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Continuous leaching modifies the surface properties and metal(loid) sorption of sludge-derived biochar

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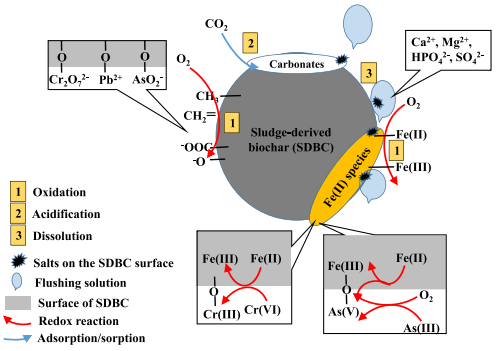
HIGHLIGHTS

- Continuous leaching increases surface acidity, CEC, and O-containing groups of SDBC.
- Sorption of Pb(II), Cr(VI), and As(III) is enhanced on SDBC after 1–4 month leaching.
- Hydroxyl/carboxyl groups and Fe(II) species play key roles in metal(loid) sorption.
- Surface oxidation/acidification by leaching is less significant than atmospheric exposure.

GRAPHICAL ABSTRACT



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ABSTRACT

After the application of sludge derived biochar (SDBC) for soil stabilization, it is subjected to continuous leaching that may change its surface properties and metal(loid) immobilization performance. This study simulated the continuous leaching through the fresh SDBC sample in columns with unsaturated and saturated zones under flushing with 0.01 M $NaNO_3$ solution (pH 5.5) and acidic solution (pH adjusted to 3.2 by HNO_3:H_2SO_4 = 1:2), respectively. The resultant changes were assessed in terms of the SDBC surface characteristics and metal(loid) sorption capacities. Continuous leaching was found to gradually decrease the density of basic functional groups and increase the density of carboxyl groups as well as cation exchange capacity on the SDBC surface. It was attributed to the surface acidification and oxidation process by the leaching process, yet it occurred to a lesser extent than the atmospheric exposure. Continuous leaching increased Pb(II), Cr(VI), and As(III) sorption capacity of the SDBC, probably because the increase in carboxyl groups promoted inner-sphere complexation and Fe oxidation as revealed by spectroscopic analysis. It was noteworthy that the SDBC in the unsaturated and saturated zones under continuous leaching displayed distinctive effects on metal(loid) sorption capacity than the atmospheric

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exposure. Future investigations are needed for understanding the fate and interactions of the SDBC under varying redox conditions and intermittent leaching process.

1. Introduction

Soil stabilization with amendment materials has been widely studied for contaminated site remediation (Tsang et al., 2014; Rajapaksha et al., 2016; Beiyuan et al., 2018). Among the stabilizing agents available for scalable applications, biochar has received extensive attention because of its benefits for improving the soil quality by replenishing nutrients, retaining moisture, and enhancing microbial community abundance (Hossain et al., 2010; Igalavithana et al., 2017; O'Connor et al., 2018), while providing high adsorption capacity for metal(loid) immobilization (Beiyuan et al., 2016; Ahmad et al., 2017). However, the efficacy of biochar is related to its physicochemical properties and surface chemistry, which are dominated by the pyrolysis conditions and feedstock used such as agricultural residues, wood waste, and sewage sludge (Wesenbeeck et al., 2014; Tan et al., 2015; Suliman et al., 2016).

Different from the plant-derived biochar, the sewage sludge-derived biochar (SDBC) has relatively low organic carbon content but high contents of inorganic minerals as well as alkali and alkaline earth metals (Bian et al., 2014; Zhang et al., 2015, 2017), which can provide active sites for metal complexation and alkalinity for precipitation. The SDBC is considered as a highly efficient sorbent for simultaneous immobilization of coexisting metals and metalloids, such as cationic Pb(II) together with oxyanionic Cr(VI) and As(III) species (Zhang et al., 2013; Fang et al., 2016). In general, the interactions between SDBC and metal(loid) species include electrostatic attraction with charged surfaces, π electron system from C═C bonds, surface complexation with oxygen-containing functional groups, as well as precipitation and/or co-precipitation with other mineral phases (Lu et al., 2012; Zhang et al., 2017). The potential risks related to metal(loid)s leaching from the SDBC itself are considered acceptable and much lower than the direct application of sewage sludge (Hwang et al., 2007; Fang et al., 2016). Hence, the SDBC seems to be one of the promising stabilizing materials for diverse areas of metal(loid)s-contaminated agricultural soils.

