

Improvement of Membrane Filtration Performance for Surface Water Treatment by Combining GAC with Both Adsorption and Antimicrobial Function

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Abstract

The combined use of silver-loaded granular activated carbon (Ag/AC) and Microfiltration (MF) was evaluated for surface-water treatment to improve permeate quality and mitigate membrane fouling. Ag/AC was prepared by AgNO₃ impregnation and thermal stabilization, resulting in strong and durable antimicrobial activity ($\geq 99\%$ bacterial inactivation) while maintaining high adsorption capacity for Dissolved Organic Matter (DOM). By suppressing microbial growth and reducing biofilm precursor formation within the carbon bed, Ag/AC pretreatment decreased biodegradable organics and delivered cleaner feedwater to the MF unit. Increasing the Empty Bed Contact Time (EBCT) from 10 to 30 min significantly enhanced DOM and aromatic compound removal, lowering DOC from 1.35 to 0.50 mg/L and reducing UV₂₆₀. When coupled with MF, Ag/AC pretreatment effectively reduced membrane resistance-related substances and stabilized permeate flux compared with direct filtration of raw water. Although slight localized resistance increases were observed relative to unmodified GAC, possibly due to silver-associated deposits, the overall improvement in permeate quality and membrane operational stability was substantial. These results demonstrate that integrating Ag/AC with MF provides a practical and efficient strategy for surface-water purification, offering synergistic benefits of adsorption and antimicrobial effects for long-term fouling control in drinking water treatment systems.

Keywords

Silver-Loaded GAC, Microfiltration, Membrane Fouling, Water Quality, Adsorption, Antimicrobial Activity, Surface Water Treatment

1. Introduction

Membrane filtration is increasingly recognized as an efficient and reliable alternative to conventional sand filtration for drinking water treatment, particularly when surface water such as rivers, lakes, or reservoirs is utilized as sources [1]-[4]. Compared with sand filtration, Microfiltration (MF) and Ultrafiltration (UF) membranes offer smaller pore sizes, enabling the effective removal of fine particles, colloids, bacteria, protozoa, and even viruses [5]-[7]. In addition, membrane systems are characterized by compact design, ease of operation, and stable water quality, which can render them increasingly attractive for advanced drinking water production [8] [9].

When MF or UF is applied to surface-water treatment, membranes face two critical challenges: 1) the progressive development of membrane resistance caused by fouling and 2) the insufficient removal of Dissolved Organic Matter (DOM), which acts as a precursor for both fouling and microbial proliferation [10] [11]. DOM in surface water is a heterogeneous mixture comprising fractions with different molecular weights, chemical characteristics, and biodegradability [12]. Among these fractions, Biodegradable Dissolved Organic Matter (BDOM), including Assimilable Organic Carbon (AOC) and biopolymer-like substances, plays a critical role in promoting microbial regrowth and biofilm formation in drinking water systems [13]. In contrast, humic- and fulvic-like substances primarily contribute to organic fouling through adsorption, pore blockage, and cake layer formation rather than directly stimulating microbial proliferation. Therefore, effective control of both biodegradable and non-biodegradable DOM fractions is essential for mitigating combined organic fouling and biofouling in membrane-based surface water treatment processes [14]. Accordingly, without effective pretreatment, MF and UF alone cannot ensure stable long-term operation when treating surface water with elevated organic and microbial loads. Fouling leads to increased trans-membrane pressure and reduced permeate flux, which can ultimately shorten membrane lifespan. It can be categorized as reversible fouling, removable through backwashing, and irreversible fouling, which persists even after cleaning and results in continuous resistance accumulation. Irreversible fouling primarily results from the deposition of DOM and biofouling associated with bacterial adhesion and biofilm formation [15] [16]. The complexity of biofouling arises because bacteria can exist as suspended solids, multiply, release extracellular polymeric substances, and generate new DOM, thereby establishing a self-reinforcing fouling cycle. Consequently, without effective pretreatment, MF and UF alone cannot ensure stable long-term operation when treating surface water with high organic and microbial

loads.

To overcome these limitations, various pretreatment strategies have been implemented to reduce organic and microbial loads prior to membrane filtration. Notably, Powdered Activated Carbon (PAC) adsorption has been extensively applied for DOM removal [17]. However, practical drawbacks such as PAC abrasion of membrane surfaces, challenges with PAC regeneration, and the need for frequent PAC disposal due to exhausted adsorption capacity [18]. When combined, PAC and coagulation can generate excessive sludge and may lead to persistent color or turbidity problems and can be discarded before adsorption equilibrium is reached [19] [20]. As an alternative, Granular Activated Carbon (GAC) provides a fixed-bed configuration that minimizes sludge production and ensures stable long-term adsorption [21]. At the same time, to suppress microbial proliferation while maintaining adsorption efficiency, silver-loaded GAC (Ag/AC) has been introduced as a dual-function pretreatment medium [22]-[24]. Silver ions (Ag^+) interact with microbial DNA and proteins, disrupt cell membranes, and inhibit bacterial replication. When immobilized on GAC, silver provides antimicrobial functionality without substantially reducing adsorption capacity, thereby enabling the simultaneous regulation of organic matter and microorganisms prior to membrane filtration [25] [26]. Recently, some researchers have explored the incorporation of silver into carbonaceous adsorbents to impart antimicrobial functionality while maintaining organic matter removal capabilities. For example, silver-loaded activated carbon synthesized by chemical reduction exhibited potent antimicrobial activity against *Escherichia coli* and *Staphylococcus aureus*, but its stability under continuous water treatment conditions remains uncertain, as significant silver release was observed under static testing [27]. An industrial demonstration comparing silver-loaded activated carbon with silver zeolite-treated activated carbon reported enhanced initial disinfection but highlighted a trade-off between antimicrobial strength and silver leaching or long-term performance [28]. Similarly, silver-doped biochar prepared from waste biomass exhibited excellent dual functionality for adsorption and microbial inhibition [25], although its evaluation was limited to batch experiments and did not consider flow-through behavior or membrane-coupled operation. Nonetheless, biological growth inevitably develops within the GAC bed during extended operation, potentially contributing to the release of microbial metabolites and biofilm fragments that re-enter the treated water and accelerate membrane fouling [23] [29].

