

Effect of chemical amendments on the concentration of cadmium and lead in long-term contaminated soils

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Abstract

The availability of metal in contaminated soil can be reduced by the addition of soil amendments. The objectives of this study are to study the effects of applying different soil amendments on the concentration of Cd and Pb in soil solution, DTPA or EDTA extractable Cd and Pb, and the uptake of Cd and Pb by wheat (*Triticum vulgare*) when growing in long-term Cd and Pb-contaminated soils, more than 20 years. The soil amendments, including check, compost, zinc oxide, calcium carbonate, calcium carbonate mixed with zinc oxide, and calcium carbonate mixed with compost, were conducted in a four replicates pot cultural study. The amended soils were incubated for six months under 60% of water holding capacity. Following incubation, wheat was grown for four months in greenhouse. Analyses of Cd concentration demonstrated a significant decrease in soil solution concentration and DTPA or EDTA extractable in soils amended with calcium carbonate or calcium carbonate mixed with ZnO (or compost) ($p < 0.01$). These amendments can significantly reduce the Cd concentration in the grain, leaf and stem, or reduce the total Cd uptake in all parts of wheat species grown in highly contaminated soil amended with calcium carbonate or calcium carbonate mixed with ZnO (or compost) ($p < 0.01$). The concentration of Cd in soil solution and extracted with DTPA or EDTA can predict the Cd concentration in wheat, especially for soil solution.

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1. Introduction

Heavy metal contamination in the natural soil environment is a major problem for crop quality, human health, and environmental quality, and most of the heavy metals are persistent in soil because of their immobile nature (Chen et al., 1996; Kabata-Pendias and Pendias, 2001). About 1200 sites were listed on the National Pri-

ority List (NPL) of USA for the treatment of contaminated soils, and approximately 63% of these sites were regarded as heavy metals contamination (Hazardous Waste Consultant, 1996). Soil pH value, organic matter, clay minerals, oxides of iron or aluminum, redox conditions, and the ageing of contamination controlled the behaviors of the heavy metals in contaminated soil and also affected the uptake of heavy metals by plants (Alloway, 1995; Naidu et al., 1997; Lock and Janssen, 2003; Vig et al., 2003). Thus the sorption of heavy metals in soils under different ionic strength and soil pH value is widely variable for different soils (Naidu et al., 1994). In Taiwan, some rural soils contaminated with Cd, Pb,

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Cu, and Zn have been identified by Taiwan EPA since 1980s. The contaminations of Cd and Pb in some rural soils of Taiwan were caused by the wastewater discharged from the chemical engineering plants in the industry parks (Chen, 1991).

Some soil remediation techniques were used to immobilize heavy metal in the contaminated soils and to reduce the soluble concentration in soils by the reactions of precipitation, adsorption, or to complex with organic matter in some case studies (Mench et al., 1994; Chen and Lee, 1997; Pierzynski, 1999; Kabata-Pendias and Pendias, 2001). Application of lime materials, compost, phosphate, oxides of iron and manganese, and zeolites significantly reduced the solubility of heavy metals in contaminated soils (McBride and Blasiak, 1979; Sommers and Lindsay, 1979; Kuo and McNeal, 1984; Kuo et al., 1985; Chen et al., 2000; Lee and Chen, 2000; Mench et al., 2000; Basta et al., 2001; Chen et al., 2001; Gupta et al., 2001; Hettiarachchi et al., 2001; Knox et al., 2001; Yang et al., 2001; Mavropoulos et al., 2002). Liming the contaminated soils to reduce the bioavailability of heavy metals is the most widely used remediation treatment. Application of lime materials to the contaminated soil leads to significantly increase the soil pH value because of the release of hydroxyl ion by the hydrolysis reaction of calcium carbonate. Liming can lead to the precipitation of metals as metal-carbonate and significantly decrease the exchangeable fraction of metals in contaminated soil (Knox et al., 2001). This reaction can reduce the bioavailability of heavy metals in soil and also reduce the uptake by plants (Street et al., 1977; Hirsch and Banin, 1990; Chlopecka and Adriano, 1996). Krebs et al. (1998) investigated heavy metal uptake by peas when growing in limed and check soils treated with mineral fertilizer (control), sewage sludge, and pig manure. The peas grown in the limed soils contained lower concentration of Cd, Cu, and Zn in above ground parts than that of plants grown on fertilized, un-limed soils. Friesl et al. (2003) also reported that applying lime significantly decreased Zn uptake by barley ($p < 0.5$). However, the labile or exchangeable form of metal in contaminated soils may increase and decrease the liming effectiveness after applying lime materials for a long time (Chlopecka and Adriano, 1996; Friesl et al., 2003). Thus frequently and repeatedly applying lime materials are required to increase the effect of lime on metal-contaminated soils.

McBride (1995) reported that the organic matters in sludge buffer soil pH and to avoid heavy metal uptake by plants. Organic matters with suitable reactive groups, such as hydroxyl, phenoxyl, and carboxyl, which effectively controlled the adsorption and complex of heavy metals with soil, and the activity of metals in soils (Allo-way, 1995; McBride et al., 1997). Application of organic matter in the metal-contaminated soil can efficiently reduce the concentration of Cd and Zn in the soil solution

(Isabelle and Alian, 2001) and also the concentrations of Cu, Mn, and Zn extracted with 0.01 M CaCl_2 (de Mora et al., 2003).

Zinc was one of the necessary trace elements for plants and rural soils have optimum Zn content for plant growth. Shen et al. (1996) indicated that the application of Zn can efficiently increase the yield of the *Thlaspi caerulescens*. Oliver et al. (1996) divided the crops into zinc-efficient and zinc-inefficient species. The zinc-inefficient crops were able to tolerate Zn deficiency and accumulated more Cd than that of zinc-efficient species. They also indicated that the application of Zn in the normal area and Zn deficient areas was effective in reducing the Cd concentration in the wheat grain. When growing in nutrient solution contained low concentration of Cd, a strong antagonistic effect of Zn on Cd accumulation was found in young leaves of lettuce or spinach (McKenna et al., 1993).

