

## Transfer of heavy metals from compost to red soil and groundwater under simulated rainfall conditions

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### ABSTRACT

It is well known that compost can be polluted with heavy metals. A self-designed soil column system was used to clarify the leaching or residue of heavy metals when compost was added to red soil. Less than 4% of Cu and more than 58.3% of Zn were transferred to groundwater. Maximum concentrations of Cu and Zn in leachate were 22 and 246  $\mu\text{g/L}$ , respectively. pH in leachate decreased during 6.75–11.25 h, comparing with the control. Electrical conductivity was higher in the treatment of compost polluted by both Cu and Zn than that polluted only by Cu. Over 75.3% of Cu and 78.2% of Zn remained in the residual formation. Maximum bioavailability parameters in soil were 0.099 for Cu and 0.160 for Zn. Long-term field experiments with cycling application are needed to study the cycling effect on heavy metal residue in soil or leaching to the groundwater in the future.

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### 1. Introduction

Composting is an effective method for changing organic solid waste into fertilizer that is rich of nutrients. Many types of waste, such as straw, leaves, sludge, manure, faeces and so on, are suitable for composting. Because compost is biologically more compatible than chemical fertilizers for soils and plants, composting has become a preferred choice for treating organic solid waste.

However, the wide distribution of heavy metals in soil, water and atmosphere, make the raw materials for compost possible sources of heavy metal pollution [1–3]. The content of heavy metals in compost ranges from one part per million (ppm) to one part per thousand (ppth) [4,5]. Addition of heavy metals in feedstuff causes poultry manure to have high content of Cu or Zn. Excessive application of chemical fertilizers leads to the accumulation of heavy metals such as Cd, Pb or Cr in soils. In the Pearl River Delta of southern China, Pb concentrations in agricultural soils are 20% higher than those in natural soils [6]. The heavy metals including Cu, Zn, Pb, and Cd varied from 7.2 to 1270 mg/kg in the municipal sludge in some cities in the middle-south region of China [7].

Heavy metals can be immobilized or aggregated by microbial hypha during composting [8,9]. Their existing formations can be changed by materials such as lime, which are added to compost materials [10]. After their introduction into the soil, the mobility or bioavailability of heavy metals in compost can be altered by many factors arising simultaneously from soil, plants or rain [11–13].

Many factors can affect the movement or alter the accumulation of heavy metals in compost. There is limited mobility of heavy metals in calcareous soil or soils with high Fe–Al content [14,15]. After loadings of approximately 2  $\mu\text{g/g}$  Cu and 6  $\mu\text{g/g}$  Zn from compost to soils, heavy metals losses differed widely with total losses of about 60  $\mu\text{g/kg}$  Zn and 30  $\mu\text{g/kg}$  Cu in sand soil and about 30  $\mu\text{g/kg}$  Zn and 15  $\mu\text{g/kg}$  Cu in sandy-loam soils under steady hydrological conditions [16]. Ion exchange between heavy metals and soil particles may lead to a change of pH [17,18]. Dissolved organic matter in soil or compost is also an important factor that affects the mobility of heavy metals in compost [19,20]. The polarity change of dissolved organic matter may promote or inhibit the movement of heavy metals in soil.

To prevent the pollution of soil or groundwater by heavy metals from compost, many countries such as Belgium, Holland and Germany have established standards for heavy metals in compost. For example, in Holland, the maximum allowed concentrations of heavy metals of Zn, Cu, Pb and Cd in compost are 75, 25, 65 and 0.7 mg/kg, respectively, while in Germany, they are 400, 100, 150 and 1.5 mg/kg, respectively. However, in China, there are no heavy

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metal limits for compost application [21], mainly because of the lack of appropriate scientific research [11].

According to the statistics of Liu et al. [21], Cu contents in commercial compost are in the range of 0.0–998.6 mg/kg. 83.3% of samples are less than 100 mg/kg, with an average of 75.4 mg/kg. The refer of other limits such as Control Standard for Pollutants in Sludges from Agricultural Use (GB 4284-84) or Control Standards for Urban Wastes for Agricultural Use (GB8172-87) maybe pose a potential heavy metal risk to soil and groundwater.