In this study, a hybrid water treatment system integrating Ag/AC pretreatment with microfiltration was employed to control microbial-related membrane fouling during drinking water production. The objectives of this study were to 1) assess the antimicrobial and adsorption performance of GAC and Ag/AC under varying Empty Bed Contact Times (EBCTs), 2) analyze the reduction of DOM and particulate matter through adsorption and coarse filtration, and 3) investigate the influence of pretreatment on membrane resistance and fouling behavior using different membrane materials (PVDF, PTFE, and MCE). This study provides a comprehensive understanding of how adsorption and antimicrobial action can act

synergistically to mitigate both membrane resistance and DOM-related fouling, serving as the two primary challenges restricting the sustainable application of membrane filtration for surface water treatment.

2. Materials and Methods

2.1. Surface Modification of GAC

Commercial coal-based and steam-activated granular activated carbon (F400, Calgon Carbon, USA) was used as the adsorbent. Silver-loaded GAC (Ag/AC) was prepared by impregnating the material in 0.1 mol/L AgNO₃ (Wako Pure Chemicals, Japan) under acidic conditions (pH ≈ 4, adjusted with HNO₃), followed by drying at 105 °C for 2 h and thermal treatment at 700 °C for 2 h to promote silver deposition and stabilization within the carbon matrix. This AgNO₃-impregnation and thermal conversion method is consistent with conventional silver-loading practices for activated carbons and has been reported to produce stable materials with combined adsorption and antimicrobial functionality [25] [26]. Unmodified F400 GAC (AC) was used as the control. As shown in **Table 1**, the BET analysis indicates that the pristine AC is predominantly microporous, with micropores accounting for the majority of the total surface area, while mesopores and macropores contribute only minor fractions. After silver loading, the total BET surface area of Ag/AC decreased from 1022.9 to 977.1 m²/g, accompanied by slight reductions in micropore, mesopore, and macropore surface areas.

Table 1. Surface properties and silver content of AC and Ag/AC determined by BET.

Sample	BET surface area (m ² /g)	Total pore volume (cm ³ /g)	Micropore area (m ² /g)	Mesopore area (m ² /g)	Macropore area (m ² /g)
AC	1022.9	0.4447	778.2	62.4	0.3
Ag/AC	977.1	0.4165	754.7	50	0.16

2.2. Column Experiments

Raw water was collected from the Nagara River (Gifu, Japan) without prior treatment. The experimental column consisted of a polyvinyl chloride tube (2.6 cm inner diameter, 30 cm height) fitted with rubber stoppers, silicone tubing, and wire mesh. A rubber stopper with a tubing thread was attached to the bottom of the tube, and a wire mesh cut to the tube's inner diameter was placed on top to prevent carbon leakage. Activated carbon was packed to a depth of 10 cm, and the column was gently tapped during packing to minimize voids. A rubber stopper with a tubing thread was attached to the top end of the tube. This procedure was repeated to construct three columns containing Ag/AC (Ag/AC1, Ag/AC2, and Ag/AC3) and three columns containing unmodified GAC (AC1, AC2, and AC3) as controls to compare both adsorption and antimicrobial performance of Ag/AC. The water flowed downward through each column. The Empty Bed Contact Time (EBCT) represents the average duration that water resides within the GAC bed, governing both adsorption and biodegradation processes. The flow rates were

controlled using peristaltic pumps (Masterflex L/S, Cole-Parmer, USA) to achieve EBCTs of 10 (AC1, Ag/AC1), 20 (AC2, Ag/AC2), and 30 (AC3, Ag/AC3) minutes. An EBCT of 10 min represents the lower range commonly adopted in full-scale GAC filters, where adsorption predominantly governs the removal of Dissolved Organic Matter (DOM). This condition enables evaluation of system performance under practical, high-throughput conditions with limited contact time. An EBCT of 20 min represents an intermediate condition that balances adsorption and emerging biological activity, corresponding to the transitional phase, in which biofilm development begins to contribute to contaminant removal. The 30-minute EBCT was chosen to investigate the effect of prolonged contact time, which promotes adsorption equilibrium and biological stabilization, thereby improving the removal efficiency of DOM and microorganisms. These EBCTs were selected to evaluate the effect of contact time on adsorption and antimicrobial performance.

2.3. Membrane Experiments

MF experiments were conducted using three types of membranes, including Polyvinylidene Fluoride (PVDF), Polytetrafluoroethylene (PTFE), and Mixed Cellulose Ester (MCE) (Advantec, Japan), each with a nominal pore size of 0.1 μm (effective filtration area 10.7 cm^2). The transmembrane pressure was maintained at 0.1 MPa throughout the experiments. To maintain uniform concentration gradients, the feed water was stirred continuously in the reservoir. Three treatment trains were employed. 1) Direct MF (Control): raw river water was directly filtered through MF. 2) GAC adsorption and MF: water was pretreated using one of the six GAC columns prior to MF. 3) GAC adsorption followed by coarse filtration and MF: effluent from the GAC columns was subjected to an additional 0.6 μm coarse filtration membrane (Advantec, Japan) to remove large, suspended solids before MF. The setup of the experimental equipment is shown in **Figure 1**.

