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Submerged Membrane System with Biofilter as a Treatment to Rainwater

N. Areerachakul · M. Kitiphatmontree · J. Kandasamy · B. Kus · C. Duangduen · S. Pivsa-Art · S. Vigneswaran

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Abstract Rainwater has been used as drinking water in Thailand for centuries especially in the rural parts and is accepted as an important water resource. From past to present, the quality of rainwater has changed with the landuse of the landscape, and its water quality is influenced by a diverse range of conditions such as the management of pollutant sources, the catchment condition, wind and meteorological conditions, and the location of rainwater collection points. In this study, the quality of rainwater collected off roofs at several locations was examined. Granular activated carbon (GAC) filtration was used as a pretreatment to microfiltration (MF) to remove the dissolved organic matter (DOC). After an initial adsorption period, the biofilm that formed on the GAC (biofilter) was found to remove DOC by up to 40%, 35%, and 15% for bed filter depths of 15, 10, and 5 cm, respectively. Biofilters also removed nitrate and phosphate by more than 80% and 35%. The hollow fiber membrane microfiltration with pore size of 0.1 μm was used to treat the effluent from biofiltration to remove the microorganisms/pathogens in the rainwater. Although there was no significant additional removal of DOC by MF, the biofilter removed all microorganisms. The use of biofilters as pretreatment to MF/UF could remove a higher amount of DOC, remove microorganisms, increase the membrane treatment efficiency, and reduce membrane fouling.

Keywords Rainwater · Characterization · Biofilters · GAC · Membrane filtration

N. Areerachakul · M. Kitiphatmontree · C. Duangduen · S. Pivsa-Art
Faculty of Science, Rajamangala University of Technology, Bangkok, Thanyaburi, Thailand

J. Kandasamy · B. Kus · S. Vigneswaran (✉)
Faculty of Engineering and IT, University of Technology, Broadway,
Sydney, NSW 2007, Australia
e-mail: s.vigneswaran@uts.edu.au

1 Introduction

Rainwater has been used as drinking water in Thailand for centuries especially in the rural parts and is accepted as an important water resource. Roofs of households and buildings are the main catchment area to harvest rainwater for consumption. Thus, rainwater quality is liable to be contaminated from bird feces, microorganisms, dust, particulates from urban pollution, wind blow dust, pesticides, herbicides, and dissolved organic gases (CO_2, NO_x, and SO_x) from industries, vehicles, etc.

Yeo et al. (2006) studied a reuse system using membrane process treating rainwater runoff from an urban parking area in Korea, which contained non-point pollutants. The rainwater reuse system consisted of a pre-filter, membrane filter, and disinfection. Hollow fiber membrane having pore size of 0.4 μm made of polyvinylidne fluoride (PVDF) was used in this system because of its strength and ability in providing a stable flux. The treated water met all the parameters of the guideline values regulated by the Korean standard for reclaimed water. Turbidity was less than 0.3 nephelometric turbidity units (NTU) in the final effluent. COD concentration decreased from 23.0 to 13.1 mg/L and BOD_5 decreased from 5.3 to 1.7 mg/L after treatment by the pre-filter and membrane process. *Escherichia coli* was completely removed by this system (Yeo et al. 2006). However, membrane fouling was the major obstacle.

Heavy metals have recently become a concern as their concentration in rainwater tanks was found to exceed the recommended levels and therefore makes it unsuitable for human consumption (Magyar et al. 2007, 2008; Han and Mun 2007; Han et al. 2006). Rainwater storage tanks also accumulate contaminants and sediments that settle to the bottom of the tank.

Changes in pH may also occur in rainwater collected in tanks. Han and Mun (2007) and Han et al. (2006) reported the results of monitoring rainwater quality, such as pH, turbidity, and metals, for a year, in the rainwater harvesting system at student dormitories at the Seoul National University, Korea. The pH of stored water changed to neutral over time, and turbidity and metal concentrations reduced through sedimentation over time. The pH of roof runoff and stored rainwater ranged from 6.5 to 9.0 and 6.8 to 8.4, respectively. It was weakly alkali but neutralized naturally in the storage tank. The turbidity of the stored rainwater showed a constant range of 1.29–2.35 NTU and metals were well within the Korean standards for drinking water.