The objectives of this study are: (1) to evaluate the effects of chemical amendments on the concentrations of Cd and Pb in soil solution and extract with DTPA or EDTA, and (2) to evaluate the effects of different chemical amendments on the Cd and Pb concentration in the different parts of wheat and total metal uptake in wheat when growing in long-term Cd- and Pb-contaminated soils, more than 20 years, of northern Taiwan.

2. Materials and methods

2.1. Basic soil properties of four soils

Four soils contaminated with Cd and Pb in northern Taiwan, highly contaminated Tatan sandy soil site and slightly contaminated Chungfu clayey soil site, were selected for this study. Tatan A and B sandy soils and Chungfu C and D clayey soils were collected from four different sites contaminated by two different chemical engineering plants located at both the Tatan village and Chungfu village for more than 20 years, respectively. The surface soils (0–20 cm) of each contaminated site were sampled, air-dried, ground through a 2 mm sieve, mixed, and stored in the plastic vessel prior to laboratory analysis and pot experiments. Soil pH values were determined using glass electrodes in a soil: water ratio of 1:1 (McLean, 1982). The particle size distribution of the two soils was analyzed by the pipette method (Gee and Bauder, 1986). Organic carbon content was determined by the Walkley–Black wet combustion method (Nelson and Sommers, 1982). Exchangeable cations (K, Na, Ca, and Mg) and cation exchange capacity (CEC) were determined using ammonium acetate (pH 7.0) (Rhoades, 1982; Thomas, 1982). Lime was applied in the study to increase the soil pH value to 7.5, and the amounts of lime used were determinate by direct determination (Thomas, 1982).

2.2. Chemical amendments design

The contaminate soils (3.5 kg, dry weight) were mixed with the following chemical amendments and then transferred to the plastic pots (16 cm diameter and 19.5 cm height) for pot experiments. The chemical amendments including (1) control (no treatment), (2) hog manure (60 g) composts (CO) per pot (equal to 40 tons/ha), (3) zinc oxide (0.08 g) (ZN) in $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ per pot (equal to 50 kg ZnO/ha) for competition for Cd uptake by wheat from the soil, (4) calcium carbonate (CA) for Tatan sandy soils (15 g/pot) and Chungfu clayey soils (30 g/pot), respectively, (equal to 10 tons/ha for Tatan soils and 20 tons/ha for Chungfu soils) to increase the soil pH value to 7.5, (5) 15 g and 30 g of calcium carbonate mixed with zinc oxide (50 kg ZnO/ha) (CA + ZN) per pot for Tatan sandy soils and Chungfu clayey soils, respectively, and (6) 15 g and 30 g of calcium carbonate mixed with compost (40 tons/ha) (CA + CO) per pot for Tatan sandy soils and Chungfu clayey soils, respectively. Each treatment was replicated four times and arranged in the greenhouse using RCBD (randomized complete block design), and incubated for six months at 60% of the water holding capacity by weighing the pots every 2 days and adding deionized water to compensate for the lost moisture.

2.3. Pot experiments and laboratory analysis

More than 20 seeds of the wheat species (*Triticum vulgare*, Taichung No. 31) were grown in each pot and controlled to 15 plants per pot after germination. Fifteen plants of wheat were harvested and weighted after growing in the pots for four months. The harvested wheat were divided into grain, leaf and stem, and husk part and then oven-dried at 60 °C for 72 h. The samples were ground to 0.4 mm size with a grinder for more homogenized condition and 0.3 g of grain, leaf and stem, or husk of wheat were digested with $\text{H}_2\text{SO}_4/\text{H}_2\text{O}_2$ digestion method (Jones and Case, 1990). The digested solutions were whole through a Whatman No. 42 filter paper and diluted to a 25 ml flask.

The concentration of Cd and Pb in the treated soils were determined by analyzing the soil solution and 0.005 M DTPA (pH 5.3) or 0.05 M EDTA (pH 7.0) extracts (Lindsay and Norvell, 1978; Mench et al., 1994). Soil solutions were collected directly with rhizon soil moisture samplers (RSMS) produced in the Netherlands. The concentration of Cd and Pb in extracts, soil solutions, and digested solutions of wheat were determined by ICP (Jobin Yuon 138 ULTRANCE). The total concentration of Cd and Pb were digested with aqua regia and determined by ICP (Jobin Yuon 138 ULTRANCE).

2.4. Statistics

The variance and significant differences of concentration of Cd and Pb in different chemical amended soils, grain, leaf and stem, and husk were analyzed by ANOVA. The statistical significance level in this study was defined at $p < 0.01$.

3. Results and discussion

3.1. Soil properties

The cation exchange capacity (CEC) of Chungfu C and D clayey soils were greater than those of Tatan A and B sandy soils because of the greater contents of organic carbon and clay in the Chungfu C and D clayey soils (Table 1). Those mechanisms involved in the adsorption of metals in soil including cation exchange, specific adsorption, co-precipitation, and organic complexation (Alloway, 1995). Adsorption of metals increased with increasing their clay and organic matter contents in the soils (Naidu et al., 1997). The CEC and organic carbon of Chungfu clayey soils were greater than that of Tatan sandy soils, which indicate that Chungfu C and D clayey soils can adsorb more Cd and Pb than Tatan A and B sandy soils. The soil texture was coarser in the Tatan A and B sandy soils than Chungfu clayey soils and leached into the groundwater

Table 1
The physical and chemical properties of four studied soils

Soil sample	pH H_2O	O.C. ^a (g/kg)	Particle size analysis (g/kg)			CEC ^b (cmol ₍₊₎ /kg soil)	Exch. base (cmol ₍₊₎ /kg soil)	Base saturation (%)
			Sand	Silt	Clay			
Tatan A soil	6.0	12.1	726	57	217	4.5	3.23	72
Tatan B soil	5.9	12.9	742	123	135	4.5	3.71	82
Chungfu C soil	5.4	23.5	113	481	406	12.4	3.82	31
Chungfu D soil	5.1	15.2	102	524	374	9.9	3.77	38

^a O.C.: Organic carbon.

^b CEC: Cation exchange capacity.

by precipitation or irrigation water was easier to occurred.

