Effect of elemental sulphur on solubility of soil heavy metals and their uptake by maize

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Abstract

A pot experiment was conducted to study the influence of elemental sulphur (S) on solubility of soil Pb, Zn and Cd and uptake by maize (*Zea mays* L.). Two rates of elemental sulphur (S) applied at 0 (S_0) and 200 (S_{200}) mmol kg⁻¹ soil with three rates of each heavy metal at Pb, 0 (Pb_0), 200 (Pb_{200}), 400 (Pb_{400}) mg kg⁻¹ soil, Zn, 0 (Zn_0), 100 (Zn_{100}), 200 (Zn_{200}) mg kg⁻¹ soil and Cd, 0 (Cd_0), 50 (Cd_{50}), 100 (Cd_{100}) mg kg⁻¹ soil, respectively. The result showed that with S application at 200 mmol S kg⁻¹, soil pH decreased about 0.3 unit and the solubility of the Zn and Cd was significantly increased, but the solubility of Pb had no significant influence. The concentration of Pb, Zn and Cd in maize shoots and roots were increased with increasing rates of heavy metals. However, the concentration of Zn and Cd in shoots and roots were higher with application of S rather than without S but no significant difference was found for Pb. The highest concentration of Zn in the shoots was 2.3 times higher with application of S rather than without at the same rate of Zn, 200 mg kg⁻¹. Plant biomass was also significantly affected by the application of S and of heavy metals. With heavy metal addition, the shoot and root biomass were decreased with the rates of those of heavy metals increased either with or without application of S. However, the shoot biomass was significantly decreased with S application at the same rate of heavy metals except that with Zn addition. The removal of Cd and Pb by maize uptake and accumulation with application of S had no significant increase compared to that without, but the removal Zn by maize uptake from the soil increased by application of S, 90.9 $\mu\text{g plant}^{-1}$ contrast to 25.7 $\mu\text{g plant}^{-1}$ at Zn_{200} within a growth period of only 40 days.

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1. Introduction

Soil contamination by heavy metals is an important issue around the world because some metals are very toxic to microorganisms, plants and animals. With a rapid increase in industry and city population in most cities of China, soil contamination has also become an important concern around the country (Wang et al., 2001). There is considerable interest in the development of techniques for remediation of sites contaminated with heavy metals at minimal cost with the least environmental side effects.

In recent years, phytoremediation (especially phytoextraction) has been considered as a cost-effective approach to remediate soil contaminated by heavy metals. The success of phytoremediation depends on several factors, e.g., plant

must produce sufficient biomass while accumulating a high concentration of the certain metal. The metal-accumulating plants also need to be responsive to agricultural practices to allow repeated planting and harvesting of the metal-rich tissues (Blaylock et al., 1997). Therefore, enhancing metal accumulation in existing high yielding crop plants without diminishing their yield is the most feasible strategy in the development of phytoremediation and maize is often used as a metal accumulator in phytoremediation experiments (Huang et al., 1997; Brennan and Shelley, 1999; Bricker et al., 2001).

In the process of phytoremediation, the bioavailability of heavy metals is an important factor. There are two main approaches that have been used to increase the bioavailability of heavy metals in soils and the mobility of heavy metals within plants: lowering soil pH (Salt et al., 1995; Chlopecka et al., 1996; Blaylock et al., 1997) and adding synthetic chelates (Blaylock et al., 1997) such as ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTA) and diethylenetriaminepentaacetic acid (DTPA). A decrease in soil

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pH can be achieved by application of mineral or organic acids or acid-producing fertilizers such as ammonium chloride (Salt et al., 1995; Huang et al., 1997; Wasay et al., 1998). However, these methods have some limitations due to negative effects on the chemical, physical and biological properties of the soil, or may lead to groundwater pollution through leaching. To avoid some of these constraints, the use of elemental S to decrease soil pH and increase the solubility of heavy metals in soils has been suggested (Tichý et al., 1997; Seidel et al., 1998; Kayser et al., 2000). However, more investigations are needed to evaluate the feasibility of this method as a tool for the enhancement of metal solubility and uptake by a metal accumulator, especially in alkaline or neutral soils contaminated by different concentrations of heavy metals. Therefore, the objectives of this study were to (1) compare the metal solubility of soil with different concentrations of Pb, Zn and Cd addition by application of S and (2) investigate the potential to increase metal uptake and accumulation by maize through application of S.

