



Capacity and mechanisms of ammonium and cadmium sorption on different wetland-plant derived biochars



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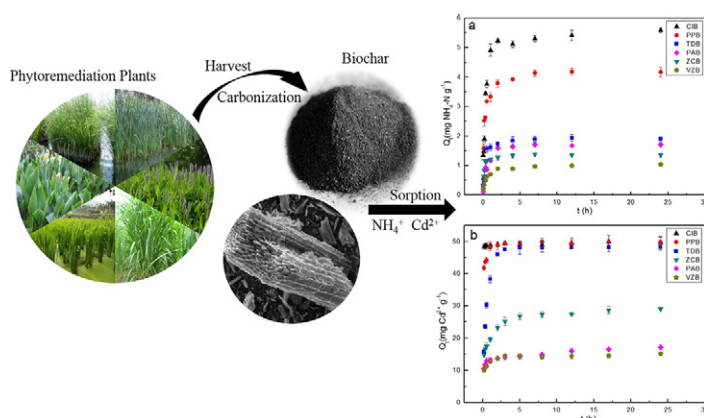
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HIGHLIGHTS

- Biochars varied in physicochemical properties and adsorption capacity.
- *Canna indica* derived biochar has a high sorption capacity for Cd^{2+} .
- NH_4^+ and Cd^{2+} sorption on biochars fits a pseudo second order and Langmuir model.
- Sorption mechanism is related to complexation, cation exchange and precipitation.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 30 March 2015

Received in revised form 13 August 2015

Accepted 1 September 2015

Available online 16 September 2015

Editor: Ajit Sarmah

Key words:

Biochar

Wetland plants

Contaminants

Sorption characteristics

Mechanisms

ABSTRACT

The objective of this study was to investigate the relationship between $\text{Cd}^{2+}/\text{NH}_4^+$ sorption and physicochemical properties of biochars produced from different wetland plants. Biochars from six species of wetland plants (i.e., *Canna indica*, *Pennisetum purpureum* Schum, *Thalia dealbata*, *Zizania caduciflora*, *Phragmites australis* and *Vetiveria zizanioides*) were obtained at 500 °C and characterized, and their sorption for ammonium and cadmium was determined. There were significant differences in elemental composition, functional groups and specific surface area among the biochars derived from different wetland plant species. Sorption of ammonium and cadmium on the biochars could be described by a pseudo second order kinetic model, and the simple Langmuir model fits the isotherm data better than the Freundlich or Temkin model. The *C. indica* derived biochar had the largest sorption capacity for NH_4^+ and Cd^{2+} , with a maximum sorption of 13.35 and 125.8 mg g^{-1} , respectively. *P. purpureum* Schum derived biochar had a similar maximum sorption (119.3 mg g^{-1}) for Cd^{2+} . Ammonium sorption was mainly controlled by cation exchange, surface complexation with oxygen-containing functional groups and the formation of magnesium ammonium phosphate compounds, whereas for Cd^{2+} sorption, the formation of cadmium phosphate precipitates, cation exchange and binding to oxygen-containing groups were the major possible

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mechanisms. In addition, the sorption of ammonium and cadmium was not affected by surface area and micro-porosity of the biochars.

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1. Introduction

With the rapid development of economy in modern society, water resources have been dramatically exhausted. Water eutrophication and heavy metal contamination are posing a threat to the existing water resources worldwide. Cadmium (Cd^{2+}) is the most concerned toxic heavy metal that has resulted in soil and water pollution due to its high mobility and persistence while ammonium (NH_4^+) is the most common nitrogen (N) form that is readily available and often results in water eutrophication (Conley et al., 2009; Mohan et al., 2007). Therefore, much attention has been paid to the removal of these contaminants from water in order to protect water resources and maintain ecological environment security. Several technologies have been applied to remove these contaminants from aqueous solutions, such as chemical precipitation, ion exchange, membrane separation and adsorption (Jorgensen and Weatherley, 2003; Sud et al., 2008; Zhang et al., 2009). In comparison, sorption is considered as a cost-effective approach for remediation of contaminated water because of its low cost, high efficiency and environmental friendliness (Chen et al., 2011b; Sun et al., 2011). In this approach, adequate sorbent is the primary factor that determines the efficiency, and several sorbents have been studied with respect to their removal efficiency of contaminants from water, including activated carbon, organic material, microbial biomass, agricultural waste materials and carbon nanotubes (Ji et al., 2010; Kumar et al., 2014; Sud et al., 2008). However, all the materials have limitations of high cost, low efficiency, or disposal restriction.

Biochar, a solid material obtained from the thermochemical conversion of biomass in an oxygen-limited environment (IBI, 2012), has been applied for soil improvement, waste management, climate change mitigation and energy production (Bell and Worrall, 2011; Lehmann, 2007). In recent years, biochars have proven to be a green environmental sorbent for contaminants in water (Ahmad et al., 2014; Cao et al., 2011). Sorption of Cd^{2+} on biochars varied greatly with source materials. Biochars derived from wood and bark had a small sorption capacity ($0.34\text{--}5.40\text{ mg Cd g}^{-1}$) (Mohan et al., 2007), and the maximum sorption of Cd^{2+} by dairy manure biochar was greater (51.4 mg g^{-1}) (Xu et al., 2013), and various crop straws derived biochars showed even greater sorption capacities for Cd^{2+} ($57.7\text{--}96.4\text{ mg g}^{-1}$) in aqueous solution. According to literature, sorption capacity of the biochars for Cd^{2+} decreased in the order of crop straw > dairy manure > wood and bark (Ahmad et al., 2014; Frišták et al., 2015; Mohan et al., 2007; Sun et al., 2014; Xu et al., 2013). Obviously, selection of feedstocks for biochars is critical as the raw material often determines sorption capacity and performance of the resultant biochars. Waste biomass has been recognized as preferable feedstocks due to its wide availability and low costs (Ahmad et al., 2014).

