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An integrated approach to safer plant production on metal contaminated soils using species selection and chemical immobilization

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ABSTRACT

In order to examine the species specific accumulation of heavy metals in medicinal crops, seven different common medicinal plants were cultivated on a Cd (55 mg kg^{-1}) and Pb (1283 mg kg^{-1}) contaminated soil. Subsequently, the effect of various immobilizing agents, applied in isolation and in combination, on Cd and Pb uptake by two medicinal plant species was examined.

Cadmium and Pb root concentrations in medicinal plants grown in the control soil varied between 0.5 and 2.6 mg kg^{-1} for Cd and 3.2 and 36.4 mg kg^{-1} for Pb. The highest accumulation occurred in Osterici Radix (*Ostericum koreanum)* and Ginger (*Zingiber officinale)* and the lowest in Yam (*Dioscorea batatas)*. Application of immobilizing agents significantly reduced both Cd and Pb concentrations in all medicinal plants examined, where the most effective single immobilizing agent was lime fertilizer (LF). Application of combination treatments involving sorption agents such as compost together with lime further decreased Cd and Pb concentrations from 1.3 and 25.3 mg kg^{-1} to 0.2 and 4.3 mg kg^{-1}, respectively, which was well below the corresponding WHO guidelines. Thus appropriate immobilizing agents in combination with species selection can be practically used for safer medicinal plant production.

1. Introduction

While medicinal plants have been utilized as traditional folk remedies for thousands of years (Palombo, 2006), they have recently received significant attention from the pharmaceutical, health food, and natural cosmetic industries (Khorasaninejad et al., 2011; Saikia et al., 2006). This recent increased interest in natural and/or organic products has consequently triggered expansion of medicinal plant cultivation in agricultural upland. For example, in Korea the area used for medicinal plant cultivation increased from 7676 ha in 2000 to 12,834 ha in 2013 (MAFRA, 2014). While medicinal plants can have advantageous human health effects, they may also pose a risk to human health if the concentrations of potentially hazardous substances, such as heavy metals (HMs), exceed safety limits. Indeed, recently HM exposure via the ingestion of medicinal plants and/or medicinal plant derived products has become a hot topic in the news and mass media. To protect human health, the World Health Organization (WHO) (1998) legislated maximum permissible limits of toxic metals including arsenic (1.0 mg kg^{-1}), cadmium (0.3 mg kg^{-1}), and lead (10 mg kg^{-1}) on a dry weight basis. Many Asian countries including China, Korea, Malaysia, Singapore and Thailand also have their own guideline values to ensure the safe management and trade of medicinal plants.

Heavy metal exposure via medicinal plants mainly occurs through the cultivation of medicinal crops on HM contaminated soils (Başgel and Erdemoğlu, 2006). The extent of metal uptake by medicinal plants from the soils is governed by the phytoavailable metal pools in the soil rather than the total metal concentration (Chojnacka et al., 2005). Since it is well known that most plants preferentially accumulate HMs in their roots rather than in their aboveground parts (Carbonell et al., 2011; Li et al., 2009; Singh and Agrawal, 2007), specific attention needs to be paid to medications derived from medicinal plant roots. In addition, as some medicinal plant species are known to be HM hyper-accumulators (Lai and Chen, 2005; Masarovičová et al., 2010; Wei et al., 2008) in some instances significant metal transport from roots to shoots can occur which is also of potential health concern when medications are

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derived from foliar plant parts. Since, HMs may be accumulated in both above and below ground plant parts, which cannot be easily removed post-harvest, it is essential to avoid any excessive metal uptake during cultivation. Also in terms of plant performance, HMs can also affect numerous biochemical and physiological processes and reduce plant chlorophyll content, photosynthesis, and biomass (Adrees et al., 2015; Keller et al., 2015; Zhang et al., 2014).

Two potential approaches currently exist to reduce metal uptake in plants. The first approach involves selection of hypoaccumulating medicinal plants, which are naturally predisposed to accumulate significantly lower amounts of HMs in their tissues (Kim et al., 2012). As each plant species exhibits genotypic variations, the extent of metal accumulation by medicinal plants would also vary with species (Yang et al., 2010). For example, Alexander et al. (2006) demonstrated that the accumulation of HMs (Cd, Cu, Pb and Zn) varied in common crop plants in the order lettuce > spinach > carrot > onion > pea > bean. The second approach to minimize metal uptake by medicinal plants involves chemical immobilization; where soil amendments act to reduce the soil's phytoavailable metal pools (Rehman et al., 2016). Among the myriad of potential immobilizing agents proposed, alkaline materials such as lime based materials, fly ash, and biochar are the most popular (Castaldi et al., 2005; Gray et al., 2006; Kim et al., 2015). These materials increase soil pH, favor deprotonation and the formation of oxides, metal-carbonate precipitates, complexes and secondary minerals (Chlopecka and Adriano, 1996; Mench et al., 1994) that all reduce HM phytoavailability. For example, application of red mud at 5% (w/w) decreased Cd and Pb phytoavailability by 52% and 85%, respectively, due to an increase in soil pH of 1.7 units (Gray et al., 2006).

