

Pilot-scale biological activated carbon filtration-ultrafiltration (BAC-UF) system for removing pharmaceutical and personal care products (PPCPs) from surface water

Qian Wang^a, Xiaobin Tang^a, Weichen Zeng^a, Feng Wang^b, Weijia Gong^b, Jingyuan Chen^a, Jinlong Wang^{a*}, Guibai Li^a and Heng Liang^a

^aState Key Laboratory of Urban Water Resource and Environment (SKLUWRE), Harbin Institute of Technology, 73 Huanghe Road, Nangang District, Harbin, 150090, P.R. China

^bSchool of Engineering, Northeast Agricultural University, 600 Changjiang Street, Xiangfang District, Harbin 150030, PR China

E-mail address: fswaterhit@163.com (Q. Wang); tang5462@163.com (X. Tang); zeng_weichen@163.com (W. Zeng); 15534946613@163.com (F. Wang); gongweijia@126.com (W. Gong); chenjingyuan2022@163.com (J. Chen); chnwangjinlong@163.com (J. Wang); liguibai@vip.163.com (G. Li); hitliangheng@163.com (H. Liang)

* Correspondence: Author: Tel.: +86 15765532017; E-mail: chnwangjinlong@163.com (J. Wang)

Abstract: Biological activated carbon (BAC) biofilter coupling ultrafiltration (UF) is a promising process for the treatment of surface water contaminated by pharmaceutical and personal care products (PPCPs). However, the long-term pilot-scale study is urge to be investigated. In this study, a BAC-UF system with treatment capacity of 0.16 m³/h was operated. The water quality was analyzed including COD_{Mn}, UV₂₅₄, NH₄⁺-N and PPCPs. The results showed that the BAC unit reduced COD_{Mn} with 35.45%, while the BAC-UF further decreased to 56.00%. The concentration of ammonia nitrogen in the raw water was closely related to the removal rate of BAC-UF on ammonia nitrogen. Nevertheless, BAC and UF were both related to the removal effects of NH₄⁺-N, NO₂⁻-N and NO₃⁻-N. Importantly, the BAC-UF process showed high efficiency in the removal PPCPs micropollutants and the average removal of total PPCPs reached 47.84%, especially Anhydroerythromycin, Sulfachloropyridazine, Sulfadiazine, Trimethoprim, and Caffeine. It was found that the BAC unit played a key role in PPCPs removal and the UF unit also degraded them by the biomass on UF membranes. Therefore, this study proved the removal performance of BAC-UF for treating popular pollutants from surface water. The BAC-UF process in this work was considered feasible to produce clean drinking water.

Keywords: ammonia nitrogen; organic compounds; PPCPs; surface water; biological activated carbon; ultrafiltration

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1. Introduction

Over the last few decades, the risks of pharmaceutical and personal care products (PPCPs) in the aquatic environment have taken a worldwide issue in environmental concern [1, 2]. The PPCPs catalog includes a variety of chemicals, such as human and veterinary drugs, fragrances and disinfectants used in personal care products (e.g., soaps, lotions, sun-screens and body cleaning products) and household chemicals [3]. These PPCPs generally present in waters at trace concentrations, ranging from several ng/L to thousands µg/L [4]. The concentrations and diversities complicate the associated detection and even create challenges for water purification processes. Generally, many PPCPs flow through conventional water treatment processes with little degradation due to their persistency or/and the continuous introduction [5].

Ultrafiltration (UF) as emerging technology, has been widely used to remove pollutants such as particles, colloids, bacteria and viruses, reducing the risk of water-borne diseases [6]. Size exclusion is considered the primary removal mechanism for the UF. However, the PPCPs with a small molecular weight (typically < 600 g·mol⁻¹), UF membranes cannot effectively reject these soluble substances [7]. Nanofiltration and reverse osmosis are able to remove these PPCPs based on the thin-film composite [8]. Nevertheless, they play an unnecessary desalination role and were operated at a much higher energy consumption than UF [9]. The combined process with ultrafiltration may be another promising choice, having a comparable removing performance as the nanofiltration and low operational cost.

Oxidation process and adsorption process, have been applied to disposal the risk of PPCPs pollution [10]. The oxidation method exhibited a fast reaction speed and high removal efficiency. Borikar et al. [11] investigated pilot-scale advanced oxidation processes (AOPs) for removing various PPCPs. The results showed superior removal rates of over 95% involving carbamazepine, fluoxetine, naproxen, gemfibrozil, and ibuprofen. Yang et al. [10] found that UV/chlorine showed superior PPCP removal but disinfection byproducts (DBPs) were formed after chlorination. The adsorption using powder activated carbon was generally used for short-term emergency treatment when PPCPs suddenly increased [12]. Biological activated carbon (BAC) combined the adsorption and biologic degradation consuming low power energy and chemicals without concern of DBPs production as well as no frequent updates for BAC media. BAC sandwich slow sand filters can remove more than 93.2% of DEET, paracetamol, caffeine and triclosan [13]. To further reveal the contribution of adsorption and biotransformation in PPCPs removal, Paredes et al. [14] compared BAC biofilters with or without biological activity. The biotransformation was critically important to improve the degradation of PPCPs and extend the lifespan of the BAC media. However, the BAC biofilters were colonized by invertebrates and thereby caused microbial leakage out from the conventional purification process, which decreased the microbiological and chemical safety of drinking water quality [15]. Studies have shown that the number of bacteria will be increased significantly along with fine activated carbon particles in the effluent of the BAC process [16]. Besides, these particles can protect bacteria against disinfection [17]. During practical operation, the surviving invertebrates threaten human health as pathogen hosts including *Rotifers*, *Copepods*, *Cladocerans*, *Oligochaetes* and *Nematodes* [18]. The operation of BAC biofilters needs assistance from other processes to build a barrier for the invertebrates and fine BAC particles.

Therefore, BAC biofilter coupling UF is a promising process for the treatment of surface water contaminated by PPCPs. The BAC biofilter can remove the PPCPs pollutants, and then the followed UF can reject microorganisms and particles flowing out from the biofilter to ensure the quality of drinking water. Such above coupling process makes up for the defects of respective operations of BAC biofilter and UF. However, the current BAC-UF system research focuses on the removal performance with the lab scale. Shanmuganathan et al. [19] confirmed the microfiltration-BAC hybrid system, achieving removal of 45–80% to hydrophobic organics as well as removal of 50–80% to hydrophilic organics. The removal of various PPCPs (e.g., diclofenac, trimethoprim, caffeine, etc.) was ranged from 33% to 92%. Snyder et al. [20] investigated the role of pilot-scale membranes and activated carbon in the advanced treatment of municipal wastewater effluents. In addition, BAC improved the threshold flux values more for the UF membranes by reducing the reversible fouling resistance [21]. As far as we investigated, the lack of enough attention to long-term pilot study is present. Although many pilot-scale setups were used to treat secondary wastewater effluent for water reclamation, this type raw water quality was different from the surface water. Therefore, in this study, a BAC-UF system was carried out several months. The water quality was analyzed including COD_{Mn}, NH₄⁺-N and PPCPs.

