Phytomanagement of a Chromium-contaminated Soil by a High-value Plant: Phytostabilization of Heavy Metal Contaminated Sites

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Phytoremediation of metal-contaminated soil can be an eco-friendly technology. However, relatively long cultivation times impedes its popularization on a commercial scale. This study evaluated the effectiveness of lavender plants (*Lavandula dentata* L.) to remediate a highly chromium (Cr)-contaminated site through a pot experiment. The lavender growing soil was mixed both with and without biochar (2.5% w/w) + oyster shell waste (2.5% w/w) and biochar (2.5% w/w) + citrus peel waste (2.5% w/w). The results indicated that Cr(VI) accounted for 19.0% to 4.7% of the total soil Cr, while Cr(III) accounted for 81.0% to 95.3%, from the beginning to the end of the cultivation. The water-soluble Cr concentration decreased from 44.6 mg/kg to 7.5 mg/kg. The biomass of the lavender growing in the contaminated soil decreased by factors in the range between 4-fold and 6-found.The addition of soil amendments significantly reduced the (potential) bioavailable Cr ($p < 0.05$) in the range of 2 to 3 fold, consequently improving the growth of lavender in the highly toxic soil. In addition, the soil amendments significantly reduced the Cr bioaccumulation and the translocation from the roots to the shoots. These results showed that the cultivation of lavender with suitable amendments can effectively be used for phytomanagement techniques in highly contaminated soil.

Keywords: Chromium; Phytoremediation; Lavender; Biochar; Oyster shells

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INTRODUCTION

Soil contamination is a global problem with potential risks to human health and ecosystem stability (Carré *et al*. 2017). Pollutants in the food chain (*e.g.*, soil-plant-human, soil-plant-animal-human, or contaminated ground water) adversely affect human health globally, with harmful effects on soil ecosystem services (Morel *et al*. 2015; Zhao *et al*. 2015; Arslan *et al*. 2016). The remediation of contaminated soils is crucial to reduce associated risks and to enhance food security (*i.e.*, food quality and quantity). Soil remediation based on physico-chemical methods is generally expensive and often results in soil deterioration. Therefore, eco-friendly technologies, such as phytoremediation, have been developed (Ali *et al*. 2013; Ding *et al*. 2018).

Phytoremediation comprises bioremediation processes that use living green plants to remove, transfer, stabilize, and/or destroy contaminants in the soil. Such methods are widely evaluated as green and cost-effective environmental restoration technologies (Wang *et al*. 2018; Yang 2018). The principles of phytoremediation is to decrease the labile pool of contaminants and to reduce the pollutant linkages that are based on two main strategies, namely phytostabilization and phytoextraction. More than 500 plant species are highlighted as the most promising hyperaccumulators. Many phytoremediation studies have been performed in the last few decades, resulting in progressive research and the development of the phytotechnologies, including innovative approaches and paradigms (Agnello *et al*. 2014; Antoniadis *et al*. 2017). The advantages of phytoremediation (*i.e*., green technology and low costs) have led to high public acceptance; generally, these methods are considered aesthetically pleasing and environmental friendly management strategies for the remediation of contaminated soils (Ali *et al*. 2013; Sarwar *et al*. 2017). However, the limitations of phytoremediation, such as the long periods required for effective metal removal, particularly in moderately and highly contaminated sites, need to be considered (Conesa *et al*. 2012; Burges *et al*. 2018). Several factors, including plant life cycles (from planting or transplanting to harvesting, namely a growth season), low plant biomass and growth, along with bioavailability and bioaccessibility of metals and metalloids in the soil, can extend the removal period (Zayed and Terry 2003; Ali *et al*. 2013). Site characteristics, such as soil properties, mixed contamination, and climatic conditions, can also considerably affect the remediation efficiency (Mendez and Maier 2008). Agricultural activities, such as fertilization and field management, can enhance the growth of phytoremediation plants and therefore stimulate the remediation capacity (Chen and Cutright 2002; Wei *et al*. 2010; Tang *et al*. 2017). Technologies, such as the use of chelating agents, transgenic plants, and soil amendments, can also be suitable options to overcome the disadvantages (Evangelou *et al*. 2007; Yadav *et al*. 2009; Zhang *et al*. 2013). Generally, the strategies to increase phytoremediation efficiencies involve logistical difficulties in large-scale applications, mainly because of additional costs and potential environmental risks (*e.g*., leaching into the groundwater), hampering the deployment of such methods on commercial scales (Conesa *et al*. 2012; Mahar *et al*. 2016).