In addition to pH change-induced immobilizing agents, decreasing HM phytoavailability may also be achieved in soils through the application of either sorption agents or materials that decrease dissolved organic carbon (DOC). Li et al. (2008) reported that application of peat to Cd and Cu contaminated soil at 90 g per pot decreased the concentrations of Cd and Cu by 32% and 87%, respectively. This decrease was attributed to the increased HM adsorption capacity of the peat. DOC is known to increase metal phytoavailability through the formation of dissolved organo-metallic complexes (Gray et al., 2006). Hence, decreases in DOC concentrations in soil solution following application of Ca-containing compounds can also decrease the phytoavailable HM concentrations (Römkens et al., 1996). Calcium can reduce the DOC concentration in the soil solution through DOC coagulation (Bolan et al., 2003).

Thus the current study was carried out to examine the effectiveness of an integrated approach to reduce Cd and Pb uptake in medicinal plants. These two metals were chosen because they are the main two HMs commonly associated with human health issues and are potentially present in high levels in medicinal plants. The current study tests the hypothesis that metal uptake in medicinal plants can be reduced via hypoaccumulating medicinal plant selection together with application of chemical immobilization, to reduce HM phytoavailability and produce a safer medicinal plant for cropping on metal contaminated soils.

2. Methods and materials

2.1. Soil, plants, and immobilizing agents

The soil used for this study was a cultivated upland soil collected from an abandoned mining site contaminated by two heavy metals (Cd and Pb). Four tons of topsoil (0∼25 cm) were collected using an excavator and thoroughly mixed prior to use in the pot trial. Selected physicochemical properties of a representative subsample of the mixed soil are summarized in Table 1.

Table 1.
pH, EC, and heavy metal (Cd and Pb) concentration of the soil and immobilizing agents used in this study.

Materials	pH	EC (mS cm^{-1})	Cd	Pb
			(mg kg^{-1})	
Soil	5.7	0.1	55	1283
Lime fertilizer	12.3	9.5	0.7	0.1
Compost	7.5	9.3	0.9	8.0
Biochar	10.3	1.6	0.1	0.8
Fly ash	12.1	6.1	1.2	2.3
Spent mushroom media	5.3	4.1	0.3	3.6
Gypsum	9.4	10.3	2.6	4.1

Seven different medicinal plants, which are all widely cultivated throughout Korea (MFDS, 2006), were selected for the pot trial to examine species specific differences in root accumulation of Cd and Pb. The seven plants selected were Osterici Radix (*Ostericum koreanum*), Ginger (*Zingiber officinale*), Atractylodes rhizome white (*Atractylodes macrocephala*), Korean Angelica (*Angelica gigas*), Pilose asiabell (*Codonopsis pilosula*), Rehmannia (*Rehmannia glutinosa*) and Yam (*Dioscorea batatas*).

Six different immobilizing agents (biochar, compost, fly ash, gypsum, lime fertilizer, and spent mushroom media) were either applied in isolation (as a single incorporation to the soil) and/or as complex mixture (applied simultaneously to the soil as a mixture of 2–3 reagents). Lime fertilizer was included as a soil pH change-inducing immobilizing agent, while compost and spent mushroom were included as sorption agents. Biochar and fly ash were two materials expected to act both as soil pH change and sorption agent. Like lime, application of biochar and fly ash has been shown to increased soil pH when used as an amendment (Kim et al., 2015; Lee et al., 2006). Finally, gypsum was included as a DOC coagulator. Mixing of the contaminated soil with an uncontaminated soil at a 4-1 ratio (hereafter refer to as “diluted soil”) was included to properly compare immobilizing agent effects. The selected chemical properties of the immobilizing agents used in this study are presented in Table 1.

2.2. Pot study setup

Variation in metal accumulation in plant roots with species was assessed by growing all seven medicinal plants in the untreated (control) soil. Selected species were also grown in soils incorporating various immobilization agents in isolation and in combination. Treatments to observe the variation in response among plant species due to changes in phytoavailable pools of Cd and Pb included: lime fertilizer treated (LF), compost treated (CO), lime + compost treated (LF-CO) soils. Two specific medicinal plant species (Korean Angelica and Atractylodes rhizome white) were also selected to examine the efficiency of all immobilization treatments (Table 2). The treatments applied (Table 2) were specifically chosen to enable comparison of the effects of each agent in isolation and also any possible synergetic effects of immobilizing agents on decreasing Cd and Pb uptake by the plants. Application rates of each agent were fixed on a dry weight basis of soil at 1% for LF and 3% for all other agents based on comparison with previous published studies (Han et al., 2013; Kim et al., 2012). All treatments were prepared in quadruplicate.

In each pot, contaminated soil (25 kg) was thoroughly mixed with the desired immobilizing agents and allowed to equilibrate for one month prior to planting. During the equilibration period, the moisture contents were maintained with natural rainfall and