A pilot bioretention system with commercial activated carbon and river sediment-derived biochar for enhanced nutrient removal from stormwater

Min Sang, Miansong Huang, Wei Zhang, Wu Che and Huichao Sun

ABSTRACT

Bioretention is an effective technology for urban stormwater management, but the nutrient removal in conventional bioretention systems is highly variable. Thus, a pilot bioretention column experiment was performed to evaluate the nutrient control of systems with commercial activated carbon and river sediment-derived biochar. Significant chemical oxygen demand (COD) and total phosphorus (TP) leaching were found with the addition of activated carbon and biochar, but total nitrogen (TN) leaching was significantly improved when activated carbon was used as the medium. During a semi-synthetic runoff experiment, the bioretention systems containing two types of fluvial biochar showed relatively better COD and TN control (average mass removal efficiencies and cumulative removal efficiencies) than commercial activated carbon. However, the average TP mass removal efficiency with commercial activated carbon (95% ± 3%) was significantly higher than biochar (48% ± 20% and 56 ± 14%). The addition of biochar in the media increased the nitrogen removal efficiency, and the addition of activated carbon significantly increased the phosphorous removal efficiency. Therefore, both biochar and activated carbon are effective materials for bioretention, and fluvial biochar provides an alternative approach to comprehensively utilize river sediment.

Key words | activated carbon, biochar, bioretention, nutrient

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INTRODUCTION

Urban stormwater runoff is one of the degradation factors for urban streams and threatens water ecological security (Walsh 2000). Stormwater runoff contains various components, including nutrients (Yu *et al.* 2015), heavy metals (Mahbub *et al.* 2010) and pesticides (Paul & Meyer 2001), that lead to aquatic environment pollution.

Bioretention is a typical stormwater management practice and can reduce the impact of pollution both on hydrology and water quality (Roy-Poirier *et al.* 2010). The effects of bioretention on nutrients, especially phosphorus and nitrogen, have been previously investigated. In Payne *et al.*'s research, a laboratory column experiment through designing variables including the plant species and parameter of the saturated zone was conducted and the results showed that the reduction of nitrogen concentrations on average was increased to over 70% through optimizing plant species and saturated designs during the wet period (Payne *et al.* 2014). However, only a 9% reduction in total nitrogen (TN) concentration was observed in a bioretention field study, as a result of obvious dissolved organic nitrogen (DON) and NO_3-N leaching from the media (Li & Davis 2014). In addition, the mass loading removal of phosphorus has shown a wide range, from a 240% increase (Hunt *et al.* 2006) to a 99% decrease (Hsieh & Davis 2005).

To improve nutrient removal, several approaches, such as enhancing bioretention media (Goh *et al.* 2017), optimizing plant species selection (Read *et al.* 2008) and providing a saturated zone (Lucas & Greenway 2011; Zhang *et al.* 2019a), have been investigated. Supplementing the ideal additive into bioretention media has also been shown to be an effective approach.

An ideal additive provides stable and effective nutrient removal and various compounds have been studied for

doi: 10.2166/wst.2019.310

bioretention (LeFevre *et al.* 2014). Extensive studies have been done on new types of material that can be mixed into bioretention systems with additives including scrap wood (e.g. wood chips) (Peterson *et al.* 2015), pyrolyzed biomass (e.g. biochar) (Iqbal *et al.* 2015) and river sediment (Zhang *et al.* 2019b). It is worth noting that pyrogenic carbonaceous materials have drawn increasing attention because of their high specific surface area and microporous structure, which is particularly relevant for biochar (Reddy *et al.* 2014).

Biochar is generally produced from a variety of agricultural or other carbon-rich waste (Kah *et al.* 2016; Sigmund *et al.* 2016), such as wood chip (Mohanty *et al.* 2014), crop straw (Ding *et al.* 2010) or manure (Wang *et al.* 2016). Some research has investigated the effects of biochar converted from waste biomass on a variety of contaminants, including pesticides (Liu *et al.* 2018), heavy metals (Chen *et al.* 2018) and antibiotics (Li *et al.* 2019). Recently, the effects of biochar treatment of contaminants from stormwater runoff have been evaluated (Iqbal *et al.* 2015; Tian *et al.* 2016). The structure and properties of biochar mainly depend on the types of feedstock (Gray *et al.* 2014). As shown in previous studies, biochar applied to stormwater treatment facilities has mostly been produced from scrap wood (Reddy *et al.* 2014; Afrooz & Boehm 2017) and there are very few studies that report on biochar derived from dredged river sediment.

