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Water Purification Using Active Charcoal with Microbes and Chelated Iron Soaked into Its Micropores

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Abstract: Urbanization in China has led to a significant increase in surface water pollution, posing a threat to the health and safety of residents and hindering sustainable economic development. Individual traditional methods have been used to purify polluted water, including the use of bamboo-derived activated charcoal, microbial material, and zero-valent iron. However, these methods have been found to have certain limitations. This study investigates the effects of an activated charcoal material combined with beneficial microbes and chelated nano-iron in removing nitrates. The experiments were conducted at various scales, including a bench-scale study, and studies of a small river, sewage plant tailwater, and artificially constructed wetlands. The microbes used included *Bacillus* spp., *Lactobacillus* spp., and yeasts. During the fermentation process, nano-scale iron powder was added, resulting in the formation of bivalent iron ions under anaerobic conditions. These ions were subsequently chelated by organic acids. Bamboo-derived activated charcoal was then soaked in the fermented liquid, allowing the microbes, chelated iron ions, and organic acids to infiltrate the pores of the activated charcoal. This activated charcoal material, containing microbes and chelated iron ions, demonstrated effective nitrate removal in laboratory experiments and sewage plant tailwater treatment, and water purification in wetlands and rivers. It is important to note that this research solely focused on the removal of nitrates, and further studies are required to confirm its effectiveness in other aspects of water purification.

Keywords: active charcoal; chelated iron ion; compound microbes; nitrate; water purification

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1. Introduction

Serious pollution of aboveground water in China has been caused by rapid and concentrated urbanization. Unfortunately, water purification technology has not kept pace with this pollution [1]. The primary sources of water pollution include industrial wastewater, agricultural and livestock runoff, and domestic sewage. In recent years, there have been strict regulations for the pre-discharge treatment of industrial sewage, and most companies have the financial resources and capabilities to treat their sewage effectively. The treatment of domestic sewage in large cities is fairly standardized. Nevertheless, in rural areas, the treatment of domestic sewage, livestock and poultry waste, and aquaculture waste falls very short of meeting discharge standards. This has led to severe eutrophication in rivers, lakes, and coastal water, characterized by the excessive growth of algae and further deterioration of water quality [2]. Additionally, due to China's rapid urbanization, especially in small and medium-sized cities, sewage treatment and other environmental infrastructure have not kept pace. While the sewage outlets in major cities are relatively complete, the sewage network is not ideal, resulting in the majority of residents' domestic sewage not being able to enter the urban sewage system.

Domestic sewage contains a significant amount of organic matter, such as cellulose, starch, sugar, fat, and protein. Under anaerobic conditions with bacteria, these substances can easily generate odorous compounds, such as hydrogen sulfide and mercaptans [3]. The effluent from sewage treatment plants is often not allowed to be discharged because it does not meet nitrate concentration standards. Excessive nitrates also affect the water quality of rivers and lakes. High nitrate concentrations lead to excessive algae growth, exacerbating water pollution and sometimes causing the death of fish and shrimps [4]. Nitrate pollution in aboveground water mainly comes from agricultural fertilization, animal husbandry, and domestic sewage. Excessive nitrate is known to cause methemoglobinemia and cancers [5,6].

Commonly used technologies in sewage treatment and water purification include activated charcoal, iron powder or iron-containing compounds, and various bacteria, including photosynthetic bacteria [7,8]. Activated charcoal possesses a highly developed pore structure and demonstrates a strong capacity for adsorption on both its surface and interior [8]. Moreover, it can be regenerated and reused multiple times. Traditionally, activated charcoal has found extensive use as an adsorbent and has been employed since 1932 to treat tap water in Chicago [9]. Over the years, it has been increasingly utilized in water treatment and has become a research focal point worldwide. Nowadays, the applications of activated charcoal widely span across various industries, including chemical smelting, pharmaceutical engineering, wastewater treatment, gas purification, drinking water purification, and the food industry, primarily for treating waste gas, wastewater, and drinking water [9]. The Japanese were the first to apply photosynthetic bacteria and *Bacillus* spp. in wastewater treatment and water purification [10]. Subsequently, lactic acid bacteria and yeast were included. The exact mechanisms of action have yet been clearly defined. Under water conditions, the symbiotic proliferation of various bacteria can fully utilize a good microecosystem to effectively complete the complex process of pollutant degradation. Microbial technology has been applied to the purification of wastewater, rivers, and lakes [11,12]. Compared to traditional biological methods, compound microbial materials not only select and domesticate microorganisms, but also combine bacteria and enzymes with different functions [13]. They can incorporate optimal strains for different environments, such as natural water bodies, domestic sewage, and industrial wastewater. These materials can be used in both anaerobic and aerobic states, even in the presence of multiple pollution sources [12]. The substances and secretions they produce during growth become the substrates for their respective or mutual growth. This symbiotic relationship forms a complex and stable micro-ecosystem that accelerates the degradation of organic matter and performs various functions. Upon activated charcoal, the probiotics are concentrated and able to interact better. This ensures that the compound microbes are securely held in the activated charcoal and not washed away during the sewage purification process.

