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# Release behavior of copper and zinc from sandy soils

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**Abstract:** The concentrations and chemical forms of copper(Cu) and zinc(Zn) in surface soils directly influence the movement of Cu and Zn. In this study, thirteen sandy soil samples with a wide range of total Cu and Zn concentrations were collected for evaluating the relationships between Cu and Zn release and extraction time, ratio of soil to water, pH and electrolyte types. The results indicated that Cu released in batch extraction that represents long-term leaching was mainly from exchangeable, and carbonate bound Cu fractions, and Zn released in the batch extraction was mainly from its carbonate bound fraction. However, the Cu and Zn leached from the soils using the column leaching that represents short-term leaching were mainly from their exchangeable fractions. Soil column leaching at different pH values indicated that the amounts of leached Zn and Cu were greatly affected by pH. The Cu and Zn release experiments with varying extraction times and ratio of soil to water suggest that long-term water-logging in the soils after rain may increase contact time of the soils with water and the release of Cu and Zn to water from the soils, and total amounts of Cu or Zn released from the soils increase, but the Cu or Zn concentration in the surface runoff decrease with increasing rainfall intensity. The increased Ca concentration in soil solution increased stability of organic matter-mineral complexes and might decrease the dissolution of organic matter, and thus decreased the release of Cu-binding component of organic matter. However, high concentration of Na in the soil solution increased the dispersion of the organic matter-mineral complexes and increased dissolution of organic matter and the release of Cu from the soils.

**Keywords:** release behavior; copper; zinc; sandy soil

## Introduction

Increased anthropogenic inputs of heavy metals in agricultural soils from the use of municipal waste, organic manure, fungicides, and pesticides have caused considerable concern relative to their impact on water contamination (Alloway, 1995; Moore, 1998). Soils contaminated with Cu and Zn can contribute to the enrichment of Cu and Zn in surface runoff and reduce quality of surface waters (Moore, 1998). Most soils in Florida under citrus production are sandy soils. Repeated applications of Cu- and Zn- containing agricultural chemicals have contributed to Cu and Zn accumulation in the soils (Zhu, 1993). Copper and Zn have moderate mobility under slightly acid soil conditions (Elliott, 1986; Jorgensen, 1991; Hesterberg, 1993). The accumulation of Cu and Zn in the agricultural soils can increase the possibility of Cu and Zn transport from soil to water. Previous studies indicated that Cu and Zn concentrations and forms in surface soil directly influenced the movement of Cu and Zn, especially in sandy soils (Scokart, 1983; Edwards, 1997; Moore, 1998; Cezary, 2001). Dissolved Cu and Zn concentrations in the surface runoff were significantly correlated with soil Cu and Zn levels extracted with 0.01 mol/L  $\text{CaCl}_2$ , but the temporal variations for Cu and Zn concentrations of the surface runoff in a specific site were great (Zhang, 2003a). Concentrations of heavy metals in soil solution are primarily controlled by sorption-desorption, and dissolution-precipitation reactions at the particle-water interface. Although numerous studies have been conducted to understand adsorption of Cu or Zn to soil minerals (Cavallaro, 1984; Atanassova, 1999), minimal information is available on release behavior of soil Cu and Zn in water from soils and factors which potentially influence Cu and Zn release. The objectives of this study were to understand Cu and Zn release behavior from Florida sandy spodosols under different conditions, including pH,

background electrolyte and extraction time.

## 1 Materials and methods

### 1.1 Soil analysis

Thirteen surface soil samples (0—5 cm) (S1—S13) were collected from St. Lucie County, Florida. Among them, ten samples were collected from different commercial citrus groves and three were collected from adjacent non-cultivated forestland with a native population of sand pine (*Pinus clausa* vasey ex). All of the soils are Wabasso sand (sandy, siliceous, hyperthermic Alfic Alaquods). Soil samples were air-dried and a sub-sample from each sample was passed through a 2 mm sieve for analysis. The remaining soil sample was used for column leaching and batch extraction studies. Selected properties of the soils are presented in Table 1. All soils are sandy soils, and differ widely in their total Cu and Zn. Higher soil pH in citrus groves than forestland was due to lime application in the citrus production, lower soil organic matter in citrus groves was resulted from mineralization of organic matter induced by frequent cultivation. Soil pH was measured in water at a ratio of soil : water (1 : 1) using a pH/ion/conductivity meter (Accumet Model 50, Fisher Scientific, Norcross, GA). Particle composition of soil sample was determined using the micro-pipette method (Miller, 1987). Total carbon (C) in the samples was determined using a CN-Analyzer (Vario MAX CN Macro Elemental Analyzer, Elemental Analysensystem GmbH, Hanau, Germany).

