

Changes of Cu, Zn, and Cd speciation in sewage sludge during composting

GAO Ding, ZHENG Guo-di, CHEN Tong-bin*, LUO Wei, GAO Wei, ZHANG Yi-an, LI Yan-xia

(Center for Environmental Remediation, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China. E-mail: chentb@igsrr.ac.cn)

Abstract: The potential toxicity risks from heavy metals depend on their chemical speciation. The four stages of the Tessier sequential extraction method were employed to investigate changes in heavy metal speciation (Cu, Zn, and Cd) of sewage sludge during forced aeration composting, and then to identify whether the composting process would reduce or enhance their toxicities. Throughout the composting process, the exchangeable, carbonate-bound, Fe-Mn oxide-bound, and organic matter-bound fractions of Cu were converted to the residual Cu fraction. The organic matter-bound Cu fraction greatly contributed to this transformation. Residual Zn fraction was transformed to the Fe-Mn oxide-bound and organic matter-bound fractions after composting. The residual Zn fraction was a major contributor to the organic matter-bound Zn fraction. The availability of Cu and Zn was reduced by composting such that the risk of heavy metal toxicity decreased with prolonged treatment times. Additionally, attention should be paid to the increased availability of Cd in sewage sludge after composting treatment.

Keywords: composting; distribution; heavy metal; sewage sludge; speciation

Introduction

Wastewater treatment plants face additional problems as the requirements of China's environmental laws for that dealing with sewage sludge have changed. Because sewage sludge is also a significant source of supplemental nitrogen, phosphorus, and other nutrients for plant growth, its disposal on agricultural lands has been strongly promoted over the past two decades (Pinamonti, 1997; Baveye, 1999). However, heavy metals are potential contaminants in sewage sludge that may affect waste treatment and processing and limit the use of sewage sludge as a soil amendment (Carbonell-Barrachina, 1999; Veeken, 2002).

For developing efficient waste disposal collection and treatment systems, the concentrations of heavy metals in sewage sludge have been reduced in recent years, relieving the environmental hazards of applying sludge to land (Chen, 2003). However, since most of the sewage sludge contains low levels of pollutants, it therefore meets the strict regulations of the countries concerned (Führacker, 1995; Chen, 2003).

Some methods, such as acid treatment, electrodialytic remediation, and bioleaching, when used prior to land application, can be used to dissolve and remove a significant fraction of heavy metals from sludge (Veeken, 1999; Solisio, 2002; Jakobsen, 2004). Nevertheless, many of these approaches are marginally cost-effective or difficult to implement in developing countries. These processes may additionally result in a significant loss of the nutritional value of sewage sludge (Shanableh, 1999).

The potential hazard of heavy metals depends on their speciation in sewage sludge (Ho, 1998; Pérez-Cid, 1999; Parkpain, 2000; Zheng, 2004); therefore it is not sufficient to evaluate the degree of metal toxicity solely on the total concentration of the heavy metal (Fuentes, 2004). A sequential extraction method has been used in an attempt to evaluate solid phase speciation, which is reported to provide useful information on the chemical associations of metals (Tessier, 1979).

Composting is one of the most efficient and environmentally friendly methods of municipal sewage sludge disposal. Through the composting process, the hazards of sewage sludge can be reduced, including pathogen death and organic matter degradation. In addition, the concentrations of the contaminating metals may change after the composting process (Planquart, 1999; Fang, 1999; Li, 2000). Due to the reduced volume and weight of the raw material after composting, the changes in toxicities of the metals cannot be directly assessed by their total concentrations alone.

Generally, Chinese sewage sludge contains higher concentrations of Zn and Cu than of other metals (Chen, 2003). Zn and Cu are often the two elements that exceed national standards (GB4284-84) for land application of sewage sludge. Cd is also a concern because of its potentially greater harmful effects than those of other elements (Chen, 2003).

In this work, the Tessier sequential extraction method was applied to operationally define metal speciation, and subsequently, to monitor the changes in metal chemical speciation distribution after sewage sludge composting. This study mainly focused on the influence of composting on Cu, Zn, and Cd speciation in order to determine how composting effects the toxicities of these heavy metals.