This study analyzed the water quality in rainwater collected at representative locations in Rajamangala University of Technology, Thanyaburi, Thailand in PVC tanks and in Ayudhaya Province, Thailand in clay rain jars. Water quality was characterized in terms of physical, chemical, and organic parameters and compared against drinking water standards. The other objective of this study was to investigate the performance of biologically active granular activated carbon (biofilters) as a pretreatment to microfiltration (MF) in terms of dissolved organic matter (DOC) removal and membrane fouling reduction.

2 Experimental Methodology

2.1 Rainwater

The rainwater samples used in these experiments were collected from three concrete roofs in PVC tanks at the Rajamangala University of Technology (RMUTT), Thanyaburi, Thailand and in clay jars at five locations in the Ayudhaya Province (Fig. 1). The catchment area of the roofs had no noticeable leaves or debris in the guttering, and none had first-flush systems installed. First-flush systems divert the first part of the rainwater runoff from the roof before it can reach the tank. However, birds were present, which may have contaminated the rainwater by their dropping.

The pH of the rainwater samples were an average of 6.3 at 30°C, and the conductivity was 78 μs/cm at 30°C.

2.2 Biofilters

Biological adsorption experiments with granular activated carbon (GAC) were conducted in fixed bed columns. The physical properties of the GAC are shown in Table 1. The GAC was washed with distilled water then dried at 103°C and desiccated before use.

The experimental setup is shown in Fig. 2. The experiments were conducted using transparent acrylic filter columns with dimensions of 2 cm in diameter and 150 cm in length. The column had outlet pipes along its length and at the bottom of the column. The GAC was packed into the column to the required

Fig. 1 Rainwater Jar in Ayudhaya province

Table 1 Physical properties of GAC

Specification	Estimated value
Iodine number, mg/(g min)	800
Nominal size, m	3×10^{-4}
Maximum moisture content	5%
Bulk density, kg/m^3	748
BET surface area, m^2/g	748

depth. The columns were operated in the downflow mode. Feed water was pumped from a water tank to the top of the columns and passed through the filter bed. An overflow outlet was placed above the filter bed to maintain a constant head above the GAC filter bed. Effluent samples were collected from the bottom of the column for analysis. Experiments were conducted with filtration velocity of 4 m/h with different bed depths of 5, 10, and 15 cm. The filters were backwashed for a period of 5 min once in 2 days to control excessive biofilm growth.

2.3 Hollow Fiber MF Membrane

The schematic diagram of the membrane filtration system is shown in Fig. 3. Short term (6 h) experiments were conducted with rainwater. The hollow fiber membrane (PVDF of 0.1 μm, Kolon membrane) was vertically submerged directly into a 10-L tank (Fig. 3). The membrane length was 48.5 cm with a

