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Performance optimization of novel multi-unit green wall system for blackwater treatment and reuse on-site

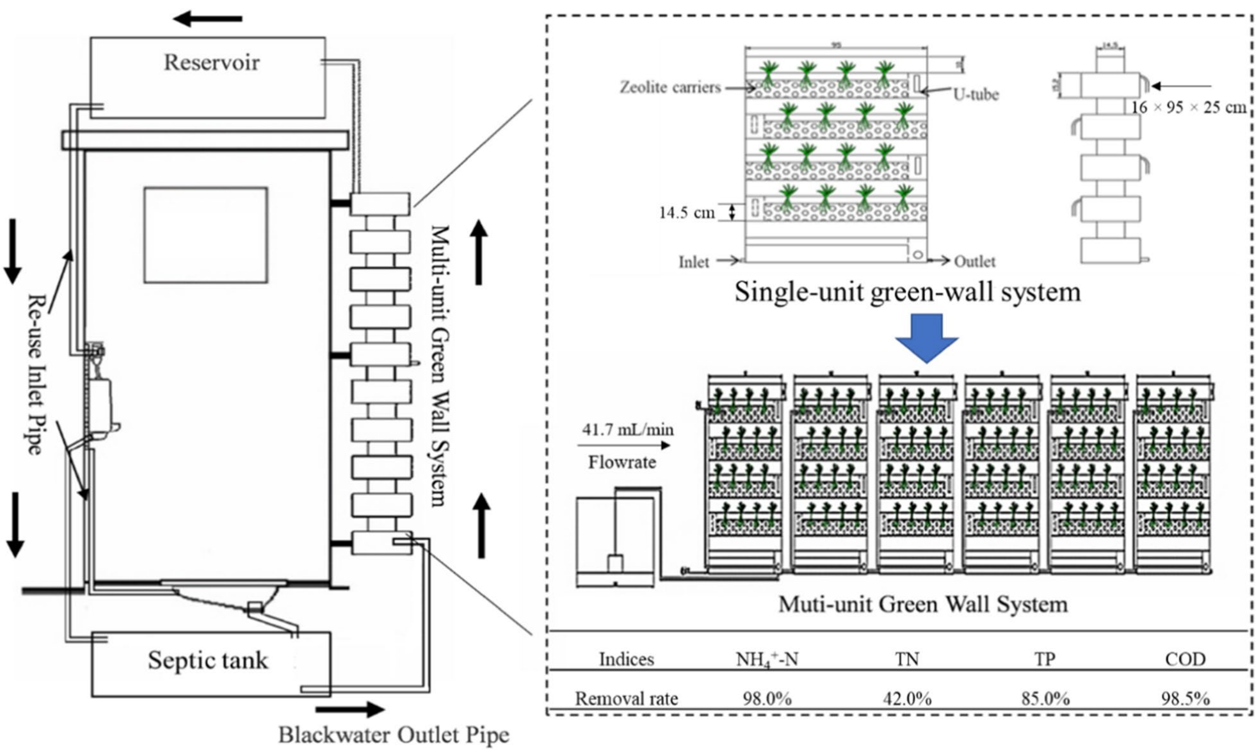
Zhiquan Wang[1,2,3], Weijie Xie[1,2,3], Fan Shi[1,2,3], Chunzhen Fan[1,2,3], Suqing Wu[1,2,3], Shengbing He[4], Hainan Kong[4], Min Zhao[1,2,3] and Xiangyong Zheng[1,2,3*]

Abstract
Blackwater constitutes a primary component of environmental pollution posing serious risk to human health; however, reusing the nutrients found in blackwater diminishes the associated pollution and promotes resource recycling. Conventional green-wall systems are not suitable for in situ treatment of high-concentration wastewater. Thereby, a novel multi-unit green-wall system with six independent treatment units was designed to achieve a cost-effective and eco-friendly in situ treatment of blackwater with high organic load. Zeolite carriers were selected for the matrix with carrier depth of 14.5 cm, ivy and chlorophytum were selected as the greening plants. Various pollutants were rapidly reduced at the initial stage and the concentration of the pollutants decreased as the number of treatment units increased. Overall, the green-wall system was more effective in removing COD and NH_4^+–N with the removal rates of 98.5% and 98%, which may be due to the good buffering capacity of the media and the roles of *Firmicutes* and *Bacteroidetes* in COD degradation and ammoniated nitrification reactions. In addition, the maximum TP and TN removal rates were observed with a value of 85% and 42%, respectively. Consequently, the novel multi-unit green-wall system is an effective method for in situ blackwater reuse.

Keywords Blackwater reuse, High organic load, Multi-unit green wall, Removal efficiency, Dominant species

*Correspondence:
Xiangyong Zheng
x.zheng@wzu.edu.cn
Full list of author information is available at the end of the article

Graphical Abstract



Introduction

Blackwater is concentrated waste stream composed mainly of feces, urine, and flushed toilet water; it is generally harmful to human health and poses great contamination risks [1–3]. Nevertheless, it is also considered as the domestic wastewater fraction with the most organic carbon and nitrogen because even though it constitutes only 30% of the total municipal wastewater volume, it includes 90%, 50%, and 80% of the total nitrogen (N), chemical oxygen demand (COD), and phosphorus (P), respectively [4]. In other words, blackwater has great reuse potential owing to its high nutrient uptake by plants. Currently, blackwater is treated along with municipal wastewater in conventional municipal wastewater treatment systems, with nutrients being discharged into water bodies and eventually lost [5]. The segregation and treatment of blackwater, regardless of their environmental and health-related benefits, are generally considered as additional expenses because of the required areas, construction costs, operation, and maintenance [6, 7]. The large-scale pipe network, high construction costs, complex implementation and operation, and low recovery of water caused by the centralized sewage treatment systems lead to the unfriendly utilization potential for agricultural production and reuse of blackwater [8]. Thereby, the recycling of nutrient in blackwater may be crucial in satisfying the needs of energy-saving, carbon emission reduction and sustainable development, especial for the developing countries [2, 9].

