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Characteristics and evaluation on heavy metal contamination in Changchun municipal waste landfill after closure

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Abstract

In the present study, comprehensive investigation on the spot and typical investigation method were used to assess Mn, Zn, Pb, Cd, Cr, Ni, As and Cu level, pH value, organic matter, total nitrogen and total phosphorus contents in soil of Changchun municipal waste landfill. The results showed that soil in the closure area of Changchun municipal waste landfill was alkaline in nature and the average value of organic matter, total nitrogen and total phosphorus contents were lower than that in normal black soil in Changchun City of Jilin Province. Single factor indices of As, Pb and Cr content was > 1, where P_{As} was 1.131, P_{Pb} 1.061 and P_{Cr} 1.092 mildly contaminated. In different sample spots but the same landfill time, the comprehensive Nemerow contamination indexes of 7a (5 #) and 7a (2 #) were $P_{2\ comprehensive}$ = 1.176 and $P_{5\ comprehensive}$ = 1.229. The performance value of of heavy metal contamination in soil was similar and there was a low ecological risk.

Key words

Changchun, Contamination index, Heavy metal contamination, Landfill

Introduction

In recent years, with a sharp increase in urban population, the urban household garbage is increasing rapidly at an annual average growth rate of 8%~10%. Currently, the volume of garbage in storage is nearly 8 billion tons, and 5.3 billion square meters of land has been occupied in China. Therefore, China has become a country with the heaviest garbage burden all over the world and urban garbage pollution has become one of the major environmental problems as well as a core concern for environmental research scientist (Chang *et al*., 2007; Hou and Ma, 2005; Wang *et al.,* 2012; Zhang *et al.,* 2003; Zhang *et al.,* 2007). At present, there are incineration, composting and sanitary landfill methods for garbage disposal at home and abroad, and landfill method has been widely used in China, thus forming a large number of landfills (Zhang *et al.,* 2003; Nakasaki *et al.,* 1993; Hou and Ma, 2005; Weng and Chang, 2001; Liu et al., 2007; Long *et al.,* 2007; Meng *et al.,* 2003; Zhou, 2012). As domestic cities do not have separate collection for garbage, high molecular synthetic materials, plastics, batteries and various building materials have been transported into the landfill, resulting in serious heavy metal contamination in landfill, meanwhile the landfill leachate also cause serious pollution to the downstream area and surrounding environment (Wang *et al.,* 2004; Jiang *et al.,* 2006; Kjeldsen *et al.,* 2002; Matejczyk *et al.,* 2011; Melnyka *et al.,* 2014;). Some garbage landfill has been reclaimed by local farmers and grazed by herdsman after being closed for two or three years, so the heavy metals in these wastes have entered human body *via* food chain, causing serious threat to human health (Hao *et al.,* 2009; Huang *et al.,* 2014b; Cui *et al.,* 2004; Hernández-Martínez and Navarro-Blasco, 2012; Jiang *et al.,* 2006: Liu *et al.,* 2006: Khan *et al.,* 2008). Therefore, it is important to master the characteristics of heavy metal contamination in landfills in order to control heavy metal contamination in landfills.

At present, heavy metal contamination at mining site has been reported by several researchers but, reports on heavy metal contamination in landfills is meagre (Lei *et al*., 2005; Huang *et al.,* 2014a; Yang *et al.,* 2012; Liu *et al.,* 2009; Bi *et al.,* 2006; Wang *et al.,* 2007). In view of this, two garbage landfills in Changchun Sandao Sanitary Landfill and Changchun Organic Waste Treatment Center were selected in March 2012 to conduct field investigation. In addition, a comprehensive investigation was carried out to survey Mn, Zn, Pb, Cd, Cr, Ni, As and Cu contamination in soil, in order to grasp the characteristics of contamination. Moreover, status of heavy metal contamination in garbage landfill was also evaluated to provide a basis for reducing heavy metal contamination of landfill in northern cities, which guarantees maximum safety recycling of landfill and to promote environmental management and environmental restoration (Zhou and Song, 2001; Guo *et al.*, 2006; Ru, 2008).