The water treatment industry has shown great interest in using iron powder, especially zero-valent iron, for sewage treatment [14]. Zero-valent iron is a cost-effective and efficient treatment agent because its standard oxidation electrode has low potential, enabling it to reduce various contaminants, including nitrate. It is also more economical and does not require additional treatment. Previous studies have extensively studied the use of zero-valent iron in wastewater treatment, showcasing its effectiveness in treating difficult wastewater, such as that contaminated with arsenic, phenol, dyes, and nitrate-eutrophicated water [8,15,16]. In recent years, nanometer zero-valent iron (nZVI) has gained attention due to its finer particle size, larger specific surface area, higher reactivity, and easy oxidization. Studies have demonstrated that nZVI eventually oxidizes to a complex of iron oxide and fibrous iron ore, especially when in an acidic environment [17]. Several mechanisms, including activated charcoal, nano-iron powder, and microbes, have been described for water purification [8,14,17]. However, there are certain disadvantages to using zero-valent iron powder for reducing nitrate in water. Firstly, the pH of the reaction must be strictly regulated. Secondly, after the reaction, there is an increase in ammonia nitrogen and nitrite nitrogen in the water, which requires additional treatment. Despite these drawbacks, the iron reduction method remains a popular choice as it does not have any specific environmental requirements other than pH regulation. Currently, there are several improvement measures that have been proposed, suggesting that using zero-valent iron with auxiliary methods to remove nitrate from water is an emerging research area [14–16].

This research promotes the concept of using waste materials, such as activated charcoal, to conserve resources and protect the environment. Meanwhile, we aim to develop a composite material using activated charcoal, nano-iron powder, and compound microbes, and investigate its effectiveness in removing nitrate from water. Our focus is on the reduction in nitrate ion concentration as an index of water purification; however, the impact on COD (chemical oxygen demand), phosphorus, and heavy metals will be explored in future research projects.

2. Materials and Methods

2.1. Preparation of Compound Microbial Liquid

The raw materials listed in Table 1 were mixed and added to a stirring tank. The raw material liquid was aerated and thoroughly stirred to ensure saturation of dissolved oxygen. The stirred fermentation liquid was then transferred to a fermenter, closed, and fermented at 36.8 °C for 72 h. During the first half of the fermentation period, aerobic fermentation took place to consume dissolved oxygen. In the second half, the fermentation automatically transitioned to the anaerobic stage. After fermentation was completed, the fermentation liquid was transferred to a ripening tank, which was then closed for an additional 4 weeks to complete residual fermentation of remaining sugar and prevent gas release, thus avoiding container expansion.

Table 1. Preparation of raw materials for fermentation broth of compound microbial fermented liquid (quantity unit: kg).

Material	Quantity	Remarks
Sucrose	10~30	50 kg of molasses (with a sucrose content of 60%) can also be used as a substitute to reduce costs
Sodium glutamate	1~3	
Salt	0.5~1.5	Kosher salt containing a variety of inorganic elements is preferred
Table salt	1~3	
Aleurone extract	50~150	Equivalent of 5 kg dried wheat bran
Tap water	600~831	
Carbamide	1~3	
Liquid tea	50~100	Dry green tea 100 g
Microbial inoculant	50~100	
Total	1000	

2.2. Preparation of Activated Charcoal Containing Microbes and Chelated Iron

Bamboo-derived activated charcoal (BAC, referred to as "activated charcoal" or "charcoal" in this article) was purchased from Dingcheng-Ou-Bao Charcoal Corporation in Changde, China. The activated charcoal was processed into blocks or rods of various sizes. The process involved sealing the bamboo-derived activated charcoal in a plastic vacuum bag, removing the air from the micropores of the charcoal, submerging the bag in fermentation liquid, breaking the bag, and allowing the liquid to be absorbed into the activated charcoal under negative pressure. This resulted in activated charcoal with microbes and chelated iron soaked in, known as ACMI.

2.3. Water Purification Experiment in the Laboratory

In the laboratory water purification experiment, contaminated water with 200 ppm of nitrate ions was placed in a 1000 mL beaker. A gauze pad absorbing 100 cm^3 of ACMI was suspended in the water. The beaker was positioned on a magnetic stirrer, which stirred the water, while maintaining a temperature of 22 °C. Water samples were taken every 10 min to determine the nitrate concentration.

