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Application of Modified Biochar in the Treatment of Pesticide Wastewater by Constructed Wetland

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Abstract: To explore the synergistic effects of modified biochar in the purification of herbicide-containing wastewater, the effect of biochar addition on the removal effect of the herbicide atrazine in wastewater was verified by the addition of biochar bags in a small reed bed-constructed wetland in the laboratory. The results showed that the addition of sulfuric acid-modified biochar could increase the removal rate of atrazine in wastewater from 50% to 70%, and the COD elimination rate in wastewater was from 66.7% to 86.7%. The addition of biochar to the constructed reed bed wetland improved the removal efficiency of total nitrogen and total phosphorus in the wastewater, and the outlet water from the constructed wetland reached the Class III level of China's surface water quality standard (the inlet water was inferior to Class V). The experimental design met the requirements of low-cost, generalized atrazine-containing wastewater treatment and thus could have the potential for wide application. The results reflected the application potential of modified biochar as a synergist in the treatment of herbicide wastewater in constructed wetlands.

Keywords: biochar addition; wastewater treatment; constructed wetland; herbicide atrazine; wetland design

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1. Introduction

With the rapid development of industry and agriculture, the amount of sewage produced is increasing daily. In China, a large volume of polluted water is discharged into the natural environment without being effectively purified, resulting in the deepening of the deterioration of the current water resources [1]. China is a large country with a huge population and limited farmland area per capita. As such, to increase food production, the use of herbicides has become increasingly frequent, resulting in a serious problem of pesticide pollution in water bodies [2,3].

Atrazine (6-chloro-N-ethyl-N′-(1-methylethyl)-1,3,5-triazine-2,4-diamine is a synthetic triazine herbicide used to control grassy and broadleaf weeds in sugarcane, wheat, conifers, sorghum, nuts, and corn crops [4]. Because corn has a detoxification mechanism for ATZ, such herbicides are often used in large quantities during corn cultivation [5,6]. However, while enjoying the increase in food production brought about by the application of herbicides, we often overlook the associated threats. For example, in a survey on the status of herbicide pollution in surface waters of key river basins in China, the detection rate of ATZ reached an astonishing 100% [7]. Meanwhile, the treatment technology of herbicide wastewater has been developed rapidly to meet the need for low-pollution or pollution-free waters. At present, the treatment methods for pesticide wastewater that have been applied include ultrasonic technology, photocatalysis, electrolysis, and biological methods [8–10]. Among these, biological methods are a current research focus in sewage purification. These methods are thought of as environmentally friendly for sewage treatment since pesticides can be degraded by animals, plants, and microorganisms without harmful effects on the environment.

Constructed wetland is an engineering technology that simulates natural wetlands and uses the physical, chemical, and biological synergy between wetland plants, substrates, and related microorganisms to purify sewage. Constructed wetland has a high pollutant removal rate, low investment and operating costs, convenient maintenance and management, and other advantages [11]. Constructed wetlands can effectively reduce herbicides/pesticides and other pollutants produced in agriculture. However, a constructed wetland has a certain threshold for a load of pollutants. When this value is exceeded, the environment deteriorates; this level is known as the "wetland capacity".

Biochar has attracted attention due to its high carbon content, large specific surface area, developed porosity, various surface functional groups, stable structure [12], and performance in strengthening nitrogen and phosphorus removal in sewage [13]. This paper aimed to modify the biochar to improve its adsorption performance for the herbicide atrazine and apply the modified biochar to the constructed wetland. This would not only strengthen the removal of nitrogen and phosphorus in the constructed wetland but also act as a buffer and adsorbent for herbicide-containing wastewater and reduce the damage to constructed wetland organisms and the spread of herbicide pollution in important water areas. The study provides a theoretical and practical basis for applying biochar to constructed wetlands and for the treatment and restoration of agricultural polluted water bodies.

2. Materials and Methods

2.1. Modification of Biochar

The biochar used in this experiment was high-temperature carbonized corn stalk in the form of filamentous powder, and its basic physical and chemical properties are shown in Table 1.