In recent years, blackwater treatment technology has been a gradual trend towards resource recovery, while chemical and physical technologies have been developed to enhance non-hazardous use and improve resource recovery efficiency [2]. Reusing the key nutrients (i.e., N and P) can not only reduce the eutrophication and potential pollution, but also realize the recycling of resources, since the reused nutrients constitute a sustainable fertilizer source [10–12]. Based on this demand, source separation and nutrient recovery of blackwater on-site have been developed as the necessary modes for avoiding long-distance collection and improving the efficiency of nutrient recovery [2, 13]. Generally, methods for energy and nutrient recoveries from blackwater mainly involve the decentralized wastewater collection and reuse on-site [10, 14]. Currently, various technologies have been used for in situ blackwater treatment, including wet composting, membrane bioreactors, anaerobic baffled reactors, and constructed wetlands [15–17]. Compared to biological treatment processes, constructed wetlands, which constitute an eco-treatment technology, have lower energy requirements, positive landscape effects, easy operation and maintenance, and significant potential regarding nutrient reduction [13, 18, 19]. Green walls have become an ideal solution that have the ability to perform the functions of constructed wetlands in wastewater treatment without occupying space due to their vertical structure, because the biological transformation and plant uptake play major roles in contaminant removal [20, 21]. Green walls have increasingly been employed in the treatment of low-concentration greywater [18, 19, 22], but it was few applied in the treatment of blackwater which may be due to the negative pressure of high organic load on plant growth [6, 22]. Therefore, the design of novel green walls with higher organic loads and stronger adaptability is essential for the reusing of high-concentration blackwater.

This study introduces a novel multi-unit green-wall system which trying to further enhance the organic load and adaptability regarding high-concentration blackwater. The novel multi-unit green-wall system has been explored as a viable blackwater treatment system with a water-cycle treatment process which was reliable, stable, thereby efficient in reusing N and P of blackwater. Throughout the process, we closely monitored the dominant species contributing to the removal of various pollutants in this novel multi-unit green-wall system.

Materials and methods

Blackwater characterizations

Blackwater was pre-treated with a septic tank and biological trickling filter to maintain a stable level before the 9:00 am collection from the student dormitory of Wenzhou University. The whole sampling cycle lasted for nearly 3 months. The blackwater characteristics are presented in Table 1.

Multi-unit green-wall system

The multi-unit green-wall system employed in this study is shown in Fig. 1(c); it consisted of six independent treatment units, which were made of five cuboids, with unit sizes of 16×95×25 cm, as shown in Fig. 1(a). A U-tube with a diameter of 1.5 cm and height of 13 cm was fixed on the right side of each cuboid. Six treatment units were all composed of four zeolite/planted containers and connected in series, blackwater was delivered from bottom to top by submersible pumps at a flowrate of 41.7 mL/min and further flowed through each cuboid from top to bottom through the U-tube, the special design will contribute to enhance the buffer capacity of multi-unit green-wall system for the blackwater with high organic loads. We also implemented an upstream equalization tank, as shown in Fig. 1(b), to enhance the adaptability of the system to sudden water-quality changes. Moreover, zeolite carriers (Table 2) were selected for the matrix because of their excellent adsorption effects in relation to N and P in wastewater. The zeolite diameters ranged between 4 and 8 mm, and the carrier depth was 14.5 cm. Ivy (*Hedera nepalensis var.sinensis*) and chlorophytum (*Chlorophytum comosum 'Variegatum'*) were selected as the greening plants due to their low requirements for light and strong vitality. Ivy and chlorophytum were evenly distributed, and four plants were planted at equal 14.5 cm intervals in each unit layer to ensure sufficient growth space. Finally, wastewater sequentially flowed through the multi-unit system for further blackwater treatment. Besides, a septic tank, biological trickling filter and submersible pumps were added at the initial end of the system to provide a stable blackwater characteristics and inlet flowrate.

Table 1 Blackwater characteristics

Indices	pH	NH_4^+–N/(mg/L)	TN/(mg/L)	TP/(mg/L)	COD/(mg/L)
Blackwater	7.42–7.95	21.92–23.72	27.53–33.01	2.26–2.93	198–205

Operation modes

Determination of the optimal idle/reaction time and hydraulic retention time (HRT)

Before treating high-concentration blackwater, the green-wall system requires stable operation with low-concentration blackwater for a month to obtain a sufficient carrier biofilm. A timer switch controlled the idle/reaction time. We analyzed various indices of raw water and effluent, including COD, total nitrogen (TN), ammonia (NH_4^+–N), nitrate (NO_3^-–N), nitrite (NO_2^-–N), and total phosphorus (TP). The optimal idle/reaction time and HRT of blackwater in the single-unit green-wall systems were evaluated based on the removal efficiency in different operation modes (Table 3**),** different modes of continuous and intermittent inlet were considered with increasing hold time and HRT in order to obtain the optimal operation mode. The maximum hold reaction time and HRT of the intermittent inlet was 300 min and 6 days, respectively. In particular, we opted for a sixfold dilution of the original blackwater for the determination of the optimal idle/reaction time and HRT. Subsequently, we evaluated the removal efficiency of the original high-concentration blackwater at the optimal idle/reaction time and HRT.

