacity of retaining fertilizers and water. The main native vegetation is a mixed broadleaved-coniferous forest, with the broadleaved as the predominated species.

The mine's coal is present mainly in the upper Cenozoic era (Tertiary) strata. The coal seam averages 50 m in thickness but is thinner in the east and thicker in the west. Its upper stratum is a 5- to 30-m-thick layer of fluvial and alluvial materials of Quaternary origin, with a mixed layer of green shale, oil shale, and basalt intrusions underlying the coal stratum at a depth of 90 to 280 m. At greater depths below the coal stratum, basalt and sandstone from the Mesozoic and Paleozoic eras can be found.

Reclamation experiment

There are a number of areas of subsidence distributed in the Fushun coalfield. The subsidence depth of the chosen area ranged from 25 to 28 m, which corresponds to a volume of about 1.2×10^6 m^3. This pit was partially refilled with gangues of green shale, refused shale and kerogen, which were solid wastes from coal mining. These gangues were then covered with covering soils of stream sediment, loess, or mixed urban garbage of about 80 cm thick at different locations to provide a range of treatment options. The elevation of the remade ground averaged 81.3 m above sea level, which is about 0.8 m higher than the surrounding area. The filled ground was then planted with several pioneer species such as radish (*Raphanus* spp.), Chinese cabbage (*Brassica pekinensis),* potherb onion (*Allium oleraceum*), potherb mustard (*Brassica juncea* var. crispifolia) etc., from 1991 to 1999. An annual fertilization with 400 kg/ha of diamonium phosphate and 60 m^3/ha of composted manure was provided. The fertilizer was applied within 5 - 30 cm soil depth in the early spring. No irrigation was supplied during the growing season.

Sample collection

Topsoil samples were collected in the experimental area applying a grid method. A total number of 27 sections in the experimental area were sampled with 9 sampling sections in each type of topsoil and 4 sample sites in each section. Samples from three different depths (0 - 30, 30 - 60, and 60 - 80 cm) at each sample site were collected, with a total number of 324 samples collected at each sampling time. Samples for 2, 4, 6 and 8 years after landfill were collected, respectively.

Analytical method

The soil samples were dried to constant weight for 24 h at 120˚C, then grounded and sieved using a 100-mesh sieve. About 0.5 g of each sample was digested using a HNO_3-HF-$HClO_4$ mixture with ratio 5:3:2, then dissolved in 2 ml HCl (2 mol/l) before being placed into a 50 ml quantitative flask for the determination of element concentrations. One national standard sample provided by Chinese Standard Materials Center, and two blanks were analyzed in each batch using the same method to provide quality control. All measurements were verified by subtracting the blanks and then comparing with the known values for the standard.

The heavy metals (that is Cu, Zn, Mn, Cr and Cd) contents were analyzed by using Shimadzu ICPS-7500 sequential type plasma emission spectroscopy. After soil samples were extracted with different methods, soil total nitrogen (TN) and available nitrogen (AV) was determined using the Kjeldahl method; total phosphorus (TP), available phosphorus (AP), total potassium (TK) and available potassium (AK) concentrations were determined by using GBC-906AA atomic absorption spectrophotometer.

RESULTS AND DISCUSSION

Initial levels of nutrient elements and heavy metals in the coal gangues and topsoil

The initial levels of nutrient elements and heavy metals in the gangues and the topsoil are shown in Table 1. These data are based on the soil analyses conducted by Fushun Mine Bureau when the gangues were used to fill the pit. All the three types of original topsoil (stream sediment, loess, and mixed urban garbage) had very low nutrient element contents, because they had not been developed to be agricultural soils. Whereas, all three kinds of topsoil have high levels of heavy metals, because these had been collected near the coalmine, and were thus contaminated with heavy metals. The gangues included green shale, kerogen, and refused shale, which contained high levels of heavy metals, such as Cu, Zn, Cd,

Table 1. Initial concentrations of nutrient elements and heavy metals in the topsoil and the coal mining wastes (mg/kg).