The total Cu and Zn in the soils were fractionated into five fractions (exchangeable, carbonate bound, organically bound, oxide bound, and residual fractions) by a modified procedure of Amacher (Amacher, 1996). Two grams of soil were sequentially extracted with 0.1 mol/L  $\text{Mg}(\text{NO}_3)_2$ , 1 mol/L NaOAc, 0.1 mol/L  $\text{Na}_4\text{P}_2\text{O}_7$  and 0.2 mol/L ammonium oxalate + 0.2 mol/L oxalic acid + 0.1 mol/L ascorbic acid (pH 3) for separation of exchangeable,

carbonate bound, organically bound, and oxides bound fractions. After each extraction, the suspension was centrifuged at  $7500 \times g$  (rcf, relative centrifuge force) for 30 min and then the supernatant was passed through a Whatman # 42 filter paper. Residual Cu and Zn contents in the soils

were determined by digesting residual soil with nitric acid and perchloric acid (Reed, 1996). Copper and Zn concentrations in the extracts and digested solutions were determined using an inductively coupled plasma atomic emission spectrometry (ICP-AES, Ultima, JY Horiba Inc. Edison, N.J.).

Table 1 Properties of the tested soils

Soils	Land use	pH	Total C, g/kg	Sand, g/kg	Silt, g/kg	Clay, g/kg	Total Cu, mg/kg	Total Zn, mg/kg
S1	Forest	4.22	13.4	921	31	48	1.7	2.1
S2	Forest	4.57	15.6	917	24	59	1.6	3.2
S3	Forest	4.13	11.8	894	56	50	3.6	4.0
S4	Citrus	7.03	12.5	932	28	40	400.1	100.6
S5	Citrus	7.07	10.6	901	42	57	266.5	68.1
S6	Citrus	7.20	9.7	924	37	39	242.4	64.0
S7	Citrus	6.76	11.7	911	40	49	239.2	40.2
S8	Citrus	5.54	14.6	876	78	46	67.7	24.4
S9	Citrus	6.76	8.7	921	47	32	97.2	40.3
S10	Citrus	7.48	7.9	907	41	56	286.1	67.8
S11	Citrus	7.60	10.2	911	47	42	281.9	32.4
S12	Citrus	7.27	11.3	921	43	36	306.2	70.1
S13	Citrus	7.46	8.7	941	28	31	352.6	43.3

## 1.2 Batch extraction

Extraction of soluble Cu and Zn in the soils was conducted in various water: soil ratios, electrolytes, and extracting times. Time-dependent changes of Cu or Zn released from the soils were conducted with eight extraction time (0.25, 0.5, 1, 4, 8, 24, 48, and 96 h) at a water: soil ratio of 5:1. For understanding the effects of water: soil ratio on the Cu and Zn release, soil samples were equilibrated with deionized water in various water: soil ratios (1, 2, 5, 10, and 20) for 24 h. In addition, four dilute electrolytes (0.02 mol/L  $\text{NH}_4\text{NO}_3$ , 0.02 mol/L  $\text{NaNO}_3$ , 0.02 mol/L  $\text{KNO}_3$ , and 0.01 mol/L  $\text{Ca}(\text{NO}_3)_2$ ) and deionized water were used as extractants to extract Cu and Zn at solution: soil ratio of 5:1 with extraction time of 24 h for understanding effects of background electrolytes on Cu and Zn release. Each extraction was performed on an end-to-end shaker ( $180 \text{ cycles min}^{-1}$ ). After each extraction, the supernatant was separated by centrifuging at  $7500 \times g$  (rcf) for 30 min and filtering through a  $0.45 \mu\text{m}$  membrane filter. The Cu and Zn concentrations in the filtrates were determined using the ICP-AES.

