



# Using biochar capping to reduce nitrogen release from sediments in eutrophic lakes

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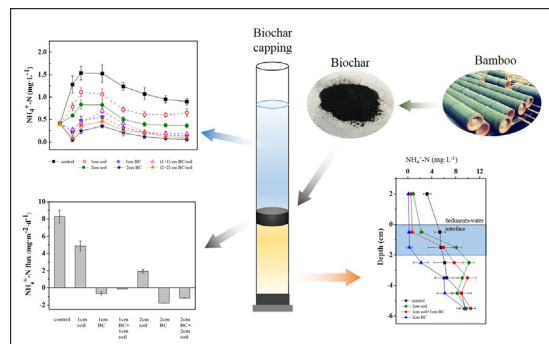
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## HIGHLIGHTS

- This study investigates the use of solid waste of *Phyllostachys pubescens*.
- We first propose the idea of using biochar for sediment nitrogen remediation.
- Biochar has a good effect on reducing nitrogen release, especially  $\text{NH}_4^+$ -N.
- We propose a good application of biochar capping technology.

## GRAPHICAL ABSTRACT



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## ABSTRACT

The effects of reduced nitrogen release from sediments were studied using biochar (BC) capping in simulated water–sediment systems. Dried solid waste of *Phyllostachys pubescens* was used to produce BC, which was then pyrolyzed at 500 °C. Subsequently, 14 sediment cores were collected, including the sediment–water interface and some overlying water, from two sites in Baiyangdian Lake (China). The sediment cores were split into two batches (A and B), and then two each were capped with soil, BC or a BC/soil mixture, and incubated for 30 days. In the BC capped cores, the ammonia nitrogen ( $\text{NH}_4^+$ -N), nitrate nitrogen ( $\text{NO}_3^-$ -N) and total nitrogen (TN) concentrations decreased from 0.90 mg·L<sup>-1</sup> to 0.05 mg·L<sup>-1</sup>, 0.88 mg·L<sup>-1</sup> to 0.18 mg·L<sup>-1</sup>, 6.93 mg·L<sup>-1</sup> to 2.81 mg·L<sup>-1</sup>, respectively, in batch A and 3.51 mg·L<sup>-1</sup> to 0.11 mg·L<sup>-1</sup>, 0.92 mg·L<sup>-1</sup> to 0.61 mg·L<sup>-1</sup>, 8.88 mg·L<sup>-1</sup> to 3.32 mg·L<sup>-1</sup>, respectively, in batch B. The sediments to water fluxes of  $\text{NH}_4^+$ -N,  $\text{NO}_3^-$ -N and TN were greatly reduced or reversed. Compared with other cappings, the BC layer was shown to absorb more  $\text{NH}_4^+$ -N from the pore water, thereby breaking the diffusion gradient of  $\text{NH}_4^+$ -N at the sediment–water interface, and has a good inhibitory effect on the endogenous release of  $\text{NH}_4^+$ -N from the sediments. Additionally, in the BC capped cores, the redox potential remarkably increased and dissolved oxygen was comparatively high. This study suggests that BC capping can reduce the amount of nitrogen released from polluted sediments because the diffusion of nitrogen to the overlying water is chemically blocked by the cap.

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## 1. Introduction

The use of artificial nitrogen (N) fertilizer (Galloway et al., 2008), release of untreated sewage (Carlson et al., 2013) and industrial effluent

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(Gibson et al., 2012), as well as increased use of specific household products (Braga et al., 2000) have resulted in large amounts of N emissions to natural water systems. N compounds form nutrients essential to all forms of life, but can trigger a series of ecological problems when there are excess N-based nutrients in aquatic environments, such as eutrophication, hypoxic or anoxic episodes of the water column, acidification of freshwater ecosystems, and increased toxicity to benthic organisms and fish (Bhatnagar and Sillanpää, 2011; Camargo and Alonso, 2006; Liu et al., 2012). With increased attention, the input of exogenous N to many lakes has been effectively controlled. Thus, research foci have shifted to the release of endogenous N from sediments. Sediments are a source of N, and the release of N from sediments can play a vital role in the quality of surface water (Beutel, 2006; Wu et al., 2001). Thus, it is especially important for water quality improvement to reduce the amount of N released into water that overlies sediments.

