Science of the Total Environment 694 (2019) 133719

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Biocarriers facilitated gravity-driven membrane (GDM) reactor for wastewater reclamation: Effect of intermittent aeration cycle

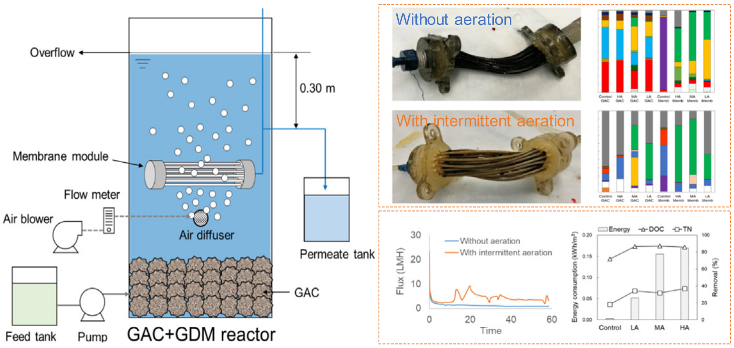
Seonki Lee [a,b], Matthias Sutter [c], Michael Burkhardt [c], Bing Wu [d,*], Tzyy Haur Chong [a,b,**]

[a] *Singapore Membrane Technology Centre, Nanyang Environment and Water Research Institute, Nanyang Technological University, 1 Cleantech Loop, Clean Tech One 06-08, 637141, Singapore*
[b] *School of Civil and Environmental Engineering, Nanyang Technological University, 50 Nanyang Avenue, 639798, Singapore*
[c] *HSR Hochschule für Technik Rapperswil, Institute of Environmental and Process Engineering (UMTEC), Oberseestrasse 10, 8640 Rapperswil, Switzerland*
[d] *Faculty of Civil and Environmental Engineering, University of Iceland, Hjardarhagi 2-6, IS-107 Reykjavik, Iceland*

HIGHLIGHTS

- Biocarriers + gravity-driven membrane (GDM) reactor improved permeate quality.
- Intermittent aeration enhanced permeate flux by reducing cake resistance.
- Microbial community compositions were influenced by intermittent aeration.
- Biocarriers + GDM reactor at low aeration intensity had a high energy efficiency.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 25 June 2019
Received in revised form 31 July 2019
Accepted 1 August 2019
Available online 05 August 2019

Editor: Huu Hao Ngo

Keywords:
Decentralized wastewater treatment
Gravity-driven membrane
Granular activated carbon
Intermittent aeration
Membrane fouling
Microbial community

ABSTRACT

This study investigated the performances of gravity-driven membrane (GDM) reactors integrated with granule activated carbon (GAC) biofilm process for wastewater treatment under different intermittent aeration cycles (intensity and frequency). The results showed the removal efficiencies of dissolved organic carbon, total nitrogen, ammonia were significantly improved under intermittent aeration conditions (~86–87%, ~29–37%, and ~83–99%, respectively) compared to non-aeration condition (~72% and ~18%, and ~17%, respectively). In addition, it was found that the intermittent aeration significantly reduced the cake layer resistance and therefore improved ~130–300% the permeate flux compared to control without aeration. Microbial community analysis indicated that prokaryotic and eukaryotic compositions in the cake layer biofilm were significantly influenced by aeration condition. Lastly, energy consumption analysis revealed that GAC + GDM with shorter aeration period and low aeration intensity could be promising as a decentralized wastewater treatment process in terms of water quality and operating energy.

1. Introduction

Globally, providing reliable wastewater treatment in rural areas with low population densities and dispersed households is a challenge (Massoud et al., 2009). Especially, in many scenarios (such as shortage of fresh water supply), the treated water from the decentralized

---

* Correspondence to: B. Wu, Faculty of Civil and Environmental Engineering, University of Iceland, Hjardarhagi 2-6, IS-107 Reykjavik, Iceland.
** Corresponding to: T.H. Chong, Singapore Membrane Technology Centre, Nanyang Environment and Water Research Institute, Nanyang Technological University, 1 Cleantech Loop, Clean Tech One 06-08, 637141, Singapore.
E-mail addresses: wubing@hi.is (B. Wu), THChong@ntu.edu.sg (T.H. Chong).

https://doi.org/10.1016/j.scitotenv.2019.133719

wastewater treatment systems needs to be adopted for non-potable reuse purposes such as toilet flushing, gardening, irrigation, etc. (Capodaglio et al., 2017; Gikas and Tchobanoglous, 2009). Conventionally, constructed wetland, media filters, lagoons, and bioreactors have been widely used as decentralized wastewater treatment or reclamation processes (Fernandes et al., 2013; Vega et al., 2003; Wu et al., 2011). However, in some situations, these traditional methods cannot guarantee the treated water to meet the increasingly strict wastewater discharge or non-potable reuse standards (Nguyen et al., 2007; Wu et al., 2015).

Recently, gravity-driven membrane (GDM) filtration have received great attention as a decentralized process in treating surface water, rainwater, greywater, and sewage water (Cecconet et al., 2019; Pronk et al., 2019; Tang et al., 2016; Wu et al., 2019). The advantages of the GDM process include that (1) it can produce superior treated water due to high membrane separation efficiency; (2) it is an economic process due to its lower capital cost (no permeate suction pump) and operation cost (without requiring physical and chemical cleaning) compared to other membrane processes such as membrane bioreactors (MBRs).

Previous studies have shown that the GDM systems could potentially treat greywater/municipal wastewater, but with a relatively low permeability (30–70 L/m^2 h/bar), which is dependent on the wastewater quality such as organic concentrations (Ding et al., 2016; Ding et al., 2017a, b; Jabornig and Podmirseg, 2015; Wang et al., 2017). In addition, the GDM systems could not fully remove dissolved organic substances, such as humic substances, building blocks, and low molecular weight substances (Ding et al., 2018a; Wu et al., 2019).

To further improve GDM performance in municipal wastewater treatment, activated carbon media were attempted to be integrated with the GDM system by coating them on membrane surface, or as pretreatment filter, or by packing them inside of GDM reactor (Ding et al., 2018a, b; Tang et al., 2018a, b). The reported studies have revealed that the presence of activated carbon could significantly improve permeate quality of the GDM systems due to the adsorption capability of activated carbon or/and biodegradation behaviors of the attached biofilm on activated carbon. However, flux enhancement in the GDM systems was only observed when activated carbon particles were performed as pretreatment filter or packed as biofilm media in the GDM reactor (Tang et al., 2018a, b). Nevertheless, nutrient removal in the reported biocarriers facilitated GDM systems was not explored, which was considered as an important parameter in municipal wastewater treatment for satisfactory discharge or reclamation.

In conventional activated sludge processes, nutrient removal is generally achieved by combining aerobic and anoxic/anaerobic reactors via nitrification and denitrification pathways. Alternatively, intermittent aeration can be applied in a single reactor to develop sequential aerobic and anoxic conditions for the simultaneous removal of carbon and nitrogen from wastewater (Yoo et al., 1999). In MBRs, several studies have illustrated high treatment efficiency under intermittent aeration conditions (Capodici et al., 2015). The same approach may also be suitable for the biocarriers facilitated GDM system in treating municipal wastewater for simultaneous carbon and nutrient removal. Although Tang et al. (2016) has emphasized the intermittent aeration could influence membrane permeate flux in the conventional GDM system, they did not examine the nutrient removal in the presence of intermittent aeration. Therefore, further optimization of intermittent aeration condition in the biofilm facilitated GDM system is necessary to maximize treatment and economic efficiencies.

This study aims to compare organic and nitrogen removal, membrane performance, microbial community, and energy consumption in granular activated carbon (GAC) facilitated GDM systems in treating real municipal wastewater under different intermittent aeration cycles (intensity and frequency).

2. Materials and methods

2.1. GAC + GDM reactor setup and operation

Two lab-scale GAC + GDM reactors were setup in parallel and the schematic diagram of the reactor was shown in Fig. 1. The reactor has a working volume of 8.6 L packed with 1.25 kg of GAC media (FiltrabsorbR 300, Calgon Carbon, US) at the bottom of the reactor. A hollow fiber membrane module (PVDF; 150 kDa; total effective surface area of 138 cm^2) was installed into the reactor and located 30 cm below the water level (i.e., a hydrostatic pressure of 30 mbar). The air diffuser was placed below the membrane modules and above the GAC layer. The wastewater was collected from the primary sedimentation tank in a municipal wastewater treatment plant in Singapore. The feed flow rate was adjusted according to permeate flow rate daily to minimize the overflow. The reactor was operated at a room temperature of 21 ± 1 °C.

