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# Variations of metal distribution in sewage sludge composting

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## Abstract

In the study, the variations of heavy metal distributions (of Cu, Mn, Pb, and Zn) during the sewage sludge composting process were investigated by sequential extraction procedures. The total content of Cu and Zn in the composted mixture increased after the composting process. Mn and Zn were mainly found in mobile fractions (exchangeable fraction (F1), carbonate fraction (F2), and Fe/Mn oxide fraction (F3)). Cu and Pb were strongly associated with the stable fractions (organic matter/sulfides fraction (F4) and residual fraction (F5)). These five metal fractions were used to calculate the metal mobility (bioavailability) in the sewage sludge and composted mixture. The mobility (bioavailability) of Mn, Pb, and Zn (but not Cu) increased during the composting process. The metal mobility in the composted mixture ranked in the following order: Mn > Zn > Pb > Cu.

## 1. Introduction

The rapid increase of world population leads to significant amounts of waste discharge. At present, environmentally friendly and cost effective methods of waste utilization have received much attention. The European Union (EU) Waste Framework Directive (91/156/EEC amending 75/442/EEC) confirms the waste management hierarchy. Preference is given to waste prevention, followed by waste reduction, re-use, recycling, and energy recovery. In 1980, the main solid waste management option in the USA was landfilling, and only a minor part was incinerated, or otherwise utilized. However, the extent of waste utilization steadily increased, and it is estimated that almost 35% of all waste in the USA will be utilized in 2010 (USEPA, 1997).

In order to improve the wastewater control process, the Taiwan Government has decided to raise the popularization of sewer systems by a rate of 3% per year. Due to the expansion of population, the number of municipal sewage treatment plants (MSTP) is increasing rapidly, and a great amount of sewage sludge will be produced. It is estimated that 832 tons of sewage sludge per day will be generated in 2010. Surveying the reports of sewage sludge production worldwide shows that the total amount of sewage sludge produced in 15 EU countries increased to at least 9.4 million tonnes of dry matter in 2005 (EEA, 2005). Therefore, the disposal of waste sewage sludge will become an environmental issue in the future. Not only in European countries, but all over the world, scientists and producers make every effort to reuse sewage sludge as a valuable bio-fertiliser. The sewage sludge usually contains a high content of organic matter and nutrients, and therefore it can be used as a fertiliser for land application. Although such application may be beneficial to soil, its negative impacts, such as groundwater pollution, pollutant accumulation in soil, etc., should be considered (Veeken and Hamelers, 2002; Peigne and Girardin, 2004; Wei and Liu, 2005). Analysis of EU Environmental Legislative documents (Directives, Regulations in force and projects) showed that the requirements of sewage sludge, composted

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material, and anaerobic digestate for land application are becoming more stringent, especially with regard to heavy metals (Horn et al., 2003).

In general, the total amount of heavy metals in the sewage sludge cannot fully reflect their ecological impact in the soil environment. In addition to the quantity of heavy metals, the mobility and bioavailability of heavy metals are considered important for predicting the release of metals into soil and subsequent absorption by plants (Song and Greenway, 2004). To date, various single-step extraction procedures are used to determine metal bioavailability (Ure, 1996; Korboulewsky et al., 2002; During et al., 2003; Hseu, 2004). However, the sequential extraction procedures used in metal bioavailability can isolate the fractions of heavy metal more specifically (Song and Greenway, 2004). The purposes of this study were twofold: (1) to investigate the metal speciation in the raw materials and composted mixture by a sequential extraction method, and (2) to evaluate the variations of metal distribution during the composting process. In addition, the metal mobilities (bioavailabilities) determined by the metal fractions before and after composting were compared.

## 2. Materials and methods

### 2.1. Composting procedures

The aerobically digested and dewatered sludges were collected from the Min-Shen MSTP in northern Taiwan. The sludge composting was carried out in two 13 $m^3$ aerated piles in the pilot-scale composting plant at the Pa-Li MSTP in Taipei, Taiwan. The weight based percentages of sludge, bulking agent (sawdust), and recycled compost (obtained from previous studies and used as inoculants) were 62% (5490 kg), 24% (2122 kg) and 14% (1265 kg), respectively. Air flow rate was controlled by a computer-regulated solenoid valve and was measured by a flow meter. The average air flow rate was 13.85 $m^3\ h^{-1}$. The composted mixture was turned once a week during the first month of the composting cycle and twice a week during the second month. The composted mixture was sampled weekly during the 2-month period. Samples were air-dried at room temperature, and were thoroughly mixed and ground to pass through a 2 mm sieve for further analyses.