2. Materials and methods

2.1. Characteristics of raw water

The experimental study was carried out in a rural area. The raw water was with-
drawn from a natural river (Foshan, Guangdong, China), which exhibited the typical char-
acteristics of a high level of $\text{NH}_4^+\text{-N}$ ($\text{mg}\cdot\text{L}^{-1}$), COD_{Mn} ($\text{mg}\cdot\text{L}^{-1}$) and PPCPs ($\text{mg}\cdot\text{L}^{-1}$) contam-
ination. The quality of raw water is summarized in Table 1.

Table 1. Raw water quality parameters of river water.

Parameter (unit).	Raw water		
	Range	Avg.	Std. Dev.
pH	6.71-8.07	7.08	0.32
Turbidity (NTU)	15.3-231.0	53.01	51.30
DO ($\text{mg}\cdot\text{L}^{-1}$)	4.34-7.46	5.94	0.79
COD_{Mn} ($\text{mg}\cdot\text{L}^{-1}$)	1.88-13.79	7.62	2.63
$\text{NH}_4^+\text{-N}$ ($\text{mg}\cdot\text{L}^{-1}$)	0.09-4.54	2.31	1.29
$\text{NO}_2^-\text{-N}$ ($\text{mg}\cdot\text{L}^{-1}$)	0.15-0.54	0.25	0.08
$\text{NO}_3^-\text{-N}$ ($\text{mg}\cdot\text{L}^{-1}$)	0.87-8.26	2.12	1.95

2.2. Pilot-scale system

The experiment was carried out with the pilot scale, which consisted of a BAC col-
umn and a gravity-driven UF system. The system was operated automatically by a pro-
grammable logic controller (PLC). A schematic diagram of the treatment process is shown
in Fig.1. The BAC column with 200 mm effective diameter and 3 m in total effective height,
contained 1.6 m granular activated carbon particles (5-10 mesh, prepared from coconut
shells) and 0.3 m cobble supporting layer. The commercial hollow-fiber modules of poly-
vinyl chloride (PVC) UF membrane were provided by Litree Purifying Technology Com-
pany (Suzhou, Jiangsu, China). These modules contained membrane fibers with an effec-
tive area of 10 m^2 and a molecular weight cutoff of 100 000 Da. All pipes for water flow
were made of PVC in the pilot-scale system.

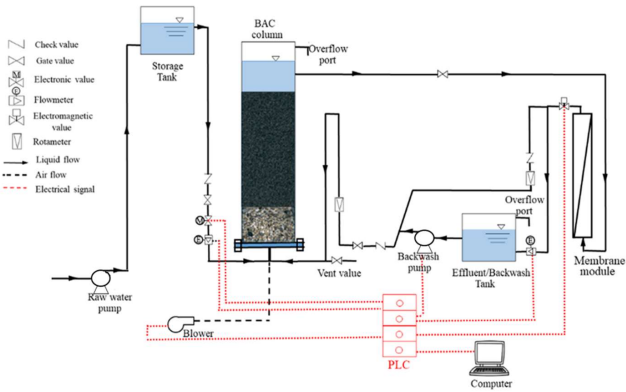


Figure 1. Schematic diagram of the experimental set-up.

2.3. Experimental protocols

The natural river water was continuously pumped from a nearby river to a storage
tank (Fig. 1). The BAC system was fed using the raw water in the storage tank. The BAC

column was operated in an up-flow mode, which maintained 25% expansion of carbon bed using the flow rate of $0.16 \text{ m}^3/\text{h}$. In order to reduce the clogging of BAC filter media, a backwash procedure was used every 7 days. This procedure was carried out as follows: the gas scrubbing was firstly performed with the backwash time and intensity of 2 min and $10 \text{ L m}^{-2} \text{ h}^{-1}$ respectively, using an air compressor. After that, the hydraulic backwashing was adopted with a duration of 1 min and an intensity of $14 \text{ L m}^{-2} \text{ h}^{-1}$, respectively. The UF system was carried out in gravity-driven mode at a constant transmembrane pressure (6.5 kPa) provided by the water level difference between the BAC column and the effluent tank [20]. The UF process was operated continuously for 40 min and then applied with forwarding flush and backwash both for 1 min respectively. The BAC-UF system was operated for 130 days, from spring to summer.

2.4. Analytical method

Characteristics of the raw water, the BAC column effluent and the UF permeate were analyzed over the course of this study. Prior to analysis, $0.45 \mu\text{m}$ cellulose acetate microfiltration membranes (Xingya Purification Material Co., Shanghai, China) were washed by nano-pure water ($18 \text{ M}\Omega\cdot\text{cm}$) and then used to filter water samples. DO concentration and pH values were measured using a multiple handheld meter (Multi 3420, WTW, Munich, Germany). Chemical oxygen demand (COD) and UV_{254} were used to evaluate removal performance. UV_{254} , representing the unsaturated organic matter like humic substances, was measured using a UV-visible spectrophotometer (UV-2450, SHIMADZU, Tokyo, Japan). The concentrations of ammonium nitrogen ($\text{NH}_4^+\text{-N}$) and nitrite nitrogen ($\text{NO}_2\text{-N}$) were determined using a spectrophotometer (722G, JK Co., Shanghai, China) with nessler's reagent and diazonium coupling reaction, respectively. The total organic carbon (TOC) and dissolved organic carbon (DOC) concentrations were measured using an automatic total organic carbon analyzer (TOC-L, SHIMADZU, Tokyo, Japan). Before the TOC determination, the samples were mixed using a magnetic stirrer. Homogenization during injection ensured measurement is performed on a representative sample.

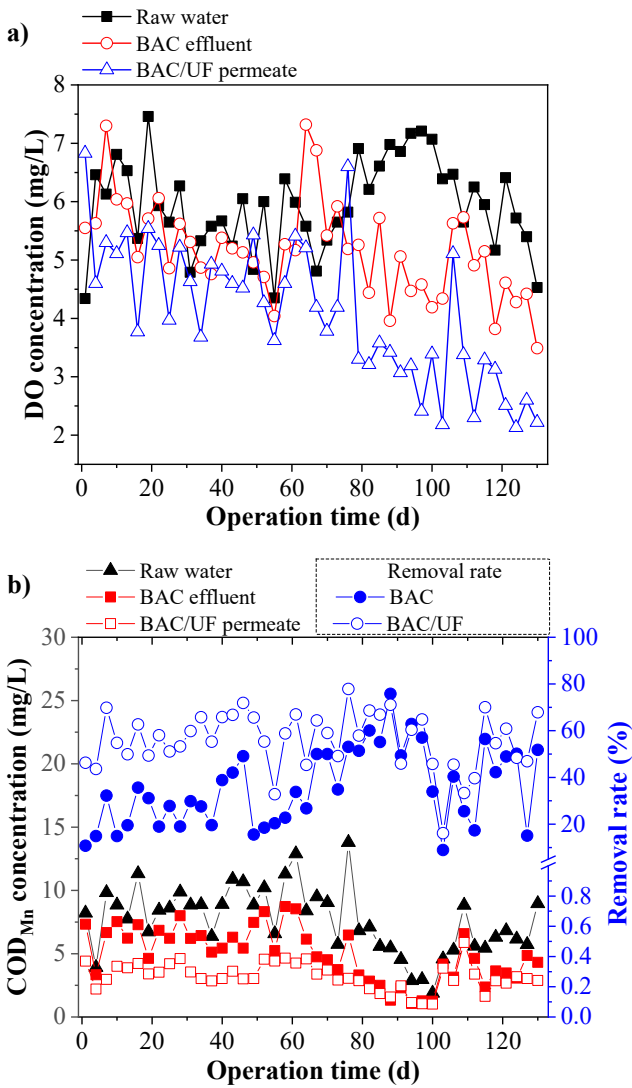
Target PPCPs were extracted using solid-phase extraction (SPE) and analyzed by high-performance liquid chromatography coupled to triple quadrupole tandem mass spectrometry (HPLC-MS/MS). Before extraction, all the water samples were vacuum filtered through $0.22 \mu\text{m}$ glass fiber filters and then spiked with corresponding internal standard solutions. Target PPCPs were determined using HPLC-MS/MS (6410B, Agilent Technologies, USA).

