

Comparison of optional process configurations and operating conditions for ceramic membrane MF coupled with coagulation/flocculation pre-treatment for the removal of NOM in drinking water production

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ABSTRACT

The increase of Natural Organic Matter (NOM) in natural water sources, partly caused by progressing climate change issues, is a growing concern for drinking water production. In recent years, membrane technology like Reverse Osmosis (RO) or Nanofiltration (NF) has been successfully applied as a robust solution for NOM removal to produce potable water. However, coagulation/flocculation pre-treatment, combined with Microfiltration (MF) ceramic membrane filtration is nowadays seen as an alternative, less-energy-consuming membrane process for NOM removal. In this study different coagulants have been used under varying coagulation/flocculation conditions to investigate the respective impact on membrane filtration performance.

Three alternative coagulation/flocculation configurations were compared. It is shown that NOM was efficiently removed independently on the chosen configuration or coagulant type. Similar and low membrane fouling rates were observed for all tests. Residual metal concentration was found to be the limiting permeate quality parameter, which limits the options of operating conditions. Furthermore, the compact inline pipe flocculator configuration has the potential of designing more compact full-scale units, using less space compared to conventional sand filtration units or even membrane filtration plants using classical tank coagulation/flocculation configurations.

Key words | ceramic membranes, coagulation, drinking water, flocculation, microfiltration, natural organic matter

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INTRODUCTION

Surface water is a common drinking water source. Approximately 90% of the population in Norway consumes drinking water which originates from surface water (80% lakes, 10% rivers). In many cases these waters are typically characterized by a high content of Natural Organic Matter (NOM), resulting in high colour, very low turbidity, low alkalinity and low hardness, due to the natural conditions (Ratnaweera *et al.* 1999). Table 1 gives an overview of typical raw water properties and the demands of the drinking water guidelines as published by The Norwegian Institute of Public Health. Colour is expressed

in Hazen units as Pt/L units and organic matter as TOC or COD in mg/L C or mg/L O, respectively.

Although the main part of NOM is not harmful, some fractions like algogenic matter can cause colour, taste and odour problems or are even toxic like the algae toxin microcystin. Special fractions of NOM in the drinking water can potentially lead to bacterial regrowth in the distribution system. Removal of NOM also reduces the load of subsequent treatment steps and facilitates the elimination of important trace compounds such as pesticides found in the NOM. Furthermore, these substances increase the

Table 1 | Typical raw water quality and drinking water guidelines in Norway (Nasjonalt folkehelseinstitutt 2004)

Raw water quality	Drinking water standards (limiting values)	Recommendations from authorities
High colour (30–80 mg Pt/L)	20 mg Pt/L	5 mg Pt/L ($VIS_{436} \approx 0.3 \text{ m}^{-1}$)
High TOC (3–8 mg/L)	5 mg/L	3 mg/L
High COD (4–8 mg/L)	5 mg/L	3 mg/L
Low turbidity (<1 NTU)	1 NTU at WTP 4 NTU at consumers	0.2 NTU
Low alkalinity (<0.5 meq/L)	No limiting value	0.6–1 meq/L
Low hardness (<5 mg Ca/L)	No limiting value	15–25 mg/L Ca
Aluminium and iron	200 µg/L for both	150 µg/L for both

necessary amount of disinfectants and are recognized as precursors for disinfection by product formation such as THMs, which can be carcinogenic. The Norwegian Institute of Public Health has subsequently provided limiting values and recommendations in the national drinking water guidelines. The removal of NOM is therefore one of the major concerns and therefore necessitates an advanced drinking water treatment.

There is a growing concern surrounding the increase of NOM in natural water sources, where recent studies have shown that the concentration of NOM in surface water might increase during the next decades, partly caused by progressing climate change issues. Due to increased precipitation in parts of Scandinavia during the last 30 years the water pathways in many catchments have changed, leading to an increased leaching of organic compounds from the upper forest floor to the lakes. In some areas a concentration increase of up to 50% of colour and DOC between 1983 and 2000/2001 has been reported (Forsberg 1992; Hongve *et al.* 2004).

Direct filtration of NOM by NF membranes has been successfully used for full-scale drinking water treatment in Norway (Ødegaard *et al.* 1999, 2000). Polymeric porous membranes, eventually coupled with an appropriate pre-treatment option for fouling minimization, such as coagulation, is an alternative process scheme that can be efficiently used for the removal of NOM and for the treatment of surface water (Lahoussineturcaud *et al.* 1990; Peuchot & Ben-Aim 1992; Kim *et al.* 2001; Machenbach 2005; Cho *et al.* 2006). Membrane filtration processes are commonly viewed as an energy-intensive treatment scheme

where more-energy-efficient, low-pressure membrane options may be preferred alternatives.

In this respect, ceramic microfiltration membranes are an interesting alternative to polymeric membranes. In addition, due to their physical properties, mechanical stability and resistance to chemicals, ceramic membranes have shown to be superior compared to organic membranes, particularly with respect to cleaning alternatives (Weber *et al.* 2003; Lee & Cho 2004). This should be considered especially since NOM was identified as a major foulant in surface water treatment (Yamamura *et al.* 2007; Kimura *et al.* 2008), causing irreversible fouling which can only be cancelled by extended chemical cleaning, not by backwashing.