In the first stage, two reactors were operated under non-aeration (hereinafter defined as control) and 60 min on/60 min off with 2 L/min aeration (hereinafter defined as HA) conditions, respectively; in the second stage, two reactors were operated under 30 min on/60 min off with 2 L/min (hereinafter defined as MA) and 0.5 L/min (hereinafter defined as LA) aeration, respectively (Table 1). In each condition, a new membrane module and fresh GAC media were used.

2.2. Water quality analysis

In this study, the water samples were periodically taken from the reactors for analysis, i.e., multi samples ($n = 4$–8) were measured during 62-day's operation. The dissolved organic carbon (DOC) and total nitrogen in the feed, reactor, and permeate were measured by a TOC/TN analyzer (Shimadzu, Japan) after the samples was filtered with a syringe membrane (0.45 μm, Millipore, USA). Ammonia (NH_4-N) and nitrate (NO_3-N) were examined using the spectrometric method with Ammonia TNT 831 kit (Hach, USA) and Nitrate TNT 835 kit (Hach, USA), respectively. The pH and dissolved oxygen (DO) measurements were conducted with a portable pH-meter (Mettler Toledo, Switzerland) and a portable DO meter (Mettler Toledo, Switzerland), respectively. To illustrate statistical significance, two-sample *t*-test was performed by comparing the data groups (different sampling times) between two reactors (i.e. control GDM reactor vs. intermittent-aerated GDM reactor) and one-way ANOVA test was performed for a comparison among three intermittent-aerated GDM reactors. The *p*-values for the two-sample *t*-test and one-way ANOVA test were calculated at a significance level at 5%.

2.3. Biofilm layer analysis

(1) Fouling resistance

The fouling resistance was evaluated using resistance-in-series model (Broeckmann et al., 2006) based on Darcy's Law as shown in Eqs. (1)–(3). R_t is the total resistance (m^{-1}), consisting of intrinsic membrane resistance (R_m), irreversible fouling resistance (R_{ir}), and cake layer resistance (R_c).

$$R_t = R_m + R_{ir} + R_c = \frac{\Delta P}{\mu J_s} \tag{1}$$

$$R_m + R_{ir} = \frac{\Delta P}{\mu J_p} \tag{2}$$

$$R_m = \frac{\Delta P}{\mu J_m} \tag{3}$$

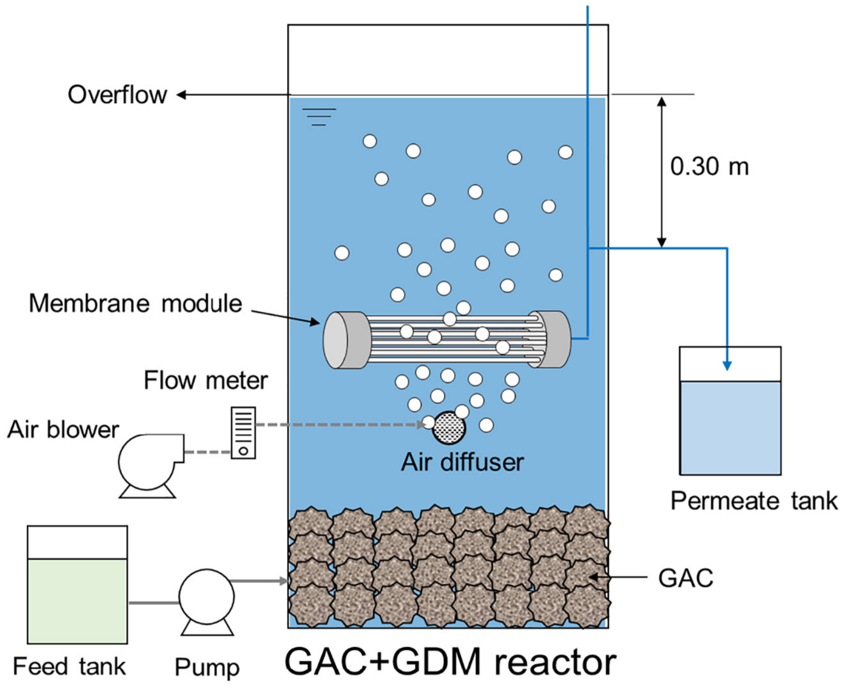


Fig. 1. A schematic diagram of the GAC + GDM reactor.

where ΔP is the transmembrane pressure (Pa), μ is the viscosity of permeate (Pa·s), J_s is the stabilized permeate flux (L/m^{2}h), J_m is the permeate flux of clean membrane (L/m^{2}h). At the end of operation, the fouled membrane was taken from the GAC + GDM reactor. After the cake layer was physically removed by rinsing with Milli-Q water for 10 min, the permeate flux (J_p) of the physically-cleaned membrane was measured at the hydrostatic pressure of 30 mbar. The cake layer resistance (R_c) was calculated based on the difference between total resistance (R_t) and the resistance after physical cleaning ($R_m + R_{ir}$). The irreversible fouling resistance (R_{ir}) was achieved based on the difference between the resistances after physical cleaning ($R_m + R_{ir}$) and intrinsic membrane resistance (R_m).

(2) Microbial community

At the end of each experiment, the membrane module was taken from the GDM reactor and the biofilm was removed from the membrane surface by rinsing with 50 mL of sterilized Milli-Q water for 10 min (i.e., membrane biofilm solution). In addition, all GAC particles were taken from the reactors and well mixed. Approximately 10 g of wet GAC particles were added into a tube with 20 mL of sterilized Milli-Q water and the mixture was vortexed for 2 min in order to remove biofilms from GAC particles (i.e., GAC biofilm solution). The biofilm solution (20 mL) was centrifuged at ×4500*g* for 10 min. Then, the biofilm pellets were collected and kept at −20 °C before DNA extraction. PowerBiofilm® DNA isolation kit (MO BIO, USA) was used to extract the genomic DNA of microorganisms in the biofilm. The microbial communities of prokaryotes and eukaryotes were analyzed using the 16S and 18S rRNA sequencing, respectively. The sequencing was performed using Illumina MiSeq platform with primers 357wF (CCTACGGGGNGGCWGCAG) and 785R (GACTACHVGGGTATCTAATCC) for prokaryotes, TAReukF (CCAGCASCYGCGGTAATTCC) and TAReukR (ACTTTCGTTCTTGATYRA) for eukaryotes. The results were analyzed by the standard de novo operational taxonomic unit (OUT)-based approach using QIIME (Caporaso et al., 2010).

2.4. Liquid chromatography-organic carbon detection (LC-OCD) analysis

Soluble organic fractions in the water and biofilm samples were measured by an LC-OCD analyzer (LC-OCD Model 8, DOC-LABOR, Germany), a size-exclusion chromatography integrated with organic carbon detector and organic nitrogen detector. The organic matters were identified into five different fraction groups according to their molecular weights, namely, biopolymers (MW > 20 kDa), humic substances (MW ~ 1000 Da), building blocks (MW ~ 300–500 Da), low molecular weight (LMW) acids and neutrals (MW < 350 Da). The mobile phase was prepared by dissolving 2.5 g KH_2PO_4 (Merck,

Table 1
Operating conditions of the GAC + GDM reactors.

Parameter	Stage 1		Stage 2	
	Control (non-aeration)	HA (high aeration)	MA (medium aeration)	LA (low aeration)
Aeration frequency	–	60 min on 60 min off	30 min on 60 min off	30 min on 60 min off
Aeration intensity	–	2 L/min	2 L/min	0.5 L/min
Average aeration rate	–	1 L/min	0.67 L/min	0.17 L/min
DO (aeration/non-aeration)	<0.5 mg/L	7.0–8.8 mg/L /3.3–4.9 mg/L	7.8–8.5 mg/L /4.4–5.2 mg/L	7.0–7.7 mg/L /3.3–4.2 mg/L
pH	6.8 ± 0.3	7.1 ± 0.4	6.6 ± 0.2	6.3 ± 0.3
HRT[a]	~490–720 h	~80–180 h	~180–240 h	~220–320 h

[a] HRT: hydraulic residence time, which was calculated by averaging the daily HRT values during day 20–62.