The bioavailability of Cd and Pb in the four contaminated soils was assessed by extracts of 0.1N HCl, 0.05M EDTA (pH 7.0), and 0.005M DTPA (pH 5.3), respectively. The extractable concentration in Tatan soils ranged from 9.80 to 32.0mg Cd/kg and ranged from 112 to 163mg Pb/kg, respectively. The extractable concentration in Chungfu soils ranged from 1.14 to 2.23mg Cd/kg and from 2.82 to 13.8mg Pb/kg, respectively (Table 2). The Taiwan EPA have identified the Tatan two soils were highly contaminated with Cd and slightly contaminated with Pb, and Chungfu two clayey soils were slightly contaminated with Cd and not contaminated with Pb. The Cd concentration in the grain of rice grown in these two Chungfu soils was higher than that of control regulation level of Cd in the grain of rice (0.5mg/kg) proposed by the Department of Health of Taiwan, and also beyond the allowable limit of Germany (0.1mg/kg) and maximum levels of Joint FAO/WHO commission (0.2mg/kg) (Joint FAO/WHO Food Standards Programme, 2002), respectively.

3.2. Effect on the soil pH

Soil pH value was significantly increased after the treatments of calcium carbonate ($p < 0.01$). Calcium carbonate (CA), calcium carbonate mixed with zinc oxide (CA + ZN), and calcium carbonate mixed with compost (CA + CO) significantly increased the soil pH value from 5.0–5.7 (control) to 7.6–7.8 in sandy soils and from 5.5–6.0 (control) to 7.5–7.8 in clayey soils ($p < 0.01$) (not shown). The soil pH value after addition of zinc oxide

also significantly changed, ranged from 5.2 to 5.5 in Tatan sandy soils and ranged from 5.2 to 5.9 in Chungfu clayey soils, respectively ($p < 0.01$). Because of the higher pH value of applied compost (pH 6.4) compared with the studied soils (pH 5.9–6.0 in Tatan sandy soils and pH 5.1–5.4 in Chungfu clayey soils), the treatment of compost significantly increased soil pH value of the studied soils, especially for Tatan B sandy soil ($p < 0.01$).

3.3. Effects on the concentration of Cd and Pb extracted with DTPA

3.3.1. Calcium carbonate

The addition of calcium carbonate (CA), calcium carbonate mixed with zinc oxide (CA + ZN), and calcium carbonate mixed with compost (CA + CO) significantly decreased DTPA extractable Cd and Pb concentration, especially in Tatan A sandy soil ($p < 0.01$) (Tables 3 and 4). The concentration of Cd in extracts was significantly decreased from 35.0 ± 2.70 mg/kg to 8.90 ± 0.62 mg/kg in Tatan A sandy soil treated with calcium carbonate. However, there was no significant effect of calcium carbonate on DTPA extractable Cd concentration in Chungfu C and D clayey soils because of the lower concentration of Cd and Pb in these two soils compared with Tatan soils. The increase of soil pH value resulted in the precipitation of heavy metals with carbonates (Naidu et al., 1994; Holm et al., 1996), and also reduced the solubility of heavy metals in the soil (Chlopecka et al., 1996; McBride et al., 1997; Krebs et al., 1998). In addition, the rising of soil pH increased the pH-depend charges and also increased the metals adsorbed with the soil particles (Pardo and Guadalix, 1996; Naidu et al., 1997). Therefore, the treatments of calcium carbonate can significantly decrease the concentration of Cd in the soil extracts, especially for Tatan A sandy soil seriously contaminated with Cd. These results are consistent with that reported by numerous studies (McBride and Blasiak, 1979; Sommers and Lindsay, 1979; Kuo et al., 1985; Chen and Lee, 1997). But, we think the liming treatment do not deliver a long-term immobilization of Cd in the soil, especially for the Tatan sandy soil, compared with previous studies (Chlopecka and Adriano, 1996; Friesl et al., 2003).

3.3.2. Organic matter

Due to the lower concentration of Cd and Pb and higher contents of organic carbon and clay in the Chungfu soils compared with Tatan soils, the addition of compost had no significant effect on decreasing the concentration of Cd in Chungfu clayey soils, and only significantly decreased the concentration of Cd and Pb in the Tatan A sandy soil ($p < 0.01$) (Tables 3 and 4). The Zn concentration extracted with 0.05M EDTA ranged from 7 to 10mg/kg (data not shown) which is considered adequate rate for wheat growth and produc-

Table 2

The concentration of Cd and Pb extracted with 0.1 M HCl, 0.05 M EDTA DTPA for four soils

Extraction methods	Cd (mg/kg)	Pb (mg/kg)
<i>Tatan A sandy Soil</i>		
0.1 N HCl	32.0	121
0.05M EDTA	26.2	115
0.005M DTPA	16.7	79.8
<i>Tatan B sandy soil</i>		
0.1 N HCl	9.96	158
0.05M EDTA	11.6	163
0.005M DTPA	9.80	112
<i>Chungfu C clayey soil</i>		
0.1 N HCl	2.17	13.8
0.05M EDTA	2.23	10.0
0.005M DTPA	1.55	4.52
<i>Chungfu D clayey soil</i>		
0.1 N HCl	1.40	9.93
0.05M EDTA	1.42	6.37
0.005M DTPA	1.14	2.82

Table 3

The concentration of Cd in soil solution or extracted with DTPA or EDTA in contaminated soils treated with different amendments