1 Materials and methods

Dehydrated, digested sewage sludge was collected from the Beixiaohe Wastewater Treatment Plant, Beijing. Sewage sludge was mixed with an inorganic bulking agent at a ration of 1:1 (v/v) and the moisture content of the reactor was adjusted to 60%.

The top of the compost pile was uncovered with dimensions of 2.5 m × 1.5 m × 1.4 m. The pile was overlaid with a 0.1 m layer of mature sludge compost to retain moisture and heat. The treatment process was automatically controlled by a forced aeration static pile system that included continuous temperature monitoring. The total composting duration was 31 d. The entire process was divided into three phases: 0–5 d: mesophilic phase (< 50°C), 5–25 d:

thermophilic phase($\geq 50^{\circ}\text{C}$), and 25—31 d: cooling phase. Samples were regularly taken from the pile at days 0, 2, 5, 9, 14, 19, 25, and 31. The samples were subsequently air-dried, crushed, and passed through a plastic sieve with mesh sizes of 0.075 and 1.0 mm in preparation for chemical testing.

The Tessier sequential extraction method, used previously in soil science, was applied to study the metal fractions of the samples(Tessier, 1979; Flyhammar, 1998; Illera, 2000). Two grams of each sample were weighed into a 50 ml polypropylene centrifuge tube and the following fractions were obtained: exchangeable(1.0 mol/L of MgCl_2 , pH 7); carbonate-bound(1 mol/L of NaOAc/HOAc , pH 5); Fe-Mn oxide-bound (40 mmol/L of $\text{NH}_2\text{OH} \cdot \text{HCl}$ in 25% HOAc); organic matter-bound(20 mmol/L of HNO_3 in 30% H_2O_2 , pH 2; 3.2 mol/L of NH_4OAc in 20% HNO_3); and residual (digested with concentrated $\text{HNO}_3 + \text{HClO}_4$). The Cu and Zn concentrations were determined using flame atomic absorption spectrophotometry and the Cd concentration was

determined using flame atomic absorption spectrophotometry with graphite furnace (AAS Vario 6, Germany). The dissolved organic carbon(DOC) concentration was analyzed by a total organic carbon analyzer(Apollo 9000, USA) and pH was determined using a pH meter (REX PHSJ-4A, China). All measurements were conducted in triplicate for each sample and the results were presented as the average of three replicates. Correlation analysis was conducted using Statistical Analysis System software.

2 Results

2.1 Concentrations and speciation distribution of Cu during composting

Table 1 shows the concentrations and distribution of various species of Cu during composting. During the treatment process, the total concentration of Cu steadily increased from 138.6 mg/kg to 169.2 mg/kg. The rates of concentration increase differed between the three composting phases.

Day	Total	F1		F2		F3		F4		F5	
	mg/kg	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%
0	138.6	3.36	2.43	5.09	3.67	2.71	1.95	89.6	64.6	37.9	27.3
2	136.0	4.03	2.96	4.38	3.22	2.86	2.10	90.2	66.3	34.5	25.4
5	141.7	2.58	1.82	4.36	3.08	2.94	2.07	87.1	61.5	44.7	31.6
9	148.2	2.64	1.78	4.22	2.84	2.55	1.72	86.4	58.3	52.4	35.3
14	159.7	2.74	1.71	3.95	2.47	2.69	1.68	92.5	57.9	57.9	36.2
19	162.0	2.68	1.65	3.85	2.38	2.63	1.63	90.1	55.6	62.7	38.7
25	166.0	2.52	1.52	3.49	2.10	2.58	1.55	82.7	49.8	74.7	45.0
31	169.2	2.59	1.53	3.10	1.83	2.18	1.29	87.3	51.6	74.0	43.7

Notes: F1. exchangeable; F2. carbonates bound; F3. Fe-Mn oxides bound; F4. organic matter bound; F5. residual; % . percentage of distribution with respect to its total concentration

At the beginning of the composting process, the concentration of exchangeable Cu increased from 3.36 mg/kg to 4.03 mg/kg, but later decreased to 2.58 mg/kg during the mesophilic phase. However, the concentration changed only slightly during the thermophilic and cooling phases through the day 26. The concentration of carbonate-bound Cu decreased steadily from 5.09 mg/kg to 3.10 mg/kg during the entire process. The concentration of Fe-Mn oxide-bound Cu increased during the mesophilic phase and decreased in the thermophilic phase. The concentrations of organic matter-bound and residual Cu were maintained at high levels during the composting process, and the concentration of residual Cu increased during the mesophilic and thermophilic phases, while remaining unchanged during the final cooling phase.

