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Short communication

# Influence of washing treatment on the qualities of heavy metal–contaminated soil

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ABSTRACT

Soil washing is an efficient method to remove heavy metals from contaminated soils; however, soil properties can be altered during remediation processes. Therefore, changes in physicochemical properties after soil-washing treatments were examined to investigate their effects on the ecological properties of soil. Two types of washing processes using different physical separation methods and extraction reagents were applied to Pb- or Zn-contaminated soil. The physicochemical properties of soil changed following physical separation and chemical extraction processes, resulting in deterioration of soil fertility. Soil texture and particle size showed more uniformity regardless of the original properties. Although sorting improved, water-holding capacity decreased because of an increase in sand content. The ecological properties of soil could not be improved completely after the removal of metals due to adverse changes in the physicochemical properties of soil. Therefore, the decision to reuse or recycle remediated soils should reflect changes in soil quality.



## 1. Introduction

Heavy metals are common contaminants in most polluted soils and their toxicity makes them hazardous to the biota (Inoue, 2013; Neuhauser et al., 1985). The presence of heavy metals reduces soil quality, increases the risk of exposure to humans and ecosystems, and restricts healthy land use (Lee et al., 2006; Oves et al., 2012). Soil remediation methods have been applied to reduce these risks and to guarantee safe land use. Soil-washing treatments have been widely used for remediation of heavy metal-polluted soils (Torres et al., 2012). This method is fast, applicable to multiple contaminants, and is easily controlled (Dermont et al., 2008; Elgh-Dalgren et al., 2009). Although soil washing is an efficient and permanent way of removing heavy metals from contaminated soils, soil properties can change during remediation processes. Soil texture might change due to the physical separation process, and its chemical properties can be altered by reagents such as acids/bases, surfactants, chelating agents, salt, or redox agents that are used to transfer metals from solid to aqueous phase (Zupanc et al., 2014). If soil properties are modified during remediation processes, the function of soil may not recover, even after the heavy metal concentration has reduced below the target concentration (Jelusic et al., 2013). Soil is an irreplaceable natural resource (Banwart, 2011), and the significance of remediated soil should be recognized because of its growing demand for reuse. To facilitate the reuse of remediated soil, not only remediation efficiency but also soil characteristics that are related to soil quality or health should be carefully considered. However, related studies have focused on the development of remediation techniques that can be applied to specific contaminants or sites, as well as those that can increase remediation efficiency or reduce the remediation time (Sierra et al., 2014; Udovic and Lestan, 2009; Villa et al., 2010). Therefore, the present study investigated the effects of soil-washing processes on the physicochemical properties of soil. Changes in the ecological properties of soil after two different soil-washing treatments were also examined. These results may aid our understanding of how soil washing affects soil quality.

## 2. Material and methods

Two types of soil-washing treatments were applied to Pb- or Zn-contaminated soils derived from two former military sites. Both soil-washing treatments involved physical separation and extraction processes that used different extraction reagents. The treatments used in case 1 involved physical separation that excluded oversized material (>3 mm) and removed the concentrated soil fraction (<0.075 mm). The heavy metals in the remaining soils were extracted using clean water. The Pb concentration decreased

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from 650 mg/kg of Pb to 62 mg/kg after soil-washing treatment, with a removal efficiency of 90.5%. In case 2, physical separation excluded gravel >2 mm and removed concentrated soil fractions <0.105 mm. The heavy metals in the remaining soils were extracted with diluted hydrochloric acid. The extraction solution was mixed at a solution/soil ratio of 10 to 20 L/kg. To treat 1 $m^3$ of contaminated soil, 28.4 kg of 35% hydrochloric acid was used. The soil used in case 2 was contaminated with 413.45 mg/kg of Zn, which decreased to 160.37 mg/kg after treatment, demonstrating 61.2% removal efficiency. Calcium hydroxide was added to neutralize the soil at the end of the process. Three subsamples of soils were collected before and after washing treatments and used to obtain a representative sample for analysis. Non-contaminated soils (NCSs) were also sampled from the surrounding area for use as control soils and analyzed to compare the ecological properties of treated soil.

The soil texture before and after washing was determined using the pipette method (Gee and Bauder, 1986) and classified according to the International Soil Society textural classification (Moeys, 2012). Soil texture, mean particle diameter (MPD), sorting, skewness, and kurtosis were calculated by the GRADISTAT program using the modified Folk and Ward graphical method (Blott and Pye, 2001). The maximum water-holding capacity (WHC) was measured by soaking 10 g of each soil sample in 50 mL of water for 3 h, which was then dried in an oven at 105 °C for 24 h (OECD/OCDE, 2008). The organic matter content was measured using the loss-on-ignition method (Nelson and Sommers, 1996). Total nitrogen (TN) concentration was measured using the micro-Kjeldahl method (NAAS, 1988), and the available phosphorous concentration ($P_2O_5$) was measured using the Bray P2 soil test procedure (Jones, 2001). The pH was measured using an Orion 4 star pH electrometer (Thermo Electron Co., USA) in 1:5 soil/distilled water suspensions. Exchangeable cation (K, Ca, and Mg) and heavy metal (Pb and Zn) concentrations in soil were analyzed using an inductively coupled plasma system (PerkinElmer, USA).

