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Mitigation of Greenhouse Gas Emissions and Enhancement of Water Quality in Constructed Wetlands: A Case Study in Cold Temperate Regions of China

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ABSTRACT

Constructed wetlands (CWs) have emerged as a green solution for wastewater treatment in many regions. However, their efficacy can be impacted by temperature fluctuations, and the potential emission of greenhouse gases may offset their environmental and ecological benefits. This study focuses on the effluent of one wastewater treatment plant in the cold temperate zone of northern China. It investigates the supplemental treatment effects of CWs on effluents from conventional sewage treatment plants using three plant species: Phragmites australis, Scirpus validus, and Typha orientalis for phytoremediation. Under 15°C, CWs showed moderate removal efficiencies for COD (35.71-40.28%) and TN (28.79-33.59%), with relatively low CO_2 emission flux (43.56-176.56 mg/m^2/h) and global warming potential (GWP,2.815-6.613 mg/m^2/h). Among the plants, Scirpus validus demonstrated superior pollutant removal and lower greenhouse gas (GHG) emissions, making it a prime candidate for future use. Additionally, it explores the incorporation of biochar into CW substrates to simultaneously enhance water quality (+9.99% for COD and +22.13% for TN) and mitigate GHG emissions (-9%). The conclusions provide insights into the potential of CWs as complementary measures for conventional wastewater treatment, particularly in reducing GHG emissions and improving water quality in cold temperate regions. These findings contribute to understanding sustainable wastewater management practices in environmentally sensitive areas.

Keywords:
Constructed Wetlands (CWs); greenhouse gas (GHG); biochar; cold temperate regions

1. Introduction

Addressing global climate change has emerged as one of the paramount issues and challenges within the international community [1,2]. While climate change exerts pressure on environmental resources and human development, it also presents new challenges for the wastewater treatment industry [3]. The carbon emissions from wastewater treatment plants constitute approximately 1.71% to 2.8% of the total anthropogenic carbon emissions in society [4]. Early estimates indicate that Methane (CH_4) and Nitrous oxide (N_2O) emissions from wastewater management account for 5% and

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3% of the total global CH_4 and N_2O emissions, respectively [5]. The vigorous promotion of Nature-Based Solutions (NBS) as a pivotal means to enhance water quality contributes significantly to the achievement of Sustainable Development Goal 6 set forth by the United Nations [6-8]. Among the exemplary NBS initiatives, constructed wetlands (CWs) are witnessing a sustained and rapid proliferation worldwide, emerging as crucial water pollution treatment technologies and pivotal types of wetland ecosystems [9].

CW technology offers several advantages, including effective water purification, low investment and operational costs, simplified management [10], and positive environmental aesthetics [11]. Its application in the advanced treatment of effluents from wastewater treatment plants not only addresses water quality pollution in receiving bodies but also helps alleviate issues such as ecological deficits and hydraulic deficiencies in river channels to some extent [12]. Therefore, the adoption of CW technology can further reduce pollutant loads in water bodies before low-pollution water enters, thus safeguarding water quality [13]. However, the efficacy of CWs in pollutant removal is significantly influenced by factors such as temperature [14]. Under lower temperature conditions, the efficiency of pollutant removal from wastewater by CWs diminishes notably [15]. This challenge poses a significant barrier to the widespread adoption and application of CWs in cold regions of northern China [16].

Amidst the continuous development and application of CW wastewater treatment technology, there is a growing concern not only about the effectiveness of pollutant removal but also about the environmental negative externality - GHG emissions [17]. Wetlands are a significant natural source of methane emissions, accounting for approximately 25% of total methane emissions [18]. Studies have indicated that the GHG emissions per unit area from CWs exceed those from natural wetlands by 2 to 10 times [19]. Therefore, while CWs serve their function in water quality purification, there is a risk of shifting "water pollution" to "air pollution," significantly reducing the overall ecological and environmental benefits of CWs [20].