### 2.2. Raw material and compost characterization

In this study, pH (USEPA, 1995), moisture, organic matter (volatile solids) (Standard Method 2540G), and total nitrogen (CHN-OS Rapid Elemental Analyzer, Herneus) of the raw materials (sludge, sawdust and recycled compost) and composted mixture were analysed. The total heavy metal content was determined according to the HF–$HNO_3$–HCl digestion method (USEPA, 1996). Heavy metals were measured by a flame atomic absorption spectrophotometer (AAS) equipped with a graphite burner (Model Z-8100, Hitachi).

### 2.3. Fractionation procedures

Generally, the fractionation of metal in sludge and composted mixture was performed by using a sequential extraction procedure (Qiao and Ho, 1996; Hsu and Lo, 2001; Song and Greenway, 2004). The sequential extraction used in this study was modified by Lin et al. (1999). This method is designed to partition the trace metals into the following five fractions:

(1) Exchangeable fraction (F1): the solid sample (1.5 g) was extracted by 1 M $MgCl_2$ at an initial pH 7 (30 mL), and was shaken at 250 rpm for 1 h at room temperature.
(2) Carbonate-bound fraction (F2): the residue from (1) was extracted by 1 M $CH_3COONa$ (45 mL) buffered to pH 5 with $CH_3COOH$, and was shaken at 250 rpm for 6 h at room temperature.
(3) Fe/Mn oxide-bound fraction (F3): the residue from (2) was extracted by 0.04 M $NH_2OH \cdot HCl$ (in 25% (v/v) $CH_3COOH$) at initial pH 2 (45 mL), and was shaken at 250 rpm for 5 h at $96 \pm 3$ °C.
(4) Organic matter/sulfide-bound fraction (F4): the residue from (3) was extracted by 0.02 M $HNO_3$ (10 mL) and 30% (v/v) $H_2O_2$ (10 mL) at initial pH 2, and was shaken at 250 rpm for 3 h at $85 \pm 2$ °C. Then 10 mL of 30% $H_2O_2$ was added into the same tube with continuous shaking for 2 h. Finally, 15 mL of 3.2 M $CH_3COONH_4$ in 20% $HNO_3$ was added into the tube, and then it was shaken at 250 rpm for 0.5 h at room temperature.
(5) Residual fraction (F5): the residue from (4) was digested with $HNO_3$, HF and HCl according to the microwave digestion method (USEPA, 1996).

The extractions were conducted in 50 mL centrifuge tubes to minimize loss of solid material. Between each extraction, separation was accomplished by centrifugation at 2000 rpm for 20 min. The supernatant was used for trace metal analysis, whereas the residue was washed by 8 mL of deionized water and then was centrifuged at 1000 rpm for 10 min. The supernatant from the washing step was mixed with the supernatant obtained from the beginning. The heavy metal concentration was determined by a flame AAS equipped with a graphite burner (Model Z-8100, Hitachi).

### 2.4. Quality assurance and quality control

During the analysis of total metal, a standard reference material (SRM 2704, National Institute of Standards and Technology) was applied to each batch of samples to ensure analytical quality. For metal fractionation, the differences between the total metal content estimated from the sum of five fractions and the total content obtained by acid digestion of the samples were all less than 10%. In this study, all data are presented as the mean values of two composting piles. During composting, all analyses were performed in triplicate for each composting pile. The standard deviations of the replications were below 10% of the mean values. The differences of mean values in different weeks were examined by ANOVA test.