2.4. Purification Experiment of Polluted River Water

An ACMI-soaked gauze pad was sewn into two layers of chemical fiber cloth to create a blanket. The blanket was equipped with fishing net foot lead drops on one side and tied to floating balls on the other side. In a 5 m wide and 100 m^2 test area of a small, slow-moving river, ACMI blankets were set every 5 m. After 24 h, the nitrate ion concentration was measured at each interval, using the water sample upstream of the ACMI blanket as a control.

2.5. Repurification of Tailwater in Sewage Treatment Plant

The tailwater from a wastewater treatment plant has a nitrate ion concentration of 200 ppm and needs to be re-treated. In the repurification of tailwater in this sewage treatment plant, a wire mesh container measuring 4 m long, 2 m wide, and 1 m high was used. ACMI blankets covered the bottom and four walls of the container, with longitudinal ACMI blankets arranged every 0.5 m along the long side, creating 6 compartments. The container was placed at a 15° tilt, and the tailwater was injected into the first compartment, flowing down the slope through 7 ACMI blankets. Water samples were taken after each blanket to determine nitrate concentration.

2.6. Artificially Constructed Five-Step Wetlands

The water purification experiment took place in artificially constructed wetlands located along a small river in the suburb of Haikou City, China. Figure 1 illustrates how the river water was directed into the constructed wetlands for purification, and then, returned to the river, similar to hemodialysis treatment for kidney patients. The first-step wetland featured *Phragmites australis* (Cav.) Trin. ex Steud. as the dominant plant. *Thalia dealbata* Fraser was the main plant in the second-step wetland, and rice (*Oryza sativa* L.) was used in the third-step wetland. *Eleocharis dulcis* (Burm. f.) Trin. ex Hensch.) was the plant in the fourth-step wetland, followed by *Oenanthe javanica* (Blume) DC in the fifth step. Each plant band had a width of 2 m and was 30 m long. A 0.5 m walkway separated the adjacent plant bands, and a mix of aquatic dwarf water plants, including *Cynodon dactylon* (L.) Persoon, *Vallisneria natans* (Lour.) Hara, and *Euphrasia pectinata* Ten., were planted in these areas. The reed belt and the dwarf water grass belt were repeated eight times, with each wetland being 20 m wide and 30 m long. At the end of each wetland step, there was an ACMI filter dam standing 0.5 m above the ground level. These dams consisted of two layers of barbed wire placed 0.3 m apart, sandwiched between bamboo-derived activated charcoal bars measuring 0.5 m. As mentioned earlier, the bamboo-derived activated charcoal was soaked with microbial liquid containing chelated iron. The water discharged from the ACMI filter dam entered a stabilization pond measuring 7 m by 3 m. This pond was planted with *Myriophyllum verticillatum* L. The purified water from the previous wetland flowed into the next wetland. Finally, the water from the fifth-step wetland was discharged into a pond containing foxtail algae (*Myriophyllum verticillatum* L.). Aquatic grasses were grown on the bottom of the pond, while slope-protecting grass (*Cynodon dactylon* (L.) Persoon) was cultivated on the slope of the stream bank. The purified water was then released into the river. After the fifth-step wetland, there was a *Myriophyllum* pond, which remained filled with water for 24 h. Subsequently, 300 m^3 of river water was added to the water inlet of the first-step wetland every day. Before refilling the water, samples were collected at five locations within each step of the wetland using the diagonal sampling method.

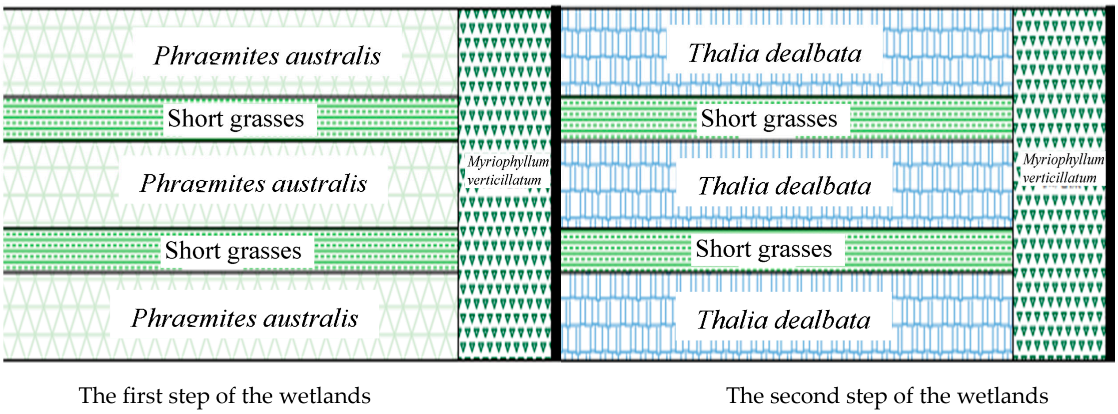

Figure 1. Illustration of the steps of the wetlands.

