



Upgrading coagulation with hollow-fibre nanofiltration for improved organic matter removal during surface water treatment



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ABSTRACT

Rising organic matter concentrations in surface waters in many Nordic countries require current drinking water treatment processes to be adapted. Accordingly, the use of a novel nanofiltration (NF) membrane was studied during a nine month period in pilot scale at a large drinking water treatment plant in Stockholm, Sweden. A chemically resistant hollow-fibre NF membrane was fed with full scale process water from a rapid sand filter after aluminum sulfate coagulation. The combined coagulation and NF process removed more than 90% of the incoming lake water dissolved organic carbon (DOC) (8.7 mg C L^{-1}), and 96% of the absorbance at 254 nm (A_{254}) (0.28 cm^{-1} incoming absorbance). Including granulated active carbon GAC filter, the complete pilot plant treatment process we observed decreases in DOC concentration ($8.7\text{--}0.5 \text{ mg C L}^{-1}$), SUVA ($3.1\text{--}1.7 \text{ mg}^{-1} \text{ L m}^{-1}$), and the average nominal molecular mass ($670\text{--}440 \text{ Da}$). Meanwhile, water hardness was practically unaffected ($<20\%$ reduction). Humic substances (HS) and biopolymers were almost completely eliminated ($6510\text{--}140$ and 260 to $10 \text{ } \mu\text{g C L}^{-1}$ respectively) and low molecular weight (LMW) neutrals decreased substantially ($880\text{--}190 \text{ } \mu\text{g C L}^{-1}$). Differential excitation emission matrices (EEMs), which illustrate the removal of fluorescing organic matter (FDOM) over a range of excitation and emission wavelengths, demonstrate that coagulation removed $35 \pm 2\%$ of protein-like material and $65 \pm 2\%$ of longer emission wavelength, humic-like FDOM. The subsequent NF treatment was somewhat less selective but still preferentially targeted humic-like FDOM ($83 \pm 1\%$) to a larger extent than protein-like material ($66 \pm 3\%$). The high selectivity of organic matter during coagulation compared to NF separation was confirmed from analyses with Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR-MS), and liquid chromatography with organic carbon detection (LC-OCD), as coagulation exclusively targeted oxidized organic matter components while NF removed both chemically reduced and oxidized components. DOC removal and change in DOC character in the GAC filters showed marked differences with slower saturation and more pronounced shifts in DOC character using NF as pre-treatment. Fluorescence derived parameters showed a similar decrease over time of GAC performance for the first 150 days but also indicated ongoing change of DOM character in the post NF GAC filtrate over time even after LC-OCD indicated steady state with respect to outgoing carbon. During our trial iron concentrations were low ($<30 \text{ ppb}$) and thus A_{254} could be directly related to the concentration of HS ($R^2 = 0.9$). The fluorescence derived freshness index ($\beta:\alpha$) proved to be an excellent variable for estimating the fraction of HS present in all samples. Given the recommended limit of 4 mg L^{-1} for chemical oxygen demand (COD) for Swedish drinking water, coagulation will need to be supplemented with one or more treatment steps irrespective whether climate change will lead to drier or wetter conditions in order to maintain sufficient DOC removal with the current increasing concentrations in raw waters.

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1. Introduction

Rising levels of dissolved organic matter (DOM) in boreal and north European surface waters (Hongve et al. 2004) pose a number of technical and chemical challenges for drinking water production. Water treatment costs have increased and are expected to continue to rise, especially when using coagulation techniques which require higher chemical doses that results in more sludge (Eikebrokk et al. 2004). In addition, climate change is expected to lead to larger fluctuations in dissolved organic carbon (DOC) concentrations, commonly used as a proxy for DOM, and thus, further degradation of raw water quality in the future (Delpla et al., 2009). Intensity, timing and duration of fluctuations in DOC concentration and composition in surface waters are of vital economic interest for drinking water producers but difficult to foresee. DOC removal through coagulation treatment, which is the most commonly used method at Swedish water treatment plants (WTPs), is affected by organic matter composition and the removal efficiency is commonly reported to increase with the DOC-normalized absorbance at 254 nm (SUVA), an indicator of aromatic carbon and a terrestrial origin (Matilainen et al. 2010; Weishaar et al. 2003). The fraction of terrestrially derived DOC decrease with water turnover time in lakes (Gondar et al. 2008; Tang et al. 2013; Köhler et al. 2013), leading to lowered removal efficiency during the coagulation treatment. Lakes with varying turnover times are thus more prone to temporal changes in DOC character. This is the case for Mälaren, the third largest lake in Sweden, which is used as raw water source for three large WTPs supplying the Swedish capital city Stockholm with drinking water. Raw water DOC concentration is currently approximately 9 mg C L^{-1} , of which only roughly half can be removed through coagulation treatment. Lake water retention time has been identified as a key driver of DOM quality in this lake (Köhler et al. 2013; Lavonen et al. 2015). In Sweden, WTPs have to comply with a recommended limit of around 4 mg C L^{-1} organic matter measured as chemical oxygen demand (COD) for drinking water. In the case of Mälaren this is equivalent to 5 mg C L^{-1} DOC. Future climate or land use driven changes in DOC concentration and composition are thus large challenges for drinking water producers. Lower outgoing DOC will lead to decreased unwanted consumption of disinfectant and, reduced microbial regrowth potential in the distribution network. Improved DOC removal prior to treatment with granular activated carbon (GAC) filters may furthermore decrease fouling by irreversibly bound DOM components which diminish the ability of GAC filters to remove micro-pollutants such as perfluorooctanesulfonic acid (PFOS), algal degradation products or fuel residues, all which may occur in Mälaren.