Table 1. Biochar properties.

Material	Raw Material	Specific Surface Area (m^2/g)	pH	Ash (%)	C (%)	N (%)	P (%)	K (%)	Cation Exchange Capacity (cmol/kg)
Biochar	Corn stalks	7.29	9.46	27.9	55.31	1.35	0.243	1.18	24.6

The prepared high-temperature carbonized corn stalk biochar was used as a raw material, and after being repeatedly rinsed with deionized water, it was placed in an oven to be dried for later use.

An appropriate amount of washed biochar was weighed, and different concentrations of phosphoric acid (H_3PO_4) [14,15], ferric chloride ($FeCl_3$) [16–18], sulfuric acid (H_2SO_4) [18–20], and potassium hydroxide (KOH) [21–23] were used to modify the biochar.

(1) H_3PO_4 modification: 11.76 mL of analytically pure (85%) H_3PO_4 was diluted to 20% H_3PO_4, and 50 mL of the solution was combined with 5 g biochar in a 50 mL beaker and allowed to soak for 3 h. Then, the obtained modified biochar was washed with pure water until the eluate pH was neutral, and solid-liquid separation was carried out by a vacuum filter for each wash. Finally, the obtained modified biochar was dried at 70 °C, ground, passed through a 60-mesh sieve, and saved for later use.

(2) Fe loading modification: 100 mL of 0.5 mol·L^{-1} ferric chloride solution and 5 g of dried biochar were combined in a 100 mL beaker, and 50 mL of ferric chloride solution was added. The beaker was placed on a stirrer for rapid stirring for 12 h, washed with deionized water until the eluate pH was neutral after each wash, and solid-liquid separation was carried out using a vacuum filter. The obtained modified biochar was dried at 70 °C, ground, passed through a 60-mesh sieve, and stored for future use.

(3) Acid (H_2SO4) or base (KOH) modification: 5 g of the prepared biochar was placed in a conical flask, and 50 mL of 10% H_2SO_4 or 3 mol·L^{-1} KOH solution was added and

stirred with a magnetic stirrer at 65 °C. The modified corn stalk biochar was washed with deionized water until the pH of the leachate was neutral, and then solid-liquid separation was carried out by a vacuum filter after each wash. The obtained modified biochar was dried at 70 °C to constant weight, ground, passed through a 60-mesh sieve, and stored for future use.

2.2. Adsorption Experiment

2.2.1. Adsorption Thermodynamic Experiment

The modified biochar (0.2 g) was placed into 50 mL polypropylene plastic centrifuge tubes and combined with 30 mL of atrazine solution; the atrazine concentrations were 0.5 mg·L^{-1}, 1 mg·L^{-1}, 5 mg·L^{-1}, 10 mg·L^{-1}, and 20 mg·L^{-1}. The tube was covered and sealed on a shaker at 25 °C (±0.5 °C) for 24 h at room temperature. The tube was then centrifuged at 4000 r·min^{-1} for 5 min on a high-speed centrifuge, filtered with a 0.45 µm membrane, and the concentration of atrazine in the supernatant was measured. The amount absorbed was calculated, and each treatment was repeated three times. The adsorption test was carried out in an airtight system with shading, and a blank control was set at the same time.

2.2.2. Adsorption Kinetic Experiments

The modified biochar (0.2 g) was placed into 50 mL polypropylene plastic centrifuge tubes and combined with 30 mL of atrazine solution with a concentration of 6 mg·L^{-1}, then covered and sealed at room temperature at 25 °C (±0.5 °C). The tube was placed in a constant temperature shaking box and oscillated at 200 r·min^{-1} for a certain period in the dark. Samples were taken at 30, 60, 180, 420, 720, and 1440 min. The sample was then centrifuged at 4000 r·min^{-1} for 5 min on a high-speed centrifuge, filtered with a 0.45 µm membrane, and the concentration of atrazine in the supernatant was measured. The adsorption amount was calculated, and each treatment was repeated three times.

