



The effectiveness of spent coffee grounds and its biochar on the amelioration of heavy metals-contaminated water and soil using chemical and biological assessments



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ARTICLE INFO

Article history:

Received 14 May 2014

Received in revised form

30 June 2014

Accepted 3 July 2014

Available online 28 August 2014

Keywords:

Acid mine drainage

Biochar

Phytotoxicity

Remediation evaluation

Trace elements

ABSTRACT

Spent coffee grounds (SCG) and charred spent coffee grounds (SCG-char) have been widely used to adsorb or to amend heavy metals that contaminate water or soil and their success is usually assessed by chemical analysis. In this work, the effects of SCG and SCG-char on metal-contaminated water and soil were evaluated using chemical and biological assessments; a phytotoxicity test using bok choy (*Brassica campestris* L. ssp. *chinensis* Just.) was conducted for the biological assessment. When SCG and SCG-char were applied to acid mine drainage, the heavy metal concentrations were decreased and the pH was increased. However, for SCG, the phytotoxicity increased because a massive amount of dissolved organic carbon was released from SCG. In contrast, SCG-char did not exhibit this phenomenon because any easily released organic matter was removed during pyrolysis. While the bioavailable heavy metal content decreased in soils treated with SCG or SCG-char, the phytotoxicity only rose after SCG treatment. According to our statistical methodology, bioavailable Pb, Cu and As, as well as the electrical conductivity representing an increase in organic content, affected the phytotoxicity of soil. Therefore, applying SCG during environment remediation requires careful biological assessments and evaluations of the efficiency of this remediation technology.

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1. Introduction

Heavy metals have been mobilized and dispersed into the environment by human actions, including metal plating, mining operations and other industrial activities, more rapidly than the natural circulation processes (Gratão et al., 2005; Han et al., 2013; Inyang et al., 2012; Lee et al., 2011). The elevated heavy metal concentrations in the environment are concerning because these deposits are not biodegradable and may accumulate in living organisms, exhibiting toxicity (Bailey et al., 1999; Martínez et al., 2006; Zeng et al., 2011).

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Many techniques have been proposed to reduce heavy metals and their effects in water systems, including ion exchange, membrane processing and electrolytic methods; however, most of these techniques are costly and might generate by-products or toxic intermediates during the treatment process (Minamisawa et al., 2004). Of these various techniques, removing heavy metals by adsorbing them has remained the most versatile because it is a relatively simple and cost effective technique (Bailey et al., 1999; Martínez et al., 2006); in particular, appropriately modified agricultural wastes have a high capacity for heavy metal adsorption (Demirbas, 2008). For soil systems, the proposed remediation technologies are based on the excavation, transport, washing, flushing, and landfilling of contaminated soils; these techniques competently reduce the risks of heavy metals but remain too expensive to apply broadly (Lee et al., 2009). Compared to other remediation technologies, chemical stabilization is less expensive, possibly providing a long-term solution for heavy metal

contamination (Lee et al., 2009). Many investigations have sought to find inexpensive and locally available adsorbents to remove and stabilize heavy metals; these materials have competitive adsorption capacities and include clay minerals, chitosan, and industrial waste products, among others (Bailey et al., 1999; Martínez et al., 2006). Demirbas (2008) examined the utility of agricultural by-products, including tea waste, fruit peels, hulls and barks, during heavy metal removal in both industrial and municipal wastewater.

Coffee is one of the most abundant agricultural products and the second most traded commodity worldwide (Boonamnuayvitaya et al., 2004; Kondamudi et al., 2008; Tsai et al., 2012); consequently, large amounts of coffee wastes, especially spent coffee grounds (SCG), are generated. SCG are the powdered organic remnants obtained after coffee is extracted from the beans using steam under high pressure. Some SCG have been utilized during the production of biodiesel, fuel pellets, and composts, as well as in animal feeds (Kondamudi et al., 2008). SCG are small particles ($\approx 20 \mu\text{m}$) of organic material composed of fiber (>50%) and complex lignin structures with large surface areas ($7.5 \text{ m}^2 \text{ g}^{-1}$) (Kondamudi et al., 2008; Fiol et al., 2008). Additionally, SCG contain an embedded tannin material that provides polyhydroxy polyphenol functional groups; these groups are able to adsorb heavy metals (Boonamnuayvitaya et al., 2004; Fiol et al., 2008; Minamisawa et al., 2004). Consequently, the applications of recycled SCG have been studied in various research fields. According to the recent review by Kim et al. (2012), recent studies using SCG focus mainly on the extraction of useful substances from the SCG or pollutant remediation (e.g., heavy metals, metalloids, organic compounds) in the aqueous phase. In addition, Fenoll et al. (2011) demonstrated that SCG is a versatile barrier that reduces the mobility of pesticides in the soil; common soil management practices in agriculture include applying organic carbon, such as SCG. Meanwhile, Limousy et al. (2013) and Silva et al. (1998) revealed that, when used as an alternative fuel, the SCG can burn easily in a boiler and a heat value (kcal/kg dry) of SCG was higher than other agriculture biomass residues (e.g., rice husk, wheat straw, corn ears, sugar cane).