Biochar has some effect on the removal of nitrogen, but it is less effective for phosphorus control (Novak *et al.* 2009; Ying *et al.* 2010; Chen *et al.* 2011; Yao *et al.* 2012). However, activated carbon has shown good adsorption effects on phosphorus in previous research (Kumar *et al.* 2010; Manjunath & Kumar 2018). Activated carbon, which has high specific surface area and developed porosity (Dias *et al.* 2007), is mainly used in drinking water and wastewater treatment, and rarely used in stormwater treatment (Björklund & Li 2017). The application of activated carbon as a soil additive sequesters a variety of contaminants (Hale *et al.* 2012), such as organic pollutants (Hale *et al.* 2012; Choi *et al.* 2014) and heavy metals (Wu *et al.* 2016). However, there has been limited investigation of the bioretention of nutrients in urban stormwater.

Accordingly, this study used river sediment-derived biochar and commercial activated carbon mixed with conventional media to investigate the control of bioretention on nutrients. In addition, the feasibility of resource utilization of dredged river sediment was explored. Laboratory-based bioretention column experiments were used to address the feasibility and mechanism of pollution control by adding river sediment-derived biochar and activated carbon into bioretention media. The objectives were to: (a) determine the leaching effect of river sediment-derived biochar and commercial activated carbon additives; (b) assess enhanced nutrient removal of bioretention systems amended with activated carbon and river sediment-derived biochar; (c) compare nitrogen and phosphorus removal when activated carbon and river sediment-derived biochar were used as additives.

MATERIALS AND METHODS

Bioretention column setup

Bioretention columns were located in a greenhouse at the Beijing University of Civil Engineering and Architecture. Bioretention column experiments were performed from January to June 2018, during which time the average temperature was 11.0 ± 11.3 °C. The columns consisted of 150-mm-diameter PVC pipes, each containing 70 cm of media with 16 cm of freeboard top section that allowed for ponding (Figure 1). Media were placed above the geotextile atop a 10-cm-thick gravel drainage layer to prevent media washout (Figure 1). Three columns (GAC, GUB and GDB) contained media consisting of 94% garden soil and 6% activated carbon (GAC) or river sediment-derived biochar (GUB and GDB). The details and characteristics of these materials are given in Table 1. All of the media was passed through a 2 mm standard sieve and gravel was sieved from 5–10 mm. All of the columns were planted with native vegetation, *Iris lactea* Pall. var. chinensis (Fisch.) Koidz., because of its adaptability and tolerance to drought (Figure 1).

Material preparation

The fluvial biochar used in the laboratory was converted from sediment deposited upstream (UB) and downstream (DB) in the Zhuanghe River, China. It is a seriously polluted river, and the main source of pollution is combined sewer overflows and domestic sewage discharge. The UB sediment was collected from the upstream river. The DB sediment was collected from an area in the downstream river, close to the river's outflow to the sea where large amounts of tidally transported seabed sediments are deposited (Zhang *et al.* 2019b). The sediment was air dried in the shade and then passed through a 2 mm standard sieve before it was placed into a porcelain crucible and capped. The porcelain