2.2.3. Parameters of Testing Wastewater

Due to the large differences in the composition of farmland runoff in different places, the basic wastewater was prepared according to the relevant literature [24] and the technical specification for constructed wetlands "HJ−2005–2010" [25]. The characteristics of the wastewater are shown in Table 2.

Table 2. Parameters of the inlet wastewater (COD: Chemical Oxygen Demand; TP: Total phosphorus; TN: Total nitrogen).

Indicators	**COD**	**TP**	**TN**	**$MgSO_4·7H_2O$**
Content (mg·L^{-1})	150	5	10	24.6

2.3. Design and Commissioning of Simulated Constructed Wetland Device

The experimental device (Figure 1) was built based on a white polyethylene plastic bucket with a height of 50 cm, an inner diameter of 43 cm in the upper part, and an inner diameter of 33 cm in the lower part as a simulated vertical subsurface constructed wetland. The total volume was about 160 L. A water outlet faucet was installed 5 cm above the bottom of the bucket. A water receiving pot was put under the water outlet faucet. After the inlet water reached the receiving pot, the water was pumped up to the surface to complete the water circulation (Figure 1).

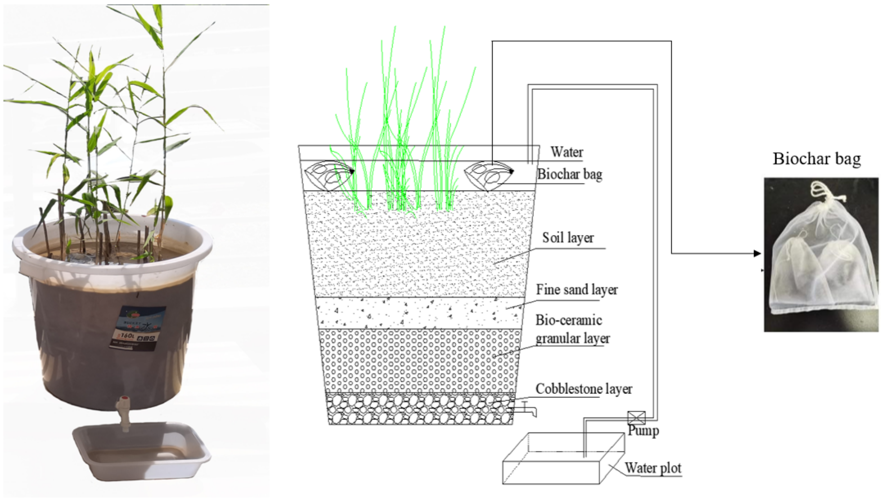

Figure 1. A schematic diagram of the simulated constructed wetland and the biochar bags.

The constructed wetland substrate consisted of a 25 cm soil layer, a 5 cm layer of fine sand, and a 10 cm layer of ceramsite, a 5 cm layer of cobblestone from top to bottom, and the layers were separated by fine gauze. Except for soil, substrates were washed with water before loading to avoid extrinsic contaminants.

The common reed (*Phragmites australis*) was selected as the constructed wetland plant. Reed seedlings (bud length 3–5cm) were bought online in January 2022. The seedlings were first placed in the culture medium at room temperature (about 10–20 °C) for 3 d of stabilization and then cultured in a green-house incubator with the addition of nutrient solution (25 °C constant temperature) for 10 d. After the buds grew significantly by 1–3 cm, the seedlings were transplanted to an outdoor soil ridge and were watered and provided with nutrient solution each day. After 50 d, the reed seedlings that had grown to 10–15 cm were transplanted into the constructed wetland device, and nine plants were evenly planted in each wetland device.