Due to the continuously expanding area of CWs and their relatively high GHG emissions per unit area, enhancing the pollutant removal capacity of CWs, quantifying the GHG content within CWs along with their controlling factors, mitigating the potential for GHG emissions, and consequently providing a basis for regulating and optimizing the operation of CWs, are increasingly worthy of attention [21]. This study investigates the removal efficiency of major pollutants and the emission patterns of greenhouse gases in CWs, utilizing treated effluent from urban wastewater treatment plants. Additionally, this study explores the impact of biochar addition on these processes. The aim is to provide insights for controlling GHG emissions and enhancing pollutant removal efficiency during the advanced treatment of wastewater effluent in cold temperate zone CWs supplemented with carbon sources.

2. Methodology

2.1 Study Area

Chengde City (115°54′ E-119°15′ E, 40°11′ N-42°40′ N) is located in northeastern Hebei Province, China and is adjacent to Beijing City and Tianjin City to the south [22]. It features a temperate continental monsoon climate, characterized by distinct four seasons. The average annual temperature is 9.0°C, with the hottest month averaging 23.0°C in summer and the coldest month averaging -10°C in winter. During the winter months (December to February), the average temperature falls below 0°C. Chengde is recognized as one of the first batch of pilot cities for carbon peaking in China, as designated by the National Development and Reform Commission (NDRC), comprising a total of 25 cities and 10 regions.

The Chengde Urban Wastewater Treatment Plant is situated in the southeastern part of the Chengde municipal area. With a treatment capacity of 150,000 cubic meters per day, it employs the Anaerobic-Anoxic-Oxic (A^2O) process. The discharged wastewater complies with the Grade 1 A level treatment standard prescribed for municipal sewage treatment plants in China. Effluent samples are collected from the plant's discharge outlet and transported to the laboratory for indoor simulated experiments. The indoor experiments utilize Horizontal subsurface flow constructed wetlands (HFCW) simulation device. The water tank has a volume of 200 liters and is filled with zeolite as the substrate. The cultivation temperature is set at 15°C. Three commonly used species of CW plants are selected for the study: Phragmites australis, Scirpus validus, and Typha orientalis. Figure 1 shows the schematic of the experimental setup.

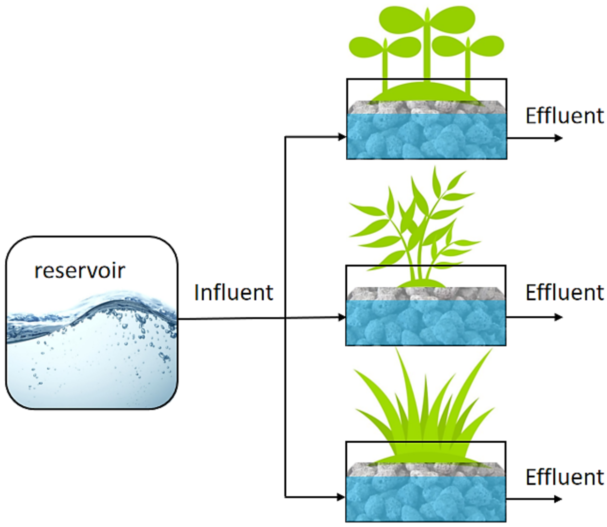

Fig. 1. The horizontal subsurface flow constructed wetlands (HFCWs) with different plant species

2.2 Sampling and Analysis

The experiment spans a period of 4 weeks, totaling 28 days, with sampling from the effluent conducted twice weekly. Samples of both water and gas are collected to determine the concentrations of pollutants such as chemical oxygen demand (COD) and total nitrogen (TN), as well as greenhouse gases including carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). The measurements of COD and TN follow the procedures outlined in Standard Methods [23]. Gas concentrations of CO_2, CH_4, and N_2O are determined using a static chamber coupled with gas chromatography (GC, Shimadzu, Japan). Gas fluxes are calculated based on a linear model derived from the temporal changes in gas concentrations within the static chamber over a 60-minute period.

The quantification of gas fluxes is achieved using the mathematical model as depicted in Eq. (1) [24].

$$J = \frac{dc}{dt} \cdot \frac{M}{V_0} \cdot \frac{P}{P_0} \frac{T_0}{T} \cdot H \tag{1}$$

In the equation, J represents the gas flux (in mg•m^{-2}•h^{-1} or μg•m^{-2}•h^{-1}), $\frac{dc}{dt}$ denotes the rate of change of gas concentration within the static chamber over time, M (in g/mol) signifies the molar mass of the gas, P (in Pa) indicates the pressure within the static chamber, T (in K) represents the temperature within the static chamber, H (in m) denotes the height of the static chamber above the water surface, V_0, P_0 and T_0 respectively denote the volume, pressure, and temperature of the gas under standard conditions.