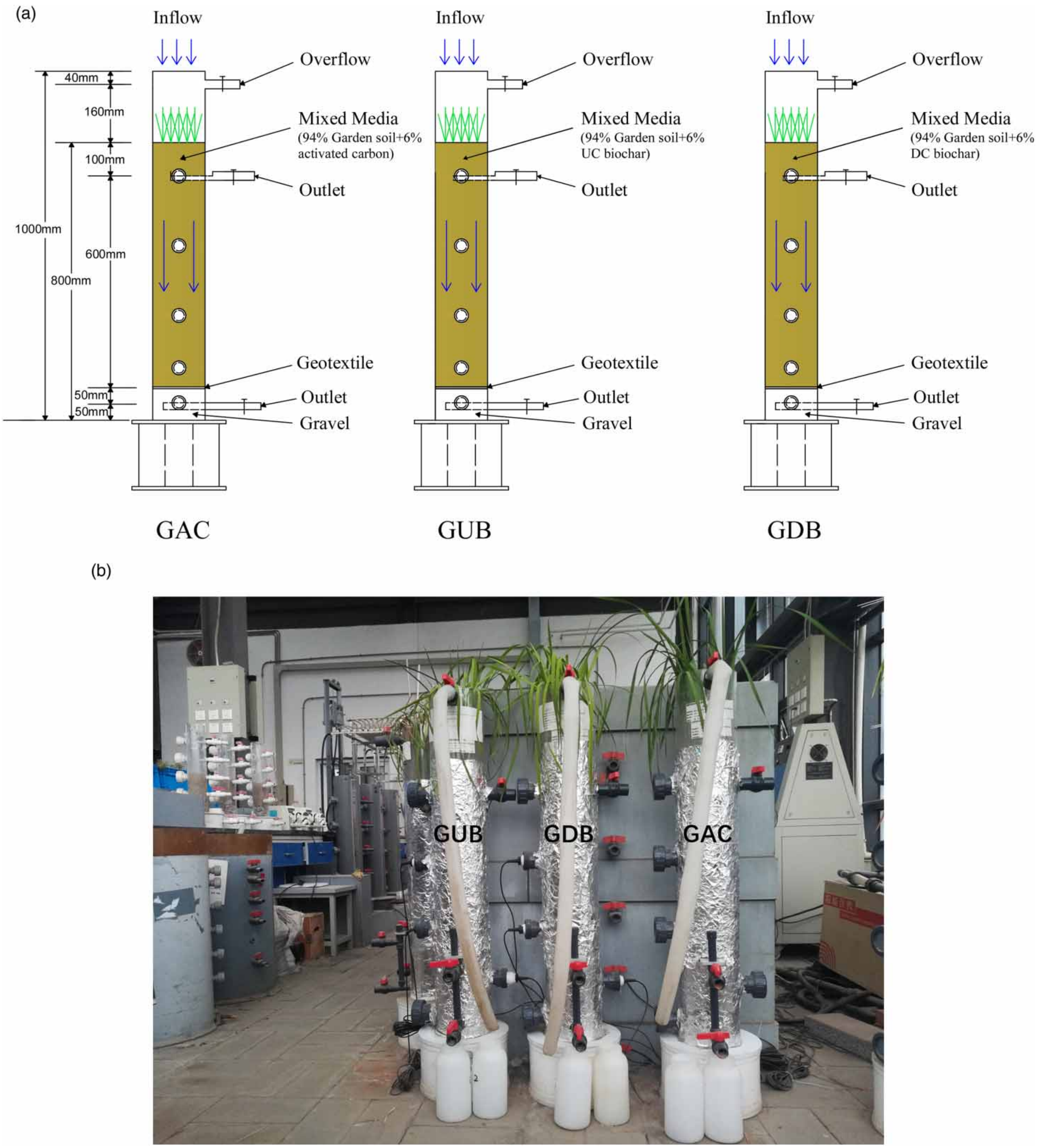

Figure 1 | Details of (a) the column structure and (b) bioretention columns.

crucibles were subjected to heating and the sediment was pyrolyzed using an electrical muffle furnace at 400 °C under hypoxic conditions for 4 h. After naturally cooling to room temperature (25 °C), the biochar was then sieved over a 2 mm standard sieve; particles less than 2 mm were collected and stored in a closed container. The activated carbon was obtained from a commercial activated carbon producer.

Table 1 | The details and characteristics of bioretention column media

Column	Media type	Bulk density/g · cm^{-3}	Available nitrogen/mg · kg^{-1}	Organic matter/g · kg^{-1}	Available phosphorus/mg · kg^{-1}
GAC	94% Garden soil + 6% activated carbon	1.18	121.22	4.36	7.16
GUB	94% Garden soil +6% UC biochar	1.01	73.39	4.68	15.56
GDB	94% Garden soil +6% DC biochar	1.27	117.22	5.14	18.57

Media characteristics

The material properties (Table 1) were measured using standard methods for soil bulk density, available nitrogen and phosphorus, and soil organic matter (Yang *et al.* 2017). The surface area was determined with the Brunauer-Emmett-Teller (BET) method through N_2 adsorption (Huggins *et al.* 2016). The BETs of garden soil, activated carbon, UB and DB were 2.5609, 17.9621, 11.9396 and 12.6899 $m^2 \cdot g^{-1}$, respectively.

Semi-synthetic runoff and experimental methods

The experiment contained two stages, a leaching stage and a semi-synthetic runoff stage. The leaching experimental stage provided tap water as inflow to estimate the media leaching characteristics. The semi-synthetic runoff experimental stage provided semi-synthetic runoff as inflow to estimate the bioretention performance.

During both the leaching experimental stage and semi-synthetic runoff experimental stage, the same inflow water quantity condition was used. The inflow volume is 4.3 L, assuming a bioretention column sized at 10% of the impervious catchment (Hunt *et al.* 2006), with a runoff coefficient of 0.9 based on the design rainfall (27.3 mm) according to an 90% capture ratio of total annual runoff volume in Guyuan, Ningxia, China.