2.7. Determination of Nitrate Concentration

Nitrate concentration was measured in rivers, in wetlands, and in the laboratory using Attenuated Total Reflectance Fourier Transform Infrared spectroscopy (ATR-FTIR) coupled with a deconvolution algorithm [18].

3. Results

3.1. Results of Water Purification in Laboratory

To purify the polluted water, a gauze pad of activated charcoal with microbes and chelated iron ions (ACMI) was placed in a 1000 mL beaker filled with the water. The beaker was then agitated using a magnetic stirrer to release the chelated iron ions, microbes, and fermented liquid into the water, promoting a reduction of nitrates. Water samples were collected every 10 min to determine the nitrate concentration. The change in nitrate concentration is shown in Figure 2. The results of the simulated experiment demonstrate that the nitrate levels steadily decreased over time (Figure 2). Initially, in the presence of chelated irons, the nitrate concentration remained unchanged for the first 0 to 5 min. However, after 5 min, the nitrate concentration decreased significantly. From 30 to 60 min, the decline in nitrate concentration was more gradual, indicating that most of the nitrate ions in the water had been degraded. At 65 min, the nitrate concentration reached its lowest value of approximately 35 mg kg^{-1}. The change in nitrate concentration (N) followed the right side of the modified Gaussian equation $N = N_M e^{-\alpha(t-\tau)^2} + N_R(1 - \beta(t-\tau)^2)$. The actual formula is $N = 160e^{-0.0036(t-3.2)^2} + 50\ (1 - 0.000085(t - 3.2)^2)$. A higher value of α corresponded to a faster reduction of nitrate, while a smaller value of β indicated a slower reduction of nitrate in the latter half of the curve.

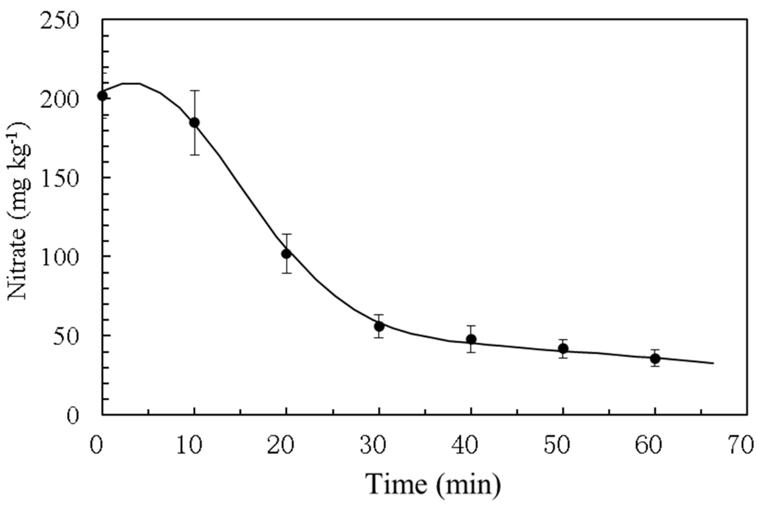

Figure 2. In the presence of the ACMI pad, nitrate concentration changed with the reaction time.

3.2. Results of Purification in Polluted River Water

In this in situ experiment, water samples were collected from an upstream location without an ACMI blanket as the control. After 24 h, additional water samples were taken to measure the nitrate concentration. The results are shown in Figure 3. The initial nitrate concentration was 200 mg kg^{-1}. When the number of ACMI blankets was increased from 0 to 3, the nitrate concentration dropped significantly to 75 mg kg^{-1}, reaching half of the initial level. However, further increasing the number of ACMI blankets did not result in a significant increase in nitrate degradation, indicating that ACMI blankets effectively reduce nitrate in polluted water. The change in nitrate concentration followed a modified negative exponential curve pattern, $N = (N_M - N_R)\ e^{-\alpha t} + N_R\ (1 + \beta t)$. The actual equation is $N = (204 - 10.5)\ e^{-0.46t} + 10.5\ (1 + 0.307t)$. A higher value of α corresponded to a faster reduction of nitrate in the initial stage, while the value of β correlated with the steepness of the second half of the curve.