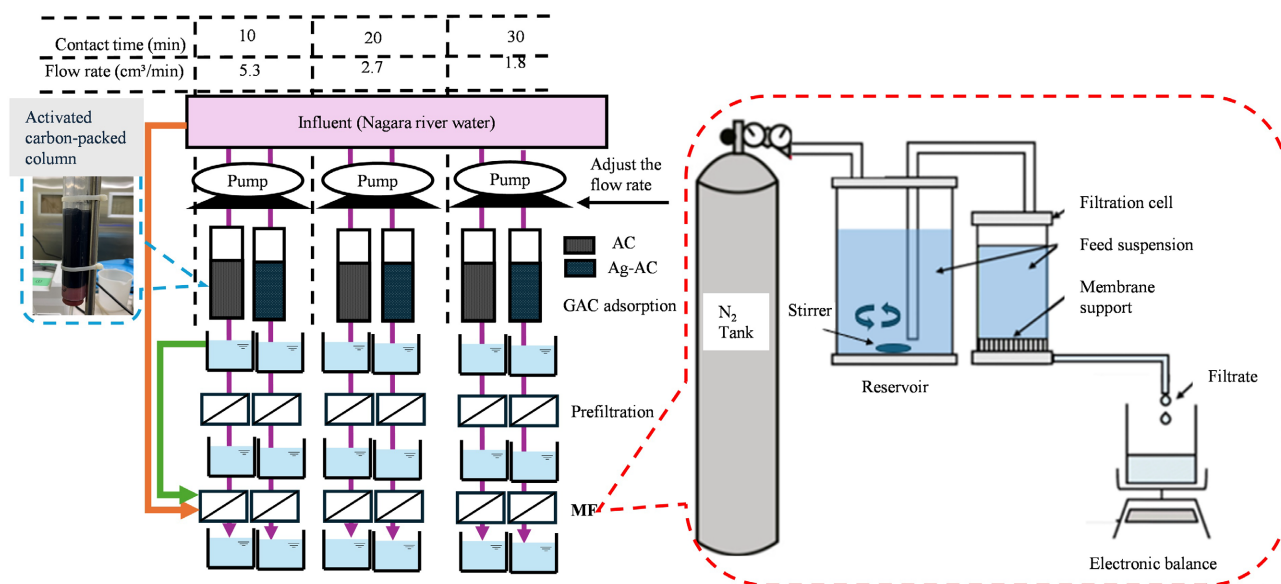


Figure 1. Experiment setup.

2.4. Analysis

Water quality parameters were measured to assess the performance under each treatment condition. Turbidity was determined by using a nephelometric turbidity meter (2100N, Hach, USA). Particle counts were obtained using a particle counter to analyze the removal of suspended solids. Dissolved Organic Carbon (DOC) was quantified using a total organic carbon analyzer (TOC-LCPH/CPN, Shimadzu Co., Kyoto, Japan). UV absorbance at 260 nm (UV260) was measured using a UV-Vis spectrophotometer (UV-2600, Shimadzu Co., Kyoto, Japan) to indicate the presence of aromatic organic compounds. The inactivation effect was assessed by comparing bacterial counts in the influent and treated water of unmodified AC and Ag/AC. General bacteria were defined as colonies capable of forming nutrient media under $35^{\circ}\text{C} \pm 1^{\circ}\text{C}$ for 48 h following APHA standard methods. Although most were non-pathogenic, higher colony counts indicated elevated microbial contamination, which could serve as a useful indicator of water quality (APHA, 2005). According to the Japanese water quality standards, the number of colonies should not exceed 100 CFU/mL. Viable bacteria were quantified using the plate culture method with standard nutrient agar composed of 5 g peptone, 2.5 g yeast extract, 1 g glucose, and 15 g agar per liter of medium. Scanning Electron Microscopy (SEM) (SU3500, Hitachi Co., Tokyo, Japan) was employed to observe fouled membranes and a clean control membrane following the protocol of Tian *et al.* [30]. Membrane fouling during filtration was quantitatively evaluated in terms of filtration resistance using the Relative Resistance Index (RRI), which represents the degree of resistance development relative to the intrinsic resistance of the clean membrane, as calculated using Equation (1) [7] [31]:

$$\text{RRI} = R_t / R_m \quad (1)$$

where R_t is the total resistance during filtration, and R_m is the clean membrane resistance. The resistance was calculated based on Darcy's law, which describes laminar flow through a porous medium by using Equation (2):

$$R = \Delta P / \mu \cdot J \quad (2)$$

where ΔP is the transmembrane pressure (0.1 MPa), μ is the water viscosity (8.9×10^{-4} Pa·s at 25°C), and J ($\text{m}^3 \cdot \text{m}^{-2} \cdot \text{h}^{-1}$) is the permeate flux which is computed based on Equation (3) [16]:

$$J = V / A \cdot T \quad (3)$$

where V (m^3) is the volume of permeate collected during the time interval T (h), and A (m^2) is the effective filtration area of the membrane (10.7 cm^2 in this study). To ensure constant water viscosity, all flux measurements were performed under steady-state conditions at 25°C .

All experiments were conducted in duplicate unless otherwise stated. Results are presented as mean values, and observed trends were interpreted qualitatively due to the limited number of replicates.

3. Results and Discussion

3.1. Antimicrobial Efficiency of AC and Ag/AC

As shown in **Figure 2**, the number of general bacteria in the raw water is 18470 CFU/ml Ag/AC, which exhibited excellent antimicrobial performance, achieving nearly complete bacterial inactivation ($\geq 99\%$) at all contact times. This demonstrated that silver loading provided strong and consistent bactericidal activity, effectively suppressing bacterial growth regardless of contact duration. The results confirmed that the incorporation of silver ions greatly enhanced the disinfection capacity of activated carbon, enabling simultaneous adsorption and antimicrobial functions. Such dual functionality could be highly beneficial in drinking water treatment, where both organic matter and microorganisms contribute to membrane fouling. In comparison, the AC samples displayed only limited antimicrobial efficiency, even when the contact time was extended (AC1 < AC2 < AC3). The efficiencies remained below 70%, indicating that adsorption alone could not effectively inactivate microorganisms. These findings demonstrate the crucial contribution of silver to microbial control and underline the superiority of Ag/AC media in suppressing biological activity and improving water quality prior to membrane filtration.