The speciation distribution of Cu changed in the following manner: the percentage of exchangeable and organic matter-bound Cu increased in the mesophilic phase and decreased in the thermophilic phase. The percentage of carbonate-bound Cu steadily decreased from 3.67% to 1.83% during the treatment process. The percentage of residual Cu decreased just prior to initiation of the mesophilic phase; but increased from the anaphase phase and ended with 43.7% of the final product. The distribution of Fe-Mn oxide-bound Cu was maintained at 2% at the initiation of the mesophilic phase and decreased slightly afterwards. The distribution of the organic matter-bound Cu decreased from 64.6% to 51.6% by the end of composting process.

2.2 Concentrations and speciation distribution of Zn during composting

The variation in concentrations and speciation distribution of Zn during composting is shown in Table 2. The concentration of Zn increased 34% in contrast to the raw material. The exchangeable Zn decreased dramatically at the initiation of the composting process and maintained a stable level of 1.5 mg/kg through the thermophilic phase. The carbonate-bound Zn fraction varied slightly at the beginning of the mesophilic phase, but increased rapidly at the end of this phase to reach its highest concentration. The concentration of Fe-Mn oxide-bound Zn increased slowly during the mesophilic phase, and then increased significantly from 101.8 mg/kg to 156.9 mg/kg during the thermophilic phase, accounting for 179% of the raw material. Through composting, the concentration of organic bound-Zn in the final product accounted for 220% of the raw material. Residual Zn decreased during the composting process. The Fe-Mn oxide-bound and residual Zn fractions were different from the other three fractions as they decreased during the cooling phase.

The distribution of exchangeable Zn decreased steadily from 0.90% to 0.29% during the composting process. The distribution of carbonate-bound Zn varied little during the process. Fe-Mn oxide-bound Zn increased during composting, especially during the thermophilic phase. While there were no significant changes during the initiation of the mesophilic phase in the organic matter-bound Zn, a rapid

increase occurred during the remaining phases of the process . The distribution of residual Zn varied little during the mesophilic phase, but gradually decreased to 30.6% .

2.3 Concentrations and speciation distribution of Cd during composting

Concentrations and speciation distribution of Cd during composting are shown in Table 3. The concentrations of total Cd and its four fractions (exchangeable, carbonate-bound,

Fe-Mn oxide-bound, and organic matter-bound) increased during the composting process, especially during the thermophilic phase. The concentration of residual Cd decreased from 203 $\mu\text{g/kg}$ to 140 $\mu\text{g/kg}$, and then increased during the thermophilic phase(the day 14) . It then decreased to 81 $\mu\text{g/kg}$ by the end of the thermophilic phase. However, the concentration of residual Cd increased to 121 $\mu\text{g/kg}$ during the cooling phase.

Table 2 Concentrations and distribution of the various species of Zn during composting

Day	Total	F1		F2		F3		F4		F5	
	mg/kg	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%
0	378.6	3.41	0.90	2.73	0.72	87.7	23.2	85.0	22.5	199.7	52.7
2	381.8	3.05	0.80	2.50	0.65	89.7	23.5	84.5	22.1	202.0	52.9
5	417.5	2.52	0.60	4.29	1.03	101.8	24.4	121.9	29.2	187.0	44.8
9	424.3	1.83	0.43	3.71	0.87	103.0	24.3	127.5	30.0	188.3	44.4
14	435.3	1.54	0.35	3.74	0.86	116.9	26.8	136.0	31.2	177.2	40.7
19	472.0	1.76	0.37	3.74	0.79	138.4	29.3	152.5	32.3	175.6	37.2
25	490.8	1.53	0.31	3.8	0.77	156.9	32.0	166.7	34.0	161.9	33.0
31	507.4	1.48	0.29	3.61	0.71	157.5	31.0	189.5	37.3	155.3	30.6

