Ceramic microfiltration; a novel and compact process for the treatment of surface water

C. Rockey, D. C. Metcalfe, G. Galjaard, H. Shorney-Darby and J. Zheng

ABSTRACT

In-line coagulation adsorption (ILCA) followed by ceramic microfiltration (CMF) was tested at pilot scale and compared to a full scale traditional process consisting of coagulation and dissolved air flotation (DAF) followed by rapid gravity sand filtration (RGF), for treating a reservoir water source which is prone to high concentrations of algae. The ILCA CMF process was shown to remove 10–16% more dissolved organic carbon (DOC) and reduced disinfection by-product formation potential (DBPFP) by 9–13% in comparison to conventional treatment (optimised coagulation). ILCA effectively controlled membrane fouling allowing the ceramic membranes to be operated at high flux (200 l/m^{2}h) with low membrane fouling (0.9–1.9 kPa/day). A process comprising ILCA and direct ceramic microfiltration was shown to provide very stable treated water quality under a range of challenging conditions. Additionally, the process is more compact showing significant reductions (circa 60%) in footprint relative to a conventional DAF/RGF process.

Key words | ceramic membrane, disinfection by-product removal, DOC removal, in-line coagulation, liquid chromatography-organic carbon detection

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INTRODUCTION

South West Water (SWW) is responsible for the production of good, safe drinking water to approximately 1.7 million consumers in the South West of England, UK. The majority of this water (>90%) is sourced from surface water. These waters require careful pre-treatment to reduce levels of dissolved organic carbon (DOC), pathogens and particulate matter prior to disinfection.

This research was undertaken to investigate the application of ceramic membranes for the treatment of surface water sources heavily influenced by algae. The raw water source in question presents very challenging conditions for conventional treatment and requires a significant upgrade. Membrane filtration is compared to sand filtration (Lee & Kim 2014), and evaluating the capability of ceramic membranes in treating such sources was desirable as it could lead to improved water quality (including the provision of an absolute barrier for *Cryptosporidium*), a more compact treatment footprint and improved operability.

Levels of DOC in UK source waters have been increasing in recent decades (Ritson *et al.* 2014), linked to reduced acid deposition, increasing temperature, changes in land use and rainfall patterns and more frequent extreme weather events associated with climate change. Some of these factors also increase the risk of *Cryptosporidium* being present in treated water due to both an increased loading of the parasite in the raw water and an increased challenge to conventional pre-treatment associated with increased levels of turbidity and DOC.

Whilst conventional treatment consisting of coagulation, clarification and sand filtration can significantly reduce the risk of chlorine resistant pathogens and produce water low in turbidity and DOC (Bolto *et al.* 2004; Bond *et al.* 2011), these processes require careful operation and optimisation to remain effective at all times. Changes in raw water quality and weather patterns noted in the region led SWW to review its approach to surface water treatment

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at a number of sites to ensure water quality could be assured now and in the future. Building on a previous investigation into the application of ceramic microfiltration (Metcalfe *et al.* 2015, 2016), this research focused on understanding the potential of in-line coagulation to control membrane fouling and reduce DOC in the membrane permeate. Achieving a high sustainable flux (and therefore a low treatment process footprint) was an important element to establish if this approach could enable an upgrade of an existing treatment facility (College Water Treatment Works (WTWs), West Cornwall, UK). As a starting point SWW reviewed and restated drinking water treatment goals for new WTWs to include the following:

- Provision of an absolute barrier to *Cryptosporidium*.
- Enhancement of water quality, particularly DOC removal, disinfection by-product (DBP) reduction and effective removal of pesticide risks where present.
- Use of efficient design to realise improved operability (compact footprint, automation, reliability and robustness of processes).
- Sustainability (chemicals, energy, waste, life expectancy).
- Application of innovative, forward looking technology where applicable.