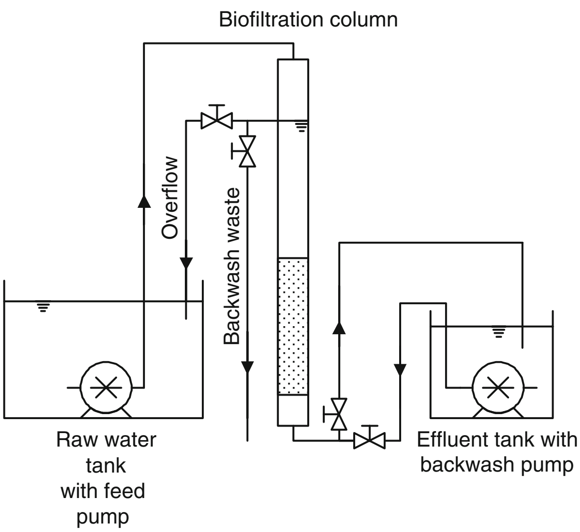


Fig. 2 Biofilter experimental setup

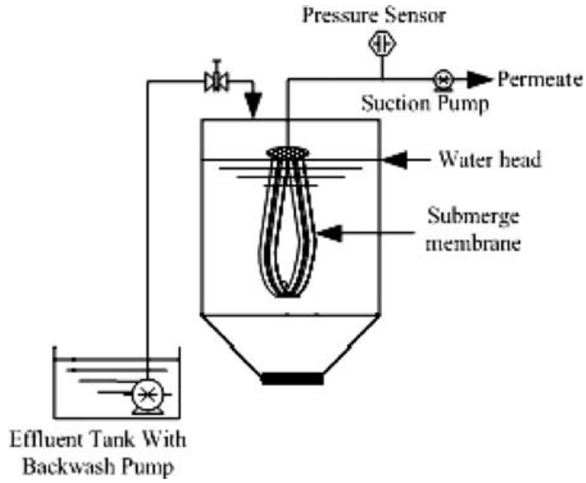


Fig. 3 Schematic of submerge membrane setup

radius of 2 mm. The combined surface area of the hollow fiber membrane was 0.030486 m^2. Constant flux experiments were conducted, and the transmembrane pressure was measured by a pressure sensor. The microfiltration unit was operated at 8 L/m^2 h.

2.4 DOC

DOC was measured using a carbon analyser (TOC-V, Shimadzu, Japan). All samples were filtered through the 0.45 μm membrane prior to the DOC measurement.

2.5 Water Quality Analysis

Detailed laboratory analyses were carried out on the eight rainwater storages to determine individual pollutants that exist in the rainwater tanks. At each location, 10 samples were taken at different times. The pollutants analyzed were heavy metals, mineral salts, nitrate, phosphate, sulfate, carbonate, total suspended solids, pH, conductivity, hardness, and turbidity. The testing methods are summarized in Table 2.

3 Results and Discussion

3.1 Characterization of Rainwater

From Table 3, the results of rainwater characterization showed that the rainwater in Ayudhaya has better

Table 2 Water quality parameters and measurement methods (Eaton et al. 2005)

Parameter	Measurement method
Heavy metals (aluminum, arsenic, cadmium, chromium, copper, iron, lead, manganese, mercury, nickel, selenium, silver, and zinc)	APHA 3120 ICPMS—inductively coupled plasma–mass spectrometry
Chloride	APHA 4500-CL-G—mercuric thiocyanate flow injection analysis
Nitrate	APHA 4500 NO3-F—automated cadmium-reduction method
Mineral salts (calcium, magnesium, potassium, sodium, and sulfate)	APHA 3120 ICPOES—inductively coupled plasma–optical emission spectrometry
pH	APHA 4500-H+—electronic method
Conductivity	APHA 2510-B—laboratory method
Water hardness	Calcium and magnesium calculation
Turbidity	APHA 2130—nephelometric method
Total suspended solids	GFC equiv. filter-APHA 2540-D—total suspended solids dried at 103–105°C
Total dissolved salts	Calculation using EC×680
Bicarbonates	Total alkalinity–APHA 2320–titration method

APHA American Public Health Association

Table 3 Rainwater characterization in various locations at Rajamangala University of Technology Thanyaburi (RMUTT) and at Ayudhaya province

Parameter	AWDG (2004)	Ayudhaya[a]	RMUTT[b]
pH	6.5–8.5	6.4	6.7
Conductivity (EC; dS/m)	<0.8	0.082	0.78
Total dissolved salts (mg/L)		55.31	160
Total suspended solids (mg/L)		400	428
Turbidity (NTU)	<5	5.07	42
Water hardness (mg/L $CaCO_3$ equivalent)	<200	47	59
Nitrate (mg/L N)	<50	14.1	18.6
Chloride (mg/L)	<400	1.45	1.35
Sulfate (mg/L)	<400	3.24	5.8
Phosphate (mg/L)		0.86	1.5
Calcium (mg/L)		10.30	21.1
Copper (mg/L)	<2	0.03	0.19
Iron (mg/L)	<0.3	0.54	0.875
Manganese (mg/L)	<0.1	0.001	0.006
Lead (mg/L)	<0.01	0.017	0.174
Zinc (mg/L)	<3	0.15	0.19
Arsenic (mg/L)		ND	ND
Cadmium (mg/L)		ND	ND
Total coliform (MPN/100 mL)	<2.2	6.8	≥1,000
Fecal coliform (MPN/100 mL)	<2.2	6.8	920
E. coli (MPN/100 mL)	ND	2	20
DOC		2.1	3.3