## 3. Results and discussion

### 3.1. Composting of sewage sludge

Tables 1 and 2 show the physical–chemical characteristics and content of heavy metals in raw materials and the composted mixture. The quantity of heavy metals varies significantly between different sources of sewage sludge (Zufiaurre et al., 1998). Locally obtained (Min-Shen MSTP, Taiwan) sewage sludge used in this study had rather low concentrations of all heavy metals. These concentrations are far below upper permissible limits of standard criteria under the EU Sludge Directive (Cu: 1750 mg kg$^{-1}$, Ni: 400 mg kg$^{-1}$, Pb: 1200 mg kg$^{-1}$, Zn: 4000 mg kg$^{-1}$, and Cd: 40 mg kg$^{-1}$) (EC, 2001). As sewage sludge was mixed with bulking agent (sawdust), heavy metal concentrations in the raw composted mixture reduced significantly. In the composting process, the temperature is gradually increased due to heat released from exothermic reactions of organic matter decomposition (Hassouneh et al., 1999). A typical temperature variation during composting obtained from many operating composting plants (Veeken and Hamelers, 2002) was observed in this study (Fig. 1a). As revealed in Fig. 1a, the temperature increased to 74 °C in the first 7 days, and temperature stayed above 60 °C for the next 21 days. The temperature dropped to 40 °C on day 35, and then decreased to 30 °C at the end of the composting cycle on day 56. Fig. 1b indicates the variation of the moisture content with time. As can be seen, the moisture decreased with time. This was probably due to the increased temperature during the composting process. Generally, the moisture content is an important parameter in the composting process because of its significance for microorganism activity. During the composting process, the amounts of organic matter slightly decreased with the composting period (Fig. 1c). It was found that about 4% of organic matter decomposed in the 56 days of the composting cycle. However, the C/N ratio was found to decrease from 30.5 at the beginning to 19.4 at the end (Table 1). A decrease in the C/N ratio could be an index for determining composting maturity.

The changes of heavy metal concentrations in composted mixtures are shown in Fig. 2. As shown in the figure, a significant increase in the total heavy metal concentration was observed in the first 28 days of composting ($p < 0.05$, $\alpha = 0.05$), and then the heavy metal concentrations became more consistent. From the beginning to the end of composting, the total concentrations of Cu and Zn increased 20% and 28%, respectively, due to the decomposition of organic matter ($p < 0.05$, $\alpha = 0.05$). Similarly, heavy metal concentrations increased by 25–40%, 15–36%, 50–100%, and 170–180% (for Cu and Zn) in spent

Table 1
Characteristics of raw materials for composting

| Material | Moisture (%) | Organic matter (%) | C/N[b] | pH[b] |
|---|---|---|---|---|
| Sewage sludge | 84.7 ± 3.4[a] | 71.9 ± 1.7 | 7.4 | 7.7 |
| Sawdust | 2.3 ± 0.4 | 98.8 ± 0.9 | 420.0 | 5.6 |
| Recycled compost | 51.4 ± 0.9 | 82.2 ± 1.6 | 19.4 | 5.8 |
| Raw composted mixture | 65.4 ± 1.8 | 91.7 ± 1.0 | 30.5 | 7.2 |
| Final compost | 52.8 ± 1.1 | 88.3 ± 1.3 | 19.4 | 6.2 |

[a] Mean ± SD ($n = 7$).
[b] Data are taken from Tung et al. (2003).

Table 2
Total heavy metal contents in raw materials for composting

| | Cu | Mn | Ni | Pb | Zn | Cd | Cr |
|---|---|---|---|---|---|---|---|
| | (mg kg$^{-1}$, dry weight) | | | | | | |
| Sewage sludge | 175 ± 10[a] | 46 ± 5 | 25 ± 3 | 64 ± 2 | 907 ± 18 | <0.02[b] | <2[b] |
| Recycled compost | | 102 ± 6 | 23 ± 4 | 50 ± 7 | | | <2 |
| Sawdust | 88 ± 2 | 13 ± 3 | 8 ± 3 | 13 ± 3 | 509 ± 6 | <0.02 | <2 |
| | 5 ± 1 | | | | 50 ± 4 | <0.02 | |
| Raw composted mixture | 71 ± 1 | 76 ± 2 | 21 ± 5 | 47 ± 8 | 315 ± 10 | <0.02 | <2 |
| Final compost | 85 ± 1 | 78 ± 2 | 23 ± 3 | 39 ± 5 | 403 ± 24 | <0.02 | <2 |
| *Limit values for heavy metals in final compost* | | | | | | | |
| EU | 100 | – | 50 | 100 | 200 | 0.7 | 100 |
| Taiwan | 150 | – | 25 | 150 | 500 | 5 | 150 |

[a] Mean ± SD ($n = 6$).
[b] Method detection limit.