A soil column experiment designed for investigating the transfer of heavy metals from compost to soil and groundwater under simulated rainfall conditions is described in this paper. Red soil was chosen because of its wide distribution in southern China and its low organic content, which requires supplements of organic fertilizer. Polluted compost was created by adding Cu or Zn before composting. Changes of heavy metals in water or the soil phase, pH, and electrical conductivity (EC) were measured to characterize the transfer characteristics of heavy metals from compost to soil or groundwater under simulated rainfall conditions.

## 2. Materials and methods

### 2.1. Soil sample

The low organic matter soil used in this experiment, sampled from an old orange orchard in the Hunan Agriculture Institute of China, is the typical agrotypic of the red soil widely distributed in southern China. Surface grass and leaves on soil were removed. A hole with about 0.8 m<sup>2</sup> of area and 1 m of depth was dugged out with a spade. The soil profile samples, consisted of a surface layer of 0–20 cm, a subsurface layer of 20–40 cm, and a deep layer of 40–60 cm, were collected, air dried and sieved with 2 mm plastic sieve. The soil is classified as Ferralsol in the Chinese soil classification system and as Ultisol in the American soil classification system. The basic characteristics of this soil are shown in Table 1.

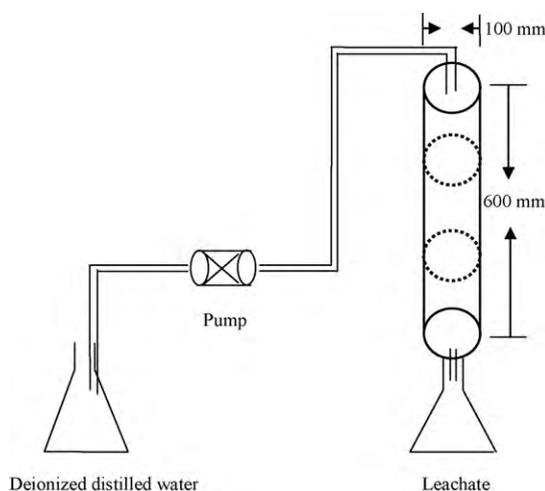
### 2.2. Compost

Five kinds of compost were prepared: four for adding with different content of heavy metals and one for the control. Leave residues (45%), pig manure (10%), rice milling product (20%) and fly ash (25%) were dried and grinded to smaller than 20 mm with a sample mill (Foss Tecator, Sweden).

For the lack of appropriate scientific research data, there are no heavy metal limits for compost application in China. Heavy metal concentrations in compost varied with the kinds of raw composting materials. Referring to the average content of 75.4 mg/kg Cu in commercial compost in China [21], the contents of Cu with 40 and 80 mg/kg were chosen to study the Cu leaching characteristics even in the presence of Zn. The heavy metal concentrations added to compost were 40 mg/kg Cu (Treatment 1), 80 mg/kg Cu (Treatment 2), 40 mg/kg Cu and 40 mg/kg Zn (Treatment 3), and 80 mg/kg Cu and 40 mg/kg Zn (Treatment 4). Heavy metals containing solutions were sprayed to the grinded mixture for composting. The solution containing heavy metals was prepared by chemical reagents of Cu(NO<sub>3</sub>)<sub>2</sub> and Zn(NO<sub>3</sub>)<sub>2</sub> of analytical grade. The control compost was water extracted with 7.15 of pH, 2427.18 mg/kg of

**Table 1**  
Basic soil characteristics.

Soil sample	Surface layer (0–20 cm)	Subsurface layer (20–40 cm)	Deep layer (40–60 cm)
pH	4.52	4.1	3.93
Organic matter (g/kg)	23.8	7.1	1.5



**Fig. 1.** Equipment of soil column experiment.

DOC, 1.97 ms/cm of EC, 0.36 mg/kg Cu and 2.51 mg/kg Zn. The total metal contents of heavy metals in compost were 3.42 mg/kg Cu and 14.37 mg/kg Zn.

### 2.3. The leaching experiment

The experiment that simulated rainfall conditions was performed in a column system produced by Haijie Manufacture Technology Co. Ltd. of Hunan University (Fig. 1). The column was made of polypropylene with an inner diameter of 100 mm and an inner height of 600 mm. To maintain a uniform fall of water on the soil column and to avoid drops of water, double plastic plates, each with 50 cavities, were interlaced with the top of the surface layer. A plastic plate with 50 cavities was also placed in the column bottom to facilitate the outflow of leachate. Double filter papers were placed on the bottom of plastic plate to avoid the outflow of soil particles from the soil column. Prior to the experiment, the equipment was washed with diluted nitric acid (3%, v/v) and distilled water with each of three times, respectively.