Leaching experiment

All of the columns received 4.3 L of tap water with constant water head as inflow. The influent tap water was added to the columns to the level of the overflow ports. A water head of 50 mm was maintained throughout the influent process. The leaching stage included 8 rainfall events, and the antecedent dry period (ADP) was 4 days. In our pre-experiment, the water content of media in a packing layer of bioretention basically returned to the initial state after 24–48 h. Hence, the ADP was determined as 4 days during this stage. The average pollutant concentrations in inflow during this stage were: chemical oxygen demand (COD) = 5.0 ± 0.8 mg · L^{-1}, T$N = 8.49 \pm 0.80$ mg · L^{-1}, total phosphorus (TP) = 0.03 ± 0.02 mg · L^{-1} and turbidity = 0.05 ± 0.01 NTU.

Semi-synthetic runoff experiment

During this stage, all of the columns received 4.3 L of semi-synthetic runoff from the same hydrologic regime as the inflow. This stage included 16 rainfall events, and the ADP was 7 days. Based on the survey of local rainfall data for past 30 years, the average antecedent dry period (ADP) was approximately 7 days. The local rainy season in Guyuan is from May to September, with an average rainfall of 430 mm during this period. Hence, the rainfall event number during the semi-synthetic runoff experimental stage was determined as 16 rainfall events, and the total 436.8 mm to simulate a full rainy season performance of the bioretention columns.

The inflow water composition considered the local urban runoff quality (Ren *et al.* 2008). Semi-synthetic runoff was simulated by adding glucose, KNO_3, NH_4Cl and KH_2PO_4, which were supplied as dissolved pollutants. Additionally, the road dust was collected from a local municipal road a brush and a vacuum cleaner, and it passed through a 0.10-mm standard sieve. This local road is bi-directional single lane, which is eight meters wide. Land use nearby this road involves residential and educational. The daily cleaning of the road is manual and mechanical, and the frequency of cleaning is once a day. Besides, the dust passed through a 0.10-mm standard sieve was added into the semi-synthetic runoff to simulate particulate pollutants. Determination of the concentrations of pollutants for semi-synthetic runoff was based on road runoff pollution reported in previous studies in China (Ren *et al.* 2008; Zhang *et al.* 2012), after adding appropriate chemical reagents, such as glucose, KNO_3, NH_4Cl, and KH_2PO_4, to simulate the soluble pollutant concentrations in urban stormwater runoff. Across all of the rainfall events during this stage, the average inflow pollutant concentrations were COD = 237 ± 31 mg · L^{-1}, T$P = 0.77 \pm 0.29$ mg · L^{-1}, PO_4-P = 0.67 ± 0.26 mg · L^{-1}, T$N = 16.36 \pm 3.34$ mg · L^{-1}, NH_4-N = 3.10 ± 1.59 mg · L^{-1}, NO_3-N = 9.84 ± 1.24 mg · L^{-1} and turbidity = 39.38 ± 14.37 NTU.

Mixed inflow and outflow samples were collected with sample bottles during both dosing periods and measured for TN, TP, COD and turbidity within 24 h using standard methods (APHA 2012).

Data analysis

The pollutant mass removal efficiency (R_L), which was used to estimate the bioretention column performance, was calculated as:

$$R_L = \frac{C_{in} \cdot V_{in} - C_{out} \cdot V_{out}}{C_{in} \cdot V_{in}} \tag{1}$$

where C_{in} is the inflow pollutant concentration ($mg \cdot L^{-1}$), V_{in} is the inflow volume (L), C_{out} is the outflow pollutant concentration ($mg \cdot L^{-1}$) and V_{out} is the outflow volume (L).

The cumulative removal efficiency was cumulative removal (in %) of the total influent pollutant mass and calculated as:

$$R_L = \frac{\sum_{i=1}^{n} C_{in,n} \cdot V_{in,n} - \sum_{i=1}^{n} C_{out,n} \cdot V_{out,n}}{\sum_{i=1}^{n} C_{in,n} \cdot V_{in,n}} \tag{2}$$

where $C_{in,n}/C_{out,n}$ is the inflow/outflow pollutant concentration ($mg \cdot L^{-1}$) for run n and $V_{in,n}/V_{out,n}$ is the inflow/outflow volume (L) for run n.

In this study, the water volume reduction efficiency only included the reduction of water from evaporation, absorption of media and the storage facility, and excluded overflow volume during rainfall events. The water volume reduction efficiency (R_v) was defined and calculated as:

$$R_v = \frac{V_{in} - V_{out}}{V_{in}} \tag{3}$$

where V_{in} is the inflow volume (L), especially the water volume ponding in depressions during a rainfall event and V_{out} is the outflow volume (L).

One-way analysis of variance (ANOVA) was applied with the media type as a factor to evaluate differences in pollutant concentrations from various columns; significant results showed $p > 0.05$.