The two primary goals for the selection of treatment processes for the new WTWs were driven by changes in UK Regulations and consumer expectations with respect to drinking water quality; to provide an absolute barrier to *Cryptosporidium* and reduce DOC and DBP formation. The requirement to minimise physical footprint was a critical factor in any future treatment upgrade at the site in question, as available development land was very limited and the re-use of the existing foundations and building was economically desirable. The objectives of this research were to:

- determine the maximum sustainable flux of a ceramic membrane microfiltration process for treating the source water;
- compare DOC removal and the residual disinfectant by-product formation potential (DBPFP) of water treated by the pilot process against that achieved by the existing conventional process;
- estimate the potential reduction in physical footprint of a new membrane process versus a refurbished and upgraded conventional process.

MATERIAL AND METHODS

Raw water source

Both the pilot test facility and existing WTWs were supplied with the same raw water from Argal impounding reservoir near Falmouth, West Cornwall, UK. The reservoir can be described as a eutrophic lowland impounding reservoir and the typical raw water quality is indicated in Table 1. These data show the extremely high average algal concentrations encountered in this reservoir, primarily associated with cyanobacteria such as *Aphanizomenon flos-aquae* and *Oscillitoria* sp. At times the concentration of algal cells at the works inlet can exceed 1,000,000 cells/ml.

The DOC and UVA data indicate that the bulk organic compounds in this water source typically have a low specific UV absorbance (SUVA) of approximately 2.5 l/mg-m which is typical for algal dominated source waters.

Pilot plant

A containerised pilot facility consisting of in-line coagulation (ILCA) and ceramic microfiltration (CMF) (PWN Technologies, The Netherlands) was operated for an extended 7 month period. A simple flow schematic of the pilot plant is shown in Figure 1.

ILCA was tested using both polyaluminium chloride (PACl) and ferric sulphate (ferric) (Water Treatment

Table 1 | Typical raw water quality supplied to the pilot facility and WTWs

Parameter **Units**	**Algae total** **cell/ml**	**Turbidity** **NTU**	**Colour filtered** **mg/l**	**TOC** **mg/l**	**DOC** **mg/l**	**UVA at 254 nm** **UVA/cm**	**pH** **units**	**Alkalinity at pH 4.5 as HCO_3 total** **mg/l**
Average	233,000	10.9	12.0	5.7	5.5	0.13	7.9	27.7

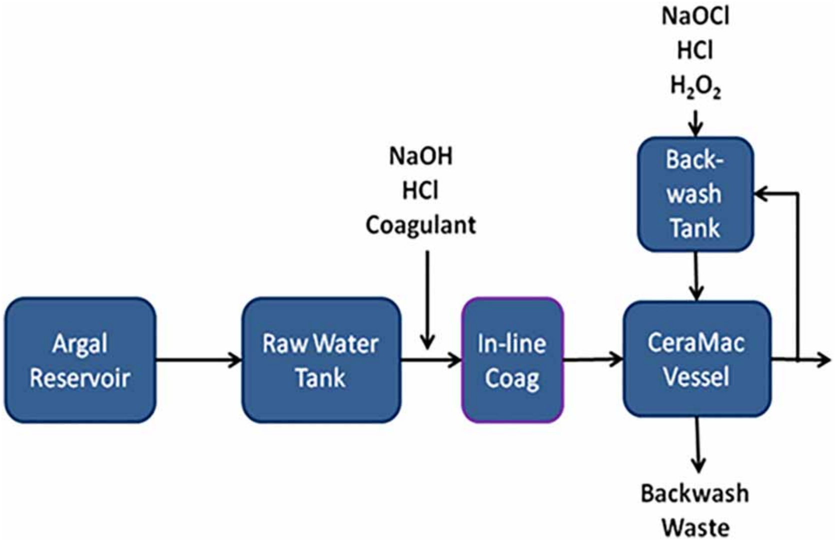

Figure 1 | Pilot facility process flow schematic.