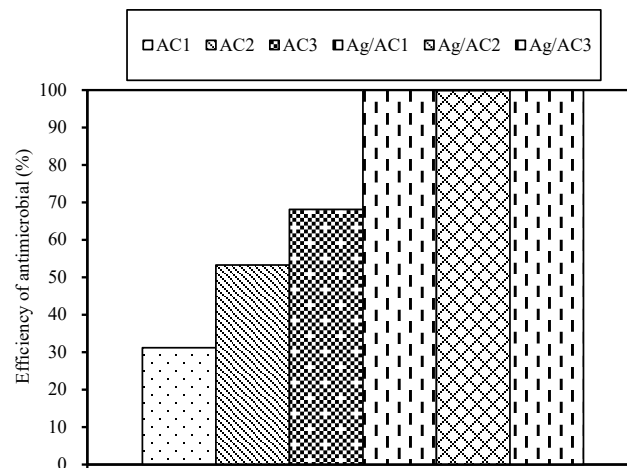


Figure 2. Antimicrobial efficiency of AC and Ag/AC.

3.2. Effect on DOM Removal Efficiency

Table 2 summarizes the physicochemical properties of the water before and after treatment by GAC adsorption and subsequent coarse filtration. The pH (7.0 - 7.4) and electrical conductivity (5.3 - 5.8 mS/m) remained nearly constant throughout all treatment steps, confirming that neither adsorption nor coarse filtration significantly altered the ionic composition of the water. In contrast, substantial reductions in turbidity and particle counts were observed after GAC adsorption and were further enhanced following 0.6 μm coarse filtration, particularly for particles larger than 1 μm . These results demonstrated the effective removal of suspended solids and colloidal matter. For DOM, both the DOC concentration and UV260

absorbance decreased markedly compared with raw water (DOC = 1.35 mg/L; UV260 = 1.72 m⁻¹). Increasing EBCT from 10 to 30 min improved the adsorption efficiency, reducing DOC to 0.55 mg·L⁻¹ for unmodified GAC and 0.50 mg·L⁻¹ for silver-loaded GAC. Coarse filtration further decreased DOC to 0.35 mg/L, confirming the complementary effect of size-based removal. The organic matter content (OM), expressed as the ratio of volatile to total suspended solids, markedly increased from 6% in the raw water to approximately 50% after adsorption by the AC column, indicating that a portion of the organic matter originated from the activated carbon column itself. In contrast, the Ag/AC column exhibited a lower OM, demonstrating that the presence of silver effectively inhibited bacterial growth and suppressed biofilm formation on the activated carbon surface, thereby exhibiting strong antimicrobial performance. Overall, extending EBCT and including coarse filtration substantially reduced both particulate and dissolved organic loads. Moreover, silver loading maintained adsorption efficiency while providing additional antimicrobial functionality, thereby improving the overall water quality prior to membrane filtration.

Table 2. Water quality before and after GAC adsorption and prefiltration.

Sample	pH	EC (mS/m)	Turbidity (NTU)	Particle number (number/mL)						OM (%) (VSS/SS)	DOC (mg/L)	UV260 (m ⁻¹)	
				0.5 - 1 mm	1 - 3 mm	3 - 5 mm	5 - 10 mm	10 - 30 mm	>30 mm				
Raw water	7.25	5.49	2.47	5.91E+05	9.36E+01	7.27E+02	1.27E+02	2.31E+01	1.60E+00	6	1.35	1.72	
Water after GAC adsorption	AC1	7.05	5.41	1.33	1.94E+05	5.78E+01	7.03E+02	3.66E+02	1.92E+02	2.00E+00	44	0.84	0.89
	AC2	7.33	5.55	1.21	7.87E+04	1.49E+02	1.29E+03	5.42E+02	2.11E+02	1.06E+01	96	0.65	0.66
	AC3	7.32	5.79	1.07	3.77E+04	3.73E+01	8.44E+02	1.40E+02	8.15E+01	8.60E+00	44	0.55	0.50
	Ag/AC1	7.24	5.35	2.28	2.52E+05	2.06E+01	2.47E+02	4.19E+01	2.07E+01	3.80E+00	78	0.72	0.86
	Ag/AC2	7.40	5.43	1.93	1.77E+05	7.98E+01	6.53E+02	3.39E+02	1.68E+02	1.60E+00	88	0.62	0.57
	Ag/AC3	7.40	5.53	1.78	7.43E+04	2.31E+01	2.23E+02	6.07E+01	8.53E+00	1.80E+00	13	0.50	0.47
Water after GAC adsorption and coarse filtration	AC1	6.82	5.46	1.17	1.07E+05	9.16E+01	3.27E+02	1.04E+02	4.93E+01	3.03E+01	33	0.73	0.86
	AC2	7.05	5.45	0.86	7.15E+04	3.73E+01	1.57E+02	5.92E+01	1.93E+01	1.67E+00	20	0.51	0.52
	AC3	6.98	5.66	0.81	1.01E+04	1.06E+01	9.53E+01	1.91E+01	1.69E+01	1.67E+00	67	0.35	0.40
	Ag/AC1	6.96	5.50	1.16	1.57E+05	1.06E+01	1.30E+02	5.19E+01	3.07E+01	4.13E+00	34	0.56	0.67
	Ag/AC2	7.06	5.51	0.92	1.40E+05	3.18E+01	2.40E+02	4.70E+01	2.71E+01	3.93E+00	38	0.35	0.43
	Ag/AC3	7.15	5.50	0.76	6.44E+04	1.93E+01	1.50E+02	4.36E+01	4.05E+01	4.27E+00	20	0.31	0.38

Note: Particle numbers are expressed in scientific notation (e.g., 1.27E+02 represents 1.27×10^2 number/mL).

The variations in DOC concentration throughout the treatment process are shown in **Figure 3(a)**. A significant reduction was observed after GAC adsorption, with the removal efficiency increasing with EBCT, confirming that the extended contact enhanced DOM adsorption on the carbon surface. Notably, Ag/AC consistently achieved a slightly higher DOC removal than AC at each EBCT, indicating a synergistic interaction between adsorption and antimicrobial activity. The presence of silver ions suppressed microbial activity within the carbon bed, pre-

vented biofilm formation, maintained active adsorption sites, and enhanced the overall DOM removal efficiency.