Great efforts have been made globally to control the release of N from sediments during the last decades, particularly in China. There are several strategies that are used, including mechanical, chemical, and microbial measures. These strategies involve dredging (Jing et al., 2013), in situ chemical injection (Liu et al., 2015), in situ capping (Gu et al., 2017; Lin et al., 2011) and bioaugmentation remediation (Jiao et al., 2011). To reduce the amount of N in a lake at larger scales, any technique employed should be quick and sustainable. Additionally, any successful technique will also need to be thorough, ecologically safe, cost effective and multifunctional. In situ capping is a traditional and widely used approach to address nutrient pollution in sediments. The core of this technology is in choosing a suitable material that acts as a barrier. Therefore, soil (Xu et al., 2012), coal ash (Kim et al., 2014), active carbon (Gu et al., 2017), natural and modified zeolite (Gibbs and Özkundakci, 2011), and other materials have been traditionally selected as capping layers. However, they are not ideal materials because they have disadvantages, such as low efficiency, high cost and adverse effects on benthic organisms. In contrast, the successful application of biochar (BC) in soil remediation has opened possibilities to also explore its potential for sediment remediation.

BC is a stable carbon-rich product synthesized through the pyrolysis of biomass (plant or animal) (Ahmad et al., 2014). Since BC was first identified in soil from the Amazon Basin, research interest in its various beneficial applications has increased (Lehmann, 2007; Lehmann et al., 2012); the “new black gold” has opened up multidisciplinary areas for science and engineering. BC has the ability to bind and sequester pollutants in sediments, especially heavy metals and organic pollutants. It adsorbs and immobilizes Cu, Pb and Hg (Wang et al., 2018), binds polycyclic aromatic hydrocarbons and reduces ecotoxicity, particularly to the benthic community (Fang et al., 2014; Han et al., 2017). Overall, BC is a natural product, has fewer ecotoxic side effects and is relatively low cost; thus, it is conducive to solid waste treatment and carbon sequestration. It has good N-fixing performance in soil (Laird et al., 2010) and good adsorption of ammonia nitrogen ( $\text{NH}_4^+\text{-N}$ ) in aqueous solution (Vu et al., 2016). Additionally, BC can promote microbial nitrification and denitrification in soil (Xu et al., 2014). These properties suggest that it could have special effects on the migration and transformation of N in sediments, which supports the use of BC in sediment N remediation. The use of activated carbon for sediment remediation could be a template for BC.

This study investigated the possibility of BC becoming sub-aqueous after capping contaminated sediments with an active barrier system. While BC has been used for nitrogen retention in soil and high  $\text{NH}_4^+\text{-N}$  concentrations in wastewater treatment, this is the first application in sediment N remediation. The BC was prepared from dried solid waste of *Phyllostachys pubescens*. The effects of N compound concentration changes in the overlying water and a reduction in N flux from the sediments were studied in cores recovered from Lake Baiyangdian in China. The objectives of this study were to explore the effects and mechanisms of N diffusion from sediments using BC capping technology.

## 2. Material and methods

### 2.1. Study sites

Lake Baiyangdian (BYDL), the sampling location, is the largest freshwater inland lake in northern China with an average water depth of 2.5 m. Two sites were cored: A ( $38^\circ49'24''\text{N}$ ,  $116^\circ0'23''\text{E}$ ) and B ( $38^\circ47'267''\text{N}$ ,  $116^\circ0'14''\text{E}$ ) (Fig. 1a). In previous investigations, the  $\text{NH}_4^+\text{-N}$  concentrations in the sedimentary pore waters of these areas were found to be high. However, the  $\text{NH}_4^+\text{-N}$  concentration in the overlying water was significantly lower than the pore water from surface sediments, which suggests  $\text{NH}_4^+\text{-N}$  has a stronger endogenous release risk. The  $\text{NH}_4^+\text{-N}$  flux from sediment ranges between 5.35 and  $48.76 \text{ mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$  (Zhu et al., 2018). The surface water content of various N compounds from the two sites is similar, but the  $\text{NH}_4^+\text{-N}$  pore water concentration of surface sediments at Site B is  $8.75 \text{ mg}\cdot\text{L}^{-1}$ , which is more than twice the concentration at Site A ( $4.09 \text{ mg}\cdot\text{L}^{-1}$ ) (Fig. 1b). These sites were chosen for incubation experiments because they represent different potential  $\text{NH}_4^+\text{-N}$  release risks and can reveal the inhibiting effect of BC on  $\text{NH}_4^+\text{-N}$  release.