Operation of the multi-unit green-wall system

Then, the six single-unit green-wall systems were further connected in series and operated at the optimal idle/reaction time and HRT for the treatment of original high-concentration blackwater. We combined six treatment

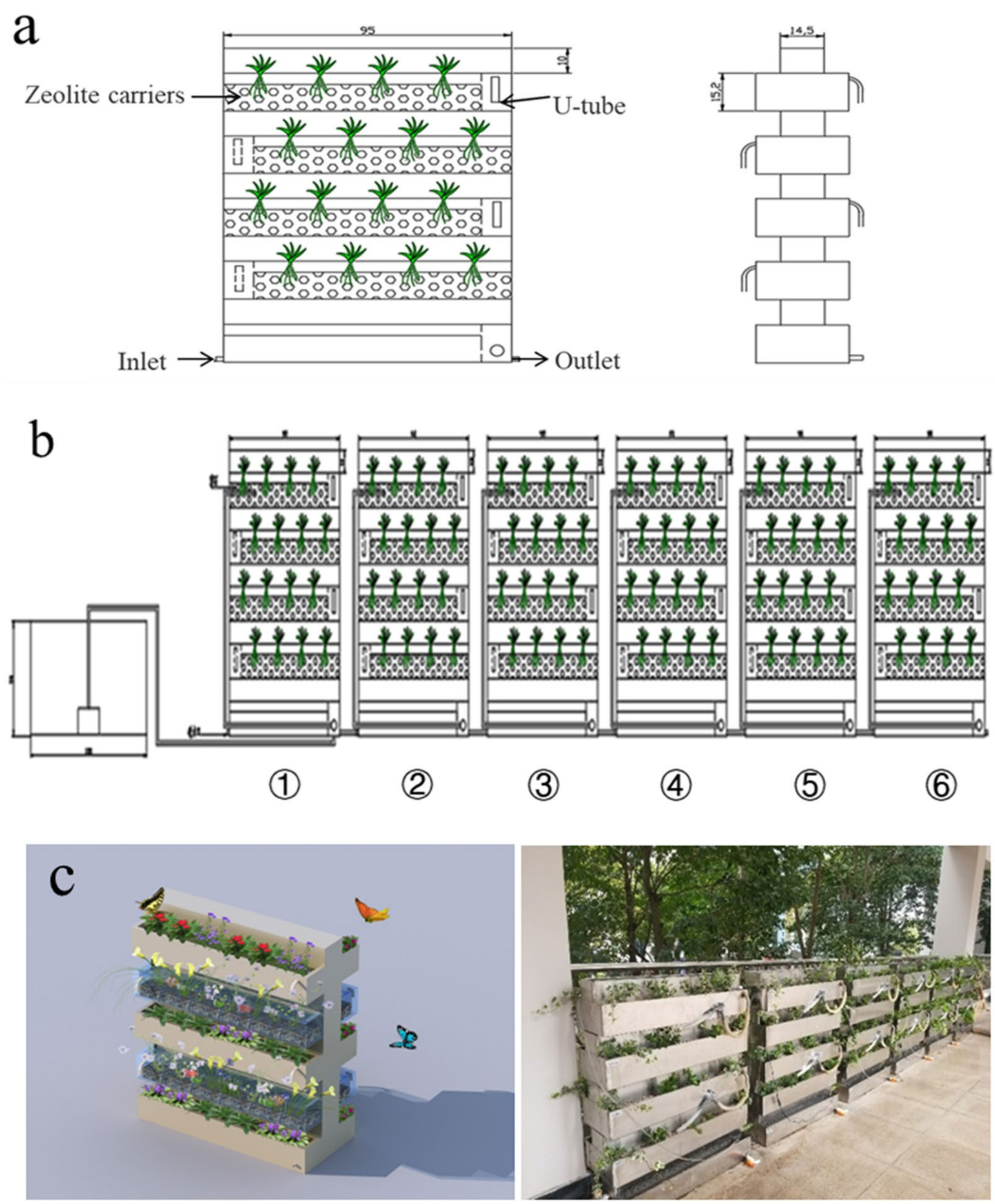


Fig. 1 Green-wall system with zeolite carriers and greening plants. **a** Single-unit green-wall system; **b** multi-unit green-wall system; **c** conceptual design and the actual system

units and analyzed water samples collected between May and December 2019, one month after the start-up stage. We also analyzed biological samples from the zeolite surfaces of the reactors; prior to that, threefold diluted blackwater as well as the original blackwater were sequentially added to allow the gradual adaptation of the carrier biofilm to the high-concentration blackwater. A timer switch controlled the onset of pumping to ensure

Table 2 Chemical composition of the zeolite carriers

Component	SiO_2	Al_2O_3	Fe_2O_3	K_2O	Na_2O	CaO	MgO	MnO	TiO_2
%	70.45	12.35	0.59	1.14	1.09	1.66	1.68	0.05	0.19

Table 3 Different operation modes

Units	Operation modes		Idle/reaction time	HRT (d)
U-1	Continuous inlet	Hold 0 min	0:24	1
U-2	Inlet 60 min	Hold 12 min	1:5	2
U-3		Hold 30 min	1:2	3
U-4		Hold 60 min	1:1	4
U-5		Hold 120 min	2:1	5
U-6		Hold 300 min	5:1	6

a fixed water intake time. The effect of the multi-unit green-wall system on the original blackwater was evaluated by measuring various effluent indices.

