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RESEARCH ARTICLE

Novel chelant-based washing method for soil contaminated with Pb and other metals: A pilot-scale study

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Abstract
Remediation with chelants can restore metal-contaminated soils for use as a natural resource. Calcareous soil from Meza Valley, Slovenia, and acidic soils from Arnoldstein, Austria, and Pribram, Czech Republic (with 1,028, 862, and 926 mg Pbkg^{-1}, respectively), were washed with 60–100 mmol EDTA per kilogram of air-dried soil in series of 30 batches (50 kg soil batch^{-1}). The approach involves a novel reaction that incorporates alkaline substitution, precipitation and adsorption of toxic metals on polysaccharides, and chelant acidic precipitation via 83% EDTA (on average) and complete process water recycling (no wastewater was generated). The pH gradient was imposed by $Ca(OH)_2$ and H_2SO_4, and excess reagent was removed with the remediated soil as $CaSO_4$, thereby preventing the salification of the recycled waters. Remediation removed 60%, 78%, and 71% of the Pb from the Meza, Arnoldstein, and Pribram soils, respectively, and reduced the Pb bioaccessibility levels in the simulated human gastrointestinal phase by 5.0, 7.7, and 8.1 times. Residual emissions (EDTA, toxic metals) were reduced with soil aging and remediated soil deposition on a reactive permeable barrier. The solid waste generated from the process totaled 10.8 kg tons^{-1} of the air-dried soil, and the material/energy costs of remediation reached 20.6 € tons^{-1}. These results demonstrate the robustness, efficiency, and safety of this novel approach.

KEYWORDS
cost efficiency, EDTA washing, pilot scale, soil remediation, toxic metals

1 | INTRODUCTION

Land degradation is frequently driven by the misuse of soil resources with agricultural practices and represents the impacts of overgrazing, soil erosion, forest fires, mining, and soil sealing (Novara et al., 2015). However, soil contamination is the main land degradation process that impacts human health and ecosystem fitness (Roy & McDonald, 2015). Soils act as integrators of decades of pollution accumulation, and Pb represents one of the most pervasive and persistent risks to human health (Cai, McBride, & Li, 2016). Pressures on limited soil resources are growing, and metal-contaminated land has been exploited for food production to compensate for the loss of agricultural surfaces (Chen, 2007). Contaminated areas are difficult to avoid, and the proximity of human populations to contaminated land increases the likelihood of ingesting soil pollutants. Pb has pathophysiologic effects on multiple organ systems, neurologic and hematologic systems, and the gastrointestinal tract (Flora, Gupta, & Tiwari, 2012).

A solution to the problem of contaminated soils may involve the reclamation and reuse of metal-contaminated land through the use of effective and cost-efficient remediation technologies. However, few remedial options meet these criteria. The potential use of aminopolycarboxylic chelants for soil Pb removal and bioaccessibility stripping is well known. The most frequently used chelant is ethylenediamine tetraacetate (EDTA) because of its strong capacities for toxic metal removal (Voglar & Lestan, 2014). EDTA is characterized by low levels of biodegradability and high levels of environmental persistence (Bloem, Haneklaus, Haensch, & Schnug, 2017). At environmental concentrations, it is relatively harmless to humans (Lanigan & Yamarik, 2002), and a concentration of 2,200 μg EDTA L^{-1} has been determined to have no effects in water (European Chemicals Bureau, 2004).

Soil washing (extraction, leaching, and flushing) is a straightforward process by which EDTA and toxic metals form water-soluble complexes (chelates), and the washing solution is separated from the solid phase. However, chelates that partly remain in washed soil are