Ceramic membranes allow an easy and complete recovery of membrane performance through a whole variety of cleaning procedures, using all kinds of chemicals if necessary, and show a potential for replacing polymeric membranes in drinking water treatment (Weber *et al.* 2003; Lee & Cho 2004; Lerch *et al.* 2005). Higher investment costs of ceramic compared to organic membranes can be diminished by operating ceramic membranes with higher fluxes and by considering higher lifetimes. Ceramic membranes are commonly manufactured as multichannel units, which promotes alternative membrane module designs and operating conditions.

This study has investigated the use of coagulation/flocculation, combined with MF ceramic membrane filtration as an alternative membrane process for the removal of NOM to produce potable water. Different coagulants have been used under varying coagulation/flocculation

conditions to investigate the impact on the membrane filtration performance. Two alternative process configurations for the pre-treatment stage were compared.

METHODS

Raw water

The raw water used in this study was prepared using a NOM concentrate from a full-scale ion exchange treatment plant, which was mixed with tap water to make up an analogue water with a turbidity of around 1 NTU, a colour of 50 ± 1.13 mg/L Pt at pH 7 (corresponding to an absorbance of 1.97 m^{-1} at a wavelength of 436 nm), a UV_{254} -absorbance of $23.8 \pm 0.8 \text{ m}^{-1}$ and a DOC concentration of 5.2 ± 0.3 mg/L. Analogue feed water ensures that the same experimental conditions apply for all experiments conducted. Previous studies have shown that this method can be successfully used and simulates real conditions quite well (Leiknes *et al.* 2004, 2005).

The NOM concentrate used in this study can be characterized as highly hydrophobic and humic acid like. This can be derived from the relatively high SUVA of 4.5 and is supported by the Fluorescence Excitation Emission Matrix (FEEM) shown in Figure 1(a). The NOM concentrate diluted in distilled water shows a very distinct peak at an excitation of 305 nm and an emission of 430 nm.

This falls in the humic acid like region, previously identified and classified in literature (Chen *et al.* 2003). The FTIR spectrum shown in Figure 1(b) is dominated by two broad peak regions. One lies between $2,900$ and $3,650 \text{ cm}^{-1}$ and includes bands of O-H stretching vibrations ($3,200$ – $3,550 \text{ cm}^{-1}$) and C-H stretching vibrations ($3,000$ – $3,340 \text{ cm}^{-1}$). The second active region is located between $1,300$ and $1,800 \text{ cm}^{-1}$ and contains signals of conjugated C=O stretching ($1,590$ – $1,750 \text{ cm}^{-1}$), C=C stretching ($1,585$ – $1,625 \text{ cm}^{-1}$) and C-H bending in methyl and ethyl groups ($1,355$ – $1,465 \text{ cm}^{-1}$). Since no very sharp and distinct peaks are visible it can be assumed that the NOM used in this study contains a variety of different organic compounds with varying functional groups.

Pilot plant configuration

Experiments were done with a membrane filtration pilot plant using a classical coagulation/flocculation pre-treatment configuration, shown in Figure 2(a). Prior to the membrane filtration unit, a two-tank coagulation/flocculation setup with a rapid mixing followed by a slow mixing step was used. After that, the coagulated water was filtered by the membrane unit. The pilot plant was equipped with three identical and independent process trains. A second option using an inline pipe flocculator was also investigated as an alternative to the classical configuration, shown in Figure 2(b).

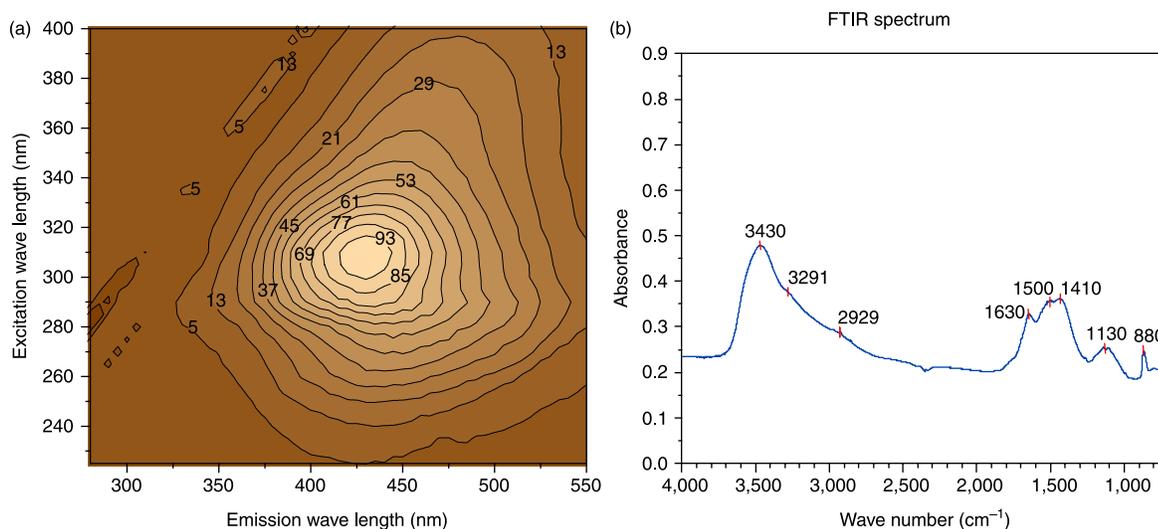


Figure 1 | NOM characterization.