RESULTS AND DISCUSSION

COD, TN and TP leaching

The cumulative inflow and outflow COD, TN and TP masses for each bioretention column during the leaching experiment are shown in Figure 2.

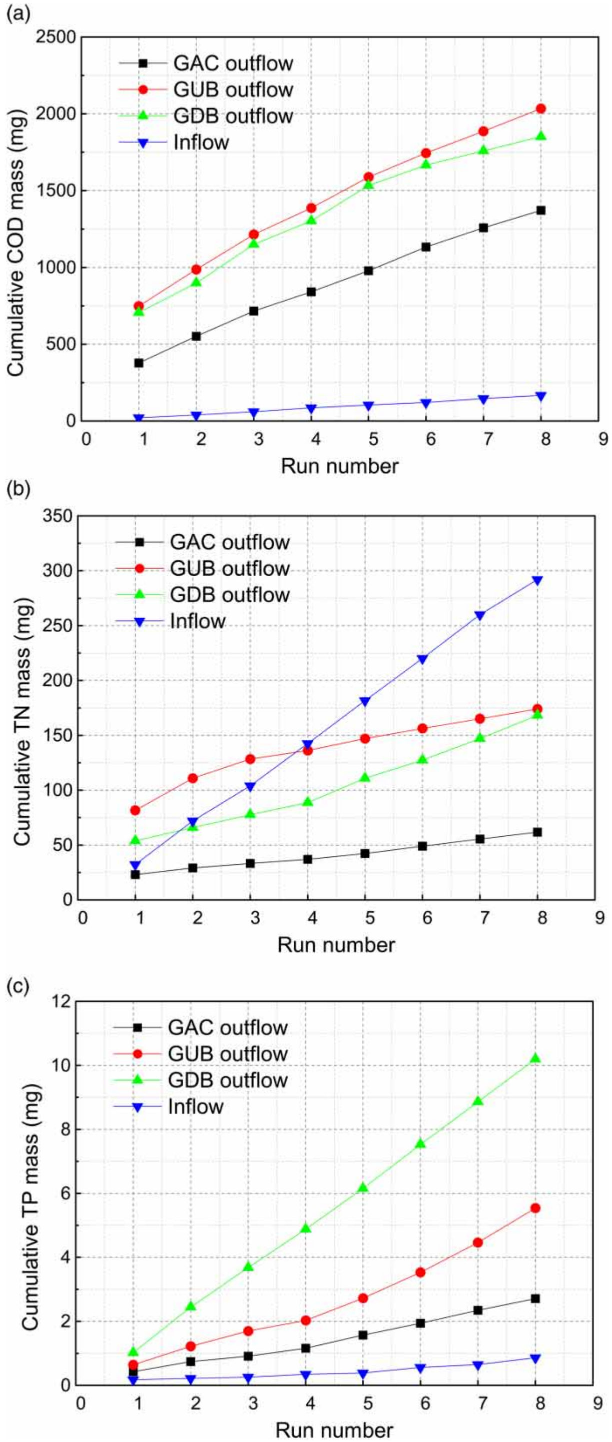

Figure 2 | Cumulative mass of pollutant inflow and outflow from each column during the leaching experiment. A run represents a rainfall event. (a) COD. (b) TN. (c) TP.

COD

The cumulative inflow and outflow COD masses from each column showed a similar performance. The cumulative inflow was far less than the outflow, which indicates there was obvious COD leaching during the initial stage of this experiment. Moreover, the cumulative outflow from GAC (1,372 mg) was lower than GUB (2,035 mg) and GDB (1,852 mg), which indicates that the accumulated leaching was less. It may be because the organic matter content of media in GAC was the lowest among the three columns (Table 1).

TN

The cumulative inflow TN mass increased steadily during the leaching stage because of nitrogen in the tap water; the average inflow concentration of nitrogen was 8.49 ± 0.80 mg·L^{-1}. The cumulative outflow from GUB and GDB was higher than the inflow before the fourth (GUB) and second (GDB) rainfall events because of vast nitrogen leaching from media during the first rainfall event. Testing of biochar has shown that it releases nitrogen into water (Yao *et al.* 2012). However, during the second rainfall event, the cumulative outflow mass from GUB and GDB were 29.29 mg and 11.93 mg, respectively, which was lower than the inflow (39.69 mg). Moreover, the cumulative TN mass from GUB and GDB exhibited a slower growth trend than the inflow. It showed that GUB and GDB removed TN from the inflow rather than leaching. In contrast, the outflow TN from GAC was always less than the inflow, demonstrating TN removal rather than TN leaching.