ND non-detectable

[a] Average of 10 samples at each of three locations at Ayudhaya

[b] Average of 10 samples at each of five locations at RMUTT

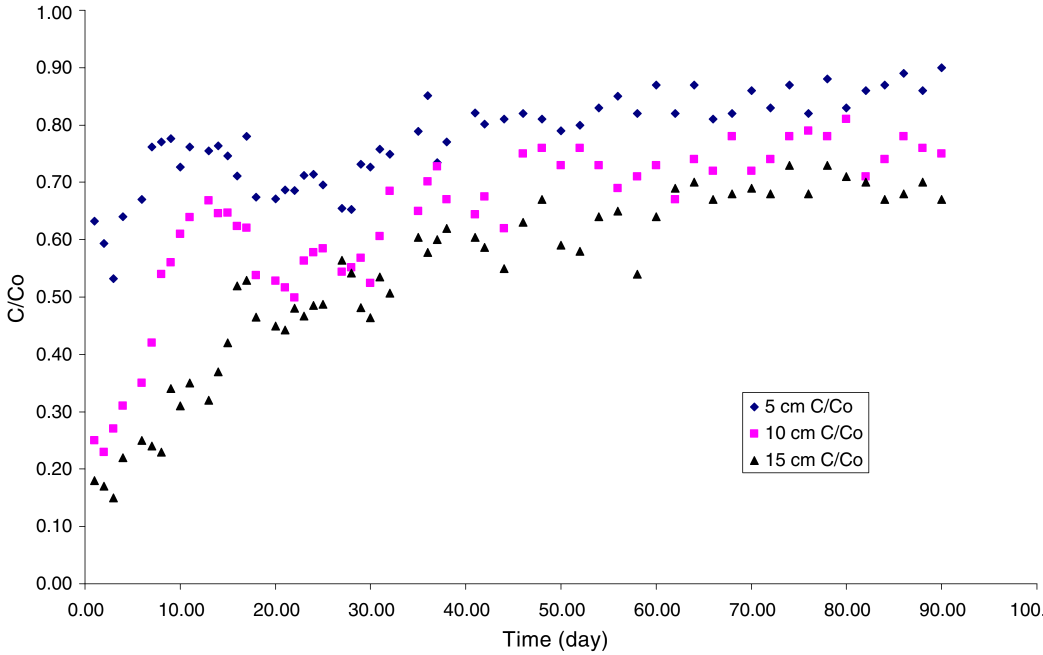


Fig. 4 DOC removal with different BAC bed depth 5, 10, and 15 cm, respectively, with V=4 m/h (where C and C_o are the effluent and influent DOC concentrations)

quality of rainwater than in RMUTT (in terms of DOC, total coliform, fecal coliform, heavy metal, and mineral). This is because of the heavier pollution in the more urban area of Thanyaburi where RMUTT is located compared to Ayudhaya, which is in the provinces and less urbanized. The results of testing show that the quality of the water in Ayudhaya meets many of the parametric standards specified in ADWS (2004). The concentrations of heavy metals were also