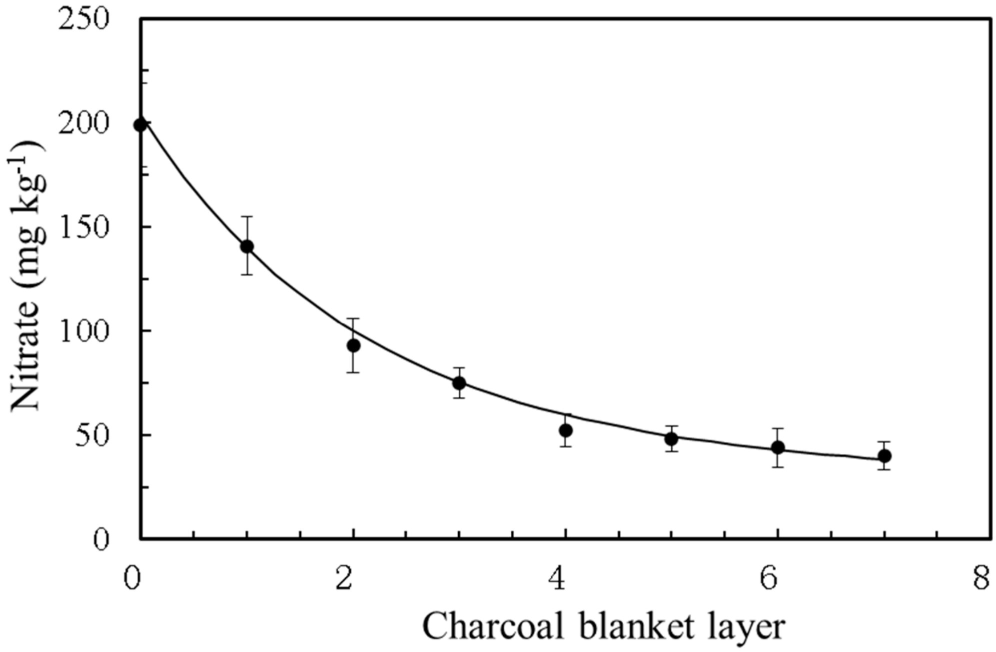

Figure 3. In the presence of the ACMI blanket, nitrate concentration decreased as the blanket layer increased.

3.3. Results of Repurification of the Tailwater in Sewage Treatment Plant

Although the sewage treatment plant successfully purifies industrial, domestic, and agricultural sewage, the nitrate concentration in the tailwater exceeds the discharge standard at 200 ppm. Therefore, the tailwater requires additional purification. In this experiment, we passed the tailwater through eight layers of boxes equipped with ACMI in succession. We then measured the nitrate concentration in the tailwater after it passed through the boxes. The results are shown in Figure 4. After passing through two layers of the ACMI box, the nitrate concentration in the tailwater sharply decreased from 150 mg kg^{-1} to 70 mg kg^{-1}. As the tailwater continued through the fourth layer, the decrease in nitrate concentration became much gentler. This indicates that ACMI effectively degrades nitrate in the tailwater of the sewage treatment plant. Similar to Figure 3, the nitrate concentration dropped rapidly from 150 mg kg^{-1} to 45.1 mg kg^{-1}, meeting the discharge standard. This change in nitrate concentration follows the modified negative exponential curve model, with the actual equation being $N = (150 - 45.1)\ e^{-0.51t} + 45.1\ (1 - 0.0098t)$. The value of α is related to the rate of nitrate reduction, while the value of β is related to the steepness of the second half of the curve.

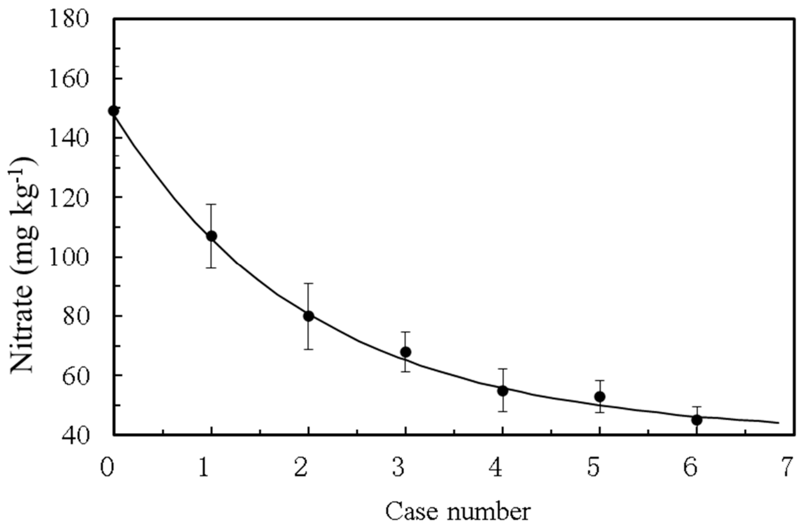

Figure 4. Nitrate concentration decreased as ACMI cases increase in the experiment of tail water repurification in sewage treatment plant.