Solutions, UK). Water was pH corrected with NaOH or HCl and injected with coagulant, mixed with a static mixer and flocculated for between 1.7 minutes (250 l/m^2 h (LMH)) and 2.9 minutes (150 LMH) prior to CMF. The coagulation pH was adjusted to 6.4 (for PACl) or 5.5 (for ferric). The coagulant dose was optimised for natural organic matter (NOM) removal by jar testing, with filtered UV absorbance at 254 nm (UVA) used as a surrogate for NOM removal.

CMF was carried out using one vertically mounted production sized 25 m^2 ceramic membrane element (Metawater, Japan, nominal pore size 0.1 µm), operating by dead end filtration. The membrane flux was varied between 150 LMH and 250 LMH. The membrane was backwashed with 75 l of membrane permeate at 4 bar pressure following a filtration load of 100 l/m^2. Periodically (between 1,000 and 1,600 l/m^2) the membrane was cleaned with a chemically enhanced backwash (EBW) using either 100 mg/l NaOCl or 100 mg/l H_2O_2/HCl at pH 2. During an EBW the chemical solution was pushed onto the membrane at 2 bar to displace the water within the membrane housing followed by a soak (the overall chemical contact time with the membrane was 10 minutes). A normal backwash with 75 l of membrane permeate at 4 bar ended the EBW sequence.

In between most tests the membrane was cleaned in place (CIP) with a recirculation with 1,000 mg/l NaOCl at pH 12 followed by an acid/H_2O_2 CIP (100 mg/l H_2O_2 at pH 2.7). Cleaning using these chemicals was consistently able to recover the membrane permeability to that of the virgin material. *Note: Test 2 (*Figure 2(b)*) was a continuation of Test 1 and therefore the membrane was not cleaned between these data sets*.

The pilot was sized to treat between 50 and 150 m^3/day and operated over an extended period under operational conditions suitable for water production, to ensure that the results of the study could be reasonably scaled up to predict future full scale performance.

Full scale WTWs

The WTWs (College WTWs, SWW, West Cornwall, UK) process consists of powdered activated carbon dosing (3–5 mg/l), coagulation using PACl (typical dose 2–5 mg/l as Al), coagulation pH adjustment with hydrated lime followed by dissolved air flotation (DAF) and rapid gravity sand filtration (RGF).

Laboratory analysis and disinfection by-product formation potential testing

Samples were collected from the pilot plant and the WTWs during stable operation of both systems for the conditions under test. UVA was measured using a Hach DR6000 spectrophotometer after samples were filtered

