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Nature-inspired water purification: Integrating riverbank filtration and biofilm-regulating nanofiltration

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ABSTRACT

Nanofiltration (NF) is an effective method for removing various emerging pollutants in drinking water. However, its conventional application, primarily adapted from desalination practices, faces challenges such as stringent pretreatment requirements, high energy consumption, and severe membrane fouling. To address these issues, we modified the NF process by transitioning from the traditional spiral-wound configuration to a submerged flat-sheet configuration and incorporating riverbank filtration (RBF) as a pretreatment. Experimental results demonstrated that the RBF-NF system could selectively remove natural organic matter (60.6 %) and various trace organic compounds (30.7 %–68.0 %), without the losses of beneficial ions. Additionally, the RBF-NF system reduced the risk of microbial regrowth in treated water by effectively lowering assimilable organic carbon and phosphorus levels, with removal of 52.1 % and 35.0 %, respectively. More importantly, a membrane biofilm naturally developed on the NF membrane surface over a 6-month filtration period, which facilitated the self-cleaning of the NF by biodegrading foulants and loosening the fouling cake structure. This resulted in stabilized filtration without an increase in transmembrane pressure, highlighting the potential for cleaning-free and low-maintenance operation. Additionally, the RBF-NF process reduced energy consumption by 94.6 % and carbon emissions by 87.4 % compared to traditional NF processes, primarily through the reduction of driving pressure and the elimination of crossflow. These findings demonstrate that RBF-NF is an efficient, chemical-free, and nature-based water treatment technology with significant operational and environmental benefits.

1. Introduction

The traditional drinking water treatment process, i.e., coagulation-sedimentation-filtration-disinfection (Na et al. 2021), is increasingly inadequate in addressing emerging pollutants (Kümmerer et al. 2018, Schwarzenbach et al. 2006), such as pharmaceuticals, per- and polyfluoroalkyl substances, and pesticides. To address this issue, advanced treatment methods like enhanced coagulation (Xiao et al. 2013), advanced oxidation (Esplugas et al. 2007), and adsorption (Yang et al. 2017) are integrated into the traditional process. These advanced methods, however, often depend on chemical additives, which not only increase operational costs but may also generate toxic by-products (Kümmerer et al. 2018, Sedlak and von Gunten 2011). Consequently, there is an urgent need for nature-based treatment methods that are both chemical-free and sustainable (Gitis and Hankins 2018).

Nanofiltration (NF) holds a significant potential as a chemical-free and sustainable method (Guo et al. 2022, Shao et al. 2022), due to its ability to physically remove a wide range of pollutants without the addition of chemicals. However, in practice, NF-based water treatment systems are not actually sustainable and chemical-free. This limitation primarily arises from membrane fouling, which leads to the need for chemical-based pretreatments (Lin et al. 2020) and chemical membrane cleaning (Li and Elimelech 2004). Moreover, existing NF system design are typically derived from desalination and may not be optimal. For example, an NF-based process generally involves stringent pre-treatments and high energy consumption (Guo et al. 2022, Lin et al.

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2021). Therefore, a key research question is whether NF-based processes can be redesigned to better suit the characteristics of drinking water, making the process more chemical-free and sustainable.

One potential solution is to regulate the NF process with a membrane biofilm (Zhou et al. 2023a), a naturally occurring biofilm that forms on the membrane surface under low-flux conditions (<10 L m^{-2} h^{-1}) (Desmond et al. 2022). This membrane biofilm can address the fouling issue by promoting the self-cleaning and self-stabilization of membrane system (Desmond et al. 2022, Peter-Varbanets et al. 2010), i.e., it can degrade foulants and engineer porous cake structures (Chomiak et al. 2015, Derlon et al. 2013). As a result, the membrane system could be continuously operated for months to years without the need for physical or chemical cleaning (Pronk et al. 2019), thereby significantly reducing energy consumption and chemical usage. In the context of the membrane biofilm, NF systems could be further optimized. First, at the low flux required by the membrane biofilm (Pronk et al. 2019), the driving pressure could be reduced to below 0.1 MPa, which allows for the use of a submerged configuration (Fujioka et al. 2024, Zhou et al. 2023a). Second, the membrane biofilm approach eliminates the need for the physical cleaning of crossflow, making it possible to replace traditional spiral-wound modules with other module types, such as flat-sheet module (Zhou et al. 2023a, Zhou et al. 2023b). These changes not only reduce energy consumption but also eliminate the need for stringent pretreatments, making biofilm-regulated NF a more sustainable and chemical-free alternative to conventional NF processes.

From the perspective of multi-barrier concept, integrating appropriate pretreatment with biofilm-regulated NF can improve water purification efficiency (Marron et al. 2019, Medeiros et al. 2020). Among various pretreatment options, riverbank filtration (RBF) stands out as a natural-based, chemical-free strategy (Wang et al. 2024, Zhai et al. 2022). The RBF directs water through riverbank soils, where processes such as soil retention, adsorption, and biodegradation occur, leading to the removal of particles, pathogens, and some emerging pollutants (Hu et al. 2016, Tufenkji et al. 2002, Zhai et al. 2022). Coupling RBF with biofilm-regulated NF creates a dual-barrier system that can not only enhance water purification efficiency but also mitigate membrane fouling (Wang et al. 2024). Thus, we propose that the coupling of RBF and biofilm-regulated NF offers a promising solution for chemical-free and sustainable water treatment.

In this study, we assessed the performance of the coupling of RBF with biofilm-regulated NF, focusing on pollutant removal efficiency, sustainability, and practical feasibility. We assessed the selective removal of emerging pollutants by the RBF-NF system and examined the biological stability of its permeate. Additionally, we monitored membrane fouling over a six-month period to investigate the potential for cleaning-free operation. Based on these investigations, we compared the RBF-NF system with the conventional NF and highlighted the feasibility of RBF-NF as an efficient, sustainable, and chemical-free process.

2. Methods and materials

2.1. Materials

The raw water used in this study was obtained from the East Lake, in Hubei Province, China. The dissolved organic carbon (DOC) and ultraviolet absorbance (UV_{254}) content was 7.5–8.3 mg L^{-1} and 0.060–0.082 cm^{-1}, respectively. The turbidity of the water generally ranged from 3 to 4 NTU except for rainy days. The total nitrogen (TN) concentration of the raw water was 1.17mg L^{-1}, and the ammonia nitrogen (NH_4^+-N) concentration was 0.55 mg L^{-1}. To evaluate the removal of trace organic compounds, diclofenac, carbamazepine, acetaminophen, and bisphenol A were introduced into the feedwater during days 142 to 151 at concentrations of 41.8, 17.7, 35.4, and 67.6 μg L^{-1}, respectively. The properties of these trace organic compounds were detailed in Table S1. The temperature of the raw water ranged from 19 to 25 °C.