Materials and Methods

Study area : Changchun City is located in mid-latitude zone of Northern Hemisphere and hinterland of Great Plains in Northeast China at the east coast of Eurasia. It lies at 43°05' ~ 45°15' N and 124°18'~127°02' E. It stretches 217.5 km from south to north and 227km from east to west. Changchun City is adjacent to Baicheng City in northwest, connected to Siping City in southwest, near Jilin City in southeast and bordered on Heilongjiang Province in northeast. It is situated along the riverside of Yitong River, which is three-level tributary of Songhua River. The annual average temperature is 4.8°C, where maximum temperature is 39.5°C and minimum temperature is -39.8°C. In Changchun City, the annual average sunshine duration is about 2866 hr, frost-free period is about 140 days and annual average rainfall is 567.0mm.

Changchun Sandao Waste Sanitary Landfill is located in Sandao Town, Erdao District of Changchun City. It covers an area of 48ha and at 7km in the south line of Changchun—Jilin Highway, and also the largest garbage disposal site in Jilin Province. It is divided into four backfilling areas with closure time of 3a, 7a, 14a, 16a respectively; Changchun Organic Waste Treatment Center is located in Luyuan District with closure time of 7a (Fig. 1). Both of them are not standardized closure, because HDPE membrane is not used between the covering layer and waste landfill layer for isolation, and the covering soil is only 30cm thick.

Soil sample collection and analytical testing method : Soil samples were taken from Changchun Sandao Waste Landfill with closure time of 3a, 7a, 14a and 16a respectively and Changchun Organic Waste Treatment Center in Luyuan District with closure time of 7a from April to September, 2012.

Fig. 1 : Distribution diagram of organic waste processing center and Sandao Waste Sanitary Landfill in Changchun

It optimized stationing and conducted S-shaped stationing and sampling according to the area and contamination characteristics of each zone in accordance with different durations of landfill and in line with the *Sampling and Physical Analysis Methods for Municipal Domestic Refuse* (CJ / T3039-95) released by the Ministry of Housing and Urban-Rural Development of the People's Republic of China. Soil at 0-30cm below ground was collected and samples coming from 5-9 points were collected together to compose a mixed sample and was kept in a clean polyethylene bag. Each polyethylene bag was marked and brought back to laboratory. Soil samples were dried naturally and then ground by agate ball after removing stones, glass, plant roots, plastic, etc.. Dried soil samples were sieved through 10 mesh and 20 mesh sieves and divided into two parts by quartering. One part was sieved through 60 mesh sieve and was used to measure pH acidometer; oil and organic matter content was determined by oil bath heating—potassium dichromate volumetric method. AA3 analyzer was used to measure total nitrogen content, flame photometer was used to measure total K content by oxygen-fluorine acid digestion method. The other part of soil was sieved through 100 mesh sieve. Cu, Zn, Pb, Cd, Ni, Mn content was estimated by atomic absorption spectrophotometry silver diethyl dithiocarbamate spectrophotometric method was used to measure As (GB7485-87), while 1, 5 Diphenyl carbohydrazide spectrophotometric method was used to estimate Cr content (Fig. 2).

