



Removal of natural organic matter through membrane filtration and subsequent effect on disinfectant decay

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ABSTRACT

This study aims to evaluate the effectiveness of membrane filtration in removing natural organic matters (NOMs) from four different source waters and the subsequent effect that it has on total chlorine (TC) demand of these waters. Source water samples were filtered sequentially through membranes with molecular weight cut-off of 3,500, 1,000 and 200 Da as well as RO membrane. The source waters and sequentially filtered samples were dosed with chlorine and the residual chlorine data were used to estimate the TC demand of these waters. A robust chlorine decay model constructed in AQUASIM software was used to do so. More than 80% of the chlorine demand in untreated surface water sources was found to be contributed mainly by NOMs that were larger than 3,500 Da. However, for water treated by granular filtration, the chlorine demand was found to be contributed by NOMs which were down to 200 Da. Sequential filtration through all four membranes reduced chlorine demand by more than 94% in surface waters and 84% in waters treated by granular filtration. Significant reduction in the formation of trihalomethane can be achieved if water is treated by appropriate membranes after granular media filtration.

Keywords: Chlorine; Chlorine demand; Disinfection by-products; Mathematical modelling; Membrane; Natural organic matter (NOM)

1. Introduction

In modern water treatment processes, chlorine is generally involved as a disinfectant [1]. Water taken from catchments is generally of poor quality due to the presence of organic and inorganic pollutants. The treatment of such water significantly improves the quality. However, during the distribution of the water from the treatment plant to the final user, the quality again declines [2]. Chlorine can react with the natural

organic matter (NOM) found in water to form carcinogenic disinfection by-products (DBPs). These by-products are known to be harmful to human health and ideally should not be present in drinking water. The most prevalent of these by-products are the trihalomethanes (THMs).

Membrane filtration is a separation process that removes particle matter from water [3]. Several forms of membrane filtration exist, with each type having different properties and applications. Common types of filtration include microfiltration, ultrafiltration (UF),

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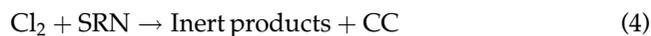
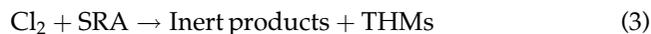
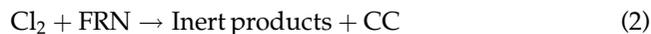
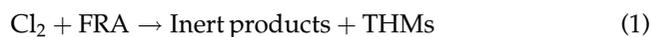
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nanofiltration (NF), and reverse osmosis (RO). Water that is to be treated by membrane filtration generally requires pre-treatment before it can pass through the membrane. Particles in the water that are much larger than what the membrane is designed to filter have a tendency to foul the system [4]. This problem can be prevented by screening the water through a standard filter upstream of the membrane [4].

This study aims to evaluate the effectiveness of membrane filtration in removing NOMs from four different source waters and the subsequent effect that it has on the total chlorine (TC) demand of these waters. UF, NF and RO membranes were used in series to treat these waters, and the permeates from each membrane were chlorinated; the residual chlorine concentrations collected over a period of time were then used to model the decay of chlorine. The modelling outputs were used to compute the chlorine demand of these permeates. Comparing the chlorine demand of permeates against the chlorine demand of source waters will be able to quantify the effectiveness of membrane filtration in removing NOMs that demand chlorine and subsequently produce DBPs.

2. Modelling the decay of chlorine

The decay of chlorine and the formation of DBPs will definitely be controlled by membrane processes. A theoretical model developed by Kastl et al. [5] was used to determine the chlorine demand of water and the formation of DBPs in the water that has been treated through different membranes. The model considers both the initial rapid decay and the continuing slow decay. The initial rapid decay is caused by fast reducing agents (FRA) and fast reducing nitrogenous compounds (FRN) while the continuing slow decay is caused by slow reducing agents (SRA) and slow reducing nitrogenous compounds (SRN) [5]. Chlorine can be considered as free chlorine (comprised of HOCl and OCl⁻ and denoted as Cl₂ herein) and can react with all four of these components. A reaction between chlorine and FRN or SRN will produce combined chlorine (CC). The combination of CC and free chlorine will produce TC. The reaction between chlorine and FRA or SRA can be considered for producing THMs. Different membranes are expected to remove different levels of FRA, SRA, FRN and SRN concentrations from the water that is treated by these membranes. The reaction schemes outlined by Jegatheesan et al. [6] can be used to model the decay of chlorine and to obtain decay rate constants for each reactions:



The rates for these reactions are denoted by k_1 , k_2 , k_3 , k_4 and k_5 respectively [5].