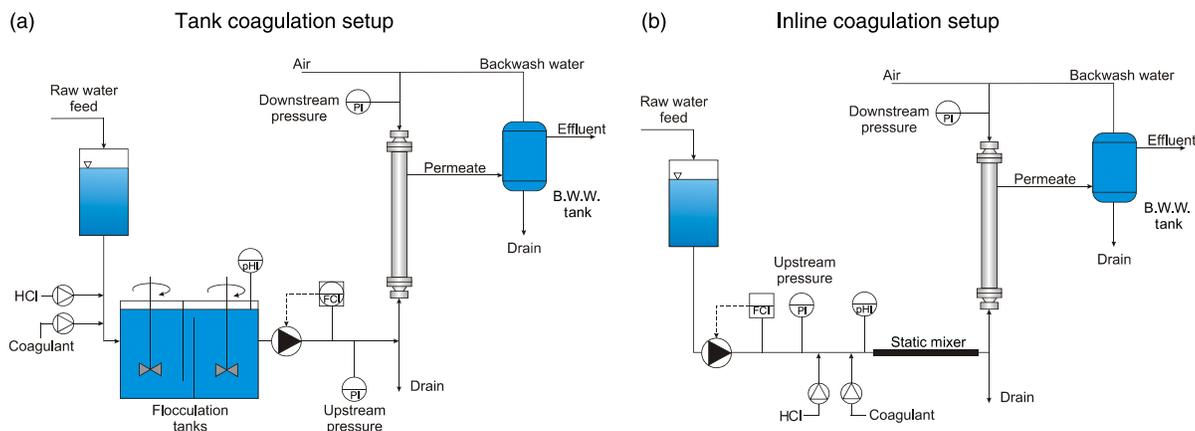


Figure 2 | Flow diagram of the different membrane filtration plant configurations.

In this study multichannel ceramic membranes, operated in dead-end, inside-out mode with a nominal pore size of $0.1 \mu\text{m}$ were investigated. Each membrane module had 55 channels, was 1 m long and had an effective area of 0.43 m^2 . The feed water was pumped in up-flow direction. The plant was operated in constant flux mode, maintaining a flux of $143 \text{ L m}^{-2} \text{ h}^{-1}$ (Table 2). Regular cleaning was performed each hour by an initial backwash just with pressurized permeate and then, after some seconds, by a simultaneous air blow into the feed channels. For the backwash a pressurized tank was used. The backwash pressure expands from initially 5 bar to about 2 bar at the end of the regular cleaning. In this way the amount of backwash water always remains the same. The pressure of the air blow is likewise 2 bar. In total the regular cleaning lasted about 10 seconds. The obtained sludge was discharged to the sewer system. The pilot plant also offers the possibility of chemical enhanced backwashing, e.g. by dosing acid, base or chlorine; however, these

Table 2 | Membrane operating conditions and module specification

Membrane type	Ceramic MF
Module length/diameter	1 m/0.03 m
Channels per module	55
Channel diameter	2.5 mm
Module area/nominal pore size	$0.43 \text{ m}^2/0.1 \mu\text{m}$
Operational flux	$143 \text{ L m}^{-2} \text{ h}^{-1}$
Regular cleaning intervals/procedure	1 h/backwash and air blow

options were not used or found necessary in the experiments conducted. After each experiment the membrane modules were cleaned intensively by soaking alternating in citric acid solution ($w = 1\%$) and sodium hypochlorite solution ($c = 3,000 \text{ ppm}$).

Two different coagulants were compared in this study, polyaluminium chloride (PAX-18) and iron chloride (PIX-111), both supplied by Kemira Chemicals. The process performance was assessed as a function of coagulant type, dosage and pH value. The DOC and colour removal, membrane fouling and the residual metal concentration in the permeate were measured for evaluation of the process performance. For the polyaluminium chloride (PACL) doses of 2, 3 and 5 mg Al/L were chosen, corresponding roughly to an alum dose between 0.4 and $1 \text{ mg Al per mg of DOC}$. The chosen iron doses were twice as high as for the alum (in order to have equivalent concentrations), 4, 6 and 10 mg Fe/L , corresponding to an iron dose range between 0.8 and $2 \text{ mg Fe per mg DOC}$. The investigated pH range was from 4.5 to 7 for both coagulants. The hydraulic residence time (HRT) in the coagulation/flocculation tanks was 21 minutes in total, 7 minutes in the rapid mixing tank and 14 minutes in the slow mixing tank, respectively. Each condition was tested for at least 72 hours. Table 3 summarizes the applied coagulation conditions.