Analytical methods

COD, TN, NH_4^+–N, NO_3^-–N, and TP concentrations were measured in triplicates, following [1]; DNA extraction, PCR, sequencing, and data analysis were conducted following [23]. The Freundlich model was used to analyze the adsorption effect because it fits the zeolite well [24], while different kinetic models were used to analyze the reaction process, as shown in Table 4.

Results and discussion

Optimal idle/reaction time and HRT of the single-unit green-wall system

Regarding the treatment of the sixfold diluted blackwater, the removal rates of various pollutants at different idle/reaction times are shown in Fig. 2(a). The highest removal rates, suggesting the best treatment performance, were observed in the U-5 mode. The COD removal rate of U-5 at different idle/reaction times exceeded 90%, with the maximum value of 94%. A similar result was obtained for the NH_4^+–N removal, with the maximum removal rate of 93%. Moreover, the TN removal rate was lower than that of NH_4^+–N, with the maximum removal rate of 60%. However, the TP removal rate first decreased and then increased until U-4, no obvious fluctuation and a high removal rate in excess of 80% was maintained with the increase in idle/reaction time. Notably, rapid and high removal efficiency was obtained immediately during the

Table 4 Reaction kinetic equations of several different orders

Orders	Kinetic equations: Differential form	Kinetic equations: Integral form	Units	Half-life $t_{1/2}$
0	$-\frac{dC}{dt} = k_0$	$C_0 - C = k_0 t$	mol*m^{-3}*s^{-1}	$\frac{C_0}{2k_0}$
1	$-\frac{dC}{dt} = k_1 C$	$ln\frac{C_0}{C} = k_1 t$	s^{-1}	$\frac{0.693}{k_1}$
2	$-\frac{dC}{dt} = k_2 C^2$	$\frac{1}{C} - \frac{1}{C_0} = k_2 t$	(mol*s)$^{-1}$	$\frac{1}{k_2 C_0}$

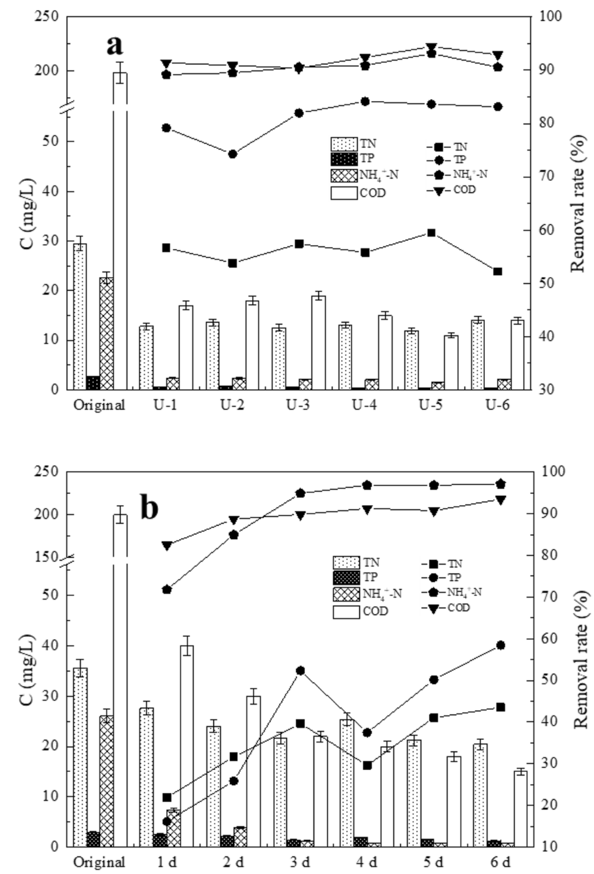


Fig. 2 Removal effects of blackwater for **a** different idle/reaction times and **b** HRT

start-up stage, typically due to the adsorption immobilization of zeolite carriers and biomass cells for various pollutants [25–27]. Furthermore, no obvious desorption fluctuation was found with an increase in idle/reaction time, indicating that follow-up biodegradation also played an important role in the removal of various pollutants.

The changes in the concentrations of various pollutants at different HRT are shown in Fig. 2(b). Similar results were observed for the COD and NH_4^+–N removals, both removal rates increased gradually until day 3 and remained at 90% and 96%, respectively. The increase in HRT did not inhibit the continuous and efficient removal process, in accordance with previous reports [7, 28], suggesting that increased HRT results in more efficient COD and NH_4^+–N removals. For the TN and TP removals, the removal rates increased from 22 to 40% and 16% to 55%, respectively, until day 3. Some fluctuations occurred after day 3, when HRT was increased, and the maximum TN and TP removal rates occurred on day 6 with values of 44% and 58%, respectively, which was slightly higher than those on day 3. Overall, the results indicated the positive correlation between removal rates and HRT, the optimal HRT should be 3 days because of the higher efficiency and lower operation time.

Treatment of the original blackwater with optimal idle/reaction time and HRT

To improve the adaptability of the green-wall system to the original blackwater, the concentration of blackwater was gradually increased. The optimal treatment conditions for low-concentration (i.e., six fold diluted) blackwater were observed in U-5 (idle/reaction time: inlet 60 min and hold 120 min) with an HRT of three days. We also considered the treatment effects for higher-concentration blackwater, i.e., the threefold diluted blackwater and original blackwater. Figure 3 presents the indicator changes of various pollutions, it is clear that the CODs of the threefold diluted blackwater and original blackwater both decreased significantly with removal rates of 83.4% and 94.5%, respectively. A higher removal rate was observed for the original blackwater with removal rates of 98.5%, primarily due to the better adsorption performance of the zeolite carriers. Similar results were obtained for the TN, NH_4^+–N, and TP removals, with the removal rates for the original blackwater being much higher than those for the threefold diluted blackwater. Consequently, the higher COD did not inhibit the decomposition of organics, while either the nitrification or denitrification suffocation processes can cause low TN removal efficiency [29]. Plant-based wetland systems, where processes such as solid sedimentation, organic matter decomposition, filtration by media, and uptake by plant roots occur, effectively provide the function of effluent treatment [30].