Using AQUASIM, the following equation can be used to determine the decay of free and combined chlorine as well as the formation of THM [5]:

$$\frac{d[\text{Cl}_2]}{dt} = -k_1[\text{Cl}_2][\text{FRA}] - k_2[\text{Cl}_2][\text{FRN}] - k_3[\text{Cl}_2][\text{SRA}] - k_4[\text{Cl}_2][\text{SRN}] \quad (6)$$

$$\frac{d[\text{CC}]}{dt} = -k_5[\text{CC}] + k_2[\text{Cl}_2][\text{FRN}] + k_4[\text{Cl}_2][\text{SRN}] \quad (7)$$

$$[\text{Total Cl}_2] = [\text{Cl}_2] + [\text{CC}] \quad (8)$$

$$\frac{d[\text{THM}]}{dt} = \alpha k_1[\text{Cl}_2][\text{FRA}] + \beta k_3[\text{Cl}_2][\text{SRA}] \quad (9)$$

where α and β are the THM formation coefficients.

Residual free and TC concentration data at different time intervals over a period of time should be obtained by dosing given water with two different chlorine concentrations. A mathematical model using Eqs. (6)–(9) can be constructed in AQUASIM. AQUASIM contains a dynamic equation solver which can perform parameter estimation to find the best fit of the model output to the experimental data [7]. Thus, the chlorine demand can be calculated by adding the chlorine equivalent of fast and slow reacting agents as well as nitrogenous compounds (FRA, SRA, FRN and SRN) that were estimated through modelling.

3. Materials and methods

3.1. Materials

Sodium hypochlorite (NaOCl) with a known concentration of chlorine was used to dose the samples. The membranes used in this experiment include RO, NF (200 Da) and UF (1,000 and 3,500 Da). The properties of the membranes as outlined by the manufacturer are shown in Table 1.

3.2. Sample collection

For this study, water was taken from four different locations around the Geelong region situated in the State of Victoria in Australia. Samples were taken from Geelong's main source of fresh water, the West Barwon Reservoir. Samples were also taken from Wurdee Boluc Reservoir, which flows directly into Geelong's drinking water treatment plant, with samples of treated water from the plant itself also taken for comparison with the help of Barwon water which is the supplier of water to the Geelong region. The Wurdee Boluc Water Treatment Plant uses dual media direct pressure filtration as the main process for filtering the water. Finally, a sample of sea water was taken in order to compare it against the samples of fresh surface water. This sample was taken along a stretch of coast adjacent to the head of the Barwon River, a river that plays a key part in Geelong's water supply. Cleaned plastic containers were used to collect the samples; samples were collected after rinsing the containers three times with the water that was to be collected. Water quality parameters of collected samples measured at the laboratory and the samples were stored in 4°C cold room until the experiments commenced.

3.3. Filtration

Using a laboratory-scale filtration unit, the samples were filtered across each successive membranes (3,500 Da, 1,000 Da, 200 Da, RO) with small samples of treated water from each membranes taken for further analysis. The RO was operated at 35 bar and all others were operated at 10 bar. Flow rate of water through all the membranes was 8 L/min.

