Enhanced Benzoﬂuoranthrene Removal in Surface Flow Constructed Wetlands with the Addition of Carbon

Qingqing Cao, Yan Li, Yan Kang,* and Zizhang Guo

ABSTRACT: Polycyclic aromatic hydrocarbons (PAHs), as hazardous pollutants, could be removed by constructed wetlands (CWs). While the traditional substrate of CWs has a weak adsorption capacity for PAHs, in this study, the carbonous fillers—activated carbon (AC) and biochar—were added into the substrate of surface ﬂow CWs to improve the removal performance of benzoﬂuoranthrene (BbFA), a typical PAH. The results showed that the BbFA removal efﬁciencies in CWs with the addition of AC and biochar were 11.8 and 1.2% higher than those in the Control group, respectively. Simultaneously, the removal efﬁciencies of NO3− were 42.8 and 68.4% in these two CWs, while the BbFA content in the substrate and plants with the addition of carbon was lower than that in the Control group. The addition of carbonous ﬁller reduced the absorption of PAHs by plants in CWs and enhanced microbial degradation. The microbial community results showed that the relative abundance of Proteobacteria, especially γ-proteobacteria, was higher with the addition of ﬁllers, which related to PAH degradation.

1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are typical persistent organic pollutants with biotoxicity and teratogenic carcinogenicity. PAHs accumulate in the biological chain and thus are harmful to the ecological environment and the health of organisms. It has been detected in watershed environments around the world, e.g., Yangtze River (China), Elbe River (Germany), Brisbane River (Australia), Ovia River (Southern Nigeria), and other rivers. PAHs (>3 rings) have a stable chemical structure with the character of lipotropy and hydrophobicity and thus are diﬃcult to be biodegraded. The current methods for PAH removal mainly include chemical methods, such as coagulation, chemical oxidation, ultrasonic, and adsorption. However, these methods are not suitable for water pollution treatment in large river basins. Constructed wetland (CW) is an ecological project for pollution treatment in an aquatic environment with low energy consumption, high removal eﬃciency, and simple operation management. It has been proved to have an eﬀective removal capacity for nitrogen, phosphorus, and organic pollutants from water by the synergistic eﬀect of plants, substrates, and microorganisms.

Previous research revealed that substrate adsorption and microbial degradation played important roles in organic contaminant treatment in CWs, which contributed to 28.67–61.00% of pollutant removal. As the main framework of CWs, the substrate provided a medium for plant roots and microorganisms’ culture and provided the location for contaminant degradation by microbes. Meanwhile, due to the sedimentation and hydrophobicity of PAHs, PAH pollutants mainly accumulated in the substrate. Thus, improving the adsorption capacity and microbial degradation of pollutants in the substrate were important for the removal of PAHs in CWs. An eﬀective method to enhance the performance of the substrate is the addition of a ﬁller. The main substrate ﬁllers in CWs are gravel, sand, and soil, but these ﬁllers showed lower adsorption capacity for PAHs. Some other absorbents, such as montmorillonite and clay, have good adsorption properties for PAHs but cause blockage of CWs when added as substrate ﬁllers.

The carbon adsorbent has stable physical and chemical properties, well-developed speciﬁc surface area, abundant surface functional groups, and shows good performance for the PAH adsorption from wastewater. As a ﬁller, the carbon adsorbent was predicted to improve the adsorption capacity of the substrate as well as promote the PAH transformation and...
degradation by microbes in the habitat provided by the well-developed pore structures. Thus, the removal efficiency of PAHs can be further improved in CWs. Moreover, carbon adsorbent played an important role in plant growth and microbial culture.

Based on the potential role of carbon adsorbent for PAH removal in CWs, the biochar and activated carbon (AC) were added to the substrate to establish CW microcosms. Benzo[a]fluoranthrene (BbFA), a typical PAH with 5 rings, was selected as a PAH pollutant in this work. The objectives of this research were to (1) observe the treatment performance of CWs with biochar and AC; (2) evaluate the influence of these two materials on PAH removal in wastewater, substrate, and plants in CWs; and (3) explore the effect of the two materials on the microbial community. This research provides useful information for PAH removal in an aquatic environment.