Notes: F1. exchangeable; F2. carbonates bound; F3. Fe-Mn oxides bound; F4. organic matter bound; F5. residual; % . percentage of distribution with respect to its total concentration

Table 3 Concentrations and distribution of the various species of Cd during composting

Day	Total	F1		F2		F3		F4		F5	
	$\mu\text{g/kg}$	$\mu\text{g/kg}$	%	$\mu\text{g/kg}$	%	$\mu\text{g/kg}$	%	$\mu\text{g/kg}$	%	$\mu\text{g/kg}$	%
0	1228	42	3.46	34	2.78	758	61.7	191	15.6	203	16.5
2	1237	44	3.61	35	2.86	818	66.1	199	16.1	140	11.3
5	1353	43	3.20	36	2.69	869	64.3	231	17.1	173	12.8
9	1386	48	3.45	38	2.76	893	64.5	250	18.0	156	11.3
14	1462	52	3.54	41	2.80	952	65.1	259	17.7	159	10.8
19	1495	55	3.69	42	2.81	1020	68.2	276	18.5	101	6.8
25	1552	59	3.77	45	2.92	1073	69.1	295	19.0	81	5.2
31	1666	70	4.17	46	2.76	1123	67.4	306	18.4	121	7.3

Notes: F1. exchangeable; F2. carbonates bound; F3. Fe-Mn oxides bound; F4. organic matter bound; F5. residual; % . percentage of distribution with respect to its total concentration

The distribution of the exchangeable and carbonate-bound Cd changed slightly during the composting process. The percentage of Fe-Mn oxide-bound Cd increased to 67.4% in the final product compared with 61.7% of the raw material. The percentage of organic matter-bound Cd increased from 15.6% to 19.0% throughout the thermophilic phase and slightly decreased throughout the cooling stage. The distribution of residual Cd decreased steadily from 16.5% to 5.2% throughout the thermophilic phase, and then increased to 7.3% in the final product.

2.4 Correlation of pH and DOC to the heavy metal fractions

Fig.1 shows that the pH of the compost decreased from 6.21 to 5.88 during the initial mesophilic phase. On the day 2, the pH of the compost increased rapidly from 5.88 to 6.53. The pH was maintained between 6.5 and 6.7 throughout the high temperature stage until the day 19. At the latter stage of the composting process, the pH increased again to a maximum value of pH 7.57 in the final product.

The dependence of the heavy metal fractions on pH was examined by linear regression and correlation as Table 4.

DOC decreased dramatically during the mesophilic and thermophilic phases (Fig.2), but did not vary much during the cooling phase, remaining stable at 5 g/kg. The

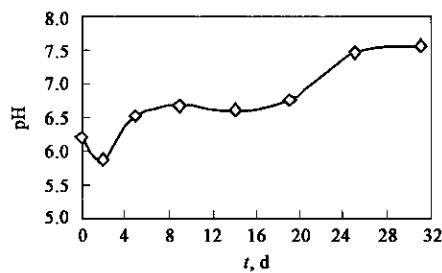


Fig.1 Variation of pH during composting

dependence of various heavy metal fractions on DOC was examined by linear regression and correlation as Table 5.

Table 4 pH correlation with the fractions distribution of heavy metals (n = 8)

Heavy metals	F1	F2	F3	F4	F5
Cu	- 0.8495 **	- 0.8874 **	- 0.8947 **	- 0.9685 **	+ 0.9678 **
Zn	- 0.8322 *	+ 0.0296	+ 0.9030 **	+ 0.9338 **	- 0.9533 **
Cd	+ 0.6732	+ 0.1399	+ 0.6410	+ 0.8540 **	- 0.7537 *

Note: F1. exchangeable; F2. carbonates bound; F3. Fe-Mn oxides bound; F4. organic matter bound; F5. residual; * $p < 0.05$; ** $p < 0.01$