3.4. Chlorination

After filtering, each collected sample had been divided into five smaller samples ready for further analysis (unfiltered as well as permeates from 3,500, 1,000, 200 Da and RO membranes). Dark brown bottles were used in chlorination studies where a portion of

each of these samples was placed in these bottles and dosed with NaOCl to obtain 2 and 4 mg/L of free chlorine. The total and free residual chlorine measurements were taken at regular time intervals up to 168 h. These measurements were taken using the DPD pillow method and a Hach Pocket Colorimeter. Using these measurements and the theoretical model [5], a value for chlorine demand could be determined for each of the samples. When the decay of chlorine was so significant that only small amounts of residual chlorine were present, re-chlorination of the samples occurred. The reason for re-chlorinating samples was to validate the chlorine decay model. The model parameters obtained using the chlorination data were used to predict the chlorine decay after re-chlorination. This was matched with the data obtained. However, the validation is not reported in this paper. Fig. 1 depicts the experimental processes used in this study.

3.5. Analysis of samples

Further to the chlorine decay analysis, testing was conducted on the samples for total organic carbon (TOC) using a Shimadzu TOC-L+ASI-L Analyser. TOC can be considered an indication of the amount of NOM present in the water. TOC analysis was conducted for the unfiltered and filtered samples before and after chlorination to ensure accuracy. The pH of the samples was regularly tested throughout the experiment using a WTW 315i pH meter as was the temperature (Hanna Instruments Minitherm HI8751 Digital Thermometer), the conductivity (WTW LF330 Conductivity Meter) and the turbidity (Hach 2100P Turbidimeter). Limited THM analysis was also conducted. Standard Method 6232 b [8] was used to analyse THMs and their species and liquid–liquid extraction gas chromatographic method was employed in this study.

3.6. Flux

Flux was an important parameter to measure as it could indicate fouling within the membrane. During the filtration stage, the mass of permeate that had

Table 1
Properties of membranes used in this study

Property	SG 1812C-47P (RO)	DK 1812C-34D (NF)	GE 1812C-34D (UF)	GK 1812C-47P (UF)
Active area (m ² (ft ²))	0.27 (3.00)	0.32 (3.50)	0.32 (3.50)	0.27 (3.00)
NaCl rejection (%)	97	–	–	–
MgSO ₄ rejection (%)	–	98	–	–
MWCO (Dalton)	–	200	1,000	3,500
pH range	3–10	3–9	2–10	2–10
Maximum pressure (kPa)	4,137	4,137	2,760	2,760

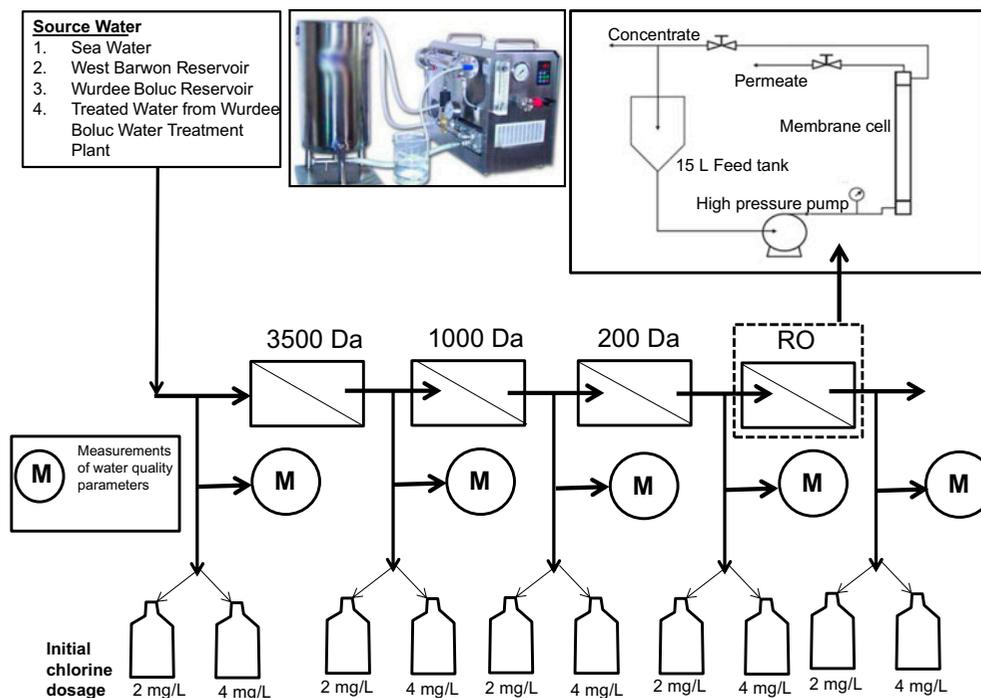


Fig. 1. Experimental processes carried out in this study.

