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Combined mild soil washing and compost-assisted phytoremediation in treatment of silt loams contaminated with copper, nickel, and chromium

Menghau Sung[a,*], Chi-Yi Lee[b], Suen-Zone Lee[c]

[a] Department of Environmental Science and Engineering, Tunghai University, Taichung 40704, Taiwan
[b] Guan-Cheng Environ. Tech. Protection Co., Ltd., Tainan 71066, Taiwan
[c] Department of Environmental Engineering and Science, Chia-Nan University, Tainan 71710, Taiwan

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ABSTRACT

A new soil remediation option, combining the soil washing process using pure water followed by the compost-assisted phytoextraction, is evaluated using silt loams contaminated with plating wastewater containing Cu, Ni, and Cr. Plants utilized in this study are the rapeseeds, sunflowers, tomatoes, and soapworts. Phytoextraction operation was carried out in pot experiments over a period of 4 months. Metal concentrations in roots and shoots of plants were analyzed upon completion of each pot experiment. Hypothesis testing was employed in assessing the significance of difference in the experimental data. Results indicated that the rapeseed, a hyperaccumulator, is most effective in extracting metals from the compost-amended silt loams. The fast-growing sunflowers and tomatoes are comparable to rapeseeds in accumulating metals despite their relatively low metal concentrations in tissues. Bioaccumulation coefficients obtained for all plants are less than one, indicating that phytostabilization rather than phytoextraction is the dominant mechanism at this simulated final-phase condition.

1. Introduction

Soil contamination by heavy metals has been a serious problem worldwide. Various techniques such as excavation, solidification, stabilization, soil washing, electroremediation, and phytoremediation have been employed to mitigate the impact of heavy metals in the soil environment [1]. Among these techniques, phytoremediation, including phytostabilization and phytoextraction, is considered to be highly cost-effective and not harmful to physical, chemical, and biological characteristics of the soil. In phytoextraction, heavy metals are removed through uptake and subsequent translocation from roots to the above-ground parts of the plants [2,3]. Although phytoextraction is an attractive alternative, it still has drawbacks emerged from recent practices, including the long treatment time to achieve specific objectives [4,5] and unfavorable soil characteristics such as highly acidic soils that can limit its performance [6]. Thus, techniques to shorten the uptake time for plants, and to add amendments to parent soils with the purpose for effective plants growth have become two important tasks in optimizing phytoremediation.

In order to speed up the phytoextraction process, various chelators have been utilized in soils to assist rapid metal translocation from soil to plants [7,8]. Although non-biodegradable chelators such as EDTA (ethylenediaminetetraacetic acid) are effective in increasing phytoextraction efficiency by solubilizing heavy metals, they also cause groundwater pollution problems since the amounts of metals leached out of the root zone are much greater than ones extracted by roots [9,10]. In recent years, less effective but biodegradable chelators such as NTA (nitrilotriacetate) and EDDS (S,S-ethylenediaminedisuccinic acid) have been used as alternatives [7,11]. In addition to chelators, composts have also been added in soils to assist phytoremediation. Their effects on properties of clay soils [12–14] as well as on plants growth [15,16] have both been examined. Compost-assisted phytoextraction in contaminated soils that are inimical to vegetation such as barren soils and mine tailings has also been conducted. For instance, in mine tailings restoration it was found that when the clay loams were amended with composted biosolids, willows growing in the medium demonstrated effective phytoextraction of Mn, Cu, and Cd [17]. Composts can not only supply nutrients to plants, but also create loose and ventilated soils for plants growing in hostile soils.

Metals in soils can distribute in various chemical pools ranging from water soluble, residual, precipitated, to various recalcitrant forms that are bound to carbonate, Fe and Mn oxides, and organic matter [18]. Metals in soluble or weakly adsorbed pools are considered as having higher phytoavailability than those in strongly adsorbed or occluded forms [19]. In fact, in modeling plant uptake of metals from soils, it has normally been assumed that the roots can only absorb ions from soil solution [20]. For adsorbed or precipitated metals, they have to dissolve to the soil solution prior to

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[*] Corresponding author.
E-mail address: mhsung@thu.edu.tw (M. Sung).

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their absorption by the roots. Consequently, in an effective field phytoextraction process the plants would first absorb soluble metals initially present in soil solution, and then extract adsorbed or precipitated metals on the soil surface. At the final-phase of a typical phytoextraction operation, the root system would be in a soil environment of minimum soluble metals. The phytoextraction efficiency at this phase is important since it determines the overall duration of the process. However, our knowledge in this aspect is still lacking since most studies of phytoextraction so far focused merely on the initial stage when soluble metals were readily available.

Soil washing with various chemical solutions has been a popular and effective technique in remediation. However, a follow-up unit designed to treat the final washing solution is always necessary. When a chelator such as EDTA is employed as the washing solution, an intense treatment unit, such as the advanced oxidation process, would be required to breakdown the complex. In this study, we proposed a new green remediation option that combined the soil washing process using pure water and the phytoremediation process. The polluted soils were washed with pure water first to remove soluble metals, and were subsequently phytoremediated. Since no chelators were added, the metal-containing washing solution can be treated by a simple unit such as metal precipitation by alkalization, or by other recent green technologies such as metal sequestration with calcite [21]. The time required to complete the entire remediation is shortened compared to a standard phytoremediation process. For clayey soils that are originally not suitable for plants, natural soil amendments such as composts could be added to assist the plants growth, further making this green remediation option viable to unfavorable soils. Although this is an attractive method, the performance of metal absorption by plants under the environment of minimum soluble metals had never been evaluated.