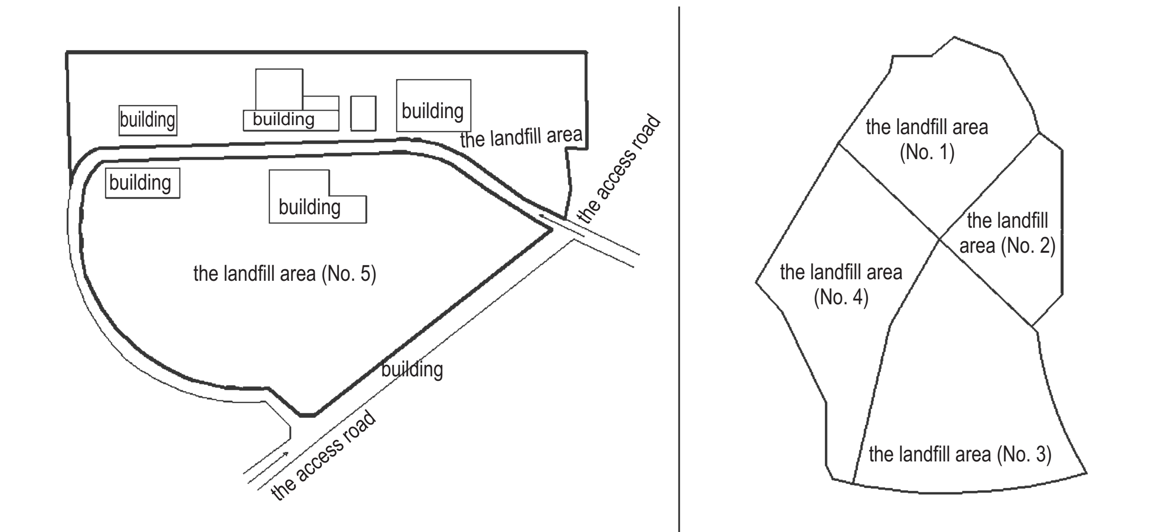
Soil environmental quality evaluation standard and method : Soil fertility evaluation was conducted to compare the measured value of organic matter, total nitrogen, total phosphorus in soil with average value of normal black soil in Changchun of Jilin Province (Qin, 2007; Wu, 1986; Wei, 1990). The evaluation methods used in this study included single factor contamination index method and Nemerow contamination index method which widely accepted. The secondary standard value of Environmental Quality Standard for Soil (GB15618-1995) was used as evaluation standard to evaluate soil pollution state (Table 3) (Xia, 1996; An *et al.,* 2006)

Single factor contamination index method :

$$P_i = C_i / S_i$$

Where, P_i represents the contamination index of i pollutants; C_i represents the actual value of i pollutants; S_i represents the evaluation standard of i pollutants. Grading standards: $P_i \leq 1$ uncontamination; $1 < P_i \leq 2$ mild contamination; $2 < P_i \leq 3$ moderate contamination; $P_i > 3$ heavy contamination.

Comprehensive contamination index method : Nemerow contamination index method is an integrated method compatible



Organic waste treatment center of Changchun City | The sandao landfill sites in Changchun

Fig. 2 : Soil sampling schematic plot

with extreme value, which not only considers the role of single element, but also highlights the importance of element with the most serious contamination.

$$P_{comprehensive} = \sqrt{\frac{1}{2}\left[(\bar{P})^2 + (P_{i,max})^2\right]}$$

where, $P_{comprehensive}$ represents soil contamination comprehensive index; represents average value of element contamination index; $P_{i,max}$ represents the maximum value of single factor contamination index in soil pollutants measured at certain site. The comprehensive contamination index can be used to assess the comprehensive contamination level of soil at each test site. Soil classification standard is given in Table 4.

Assessment of soil ecological risk : Soil ecological risk was evaluated according to the ecological risk assessment reference values provided in Regulations on Soil Contamination Risk Assessment Technique in Key Areas of National Survey of Soil Contamination Status (HF [2008] No. 115) (Table 5) and uses risk quotients to assess the ecological risk of soil by the following formula :

$$Q_i = C_i/R_i$$

where, Q_i : risk index of soil pollutant i (risk quotents); C_i : actually measured concentration of pollutant i at survey point (mg kg^{-1}); R_i : risk assessment reference value of pollutant i (mg kg^{-1}). According to Q_i, degree of risk of soil can be divided into four levels: I : $Q_i \leq 1$ as risk-free; II: $1 < Q_i \leq 3$ as low risk; III: $1 < Q_i \leq 6$ as moderate risk and IV: $Q_i > 6$ as high risk.

