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Research article

A multi-technique phytoremediation approach to purify metals contaminated soil from e-waste recycling site

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ABSTRACT

Multiple techniques for soil decontamination were combined to enhance the phytoremediation efficiency of *Eucalyptus globulese* and alleviate the corresponding environmental risks. The approach constituted of chelating agent using, electrokinetic remediation, plant hormone foliar application and phytoremediation was designed to remediate multi-metal contaminated soils from a notorious e-waste recycling town. The decontamination ability of *E. globulese* increased from 1.35, 58.47 and 119.18 mg per plant for Cd, Pb and Cu in planting controls to 7.57, 198.68 and 174.34 mg per plant in individual EDTA treatments, respectively, but simultaneously, 0.9–11.5 times more metals leached from chelator treatments relative to controls. Low (2 V) and moderate (4 V) voltage electric fields provoked the growth of the species while high voltage (10 V) had an opposite effect and metal concentrations of the plants elevated with the increment of voltage. Volumes of the leachate decreased from 1224 to 134 mL with voltage increasing from 0 to 10 V due to electroosmosis and electrolysis. Comparing with individual phytoremediation, foliar cytokinin treatments produced 56% more biomass and intercepted 2.5 times more leachate attributed to the enhanced transpiration rate. The synergistic combination of the individuals resulted in the most biomass production and metal accumulation of the species under the stress condition relative to other methods. Time required for the multi-technique approach to decontaminate Cd, Pb and Cu from soil was 2.1–10.4 times less than individual chelator addition, electric field application or plant hormone utilization. It's especially important that nearly no leachate (60 mL in total) was collected from the multi-technique system. This approach is a suitable method to remediate metal polluted site considering its decontamination efficiency and associated environmental negligible risk.

1. Introduction

Concerns on growing occurrence of contamination accidents in various media including atmosphere, aquatic, soil, livestock and crops with rapid industrialization and urbanization increased in recent years (Laidlaw et al., 2015; Stingu et al., 2012). Comparing to other contaminants, heavy metals can cause long term environmental influence due to their non-biodegradable characteristic (Bennedsen et al., 2012). The main natural sources of heavy metals are mineral weathering, soil erosion and volcanism. Distribution and composition of metals derived from human activities mainly depended on their sources including ore mining, ironware smelting, equipment manufacturing, traffic emission, waste disposing and agricultural management (Ginneken et al., 2007; Zhao et al., 2015). Electric waste (e-waste) dismantling and disposing is one of the most concerned issues among anthropogenic sources because a surprisingly large amount of inorganic and organic contaminants were generated and spread during e-waste recycling processes (Quan et al., 2014; Tue et al., 2014).

Relative to conventional treatments which can rapidly remediate soils but irreversibly damage the ecological system and the agricultural function of substrates (Cameselle et al., 2013; Heckenroth et al., 2016), including incineration, landfill, chemical washing, physical covering, solidification and vitrification, phytoremediation is a solar driven and environmentally friendly technique because it decontaminates contaminants along with the metabolism processes of species without disturbing the physical, chemical and ecological characteristics of soils (Mojiri et al., 2016; Salam et al., 2017). Low survival and biomass production of plants

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grown on contaminated soils and long time required for species to remove exceeding contaminants from substrates were the two major factors restricting the success of phytoremediation (Luo et al., 2016; Niazi et al., 2012).

Various assisting techniques including gene modification (Bech et al., 2002; Gomes et al., 2016), growth facilitating rhizosphere and endosphere bacteria inoculation (Fatima et al., 2015; Teixeira et al., 2014), chelating agent application (Guo and Cutright, 2015; Luo et al., 2016) and electric field assistance (Lim et al., 2012; Mao et al., 2016) were used to enhance the efficiency of phytoremediation, but with problems like plant growth suppression (Kim and Lee, 2010), groundwater pollution (Martinez-Alcala et al., 2016) and indigenous species disturbing (Marques et al., 2013; Ranjard et al., 2006). EDTA treatment increasing the bioavailability of various elements and electric field application stimulating the growth of plants were widely accepted supplementary means for phytoremediation, despite of some imperfection (Cui et al., 2015; Lim et al., 2012). Comparing with direct soil treatment, cytokinin application can improve the transpiration rate and growth ability of plants without direct chemical reaction with soil matters and consequently, increase phytoremediation efficiency (Cassina et al., 2011).

In the present study, a multi-technique phytoremediation approach was designed for heavy metal decontamination from polluted soils. Methods with minimum interference with the soil system which generated inverse gravitational migration of metal ions including electro-osmosis production and transpiration rate increase were developed and combined to elevate the phytoremediation efficiency and alleviate the leaching risk. The main objectives of this study are to evaluate (1) the influences of individual and combined methods on phytoremediation; (2) the synergistic effects of chelator, electric field and cytokinin on metal leaching prevention; and (3) the mechanism associated with phytoremediation.