2.2. Experimental setup

The experimental setup is shown in Fig. S1 in the Supporting Information. The RBF column, a cylinder with a height of 100 cm and an inner diameter of 5.9 cm, had an effective volume of 2.7 L. Riverbank soil was fetched from the bank of the East Lake near the Wuhan University. Large particles in the soil, such as tree roots, were removed before loading into the RBF column. The raw water was pumped into the RBF columns upwards using a peristaltic pump (BT100-2J, Longerpump, China) at a flow rate of 2.5mL min^{-1}, resulting in an empty bed contact time of 18.75 h. The effluent of the RBF column flowed to the submerged membrane tank.

Flat-sheet membrane modules were used in the biofilm-regulated NF. NF membrane coupons were originated from the commercial polyamide NF membrane (NF270, Dow Filmtec, USA), which had a water permeance of 15.5 L m^{-2} h^{-1} bar^{-1}. To better analyze the effectiveness of NF and the role of membrane biofilm, a UF membrane (UP150, Microdyn-Nadir, Germany) was also run in parallel. The UF membrane has a molecular weight cut-off of 150 kDa and a permeance of 790 L m^{-2} h^{-1} bar^{-1}. The UF and NF membrane modules were circular and had an effective surface area of 28 cm^2 (Fig. S1). For the first 55 days of the process, the influent was a dilution of East Lake water, with a ratio of East Lake water to tap water of 2:1. After that, until the end of the operation at day 188, the influent was raw East Lake water. The UF and NF membrane modules were directly submerged into a membrane tank, and continuously operated at the same flux of 4 L m^{-2} h^{-1} using peristaltic pumps (BT100-2J, Longerpump, China). This low flux was expected to facilitate the formation of membrane biofilm on the surfaces of both the UF and NF membranes. During the whole filtration, no physical and chemical cleaning was conducted.

2.3. Organic matter analysis

The fluorescent components of organic matter were investigated using a fluorescence spectrophotometer (F-7000, Hitachi, Japan). The obtained excitation-emission matrix (EEM) were quantified using a parallel factor analysis (PARAFAC) with the DOM Fluor v.1.7 toolbox (Shao et al. 2014, Stedmon and Bro 2008). The molecular weight distribution of organic matter were analyzed using a high-performance liquid chromatograph (Waters E2695, USA) with an ultraviolet absorbance detector (Waters 2486, USA). The organic matter was separated by a size exclusion chromatography to different organic compounds including biopolymers (BP), humic substances (HS), building blocks (BB), low molecular weight (LMW) acids, and neutrals (Huber et al. 2011). Paracetamol, bisphenol A, sulfamethoxazole, and carbamazepine concentrations were quantified using a high-performance liquid chromatography (HPLC, 1220Infinity II, Agilent, USA).

2.4. Biological stability analysis

The biological stability was evaluated using the indicators including assimilable organic carbon (AOC), total phosphorus (TP), and bacterial growth potential (BGP) (Prest et al. 2016). The AOC measurement generally involved incubating a water sample under controlled conditions with an indigenous bacterial inoculum (Hammes and Egli 2005). Specifically, the water samples were first filtered using 0.22 μm membranes and subsequently pasteurized. Then, 20 mL of the sample and 1 mL of the indigenous bacterial community (i.e., bacteria from the East Lake) were added into carbon-free vials, and these vials were then incubated for 72 h at 30°C. Bacterial counts were determined using a flow cytometry (FCM) (BD Accuri C6, US), following the protocols outlined in the previous studies (Hammes and Egli 2005, Li et al. 2025b). Sodium acetate was used as the AOC standard, with the corresponding standard curve depicted in Fig. S2. Phosphorus is an essential element for bacteria growth, and sufficient phosphorus can promote biomass growth in drinking water, thereby increasing the microbial risk

(Nescerecka et al. 2018, Prest et al. 2016). The phosphorus level was quantified using TP according to the standard method (Chinese SEPA, 2002). The BGP was monitored using the FCM (BD Accuri C6, US) as described previously (Farhat et al. 2018). The staining process involved mixing 500 μL of sample with 5 μL of SYBR® Green I, and incubating in the dark for 10 min. Measurement was then performed with FCM at a flow rate of 35 μL min^{-1} and an analyzed volume of 30 μL.

2.5. Characterization of membrane biofilm

The formation of membrane biofilm is very important for the sustainable filtration of NF. The membrane biofilm generally has heterogeneous morphologies, and therefore, the morphology of the membrane biofilm was in situ observed using an Optical Coherence Tomography (OCT) system (GAN210C1, Thorlab, USA). In each observation using OCT, ten spots were evenly selected on the membrane surface, and the obtained images were used to analyze the thickness and roughness of the membrane biofilm, according to the method provided in the previous studies (Shi et al. 2020, Wagner and Horn 2017).

The composition of the membrane biofilm was determined at the end of the filtration. The membrane biofilms were physically collected from the membrane surface, and their composition were analyzed in terms of the levels of microorganisms and extracellular polymeric substances (EPS). The level of microorganisms was quantified using adenosine triphosphate (ATP), which was measured using a luminometer (GloMax 20/20, Promega, USA) with an Enhanced ATP Assay Kit (Beyotime Biotechnology, China) (Hu et al. 2024). EPS was extracted by a heating method and quantified using total organic carbon (TOC).

2.6. Microbial community analysis

The membrane biofilm was collected for microbial analysis at the end of the filtration. DNA extraction was conducted by using the OMEGA E.Z.N.A ™ Mag-Bind Soil DNA Kit (OMEGA, Georgia, USA). Primers 338F and 806R were used to amplify the V3 and V4 sections of the 16S rRNA gene, while primers 528F and 706R were used to amplify the V4 region of the 18S rRNA gene for eukaryotes in the membrane biofilm. The purified PCR products were detected for high-throughput sequencing with the Illumina HiSeq platform. Clustering analysis of all communities was done and visualized using the "ggtree" package. All analyses were performed in R 4.0.5.

2.7. Economic and environmental feasibility analysis

To evaluate the economic feasibility of the RBF-NF process, the operating cost was calculated and benchmarked against the conventional drinking water treatment process and the conventional NF process (detailed in Fig. S3, Eq. S1-S4, and Table S2). The capital cost was not calculated due to its strong regional variability. The environmental feasibility of the RBF-NF process is related to its carbon emissions, which were calculated using a life cycle assessment (LCA) method (calculated by the Intergovernmental Panel on Climate Change 2013) (Table S3). The LCA was conducted using the openLCA 1.11.0 program. The impact assessment method used in this study was the CML-IA baseline (Xiong et al., 2025).

