



# Remediation of lead-contaminated soil using dissolved organic carbon solutions prepared by wine-processing waste sludge



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## ARTICLE INFO

### Article history:

Received 16 January 2014

Received in revised form 1 July 2014

Accepted 5 July 2014

Available online 26 July 2014

### Keywords:

Calcium

Dissolved organic carbon

Lead

Magnesium

Sludge

Soil washing

## ABSTRACT

Wine-processing waste sludge and alkaline substances, including KOH, NaOH, Ca(OH)<sub>2</sub>, and Mg(OH)<sub>2</sub>, were employed to prepare various dissolved organic carbon (DOC) solutions for remediating a Pb-contaminated cropland by soil washing. The Pb concentrations were 5340 and 4920 mg kg<sup>-1</sup> in the topsoil and subsoil, respectively. Aqueous solution treatment, general DOC treatment, and enhanced DOC treatment were proposed as treatments for the reclamation and determination of the factors affecting the removal of Pb, such as pH, initial concentration of the DOC solution, temperature, and washing frequency. The aqueous solution treatment could not remove sufficient Pb and therefore failed in the reclamation. Approximately 86.5% Pb was removed from the topsoil at pH 2.0 by the general DOC treatment undergoing two wash cycles. However, over 90% Pb was removed from the subsoil under the same conditions; only the final Pb concentration in the subsoil met the control standard, 500 mg kg<sup>-1</sup>. About 93% Pb in the topsoil could be removed at pH 3.0 by the enhanced DOC treatment, which was performed using a 2500 mg L<sup>-1</sup> DOC solution containing abundant Ca<sup>2+</sup> and Mg<sup>2+</sup> at an 80:1 liquid/soil ratio (v/v) at 40 °C for 60 min. Meanwhile, organic matter, cation exchange capacity, NH<sub>4</sub><sup>+</sup>-N, as well as exchangeable K, Ca, and Mg contents increased by 125%, 22.6%, 107%, 577%, 55.3%, and 306%, respectively, and the pH in the topsoil decreased to 3.7. The use of Ca(OH)<sub>2</sub> and Mg(OH)<sub>2</sub> not only prevented washing losses of the exchangeable Ca and Mg but also increased Pb removal by 20%. However, its use decreased the availability of P in the soil.

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## 1. Introduction

In Taiwan, illegal metal-processing factories, without any wastewater treatment plant, have extensively discharged plating wastewater into the neighboring canals. Heavy metals, including Pb, Cd, Cu, Zn, Ni, and Cr, are commonly found in the plating wastewater. These toxic metals are generally suspended in the water or accumulated in the sediments of the canals. Water from the contaminated canals is used for irrigation by the farmers; the toxic metals present in the water deteriorate the croplands.

According to the recent literature review, the technologies mostly selected for the remediation of metal-contaminated soils include: (i) metal extraction techniques: soil washing (Dermont et al., 2008a), phytoextraction (Dermont et al., 2008b; Vangronsveld et al., 2009), and electrokinetic remediation (Lageman et al., 2005; Virkutyte et al., 2002); (ii) metal immobilization techniques: Stabilization/solidification, phytostabilization, in-situ chemical stabilization, and bioremediation (Dermont et al., 2008b; Kumpiene et al., 2008; Vangronsveld et al., 2009). Soil washing using chemical extraction techniques (with acids

or chelating agents) has been frequently studied at lab-scale and pilot-scale, but these methods have not been selected for field remediation projects (for metal-contaminated sites) as often as other remediation methods such as excavation and landfill, solidification/stabilization, phytoremediation, electrokinetics, and soil washing using physical separation technologies (CLARINET, 2002; Dermont et al., 2008b; USEPA, 2007). Nevertheless, such soil washing is still considered as a useful method because of highly efficient removal of metals and rapid cleanup of a contaminated site (Gusiatin and Klimiuk, 2012). In fact, acid washing and chelate washing are the two most common methods (Ko et al., 2005). Although strong acids have a high extracting efficiency, it can result in massive nutrient loss and low activity of microbes in the soil. Eventually, the soil productivity shows a severe decline and the restoration of the soil becomes very expensive (Sung et al., 2011). Therefore, remediating the soil at a higher pH should be considered. Some chelating agents have been demonstrated to be effective in removing cationic heavy metals because they form stable anionic chelate complexes (Isoyama and Wada, 2007). EDTA is a popular chelating agent that has been widely used to extract heavy metals from the soil (Zhang et al., 2007). However, its low biodegradability may lead to residual EDTA and metal-EDTA complexes in the soil, which may eventually cause ecological problems (Luo et al., 2006; Yang et al., 2012). Several

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biodegradable washing agents may be considered as the alternatives to replace EDTA, including EDDS and MGDA (Arwidsson et al., 2010a; Hauser et al., 2005), biosurfactants (Mulligan and Wang, 2006), organic metabolites from Fungi (Arwidsson et al., 2010b), and dissolved organic carbon (DOC) solutions (Liu and Chen, 2013; Liu and Lin, 2013; Lee and Chen, 2010). It is believed that employing an improved DOC solution as the agent will have more advantages for soil washing.