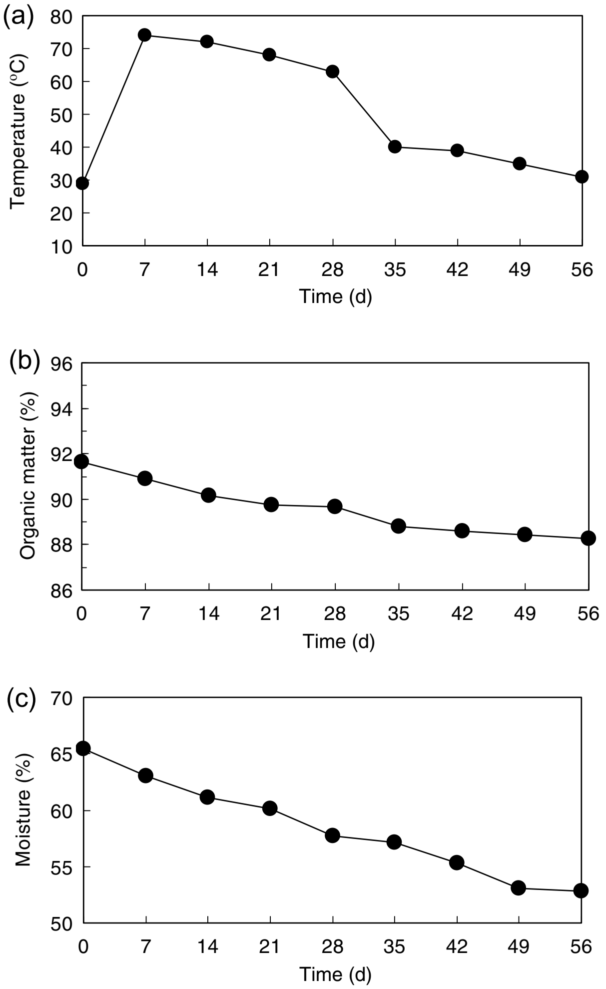

Fig. 1. Changes in characteristics of composted mixture during composting: (a) temperature, (b) organic matter, and (c) moisture.

pig litter, cattle manure, municipal solid waste, and swine manure, respectively (Hsu and Lo, 2001). As has been known, the composting types and the composting materials have significant effects on metal condensation. However, the heavy metal content affects the final quality of the compost. As shown in Table 2, the contents of all heavy metals were below the standard limits in Taiwan. In comparison to the EU's limitation, only the Zn content was higher than the standard limit.

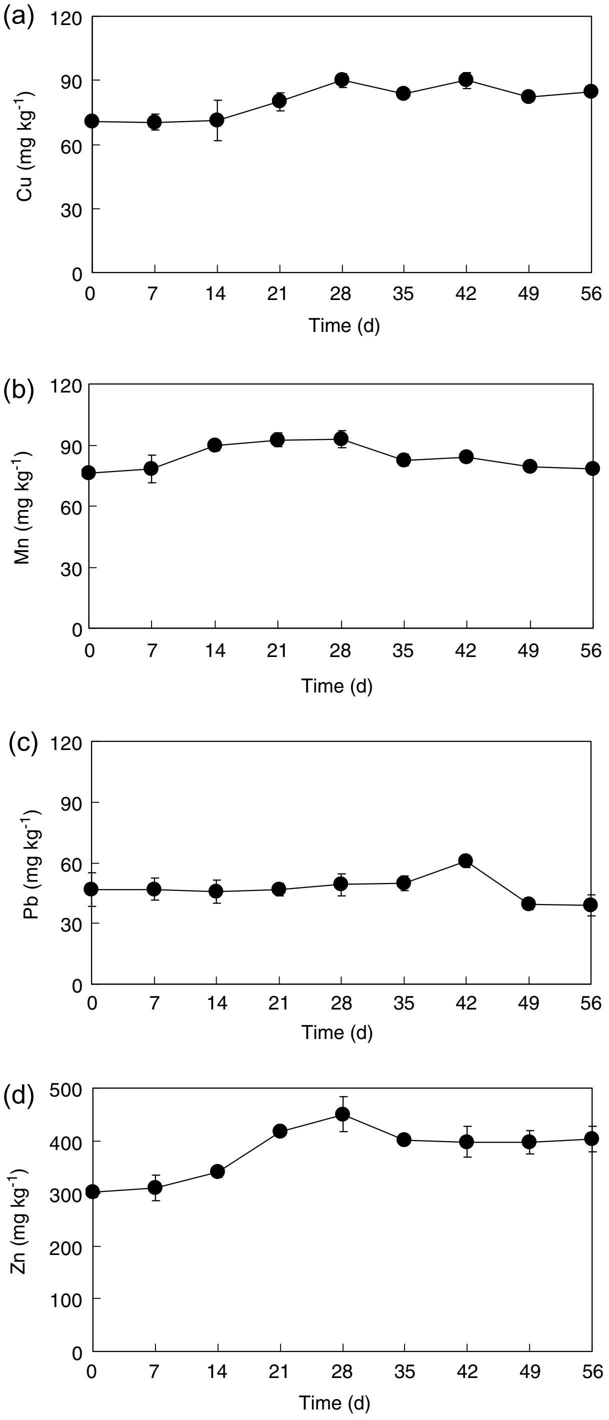

Fig. 2. Changes of total heavy metal contents during composting.