2.8. Other analytical methods

DOC and TOC were measured by a total organic carbon analyzer (Multi N/C 2100, Jena, Germany). UV_{254} was measured using a UV/Vis spectrophotometer (PGENERAL, China). The turbidity of the water sample was monitored using 2100N turbidity meter (Hach, USA). The conductivity was measured using a conductivity meter (4PII, Myron L®, USA). The concentrations of ions (e.g., Ca^{2+}, Mg^{2+}) were determined utilizing an inductive coupled plasma emission spectrometer (Avio200, PerkinElmer, USA).

3. Results and discussions

3.1. RBF-NF effectively removes pollutants without the losses of beneficial ions

Because of the efficient removal of particulate matter by NF membranes, the permeate turbidity of the RBF-NF was very low (< 0.2 NTU) (Fig. 1a). This effective removal of particulate matter indicates a significant reduction in pathogens, suggesting that the NF membrane could provide physical disinfection and thereby reducing the microbial risk in the permeate water (Crittenden et al. 2012). For organic matter (Fig. 1b and Fig. S4), the RBF-NF process achieved a stable removal of DOC during the whole filtration, with an average of 60.6 %, which exceeds the typical ~30 % removal in the conventional drinking water treatment process (Kim and Yu 2005). A comparison with NF without RBF pretreatment (NF alone) indicates that, although RBF pretreatment could remove a certain amount of organic matter, the NF membrane played a dominate role. The removal of UV_{254} was similar to that of DOC (Fig. 1c), but with higher efficiencies, e.g., an 80.8 % removal for the RBF-NF process. Because UV_{254} is often used to indicate disinfection byproduct precursors (Fu et al. 2017), its efficient removal suggests that the RBF-NF process preferentially removed disinfection byproduct precursors, which could help to reduce the formation of disinfection byproducts.

Trace organic compounds are emerging pollutants of significant concern due to their potential threats to aquatic ecosystems and human health (Kümmerer et al. 2018, Schwarzenbach et al. 2006). Fig. 1d indicates that the RBF-NF process could remove the trace organic compounds to varying extents (38.9 %–70.3 %), depending on their degradability and retentivity properties. For example, Bisphenol A, which is difficult to biodegrade or retain by NF membranes, was removed by approximately 15 % with RBF alone and less than 20 % with NF alone. However, the RBF-NF process enhanced its removal to 40 %. For Diclofenac, which is difficult to biodegrade but easily retained by NF, the primary removal contribution was from NF. Conversely, for Acetamidophenol, which is highly biodegradable but difficult to reject, the biodegradation in RBF played a more significant role. These results also suggested that biodegradation and membrane rejection played major roles in the removal of trace organic compounds. To achieve better removal of trace organic compounds, enhancing the biological activity in RBF or selecting a denser NF membrane, such as NF90, may be beneficial.

Interestingly, despite the RBF-NF process effectively removes particulates, natural organic matter, and trace organic compounds (Fig. 1a–d), it allows most salt ions to pass through, resulting in only a slight reduction in total conductivity (Fig. 1e). The RBF-NF process also showed minimal removal of Ca^{2+} and Mg^{2+}. Considering that the NF270 membrane typically retains salt ions under standard operating conditions (50 %-99 %), this minimal removal can be attributed to the low-flux operating condition in our study. Under this condition, the water flux decreases while the salt flux remains relatively constant, leading to a reduction in salt rejection (Zhou et al. 2023b), based on the solute-diffusion transport mechanism of NF membranes (Crittenden et al. 2012). The low salt removal observed in the RBF-NF process suggests that it could keep beneficial mineral ions and alkalinity, and thus enhance the chemical stability of the permeate water (Meride and Ayenew 2016). This also avoids the need for complex posttreatment to re-introduce ions, as required in typical NF and reverse osmosis processes (Crittenden et al. 2012, Shenvi et al. 2015). Additionally, low salt retention reduces concentration polarization and prevents the generation of high-salinity brine. In summary, the RBF-NF process effectively addresses numerous emerging pollutants in water, and significantly improves drinking water quality.