Results and Discussion

The results of physico-chemical properties showed that

Table 3 : Environmental Quality Standard for Soils GB15618-1995 (mg kg^{-1})

Elements	I	II			III
	Natural background	**pH<6.5**	**pH (6.5~7.5)**	**pH>7.5**	**pH>6.5**
Cu Farmland ≤	35	50	100	100	400
Orchard ≤	-	150	200	200	400
Zn ≤	100	200	250	300	500
Cr Paddy field ≤	90	250	300	350	400
Dry farmland ≤	90	150	200	250	300
Cd ≤	0.20	0.30	0.30	0.60	1.0
Pd ≤	35	250	300	350	500
Ni ≤	40	40	50	60	200
As Paddy field ≤	15	30	25	20	30
Dry farmland ≤	15	40	30	25	40

as compared to normal black soil, the pH values of soil in Changchun Sandao Waste Landfill with closure time of 3a(1#), 7a(2#), 14a(3#), 16a(4#) and Changchun Organic Waste Treatment Center with closure time of 7a (5#) was alkaline. Viewing from the closure time, as compared to 2# and 5# garbage dumps of landfill 7a, the pH value, organic matter, total nitrogen, total phosphorus contents of 3a, 14a and 16a landfills were relatively high. The pH value, organic matter, total phosphorus, total nitrogen content of 2# and 5# garbage dumps of landfill 7a were higher than the normal average value of black soil in Changchun City of Jilin Province; while the average values of organic matter, total nitrogen, total phosphorus content of five garbage dumps were lower than the average value of normal black soil in Changchun City of Jilin Province (Table 6).

As shown in Table 7, contamination of heavy metal in landfill at different closure time was significantly different. Heavy metal content in 3a (1#) garbage dump was lower, and Cd, As, Pb, Cr, Cu, Zn, Ni, Mn content was 0.16, 26.43, 60.24, 178.29, 20.71, 59.75, 19.83, 614.57 mg kg^{-1} respectively, which might be due to short duration of landfill, so heavy metals in garbage were not fully released. At the same landfill site but at different landfill time, Cd, As, Pb, Cr, Cu, Zn, Ni and Mn content of 7a (2#) garbage dump was 0.45, 44.95, 110.56, 210.29, 29.48, 85.97, 42.76 and 639.45 mg kg^{-1} respectively, and Cd, As, Pb, Cr, Cu, Zn, Ni and Mn content of 7a(5#) garbage dump was 0.42, 43.62, 102.43, 201.36, 31.56, 94.52 , 58.64 and 659.58 mg.kg^{-1} respectively. Heavy metal contents in soil at the same landfill site but different landfill time were similar, most of the heavy metals at 7a(2#) garbage dump were spread, so heavy metal content increased significantly. In addition, heavy metal content was evaluated referring to the pollution limits of various heavy metals in dry farmland with secondary standard as stipulated in the Environmental Quality Assessment Standard for Soils, and the results indicated that As, Pb and Cr contents exceeded the standard values (Xia, 1996). Heavy metal content in 14a(3#) and 16a(4#) garbage dump decreased progressively, where as Cd, As, Pb, Cr, Cu, Zn, Ni and Mn content in 14a(3#) garbage dump was 0.26, 28.27, 76.42, 189.78, 28.72, 67.46, 48.45 and 582.15 mg.kg^{-1} respectively; Cd, As, Pb, Cr, Cu, Zn, Ni and Mn content in 16a(4#) garbage dump was 0.23, 29.34, 74.61, 182.48, 26.69, 66.66, 35.65 and 541.19 mg.kg^{-1} respectively, which might be related to natural vegetation in landfill.

Single factor contamination index method and Nemerow integrated contamination index method were used to evaluate five heavy metals in soil in accordance with the background values of heavy metals in soil of Changchun. The evaluation results are shown in Table 8. As shown in Table 8, contribution order of heavy metal contamination in soil was As > Cr> Pb> Ni>

Table 4 : Classification standard of soil

Classification	Comprehensive contamination index	Class of contamination	Contamination level
1	P≤0.7	Safety	Clean
2	0.7<P≤1.0	Warning line	Clean
3	1.0<P≤2.0	Mild contamination	If the soil contamination exceeds the background value, it will be considered as mild contamination, then the crops will be contaminated.
4	$2.0<P_{comprehensive}\leq 3.0$	Moderate contamination	The soil and crops are suffered from moderate contamination
5	p>3	Heavy contamination	The soil and crops have been heavily contaminated.