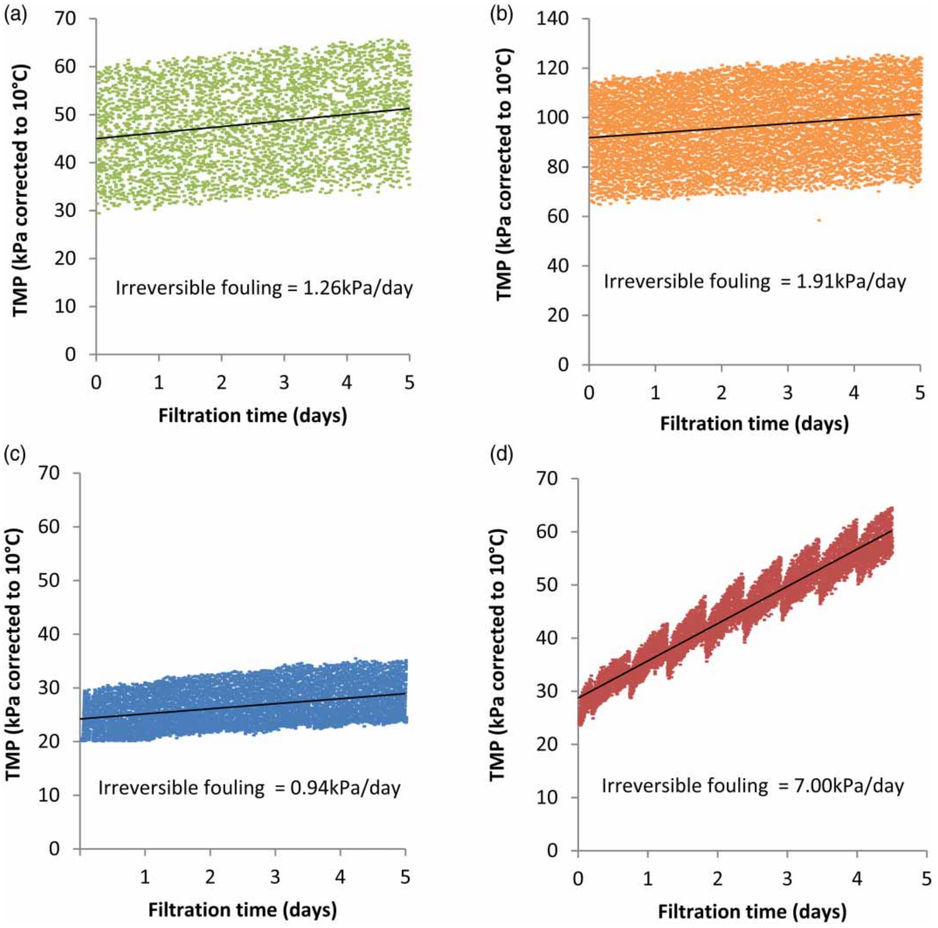

Figure 2 | Irreversible membrane fouling rates for: (a) Test 1–150 LMH relatively low algal conc., winter quality, (b) Test 2–200 LMH relatively low algal conc., winter quality, (c) Test 3–200 LMH high algal conc., autumn quality, (d) Test 4–250 LMH high algal conc., autumn quality. All tests were carried out using PACl coagulant and please note the scale in (b) is different.

through a pre-flushed 0.45 μm filter. Other analyses were undertaken using standard methods at SWW Laboratories (Exeter, UK).

Trihalomethane (THM) formation potential tests (THMFP) were performed at SWW Laboratories using an adapted version of the Standard Method 5710B (Eaton *et al.* 2005) from the American Public Health Association (APHA), as described by Metcalfe *et al.* (2015).

Liquid chromatography-organic carbon detection (LC-OCD) was performed at Het Water Laboratorium (The Netherlands). This analysis characterises the organic compounds into a series of different molecular weight (MW) fractions classified as biopolymers, humic substances, building blocks, low molecular weight (LMW) neutrals and LMW acids as described by Huber *et al.* (2011).

Irreversible membrane fouling rates (e.g. fouling not removed by routine physical and chemical cleaning) were assessed by linear regression of all trans-membrane pressure (TMP) data during the run with data from approximately 5 days of membrane operation.

RESULTS AND DISCUSSION

A series of extended tests were performed to establish the membrane fouling rates associated with different membrane flux rates whilst operating the ceramic membrane with ILCA pretreatment. The maximum sustainable flux was determined from the membrane critical flux data obtained during these tests. The maximum sustainable flux data were subsequently used to establish the approximate footprint required for the main process element to produce a maximum daily output of 12 million litres per day (MLD). This footprint was compared with the footprint required for a conventional 'high rate' DAF-RGF process to produce the same output. In addition, tests were performed to compare the treated water quality data (specifically DOC and disinfection by-product formation potential (DBPFP) removal) with the conventional treatment process.

The key findings of this research were that a maximum sustainable flux of 185 LMH could be obtained, whilst maintaining low fouling rates (<2 kPa/day). This flux led to a 55% reduction in the footprint of a full scale ILCA-CeraMac design when compared to a DAF-RGF design. The ILCA-CeraMac process also led to improved DOC removal and lower DBP formation potential.

Figure 2 summarises the irreversible fouling rates associated with some of the tests performed at different fluxes (150, 200 and 250 LMH) when treating water with moderate algal concentrations (10,000 cells/ml, winter water quality) and during periods of very high algal concentrations (500,000 cells/ml, autumn water quality).