passed through the membrane was measured at one minute intervals. The flux was then determined using the following equation:

$$J_i = \frac{(m_i - m_{i-1})}{\rho A(t_i - t_{i-1})} \quad (10)$$

where J_i = flux at interval between time t_i and $t_{(i-1)}$ ($L/m^2 h$), $m_i - m_{i-1}$ = mass of sample collected at interval between time t_i and $t_{(i-1)}$ (kg), ρ = density of the solution (kg/m^3), A = area of the membrane used for filtration (m^2).

The average flux for each experiment can be calculated using the following equation:

$$J = \frac{\sum_{i=1}^{t_n} J_i}{t_n} \quad (11)$$

where J = average flux ($L/m^2 h$) from the start to time = t_n , A density of a $1,000 kg/m^3$ was used for the water.

4. Results and discussion

4.1. Changes in water quality due to filtration

Table 2 shows the water quality parameters in terms of pH, turbidity, conductivity and TOC for water before and after passing through all four

membranes. From the table, it can be seen that the pH of the samples generally have a relatively neutral pH close to 7. The exceptions to this are the sea water samples that had slightly alkaline pH levels closer to 8 and the RO-treated samples that were moderately acidic. Samples were stored in a cold room until the commencement of experiments and, therefore, variations in temperature were observed as can be seen in Table 2.

The turbidity values are a measure of the suspended solids present in the water. Among the unfiltered samples, the two reservoir samples had significant turbidity readings. Although not as high as the reservoir samples, the sea water sample still gave a noticeable turbidity reading. All three samples gave readings above the Australian drinking water guidelines (ADWG) recommended value of 5 NTU. As expected, the water treated by the Wurdee Boluc WTP gave a low reading. All of the membrane-treated samples had low turbidity levels. Further, for the treated water from the Wurdee Boluc WTP, mixing of the samples before membrane experiments could have increased the turbidity of the feed water to the membranes. However, drop in turbidity after passing through each membranes indicate that each of the membranes remove certain amount of turbidity-causing particles.

The conductivity values are a measure of the mineralization (total dissolved solids) in the solution. With the exception of the sea water sample, the

Table 2
Water quality changes when passing through all four membranes

Source	Condition	Temp. (°C)	pH	Turbidity (NTU)	Conductivity (mS/cm)	UV ₂₅₄ (AU)	TOC (mg/L)
Sea water	Unfiltered	17.2	7.93	5.25	49.300	0.033	2.92
	Through 3,500 Da	18.4	7.78	0.27	48.500	0.013	
	Through 1,000 Da	22.4	7.88	0.15	52.500	0.030	1.55
	Through 200 Da	24.1	7.60	0.11	53.000	0.010	1.85
	Through RO	23.6	4.05	0.12	5.590	0.010	0.41
West Barwon Reservoir	Unfiltered	19.0	7.07	13.50	0.130	0.300	5.91
	Through 3,500 Da	22.5	7.68	0.15	0.245	0.190	1.47
	Through 1,000 Da	16.0	6.92	0.15	0.081	0.005	0.79
	Through 200 Da	16.1	6.77	0.07	0.075	0.013	0.38
	Through RO	16.4	7.05	0.09	0.009	0.004	0.19
Wurdee Boluc Reservoir	Unfiltered	15.0	6.95	13.30	0.160	0.270	5.08
	Through 3,500 Da	15.8	7.14	0.11	0.149	0.050	1.98
	Through 1,000 Da	15.8	6.92	0.10	0.102	0.000	0.62
	Through 200 Da	15.9	6.60	0.08	0.075	0.000	0.36
	Through RO	16.9	5.41	0.08	0.008	0.000	0.28
Treated water from Wurdee Boluc WTP	Unfiltered	17.4	7.33	0.19	0.265	0.046	2.41
	Through 3,500 Da	18.2	7.21	0.49	0.248	0.030	1.44
	Through 1,000 Da	17.8	7.23	0.33	0.203	0.009	0.36
	Through 200 Da	18.5	6.71	0.21	0.117	0.000	0.48
	Through RO	19.0	5.77	0.10	0.014	0.001	0.49

