



Understanding the potential for selective natural organic matter removal by ion exchange

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ARTICLE INFO

Article history:

Received 10 July 2018

Received in revised form

21 September 2018

Accepted 24 September 2018

Available online 27 September 2018

Keywords:

Ion-exchange

Organic carbon

Disinfection by-products

Coagulation

Natural organic matter

ABSTRACT

Dissolved organic carbon (DOC) removal from a river water source was investigated using ion exchange (IEX), coagulation and membrane filtration. This research linked the variable charge characteristics of the organic compounds present in the source water with removal by IEX and coagulation. The raw water charge density fluctuated considerably (between 5.4 and 10.7 meq mg⁻¹DOC) and controlled removal of the charge loading. Importantly, charge density was not correlated with the organic carbon concentration. The combined IEX and coagulation process reduced the specific DBP-FP (sDBP-FP) of the final water, with values as low as 18 µg mg⁻¹DOC for both haloacetic acids and trihalomethanes. IEX removed a particular fraction of NOM that 1) enhanced coagulation efficiency, providing increased removal of overall DOC; and 2) enabled coagulation to subsequently remove higher levels of specific components of NOM that have a high DBP-FP. The component of NOM removed by IEX that had a positive impact on coagulation was identified to be charged low molecular weight organic compounds of all hydrophobicity levels, resulting in a reduced specific DBP-FP compared to coagulation alone.

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1. Introduction

Natural organic matter (NOM) has increased in many surface waters across the world, which has impacted on the effective operation of drinking water treatment systems (Matilainen et al., 2010). In many cases this has increased the organics loading onto the works above its treatment capacity, meaning that existing assets struggle to remove enough NOM. In addition to aesthetic impacts, process fouling, biofilm formation, and bacterial regrowth in the distribution network, residual NOM can cause the formation of disinfection by-products (DBPs) when disinfectants are added to the water (Pramanik et al., 2017; Winter et al., 2018). Some DBPs are potentially harmful to humans and have been linked to cancer (Richardson et al., 2007). Trihalomethanes (THMs) and haloacetic acids (HAAs) are the DBPs which are most routinely studied. These compounds are typically the DBPs that are dominant by mass and are used as indicators of a wide range of other DBPs (Li and Mitch, 2018). Therefore, the concentration of these compounds in drinking water are regulated worldwide. In the US, THMs are limited to 80 µg L⁻¹ and HAA5 at 60 µg L⁻¹ while in the European Union THMs

are regulated at 100 µg L⁻¹, and a recent proposal for HAA9 of 80 µg L⁻¹ has been released (European Commission, 2018; USEPA, 1998).

Ion exchange (IEX) is an increasingly applied process for the reduction of NOM, particularly in circumstances where coagulation alone is unable to achieve the required level of NOM removal. In this process, an anionic exchange resin is used to remove negatively charged organic compounds by the exchange of a counter ion, usually chloride, from the resin surface. A large proportion of NOM carries a negative charge, mostly attributed to acidic functional groups mainly present in an ionic form at pH > 4 (Bolto et al., 2004).

IEX resins can be used in fixed columns or mixed in the water in suspended reactors in commercial systems such as suspended ion exchange (SIX) and magnetic ion-exchange (MIEX) processes (Cornelissen et al., 2009; Watson et al., 2015; Metcalfe et al., 2015). For NOM removal applications, suspended systems have proven to be more effective as a pre-treatment prior to coagulation. This configuration allows treatment of waters containing suspended solids and shorter contact times through increased rates of mass transfer (Kishore and Verma, 2006; Bazri and Mohseni, 2016). In the SIX process, the resin is suspended in the raw water and is separated after a defined contact time in a plug flow contactor. The used resin is regenerated before it is reinjected into the system

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(Shorney-Darby et al., 2014). A reduction in DBP formation potential (DBP-FP) in waters treated by IEX has been observed and some studies have found a similar or even superior reduction of DBP-FP compared to coagulation (Boyer and Singer, 2005; Metcalfe et al., 2015). However, reduction in both overall DBP concentration and the specific THM-FP (mg of DBP per g of dissolved organic carbon (DOC)) by IEX alone can be higher or lower than coagulation depending on the raw water composition (Metcalfe et al., 2015).

When IEX processes are used in combination with coagulation, higher removal of NOM and reduced DBP-FP compared to conventional coagulation has been consistently observed (Boyer and Singer, 2005; Shorney-Darby et al., 2014; Watson et al., 2015). This is because different groups of organic compound are being targeted by the two processes to provide higher combined removal of NOM (Mergen et al., 2009). Several studies have also shown that IEX combined with coagulation reduces the specific DBP-FP, and some have found a further reduction than that by optimised coagulation alone (Drikas et al., 2011), in certain cases even when there was no reduction in the specific DBP-FP after the IEX stage (Metcalfe et al., 2015).

Additionally, the amount of coagulant used has been shown to be considerably reduced in a combined IEX coagulation process (Crittenden et al., 2012) and an increase in floc strength was found by Jarvis et al. (2008). This suggests selective removal of organic compounds by IEX that improves coagulation performance. However, this may be occurring in a number of different ways and understanding these mechanisms is an important knowledge gap.

Investigations on DOC removal by IEX often look at MW and hydrophobicity to understand removal behaviour (Grefte et al., 2013). However, information linking NOM removal by IEX the charge characteristics of the organic compounds present in real water sources is often overlooked. This is important given that NOM charge density is seasonally variable and can change rapidly and may not always be coincidental with a change in UV₂₅₄ or DOC concentration, often the main way by which IEX dosing is controlled. In the current work the combined influence of NOM hydrophobicity, MW and charge were investigated to understand the selective removal of organics by IEX to determine the importance of variable charge load and charge density on the removal process.