#1.04873) and 1.5 g Na_2HPO_4 (Merck, #1.06580) into 1 L of Milli-Q water. The acidification solution was prepared by adding 4 mL of o-phosphoric acid (85%, Merck, #1.00573) and 0.5 g of potassium peroxodisulfate (Merck, #1.05091) into 1 L of Milli-Q water. The detailed LC-OCD operation and analysis methods were described in the literature (Huber et al., 2011).

3. Results and discussion

3.1. Permeate quality

3.1.1. Organic carbon removal

The DOC concentrations in the feed wastewater, reactor, and permeate were periodically monitored. As shown in Fig. 2a, the feed wastewater concentration considerably fluctuated (~16–68 mg DOC/L) in both stages. Nevertheless, in the reactor, DOC removal ratios of GAC + GDM reactors with intermittent aeration (HA: 87.3%, $p < 0.005$; MA: 79.6%, $p < 0.05$, LA: 82.3%, $p < 0.05$, two-sample *t*-test) were significantly higher compared to the control reactor without aeration (65.0%). The observations indicated that the presence of aeration promoted the DOC removal, possibly due to (1) the enhanced biodegradation activity as well as bacterial propagation in the presence of sufficient DO (Ding et al., 2017a; Dong et al., 2009) and (2) improved organic sorption capability of the top-layer GAC particles exposing to aerobic scenarios (Karanfil et al., 1996).

While, the DOC removal ratio in the GAC + GDM reactors was independent with intermittent aeration cycles ($p > 0.1$, one-way ANOVA test). It was probably due to their similar DO levels, despite different aeration intensities (0.5 or 2 L/min) and aeration frequencies (30 or 60 min on/60 min off). As shown in Table 1, DO level during aeration was 7.0–8.8, 7.8–8.5, and 7.0–7.7 mg/L in the HA, MA, and LA reactors, respectively, indicating sufficient DO for microbial metabolism under different aeration conditions. Also, during non-aeration mode, DO levels remained over ~3 mg/L in all intermittent aeration reactors. It may be attributed to the low concentrations of suspended biomass in reactors (<100 mg/L) because of the nature of GAC + GDM reactor.

To further investigate the effect of aeration on biodegradation of DOC, compositions of DOC were analyzed by LC-OCD. Fig. 3 shows that the removal efficiencies of small organic fractions, i.e., humic substances (~84–93%), building blocks (~82–93%), LMW neutrals (~81–90%), and LMW acids (~93–100%) in the GDM reactors were significantly greater than that of biopolymers (~56–69%), regardless of aeration cycle intensity and frequency. In addition, intermittent aeration enhanced removal performance for building blocks, LMW neutrals, LMW acids by ~3–10%, ~7–9%, ~5–7%, respectively, compared to control reactor without aeration. While, biopolymers (~56–69% with intermittent aeration vs. ~58% without aeration) and humic substances (~84–93% with intermittent aeration vs. ~84% without aeration), appear to be relatively less affected by aeration conditions.

Furthermore, the permeate quality of the GDM reactors were compared. The presence of intermittent aeration improved permeate quality (~2.6–4.4 mg DOC/L vs. ~9.4 mg DOC/L in the control), mainly due to enhanced biodegradation/biosorption roles (Fig. 2a). While, the membrane separation only contributed <8% of DOC removal. In detail, biopolymer was significantly rejected by the membrane (>25%) regardless of aeration condition (Fig. 3), due to their relatively greater sizes than membrane pore size. While, LMW substances in the permeate were greater than those in the reactor. This phenomenon was also observed in the previous studies (Chomiak et al., 2015; Derlon et al., 2014; Wu et al., 2017). It was speculated that the biofilm on the membrane surface utilized the greater-sized organics and produced such LMW substances, which could not be effectively retained by the membrane and therefore were present in the permeate.

3.1.2. Nitrogen removal

Fig. 2b shows the concentrations of TN in feed, reactor, and permeate and removal ratios. The TN removal ratios attributed by biodegradation/biosorption in the GAC + GDM reactors with intermittent aeration (~29.3–37.3%) was approximately 2–3 times greater than that without aeration (~13.3%), revealing that intermittent aeration significantly enhanced the TN removal. While, the employed aeration frequency and intensity did not lead to dissimilar TN removal performance ($p > 0.2$, one-way ANOVA test).

To examine the nitrogen removal mechanism, ammonia and nitrate were analyzed and presented in Fig. 4a and b, respectively. The ammonia removal in the control reactor without aeration was only ~10.6% due to its limited DO (< 0.5 mg/L) for nitrification (Ruiz et al., 2003). Meanwhile, the nitrate was not detected in the control reactor, implying that

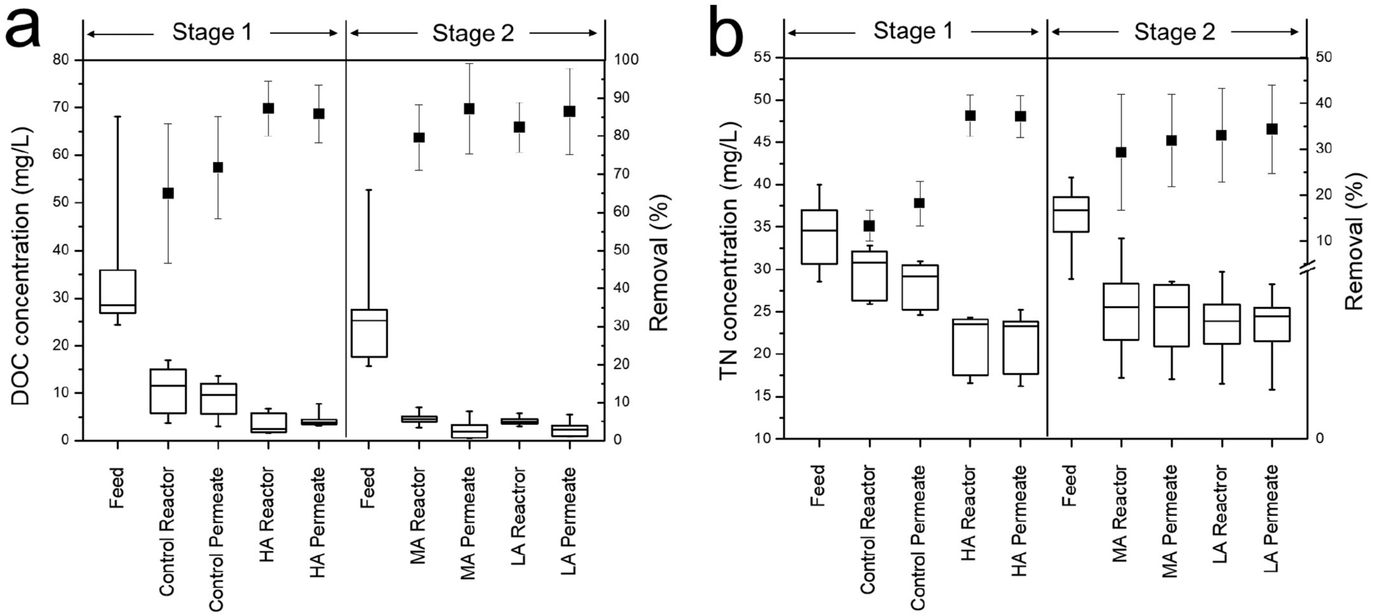


Fig. 2. (a) DOC and (b) TN concentrations in the feed, reactor, and permeate during day 15–62 and their removal efficiencies (n = 8). The central line of each box is the median value of the concentration, while the top and bottom of each box represent the third and first quartile, respectively. The vertical line extends from the minimum to the maximum values. Square dots indicate the average removal efficiencies, which were calculated based on the data in the feed.