2. Materials and methods

2.1. Soil characteristics

Soil (clay loam) was collected from the top 20 cm of the profile in Qinghe, Beijing, China. The physicochemical properties of the soil before the experiment were: pH 7.7, organic matter content 22 g kg⁻¹, total Pb 130 mg kg⁻¹, total Zn 120 mg kg⁻¹, total Cd 0.05 mg kg⁻¹, CaCl₂-extractable Pb 0.45 mg kg⁻¹, CaCl₂-extractable Zn 0.98 mg kg⁻¹.

2.2. Plants and cultivation

Maize (*Zea mays* L.) seeds were sown at a rate of 4 pot⁻¹ and thinned to leave two seedlings in each pot 1 week after germination. The experiment was carried out in a greenhouse with a 14 h (26 °C)/10 h (13 °C) day/night cycle. Soil moisture content was adjusted regularly by weight to about 60% of water holding capacity and the plants were grown for 40 days.

2.3. Experimental design and treatments

The experimental designs were employed with elemental sulphur (S) and Pb, Zn or Cd. Sulphur was applied and mixed thoroughly with the soil at two rates, 0 (S₀), and 200 (S₂₀₀) mmol kg⁻¹ soil and three heavy metals applied with three rates, Pb at 0 (Pb₀), 200 (Pb₂₀₀), 400 (Pb₄₀₀) mg kg⁻¹ soil, Zn at 0 (Zn₀), 100 (Zn₁₀₀), 200 (Zn₂₀₀) mg kg⁻¹ soil and Cd at 0 (Cd₀), 50 (Cd₅₀), 100 (Cd₁₀₀) mg kg⁻¹ soil, respectively. There were 18 treatments in total and each one with three replicates. The individual metals were added by metal solutions (Pb(NO₃)₂, Zn(NO₃)₂ or Cd(NO₃)₂ after S

was added, the adding concentrations of the three metal solutions were 38.6, 61.2 and 17.8 mmol l⁻¹, respectively. Fertilizer was applied at the rates of 120 mg N (as NH₄NO₃), 35 mg P (as KH₂PO₄) and 100 mg K (as KCl and KH₂PO₄) kg⁻¹ soil before seeding, which was about 12 days after metal contaminants were added in. Each pot received 1 kg of soil that was allowed to equilibrate for a period of 14 days after S was put in then the plant seeds were sown in the greenhouse.

2.4. Sampling and analysis

The soil samples were taken before and after the experiment, then air dried, ground and sieved passing through a 2-mm sieve. Soil pH was measured with subsamples using a 1:2.5 (soil/0.01 M CaCl₂) ratio by a pH meter. For soil extractable metals, 20 ml of 0.01 M CaCl₂ were added to 2.00 g of air-dried soil in a 50-ml polypropylene centrifuge tube. The tube was shaken end-over-end for 16 h at 25 °C, centrifuged at 4000 rpm for 10 min. The supernatant was decanted and saved for Pb, Zn and Cd determination by atomic absorption spectrophotometry (AAS: Z-6100 Hitachi, Japan) and inductively coupled plasma-optical emission spectroscopy (ICP-OES: Optima-2000, Perkin-Elmer, USA) in order to ensure the results are identical (Novozamsky et al., 1993; Degryse et al., 2003). For the total amount of metals, the air-dried soil was ground and sieved by passing through a 0.25-mm sieve and the subsamples were digested with a mixture of concentrated HNO₃/HClO₄/HF (3:1:1) (the concentrations of HNO₃, HClO₄ and HF were 15.6, 11.7 and 19.7 mol l⁻¹, respectively) and then the total concentrations of Pb, Zn and Cd were determined by AAS. After plant harvest, the soil samples were taken, air dried, ground and sieved by passing through a 2-mm sieve. Soil pH, CaCl₂-extractable Pb, Zn and Cd were also determined with the same methods as described above.