## 1.3 Column leaching

Soil Cu and Zn leachability was determined using soil columns. Each column was prepared in the laboratory using a plexiglass leaching column (10 cm long and 7.5 cm inner diameter). The bottom of the column was consisted of a plexiglass plate containing several 5 mm wide holes. The top of the plate was covered with a nylon cloth and glued to the bottom of the column. Each 300 g soil sample was packed to form a column 5 cm high. Two disks of filter paper (Whatman # 2) were placed on the top of soil prior to leachings to prevent disturbance by applied water. Prior to setting up the leaching experiment, the soil columns were slowly saturated from the bottom upward with deionized water to remove air pockets. The columns were then set up in stands for two days at room temperature. About 110 ml (1 pore volume) of deionized water (pH 7) was applied to each soil column on a daily basis and repeated for 10 d at a rate of 2 ml/min. This application rate did not allow any ponding on the top of the

column. Leachates were collected in 1000 ml beakers below the soil columns, filtered through a  $0.45 \mu\text{m}$  membrane filter for analysis. Copper and Zn concentrations in the filtrate were determined using the ICP-AES. In order to understand the effects of pH on Cu and Zn leaching, three soils (S1, S4, and S11) were leached using different pH leachants (deionized water adjusted to pH 3, 4, 5, 7, and 9 with dilute  $\text{HNO}_3$  and  $\text{NaOH}$  solution). All the above measurement, including the studies of fractionation of Cu and Zn, batch extraction and column leaching, were run on triplicate samples.

## 1.4 Statistical analysis

Least squares multiple regression analysis was used to assess relationships of Cu and Zn amounts released from the soils and their fractions. Data analyses were conducted using SAS program procedures (SAS Institute, 1998).

## 2 Results and discussion

### 2.1 Fractions of copper and zinc in the soils

Total Cu and Zn in the soils ranged from 1.6 to 400.1 mg/kg and from 2.1 to 100.6 mg/kg, respectively (Table 1). The Cu and Zn concentrations in the citrus soils (67.7—400.1 mg/kg for Cu and 24.4—100.6 mg/kg for Zn) were higher than those in the forest soils (1.6—3.6 mg/kg for Cu and 2.1—4.0 mg/kg for Zn), indicating that Cu and Zn were significantly accumulated in the citrus soils. The Cu and Zn accumulation in the citrus soils was attributed to repeated applications of Cu- and Zn-containing agricultural chemicals, such as organic manure, chemical fertilizers, fungicides, and pesticides (Zhu, 1993; Zhang, 2003b). The fractions of Cu and Zn varied among the soils (Fig. 1). For the forest soils, Cu was mainly present in organically and oxides bound fractions, no carbonate bound Cu fraction was detected in the soils. Mean proportions of total Cu as various fractions decreased in the order of organically bound (60.1%) > oxides bound (21.8%) > residual (15.5%) > exchangeable (2.7%). Zinc was primarily presented in residual and exchangeable fractions in the forest soils (Fig. 1), the proportion of total Zn as exchangeable fraction was

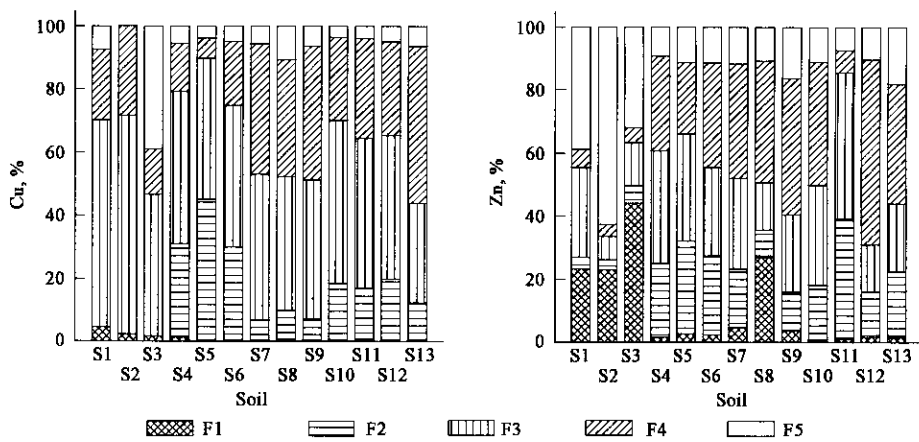


Fig.1 Fractionations of Cu and Zn in the soils  
F1: exchangeable; F2: CaCO<sub>3</sub>-bound; F3: organic matter bound; F4: oxides-bound; F5: residual

23.2%—44.13%. Mean percentages of total Zn as various fractions decreased in the order of residual (44.4%) > exchangeable (30.1%) > organically bound (16.6%) > oxides bound (4.8%) > carbonate bound (4.1%).