TP

The cumulative outflow TP mass from each column was always higher than the inflow. Except for inflow phosphorus, excess TP in the outflow from each column was probably attributed to leaching from media. Outflow from GAC, GUB and GDB increased with run numbers during the leaching stage and finally reached 2.71 mg, 5.54 mg and 10.19 mg, respectively. GAC showed the smallest amount of accumulated leaching among the three columns. This is attributed to less available phosphorus in mixed media of GAC (7.2 mg · kg^{-1}) than GUB (15.6 mg · kg^{-1}) and GDB (18.6 mg · kg^{-1}).

COD, nitrogen and phosphorus removal

The average mass and cumulative removal efficiencies of TN, TP and COD across all of the semi-synthetic runoff experiments are illustrated in Figure 3.

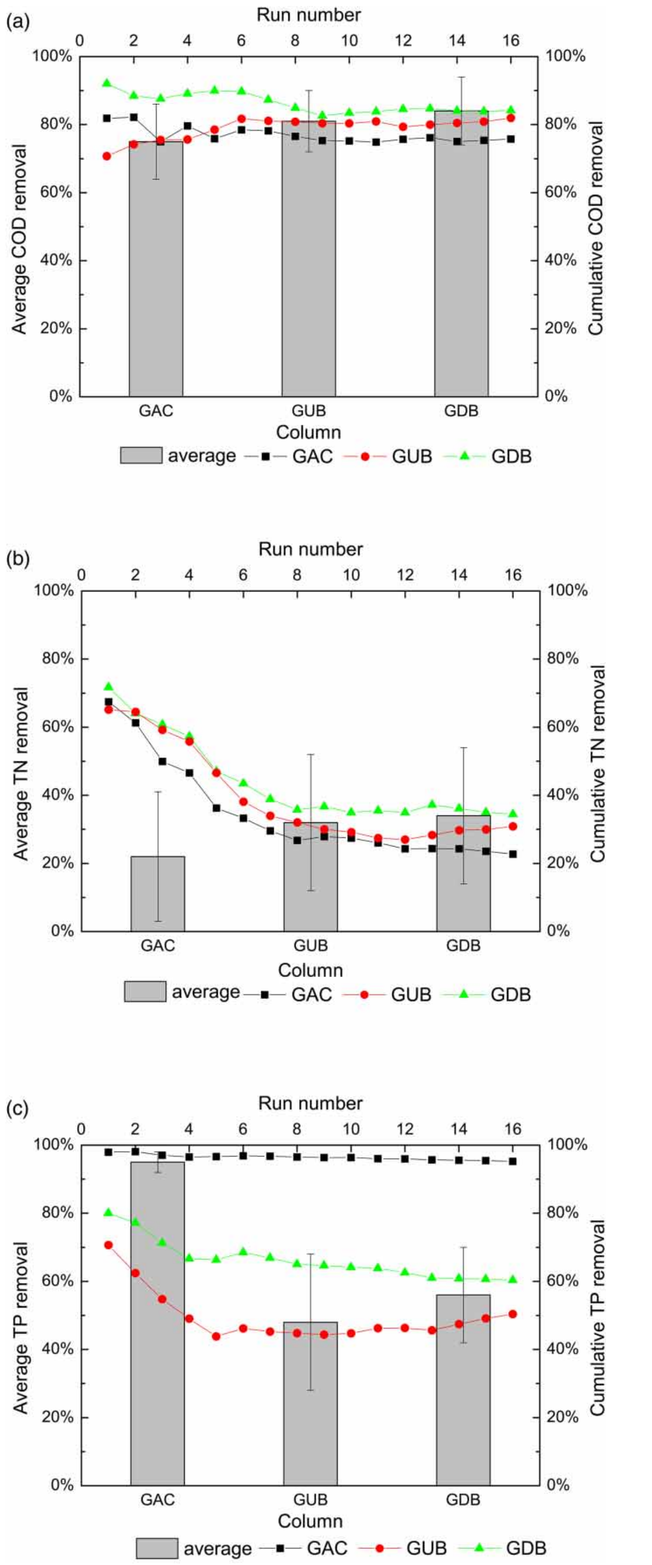

Figure 3 | Cumulative COD, TN and TP mass removal of three columns and average removal during the semi-synthetic runoff experiment. Each column and error bar represent the mean value and standard deviation. A run represents a rainfall event. (a) COD. (b) TN. (c) TP.