2. RESULTS AND DISCUSSION

2.1. Performance of CWs in Wastewater. The BbFA concentration in the wastewater of each group is shown in Figure 1a. The BbFA removal efficiencies in the experimental treatments followed the order A-CW > B-CW > Control. The average removal efficiency of BbFA in A-CW was up to 99.0 ± 0.23%, which was 10.6 and 1.2% higher than that in the Control and B-CW groups, respectively (p > 0.05). The results reflected that both AC and biochar promoted the removal of BbFA in CWs, and could be attributed to the immobilization and adsorption of organic contaminants due to their hydrophobic properties. Brennan et al. found that the AC has better performance for PAH reduction in porewater than biochar, while the biochar had larger particle sizes.

The concentrations of the main pollutants were also measured to evaluate the operation conditions of CWs. As shown in Figure 1, there was a great variation in concentrations of each inorganic nitrogen among the three groups. Both biochar and AC showed better performance for nitrate (NO$_3^-$−N) removal, especially B-CW, when the NO$_3^-$−N concentration in the effluent was less than 16.9 mg/L; the removal efficiency was 30.4 ± 2.77%, which was 68.4% higher than that in the Control group. The addition of AC in CWs also increased the NO$_3^-$−N removal efficiencies, which was 42.8% higher than that of the Control group. For ammonium (NH$_4^+$−N) concentrations, as shown in Figure 1c, the B-CW group showed better performance of NH$_4^+$−N reduction with an average removal efficiency of 86.3 ± 3.29%, while no significant difference of NH$_4^+$−N reduction in A-CW was observed when compared to the Control group (p > 0.05). As shown in Figure 1d, the CWs also has a good reduction performance of total phosphorus (TP), especially B-CW. Approximately 91.6 ± 3.82% of the phosphorus was removed in B-CW. Generally, the B-CWs showed a better reduction of nitrogen and phosphorus than A-CWs. The ion-exchange reactions between the functional groups and contaminants make biochar an effective adsorbent for inorganic nitrogen and phosphorus.
uptake by plants in CWs involved several steps. First, the PAHs pollutants in the substrate settled from wastewater. The BbFA that the uptake of BbFA in the root was higher than that in the stems.24 Further, the higher content of BbFA retained in the finally transferred to fillers such as AC or biochar according to ACE and Chao 1 values, which showed the following order: Control > B-CW > A-CW; a part of the abundance was also found in CWs with the addition of AC or biochar. Hence, the addition of AC and biochar could reduce the microbial species, which might be due to the re-establishment of dominant microbial groups under the changed substrate. Also, biochar and AC might increase the organic carbon content in the substrate by the release of carbon and polymerization, which was negative to microbial richness.29 A little lower community abundance was also found in CWs with the addition of AC or biochar according to ACE and Chao 1 values, which showed the following order: Control > B-CW > A-CW; a part of the community was also affected by the addition of carbon.30 As illustrated by the Shannon values, A-CW had the highest community diversity and correlated with higher metabolic versatility. B-CW had a lower Simpson value and also reflected higher community diversity. As reported in a previous research study, higher community diversity reflected that the microbes could utilize different organic sources.31

Previous studies showed that the nitrogen element contents in AC and biochar were decreased due to the denitrification process in CWs.21 Both AC and biochar could reduce the water-soluble fraction of pollutants and store the organic matter in the pores of the carbon. As shown in Figure 2, the Control group had the highest BbFA content in the substrate (1.10 ± 0.04 μg/g), followed by A-CW (0.84 ± 0.02 μg/g) and B-CW (0.64 ± 0.03 μg/g). The reduction in the BbFA content of the substrate treated with AC or biochar can be attributed to the immobilization of the bioavailable fraction of organic contaminants. Meanwhile, both AC and biochar could adsorb and retain contaminants due to their hydrophobic character and micropores.22 Hence, the addition of AC and biochar in the substrate could reduce the adverse impact of PAHs in plant growth and microbial reproduction.