The comprehensive impact of GHG emissions from different CW systems is assessed using the Global Warming Potential (GWP). Methane (CH_4) and nitrous oxide (N_2O) cumulative emissions are converted into CO_2 equivalents (CO_2-eq) for this purpose. Therefore, the formula for calculating the comprehensive GWP of each wetland is provided in Eq. (2) [25].

$$GWP = (34 \times CH_4) + (298 \times N_2O) \tag{2}$$

2.3 Biochar-Amended CWs

Conducting biochar substrate experiments with the selected optimal plant species among the aforementioned three. The substrate used in unamended CWs consisted of zeolite (with an approximate diameter of 2 cm). In contrast, the substrate employed in biochar-amended CWs comprised a mixture of zeolite and biochar (with an approximate diameter of 2-4 mm and a specific surface area of 800 m^2/g) at a ratio of 5:1 (V: V). The biochar was procured from Pingquan, China, derived from the pyrolysis of apricot shells.

3. Results
3.1 Pollutants Removal Performance
3.1.1 COD removal

The effluent COD concentrations of all treatment groups exhibited a decreasing trend with prolonged operation time (Figure 2), indicating a generally stable removal efficiency, with final removal rates of 35.71% for Phragmites australis, 40.28% for Scirpus validus, and 38.39% for Typha orientalis, respectively. Different plant species demonstrated varying degrees of COD removal effectiveness, with Scirpus validus exhibiting superior performance compared to the other two species.

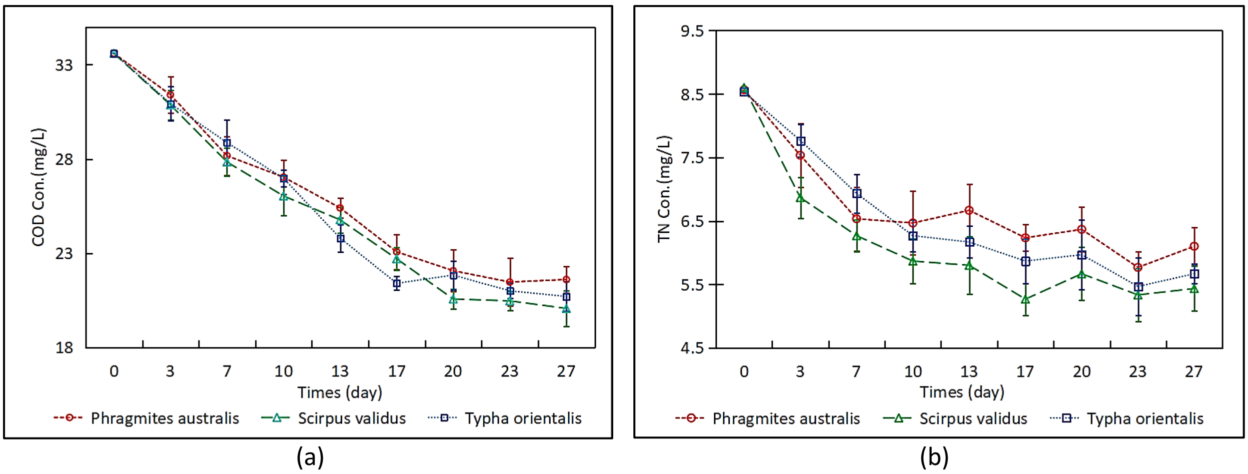

Fig. 2. (a) The effluent concentrations of COD (b) TN with increasing operation time

COD represents the concentration of organic pollutants in water, and in CW systems, various complex pathways contribute to organic matter removal. These pathways can be categorized into non-degradation and degradation pathways, involving the collective actions of plants, microorganisms, and substrates [26]. Among these, microorganisms play a predominant role in organic matter removal in CWs. Temperature is a crucial factor influencing microbial pollutant degradation; it not only affects the metabolic rate of microorganisms within wetland systems but also influences microbial population dynamics, community structure, and functionality, thereby impacting the efficiency of organic matter degradation [27].