3. Results and discussion

3.1. Performance of organic matter removal

Fig. 2 shows the DO consumption and the removal performance of the BAC and the BAC-UF system on organic matter. The DO concentrations (Fig. 2a) in raw water varied with approximately $4.35\text{--}7.46 \text{ mg/L}$. Prolonging the operation time, the DO concentrations in the BAC effluent and UF permeate were gradually decreased, indicate the increase of biomass in this pilot-scale system. As seen in Fig. 2b, the COD_{Mn} of the raw water was $1.88\text{--}13.79 \text{ mg/L}$. As the concentration variation of organic matter in the raw water, the COD_{Mn} concentrations also waved in the BAC effluent. However, the UF membrane controlled the COD_{Mn} concentration in permeate stable, keeping the removal rate mainly above 20%. This enhancement was related to the bio-fouling layer on the UF membranes [22]. The average removal rate of COD_{Mn} was 35.45% for BAC, while the removal rate of BAC-UF had been increased to 56.00%. This finding implied that both BAC and UF could effectively remove organic matter. The results might also support the evidence of PPCPs removals. Similar to the COD_{Mn} , the UV_{254} value in the raw water was pulsed between 0.06 cm^{-1} and 0.49 cm^{-1} due to the pollution from the upstream of river. While the UV_{254} values were increased in the raw water, the removal rates of BAC and UF were also improved. Averagely, the removal rate of BAC reached about 34.98%, and that of the BAC-UF system

achieved 55.25%. This finding showed that further process after BAC using the UF membrane was necessary to secure the clean of potable water.



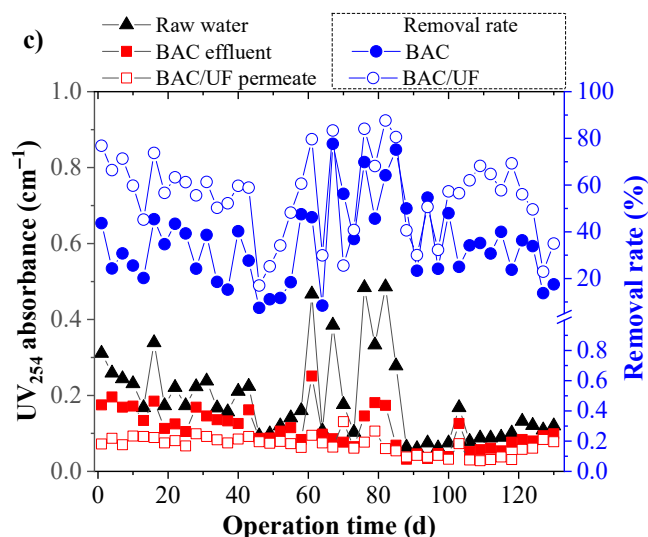


Figure 2. The removal performance of COD_{Mn} and UV_{254} by BAC-UF system: a) DO, b) COD_{Mn} , c) UV_{254} .

In order to confirm the removal mechanism of organic substances, fluorescent substances were analyzed at the end of the operation. Four peaks are illustrated in Fig.3: Peak A (Ex/Em = 230.0 nm/330.0 nm) represented aromatic protein, Peak B (Ex/Em = 270.0 nm/298.0 nm) represented soluble microbial production (similar to the protein), Peak C (Ex/Em = 260.0 nm/ 418.0 nm) represented fulvic acid-like substances, and Peak D (Ex/Em = 320.0 nm/410.0 nm) represented humic acid-like substances [23]. As seen, after the BAC treatment, intensities of four peaks were obviously decreased. This system showed a similar trend of fluorescent substances removal with previous studies where the synergistic behaviors of adsorption and microbial degradation effectively removed various pollutants [24]. However, after treating by UF, the intensities of Peak C and Peak D were increased, which might be caused by the conversation from proteins to humic substances. The decomposition process of microorganisms including fungi on the surface of the UF membrane could contribute to such conversation [25]. Therefore, the above results proved the effective bio-degradation of organic matter in the BAC-UF system.

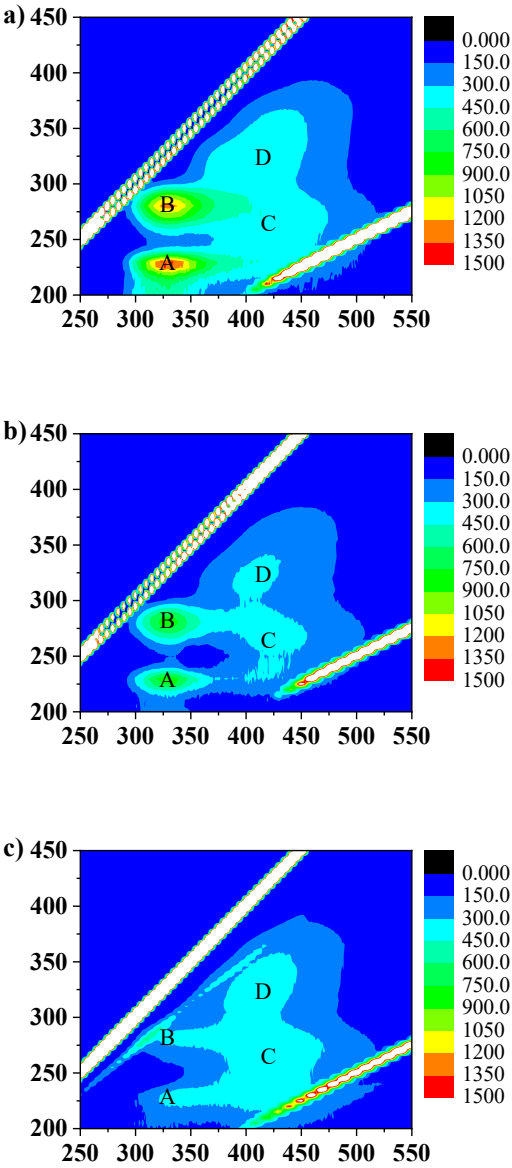
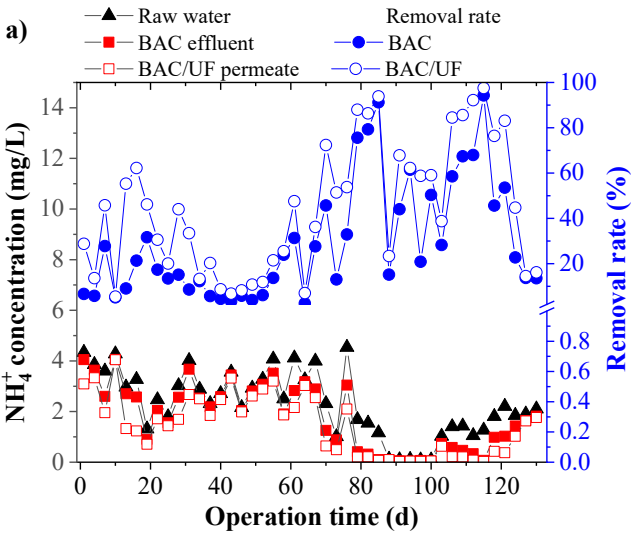