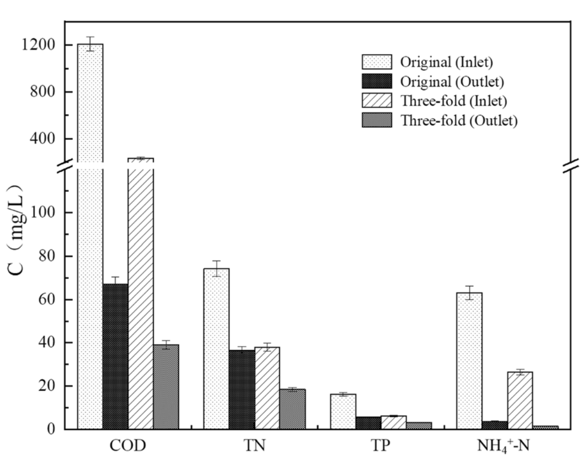


Fig. 3 Indicator changes for higher concentration blackwater in optimal idle/reaction time and HRT

Kinetics process of the single-unit green-wall system on blackwater treatment

The single-unit green-wall system had a better removal effect on the original blackwater, especially for COD and NH_4^+–N. Similar results were also obtained by Cheng et al. [31] in the treatment of secondary effluents of municipal domestic wastewater, with high COD and NH_4^+–N removal efficiencies of 92.6% and 96.1%, respectively. Thus, we further analyzed the dynamic fitting of different indices at the start-up stage to comprehensively analyze the possible reaction process. As shown in Fig. 4, the Freundlich model was used to define the biosorption of an absorbate on a heterogeneous surface of a biosorbent [32], with higher R^2 values indicating higher correlations with the adsorption effect. The significantly positive results in Fig. 4(a) suggest that the rapid and high removal efficiency at the start-up stage was primarily due to the large adsorption capacity of zeolite carriers and high original concentrations of pollutants in the single-unit green-wall system, particularly for COD and NH_4^+–N. The COD and NH_4^+–N concentrations were significantly reduced from 1200 to 18 mg/L and 78.5 to 1.53 mg/L within 20 h, with significant removal rates of 98.5% and 98.0%, respectively. The high removal rates indicated the powerful adsorption of zeolites carriers for COD and NH_4^+–N; however, the TN concentration decreased from 94.0 to 56.2 mg/L, with a removal rate of only 40.2%, while the NO_3^-–N concentration markedly increased from 1.08 to 52.10 mg/L, thereby revealing that the TN removal not only involves the adsorption effect, but also the conversion of NH_4^+–N and NO_3^-–N based on nitrification and denitrification [33, 34]. The TP concentration eventually decreased from 12.40 to 1.85 mg/L, with a removal rate of 85%, also mainly due to adsorption by the zeolites at the start-up stage. After some time, no obvious decrease was further observed in the later stage,this may be due to the lower original TP concentration compared to those of the various other pollutants.

The removal dynamics of COD and NH_4^+–N were further analyzed using different reaction models suggested by Attri et al. [35] because of their excellent removal effects. Figure 4b–d shows the fitting results, including the zero-order, first-order, and second-order reactions. The largest R^2 values of 0.877 and 0.943 were obtained by fitting the second-order dynamic reaction model for NH_4^+–N and COD, respectively. Therefore, the dynamic processes of COD and NH_4^+–N in the single-unit green-wall system can be described well using the second-order reaction model. Based on the second-order reaction model, the degradation half-lives of NH_4^+–N ($t_{1/2\text{-}NH4^+\text{–}N}$) and COD ($t_{1/2\text{-}COD}$) were 3.75 and 0.15 h, respectively (Table 4 and Fig. 4d); this is consistent with the higher R^2 of the Freundlich fitting and indicates the