Amendment ^a	Cd soil solution (mg/l)	Cd DTPA (mg/kg dry soils)	Cd EDTA (mg/kg dry soils)
<i>Tatan A sandy soil</i>			
Control	5.11 ± 2.50 ab	35.0 ± 2.70 a	48.5 ± 2.25 a
CO	4.38 ± 5.06 ab	20.9 ± 4.38 b	33.6 ± 2.03 b
ZN	8.48 ± 5.24 a	35.5 ± 2.95 a	45.8 ± 2.58 a
CA	0.07 ± 0.01 b	8.90 ± 0.62 c	21.3 ± 2.52 c
CA + ZN	0.06 ± 0.01 b	9.19 ± 0.74 c	21.1 ± 3.52 c
CA + CO	0.09 ± 0.01 b	9.79 ± 1.17 c	22.1 ± 3.85 c
<i>Tatan B sandy soil</i>			
Control	0.61 ± 0.64 a	7.60 ± 1.41 ab	8.83 ± 0.08 a
CO	0.60 ± 0.39 a	7.32 ± 0.97 ab	8.80 ± 0.53 a
ZN	3.84 ± 5.22 a	9.10 ± 0.80 a	10.2 ± 2.34 a
CA	0.01 ± 0.00 a	7.12 ± 0.52 b	9.66 ± 0.46 a
CA + ZN	0.01 ± 0.01 a	7.38 ± 0.34 ab	9.49 ± 1.25 a
CA + CO	0.03 ± 0.01 a	6.80 ± 0.84 b	8.92 ± 1.52 a
<i>Chungfu C clayey soil</i>			
Control	0.01 ± 0.01 abc	1.43 ± 0.02 a	1.83 ± 0.11 a
CO	0.02 ± 0.01 a	1.38 ± 0.07 a	1.81 ± 0.06 a
ZN	0.01 ± 0.01 ab	1.47 ± 0.04 a	1.83 ± 0.13 a
CA	0.004 ± 0.001 c	1.07 ± 0.03 a	1.68 ± 0.05 a
CA + ZN	0.004 ± 0.001 c	1.37 ± 0.48 a	1.69 ± 0.07 a
CA + CO	0.01 ± 0.001 bc	1.29 ± 0.29 a	1.74 ± 0.04 a
<i>Chungfu D clayey soil</i>			
Control	0.01 ± 0.00 bc	1.26 ± 0.74 a	1.16 ± 0.04 a
CO	0.02 ± 0.01 a	1.16 ± 0.50 a	1.20 ± 0.02 a
ZN	0.011 ± 0.003 ab	0.93 ± 0.09 a	1.20 ± 0.06 a
CA	0.004 ± 0.002 c	0.59 ± 0.02 a	0.96 ± 0.03 c
CA + ZN	0.003 ± 0.000 c	0.62 ± 0.03 a	0.98 ± 0.02 bc
CA + CO	0.004 ± 0.000 c	0.49 ± 0.28 a	1.03 ± 0.04 b

Data are expressed as mean value of four replicates.

Different letters in a column indicate significant difference between amendments at $p < 0.01$.

^a Control: No treatment; CO: applying composts 40 tons/ha; ZN: applying zinc oxide 50 kg ZnO/ha; CA: applying calcium carbonate to increase the soil pH to 7.5; CA + ZN: applying calcium carbonate mixed with zinc oxide; and CA + CO: applying calcium carbonate mixed with compost.

tion. In this study, Zn has no effects on competitive uptake of Cd for plant's growth in terms of plant physiology. This maybe attributed to the adequate Zn concentration for wheat growth in the original contaminated soils. No competitive uptake between Cd and Zn occurred in the amendment of zinc oxide in Tatan or Chungfu soils. The addition of the zinc oxide can not significantly reduce the DTPA extractable Cd and Pb concentration in soils.

3.4. Effects on the concentration of Cd and Pb extracted with EDTA

Similar concentrations of Cd and Pb were detected in 0.05 M EDTA and 0.005 M DTPA extracts of four soils (Table 3 and 4). The application of calcium carbonate (CA), calcium carbonate mixed with zinc oxide (CA + ZN), and calcium carbonate mixed with compost

(CA + CO) significantly decreased the concentration of Cd in the extracts of the Tatan A sandy soil and Chungfu D clayey soil ($p < 0.01$), but there was no effect of compost compared with calcium carbonate except for Tatan A sandy soil. The concentration of Cd in extracts was significantly decreased from 48.5 ± 2.25 mg/kg to 21.3 ± 2.52 mg/kg in Tatan A sandy soil and decreased from 1.16 ± 0.04 mg/kg to 0.96 ± 0.03 mg/kg in Chungfu D clayey soil treated with calcium carbonate, respectively ($p < 0.01$).

After comparison the concentration of Cd and Pb extracted with DTPA and EDTA, 0.05 M EDTA extracted more amounts of Cd and Pb in contaminated soils than 0.005 M DTPA (Tables 3 and 4). We supposed that 0.05 M EDTA (pH 7.0) was a stronger extracting reagent than 0.005 M DTPA (pH 5.3) and can extract more quantity of Cd and Pb of the contaminated soils in this study.

Table 4

The concentration of Pb in soil solution or extracted with DTPA or EDTA in contaminated soils treated with different amendments

Amendment ^a	Pb soil solution (mg/l)	Pb DTPA (mg/kg dry soils)	Pb EDTA (mg/kg dry soils)
<i>Tatan A sandy soil</i>			
Control	0.04 ± 0.00 b	93.5 ± 3.33 a	130 ± 2.36 ab
CO	0.04 ± 0.00 b	84.9 ± 5.80 b	120 ± 1.28 b
ZN	0.04 ± 0.00 b	91.6 ± 3.27 a	124 ± 4.95 b
CA	0.05 ± 0.00 ab	75.5 ± 0.91 c	137 ± 7.11 a
CA + ZN	0.05 ± 0.01 ab	75.9 ± 1.75 c	131 ± 9.12 ab
CA + CO	0.05 ± 0.01 a	73.9 ± 0.92 c	130 ± 5.62 ab
<i>Tatan B sandy soil</i>			
Control	0.04 ± 0.00 b	137 ± 1.22 ab	176 ± 1.32 a
CO	0.04 ± 0.00 b	124 ± 5.66 bc	175 ± 5.05 a
ZN	0.04 ± 0.00 b	145 ± 5.23 a	190 ± 22.3 a
CA	0.04 ± 0.00 b	120 ± 5.56 c	191 ± 8.12 a
CA + ZN	0.06 ± 0.00 ab	121 ± 2.95 c	185 ± 13.3 a
CA + CO	0.06 ± 0.01 a	117 ± 8.53 c	181 ± 16.5 a
<i>Chungfu C clayey soil</i>			
Control	0.04 ± 0.00 b	6.24 ± 0.14 a	11.0 ± 0.42 cd
CO	0.04 ± 0.00 b	5.80 ± 0.23 ab	11.6 ± 0.53 bc
ZN	0.04 ± 0.00 b	6.27 ± 0.18 a	10.6 ± 0.37 d
CA	0.05 ± 0.01 ab	4.94 ± 0.17 bc	12.1 ± 0.35 ab
CA + ZN	0.04 ± 0.00 b	5.78 ± 1.10 ab	12.2 ± 0.48 ab
CA + CO	0.06 ± 0.01 a	4.52 ± 0.50 c	12.6 ± 0.24 a
<i>Chungfu D clayey soil</i>			
Control	0.04 ± 0.00 a	4.18 ± 2.45 a	4.59 ± 0.95 a
CO	0.04 ± 0.00 a	2.68 ± 0.70 a	4.52 ± 0.15 a
ZN	0.04 ± 0.00 a	3.42 ± 1.80 a	5.13 ± 1.97 a
CA	0.04 ± 0.00 a	2.15 ± 0.23 a	4.99 ± 0.42 a
CA + ZN	0.04 ± 0.00 a	2.03 ± 0.12 a	4.89 ± 0.15 a
CA + CO	0.04 ± 0.00 a	1.92 ± 0.10 a	5.34 ± 0.06 a

Data are expressed as mean value of four replicates.