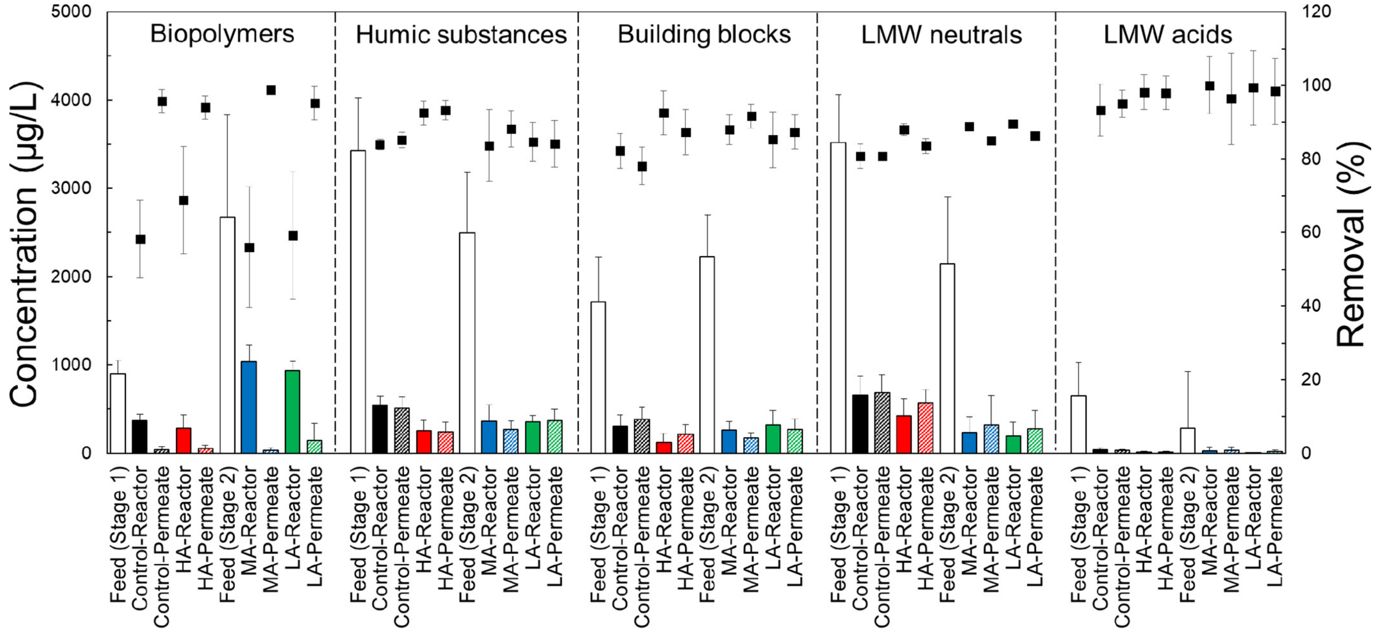


Fig. 3. Soluble organic compositions analyzed by LC-OCD (n = 4–5). Columns indicate the concentrations while square dots indicate the average removal efficiencies calculated based on the data in the feed.

the ammonia-converted nitrate was completely denitrified to nitrogen gas in the control reactor. Under intermittent aeration conditions, the ammonia removal ratios (~84–98%) were greater than that in the control reactor, attributing to improved nitrification under sufficient DO conditions during aeration period (Ossenbruggen et al., 1996). While, nitrate (5–28 mg/L) was detected in the three GDM reactors with intermittent aeration, showing incomplete denitrification during non-aeration period (DO > 3 mg/L).

Furthermore, the ammonia removal in the HA reactor (~98%) was significantly greater than those in MA (~84%, $p < 0.001$, two-sample *t*-test) and LA (~85%, $p < 0.05$, two-sample *t*-test) reactors, implying higher ammonia oxidation activity in the HA reactor. However, the average NO_3-N concentration in the HA reactor (~12 mg/L) was lower than those in the MA (~21 mg/L) and LA (~21 mg/L) reactors. Thus, it may be attributed by a higher C/N ratio in the HA feed (TOC/TN: 1.0 ± 0.3 in Stage 1) than that in the MA and LA feed (TOC/TN: 0.7 ± 0.2 in Stage 2), which could promote ammonia assimilation by heterotrophic bacteria (Matsumoto et al., 2007).

In addition, it is noted that during non-aeration period, the DO levels in the GAC + GDM reactors were kept at 3.3–5.2 mg/L (Table 1). The presence of denitrification under such a higher DO level may be associated with two possibilities: (1) coexistence of anoxic and aerobic zones within GAC bed and/or (2) the presence of aerobically denitrification process (Ji et al., 2015). Overall, similar to the DOC removal patterns,

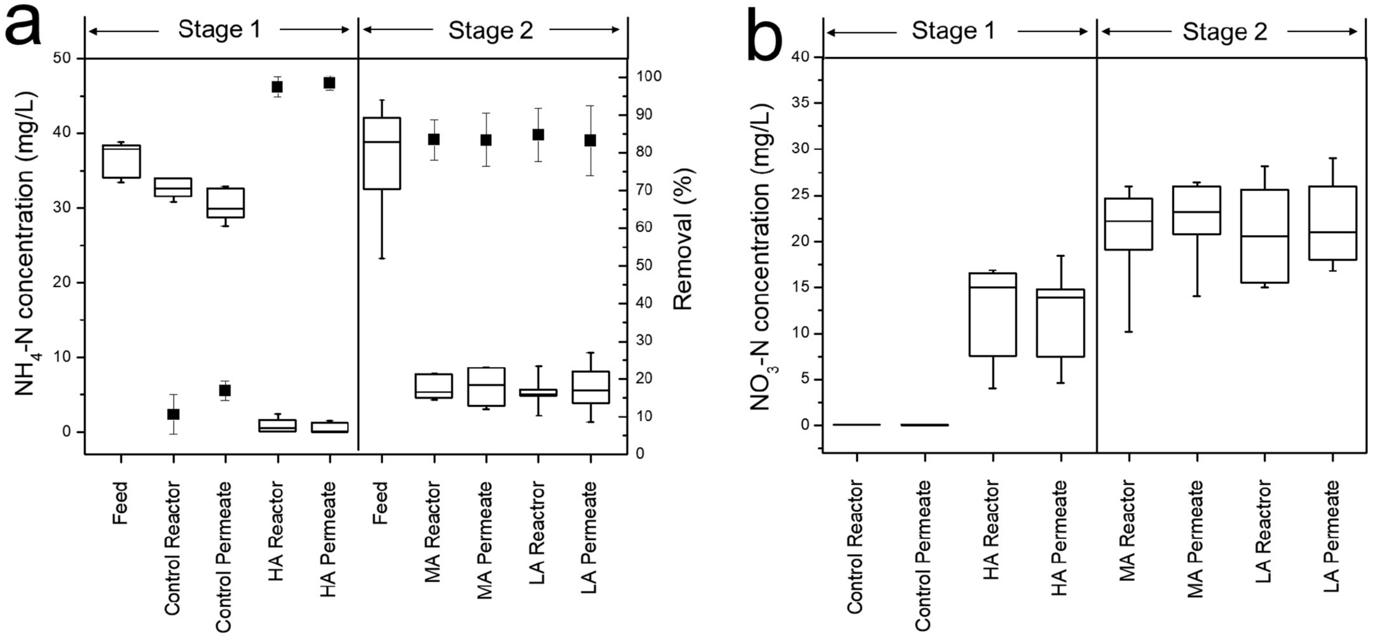


Fig. 4. (a) Ammonia concentrations and removal ratios (n = 5–7) and (b) nitrate concentrations (n = 4–6) in the feed, reactor, and permeate during day 15–62. The central line of each box is the median value of the concentration, while the top and bottom of each box represent the third and first quartile, respectively. The vertical line extends from the minimum to the maximum values. Square dots in panel a indicate the average ammonia removal efficiencies, which were calculated based on the data in the feed.

the nitrogen removal was mostly via biological nitrification and denitrification pathways in the GAC + GDM reactor instead of membrane separation.

As shown in Fig. 2, relatively fluctuations of organic removals were also noticed. In this study, the real municipal wastewater was periodically taken from the wastewater reclamation plant and only dissolved organic substances in the feed water were monitored (i.e., DOC and dissolved TN). The particulate organics in the feed water were not examined, which may convert to dissolved organics during GDM reactor operation. Part of these dissolved organics (from particulate organics) may pass through the membrane and be present in the permeate. Therefore, the fluctuations of organic removal ratios may be associated with the facts: (1) fluctuations of dissolved organics (Fig. S1 in supplementary data) and particulate organics in the feed water; (2) dynamic microbial development with extending reactor operation time.