However, compared to the COD removal efficiencies (> 80%) observed in other CWs [28], the COD removal efficiency in this experiment was relatively low. The experimental setup simulated the winter operation conditions of CWs, where under low temperature conditions, microbial communities that are usually effective in COD removal may become inactive [29]. Additionally, the decreased oxygen transport capacity of plant roots under low temperature conditions further affects COD removal effectiveness [30]. Moreover, the influent used in this experiment was treated effluent from a wastewater treatment plant, characterized by poor biodegradability and a low carbon-to-nitrogen ratio (< 6), which increases the difficulty of COD removal.

3.1.2 TN removal

In this study, all treatment groups demonstrated some degree of nitrogen removal effectiveness (Figure 2), with final TN removal rates of 28.79% for Phragmites australis, 36.82% for Scirpus validus, and 33.59% for Typha orientalis, respectively. Among the three plant species tested, Scirpus validus exhibited the most optimal removal effectiveness for TN. Typically, nitrogen removal in CWs occurs through two main pathways: absorption by plants and microbes, and biochemical transformation of nitrogen-containing compounds by microorganisms [31]. The impact of plants on nitrogen transformation and removal in CWs is primarily manifested in two ways: firstly, by providing attachment sites and oxygen for nitrifying bacteria through root systems to facilitate nitrification reactions; secondly, by providing carbon sources through root exudates to promote denitrification reactions by denitrifying bacteria [32].

However, compared to the TN removal rates (>40%) observed in certain CWs [33,34] the denitrification effectiveness in this experiment did not reach an optimal level. Firstly, as the influent used in the experiment was treated effluent from a wastewater treatment plant, characterized by poor biodegradability and a low carbon-to-nitrogen ratio (<6), which is generally considered insufficient for denitrification processes, this condition partially limited the denitrification process. Secondly, the experiment was conducted during winter under low temperature conditions, with an average temperature of 15°C. Studies suggest that nitrogen removal is inhibited when temperatures fall below 15°C, and the optimal temperature range for denitrification typically falls between 20°C and 40°C [29].

3.2 GHG Emissions

3.2.1 CO_2 emission

In the CW planted with Typha orientalis, the average CO_2 emission flux (176.56 mg/m^2/h) was higher than that in the wetlands planted with Phragmites australis (43.56 mg/m^2/h) and Scirpus validus (96.89 mg/m^2/h). It is speculated that plant respiration accounts for a significant proportion of the overall ecosystem respiration, thus resulting in differences in CO_2 emission fluxes among different plant species. Additionally, plants can enhance aerobic conditions by accumulating oxygen in the rhizosphere, thereby promoting aerobic degradation of organic matter and root respiration, consequently increasing the overall respiratory rate of the system [24].

3.2.2 CH_4 emission

The CH_4 emission fluxes vary among CWs with different plant species (Figure 3). In the CW planted with Phragmites australis, consistently positive CH_4 emission fluxes were observed. Conversely, in the CW planted with Typha orientalis, the CH_4 emission fluxes were positive at the beginning of the experiment, but subsequently transitioned to negative values, maintaining negative emission fluxes thereafter. Meanwhile, in the CW planted with Scirpus validus, CH_4 emission fluxes fluctuated around zero, with both positive and negative values observed. Regarding the average CH_4 emission flux, the CW planted with Typha orientalis (-22.67 μg/m^2/h) exhibited lower emissions compared to those planted with Phragmites australis (15.89 μg/m^2/h) and Scirpus validus (-3.89 μg/m^2/h). The amount of CH_4 emitted by all treatment groups accounted for only a small fraction of COD removal (< 0.4%).

The CH_4 emission flux in CWs results from the combined effects of CH_4 production, transport, and oxidation processes [35]. This study demonstrates that different plant species have a significant impact on CH_4 emission flux. Compared to other studies [36], the CH_4 emission fluxes observed in this research were relatively low. This phenomenon is speculated to be attributed to the thermodynamic process of denitrification, which occurs more readily than methane production. Denitrifying bacteria compete with methanogenic bacteria for organic substrates, thereby inhibiting CH_4 production [37]. Additionally, differences in plant root growth and development, root density, length, and microbial ecology among different plant species can also influence the biogeochemical processes in CWs [38].