Chromium (Cr), particularly in its hexavalent form, is one of the widespread heavy metals that causes serious environmental problems in soil and groundwater (Ertani *et al*. 2017; Shahid *et al*. 2017). The trivalent (Cr(III)) and hexavalent (Cr(VI)) forms are the stable chemical forms found in the environment, with Cr(VI) causing the greatest concern because of its toxicity to all forms of life (Ertani *et al*. 2017). Additionally, Cr(VI) is more water-soluble and more mobile than Cr(III) in soil and therefore has a comparatively high soil-plant transfer index (Han *et al*. 2004; Shahid *et al*. 2017). Numerous studies have been conducted with several plant species to evaluate the phytoremediation of Cr-contaminated soils (Shahandeh and Hossner 2000; Sinha *et al*. 2018). These species include both native genotypes and agronomic species. For example, *Portulaca oleracea* (an annual, succulent herb) is considered a prospective plant for the phytoremediation of Cr-contaminated sites, and *Zea mays* L. shows high tolerance towards Cr with enhanced efficiency of Cr(VI) phytoextraction when amended with phosphate (Kale *et al*. 2015; Gheju and Balcu 2017). However, most of these plants require regular and precise management practices to promote the phytoremediation efficiency. As a result, sufficient financial support is crucial to establish the phytoremediation technology in the initial stage. In most parts of the world, funding for remediation projects, particularly for the rehabilitation of contaminated farmland is scarce (Qu *et al*. 2016). Therefore, involving the public in soil remediation programs can be a viable alternative to mitigate soil pollution, particularly in remote and less developed regions (Conesa *et al*. 2012; Gupta *et al*. 2013). Such an approach represents an opportunity for the public to obtain economic benefits. Plant species that can provide financial returns (*e.g.*, energy crops, cash crops, potential biochar sources, and medicinal plants) are currently being evaluated in phytomanagement studies (Pandey *et al*. 2016; Thijs *et al*. 2017; Venkatachalam *et al*. 2017; Verma *et al*. 2017). However, the relatively low economic benefits are still an obstacle for public involvement in the rehabilitation of contaminated areas. In this context, this study used lavender (*Lavandula dentata* L.) for the phytoremediation of a soil polluted with high levels of chromium. Lavender is not a hyperaccumulator. Rather, it is a commercially grown plant. Accordingly it ordinarily is not used for extracting chromium from soil, but for stable management of chromium pollutants in soil. Specifically, whether this high-value plant can be successfully cultivated in such a site was tested. The selection of this plant species was based on its use for producing an essential oil that is frequently used in soaps, detergents, and cosmetics. Studies have shown that the essential oils produced from plants in metal-contaminated soils are generally free of heavy metals (Zheljazkov *et al*. 2006; Gupta *et al*. 2013; Verma *et al*. 2017). Agricultural wastes (or their derived products), such as oyster shell waste, citrus peel waste, and biochar, are used as soil amendments to reduce the water-soluble fraction of Cr, which has a strong relationship with soil phytotoxicity. Biochar is widely recommended as a cost-effective soil amendment for metal-contaminated soils because of its high sorption capability (Ahmad *et al*. 2014). Oyster shells (*Crassostrea angulata*) and citrus peels (*Citrus maxima*) are common agricultural wastes in the Fujian province of China and have become a serious environmental problem due to their local and random disposal. Oyster shell waste is particularly rich in $CaCO_3$ and CaO components, which can serve as a liming material for the stabilization of metal-contaminated soil due to the formation of insoluble metal hydroxides at alkaline pH levels (Moon *et al*. 2013). Citrus peel waste was also considered as an efficient biosorbent in the removal of metal(loid)s from waste (Njikam and Schiewer 2012; Bhatti *et al*. 2016). So these agricultural wastes were added in chromium polluted soils to reduce Cr(VI) to Cr(III) for decreasing the toxicity to improve the lavender growth.

The aim of the present study was to reveal the effects of agricultural waste on the growth of lavender, the accumulation characteristics of chromium and the toxicity of chromium in soil, and to analyze the potential of lavender in the management of high chromium contaminated soil. The assumption in the study was based on lavender has relatively high biomass in high Cr concentration soil after adding agricultural wastes, which can be used to extract essential oil after harvest, and its residue can be burned to extract chromium or disposed as hazardous waste if it contained high Cr.

EXPERIMENTAL

Materials

Soil collection

Soil samples were collected from a Cr-contaminated slag site at a depth of 0 cm to 20 cm from Qingdao in Eastern China, with an average Cr concentration of 8,700 mg/kg ± 200 mg/kg. Approximately 200,000 tons of chromite ore processing residues (COPRs) had been deposited at this contaminated site (14,000 m^2) from 1959 to 2004, generated from a chromate production enterprise. Due to the leaching of rainfall, chromium may dissolve into the leachate and cause serious soil pollution. After 2007, all the COPRs were safely removed, and no vegetation covers were found before soil sampling. The soil, classified as a fluvo-aquic soil, was characterized by a silty loam texture (with 77.6% silt, 15.3% sand, and 7.1% clay) with an average pH of 8.87 ± 0.08. The soil prior to the experiment contained 25.0 g/kg of soil organic matter and 12.9 cmol/kg cation exchange capacity (CEC). Other contaminants, such as heavy metals, were below the safe limits in agricultural soil (see table S1). For example, the total Ni concentration in the studied soil ranged from 32.6 mg/kg to 33.1 mg/kg, Cu (34.1mg/kg - 51.5 mg/kg), Zn from 534.1 mg/kg to 654.0 mg/kg, and As from 29.7 mg/kg to 34.2 mg/kg, respectively. Therefore, only Cr toxicity was considered during plant growth in this study. Soil samples were air-dried at room temperature and crushed to pass through a 2-mm sieve prior to the application of various soil amendments and to transplanting.

Soil amendment preparation

Biochar was produced in a muffle furnace with N_2 flow, using rice straw combusted at 400 °C for approximately 4 h. Before being mixed with the soil, the biochar was passed through a 2-mm stainless steel sieve. Oyster shell waste (*Crassostrea angulata*) was collected from a local oyster farm (Quanzhou City, China), crushed, and ground to pass through a < 0.3-mm mesh. Citrus peel waste (*Citrus maxima*) was collected from a local fruit market (Quanzhou City, China) and was ground and sieved to a 1-mm to 1.1-mm diameter. The selection of these three bio-wastes was based on their local availability and their disposal's association with considerable environmental impacts. In addition, these three amendments were easily obtained locally at low costs.

Pot experiment

The greenhouse pot experiment was conducted to investigate the remediation efficiency of *Lavandula dentata* L. The experiment was performed in the greenhouse of Quanzhou Normal University in Fujian, China, using 12 foamed boxes (80 cm length × 60 cm width × 40 cm height). The following treatments were tested: (T1) contaminated soil amended and mixed with 2.5 wt% biochar and 2.5 wt% oyster shell waste; (T2) contaminated soil amended and mixed with 2.5 wt% oyster shell waste and 2.5 wt% citrus peel waste; (T3) contaminated soil without any amendments; and (T4) uncontaminated soil from a local (Quanzhou, China) agricultural farm, which was under more than 5 yr of continuous cultivation of vegetables and free from heavy metal pollution (*i.e.*, < 75.0 mg/kg Cr).