Biochar is produced from the thermal degradation of organic substances in the absence of air; this combustible solid may be incinerated to generate heat energy (Griffith et al., 2013; Laird et al., 2009) alongside other agricultural waste. From these studies, biochar has demonstrated superior pollutant removal efficiency in solution and enhanced the fertility and carbon storage in soil by reducing the acidity and increasing the water holding capacity (Griffith et al., 2013; Han et al., 2013; Inyang et al., 2012; Laird et al., 2009). However, Laird et al. (2009) disclosed that plant growth decreases in soils treated with biochar containing large quantities of volatile matter; these volatile components immobilized the available nutrients, causing stagnation in the yield or biomass. Nevertheless, the research related to biochar from SCG has grown more popular in various fields for its emerging and promising applications in treatment technology (Felber et al., 2012; Inyang et al., 2012; Limousy et al., 2013; Tsai et al., 2012).

The SCG consist of organic material that requires large amounts of oxygen to decompose; when discharged directly into the environment, SCG may cause ecological toxicity because it contains various organic compounds (e.g., phenol, chlorogenic acid, hydroxycinnamic acid, quinic acid derivatives) (Clifford, 2000; Limousy et al., 2013; Silva et al., 1998). Additionally some studies indicated that certain organic compounds may be deleterious to plant growth (Griffith et al., 2013). To our knowledge, most studies using SCG or biochar utilized chemical analysis targeted only on the pollutants; however, studies describing the biological toxicity remain incomplete.

Therefore, in this study, we sought to evaluate the effects of SCG and biochar on the amelioration of heavy metal contaminated water (acid mine drainage) and soil (abandoned mine area) using both chemical and biological assessments. For the chemical assessment, the chemical species were quantitatively analyzed, and the biological assessment was performed via phytotoxicity tests.

2. Materials and methods

2.1. Preparation of SCG and SCG-char

The spent coffee grounds (SCG) were obtained from a local coffee shop in Seoul, Korea. The SCG were air dried and passed through a 0.5 mm sieve. The sieved SCG were stored without any physical or chemical pretreatment in polyethylene bottles until used. The pH and EC were measured in a 1:5 of solid:water suspension with a combination pH-EC meter (Thermo Orion 920A); the amount of organic matter was determined using the mass lost upon ignition at 400 °C for 16 h. To produce SCG-char, the SCG were slowly pyrolyzed in a temperature-controlled electric furnace. The SCG were heated at approximately 20 °C/min up to 400 °C and held for 30 min (Bruun et al., 2011). The SCG-char yield was 36% and the resulting SCG-char was ground and sieved to less than 0.5 mm before stored in polyethylene bottles.

2.2. Characterization of water and soil samples

Acid mine drainage (AMD) samples were obtained from the Munbaek gold mine in Chungcheongbuk-do, South Korea. The AMD sample was collected in new polyethylene bottles and moved to the laboratory in a cooler. Raw AMD samples were filtered (0.45 μm) to remove any suspended particles and stored at 4 °C until used. The pH and EC of the AMD were determined with a combination pH-EC meter. The total dissolved organic carbon (DOC) was analyzed using an automatic total organic carbon analyzer (Shimadzu TOC-VCPH). The AMD's heavy metal concentrations were determined using ICP-OES (Vista PRO, Varian, USA) after acidification with 2% (v/v) HNO₃.

Three soil samples were collected from agricultural land adjacent to the abandoned Gahak zinc mine in Gyeonggi-do, South Korea. The soil samples were air-dried and passed through a 2 mm sieve. The soil pH and electrical conductivity were determined using a 1:5 soil:water suspension. The total organic carbon and Kjeldahl nitrogen were determined using the Walkely-Black and Kjeldahl methods, respectively (Bremner, 1996; Nelson and Sommers, 1996). The exchangeable cations were identified using the saturated ammonium acetate method (Thomas, 1982). The pseudo-total trace element concentrations were determined by digesting samples with aqua regia according to ISO 11466 (ISO, 1995) and using ICP-OES.

2.3. Chemical assessment of AMD and soil samples treated using SCG and SCG-char

Batch sorption tests were conducted to remove the heavy metals from AMD by mixing with adsorbents (SCG and SCG-char, respectively) in a 1:10 (solid-solution) ratio for 2 h using an end-over-end shaker. The optimum adsorption conditions were elucidated by preliminary experiments using synthetic solutions; the maximum adsorption capacities of the adsorbents were calculated using their Langmuir isotherms. The suspension was passed through a 0.45 μm filter and the remaining heavy metal content in the filtrate was determined using ICP-OES after acidification. Every batch test was conducted in triplicate at a constant temperature (25 °C).

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