Different letters in a column indicate significant difference between amendments at $p < 0.01$.^a The amendments are shown in Table 3.

3.5. Effects on the concentration of Cd and Pb in soil solution

Consistent with the trends recorded above for DTPA and EDTA extractable Cd and Pb concentration, adding of calcium carbonate (CA), calcium carbonate mixed with zinc oxide (CA + ZN), and calcium carbonate mixed with compost (CA + CO) decreased the concentration of Cd in soil solution, but were not statistically significant. However, the concentration of Cd and Pb in soil solution are much less than those of DTPA or EDTA extracts, especially for the highly contaminated Tatan A and B sandy soils (Tables 3 and 4). The addition of calcium carbonate reduced the concentration of Cd in the soil solutions of Tatan A sandy soil (changed from 5.11 ± 2.50 to 0.07 ± 0.01 mg/l). The addition of compost (40 tons/ha) only slightly decreased the concentration of Cd in Tatan A sandy soil (changed from 5.11 ± 2.50 to 4.38 ± 5.06 mg/l), but there was no effect on Chungfu clayey soils which were only slightly con-

taminated with Cd. Because the concentrations of metals in soil solutions are significantly related to the metals uptake by plants (Lock and Janssen, 2003), we proposed that the application of calcium carbonate, calcium carbonate mixed with zinc oxide (or compost) can decrease the concentrations of metals both in soil solutions and within plants.

3.6. Effects on the growth and heavy metals uptake of wheat

Wheat was one of main crops for food stuff, therefore the concentration of heavy metal in this crop was seriously monitored to control the health of the public. It is well known from many previously studies that wheat can uptake and accumulate heavy metals from the soil. Wheat was a xerophyte, simply to control the soil water content, and the concentration of heavy metal in the soil solution will not be easily increased through the practices of drainage and irrigation system.

The application of calcium carbonate (CA), calcium carbonate mixed with zinc oxide (CA + ZN), or calcium carbonate mixed with compost (CA + CO) significantly reduced grain Cd concentration of the wheat species under amendments (Table 5). The grain Cd content of wheat grown in the soil treated with calcium carbonate was significantly decreased from 11.0 ± 0.93 mg/kg to 4.43–4.63 mg/kg for Tatan A soil, 10.1 ± 2.45 mg/kg to 3.53–4.32 mg/kg for Tatan B soil, 1.49 ± 0.13 mg/kg to 0.36–0.52 mg/kg for Chungfu C soil, and from 1.46 ± 0.19 mg/kg to 0.13–0.23 mg/kg for Chungfu D soil, respectively ($p < 0.01$).

However, the application of calcium carbonate did not significantly reduce grain Pb concentration of wheat grown in the four contamination sites because of the studied soil were slightly contaminated with Pb or with no Pb contamination (maximum concentration extracted with 0.05 M EDTA was 163 mg Pb/kg in Tatan B sandy soil). The Pb concentration in the grain of the

wheat in the four studied soils (< 5 mg/kg) were all below the safety level of 10 mg/kg reported by Lin et al. (1993), but were all beyond the allowable limit of Germany (0.3 mg/kg) and maximum levels of Joint FAO/WHO Food Standards Programme Codex alimentarius commission (0.2 mg/kg). The Cd concentration in the grain of wheat, after growing in Chungfu C and D clayey soils which were slightly contaminated with Cd and treated with calcium carbonate, was significantly lower than the regulation of Taiwan (0.5 mg/kg), surveillance level of Japan (0.5 mg/kg) (Asami, 1986), and maximum allowable limit of Japan (1 mg/kg) (Asami, 1986). However, most of the concentration of Cd in the grain of wheat were beyond the allowable limit of Germany (0.1 mg/kg) and maximum levels of Joint FAO/WHO Food Standards Programme Codex alimentarius commission (0.2 mg/kg) (Table 5). The treatment of calcium carbonate significantly decreased the Cd concentration in the grain of the wheat grown in highly contaminated

Table 5

The yield of grain and concentration of Cd in the grain, leaf and stem, and husk of wheat grown in contaminated soils treated with different amendments