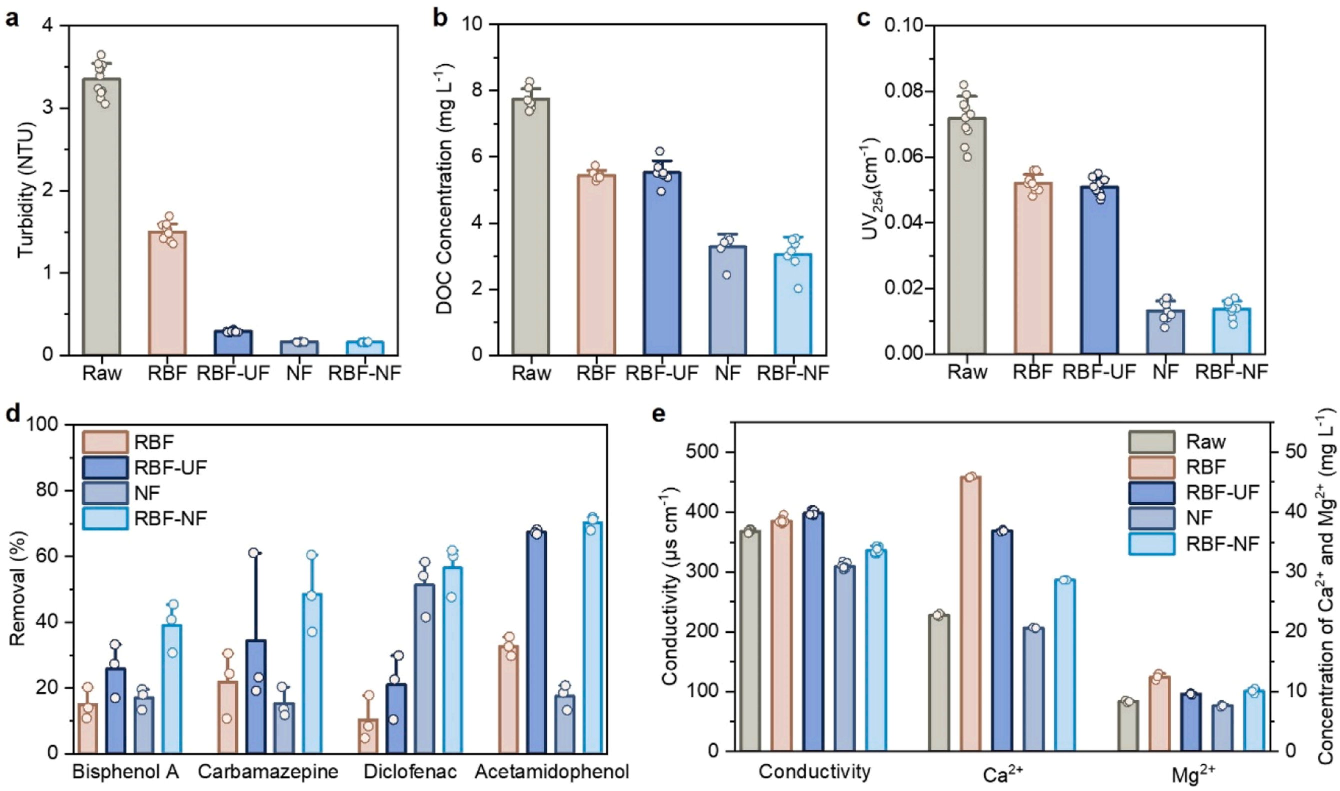

Fig. 1. Pollutant removal in the RBF alone, RBF-UF, NF alone, and RBF-NF processes: (**a**) Turbidity; (**b**) DOC; (**c**) UV_{254}; (**d**) trace organic compounds; and (**e**) major ions. The concentration of bisphenol A, carbamazepine, diclofenac, and acetaminophen in the raw water was 67.6, 17.7, 41.8, and 35.4 μg L^{-1}, respectively.

3.2. RBF-NF produces water with improved biological stability

To mitigate disinfection byproducts, besides reducing their precursors (Fig. 1c), minimizing the addition of disinfectants is also crucial (Ding et al. 2024). If water exhibits high biological stability, microbial regrowth can be inhibited, potentially allowing for reduced or no disinfectant usage (Rosario-Ortiz et al. 2016). Previous studies have shown that water with minimal nutrients for microbial growth, such as AOC and TP, generally exhibits higher biological stability (Prest et al. 2016). Fig. 2a shows that the RBF-NF process achieved a 52.1 % removal of AOC, with contributions from both RBF and NF. Microorganisms in the RBF degraded and consumed a portion of the AOC, while NF removed another portion through rejection. Additionally, RBF-UF results suggest that the membrane biofilm also contributed to AOC reduction (Shao et al. 2020a). For TP (Fig. 2b), the RBF-NF process achieved a removal efficiency of 35 %. Considering the soil used in this study released a certain amount of TP, selecting soil with lower TP concentrations (e.g., from areas far from residential zones) would significantly enhance the TP removal efficiency of the RBF-NF process; at least, the TP removal rate of RBF-NF could exceed that of NF alone, i.e., 62.5 %. In addition to evaluating AOC and TP removal, bacterial regrowth potential was also investigated to further assess biological

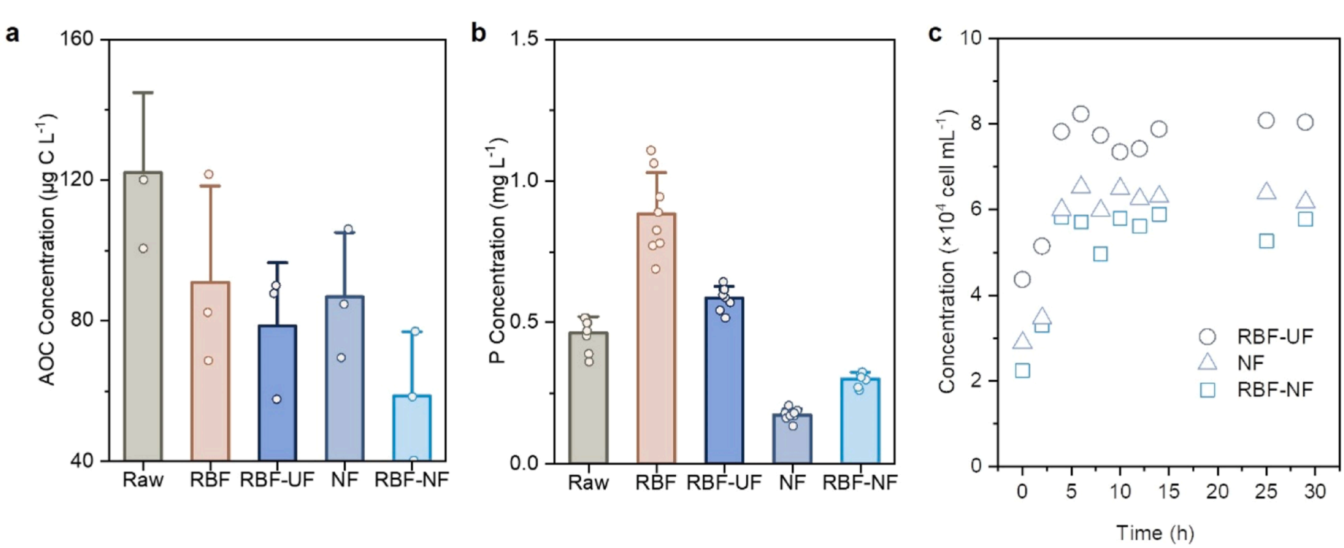

Fig. 2. Biological stability of the permeate water of RBF alone, RBF-UF, NF alone, and RBF-NF: (**a**) Assimilable organic carbon (AOC), (**b**) total phosphorus (TP), and (**c**) trend of bacterial cell regrowth. (n = 3, and error bars show the standard deviation).

stability (Fig. 2c). Bacterial regrowth exhibited a relatively short lag phase of about 4 h, reaching a stable phase around 30 h, which was consistent with the previous studies (Hammes and Egli 2005). Among the three systems, the RBF-NF permeate had the lowest bacterial counts, indicating its highest biological stability. Given that the raw water was highly polluted, employing higher-quality raw water is expected to enable treated water with higher biological stability.

3.3. RBF-NF could be stably operated without physical/chemical cleaning

3.3.1. Membrane biofilm leads to a stabilized filtration

A key feature of the membrane biofilm is its rough and loose morphology (Desmond et al. 2022, Li et al. 2025a), which differs from the dense and uniform morphology typically observed in traditional fouling layers. Cross-sectional observations using OCT (Fig. 3a) confirm the successful formation of the membrane biofilm on both the NF and UF membrane surfaces. These rough and loose morphologies are derived from microbial activities, such as degradation and predation (Chomiak et al. 2015, Derlon et al. 2013, Jiang et al. 2025). Moreover, the low-flux condition can prevent the compaction of these loose structures by water flow (Hu et al. 2023, Shao et al. 2020b). Consequently, a membrane biofilm with loose and heterogeneous structure is gradually formed on the membrane surface (Shao et al. 2022). It is worth noting that the formation of membrane biofilms typically requires several weeks, and crossflow conditions and chemical cleaning are generally unfavorable for its development (Zhou et al. 2023b). As a result, membrane biofilm is difficult to establish under the conventional operating conditions of NF membranes.