All the plants were harvested after the experiment and the plant samples were rinsed with deionized water, oven dried at 70 °C for 48 h, ground with an agate mill, digested with a mixture of concentrated HNO₃/HClO₄/HF (3:1:1 in volume), and analyzed by AAS. The data were subjected to ANOVA and subsequently to Duncan's multiple range test by application of SAS package (version 8.1, SAS, Cary, NC, USA).

3. Results and discussion

3.1. Soil pH

Soil pH decreased in all the treatments with application of S compared to the control (Table 1). One important fact is the oxidation of S by certain groups of acidophilic bacteria, notably *Thiobacillus* spp. in the soil (Lee et al., 1988; Seidel et al., 1998; Tichý et al., 1997; Kayser et al., 2000). Tichý et al. (1997) reported that 156 mmol of different types of

Table 1
Changes in soil pH under different treatments of heavy metals and S application after 14 and 54 days

Treatment	14 Days		54 Days	
	S ₀	S ₂₀₀	S ₀	S ₂₀₀
Pb ₀	7.69	7.53	7.49	7.26
Pb ₂₀₀	7.63	7.46	7.43	7.29
Pb ₄₀₀	7.63	7.42	7.40	7.27
Zn ₀	7.69	7.53	7.49	7.26
Zn ₁₀₀	7.60	7.48	7.46	7.26
Zn ₂₀₀	7.63	7.37	7.46	7.23
Cd ₀	7.69	7.53	7.49	7.26
Cd ₅₀	7.63	7.47	7.49	7.32
Cd ₁₀₀	7.62	7.46	7.49	7.30

Analysis of variance for elemental sulphur (S), metals, time (days (D)) and their interactions

S Lead (Pb), Day		S Zinc (Zn), Day		S Cadmium (Cd), Day	
S level (S)	$P < 0.001$	S level (S)	$P < 0.001$	S level (S)	$P < 0.001$
Pb level (Pb)	$P = 0.01$	Zn level (Zn)	$P = 0.003$	Cd level (Cd)	NS
Day (D)	$P < 0.001$	Day (D)	$P < 0.001$	Day (D)	$P < 0.001$
S*Pb	NS ^a	S*Zn	NS	S*Cd	NS
S*D	NS	S*D	NS	S*D	NS
Pb*D	NS	Zn*D	NS	Cd*D	$P = 0.007$
S*Pb*D	NS	S*Zn*D	NS	S*Cd*D	NS

^a NS stands for no significant difference.

elemental sulphur per kilogram of soil were added to a gleyic cambisol with an initial soil pH between 5 and 5.5, the soil pH decreased to 3.3 within 80 days. Kayser et al. (2000) reported that adding 36 mol S m⁻² to the soil led to a decrease in soil pH from 7.2 to 6.9. In the present experiment, adding S also acidified the soil, which caused the soil pH to decrease about 0.3 unit within 54 days.

3.2. Soil extractable Pb, Zn and Cd

As shown in Table 2, soil CaCl₂-extractable Pb concentrations changed a little with or without S application, but the concentrations of CaCl₂-extractable Zn and Cd markedly increased with soil pH decrease for all the treatments with Zn and Cd addition by application of S. The highest concentration of CaCl₂-extractable Zn was 2.28 mg kg⁻¹ at the treatment of Zn₂₀₀ associated with application of S, 1.8-fold higher than that with the same rate of Zn without application of S. The highest concentration of CaCl₂-extractable Cd was 2.07 mg kg⁻¹ at the treatment of Cd₁₀₀ with application of S, 9.9-fold higher than that with the same rate of Cd without application of S.

The mobile fraction of soil heavy metals is greatly influenced by soil pH and generally increases as soil pH decreases (Kayser et al., 2000; Martinez and Motto, 2000). Kayser et al. (2000) used NaNO₃ to extract metals from soil who found that NaNO₃-extractable Cd and Zn increased 35- and 8-fold, respectively, when soil pH reduced from 7.2 to 6.9. In our experiment, the CaCl₂-extractable fractions of Zn

and Cd increased 0.8 and 8.9 times, respectively, when 200 mmol S kg⁻¹ was applied and when soil pH decreased about 0.3 unit with the treatments of Zn₂₀₀ and Cd₁₀₀. Reddy et al. (1995) found that at low pH, dissolved metal concentration in soil water extracts was dominated by free ionic forms (e.g. Zn²⁺) followed by ion pairs (e.g. ZnSO₄⁰). The availability and mobility of metal ions increased with the soil pH decreased due to these chemical forms increased in soil solutions. Since Pb is a relatively immobile metal in soils due to complexation with organic matter, sorption on oxides and clays, and precipitation as carbonates, hydroxides and phosphates, therefore the concentration of extractable Pb did not show any increase with a decrease in soil pH.