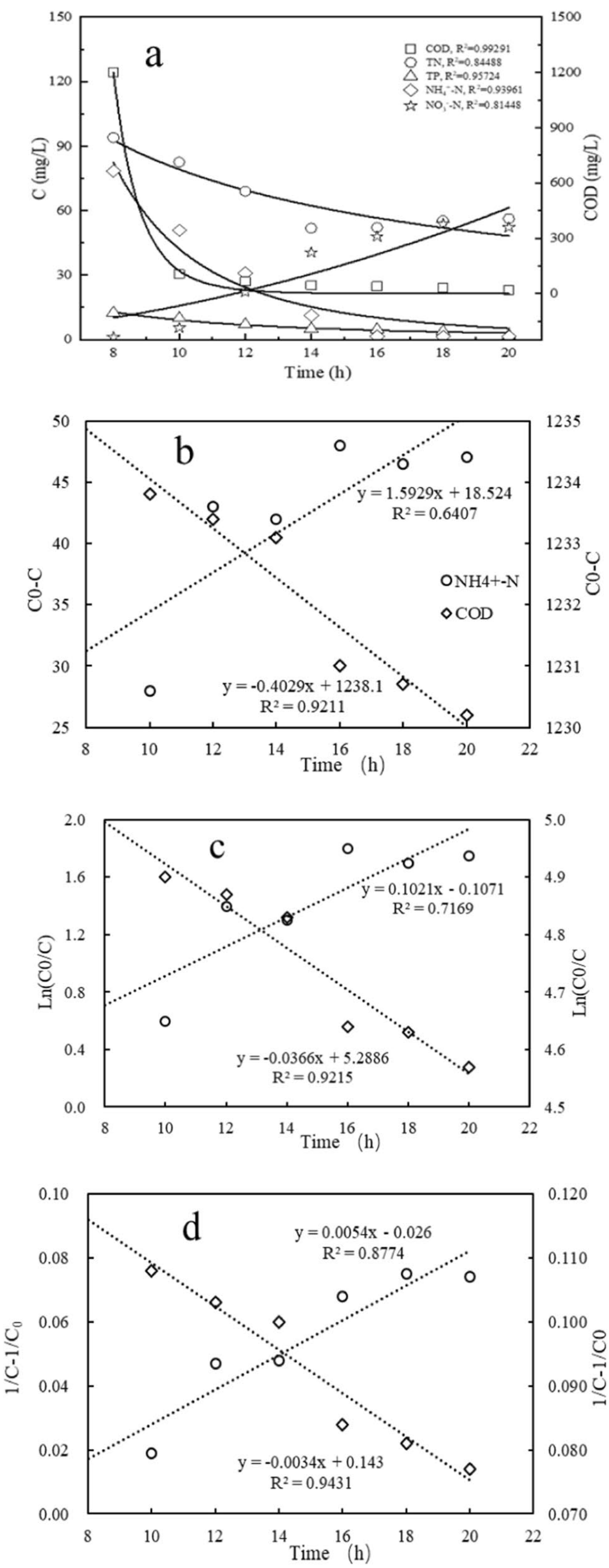


Fig. 4 Reaction kinetics model fitting. **a** Freundlich fitting of various pollutants at the start-up stage; **b** zero-order reactions of NH_4^+–N and COD; **c** first-order reactions of NH_4^+–N and COD; **d** second-order reactions of NH_4^+–N and COD

high load acceptance of the single-unit green-wall system for blackwater with a higher organic load.

Blackwater treatment in the multi-unit green-wall system

At the start-up stage, the single-unit green-wall system showed a powerful adsorption performance for various pollutants, which led to the high blackwater removal efficiency. However, continuous biodegradation is more significant for the eventual removal of adsorbed pollutants [36], with the extent of degradation of adsorbed pollutants depending on the reversibility of adsorption and the potential of the biofilm to biodegrade pollutants [37]. Thus, to further improve the adaptability of the green-wall system to sudden wastewater changes with high organic load, a multi-unit green-wall system was considered for the continuous degradation of adsorbed pollutants in the original blackwater.

We considered a multi-unit green-wall system combining six treatment units to evaluate the continuous degradation and reuse performance of nutrients that were present in the original blackwater. The plant growth change in the multi-unit green-wall system after treatment with the original blackwater is shown in Additionl file 1: Fig. S1, the plants grew very strong with a well-developed root system, indicating that a large amount of nutrients in the blackwater had been used effectively and highlighting the significant treatment potential in nutrient reduction. The changes in various pollutants in the original blackwater after the multi-unit green-wall system are shown in Fig. 5. First, the COD change with increasing the number of system units is shown in Fig. 5a, though the influent COD fluctuated greatly, the effluent COD was stable at 50 mg/L, indicating the good buffer capacity of the multi-unit green-wall system. Notably, the COD removal rates were improved greatly in the same running time with increasing number of system units, the removal efficiency was maintained at the highest level (i.e., above 92%) when more than three system units were combined and no obvious fluctuation was observed with the increase in HRT. Piai et al. [36] distinguished adsorption from biodegradation in an activated carbon biofilm system, where biodegradation is the eventual pathway for removing pollutants from carrier materials. Thus, the constant high removal rate suggests continuous degradation of adsorbed pollutants in the