Note: Measurement accuracies for temperature, pH, turbidity, conductivity, UV₂₅₄ and TOC are $\pm 0.3^\circ\text{C}$, ± 0.03 , ± 0.03 NTU, $\pm 3 \mu\text{S}/\text{cm}$, ± 0.003 and $0.05 \text{ mg}/\text{L}$.

conductivity decreased with pore size. Only the RO membrane had any significant impact on the conductivity of the sea water sample as expected.

The expectation was that TOC concentration would decrease when filtered through the membranes. Improved TOC removal occurred with smaller pore sizes with the samples filtered through the RO membrane performing the best. All of the samples filtered through membranes performed significantly better than the unfiltered samples in respect to TOC concentration.

4.2. Flux

Table 3 shows the average water flux obtained for all four membranes. The sea water sample had a significantly lower flux through the 200 Da and RO membranes compared to the other samples. This can be attributed to the effect of osmotic pressure of sea water which reduces the applied pressure which in turn reduces the flux. Also the flux of the sea water sample through the 200 Da membrane declined quite rapidly after around 40 min, indicating the occurrence of significant fouling of the membrane. However, the interesting point to observe is the significant increase in the flux through 200 Da membrane for all the surface waters. This implies that both 3,500 and 1,000 Da membranes were effective in removing impurities and

in the absence of adverse osmotic pressure effect the water that was fed into the 200 Da membrane had minimal fouling effects on the membrane. The effectiveness of 3,500 and 1,000 Da membranes is also reflected in the chlorine demand of these waters which will be discussed in the next section. Further, the flux values were obtained for every 1 min intervals and averaged for an entire experimental run. Thus, there will always exist experimental/measurement errors which are well within the acceptable range (<5%). Thus, we could say that the flux for all three surface water samples were similar through 3,500 and 1,000 Da membranes. This helps to conclude that the fouling of these membranes by the NOM fractions above 1,000 Da in the above-mentioned surface waters are similar.

4.3. Chlorine demand

Using data obtained from the chlorine decay experiments and the model developed by Kastl et al. [5], the parameters of the model were estimated using Aquasim software. Using these estimations, the TC demand for each set of experiments was also predicted. The chlorine demand was calculated by adding the chlorine equivalent of fast and slow reacting agents as well as nitrogenous compounds (FRA, SRA,

Table 3
Average flux through the membranes used in this study

Membrane	Average flux (L/m ² h)			
	Sea water	West Barwon Reservoir	Wurdee Boluc Reservoir	Wurdee Boluc WTP
3,500 Da	34.74	31.73	30.24	30.74
1,000 Da	18.81	18.44	20.65	19.91
200 Da	22.62	83.60	77.81	77.11
RO	15.21	49.88	51.06	54.44

Note: Operating conditions—the RO was operated at 35 bar and all others were operated at 10 bar. Flow rate of water through all the membranes was 8 L/min.

FRN and SRN) that were estimated through modeling. Fig. 2(a) and (b) show the free and TC decay data along with model fitting for the treated water samples (unfiltered in the laboratory) obtained from Wurdee Boluc water treatment plant. The parameters estimated in AQUASIM in order fit the model to these data given in Tables 4 and 5 which show the TC demand of source waters as well as for samples passed through all the four membranes.

The chlorine decay studies reveal that there is a trend towards lower rates of decay for samples filtered through smaller pore sizes. This outcome was expected due to the relationship between NOM and chlorine decay. Filtering through membranes lowers the NOM concentration and hence, lowers the rate of chlorine decay. Some detailed analysis could also be made from this study as discussed subsequently. Table 6 is derived from the data shown in Table 5.