It is noteworthy that the plants used in this study have rhizomatous root systems, which help them spread horizontally and establish colonies in aquatic habitats. Moreover, the roots are sufficiently long (~20 cm) to reach the bottom of the simulated setup. These two characteristics indicate that the plants possess a higher root oxygenation capacity, creating a more widespread aerobic environment in the CW [39]. Consequently, CH_4 and/or atmospheric CH_4 produced at the bottom of the setup are more susceptible to oxidation by methane-oxidizing bacteria, leading to the consumption of CH_4.It is hypothesized that the zeolite used in this study contains numerous pores, facilitating easier oxygen access and promoting the process of CH_4 oxidation [40]. The experiment demonstrates that Phragmites australis serves as an efficient conduit for gas transport (transferring CH_4 from sediment to the atmosphere) compared to Scirpus validus and Typha orientalis.

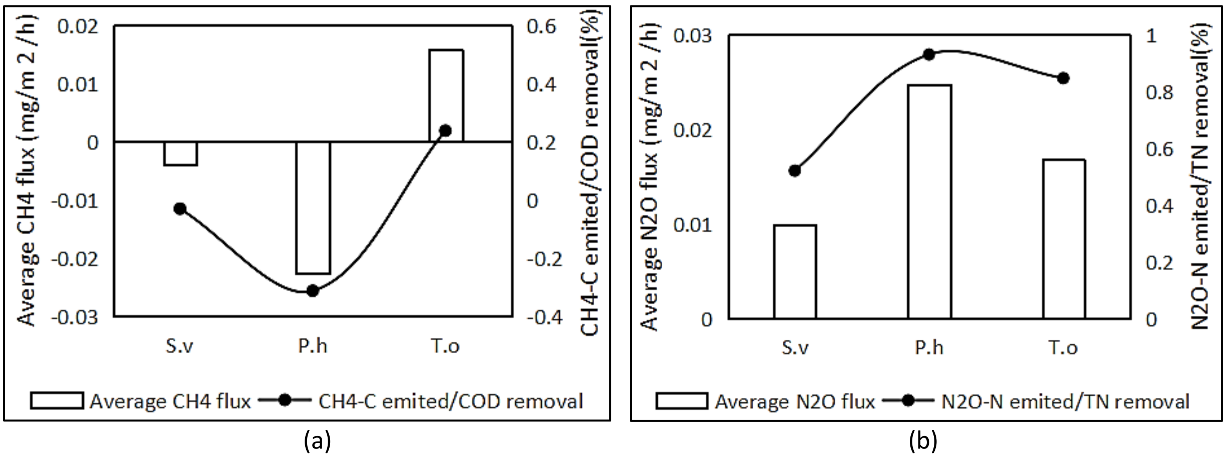

Fig. 3. (a) The average CH_4 flux and the CH_4-C emitted/COD removal (b) The average N_2O flux and the N_2O-N emitted/TN removal of CWs

3.2.3 N_2O emission

The emissions of N_2O varied among different plant-based CWs (Figure 3). Unlike CH_4 emission fluxes, which exhibited negative values, N_2O emission fluxes remained positive throughout the observation period. Regarding the average N_2O emission fluxes, the CW planted with Typha orientalis (24.78 μg/m^2/h) significantly exceeded those of wetlands planted with Phragmites australis (16.78 μg/m^2/h) and Scirpus validus (9.89 μg/m^2/h), with a difference of approximately 2.5 times. The proportion of N_2O emissions relative to TN removal was consistently less than 1%. During the nitrogen removal process in CWs, N_2O is generated as a byproduct of nitrification and an intermediate product of denitrification and plants play a crucial role in N_2O emissions from CWs [35].

Compared to the range of N_2O emission flux reported in previous studies [41-42], the average N_2O emission flux obtained in this study falls within the lower range. Denitrification processes primarily account for N_2O uptake [35]. The presence of certain plants, such as Typha orientalis and Phragmites australis used in this study, can absorb nitrogen from water, reducing its availability and thereby diminishing the substrates for denitrifying bacteria, leading to reduced N_2O emissions.