Table 5 : Reference value of ecological risk assessment in key area (mg kg^{-1})

Items	Cd	As	Pb	Cr	Cu	Zn	Ni	Mn
Reference value of ecological risk assessment	0.4	18	56	100	100	160	130	4000

Table 6 : Monitoring data of fundamental physical and chemical properties of soil (g kg^{-1})

Closure time /a	Sample number	pH	Organic matter	Total nitrogen	Total phosphorus
3a	1#	7.67	21.01	1.27	0.22
7a	2#	7.65	24.58	1.4	0.45
14a	3#	7.48	12.26	1.32	0.27
16a	4#	7.40	6.03	0.53	0.15
7a	5#	7.32	24.02	1.42	0.49
Average value		7.50	17.58	1.19	0.32
Normal black soil (average value)	6.5—7.0	23.6	1.43	0.33	

Cd> Mn> Zn> Cu. At 7a($2^{\#}$) and 7a($5^{\#}$), single factor index of As, Pb and Cr was > 1. Average value of single factor index of As in P_1~P_5 soil was P_{As}=1.131, P_{Pb}= 1.061, P_{Cr}=1.092 (between 1 and 2), indicating that soil was slightly contaminated by As, Pb and Cr, where As pollution was high. However, in P_3 and P_4 soil, As, Pb, Cr were within the threshold values of slight contamination, indicating potential contamination.

Nemerow comprehensive contamination index method was used to evaluate the soil environmental quality, and the order of degree of soil contamination was 7a($5^{\#}$)>7a($2^{\#}$)>14a($3^{\#}$)>16a ($4^{\#}$)>3a($1^{\#}$) respectively. Where as Nemerow comprehensive contamination indices of 7a($5^{\#}$) and 7a($2^{\#}$) were $P_{2\ comprehensive}$=1.176, and $P_{5\ comprehensive}$=1.229, respectively (between 1.0 and 2.0), so they belonged to light contamination category; the Nemerow contamination indices of the rest sites were between 0.7 and 1.0, and they belonged to warning category.

According to the calculation results and rating standard as shown in Table 9, it could be seen that As, Pb and Cr exceeded the standard value in $1^{\#}$, $2^{\#}$, $3^{\#}$, $4^{\#}$ and $5^{\#}$ landfill sites, indicating a secondary low ecological risk. However, As contamination values in $2^{\#}$ and $5^{\#}$ landfill sites were 2.33 and 2.42, indicating a low ecological risk, but the ecological contamination indices in $2^{\#}$ and $5^{\#}$ landfill sites were higher. At different locations but at same closure time, the ecological risk values in $2^{\#}$ and $5^{\#}$ landfill sites were basically same, where as Cd, As, Pb, Cr, Cu, Zn, Ni and Mn values were 0.80, 2.33, 1.97, 2.80, 0.29, 0.54, 0.33, 0.16; 0.59, 2.42, 1.83, 2.61, 0.32, 0.59, 0.45 and 0.16 respectively.

The soil in Changchun Municipal Solid waste landfill was alkaline and the pH value of soil outside the landfill was between 6.5 and 7.0. At 7a landfill, the pH value, organic matter, total phosphorus and total nitrogen contents at $2^{\#}$ and $5^{\#}$ landfill sites in Changchun City of Jilin Province were higher than the average value of the normal black soil; the average values of organic matter, total nitrogen and total phosphorus contents at $1^{\#}$ to $5^{\#}$ landfill sites were lower than the average value of normal black soil in Changchun City of Jilin Province. In soil of Changchun Municipal Solid Waste Landfill, As, Pb and Cr belonged to light composite contamination. Heavy metal contamination in soil with 3a closure time was minimum, which might be due to short duration of landfill, so that the heavy metals in garbage were still not fully released. Heavy metal contamination in soil with 7a

Table 7 : Heavy metal content in soil (mg kg^{-1})

Sample number	Closure time/a	Cd	As	Pb	Cr	Cu	Zn	Ni	Mn
$1^{\#}$	3a	0.16	26.43	60.24	178.29	20.71	59.75	19.83	614.57
$2^{\#}$	7a	0.32	41.95	110.56	280.29	29.48	85.97	42.76	639.45
$3^{\#}$	14a	0.26	28.27	76.42	189.78	28.72	67.46	48.45	582.15
$4^{\#}$	16a	0.23	29.34	74.61	182.48	26.69	66.66	35.65	541.19
$5^{\#}$	7a	0.38	43.62	102.43	261.36	31.56	94.52	58.64	659.58

