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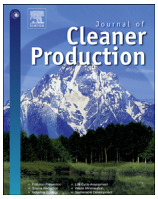

Biological pre-treatments enhance gravity-driven membrane filtration for the decentralized water supply: Linking extracellular polymeric substances formation to flux stabilization

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ABSTRACT

Gravity-driven membrane (GDM) filtration is a promising technology for decentralized drinking water treatment due to its low energy requirements and simple operation. The reduced flux observed in GDM systems relative to other membrane treatment techniques remains a vital obstacle to their wider application. With the goal of further improving membrane flux and permeate quality, three simple and practical technologies have been examined as pre-treatments for GDM systems, (i) slow filtration with granular activated carbon (GAC), expected to remove both biodegradable and non-biodegradable contaminants, (ii) slow filtration with modified fiber ball (MFB), expected to promote biodegradation of organic compounds and (iii) microfiltration (MF), expected to reject suspended substances and particles. Results indicated that stable permeability increased by approximately 230%, 150% and 100% in GDM systems employing GAC, MFB and MF pre-treatments compared to the control. The dissolved organic compounds (DOC) were removed by approximately 60% and 30% in GAC/GDM and MFB/GDM, while MF/GDM and GDM control were not effective at DOC removal. Correlations between the stable flux and extracellular polymeric substances (EPS) concentration ($R^2 > 0.9$) indicated that reduction of EPS should be responsible for the flux improvements. Tracing the fate of florescent foulants illustrated that the EPS was mainly secreted/converted by the microbes colonizing within the bio-fouling layer of the GDM system. During long-term filtration, assimilable organic compounds (AOC) which can serve as nutrients for the growth of microbes within the bio-fouling layer were efficiently consumed (>50%) by the biological pre-treatments. This reduction in AOC was linked with a reduction in the growth/activity of bacteria within the bio-fouling layer of GDM system and a reduction in EPS accumulation. Overall, biological pre-treatments can significantly enhance the flux and water quality produced by a GDM system without significantly increasing the operation and maintenance. These improvements in flux and water quality can hopefully spark wider adoption of GDM as an economical and environmentally-friendly technology for decentralized drinking water treatment.

1. Introduction

Membrane fouling and the complexity of operation and maintenance have presented grand challenges for membrane-based water treatment in a decentralized setting. Recently, gravity-driven membrane (GDM) filtration has emerged as a potential membrane operation strategy that is better suited for decentralized treatment. GDM process has been widely investigated in surface water treatment (Peter-Varbanets et al., 2010), grey water treatment (Ding et al., 2016a), rainwater recycling (Ding et al., 2017b), sea water pre-treatment (Wu et al., 2016) and wastewater effluent treatment (Wang et al., 2017). In GDM filtration, the ultrafiltration (UF) membrane is driven by an ultra-low gravitational pressure (40–100 mbar) without any backwash or chemical cleaning and the

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flux can remain constant (typically 5–10 L m^{-2} h^{-1}) during long-term filtration, endowing it with many inherent advantages compared to the traditional UF filtration, e.g. low energy supply, simple operation and low maintenance (Tang et al., 2016).

Noticeably, the flux stabilization in GDM filtration should not be confused with the "critical flux", both of which are well-differentiated concepts in their definitions and measurements. The "critical flux" was obtained during a short-term filtration in absence of long-term assessments, and can only be found in a cross-flow based membrane filtration and cannot be observed in a dead-end filtration (Bacchin et al., 2006). Furthermore, the "critical flux" just refers to the physical or physiochemical process without any involvements of biological activity. Unlike cross-flow systems where "critical flux" is applicable, GDM systems are configured in a dead-end filtration orientation, and do not require backwash, cross-flow or chemical cleaning, even during long-term operation. The flux gradually stabilizes after approximately one-week due to the biological activity induced formation of a bio-fouling layer with porous and heterogeneous structures on the membrane surface. A significantly improved stable flux can be achieved via enhancing the predation by higher organisms within the bio-fouling layer which results in a more porous and heterogeneous structure (Derlon et al., 2012).