After adding the amendments, the treated soils were subjected to a 1-week equilibrium period. The lavender (*Lavandula dentata* L.) seedlings used in this study were purchased from the Quanzhou Horticulture Company in Fujian, China. Healthy and uniform-sized seedlings with an average height of 4 cm were selected and transplanted into the three treatments (10 plants in each pot), with three replicates per treatment, following the same arrangement for each treatment. Prior to transplanting, each pot was fertilized with 120 kg N/ha as $CO(NH_2)_2$ as well as 117 kg P/ha and 76.5 kg K/ha as KH_2PO_4. The plants were grown under natural sunlight with a light/dark cycle of approximately 16 h / 8 h, and the temperature in the greenhouse was 18 °C to 30 °C. Each treatment was irrigated daily to maintain similar soil moisture values (60% to 70% water holding capacity) in all treatments. The experiment was started on November 4, 2016, and lasted for 104 d.

Soil and plant sample collection

Soil samples were taken at 0 d, 41 d, and 104 d from each plot after transplanting. The soil was air-dried at room temperature, ground into 2-mm and 0.15-mm particles, and stored until analysis. After the cultivation, all plant roots and aerial parts (considered as shoots) in each pot were harvested, washed with tap water, rinsed with deionized water, and oven-dried at 60 °C to 70 °C for more than 48 h. Subsequently, the plant biomass was determined, and the dried plant material was ground into a powder using mortar and pestle for chemical analysis.

Methods

Chemical analysis

Soil pH was measured in a 1:2.5 (w/v) mixture of soil and water with a pH-EC meter (Excel XL60; Thermo Fisher Scientific, Waltham, MA, USA). The total Cr concentration of the dried samples was determined *via* microwave-assisted acid digestion, using trace-pure HNO_3 and hydrofluoric acid (HF) and a closed-vessel, high-pressure microwave digester (Multiwave GO; Anton Paar, Graz, Austria). The digested solution was then quantitatively transferred to a sterile tube and diluted to volume with distilled water (containing 1.5% HNO_3, v/v). Standard solutions were prepared by dilution of 1,000 mg/L stock solutions, and the calibration curve was obtained using seven points, including the blank. Inductively coupled plasma mass spectrometry (Agilent 7500cx; Agilent Technologies, Inc., Santa Clara, CA, USA) was applied to qualify and quantify the metal concentrations and the correlation coefficients ($r^2 = 0.999$ or better) of the linear calibration curves. The accuracy of the analysis methods was evaluated through repeated analysis of the standard reference materials (GBW07317 for soils and GBW007603 for plants obtained from the Center of National Standard Reference Material of China); recoveries of Cr were 106% and 91.7% for soil and plants, respectively. The laboratory control sample, the blank, and the duplicate samples (relative percent difference was less than 17%) were prepared and analyzed as quality controls in each procedural batch.

Hexavalent chromium (Cr(VI)) in the soil was extracted using the alkaline digestion method (Na_2CO_3/NaOH) according to USEPA Method 3060A (1996) and determined by colorimetry with diphenylcarbazide according to USEPA Method 7196A (1992). Trivalent chromium (Cr(III)) in the soil was calculated from total Cr and Cr(VI), for which it was assumed that the total Cr was composed of Cr(VI) plus Cr(III) (Banks *et al*. 2006). Modified Community Bureau of Reference (BCR) sequential extraction (F1: water-soluble fraction; F2: acid-soluble fraction; F3: reducible fraction; F4: oxidizable fraction; and F5: residual fractions) of the soil was conducted using the procedure of Arain *et al*. (2008). All extraction steps were performed in 50-mL polypropylene centrifuge tubes with screw caps. The detailed technical procedures for chemical fractionation can be found elsewhere (Tokalıoğlu *et al*. 2010; Hasan *et al*. 2018).

Statistical analysis

Statistical analysis was performed using SPSS 13.0 software for Windows (SPSS Inc., Chicago, IL, USA). One-way analysis of variance (ANOVA) and *post hoc* multiple comparisons (least significant difference) were used to determine the differences between treatments at a significance level of $p < 0.05$. The bioconcentration factor (BCF) is an index to depict the ability of the plants to accumulate particular heavy metal from soil, and the translocation factor (TF) indicates the ability of plant to translocate the heavy metal from roots to the aerial part of plant (Mattina *et al*. 2003; Ghosh and Singh 2005). These are shown in Eqs. 1 and 2,

$$BCF = \frac{C_{\text{pt}}}{c_{\text{s}}} \tag{1}$$

$$TF = \frac{c_{\text{ap}}}{c_{\text{r}}} \tag{2}$$

where c_{pt} is the average metal concentration in the plant tissue (mg/kg), c_{s} is the metal concentration in the soil (mg/kg), c_{ap} is the metal concentration in the aerial part of plant (mg/kg), and c_{r} is the metal concentration in the root of plant (mg/kg).

RESULTS AND DISSCUSSION

Cr-contaminated Soils

The studied soil was highly polluted with chromium, with Cr concentrations greater than 8,000 mg/kg, which was 80 times greater than the natural background value of Cr in Qingdao, China (91 mg/kg to 108 mg/kg) and much higher than the three-level standard of Soil Environmental Quality Standard in China (the recommendations for paddy fields and dry land areas are 500 mg/kg and 300 mg/kg, respectively). Such soil should be carefully managed to prevent leaching and runoff. Based on the total Cr concentration and alkaline extraction, Cr(VI) accounted for at least 4.7% to 19.0% of the total soil Cr, while Cr(III) accounted for 81.0% to 95.3%, from the beginning to the end of the cultivation period, assuming that all the Cr(VI) was extracted by the alkaline digestion (James *et al*. 1995). It was noted that CrO_4^{2-} and $HCrO_4^-$ were the substantial species in Cr(VI) aqueous solutions and that the relative distribution of each varied with pH, with CrO_4^2 being the dominant species in alkaline environments, thereby decreasing the diffusion of chromate ions in the soil environment due to the competitive adsorption between OH^- ions and oxyanions of chromium (Sengupta *et al*. 1986; Mohamed *et al*. 2016). One reason for the decreased Cr(VI) concentration in the cultured soils (Fig. 1) might have been the high pH value (alkalinity) of the soil and the changed redox conditions following cultivation (Banks *et al*. 2006; Choppala *et al*. 2018).