Amendment ^a	Grain yield (dry weight) (g/pot)	Cd in grain (mg/kg)	Cd in leaf and stem (mg/kg)	Cd in husk (mg/kg)
<i>Tatan A sandy soil</i>				
Control	0.09 ± 0.04 b	11.0 ± 0.93 a	136 ± 15.8 a	27.2 ± 3.72 b
CO	0.59 ± 0.30 b	7.14 ± 0.42 ab	56.0 ± 5.22 b	16.1 ± 1.36 c
ZN	0.08 ± 0.02 b	10.3 ± 7.56 ab	125 ± 21.8 a	37.6 ± 2.59 a
CA	3.04 ± 0.19 a	4.63 ± 0.63 b	28.4 ± 1.30 c	10.2 ± 1.53 d
CA + ZN	3.15 ± 0.32 a	4.43 ± 0.16 b	27.2 ± 1.65 c	11.3 ± 0.62 d
CA + CO	3.64 ± 0.71 a	5.58 ± 0.33 ab	38.8 ± 4.46 bc	12.0 ± 0.21 d
<i>Tatan B sandy soil</i>				
Control	0.49 ± 0.19 bc	10.1 ± 2.45 a	62.0 ± 16.6 a	19.5 ± 6.95 a
CO	1.31 ± 0.29 bc	6.79 ± 0.84 b	34.8 ± 4.55 bc	13.0 ± 0.88 b
ZN	0.11 ± 0.06 c	8.10 ± 1.36 ab	49.4 ± 13.5 ab	16.6 ± 1.91 b
CA	2.57 ± 0.65 a	4.32 ± 0.19 c	18.1 ± 2.08 c	8.13 ± 1.33 c
CA + ZN	1.50 ± 1.08 ab	3.74 ± 0.33 c	16.0 ± 0.75 c	7.46 ± 0.71 c
CA + CO	2.64 ± 0.19 a	3.53 ± 0.23 c	20.4 ± 1.79 c	7.19 ± 0.70 c
<i>Chungfu C clayey soil</i>				
Control	3.84 ± 1.13 a	1.49 ± 0.13 b	8.09 ± 0.69 a	6.50 ± 0.56 a
CO	2.06 ± 1.22 a	1.75 ± 0.22 a	9.62 ± 5.31 a	6.98 ± 0.82 a
ZN	3.55 ± 1.50 a	1.53 ± 0.06 b	7.47 ± 0.39 a	6.78 ± 1.08 a
CA	4.47 ± 1.22 a	0.52 ± 0.15 c	1.79 ± 0.17 b	1.32 ± 0.28 b
CA + ZN	4.57 ± 1.38 a	0.36 ± 0.06 c	1.25 ± 0.1 b	1.00 ± 0.39 b
CA + CO	4.05 ± 2.00 a	0.47 ± 0.13 c	1.73 ± 0.27 b	1.40 ± 0.38 b
<i>Chungfu D clayey soil</i>				
Control	2.08 ± 1.15 b	1.46 ± 0.19 b	7.56 ± 0.62 b	5.97 ± 1.55 b
CO	1.58 ± 0.81 b	1.87 ± 0.15 a	9.82 ± 0.77 a	8.64 ± 1.07 a
ZN	2.15 ± 1.01 b	1.76 ± 0.21 a	8.04 ± 0.67 b	7.07 ± 0.79 b
CA	4.79 ± 0.86 a	0.16 ± 0.07 c	0.72 ± 0.04 c	0.41 ± 0.26 c
CA + ZN	4.52 ± 0.60 a	0.13 ± 0.06 c	0.71 ± 0.37 c	0.32 ± 0.26 c
CA + CO	3.20 ± 1.07 ab	0.23 ± 0.08 c	1.41 ± 0.96 c	0.66 ± 0.28 c

Data are expressed as mean value of four replicates.

Different letters in a column indicate significant difference between amendments at $p < 0.01$.

^a The amendments are shown in Table 3.

Table 6

The concentration of Pb in the grain, leaf and stem, and husk of wheat grown in contaminated soils treated with different amendments

Amendment ^a	Pb in grain (mg/kg)	Pb in leaf and stem (mg/kg)	Pb in husk (mg/kg)
<i>Tatan A sandy soil</i>			
Control	0.80 ± 0.00 a	4.79 ± 0.00 a	6.68 ± 0.00 a
CO	2.00 ± 1.31 a	4.30 ± 0.00 a	7.22 ± 0.00 a
ZN	0.80 ± 0.00 a	4.84 ± 0.00 a	8.82 ± 0.00 a
CA	4.24 ± 0.00 a	0.80 ± 0.00 a	8.90 ± 0.00 a
CA + ZN	0.01 ± 0.00 a	0.80 ± 0.00 a	7.65 ± 0.00 a
CA + CO	0.81 ± 0.12 a	0.80 ± 0.00 a	8.48 ± 0.00 a
<i>Tatan B sandy soil</i>			
Control	1.57 ± 0.71 a	0.80 ± 0.00 a	0.80 ± 0.00 a
CO	3.98 ± 1.48 a	4.93 ± 0.00 a	4.18 ± 0.36 a
ZN	0.80 ± 0.00 a	0.80 ± 0.00 a	0.80 ± 0.00 a
CA	2.59 ± 0.28 a	4.64 ± 0.00 a	5.33 ± 0.00 a
CA + ZN	5.66 ± 6.20 a	0.80 ± 0.00 a	7.18 ± 5.05 a
CA + CO	3.14 ± 0.84 a	4.15 ± 0.00 a	6.23 ± 0.00 a
<i>Chungfu clayey soil</i>			
Control	2.81 ± 0.19 b	4.66 ± 0.69 a	4.15 ± 0.65 a
CO	3.51 ± 0.82 ab	0.80 ± 0.00 a	5.18 ± 0.00 a
ZN	2.68 ± 0.16 b	0.80 ± 0.00 a	5.65 ± 0.00 a
CA	4.39 ± 0.71 a	5.33 ± 0.26 a	4.73 ± 0.00 a
CA + ZN	4.34 ± 0.72 a	6.49 ± 0.00 a	5.06 ± 0.00 a
CA + CO	3.53 ± 0.61 ab	0.80 ± 0.00 a	4.37 ± 0.00 a
<i>Chungfu clayey soil</i>			
Control	3.49 ± 0.48 a	0.80 ± 0.00 a	5.50 ± 0.00 a
CO	4.04 ± 0.78 a	4.35 ± 0.20 a	5.51 ± 0.00 a
ZN	3.21 ± 0.61 a	0.80 ± 0.00 a	4.44 ± 0.64 a
CA	2.96 ± 0.64 a	6.56 ± 3.08 a	5.33 ± 1.98 a
CA + ZN	3.19 ± 0.68 a	0.80 ± 0.00 a	4.61 ± 0.00 a
CA + CO	3.07 ± 0.23 a	4.45 ± 0.00 a	5.98 ± 0.00 a

Data are expressed as mean value of four replicates.

Different letters in a column indicate significant difference between amendments at $p < 0.01$.^a The amendments are shown in Table 3.