Three layers of soils, each of 1200 g (dry weight), were placed in the column with 20 cm per layer and were with similar density to natural situation. In the surface layer with an area of 78.5 mm<sup>2</sup>, the soil was mixed with 51 g of compost, which was equivalent to 65 Mg/ha under field conditions (Fig. 1). For the changes of complicated components with the location site in real rain, not the real rainfall but the DDI water or tag water was preferred to choose as the simulated rainfall in many studies [22–25]. Referring to the design of Kaschl et al. [22], deionized distilled water was used as the simulated rain in the leaching experiment, with pH 6.5 and 1.07 μs/cm of electric conductivity (EC). Total volume of the deionized distilled water pumping to the column was 1500 mL, equal to 200 mm of rainfall, and divided into five aliquots, with 300 mL each time.

The leachate from the soil column was to determine heavy metals concentration, pH, dissolved organic matter, and electrical conductivity. After leaching, each layer of soil was collected to analyze the residue of heavy metals using the method of Tessier et al. [26] and to evaluate the biological availability of heavy metals.

### 2.4. Analytical methods

Heavy metal concentration in the leachate from the soil column was determined by the method of atomic absorption spectrometry (AA700, PerkinElmer, USA).

To determine the content of heavy metals in soil, each layers of soil was air dried at room temperature and milled to sieve with 2 mm plastic mesh. The formation of heavy metals was analyzed according to the sequence extraction method of Tessier et al. [26]. The extraction steps were in the order of 1.0 mol/L  $MgCl_2$  for exchangeable formation, 1 mol/L NaOAc for formation bound to carbonates, 0.04 mol/L  $NH_2OH \cdot HCl$  for formation bound to iron and manganese oxides, 0.02 mol/L  $HNO_3$  and 10 mL 30%  $H_2O_2$  for formation bound to organic matters, and  $HNO_3-HClO_4$  for residual formation. The extracted solution was centrifuged, filtered with a 450  $\mu m$  filter paper, and kept in a volumetric flask under acidic condition. Final concentrations of heavy metals were measured with atomic absorption spectrometry method.

pH value in soil was determined at a soil and water ratio of 1:1 with a pH electrode (E-201-C, Shanghai Precision & Scientific Instrument Co. Ltd., China). The pH in the leachate was directly detected after filtering with filter membrane of 450  $\mu m$ . EC in leachate was measured with a conductivity meter (DDS-307, Precision & Scientific Instrument, Shanghai, China).

Fresh deionized distilled water was used throughout the experiments, which was produced with a hyperfiltration pure water system (Labconco, Water Pro Plus, USA). All chemical reagents were of analytical grade purchased from Shanghai Pharmaceutical Co. Ltd. in China. All data were processing with Sigma plot 10.0 or Microsoft Excel.

### 3. Results and discussion

#### 3.1. Heavy metal transfer from compost to groundwater

Fig. 2 shows the leaching of heavy metals under simulated rainfall conditions. The maximum concentration of Cu was 0.022 mg/L in leachate. Only a little amount of copper from the compost was transferred to the leachate, regardless of 40 mg/kg Cu, 80 mg/kg Cu, or even 40 mg/kg Zn in the compost. However, the situation was different for Zn. Though only a small amount of Zn was in the effluent from the soil column before 8 h, the Zn concentration in the leachate increased from 0.029 mg/kg to a peak of 0.175 mg/kg in the treatment with 40 mg/kg Cu and 40 mg/kg Zn. It decreased to a low concentration of 0.017 mg/kg when the duration of simulated rainfall exceeded 15 h. The higher Zn peak of 0.246 mg/kg in leachate was found in the treatment of compost with 80 mg/kg Cu and 40 mg/kg Zn.