The goal of this study is to understand the performance of the compost-assisted phytoextraction of contaminated silt loams devoid of soluble heavy metals, obtained by mild soil washing using pure water. The washed soils are assumed to represent the late-phase soil conditions in a typical phytoextraction process. Field silt loams contaminated by plating waste water are used in this investigation. Specific objectives of this study are: (1) to understand the effects of composts on phytoextraction in inimical silt loams, (2) to mimic the phytoextraction performance at the late phase of a phytoextraction process, and (3) to determine the concentration and mass of heavy metals that plants are able to accumulate during the operations. Plants tested in this study include tomatoes (*Solanum lycopersicum*), sunflowers (*Helianthus annuus*), rapeseeds (*Brassica napus*), and soapworts (*Saponaria officinalis*). The first two are frequently used as fast-growing species, the third one is a hyperaccumulator [22], and the last one is a perennial species that has not been tested for phytoextraction before.

2. Materials and methods

2.1. Soils and composts

Field soils used in this study are from a site right next to a plating plant in southern Taiwan. This site is known to contamination with Cr, Ni, and Cu, which are all ingredients used in plating. Collected soil samples were first dried in the air for two weeks, crushed, and sieved through a 2.0 mm screen to remove stones and undesirable debris. After this pretreatment procedure, the soils were put into a tank and deionized water of pH 7 was added as mild washing solution at the soil to water mass ratio of 1:100. An impeller mixer was placed in the tank to start washing soils by mixing. The washing process was completed after 24 h. Then, the soils were separated from the water, dried in the air for another two weeks, and then saved for later pot experiments. The compost was obtained using wasted tea leaves as the main carbon source and swine manure as the nitrogen source by mixing at a mass ratio of 20 to 1. The composting process was carried out in the high rate composter maintaining at 60 °C for 30 days. The mature compost had a C/N ratio close to 9. The compost was dried in air, ground, and then passed through 2.0 mm screen prior to use.

2.2. Pot experiments

Four different test soils were prepared by mixing 0%, 5%, 10%, and 15% of compost, based on dry weight, into the washed soils. The pot used in the experiments had a diameter of 8.5 cm and a height of 7.0 cm. A total of 170 g of dry test soil was used in each pot. The experiments were carried out in triplicate pots. Seeds of sunflowers, tomatoes, and rapeseeds were first cultivated in peat moss for about one to two weeks until their seedlings emerged. Then they were transferred to pots filled with test soils. As to soapworts, they did not start from seeds, but from grown ones by transferring a single root system to each test pot. All prepared pots were placed outdoors regularly, but were kept indoors on rainy days. A total of 15 rainy days was recorded during the growing period of four months. Each pot was watered three times a day with the mist sprayer. The volume sprayed was kept at minimum so as not to cause any significant infiltration. After growing for four months, the whole plant in each pot was taken out of the soil, and the root was cleaned and cut. Subsequently, both the roots and shoots were drying in a 50 °C oven for three days, weighed on the balance, and were then ground into powders for further chemical analyses. Pot experiments using uncontaminated garden soils were also conducted as controls to understand the background accumulation levels of these metals in plants.

2.3. Chemical analysis

Conventional aqua regia digestion method [23] was used to determine concentrations of heavy metals in test soils, roots, and shoots. In digestion, a well-mixed sample of 3 g was first mixed with 28 mL of aqua regia in a 250 mL beaker. Then the beaker was connected to a reflux system, which was kept at the room temperature for 16 h and then under boiling for 2 h to complete the extraction. After the digestion, the supernatant was filtered and the filtrate was analyzed by FAA spectrophotometer (Z-5300, Hitachi Co., Japan) for Cu, Ni, and Cr. For the organic matter determination using the Walkley–Black method [24], 0.5 g of the sample was mixed with 5 mL of 0.5 N $K_2Cr_2O_7$ plus 5 mL of concentrated H_2SO_4 in a 250 mL beaker for oxidation. The concentration of organic matter was then determined by back titration of the remaining $K_2Cr_2O_7$ with 0.5 N ferrous sulphate. The cation exchange capacity (CEC) was determined by the standardized method of extraction with ammonium acetate [25]. The total nitrogen was determined by the Kjeldahl method [26]. The available phosphate and potassium were determined by the colorimetric method [27] and the FAA spectrophotometer [28], respectively. Extractants used for phosphate and potassium were concentrated H_2SO_4 and Bray's reagent, respectively.

2.4. Statistical analysis

Once the means as well as standard deviations of metal concentrations accumulated in plants from the aforesaid triplicates were obtained, the inter-species and intra-species difference between two selected means was statistically assessed. The two-sample *t*-test for independent samples was used for the hypothesis testing. Prior to performing the *t*-test, the equality of the two variances