## 3.2. Changes of metal distribution during composting

The environmental impacts of particulate metal speciation are highly related to the behaviours of remobilization. Heavy metals in exchangeable (F1), carbonate-bound (F2), and Fe/Mn oxide-bound (F3) fractions are considered to be more mobile and bioavailable. The organic matter/sulfide-bound (F4), and residual (crystal lattice-held) (F5) metals are stable and non-bioavailable (Perin et al., 1997). The metal distribution in raw materials prior to composting is shown in Table 3. It was found that Zn mainly appeared in the F3 (51%) and F4 (31%) fractions in sewage sludge. Most of Cu (89%) existed in fraction F4. Pb (70%) in the sludge was found to be more prominent in the residual fraction (F5). The dominant speciation of Mn in the sludge was in the exchangeable (F1) (37%) fraction. Similar to sewage sludge, a high percentage of Zn was found in the F1 (36%), F3 (27%), and F4 (19%) fractions in the recycled compost. Cu in the F4 fraction (83%) represented the most significant speciation, compared to other fractions in the recycled compost. Mn was

Table 3
Original heavy metal distribution in raw materials for composting

| Fraction (mg kg$^{-1}$) | Sewage sludge | | | | Recycled compost | | | | Sawdust | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Cu | Mn | Pb | Zn | Cu | Mn | Pb | Zn | Cu | Mn | Pb | Zn |
| F1 | 0.9 ± 0.1[a] (1%)[b] | 16.2 ± 0.1 (37%) | 1.0 ± 0.1 (2%) | 32.5 ± 2.4 (4%) | 0.8 ± 0.1 (1%) | 66.7 ± 7.0 (67%) | 2.5 ± 0.7 (5%) | 163.4 ± 9.7 (36%) | 0.2 ± 0.1 (4%) | 6.9 ± 1.7 (58%) | 1.3 ± 0.2 (10%) | 14.9 ± 1.7 (33%) |
| F2 | 1.6 ± 0.5 (1%) | 7.1 ± 1.7 (16%) | 4.5 ± 0.8 (7%) | 87.7 ± 13.4 (10%) | 1.2 ± 0.1 (1%) | 8.6 ± 0.7 (9%) | 2.5 ± 0.2 (5%) | 51.4 ± 3.0 (11%) | 0.9 ± 0.1 (17%) | 1.9 ± 0.1 (16%) | 2.5 ± 0.2 (20%) | 7.6 ± 2.5 (17%) |
| F3 | 2.6 ± 0.2 (2%) | 6.4 ± 0.7 (14%) | 5.6 ± 0.3 (9%) | 453.2 ± 15.1 (51%) | 3.3 ± 0.2 (4%) | 11.8 ± 2.1 (12%) | 4.2 ± 0.7 (9%) | 124.6 ± 24.9 (27%) | 1.2 ± 0.2 (23%) | 1.1 ± 0.2 (10%) | 2.5 ± 0.1 (20%) | 12.5 ± 1.9 (28%) |
| F4 | 181.5 ± 11.3 (89%) | 5.9 ± 0.7 (13%) | 7.4 ± 1.2 (12%) | 275.2 ± 67.7 (31%) | 69.4 ± 3.8 (83%) | 3.7 ± 0.1 (4%) | 26.9 ± 2.0 (56%) | 85.7 ± 2.2 (19%) | 2.2 ± 0.9 (42%) | 0.2 ± 0.1 (1%) | 2.3 ± 0.2 (18%) | 3.8 ± 1.9 (9%) |
| F5 | 12.4 ± 0.1 (7%) | 8.8 ± 1.2 (20%) | 42.5 ± 1.8 (70%) | 37.1 ± 6.0 (4%) | 8.9 ± 0.1 (11%) | 8.4 ± 0.6 (8%) | 12.2 ± 1.4 (25%) | 38.8 ± 19.9 (7%) | 0.7 ± 0.2 (14%) | 1.8 ± 0.6 (15%) | 4.0 ± 1.7 (32%) | 6.0 ± 1.6 (13%) |

[a] Mean ± SD ($n = 6$).

[b] Percentage of total metal.

also mostly found in the F1 (67%) fraction and a high level of Pb (56%) was found in the F4 fraction. In sawdust, the metal distribution was similar to that in recycled compost although the metal content was very low.