COD

The average mass removal efficiencies of GUB (81% ± 9%) and GDB (84% ± 10%) were slightly larger than GAC (75% ± 11%). Across the 16 rainfall events, all of the columns showed basically a stable COD mass removal, which indicates a consistent removal effect on COD. During the last rainfall event, cumulative COD removals from GAC, GUB and GDB were 76%, 82% and 84%, respectively, which indicated effective removal of COD. Compared with activated carbon that mainly contains micropores (>1 nm), biochar is comprised primarily of larger pore sizes (Huggins *et al.* 2014). These macropores connect with smaller micropores and contribute to the adsorption of larger organic molecules (Huggins *et al.* 2014). Huggins *et al.* (2016) also observed better COD control in biochar because of its macroporous structure, compared with microporous activated carbon.

Nitrogen

The average TN mass removal efficiencies of GAC, GUB and GDB were 22 ± 19%, 32 ± 20% and 34 ± 20%, respectively (Figure 3(b)). The cumulative TN removal efficiencies for all of the columns decreased during the semi-synthetic runoff experiment, indicating that effective nitrogen control decreased with the run number. The cumulative removal efficiencies decreased 34% and 37% in GUB and GDB, respectively, which was slow compared to the 45% decrease in GAC. GUB and GDB removed approximately 1.35 times (31% vs 23%) and 1.48 times (34% vs 23%) more nitrogen, respectively, than GAC. Since the second rainfall event, the cumulative TN removal with biochar (GUB and GDB) was consistently higher than with activated carbon (GAC). Based on the average TN mass removal efficiencies and cumulative TN removal, the addition of biochar increased the nitrogen removal efficiency of the bioretention system. Biochar can adsorb ammonium cations, mainly through cation exchange, and low temperature biochar (e.g. 400 °C) probably had a relatively stable aromatic backbone from pyrolysis and more C = O and C-H functional groups, which may act as nutrient exchange sites after oxidation (Glaser *et al.* 2002; Novak *et al.* 2009; Ying *et al.* 2010).

Nitrogen and phosphorus for the inflow and outflow monitored across all events during the runoff experiment are illustrated in Figure 4. Among all samples collected from the bioretention columns, inflow TN was largely composed of NO_3-N and NH_3-N and together accounted for

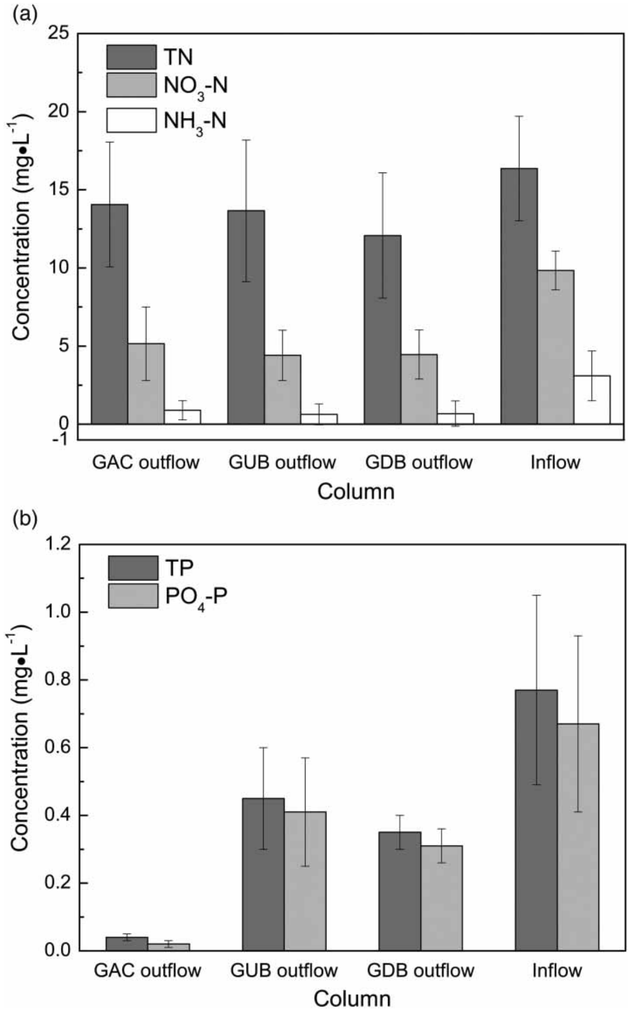

Figure 4 | Inflow and outflow, monitored across all events during the runoff experiment, for nitrogen and phosphorus species in different bioretention columns. Each column and error bar represents the mean and one standard deviation. (a) Nitrogen. (b) Phosphorus.