2. Materials and methods

An IEX pilot plant was used consisting of suspended ion exchange (SIX process from PWN Technology, Netherlands), in-line coagulation and ceramic membrane filtration (CCMF) (Supplementary Information, Figure S1). This was compared with a full scale water treatment works (WTW) comprising coagulation and membrane ultrafiltration. Water feeding both plants was from a holding reservoir containing water from the river Dee at Invercarnie WTWs. This study was carried out between May and October 2017, following a commissioning and optimisation stage. Fresh resin was added to the SIX at the start of the study to enable understanding of the change in treatment performance as the resin was used multiple times. In this period, the pilot plant was operated with both units (SIX + CCMF). For a period of three weeks (14th August till 04th September) no SIX pre-treatment was used and raw water was directly coagulated and treated by the membrane (CCMF).

2.1. SIX and CCMF pilot plant

The process was run with and without the SIX process, enabling understanding of the influence of IEX on NOM removal by coagulation and membrane filtration. The IEX resin was a gel-type,

strongly basic anion exchange resin with an acrylic quaternary amine backbone in the chloride form (Lewatit S5128, Lanxess, Germany). The resin was selected based on a previous pre-screening process for the water source. Raw water contacted with the resin under plug flow conditions at a flow rate of 7 m³ h⁻¹. A resin concentration of 25 mL L⁻¹ was applied at a contact time of 30 min based on data from a preliminary study for the water. The resin dose, contact time and flow was kept constant for the duration of the study. The resin was separated from the water by a lamella contactor and regenerated with sodium chloride (25 g L⁻¹ Cl⁻). Following IEX, water was adjusted to pH 6.4 using caustic soda and sulphuric acid. Polyaluminium chloride (PACl) was added as a coagulant at a dose based on the UV transmittance (UVT) of the water from an online solids-compensated UVT unit (Spectro:lyser, S:CAN, Austria). The PACl had a specific gravity of 1.22, a basicity of 37.9% and an aluminium content of 5.3%. The coagulant dose was calculated from algorithms developed from jar testing of the water (Table S1). The average coagulant dose for SIX + CCMF was 2.13 mg L⁻¹ or 0.47 mg_{Al3+} mg_{DOC}⁻¹ (range 0.19–6.04 mg L⁻¹ or 0.07–0.82 mg_{Al3+} mg_{DOC}⁻¹) and for CCMF 3.73 mg L⁻¹ or 0.75 mg_{Al3+} mg_{DOC}⁻¹ (range 1.58–6.06 mg L⁻¹ or 0.44–1.19 mg_{Al3+} mg_{DOC}⁻¹). After in-line coagulation (ILCA), the water was filtered through a ceramic membrane (pore size 0.1 μm, surface area 25 m²) at a flux of 150–250 L m⁻² h⁻¹ (LMH). The membrane was backwashed (BW) after loading between 50 and 167 L m⁻² and underwent an enhanced BW (EBW) after 500–1500 L m⁻². The EBW was carried out using NaOCl (100 mg L⁻¹ free Cl₂) or H₂O₂ (100 mg L⁻¹, pH 2) (details in Table S1). Samples were taken after treatment by IEX and then following membrane treatment.

2.2. WTW

The WTWs operated at an average of 39 ML day⁻¹. Raw water was adjusted to pH 6.9 using lime. PACl was added as the coagulant and had the same properties as that used in the pilot plant. PACl dose was automatically optimised based on raw water colour, approximately 1.2 mg L⁻¹ Al³⁺ per 20 hazen, resulting in an average of 1.62 mg L⁻¹ or 0.37 mg_{Al3+} mg_{DOC}⁻¹ (range 0.35–3.34 mg L⁻¹; 0.22 to 0.70 mg_{Al3+} mg_{DOC}⁻¹). The coagulated water was filtered from out-to-in, using polyvinylidene difluoride S10V Evoqua membranes with a pore size of 0.04 μm. A total membrane surface area of 11,048 m² was available (6 cells, containing 396 elements each) allowing a flux of 21.4–30.2 LMH. Samples of raw water and permeate were taken for analyses.

2.3. Sample analysis

Samples for UVA₂₅₄ measurement were pre-filtered with a 0.45 μm syringe filter and analysed using a Hach DR6000 spectrophotometer (Hach Lange, Germany, 5 cm quartz cuvette cell). DOC was measured using a non-purgable organic carbon method on a Shimadzu TOC-L analyser (EQS 24H) or a Skalar Formacs HT TOC analyser. All samples were passed through a 0.45 μm filter prior to analysis. A series of XAD7 and XAD4 resin were used to separate the DOC into hydrophobic (HPO), transphilic (TPI) and hydrophilic (HPI) fractions using adapted methods described in Bond et al. (2010). HAA- and THM-FP were determined from pre-filtered (0.45 μm) samples diluted to 1 mg L⁻¹ DOC. Water was spiked with NaOCl to give 5 mg L⁻¹ free chlorine and stored for 7 days in the dark at 25 °C. After quenching with thiosulfate, the samples were analysed with Gas chromatography–mass spectrometry. Liquid chromatography–organic carbon detection (LC-OCD) was carried out to determine the relative molecular weight distribution of organic compounds in water as it passed through various treatment stages. Samples were analysed by Het Water

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