Summarizing the above results, it was found that most (>60%) of Zn and Mn were associated with the mobile fractions (F1–F3) in sewage sludge, recycled compost, and sawdust. More than 93% of Cu was distributed in the stable fractions (F4 and F5) in both sewage sludge and recycled compost, though this number was only 56% in sawdust. Meanwhile, the distribution of Pb was similar to that of Cu. The percentage of Pb in the stable fractions was 82%, 81%, and 51% in sewage sludge, recycled compost, and sawdust, respectively. These results suggest that Zn and Mn were more mobile, and Cu and Pb were more immobile in raw materials. The findings of this study were in agreement with those of Lin et al. (1999) and Solis et al. (2002).

The variations of heavy metal distribution during the composting process are presented in Fig. 3. As shown in Fig. 3a, Cu prevailed in the organic matter/sulfide-bound fraction (F4) in the composted mixture (>82%). In general, the organometallic complexes follow the Irwing–William series: Cu > Pb > Zn > Cd > Fe (Coquery and Welbourn, 1995). Due to a high affinity for organic matter, Cu was not easily mobilized in the composting process. Cu ions are directly bound to two or more organic functional groups, mainly carboxylic, carbonyl, and phenolic groups, so that the ions are immobilized in a rigid inner-sphere complex (McBride, 1989). In addition, some parts of Cu were significantly transferred from mobile fractions (F1-F3) and F5 (residue fraction) to F4 during the composting process ($p < 0.005$, $\alpha = 0.05$). Because of acid production in the composting process (Hassouneh et al., 1999), there was a trend of decreased pH by the end of the composting process (Table 1). However, the decrease in pH accelerated the mobilization of heavy metals, especially those in mobile fractions (Yoo and James, 2002; Simpson et al., 2002). These released heavy metals were easily complexed with organic matter in the composted mixture. Therefore, Cu in fraction F4 increased from 82% to 97% by the end of the composting cycle. Similar results were also found in previous studies (Qiao and Ho, 1996; Hsu and Lo, 2001).

Due to the decrease of moisture and pH during the composting process, Mn in the F1 and F5 fractions decreased ($p < 0.01$, $\alpha = 0.05$), but increased significantly in the F2, F3, and F4 fractions ($p < 0.001$, $\alpha = 0.05$). Since Mn already existed in the mobile fractions (F1–F3), the total percentage of Mn in these fractions increased slightly (84–90%) in the composting process. Pb in the sewage sludge was found mainly in the residual fraction (F5), and it decreased from 32% to 17% ($p < 0.001$, $\alpha = 0.05$) in the residual fraction. Although the amount of Pb bound to the organic-sulfide fraction increased in the first 28 days ($p < 0.01$, $\alpha = 0.05$), the transformation of Pb to more mobile (bioavailable) fractions is clearly observed in the last composting period. Pb in mobile (F1–F3) fractions