In the presence of membrane biofilm, the TMP of the NF membrane in the RBF-NF system stabilized at approximately 30 kPa over 160 d (Fig. 3b). The thickness of membrane biofilm in RBF-NF increased only in the first 100 days (Fig. 3c), after which it stabilized and even showed a decreasing trend. The relative roughness of the biofilm in the RBF-NF also gradually increased after 100 days (Fig. S5). This membrane biofilm could remove the foulants by 1) biodegrading the organic foulants (Chomiak et al. 2015), and 2) facilitating the detachment of fouling layer (Fig. S6) (Hu et al. 2023, Xiong et al. 2025). When the removal of foulants and the loading of foulants reach a balance, the thickness of the membrane biofilm is stabilized. In a typical membrane biofilm, microorganisms can degrade the rejected foulants, resulting in a much lower TOC/ATP ratio compared to the conventional cake layer. The TOC/ATP ratio observed in the three membrane systems (Fig. 3d) were comparable to those of previously reported membrane biofilms (Hu et al. 2024b, Zhou et al. 2023b), further supporting the formation of a membrane biofilm. For RBF-UF, the stabilization of the TMP and fouling layer was also observed. For NF alone, however, the TMP continuously increased from 20 to 60 kPa without RBF pretreatment, suggesting the importance of RBF in the fouling control (will be discussed in detail in the next section). Overall, the membrane biofilm led to the self-cleaning and self-stabilization of the fouling layer, and consequently, the NF could be stably operated without the need for physical or chemical cleaning (Liu et al. 2023). Consequently, the coupling of BRF and biofilm-regulated NF could be low-maintenance, chemical-free, and sustainable.

3.3.2. RBF pretreatment leads to a more efficient membrane biofilm

Intuitively, RBF pretreatment can remove certain foulants, thereby reducing the foulant load to the membrane biofilm. To better understand this, we analyzed the removal of key foulants using RBF (Fig. 4). Generally, membrane fouling is caused by a small fraction of organic matter in water, particularly protein-like components and macromolecular substances (Shao et al. 2014, Tian et al. 2013). According to

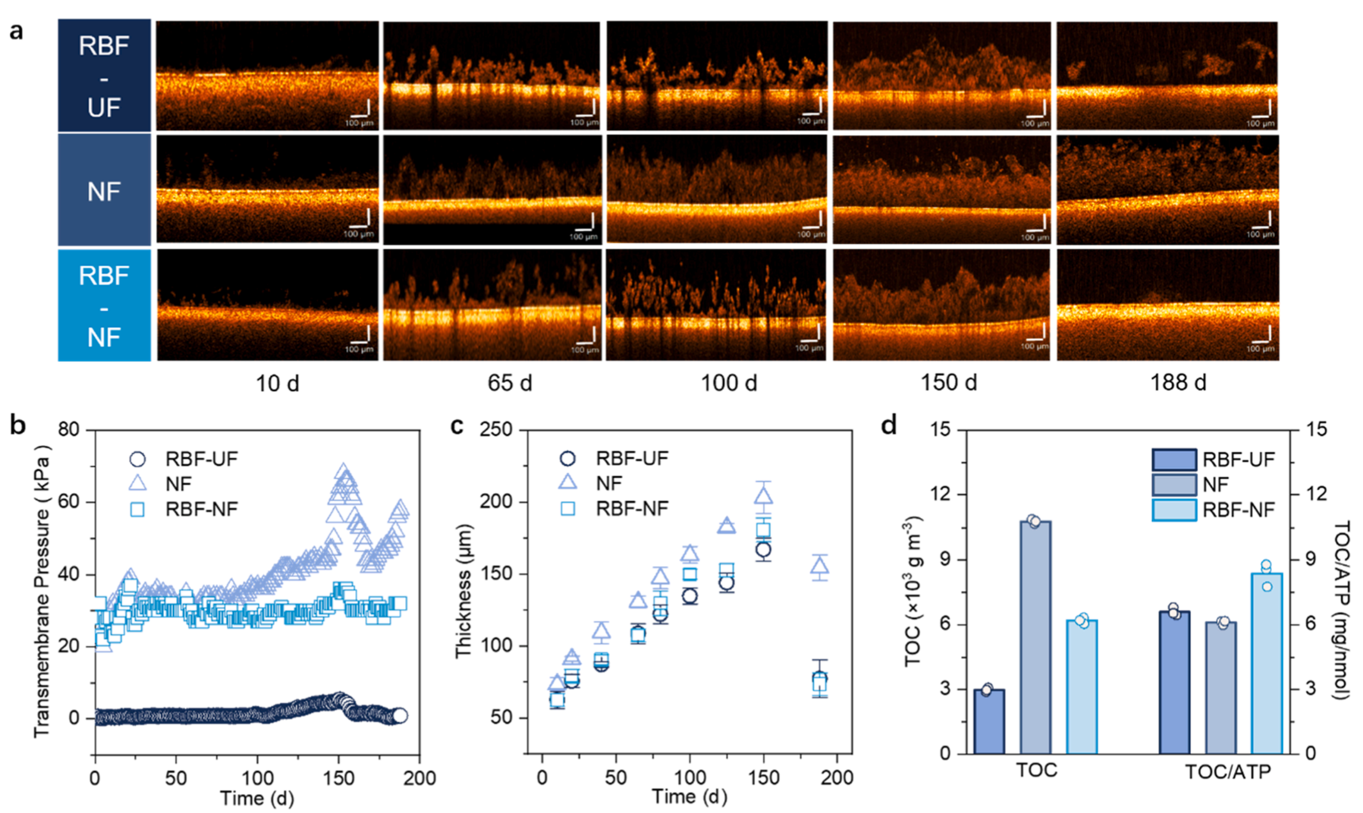

Fig. 3. Membrane fouling in the RBF-UF, NF alone, and RBF-NF. **(a)** Typical cross-sectional images of the fouling layers (membrane biofilms); **(b)** evolution of transmembrane pressure (TMP) at the flux of 4 L m^{-2} h^{-1}; **(c)** thickness of the fouling layer (membrane biofilm) during the filtration; **(d)** TOC and TOC/ATP in the fouling layers (membrane biofilm). The images were obtained in situ using OCT. The sudden change of TMP in days 142-151 was due to the addition of a methanol solution containing trace organic compounds.