The constructed wetlands were set up on 1 March 2022. Plant nutrient solution was added to the wetlands in the early stages of construction to promote the growth of the plants on the premise of ensuring a high survival rate for the wetland plants. After 15 d, the plants reached a height of 20–30 cm, and then the next experimental step was started. The plants were cultivated with tap water for 1 week, and the water level did not exceed 5 cm above the soil surface. Due to water evaporation, plant absorption, and soil adsorption, the water level continued to decrease. It was necessary to supply water to the bucket at certain times and then empty the system, and the saturated water volume of the system was measured to determine the maximum water intake in the subsequent test process. After 1 week, the water level was set to 15 cm below the surface to promote the root system to go deeper into the ground for water uptake. After 2 weeks of cultivation, the basic wastewater was poured into the wetland system to have the plants adapt to the wastewater environment. During the experiment, the water level in the bucket was kept 5 cm above the soil surface by adding tap water to supplement the water consumed by evaporation, plant transpiration, and sampling. During the operation of the wetland system, the detection of DO, pH, COD, TN, and TP in the effluent was carried out every day to maintain control of the basic water quality indicators of the effluent.

In Figure 2, the four constructed wetlands are numbered 1, 2, 3, and 4 from right to left. In constructed wetlands No. 1 and 3, biochar bags (20 g biochar modified by H_2SO_4) were added. The bag was added to the overlying water layer above the soil surface, and the additional amount of the biochar bag was calculated according to the content of atrazine in the influent water body and the treatment amount of atrazine-containing wastewater. In this simulation experiment, two biochar bags were added to each simulated constructed wetland. Nos. 2 and 4 without biochar addition were used as controls to explore the removal effect of modified biochar on atrazine and the effect of modified biochar on nitrogen and phosphorus removal.

Figure 2. Simulated constructed wetland devices (from right to left, wetlands No. 1, 2, 3, and 4).

First, basic wastewater containing 0.3 mg/L atrazine herbicide was added to four groups of constructed wetlands, and the water quality in the four groups of constructed wetlands was monitored for 7 d.

2.4. Analysis Methods

During the constructed wetland experiment, the indicators COD, TN, and TP of the water were regularly measured. COD was determined by a Hach instrument, and TN was determined by potassium persulfate oxidation-UV spectro-photometry and TP ammonium molybdate spectrophotometry. DO and pH were measured with a pH meter (Shanghai Youke, PHS-3E).

2.5. Atrazine Determination

The presence of atrazine herbicide was detected by HPLC (high-performance liquid chromatography) (Thermo Fisher Scientific, System UltiMate 3000, Waltham, MA, USA). The detection conditions were as follows: the chromatographic column was the AR-C18 model; the particle size was 5 um, and the mobile phase was acetonitrile: water = 50:50 (v/v). The flow rate was 1.0 mL/min; the detection wavelength was 220 nm; the injection volume was 20 μL, and the column temperature was 30 °C.

3. Results and Discussion

3.1. Adsorption Experiment of Modified Biochar

As shown in Figure 3, at 25 °C and 0–12 mg·L^{-1} of equilibrium concentration, the adsorption capacities of modified biochars prepared by different modification methods for ATZ were varied. The biochar modified with sulfuric acid had the strongest adsorption capacity for ATZ.

As shown in the right panel of Figure 3, the equilibrium adsorption capacity of modified biochars prepared by different modification methods for ATZ showed a gradually increasing trend with time. Compared with the original biochar, the adsorption rate of ATZ by the modified biochar was significantly faster, and the equilibrium adsorption capacity of the modified biochar was higher than that of the original biochar. When the adsorption time reached 24 h, the adsorption amount of ATZ by each biochar remained stable and reached the adsorption equilibrium state.

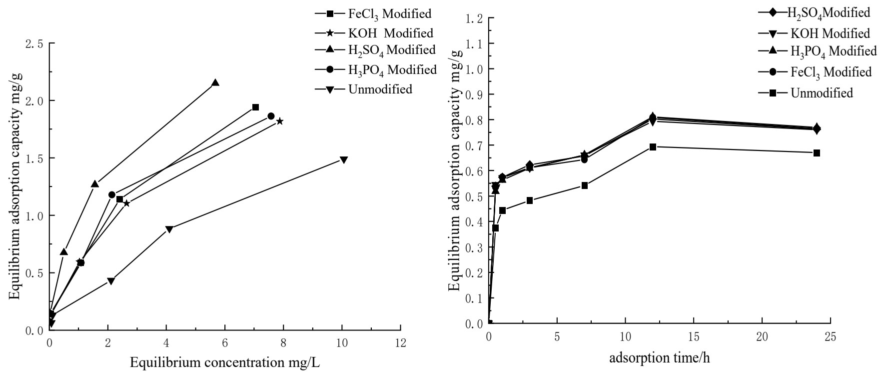

Figure 3. Adsorption isotherm curve (**left**) and adsorption kinetics curve (**right**).