3.4. Results of Water Purification in Artificially Constructed Five-Step Wetlands

In our study of the artificially constructed five-step wetlands as a water purification method, we observed that the initial river water entering the first-step wetland belonged to the national Level IV water category. After going through the second-step wetland, the nitrate concentration was already very low, reaching 3 mg kg^{-1}. As the wetland steps increased, the nitrate concentration continued to decrease, following a negative exponential curve model. The actual equation for this trend is $N = (24.5 - 0.8)\, e^{-1.33t} + 0.8\, (1 - 0.045t)$ (Figure 5). Although this experiment did not measure the harvest of wetlands as cultivated land, we were able to harvest reeds, aquatic flowers, feed rice, and vegetables.

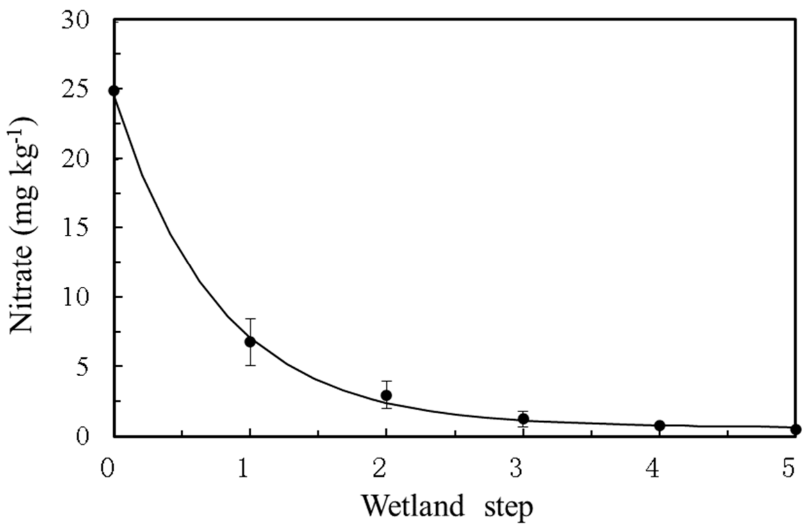

Figure 5. Nitrate concentration decreased as the wetland steps increased in the experiment of water repurification in five-step wetlands.

4. Discussion

The Chinese government has implemented strict environmental protection policies to improve industrial sustainability through cleaner production and a circular economy. Small chemical businesses struggle to meet national wastewater discharge standards due to funding and technology limitations, resulting in closures. However, in China, more than 1/3 of rivers and a higher proportion of urban water are seriously polluted, affecting drinking water quality and public health [19,20]. Water pollution is mainly caused by inadequate sewage treatment with low technology. Activated charcoal is often considered an ideal material for sewage treatment due to its abundant pore structure and strong adsorption capacity on its inner surface [21]. Additionally, it can be reused multiple times.

In this study, a new material called ACMI (activated charcoal soaked with microbial fermented liquid and chelated iron ions) was developed. We conducted a comprehensive study on the purification of wastewater from various sources using ACMI.

ACMI contains organic acids that effectively chelate iron ions and prevent their oxidation. By soaking activated charcoal in microbial fermented liquid, compound microbes, chelated iron, and organic acids attach to the inner micropores of ACMI. ACMI acts as a barrier, preventing chelated iron, microbes, and their secretions from being washed away by flowing water.

In the laboratory water purification experiments, the nitrate concentration decreased significantly within 60 min, indicating that most of the nitrate ions were degraded in the absence of chelated irons.

Afterwards, ACMI was used to create an ACMI blanket, which consisted of two layers of chemical fibers and was suspended in polluted river water. The nitrate ion concentration dropped significantly with the increment of the blankets, indicating that ACMI blankets were able to effectively reduce nitrate.

ACMI was also found to be effective in repurifying the tailwater of a sewage treatment plant. The nitrate concentration sharply decreased after passing through ACMI layers, which indicates the effectiveness of ACMI in degrading nitrate in the tailwater.

Furthermore, the ACMI material was applied in a five-step wetlands system, which significantly reduced nitrate levels in river water. In the wetlands experiment, ACMI was sandwiched between two pieces of wire net to form a wall or dam. Water was first purified by aquatic plants, and then, further purified by the ACMI dam or wall. The purification effects were attributed to the factors within the ACMI material and the combination of ACMI and wetland aquatic plants.