3.3. Shoot and root yield

Plant dry matter biomass was also significantly affected by the application of S and heavy metals (Fig. 1). With heavy metal addition, the shoot and root biomass decreased when the concentration of heavy metals increased either with or without application of S. Compared to the control (without S application), the shoot biomass was significantly decreased with the S application at the same rate of heavy metals except for the treatments with Zn addition. Regarding the root biomass, except for the control and the treatment with Zn addition, it was lower with S application than without. Kayser et al. (2000) reported that shoot weight in some plants did not change significantly with S application. However, in the present experiment, application of S decreased the weight of plant shoots except the treatment of Zn addition. One of the possibilities was that the soil pH

Table 2
Soil CaCl₂-extractable Pb, Zn and Cd (mg kg⁻¹) after maize harvest

Treatment	S ₀	S ₂₀₀
Pb ₀	0.43 (Pb) ^a	0.63 (Pb)
Pb ₂₀₀	0.80 (Pb)	0.79 (Pb)
Pb ₄₀₀	0.81 (Pb)	0.81 (Pb)
Zn ₀	0.99 (Zn)	1.24 (Zn)
Zn ₁₀₀	1.05 (Zn)	1.75 (Zn)
Zn ₂₀₀	1.26 (Zn)	2.28 (Zn)
Cd ₀	BDL ^b	BDL
Cd ₅₀	0.13 (Cd)	0.86 (Cd)
Cd ₁₀₀	0.21 (Cd)	2.09 (Cd)

Analysis of variance for elemental sulphur (S), metals and their interactions

S Lead (Pb)		S Zinc (Zn)		S Cadmium (Cd)	
S level (S)	NS ^c	S level (S)	$P = 0.02$	S level (S)	$P < 0.001$
Pb level (Pb)	$P = 0.077$	Zn level (Zn)	NS	Cd level (Cd)	$P < 0.001$
S*Pb	NS	S*Zn	NS	S*Cd	$P < 0.001$

^a Data showed the concentration of CaCl₂-extractable metal that is in parenthesis.

^b Below detection limit.

^c NS stands for no significant difference.

Fig. 1. Shoot and root biomass of maize influenced by different treatments of heavy metals and S application: (a) shoot and (b) root. $\square$ -S₀, $\square$ -S₂₀₀.

decreased, which may have introduced phytotoxicity by increasing the solubility and bioavailability of the heavy metals. The concentration of Zn addition did not reach the

toxic concentration to the plant. However, another reason might be that different plants have different sensitivity to the toxicity of the metals or the amendment.

Fig. 2. The concentration (mean $\pm$ S.E.*) of Pb, Zn and Cd in shoots and roots of maize. (a) Shoot Pb, (b) shoot Zn and (c) shoot Cd. (A) Root Pb, (B) root Zn and (C) root Cd. $\diamond$ -S₀, $\square$ -S₂₀₀. (*S.E. represents standard error of the means).

3.4. Accumulation of Pb and Zn in plant shoots and roots

The concentrations of Pb, Zn and Cd in shoots and roots increased with the increased application of heavy metals associated with S (Fig. 2). The concentrations of Zn and Cd in the shoots and roots were higher with application of S than without at the same rates of Zn and Cd addition (Fig. 2a and A, b and B), however, Pb concentration in the shoots and roots increased slightly with application of S compared to that without the same treatment of Pb (Fig. 2c and C).

It is well known that Pb is an immobile metal in soil since it readily forms a precipitate with a low aqueous solubility within the soil matrix, and in many cases it is not readily bioavailable, especially in alkaline or neutral soil (Blaylock et al., 1997; Martínez and Motto, 2000). In addition, many plants retain Pb in their roots via sorption and precipitation with only minimal transport to the aboveground harvestable plant portions (Kumar et al., 1995; Huang and Cunningham, 1996; Brennan and Shelley, 1999). In the present experiment, the increase of Pb concentration in maize shoots and roots is lower than that of Zn and Cd.