It is possible that the spiking of copper ions to the organic matter in the compost formed a stable soil organic-Cu compound, and so little copper filtered out of the soil column. In compost-amended

sandy soil, the majority of water-soluble Cu from compost was accumulated in the soil rather than washed out with the leachate (less than 100  $\mu g/L$ ) [22]. Dissolved organic matter (DOM) in compost disrupts the binding intensity of soils to Cu [27]. Results of Pédrot et al. [28] show that three groups of elements can be distinguished based on their relationships with the colloidal phases: the first as the “truly” dissolved group (K, Na, Mg et al.), the second as an intermediate group (Cu, Cd, Co, and Ni), and the third group as the organic colloidal pool (Al, Cr, Pb, Fe et al.). For the intermediate group of Cu, the fulvic acid appears to be a major organic carrier phase. Red soil rich of Fe–Al oxide can strongly adsorb organic matter [20]. Cu in the compost may be transferred with the fulvic acids to the soil and adsorbed by abundant Fe–Al oxide. Therefore, there was little Cu in the effluent from the red-soil column.

As noted earlier, little Zn was leached from the soil column in the first 8 h of the experiment. Probably this was the time taken by Zn to move from the upper surface layer to the bottom of the soil column and by Zn bound compounds to functionalize with soil components. After 8 h, more Zn flowed out from the soil with the highest concentration of 246  $\mu g/L$  in the treatment with 80 mg/kg Cu and 40 mg/kg Zn. It appears that Zn in the compost escaped more easily from the soil column than Cu. Similar results were obtained by Gove et al. [16] that in sandy or sandy-loam soils, more Zn flowed out from soil column than Cu in the compost treatment.

The maximum concentrations of Cu and Zn in leachate were 22 and 246  $\mu g/L$ , respectively. However, it was only an indoor simulated experiment of applying compost to red soil. Long-term field experiments applying compost with multiple times are needed to study the cycling effect on heavy metal residue in soil or leaching to the groundwater.

#### 3.2. Changes in pH during filtration

Ion-exchange reactions commonly occur among heavy metals, cations, and protons in soils. The pH was an important factor that affected the heavy metal formation changes when heavy metals were transported from the soil column to the leachate. The changes of pH values in the leachate are shown in Fig. 3. For the red soil in South China is a kind of acid soil, the initial pH in leachate was low with 3.9–4.4. In the compost treatments in which heavy metal was added, pH was in the range of 3.34–4.21, which was lower than that in the control. The greatest decrease of pH was 0.3 in Cu-added compost treatments and 0.6 in Cu- and Zn-added compost treatments. Ion exchange must have occurred between Cu in the compost and the protons in the inorganic–organic compounds of

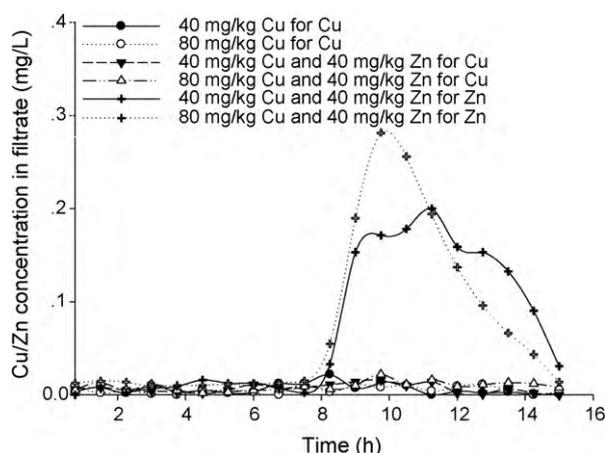


Fig. 2. Heavy metals in leachate under simulated rainfall condition.

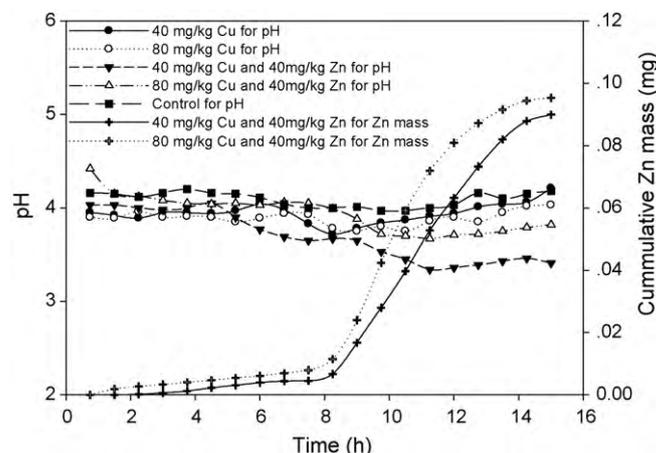


Fig. 3. Relationships of pH and Zn leaching in filtrate.