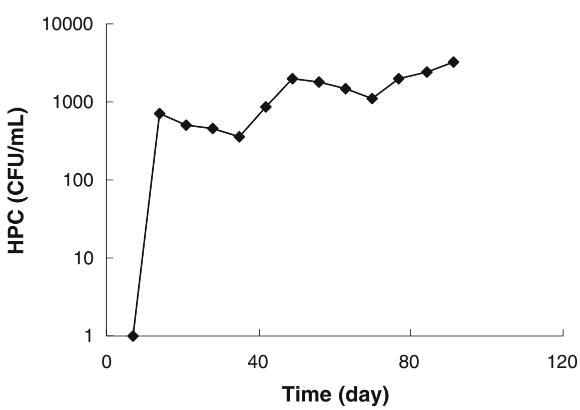


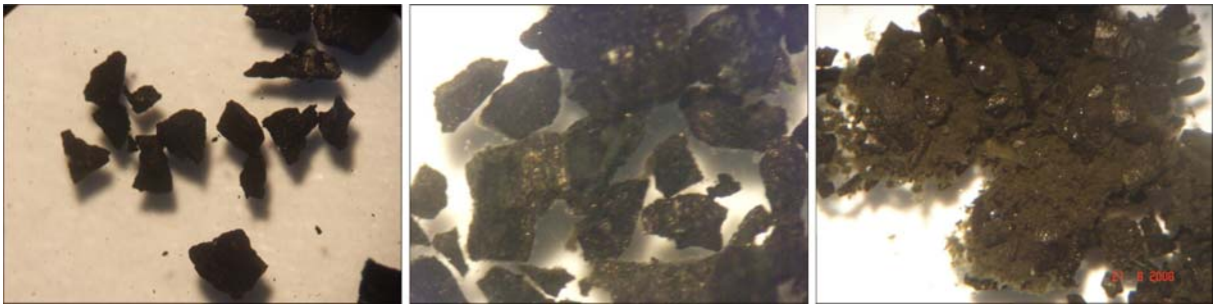
Fig. 5 Increase in HPC pour plate (CFU/mL) as a function of time (day) with 15 cm bed depth and v=4 m/h

at or below water quality standards (ADWS 2004). However there is still a need for treatment methods to improve rainwater quality especially at RMUTT, Thanyaburi. The results also imply that diverting the first flush off the roof, which is heavily polluted, can improve the water quality of the rainwater collected in the tank.

In this study the submerged microfiltration was used to treat the rainwater collected from RMUTT with and without GAC biofilter as a pretreatment.

3.2 Biofilter

Figure 4 shows the DOC removal by the biofilters during 3 months of operation. The DOC removal was 10%, 25%, and 40% with 5-, 10-, and 15-cm filter bed depth, respectively. The removal rates in the early stages were relatively high due to the adsorption by granular activated carbon. However as the granular activated carbon became exhausted and as biological activity on the granular activated carbon increased, a relatively steady removal rate, albeit with some fluctuation, in DOC was established. The biofilter operated under this steady condition for 70 days after an initial period of adsorption. The results show that



A. Pure GAC B. 30 days operation C. 90 days of operation

Fig. 6 Biomass growth on GAC in different period. **a** Pure GAC, **b** 30 days of operation, **c** 90 days of operation

the biofilter can remove organics for a prolonged period of time without the need to regenerate the activated carbon.

It had been previously estimated that the approximate time for colonization of biomass into a steady-state condition could take nearly 3 months (Cauchi et al. 1993). Figure 5 shows the biomass growth on GAC. During the first month the growth of the colonization using the indirect effluent measure of hetrotropic plate count (HPC) showed that the colonization of microorganism was 400 CFU/mL. It rose rapidly, and after 90 days, the microorganism count was more than 3,000 CFU/mL. This may be due to the presence of first flush in the rainwater used in this study.

The growth of biomass on the GAC is shown in Fig. 6. The biomass growth in terms of colony count was not detected initially (0 day), and it increased to the third order (10^3 CFU/gm GAC) after 30 days of operation and then to more than 10^5 CFU/g GAC after 90 days of operation (Table 4). The biomass was taken from the GAC column by backwashing, and GAC weight was calculated from bed height. The use of Gram's stain showed that the colony had both Gram positive and Gram negative. However the colony was predominantly Gram positive and rod-shaped.