Compared to without application of S, concentrations of Zn in maize roots and shoots increased by 4.5 and 2.3 times with application of S at the same treatment of 200 mg Zn kg⁻¹ soil (Fig. 2b and B). Concentrations of Cd in maize roots and shoots increased by 2.9 and 2.2 times with application of S at the same treatment of 100 mg Cd kg⁻¹ soil (Fig. 2c and C). There are a number of reasons for this result in high plant uptake and accumulation of these heavy metals; however, the main reason is most probably that the increase of solubility of Zn and Cd leads to an improvement in metal bioavailability with a decrease of soil pH by application of S. There are certain groups of acidophilic soil bacteria in the soil, predominantly the genus *Thiobacillus*, that can oxidize S and change it to SO₄²⁻ when soil pH has decreased (Lee et al., 1988; Tichý et al., 1997; Kayser et al., 2000). McLaughlin et al. (1998) reported that increasing SO₄²⁻ in soil could increase shoot Cd concentration, in the present experiment, the increasing SO₄²⁻ in soil (data were not given) may be another cause that increased the shoot Cd content.

3.5. Removal of heavy metals

Heavy metals removed by maize were shown in Table 3. It is well known that a good phytoextraction method depends on plants producing sufficient biomass while accumulating high concentrations of heavy metals. In the present experiment, the shoot Zn and Cd concentrations increased with the application S at the same treatment of Zn and Cd addition, but the shoot biomass of maize was significantly decreased by application of S and Cd due to the plant sensitivity to the toxicity of Cd. Therefore, the removal of Cd with application of S had no significant impact compared to without S application. The increased

Table 3

Heavy metal content in the shoot of maize (μg plant⁻¹)

Treatment	S ₀	S ₂₀₀
Pb ₀	3.02 (Pb) ^a	2.34 (Pb)
Pb ₂₀₀	18.52 (Pb)	12.67 (Pb)
Pb ₄₀₀	22.40 (Pb)	13.65 (Pb)
Zn ₀	17.16 (Zn)	11.64 (Zn)
Zn ₁₀₀	16.31 (Zn)	46.92 (Zn)
Zn ₂₀₀	25.73 (Zn)	90.89 (Zn)
Cd ₀	0.24 (Cd)	0.18 (Cd)
Cd ₅₀	3.12 (Cd)	3.73 (Cd)
Cd ₁₀₀	5.87 (Cd)	5.66 (Cd)

Analysis of variance for elemental sulphur (S), metals and their interactions

S Lead (Pb)		S Zinc (Zn)		S Cadmium (Cd)	
S level (S)	NS ^b	S level (S)	P=0.002	S level (S)	NS
Pb level (Pb)	P<0.001	Zn level (Zn)	P=0.002	Cd level (Cd)	P<0.001
S*Pb	NS	S*Zn	P=0.009	S*Cd	NS

^a Data showed the content of metal that is in parenthesis.^b NS stands for no significant difference.

solubility of Zn had no negative effect on yield of maize but increased metal accumulation by maize directly translated into equivalent increases in metal removal. As for Pb removal, the decreased shoot biomass and Pb concentration in shoots of maize had no significant effects with the application of S. In fact, it is lower than without application of S (Table 3).

4. Conclusions

Soil pH decreased with S application and the solubility of the Zn and Cd was significantly increased in all treatments with S application. However, the solubility of Pb increased little by the treatment with S. Removal of Zn by maize was significantly increased with the application of S at the same rates of Zn addition, which implied that elemental sulphur can enhance uptake of Zn by maize. Removal of Pb and Cd did not significantly increase with the application of S for the same Pb/Cd-amended soil because maize was sensitive to the toxicity of Pb/Cd, which results in a severe loss of biomass production.

Although the application of S can increase uptake of some metals by plants by enhancing metal mobility, there is also a potential risk of movement of metals into the groundwater; with consequently environmental risk, an appropriate rate of S must therefore be considered in application in the field so that environmental risk can be minimized.

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