High pressure ($>10 \text{ bar}$), NF membranes with low molecular weight cut-off ($<300 \text{ Da}$) have been used for a number years to remove organic matter for drinking water purposes (Meylan et al. 2007). These tight membranes are efficient in removing DOC and hardness (e.g. Ca^{2+} , Mg^{2+} , $>80\%$ removal) as well as a number of organic micro-pollutants (Zhang et al. 2006). Commercially available spiral wound NF membranes are designed for DOC removal at the expense of undesirable retention of hardness for drinking water production from soft raw waters. Furthermore, the spiral wound membranes are characterized by low chlorine stability, limited disinfection and chemical cleaning possibilities, e.g. pH 3–8 for cellulose acetate filters as compared to pH 2–12 for polysulfonate (Regula et al. 2014). Intensive pre-treatment is necessary due to limited hydraulic cleaning options. Capillary, hollow fibre NF membranes have been applied for direct filtration of highly colored surface water during the last decade (Meylan et al. 2007). One of the latest concepts in NF for highly effective removal of organic matter, the so called Color Removal Package, is based on capillary

NF membranes, combining the chemical resistance of hollow fibre membranes with the organic carbon retention of spiral wound units (De Groot, 2015). These membranes are modified for enhanced organic matter removal and limited retention of bivalent metal ions from the feed water. As they are operated using outside in flow, they may be flushed inside out which is ideal when retrofitting an existing treatment scheme. They do not require pre-treatment other than 300 micron safety screen and can directly be fed with raw surface water. In summer 2013, a HFW 1000 membrane pilot plant was installed at Görväln WTP, situated in Stockholm, Sweden, to evaluate the organic matter removal and performance of NF filtration following conventional coagulation and rapid sand filtration. Organic matter quality and quantity in all steps from raw to drinking water were evaluated with a large range of analytical techniques. These included total and dissolved organic carbon (TOC, DOC), online ultraviolet and visible (UV-VIS) absorbance (250–700 nm), 3D fluorescence, Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR-MS) and liquid chromatography with organic carbon detection (LC-OCD).

This study aims to a) evaluate the performance of a retrofitted new generation HFW 1000 nanofilter membrane, b) study the selective removal of DOM fractions in the combined coagulation NF process and the currently used coagulation technique; c) identify DOM characterization techniques that are informative for validating membrane and GAC performance, and d) use the acquired data to estimate removal of DOC with varying composition in the studied raw water as a function of lake water retention time.

2. Material and methods

2.1. NF pilot plant

The membrane material is composed of sulfonated polyether sulfone. The presence of sulfonate groups on the benzene ring structure renders them hydrophilic and leads to a negative zeta potential at pH above 5. The zeta potential further decrease to around -20 mV with the raw water pH 7.5 used in our study and the membrane therefore effectively rejects molecules with negatively charged functional groups such as DOC (De Groot, 2015). The separating layer of the membrane is on the inside of the fibres, thus allowing operating inside-out. The internal hydraulic diameter of the membrane fibres is 0.8 mm and the molecular weight cut-off is approximately 1000 Da based on Dextran permeation.

The HFW 1000 NF membrane module is 0.20 m (8") in diameter and 1.54 m (60.5") long. It contained 40 m^2 of membrane surface area with the 0.8 mm fibres. The test module was equipped with surface flow collectors. The test facility (a QuickScan pilot plant, supplied by Pentair X-Flow) had a capacity of 600 L h^{-1} (permeate production). During the pilot trials the test membrane was continuously operated at feed flow rate of $1.2 \text{ m}^3 \text{ h}^{-1}$ with a flux range of around $15 \text{ L m}^{-2} \text{ h}^{-1}$, a cross-flow velocity of 0.5 m s^{-1} , an intermittent forward flushing interval of every 60 min and a recovery rate of 50%. We chose to protect the membrane and operate at this low recovery even if higher recoveries of up to 80 were successfully tested, With operating pressures around 4 bar and permeability rates of $10 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ the transmembrane pressure typically increased with 0.17 bar and the permeability decreased with $1.0 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ at low water temperatures of around 1–3 °C during a filtration period of 42 days. The cleaning procedure is explained in detail in the appendix.

2.2. Mälaren as drinking water source

Görväln WTP is located in the eastern part, close to the outlet of Lake Mälaren, Sweden, where water from a northern basin (16%)

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