These data indicate that at fluxes up to 200 LMH low fouling rates were achievable (1.25 kPa/day at 150 LMH and 1.91 kPa/day and 0.94 kPa/day at 200 LMH on the low and high algal laden source water respectively) with higher fouling rates observed when algal concentrations were low. Although this finding may be counterintuitive it can be related to the floc quantity and size which was observed to vary during the autumn and winter periods; significantly larger flocs and greater quantity of floc was seen during periods of algal challenge and these are likely to have resulted in the more rapid subsequent formation of a more permeable cake layer on the membrane surface.

Due to the short contact times within the ILCA, water temperature and the number of nucleation sites significantly affect floc formation. The permeability of the cake layer and the time prior to formation of a cake layer can affect the level of irreversible fouling. As a cake layer forms it has the ability to protect the membrane from unflocculated organic compounds or fine flocs/colloids which could otherwise plug the pores (Guigui *et al.* 2002; Dong *et al.* 2005). Additionally, when algal concentrations were low a greater pressure build-up between backwashes was observed indicating that the forming cake layer was less permeable in these circumstances. Greater pressure build-up during the run might lead to stronger interaction between foulants and the membrane increasing the rate of irreversible fouling. These factors likely explain the observations between autumn and winter fouling.

At the very high flux of 250 LMH the fouling rate was moderate and greater than could be tolerated for practicable and economic extended operation. The level of fouling at this flux was deemed acceptable for short periods of operation, for example during an emergency situation or unplanned asset failure in a period of peak or high demand.

Having reviewed the options regarding full scale design, (including the number of membrane elements within a vessel and the number of vessels) a maximum operating flux of 185 LMH with all vessels in service was deemed most appropriate and taken forward for the full scale design.

In addition to the data shown in Figure 2 where PACl was used as the coagulant, tests were performed to compare the membrane performance when using PACl or ferric. These tests showed that membrane fouling rates were similar with either coagulant suggesting that the coagulant used has little impact upon membrane fouling (0.63 vs 1.44 kPa/day PACl/ferric respectively at 150 LMH and 11.58 vs 10.56 kPa/day for PACl/ferric respectively at 250 LMH). The operational flexibility provided by the ability to choose from a range of coagulants depending on water quality, availability, waste disposal or economic drivers is an important consideration for utilities.

The removal of DOC was also assessed by LC-OCD and these data are illustrated in Figure 3. Almost complete removal of the highest MW organic compounds was noted for all of the different coagulation processes in keeping with previous results which have reported effective removal of this fraction by coagulation (Fabris *et al.* 2007; Humbert *et al.* 2007). LC-OCD analysis also revealed improved DOC removal by the pilot plant process in comparison with the WTW process and showed that ferric provided improved removal of organics relative to PACl. This is in keeping with the results of previous studies (Matilainen *et al.* 2005) where lower pH was shown to improve the removal of LMW organic compounds when using ferric coagulants.

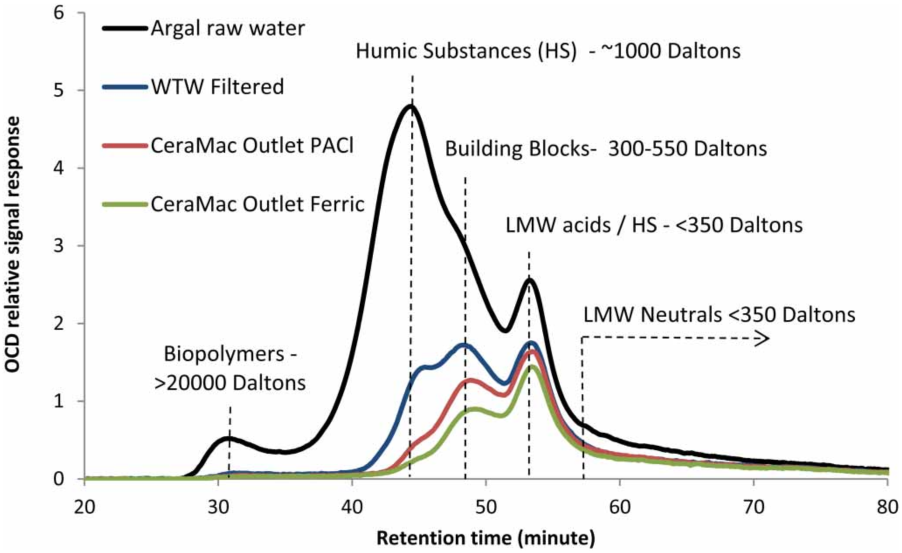

Figure 3 | Liquid chromatography-organic carbon detection (LC-OCD) chromatograph – initial raw water and residual organic fractions following different treatment approaches. Organic peak/fraction classification is shown for reference.

