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Correlations of relevant membrane foulants with UF membrane fouling in different waters

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ABSTRACT

Correlations between potential fouling-relevant substances and membrane fouling during ultrafiltration (UF) of different waters were investigated, including water samples from Lake Tegel, from a Berlin canal (Landwehrkanal) and from a wastewater treatment plant (WWTP) secondary effluent. The biopolymers quantified with liquid chromatographyorganic carbon detection (LC-OCD) showed a remarkable correlation with UF membrane fouling for all the three water sources at different seasons. This finding suggests that the biopolymer content in water can be employed as a universal indicator for predicting membrane fouling potential in UF processes. The particulate matter in the two surface waters Lake Tegel and Berlin canal, as characterized by suspended solids and turbidity, also exhibited a distinct correlation with UF membrane fouling, although its correlation was slightly weaker than that of biopolymers. However, the humic substances, which are generally believed to be major membrane foulants, did not show any reliable correlation with the UF membrane fouling of the different waters. This work may provide useful information for the development of optimized fouling control strategies for sustainable UF operation.

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1. Introduction

Ultrafiltration (UF) has been recognized as an established technology in drinking water treatment [\(Cai and Benjamin,](#page--1-0) [2011;](#page--1-0) [Huang et al., 2007](#page--1-0); [Kimura et al., 2004](#page--1-0); [Liu et al., 2011](#page--1-0); [Yang et al., 2011\)](#page--1-0), due to the development of membrane materials and the decrease of costs in the past decade. UF can effectively remove colloids, particles, pathogenic microorganisms, and viruses from water [\(Peter-Varbanets et al., 2009\)](#page--1-0), thus demonstrating the potential to replace the whole traditional treatment chain for drinking water production (coagulation/sedimentation, sand filtration, and disinfection). In the past decade, UF has also been extensively employed for the treatment and reuse of secondary effluent from wastewater treatment plants (WWTPs) ([Hatt et al., 2011a;](#page--1-0) [Huang et al.,](#page--1-0) [2012;](#page--1-0) [Zheng et al., 2011\)](#page--1-0), to address the problem of global water shortage. However, membrane fouling encountered in both drinking water treatment and reclamation of WWTP secondary effluent still remains as a major obstacle that restricts the more widespread application of UF ([Laabs et al.,](#page--1-0) [2006;](#page--1-0) [Peiris et al., 2010](#page--1-0)).

Operationally, membrane fouling can be classified as hydraulically reversible and irreversible fouling. The hydraulically reversible fouling, which is loosely attached to

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the membrane and can be eliminated through permeate backwashing, leads to the reduction of membrane productivity and increase of operational costs ([Huang et al., 2007;](#page--1-0) [Peiris et al., 2011](#page--1-0)). The hydraulically irreversible fouling, which is tightly bound to the membrane and can only be removed by chemical cleaning, results in not only the increase of operational complexity, but also the reduction of membrane lifetime [\(Peldszus et al., 2011](#page--1-0); [Raffin et al., 2011a](#page--1-0)). To overcome the fouling issue during UF process, a variety of pre-treatment technologies have been proposed and investigated, including coagulation, adsorption, pre-oxidation and pre-filtration, etc. ([Huang et al., 2009](#page--1-0); [Gao et al., 2011](#page--1-0)).

For selecting suitable pre-treatment methods to improve UF performance, first of all, it is necessary to identify the major membrane foulants. Generally, natural organic matter (NOM) in surface water and effluent organic matter (EfOM) in WWTP secondary effluent are considered as the most relevant membrane foulants in UF processes. In NOM/EfOM, the fraction of humic substances (HS) has been considered as severe membrane fouling substances, possibly because HS constitute the main part of NOM/EfOM [\(Zularisam](#page--1-0) [et al., 2006](#page--1-0)). With the development of innovative analytical techniques,NOM/EfOM fractions can be separated more efficiently and detected more precisely according to their physical and chemical properties. Among these advanced characterization techniques is the high performance liquid chromatography with on-line organic carbon detection (LC-OCD) [\(Huber et al., 2011\)](#page--1-0).