Additional experiments were performed to evaluate the influence of the pre-treatment process configuration on the overall process performance. Prior to the membrane filtration unit three different coagulation configurations were compared, a classical two-tank coagulation/flocculation

Table 3 | Coagulation conditions

All experiments	
Rapid mixing time/G-value	6.7 min/400 s ⁻¹
Slow mixing time/G-value	13.3 min/100 s ⁻¹
PACL coagulation	
Coagulant dose	2, 3 and 5 mg Al/L
Investigated pH values	4.5; 5.0; 5.5; 6.0; 6.5; 7.0
Iron coagulation	
Coagulant dose	4, 6 and 10 mg Fe/L
Investigated pH values	4.5; 5.0; 5.5; 6.0; 6.5; 7.0

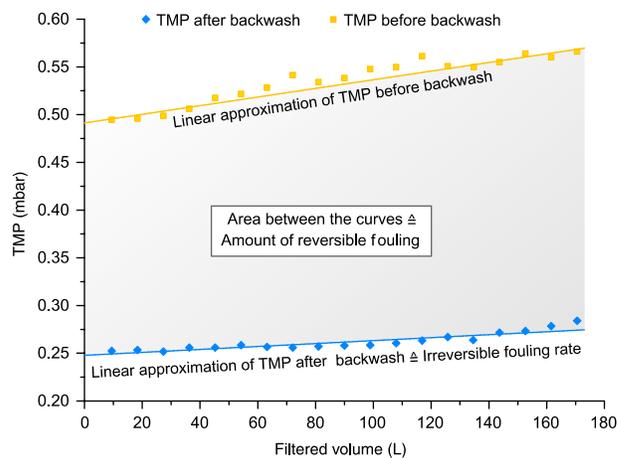
setup with a rapid mixing followed by a slow mixing step, a simplified setup containing only rapid mixing and bypassing the slow mixing, and finally an even more simplified inline mixing and pipe coagulation setup. In the inline setup (Figure 2b) the initial mixing of the coagulant with the raw water took place in the membrane feeding pump, followed by a pipe flocculator, which further ensured mixing (G-value 330 s⁻¹) until the water reached the membrane. The total HRT in the inline mixing was 45 s. For this initial set of experiments conditions beneficial for DOC removal were selected. Iron chloride (PIX-111) was chosen as coagulant, operated at a pH of 5 and an iron dose of 10 mg Fe/L.

Experimental analysis

The membrane performance was monitored by logging the transmembrane pressure (TMP) development for constant flux operation. Samples were taken before the coagulation and after the membrane filtration twice a day during different stages in the process cycles. Turbidity measurements (90° scattered light method, Turbidimeter 2,100N, Hach) and the residual metal concentration (measured by High Resolution ICP-MS at the Institute for Chemistry at NTNU) in the permeate were analysed for each sample. The removal of organic matter was monitored by measuring colour, UV-absorption at 254 and 436 nm (Spectrophotometer U-3,000, Hitachi) and DOC (Laboratory analyser: Dohrmann Apollo 9,000, Teledyne-Tekmar; On-line spectrometric probe: Spectro::lyser[®], s::can Meßtechnik GmbH, Vienna, Austria).

In this study measurements were conducted to distinguish between reversible and irreversible fouling. By definition, reversible fouling can be removed by regular cleaning procedures without using chemicals, whereas irreversible fouling has to be removed by enhanced chemical cleaning. This is usually done when a defined level of the system permeability is undercut.

In the conducted experiments the TMP was measured for each cycle before and after a backwash. The resulting data were linear approximated as shown in Figure 3 and the slope of the regression curves calculated. The slopes represent the actual fouling rate during an experiment, expressed as pressure increase per amount of time with the unit mbar/h. The area under the regression curves can be defined as the total amount of fouling and was estimated by integration of the regression curves. In contrast to the fouling rates the total amount of fouling has the unit mbar h. By calculating the area between the two regression curves the amount of reversible fouling in an experiment can be quantified. The obtained values are a measure for the distance between the linear approximated TMP curves and thus a quantification for the degree of reversible fouling during an experiment. At the end, all irreversible fouling rates were subsequently compared to each other and related to the different flocculation conditions applied. For the overall evaluation of the reversible fouling, the total amount of fouling, which contains more information about the filtration resistance build-up during an entire experiment compared to the actual fouling rate, was used rather than the fouling rate.

**Figure 3** | Exemplary calculation of reversible and irreversible fouling.

RESULTS AND DISCUSSION

Tank coagulation/flocculation configuration

Overall, the applied treatment combinations investigated in this study worked very efficiently. After a short initial phase at the beginning of the experiments, the TMP measured was stable for the remaining experimental period. Good colour and DOC removal were achieved.

As shown in Figure 4, more than 90% of the raw water colour (VIS_{436} : 1.97 m^{-1}) was removed at most of the chosen conditions. For the PACL (Figure 4(a)) a removal maximum was observed around pH 5.5. Above and below that the colour removal decreased. Even with the low alum dose of 2 mg/L a colour removal higher than 90% was achievable at optimal pH. For the iron chloride the colour removal was a bit lower compared to PACL (Figure 4(b)). To achieve a removal above 90% a minimum iron dose of 6 mg/L was required. A removal maximum was observed at pH 5 and below.

The DOC removal followed a similar pattern as for the colour (Figure 5). For PACL the removal optimum was at pH 5.5 and with a decrease at lower or higher pH values, respectively. The removal varied between 50 and 85% at these pH levels and increased with higher coagulant doses. For iron chloride the DOC removal optimum was at pH 4.5. At a dose of 3 mg/L only up to 48% of the DOC was

removed. Dosages of 6 mg Fe/L and higher were necessary to remove more than 70%. The highest removal with iron chloride was 87% at a pH of 4.5 and an iron dose of 10 mg/L.