The improved removal of DOC by the pilot membrane process was primarily related to enhanced removal of the humic and building block fractions and smaller improvements in the removal of the LMW acids/HS fraction.

The conventional WTWs process achieved 55% removal of DOC (Figure 4) from the raw water indicating reasonable performance from the PACl and coagulation processes for the relatively low SUVA raw water.

DOC removal by the pilot CMF process was significantly higher than the conventional WTWs process when using either PACl (65%) or ferric sulphate coagulants (71%). The increased removal of DOC when operating with PACl, the same coagulant as the WTWs, could be attributed to the coagulant dose being better optimised for DOC removal, a small degree of additional adsorption of organic compounds within the cake layer, better mixing and/or the very fine filtration (<0.1 μm) provided by the membrane. Despite the slightly higher (c20%) PACl dose used in the pilot plant, the additional DOC removal by the ILCA/CMF process is noteworthy as it was achieved in the absence of PACl.

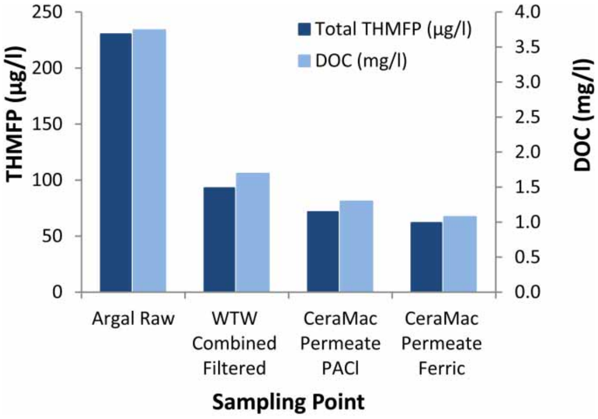

Figure 4 | THMFP and DOC in raw water, WTWs operating on PACl, pilot plant operating on PACl and ferric.

A reduction in THMFP of 60% was achieved by the WTWs process whereas the pilot process achieved 69 and 73% when using PACl and ferric coagulants respectively.

As the THMFP reductions were greater than the DOC removals, the coagulants were shown to be selectively removing UV absorbing organics, as has commonly been reported in previous studies of coagulation processes.

A comparison of the performance of the pilot plant processes against the baseline performance from the WTWs process (to establish likely improvements to treated water quality) revealed that DOC reductions of 23 and 37% and THMFP reductions of 23 and 33% were possible for PACl and ferric coagulant respectively. The specific reactivity of the residual organics was similar for all of the samples being between 55 and 58 μgTHM-mgC, therefore the reductions in THMFP were primarily attributable to the improved DOC removal by the membrane process.

Membrane integrity (pressure decay) tests were undertaken periodically throughout the pilot testing which indicated that the integrity of membrane remained for the duration of the study. The turbidity of the membrane permeate was also monitored by an on-line instrument (Swan, Switzerland) which also confirmed the integrity of the membrane with results being consistently <0.05 NTU. These turbidity data were supported by regular laboratory measurements where the level of turbidity was reported as less than the limit of detection in all but one sample.

IMPACT ON FULL SCALE DESIGN FOOTPRINT

One of the benefits of membrane processes is their compact footprint in comparison with conventional rapid gravity sand filters (RGFs). In addition, clarification processes require large footprints to enable sufficiently low rise rates for effective settlement. Higher rate DAF processes occupy a reduced footprint in comparison to conventional clarification, however the footprint is still significant.