3 Discussion

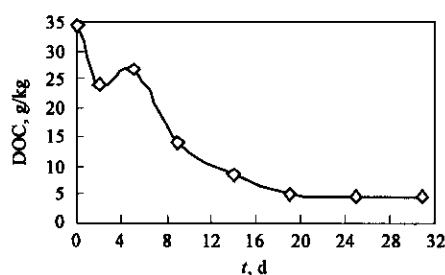


Fig. 2 Variation of DOC during composting

Throughout the composting treatment process, the total concentrations of Cu, Zn, and Cd increased to 22.1%, 34.0%, and 35.6%, respectively. Composting is the solid-phase decomposition of organic material by microorganisms to produce heat, carbon dioxide, and water vapor (Hackett, 1999). With the volatilization of gases and water, the heavy metals in sewage sludge are enriched and their total concentrations in sewage sludge are commonly increased during composting. Due to differential recovery of the heavy metal fractions, the total concentration percentages exhibited different rates of increase (Ho, 1998).

Table 5 DOC correlation with the fractions distribution of heavy metals ($n = 8$)

Heavy metal	F1	F2	F3	F4	F5
Cu	+0.7344*	+0.9499**	+0.8546**	+0.8794**	-0.8890**
Zn	+0.9457**	+0.0735	-0.8620**	-0.8646**	+0.8897**
Cd	-0.6620	-0.3622	-0.8303*	-0.9274**	+0.9119**

Note: F1. exchangeable; F2. carbonates bound; F3. Fe-Mn oxides bound; F4. organic matter bound; F5. residual; * $p < 0.05$; ** $p < 0.01$

By composting, the total concentrations of the heavy metals were enriched and the concentrations of the various heavy metal fractions were changed. Total concentrations of the heavy metals are usually used to assess their potential toxicity hazards for land application of sewage sludge compost. However, research has shown that the degree of heavy metal hazard to the environment is not only related to its total concentration in sewage sludge, but also to its chemical speciation (Parkpain, 2000). As the quantity of the heavy metal in sludge appears unchanged, the changes in the percentage of that heavy metal's various fractions compared to the total can be used to assess changes in its availability. The variations in the heavy metal fractions' distribution may be a better tool to assess sewage sludge hazard compared to its total concentration.

Using the percentages of the distribution of five different

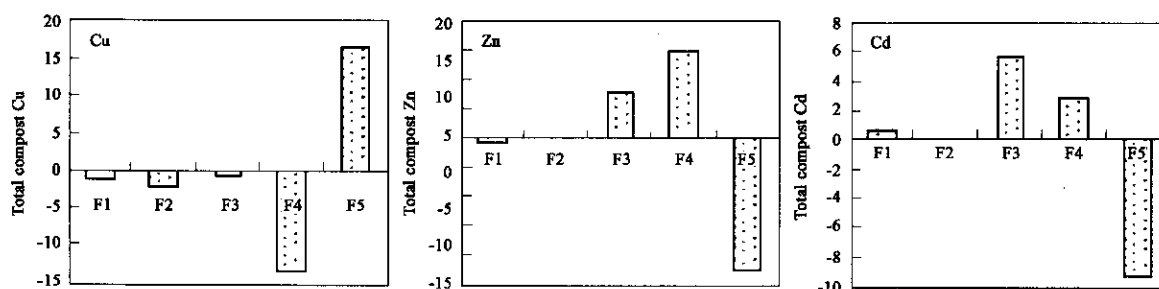


Fig. 3 Distribution changes of the heavy metals various fractions after composting

F1. exchangeable; F2. carbonates bound; F3. Fe-Mn oxides bound; F4. organic matter bound; F5. residual; changes of fractions distribution (%): the percentages of five different fractions distribution in the raw sludge subtracted from their percentages in the final product

fractions in the raw sludge subtracted from their percentages in the final product, we obtained the variation in the heavy metal fractions' distribution throughout composting (Fig. 3). This analysis will aid the understanding of the chemical transformation among the five fractions and assess if composting can limit or enhance their toxicity risk.

The exchangeable fractions of heavy metals are the most toxic and are released only after increasing the ionic strength of the medium. The metals present in the exchangeable fraction are considered to be the most available fraction for uptake by plants. Additionally, metals bound to carbonates are very sensitive to pH changes and are leached by lowering the pH (Pérez-Cid, 1999), and therefore, heavy metals present in these two fractions are considered to be the most available fractions for plant uptake.