3.2. Membrane performance

Fig. 5a describes the flux development in the GAC + GDM reactors under different aeration conditions. During the initial filtration stage, the permeate flux dramatically dropped in all GAC + GDM reactors, regardless of aeration intensity and frequency. Possibly pore blocking and narrowing were predominant fouling during this period of time (Wu et al., 2016), as a result, the foulants in the membrane pores could not be effectively removed by air scouring (i.e., irreversible fouling). After 5-day operation, the permeate fluxes in the control, LA, and MA reactors were gradually stabilized over the time. While, in the reactor with higher aeration intensity and extended aeration time (i.e., HA reactor), the permeate flux fluctuated more obviously. It may be attributed to the fact that the stronger shear force induced by aeration with higher intensity and longer time periodically removed the formed loosely-attached cake layers, which could enhance membrane filtration.

On average, the stabilized flux (calculated based on the flux values during last 20 days) was achieved at ~0.9, 2.0, 2.6, and 3.5 L/m^2 h in the control, LA, MA, and HA reactors, respectively. This reveals that (1) the presence of aeration could significantly improve the stabilized flux by 130–300%; (2) under the same aeration frequency (30 min on/60 min off), with increasing aeration intensity from 0.5 to 2 L/min, the stabilized flux improved 30%; (3) under the same aeration intensity (2 L/min) and non-aeration period of time (60 min off), with extending aeration time from 30 min to 60 min, the stabilized flux increased 35%.

Our finding was consistent with the observation in a previous study that the permeate flux of the GDM reactor in treating surface water could be improved by intermittent aeration (Tang et al., 2016). However, several previous studies also revealed that continuous aeration (i.e., continuous shear force) seems not to be beneficial for improving permeate flux due to the formation of thinner, denser, smoother and less permeable biofilm layers on GDM membranes (Ding et al., 2016; Jabornig and Podmirseg, 2015). In non-aeration GDM systems, it has been well recognized that the gazing and predation behaviors of eukaryotes could induce the heterogeneous and porous biofilm layer on the membrane surface, which facilitates a higher permeability (Derlon et al., 2013; Klein et al., 2016). In the presence of continuous aeration shear force, the loosely-attached biofilm layer could be removed from the membrane surface and the number of eukaryotes in the biofilm layer may decrease with biofilm detachment, which may have a negative impact on the biofilm permeability. However, in this study, the eukaryotic activity on the membrane could be decreased by GAC layer where can reject eukaryotes with large sizes. On the other hand, under intermittent aeration conditions, non-aeration period could provide a chance for eukaryotes to attach on the biofilm and perform gazing and predation roles. Possibly, the intermittent aeration intensity and frequency could determine the biofilm cake layer formation and detachment situations, which are associated with permeate flux in the GDM system.

To further explore the membrane fouling mechanism in the GAC + GDM reactors, the membrane module was taken out of the reactor at Day 62 and the fouling resistance was evaluated, shown in Fig. 5b. Overall, the total resistance decreased with an increase of aeration input. Obviously, the cake layer formation contributed majorly to the total fouling in all reactors, ranging from 55% to 85%. Compared to the GAC + GDM reactor without aeration, the presence of aeration significantly reduced the cake layer resistance ($2.4–4.0 \times 10^{-12}$ m^{-1} vs. 12.8×10^{-12} m^{-1}), but only led to a slight decrease in irreversible fouling resistance ($0.23–1.71 \times 10^{-12}$ m^{-1} vs. 1.87×10^{-12} m^{-1}). Our results suggest that the intermittent aeration could majorly influence the formation of biofilm cake layer attached on the membrane surface in the reactor.

Under the same aeration frequency (30 min on/60 min off), with increasing aeration intensity from 0.5 to 2 L/min, the cake layer resistance decreased by 40%, but irreversible fouling increased 53%. This may be attributed to the fact that higher shear force induced by a higher air flow rate could more effectively remove the formed cake layer from the membrane surface. Accordingly, the lack of the biofilm cake layer (i.e., considered as a secondary membrane) may promote membrane pore blocking/narrowing (i.e., irreversible fouling), which could not further be removed by shear force. However, under the same aeration intensity (2 L/min) and non-aeration period of time (60 min off), with extending aeration time from 30 min to 60 min, the cake layer resistance increased by 33%, but irreversible fouling decreased by 87%. It might imply that under a higher shear force condition, with extending air scouring time, the foulants that strongly-attached on the membrane surface (i.e., irreversible foulants) tended to be readily removed

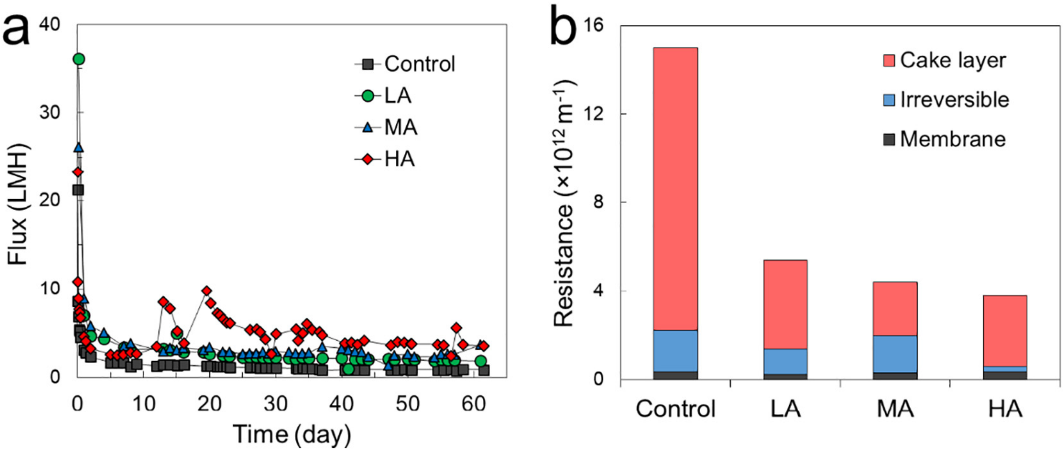


Fig. 5. (a) Permeate flux developments and (b) filtration resistance profiles in the GAC + GDM reactors.

(leading to flux fluctuation in the HA reactor during 8–20 days). As a result, the contribution of irreversible fouling to overall filtration resistance was reduced, while that of reversible fouling was increased.

3.3. Characterization of biofilm cake layer

3.3.1. Soluble organic substances in the biofilm cake layer

In conventional MBRs, it has been well illustrated that the soluble organic substances (such as extracellular polymeric substances) accumulated on the membrane surface contribute greatly to membrane fouling (Gao et al., 2012; Meng et al., 2009). In this study, the fractions of soluble organic substances in the biofilm cake layer were examined by LC-OCD and described in Table S1.

Biopolymers were identified as the major organic foulants in the control reactor (72%) and the reactors with low aeration rate and shorter aeration duration (83–87%). While, they only accounted for 25% of the total soluble organic foulants in the reactor with high aeration intensity (Table S1). Nevertheless, the amount of biopolymers was highly correlated with the cake layer resistance ($R^2 = 0.997$), rather than the amounts of small-sized organic fractions (i.e., building blocks and LMWs). Furthermore, it was observed that the amounts of biopolymers on the membranes in the intermittent aerated reactors (2.2–6.2 μg/cm^2) were remarkably lower than that in the control reactor (43.4 μg/cm^2). This result can be explained by two aspects: (1) biopolymer aggregates could be detached through shear stress induced by aeration from the membrane surface (Chua et al., 2002) and/or (2) low resistance of biofilm with thin and less extracellular polymeric substances (EPS, i.e., biopolymers) could be formed under the aerobic (i.e. high DO) conditions compared to non-aeration (i.e. low DO) (Ding et al., 2017a).

3.3.2. Microbial community

(1) Prokaryotic community

At the end of filtration operation, the biofilm samples were collected from the GAC particles and the fouled membranes. Subsequently, the microbial community compositions of these samples were analyzed in terms of prokaryotic (Fig. 6a) and eukaryotic community (Fig. 6b).