2. Materials and methods

2.1. Soil description

The soils were collected from the agricultural land still used for crop cultivation in Guiyu, Guangdong Province, a notorious electronic waste (e-waste) disposing and reclamation capital. Seven hundred surface soils (20 cm) were sampled from a 10 m × 10 m grid and sieved through 2 mm meshes after air dried and crushed. The pretreated soil was thoroughly mixed once a week for three cycles to eliminate the heterogeneous distribution of metals impacting the phytoremediation efficiency evaluation as the remediation assessment was generally on a point-by-point basis (Luo et al., 2016). The initial soil metal concentrations were analysed to calculate the time consumption of phytoremediation for exceeded contaminant decontamination.

2.2. Experimental design

Experiments were performed in a fully controlled greenhouse installed with fluorescent lamps with a photoperiod of 8/16 h dark/light and corresponding temperature of 18/26 °C. Eight experimental conditions (Table 1) were randomly designated to 40 cylindrical PVC containers with 20 cm in diameter and 80 cm in height (five replicates). Thirty-two kg of the completely mixed soil was padded in each pot to 75 cm for similar soil density. *Eucalyptus globulus*, recommended as a candidate to phytoremediate multi-metal polluted soil (Arriagada et al., 2007), was selected in the present study, although its metal concentrations were lower than conventional hyperaccumulators in unit biomass. In real scale field, *E. globulus* can accumulate more metals from soil relative to known hyperaccumulators including *Brassica juncea*, *Lupinus albus* and *Thlaspi caerulescens* because the low metal concentrations in unit biomass can be compensated by its large biomass yield (Peng et al., 2012). Developed *E. globulus* with similar appearance grown on the clean place were transplanted to containers except the bare controls.

Table 1
Experimental designs.

	E1	E2	E3	E4	E5	E6	E7	E8
Planting	–	+	+	+	+	+	+	+
EDTA	–	–	+	–	–	–	–	+
DC field	–	–	–	2 V	4 V	10 V	–	4 V
cytokinin	–	–	–	–	–	–	+	+

Experiment E1 was bare control. Experiment E2 was individual phytoremediation process. In experiment E3, appropriate content of EDTA (0.5 mmol L^{-1}) for metal bioavailability increase in phytoremediation (Cui et al., 2015) was applied with a single dose of Na_2-EDTA (250 mL) to the soil surface using a syringe after *E. globulus* were transplanted. In experiments E4 to E6, direct-current electric fields with different voltages (2, 4 and 10 V) were applied on planting containers and sustained for two weeks (6 h per day). Six identical graphite electrodes (1 cm in width and 65 cm in height) were set with equal interval on the vessel circumference as anodes while the species was negatively charged using a stainless-steel sheet sticked on the aerial part of the plant just above the soil surface. E7 was plant hormone foliar application experiment; 20 mg cytokinin kg^{-1} was sprinkled on the whole leaves of *E. globulus* every 7 days for two cycles. After the termination of the above 6 experiments, the optimal voltage (4 V) for biomass production and metal accumulation of the plant was assured by E3 to E5 and chelator and cytokinin were combined to constitute a multi-technique approach (E8) with the same time schedule of each individual experiment. All assisted methods were applied after 30 days of species transplant.

To simulate a precipitation of 25 mm per day, 785 mL of deionized water was sprayed on the soil surface of each container 1 h after the above assisted methods were applied for 14 days.

2.3. Sample analysis

Soil samples were collected from each container around the plant to confirm the homogeneity of the substrate just after the *E. globulus* transplant. Plants were harvested and segmented to roots and aerial parts at the end of the experiment. The species was shaken slightly to strip the loose attachments and then washed by running water. To eliminate the tightly bound material, the plants were scrubbed using deionized water. The cleaned species were dried to constant weight at 80 °C.

The dried soil and species samples were pulverized and then sieved with 200 meshes. The powders of treated samples were digested using concentrated HCl and HNO_3 mixture at a volume ratio of 3:1 for 45 min in a crucible at 150 °C. The solution was mixed with 10 mL of hydrogen peroxide after cooling and heated to nearly dry, followed by a 20 mL of 1% HNO_3 dilution. The diluted solution was heated to boiling and lasted for 10 min, and then diluted to 50 mL precisely using distilled water. The prepared solution was filtered with polyethersulfone membrane filters for further analysis. Inductively coupled plasma-mass spectrometry (ICP-MS) was performed to determine the contents of Cd, Pb, Hg, Cr, Zn and Cu.

The leachate collected at the bottom of each container, if any, was stored in polyethylene plastic bottles after volume measurement. After shaked and filtered with No. 42 Whatman filter paper,