



Original Research

Ecological filter walls for efficient pollutant removal from urban surface water



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ABSTRACT

Urban surface water pollution poses significant threats to aquatic ecosystems and human health. Conventional nitrogen removal technologies used in urban surface water exhibit drawbacks such as high consumption of carbon sources, high sludge production, and focus on dissolved oxygen (DO) concentration while neglecting the impact of DO gradients. Here, we show an ecological filter walls (EFW) that removes pollutants from urban surface water. We utilized a polymer-based three-dimensional matrix to enhance water permeability, and emergent plants were integrated into the EFW to facilitate biofilm formation. We observed that varying aeration intensities within the EFW's aerobic zone resulted in distinct DO gradients, with an optimal DO control at $3.19 \pm 0.2 \text{ mg L}^{-1}$ achieving superior nitrogen removal efficiencies. Specifically, the removal efficiencies of total organic carbon, total nitrogen, ammonia, and nitrate were 79.4%, 81.3%, 99.6%, and 79.1%, respectively. Microbial community analysis under a 3 mg L^{-1} DO condition revealed a shift in microbial composition and abundance, with genera such as *Dechloromonas*, *Acinetobacter*, unclassified_f_Comamonadaceae, *SM1A02* and *Pseudomonas* playing pivotal roles in carbon and nitrogen elimination. Notably, the EFW facilitated shortcut nitrification-denitrification processes, predominantly contributing to nitrogen removal. Considering low manufacturing cost, flexible application, small artificial trace, and good pollutant removal ability, EFW has promising potential as an innovative approach to urban surface water treatment.

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1. Introduction

The persistent discharge of untreated or inadequately treated municipal sewage has significantly contaminated urban surface water in many Chinese cities, leading to anoxia due to excessive oxygen overconsumption. Under anoxic conditions, pollutants decompose, releasing H_2S and CH_4 , which result in the blackening and malodor of urban water [1]. Eliminating urban black and odorous water is the main task at present. Physical and chemical methods, such as filtration [2], aeration [3], flocculation [4], and advanced oxidation [5], were universally applied to treat contaminants in the wastewater treatment plant. However, these methods prove inefficient for the remediation of urban water with complex

pollution components, large water volume, and wide distribution.

In contrast to other methods, bioaugmentation and ecological remediation can play an effective role in polluted urban water remediation to remove carbon [6–9], nitrogen [10,11], and phosphorus [12]. Bioaugmentation is notable for its cost-effectiveness [13], sustainability [14], and high removal efficiency [15]. However, the microbial species introduced by bioaugmentation were difficult to maintain at a high level over the long-term operation [16], limiting its widespread application. Meanwhile, despite its low construction and operation costs, convenient maintenance, and positive ecological benefits, ecological remediation faced long remediation cycles and seasonal variations [17].

Combining environmental engineering with ecological theory, bio-ecological remediation technologies present a sustainable approach to converting polluted environments into benign ecosystems [18]. The microorganisms present in these systems promoted organics and nitrogen removal, while the plants assisted in

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nitrogen and phosphorus absorption. Additionally, the roots of plants secreted oxygen [19–21], which was beneficial for nitrifying bacteria enrichment. So far, a large number of bio-ecological remediation technologies have been reported for the removal of organics [22,23], nitrogen [24–26], and phosphorus [27]. However, some limitations were still present in applying existing bio-ecological technologies in urban water remediation. For instance, constructed wetlands require a large footprint during remediation [28–30]. The contacting area of ecological floating beds [31–33] and ecological revetments [34,35] with water was limited. Furthermore, the role of microorganisms was often underutilized. Besides, existing bio-ecological technologies, such as ecological floating beds and constructed wetlands, exhibited a natural dissolved oxygen (DO) gradient. Whether the gradient had a significant impact on nitrogen removal deserved further exploration. In recent years, Yujie Feng's group focused on bio-ecological remediation technologies development for urban water, such as ecological floating beds [36,37]. However, the removal pathway of nitrogen within polyester fiber floating beds was still unclear. In the pilot scale experiment, we detected that the ecological floating bed would form the DO gradient from the outside to the inside due to its thickness. The present study continued previous research, using polyester fiber as the matrix material, and reported a novel bio-ecological remediation technology: the ecological filter walls (EFW). The EFW stands out for its low manufacturing cost, flexible application, small artificial trace, and stable water quality. It provides a new method for *in situ* remediation of urban water, demonstrating significant potential for practical application.

The present research aims to (1) study the response of EFW to DO in the removal of pollutants, (2) investigate the gradient of DO concentration generated by EFW, and (3) clarify the removal mechanism and pathway of EFW for nitrogen. The paper can provide theoretical support and technical guidance for the design and construction of EFW to maintain the long-term cleaning of urban water.