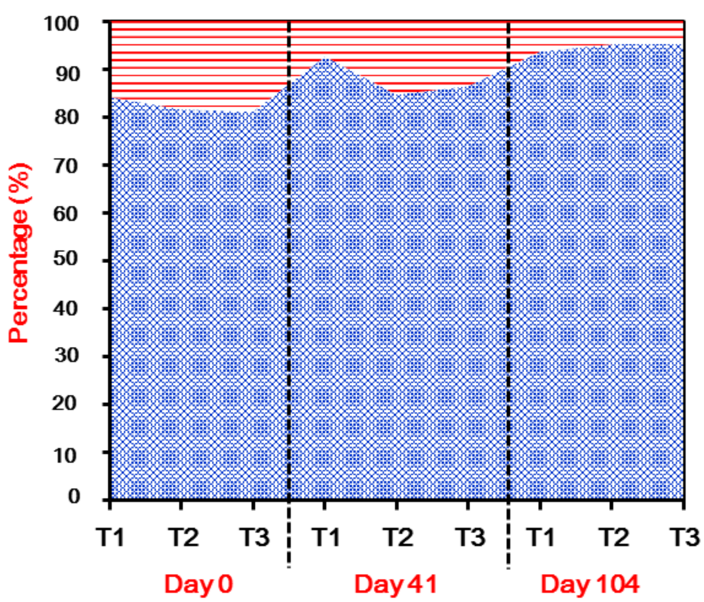

Fig. 1. The percentage of extracted Cr(VI) (the red part) and computed Cr(III) (blue part) in the soils depending on elapsed time – T1: contaminated soil was amended with 2.5 wt% biochar and 2.5 wt% oyster shell waste; T2: contaminated soil was amended with 2.5 wt% oyster shell waste and 2.5 wt% citrus peel waste; and T3: control soil without any amendments

Throughout the entire cultivation period, the soil pH was rather alkaline (8.30 to 8.92), which increased the retention of Cr(III) because of the low solubility of $Cr(OH)_3$ and (Fe ,Cr)$(OH)_3$ in the pH range of 7.0 to 9.0 (Rai *et al*. 1989). Under natural conditions, most of the Cr in soils is bound by iron oxides and organic matter, and only a small proportion occurs in the exchangeable form (Shahid *et al*. 2017). However, Cr(VI) mainly occurs as an anion, and its activities are controlled both by hydroxide minerals and organic matter in Cr-contaminated soils (Jardine *et al*. 2013; Hausladen and Fendorf 2017). Another reason may be that organic matter and biochar in the soil adsorb Cr (VI), some functional groups reacted with hexavalent chromium in the adsorption process (C-O was oxidized to C=O), which led to the reduction of Cr(VI) to Cr(III) (Yin *et al*. 2019).

Table 1 shows the chromium fractions in this study, where chromium was mainly associated with the residual phase. The water-soluble fraction (F1) showed Cr levels in the range of 5.0 mg/kg to 48.1 mg/kg. The significant decrease of this Cr fraction (F1) in the contaminated soil during plant cultivation indicated effective remediation, including the amendments' sorption and plant uptake, which were likely based on the bioavailability of Cr in the aqueous phase.

The lower concentration of the F1 fraction in the T1 treatment (compared to T2 and T3) might be explained by the adsorption of water-soluble Cr to the biochar (Ahmad *et al*. 2014; Herath *et al*. 2017). Oyster shell waste also has a high sorption capacity for water-soluble metals, which probably led to the high concentration of the acid-soluble fraction (F2) in the T2 treatment (Table 1) (Moon *et al*. 2013). This fraction was predominantly extracted in the first step of the modified sequential extraction, representing the metal bound to acid-soluble fractions such as carbonates or sorbed/exchangeable phases (Arain *et al*. 2008).

Meanwhile, in all the treatments, the Cr levels in the leachate were related to the acid-soluble fraction, suggesting that the bioavailability of Cr (water-soluble fraction) in highly contaminated soil strongly depends on the sorbents and the soil pH (Zayed and Terry 2003; Kumpiene *et al*. 2008). Chromium extracted from the reducible phase accounted for 1,010 mg/kg to 1,832 mg/kg. Alkaline soil conditions enhance the affinity of Fe/Al oxides to cations because of the high sorption capacity of Fe/Al hydroxides to metal elements (Langlois and James 2015).

In this study, alkaline pH values indicated the predominant negative charges on mineral surfaces, which would lead to appreciable electrostatic repulsion of the chromate oxyanions (He and Traina 2005; Gu *et al*. 2017). However, it was also noted that Cr(VI) would be firmly retained by Fe(III)/Cr(III) hydroxides, likely due to the formation of surface precipitates or complexes between Cr(VI) and Cr(III) sites, which could hardly be substituted by OH^- during the pH increase (Tzou *et al*. 2003). The range of extractable Cr in the oxidizable fractions was 1,147 mg/kg to 1,830 mg/kg, depending on the treatment, which showed increased concentration of this element in the soil, particularly in the T2 treatment that was amended with citrus peel waste. Organic matter can play a crucial role in Cr retention in the soil environment, because of the strong interaction with Cr(III), and in its ability to transform Cr(VI) to Cr(III) (Rai *et al*. 1989; Gustafsson *et al*. 2014). In addition, the low variability of the three main Cr fractions (*i.e.*, reducible, oxidizable, and residual) during plant cultivation suggests a relatively successful stabilization of Cr due to the soil amendments.