The stabilized flux of a GDM process is based on a double-layer structure, including the bio-fouling layer and UF membrane. Peter-Varbanets et al. (2011a) even elucidated that the properties of the membrane did not significantly influence the operating flux following stabilization, highlighting the importance of understanding the bio-fouling layer. However, most previous studies (Derlon et al., 2013; Klein et al., 2016) investigating the bio-fouling layer in GDM treatment focused on the influence of biofilm structures on the flux stabilization, and how to manipulate the structures of biofilm to obtain a higher flux. Few studies (Ding et al., 2016b; Tang et al., 2018) investigated the impacts of the extracellular polymeric substances (EPS) on the stabilized flux. As well understood, EPS plays a fundamental role in the membrane fouling due to its preference to form ligand complexes with metals or other cations (Kavitha et al., 2014; Lee et al., 2008). Furthermore, EPS can also impact the compressibility and structures of cake layer to make it denser and more complex (Lin et al., 2014). Zhou et al. (2015) even suggested that the EPS produced by the microbial cells deposited in the cake layer were the dominant foulants observed in a long-term membrane filtration system. In GDM filtration, a previous study (Tang et al., 2018) also expounded that EPS accumulated in the bio-fouling layer and deposited in the membrane pores conferred a severe flux reduction. Recent investigation (Desmond et al., 2018) further pointed out that the meso-scale physical structures of bio-fouling layer was apparently determined by the composition of the EPS matrix. Another study (Ding et al., 2017c) even elucidated that the higher concentration of EPS facilitated a denser fouling layer in GDM system, which should be responsible for the lower flux and severe membrane fouling. In addition, the EPS formation is an inevitable process due to the toleration of bio-fouling layer on the membrane surface in GDM long-term filtration. Therefore, it's of great significance to gain a deeper understanding of the role of EPS (proteins, polysaccharides and florescent foulants) in GDM filtration, and develop some new approaches to reduce the EPS production and improve the permeability of GDM systems.

Granular activated carbon (GAC) filtration has been regarded as a sustainable and environmentally-friendly technology for drinking water treatment and has been extensively applied to improve the permeate quality and mitigate membrane fouling in membrane-based systems (Rasouli et al., 2017). Naidu et al. (2013) found that a GAC filter can significantly remove dissolved organic compounds (DOC) (especially the low-molecular weight (low-MW) compounds) and assimilable organic compounds (AOC). In addition, GAC pre-treatment can observably remove the biopolymers (proteins and polysaccharides) prior to UF filtration to mitigate the membrane fouling (Hallé et al., 2009). Another previous study (Tang et al., 2018) further revealed that coupling GAC to GDM systems significantly reduced the accumulation of organic foulants and engineered an apparently porous structure of bio-fouling layer, resulting in a higher stable flux.

Modified fiber ball (MFB) filtration has been widely applied due to its simple operation, low maintenance and efficient removal performance, which is proposed as an appropriate approach to minimize the fouling of UF filtration (Huang et al., 2011). Suspended substances, such as particles and colloids can be effectively removed by MFB filtration (Gao et al., 2012). Wu et al. (2016) indicated that the MFB filtration prior to the GDM process can efficiently improve both flux and permeate quality.

In addition, microfiltration (MF) has also been increasingly implemented in water treatment because it also presents and efficient barrier to particulate and colloidal contaminants (Rasouli et al., 2017). Zhang et al. (2013) reported that MF membrane can reject the carbohydrates, proteinaceous and algal organic substances which have been regarded as the typical membrane foulants. El Rayess et al. (2011) further indicated that MF process can efficiently retain the unicellular eukaryotic micro-organisms (i.e. yeasts) to improve the microbiological stabilization.

Numerous studies demonstrated that GAC, MFB and MF filtration, each conferred efficient improvements when paired with traditional UF processes, which are also expected to confer improvements when paired with GDM processes. It's hypothesized that the adsorption/biodegradation of GAC and the biodegradation exhibited by MFB treatment, will significantly consume nutrients (e.g. AOC) from the water which may limit the development of biofilm in the GDM system, production of EPS and ultimately hydraulic resistance. Besides, MF treatment can be anticipated to efficiently reject the particles and high-MW organics, which may also limit the development of the biofilm in GDM system. The flux, fouling behavior and achieved water quality were characterized. The mass conversion of constituents in the water and EPS formation were investigated based on the fate of florescent compounds among the feed water, permeate and bio-fouling layer. Besides, the respective roles of different pre-treatments in impacting the EPS accumulation and structures of bio-fouling layer were estimated accordingly. Finally, potential mechanisms of flux improvement of GDM in the nutrient-limiting conditions were discussed.

2. Materials and methods

2.1. Characteristics of raw water

Songhua River water was used as the feed water to evaluate the impacts of different biological pre-treatments on the long-term performance of GDM filtration. The raw water qualities are listed as below: turbidity: 3–10 NTU (approximately 30–100 NTU during rainstorms); DOC: 3–7 mg L^{-1}; TOC: 4–8 mg L^{-1}; ammonia nitrogen (NH_3-N): 0.3–1.5 mg L^{-1}; nitrite (NO_2^{-1}-N): 0–0.11 mg L^{-1}; nitrate (NO_3^{-1}-N): 0.63–2.25 mg L^{-1}; dissolved oxygen (DO): 5–8 mg L^{-1}; temperature: 4–30 °C; pH: 6–8.

2.2. Experimental setups

The GDM systems in presence/absence of pre-treatments are presented in Fig. S1, mainly including a raw water tank, pre-treatment tank, GDM tank, overflow tank, permeate tank, recirculation tank and pump. The Songhua River water was added daily into the raw water tank and then gravitationally flowed into the