Among the citrus soils used in this study, the proportion of total Cu present as exchangeable fraction ranged from 0.29%—1.2%. The organically bound fraction was the dominant fraction of Cu, which varied from 31.6%—51.7% of total Cu. Only 3.5%—10.6% of total Cu was present in the residual fraction. The mean percentages of the total Cu as various Cu fractions in the citrus soils were organically bound (44.8%) > oxides bound (30.0%) > carbonate bound (19.1%) > residual (5.5%) > exchangeable (0.56%). Distribution of Zn in the citrus soils among the various fractions was different from that of Cu. The proportion of the total Zn as exchangeable Zn fraction was much greater than that of Cu, while percentage of the total Zn as organically bound Zn fraction was lower than that of Cu. The mean proportions of the total Zn as various Zn fractions in the citrus soils were oxides bound (34.7%) > organically bound (28.1%) > carbonate bound (20.9%) > residual (11.7%) > exchange (4.7%).

Correlation analysis using the data from 13 soils indicated that concentrations of all five Cu fractions were significantly correlated with their total Cu ( $r = 0.75$ — $0.97$ ,  $p < 0.01$ ). The concentrations of carbonate bound, organically bound, oxides bound and residual Zn fractions were significantly correlated with their total Zn ( $r = 0.87$ — $0.92$ ,  $p < 0.01$ ), but there was no significant correlation between exchangeable Zn and total Zn ( $r = -0.07$ ). pH had significant effects on the percentages of exchangeable and carbonate bound Cu and Zn fractions, and was negatively correlated with exchangeable Cu and Zn ( $r = -0.76$  and  $-0.93$ , respectively,  $p < 0.01$ ) and positively correlated with carbonate bound Cu and Zn fractions ( $r = 0.66$  and  $0.81$ , respectively,  $p < 0.01$ ).

2.2 Changes of Cu and Zn released from soils with extraction time

Copper and Zn released from the soils increased with extraction time and varied among the soils (Fig. 2). The amounts of Cu and Zn released from the forest soils (S1—S3) using the shaking extraction were very low and increased slightly with the extraction times. However, the amounts of

Cu and Zn released from the citrus soils (S4—S13) were much higher than those from the forest soils. The soil with highest amounts of Cu and Zn released was S4 that had the highest levels of Cu and Zn accumulation (Table 1). Multiple regression analysis showed that the three fractions correlated to the Cu amounts released from the soils in 96 h ( $Y$ ) were exchangeable ( $X_1$ ), carbonate bound Cu ( $X_2$ ), and organically bound Cu ( $X_3$ ). The relationship could be described by a linear model:  $Y = 0.49 + 1.04 X_1 + 0.026 X_2 + 0.0011 X_3$  ( $R^2 = 0.94^{**}$ ), in which exchangeable Cu and carbonate bound Cu accounted for 84% and 9%, respectively, of the total variance in the released Cu amount (Table 2). The associations between the released Cu and the fractions suggest that Cu released in batch extraction is mainly from exchangeable and carbonate bound Cu fractions.

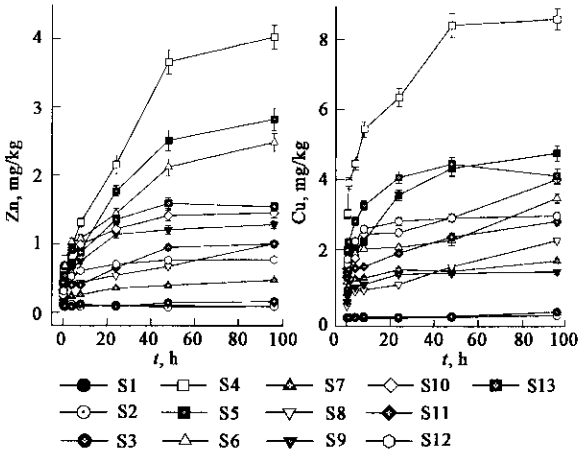


Fig. 2 Changes of Cu and Zn released from the soils with shaking time

The amount of Zn released from the soils ( $Y$ ) was related with exchangeable Zn ( $Z_1$ ), carbonate bound Zn ( $Z_2$ ), organically bound Zn ( $Z_3$ ), the linear relationship between the Zn released from the soils and the Zn fractions was:  $Y = -0.23 + 0.14 Z_1 + 0.089 Z_2 + 0.042 Z_3$  ( $R^2 = 0.88$ ). The carbonate bound Zn accounted for 83% of the total variance in the released Zn. However, simple correlation analysis showed that no significant correlation was found between the amounts of Zn from the soils and

exchangeable Zn. The results suggest that carbonate bound Zn has major contribution to Zn released from the soils using the shaking extraction. During the extraction, carbonate bound Zn could be transformed into soluble form and partially released.