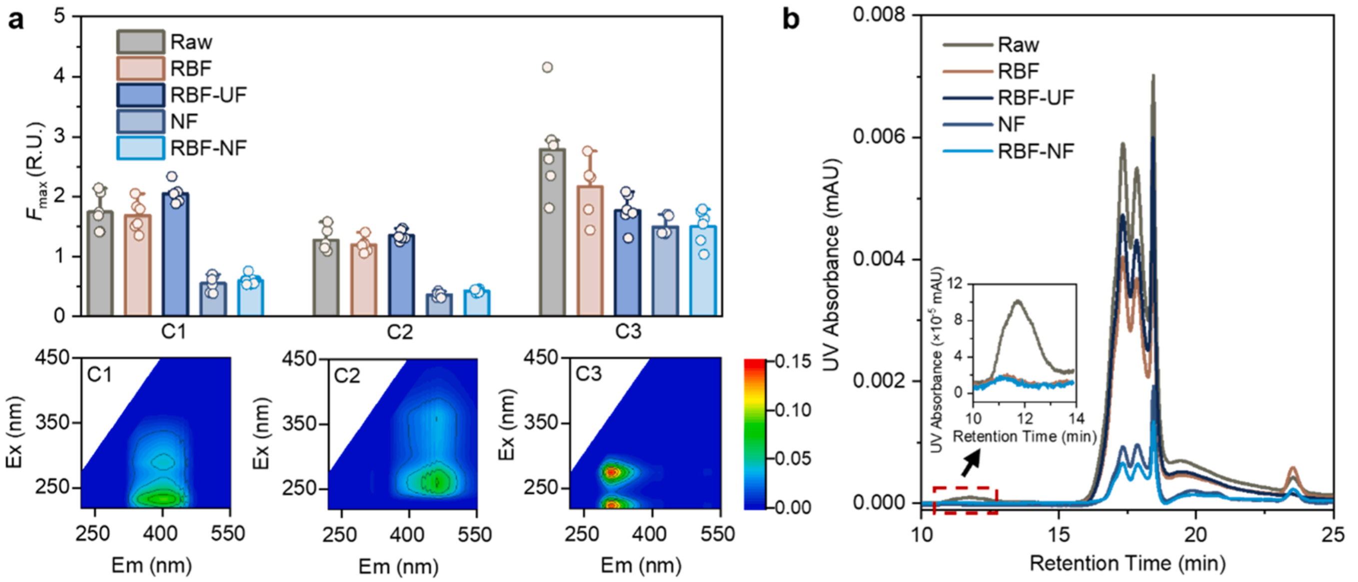

Fig. 4. Variation in organic compound components and concentrations in the RBF alone, RBF-UF, NF alone, and RBF-NF processes. **(a)** Fluorescent components; and **(b)** organic components with different molecular weight (macromolecular compounds are indicated by the peaks at the retention time of 11 min). The fluorescent components were identified using PARAFAC-EEM analysis, and three fluorescent components were identified: C1 represents microbial-derived humic substances; C2 represents terrestrial-derived humic substances; and C3 represents protein-like substances. F_{max} (the maximum fluorescence intensity) was used to indicate the concentration of the fluorescent components. C1 has excitation peaks at 240 nm and 310 nm, with emission between 400 and 450 nm; C2 has excitation peaks at 270 nm and 360 nm, with emission at 470 nm; and C3 has excitation peaks at 225 nm and 280 nm, with emission at 340 nm.

fluorescence analysis (Fig. 4a and Fig. S7), RBF selectively removed the protein-like C3 components. In contrast, the removal of humic-like C1 and C2 compounds by RBF was limited, though these compounds were efficiently rejected by the NF membrane. Additionally, RBF effectively removed macromolecular compounds (Fig. 4b), with the corresponding peak (retention time = 11 min) decreasing from 9.9×10^{-5} mAU in the raw water to 1.3×10^{-5} mAU in the RBF effluent. By effectively removing these key foulants (protein-like and macromolecular components), RBF significantly reduced the foulant loading on the subsequent NF membrane biofilm, making it easier to achieve equilibrium and self-stabilization, and thus enabling stabilized filtration (Fig. 3b). The peak of small molecular organic compounds (retention time = 11 min) increased after RBF treatment, which also indicated that microorganisms might hydrolyze some macromolecular organic compounds into small molecular organic compounds. In addition to removing key foulants, the RBF process also enriched the microbial community in the membrane biofilm (Fig. S6), enabling the biofilm to more effectively remove foulants and maintain a loose cake structure. Therefore, in the coupling of BRF and biofilm-regulated NF systems, RBF pretreatment not only enhances pollutant removal but also leads to a more efficient membrane biofilm.

3.3.3. Related the performance of RBF-NF to microbial community

The microbial communities of bacteria and eukaryotes in the RBF column and the membrane biofilms of RBF-UF, NF, and RBF-NF were compared at the end of the experiment. At the phylum level (Fig. 5a), *Firmicutes* were most abundant in the membrane biofilm of the RBF-NF system, accounting for 41.1 %, which is significantly higher than in the NF system (10.7 %). *Firmicutes* are known for their ability to rapidly utilize carbohydrates as a nutrient source (Liu et al. 2018), making them dominant bacterial groups. *Acidobacteriota* is most abundant in RBF, and it is associated with the degradation of polysaccharides and proteinaceous substances (Flieder et al. 2021), which potentially contributed to the reduction in protein-like components and macromolecular substances (Figs. 3e and Fig. 4b). Eukaryotes promote the formation of porous and heterogeneous biofilms through predation and grazing. The relative abundance of *Ascomycota* was higher in RBF (37.6 %) and RBF-NF (40.0 %) systems compared to NF (36.3 %) (Fig. 5b). *Ascomycota* is an important fungal phylum in water purification, as it actively participates in the assimilation of organic matter (Grodnitskaya et al. 2023, Sha et al. 2024).

Microbial abundance patterns are influenced by water quality (Tang et al. 2018), therefore the dominant bacterial phyla were correlated with five water quality indices: AOC, DOC, turbidity, TP, and conductivity (Fig. 5c). *Nitrospirota* and *Desulfobacterota* showed significant positive correlation with all five indices. This is likely due to their ability to tolerate adverse environmental conditions and degrade dissolved and colloidal biodegradable organic compounds (Feng et al. 2022, He et al. 2022, Rincón-Tomás et al. 2024). *Chloroflexi* phylum was positively correlated with AOC, indicating its primary role in carbon metabolism (Wu et al. 2023). However, *Verrucomicrobiota* and *Planctomycetota* were negatively correlated with all five water indices. *Verrucomicrobiota* exhibit low environmental tolerance (Bar-Shalom et al. 2023), and *Planctomycetota* are likely associated with complex lifestyles and biofilm-associated habitats (Godinho et al. 2024). These results indicated that the performance of RBF-NF is highly related to the microbial communities in the RBF and the membrane biofilm.