Activity of dehydrogenase (Casida et al., 1964), β-glucosidase (Eivazi and Tabatabai, 1988), acid phosphatase (Eivazi and Tabatabai, 1977), and arylsulfatase (Tabatabai and Bremner, 1970) were determined. A plant germination and shoot growth experiment was performed using *Brassica juncea* as previously described (Wang et al., 2010). The number of seeds that germinated and the height of the aboveground plant parts were measured at day 14. Changes in earthworm biomass can be a useful indicator of soil quality (Waterhouse et al., 2014). *Eisenia andrei*, a test species recommended by the Organization for Economic Cooperation and Development (OECD), was used in the experiment (OECD/OCDE, 2004). After 21 days of incubation, the weight of individual earthworm was measured. All experiments were performed in triplicate.

Student's *t*-test was used to determine the statistical significance of the effects of soil-washing treatment on the chemical properties of the soil. Significant differences between the NCS and the contaminated soil, before and after soil washing, were established using analyses of variance (ANOVA). ANOVA and *t*-tests were performed using SPSS 18 (SPSS Inc., USA) and a 95% significance level was used. Principal component analysis (PCA) was conducted using CANOCO for Windows (Ter Braak and Šmilauer, 2012), and an ordination diagram was drawn to analyze the physical, chemical, and ecological soil properties.

**Table 1**
Soil texture variables of soil before and after washing.

| | Case 1 | | Case 2 | |
|---|---|---|---|---|
| | Before | After | Before | After |
| Sand (%) | 75.5 | 82.5 | 40.4 | 86.7 |
| Silt (%) | 5.61 | 11.7 | 39.4 | 8.2 |
| Clay (%) | 18.9 | 5.8 | 20.2 | 5.1 |
| MPD (μm) | 67.92 | 185.1 | 32.55 | 357.0 |
| Sorting | 19.74 | 7.829 | 11.48 | 4.624 |
| Skewness | −0.578 | −0.476 | 0.104 | −0.437 |
| Kurtosis | 1.024 | 1.445 | 0.774 | 1.689 |
| WHC (%) | 52.8 | 38.4 | 56.4 | 32.3 |

MPD: mean particle diameter, WHC: maximum water-holding capacity.

**Table 2**
Changes in soil chemical characteristics before and after washing.

| | Case 1 | | Case 2 | |
|---|---|---|---|---|
| | Before | After | Before | After |
| EC | 0.60 | 0.49 | 0.40 | 4.98** |
| pH | 6.31 | 6.35* | 6.71 | 4.77* |
| Organic matter (%) | 3.84 | 2.15** | 5.38 | 3.81* |
| TN (mg/kg) | 243.21 | 119.93 | 89.50 | 54.92 |
| $P_2O_5$ (mg/kg) | 59.92 | 103.28** | 27.97 | 33.88 |
| Exchangeable K | 0.17 | 0.11** | 0.14 | 0.13 |
| Exchangeable Ca | 4.89 | 4.41** | 6.36 | 5.82* |
| Exchangeable Mg | 1.01 | 0.80** | 2.94 | 1.30* |

*Indicates a significant difference at $p < 0.05$ (two-tailed) between soil samples before and after soil washing.
**Indicates a significant difference at $p < 0.01$ (two-tailed).

## 3. Results and discussion

### 3.1. Changes in soil physicochemical properties

Soil-washing treatment affected the physicochemical properties of soil (Tables 1 and 2). The soil texture in cases 1 and 2 changed from sand clay loam to sandy loam, and from clay loam to loamy sand, respectively. Sand content increased in both cases and soil textures became similar after soil-washing treatment regardless of the original texture (Table 1). Physical separation processes during soil-washing treatment affected the overall particle size of the soil used in the study. Mean soil particle diameter increased after soil-washing treatment and changed from very fine sand to sand in case 1, and from very coarse silt to fine sand in case 2. Sorting improved after washing in both cases. Decreased skewness and kurtosis of soil particle distribution also indicated that soil washing improved soil uniformity and reduced variation. Maximum WHC decreased by 27.4% and 42.6% for cases 1 and 2, respectively, after soil-washing treatments. An increased sand fraction facilitates the downward movement of water and of dissolved nutrients in the soil water (Slater and Lesmes, 2002). A decreased clay fraction reduces surface tension and the absorption capacity for water and nutrients in soil. Therefore, decreased WHC can affect the fertility and productivity of plants.

Overall, there was a decrease of 43.95% and 29.15% of organic matter content and 50.69% and 38.64% of TN concentration after soil washing in cases 1 and 2, respectively (Table 2). Exchangeable Ca, Mg, and K concentrations also decreased after washing ($p < 0.05$). Extraction processes used to transfer heavy metals from a solid phase into an aqueous solution affected the organic matter content and nutrient concentration in soil. Exchangeable cations in soil can be simultaneously removed because exchangeable forms of metals are amenable to soil washing (Peters, 1999). The extraction process decreased the organic matter content of soil by removing the soluble phase and particulate organic matter (Ko et al., 2005).

Not all chemical properties were similarly affected. Only case 2 showed a significant increase in electrical conductivity (EC) ($p < 0.01$) and soil pH decreased by 28.9% ($p < 0.05$). This is because different extraction reagents were used in the soil-washing treatments. For case 2, diluted hydrochloric acid solution was used, which decreased soil pH. The reagent solution used in the