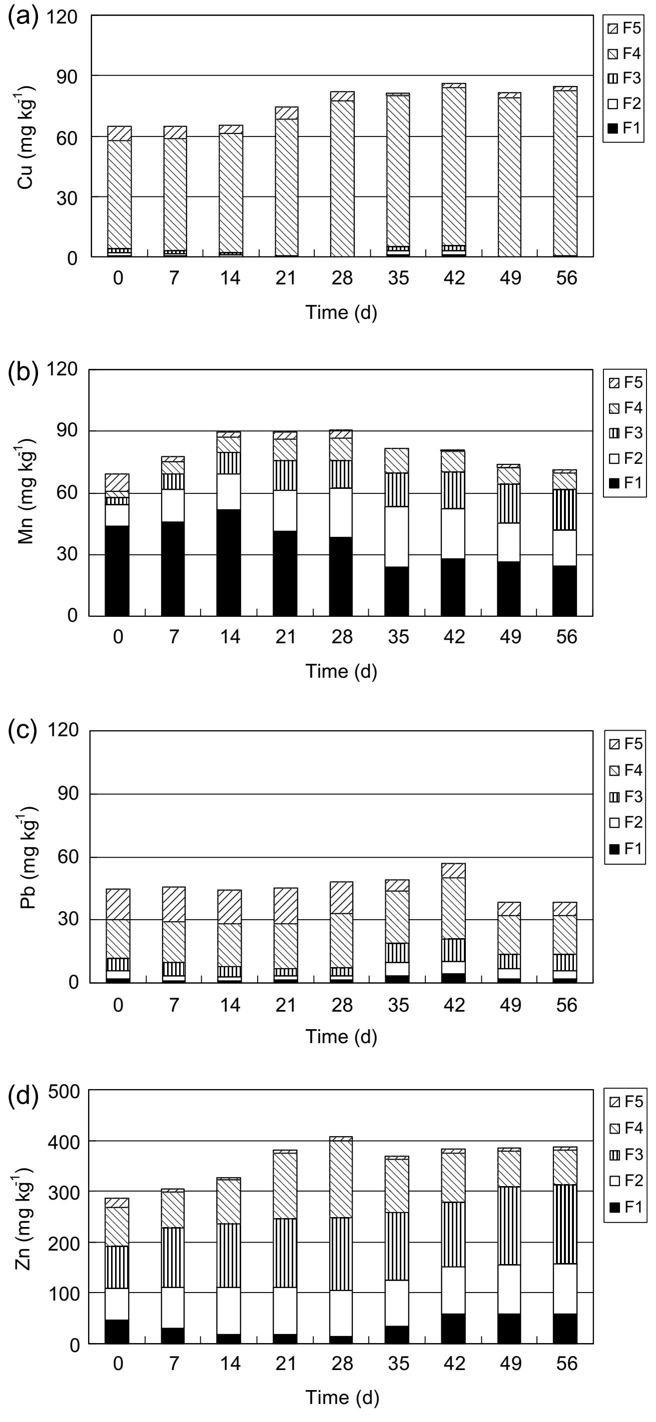

Fig. 3. Changes of heavy metal distribution during composting: (a) Cu, (b) Mn, (c) Pb, and (d) Zn.

increased from 26% to 36% after the composting process ($p < 0.001$, $\alpha = 0.05$). In contrast, Pb in the stable fractions (F4 and F5) significantly decreased in the composting process ($p < 0.001$, $\alpha = 0.05$). Similar results were also obtained by Bordas and Bourg (1998). As the composted mixture degraded and dried during the composting process, the iron oxyhydroxides formed. These hydrous oxides scavenge heavy metals from the composted mixture, owing to the charges on their surfaces (Stenphens et al., 2001). Meanwhile, the co-precipitation of heavy metals with iron oxides may also explain the increase of heavy metals (Mn, Pb, and Zn) appearing in the F3 fraction (Alloway, 1995).

Similarly, Zn significantly moved from stable fractions (F4 and F5) to mobile fractions (F1–F3) ($p < 0.001$, $\alpha = 0.05$). The percentage of Zn in mobile fractions increased from 67% to 81% after composting ($p < 0.005$, $\alpha = 0.05$). Due to the oxidation of organic matter and sulphide, as well as its high oxidation–reduction potential (Saeki et al., 1993), Zn was transferred from organically bound (F4) into more mobile fractions (F1–F3). The metal sulfides in the composted mixture were probably oxidized to metal sulphates. Therefore, metal bound with sulfides might have been released and then adsorbed on sulphate surfaces or precipitated as oxides (Stenphens et al., 2001).

### *3.3. Mobility of metal in compost*

Generally, metals bound in the first three fractions (F1–F3) were considered more mobile and bioavailable, and metals in fractions F4 and F5 were believed to be stable and non-bioavailable (Perin et al., 1997). Accordingly, the sum of heavy metal bound in the first three fractions (F1–F3) was therefore used to evaluate the potential mobility and bioavailability of metals in this study. Fig. 4 shows the percentages of heavy metals in mobile and stable fractions with respect to sewage sludge, raw composted mixture, and final compost. As revealed in Fig. 4, the percentages of heavy metals in the mobile fractions in the raw composted mixture were higher than those in sewage sludge. Apart from Cu, the percentages of Mn, Pb, and Zn in the mobile fractions increased obviously after composting. It was also found that Mn and Zn generally had greater potential mobility (bioavailability) in the sewage sludge and composted mixture. Nevertheless, Cu and Pb had higher proportions in the stable fractions. This indicates that these metals were not readily released from the sewage sludge and composted mixture. This agrees with the results reported by other authors (Obrador et al., 2001).

In this study, the ratio of the sum of metal bound in mobile fractions (F1–F3) to that in stable fractions (F4 and F5) was used to evaluate the potential mobility of heavy metals in sewage sludge and composted mixture. The mobility of heavy metals in sewage sludge and composted mixture is shown in Table 4. As is seen, the mobility of the investigated heavy metals in sewage sludge and composted mixture were different from each other. The more mobile heavy metals in sewage sludge and composted mixture were Zn and Mn, and their mobility increased remarkably after composting. The mobility of Pb and Cu in sewage sludge and composted mixture was relatively low (<1). The mobility of Pb, unlike that of Cu, also increased in composted mixture after composting. These results suggest that the heavy metal mobility (bioavailability) in sewage sludge and composted mixture ranked in the order of Mn > Zn > Pb > Cu.