2. Materials and methods

2.1. Reactors construction

The reactors, which contained a liquid volume of 30 L, were rectangular water tanks made of polymethyl methacrylate (length \times width \times height = 35 \times 35 \times 36 cm). The porous three-dimensional matrix material weaved by polyester fiber (Wenzhou, Zhejiang East Abrasives) was 35 \times 10 \times 24 cm (length \times width \times height). The porosity of the matrix material was 92.06%, with good water permeability (Fig. S2a). The matrix materials had four through holes with a diameter of 5 cm, where emergent plants were fixed with crushed stone in the holes. *Acorus calamus*, which has a large root system and can absorb nitrogen and phosphorus, was selected as the vegetation [38]. The *Acorus calamus* from Zhejiang Qinyuanchun Gardening Store, with a fresh weight of 100 g, was planted on the matrix materials. Besides, these matrix materials were retrofitted to determine the DO concentration. In this modification process, a 3 cm diameter hole was made in the upper part of each matrix material to allow the DO probe to be attached. The reactor and matrix material design details are provided in Fig. 1. The experiments were operated in continuous lighting (12 h d⁻¹) with a light intensity of 4000 lux provided by pure blue and pure red mix-light-emitting diodes [39,40].

2.2. System operation

2.2.1. EFW start-up in batch mode

EFW was inoculated with domestic sewage and fed five times

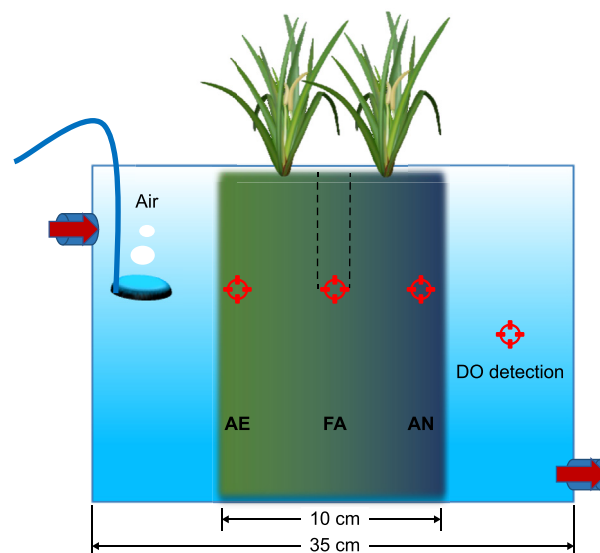


Fig. 1. Schematic diagram of ecological filter walls (EFW) and detection points for dissolved oxygen (DO) concentration in the aerobic (AE), facultative (FA), and anoxic (AN) zones.

diluted domestic sewage in a batch influent at three-day intervals. The EFW was started up successfully until the system removal efficiency of total organic carbon (TOC) and total nitrogen (TN) was stable (Fig. S1). At the same time, the DO of diluted domestic sewage was controlled as 1, 2, 3, and 4 mg L⁻¹ by pumping N₂, respectively. The experiment of nitrogen uptake by emergent plants was operated in 5 L beakers before systems ran, and the DO was also controlled as 1, 2, 3, and 4 mg L⁻¹. Then, the emergent plants were put into the EFW holes.

2.2.2. EFW running in continuous mode

The synthetic water was prepared with sodium acetate (68.33 mg L⁻¹), NH₄Cl (15.28 mg L⁻¹), KNO₃ (43.28 mg L⁻¹), and KH₂PO₄·3H₂O (2.19 mg L⁻¹). All chemicals were of analytical grade. The major characteristics of the simulated influent are shown in Table S1. The influent was stored in the tanks, and DO concentration was controlled as 1, 2, 3, and 4 mg L⁻¹ by pumping N₂, respectively. The EFW in the aerobic zone was intermittent aeration, and the DO concentration was set as 1, 2, 3, and 4 mg L⁻¹, respectively. The systems were named 1, 2, 3, and 4 mg L⁻¹-EFW. The DO in the aerobic, facultative, and anoxic zones was detected (Fig. 1), and the aeration rate was adjusted according to the DO value of the aerobic zone. The experiment was operated in continuous mode. The hydraulic retention time (HRT) was three days, and all reactors were operated at 28 \pm 1 °C. The effluent of TOC and TN was stable after 15 days, and the mass balance of nitrogen (NH₄⁺-N, NO₃⁻-N, and NO₂⁻-N) was done by calculating the nitrogen content in influent, plant uptake, nitrification, denitrification, and effluent.

2.3. Analytical methods

The functional group of the matrix material was determined by attenuated total reflection spectrum (ATR, PerkinElmer Frontier, USA) (Fig. S2b). X-ray photoelectron spectroscopy (XPS) was performed by the X-ray excitation source of Al K α by using electron energy (Fig. S2c). The hydrophilic property of the polyester fiber was measured with the wetting angle of distilled water by a YH-168A contact angle tester (YHDO, China). (Fig. S2d). The morphology of the polyester fiber was observed by the scanning

electron microscope (SEM) (Sigma 500, ZEISS, Germany) (Fig. S2e and f).

Water samples were obtained from the influent tanks and three sampling points in EFW. DO concentration was monitored by the dissolved oxygen meter (HQ30d, HACH Co. Ltd., USA). $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and $\text{NO}_2^-\text{-N}$ concentrations were measured by microplate spectrophotometer (Epoch, Bio Tek Instrument Inc, USA) according to the Standard Methods of Ministry of Ecology and Environment of the People's Republic of China (Table S2). TOC and TN were tested by the total organic carbon/total nitrogen analyzer (Multi C/N2100, Germany).

