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The interactive effects between chelator and electric fields on the leaching risk of metals and the phytoremediation efficiency of *Eucalyptus globulus*

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ABSTRACT

The single and combined effects of phyto and electro remediation to decontaminate metal contaminated soil were evaluated in this study. Leaching risks of metals from top soil to groundwater after chelator application were compared under different conditions. Soil from Guiyu (China), a notorious e-waste recycling centre, was remediated by different methods including chelator addition, electric field application and their combination. Ethylene diamine tetraacetic acid (EDTA) was applied as chelator. In electrokinetic and combined remediation experiments, stainless steel sheets were adhered to stems to charge the plant negatively. Chelator can significantly enhance the plant's ability to absorb metals and impel the metals moving from top to deep soil layers. Individual effects of electric field with varied voltages on phytoremediation efficiency were different. The ability of plants to accumulate metals increased with the increase of voltages. The biomass production of plants was enhanced at low (2 V) and moderate (4 V) voltages and decreased at high (10 V). Metal leaching in electrokinetic remediation processes was less significant than in EDTA treatment and the control. Phytoremediation efficiency improved by combining chelator and electric field was up to twice as much as by individual chelator. These results indicated that phytoremediation assisted by electric field is a potential technology to decontaminate large scale fields.

1. Introduction

With the rapid growth of human population, urbanization and manufacturing industries, all kinds of pollutants have been released to soil, atmosphere and aquatic system (Lotfy and Mostafa, 2014). In recent decades, exposure to heavy metals has gained particular attention worldwide because they are not biodegradable and can finally be taken up by human through food chains (Azad et al., 2018). Geologic processes including magmatism, metamorphism, deposition, weathering and erosion determine the concentrations and spatial-temporal distribution characteristics of metals at regional scale (Falkowska et al., 2016), while anthropogenic activities including mining, fertilization, transportation and waste disposition can gather metals within a range (Busico et al., 2017). Electronic waste (e-waste) dismantling is one of the most pernicious industries among anthropogenic activities because a great amount of contaminants are released into the surrounding environment during the recycling process (Liu et al., 2018; Polat et al., 2018). Soil can be regarded as a contaminant sink because it has strong self-purification capacity (particulate matter in soil can adsorb toxins, microorganism degrade organic contaminants and amphoteric colloid neutralize acidic or alkalinity substances), but it becomes a crucial contaminant source when pollutants overwhelm its buffering ability (Song et al., 2018).

Other than conventional remediation methods including excavation, chemical leaching, vitrification and solidification which can destroy the structure and biodiversity of the soil irreversibly (Luo et al., 2015), phytoremediation can remove contaminants from soil without disruption (Rodrigues et al., 2016).

However, this environmentally friendly remediation technique still has some deficiencies. Compared to traditional methods, the

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major drawback of phytoremediation is that it is time-consuming. The critical factors of successful phytoremediation are biomass production and pollutant absorption (Pandey et al., 2015). Hyperaccumulators which can tolerate and accumulate plenty of specific contaminants cannot effectively produce sufficient biomass to extract metals from contaminated soil (Van der Ent et al., 2013), while it is difficult for high biomass producing species to mobilize and absorb metals in soil (Redondo-Gómez, 2013).

The application of chelator to improve metal bioavailability is a universal practice to promote phytoremediation efficiency (Wang et al., 2007). The disadvantages of chelator addition are obvious, i.e. plants cannot accumulate instantly mobilized metals in the soil solution in time and therefore soluble metals would leach into deep soil layers and groundwater (Lim et al., 2012). Previous studies reported the transfer, accumulation and transformation of metals and their fractions in soil and plant tissues (Cang et al., 2011; Giannis et al., 2008). However, published studies concerning phytoremediation assisted by electric field have mostly focused on plant growth promotion (Bi et al., 2010), metal accumulation (Lim et al., 2012), varying physical, chemical and biological property of the soil (Cang et al., 2012), and improvement of decontamination effect (Bi et al., 2011) during the remediation process. Only Zhou et al. (2007) discussed the effect of vertical electrical field on alleviating the leaching loss of Cu and Zn in chelator assisted phytoremediation processes, which was attributed to increased extraction capacity of ryegrass induced by electric field. As electric field can be considered as the force to drive soluble metals towards the opposite charges, it is safe to assume that counter gravitational transport of metal rich solution can be generated by electroosmotic flow, resulting in alleviation of leaching risks caused by chelator (Tahmasbian and Sinegani, 2016).

Based on the above assumption, a series of pot experiments were designed (1) to assess the individual effect of chelator and electric field as well as their interactive effects on phytoremediation efficiency; (2) to evaluate the alleviation effect of electric field on solution leaching and metal loss caused by chelator application during the remediation; and (3) to develop the optimum methodology to achieve higher electro-phytoremediation efficiency. An economical and feasible remediation technique for real scale field can be established on the basis of this study.