As shown in Fig. 6a, the prokaryotic community compositions in the biofilm cake layer was significantly influenced by the aeration condition. In detail, Chlorobi (*Chlorobaculum* genus), phototrophic bacteria that grow under strictly anoxic conditions (Bryant and Frigaard, 2006), was predominant in the biofilm cake layer in the absence of aeration (87.8%). Not surprisingly, Chlorobi was almost not present on the membrane surface in the presence of intermittent aeration. While, high abundance of Nitrospirae (*Nitrospira* genus) were found in the biofilm cake layers in all three reactors with intermittent aeration, accounting for 33.6–59.5%. It is well known that *Nitrospira* belong to nitrite-oxidizing bacteria group and can produce the nitrate under aerobic condition (Koch et al., 2015; Lücker et al., 2010). These observations correlated with the results of ammonia removal (~84–98% under intermittent conditions vs. ~10.6% under control condition). In addition, with increasing aeration intensity, the abundance of Proteobacteria in the biofilm cake layer decreased, but the abundance of Planctomycetes showed an increased trend.

In contrast, the bacterial communities derived from GAC media were considerably similar in all reactors, almost regardless of aeration condition. Bacteroidetes (22.3–39.1%), Firmicutes (15.7–38.2%), and Proteobacteria (8.4–29.1%) were major phyla grown on the GAC

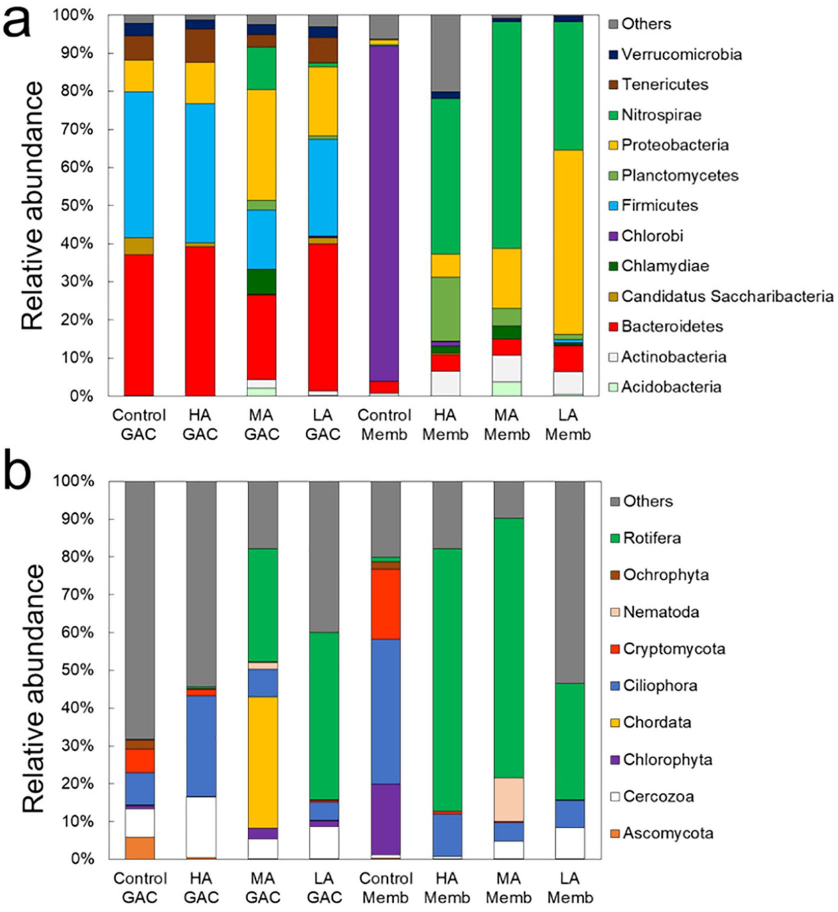


Fig. 6. Prokaryotic (a) and eukaryotic (b) community analysis at the phylum level. "Others" represents all classified taxa that were <1% and unclassified taxa.

particle. In addition, Clostridia class (10.2–26.8%) and *Anaeroplasma* (3.2–8.7%), *Lactobacillus* (1.5–6.0%), *Bacteroides* (0.9–2.3%) genera, which are obligate anaerobic bacteria or facultative anaerobic bacteria, were also found on the GAC media in all reactors. As the air diffuser was located above GAC media layer, limited dissolved oxygen was present in the GAC media layer (i.e., anoxic condition). Especially, the bacteria located inside biofilm layer or porous structure on GAC media could be more restricted from the effect of surrounding dissolved oxygen (Walters et al., 2009).

(2) Eukaryotic community

As shown in Fig. 6b, Ciliophora (38.4%), Chlorophyta (18.6%), and Cryptomycota (18.5%) were predominant eukaryotes on the membrane biofilm layer in the reactor without aeration, but they showed relatively low abundances in those reactors with intermittent aeration. While, Rotifera, a type of aerobic and oligotrophic eukaryote (Sládeček, 1983), was the major phylum in the membrane biofilm layers of the reactors with intermittent aeration (30.8–69.6%), whereas it accounted for only 1.1% in the membrane biofilm layer of the control reactor. In addition, in the presence of aeration, (1) Cercozoa showed a decreased trend with increasing aeration intensity; and (2) a high abundance of Nematoda was noticed in the MA reactor. The differences in compositions of eukaryotic community developed on the membrane surface might be attributed to the greatly dissimilar prokaryotic communities in the biofilm layer (Fig. 6a), considering the predator-prey relationship between eukaryotes and prokaryotes (Langenheder and Jürgens, 2001).

Interestingly, in the GAC biofilm layer, although a highly similarity of prokaryotic communities was present in the reactors with different aeration conditions, the eukaryotic communities appeared to be significantly different. Generally, in the wastewater, eukaryotes have a relatively lower population number with slower growth rate compared to prokaryotes (Bricheux et al., 2013). Thus, eukaryotes might not be evenly distributed within GAC layer as prokaryotes, which could influence sequencing results if only several grams of GAC particles were sampled.

3.4. Economic analysis

Although increasing intermittent aeration intensity could improve permeate flux and nutrient removal in the GAC + GAM reactor, aeration is an energy intensive process, consuming ~70% of energy in conventional MBRs (Judd and Judd, 2006). Therefore, it is necessary to further evaluate energy consumption effectiveness of GAC + GDM systems under the tested aeration conditions in order to ensure its economic feasibility for wastewater reclamation. As a GAC + GDM reactor was driven by gravity and operated without chemical cleaning, only feed pump energy (E_1) and aeration energy (E_2) were considered and calculated using Eqs. (4) and (5), respectively.

$$E_1 = \frac{\rho g h}{\eta} \tag{4}$$

$$E_2 = \frac{\varepsilon Q_{air}}{Q_p} \tag{5}$$

ρ is the density of wastewater (assuming 1000 kg/m^3); g is gravitational acceleration (9.81 m/s^2); h is the height of water level (0.65 m); η is the efficiency of feed pump (assuming 0.6); ε is energy consumption of aeration (assuming 0.019 kWh/m^3 of air) (Maere et al., 2011); Q_{air} is average air flow rate (m^3/s); Q_p is permeate flow rate (m^3/s). In addition, in this study, the packing density of lab-scale membrane module (1.6 m^2 membrane area/m^3 reactor volume) was extremely lower than that of conventional hollow fiber membrane modules (<450 m^2/m^3) (Peinemann and Nunes, 2010). To avoid overestimation for energy consumption, the packing density of 225 m^2/m^3 (i.e., 50% of packing density of conventional modules; because of the additional space for the packed GAC media) was adopted for calculation.

Shown in Table S2, the total energy consumption was estimated at 0.003, 0.052, 0.155, and 0.170 kWh/m^3 in the control, LA, MA, and HA reactor, respectively. It indicates that the energy consumption of intermittent aeration reactors was remarkably higher than that of control reactor since aeration energy accounted for >94% of total energy consumption. However, in terms of permeate quality, the control reactor (without intermittent aeration) may be not promising as a decentralized wastewater treatment process, especially for reuse purpose that requires superior water quality (in particular, nitrogen removal). In addition, the GAC + GDM reactors with intermittent aeration (0.052–0.170 kWh/m^3) appeared to have a competitiveness compared to the conventional activated sludge (~0.3–0.4 kWh/m^3) (Van Dijk and Roncken, 1997) and MBR processes (~0.6–6.1 kWh/m^3) (Fenu et al., 2010; Gil et al., 2010) for municipal wastewater treatment. Furthermore, lower initial capital cost and easier maintaining/operating are additional advantages of GAC + GDM reactors. Among the tested intermittent aeration conditions, the GAC + GDM reactor with low aeration can be more promising in terms of water quality and operating energy (Fig. 7).