Nutrient elements/ Heavy metals	Covering soils			Gangues		
	Stream sediment	Loess	Mixed urban garbage	Green shale	Refused shale	Kerogen
TN*	0.059 ± 0.045	0.052 ± 0.18	0.079 ± 0.023	0.418 ±0.25	0.197 ± 0.12	0.403 ± 0.15
TP*	0.046 ± 0.023	0.058 ± 0.014	0.062 ± 00.05	0.116 ± 0.056	0.124 ± 0.10	0.062 ± 0.04
TK*	1.594 ± 0.887	1.618 ± 0.57	1.841 ± 0.16	3.056 ± 0.99	2.991 ± 0.67	2.956 ± 0.98
AN	56.11 ± 11.10	61.95 ± 9.63	44.35 ± 7.36	32.11 ± 3.14	10.22 ± 2.18	26.27 ± 7.10
AP	12.96 ± 4.87	14.91 ± 4.51	3.53± 1.04	1.31 ± 0.20	1.40 ± 0.66	1.11 ± 0.19
AK	62.91 ± 12.74	50.52 ± 5.44	46.33 ± 7.18	14.44 ± 2.75	10.48 ± 3.34	12.30 ± 5.33
Cu	20.81 ± 7.35	23.05 ± 5.87	59.12 ± 3.45	32.61 ± 2.45	35.01 ± 10.35	31.69 ± 9.50
Zn	62.79 ± 14.76	62.48 ±13.55	71.52 ± 13.27	93.58 ± 22.98	86.97 ± 14.66	101.10 ± 23.17
Cd	0.023 ± 0.016	0.021 ± 0.010	0.011 ± 0.01	2.000 ± 0.17	2.212±0.96	1.531 ± 0.84
Mn	357.36 ±19.68	306.65 ± 29.87	368.40 ± 18.78	453.61 ± 17.90	435.53±23.44	413.22 ± 27.22
Cr	72.41 ± 23.71	70.92 ± 10.77	74.13 ± 12.19	82.00 ± 11.36	100.03±13.68	87.31 ± 10.75

* Unit of TN, TP and TK is %.

Mn and Cr.

Temporal variations in nutrient elements and heavy metals in the topsoil

The variation in the levels of nutrient elements and heavy metals in different kinds of 0 – 30 cm topsoil is shown in Figures 1 and 2, respectively, as a function of the age of the soil.

Figure 1 shows that the levels of nutrient elements in the topsoil increased with time, and that the available N, P, and K increased more rapid than the total (T) N, P and K. This phenomenon indicates that soil fertility has increased as a result of several years of cultivation, with fertilizer and composted manure applied every year. The levels of nutrient elements in all three kinds of topsoil increased rapidly in the first 2 years and much slower later on. The levels of available P and K and TN in soils after 2 years reclamation reached or exceeded the middle standard for soil fertility (National Soil Management Bureau, 1995).

Figure 2 shows that the heavy metal contents of all three kinds of topsoil decreased with time, with the exception of Cd and Zn in stream sediment which was at certain value after 4 years reclamation. It is clear that the levels of heavy metals in the topsoil gradually decreased as a result of cultivation, probably because the heavy metals were accumulated in the cover crops through biological processes and were removed from the topsoil (Chen, 2001; Parkpain et al., 2000; Uhlig et al., 2001). Alternatively, they may have been leached out (Li and Ji, 1995). There were relatively big fluctuations in the Cd content and similar fluctuations in TP in all three types of topsoil. The yearly variation in TP content was similar to that of Cd in the three types of topsoil. This can be attributed to variations in the quantity and quality of the fertilizers used, because the Cd content of phosphate fertilizer is considerably higher than other fertilizers (Bian and Zhang, 1999).

Vertical variation of nutrient elements and heavy metals in the topsoils

The topsoil became rich in nutrients, after 8 years of continuous tillage and fertilizer application, as result of the presence of plant root systems, and strong activity by microorganisms (Duan, 1999; Tu et al., 2000). Consistently, the nutrient levels decreased with depth (Figure 3). One-way ANOVA results show that the differences of nutrients but TP in different soil depth were significant (< 0.05).

The levels of heavy metals were different in profiles (Figure 4) because of the differences in tillage and other external factors including rainfall, evaporation, and leaching, and initial contents in the source materials (both gangues and topsoil). However, with similar cultivation, field management, and other external conditions, the vertical trends in heavy metal contents were similar for each soil type (Figure 4). With the exception of Mn, the heavy metal content gradually decreases with depth. The potential sources of heavy metal in the topsoil include: (1) heavy metals present in the gangues parent material, which were released by soil microorganisms and by plants during cultivation; (2) heavy metals initially present in the topsoil; (3) external heavy metal inputs from automobile exhaust gases, waste gases, and solid wastes from power stations, and (4) inputs from fertilization. Examples include the high Cu levels in pig manure, the high concentrations of Cd, Cr and V in phosphate fertilizers, and the high concentrations of Pb,