From Table 6 it can be seen that the contribution towards chlorine demand by species larger than 3,500 Da (or species filterable by 3,500 Da UF membrane) is nearly 40% in sea water but more than 80% in the two reservoirs situated in Geelong. This again reduces back to 34% in water produced by the treatment plant. The chlorine demand by species smaller than 3,500 Da and larger than 1,000 Da is around 6–10% for water from both reservoirs. However, the water produced by the treatment plant contains 17% of those species. Further, another 38% of chlorine demand in the water from the treatment plant is due to species that are in between 200 and 1,000 Da. Also adding a membrane treatment train to the existing water treatment plant has the potential to reduce the chlorine demand by another 85%.

A summary similar to reduction in chlorine demand was developed for TOC reduction as well which is shown in Table 7. From both Tables 6 and 7, it can be seen that a reduction in TOC in surface waters tends to reduce the chlorine demand proportionally when the total percentage reductions

are considered. The TOC in 200 Da and RO membrane effluent were very small (less than 0.5 mg/L) and, therefore, it is reasonable that there can be no good

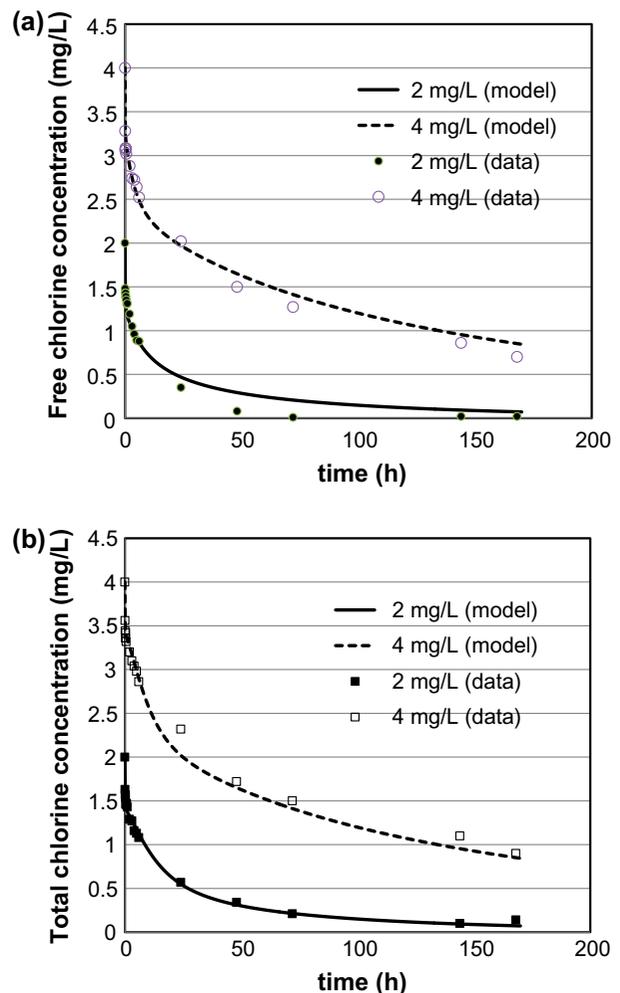


Fig. 2. Model fitting for the chlorine decay data obtained in the laboratory for the treated water samples (unfiltered in the laboratory) from Wurdee Boluc water treatment plant. (a) Free chlorine; (b) TC.

Table 4
Estimated parameters for the treated water sample from the Wurdee Boluc water treatment plant

Parameter	Estimated value
FRA (mg/L chlorine equivalent)	0.201
FRN (mg/L chlorine equivalent)	0.349
SRA (mg/L chlorine equivalent)	0.322
SRN (mg/L chlorine equivalent)	2.469
k_1 (L/mg h)	250
k_2 (L/mg h)	0.427
k_3 (L/mg h)	157
k_4 (L/mg h)	0.01
k_5 (L/mg h)	4.128

relationship between the reduction in chlorine demand and the reduction in TOC for the effluent from those membranes. Further, at lower TOC concentrations (less than 3 mg/L) the chlorine demand is proportional to TOC (with a proportionality constant nearly equal to 1).