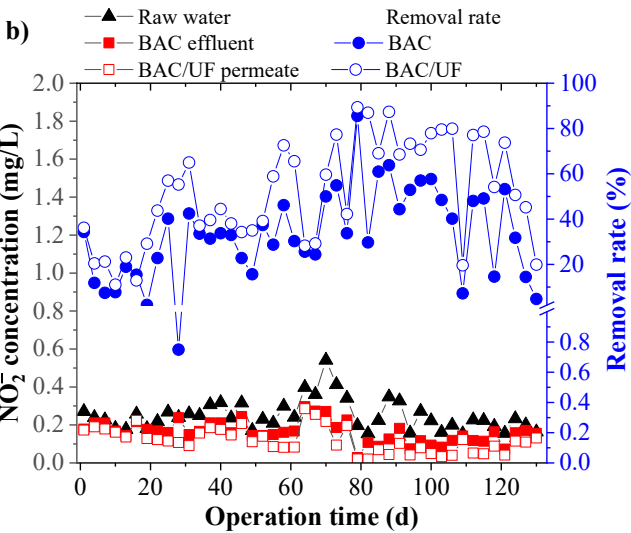
Figure 3. The removal performance of fluorescent substances by BAC-UF system: a) raw water, b) effluent of BAC, c) BAC-UF permeate.

3.2. Performance of nitrogen removal

Fig.4 shows removal of $\text{NH}_4^+\text{-N}$, $\text{NO}_2^-\text{-N}$ and $\text{NO}_3^-\text{-N}$ via the BAC-UF system. As for $\text{NH}_4^+\text{-N}$ concentration in the first 76 days (Fig. 4a), it was averagely higher than 3.10 mg/L in the raw water. Due to the limitation of dissolved oxygen concentration (Fig. 2a), the BAC removed ammonia nitrogen with a low efficiency (15.14%). After that, while the ammonia nitrogen concentration ~~downed~~ below 2 mg/L, the removal rate was increased to 50.19% in the BAC. Overall the operation, the trends of removal performance in Fig. 2a showed in parallel for the BAC and the BAC-UF, ~~showing~~ the BAC dominated the ammonia nitrogen removal. Specifically, the average removal rate on $\text{NH}_4^+\text{-N}$ was 29.48% for the BAC, and UF further increased the performance up to 44.38%. Thus, the gravity-driven UF was able to remove ammonia nitrogen, which was consistent with the previous study [22]. As for ($\text{NO}_2^-\text{-N}$), the concentration in raw water stabilized among 0.15-0.54 mg/L. With the operation time increased, its removal performance was improved for the BAC-UF system. Two possible reasons ~~might contribute to this phenomenon:~~ a) the reduction of $\text{NH}_4^+\text{-N}$ concentration decreased the competitive behavior of functional microbiome and promoted the oxidation of nitrite; b) the biomass gradually increased over time and their removal performance was increased. ~~In addition,~~ it can be seen from Fig. 4c that gravity-driven UF also ~~had~~ a significant removal effect on nitrite and improved the removal efficiency of BAC-UF. As for nitrate, the process also showed the above phenomenon in removal trend, and both BAC and UF were related to the removal effects.



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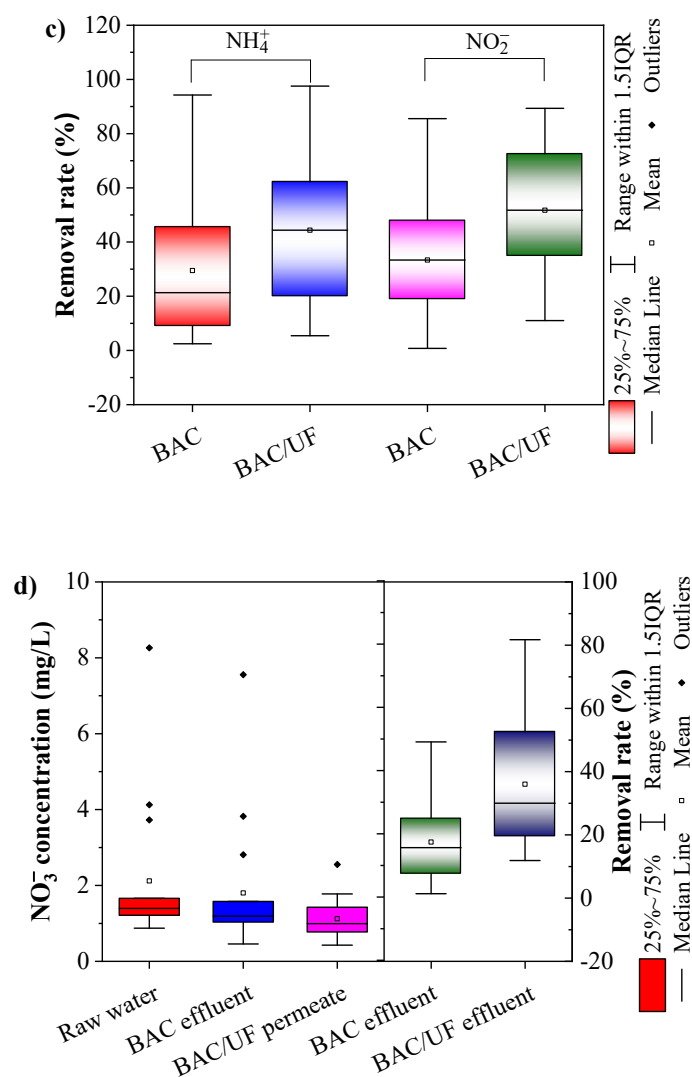


Figure 4. The removal performance of $\text{NH}_4^+\text{-N}$, $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ by BAC-UF system: a) $\text{NH}_4^+\text{-N}$, b) $\text{NO}_2\text{-N}$, comparison of BAC and BAC-UF for removals of $\text{NH}_4^+\text{-N}$ and $\text{NO}_2\text{-N}$, d) $\text{NO}_3\text{-N}$.