**Table 2** Regression models for Cu and Zn released from the soils as related to various soil Cu or Zn fractions estimated by the sequential extraction procedure (*n* = 13)

Y	Variables <sup>†</sup>	Coefficient	Partial R <sup>2</sup>	R <sup>2</sup>
Cu released by shaking extraction in 96 h	Constant	0.49		0.94
	X <sub>1</sub>	1.04	0.84	<i>p</i> < 0.0001
	X <sub>2</sub>	0.026	0.09	
	X <sub>3</sub>	0.0011	0.01	
Zn released by shaking extraction in 96 h	Constant	- 0.23		0.88
	Z <sub>1</sub>	0.14	0.04	<i>p</i> < 0.0001
	Z <sub>2</sub>	0.089	0.83	
	Z <sub>3</sub>	0.042	0.01	
Cu leached in ten leachings	Constant	0.079		0.96
	X <sub>1</sub>	0.35	0.91	<i>p</i> < 0.0001
	X <sub>3</sub>	0.0013	0.04	
Zn leached in ten leachings	Constant	- 0.16		0.97
	Z <sub>1</sub>	0.68	0.97	<i>p</i> < 0.0001

Notes: <sup>†</sup> X<sub>1</sub>: exchangeable Cu; X<sub>2</sub>: carbonate bound Cu; X<sub>3</sub>: organically bound Cu; Z<sub>1</sub>: exchangeable Zn; Z<sub>2</sub>: carbonate bound Zn; Z<sub>3</sub>: organically bound Zn

For most of the soils (particularly the soils with low Cu and Zn release capacity), the extraction time to reach extraction equilibrium was about 24—48 h. The results may suggest that long-term waterlogging in the soils after rain may increase contacting time of the soils with water and the release of Cu and Zn to water from the soils.

2.3 Effects of water-to-soil ratios on copper and zinc released from soils

Soil samples were equilibrated with deionized water in various water/soil ratios for 24 h. The concentrations of Cu and Zn in the extracts decreased significantly with increasing water/soil ratio (Fig. 3). However, the total quantity of Cu and Zn released per unit of soil weight increased with increasing the ratio (Fig. 4). The changes in Cu and Zn released with water/soil ratio were more significant in the soils with high levels of Cu and Zn (citrus soils) than the soils with low levels of Cu and Zn (forest soils). The results may suggest that total amounts of Cu or Zn released from the soils increase and the Cu or Zn concentration in the surface runoff

decrease with increasing rainfall. Because higher rainfall increases soil moisture content and dilutes composition of soil solution, leading to a marked decrease of Cu and Zn concentration in the soil solution.

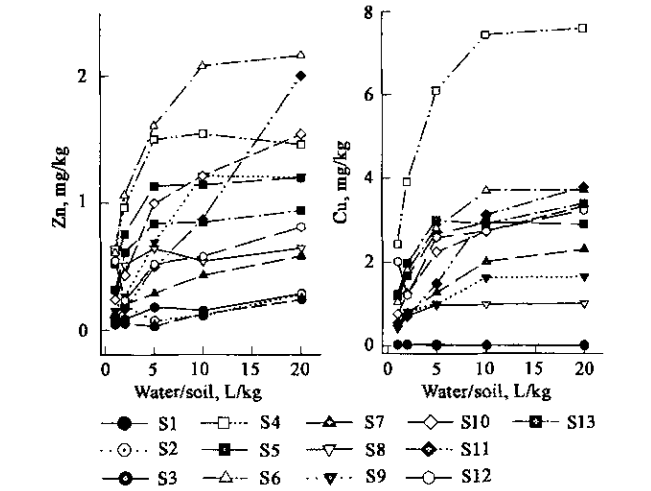


Fig.4 Amounts of Cu and Zn released from the soils as a function of the water/soil