Table 7

The total uptake of Cd and Pb by wheat grown in contaminated soils treated with different amendments

Amendment ^a	Tatan A Sandy soil	Tatan B Sandy soil (µg/pot)	Chungfu C Clayey soil (µg/pot)	Chungfu D Clayey soil
<i>Cadmium</i>				
Control	282 a	393 a	62.4 a	34.9 a
CO	203 a	229 b	40.5 ab	38.8 a
ZN	293 a	227 b	55.0 a	38.9 a
CA	219 a	152 bc	17.1 b	5.09 b
CA + ZN	229 a	129 bc	12.4 b	5.87 b
CA + CO	294 a	125 c	13.2 b	7.00 b
<i>Lead</i>				
Control	6.77 a	0.70 a	25.3 a	7.66 b
CO	11.6 a	26.1 a	7.64 a	14.0 b
ZN	15.3 a	0.80 a	12.0 a	9.18 b
CA	30.1 a	17.6 a	40.7 a	36.0 a
CA + ZN	15.0 a	21.5 a	32.4 a	15.6 ab
CA + CO	6.82 a	14.8 a	15.2 a	15.3 ab

Data are expressed as mean value of four replicates.

Different letters in a column indicate significant difference between amendments at $p < 0.01$.^a The amendments are shown in Table 3.

Tatan two soils ($p < 0.01$), but the Cd concentration of wheat grain is still too high, ranging from 3.5 to 5.6 mg/kg and have risk for food security.

In the leaf and stem of the wheat specie, the application of calcium carbonate (CA), calcium carbonate mixed with zinc oxide (CA + ZN), or calcium carbonate mixed with compost (CA + CO) significantly reduced the Cd concentration ($p < 0.01$), this result was similar to that of wheat grain. Because of the lower concentration of metals and the higher contents of organic carbon and clay in Chungfu soils compared with Tatan sandy soils, more amounts of metals were adsorbed with organic carbon or clay particles in the Chungfu clayey soils. Thus the addition of compost can only significantly reduce the Cd concentration in the leaf and stem of wheat grown in the Tatan sandy soils ($p < 0.01$), but it was not significant for Chungfu clayey soils. Similar to the concentration of Pb in the wheat grain, there was no effect

on the concentration of Pb in leaf and stem of wheat among those chemical treatments because of the low concentration of Pb in the studied soils (Table 6).

Because the treatments of the calcium carbonate in four soils significantly increased the soil pH value ($p < 0.01$), the concentration of Cd in the husk reduced significantly ($p < 0.01$) (Table 5). This result was similar to that reported by Gavi et al. (1997) and Mench et al. (1997). Because of the lower concentration of metals and higher contents of organic carbon and clay in Chungfu soils compared with Tatan sandy soils, more amounts of metals were adsorbed with organic carbon or clay particles in the Chungfu clayey soils. The addition of compost only significantly reduced the Cd concentration in the husk of wheat grown in the Tatan sandy soils ($p < 0.01$). Similar to the results of the grain or leaf and stem, there was no different of Pb concentration in the husk of wheat among those chemical

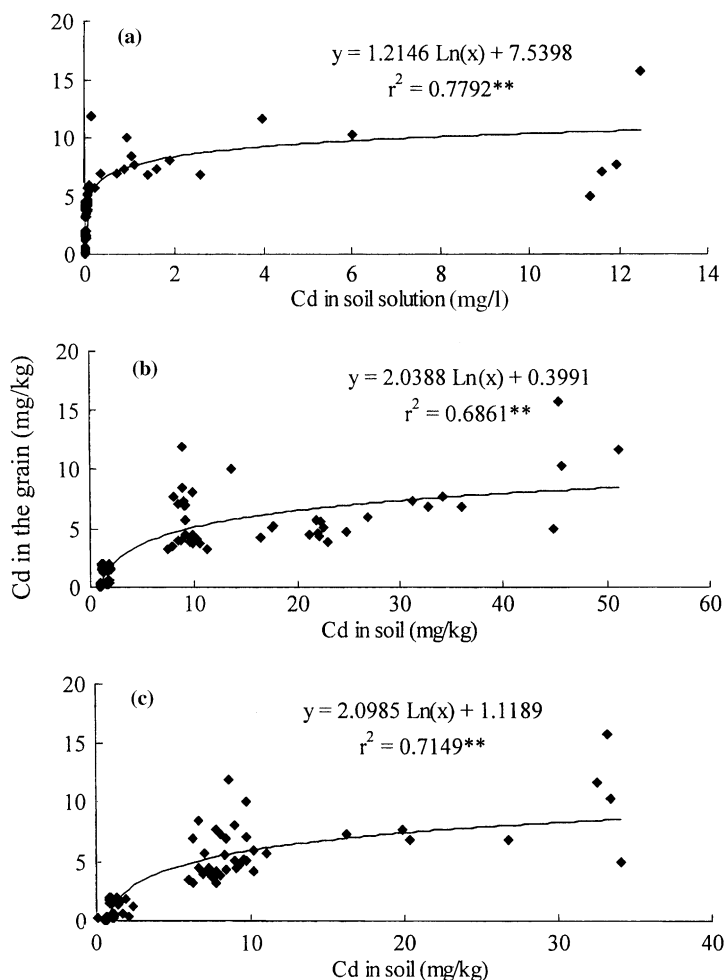


Fig. 1. The relationship between cadmium concentration in the grain of wheat and in soils extracted with different methods including (a) soil solution, (b) EDTA, and (c) DTPA (**: Significant at $p = 0.01$).

treatments because of the low concentration of metals in the studied soils (Table 6).

There was no significant effect of zinc oxide on decreasing the Cd concentration in wheat species. This may be attributed to (a) the wheat specie used in this study may be Zn-efficient plant suggested by Oliver et al. (1996), which was not sensitive to the deficient of Zn and can accumulate more Cd, (b) the tested soils were not deficient of Zn, there was no effect on the application of Zn oxide, and (c) higher content of Cd in the tested soils (1.1–32 mg/kg) compared with normal soils (<0.4 mg/kg), which lead to accumulate more Cd by wheat from the soils. The four tested soils had high content of Cd and were not deficient of Zn for wheat. Therefore, there was no significant reduction on the concentration of Cd in the grain, leaf and stem, and husk of the tested wheat species after applying zinc oxide.