The adsorption effect of the sulfuric acid-modified biochar was superior, and the modified biochar reached an adsorption equilibrium after 24 h of static adsorption, while the equilibrium adsorption capacity of various other modified biochars was nearly the same. In summary, the sulfuric acid-modified biochar was more suitable as an additional adsorbent for ATZ in constructed wetlands.

3.2. Change of Effluent Quality over Time in the Initial Stage of Simulated Constructed Wetland Operation

After the constructed wetland system was adapted to the wastewater environment, the first test was carried out on the effect of the wetland system on purifying ATZ-free wastewater. The change curves of pH, DO, COD, TN, and TP are shown in Figure 4.

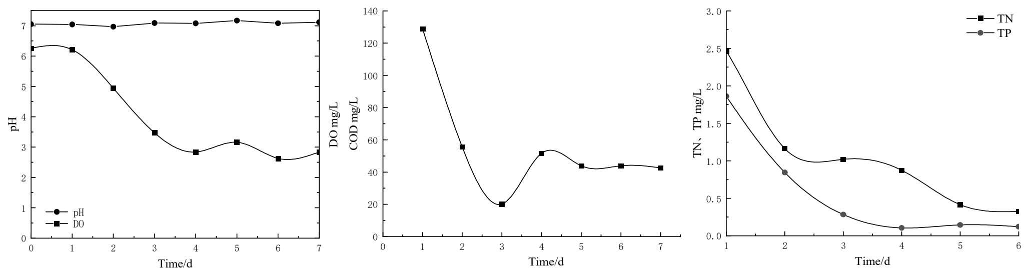

Figure 4. Variation in pH, DO, COD, TN, and TP of outlet water with time in the early stage of constructed wetland operation.

The above results were the first attempt to explore the purification effect of constructed wetlands on ATZ-free wastewater. The hydraulic retention time (HRT) of constructed wetlands was determined to be 3 d. As shown in Figure 4, according to the national water quality standards for surface water, the purification effect of constructed wetlands for TN and TP in water bodies can be upgraded from worse than class V level to class IV level, and the removal effect of COD was basically upgraded to class V level. The removal effect of COD was not as efficient as those for TN and TP; this may have been due to the plants in the wetland not being completely stabilized in the early stages of construction and operation of the constructed wetland and the low numbers of microorganisms in the wetland resulted in weak degradation of conventional pollutants. Therefore, after the test at this stage was completed, the constructed wetland was again cultivated with the basic wastewater, and the plants and microorganisms were further allowed to grow and reproduce. Before adding ATZ to the basic wastewater, the wetland could achieve an ideal effect on the removal of the basic wastewater.

3.3. Changes in Effluent Quality over Time during the Stable Operation of Simulated Constructed Wetlands

After one month of cultivation, the average height of the plants in the constructed wetland reached more than 30 cm. As described above, wastewater with 0.3 mg·L^{-1} ATZ was added to four groups of constructed wetlands, and the water quality in the four groups of constructed wetlands was monitored for 7 d. During the hydraulic retention time of 7 d, the impact of constructed wetlands on conventional water quality indicators and the removal of target herbicide ATZ was as follows (Figure 5).