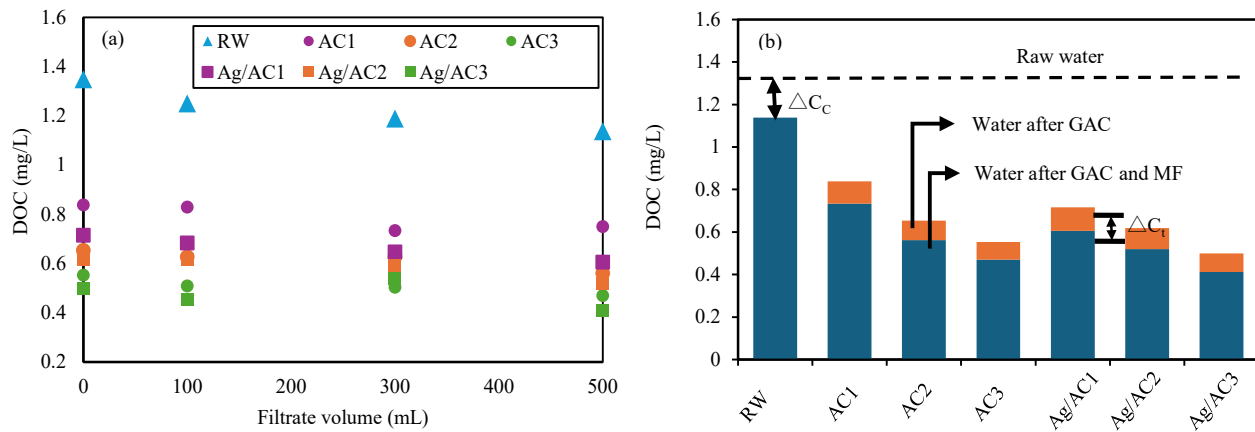


Figure 3. Variation in DOC concentration with cumulative filtrate volume using PVDF membrane (a); DOC concentration in water after MF treatment of raw water and water after pretreatment of GAC (b) (RW: Filtration using raw water without preadsorption).

Table 3. Treated Water quality after MF treatment of raw water and water after pretreatment of GAC, and reduction efficiency for DOM-related fouling.

Sample	pH	EC (mS/m)	Turbidity (NTU)	Particle number (number/mL)		DOC (mg/L)	UV260 (m ⁻¹)	Reduction efficiency (%)	
				0.5 - 1 mm	>1 mm			DOC	UV260
Raw water	7.63	5.75	0.10	9.54E+00	8.80E+00	1.14	1.54	—	—
PVDF AC1	7.88	6.03	0.10	6.40E+00	5.22E+00	0.73	0.82	49	42
PVDF AC2	7.97	5.87	0.10	3.60E+00	1.31E+00	0.56	0.60	44	37
PVDF AC3	8.01	6.09	0.14	3.40E+00	3.17E+00	0.47	0.44	40	34
PVDF Ag/AC1	7.87	5.79	0.10	4.20E+00	—	0.61	0.78	52	43
PVDF Ag/AC2	7.90	5.96	0.10	3.53E+00	7.42E+00	0.52	0.50	47	40
PVDF Ag/AC3	8.04	5.96	0.12	3.07E+00	1.75E+00	0.41	0.40	42	36
Raw water	7.62	5.85	0.10	2.53E+00	8.24E+00	1.14	1.58	—	—
PTFE AC1	7.56	6.02	0.10	1.25E+00	—	0.73	0.83	53	45
PTFE AC2	7.89	5.68	0.08	4.52E+00	1.31E+00	0.55	0.61	51	37
PTFE AC3	7.98	5.96	0.14	2.56E+00	2.71E+00	0.45	0.44	51	34
PTFE Ag/AC1	7.54	5.77	0.10	5.45E+00	—	0.60	0.78	57	55
PTFE Ag/AC2	7.59	5.36	0.10	1.25E+00	6.88E+00	0.50	0.51	57	40
PTFE Ag/AC3	7.58	5.94	0.10	1.24E+00	1.16E+00	0.39	0.40	54	37
Raw water	7.81	6.05	0.12	4.06E+00	9.53E+00	1.08	1.52	—	—
MCE AC1	7.65	5.92	0.09	1.05E+00	8.73E+00	0.75	0.86	31	21
MCE AC2	7.68	5.97	0.09	4.60E+00	3.27E+00	0.59	0.63	24	18
MCE AC3	7.66	6.03	0.08	6.27E+00	2.12E+00	0.50	0.46	19	10
MCE Ag/AC1	7.56	5.90	0.10	8.41E+00	—	0.62	0.82	35	23
MCE Ag/AC2	7.86	5.89	0.07	4.83E+00	7.96E+00	0.55	0.54	26	18
MCE Ag/AC3	7.58	6.16	0.07	3.39E+00	—	0.44	0.41	23	12

Note: Particle numbers are expressed in scientific notation (e.g., 9.54E+00 represents 9.27×10^0 number/mL).

Table 4. Treated Water quality after MF treatment of raw water and water after pretreatment of GAC and prefiltration, and reduction efficiency for DOM-related fouling.