After composting, the distribution of exchangeable, carbonate-bound, Fe-Mn oxide-bound, and organic matter-bound Cu decreased while the distribution of residual Cu increased. These results imply that the four fractions of Cu transform to the residual Cu fraction during composting, especially the organic matter-bound fraction. The composting process dramatically diminishes the distribution of residual Zn and increases the distribution of the Fe-Mn oxide-bound and organic matter-bound Zn fractions. This means that residual Zn is possibly transformed to the Fe-Mn oxide-bound and organic matter-bound Zn fractions. The transformation trend of Cd was quite similar to that of Zn, except that the distribution of exchangeable Zn increased while the distribution of exchangeable Cd decreased.

DOC is generally very mobile and, in leachates of municipal materials, is an important factor relating to solubility (Bolton, 1991). DOC has the ability to form stable, soluble complexes with heavy metals (Ashworth, 2004) and significantly increases the extractability of metals and their uptake by plants (Antoniadis, 2002). There are several high molecular weight molecules of DOC in sewage sludge, such as polyphenols, simple aliphatics and amino acids. These molecules have stronger adsorption abilities than lower molecular weight molecules. The quantity of adsorption increases through DOC integration with heavy metals (Chen, 1998). Mobility and availability of Cu increases with increasing complex formation with organic ligands. Perhaps the reason why exchangeable Cu was reduced more significantly during the period of the mesophilic and the thermophilic phases was because of organic matter binding.

Hsu and Lo (Hsu, 2001) found that metal distribution of different chemical fractions was generally independent of

composting time, and thus, independent of total metal concentration in swine manure compost. However, the results of our study showed that availability of heavy metals in sludge could be reduced over time by composting. The process of composting appears to transform high availability fractions to low availability fractions. Therefore, the availability and distribution of Cu and Zn fractions were reduced by composting. In contrast, the distribution of exchangeable Cd increased during composting and increased its availability. The concentrations and distribution of exchangeable Cu, Zn, and carbonate-bound Cu were higher in young sludge compost, especially during the mesophilic and the thermophilic phases. The potential toxicity hazards of these metals in young sludge compost were demonstrated to be more serious than those in mature sludge compost and the composting process reduced metal bioavailability.

4 Conclusions

After composting, the heavy metals Cu, Zn, and Cd, were enriched in sewage sludge and their concentrations increased by 22.1%, 34.0%, and 35.7%, respectively. During this process, the exchangeable, carbonate-bound, Fe-Mn oxide-bound and organic matter-bound Cu was transformed to residual Cu. Residual Zn was transformed to the Fe-Mn oxide-bound and organic matter-bound Zn fractions. Residual Zn was a major contributor to the fraction of organic matter-bound Zn. Composting reduced the availability of Cu and Zn, and increasing composting treatment time is predicted to decrease their toxicity hazards. More attention should be paid to the increased availability of Cd in sewage sludge compost resulting from composting treatment.

References:

- Antoniadis V, Alloway B J, 2002. The role of dissolved organic carbon in the mobility of Cd, Ni and Zn in sewage sludge-amended soils[J]. *Environ Pollut*, 117(3): 515—521.
- Ashworth D J, Alloway B J, 2004. Soil mobility of sewage sludge-derived dissolved organic matter, copper, nickel and zinc[J]. *Environ Pollut*, 127(1): 137—144.
- Baveye P, McBride M B, Bouldin D *et al.*, 1999. Mass balance and distribution of sludge-borne trace elements in silt loam soil following long-term applications of sewage sludge[J]. *Sci Total Environ*, 227(1): 13—18.
- Bolton K A, Evans L J, 1991. Elemental composition and speciation of some landfill leachates with particular reference to cadmium[J]. *Water Air Soil Poll*, 60(1): 43—53.
- Carbonell-Barrachina A A, Jugsujinda A, Burlo F *et al.*, 1999. Arsenic chemistry in municipal sewage sludge as affected by redox potential and pH[J]. *Water Res*, 34(1): 216—224.
- Chen T B, Chen Z J, 1998. Dissolved organic matter and its effects on adsorption and desorption of pollutants in soils[J]. *Plant Nutrition and Fertilizer Science*, 4(3): 201—210.
- Chen T B, Huang Q F, Gao D *et al.*, 2003. Heavy metal concentrations and their decreasing trends in sewage sludges of China[J]. *Acta Scientiae Circumstantiae*, 23(5): 561—569.
- Fang M, Wong J W C, 1999. Effect of lime amendment on availability of heavy metals and maturation in sewage sludge composting[J]. *Environ Pollut*, 106(1): 83—89.
- Flyhammar P, 1998. Use of sequential extraction on anaerobically degraded municipal solid waste[J]. *Sci Total Environ*, 212(2/3): 203—215.
- Fuentes A, Lloréns M, Sáez J *et al.*, 2004. Phytotoxicity and heavy metals speciation of stabilised sewage sludges[J]. *J Hazard Mater*, 108(3): 161—169.
- Fürhacker M, Haberl R, 1995. Composting of sewage sludge in a rotating vessel[J]. *Wat Sci Tech*, 32(11): 121—125.
- Hackett G A R, Easton C A, Duff S J B, 1999. Composting of pulp and paper mill fly ash with wastewater treatment sludge[J]. *Bioresource Technol*, 70(3): 217—224.
- Ho G, Qiao L, 1998. Chromium speciation in municipal solid waste: Effects of clay amendment and composting[J]. *Wat Sci Tech*, 38(2): 17—23.
- Hsu J H, Lo S L, 2001. Effect of composting on characterization and leaching of copper, manganese, and zinc from swine manure[J]. *Environ Pollut*, 114(1): 119—127.
- Illera V, Walter I, Souza P *et al.*, 2000. Short-term effects of biosolid and municipal solid waste applications on heavy metals distribution in a degraded soil under a semi-arid environment[J]. *Sci Total Environ*, 255(1/3): 29—44.
- Jakobsen M R, Fritt-Rasmussen J, Nielsen S *et al.*, 2004. Electrodialytic removal of cadmium from wastewater sludge[J]. *J Hazard Mater*, 106(2/3): 127—132.
- Li G X, Zhang F S, 2000. Solid waste composting and production of fertilizer[M]. Beijing: Chinese Chemical Industry Press. 65—67.
- Parkpain P, Sreesai S, Delaune R D, 2000. Bioavailability of heavy metals in sewage sludge-amended Thai soils[J]. *Water Air Soil Poll*, 122(1/2): 163—182.
- Pérez-Cid B, Lavilla I, Bendicho C, 1999. Application of microwave extraction for partitioning of heavy metals in sewage sludge[J]. *Anal Chim Acta*, 378(1/3): 201—210.
- Pinamonti F, Stringari G, Gasperi F *et al.*, 1997. The effect of compost: its effects on heavy metal levels in soil and plants[J]. *Resour Conserve Recy*, 21(2): 129—143.
- Planquart P, Bonin G, Prone A *et al.*, 1999. Distribution, movement and plant availability of trace metals in soils amended with sewage sludge composts: application to low metal loadings[J]. *Sci Total Environ*, 241(1/3): 161—179.
- Shanableh A, Ginige P, 1999. Impact of metals bioleaching on the nutrient value of biological nutrient removal biosolids[J]. *Wat Sci Tech*, 39(6): 175—181.
- Solisio C, Lodi A, Veglio F, 2002. Bioleaching of zinc and aluminium from industrial waste sludges by means of *Thiobacillus ferrooxidans*[J]. *Waste Manage*, 22(6): 667—675.
- Tessier A, Campbell P G C, Bisson M, 1979. Sequential extraction procedure for the speciation of particulate trace metals[J]. *Anal Chem*, 51(7): 844—851.
- Veeken A, Hamelers B, 2002. Sources of Cd, Cu, Pb and Zn in biowaste[J]. *Sci Total Environ*, 300(1/3): 87—89.
- Veeken A H M, Hamelers H V M, 1999. Removal of heavy metals from sewage sludge by extraction with organic acids[J]. *Wat Sci Tech*, 40(1): 129—136.
- Zheng G D, Chen T B, Gao D *et al.*, 2004. Dynamic of lead speciation in sewage sludge composting[J]. *Wat Sci Tech*, 50(9): 75—82.

(Received for review February 6, 2005. Accepted April 22, 2005)