### 2.2. Material preparation

BC was produced using a previous preparation method (Zhang et al., 2017); in brief, dried solid waste of *Phyllostachys pubescens* (a bamboo species) was slowly pyrolyzed at  $500^\circ\text{C}$  in a laboratory-scale quartz tube furnace with a trace gas mixed-gas supply system. Clean soil was collected from the bank around the sampling sites. The resultant *Phyllostachys pubescens* BC (PPBC) and soil were sieved through 80 mesh sieves. Fourteen plexiglass cylinders (diameter: 6 cm; height: 60 cm) were used as sampling and incubation vessels for the experiment. Lake water (0 to 30 cm above the sediment–water interface), together with the upper 20 cm of sediments, was sampled using a stainless steel gravity sampler in November 2017. The initial  $\text{NH}_4^+\text{-N}$  surface water concentrations at the two sites (A:  $0.41 \text{ mg}\cdot\text{L}^{-1}$  and B:  $0.33 \text{ mg}\cdot\text{L}^{-1}$ ), as well as nitrate ( $\text{NO}_3^-\text{-N}$ ) (A:  $0.26 \text{ mg}\cdot\text{L}^{-1}$  and B:  $0.23 \text{ mg}\cdot\text{L}^{-1}$ ) and total N (TN) (A:  $1.94 \text{ mg}\cdot\text{L}^{-1}$  and B:  $2.69 \text{ mg}\cdot\text{L}^{-1}$ ) were measured before treatment.

### 2.3. Incubation experiments

Approximately 1 h after sediment core retrieval, 1 cm soil, BC, or a mixture of BC with soil, and 2 cm of soil, BC, or BC with soil were used to cap the top of the sediments in the plexiglass cylinders; a control without capping was also used (Fig. 1c). All cylinders were divided into two batches (batch A and batch B) corresponding to Site A and Site B. The incubation experiments were conducted at  $25 \pm 1^\circ\text{C}$  in the dark. Overlying water samples (50 mL) from the incubation experiment were collected every 5 days (including the 3rd day) until day 30. After the 50 mL water samples were collected with a syringe (15 cm above the sediment–water interface), same volume of filtered distilled water was added to maintain the total overlying water volume. The volume of distilled water can be ignored because it is insignificant compared with the total volume of the overlying water ( $3392.9 \text{ mL}$ ).

The concentrations of TN,  $\text{NH}_4^+\text{-N}$ , and  $\text{NO}_3^-\text{-N}$  in the water samples were measured. The TN was determined using alkaline potassium persulfate digestion with an ultraviolet spectrometer.  $\text{NH}_4^+\text{-N}$  was measured by Nessler's colorimetric method and  $\text{NO}_3^-\text{-N}$  by an ultraviolet colorimetric method. The water samples were filtered through  $0.45\text{-}\mu\text{m}$  membrane filters before  $\text{NH}_4^+\text{-N}$  and  $\text{NO}_3^-\text{-N}$  determination. The oxidation–reduction potential (ORP) at the sediment–water interface and dissolved oxygen (DO) in the water were measured using a water quality multifunctional device (HACH HQ30d, US) with different probes. On day 30, the sediment cores were sectioned into 1 cm intervals from the surface to bottom. All of the sections were centrifuged to extract the pore water, and then the pore water concentrations of  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_3^-\text{-N}$

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