Table 1. Sequential Extractable Cr Concentrations in the Experimental Soils

		Water-soluble Fraction	Acid-soluble Fraction	Reducible Fraction	Oxidizable Fraction	Residual Fraction	Total Cr
T1	Day 0	23.7 ± 1.5^{a}	35.2 ± 4.5^{a}	1,315.8 ± 150.6^{a}	1,444.6 ± 96.8^{a}	5,105.5 ± 117.1^{ab}	7,924.7 ± 365.7^{a}
	Day 41	14.6 ± 1.6^{b}	23.9 ± 1.1^{b}	1,142.8 ± 119.9^{a}	1,302.8 ± 138.2^{a}	4,884.9 ± 189.9^{a}	7,368.8 ± 139.7^{b}
	Day 104	7.5 ± 2.2^{c}	20.6 ± 2.8^{b}	1,098.3 ± 148.2^{a}	1,280.7 ± 155.3^{a}	5,254.9 ± 170.6^{b}	7,662.0 ± 85.2^{ab}
T2	Day 0	32.1 ± 2.6^{a}	46.9 ± 5.0^{a}	1,456.8 ± 122.0^{a}	1,752.2 ± 89.7^{a}	5,112.5 ± 251.7^{a}	8,400.5 ± 126.4^{a}
	Day 41	24.8 ± 1.5^{b}	33.9 ± 0.6^{b}	1,287.0 ± 97.9^{a}	1,462.2 ± 72.5^{b}	4,861.1 ± 147.8^{a}	7,669.0 ± 8.7^{b}
	Day 104	19.9 ± 2.0^{c}	40.8 ± 0.2^{c}	1,229.3 ± 59.2^{a}	1,387.0 ± 95.6^{b}	5,202.7 ± 212.8^{a}	7,949.6 ± 177.0^{c}
T3	Day 0	44.6 ± 4.1^{a}	28.2 ± 1.6^{a}	1,735.8 ± 89.4^{a}	1,713.5 ± 73.8^{a}	5,218.6 ± 129.3^{a}	8,740.7 ± 264.0^{a}
	Day 41	33.1 ± 2.9^{b}	24.1 ± 3.0^{a}	1,335.3 ± 121.6^{b}	1,441.7 ± 121.1^{b}	5,354.1 ± 230.1^{a}	8,178.4 ± 179.7^{b}
	Day 104	33.9 ± 3.4^{b}	25.5 ± 3.0^{a}	1,501.0 ± 21.7^{b}	1,425.1 ± 43.7^{b}	5,529.5 ± 106.1^{a}	8,515.1 ± 121.8^{b}

Values represent mean ± standard deviation (SD) (mg/kg dry mass). Different letters (a, b, c) represent significant differences ($p < 0.05$) among sampling days in each treatment. T1: contaminated soil was amended with 2.5 wt% biochar and 2.5 wt% oyster shell waste; T2: contaminated soil was amended with 2.5 wt% oyster shell waste and 2.5 wt% citrus peel waste; and T3: control soil without any amendments

Growth of Lavender Plant

Chromium significantly inhibited plant development (Fig. 2A), which was probably due to the toxic effects of Cr and the high bioavailability of the element in contaminated soils (Shahid *et al*. 2017; Velez *et al*. 2017). Here, the soil amendment combined with biochar and oyster shell waste (T2) reduced the toxic effects on the lavender. Plants growing in the untreated contaminated soil showed a decrease in shoot and root dry weight (DW) yields of more than 80% (compared with uncontaminated soil) (Fig. 2B), demonstrating a severe phytotoxicity that might be related to Cr excess greater than 8,000 mg/kg. It has been reported that a level of Cr > 5.2 mg/kg in soil can induce toxicity in plants such as *Helianthus annuus* (Davies Jr. *et al*. 2002). However, the stress effects on plant growth depend on a variety of soil conditions, such as pH, organic matter, and oxidation-reduction potential (Nirola *et al*. 2018). In general, the Cr(VI) forms of chromate and dichromate are highly soluble in water, while Cr(III) is less soluble due to higher mobility of Cr(VI) in the soil environment. Several studies have evaluated the phytotoxicity of both Cr(III) and Cr(VI) in many plants with greater and lower biomasses, and found that Cr(VI) is more phytotoxic than Cr(III), particularly in retarding plant growth (Panda and Choudhury 2005). In this study, lavender growth was significantly affected by Cr at concentrations greater than 8,000 mg/kg, which might have been mainly due to the excess 1,400 mg/kg Cr(VI) in the initial stage. Similar results in previous studies also demonstrated a linear decrease in shoot and root dry weight resulting from increasing Cr(VI) concentrations (Velez *et al*. 2017). However, the reduction in plant growth is not only a factor of metal concentration, but it also depends on the chemical forms, which can play a crucial role in the heavy metal detoxification mechanisms (Ali *et al*. 2004). Previous research has shown that binding effects (*i.e.*, metals binding to oxalates and residuals) can reduce the metals' toxicity to terrestrial plants (Clemens *et al*. 2002). Therefore, the application of soil amendments (*i.e.*, biochar, oyster shells, and citrus peel waste) in this study could provide more sorption sites for Cr binding and consequently mitigate retardation of plant growth. In addition, none of the plants died during cultivation, indicating the stress tolerance of the lavender plants to Cr contamination.

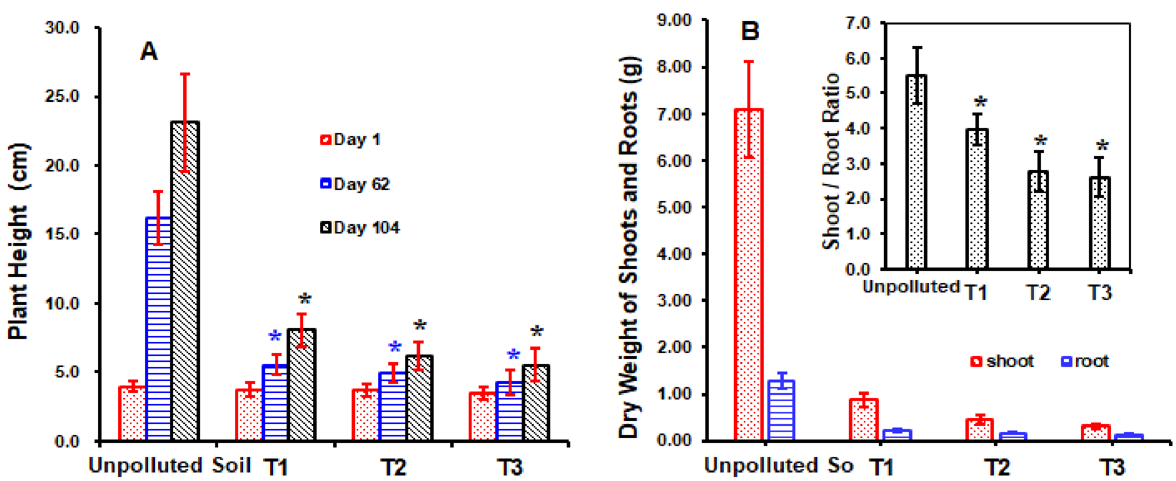

Fig. 2. A: Growth performance of lavender (*Lavandula dentata* L.) planted in soil with and without high concentrations of Cr. B: Dry biomass weight was determined at the end of the experiment. Asterisks (*) indicate statistically significant differences with respect to control plants grown without Cr ($p < 0.05$).