**Table 2**  
Heavy metal residues in soil column.

Heavy metal	Cu				Zn	
	1	2	3	4	3	4
Theoretical metal added from compost to soil column (mg/kg)	0.0667	0.1333	0.0667	0.1333	0.0667	0.0667
Metal measured in water phase (mg)	0.0087	0.079	0.0101	0.0136	0.0899	0.0953
Metal residues in soil column (mg/kg, by subtraction)	0.0643	0.1311	0.0639	0.1296	0.0417	0.0402

**Table 3**  
Bioavailabilities of heavy metals in soil.

Heavy metal	Cu				Zn	
	1	2	3	4	3	4
$M_{\text{exchangeable formation}} + M_{\text{binding to carbonate}}$ (mg/kg)	0.00598	0.00573	0.00600	0.00398	0.01055	0.00638
Bioavailability parameter ( $k$ )	0.093	0.048	0.099	0.096	0.086	0.160

the red soil, and then the outflow of  $H^+$  from the soil column led to the decrease of acidity in the leachate.

Though a slight change in the  $H^+$  concentration was detected in the first 6.75 h, there was a clear decrease of pH from 3.94 to 3.41 in the treatment with 80 mg/kg Cu, from 3.69 to 3.34 in the treatment with 40 mg/kg Cu and 40 mg/kg Zn, and from 3.76 to 3.37 in the treatment with 80 mg/kg Cu and 40 mg/kg Zn from 6.75 to 11.25 h. Lower pH was observed (except in the treatment with 80 mg/kg Cu) from 11.25 h to 15 h, the last period of the simulated rainfall.

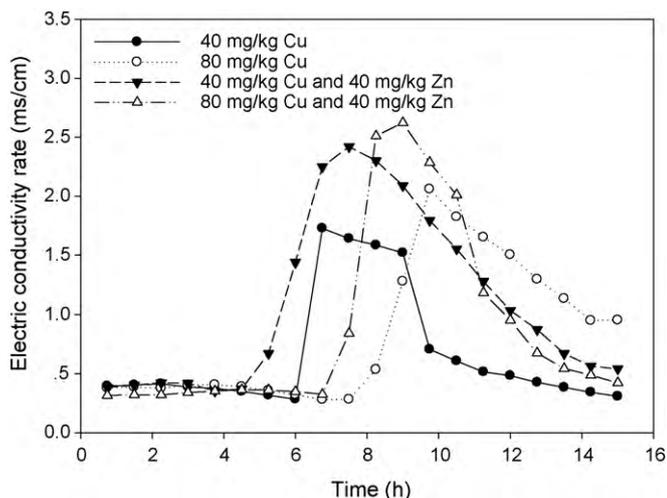
The pH decrease was highly related to Zn effluent in the leachate (Fig. 3). The cumulative Zn mass showed a large increase from 7.5 h to the end of the experiment, when pH decreased linearly. The pH changes were directly related to the ion exchange in the soil column between protons in soil particles and other cations such as heavy metal ions in soils. If the residue of heavy metals in the soil was estimated by subtraction of metal measured in water phase from theoretical metal added to compost, it was found that much Cu and Zn existed in soils assuming that the compost added was a part of soil after mixed (Table 2). Maybe it was the movement of soluble ions such as  $K^+$ ,  $Ca^{2+}$ ,  $H^+$  and so on (not measured). Low molecular organic acids might be another matter participating in the ion exchange. The replaced low molecular organic acids were moving with the leachate, which presented lower acid in higher heavy metal treatments. Moreover, these metals are known as weak Lewis acids, which also led to a decrease of pH in solution.