Table 8 : Single factor contamination indexes and Nemerow comprehensive contamination indexes of heavy metal in soil (mg kg^{-1})

Single factor contamination index	3a($1^{\#}$)P_1	7a($2^{\#}$)P_2	14a($3^{\#}$)P_3	16a($4^{\#}$)P_4	7a($5^{\#}$)P_5	$P_{average}$
Cd	0.533	1.067	0.867	0.767	1.267	0.900
As	0.881	1.398	0.942	0.978	1.454	1.131
Pb	0.753	1.382	0.955	0.933	1.280	1.061
Cr	0.891	1.401	0.949	0.912	1.307	1.092
Cu	0.207	0.295	0.287	0.267	0.316	0.274
Zn	0.239	0.344	0.270	0.267	0.378	0.299
Ni	0.397	0.855	0.969	0.713	1.173	0.941
Mn	0.410	0.426	0.388	0.361	0.440	0.405
Nemerow comprehensive contamination index $P_{comprehensive}$	0.736	1.176	0.847	0.830	1.229	

Table 9 : Ecological risk index value of soil (mg kg^{-1})

Sample number	Closure time /a	Cd	As	Pb	Cr	Cu	Zn	Ni	Mn
$1^{\#}$	3a	0.40	1.47	1.08	1.78	0.21	0.37	0.15	0.15
$2^{\#}$	7a	0.80	2.33	1.97	2.80	0.29	0.54	0.33	0.16
$3^{\#}$	14a	0.65	1.57	1.36	1.90	0.29	0.42	0.37	0.15
$4^{\#}$	16a	0.58	1.63	1.33	1.82	0.27	0.42	0.27	0.14
$5^{\#}$	7a	0.95	2.42	1.83	2.61	0.32	0.59	0.45	0.16

closure time reached peak value since most of the heavy metals in garbage dump were spread, heavy metal content in soil with 14a-16a closure time decreased progressively, indicating that as compared to other closure time, heavy metal contamination in soil with 7a closure time had reached an order of severity. At the same landfill time but different locations, the performance values of heavy metal contamination index was similar.

In soil, the contribution of contamination of As was maximum (Li *et al*., 1995). In soil of $2^{\#}$ and $5^{\#}$ landfill sites, As, Pb and Cr content exceeded the standard for dry farmland with secondary standard as stipulated in the Environmental Quality Assessment Standard for Soils (Zhang *et al*., 2007a; Gao *et al*., 2006). Soil in $1^{\#}$, $3^{\#}$ and $4^{\#}$ sites belonged to warning level and could not be used for farmland.

The $1^{\#}$, $2^{\#}$, $3^{\#}$, $4^{\#}$ and $5^{\#}$ landfill sites belonged to secondary low ecological risk. $2^{\#}$ and $5^{\#}$ landfill sites were low ecological risk, but the ecological contamination indices were higher. Within 3 to 4 years, the closure area of waste landfill was in a strong degradation period, so $1^{\#}$ landfill site was in an unstable state. Although $1^{\#}$ landfill site was free from soil pollution, but there was a low ecological risk caused by As, Pb and Cr.

It is necessary to conduct land restoration through various channels to control heavy metal contamination in the existing wastes in Changchun Municipal Solid Waste Landfill. For example, plants with ability to accumulate heavy metals can be cultivated on the waste landfill and peripheral areas for re to make directional transfer of heavy metal pollutants toward plants in order to facilitate centralized processing (Alkorta *et al*., 2001; Dahmani-Muller *et al*., 2000; Dahmani-Muller *et al*., 2001; Garbisu *et al*., 2001; Hitchcock *et al*., 2003; Madrid *et al*., 2003). In addition, it can moderately increase inhibitors in the filling process of garbage, so that the contents of heavy metals in landfill were stable.

To sum up, it is of much significance to conduct ecological restoration of landfill site and re-use land after closure.

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