Table 7 showed the total uptake of heavy metal by wheat species for various chemical treatments. We can find that there was no variation in the total Cd uptake among these treatments for highly contaminated Tatan A sandy soil. The addition of these chemical amendments in Tatan A sandy soil did not decrease the total uptake of Cd and Pb in the wheat because of the higher heavy metals concentration caused toxicity to the wheat species and thus decreased the yield of the wheat. The suitable soil pH value for the growth of wheat was ranged from 6 to 7, and the addition of calcium carbonate raised the soil pH from 5.0 to 7.8 in the Tatan A soil and thus increased the biomass of wheat. This increase in the yield diluted the concentration of Cd in the wheat. However, the total uptake of wheat in the Tatan A soil was not changed. In the Tatan B sandy soil, the application of calcium carbonate significantly reduced the total

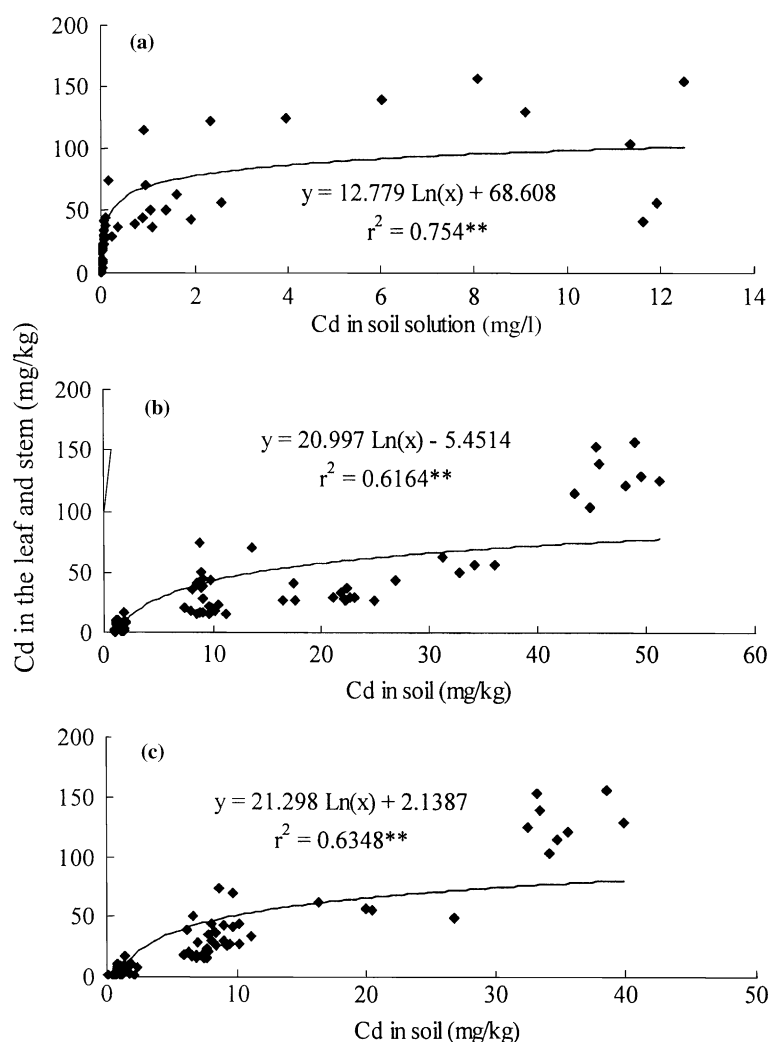


Fig. 2. The relationship between cadmium concentration in the leaf and stem of wheat and in soils extracted with different methods including (a) soil solution, (b) EDTA, and (c) DTPA (**: Significant at $p = 0.01$).

uptake of the Cd from 393 ± 86.0 to $125\text{--}152\text{ }\mu\text{g/pot}$ ($p < 0.01$).

In the Chungfu C and D clayey soils, the application of calcium carbonate (CA), calcium carbonate mixed with zinc oxide (CA + ZN), or calcium carbonate mixed with compost (CA + CO) significantly reduced the total uptake of Cd by wheat ($p < 0.01$) (Table 7). In the four studied soils, the application of chemical amendments had no significantly effect on the reduction of the total uptake of Pb because of the low concentration of Pb in the studied soils.

3.7. The relationship between the concentration of metals in the wheat and in the extracts

Of the three indicators of metal bioavailability used in this study, Cd concentration in soil solution showed the stronger relationship with grain Cd concentration (Fig. 1 and 2). These findings are consistent with many previous studies (Lund et al., 1981; Hornburg and Brümmer, 1986; Kabata-Pendias et al., 1993). Regressing the concentration of Cd below 60 mg/kg extracted with 0.05 M EDTA, Cd concentration in the grain of wheat showed good correlation with that in the soil solution and EDTA or DTPA extracts ($p < 0.01$) (Fig. 1). The results indicated that all these indicators can effectively estimate grain Cd concentration. As expected, the concentration of Cd in soil solution is the best indicator of Cd accumulated in the grain of wheat. The concentration of Cd in the leaf and stem of wheat also showed the same tendency with that of grain (Fig. 2). The concentration of Pb in soil solution and extracts of EDTA or DTPA did not show any significant correlation with the that in the grain, leaf and stem, or husk of wheat when the concentration of Pb extracted with EDTA was below 200 mg/kg (data not shown).

When we regressed the concentration of Cd below 2.5 mg/kg which was extracted with 0.05 M EDTA, the concentration of Cd in the grain of wheat have good correlation only with that in the soil solution ($r^2 = 0.74, p < 0.01$), but not for the extracts of EDTA or DTPA ($r^2 = 0.10$ for EDTA and $r^2 = 0.19$ for DTPA). The concentration of Pb in soil solution and extracts of EDTA or DTPA did not show any significant correlation with the that in the grain, leaf and stem, or husk of wheat. We proposed that when the concentration of Cd extracted with EDTA was below 2.5 mg/kg , only the concentration of Cd in soil solution directly sampled with RSMS can predict that in the grain of wheat.

4. Conclusions

The application of calcium carbonate and calcium carbonate mixed with zinc oxide (or compost) can significantly reduce the concentration of Cd in soil solution

directly sampled with RSMS, and in soil extracted with 0.005 M DTPA or 0.05 M EDTA, or reduce the concentration of Cd in the grain, leaf and stem, husk, or total Cd uptake in all parts of wheat species ($p < 0.01$). The results of this study also indicated that Cd concentration in soil solution and in soil extracted with DTPA or EDTA were effective to predict the Cd concentration or uptake by the wheat, especially for the soil solution.

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