2. Materials and methods

2.1. Soil sampling and preprocessing

Soil in the experiment was collected from the agricultural land of Guiyu, a notorious electronic waste (e-waste) dismantling and recycling centre in Eastern Guangdong, China. According to previous studies, Guiyu can be divided into five functional areas including e-waste recycling, open burning, wasteland, cultivation and residential, each with different pollution levels; for example, the e-waste recycling zone was moderately to heavily polluted whilst the cultivation area was only lightly polluted (Luo et al., 2016; Quan et al., 2014). The pollution concentrations reduced with distance from e-waste dismantling areas (Wong et al., 2007). The soil was ground and sieved through a 4 mm mesh to remove coarse material and plant debris after which it was air dried and mixed thoroughly three times for homogeneous distribution of metals.

Bahadur et al. (2017) and Lacalle et al. (2018) suggested that the equilibrium between available and stable fractions of metals was important for phytoremediation efficiency evaluation, especially in pot experiment phase, because the success of remediation was judged on the point to point basis instead of the mean values of data obtained from the experiment. It was reported that the equilibrium between available and stable fractions of metals in soil would be achieved within 15–30 days. The initial metal concentrations of the soil sample were analyzed after 3 weeks' homogenization for metal equilibrium. Soil pH values were determined with a pH meter by inserting the electronic probe into the mud (mixture of soil and aqua destillata at a volume ratio of 1:2.5); ferrisulfates was applied to titrate total organic carbon (TOC) in soil oxidized by $K_2Cr_2O_7$ (0.8 mol L^{-1}). Cation exchange capacity (CEC) of the soil was titrated using HCl after cations were exchanged with ammonium acetate (25 mL, 1.0 mol L^{-1}). The soil pH, CEC and TOC were 6.3, 14.6 cmolc kg^{-1} and 48.3 g kg^{-1}, respectively. Cadmium, Hg, Pb, Cu, Cr and Zn of the soil were 0.79, 0.26, 89.2, 68.2, 61.33 and 128.6 mg kg^{-1}, respectively. Concentrations of Cd, Pb and Cu exceeded the acceptable threshold limits of 0.3, 80 and 50 mg kg^{-1} for agricultural land (CNEPA, 2008; GB15618-2008). Contents of other metals were considerably lower than the permissible criterion.

2.2. Plant choosing

Using hyperaccumulators to remediate metal polluted soil has obtained extensive attention and made some achievements in small to moderate scale experiment (Bian et al., 2017). It takes years to centuries for hyperaccumulators to clear up metal influenced soil, depending on pollution levels, metal fractions and plant species (Bi et al., 2011; Gao et al., 2012). However, some limits like low biomass yield and disposition of harvested tissues restricted the wide use of hyperaccumulators, besides which, Li et al. (2018) suggested that hyperaccumulators mobilized all metals through acidifying the soil but could hyperaccumulate only particular elements, and therefore increased the environmental risk. To overcome the drawbacks of hyperaccumulator, productive trees like salix (Salam et al., 2016) and poplar (Zarubova et al., 2015) were utilized for soil detoxification because their high biomass yield can make up the relatively low metal contents. *Eucalyptus globulus* was chosen in this study as it grew well on marginal lands (Salas-Luevano et al., 2017).

2.3. Experimental design

The experiments were performed in a greenhouse with a controlled 16/8 h day/night cycle and corresponding temperature of 26/18 °C. Eight treatments were arranged randomly in 40 cylindrical transparent PVC containers (20 cm in diameter and 80 cm in height) in five replicates (Fig. 1). The mixed 32 kg soil was filled into each container with a height of 75 cm. After 6 months of

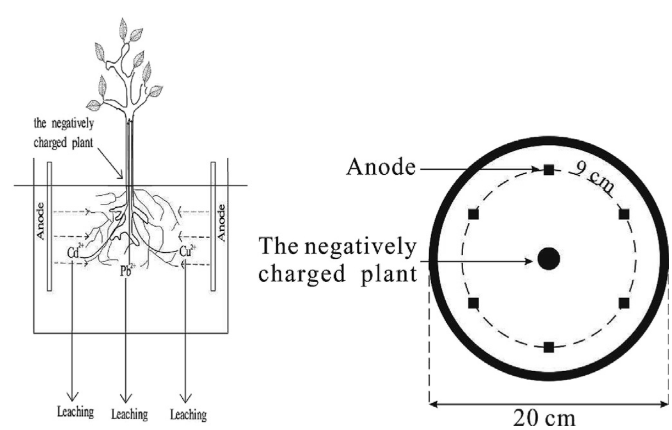

Fig. 1. Schematic of electric field assisted phytoremediation.