To investigate the phytomanagement potential of lavender plants in soils to total Cr concentrations, the cost-effective biosorbents biochar, oyster shell waste, and citrus peel waste were combined as soil amendments because of their high capacities to directly or indirectly react with heavy metals in the soil environment, thereby reducing metal bioavailability (Njikam and Schiewer 2012; Ahmad *et al*. 2014). After the incorporation of the soil amendments, the water-solution Cr fraction decreased almost 2 fold for the T1 treatment and 3 fold for T2 treatment (Table 1). In the un-amended soil, although plant growth was completely inhibited, the water-solution Cr fraction in the soil decreased 23.9%, which might have been because lavender plants (shoots and roots) bioaccumulate Cr. The total amounts of Cr uptake by the lavender plants (all plants in a pot, including shoots and roots) were approximately 2.82 mg Cr, 3.14 mg Cr, and 3.59 mg Cr for the T1, T2, and T3 treatments, respectively. This concentration removal of Cr from contaminated soil accounted for 0.87% to 1.67% of the decrease of the water-solution Cr fraction, indicating the significant binding effects of soluble Cr with amendments and soil minerals (*i.e*., metals binding to residuals). The addition of amendments resulted in a significant reduction ($p < 0.05$) in the water-soluble Cr fraction, which was more obvious with the biochar treatment (Table 1). The interaction of water-soluble Cr with amendments is controlled by a variety of factors, such as ion composition, metal species, and the charge on the sorptive surface, which are strongly influenced by pH levels (Egene *et al*. 2018; Wang *et al*. 2018). The significant decrease of this fraction indicates the sorption capacity of the selected materials (Njikam and Schiewer 2012; Moon *et al*. 2013; Ahmad *et al*. 2014). For example, shell waste showed its sorption capacity of arsenic and was shown to have positive effects on soil arsenic retention, while citrus peel waste was also considered as an efficient biosorbent in the removal of metal(loid)s from waste (Njikam and Schiewer 2012; Seco-Reigosa *et al*. 2013; Bhatti *et al*. 2016). The results suggested that these cost-effective soil amendments can be used to improve the phytomanagement efficiency of high-value plants, such as lavender, and they represent an incentive for farmers to become actively involved in soil remediation projects due to the high-value returns (*e.g*., essential oil) with low input (*i.e*., cheap amendments) (Hashemi *et al*. 2017).

Phytomanagement Efficiency of Lavender

Figure 3 shows the bioaccumulation of Cr in the roots and shoots of the lavender plants. The treatments with soil amendments resulted in low Cr concentrations in the plant tissues. In contrast, the un-amended soil (T3) led to high Cr concentrations (mg/kg DW) in roots (817.5 ± 52.1) and shoots (843.9 ± 47.0) as compared to the T1 and T2 treatments (184.6 ± 42.7 and 235.4 ± 43.3 for T1; 631.5 ± 75.4 and 450.3 ± 58.8 for T2, respectively). However, the balance masses of Cr removal by the plants (sum of shoots and roots) were only $0.87\% \pm 0.13\%$, $1.29\% \pm 0.16\%$, and $1.68\% \pm 0.10\%$ of total dissipated amounts of soil water-soluble Cr for T1 (323.3 mg Cr $\pm$ 42.6 mg Cr), T2 (243.9 mg Cr $\pm$ 28.7 mg Cr), and T3 (214.2 mg Cr $\pm$ 22.1 mg Cr), respectively. These results demonstrated that Cr uptake by the lavender was significantly controlled by its bioavailability, such as the water-soluble fraction in the soil, which was reduced by the application of soil amendments, particularly in the treatment with biochar combined with oyster shell waste (Table 1). In general, the accumulation and distribution of metal(loid)s in plant tissues are important aspects to evaluate the role of plants in the remediation of metalliferous soils (Pichtel and Bradway 2008). In terms of removing metals from contaminated sites, a greater metal concentration in the aboveground parts is preferred for enhancing the phytoremediation efficiency (Ma *et al*. 2016). The high Cr concentrations in the lavender shoots (843.9 mg/kg

± 47.0 mg/kg) grown in highly Cr-contaminated soils were close to the threshold concentration of Cr hyperaccumulators (1,000 mg/kg) (Baker and Brooks 1989). The results demonstrate that the lavender plant could be considered as a potential hyperaccumulator in Cr-contaminated sites, with the high toxicity of Cr to plants resulting in a rare ideal option for Cr phytoremediation (Nirola *et al*. 2018). However, the low plant yield and low growth rate of lavender in such highly Cr-contaminated soil may restrict its phytoremediation efficiency. However, as a high-value plant, lavender is usually grown as a landscape plant and harvested to produce essential oil, which tends to be free from heavy metals. These financial returns from lavender growth may stimulate public involvement in the rehabilitation and management of contaminated farmland, particularly in remote and less developed regions. Therefore, lavender cultivation, combined with the suitable application of soil amendments, is a novel option to phytomanage highly Cr-contaminated soils.