2.2. Performance of CWs in the Substrate. Previous studies proposed that wetland plants could accelerate PAH degradation in sediments due to the rhizosphere exudates and radial oxygen loss.25 As an important removal pathway, the PAH content stored in plants was determined in each microcosm. The uptake of BbFA in the stem and root parts varied in each group. As shown in Figure 3, the BbFA content in the plant stem in the Control group was highest (3.15 ± 0.78 μg/g), followed by A-CW (1.76 ± 0.44 μg/g) and B-CW (1.58 ± 0.39 μg/g). The BbFA content in plant roots of each group showed a similar order: Control (9.23 ± 1.02 μg/g) > A-CW (7.41 ± 1.4 μg/g) > B-CW (6.16 ± 0.93 μg/g). The results reflected that the uptake of BbFA in the root was higher than that in the stem because the roots were in direct contact with the pollutants in the substrate settled from wastewater. The BbFA uptake by plants in CWs involved several steps. First, the PAHs were absorbed into the substrate and fillers such as AC or biochar, then absorbed by roots, and finally transferred to stems.26 Further, the higher content of BbFA retained in the roots was also due to the lower weight of the roots. Among the three groups, the total amount of BbFA was highest in the Control group, indicating that the substrate modification by AC and biochar in CWs may reduce BbFA accumulation in plants. There are two possible reasons for lower BbFA contents accumulated in plants with biochar or AC: first, the BbFA settled in the substrate and adsorbed into biochar or AC. Hence, the pollutants retained in the substrate were relatively lower and could be absorbed by wetland plants.25 Second, the release of carbon source from the biochar or AC enhanced the root exudates of plants, mainly due to the modified surface structures or provided the organic matter for BbFA.26 The root exudates could change the rhizosphere microbial activity and enhance PAH desorption in the substrate.27 Meanwhile, the adsorption of carbon fractions on the root surface might prevent plants from absorbing pollutants.25 The lower BbFA accumulated in plants with biochar and AC could reduce the toxic effects of PAH pollutants on plants.

2.3. Performance of CWs in Plants. Previous studies investigated the effects of PAH pollutants on plants.28 The biological scanning electron microscopy (SEM) images of AC and biochar before and after the experiment showed a rough surface morphology and high internal surface area (Figure 4). A large number of microbes were obviously attached in the pores and outside of AC and biochar after the experiment period, presented in all randomly selected images, including rod-shaped, micrococcus, and hyphal microorganisms (Figure 4c,d). Hence, both the AC and biochar possessed porosity for pollutants’ absorption and provided habitat for microbial growth.

Figure 2. BbFA content in the dry substrate of each group.

Figure 3. BbFA content in stem (upper) and roots (bottom) of wetland plants.
The 16S rRNA gene sequencing analysis reflected the microbial community in each group. As shown in Figure 5a, Proteobacteria and Actinobacteria were predominant at the phylum level and contributed to 50.7 ± 11.9 and 29.3 ± 6.2% of all detected OTUs in the three groups, respectively. Among them, the phylum of Proteobacteria was most abundant in A-CW (63.6%), followed by B-CW (48.4%). Both of them were higher than those in the Control group (40.2%). The Proteobacteria was highly related to nitrogen or carbon cycling, which was predominant in most wastewater treatment processes, including CWs. Proteobacteria contains many denitrifiers, which contributed to NO₃⁻–N removal with the addition of AC and biochar.33 As an important microbial type, the subgroups of Proteobacteria were analyzed under the class levels. Significant differences in Proteobacteria community composition were observed between three groups, as shown in Figure 5b. Among the Proteobacteria phylum, γ-proteobacteria and Actinobacteria were identified as the most abundant classes with the relative abundance of 44.2 ± 13.8 and 23.1 ± 3.7% of the three groups, respectively. Obviously, the A-CW and B-CW groups contained more γ-proteobacteria than the Control group, accounting for 59.9 and 38.5% of the microbial community, respectively, which contributed to the enhanced NO₃⁻–N removal performance with AC and biochar. The γ-proteobacteria also enhanced the biodegradation of organic matter and is presented as potential PAH degraders.34 The results for microbial community changed by AC and biochar in CWs were highly toward enhanced nitrogen and carbon removal performance.35