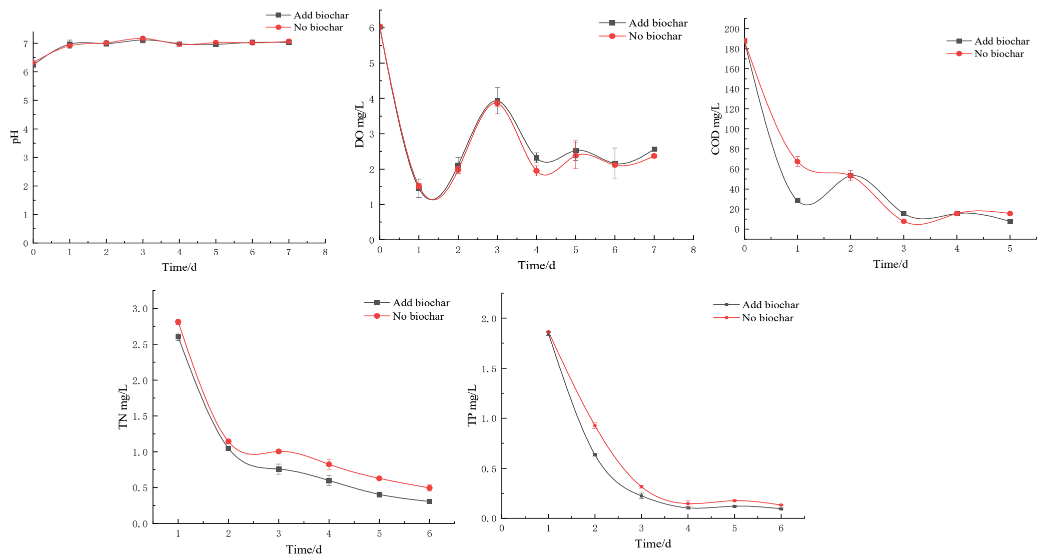

Figure 5. Comparison between with and without biochar addition in the variation of pH, DO, COD, TN, and TP of outlet water.

By comparing the effluent water quality with the national water quality standard, we found that when the HRT was 3 d, the removal effects for COD, TP, and TN by the constructed wetland reached the surface water quality standard of class III water (GB3838-2002) for the ATZ wastewater in which the inlet water pollution degree was worse than class V.

After the initial trial operation followed by one month of cultivation, the life activities of plants and microorganisms in the constructed wetland were more intense, and the degradation effect of pollutants was improved. At the same time, compared with the effluent water quality results from the early stages of the constructed wetland test, the reduction of TN, TP, and COD in the constructed wetland without biochar was significantly improved, and the removal effect of TN, TP, and COD in the constructed wetland with biochar had reached a higher level.

3.4. Removal of Herbicide Atrazine and Mechanism Analysis

According to the test results for effluent quality of the constructed wetland (Figure 6), the removal of the herbicide atrazine by the constructed wetland with modified biochar reached 70%, while that by the constructed wetland without modified biochar reached only 50%, the removal rate of which was at a medium level [26,27]. The herbicide content in the effluent of the constructed wetland was at a relatively higher level compared to those in the natural watershed since the reported highest atrazine content in surface water in some key watersheds in China was 1289.5 ng/L [7,24].Therefore, the constructed wetland we built can be applied well to the treatment of atrazine pesticide-containing wastewater.

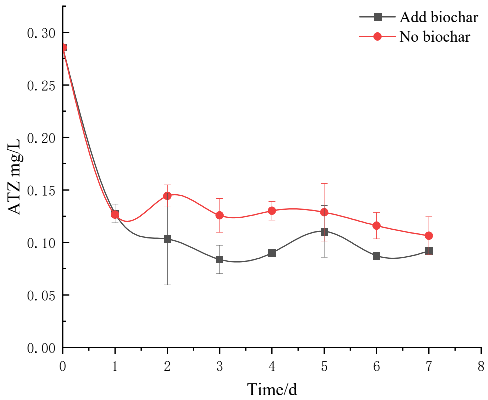

Figure 6. Changes in ATZ content in the outlet water with time.

The removal of ATZ herbicide in the experiment primarily depends on the adsorption of constructed wetland substrates and plants as well as the degradation by microorganisms, and a small fraction of ATZ pesticides can naturally degrade via physical, chemical, and biological degradation [28]. Therefore, the removal of herbicides by constructed wetlands in this experiment was divided into the following aspects.