Sample	pH	EC (mS/m)	Turbidity (NTU)	Particle number (number/mL)		DOC (mg/L)	UV260 (m ⁻¹)	Reduction efficiency (%)		
				0.5 - 1 mm	>1 mm			DOC	UV260	
Raw water	7.63	5.75	0.10	9.54E+00	8.80E+00	1.14	1.54	—	—	
AC1	8.07	5.95	0.05	2.60E+00	8.60E+00	0.62	0.79	48	39	
AC2	8.13	5.90	0.04	2.14E+00	3.17E+00	0.42	0.46	43	36	
PVDF	AC3	8.05	6.17	0.02	1.07E+00	4.49E+00	0.27	0.34	39	33
	Ag/AC1	8.10	5.95	0.05	2.40E+00	—	0.45	0.60	51	41
	Ag/AC2	8.16	5.96	0.02	1.63E+00	2.62E+00	0.26	0.37	45	37
	Ag/AC3	8.04	6.04	0.10	1.00E+00	1.36E+00	0.23	0.32	42	34
Raw water	7.62	5.85	0.10	2.53E+00	8.24E+00	1.14	—	—	—	
AC1	8.04	5.66	0.05	5.45E+00	8.15E+00	0.63	0.79	49	50	
AC2	8.05	5.94	0.06	1.23E+00	2.61E+00	0.42	0.46	41	42	
PTFE	AC3	8.16	5.97	0.10	5.35E+00	4.49E+00	0.29	0.35	30	34
	Ag/AC1	8.15	5.94	0.08	5.25E+00	4.95E+00	0.45	0.59	53	57
	Ag/AC2	8.03	5.87	0.02	1.25E+00	—	0.26	0.36	45	50
	Ag/AC3	8.15	5.89	0.05	5.35E+00	1.83E+00	0.24	0.33	35	39
Raw water	7.81	6.05	0.12	4.06E+00	9.53E+00	1.08	—	—	—	
AC1	8.04	5.87	0.12	8.27E-01	9.14E+00	0.65	0.83	29	18	
AC2	8.15	5.84	0.09	6.18E+00	9.26E+00	0.43	0.49	27	17	
MCE	AC3	8.04	5.79	0.09	1.84E+00	8.12E+00	0.27	0.38	30	12
	Ag/AC1	8.02	5.94	0.09	5.69E+00	2.26E+00	0.47	0.62	32	31
	Ag/AC2	8.13	5.98	0.07	4.19E+00	3.08E+00	0.27	0.40	31	39
	Ag/AC3	8.06	6.01	0.05	5.52E-01	—	0.23	0.35	31	14

Note: Particle numbers are expressed in scientific notation (e.g., 9.54E+00 represents 1.27×10^0 number/mL).

Table 3 and **Table 4** summarize the variations in multiple water quality parameters, including turbidity, DOC, UV260, and particle counts, measured after MF under different pretreatment conditions. The reduction efficiency (E) determined as the ratio between the difference in concentrations before and after MF (ΔC_t) and the difference between raw water before and after MF (ΔC_c) (**Figure 3(b)**) represents the proportion of DOM retained on the membrane. Smaller E values indicate lower fouling potential. Overall, GAC pretreatment significantly improved the permeate water quality compared with direct MF of raw water. As shown in **Table 2**, the turbidity, DOC, UV260, and particle counts all decreased with increasing EBCT, indicating that longer adsorption enhanced the removal of both particulate and dissolved organic matter. Ag/AC exhibited slightly greater reductions in these parameters than AC, reflecting a synergistic interaction between adsorption and antimicrobial activity, as silver ions suppressed microbial

growth within the carbon bed and reduced the formation of biodegradable organics. Among the three membranes, PVDF consistently exhibited the lowest turbidity and DOC values. Among the three membrane materials evaluated, the PVDF membrane consistently exhibited lower turbidity and DOC values in the permeate. This behavior can be attributed to the relatively higher hydrophilicity and more uniform pore structure of PVDF membranes, which reduce hydrophobic interactions with natural organic matter and mitigate pore blockage compared with PTFE and MCE membranes. Similar material-dependent fouling behavior has been reported in previous studies on low-pressure membrane filtration of surface water [3] [4]. When coarse filtration was included (Table 3), further improvements in water quality were observed across all membranes, as the fine particles and colloids remaining after GAC adsorption were effectively removed. The combination of Ag/AC pretreatment and coarse filtration produced the cleanest effluent, confirming that silver loading and coarse filtration cooperatively enhanced both the physical and biological removal efficiencies. Parameter E further substantiates these findings. Under Ag/AC conditions, E values were slightly higher than those with unmodified GAC, suggesting that additional resistance-related substances accumulated on the membrane surface, which could be due to the deposition of fine silver particles or silver-associated organic complexes. Nevertheless, the permeate quality under Ag/AC conditions remained superior as the overall removal of organics outweighed this slight increase. With increasing EBCT, the E values gradually decreased, confirming that the extended adsorption contact effectively reduced the transport of residual organics to the membrane. A similar trend was observed when coarse filtration was applied, further lowering turbidity, DOC, and UV260 while maintaining low E values. These results demonstrate that although silver loading may slightly increase the retained matter on the membrane, its combined adsorption and antimicrobial functions, particularly when supported by coarse filtration, greatly improve the water quality and reduce the fouling potential. Although silver loading effectively suppresses microbial activity, the potential deposition of silver-associated species on the membrane surface may contribute to localized resistance development. Silver nanoparticles immobilized on substrates have been shown to provide sustained antimicrobial activity with minimal leaching when strongly bound to the support structure, although surface deposition of silver particulates can alter local fouling resistance [32]. Nevertheless, the overall fouling mitigation achieved through reduced microbial growth and lower biodegradable organic loading outweighed this minor resistance increase, consistent with the enhanced antibacterial performance observed for nano-silver functionalized carbon-based adsorbents [33]. Silver leaching was not quantified in the present study. However, previous studies have reported minimal silver release from silver-loaded activated carbon materials under appropriate preparation and conditioning conditions, suggesting that properly stabilized Ag/AC composites can exhibit low effluent silver concentrations in continuous operation [34]. Future work will focus on quantifying effluent silver concentrations and evaluating long-term antimicrobial durability to ensure compliance with drinking

water safety standards.