These findings correspond with results reported in previous studies (Lerch *et al.* 2005; Konieczny *et al.* 2006). The observed optimal pH values for the removal of NOM are lower than commonly reported for treatment facilities targeting turbidity removal. This is caused by the formation of strongly positively charged hydroxo- and polyhydroxo-complexes, which takes place at low pH values. Especially the poly-complexes show a very strong tendency to attach to the negatively charged surface of NOM. The optimal pH for PIX 111 is 4.5 lower than for the pre-polymerized PAX 18 of 5.5. However, both coagulants remove colour and DOC over a wider pH range.

Residual metal concentrations in the permeate are strongly depending on the coagulant dosage and the pH values (Figure 6). The higher the coagulant dose or the lower the pH, the higher is the residual metal concentration. At pH values of 6 and below the residual metal concentration exceeds $150\text{ }\mu\text{g/L}$, the limiting value defined by the Norwegian regulations for both aluminium and iron. However, low residual iron concentrations can be achieved also at higher coagulant doses, at pH values above 6.

The residual metal concentrations correlate well with the solubility of aluminium and ferric-based metal salts.

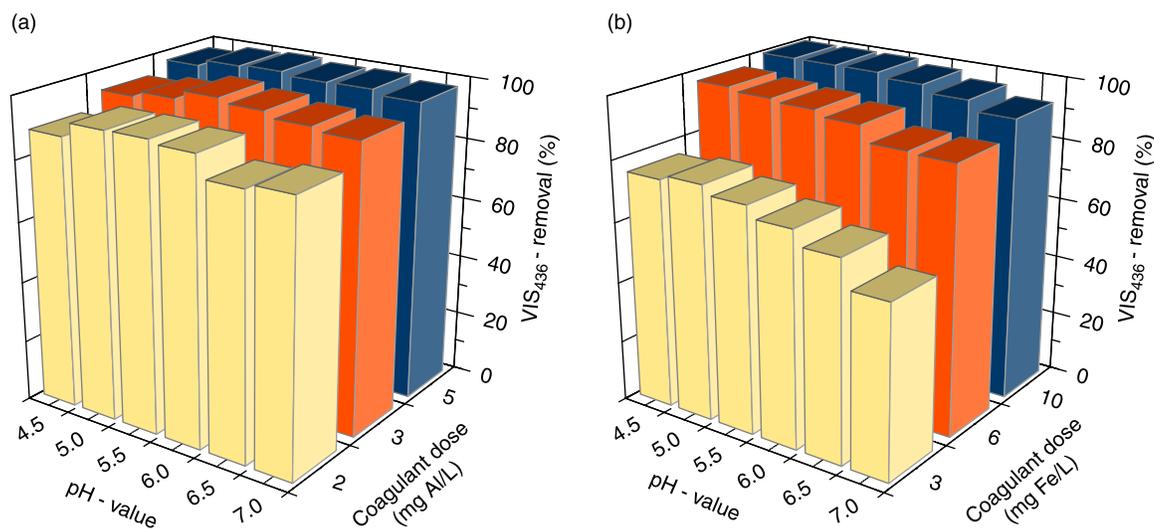


Figure 4 | Colour removal expressed as percentage VIS_{436} reduction in dependence on pH and coagulant dose.

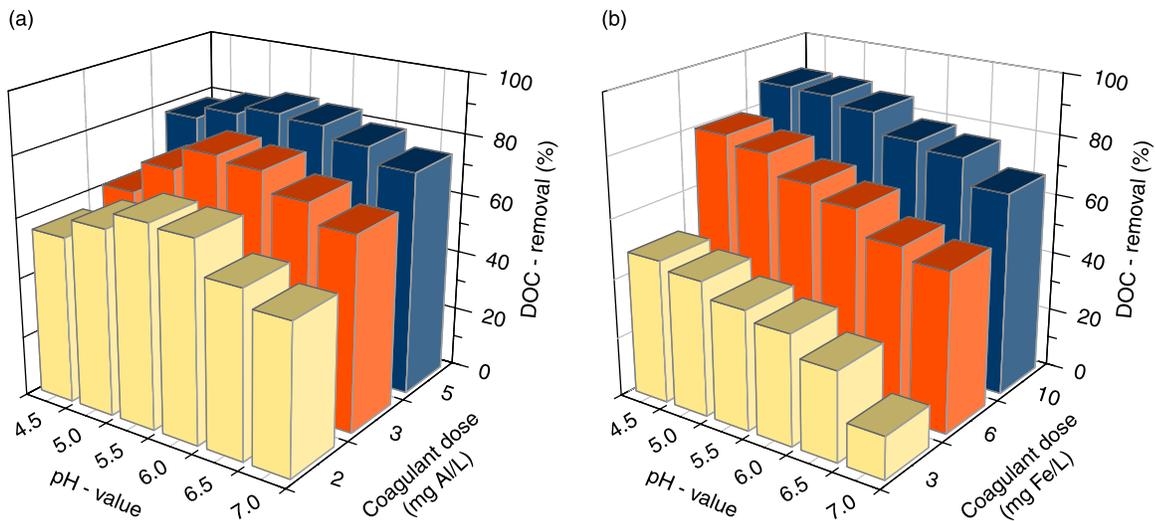


Figure 5 | Percentage DOC removal in dependence on pH and coagulant dose (raw water DOC = 5.2 mg/L).