### 3.3. Change of electrical conductivity rate in leachate

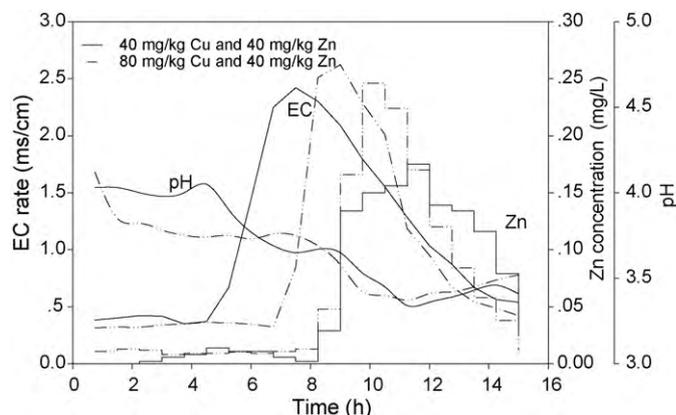
The EC indicates the content of salts in solution and is often used as a measure of ions changes in complicated system such as soil, compost, etc [29–31]. Salinity changes related to ion exchange can be estimated by the measurement of EC in the leachate during the simulated rainfall. Fig. 4 shows the change of EC in the leachate during the course of the experiment.

There was little change in EC in the first 5 h. It increased to a peak during 6.75–9.75 h and then decreased to the final value. The results indicate that there was hardly any salinity increase in the first 5 h, and the maximum salinity occurred coincident with the EC peak. EC was higher in the treatments of compost polluted with Cu and Zn than in the treatment polluted only with Cu or in the treatment with the higher heavy metal concentrations. Though nearly no Cu was present in the effluent from the soil column with the treatments of compost polluted with 40 mg/kg and 80 mg/kg Cu, other processes involving ion exchange must have occurred and led to the increase in EC. Maybe it was the movement of soluble ions such as  $K^+$ ,  $Ca^{2+}$ ,  $H^+$ , and so on. Similar results were observed by Kaschl et al. [22]. A slightly higher EC was observed in treatments with higher heavy metal concentrations during the leaching of municipal solid waste compost in calcareous soils.

Even though the ions were not measured, the breakthrough of the EC rate highlighted heavy metal outflow and  $H^+$  increased in the leachate (Fig. 5). EC peaks were found at 7.5 h or 9.75 h in treatments with 40 mg/kg Zn. The EC peaks appeared earlier than the maximum leaching of heavy metals and the lowest pH, which indicated that the main components of the leachate at the occurrence of EC peaks were not primarily induced by the ingredient change of Zn in the effluent, but by the ion exchange between Zn residing



**Fig. 4.** Changes of electric conductivity rates in leachate.



**Fig. 5.** Relationships of EC rates with pH and Zn concentrations in leachate.

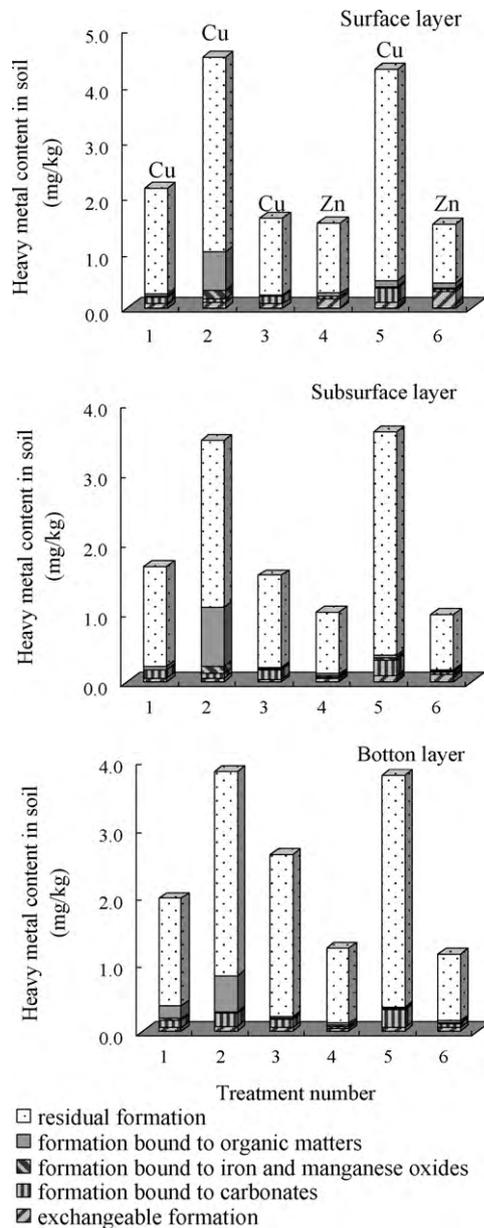


Fig. 6. Formation of heavy metal residues in soil profile column.

in the soil and other cations in the soil column. When the competition of heavy metals from compost with soil cations was finished, the leaching of heavy metals then occurred during the simulated rainfall experiments.