The key component of ACMI was the chelated iron ion, which served as the electron donor in redox reactions. Previous studies have focused on the use of zero-valent iron in water purification, but it requires a high pH for nitrate removal. ACMI, on the other hand, does not change the pH of river water, but its internal pH is stable, ensuring nitrate reduction. The components and conditions within the micropores of ACMI are not easily affected by external water, allowing iron ions and microbes to attach to the inner surface of the micropores. The compound microbial fermented liquid used in this study stabilizes iron ions and serves as an electron donor for redox enzymes. This liquid enters the micropores of activated charcoal, avoiding removal by running water and providing long-term effects.

China, being rich in bamboo resources with a short growth cycle, can utilize bamboo charcoal as a functional and environmentally friendly biological adsorption material. Bamboo charcoal has a wide range of sources, low production costs, high mechanical strength, and significant adsorption effects. By combining microbial technology with bamboo charcoal material, the quality of water can be effectively improved, greatly enhancing the purification efficiency of bamboo charcoal water. Particularly, our results show that ACMI has a significant effect on nitrate removal and is cost-effective. This approach has broad application prospects and significant potential for development in the field of sewage treatment [9].

Currently, bamboo charcoal material is widely used in the treatment of industrial and agricultural sewage as well as domestic sewage [8]. It has become a substitute for relatively expensive wood-activated carbon, addressing the development limitations of the wood-activated carbon industry caused by wood shortages. The development and utilization of bamboo charcoal material can deepen the theoretical research on water quality purification, thereby protecting the environment, promoting local agricultural and forestry economies, and driving the industrial development and sustainable utilization of bamboo resources.

5. Conclusions

Sewage with high levels of chloroacetic acid, nitrate, nitrite, heavy metals, and organic matter poses a risk to human health [4,22]. Therefore, traditional sewage purification methods need improvement to be more effective and cost-efficient. In this study, we used a micro-ecological microbial solution that contains both aerobic and anaerobic microorganisms. These microorganisms coexist and produce active substances and secretions during fermentation, which serve as substrates and nutrients for their growth [23]. The solution also contains various organic acids that effectively chelate iron ions, preventing them from losing their properties due to oxidation or reduction.

This symbiotic relationship speeds up the degradation of organic matter and the reduction of nitrate. By immersing activated charcoal in the micro-ecological microbial solution, the active ingredients attach to its micropore structure. This not only enhances the interaction between the microorganisms, but also prevents them from being washed away during the experiment. The resulting ACMI can be combined with two layers of chemical fiber to create an activated charcoal blanket that can be suspended in various polluted water bodies to assess its ability to reduce nitrate. ACMI utilizes a combination of biological and physical methods, effectively purifying water quality and avoiding secondary pollution.

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References

1. Zhou, L.; Li, Y. A study on China's water pollution status and water environment management strategy. *Environ. Develop.* **2018**, *30*, 51–52.
2. Qin, B.Q.; Zhan, Y.L.; Zhu, G.W.; Gao, G. Eutrophication control of large shallow lakes in China. *Sci. Total Environ.* **2023**, *881*, 163494. [CrossRef]
3. Chen, F. Current situation and treatment measures of urban environmental pollution in China. *Leath. Manuf. Environ. Tech.* **2022**, *3*, 117–119.
4. Xu, J.N.; Chen, Y. Water pollution in our country in the current situation. *Guangdong Chem. Indus.* **2014**, *41*, 143.
5. Shrock, D.; Krasowski, M. Methemoglobinemia due to dietary nitrate. In *Toxicology Cases for the Clinical and Forensic Laboratory*; Ketha, H., Ed.; Academic Press: New York, NY, USA, 2020. [CrossRef]
6. Giles, E.; Purcell, S.; Olson, J.; Vrieling, A.; Hirko, K.; Woodruff, K.; Playdon, M.; Thomas, G.; Gilmore, L.; Moberly, H.; et al. Trends in diet and cancer research: A bibliometric and visualization analysis. *Cancers* **2023**, *15*, 3761. [CrossRef] [PubMed]
7. Shao, Y.; Zhong, H.; Wang, L.; Elbashier, M.M.A. Purification Effect of the Aquaculture Wastewater and Sediment by Microbial Nanospheres with Different Material Ratios and Dosing Methods. *Sustainability* **2020**, *12*, 1462. [CrossRef]
8. Sen, T.K. Application of Synthesized Biomass Bamboo Charcoal–Iron Oxide "BC/Fe" Nanocomposite Adsorbents in the Removal of Cationic Methylene Blue Dye Contaminants from Wastewater by Adsorption. *Sustainability* **2023**, *15*, 8841. [CrossRef]
9. Saadallah, A.; Alsultan, M.F.; Hameed, W.; Swiegers, G.F. High surface area activated charcoal for water purification. *J. Composites Sci.* **2022**, *6*, 311. [CrossRef]
10. Kobayashi, M. Utilization and disposal of wastes by photosynthetic bacteria. In *Microbial Energy Conversion*; Schlegel, H.G., Barnea, B., Eds.; Pergamon Press: Oxford, UK, 1977; pp. 442–445.