In this study, 29.3–37.1% of nitrogen removal was achieved in the reactors with intermittent aeration. In order to further improve nitrogen removal of GAC + GDM reactors, several strategies can be adopted. For example, (1) further lowering aeration rate and extending non-aeration period was suggested. This could benefit to improve denitrification efficiency and reduce energy consumption; (2) internal recirculation can be adopted to promote nitrogen removal by delivering the produced nitrate from the intermittent aeration zone to the anoxic GAC zone. The energy consumption of the recirculation is estimated to be 0.006–0.012 kWh/m^3 (assuming the recirculation rate of 2–4 times the feed flow rate), which only accounts to 10–20% of total energy consumption of GAC + GDM systems.

4. Conclusions

The effect of intermittent aeration on the performance of GAC + GDM reactor was investigated in terms of water quality, permeate flux, microbial community, and energy consumption. The results showed the presence of intermittent aeration could improve removal efficiencies of DOC and TN due to enhanced microbial biodegradation/bioadsorption of organic substances. In addition, increasing aeration shear force and duration could benefit cake layer removal, leading to an improved permeate flux and dissimilar microbial community compositions of the biofilm on the membrane surface. The predation behaviors of eukaryotes played a limited role in the GAC + GDM system due

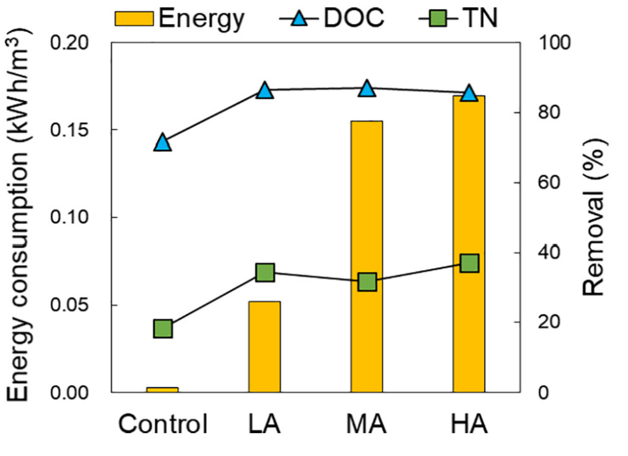


Fig. 7. Comparison of energy consumption and removal performance in GAC + GDM reactors with different aeration conditions.

to their retention in the GAC bed. The energy consumption analysis indicated that the GAC + GDM system with low aeration is promising as a decentralized municipal wastewater treatment process in terms of water quality and operating energy.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The Economic Development Board (EDB) of Singapore is acknowledged for funding the Singapore Membrane Technology Centre (SMTC), Nanyang Technological University.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.133719.

References

Bricheux, G., Morin, L., Moal, G., Coffe, G., Balestrino, D., Charbonnel, N., Bohatier, J., Forestier, C., 2013. Pyrosequencing assessment of prokaryotic and eukaryotic diversity in biofilm communities from a French river. MicrobiologyOpen 2 (3), 402–414.

Broeckmann, A., Busch, J., Wintgens, T., Marquardt, W., 2006. Modeling of pore blocking and cake layer formation in membrane filtration for wastewater treatment. Desalination 189 (1), 97–109.

Bryant, D.A., Frigaard, N.-U., 2006. Prokaryotic photosynthesis and phototrophy illuminated. Trends Microbiol. 14 (11), 488–496.

Capodaglio, A.G., Callegari, A., Cecconet, D., Molognoni, D., 2017. Sustainability of decentralized wastewater treatment technologies. Water Pract. Technol. 12 (2), 463–477.

Capodici, M., Di Bella, G., Di Trapani, D., Torregrossa, M., 2015. Pilot scale experiment with MBR operated in intermittent aeration condition: analysis of biological performance. Bioresour. Technol. 177, 398–405.

Caporaso, J.G., Kuczynski, J., Stombaugh, J., Bittinger, K., Bushman, F.D., Costello, E.K., Fierer, N., Peña, A.G., Goodrich, J.K., Gordon, J.I., Huttley, G.A., Kelley, S.T., Knights, D., Koenig, J.E., Ley, R.E., Lozupone, C.A., McDonald, D., Muegge, B.D., Pirrung, M., Reeder, J., Sevinsky, J.R., Turnbaugh, P.J., Walters, W.A., Widmann, J., Yatsunenko, T., Zaneveld, J., Knight, R., 2010. QIIME allows analysis of high-throughput community sequencing data. Nat. Methods 7, 335.

Cecconet, D., Callegari, A., Hlavínek, P., Capodaglio, A.G., 2019. Membrane bioreactors for sustainable, fit-for-purpose greywater treatment: a critical review. Clean Techn. Environ. Policy 21 (4), 745–762.

Chomiak, A., Traber, J., Morgenroth, E., Derlon, N., 2015. Biofilm increases permeate quality by organic carbon degradation in low pressure ultrafiltration. Water Res. 85, 512–520.

Chua, H.C., Arnot, T.C., Howell, J.A., 2002. Controlling fouling in membrane bioreactors operated with a variable throughput. Desalination 149 (1), 225–229.

Derlon, N., Koch, N., Eugster, B., Posch, T., Pernthaler, J., Pronk, W., Morgenroth, E., 2013. Activity of metazoa governs biofilm structure formation and enhances permeate flux during gravity-driven membrane (GDM) filtration. Water Res. 47 (6), 2085–2095.

Derlon, N., Mimoso, J., Klein, T., Koetzsch, S., Morgenroth, E., 2014. Presence of biofilms on ultrafiltration membrane surfaces increases the quality of permeate produced during ultra-low pressure gravity-driven membrane filtration. Water Res. 60, 164–173.

Ding, A., Liang, H., Li, G., Derlon, N., Szivak, I., Morgenroth, E., Pronk, W., 2016. Impact of aeration shear stress on permeate flux and fouling layer properties in a low pressure membrane bioreactor for the treatment of grey water. J. Membr. Sci. 510, 382–390.

Ding, A., Liang, H., Li, G., Szivak, I., Traber, J., Pronk, W., 2017a. A low energy gravity-driven membrane bioreactor system for grey water treatment: permeability and removal performance of organics. J. Membr. Sci. 542, 408–417.

Ding, A., Wang, J., Lin, D., Tang, X., Cheng, X., Li, G., Ren, N., Liang, H., 2017b. In situ coagulation versus pre-coagulation for gravity-driven membrane bioreactor during decentralized sewage treatment: permeability stabilization, fouling layer formation and biological activity. Water Res. 126, 197–207.

Ding, A., Wang, J., Lin, D., Cheng, X., Wang, H., Bai, L., Ren, N., Li, G., Liang, H., 2018a. Effect of PAC particle layer on the performance of gravity-driven membrane filtration (GDM) system during rainwater treatment. Environ. Sci.: Water Res. Technol. 4 (1), 48–57.

Ding, A., Wang, J., Lin, D., Zeng, R., Yu, S., Gan, Z., Ren, N., Li, G., Liang, H., 2018b. Effects of GAC layer on the performance of gravity-driven membrane filtration (GDM) system for rainwater recycling. Chemosphere 191, 253–261.

Dong, W.-y., Wang, H.-j., Li, W.-g., Ying, W.-c., Gan, G.-h., Yang, Y., 2009. Effect of DO on simultaneous removal of carbon and nitrogen by a membrane aeration/filtration combined bioreactor. J. Membr. Sci. 344 (1–2), 219–224.

Fenu, A., Roels, J., Wambecq, T., De Gussem, K., Thoeye, C., De Gueldre, G., Van De Steene, B., 2010. Energy audit of a full scale MBR system. Desalination 262 (1–3), 121–128.

Fernandes, H., Jungles, M.K., Hoffmann, H., Antonio, R.V., Costa, R.H.R., 2013. Full-scale sequencing batch reactor (SBR) for domestic wastewater: performance and diversity of microbial communities. Bioresour. Technol. 132, 262–268.

Gao, W.J., Qu, X., Leung, K.T., Liao, B.Q., 2012. Influence of temperature and temperature shock on sludge properties, cake layer structure, and membrane fouling in a submerged anaerobic membrane bioreactor. J. Membr. Sci. 421-422, 131–144.