3.3. Performance of PPCPs removal

Table 2 summarizes the concentrations of the dominant PPCPs in the raw water. The poultry on factory farms was routinely fed these ten PPCPs, causing such surface water pollution. Erythromycin (EM) as a macrolide antibiotic reagent was widely used in industrial poultry breeding, which contributed to the high concentrations (500.56–3994.07 ng/L)

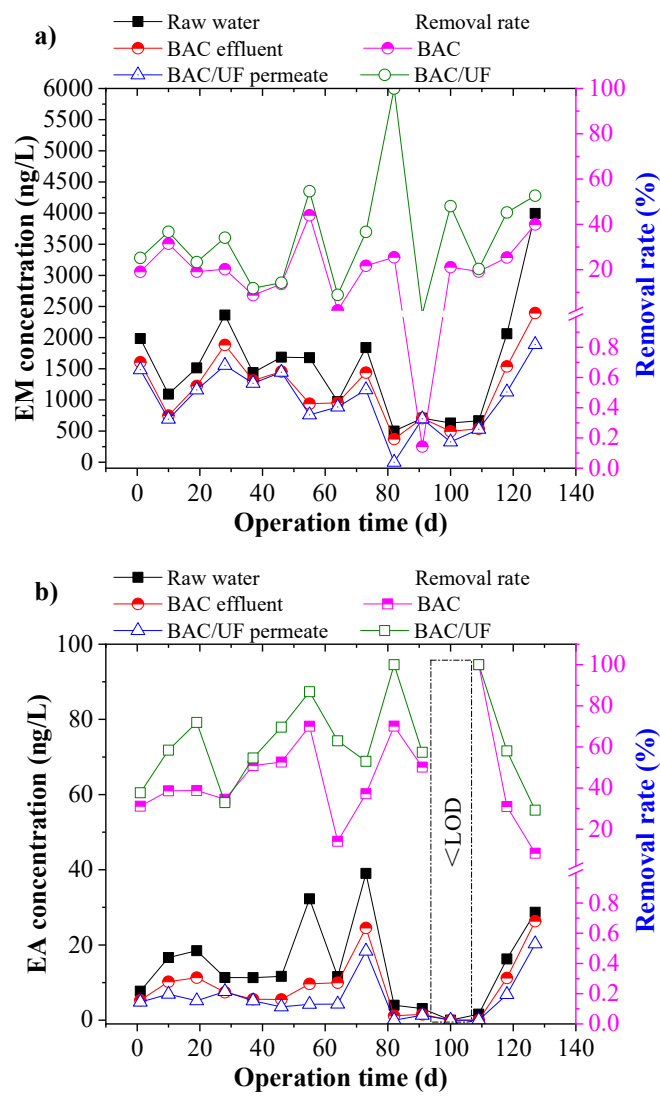
in the raw water. Other five PPCPs, including Sulfadimidine (SM2), Sulfamethoxypyridazine (SMP), Sulfametoxydiazine (SMD), Sulfadiazine (SDZ) and Caffeine (CF), existed in the raw water with maximum concentrations of above 100 ng/L. Besides, Anhydroerythromycin (EA), Sulfamethoxazole (SMX), Sulfachloropyridazine (SCP), Trimethoprim (TMP) were often detected despite relatively low concentration.

Table 2. Concentrations of typical antibiotics in the raw water.

PPCPs	Raw water (ng/L)		
	Range	Avg.	Std. Dev.
Erythromycin (EM)	500.56-3994.07	1542.43	894.38
Anhydroerythromycin (EA)	0.00-39.05	14.24	11.48
Sulfadimidine (SM2)	5.49-127.50	46.08	31.86
Sulfamethoxazole (SMX)	8.76-31.88	15.63	6.03
Sulfamethoxypyridazine (SMP)	0.75-200.84	78.04	63.91
Sulfametoxydiazine (SMD)	7.16-203.03	75.09	58.71
Sulfachloropyridazine (SCP)	0.00-58.56	19.78	15.70
Sulfadiazine (SDZ)	10.48-175.21	87.58	53.21
Trimethoprim (TMP)	0.00-16.23	3.93	5.80
Caffeine (CF)	25.41-125.34	75.13	28.23

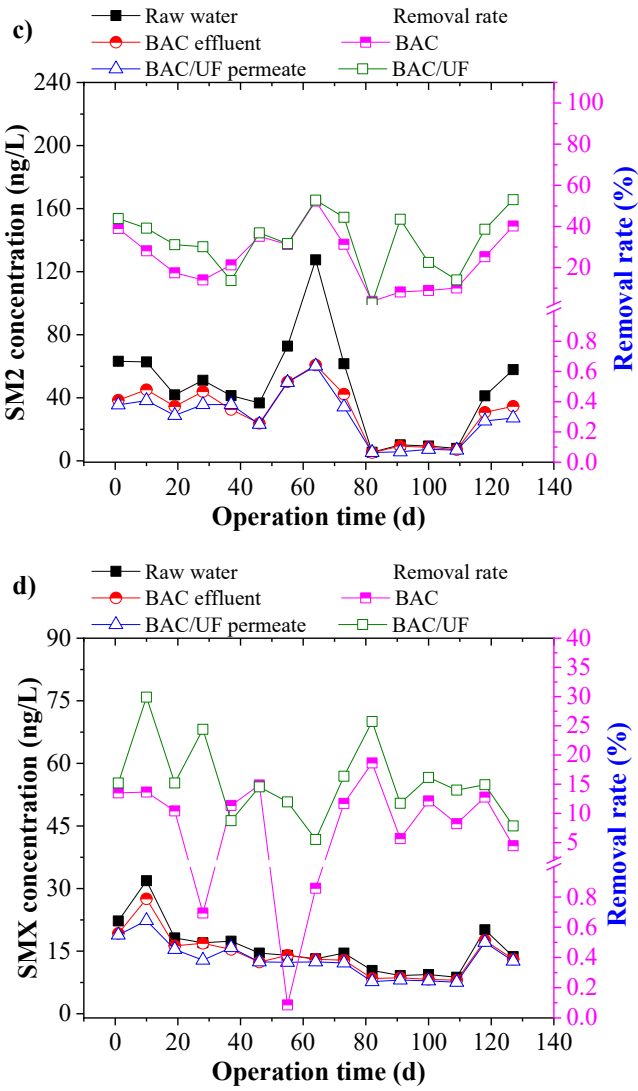
Fig. 5 depicts the removal of ten typical PPCPs using the BAC-UF system. This work was carried out during the rainy season in South China, and the rainfall played an important factor in influencing the PPCP concentration due to the dilution effect. Thus, the PPCPs concentration in the raw water showed a decreasing trend from March to July. As seen from Fig. 5b and 5i, the concentrations of EA and SCP, and TMP sometimes were measured lower than the limitation of HPLC-MS/MS spectrometry due to the rainfall dilution. In most measurements, various PPCPs were detected by the HPLC-MS/MS instrument, especially the EM. Overall, the BAC-UF process showed high efficiency in the removal PPCPs micropollutants and the average removal of total PPCPs reached 47.84%.