11. Chen, D.Q.; He, H.; Chen, Y.Q. Purification of nitrogen and phosphorus in lightly polluted landscape river by effective microorganisms combined with submerged plants. *App. Mech. Mate.* **2013**, *316–317*, 430–434. [CrossRef]
12. Tekebayeva, Z.B.; Bazarkhankyzy, A.; Bissenova, G.; Serikovna, S.Z.; Temirkhanov, A.Z. Isolation of new strains of microorganisms for bio-purification of polluted reservoirs of Northern Kazakhstan. *Intl. J. Des. Nat. Ecodynamics* **2023**, *18*, 661–668. [CrossRef]
13. Liu, L.; Wang, J.; Gao, J.; Wang, Q.; Lao, T. Research on Rural Wastewater Treatment Technology in Northwest China Based on Anaerobic Biofilm Coupled with Anaerobic Baffle Plate Reactor (ABR) Technology. *Sustainability* **2023**, *15*, 8957. [CrossRef]
14. Tichem, T.M.; Wang, Y.; Gameli, R.B.H.; Mbage, B.; Li, B. Multicomponent Adsorption of Pollutants from Wastewater Using Low-Cost Eco-Friendly Iron-Modified Rice Husk Biochar in the Era of Green Chemistry. *Sustainability* **2023**, *15*, 16348. [CrossRef]
15. Boudia, S.; Fernane, F.; Sharrock, P.; Fiallo, M. Plaster of Paris containing zero valent iron particles: Designing a permeable reactive barrier, used for remediation of 4-nitroaniline pollution. *J. Sust. Constr. Mater. Tech.* **2021**, *4*, 124–134.
16. Al-Rabadi, S.J.; Alawamleh, K.A.B.; Alawamleh, H.L.; Al-Samrraie, A.; Al-Tabbal, J. Synergistic impact of nanoscale zero-valent iron in anaerobic co-digestion treatment and biogas upgrading from the organic fraction of municipal solid waste-activated sludge. *Waste Biomass Valor.* **2023**, 1–16. [CrossRef]
17. Greenlee, E.; Maw, E.L. Neuroergonomics and vigilance: Probing the event-related cerebral hemodynamic response. *Proc. Human Fact. Ergon. Soc. Ann. Meet.* **2019**, *63*, 1674–1675. [CrossRef]
18. Wu, K.; Fei, M.; Li, Z.; Wei, C.; Gan, F.; Du, C. In-situ rapid monitoring of nitrate in urban water bodies using Fourier transform infrared attenuated total reflectance spectroscopy (FTIR-ATR) coupled with deconvolution algorithm. *J. Environ. Manag.* **2022**, *317*, 115452. [CrossRef] [PubMed]
19. Zhang, Q.N. City suburb of water environment management reform. *Sci. Tech. Innov.* **2014**, *18*, 56.
20. Zhou, Y.; Wang, X.M.; Li, W.Y.; Zhou, S.Y.; Jiang, L.Z. Water quality evaluation and pollution source apportionment of surface water in a major city in Southeast China using multi-statistical analyses and machine learning models. *Int. J. Environ. Res. Public Health* **2023**, *20*, 881. [CrossRef]
21. Reza, M.S.; Afroze, S.; Kuterbekov, K.; Kabyshev, A.; Bekmyrza, K.Z.; Haque, M.N.; Islam, S.N.; Hossain, M.A.; Hassan, M.; Roy, H.; et al. Advanced Applications of Carbonaceous Materials in Sustainable Water Treatment, Energy Storage, and CO_2 Capture: A Comprehensive Review. *Sustainability* **2023**, *15*, 8815. [CrossRef]
22. Lu, N.; Villa, K. Agricultural support and contaminated spillovers: The effects of agricultural water pollution on adult health in China. *Appl. Econ. Perspect. Policy* **2021**, *44*, 788–821. [CrossRef]
23. Suez, J.; Zmora, N.; Segal, E.; Elinav, E. The pros, cons, and many unknowns of probiotics. *Nat. Med.* **2019**, *25*, 716–729. [CrossRef] [PubMed]