As one of the main components in wetlands, plants play an important role in the degradation of pollutants [29]. Lv et al. [30] employed different plants in a vertical subsurface flow-constructed wetland and found that the plants (compared with no plants controls) had a strong influence on the removal of tebuconazole under the same influent concentration. This illustrated the promoting effect of plants on pesticide removal and the differences in the mechanism of pesticide removal by different plant species.

In the stress and physiological response test of reed to atrazine conducted by Wang et al. [31], when the concentration of atrazine was less than 8 mg/L, the reed could normally survive 1 week, but its normal growth was inhibited and stopped with time. At the same time, the peroxidase (POD) enzyme activity in reed plants increased under atrazine stress, and the POD enzyme activity showed a process of first increasing and then decreasing with the change in atrazine concentration, i.e., under mild stress, the plant itself could increase the activity of protective enzymes such as POD to remove harmful substances. In this experiment, the atrazine concentration was 0.3 $mg{\cdot}L^{-1}$, a state conducive to the degradation of atrazine by plants using their own mechanisms. Therefore, in this experiment, reed plants played a role in the degradation of atrazine herbicide.

The main role of microorganisms is to decompose organic matter in soil and water. There are many types of microorganisms in the soil, including bacteria, fungi, actinomycetes, algae, and protozoa, and the number is extremely large. Du [32] studied the bio-enhanced remediation of atrazine-contaminated soil, using the high-efficiency degrading bacteria HB-5 to degrade atrazine in the soil; the degradation rate was 3.3 times the rate under natural conditions. It can be seen that microorganisms had a significant promoting effect on the degradation of atrazine. The same results were also presented in the study of Kolekar [33].

The removal of atrazine by the matrix in this experiment can be divided into two parts, the degradation and adsorption of atrazine by soil and the ad-sorption of the constructed wetland filler matrix (in this experiment, this refers to the ceramsite adsorption). The adsorption in the soil is divided into organic matter adsorption (dominant effect) and clay mineral adsorption. Solid organic matter adsorption plays a major role in organic matter adsorption; at the same time, the process of herbicide adsorption in the soil is also affected by the soil adsorption coefficient and the influence of competitive adsorption of pollutants [29]. In this experiment, although atrazine herbicide was affected by the competition between the soil adsorption coefficient and pollutant adsorption, most were adsorbed by solid organic matter in the soil, which was one of the reasons for the rapid removal of atrazine from basic wastewater in the simulation [28]. In addition, the ceramsite of the constructed wetland filler matrix in this experiment may also play a role in the adsorption of ATZ [24]. Ceramsite is a new type of biofilm carrier filter material with light weight, large specific surface area, and strong adsorption capacity. A larger specific surface area and more internal pores provide more adsorption sites for ATZ adsorption, and this is helpful for the removal of ATZ from water.

Because of its pore structure, large specific surface area, and abundant surface functional groups, biochar is often used in modern scientific research for the adsorption treatment of various types of wastewater [34]. In recent years, biochar has gradually gained attention in fields such as environmental engineering. According to relevant studies [35–37], the types of wastewater treated by biochar and the raw materials of biochar are different, and the adsorption mechanism of biochar for solutes is also different. The removal mechanisms include the following: pore filling, diffusion and distribution, aromatic π or cationic π interaction, hydrogen bonding, electrostatic interaction and complexation, and surface adsorption.

Jiang [35] used the biochar prepared from palm to be modified by different methods to adsorb glyphosate and sulfamethazine, and results showed that the two materials could remove 99% of glyphosate (Gly) and sulfamethazine (SMT) at a concentration of 5 $mg \cdot L^{-1}$. The adsorption rate data can be well fitted by the pseudo-second-order kinetic model, and the adsorption of the modified biochar on herbicide/pesticide was primarily via complexation and electrostatic interaction.

