



## Cadmium adsorption on plant- and manure-derived biochar and biochar-amended sandy soils: Impact of bulk and surface properties



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### HIGHLIGHTS

- The minerals of biochar-amended soils were mainly covered by biochars.
- The polarity of the biochars influenced the adsorption of Cd<sup>2+</sup>.
- Plant- and manure-derived biochars had different impact on Cd<sup>2+</sup> adsorption on soil.
- Swine manure-derived biochars were effective in immobilizing Cd<sup>2+</sup> in soil.

### ARTICLE INFO

#### Article history:

Received 25 October 2013

Received in revised form 7 April 2014

Accepted 13 April 2014

Handling Editor: Klaus Kümmerer

#### Keywords:

Biochar

Cd<sup>2+</sup>

Adsorption

Polarity

Biochar-amended soil

### ABSTRACT

To investigate the role of the bulk and surface composition of both biochar and biochar-amended soils in the adsorption of Cd<sup>2+</sup>, as well as the influence of different biochars added to the soils on Cd<sup>2+</sup> adsorption, swine-manure-derived biochars (BSs) and wheat-straw-derived biochars (BWs) were produced at 300, 450, and 600 °C. These biochars were added to a sandy soil to investigate the effect of biochars on the adsorption of Cd<sup>2+</sup> by soil. The significantly higher surface C content of the amended soils compared to their bulk C content suggests that the minerals of the biochar-amended soils are most likely covered primarily by biochars. The maximum adsorption capacity ( $Q_{max, total}$ ) of the BSs was 10–15 times higher than that of the BWs due to the high polarity and ash content of the BSs. The polarity ((N + O)/C) of the low-temperature biochars greatly affected their Cd<sup>2+</sup> adsorption. The  $Q_{max, total}$  of the BS-amended soils increased with increasing dose, whereas the  $Q_{max, total}$  of the BW-amended soils showed the opposite behavior, which was attributed to the different surface composition characteristics of the two types of soil. The BSs were more effective in immobilizing Cd<sup>2+</sup> upon application to the soil relative to the BWs. This study elucidates the spatial distribution of biochars in biochar-amended soils and highlights the importance of the surface composition of the investigated samples in Cd<sup>2+</sup> adsorption.

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### 1. Introduction

Because heavy metals are not degraded in soil, modern remediation approaches have increasingly focused on in situ environmentally friendly techniques, such as the application of soil amendments (Fellet et al., 2011) or phytoextraction (Meers et al., 2005), to stabilize heavy metals and thereby reduce their toxicity. Heavy metals in the soil are stabilized by these soil amendments via the processes of adsorption, binding, or co-precipitation, which have been extensively researched (Komarek et al., 2013). In recent

years, scientists have paid considerable attention to carbon-rich materials (e.g., biochar) as soil amendments (Cao et al., 2009; Uchimiya et al., 2010). According to the International Biochar Initiative (IBI), biochar is a charcoal that can be applied to soil for both agricultural and environmental gains (Manyà, 2012). The addition of biochars to soils has been assessed to be a soil remediation method due to the potential of biochars to sequester carbon (Lehmann, 2007) while increasing soil fertility (Van Zwieten et al., 2010). Recently, attention has been paid to biochars not only with respect to the agro-environmental benefits of adding biochar to soil but also the expansion of the use of biochar as a sorbent for organic (Chen and Yuan, 2011; Sun et al., 2011) and inorganic contaminants (Beesley and Marmiroli, 2011; Xu et al., 2013).

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Unlike the study of the adsorptive behavior of biochar with respect to heavy metals, information about the adsorption of heavy metals by soils amended with biochar is still very limited. Fellet et al. (2011) suggested that the bioavailability of Cd, Pb, Tl, and Zn in mine tailings decreased after adding biochar derived from orchard prune residue. Jiang et al. (2012) demonstrated that the enhancement of Pb adsorption by soil amended with rice-straw-derived biochar increased with the amount of biochar added. Uchimiya et al. (2012) found that the addition of oxidized biochars to soil significantly improved Pb, Cu, and Zn stabilization compared to that of unoxidized biochars due to the plentiful carboxyl functional groups of the oxidized biochars. However, the difference in the adsorption properties between plant-derived biochars (C-dominated) and manure-derived biochars (ash-dominated), as well as their influence on the adsorption of heavy metals after their application to soil, has not been addressed in detail. Moreover, a previous study reported that most minerals in soil and sediment particles were mainly covered by organic matter (OM) (Mikutta et al., 2009); however, another study was supported by abundant evidence that substantial parts of the mineral surfaces are not covered by OM (Kleber and Johnson, 2010). Biochar generally consists of abundant OM because it is a carbon-rich product (Lehmann and Joseph, 2009). With the addition of biochar to soil, it is expected that biochars interact with the minerals in soil, according to previous studies (Brodowski et al., 2006; Laird et al., 2008; Liang et al., 2008); however, the spatial distribution caused by the interaction with minerals after addition to soil remains unclear. It is hypothesized that the spatial distribution of biochar within soils should affect the adsorption of heavy metals. We hypothesize that the minerals (e.g., ash) within high-mineral biochars and the spatial distribution of biochar within soils should affect the adsorption of heavy metals. Additionally, the difference in the bulk and surface elemental contents (e.g., C, O, and N) of soil OM samples has been identified (Lehmann et al., 2008; Yang et al., 2011; Sun et al., 2013a). Furthermore, the surface polarity of OM within soil samples has been observed to have a considerable effect on the sorption of organic compounds (Yang et al., 2011). To the best of our knowledge, the surface compositions of biochar-amended soils have been minimally investigated. The surface properties of biochar-amended soils and their roles in the sorption of heavy metals are also poorly understood.