2.4 Amounts of copper and zinc leached from the soils

The cumulative amounts of Cu leached from the soils increased linearly with the numbers of the leaching (Fig. 5). Change trend in the amounts of Cu leached among the soils was similar to that in amounts of Cu released from the shaking extraction. The amounts of Cu leached were high in the citrus soils than in the forest soils. The S4 had the highest amount of Cu leached. Based on intensities of soil-water reaction, above batch extraction can represent long-term leaching whereas column leaching can represent short-term leaching. Therefore, the amounts of Cu leached from the soils using the column leaching were much lower, as compared with the amounts of Cu released with the soils using the shaking extraction. The amounts of Cu leached from the soils using the column leaching were significantly correlated with its exchangeable Cu (X<sub>1</sub>), and organically bound Cu (X<sub>3</sub>) fractions (Table 2). The relationship was:  $Y = 0.079 + 0.35 X_1 + 0.0013 X_3$  ( $R^2 = 0.96$ ), in which the X<sub>1</sub> and X<sub>3</sub>

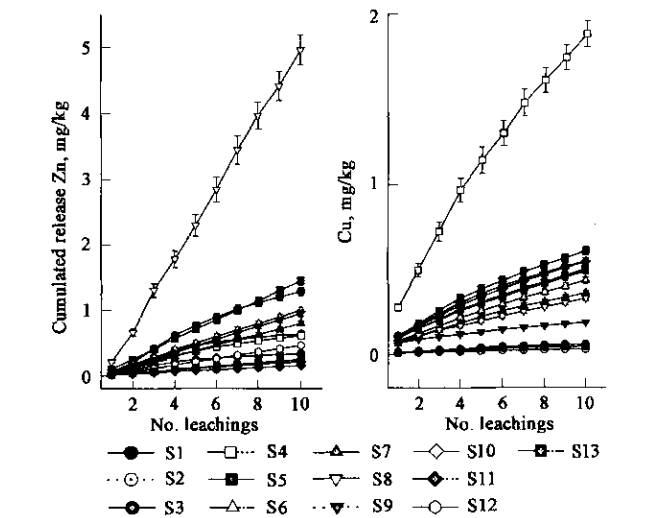


Fig.5 Cumulative amounts of Cu and Zn leached from the soils as a function of leaching number

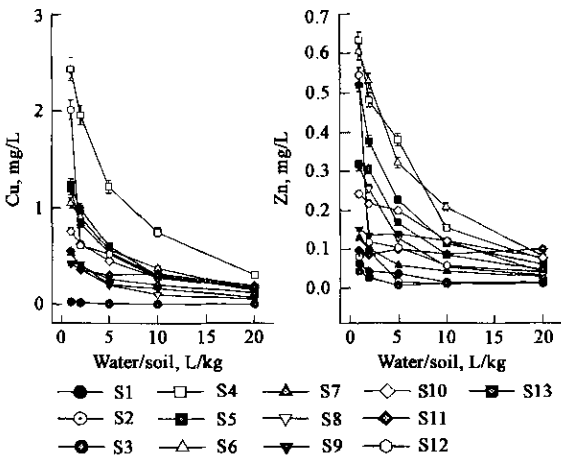


Fig.3 Copper and Zn concentrations in the equilibrium solution as a function of the water/soil

accounted for 91% and 5%, respectively, of the total variance in the leached Cu amounts.

Change trend in the amounts of Zn leached among the soils was very different from that in amounts of Zn released with the shaking extraction (Fig. 5). The highest cumulative amount of Zn leached from the soils was S8 that contained the highest level of exchangeable Cu (6.58 mg/kg) but the lowest pH (5.54) and the lowest total Zn (24.4 mg/kg) among the citrus soils (Table 1). Total amounts of Zn leached from the soils in ten leachings decreased in the order of  $S8 > S5 > S3 > S6 > S9 > S7 > S2 > S4 > S12 > S1 > S13 > S10 > S11$ . Although total Zn concentrations were much higher in the citrus soils than in the forest soils, there were no significant difference in the amount of Zn leached between the forest soils (S1–S3) and the citrus soils with an exception of S8 (Fig. 5). This may be due to higher pH and low percentages of exchangeable Cu in the citrus soils than in the forest soils. The cumulative amounts of Zn leached from the soils was significantly correlated only with exchangeable Zn fraction (Table 2), suggesting that the Zn leached from the soils in short-term leaching was mainly from the exchangeable fraction, which was different from the Zn released from the soils using the shaking extraction in long-term leaching.