The fresh weight of emergent plants was tested by an analytical balance (AL204, Mettler Toledo Instrument Co. Ltd., China). 5 g of aboveground and belowground plant biomass were respectively collected on the 15th day of reactor operation and after the end of the operation. After collection, the samples were cleaned, dried, and milled into homogeneous powder. Nitrogen and phosphorus content was analyzed using an elemental analyzer (Vario micro cube, Kester Technology Co. Ltd., China). Total biomass was estimated by measuring the number and height of stems. The relationship between the dry weight of biomass and the height of stems was obtained by measuring the height and dry weight of all the stems in EFW after the end of the experiment. Plant uptake was calculated using the total aboveground and belowground biomass and N and P content. The luminous intensity was tested by a digital lux meter (PP730, Dongguan Three Measuring Tools Co., Ltd., Japan).

2.4. Microbial community analyses

The microbial community attached to the matrix material in the 3 mg L^{-1} -EFW after the stable operation was analyzed by the 16S rDNA MiSeq Illumina platform. The microbial community samples were obtained from the aerobic, facultative, and anoxic zones. The total DNA contents of different microbial samples were extracted after pretreatment using the E.Z.N.A.TM Mag-Bind Soil DNA Kit (Omega Bio-Tek, USA) according to the manufacturer's instructions. The hypervariable V3–V4 region of microbial 16S rRNA gene was amplified with the 16S primer pair 341F (5'-CCTACGGGNGGCWGCAG-3') and 805R (5'-GACTACHVGGGTATCTAATCC-3'). The PCR procedure was according to our previous study. After two amplifications per sample, the PCR products were gel-purified. Amplification and pyrosequencing were performed on the Illumina MiSeq 2 × 300 bp platform (Shanghai Majorbio Bio-Pharm Technology Co., Ltd). The diversity indexes, rarefaction curves, and hierarchy cluster heat map of the genus were generated for three samples. According to the Kyoto Encyclopedia of Genes and Genomes (KEGG) database, the abundance of functional enzymes related to nitrogen metabolism was predicted with PICRUSt functional analysis of 16 s rRNA gene abundance [41].

2.5. Calculations

The carbon/nitrogen ratio (C/N) was calculated according to equation (1):

$$C/N = \frac{\text{TOC}}{\text{TN}} \quad (1)$$

where C/N represents the carbon/nitrogen ratio, TOC (mg L^{-1}) is total organic carbon, and TN (mg L^{-1}) is total nitrogen.

The mass balance of nitrogen content in four reactors was calculated according to equations (2)–(5):

$$m_N = C_N \times V \quad (2)$$

$$N_{\text{nitrification}} = N_{\text{influent}} (\text{NH}_4^+\text{-N}) - N_{\text{effluent}} (\text{NH}_4^+\text{-N}) - N_{\text{plant uptake}} (\text{NH}_4^+\text{-N}) \quad (3)$$

$$N_{\text{denitrification}} = N_{\text{influent}} (\text{NO}_x^-\text{-N}) - N_{\text{effluent}} (\text{NO}_x^-\text{-N}) - N_{\text{plant uptake}} (\text{NO}_x^-\text{-N}) \quad (4)$$

$$N_{\text{influent}} = N_{\text{plant uptake}} + N_{\text{nitrification}} + N_{\text{denitrification}} + N_{\text{effluent}} \quad (5)$$

where m_N (mg) was the total nitrogen content, C_N (mg L^{-1}) was the total nitrogen concentration, and V (L) was the net water volume of 30 L in the reactors. The net water volume of 30 L was measured using a measuring cylinder.

The fate of influent nitrogen was considered in the following parts: plant uptake, nitrification, denitrification, and effluent. The mass balance of nitrogen content was calculated according to the following conditions [42]: (1) ammonia volatilization and microbial assimilation were negligible; (2) the increase of nitrite was attributed to the nitrification of ammonia nitrogen and the denitrification of nitrate; (3) the nitrate and nitrite produced from ammonia nitrogen nitrification were converted into gaseous compounds through denitrification, and the nitrogen removal contribution of this part belonged to the nitrification; (4) influent nitrogen was removed mainly through emergent plant uptake and biological removal; (5) the experiment of nitrogen removal by emergent plants uptake was operated in beakers before systems running (Fig. S3).

3. Result and discussion

3.1. Performance of EFW for pollutants removal

The four reactors with DO concentrations of 1, 2, 3, and 4 mg L^{-1} were named 1, 2, 3, and 4 mg L^{-1} -EFW. Fig. 2 shows the influent TOC concentration ranged from 18.2 to 20.0 mg L^{-1} , with an average of 18.9 mg L^{-1} . The effluent TOC was 6.5 ± 1.2 , 4.2 ± 0.2 , 3.9 ± 0.2 , and 2.7 ± 0.3 mg L^{-1} with removal efficiencies of 65.6%, 77.6%, 79.4%, and 85.7% for the 1, 2, 3, and 4 mg L^{-1} -EFW. The effluent TOC of each operating cycle met the grade III standard of TOC for municipal environment quality for urban water in China. The removal of TOC was mainly induced by aerobic degradation and denitrification. The detected average DO concentration for the 1 mg L^{-1} -EFW was 1.02 mg L^{-1} , and denitrification dominated the consumption of organic matter. As the aeration intensity increased, the removal proportion of organic matter by aerobic degradation gradually increased.