81% ± 13%. The outflow NO_3-N and NH_3-N concentrations from each column were much lower than the inflow, indicating the bioretention columns had better control over ammonia and nitrate through nitrification and denitrification (Wan *et al.* 2018). Previous research proposed that the transformation of nitrogen species, especially through ammonification and nitrification, is the main reason for variable nitrogen removal (Li & Davis 2014). In addition to nitrate and ammonium, dissolved organic nitrogen (DON) probably was another factor that caused poor TN removal. The significant DON increase in outflow from bioretention underdrains has been observed in previous studies (Hatt *et al.* 2009; Hunt *et al.* 2006). The increased amount of other nitrogen species in the outflow, excluding NO_3-N and NH_3-N, is probably attributed to DON.

Phosphorus

The average TP mass removal efficiencies of GAC, GUB and GDB were 95% ± 3%, 48% ± 20% and 56 ± 14%, respectively (Figure 3(c)), and the efficiency of GAC was better than columns GUB and GDB ($p < 0.05$). From the first to 16th rainfall event, the cumulative TP removal efficiencies for GUB and GDB decreased approximately 20% compared to a 3% decrease in GAC. In addition, the cumulative TP removal for GAC was 95% during the last rainfall event, which was higher than GUB (50%) and GDB (60%). Overall, the water volume reduction efficiency of GAC was low (11% ± 5%) compared with GUB (18% ± 13%) and GDB (12% ± 6%). However, the addition of activated carbon significantly increased the phosphorous removal efficiency of the bioretention system.

The outflow TP and PO_4-P concentrations from all of the columns were significantly lower than inflow concentrations (Figure 4(b)). PO_4-P was the major form in the inflow as well as the outflow of all of the columns. Activated carbon is effective in removing phosphates from aqueous solution (Kumar *et al.* 2010) and may explain the better removal performance of phosphorus in GAC.

The total inflow volume during the two experimental stages was 103.2 L, which is equivalent to total precipitation of 655.2 mm. It is approximately equivalent to 1.3 times the average annual rainfall of Guyuan, Ningxia, West-China (semiarid areas). It can provide a reference for evaluating the bioretention performance for the initial 1–2 years' operation. However, researches on the medium- and long-term performance of the bioretention is needed in the future.

CONCLUSIONS

This laboratory study was conducted to investigate water volume reduction efficiency and nutrient removal of river sediment-derived biochar and commercial activated carbon. The main findings were as follows:

(1) For COD and nutrient leaching, the cumulative COD mass in the outflow of GAC (1,372 mg) was less than GUB (2,035 mg) and GDB (1,852 mg). Moreover, cumulative outflow TP from GUB and GDB largely increased with run numbers during the leaching experiment, finally reaching 5.54 mg and 10.19 mg, respectively; this was much higher than GAC (2.71 mg). Generally, river sediment-derived biochar and commercial activated carbon significantly leached TP and COD, but the control over TN leaching significantly improved when commercial activated carbon was used as bioretention media. Under this condition, cumulative TN mass in the outflow was always less than the inflow. Compared with commercial activated carbon, river sediment-derived biochar, GUB and GDB, released nitrogen into solution during the initial experimental stage.

(2) During the semi-synthetic runoff experiment, bioretention with the two types of fluvial biochar exhibited relatively better COD and TN control (average mass removal efficiencies and cumulative removal efficiencies) than bioretention with commercial activated carbon. However, the average TP mass removal efficiency with commercial activated carbon (95% ± 3%) was significantly higher than with biochar (48% ± 20% and 56 ± 14%). The addition of biochar in bioretention media increased the nitrogen removal efficiency and the addition of activated carbon significantly increased the phosphorous removal efficiency.

(3) Biochar and activated carbon are feasible and effective materials for bioretention, and river sediment-derived biochar provides an alternative approach to comprehensively utilize fluvial sediment. Further studies of nutrient removal mechanisms of biochar and activated carbon in bioretention systems should be conducted to facilitate their application. The experimental results may provide a reference for predicting operation efficiency of bioretention in semiarid areas, which is like the rainfall condition in Guyuan. Limited to the experimental period, the first year of bioretention operation was simulated and assessed in this study, and researches on the medium- and long-term performance of the bioretention are needed in the future.

ACKNOWLEDGEMENTS

This study was financially supported by the National Natural Science Foundation of China (No. 51608026), the Scientific Research Project of Beijing Educational Committee (No. KM201910016012), Beijing Advanced Innovation Center for Future Urban Design Projects (No. UDC2016040100) and the Research Program for Key Technologies of Sponge City Construction and Management in Guyuan City (No. SCHM-2018). The authors thank the research team members for their enthusiastic support. The authors also thank Kara Bogus, PhD, from Liwen Bianji, Edanz Editing China (www.liwenbianji.cn/ac), for editing the English text of a draft of this manuscript.

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First received 24 May 2019; accepted in revised form 2 September 2019. Available online 11 September 2019