However, there remain questions about the change of surface properties of biochar over time when exposed to the natural environment due to ageing via a series of geochemical and biogeochemical reactions (Peak et al., 2014; Wang et al., 2017). Natural oxidation and ageing with higher temperature or longer incubation time was found to increase the oxygen content, surface acidity, and negative surface charge, while it decreased the carbon content, pH, surface basicity, and the point of zero net charge (Cheng et al., 2008; Cheng and Lehmann, 2009). The long-term geochemical weathering also led to the release of the mineral fraction from the biochar and the formation of carbonyl and carboxylic functional groups (Yao et al., 2010). These changes in the surface properties of biochar, especially the increase in the O/C ratio and introduction of carboxyl groups, changed its affinity and sorption behaviour for metal(loid)s (Qian and Chen, 2014; Ghaffar et al., 2015; Qian et al., 2015). Recent results further suggested that ageing of biochar changed its functionality and capacity for the adsorption of cationic and anionic species because chemical oxidation introduced carboxyl and phenol groups, reduced oxonium groups, and transformed pyridine to pyridone (Mia et al., 2017). Furthermore, intermittent flushing and drainage was often encountered during biochar application in field (Lau et al., 2017), and the change of redox conditions affected metal(loid) immobilization in contaminated sites with biochar, such as the dissolution of Fe/Mn (hydr)oxides and (co-)precipitation between sulphides and metal(-loid)s under lower Eh conditions (Beiyuan et al., 2017).

Therefore, in this study the SDBC samples were exposed to the air in the room temperature and flushed with different solutions for varying duration to simulate the ageing processes in the atmosphere, unsaturated and saturated zones, respectively. The aged SDBC samples were extensively characterized with physicochemical and spectroscopic methods, and the corresponding effects on the Pb(II), Cr(VI), As(III) sorption were evaluated.

2. Materials and methods

2.1. SDBC preparation

The SDBC samples used in this study were prepared through pyrolyzing the dried municipal sewage sludge from Lijiao sewage treatment plant in Guangzhou (23°20′N, 113°30′E) in a LTKC-6-12 pipe oven (Blue Sky, Hangzhou, China) filled with N_2 gas at 400 °C for 2 h. The details were given in our previous studies (Zhang et al., 2015; Wang et al., 2017). The produced SDBC samples were stored airtight in a freezer before characterization, column leaching, and batch sorption experiments.

2.2. Continuous leaching in column with unsaturation and saturation zones

To simulate the effect of continuous leaching in soil application, the SDBC samples were loaded in a leaching column (as illustrated in Fig. 1) and continuously flushed with various solutions. Two packages of 40.0 g of fresh SDBC samples (wrapped with 200 mesh-glass fiber cloth) were respectively placed in unsaturated and saturated zones of the column leaching device (25 mm internal diameter; 500 mm length). The bottom section (30 mm) of the column was packed with glass beads to maintain even flow distribution for effluents. The SDBC samples were then continuously flushed from top with 0.01 mol L^{-1} $NaNO_3$ solution (pH 5.5) and the synthetic acid solution (0.01 mol L^{-1} $NaNO_3$ solution with pH 3.2 adjusted by HNO_3:H_2SO_4 = 2:1), respectively. The flushing rate was maintained to 275 ± 5 μL min^{-1} by a DDB 320 peristaltic pump (Zhixin, Shanghai, China). At the end of 15, 30, 60, and 120 d of flushing, approximately 3.0 g of SDBC samples were collected for characterization and subsequent sorption experiments.

To probe into the influence of atmospheric exposure on SDBC in the unsaturated zone, 40.0 g of the fresh SDBC sample was uniformly placed in a 20 cm × 30 cm plate with 1.5 mm thickness, and then exposed to air at 25 ± 1.0 °C and relative humidity of 75% in ETDH-9503 N humidifier (Encyclopedia Theo, Hangzhou, China). The SDBC samples were daily mixed for homogeneity, and the experimental details were reported earlier (Wang et al., 2017). Approximately 3.0 g of SDBCs was similarly collected after 30 d for characterization and subsequent sorption experiments. Our previous study (Wang et al., 2017) reported that the change of surface properties induced by atmospheric exposure was substantial only in the first 30 d, and after that the corresponding change became less significant. Therefore, the SDBC sample subjected to 30-d atmospheric exposure was herein used as the benchmark to explain the different changes of surface properties on the SDBC located in the saturated and unsaturated zones during continuous flushing process.

2.3. SDBCs characterization

Particle size distribution of SDBC samples was evaluated by using a LS-230 Particle Size Analyzer (Beckman Coulter, Germany). The surface area was determined by a NOVA4200e-Quanta specific surface area and pore size distribution analyzer (Quantachrome Instrument, USA). The pH values of SDBC samples were determined by FE20K digital pH meter (Mettler-Toledo, Switzerland) in the supernatant of the mixture