The average influent TN concentration was 9.54 ± 0.3 mg L^{-1} . The effluent TN concentration significantly reduced in 3 mg L^{-1} -EFW with an average removal efficiency of 81.3%, which was 9.6%, 10.1%, and 19.0% higher than that in 1, 2, and 4 mg L^{-1} -EFW, respectively. The effluent TN concentration in 3 mg L^{-1} -EFW was 1.79 mg L^{-1} , which met the grade V standard of TN for municipal environment quality for urban water in China ($\text{TN} \leq 2$ mg L^{-1}) (Fig. 2c). In the facultative zone, the TN concentrations were 6.3, 5.6, 5.8, and 4.9 mg L^{-1} with removal efficiencies of 34.0%, 41.5%, 39.5%, and 48.7% for 1, 2, 3, and 4 mg L^{-1} -EFW, respectively. Therefore, aerobic denitrification accounted for 30–50% of TN removal. The carbon/nitrogen ratio (C/N) of 3 mg L^{-1} -EFW decreased from 1.98 to 1.36 as carbon sources were consumed in the facultative zone. However, with extremely low carbon source levels, the effluent C/N of 3 mg L^{-1} -EFW unexpectedly raised from 1.36 to 1.90 after passing through the anoxic zone. This suggested the presence of a low-carbon denitrification process, such as nitrite denitrification or anammox in the anoxic zone.

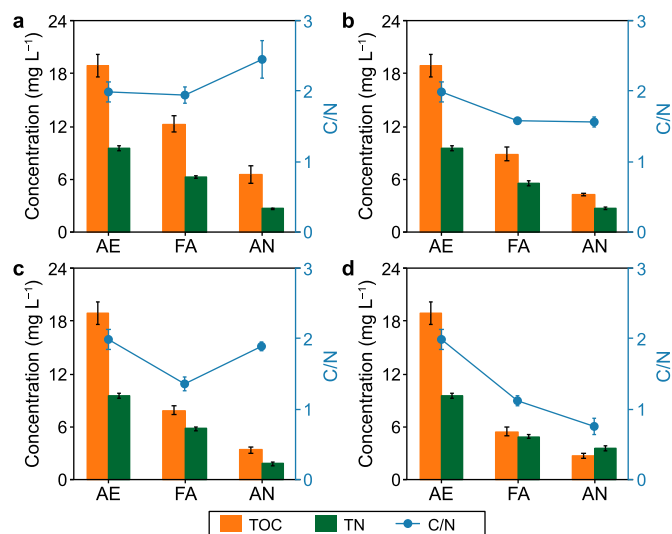


Fig. 2. Total organic carbon (TOC), total nitrogen (TN) concentrations and the carbon/nitrogen ratio (C/N) of 1 mg L⁻¹-EFW (a), 2 mg L⁻¹-EFW (b), 3 mg L⁻¹-EFW (c), and 4 mg L⁻¹-EFW (d).

NH₄⁺-N, NO₃⁻-N, and NO₂⁻-N were tested to determine the fate of influent nitrogen (Fig. 3). NH₄⁺-N and NO₃⁻-N in the influent tank were 3.6 and 5.9 mg L⁻¹, respectively. The effluent of NH₄⁺-N, NO₃⁻-N, and NO₂⁻-N in 3 mg L⁻¹-EFW was 0.03, 1.2, and 0.56 mg L⁻¹, which met the grade III standard for municipal environment quality for urban water in China (NH₄⁺-N ≤ 1 mg L⁻¹, NO₃⁻-N, and NO₂⁻-N don't have standard limits). Although the system in the study only had a small carbon source as electron donor, nitrogen removal efficiency was significantly superior to other bio-ecological remediation systems (Table S3). In other systems, most studies only focus on the effect of DO in the system but overlook the role of DO gradient in nitrogen removal process. The comparatively high pollutant removal efficiency of 3 mg L⁻¹-EFW might be due to the more efficient removal pathway of nitrogen than traditional nitrification-denitrification. The removal proportion of NH₄⁺-N in the aerobic zone positively correlated with DO in the systems, accounting for 66.2%, 87.4%, 90.3%, and 99.4% of 1, 2, 3, and 4 mg L⁻¹-

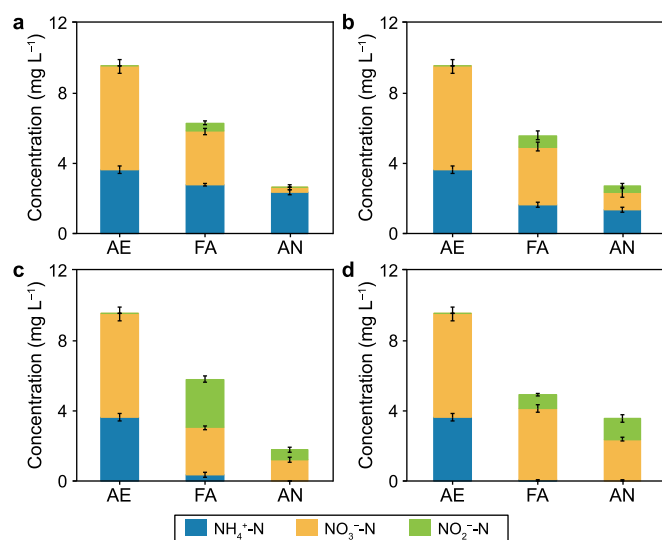


Fig. 3. The concentrations of NH₄⁺-N, NO₃⁻-N, and NO₂⁻-N in 1 mg L⁻¹-EFW (a), 2 mg L⁻¹-EFW (b), 3 mg L⁻¹-EFW (c), and 4 mg L⁻¹-EFW (d).