An optimal pH for solubility values can be defined due to the solubility of the positively charged metal-hydroxo complexes that are present in the water at lower pH values, compared to the insoluble neutral metal compounds found at a higher pH. However, the optimal pH values for the removal of NOM are lower than this pH range. The consequence is therefore a narrower pH range of operation to comply with both requirements, removal of NOM and

residual metals, making the residual metal concentration a limiting parameter for process optimization.

Assessment of the membrane filtration performance was made by evaluating TMP measurements for the various operating conditions. Even if the observed irreversible fouling, which is represented by TMP measurements after a backwash, was very low, it was still detectable and depended on the operating conditions. Figure 7(b) shows a

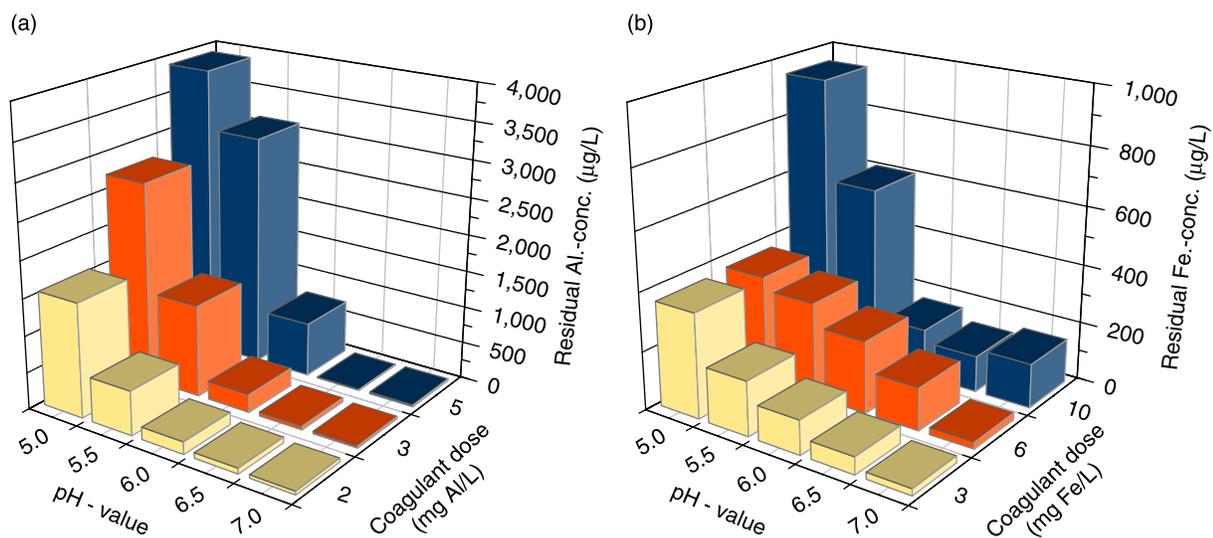


Figure 6 | Residual metal concentration in the permeate in dependence on coagulant dosage and pH value.

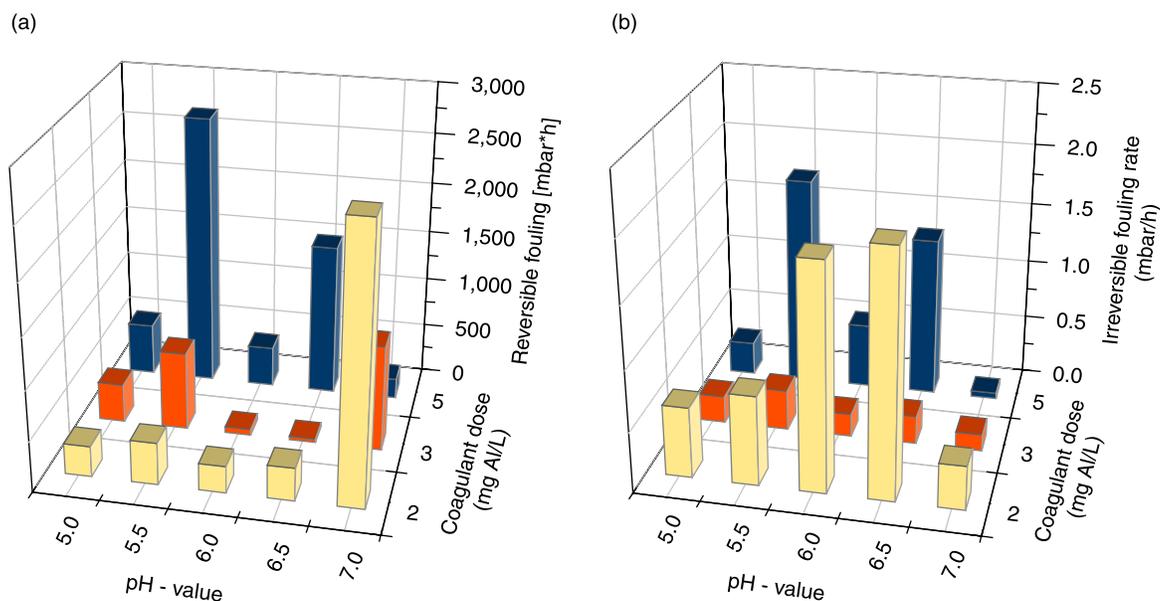


Figure 7 | Membrane performance in dependence on pH and coagulant dose (for PACL).

summary of the membrane performance, expressed as irreversible fouling, in dependence on the different coagulation/flocculation conditions for PACL.