At regular intervals after chlorination, samples of Wurdee Boluc Reservoir and treated water were taken for analysis of THM formation. The results of this analysis can be seen in Fig. 3. The performance of 3,500 Da membrane in reducing THM formation was also considered as it will be economical to use this membrane in a water treatment plant when further upgrade is required. In this study, only a limited amount of THM analysis was conducted and, there-

fore, the modelling of THM formation was not attempted. It was always expected that membrane filtration would have an impact on the level of THM formation and the analysis indicates that this was generally the case. Membrane filtration had an obvious improvement in lowering THM formation in the Wurdee Boluc WTP samples. Fig. 3 shows that all of the THM measurements recorded were below the ADWG recommended value of 0.25 mg/L.

It is also interesting to note that the total THM was comprised of chloroform, dichlorobromomethane, dibromochloromethane and bromoform. The formation of these individual species with time when dosed with 2 mg/L of chlorine are shown in Fig. 4. Dibromochloromethane and dichlorobromomethane in treated water tend to form more than the other two species and bromoform had the least rate of growth.

5. Conclusion

In this study, sea water, two surface waters (West Barwon Reservoir and Wurdee Boluc Reservoir) and treated water from a filtration plant (Wurdee Boluc WTP) were passed through membranes with 3,500, 1,000 and 200 Da molecular weight cut-off as well as a RO membrane in series. Raw water and permeates from each membrane were chlorinated with 2 and 4 mg/L of chlorine. Initial concentrations of fast and slow reacting agents as well as nitrogenous

Table 5
TC demand of water after passing through the membranes

	Chlorine demand (mg/L)				
	Source water	3,500 Da	1,000 Da	200 Da	RO
Sea water	2.489	1.519	1.504	1.104	1.171
West Barwon Reservoir	17.97	2.489	1.344	0.971	0.959
Wurdee Boluc Reservoir	11.955	2.388	1.119	1.151	0.218
Wurdee Boluc WTP	3.342	2.214	1.636	0.357	0.512

Table 6
Reduction in chlorine demand of different source waters after passing through four membranes

	Reduction in chlorine demand (mg/L) after passing through				Percentage reduction in chlorine demand after passing through				Total reduction %
	3,500 Da	1,000 Da	200 Da	RO	3,500 Da	1,000 Da	200 Da	RO	
Sea water	0.97	0.01	0.40	-0.07	39.0	0.6	16.1	-2.7	53.0
West Barwon Reservoir	15.48	1.15	0.37	0.01	86.1	6.4	2.1	0.1	94.7
Wurdee Boluc Reservoir	9.57	1.27	-0.03	0.93	80.0	10.6	-0.3	7.8	98.2
Wurdee Boluc WTP	1.13	0.58	1.28	-0.16	33.8	17.3	38.3	-4.6	84.7

Table 7

Reduction in TOC of different source waters after passing through four membranes

	Reduction in TOC (mg/L) after passing through				Percentage reduction in TOC demand after passing through				Total reduction %
	3,500 Da	1,000 Da	200	RO	3,500	1,000	200	RO	
			Da				Da		
Sea water	Incomplete data	Incomplete data	1.07	1.44	–	–	36.6	49.3	86.0
West Barwon Reservoir	4.44	0.68	0.41	0.19	75.1	11.5	6.9	3.2	96.8
Wurdee Boluc Reservoir	3.10	1.36	0.26	0.08	61.0	26.8	5.1	1.6	94.5
Wurdee Boluc WTP	0.97	1.08	–0.12	–0.01	40.2	44.8	–5.0	–0.4	79.7