Using an ILCA process hugely reduces the footprint required due to mitigating the need for a clarification step in the solids liquid separation stage and reducing the contact time required for mixing and flocculation as a settleable or floatable floc does not need to be formed (Meyn *et al.* 2012).

A footprint comparison between the main process elements for a conventional DAF-RGF process and an ILCA-CeraMac (CMF) process was undertaken for the College WTWs situation (Table 2). This assessment was based on a maximum output of 12 MLD with all process elements in service and a nominal works flow of 9 MLD with one RGF or membrane vessel out of service for maintenance. The designs indicated a reduction of 68% in footprint requirement for the main process units associated with the ILCA-CMF option. When ancillary equipment was taken into account a footprint reduction of 55% was realised.

A design flux of 185 LMH was used to achieve the maximum works output of 12 MLD which reduced to 136 LMH for the nominal flow (9 MLD) with all vessels in service. With one membrane vessel out of service the operating flux increased to 163 LMH. For the conventional design a flocculation time of 15 mins, and DAF and RGF loading rates of 10 and 6 m/h at nominal flow were used as the basis of the design.

Table 2 | Design footprint comparison between the main process elements of a DAF-RGF and an ILCA-CMF process for a 12 MLD WTWs

DAF-RGF option	**Area (m^2)**
Flocculation tanks and DAF cells	300
RGFs and filtered water channel	238
DAF-RGF ‘process footprint’	538
Ancillary equipment (flash mix tank, blowers, piping saturators, panels)	240
‘Total’ footprint – DAF-RGF	778
ILCA-CMF option	**Area (m^2)**
Membrane vessels (6 No. C19) + piping galleries front and back	90
ILCA contactor and CeraMac pumps	80
ILCA-CeraMac ‘process footprint’	170
Ancillary equipment (dosing sets, piping, pumps)	180
‘Total’ footprint (ILCA-CMF)	350
Comparison	**% Reduction**
% reduction ‘process’	68
% reduction total	55

CONCLUSIONS

Long term pilot testing established that high sustainable fluxes were possible using short contact time ILCA in front of CMF. A flux of c200 LMH resulted in an irreversible fouling rate of less than 2 kPa/day even during periods when the source water contained high concentrations of algae. For a process using ILCA in front of microfiltration membranes a settleable or floatable floc is not required. This reduces the time and infrastructure required for flocculation (Meyn *et al.* 2012) whilst not adversely affecting DOC removal due to coagulation reactions occurring rapidly.

Operation with both aluminium and iron based coagulants was possible without significant differences in membrane fouling rates, providing a valuable choice over which coagulant to use depending on specific local circumstances. The coagulant could be tailored to the raw water quality and/or other factors, such as the need to minimise disinfection by-product formation, local availability, cost and waste disposal options. Ferric coagulants are typically more effective for DOC and DBPFP removal at lower pH and this was also seen in this study. Further work to optimise ferric coagulation conditions (dose and pH) focusing on membrane performance and treated water quality could provide additional useful insight.

The primary treated water quality goals set at the start of the study were met and the treated water quality achieved in terms of DOC and DBPFP with the ILCA membrane process outperforming the conventional DAF-RGF process. Even where the same coagulant was used and despite a small dose of PACl being used in the conventional process, the ILCA membrane process produced water with a lower DBPFP and lower DOC.

The ceramic microfiltration membranes provide significant solids loading capacity and complete removal of suspended particles (online turbidity measurements typically 0.01–0.02 NTU, laboratory results typically <0.1 NTU limit of detection) therefore providing an absolute barrier against chlorine resistant pathogens, such as *Cryptosporidium*.

Membrane processes also lend themselves to automation and occupy a small footprint relative to conventional RGF approaches making them suitable for constrained sites and where the climate requires a high level of 'winterisation' (insulation/heating). Significant reductions in footprint (c60%) can be realised where membrane fouling is controlled sufficiently to allow for relatively high operational fluxes to be achieved.

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