Gikas, P., Tchobanoglous, G., 2009. The role of satellite and decentralized strategies in water resources management. J. Environ. Manag. 90 (1), 144–152.

Gil, J., Túa, L., Rueda, A., Montaño, B., Rodríguez, M., Prats, D., 2010. Monitoring and analysis of the energy cost of an MBR. Desalination 250 (3), 997–1001.

Huber, S.A., Balz, A., Abert, M., Pronk, W., 2011. Characterisation of aquatic humic and non-humic matter with size-exclusion chromatography – organic carbon detection – organic nitrogen detection (LC-OCD-OND). Water Res. 45 (2), 879–885.

Jabornig, S., Podmirseg, S.M., 2015. A novel fixed fibre biofilm membrane process for on-site greywater reclamation requiring no fouling control. Biotechnol. Bioeng. 112 (3), 484–493.

Ji, B., Yang, K., Zhu, L., Jiang, Y., Wang, H., Zhou, J., Zhang, H., 2015. Aerobic denitrification: a review of important advances of the last 30 years. Biotechnol. Bioprocess Eng. 20 (4), 643–651.

Judd, S., Judd, C. (Eds.), 2006. The MBR Book: Principles and Applications of Membrane Bioreactors in Water and Wastewater Treatment. Elsevier, Amsterdam; Boston; London.

Karanfil, T., Schlautman, M.A., Kilduff, J.E., Weber, W.J., 1996. Adsorption of organic macromolecules by granular activated carbon. 2. Influence of dissolved oxygen. Environ. Sci. Technol. 30 (7), 2195–2201.

Klein, T., Zihlmann, D., Derlon, N., Isaacson, C., Szivak, I., Weissbrodt, D.G., Pronk, W., 2016. Biological control of biofilms on membranes by metazoans. Water Res. 88, 20–29.

Koch, H., Lücker, S., Albertsen, M., Kitzinger, K., Herbold, C., Spieck, E., Nielsen, P.H., Wagner, M., Daims, H., 2015. Expanded metabolic versatility of ubiquitous nitrite-oxidizing bacteria from the genus Nitrospira. Proc. Natl. Acad. Sci. 112 (36), 11371–11376.

Langenheder, S., Jürgens, K., 2001. Regulation of bacterial biomass and community structure by metazoan and protozoan predation. Limnol. Oceanogr. 46 (1), 121–134.

Lücker, S., Wagner, M., Maixner, F., Pelletier, E., Koch, H., Vacherie, B., Rattei, T., Damsté, J.S.S., Spieck, E., Le Paslier, D., 2010. A Nitrospira metagenome illuminates the physiology and evolution of globally important nitrite-oxidizing bacteria. Proc. Natl. Acad. Sci. 107 (30), 13479–13484.

Maere, T., Verrecht, B., Moerenhout, S., Judd, S., Nopens, I., 2011. BSM-MBR: a benchmark simulation model to compare control and operational strategies for membrane bioreactors. Water Res. 45 (6), 2181–2190.

Massoud, M.A., Tarhini, A., Nasr, J.A., 2009. Decentralized approaches to wastewater treatment and management: applicability in developing countries. J. Environ. Manag. 90 (1), 652–659.

Matsumoto, S., Terada, A., Tsuneda, S., 2007. Modeling of membrane-aerated biofilm: effects of C/N ratio, biofilm thickness and surface loading of oxygen on feasibility of simultaneous nitrification and denitrification. Biochem. Eng. J. 37 (1), 98–107.

Meng, F., Chae, S.-R., Drews, A., Kraume, M., Shin, H.-S., Yang, F., 2009. Recent advances in membrane bioreactors (MBRs): membrane fouling and membrane material. Water Res. 43 (6), 1489–1512.

Nguyen, A.V., Pham, N.T., Nguyen, T.H., Morel, A., Tonderski, K., 2007. Improved Septic Tank With Constructed Wetland, a Promising Decentralized Wastewater Treatment Alternative in Vietnam. pp. 1–17.

Ossenbruggen, P.J., Spanjers, H., Klapwik, A., 1996. Assessment of a two-step nitrification model for activated sludge. Water Res. 30 (4), 939–953.

Peinemann, K.-V., Nunes, S.P., 2010. Membranes for Water Treatment. John Wiley & Sons.

Pronk, W., Ding, A., Morgenroth, E., Derlon, N., Desmond, P., Burkhardt, M., Wu, B., Fane, A.G., 2019. Gravity-driven membrane filtration for water and wastewater treatment: a review. Water Res. 149, 553–565.

Ruiz, G., Jeison, D., Chamy, R., 2003. Nitrification with high nitrite accumulation for the treatment of wastewater with high ammonia concentration. Water Res. 37 (6), 1371–1377.

Sládeček, V., 1983. Rotifers as indicators of water quality. Hydrobiologia 100 (1), 169–201.

Tang, X., Ding, A., Qu, F., Jia, R., Chang, H., Cheng, X., Liu, B., Li, G., Liang, H., 2016. Effect of operation parameters on the flux stabilization of gravity-driven membrane (GDM) filtration system for decentralized water supply. Environ. Sci. Pollut. Res. 23 (16), 16771–16780.

Tang, X.B., Ding, A., Pronk, W., Ziemba, C., Cheng, X.X., Wang, J.L., Xing, J.J., Xie, B.H., Li, G.B., Liang, H., 2018a. Biological pre-treatments enhance gravity-driven membrane filtration for the decentralized water supply: linking extracellular polymeric substances formation to flux stabilization. J. Clean. Prod. 197, 721–731.

Tang, X.B., Pronk, W., Ding, A., Cheng, X.X., Wang, J.L., Xie, B.H., Li, G.B., Liang, H., 2018b. Coupling GAC to ultra-low-pressure filtration to modify the biofouling layer and bio-community: flux enhancement and water quality improvement. Chem. Eng. J. 333, 289–299.

Van Dijk, L., Roncken, G., 1997. Membrane bioreactors for wastewater treatment: the state of the art and new developments. Water Sci. Technol. 35 (10), 35–41.

Vega, E., Lesikar, B., Pillai, S.D., 2003. Transport and survival of bacterial and viral tracers through submerged-flow constructed wetland and sand-filter system. Bioresour. Technol. 89 (1), 49–56.

Walters, E., Hille, A., He, M., Ochmann, C., Horn, H., 2009. Simultaneous nitrification/denitrification in a biofilm airlift suspension (BAS) reactor with biodegradable carrier material. Water Res. 43 (18), 4461–4468.

Wang, Y., Fortunato, L., Jeong, S., Leiknes, T., 2017. Gravity-driven membrane system for secondary wastewater effluent treatment: filtration performance and fouling characterization. Sep. Purif. Technol. 184, 26–33.

Wu, S., Austin, D., Liu, L., Dong, R., 2011. Performance of integrated household constructed wetland for domestic wastewater treatment in rural areas. Ecol. Eng. 37 (6), 948–954.

Wu, H., Fan, J., Zhang, J., Ngo, H.H., Guo, W., Hu, Z., Liang, S., 2015. Decentralized domestic wastewater treatment using intermittently aerated vertical flow constructed wetlands: impact of influent strengths. Bioresour. Technol. 176, 163–168.

Wu, B., Hochstrasser, F., Akhondi, E., Ambauen, N., Tschirren, L., Burkhardt, M., Fane, A.G., Pronk, W., 2016. Optimization of gravity-driven membrane (GDM) filtration process for seawater pretreatment. Water Res. 93, 133–140.

Wu, B., Christen, T., Tan, H.S., Hochstrasser, F., Suwarno, S.R., Liu, X., Chong, T.H., Burkhardt, M., Pronk, W., Fane, A.G., 2017. Improved performance of gravity-driven membrane filtration for seawater pretreatment: implications of membrane module configuration. Water Res. 114, 59–68.

Wu, B., Soon, G.Q.Y., Chong, T.H., 2019. Recycling rainwater by submerged gravity-driven membrane (GDM) reactors: effect of hydraulic retention time and periodic backwash. Sci. Total Environ. 654, 10–18.

Yoo, H., Ahn, K.-H., Lee, H.-J., Lee, K.-H., Kwak, Y.-J., Song, K.-G., 1999. Nitrogen removal from synthetic wastewater by simultaneous nitrification and denitrification (SND) via nitrite in an intermittently-aerated reactor. Water Res. 33 (1), 145–154.