Specifically, the BAC effectively removed EM, EA, SM2, SMX, SMP, SMD, SCP, SDZ, TMP, and CF, reaching removal rates of 20.81%, 44.84%, 24.40%, 9.28%, 29.84%, 21.85%, 42.64%, 48.69%, 57.17%, and 45.02%, respectively. The gravity-driven UF improved the removal performance to 34.27%, 62.35%, 33.20%, 15.48%, 37.84%, 26.77%, 56.21%, 55.88%, 100.00%, and 56.42%, respectively. Therefore, the BAC-UF system showed significant removal rates of EA, SCP, SDZ, TMP, and CF, both above 50%, whereas the system showed a limited effect on the removal rate of SMX (15.48%). This removal difference may be related to the chemical structure, which favored adsorption and bioprocessing [26]. Besides, the SM2, SMP and SMD might convert to the similar structure like the SMX by BAC system, causing the low removal rate [27].



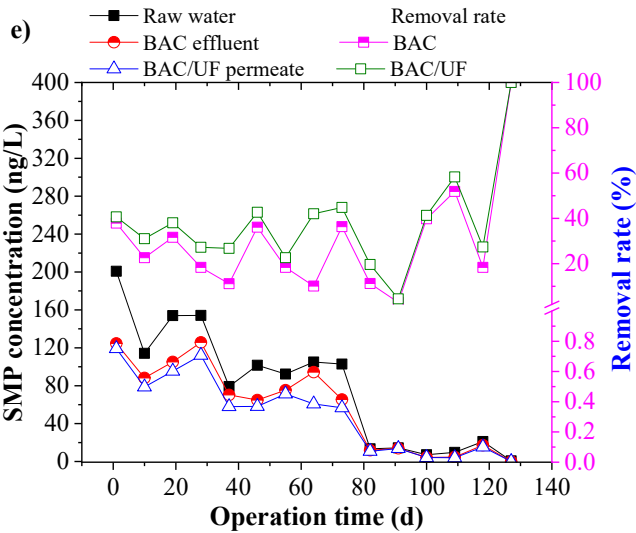
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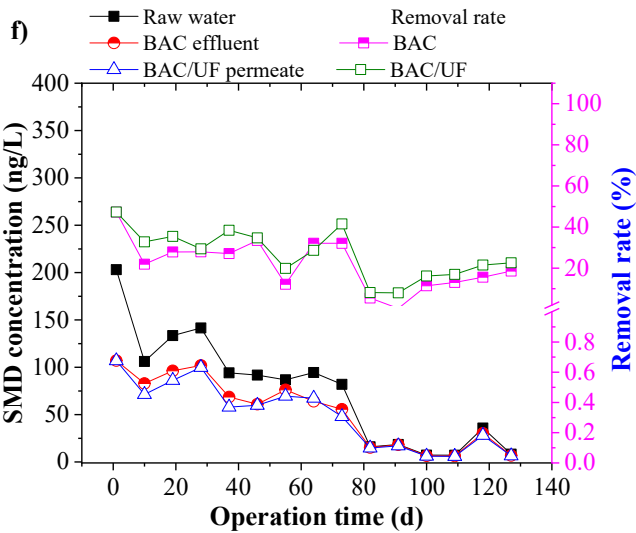


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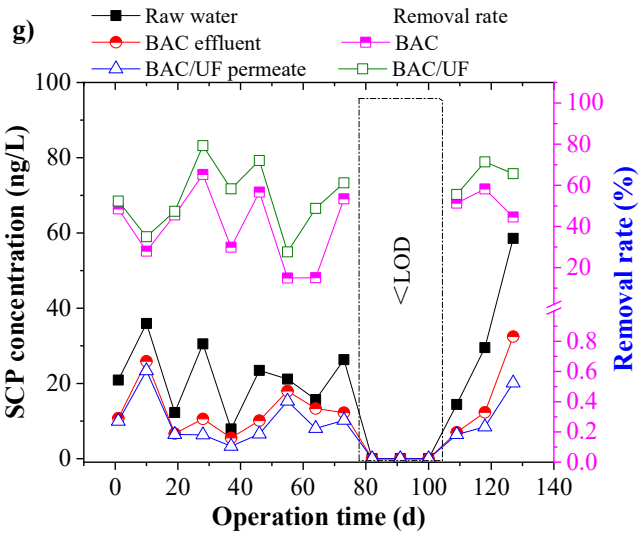
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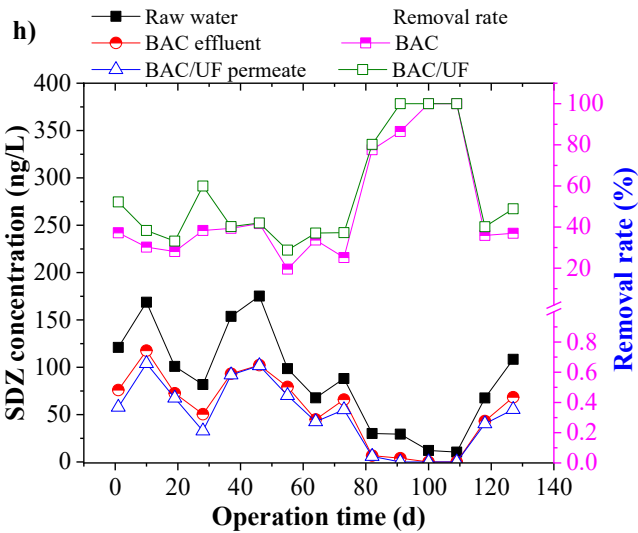
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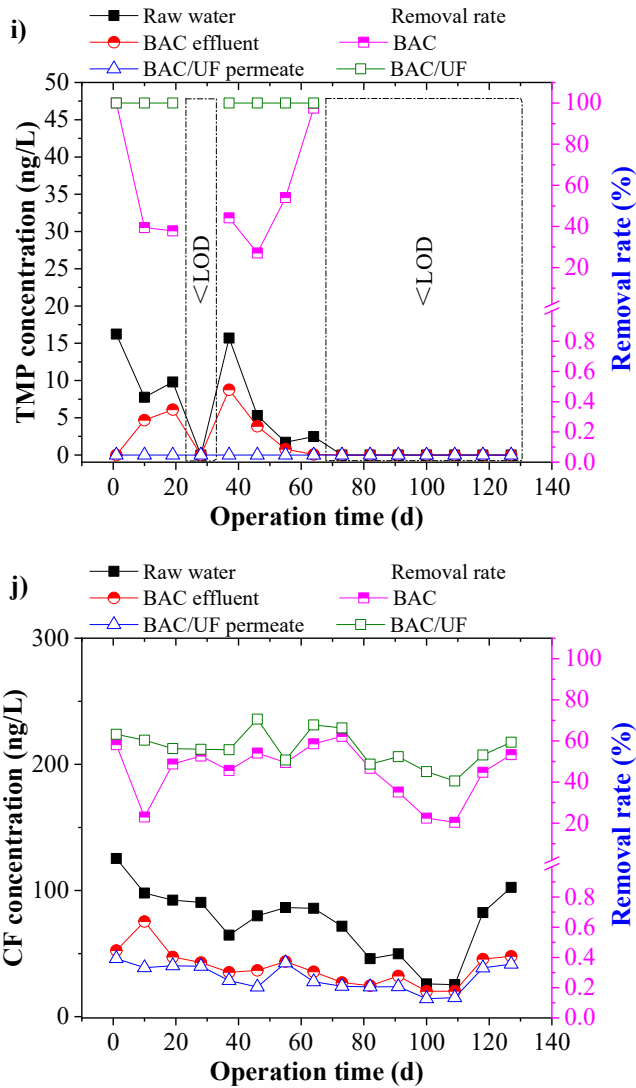