However, the average N_2O emission fluxes in the Typha orientalis treatment group were significantly higher than those in the Phragmites australis and Scirpus validus treatment groups. This disparity may be attributed to the abundant fine roots and high underground biomass of Typha orientalis, which can secrete a large amount of organic carbon from the roots. These organic carbons, largely unstable and readily degradable organic compounds, serve as substrates for denitrifying bacteria, resulting in increased N_2O emissions [43]. It is noteworthy that the average N_2O emission fluxes in the Scirpus validus treatment group were comparatively lower. The underlying reason for this difference might be the strong nitrogen uptake capability of Scirpus validus, which, coupled with sufficient carbon supply after efficient nitrification (complete denitrification), can significantly reduce N_2O emissions [44]. In summary, Scirpus validus may serve as a suitable plant species for reducing N_2O emissions in CWs.

3.2.4 GWP

When calculating the Global Warming Potential (GWP), only the emissions of CH_4 and N_2O were considered, while CO_2 emissions were excluded [45]. Given that the GWP of N_2O is 298 times that of CO_2, the Scirpus validus treatment group, with the lowest N_2O emission fluxes, also exhibited the lowest GWP, while the Typha orientalis treatment group had the highest GWP and CO_2 flux, as shown in Table 1.

Table 1
The average CO_2 fluxes and corresponding CO_2 equivalent (mean ± SD) in CWs

Plant species	CO_2flux (mg/m^2/h)	CO_2 (eq-CH_4) flux (μg/m^2/h)	CO_2 (eq-N_2O) flux (μg/m^2/h)	GWP (mg/m^2/h)
Scirpus validus	96.89±11.55	-3.89±6.15	9.89±2.42	2.815±0.638
Typha orientalis	176.56±10.47	-22.67±12.48	24.78±3.60	6.613±0.897
Phragmites australis	43.56±8.31	15.89±6.70	16.78±5.97	5.540±1.946

Overall, among the three selected plant species for CWs in this study, Scirpus validus was chosen for the subsequent construction of carbon-based CWs due to its efficient pollutant removal performance and low GHG emission characteristics.

3.3 Comparison of CWs and Biochar-Amended CWs

Biochar is a carbonaceous material produced by the pyrolysis of wood or other organic materials under limited oxygen conditions [46]. Due to its low cost and effective adsorption of pollutants, biochar has been widely applied in soil improvement, soil remediation, carbon sequestration, and wastewater treatment [47]. This part of study aims to elucidate the mechanisms by which biochar-amended CWs control GHG emissions, providing new insights for the design of CWs. The experimental setup utilized in this study was based on the CW system in section 3.1, with Scirpus validus selected as the experimental plant. Two treatment groups were established: one without biochar addition (a) and one with biochar addition (b). A detailed comparison of the treatment effects is presented in Table 2.

Table 2
Comparison of treatment effects of CWs without (a) and with (b) biochar addition

Type	COD (mg/L)	TN (mg/L)	CO_2flux ($mg/m^2/h$)	CO_2 (eq-CH_4) flux (μg/m^2/h)	CO_2 (eq-N_2O) flux (μg/m^2/h)	GWP ($mg/m^2/h$)
a	20.07±0.95	5.43±0.35	96.89±11.55	-3.89±6.15	9.89±2.42	2.815±0.638
b	16.71±0.05	3.53±0.14	91.22±10.16	4.14±1.78	8.11±1.54	2.558±0.432

3.3.1 Pollutants removal performance

The addition of biochar in CWs significantly enhances the removal efficiency of pollutants. The promotion effect of biochar addition on the removal rates of various pollutants are as follows: COD (9.99%) and TN (22.13%). This study found that the COD removal rate in the biochar-amended CW was higher than that in the non-biochar-amended CW. The increase in COD removal rate due to biochar addition can be attributed to the abundant π bonds on the surface of biochar. These π bonds facilitate the easy adsorption of organic molecules onto biochar through electrostatic attraction and hydrogen bonding, leading to a high removal rate of organic substances [48]. Furthermore, the porous structure and large surface area of biochar enable both direct adsorption of COD and provision of ample space for microbial growth, thereby promoting microbial activity and abundance, and subsequently enhancing COD removal [49]. Previous studies have demonstrated the significant role of biochar adsorption in the COD removal process [50,51].