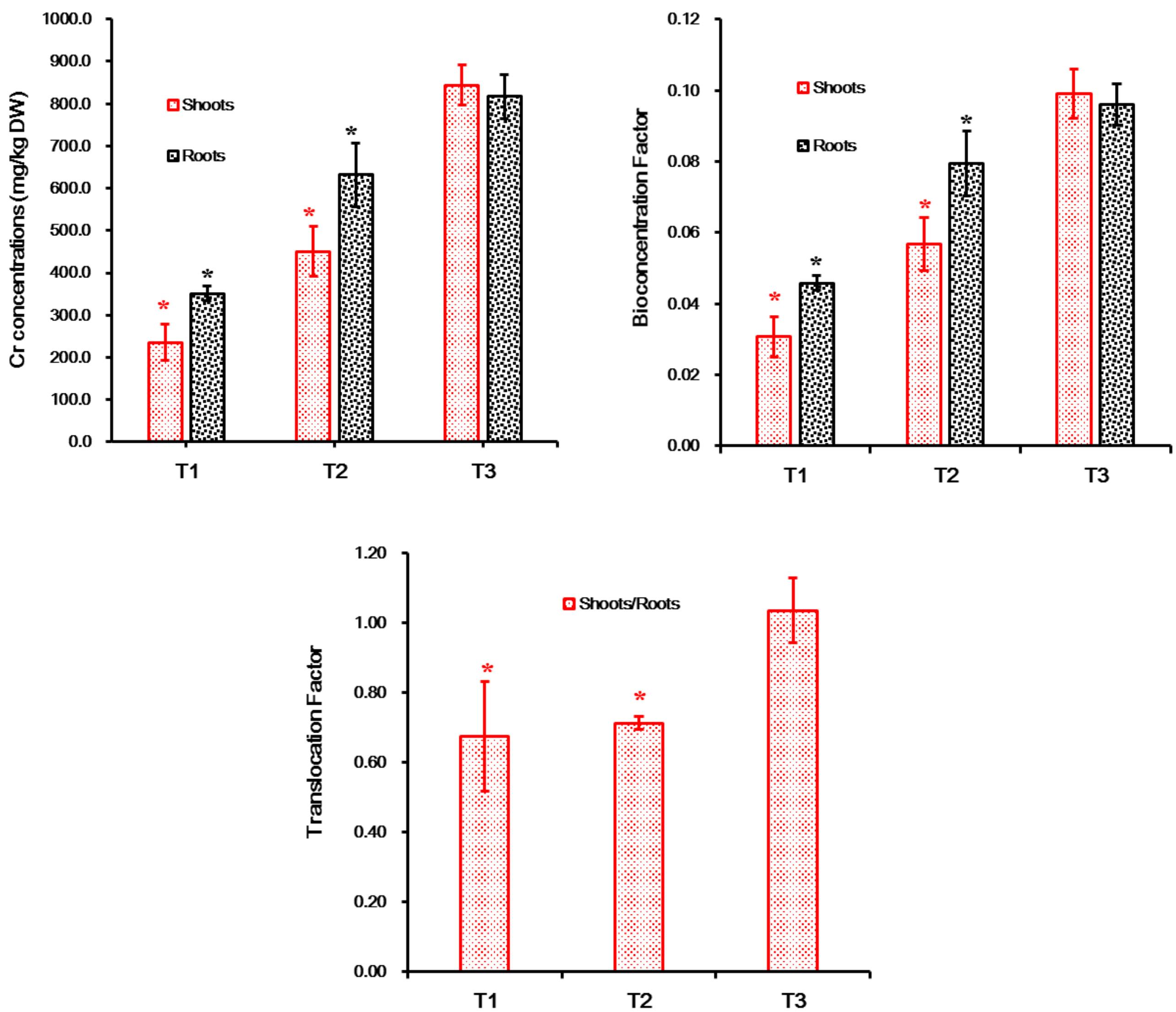

Fig. 3. Bioaccumulation and bioconcentration of lavender (*Lavandula dentata* L.) planted in soil with and without high soil amendments. Asterisks (*) indicate statistically significant differences with respect to the treatment without any amendments ($p < 0.05$).

The BCF represents the capacity of a plant to accumulate elements such as metals. In this study, the BCF values of plants grown in un-amended metal-contaminated soil (T3) were significantly greater than those of plants grown in amended soils. Moreover, the BCF values became significantly decreased as a result of the increasing sorption capacity (*i.e.*, biochar *vs*. citrus peel waste) of the amendments (Fig. 3).

The concentrations of water-soluble Cr were directly proportional to the metal bioconcentration factors of the plants (Fig. 4), suggesting that the application of amendments stabilized the heavy metals in the soil and reduced the bioavailable fraction of Cr. The TF of the Cr from the lavender roots to the shoots was greater in plants grown in un-amended soil. Reductions in the root-to-shoot translocation of heavy metals in the plant after the addition of soil amendments have been previously reported and are most likely a result of altered metal speciation in the solution, which changes the phytoavailability by increasing the soil pH or influencing plant growth and plant physiology (Clemens *et al*. 2002; Yadav *et al*. 2009; Shahid *et al*. 2014; Vargas *et al*. 2016; Hashemi *et al*. 2017). Whenever phytostabilization or phytoextraction are chosen as a remediation technique, the risk of pollutant dispersal into the environment should be minimized. In addition, it has been suggested to cultivate non-food crops on metal-contaminated soils (*e.g.*, aromatic plants) that can be used to produce essential oils (Gupta *et al*. 2013; Verma *et al*. 2017).

The percentage of heavy metal translocation to the harvestable parts is an important feature for the selection of a phytoremediation plant. However, in many Cr hyperaccumulators, native species from Cr-contaminated sites are frequently of low productivity and growth rate, which restricts their commercial use (Kale *et al*. 2015). The crops, such as *Zea mays*, must carefully consider the security of the food chain, thus reducing their application in open areas (Gheju and Balcu 2017). For lavender, although metal accumulation was observed in shoots, the essential oil is free from heavy metals. In addition, lavender plants can be cultivated as attractive ornamental elements on abandoned fields and cannot be damaged or eaten by wild animals due to its essential oil (Gupta *et al.* 2013). Therefore, lavender growth can offer a feasible alternative for the management of highly Cr-contaminated sites, especially where financial support to establish a remediation technology is not available (Zheljazkov *et al*. 2006).

Biochar and other agricultural wastes could absorb chromium ions and combine with them through porous filtration, ion exchange, electrostatic attraction and complexation reaction (Yin *et al.* 2019), which can prevent the migration and leakage of chromium ions in the soil and reduce the risk of chromium ion pollution to groundwater and surface water. In fact, lavender is a perennial plant.