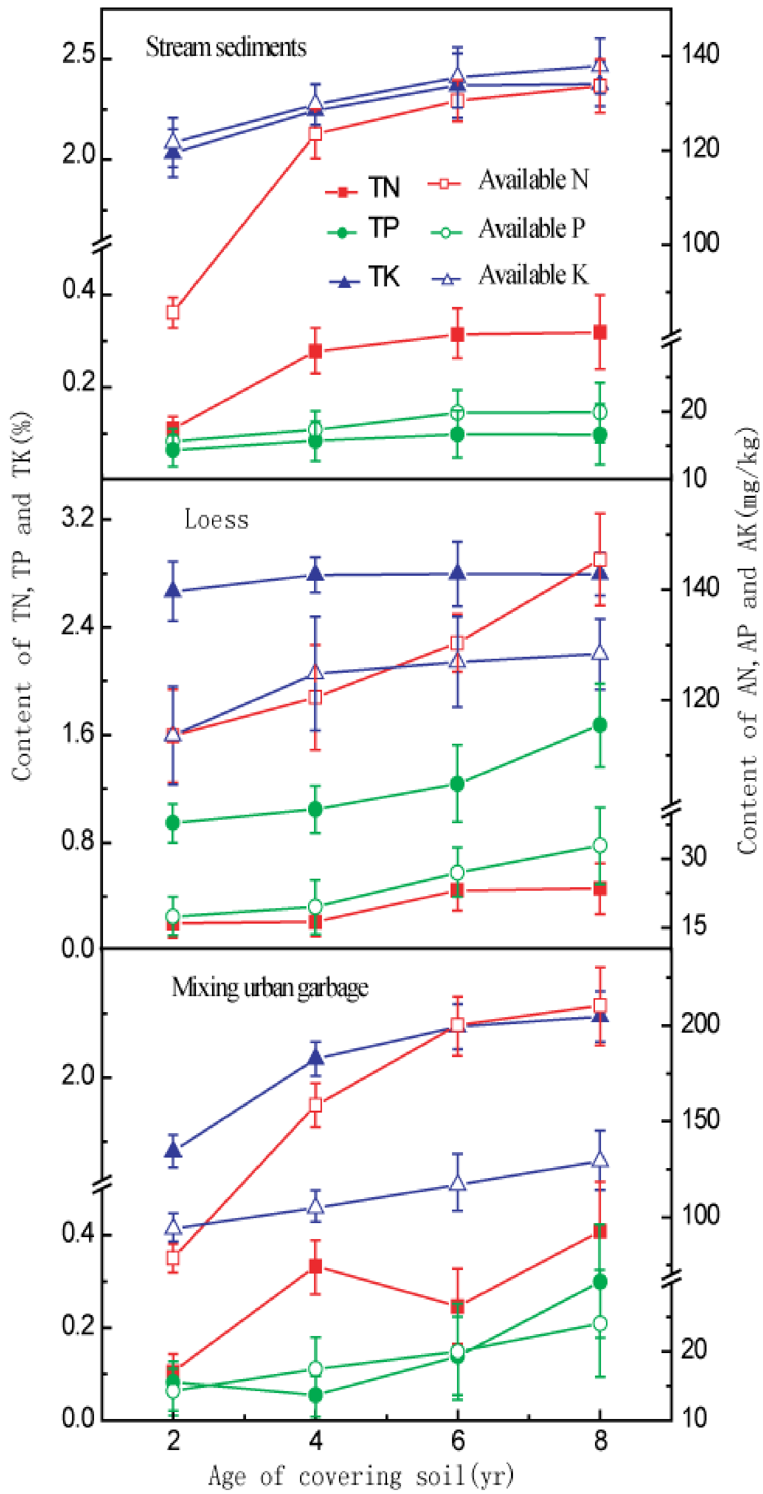

Figure 1. Annual variations of nutrient elements in the topsoil. Vertical bars represent standard deviations of the mean (n = 108).

Zn, Cr and Cu in sewage and sludge.

The contents of all heavy metals, with the exception of Mn, were lower at a depth of 60 to 80 cm than at a depth of 30 to 60 cm. The differences of heavy metal content between soil layers were pronounced. The Cu and Zn contents of topsoil created from stream sediments were 28.1 and 95.4 mg/kg, respectively, about 7.29 and 32.6 mg/kg higher than their initial values, respectively. These