EFW, respectively. The results indicated that nitrifying bacteria had been directionally enriched in the EFW, which caused the high removal efficiency of NH₄⁺-N. Inspiringly, the DO concentration in the facultative and anoxic zones was low, which could meet the anoxic environment for denitrification at the same time. With a similar level of NO₃⁻-N in influent, the average NO₃⁻-N level in effluent was 1.2 and 2.3 mg L⁻¹ in 3 and 4 mg L⁻¹-EFW, respectively. The removal efficiency of NO₃⁻-N in 3 mg L⁻¹-EFW was 18.8% higher than that of 4 mg L⁻¹-EFW. Due to incomplete nitrification, there was an interesting phenomenon in the accumulation of NO₂⁻-N (2.72 mg L⁻¹) in the facultative zone of 3 mg L⁻¹-EFW. The ammonia-oxidizing ratio of NH₄⁺-N was 13.2%, 17.6%, 74.6%, and 21.0% for 1, 2, 3, and 4 mg L⁻¹-EFW in the facultative zone. The lower or higher DO concentration gradients of 1, 2, and 4 mg L⁻¹-EFW made it difficult to accumulate the nitrite.

Based on the mass balance of nitrogen (Fig. S6, Table 1), the effluent of 1, 2, 3, and 4 mg L⁻¹-EFW accounted for 27.8%, 28.4%, 18.7%, and 37.3% of TN in influent, respectively. Nitrogen removed by emergent plants accounted for 7.2–10.6% of the EFW, meaning nitrogen adsorption was not the dominant removal pathway (Fig. S3). Chen et al. reported that plant uptake contributed 7.5–14.3% to nitrogen removal by mass balance in constructed wetlands [39], similar to the paper's result. The nitrification in 3 mg L⁻¹-EFW accounted for 35.8%, which was 2.77 times higher than that in 1 mg L⁻¹-EFW and approached 4 mg L⁻¹-EFW, indicating that NH₄⁺-N effectively was converted to NO₃⁻-N in these two reactors. The nitrogen removal involved denitrification was 30.1% in 3 mg L⁻¹-EFW, which was higher than in 4 mg L⁻¹-EFW (16.2%). In conclusion, 3 mg L⁻¹-EFW showed excellent performance in simultaneously removing NH₄⁺-N and NO₃⁻-N, illustrating that suitable DO concentration in the EFW could enhance NH₄⁺-N and NO₃⁻-N removal by forming the DO gradient and affecting the microbial community.

3.2. Distribution of DO through the wall

As shown in Fig. 4, the detected DO in the aerobic zone was 1.07, 2.09, 3.19, and 4.07 mg L⁻¹ for the 1, 2, 3, and 4 mg L⁻¹-EFW, respectively. The variation of DO concentrations across the facultative and anoxic zones indicates establishing the DO concentration gradient within the EFW. In particular, the average DO in the facultative and anoxic zones of 3 mg L⁻¹-EFW was 1.19 and 1.01 mg L⁻¹, respectively. The average effluent TN in 3 mg L⁻¹-EFW was 1.79 mg L⁻¹, with the removal efficiency of 81.2%. Aeration promoted the nitrification of microorganisms, and nitrification was positively correlated with DO concentration. NH₄⁺-N was completely removed in 3 and 4 mg L⁻¹-EFW. The average DO detected in the facultative zone was 0.77, 1.04, and 1.19 mg L⁻¹ for the 1, 2, and 3 mg L⁻¹-EFW, providing the anoxic denitrification environment. The average DO of 4 mg L⁻¹-EFW in the facultative and anoxic zones was 2.89 and 2.64 mg L⁻¹, respectively. The denitrification efficiency of 4 mg L⁻¹-EFW was only 23.4%. There was a significant linear correlation between DO concentration and denitrification (Fig. 5). With the increase of DO in the aerobic zone, the denitrification efficiency of the systems decreased gradually.

The gradient of DO concentration generated by the EFW has an essential influence on pollutant removal performance because it affects the attachment of nitrifying and denitrifying bacteria. However, not all gradients have good removal for nitrogen. When the detected DO concentration was maintained at 3.19 mg L⁻¹ in the aerobic zone, the satisfying simultaneous nitrification and denitrification (SND) was achieved in 3 mg L⁻¹-EFW. In 3 mg L⁻¹-EFW, ammonia was converted to nitrite by ammonia-oxidizing bacteria (AOB) in the aerobic zone, which created a suitable environment for shortcut nitrification. The remaining organics

Table 1
Mass balance of nitrogen in 1, 2, 3, and 4 mg L⁻¹-EFW.