In order to improve the remediation efficiency of polluted soil, the aboveground part and the underground part should be harvested together in slightly or moderately chromium contaminated soil, because the root system of lavender is developed and contains high content of chromium. However, it may be recommended to harvest only the above ground part in an area with serious soil erosion or in the soil seriously polluted by chromium, because the strong root system of lavender is conducive to prevent the diffusion of pollutants with the surface runoff. After the extraction of essential oil, lavender residues should be burned and disposed of safely as hazardous waste because they contained high concentrations of chromium (Lu *et al*. 2012).

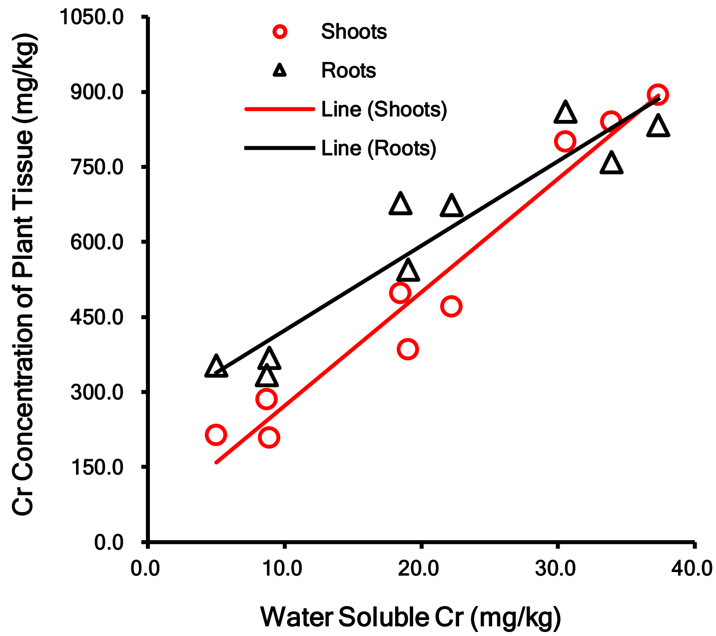

Fig. 4. Relationship between Cr concentration in the plant tissue (shoots and roots) and the concentration of the water-soluble Cr fraction

CONCLUSIONS

1. High Cr concentration (greater than 8,000 mg/kg) in the soil affected the lavender plants' growth by significantly decreasing plant height and biomass. The high proportion of Cr(VI) of total soil Cr in the initial stage was the main reason for these inhibitions.
2. Soil amendments, including biochar (2.5 wt%) + oyster shell waste (2.5 wt%) and oyster shell waste (2.5 wt%) + citrus peel waste (2.5 wt%), reduced the water-soluble soil Cr fraction and thereby impeded the Cr uptake by the plants. As a result, the growth of the lavender plants was significantly enhanced *via* the additions of the soil amendments, particularly in the treatment combining biochar with oyster shell waste. So biochar, oyster and citrus peel wastes were good soil amendments to improve phytoremediation plant growth in sever Cr polluted soil.
3. In addition, the relatively high Cr concentrations in the lavender shoots (up to 843.9 mg/kg ± 47.0 mg/kg) and roots (up to 817.5 mg/kg ± 52.1 mg/kg) in un-amended soil suggested lavender was a chromium tolerant plant and it could be used for phytomanagement of Cr serious polluted soil. Based on its special uses (*i.e.*, as an essential oil), lavender (*Lavandula dentata* L.) represented a prospective plant for the phytoremediation of Cr-contaminated soils, and its biomass could be enhanced with the addition of soil amendments. Such an approach combines economic viability with the low risk of metal accumulation in the plant biomass. In the process of lavender stabilization management of high concentration chromium contaminated soil, it is recommended to harvest only the above ground part of lavender for essential oil extraction, and the residue should be disposed according to the hazardous waste management method after incineration.

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APPENDIX

SUPPLEMENTARY INFORMATION

Table S1. Total and Tow Extractable Concentration of Toxic Metals in the Experimental Soils

		Cr	Ni	Cu	Zn	As	Cd	Pb
T1	Total	7,924 ± 365.7	32.6 ± 2.4	34.2 ± 1.6	534.1 ± 63.6	29.7 ± 2.7	0.9 ± 0.1	68.7 ± 4.9
	water-soluble	23.7 ± 1.5	0.17 ± 0.03	0.20 ± 0.02	0.54 ± 0.23	0.14 ± 0.06	0.00 ± 0.00	0.04 ± 0.02
	acid-soluble	35.2 ± 4.5	12.05 ± 2.13	0.42 ± 0.07	39.51 ± 6.51	0.15 ± 0.01	0.08 ± 0.03	0.32 ± 0.08
T2	total	8,400 ± 126.4	33.1 ± 1.6	51.5 ± 3.2	612.7 ± 30.1	34.2 ± 2.0	1.0 ± 0.0	73.5 ± 3.1
	water-soluble	32.1 ± 2.6	0.03 ± 0.01	0.02 ± 0.01	0.33 ± 0.26	0.08 ± 0.01	0.00 ± 0.00	0.02 ± 0.02
	acid-soluble	46.9 ± 5.0	14.54 ± 3.32	0.10 ± 0.05	38.41 ± 0.63	0.12 ± 0.00	0.05 ± 0.00	0.19 ± 0.02
T3	total	8,740 ± 264.0	32.8 ± 0.3	36.0 ± 0.8	654.0 ± 10.8	33.0 ± 2.0	1.0 ± 0.0	78.1 ± 1.3
	water-soluble	44.6 ± 4.1	0.06 ± 0.01	0.10 ± 0.01	0.54 ± 0.13	0.09 ± 0.03	0.00 ± 0.00	0.04 ± 0.01
	acid-soluble	28.2 ± 1.6	14.96 ± 1.57	0.41 ± 0.03	54.59 ± 6.36	0.10 ± 0.00	0.06 ± 0.00	0.26 ± 0.02

(n = 3); values represent mean ± S.D (mg/kg dry mass)