Various alkaline mixtures were used to extract the organic constituent from air-dried wine-processing waste sludge (WPWS) for preparation of the DOC solutions used in this study. However, when the component of the alkaline solution dominated the properties of the DOC washing reagent, the soil fertility after washing and metal removal efficiency were affected. For instance, if only NaOH was used to prepare the alkaline solution, a surplus  $\text{Na}^+$  appeared in the washing reagent and the soil became sodium rich after washing. When an excess of alkaline substances was used for the extraction, a substantial amount of soluble base ions were dissolved in the washing reagent, thereby causing a significant increase in the electrical conductivity of the soil. In contrast, though WPWS provided the washing reagent with abundant  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , the loss of exchangeable Ca and Mg was observed during the washing process (Liu and Chen, 2013). This reveals that the amount of WPWS provided was not sufficient to facilitate the recovery of the Ca and Mg contents in the soil back to their original levels. Therefore, the addition of extra Ca and Mg is postulated to improve the washing reagent.  $\text{Ca}(\text{OH})_2$  and  $\text{Mg}(\text{OH})_2$  are thought to serve as a source of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , respectively as well as suitable substitutes of the alkaline substances.

In this investigation, three major methods, termed *aqueous solution treatment*, *general DOC treatment*, and *enhanced DOC treatment*, were proposed to examine the reclamation of highly Pb-contaminated soil. The general DOC treatment, which is a relatively economic operation, was employed to examine the feasibility for remediation of Cd-contaminated and Cu-contaminated soil (Liu and Chen, 2013; Liu and Lin, 2013). However, it was found that the general DOC treatment did not efficiently decontaminate the highly Pb-contaminated soil. The enhanced DOC washing that involved a higher DOC concentration, higher liquid/soil (L/S) ratio, higher reaction temperature, and longer reaction time, was proposed to solve this problem. The objectives of this study were: (i) to assess the feasibility of using  $\text{Ca}(\text{OH})_2$  and  $\text{Mg}(\text{OH})_2$  for substitution of half the KOH, (ii) to identify whether the majority of Pb could be removed from the contaminated soil by the enhanced DOC washing under a higher pH, (iii) to examine the recovery of soil fertility after all treatments, and (iv) to observe the influence of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  upon reclamation of the soil.

## 2. Materials and methods

### 2.1. Preparation of soil sample and fertility analysis

Soil samples were obtained from a pedon in a fallow rice paddy with serious Pb pollution, located in Luchu, Taoyuan County, Taiwan (TWD97: 276114, 2767129). Approximately 10 kg of soil was collected from topsoil layer (0–15 cm in depth) and subsoil layer (15–30 cm in depth) by an auger. After being air-dried and ground, the samples were passed through a 2.0 mm sieve and stored in plastic cans at room temperature. The Pb concentrations were analyzed by a flame atomic absorption spectrometer (AAS, GBC 932 Plus, Australia) after digestion in aqua regia solution (1:3 conc.  $\text{HNO}_3$ :conc. HCl ratio v/v). Soil texture was determined by the pipette method as described by Gee and Bauder (1986). Soil fertilities before and after the washing treatments are respectively termed *original fertility* and *final fertility* in this study. Several examined fertility indices include pH; organic matter; cation exchange capacity; electrical conductivity; available ammonium; available phosphorus; exchangeable potassium, calcium and magnesium; exchangeable sodium percentage (ESP); and  $\text{Ca}^{2+}/\text{Mg}^{2+}$  ratio. Soil pH was measured at a 1:1 soil–water ratio (w/v) with a pH meter (SUNTEX

TS-110). Total organic matter was determined by the wet dichromate oxidation method (Nelson and Sommers, 1982). The electrical conductivity (EC) values were measured at a 1:5 soil–water ratio (w/v) with an electrical conductivity meter (HACH HQ14d). Available ammonium ( $\text{NH}_4^+-\text{N}$ ) was extracted from the soil at a soil solution ratio of 1:5 using 1 M KCl for 1 h, and its concentration was determined by the titrimetric distillation method (EPA Method 350.2, U.S.). Available P was extracted using the Bray-1 method and determined by the molybdenum blue colorimetric method (Okalebo et al., 2002). Exchangeable base ( $\text{K}^+$ ,  $\text{Na}^+$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ ) and cation exchange capacity (CEC) were obtained by a conventional ammonium–sodium exchange method at pH 7 (Gogo et al., 2010). The ESP was calculated as the ratio of exchangeable Na to the CEC. The  $\text{Ca}^{2+}/\text{Mg}^{2+}$  ratio was calculated by dividing the exchangeable Ca by exchangeable Mg.