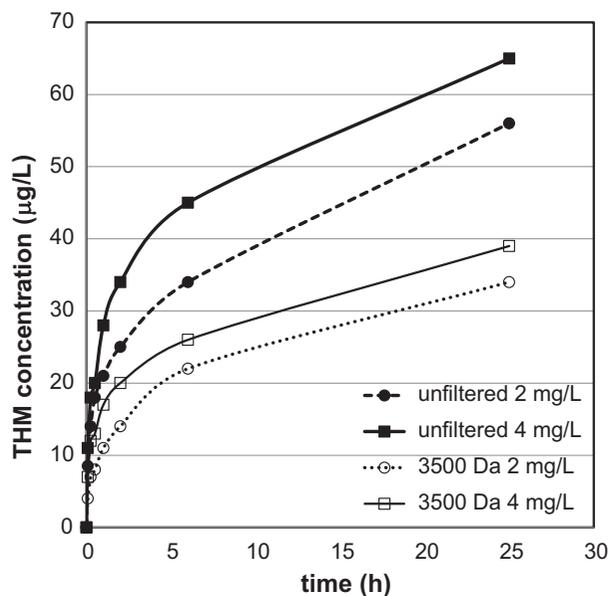


Fig. 3. Experimental data on THM formation in Wurdee Boluc WTP water when chlorine is dosed to the water as it is as well as after passing through 3,500 Da membrane.

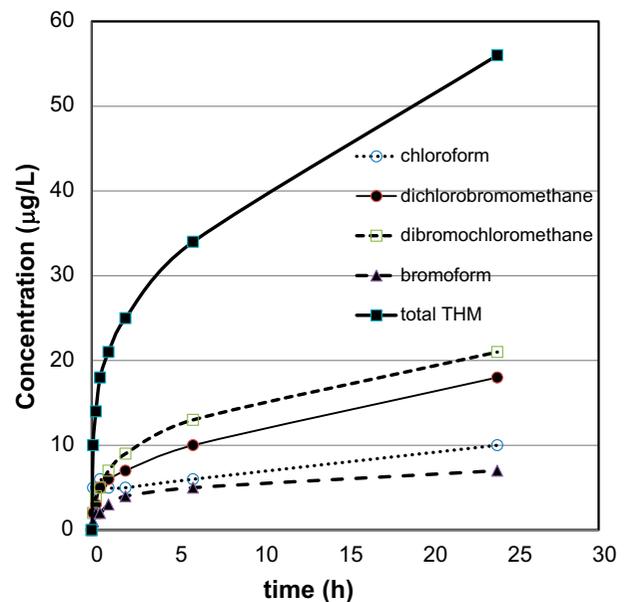


Fig. 4. Experimental data on the rate of formation of four THM species in the water from Wurdee Boluc water treatment plant (prior to the filtration at the laboratory) when dosed with 2 mg/L of chlorine.

compounds were estimated through a chlorine decay model by fitting the model output with experimental data. The summation of the initial concentrations of above four chlorine consuming groups will give the TC demand of the water tested. The following could be concluded from the results of this study:

The chlorine demand of sea water, West Barwon Reservoir, Wurdee Boluc Reservoir and treated water from Wurdee Boluc WTP were 2.49, 17.97, 11.96 and 3.34 mg/L, respectively. This indicates that Wurdee Boluc WTP removes 72% of chlorine consuming NOMs and other compounds from the source water

coming from Wurdee Boluc Reservoir to the WTP. The final chlorine demand of sea water, West Barwon Reservoir, Wurdee Boluc Reservoir and treated water from Wurdee Boluc WTP after passing through all four membranes were 1.17, 0.96, 0.22 and 0.51 mg/L, respectively. Thus the membrane series further removes 53.0, 94.7, 98.1 and 84.7% of chlorine demand from those waters, respectively.

The initial TOC of sea water, West Barwon Reservoir, Wurdee Boluc Reservoir and treated water from Wurdee Boluc WTP were 2.92, 5.91, 5.08 and 2.41 mg/L, respectively. The TOC of these waters after passing

through all four membranes were 0.41, 0.19, 0.28 and 0.49 mg/L, respectively. Thus, the Wurdee Boluc WTP removes 53% of TOC from the source water and the addition of all four membranes removes 80% of the remaining TOC of the water from the Wurdee Boluc WTP. TOC is reduced significantly from all four waters when passed through the membrane with 3,500 Da MWCO (more than 60% for both surface waters and 40% for treated water) and both 3,500 and 1,000 Da membranes together remove more than 85% of TOC from all three surface waters. Therefore, a reduction in TOC in surface waters tends to reduce the chlorine demand proportionally when the total percentage reductions are considered. Further, membrane filtration had an obvious improvement in lowering the THM formation in the Wurdee Boluc WTP samples.

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