## 2.5 Changes of copper and zinc leached from soils with pH

Leachant pH had the significant effect on potential leachability of Cu and Zn (Fig. 6). The cumulative amounts of Zn leached increased with decreasing pH for all three tested soils. Chuan *et al.* (Chuan, 1996) also reported that solubility of Zn in a contaminated soils increased with decreasing pH. However, the changes of Cu leached with pH were different from that of Zn, the lower leaching of Cu was found at pH 5–7. Further increase or decrease in pH increased leaching of Cu from the soils (Fig. 6). Increased amount of Cu leached in high pH was probably due to increase in organic matter dissolution under high pH condition and thus increased release of organically bound Cu fraction.

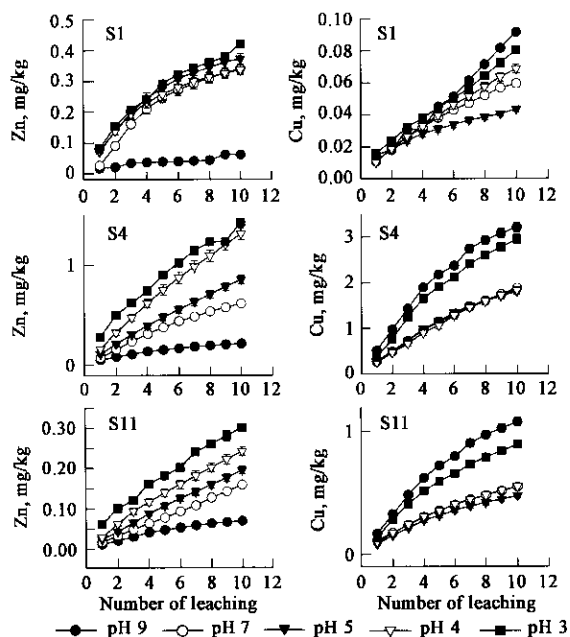


Fig. 6 Effects of leachant pH on Cu and Zn leached from the soils

## 2.6 Effects of electrolyte on copper and zinc released from soils

Types of cations in electrolyte had significant influences on amounts of Cu and Zn released from the sandy soils (Fig. 7). The influences on release of the two metals varied among the soils, and were different between Cu and Zn. Compared with the amount of Cu released in deionized water, use of 0.01 mol/L  $\text{Ca}(\text{NO}_3)_2$  as an extractant decreased significantly the release of Cu from the citrus soils (S4–S13). On the contrary, use of 0.02 mol/L  $\text{NaNO}_3$  as an extractant increased significantly the release of Cu from most of the soils (S1, S2, S3, S4, S6, S7, S8, S11, S12, and S13). Except for S4, S12 and S13, use of 0.02 mol/L  $\text{NH}_4\text{NO}_3$  or 0.02 mol/L  $\text{KNO}_3$  tended to decrease the release of Cu from the soils. The results might imply that soluble Cu released from the soils using the shaking extraction was partially from organically bound Cu in the soils. Because the soils contained high percentage of organically bound Cu (Fig. 1), increased Ca concentration in soil solution increased stability of organic matter-mineral complexes and decreased the dissolution of organic matter, and thus decreased the release of Cu-binding component of organic matter. However, high concentration of  $\text{NaNO}_3$  in the soil solution increased the dispersion of the organic matter-mineral complexes and increased dissolution of organic matter and the release of Cu from the soils.

The influences of electrolyte types on the release of Zn were different from that of Cu (Fig. 7). For the soils with the percentages of exchangeable Zn in the total Zn  $> 2.5\%$  (S1, S2, S3, S5, S6, S7, S8, and S9), use of 0.01 mol/L  $\text{Ca}(\text{NO}_3)_2$  as an extractant increased significantly the release of Zn from the citrus soils. However, the influences were not significant for the soils with the percentages of exchangeable Zn in the total Zn  $< 2.5\%$  (S4, S10, S11, S12, and S13). For the soils with the percentages of the total Zn as exchangeable Zn  $> 23\%$  (S1, S2, S3, and S8), use of 0.02 mol/L  $\text{NH}_4\text{NO}_3$ , 0.02 mol/L  $\text{NaNO}_3$  or 0.02 mol/L  $\text{KNO}_3$  also increased the release of Zn from the soils. The influences might be related to a fact that the release of Zn from the soils using the shaking extraction was partially from the exchangeable fraction. Increased Ca, K, Na, or  $\text{NH}_4$  concentrations in the soil solution increased the release of Zn from the soils by cation exchange reaction.