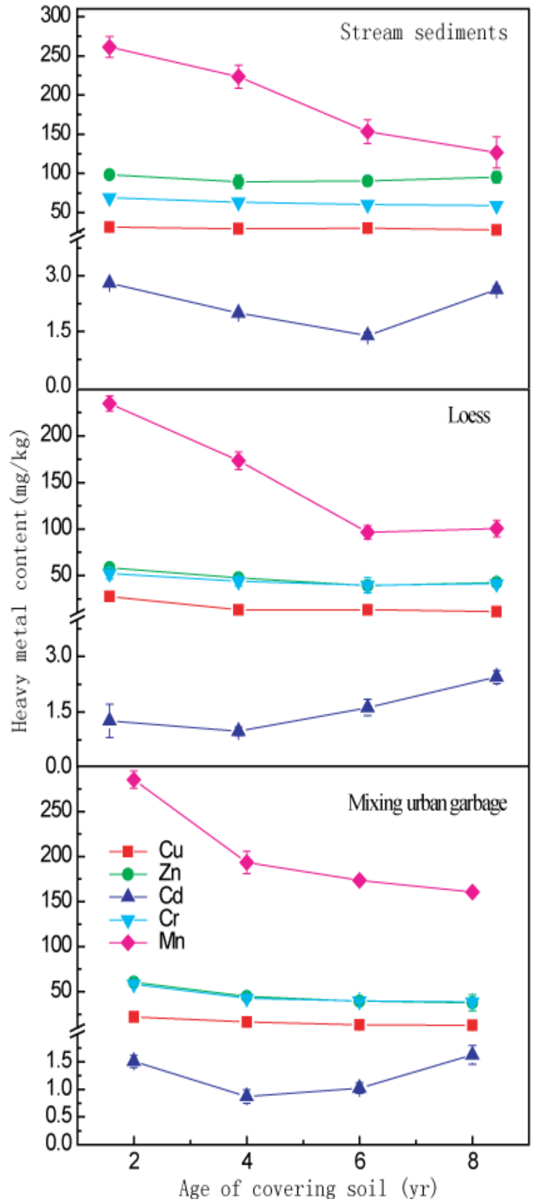

Figure 2. Annual variations of heavy metals in the topsoil. Vertical bars represent standard deviations of the mean (n = 108).

results indicate that the heavy metals did not come from the underlying material (the gangues), but rather from external sources such as the fertilizers used, pollution of the topsoil from the atmosphere, and deposition in dust. If eluviation and evapotranspiration are responsible for the difference, then profiles from previous sampling times should show similar behavior, but with smaller concentrations in the lower soil layers (Zhang, 1992). These indicate that high heavy metals were from external contamination, but not from the gangues, even contaminants can be released after self-ignition and weathering of coal mining wastes (Bian et al., 2009).

Distribution of nutrient elements and heavy metals in the topsoil

Nutrient and heavy metal contents in the 8-year tillage topsoil sampled from depths of 0 to 30 cm are shown in Table 2. It is clear that the TN content in all three kinds of topsoil exceeds China's middle fertility standard and is higher than that of the initial level in the three topsoils. The TN contents in the three topsoils were currently 2.5 to 3.7 times of the initial level.

There is no obvious variation in TK or available P among the three kinds of topsoil. However, TP and available N are higher in the soil derived from mixed urban garbage than in the stream sediment and loess soils. The available K content in mixed urban garbage is lower (by ca. 35 mg/kg) than in the other topsoils. Nonetheless, the nutrient levels in these topsoils is sufficient to grow several plant species.

The heavy metal contents of the three kinds of topsoil differed greatly after 8 years of tillage. With the exception of Mn, most heavy metal contents are lower in mixed urban garbage than in the stream sediment and loess soils, indicating that mixed urban garbage has an advantage as topsoil compared with stream sediment and loess.

Conclusion

In our field experiment, the variations in the nutrient element and heavy metal contents of the topsoils were significantly affected by cultivation and fertilizer application. The nutrient contents of the topsoil increased substantially after 2 years, then continued increasing at a slower rate for the next 6 years of reclamation. The TN contents in all three topsoils increased to 2.5 to 3.7 times that of the original level. The values for TP and available N in mixed urban garbage were higher than in the stream sediment and loess soils. However, the available K in mixed urban garbage was lower (by 35 mg/kg) than in the other topsoils. Levels of nutrient elements are higher in shallower depths than at deeper depths in all types of topsoil. It is significant that the heavy metal contents had decreased after 8 years of reclamation. Most heavy metal contents were lower in the mixed urban garbage soil than in the other topsoils, with the exception of Mn. In contrast with the nutrient elements, the heavy metal contents were lower in the topsoil than in deeper soil layers in all three kinds of topsoil. Based on the observed changes in nutrient elements and heavy metals in the three kinds of topsoil, the mixed urban garbage appears to have advantages over steam sediment and loess soils.