Reactors	Plant uptake (mg)	Nitrification (mg)	Denitrification (mg)	Effluent (mg)
1 mg L ⁻¹ -EFW	102.42	179.13	751.10	398.34
2 mg L ⁻¹ -EFW	123.51	323.69	577.31	406.50
3 mg L ⁻¹ -EFW	120.51	504.78	538.18	267.53
4 mg L ⁻¹ -EFW	152.30	497.83	247.40	533.48

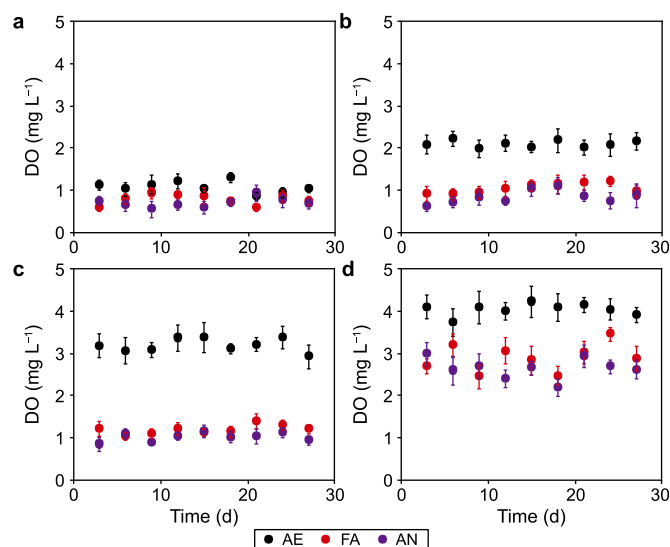


Fig. 4. DO concentration distribution of 1 mg L⁻¹-EFW (a), 2 mg L⁻¹-EFW (b), 3 mg L⁻¹-EFW (c), and 4 mg L⁻¹-EFW (d).

contributed electrons to nitrite for denitrification in the facultative and anoxic zones. The gradient distribution of DO in 3 mg L⁻¹-EFW benefited nitrogen removal. Therefore, the EFW, which formed the biofilm-like structure, was conducive to pollutant removal in the black and odorous water with low DO concentration. Such a suitable DO gradient for shortcut nitrification-denitrification (SCND) by EFW was not reported before, providing a new idea for its application. For future implementations, particularly in scenarios where water exhibits high DO levels, creating an optimal DO gradient could be achieved by adjusting the thickness of the EFW.

3.3. Microbial diversity and community distribution of 3 mg L⁻¹-EFW

DO gradient was characterized as the main factor driving the bacterial community composition. The bacterial community of 3 mg L⁻¹-EFW for different zones was analyzed by high-throughput sequencing. At the phylum level, 20 phyla were detected in the EFW (Fig. S7). Proteobacteria was the most abundant phylum, with relative abundances of 45.98%, 59.15%, and 58.20%. The phyla Cyanobacteria was the predominant bacteria in the aerobic zone (16.81%), whereas the phyla Bacteroidota and Firmicutes survived better in the facultative zone and anoxic zone. Cyanobacteria could contribute to both nitrogen reduction and organic biodegradation in the aerobic environment [43]. Besides, Bacteroidota and Firmicutes were the anaerobic bacteria involved in the nitrogen cycle and anaerobic fermentation [44,45]. These results demonstrated the crucial role of DO gradient in enriching the abundance of nitrogen conversion bacteria for superior nitrogen removal in EFW. Moreover, the bacterial community structure on the genus level showed important information on microbial populations (Fig. 6). The dominant genera in the aerobic zone included

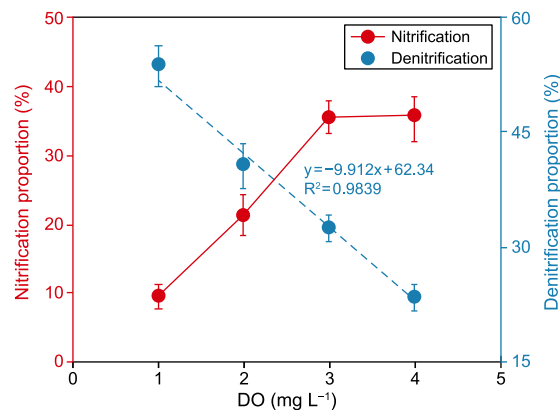


Fig. 5. The correlation between the proportion of nitrification (denitrification) and the concentration of DO in the water.

norank_f_norank_o_Chloroplast, *Zoogloea*, norank_f_Microscillaceae, UKL13-1, *Flavobacterium*, unclassified_f_Sphingomonadaceae, *Terrimonas*, and *Sphaerotilus*. On the one hand, *flavobacterium*, norank_f_Fimbriimonadaceae, *Dechloromonas*, *Zoogloea*, *Terrimonas*, *Thauera*, and unclassified_f_Sphingomonadaceae detected in the aerobic zone were related to organics degradation. Unclassified_f_Sphingomonadaceae could also degrade organics such as aromatic compounds [46]. On the other hand, norank_f_Microscillaceae (3.8%), UKL13-1 (3.7%), *Sphaerotilus* (2.3%), *Haliangium* (1.8%), and *Paracoccus* (1.8%) existed in 3 mg L⁻¹-EFW were related to nitrification [47]. In particular, unclassified_f_Comamonadaceae (6.53%) that existed in the aerobic zone was AOB, which oxidized ammonia to nitrite [48]. Besides, *Dechloromonas*, *Zoogloea*, and *Terrimonas* detected in the aerobic zones were related to aerobic denitrification.