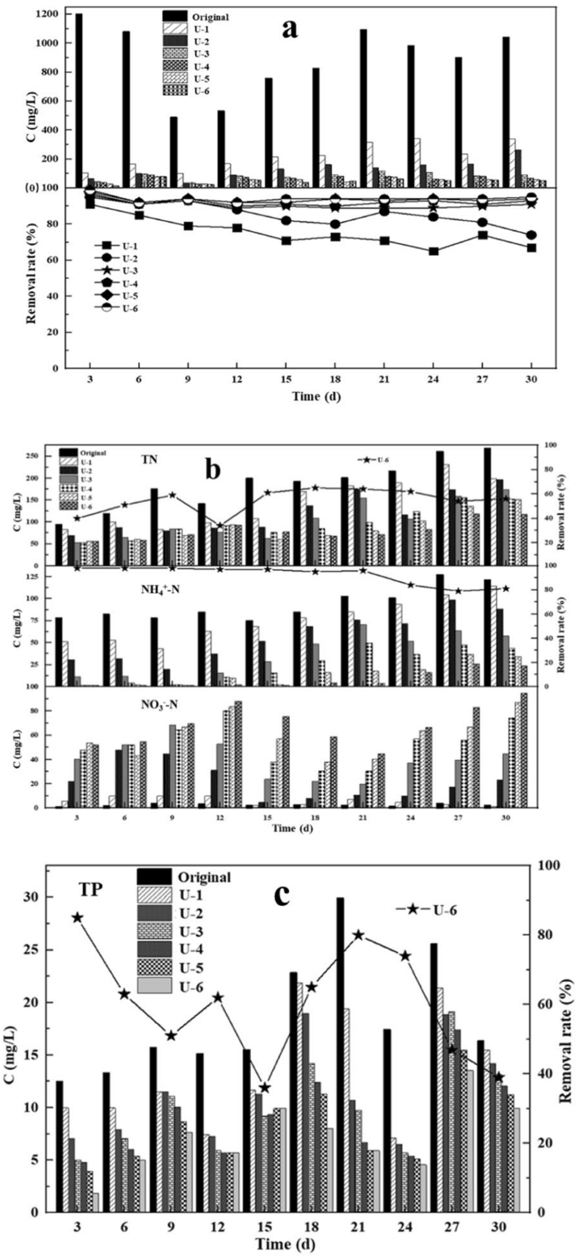


Fig. 5 Changes of pollutant indices for the original blackwater over time after the multi-unit green-wall system. **a** COD; **b** TN, NH_4^+–N and NO_3^-–N; **c** TP

multi-unit green-wall system. Importantly, there was a positive relationship between removal effects and treatment units, highlighting that the increasing number of treatment units not only improved the COD removal efficiency, but also promoted the continuous degradation of adsorbed pollutants.

Figure 5b shows the changes in N-containing organic matter, including TN, NH_4^+–N, and NO_3^-–N. During the observational period, students returned to school, resulting in a gradually increase of average TN and NH_4^+–N concentrations. However, the TN and NH_4^+–N concentrations in the effluent flowing through the multi-unit green-wall system decreased continuously with increasing number of treatment units, with average removal rates of 60% and 98%, respectively. In contrast, the NO_3^-–N concentration increased significantly, following the opposite trend to that of NH_4^+–N and indicating the strong nitrification–reaction performance of the system [33, 38]. Overall, the concentration of N-containing compounds decreased with increasing number of treatment units. The microbial community might be the main contributor to the TN removal, with the high organic content of the influent blackwater further enhancing the bacterial activity, as shown in the literature [19, 39].

As shown in Fig. 5c, the TP concentration in the overall trend fluctuated considerably over time. An obvious decrease was observed in the first 15 days; a subsequent significant increase occurred from day 15 to day 21, in accordance with Fig. 5a and the return of the students, indicating that the system has a poor buffering capacity for TP. The maximum TP removal rate was observed on day 3, with a value of 85%; after day 3, the removal efficiency was lowered mainly due to the adsorption saturation at the start-up stage and the subsequent desorption [40].

Overall, increasing the number of treatment units improved the removal efficiencies of various pollutants, especially those of COD and NH_4^+–N, and maintained them at high levels. No obvious fluctuations were observed in the removal efficiencies except for that of TP, suggesting a good buffer capacity and continuous degradation performance of adsorbed pollutants.

Microbial distribution for the blackwater in the multi-unit green-wall system

We further analyzed the main microbial components of the zeolite surfaces in the multi-unit green-wall system. We conducted principal component analysis (PCA) to analyze the relationships between the relative abundances of the predominant bacteria and environmental variables, as shown in Fig. 6a. In the figure, G represents the non-flooded area, S represents the flooded area, and 1–4 represent the cuboids from top to bottom, as shown in Fig. 1a. The accumulated percentage difference between PC 1 and PC 2 was 79.6%, with less variability in the structure of the zeolite microbial community in the non-flooded area and more similar community structures in layers 3 and 4. In addition, the microbial community structures in the flooded areas showed fewer differences between layers 1 and 2 than those in the non-flooded