With the application of PACL two trends are clearly visible. Fouling is much lower at an alum dose of 3 mg/L compared to the higher (5 mg/L) and lower dose (2 mg/L). That means that there is an optimum dosage in terms of membrane fouling for this kind of raw water and process configuration used. Since the turbidity in the raw water is very low it can be assumed that the fouling is mostly driven by the organic matter contained in the raw water and by the coagulant itself. At the low alum dose of 2 mg/L possibly the formed aggregates are smaller than at higher doses as shown by Gregory *et al.* (2000). The coagulant dose might be too low for neutralizing the negative charge of the organic matter molecules and thus decreasing aggregation and precipitation. Both effects could result in increased membrane pore blockage and lower filterability. At a higher alum dose of 5 mg/L aggregation is improved and the material deposition on the membrane surface is increased. Hence, the load of the membrane might be too high for a 1-hour filtration time. Another possibility is that a re-stabilization by charge reversal takes place, again resulting in a decrease of aggregation and precipitation, leading to increased fouling. This explanation would be

valid only for pH values below 6 to 6.5, since only in that pH range charge neutralization is the dominating removal mechanism. Above that, the dominating removal mechanism changes to enmeshment and metal hydroxide precipitation.

Secondly, using PACL a maximum of irreversible fouling can be observed around a pH range of 5.5 to 6.5. Below and above that range the fouling rates decrease. Since the fouling in this study is mostly caused by NOM, a good correlation between the removal of organic matter (Figure 5(a)) and the reversible fouling rates (Figure 7(b)) can be observed. We also observe a DOC removal maximum in that pH range. However, it can be stated that at an operating flux of $143 \text{ L m}^{-2} \text{ h}^{-1}$ and dependence on the selected coagulation conditions, irreversible membrane fouling can be kept at a low level and stable operation is achievable.

Reversible fouling, which is defined as the amount of fouling that can be cancelled by normal backwashing and quantified in this study by calculating the area between the two linearly approximated TMP curves before and after backwashing (compare with Figure 3), was detected only in small amounts during all experiments (Figure 7(a)). At some conditions the reversible fouling was higher than the average. At a pH of 7 and an aluminium dose of 2 mg/L,

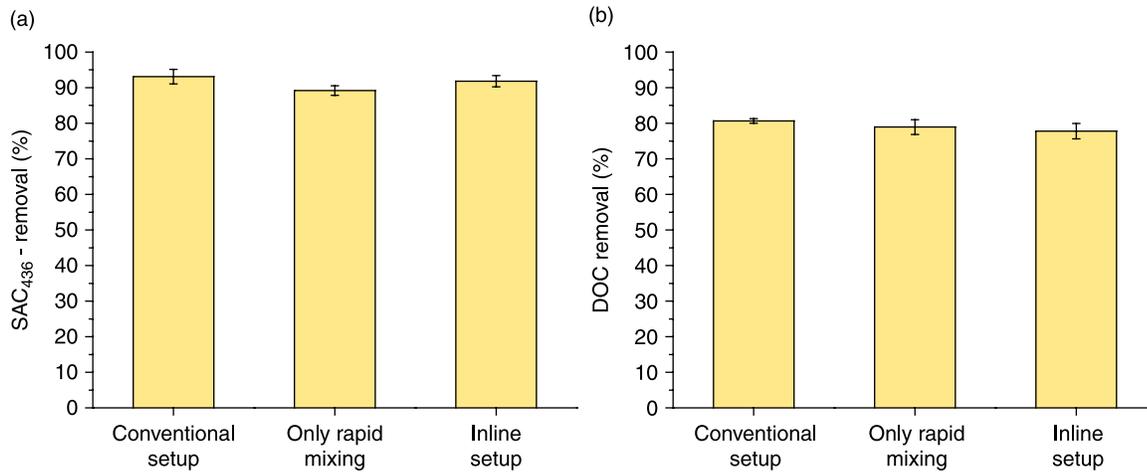


Figure 8 | Colour and DOC removal in dependence on the pre-treatment setup (iron dose of 10 mg/L, pH 5).

the amount of reversible fouling was 2,788 mbar h. Keeping in mind that the cycle length was 1 hour and the experimental duration 72 hours, this means that the average pressure build-up during a cycle was around 40 mbar, which is rather low. Thus, not much filtration resistance was built up during a filtration cycle. Therefore, the applied cycle length of one hour might be extendable and consequently an increased process efficiency achievable.

The permeate turbidity was consistently measured below 0.1 NTU for all experimental runs, indicating a very good particle removal capacity of the applied process combination.

Summarizing, it can be said that efficient operation, meaning a maximization of organic matter removal and a simultaneous minimization of membrane fouling and

residual metal concentration, is very achievable with the investigated process combination. Comparing with Figure 5 again, an efficient removal of DOC can be achieved at higher pH values and with a very low dosage of coagulant. This, combined with not too high or too low coagulant dose, minimizes also the membrane fouling (Figure 7). The coagulation/membrane filtration process therefore can be operated at higher pHs and low coagulant dosages with low membrane fouling and still meet the required efficiency.