Figure 5. Removal performance of typical PPCPs by BAC-UF system: a) EM, b) EA, c). SM2, d) SMX, e) SMP, f) SMD, g) SCP, h) SDZ, i) TMP, j) CF.

Fig.6 shows the PPCPs removal distribution by the BAC and UF. BAC played the predominant role for the removal of the PPCPs, whereas UF showed a limited effect. During 130-days operation, the contribution of BAC to removal of EM, EA, SM2, SMX, SMP, SMD, SCP, SDZ, TMP, and CF was 60.72%, 71.91%, 73.50%, 59.92%, 78.84%, 81.65%, 75.85%, 87.13%, 57.17%, and 79.80%, respectively. Compared with the UF process, the

BAC process demonstrated a higher contribution to the PPCPs removal. The PPCPs removal in the BAC process was mainly attributed to adsorption and biodegradation, whereas the UF process was mainly attributed to biomass on the UF membrane. The results showed that biodegradation played an important role in the enhancement of PPCPs removal in the BAC-UF system.

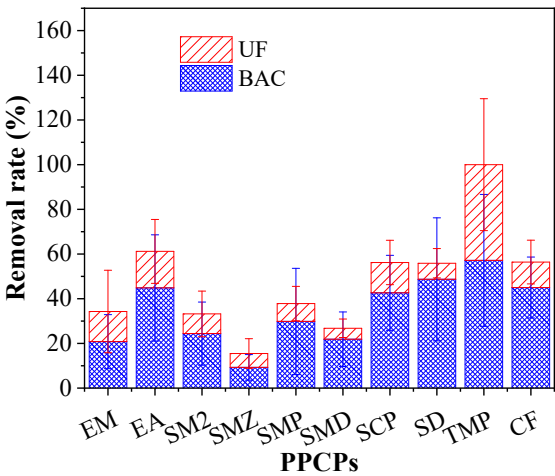


Figure 6. Typical antibiotics removal contribution by BAC and UF.

4. Conclusions

- In the pilot-scale BAC-UF process, the removal performance of BAC and UF was systematically investigated. The following conclusions can be drawn:
- (1) Based on the removal rate on organic matter, the UF membrane controlled the indexes of COD_{Mn} and UV₂₅₄ in permeate stable. Further process after BAC using the UF membrane was necessary to secure the clean of potable water.
 - (2) Both BAC and UF were related to the removal effects of NH₄⁺-N, NO₂⁻-N and NO₃⁻-N. The removal rate of UF was lower than that of BAC unit. The concentration of ammonia nitrogen in the raw water was closely related to the removal rate of BAC-UF on ammonia nitrogen.
 - (3) The BAC-UF process showed high efficiency in the removal PPCPs micropollutants and the average removal of total PPCPs reached 47.84%. Especially, removal rates of Anhydroerythromycin, Sulfachloropyridazine, Sulfadiazine, Trimethoprim, and Caffeine were above 50%.
 - (4) The BAC unit played the key role in PPCPs removal and the UF unit was mainly attributed to biomass on the UF membrane, synergistically degrading various PPCPs pollutants.