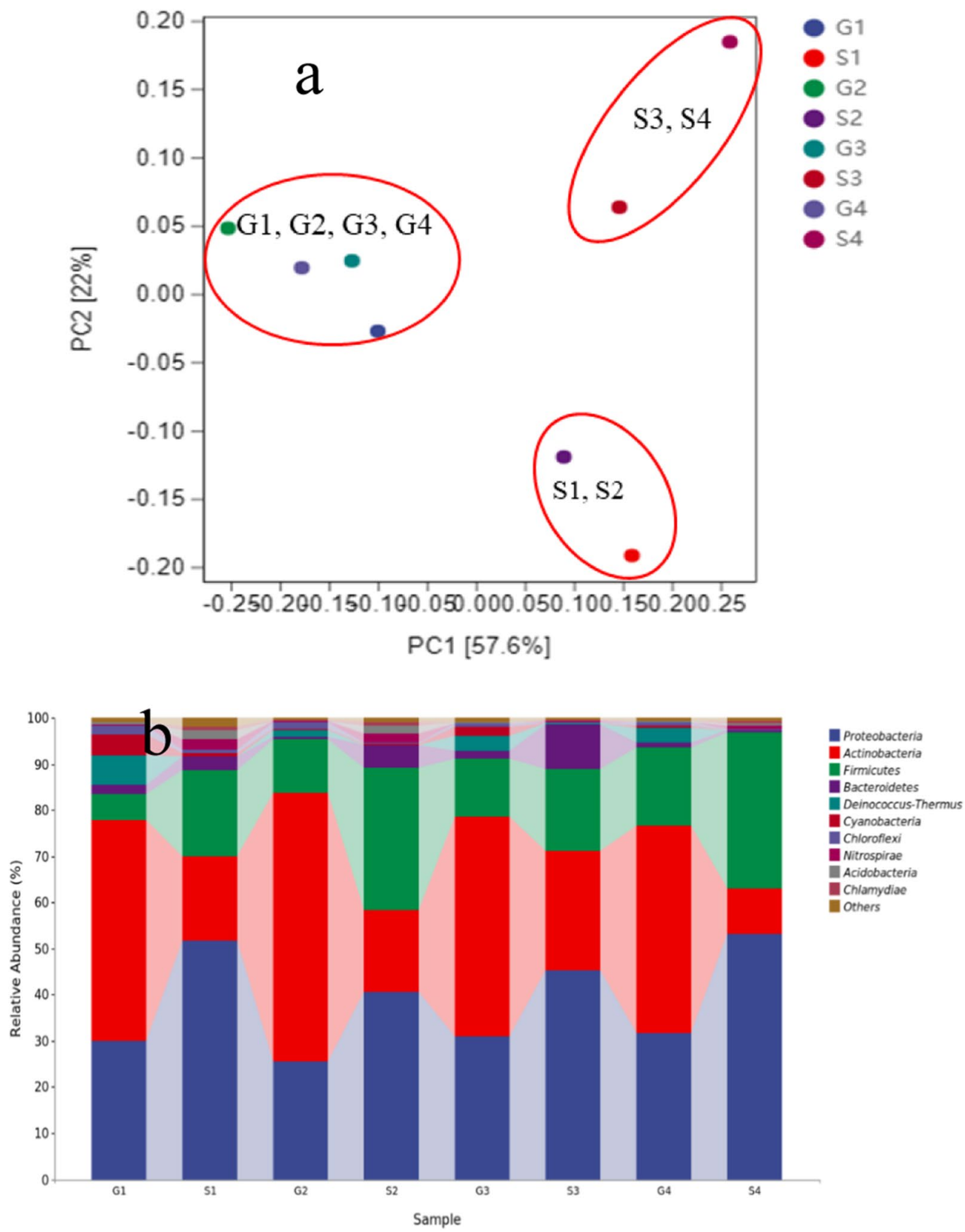


Fig. 6 Analysis of the changes of the microbial communities. **a** PCA analysis; **b** microbial distribution at phylum level

areas. In addition, the long distance between S and G indicated that the predominant bacterial community in the flooded area could be markedly changed after treatment compared to that of the non-flooded area.

Figure 6b shows the microbial distribution for the multi-unit green-wall system at the phylum level. The microorganisms with high relative abundances at the phylum level were *Proteobacteria, Actinobacteria, Firmicutes, Bacteroidetes, Deinococcus–Thermus, Cyanobacteria, Chloroflexi, Nitrospirae, Acidobacteria,* and *Chlamydia*. Among these, *Proteobacteria* are beneficial for the removal of biological nitrogen and phosphorus, and *Actinobacteria* can remove organic matter under anaerobic conditions and absorb phosphorus under aerobic conditions. In addition, *Firmicutes* and *Bacteroidetes* play important roles in the degradation of COD, including sugars and proteins [41, 42]. *Proteobacteria* were mainly concentrated in the flooded area, accounting for 51.7%, 40.6%, 45.3%, and 53.0% of layers 1–4, respectively. The relative abundance of *Proteobacteria* was highest in layer 4, indicating a stronger growth capacity with an increase in idle/reaction time. The *Actinobacteria* content did not differ significantly in the non-flooded areas, but the relative abundance was the smallest in layer 4, probably because large quantities of organic matter were degraded in the first three layers, and the lowest organic matter content in layer 4 caused a decrease in *Actinobacteria*. Besides, the *Firmicutes* and *Bacteroidetes* contents were consistent with the high ammonia removal rate of the green-wall system because of the promoting role of the ammoniated nitrification reaction [43].

Conclusions

A novel multi-unit green-wall system was designed and optimized for reusing blackwater that is rich in N and P. The optimal operating mode was observed in U-5 with the inlet time of 60 min, hold time of 120 min and HRT of 3 days. At the start-up stage, the adsorption immobilization effect of the carriers led to high removal efficiencies of various pollutants, which is benefit for the continuous degradations of the adsorbed pollutants. Then, the performance of the system was excellent in the case of blackwater with high organic load, especially for the COD and $NH4^{+}$–N with the removal rates of 98.5% and 98% because of the good buffer capacity and promoting roles of *Firmicutes* and *Bacteroidetes* which are regarding the COD degradation and ammoniated nitrification reactions. Besides, the maximum TP and TN removal rates were observed with a value of 85% and 42%, respectively. Consequently, the novel multi-unit green-wall system is an effective method for in situ blackwater reuse.

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s12302-024-00880-2.

Additional file1: Fig. S1. Plant growth change in the multi-unit green wall system. **a** Before blackwater treatment; **b** After blackwater treatment. **Fig.S2.** The equalization tank upstream designed in the muti-unit green wall system. **Fig. S3.** The COD change of effluent between unit green wall and muti-unit green wall system. **Fig. S4.** The design of each treatment unit. **Fig. S5.** The removal efficiency of greywater by the novel multi-unit green wall system (unpublished data).

Author contributions

XZ, HK and MZ were contributed to the study's conception and design; material preparation and data collection were performed by ZW, WX and FS; data analysis was performed by CF and SW. The first draft of the manuscript was written by ZW, and the manuscript was further revised by SH. All the authors read and approved the final manuscript.

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Availability of data and materials

No datasets were generated or analysed during the current study.

Declarations

Competing interests

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.

Author details

[1]College of Life and Environmental Science, Wenzhou University, Wenzhou 325035, Zhejiang, People's Republic of China. [2]State and Local Joint Engineering Research Center of Ecological Treatment Technology for Urban Water Pollution, Wenzhou 325035, Zhejiang, People's Republic of China. [3]Key Laboratory of Zhejiang Province for Water Environment and Marine Biological Resources Protection, Wenzhou 325035, Zhejiang, People's Republic of China. [4]School of Environmental Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, People's Republic of China.

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