Comparison of different pre-treatment configurations

NOM was efficiently removed independently from the chosen pre-treatment options. Direct comparison of the

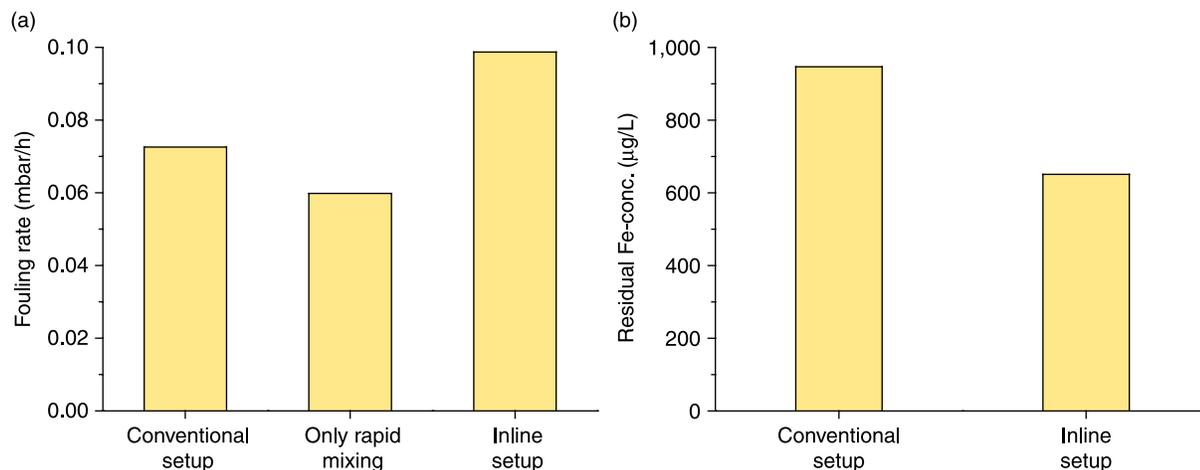


Figure 9 | Membrane performance and residual metal concentration in the permeate in dependence on the pre-treatment setup.

three different setups showed a similar colour and DOC removal rate, i.e. of around 80% at an iron dose of 10 mg/L and a pH value of 5, as shown in Figure 8(a) and (b). This is especially remarkable considering the very short hydraulic retention time of only 45 s in the pipe flocculator using the inline configuration, compared to 21 minutes in the conventional setup and 7 minutes in the configuration containing only the rapid mixing.

The irreversible membrane fouling also did not show a significant difference as shown in Figure 9(a). Although the irreversible fouling rate was 0.10 mbar/h higher in the inline configuration than in the other two setups, it was still well within the range of fouling rates found in the first set of experiments using the conventional setup, shown in Figure 7(b). However, the problem with high residual metal concentrations in the permeate at low pH values and high coagulant doses remains. Since in this experiment a rather low coagulation pH of 5 was chosen, high metal residues were found independent from the pre-treatment configuration (Figure 9(b)). The example shown is to illustrate the relationships found. However, for practical implementation it is necessary to choose operating conditions that comply with the residual metal concentration regulations.

It can be assumed that the particles formed in the pipe flocculator are large enough to be retained by the membrane, do not cause pore blocking and do not have a characteristic that increases membrane fouling due to changes in cake-layer properties. This observation can be supported by the consideration that the 1 metre long and 2.5 mm membrane channels have such a flow pattern that they can work as a pipe flocculator themselves (Yonekawa *et al.* 2004). They therefore support a further floc development and promote a dense columnar cake layer at the dead-end points, but alleviate cake-layer formation on the rest of the membrane surface.

CONCLUSIONS

The conducted experiments confirm that the combination of coagulation/flocculation and ceramic membrane filtration is an efficient and reliable option for the drinking water treatment of surface waters with high NOM concentrations. Very good DOC and colour removal can be

achieved at relatively low coagulant dosages. This removal, however, strongly depends on the coagulation pH value in which a pH range is preferred between 5 and 6. Residual metal concentration in the permeate increases in this pH region, and operation at higher pH values is therefore necessary to comply with regulation standards on acceptable concentration levels. Sufficient DOC and colour removal is also obtainable at elevated pH and low coagulant dosage. The residual metal concentration is one limiting parameter of the treatment concept investigated in this study and narrows down the choice of operating conditions.

Too low and too high coagulant doses cause increased irreversible fouling and therefore reduce the operation time of the membrane modules. Coagulant dose optimization is therefore mandatory. Even though at higher coagulant doses more DOC can be removed, the simultaneously increased irreversible fouling has to be taken into account, since the additional DOC removal might not be that high and strongly depends on other operation conditions like the pH.

An initial experiment testing a simplified inline configuration showed very good process performance compared to a classical tank coagulation/flocculation setup. This may be facilitated due to a unique flow pattern inside the membrane modules. Using inline coagulation, more compact full-scale units can be built, using less space compared to conventional sand filtration units or even membrane filtration plants using tank coagulation/flocculation.

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