3. CONCLUSIONS
The addition of AC and biochar in the substrate can effectively enhance the removal performance of PAHs in surface-flow CW microcosms. At the same time, NO₃⁻–N removal performance was enhanced with these two carbonous materials. The BbFA contents absorbed by plants and retained in the substrate were decreased due to the addition of carbonous fillers. The carbonous materials could provide a breeding habitat for microbes and improve the microbial community toward enhanced denitrification and the PAH removal performance.
4. MATERIALS AND METHODS

4.1. Construction and Operation of CW Systems. To stimulate nature CWs, three surface flow CW microcosms were established under a transparent canopy. The CW microcosms were of PVC material with a length of 60 cm, a width of 40 cm, and a depth of 50 cm. The substrate was washed gravel (particle size 1−2 mm) with 30 cm depth. In experimental groups, equal volumes of biochar and AC with 20 cm depth were added to the substrate. The typical wetland plants acorus calamus were planted in microcosms, with a density of 10 individuals/per unit. The microcosms were divided into three groups as follows: (1) Control group, with no biochar or AC added; (2) A-CW, with AC added as the substrate; and (3) B-CW, with biochar added as the substrate.

The batch-operated procedure was implemented in the experiment, with the hydraulic retention time (HRT) of 3 days. After planting, the synthetic wastewater was added to each microcosm. The pollutant concentrations were according to the Wastewater Discharge Standard (GB 18918-2002), detailed as follows: NO₃⁻−N, 24.4 ± 0.48 mg/L; NH₄⁺−N, 14.3 ± 0.38 mg/L; TP, 3.21 ± 0.11 mg/L; and chemical oxygen demand (COD), 79.5 ± 2.86 mg/L. During the experimental period, the depth of overlying wastewater was maintained at 15 cm. The concentration of BbFA was 0.08 ppm in each microcosm. The BbFA was dissolved in acetonitrile before adding it to the synthetic wastewater due to its hydrophobicity.

4.2. Sample Collection and Analysis. 4.2.1. Wastewater Samples. The overlying wastewater was collected using a 100 mL polyethylene bottle. After the filtration through a 0.45 μm filter membrane, the water quality indexes such as TP, TN, NH₄⁺−N, NO₃⁻−N, NO₂⁻−N, COD, etc., were determined according to the standard method. The BbFA in the wastewater was extracted using a solid-phase extraction method.

Figure 5. Bacterial community composition at the phylum level (a) and subgroups of Proteobacteria at the class level (b), as revealed by high-throughput sequencing analyses.
The concentration of BbFA in organic solution was determined by GC/MS.

4.3. Chemicals. The biochar and AC were prepared by wetland plants according to the previous methods.38,39 The BbFA (>98% purity) was purchased from the Aladdin Reagent (Shanghai, China). The standard substitute for pretreatment extraction and internal standards were obtained from the ANPEL Laboratory Technologies (Shanghai, China) Inc. The solid-phase extraction membrane (SPMEM, C18 Disks) was purchased from Horizon Technology Inc. All of the organic solvents used in the experiment were of chromatographic grade and were purchased from Shanghai Sinopharm Pharmaceutical Co. LTD.

4.4. Statistic Analysis. The experimental results were analyzed by SPSS 11.0 (SPSS Inc., Chicago), and analysis of variance (ANOVA) was used for statistical analyses. The results were considered to be statistically significant when \( p < 0.05 \).

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**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

This work was supported by the National Natural Science Foundation of China (No. 51908326), the Science Foundation of Shandong Jianzhu University (Grant No. X180477X), and the Science Foundation of Qingdao University of Science and Technology (No. 120304300357S).

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