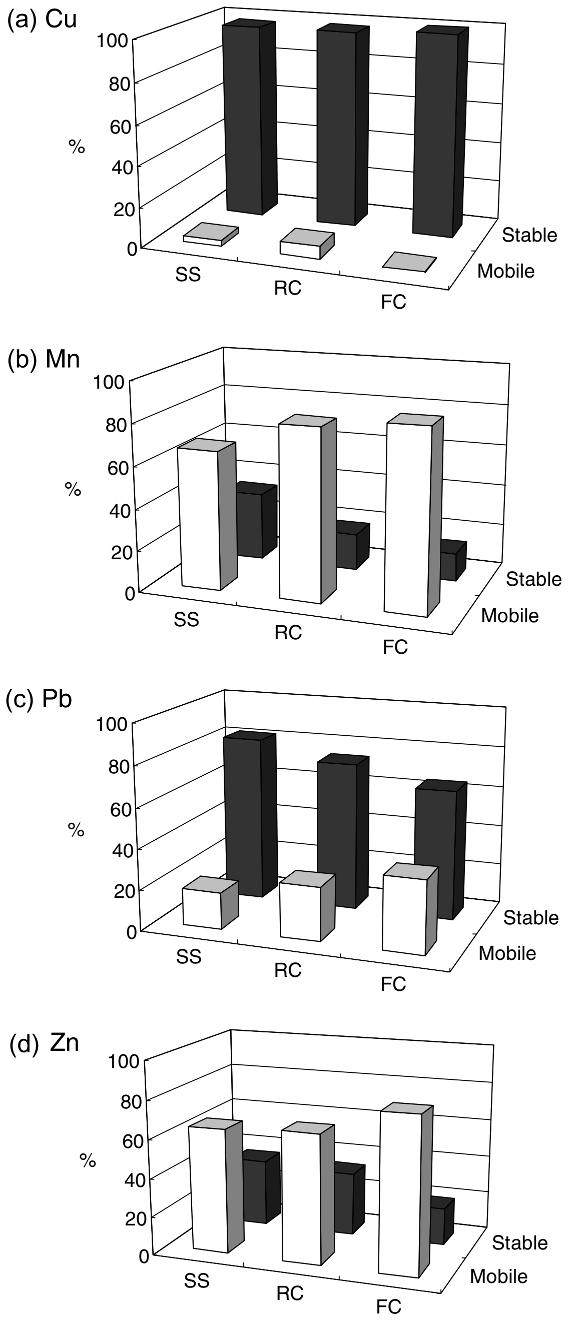

Fig. 4. Heavy metal amounts in mobile (F1–F3) and stable (F4 + F5) fractions in sewage sludge (SS), raw composted mixture (RC), and final compost (C).

Table 4
Mobility of heavy metals in sewage sludge and final compost

| Sample | Cu | Mn | Pb | Zn |
|---|---|---|---|---|
| Sewage sludge | 0.03 | 2.02 | 0.22 | 1.83 |
| Raw composted mixture | 0.07 | 5.29 | 0.36 | 2.03 |
| Final compost | <0.01 | 6.12 | 0.55 | 4.16 |

## 4. Conclusions

The application of composted municipal sewage sludge to agriculture land could be an alternative for solving the problem of the increase of excess sludge discharge. To investigate the mobility and bioavailability of heavy metals in the environment, a study of their kinetic behaviour in the composted mixture was performed using sequential extraction procedures. The heavy metal distribution in sewage sludge and composted mixture varied depending on the sources and properties of heavy metals. The increase of total Cu and Zn in the composted mixture was found after the composting process. Mn and Zn were mainly found in mobile fractions (F1–F3), whereas Cu and Pb had a greater affinity for stable fractions (F4 and F5) in sewage sludge and composted mixture. Overall, Mn, Pb, and Zn in the stable fractions were transformed into mobile fractions during the composting process, except for Cu. After composting, the potential mobility and bioavailability of Mn, Pb, and Zn increased significantly, but the opposite result was observed for Cu. Generally, Cu and Pb belonged to relatively stable heavy metals; however, Mn and Zn were more mobile in the composting process.

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