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Research article

Impacts of biochar and oyster shells waste on the immobilization of arsenic in highly contaminated soils

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ABSTRACT

Soil contamination is a serious problem with deleterious impacts on global sustainability. Readily available, economic, and highly effective technologies are therefore urgently needed for the rehabilitation of contaminated sites. In this study, two readily available materials prepared from bio-wastes, namely biochar and oyster shell waste, were evaluated as soil amendments to immobilize arsenic in a highly As-contaminated soil (up to 15,000 mgAs/kg). Both biochar and oyster shell waste can effectively reduce arsenic leachability in acid soils. After application of the amendments (2–4% addition, w/w), the exchangeable arsenic fraction decreased from 105.8 to 54.0 mg/kg. The application of 2%biochar +2% oyster shell waste most effectively reduced As levels in the column leaching test by reducing the arsenic concentration in the porewater by 62.3% compared with the treatment without amendments. Biochar and oyster shell waste also reduced soluble As(III) from 374.9 ± 18.8 μg/L to 185.9 ± 16.8 μg/L and As(V) from 119.8 ± 13.0 μg/L to 56.4 ± 2.6 μg/L at a pH value of 4–5. The treatment using 4% (w/w) amendments did not result in sufficient As immobilization in highly contaminated soils; high soluble arsenic concentrations (upto193.0 μg/L)were found in the soil leachate, particularly in the form of As(III), indicating a significant potential to pollute shallow groundwater aquifers. This study provides valuable insights into the use of cost-effective and readily available materials for soil remediation and investigates the mechanisms underlying arsenic immobilization in acidic soils.

1. Introduction

Soil contamination is a significant global problem and poses considerable risks to human health and the environment (Carré et al., 2017). The removal of pollutants from contaminated soils is a complex and lengthy process and associated with high costs, in addition with potential deleterious effects on the soil environment, often compromising crop production (Khalid et al., 2017). Therefore, the development of cost-effective and environmentally friendly soil remediation techniques is crucial to support global sustainability. Reducing the mobility or bioavailability of soil contaminants, referred to as immobilization/stabilization, represents an effective remediation technology for a wide variety of soil contaminants (Bolan et al., 2014). In this sense, materials which are cost-effective and readily available are needed to develop efficient immobilization technologies at commercial scales.

Arsenic (As) contamination is a ubiquitous environmental problem worldwide (Singh et al., 2015). Elevated As-levels in soils are a result of both natural processes (i.e., weathering and volcanic emissions) and anthropogenic activities such as mining, fossil fuel combustion, and the disposal of industrial waste (Gupta et al., 2017). In addition, the use of As as an additive to livestock feed, particularly for poultry, has resulted in high As concentrations in farmyard manure, potentially leading to the spread of As into agricultural fields (Yang et al., 2017). Soil As can be easily solubilized in water in the reduced form, which explains the ubiquitous presence of As in shallow aquifers and groundwater (Masscheleyn et al., 1991; Nordstrom, 2002). Reducing the solubility potential is therefore the key to protect shallow groundwater aquifers from As

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contamination. Several studies have focused on the reduction of mobilizable and bioavailable arsenic fractions in soil environments, using a variety of stabilization agents such as (hydro) oxides, clay minerals, biochar, and solid waste compost (Garcia-Sanchez et al., 2002; Hartley et al., 2009; Beesley et al., 2013). In the case of in-situ remediation, metal oxides and their precursors have been widely tested and used as stabilizing amendments for As-contaminated soils (Komárek et al., 2013). However, this approach is still challenging and costly due to the large impacts of many factors on specific immobilization mechanisms, such as soil properties (i.e., pH, oxidation-reduction potential), which play a crucial role in interaction mechanisms between amendments and arsenic in any specific treatment (Karna et al., 2017). Some wastes materials such as composts (derived from sewage sludges and other municipal sources) and industrial by-products (i.e., red-mud) can also achieve effectively arsenic immobilization by breaking the pollutant receptor linkage (reviewed by Gadepalle et al., 2007). These remediation techniques are cost-effective and socially acceptable compared to most traditional methods, as the materials they utilized were from the waste stream and could thereby make a dual contribution to environmental sustainability.