Notably, due to less DO, the relative abundances of anoxic microorganisms in the facultative and anoxic zones were higher than that of the aerobic zone. The dominant genera in the facultative zone included *Diaphorobacter*, *Azospira*, SM1A02, *Pseudomonas*, *Acinetobacter*, *Comamonas*, unclassified_f_Rhodobacteraceae, and unclassified_f_Comamonadaceae. At the same time, the dominant genera in the anoxic were *Dechloromonas*, SM1A02, *Zoogloea*, *Acidovorax*, unclassified_f_Sphingomonadaceae, and *Ignavibacterium*. The relative abundance of conventional denitrifying bacteria in the facultative and anoxic zones was 37.5% and 21.1%, indicating that lower DO and higher carbon source concentrations were suitable for conventional denitrifying bacteria. The abundance of chemoautotrophic denitrifying bacteria in the anoxic zone was 8.2%, higher than in the facultative zone. As sewage passed through the EFW, carbon source concentration gradually decreased. The chemoautotrophic denitrifying bacteria was enriched directionally in the anoxic zone.

At the same time, *Acinetobacter* (5.5%), *Paracoccus* (2.0%), and *Pseudomonas* (5.9%) were found in EFW to simultaneously oxidize organic matter and oxidize ammonia to nitrite and nitrate, which were denitrified to N₂ under the aerobic condition [49–52]. For instance, Gu et al. reported that the increase in the C/N ratio

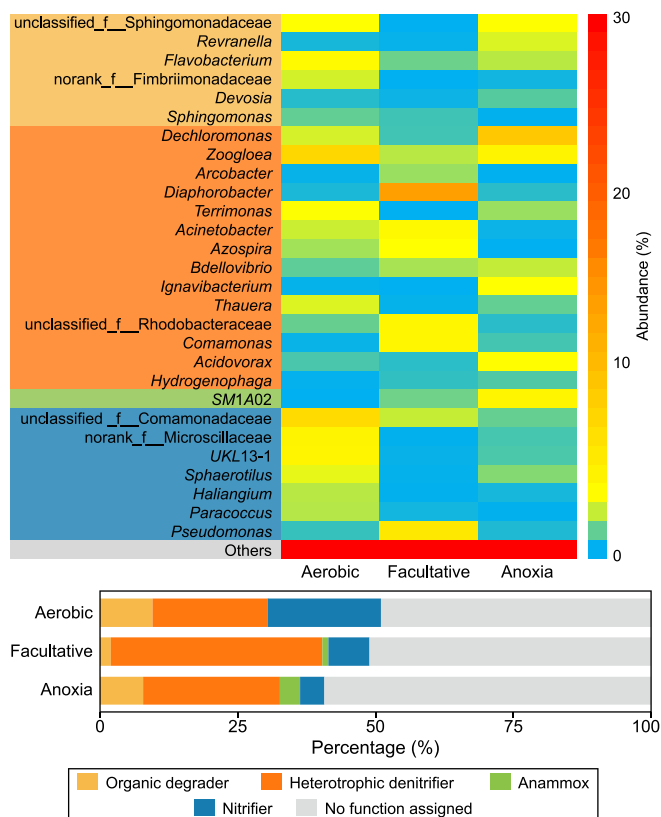


Fig. 6. The heat map graph of bacterial community structure related to carbon and nitrogen removal in different samples according to high-throughput sequencing.

enhanced the NADH/NAD(+) (nicotinamide adenine dinucleotide reduced form/nicotinamide adenine dinucleotide) ratio, NADH concentration, and enzymatic activities of *Acinetobacter* [53]. Shi et al. reported that *Paracoccus* could remove ammonia and convert over 95% nitrate to gaseous products [54]. Yang et al. studied that *Pseudomonas* showed superior heterotrophic nitrification aerobic denitrification (HNAD) ability in treating nitrogenous wastewater, with maximum removal efficiency of 10.0 and 8.9 mg L⁻¹ h⁻¹ for NH₄⁺-N and NO₃⁻-N [55]. Besides, the abundance of anaerobic ammonium oxidation (anammox) bacteria like SM1A02 was 1.1% and 3.7% in the facultative and anoxic zone, respectively [56,57]. It was reported that SM1A02 was found to be enriched in the anammox sludge [58]. In summary, NH₄⁺-N and NO₃⁻-N were degraded simultaneously under aerobic conditions. In addition, conventional denitrification, chemoautotrophic denitrification, and anammox existed in the facultative and anoxic zones.

3.4. The proposal of pollutants removal pathways in 3 mg L⁻¹-EFW

Based on the removal efficiency of pollutants and microorganisms detected in the aerobic, facultative, and anoxic zones, we proposed carbon and nitrogen removal pathways in 3 mg L⁻¹-EFW. The removal pathways mainly contained microbial degradation and emergent plant uptake. The organics were mainly degraded by aerobic and facultative aerobic bacteria. The chemoorganotrophic bacterium (such as unclassified_f_Sphingomonadaceae, *Zoogloea*, *Flavobacterium*, *Sphingomonas*, and norank_f_Fimbriimonadaceae) detected in the aerobic zone degraded the organics. At the same time, the heterotrophic denitrifying bacteria, such as *Diaphorobacter*, *Azospira*, unclassified_f_Rhodobacteraceae, and *Zoogloea*, detected in EFW degraded the organic to provide electrons for

nitrate and nitrite.