In the study treating ATZ with biochar prepared from bagasse by Yu [37], it was found that the adsorption mechanism of biochar on ATZ included distribution and surface adsorption. At the same time, the adsorption of ATZ by biochar was spontaneous and belonged to an endothermic reaction. Zhao [36] used zinc chloride and phosphoric acid-modified biochar to treat ATZ and found that the removal mechanism of modified biochar for ATZ included physical adsorption and chemical bonding (π-electron interaction, hydrogen bonding). Isothermal adsorption experiments showed that after the biochar was modified with zinc chloride and phosphoric acid, the adsorption effect for ATZ was 8–9 times that of the unmodified biochar. The adsorption reaction of unmodified biochar to pesticide ATZ was endothermic, while the adsorption process of modified biochar to pesticide ATZ was an exothermic reaction, similar to the results of Yu [37].

In this experiment, as the exogenous adsorbent modified biochar also had strong adsorption ability for ATZ, compared with the constructed wetland without modified biochar, the pesticide removal effect of the constructed wetland was increased by 20%, showing that the modified biochar in this experiment was one of the main factors affecting the removal of pesticide atrazine in the constructed wetland.

In summary, under the combined action of modified biochar, constructed wetland plants, wetland substrates, microorganisms, and other influencing factors, ATZ was rapidly removed in a relatively short period. According to the above experimental data, when the hydraulic retention time (HRT) was 3 d, the herbicide ATZ removal rate of the constructed wetland without biochar was 50%, and the pesticide removal rate of the constructed wetland with modified biochar was 70%. At the same time, the removal rates of COD, TN, TP, and other water quality indicators reached more than 80%. From the perspective of its ecological benefits, biochar can not only purify water but can also adsorb and remove pesticides in the environment, effectively reduce the accumulation of toxic substances in the environment, and reduce toxic and harmful pollutants. This can protect not only the environment but also benefit human life and health. Based on our initial progress on microbial community assembly [38], we will further analyze the internal microcosmic mechanism of biochar improving the removal rate of atrazine in constructed wetlands in consideration of the microbial communities' responses.

4. Conclusions

In this experiment, the preparation process of modified biochar was explored, and the preparation method of modified biochar with the best adsorption performance was obtained by comparing five biochar adsorption kinetic curves and isothermal adsorption curves. At the same time, a vertical subsurface-flow constructed wetland was built, and the feasibility of applying sulfuric acid-modified biochar to constructed wetlands was verified through the results of herbicide-containing wastewater purification experiments. The following conclusions were drawn.

(1) The successful modification of biochar was demonstrated by the analysis results of adsorption kinetics curves and isotherm adsorption curves.
(2) Through the analysis and discussion of the best modification conditions, it was shown that the adsorption performance of modified biochar was better when sulfuric acid was selected as the modifier of biochar, and the modified biochar had better adsorption performance after magnetic stirring for 1 h at 65 °C.
(3) The test results for the removal effect of TP, TN, and COD in the constructed wetland in the later stages of the constructed wetland cultivation showed that without the addition of modified biochar, the constructed wetland had a good purification effect on the basic wastewater.
(4) The experimental results of adding modified biochar to the constructed wetland to simulate the removal of agricultural runoff wastewater showed that the constructed wetland with modified biochar in this design has a good adsorption and removal potential for herbicides/pesticides, results that verified the theoretical feasibility of adding sulfuric acid modified biochar to the constructed wetland for pesticide removal.
(5) Modified biochar and the constructed wetland formed a safe treatment system for herbicides/pesticides and other agricultural chemical pollutants in farmland runoff and wastewater; thus, the system provided a reference for the design of constructed wetlands for purifying herbicide/pesticide-containing agricultural wastewater.

Author Contributions: J.B.: Conceptualization, Funding acquisition. M.A.J., J.J.A. and W.P.: Conceptualization. R.X.: Conceptualization, Methodology, Validation. Y.H. (Yong Hu): Writing—original draft, Formal analysis, Visualization, Investigation. B.K.: Formal analysis, Visualization. C.W.: Data curation. Z.W.: Visualization. L.Z.: Data curation, Project administration. K.Z.: Methodology, Project administration. Y.H. (Yanping Hu) and Y.W.: Software, Investigation. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: All data generated or analyzed during this study are included in this published article.

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Conflicts of Interest: The authors have no relevant financial or non-financial interest to disclose.

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