3.3. Effect on Membrane Resistance Control

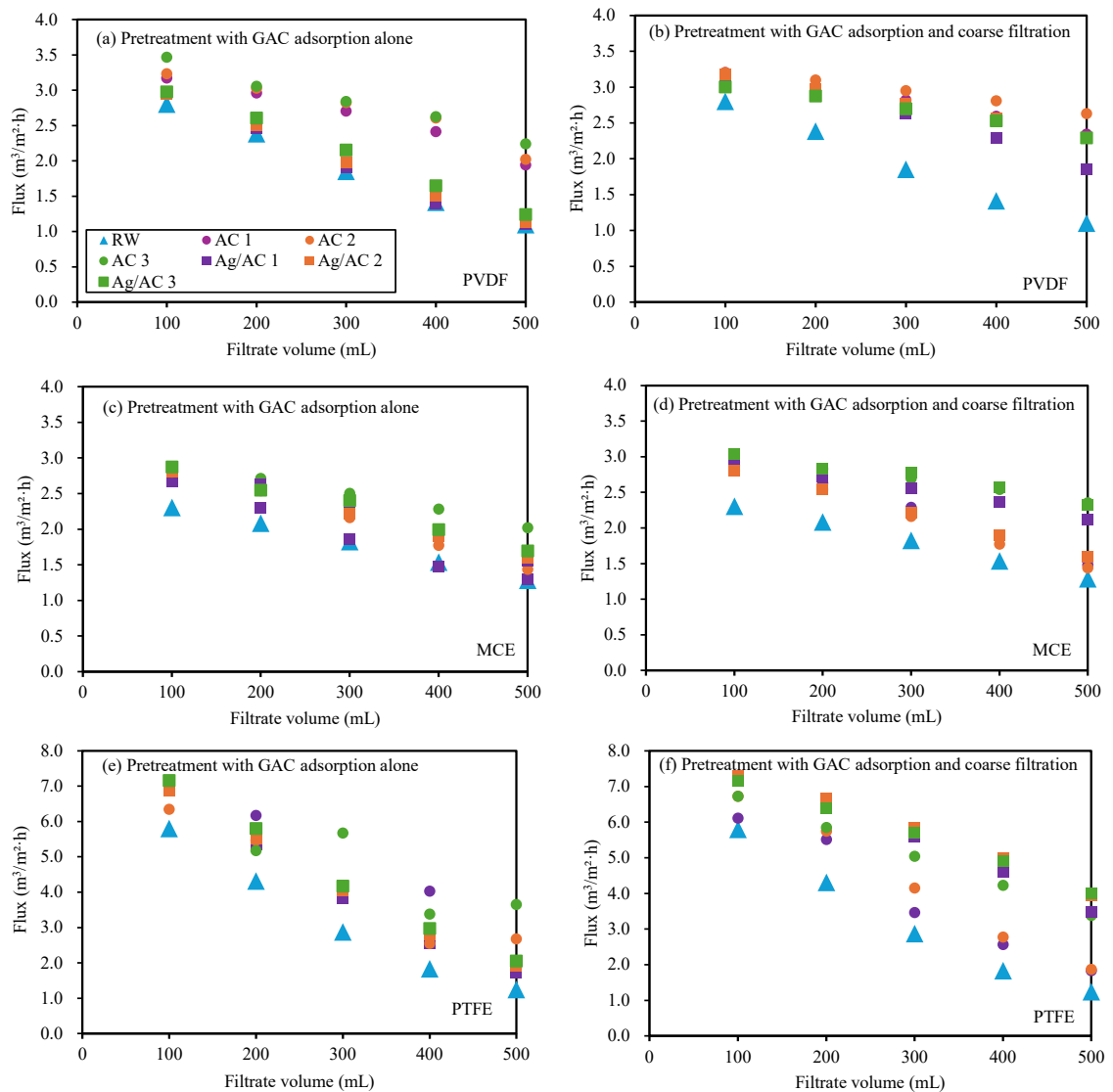


Figure 4. Variation of flux with cumulative filtrate volume after MF treatment of raw water and GAC adsorption (a, c, e), and water after GAC adsorption followed by coarse filtration (b, d, f), using 3 types of membranes.

Figure 4 illustrates the variation in permeate flux with cumulative filtrate volume under different pretreatment conditions using different membranes. As shown in **Figure 4(a)**, **Figure 4(c)**, **Figure 4(e)**, during the process in which raw water was pretreated by GAC adsorption followed by MF using a PVDF membrane, the flux decreased more gradually with increasing EBCT. Compared with the 70% flux decline observed during direct MF of raw water, the flux decline reduction was only approximately 40% at an EBCT of 30 min. This indicated that longer adsorption contacts effectively reduced the concentration of dissolved and particulate foulants entering the membrane. The Ag/AC maintained comparable or slightly

higher initial fluxes but exhibited a marginally faster decline at later stages. This suggested that although silver imparted strong antimicrobial functionality, fine silver particles or silver-associated organic complexes may have accumulated on the membrane surface, contributing slightly to additional resistance. When a coarse filtration step (0.6 μm) was included prior to MF (**Figure 4(b)**, **Figure 4(d)**, **Figure 4(f)**), the overall flux decline was further mitigated by all EBCT conditions. Coarse filtration effectively removed the residual fine particles and colloids after GAC adsorption, thereby minimizing pore blockage and surface deposition. The combination of Ag/AC pretreatment and coarse filtration produced the most stable flux profiles, demonstrating that the synergistic effects of adsorption, antimicrobial action, and particle removal effectively suppressed irreversible fouling. Similar flux trends were also observed with PTFE and MCE membranes, confirming that the beneficial effects of GAC pretreatment, silver loading, and coarse filtration on fouling mitigation were consistent across different membrane materials. Although the absolute flux values varied with membrane hydrophobicity and pore structure, the overall trend of improved flux stability with longer EBCT and with the inclusion of coarse filtration remained the same.

Table 5. Resistance Relative Index (RRI) and R (%) under different pretreatment and membrane conditions.

Sample	PVDF		PTFE		MCE		
	RRI	R (%)	RRI	R (%)	RRI	R (%)	
Raw water	2.27	—	4.72	—	1.76	—	
Water after GAC adsorption	AC 1	0.79	65	3.09	35	0.19	89
	AC 2	0.71	69	2.01	57	1.03	42
	AC 3	0.47	79	1.16	76	1.44	19
	Ag/AC 1	2.24	1	3.51	26	0.81	54
	Ag/AC 2	2.02	11	3.05	35	0.80	55
	Ag/AC 3	1.70	25	2.70	43	0.64	64
Water after GAC adsorption and coarse filtration	AC 1	0.52	77	1.27	73	0.07	96
	AC 2	0.37	84	1.12	76	0.69	61
	AC 3	0.30	87	0.97	79	0.97	45
	Ag/AC 1	0.89	61	1.34	72	0.39	78
	Ag/AC 2	0.58	74	1.14	76	0.29	83
	Ag/AC 3	0.49	78	1.05	78	0.24	86