Table 4 CFU count/g GAC at different operation period of biofilter

Date	Dilution	Colony count		Average	CFU/ g GAC
		1 replication	2 replications		
0	10^{-1}	3	5	4	<30
	10^{-2}	0	2	1	<30
	10^{-3}	0	0	0	0
	10^{-4}	0	0	0	0
	10^{-5}	0	0	0	0
30	10^{-1}	291	289	290	2.9×10^3
	10^{-2}	70	56	68	0.68×10^3
	10^{-3}	2	17	8.5	<30
	10^{-4}	3	13	8	<30
	10^{-5}	0	0	0	0
90	10^{-1}	>300	>300		
	10^{-2}	>300	>300		
	10^{-3}	127	169	148	1.48×10^5
	10^{-4}	14	9	11.5	
	10^{-5}	2	0	1	

Table 5 Average anion concentrations in the effluent of the biofilter

Anion	10 cm mg/L (% removal)	15 cm mg/L (% removal)
F^-	0.0701 (0%)	0.0805 (0%)
Cl^-	1.407 (0%)	1.253 (0%)
NO_3^-	3.451 (81%)	1.875 (90%)
SO_4^-	3.442 (41%)	3.214 (44%)
PO_4^{3-}	1.031 (31%)	0.805 (46%)

Samples were taken after a biofilter operation time of more than 1 month

Daily samples of effluent from the biofilter taken after an operation time of more than 1 month were tested for anion concentrations of F^-, Cl^-, NO_3^-, SO_4^-, and PO_4^{3-}. Table 5 shows the results of these tests. The reduction of nitrate and phosphate was 80% and 30%, respectively, in the 10-cm filter bed depth (Table 5). The reductions were much greater in the 15-cm filter bed. The reduction of F^- and Cl was negligible as these anions were not consumed by the microbes in the biofilter.

3.3 MF

The MF alone achieved only a 10% removal of DOC with rainwater. When this was used with a pretreatment of biofiltration, the DOC removal increased to 45–50%. The majority of the DOC removal was by biofiltration, which is about 40%. However HPC analysis revealed that no bacteria were detected in the effluent water following membrane filtration. The microfiltration unit was operated at 8 L/m^2 h, and the pressure development was almost negligible (less than 5 Pa) during a filter run period of 6 h.

3.4 Fouling Index with Biofilter Effluent

The two commonly used fouling indices, namely SDI and MFI (Boerlage et al. 1998), were used to measure the membrane fouling reduction by the pretreatment of biofiltation. The SDI procedure is described in American Standards Testing and Methods (ASTM) D4189-95.

The MFI is an extension of the SDI and was developed by Schippers and Verdow (1980). The MFI can be used to predict the fouling potential of the feed in membrane systems and assumes that the particulate fouling of membranes is dominated by cake filtration. The MFI is determined from the gradient of the general cake filtration region for constant pressure in a plot of t/V versus V (Boerlage et al. 1998).

The MFI values with and without biofilter pretreatment were 360 and 863 s/L^2, respectively. The SDI values were also investigated. A pretreatment with biofilter decreased the SDI value from 6.2 to 4.1. Thus, the biofilter as pretreatment to membrane filtration is effective in reducing membrane fouling potential.

4 Conclusion

The results of testing of samples of rainwater collected from roofs in Ayudhaya and in RMUTT, Thanyaburi, showed that although some parameters comply with water quality standards, there is still a need for treatment methods to improve rainwater quality. In this study various treatment methods were used, and the results are summarized below:

1. The initial DOC concentration of rainwater was 3.3 mg/L and contained nutrients such as nitrate, sulfate, and phosphate at concentrations of 18.6, 5.8, and 1.5 mg/L, respectively.
2. A GAC filter with a depth of 15 cm could remove up to 40% of DOC and could operate for at least for 3 months. It could remove a significant amount of nutrients such as nitrate and phosphate.
3. The use of MF/UF to filter the effluent of the biofilter showed only a marginal increase in DOC removal by another 5–15% but it could effectively remove microorganisms from the effluent of the biofilter.
4. The MFI values decreased from 863 to 360 s/L^2 after biofilteration. The SDI values decreased from 6.2 to 4.1. The use of biofilters as pretreatment to MF could reduce membrane fouling in addition to removing a higher amount of DOC and increasing the membrane treatment efficiency.

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