In this study, the effluent TN concentrations in the biochar-amended CWs were significantly lower than those in the non-biochar-amended wetlands, indicating that biochar can serve as a potential readily degradable carbon source. These carbon sources are either encapsulated within the porous structure of biochar or released directly from biochar through bacterial chemical metabolism, thereby promoting efficient removal of nitrogen oxides [52]. Additionally, the high porosity and large surface area of biochar facilitate the formation of denitrifying bacterial biofilms, increasing the quantity and activity of denitrifying bacteria, and thereby promoting denitrification, leading to high TN removal rates in biochar-amended CWs [53]. In summary, the addition of biochar significantly improves the pollutant removal efficiency of CWs, making it an effective enhancement technique for conventional pollutant removal in CWs.

3.3.2 GHG emissions

The average CO_2 emission flux was higher in the non-biochar-amended CW (96.89 $mg/m^2/h$) compared to the biochar-amended one (91.22 $mg/m^2/h$), resulting in 5.67 $mg/m^2/h$ reduction in the average CO_2 emission flux. Overall, the impact of biochar addition on CO_2 emission flux in the CW systems was minor. The biochar-amended CW consistently exhibited positive CH_4 emission flux, with an average CH_4 emission flux of 4.14 μg/m^2/h, which was higher than that of the non-biochar-amended CW (-3.89 μg/m^2/h). Regarding the proportion of average CH_4 emission flux to COD removal, the biochar-amended CW (0.044%) surpassed the non-biochar-amended wetland (0.031%). This study revealed a significant increase in CH_4 emission flux with the addition of biochar. Biochar, acting as a conductive material, facilitates interspecies electron transfer between methanogenic and acidogenic bacteria, thereby promoting CH_4 generation. Additionally, biochar provides unstable organic carbon for methanogens, further enhancing CH_4 production [54].

Regarding N_2O emission flux, the average N_2O emission flux and the proportion of emitted N_2O-N to TN removal were higher in the non-biochar-amended CW (9.89 μg/m^2/h and 0.521%, respectively) compared to the biochar-amended wetland (8.11 μg/m^2/h and 0.398%, respectively), resulting in an 18% reduction in the average N_2O emission flux with biochar addition. This study demonstrates that biochar addition significantly reduces N_2O emission flux. The adsorption capability of biochar reduces the effectiveness of nitrogen oxides in water, leading to a decrease in N_2O emission rates. Moreover, biochar with its high surface area can directly absorb N_2O generated by the system, thereby reducing N_2O emissions [55]. In addition, the addition of biochar in CW may inhibit the denitrification rate of the system by increasing the porosity of the substrate, thereby promoting the diffusion and transfer of oxygen. Denitrification rate is considered to be the primary process in CWs that leads to N_2O production [42].

3.3.3 GWP

The GWP of the two treatment groups was as follows: for the biochar-amended CW, it was 2.558 mg/m^2/h, whereas for the non-biochar-amended wetland, it was 2.815 mg/m^2/h. This indicates that the addition of biochar resulted in a 9% reduction in GWP values. Overall, the addition of biochar in CWs significantly decreased the GWP values. This implies that incorporating biochar in CWs can reduce GHG emissions, thereby contributing to mitigating climate change.

4. Conclusions

This study investigated the removal efficiency of typical pollutants and GHG emissions from designed CWs under low-temperature conditions (15°C). Under the experimental conditions, CWs demonstrated certain removal efficiencies for COD and TN, albeit less effective compared to other studies. The CO_2 emission flux and GWP were also within relatively low ranges compared to other research findings. This suggests that the cold winter environment and low carbon-to-nitrogen ratio of experimental sewage partially constrained both the water purification efficacy and GHG emissions of CWs. Based on the experimental results on three plant species, Scirpus validus should be prioritized for future scientific research and practical applications due to its efficient pollutant removal performance and low greenhouse gas emission characteristics.

The addition of biochar improved the removal efficiency of pollutants COD and TN in CWs. However, the effect on different GHG emissions was heterogeneous: while there was no significant impact on CO_2 emissions, it reduced N_2O emission flux while concurrently promoting CH_4 emissions. Although the decrease in N_2O flux was offset by the increase in CH_4 flux, biochar modification lowered the GWP of CWs. Amending CWs with biochar can thus be a priority strategy for designers to enhance pollutant removal and reduce GHG emissions. Given that this study was based on indoor simulation experiments, it is recommended for future research to conduct field assessments to further validate the conclusions drawn from this experiment.

Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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