### 2.2. Preparation and characterization of the dissolved organic carbon solutions

Three types of alkaline mixture solutions were previously formulated to extract the DOC from WPWS samples for preparation of the washing reagents (Table 1). The first solution, used to prepare reagents B and C, was prepared by mixing 0.5 M KOH and 0.5 M NaOH in a 4:1 ratio (v/v). The second solution, used to prepare reagent D, was prepared by mixing 0.8 M KOH, 0.2 M NaOH, a saturated  $\text{Ca}(\text{OH})_2$  solution, and a saturated  $\text{Mg}(\text{OH})_2$  solution in a 1:1:1:1 ratio (v/v). In this case,  $\text{Ca}(\text{OH})_2$  and  $\text{Mg}(\text{OH})_2$  substituted half of KOH as used in the first alkaline mixture solution for extraction of the DOC. However, visible precipitations occurred in both the  $\text{Ca}(\text{OH})_2$  solution (50.32 g  $\text{L}^{-1}$ , equal to 0.68 M) and the  $\text{Mg}(\text{OH})_2$  solution (18.66 g  $\text{L}^{-1}$ , equal to 0.32 M) owing to their low solubility in water; the utilization ratio of  $\text{Ca}(\text{OH})_2$  to  $\text{Mg}(\text{OH})_2$  was decided according to the washing results after treatment with the reagent C. The third alkaline mixture solution, used to prepare reagent E, was prepared by mixing 0.53 M KOH, 0.13 M NaOH, a saturated  $\text{Ca}(\text{OH})_2$  solution, and a saturated  $\text{Mg}(\text{OH})_2$  solution in a 1:1:1:1 ratio (v/v). The saturated solutions were obtained from the supernatants of a  $\text{Ca}(\text{OH})_2$  solution (33.30 g  $\text{L}^{-1}$ , equal to 0.45 M) and an  $\text{Mg}(\text{OH})_2$  solution (11.66 g  $\text{L}^{-1}$ , equal to 0.2 M). The amount of moles of  $\text{K}^+$ ,  $\text{Na}^+$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$  in this alkaline mixture solution were only two-thirds of that in the second alkaline mixture solution. The precipitated  $\text{Ca}(\text{OH})_2$  and  $\text{Mg}(\text{OH})_2$  solids were collected for preparation of the following reagent. The procedure of extraction and preparation of dissolved organic carbon solutions were very similar to those in an earlier study (Liu and Chen, 2013). Whole  $\text{Ca}(\text{OH})_2$  and  $\text{Mg}(\text{OH})_2$  solids collected previously were added into reagents D and E as their acidities were adjusted to pH 3.0. The solids entirely dissolved under these conditions, and consequently both reagents D and E became rich in  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ .

Five types of solutions with different conditions were employed as the washing reagents for this investigation (Table 1). Both reagent A (aqueous solutions) and reagent B (1500 mg  $\text{L}^{-1}$  of the DOC solutions) at various pH values (2.0, 3.0, 4.0, 5.0, 6.0, 7.0, 8.0, 9.0, 10.0, 11.0, and 12.0) were prepared for optimizing the experimental conditions. The washing experiments of the soil were conducted twice only for the reagents at pH 2.0 and 3.0, in an attempt to remove more Pb and achieve the remediation target. Reagents C, D, and E, having high concentration (2500 mg  $\text{L}^{-1}$ ) at pH 3.0, were also prepared for removing more Pb, but under a less acidic condition. Both reagents D and E were prepared in order to change the composition of the DOC solution, which was used in the reagent C treatment; their uses would decrease the  $\text{Na}^+$  content and increase the  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  contents in soil. In addition, the use of reagent E would diminish the content of the soluble salts in the soil after washing. Experiments with reagents C, D, and E (enhanced DOC treatment) were conducted with a higher L/S ratio (80:1), higher operational temperature, and longer reaction time in comparison with both reagents A (aqueous solution treatment) and B (general DOC treatment). These factors facilitated the removal of metals from the soil. The EC values and

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