Table 5 presents the Resistance Relative Index (RRI) and R (%) for different pretreatment and membrane conditions. R (%) represents the ratio of the RRI value for pretreated water (including GAC adsorption or GAC adsorption followed by coarse filtration) to that for the direct MF of raw water, expressed as a percentage. A smaller R (%) denotes a smaller reduction in the total filtration resistance, implying that a greater fraction of resistance remained on the membrane and thus more severe fouling occurred. Overall, GAC pretreatment significantly

reduced membrane fouling compared with the direct MF of raw water. As EBCT increased from 10 to 30 min, RRI decreased steadily. Notably, at an EBCT of 30 min, the RRI decreased by 79%, indicating that a longer adsorption contact effectively removed both dissolved and particulate organic matter and alleviated fouling. However, under Ag/AC conditions, the R (%) values were slightly smaller than those of the unmodified GAC, showing only around a 30% decrease. Although silver loading suppresses microbial activity, fine silver particles or Ag-organic complexes may be deposited on the membrane surface, resulting in slightly higher resistance. Nevertheless, the permeate quality under Ag/AC conditions remained superior, reflecting the effective control of microbial growth and improved water quality despite a minor increase in resistance. When a coarse filtration step (0.6 μm) was added before MF, the RRI values decreased further and R (%) remained lower for all EBCTs, demonstrating that the coarse filtration effectively removed the fine particles and colloids that persisted after GAC adsorption.

These results confirmed that while silver loading could slightly increase local membrane resistance, its antimicrobial effect combined with the additional benefit of coarse filtration substantially enhanced the overall fouling control efficiency, providing a balanced improvement in both water quality and membrane performance. The introduction of a coarse filtration step (0.6 μm) inevitably adds an additional unit operation to the treatment train; however, its operational implications are relatively minor compared with those of the downstream MF membrane. Due to its larger pore size, the coarse filter primarily retains suspended solids and large particulate matter and is therefore less susceptible to irreversible fouling caused by dissolved organic matter or microbial attachment. In practice, fouling of the coarse filter is dominated by surface particle accumulation and can be effectively managed through periodic physical replacement rather than chemical cleaning. From an operational perspective, the coarse filter serves as a sacrificial and low-cost protective barrier that reduces particulate and microbial loading to the MF membrane. Although it may require periodic replacement, its replacement frequency is expected to be significantly lower than the cleaning frequency of MF membranes operating without such pretreatment. Consequently, the inclusion of the coarse filtration step contributes to reduced membrane fouling rates, extended membrane lifespan, and lower overall maintenance demands of the MF system.

3.4. SEM Observation of Fouled Membranes

SEM images (**Figure 5**) of fouled membrane surfaces after filtration with different pretreatment conditions: (a) AC1 pretreatment and (b) Ag/AC1 pretreatment shown in **Figure 5**. The membrane surface after AC pretreatment was extensively covered by rod-shaped microbial cells and organic deposits, forming a relatively dense fouling layer. In contrast, the membrane surface after Ag/AC pretreatment exhibited a markedly reduced presence of microbial cells and lacked continuous biofilm-like structures. Only sporadic granular deposits were observed, suggesting effective suppression of biologically related fouling under Ag/AC pretreatment.

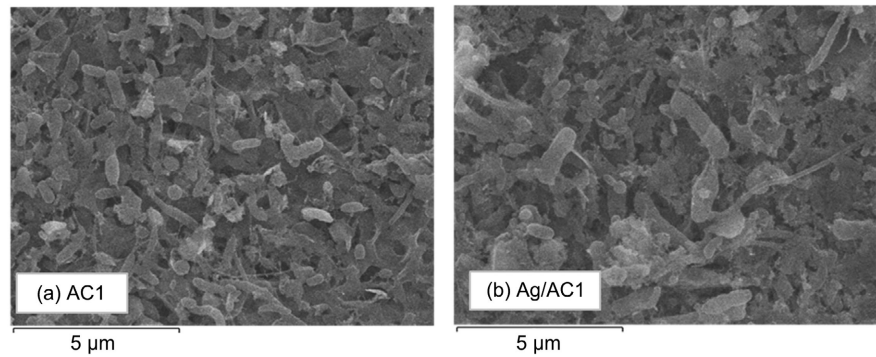


Figure 5. SEM images of fouled membrane surfaces after filtration with different pretreatment conditions: (a) AC1 pretreatment and (b) Ag/AC1 pretreatment.

4. Summary

This study evaluated the combined application of GAC with both antimicrobial and adsorption functions and MF for drinking water treatment using surface water as the source.

The results demonstrated that integrating Ag/AC pretreatment with MF effectively improved both water quality and membrane fouling control through the synergistic actions of adsorption and antimicrobial activity. Silver loading imparted strong and durable antimicrobial properties to GAC, achieving nearly complete bacterial inactivation ($\geq 99\%$) while preserving its adsorption capacity for organic matter. By suppressing microbial activity and biofilm precursor formation within the carbon bed, Ag/AC reduced the release of biodegradable organic compounds and provided cleaner feedwater for membrane filtration.

The combined action of adsorption and antimicrobial activity enhanced the removal of both particulate and dissolved organic matter, as evidenced by significant reductions in turbidity, DOC, and UV₂₆₀. When applied as a pretreatment prior to MF, Ag/AC substantially improved the permeate quality and stabilized the filtration performance. The flux decline and resistance development were greatly mitigated compared with the direct filtration of raw water, confirming that the reduction of microbial and organic foulants at the pretreatment stage effectively lessened fouling on the membrane.

Although minor increases in localized resistance were observed under Ag/AC conditions, likely owing to the limited deposition of silver-associated species, the overall benefits of water quality and membrane stability clearly outweighed these effects. The remaining challenges include monitoring potential Ag leaching and further evaluating long-term antimicrobial durability and adsorption efficiency. Overall, this approach offers valuable implications for upgrading existing membrane-based drinking water systems and promoting sustainable operation with reduced maintenance frequency and improved efficiency.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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