## 3 Conclusions

Copper released in batch extraction that represents long-term leaching was mainly from exchangeable, and carbonate bound Cu fractions, and Zn released in the batch extraction was mainly from its carbonate bound fraction. However, the Cu and Zn leached from the soils using the column leaching that represents short-term leaching were mainly from their exchangeable fractions. Leaching of Zn decreased with increase of pH from 3.0 to 9.0. Leaching of Cu was lower at pH 5–7, further increase or decrease in pH increased leaching of Cu from the soils. The releases of Cu and Zn increased with increasing soil-to-water time and ratio of water to soil, but Cu and Zn concentrations in the extracts decreased with increasing ratio of water to soil, suggesting that long-term waterlogging in the soils after rain may increase the loss of Cu and Zn to water from the soils, but the Cu or

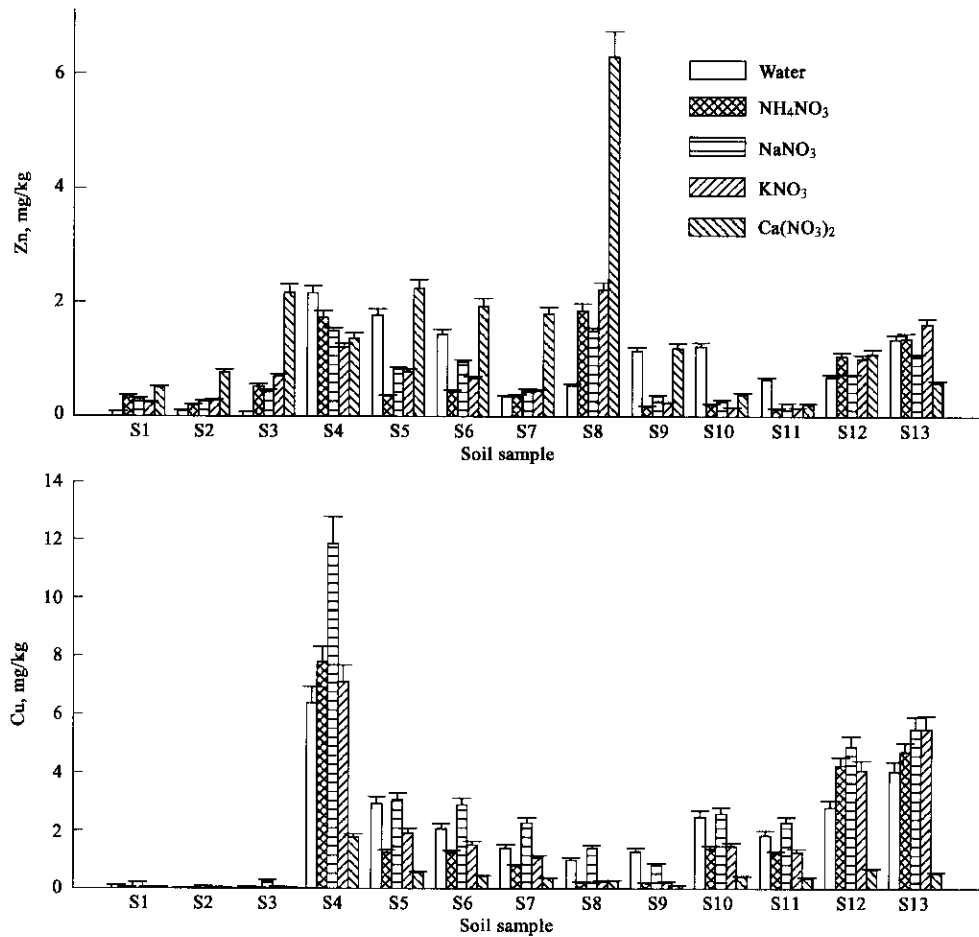


Fig.7 Effects of electrolyte types on release of Cu and Zn from the soils

Zn concentration in the surface runoff decrease with increasing rainfall intensity. Species of background electrolyte had significant effects on releases of Cu and Zn from the soils. The increased Ca concentration in soil solution increased stability of organic matter-mineral complexes and thus decreased the release of Cu-binding component of organic matter. In contrast, high concentration of Na in the soil solution increased the dispersion of the organic matter-mineral complexes and increased release of Cu from the soils. Increased Ca, K, Na, or NH<sub>4</sub> concentrations in the soil solution increased the release of Zn from the soils by cation exchange.

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