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1. [1] J.M. Brausch, G.M. Rand, A review of personal care products in the aquatic environment: Environmental concentrations and toxicity, *Chemosphere* 82(11) (2011) 1518-1532. <https://doi.org/https://doi.org/10.1016/j.chemosphere.2010.11.018>.
2. [2] P. Chaturvedi, P. Shukla, B.S. Giri, P. Chowdhary, R. Chandra, P. Gupta, A. Pandey, Prevalence and hazardous impact of pharmaceutical and personal care products and antibiotics in environment: A review on emerging contaminants, *Environ. Res.* 194 (2021) 110664. <https://doi.org/https://doi.org/10.1016/j.envres.2020.110664>.
3. [3] M. Xu, H. Huang, N. Li, F. Li, D. Wang, Q. Luo, Occurrence and ecological risk of pharmaceuticals and personal care products (PPCPs) and pesticides in typical surface watersheds, China, *Ecotoxicology and Environmental Safety* 175 (2019) 289-298. <https://doi.org/https://doi.org/10.1016/j.ecoenv.2019.01.131>.
4. [4] Y. Zhou, J. Meng, M. Zhang, S. Chen, B. He, H. Zhao, Q. Li, S. Zhang, T. Wang, Which type of pollutants need to be controlled with priority in wastewater treatment plants: Traditional or emerging pollutants?, *Environ. Int.* 131 (2019) 104982. <https://doi.org/https://doi.org/10.1016/j.envint.2019.104982>.
5. [5] Q. Bu, B. Wang, J. Huang, S. Deng, G. Yu, Pharmaceuticals and personal care products in the aquatic environment in China: A review, *J. Hazard. Mater.* 262 (2013) 189-211. <https://doi.org/https://doi.org/10.1016/j.jhazmat.2013.08.040>.
6. [6] S. Al Aani, T.N. Mustafa, N. Hilal, Ultrafiltration membranes for wastewater and water process engineering: A comprehensive statistical review over the past decade, *J. Water Process Eng.* 35 (2020) 101241. <https://doi.org/https://doi.org/10.1016/j.jwpe.2020.101241>.
7. [7] Y. Li, C. Zhang, Z. Hu, Selective removal of pharmaceuticals and personal care products from water by titanium incorporated hierarchical diatoms in the presence of natural organic matter, *Water Res.* 189 (2021) 116628. <https://doi.org/https://doi.org/10.1016/j.watres.2020.116628>.
8. [8] L. Bai, Y. Liu, N. Bossa, A. Ding, N. Ren, G. Li, H. Liang, M.R. Wiesner, Incorporation of Cellulose Nanocrystals (CNCs) into the Polyamide Layer of Thin-Film Composite (TFC) Nanofiltration Membranes for Enhanced Separation Performance and Antifouling Properties, *Environ. Sci. Technol.* 52(19) (2018) 11178-11187. <https://doi.org/10.1021/acs.est.8b04102>.
9. [9] J. Wang, X. Tang, X. Cheng, J. Xing, H. Wang, G. Li, H. Liang, In-situ crystallization generated by CEM electrolysis for NF concentrate softening along with the alleviation of ceramic membrane fouling, *Desalination* 516 (2021) 115243. <https://doi.org/https://doi.org/10.1016/j.desal.2021.115243>.
10. [10] Y. Yang, Y.S. Ok, K.-H. Kim, E.E. Kwon, Y.F. Tsang, Occurrences and removal of pharmaceuticals and personal care products (PPCPs) in drinking water and water/sewage treatment plants: A review, *Sci. Total Environ.* 596-597 (2017) 303-320. <https://doi.org/https://doi.org/10.1016/j.scitotenv.2017.04.102>.
11. [11] D. Borikar, M. Mohseni, S. Jasim, Evaluations of conventional, ozone and UV/H₂O₂ for removal of emerging contaminants and THM-FPs, *Water Quality Research Journal* 50(2) (2014) 140-151. <https://doi.org/10.2166/wqrj.2014.018>.
12. [12] P. Yu, X. Li, X. Zhang, H. Zhou, Y. Xu, Y. Sun, H. Zheng, Insights into the glyphosate removal efficiency by using magnetic powder activated carbon composite, *Sep. Purif. Technol.* 254 (2021) 117662. <https://doi.org/https://doi.org/10.1016/j.seppur.2020.117662>.
13. [13] J. Li, Q. Zhou, L.C. Campos, The application of GAC sandwich slow sand filtration to remove pharmaceutical and personal care products, *Sci. Total Environ.* 635 (2018) 1182-1190. <https://doi.org/https://doi.org/10.1016/j.scitotenv.2018.04.198>.
14. [14] L. Paredes, E. Fernandez-Fontaina, J.M. Lema, F. Omil, M. Carballa, Understanding the fate of organic micropollutants in sand and granular activated carbon biofiltration systems, *Sci. Total Environ.* 551-552 (2016) 640-648. <https://doi.org/https://doi.org/10.1016/j.scitotenv.2016.02.008>.
15. [15] H. Schreiber, D. Schoenen, W. Trautspurger, Invertebrate colonization of granular activated carbon filters, *Water Res.* 31(4) (1997) 743-748. [https://doi.org/https://doi.org/10.1016/S0043-1354\(96\)00312-0](https://doi.org/https://doi.org/10.1016/S0043-1354(96)00312-0).
16. [16] L. Han, W. Liu, M. Chen, M. Zhang, S. Liu, R. Sun, X. Fei, Comparison of NOM removal and microbial properties in up-flow/down-flow BAC filter, *Water Res.* 47(14) (2013) 4861-4868. <https://doi.org/https://doi.org/10.1016/j.watres.2013.05.022>.
17. [17] T. Lin, W. Chen, L. Wang, Particle properties in granular activated carbon filter during drinking water treatment, *J. Environ. Sci.* 22(5) (2010) 681-688. [https://doi.org/https://doi.org/10.1016/S1001-0742\(09\)60163-7](https://doi.org/https://doi.org/10.1016/S1001-0742(09)60163-7).
18. [18] Q. Wang, W. You, X. Li, Y. Yang, L. Liu, Seasonal changes in the invertebrate community of granular activated carbon filters and control technologies, *Water Res.* 51 (2014) 216-227. <https://doi.org/https://doi.org/10.1016/j.watres.2013.10.064>.
19. [19] S. Shanmuganathan, M.A.H. Johir, T.V. Nguyen, J. Kandasamy, S. Vigneswaran, Experimental evaluation of microfiltration-granular activated carbon (MF-GAC)/nano filter hybrid system in high quality water reuse, *J. Membr. Sci.* 476 (2015) 1-9. <https://doi.org/https://doi.org/10.1016/j.memsci.2014.11.009>.
20. [20] S.A. Snyder, S. Adham, A.M. Redding, F.S. Cannon, J. DeCarolis, J. Oppenheimer, E.C. Wert, Y. Yoon, Role of membranes and activated carbon in the removal of endocrine disruptors and pharmaceuticals, *Desalination* 202(1) (2007) 156-181. <https://doi.org/https://doi.org/10.1016/j.desal.2005.12.052>.
21. [21] B. Wu, F. Zamani, W. Lim, D. Liao, Y. Wang, Y. Liu, J.W. Chew, A.G. Fane, Effect of mechanical scouring by granular activated carbon (GAC) on membrane fouling mitigation, *Desalination* 403 (2017) 80-87. <https://doi.org/10.1016/j.desal.2015.12.003>.
22. [22] X. Tang, W. Pronk, J. Traber, H. Liang, G. Li, E. Morgenroth, Integrating granular activated carbon (GAC) to gravity-driven membrane (GDM) to improve its flux stabilization: Respective roles of adsorption and biodegradation by GAC, *Sci. Total Environ.* 768 (2021) 144758. <https://doi.org/https://doi.org/10.1016/j.scitotenv.2020.144758>.

23. [23] J. Xing, H. Wang, X. Cheng, X. Tang, X. Luo, J. Wang, T. Wang, G. Li, H. Liang, Application of low-dosage UV/chlorine pre-oxidation for mitigating ultrafiltration (UF) membrane fouling in natural surface water treatment, *Chem. Eng. J.* 344 (2018) 62-70. <https://doi.org/https://doi.org/10.1016/j.ccej.2018.03.052>.
24. [24] N. Moona, K.R. Murphy, M. Bondelind, O. Bergstedt, T.J.R. Pettersson, Partial renewal of granular activated carbon biofilters for improved drinking water treatment, *Environ. Sci.: Water Res. Technol.* 4(4) (2018) 529-538. <https://doi.org/10.1039/C7EW00413C>.
25. [25] J. Wang, X. Tang, Y. Liu, B. Xie, G. Li, H. Liang, Self-sustained ultrafiltration coupling vermifiltration for decentralized domestic wastewater treatment: Microbial community and mechanism, *Resour., Conserv. Recycl.* 177 (2022) 106008. <https://doi.org/https://doi.org/10.1016/j.resconrec.2021.106008>.
26. [26] Y. Jia, S.K. Khanal, H. Zhang, G.-H. Chen, H. Lu, Sulfamethoxazole degradation in anaerobic sulfate-reducing bacteria sludge system, *Water Res.* 119 (2017) 12-20. <https://doi.org/https://doi.org/10.1016/j.watres.2017.04.040>.
27. [27] W. Xue, F. Li, Q. Zhou, Degradation mechanisms of sulfamethoxazole and its induction of bacterial community changes and antibiotic resistance genes in a microbial fuel cell, *Bioresour. Technol.* 289 (2019) 121632. <https://doi.org/https://doi.org/10.1016/j.biortech.2019.121632>.

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