3.4. Practical feasibility assessment

The practical application of RBF-NF is straightforward. One approach involves constructing a membrane tank and submerging NF membrane modules within it (Fig. 6a). By introducing water from the RBF system into the membrane tank and filtering it through the submerged NF membrane, purified water can be obtained. The NF can be driven either by a water head difference or by a pump. Because pollutants rejected by the NF membrane will be accumulated in the membrane tank, the disposal of this concentrated water must be considered. The concentrated water can either be directly discharged or treated with a membrane biofilm-regulated UF membrane for low-water-quality usage (Fig. 6a) (Zhou et al. 2023a). In fact, the RBF well itself can potentially serve as the membrane tank (Fig. 6b), which could simplify the process configuration and reduce the construction costs. Additionally, pollutants retained in the well can diffuse back into the soil aquifer, where they

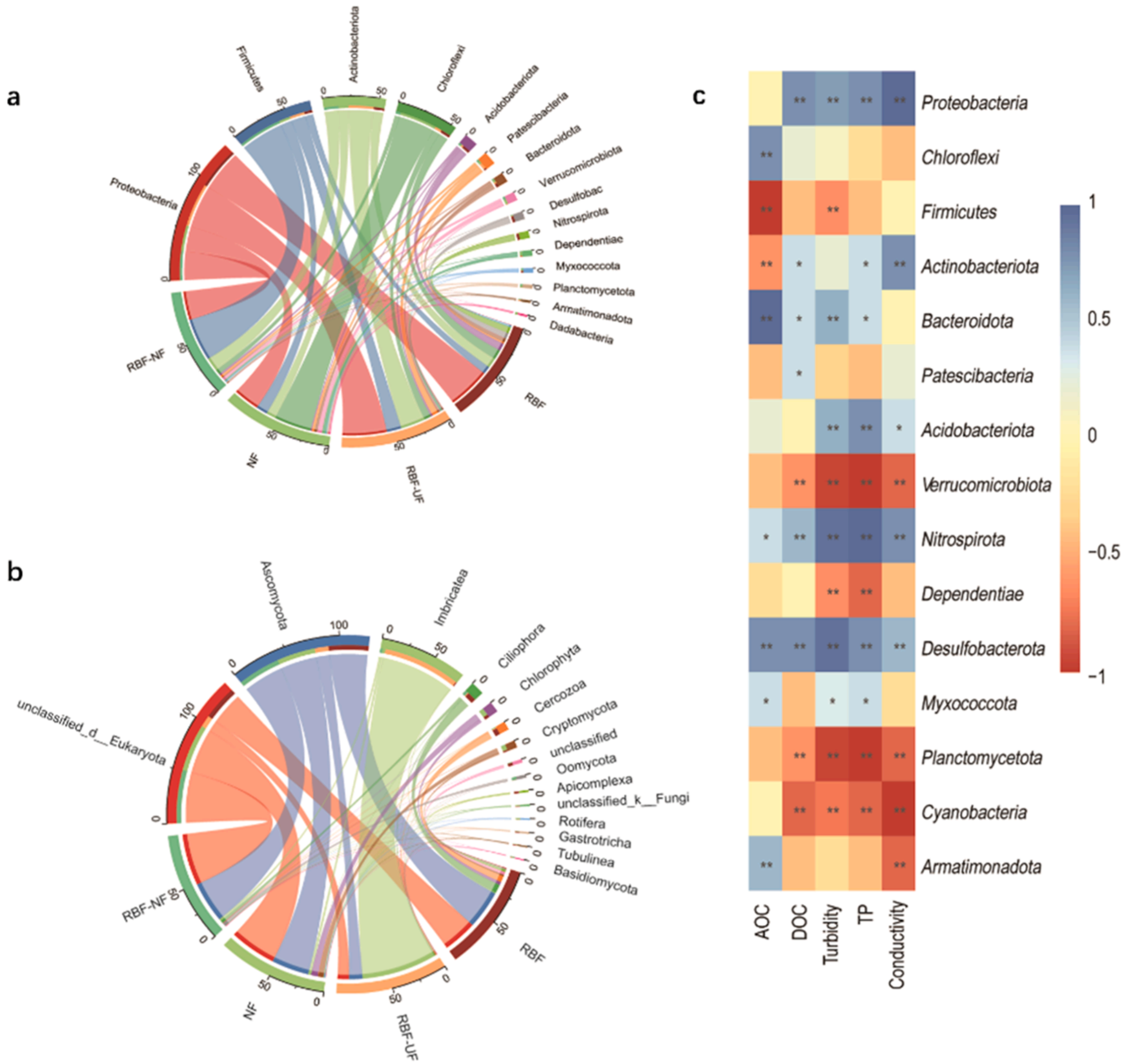

Fig. 5. Microbial community composition at the phylum level and correlation with water quality: (a) bacterial community; (b) eukaryotic community; and (c) Pearson correlation coefficient matrix between microbial phyla and water quality. Asterisks indicate correlation strength: * for low correlation ($0.3 < r < 0.5$), ** for high correlation ($0.5 < r < 1$), and no mark for weak or negligible correlation ($r < 0.3$). Blue circles represent positive correlations, while red circles indicate negative correlations.

may be removed through adsorption and biodegradation, potentially eliminating the need for concentrated water disposal. It is important to note that RBF should be employed in areas with favorable hydrogeological conditions, such as continuous river flow and permeable aquifers (Hu et al. 2016, Tufenkji et al. 2002). In locations where RBF is impractical, artificial bank filtration can serve as a viable alternative (Li et al. 2025b, Zhai et al. 2022).

The RBF-NF process significantly reduces energy consumption compared to traditional NF systems (0.0098 kWh m^{-3} vs. 0.33 kWh m^{-3}, Fig. 6c), due to the relatively low driving pressure (< 0.5 bar) and the absence of crossflow. In fact, the energy consumption of RBF-NF is only slightly higher than that of the traditional coagulation-sedimentation-filtration process, with an increase of just 0.0033 kWh m^{-3}. From an operating cost perspective, the flux used in RBF-NF (4 L m^{-2} h^{-1}) is lower than that in the traditional NF (20 L m^{-2} h^{-1}), leading to a substantial increase in NF membrane requirements and, consequently, higher membrane costs. However, RBF-NF does not require the addition of chemicals such as coagulants and membrane cleaning agents, and it also incurs lower labor costs. As a result, the overall operating cost of RBF-NF (0.58 CNY m^{-3}) is lower compared to that of traditional NF (0.86 CNY m^{-3}). Regarding carbon emissions, although RBF-NF increased the carbon emissions derived from membrane fabrication and deposal, it significantly reduced the carbon emissions derived from energy consumption and chemical usage. Therefore, the overall carbon emissions are significantly reduced by 95.1 % compared to traditional NF systems and reduced by 70.8 % compared to the traditional coagulation-sedimentation-filtration process.