Therefore, the objectives of this study were to investigate the role of the bulk and surface composition of biochar and biochar-amended soils in the adsorption of heavy metals and to identify the differences in the adsorption of heavy metals between plant-derived biochar and manure-derived biochar, as well as the differences with respect to the biochars' effect on heavy metal adsorption after being added to soil. In this study, Cd<sup>2+</sup>, as a model heavy metal, was selected to investigate the adsorption of heavy metals on biochar and biochar-amended soil due to its importation into soils via the process of fertilizing, manufacturing and mining (Liu et al., 2008) leading to Cd pollution in soils.

## 2. Materials and methods

### 2.1. Chemicals

All chemical reagents were of analytical reagent grade. Distilled, deionized water (DDW) with a resistivity of 18 MΩ cm (Millipore Corp., Milford, MA) was used in all procedures. Stock solution (1000 mg L<sup>-1</sup>) of Cd<sup>2+</sup> were prepared with Cd(NO<sub>3</sub>)<sub>2</sub>·4H<sub>2</sub>O and DDW.

### 2.2. Sorbents

The sandy soil used in this study was collected from the TongZhou district of Beijing, China. The soil was air-dried at room

temperature, and its soil moisture content was 0.45%. Biological remnants and large debris were removed from the soil, and the soil was passed through a 2-mm nylon sieve for further use. The pH value of the soil was 8.63, and its elemental composition is listed in Table 1.

Wheat straw and swine manure were pyrolyzed at different temperatures (i.e., 300, 450, and 600 °C) and were used to produce three wheat-straw-derived biochars (BW<sub>s</sub>) and three swine-manure-derived biochars (BS<sub>s</sub>). The samples are hereafter referred to as BW<sub>X</sub> and BS<sub>X</sub>, with X indicating the final heating treatment temperature (HTT) (i.e., 300, 450, or 600 °C). The process used to prepare the biochars is described elsewhere (Sun et al., 2013b). Briefly, the dried feedstocks were rinsed with deionized water and ground with a disintegrator. The feedstocks were then passed through a 60 mesh for further use. The feedstocks were then charred at different temperatures (i.e., 300, 450, or 600 °C) for 1 h under oxygen-limited conditions in a muffle furnace. The HTTs were increased to the desired values (300, 450, or 600 °C) at a ramp rate of 10 °C min<sup>-1</sup>. Then, the biochars were washed with 0.1 M HCl followed by deionized water until they became neutral and were subsequently oven-dried at 105 °C and gently ground and homogenized to pass through a 0.25-mm sieve (60 mesh).

The biochar-amended soils used in the experiments were prepared by mixing the abovementioned soil and the two types of biochar in different ratios. The contents of the biochar materials in the amended soils were 5% and 20% for the BW<sub>s</sub> and BS<sub>s</sub>, respectively, produced at 300, 450, or 600 °C. The biochar-amended soils were mixed thoroughly on a rotary shaker for 30 d at room temperature before being used as sorbents for sorption experiments (Yu et al., 2006). The biochar-amended soils containing the BW<sub>s</sub> and BS<sub>s</sub> were denoted SW<sub>s</sub> and SS<sub>s</sub>, respectively.

### 2.3. Sorbent characterization

The bulk C, H, O, and N content of the samples was determined by an Elementar Vario ELIII elemental analyzer via complete combustion. Ash content of the biochars was measured by heating samples at 750 °C for 4 h. The surface area (SA-CO<sub>2</sub>) of the samples was obtained by gas adsorption using an Autosorb-1 gas analyzer (Quantachrome Instrument Corp., Boynton Beach, FL) using CO<sub>2</sub> isotherm at 273 K, and surface elemental composition of all sorbents were determined using X-ray photoelectron spectroscopy (XPS). Also, the spectra of both the solid-state cross-polarization magic-angle-spinning <sup>13</sup>C NMR and the Fourier transform infrared (FTIR) of the biochars were characterized. A more detailed account of these characterization procedures and peak assignments for the <sup>13</sup>C NMR procedure is provided elsewhere (Sun et al., 2013a, b).

### 2.4. Adsorption experiments

Because the adsorption capacities of BW300 and BW450 were similar in preliminary experiments, the adsorption of BW300 was not tested in this study. A batch equilibration technique was used to obtain the adsorption isotherm of Cd<sup>2+</sup> by 19 sorbents. The test solutions contained Cd<sup>2+</sup> concentrations ranging from 0.3 to 100 mg L<sup>-1</sup>. The background solution contained 0.01 M NaNO<sub>3</sub> in deionized water to maintain a constant ionic strength. The pH of the Cd<sup>2+</sup> solutions was adjusted to 4.5 with either HNO<sub>3</sub> or NaOH to prevent Cd<sup>2+</sup> from precipitating after being added to the samples. All of the sorbents (2–160 mg) were weighed into 8-mL polypropylene centrifuge tubes, and the Cd<sup>2+</sup> solutions were added to these tubes and horizontally shaken at 120 rpm for 24 h at room temperature. Preliminary experiments showed that the adsorption equilibrium was reached within 24 h. The blanks consisted of Cd<sup>2+</sup> solution without sorbents. After 24 h, all tubes were centrifuged (11 000g for 20 min). The supernatant was withdrawn from each

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