### 3.4. Residues of heavy metals in soil

The simulated rainfall in the experiment caused very little Cu or partial Zn in the compost to be transported from the soil column to the leachate (Fig. 2). The residues of Cu or Zn in soil or water phase are shown in Table 2. Heavy metals contents of “theoretical metal added from compost to soil column (mg/kg)” and “metal residues in soil column (mg/kg, by subtraction)” in Table 2 are calculated based on the total 3 layers of soil mass in column. Over 96% of Cu remained in the soil phase. Copper in the soil increased with the addition of Cu in the compost. 62.5% of Zn remained in the soil with the treatment of 40 mg/kg Cu and 40 mg/kg Zn, and 58.3% in the treatment with 80 mg/kg Cu and 40 mg/kg Zn.

To clarify the formation of heavy metals in soil, heavy metals in the soil profile were detected by the method of Tessier et al. [26]. The components of exchangeable formation, formation bound to carbonates, formation bound to iron and manganese oxides, formation bound to organic matters and residual formation are shown in Fig. 6. Over 75.3% of Cu and 78.2% of Zn was in the residual formation in the soil column. Lower than 17.6% of heavy metals existed in other formations. Average ratios of exchangeable formations bound to carbonates, iron or manganese oxides, and organic matters were 4.9%, 4.8%, 1.1%, and 5.2% in soil columns, respectively.

Residual formation of heavy metals is the left section after other four fractions have been removed by the method of Tessier et al. [26], which is the trace metals existed mainly within the crystal structure of primary and secondary minerals. This formation of metals is commonly not expected to be released in solution over a reasonable time span under the conditions normally encountered in nature. In each layer of the soil column, the exchangeable formation, bound to carbonates or iron and manganese oxides were lower than 0.264 mg/kg. The results indicate that the heavy metals Cu and Zn in the compost were mostly changed to the immobile form in the soil profile under the simulated rainfall conditions. There is significant difference between residue formation and any other formations among the three layers ( $p < 0.01$ ), while there is not significant difference between any other four formations except the residue formation ( $p > 0.05$ ).

The classification method of Tessier et al. [26] is used to estimate the bioavailabilities of heavy metals in soils. Heavy metals of exchangeable formation and formation bound to carbonates are considered adsorbing on sediments (or on their major constituents) or associating with sediment carbonates. These fractions are classified as the components that could be obtained or even have poisonous effects on microbes or plants. Based on the exchangeable formation and formation bound to carbonates, the bioavailability of heavy metals could be expressed by the parameter  $k$  [32]:

$$k = \frac{C_{\text{exchangeable formation}} + C_{\text{bounding to carbonates}}}{\text{total}} \quad (1)$$

The bioavailabilities of heavy metals in the soil profile after the application of compost are shown in Table 3. The bioavailabilities of Cu and Zn were very low with the maximum of 0.16 for Zn in the treatment of 80 mg/kg Cu and 40 mg/kg Zn. There was little difference in the bioavailability between the treatment with 40 mg/kg Cu and 40 mg/kg Zn, treatment with 40 mg/kg Cu, and treatment with 80 mg/kg Cu and 40 mg/kg Zn. The bioavailability of Zn was higher than that of Cu in the treatment, which was polluted with both Cu and Zn.

## 4. Conclusions

Very little Cu and over a half of Zn were leaching out of the soil column when heavy metal contaminated compost was applied to red soil. That obvious pH decreased and EC peak appeared before maximum leaching of heavy metals implied ion-exchange reactions might occur during leaching period. Low molecular organic acids might be another matter participating in the ion exchange, which should be confirmed in the next experiments. After leaching, about three quarters of Cu and Zn were in the presence of the residual formation in the soil. That the bioavailability of Zn was higher than that of Cu in the soil implied lower phytotoxicity of Cu than Zn after leaching from compost. These results should be confirmed under field conditions with long-term cycling experiments in future studies.

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