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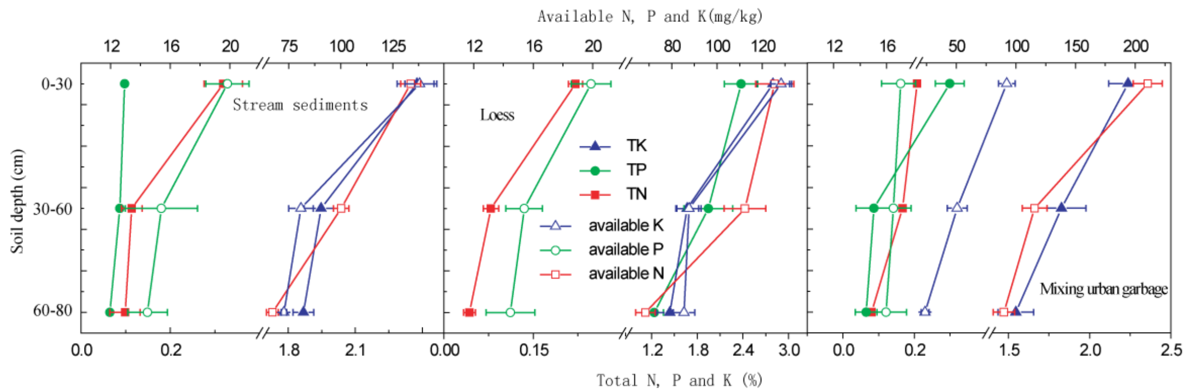

Figure 3. Vertical variation of nutrient elements in the three types of topsoil. Vertical bars represent standard deviations of the mean (n = 144).

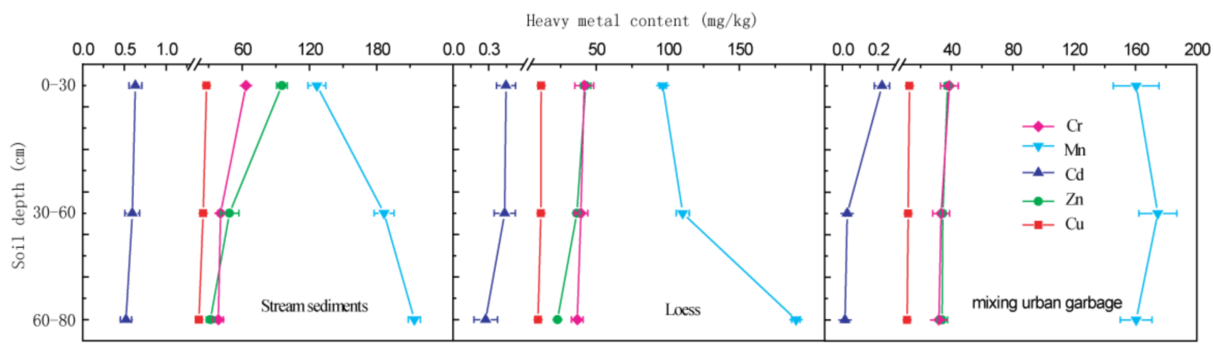

Figure 4. Vertical variation of heavy metal contents in different kinds of topsoil. Vertical bars represent standard deviations of the mean (n = 144).

Table 2. The average nutrient element and heavy metal contents at a depth of 0 - 30 cm in 8-year tillage topsoils and China's middle fertility standard (mg/kg).

Nutrient elements/ Heavy metals	Covering soils			Middle fertility standard
	Stream sediment	Loess	Mixed urban garbage	
TN*	0.217 ± 0.10	0.150 ± 0.08	0.188 ± 0.08	0.05 - 0.08
TP*	0.093 ± 0.04	0.176 ± 0.03	0.193 ± 0.05	
TK*	2.162 ± 0.76	2.232 ± 0.45	2.032 ± 0.77	
AN	116.94 ± 15.14	118.91 ± 22.54	163.11 ± 18.97	
AP	17.62 ± 0.79	17.65 ± 1.15	16.79 ± 2.19	10 - 20
AK	109.42 ± 20.46	107.97 ± 23.76	71.49 ± 0.10	50 - 70
Cu	26.57 ± 7.19	11.25 ± 4.56	10.86 ± 2.33	
Zn	63.47 ± 11.45	39.40 ± 8.56	36.02 ± 5.19	
Cd	0.613 ± 0.34	0.442 ± 0.23	0.217 ± 0.12	
Mn	156.69 ± 17.66	103.43 ± 13.45	167.57 ± 20.77	
Cr	51.93 ± 7.19	40.32 ± 5.85	36.07 ± 3.19	

* Unit of TN, TP and TK is %.

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