Wetland plants have been widely grown in artificial floating island and constructed wetlands for water purification (Cheesman et al., 2010; Huett et al., 2005; Sun et al., 2009). Previous studies reported a number of wetland plant species that have been applied to phytoremediation of eutrophic water using constructed wetland, including *Miscanthus sinensis* Anderss (sp.), *Vetiveria zizanioides*, *Acorus calamus* and *Thalia dealbata* (Zhao et al., 2012a; Zhao et al., 2012b). However, the biomass of the plants must be properly harvested in order to sustain nutrient removal efficiency of the wetland since decomposition of plant litters/biomass may return all the nutrients back into the water. As a consequence, disposal of a large quantity of plant biomass has become a challenge. Extensive studies have been conducted to evaluate biochars derived from conventional biomass materials such as corn straw, bamboo and cottonwood with respect to their application in soil amendment and water remediation (Chen et al., 2011b; Xu

et al., 2012; Zhang and Gao, 2013), but much less focused on the biochars obtained from wetland plant feedstock, particularly their application for water remediation as a sorbent.

In this research, biochars were obtained from six wetland plant species (i.e. *C. indica*, *P. purpureum* Schum, *T. dealbata*, *Z. caduciflora*, *P. australis* and *V. zizanioides*) through slow pyrolysis at $500\text{ }^\circ\text{C}$. This temperature was selected based on previous studies (Lehmann, 2007; Ahmad et al., 2014; Zeng et al., 2013), at which the produced biochars had a greater sorption capacity for contaminants and better balance between the yield and energy costs. The samples (before and after sorption) were characterized using Fourier transform infrared spectroscopy, elemental analysis, BET- N_2 surface area, and scanning electron microscopy-energy dispersive spectrometer. Sorption kinetics and isotherms of ammonium (representing nutrient) and cadmium (representing heavy metal) on the biochars were determined to assess the capacity and mechanisms of NH_4^+ and Cd^{2+} sorption on the biochars in aqueous solution. The objectives were to: 1) characterize physico-chemical properties of the biochars; 2) determine the sorption capacity of the biochars for some common contaminants; and 3) understand the mechanisms of contaminant removal from water by the biochars.

2. Materials and methods

2.1. Materials

Six species of wetland plants, i.e. *C. indica*, *P. purpureum* Schum, *T. dealbata*, *Z. caduciflora*, *P. australis* and *V. zizanioides* were obtained from Lin'An constructed wetland, Zhejiang, China. The materials were rinsed with water and air-dried for a week, then ground and sieved to $<2.0\text{ mm}$ particles using a stainless grinding machine (Retsch MM400, Germany). The powdered biomass was tightly placed in a ceramic pot, and then pyrolyzed in a muffle furnace under N_2 atmosphere, the pyrolysis was programmed to drive the internal biomass chamber temperature to $500\text{ }^\circ\text{C}$ at a rate of $5\text{ }^\circ\text{C}/\text{min}$ and held at the peak temperature for 2 h before cooled to room temperature. The biochars (i.e. the solid residues from the pyrolysis) were referred to as CIB, PPB, TDB, ZCB, PAB, and VZB after the name of each plant species used for the feedstock. All of the biochar samples were ground to pass through a 0.5-mm sieve prior to use.

2.2. Characterization

Elemental (C, H, N) analyses were conducted using a CHN Elemental Analyzer (Flash-EA112, Thermo Finnigan). The oxygen content was determined by a mass balance. The H/C and (O + N)/C atomic ratios were calculated to evaluate the aromaticity and polarity of biochars, respectively. Scanning electron microscope (SEM) imaging analysis was conducted using a scanning microscope (FEI QUANTA FEG 650) to compare the structure and surface characteristics of biochars. Surface element analysis was conducted simultaneously with the SEM at the same surface locations using energy dispersive X-ray spectroscopy (EDS, EDAX Inc. Genesis XM). Fourier transform infrared (FTIR) analysis (Nicolet 6700) was performed in the $400\text{ and }4000\text{ cm}^{-1}$ region with 50 scans being taken at 2 cm^{-1} resolution to identify the surface functional groups. The specific surface area and porosity properties of the biochars were measured by N_2 adsorption isotherms at 77 K with the Brunauer–Emmett–Teller (BET) method and by CO_2 isotherms at 273 K using a Quadrasorb Si-MP surface area analyzer. Zeta-potential measurements were performed at different pH values with a potential analyzer (Malvern Nano-ZS90, England). Ash content was measured by heating the biochar samples at $750\text{ }^\circ\text{C}$ for 5 h. The pH of biochars was measured

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