Ammonia was oxidized by nitrifying bacteria (such as unclassified_f_Comamonadaceae, norank_f_Microscillaceae, *Sphaerotilus*, UKL13-1, and *Haliangium*), and aeration provided oxygen as the electron acceptor in the aerobic zone. Due to nitrite accumulation in the facultative zone, shortcut nitrification has occurred in the aerobic and facultative zone. Enzymes related to nitrification, such as *pomA-amoA*, *pomB-amoB*, *pomC-amoC*, *narG*, and *narH*, were detected in the aerobic zone (Table S4). Besides, anammox bacteria (such as SM1A02) detected in the facultative and anoxic zone degraded ammonia and nitrite simultaneously. There were three pathways to reduce NO₃⁻-N and NO₂⁻-N, including denitrification, dissimilatory nitrate reduction (DNRA), and assimilatory nitrate reduction (ANRA). For denitrification, genes related to denitrification, such as *narI*, *napA*, *napB*, *nirK*, *nirS*, *norB*, *norC*, *nosZ*, were detected in EFW. Aerobic denitrifying bacteria (such as *Dechloromonas*, *Zoogloea*, *Terrimonas*) used carbon source as the electron donor for denitrification in the aerobic zone. Moreover, *napA* (nitrate reductase, cytochrome) and *napB* (nitrate reductase, cytochrome) related to aerobic denitrification showed a high expression abundance in the aerobic zone. Conventional denitrifying bacteria in the facultative zone used the remaining carbon source for denitrification. For DNRA, *nrfA* (nitrite reductase, cytochrome c-552) and *nrfH* (cytochrome c nitrite reductase small subunit) showed extremely low abundance, implying that the nitrite reduction might limit the DNRA in EFW. In addition, the abundances of *nasB* (assimilatory nitrate reductase electron transfer subunit) and NR (nitrate reductase (NAD(P)H)) were almost undetectable, which might insinuate that the restriction step of the ANRA might be the electron transfer of nitrate reduction. Thus, aerobic denitrification and conventional denitrification might be the main pathway to remove NO₃⁻-N and NO₂⁻-N in 3 mg L⁻¹-EFW and the genes coding of key denitrification *nir* enzymes also revealed the same conclusion.

In this process, the organic was primarily oxidized by chemoorganotrophic and heterotrophic denitrifying bacteria. For nitrogen, emergent plants took up a small amount. The DO gradient generated by 3 mg L⁻¹-EFW was crucial in nitrogen removal. On the one hand, the SCND process was the dominant removal pathway for ammonia in the aerobic and facultative zones. On the other hand, nitrate and nitrite were removed through aerobic denitrification and conventional denitrification (Fig. 7). Ammonia and

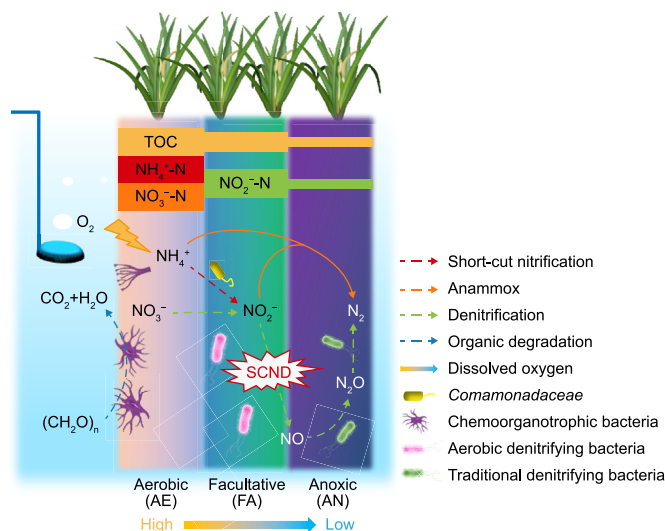


Fig. 7. The proposal of pollutants removal pathways and transformation mechanism in 3 mg L⁻¹-EFW.

nitrate were simultaneously degraded in the aerobic and facultative zones. Besides, nitrite and nitrate were denitrified by denitrifying bacteria in the anoxic zone.

4. Conclusions

An innovative ecological filter walls (EFW) was first constructed to explore the impact of the DO gradient on removing carbon and nitrogen from polluted urban water. The removal efficiencies of TOC, TN, $\text{NH}_4^+\text{-N}$, and $\text{NO}_3^-\text{-N}$ were 79.4%, 81.3%, 99.6%, and 79.1% in 3 mg L^{-1} -EFW. The organic matter was oxidized by chemo-organotrophic and heterotrophic denitrifying bacteria. The DO gradient formed by the EFW was primarily responsible for nitrogen removal, with particularly effective SND was observed in 3 mg L^{-1} -EFW. Ammonia and nitrate were simultaneously degraded in the aerobic and facultative zones, while the remaining nitrite and nitrate were denitrified by denitrifying bacteria in the anoxic zone. Shortcut nitrification-denitrification (SCND) was the primary nitrogen removal mechanism in 3 mg L^{-1} -EFW. In summary, the suitable DO gradient generated by EFW for SCND provides an innovative technology for *in situ* remediation of polluted urban water.

CRedit authorship contribution statement

Menglong Liao: Data curation, Formal analysis, Methodology, Writing – original draft. **Ye Qiu:** Methodology. **Yan Tian:** Project administration. **Zeng Li:** Conceptualization. **Tongtong liu:** Investigation. **Xinlei Feng:** Validation. **Guohong Liu:** Supervision, Validation. **Yujie Feng:** Funding acquisition, Resources, Writing – review & editing.

Declaration of competing interest

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of, the manuscript entitled, "Ecological Filter Walls for Efficient Pollutant Removal from Urban Surface Water".

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ese.2024.100418>.

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