According to a 9-year survey, about 26 million ha of farmland in China were polluted with heavy metals and metalloids (Zhao et al., 2014). These contaminated areas pose chronic risks to human health via various exposure pathways such as groundwater and the food chain. However, the current remediation strategies are not satisfying, for a large challenge in the rehabilitation of contaminated sites nationwide is not only the requirement of ongoing funding, but also the development of cost-effective and readily available technologies, especially in remote and less developed regions (Qu et al., 2016). Such areas are more susceptible to soil contamination due to the directly shallow groundwater drinking and less intentional pollution prevention. In this context, we evaluated the use of two waste by-products as soil amendments to reduce leachability from highly As-contaminated soil, with the aim to develop a cost-effective and readily available in-situ technology for the protection of shallow groundwater in sites contaminated with metals/metalloids. The biochar we selected is highly recommended as a soil amendment for contaminated soils because of its high sorption capability to various organic/inorganic contaminants. It is widely accepted as a green environmental sorbent due to its cost-effective production from biowaste resources (Beesley et al., 2011; Gwenzi et al., 2017). Oyster shells are a waste product of oyster farming and have become a serious environmental problem due to the locally random disposal in China. Oyster shell waste is particularly rich in $CaCO_3$ and CaO components, which might serve as a liming material for the stabilization of metal-contaminated soil due to the formation of insoluble metal hydroxides at alkaline pH levels (Ok et al., 2011; Moon et al., 2016). In addition, Shell waste has also shown its capacity of adsorption of arsenic and has proved to have positive effects on soil arsenic retention on forest and vineyard soils (Seco-Reigosa et al., 2013).

2. Material and methods

2.1. Soils and amendment preparation

Soil samples (about 0–30 cm depth) were taken from one site with frequently mining activities including mineral acquisition and smelting in Guangxi of China (23–25° N, 106–109° E). Arsenic concentrations reach 15,076.8 ± 726.4 mg/kg; to assess the leaching potential into shallow aquifers in the contaminated site, uncontaminated soil from local arable land was also collected and used in column leaching experiments. The specific soil properties are presented in Table 1. After removing small stones and other debris, the soil was air-dried and crushed to pass through a 2-mm sieve before amendment application. As soil amendments, we used biochar and oyster shell waste. The biochar was produced from rice straw in a muffle furnace with N_2 flow and 400 °C for around 4 h; subsequently, it was forced through a 2-mm stainless steel sieve before being mixed with the soil. Oyster shell waste was collected from a local oyster farm, air-dried, crushed, and ground to a fine powder (<0.3-mm mesh).

2.2. Column leaching experiment

A column leaching study was used to evaluate the potential of biochar and oyster shell waste to immobilize arsenic in contaminated soil, thereby preventing it from reaching the shallow groundwater aquifer. The procedure was carried out under saturated conditions in polyethylene columns (length 130 cm × internal diameter 11 cm) packed with highly As-contaminated soil (either unamended or thoroughly mixed with amendments.) and uncontaminated soil (see Fig. 1). The two amendments were tested at varying ratios (w/w dry weight amendment: soil), with each treatment being performed in triplicate. The application rates were set up according to the following treatments:

- T1, contaminated soil was mixed well with 2.0% (weight) biochar;
- T2, contaminated soil was mixed well with 4.0% (weight) biochar;
- T3, contaminated soil was mixed well with 1.0% biochar and 1.0% oyster shell waste;
- T4, contaminated soil was mixed well with 2.0% biochar and 2.0% oyster shell waste;
- T5, control treatment without any amendments.

The columns were divided into two layers (Fig. 1): the upper layer (5–35 cm) was packed with As-contaminated soil with or without amendments, and the lower layer (35–110 cm) was packed with uncontaminated soil. The top and bottom of the column were filled with a 5-cm layer of quartz sand (grain size <0.5 mm) to protect the column from leaching. The columns were equipped with Rhizon Soil Solution Samplers (10 cm long Rhizon® samplers, Rhizosphere Research Products, the Netherlands) at every 30 cm to collect porewater for the assessment of the leaching potential of arsenic. After 48 h equilibrium, ultra-pure water (milli-Q water purification system, Millipore Corp., USA) was then added slowly to the columns until the appearance of the leaching solution at the end of each column to simulate saturated conditions. The porewater samples were then collected from the Rhizon Soil Solution Samplers, filtered using 0.45-μm nylon membranes, and stored at 4 °C prior to analysis. After the leaching test, the soil column was evenly cut into twenty four 5-cm sections, air-dried, ground to a size of 0.15 mm, and stored until analysis.

2.3. Batch leaching experiment

A batch leaching experiment was performed to evaluate the immobilization of arsenic after soil amendment with biochar and oyster shell waste. Solutions with different pH levels (pH4.0, 4.5, 5.0, 5.5, 6.0, 6.5, and 7.0) were applied to simulate the acidity conditions of shallow aquifers in the southern regions of China, which are mainly a product of frequent acid rain (Larssen et al., 1999; Duan et al., 2016). A liquid/solid ratio (L/S) of 10 (according to EN-12457-2, 2002) was used to estimate the leachable As concentrations in the untreated and amended soils. For the leaching tests, 2.00 g of treated soil were suspended in 20 mL of simulated solution in 50-mL plastic centrifuge tubes and shaken at room