Overall, RBF-NF can offer significant benefits (Fig. 6d). Compared to traditional NF, it greatly reduces energy consumption and carbon emissions. Compared to the traditional coagulation-sedimentation-filtration process, it significantly improves water quality by removing various pollutants, reducing disinfection by-products, and enhancing biological stability, with only a slight increase in operating costs and energy consumption. Notably, RBF-NF is easy to operate and requires minimal specialized personnel for management. With the incorporation of photovoltaic power, it can potentially operate independently of public electricity. Therefore, RBF-NF is highly suitable for decentralized water treatment. In summary, RBF-NF is an efficient, low-maintenance, and chemical-free process that can effectively improve the safety of drinking water.

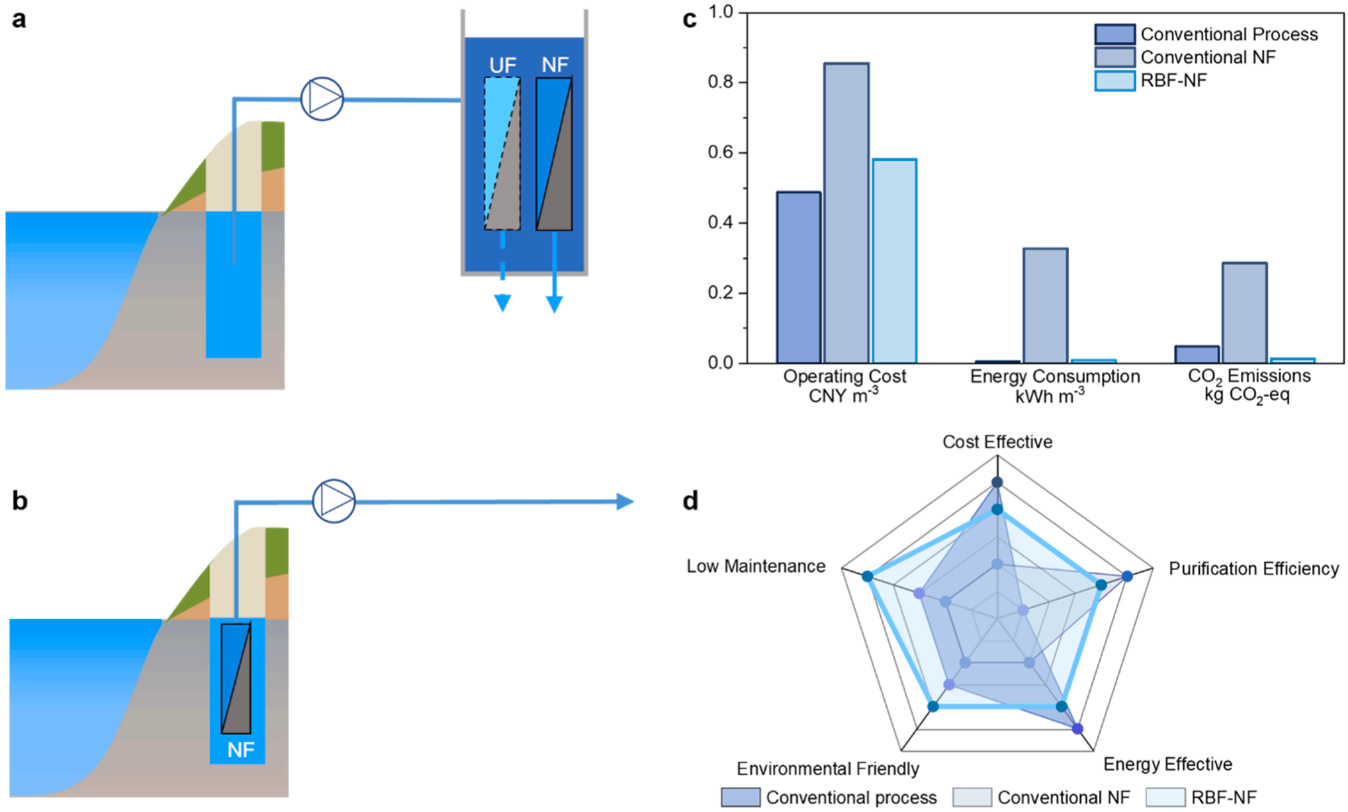

Fig. 6. Application scenarios and feasibility assessment of the RBF-NF process. **(a)** The RBF unit and the biofilm-regulated NF unit was separated; **(b)** the RBF unit and the biofilm-regulated NF unit was combined with directly submerging a NF module into the well of RBF; **(c)** comparison of operational costs, energy consumption, and carbon emissions among the conventional drinking water treatment process, the conventional NF, and the RBF-NF (detailed calculations are provided in Table S2 and Table S3); and **(d)** feasibility evaluation of the RBF-NF process.

4. Conclusions

NF is gaining increasing attention in drinking water treatment due to its ability to remove a wide range of pollutants. However, its widespread application is challenged by stringent pretreatment requirements, high energy consumption, and complex maintenance. In our study, we focused on the low-salinity characteristics of drinking water and replaced the commonly used spiral-wound NF configuration (originally designed for high-salinity desalination) with a submerged flat-sheet configuration. This configuration not only eliminates the need for strict pretreatment (e.g., simple BRF can be used), but also facilitates the effective integration of membrane biofilms with NF, enabling stabilized filtration without the need for physical or chemical cleaning. Our study highlights the importance of redesigning NF systems for low-salinity water treatment, given that NF was initially developed and optimized for high-salinity desalination.

Additionally, our study suggests that nature-based and chemical-free approaches may offer unique advantages alongside the increasing use of novel chemicals and advanced materials in water treatment. For instance, we demonstrate that RBF, a nature-based and chemical-free method, can serve as a robust pretreatment comparable to other chemical additive-based methods. Moreover, naturally grown membrane biofilms can promote the self-stabilization of fouling layers, potentially eliminating the need for complex cleaning and contributing to more sustainable filtration. Therefore, while advancing water treatment technologies with new chemicals and materials, it is crucial to also consider the benefits of nature-based approaches.

CRediT authorship contribution statement

Na Li: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis. **Yongwang Liu:** Writing – review & editing, Formal analysis. **Hongting Wan:** Formal analysis. **Li Long:** Methodology, Investigation. **Juntao Xing:** Investigation. **Senlin Shao:** Writing – review & editing, Project administration, Funding acquisition, Conceptualization. **Gang Liu:** Writing – review & editing, Conceptualization. **Walter G.J. van der Meer:** Writing – review & editing, Methodology, Conceptualization.

Declaration of competing 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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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2025.124077.

Data availability

Data will be made available on request.

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