By LC-OCD, the NOM/EfOM can be separated into certain fractions of different molecular sizes and functional groups, including biopolymers, humic substances, building blocks, low molecular-weight acids and neutrals. The "biopolymer" fraction as quantified with LC-OCD, have recently been identified as major membrane foulant in some individual cases, although the concentration of biopolymers was found to be much lower than that of HS ([Zheng et al., 2009](#page--1-0)). This is in accordance with the observations of [Costa et al. \(2006\)](#page--1-0) and [Jermann et al. \(2007\),](#page--1-0) who found that the colloidal organic matter (i.e. biopolymer component) plays a crucial role in determining the membrane fouling potential during UF process.

Recently, the synergistic fouling effect of particulate matter and different organic fractions such as HS and biopolymers on UF membranes has also been evaluated and emphasized using model substances [\(Contrerasa et al., 2009;](#page--1-0) [Jermann et al., 2008\)](#page--1-0). Therefore, particulate matter in water is also assumed to be a relevant membrane foulant, although particles alone cannot severely decrease the permeability of UF membranes. However, the extents by which the particles contribute to membrane fouling in different "real" waters remain unclear.

This study aims to investigate the relationship between relevant membrane foulants and UF membrane fouling for different water sources $-$ lake water, river water and WWTP secondary effluent at different seasons, to check if there are common correlations of relevant foulant components $$ biopolymers, HS and particulate matter with the membrane fouling potential of different waters regardless of seasonal changes, with the purpose to better elucidate the contribution of different components to UF membrane fouling.

2. Materials and methods

2.1. Raw waters

Water samples from three typical water sources were used in the investigation: (i) Lake Tegel, which serves as a drinking water source for Berlin via bankfiltration; (ii) Berlin canal (Landwehrkanal), a canal river that flows through the city of Berlin and is impacted by treated municipal effluents; and (iii) WWTP secondary effluent from the Ruhleben WWTP in Berlin. These waters represent three typical water sources for a city. Raw water samples were collected from these waters monthly during the year 2011, for the UF experiments and corresponding water quality analyses.

2.2. UF experimental setup

Unstirred cell test was employed to evaluate the fouling potential of different waters on UF membrane. The cell (Amicon 8200, Millipore, USA) has a volume of 200 mL and an effective filtration area of 28.7 $cm²$. With an attached feed reservoir, up to 4 L of water can be filtered in every run of the UF experiments. The filtration test was performed in dead-end mode at room temperature (~22 °C), with a constant transmembrane pressure (TMP) of 1.0 bar. During the experiments, the permeate flux was recorded using an electronic balance connected to a computer.

For each UF experiment, a new hydrophilized polyethersulphone (PES) UF membrane (NADIR[®] UP150 P, MWCO 150 kDa, Germany) was used. Prior to the filtration test, each membrane was soaked in ultrapure water for 24 h and then rinsed thoroughly by filtering at least 2 L ultrapure water, to remove organic residues and wetting agents on the membrane.

2.3. Determination of membrane fouling

Two sets of UF experiments were conducted for each water sample: direct UF of the raw water sample and UF of the prefiltered water sample using a $0.45 \mu m$ membrane. In the former UF experiment, the "total fouling resistance" caused by the synergy of organic matrix and particulate matter in the water sample can be obtained. In the latter UF experiment, the "organic fouling resistance" can be obtained, assuming that the particulate matter in water has already been removed by the 0.45 μ m membrane. The difference between "total fouling resistance" and "organic fouling resistance" was designated as the "particle fouling resistance", which pertains to the contribution of particles to the synergistic fouling of organic matrix and particulate matter on UF membrane, but not the fouling resistance caused by particles alone.

Immediately before the filtration test, pure water flux of the UF membrane was determined by filtration of 150 mL ultrapure water. Subsequently, 500 mL of the water sample was filtered, with the permeate volume automatically recorded by a data acquisition system. After each trial, the UF membrane was backwashed with 50 mL ultrapure water (at a TMP of 1.0 bar) to remove the reversible fouling accumulated on the membrane. Then, pure water flux of the membrane

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