Pilot scale testing of biofilter post-treatment of MIEX® treated water

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ABSTRACT

The MIEX® (Magnetic Ion Exchange) process, which employs an anion exchange resin for removal of dissolved organic carbon (DOC), was introduced at the Wanneroo Groundwater Treatment Plant in Western Australia in 2001. In this pilot-scale study we examined a range of operational parameters for optimisation of biofiltration of MIEX®-clarified water. Granular Activated Carbon (GAC) outperformed anthracite as a filter medium. Increasing the empty bed contact time (EBCT) from 8 to 16 minutes improved performance. The GAC biofilters removed up to 20% of DOC and up to 25% of Biodegradable Dissolved Organic Carbon (BDOC), once they had stabilised in biological mode. Chlorine demand was reduced by 51 to 55% and trihalomethane formation potential (THMFP) was reduced by 35 to 50% in GAC biofilter effluent waters at 16 minutes EBCT when compared with their MIEX®-treated influent water. GAC biofilters developed more biomass on the surface than anthracite biofilters and this was associated with the greatest BDOC and DOC removals. Interestingly, neither biofilters developed populations of protozoans. Use of chlorinated influent water severely restricted biomass development in all biofilters at their surface. Biofilter treatment of chlorinated influent water resulted in the poorest removal of Assimilable Organic Carbon (AOC). Biofiltration improved the water quality of MIEX®-clarified waters.

Key words | AOC, BDOC, biofiltration, chlorine demand, DOC, MIEX®, trihalomethane formation potential

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INTRODUCTION

Water quality deteriorates between the treatment plant and the customer within urban water distribution systems. This deterioration is caused by leakage of chemicals from biofilms that grow on surfaces in the distribution system, reactions of the disinfectant with natural organic matter and other chemicals in the water, and reactions between infrastructure materials and the water. The distribution system of the Wanneroo Groundwater Treatment Plant, north of Perth, Western Australia, suffered from water quality problems over many years, especially taste and odour problems associated with biofilm development (Franzmann *et al.* 2001). To overcome these problems, the Water Corporation of Western Australia commissioned the first large-scale MIEX® (Magnetic Ion Exchange) plant at Wanneroo in November 2001. MIEX® is an anion exchange resin designed to remove a proportion of the DOC from drinking water and its implementation at the Wanneroo Groundwater Treatment Plant has improved substantially the quality of the distributed water (Warton *et al.* 2007) and has reduced biofilm growth in the distribution system (Zappia *et al.* 2004).

Biofiltration is used in water treatment to remove the biodegradable fractions of the DOC, which are commonly estimated by measuring BDOC and AOC concentrations. A number of side benefits can ensue through microbially driven processes, such as iron and manganese oxidation and removal (Rittmann & Snoeyink 1984), reduction of disinfection by-product formation (Chaiket *et al.* 2002), and reduction in the concentration of taste and odour-producing compounds (Elhadi *et al.* 2004). A range of parameters are known to affect the efficacy of biofiltration, including the quality of influent water, the type of biofilter matrix material, the use of disinfectants and the frequency of backwash, however, comparisons of studies can show conflicting results. For example, backwash of biofilters with chlorinated water may (Urfer 1998) or may not (Liu *et al.* 2001) affect substantially the biodegradation of organic matter when GAC is used as the biofilter support matrix. Because of the potential for a unique biological response to combinations of biofiltration parameters, especially with changing water quality, optimisation of biofiltration for the treatment of water which originates from a new treatment process requires pilot-scale experimentation.

As far as the authors are aware, MIEX® treatment has been used in combination with a number of other processes for the treatment of drinking water, including alum coagulation (Drikas *et al.* 2003) and ferric coagulation (Fearing *et al.* 2004), but not in combination with biofiltration. Until now, many studies which have examined the combination of MIEX® treatment with other treatment processes have done so using jar tests. This study determined if biofiltration, at pilot scale, could be used after MIEX® treatment, used at full scale, to further enhance water quality and, of the few biofiltration parameters which could be adjusted, which settings produced the best outcomes.

This study was designed to evaluate:

- Whether biofiltration would further enhance the quality of water after it was treated by MIEX®.
- Whether anthracite or GAC was the best support medium for biofiltration post-MIEX®, and which of two different GAC types provided the best support.
- If variation of the empty bed contact time (EBCT) changed filter performance substantially.
- How biofilters fed with MIEX®-clarified water performed when chlorinated backwash water was used.

- How biofilters fed with MIEX®-clarified water performed when chlorinated influent and backwash water was used.

For the treatment of post-MIEX®-clarified water, biofilter performance under each regime was assessed for changes in a range of water quality parameters including concentrations in DOC, AOC and BDOC, UV_{254}, chlorine demand, oxygen concentration and trihalomethane formation potential. We also compared levels of biomass in the biofilters under different biofilter operational settings. An extensive range of other parameters were routinely measured but are not reported here.

Understanding these outcomes will assist operators of treatment plants which utilise MIEX® treatment as a treatment option to evaluate the potential advantages of combining biofiltration with MIEX® treatment. It will also help them decide which operational parameters to use in their biofiltration process.

MATERIALS AND METHODS

Water source

The water treated at the Wanneroo Groundwater Treatment Plant is sourced from both confined and unconfined aquifers. The quality of the water influent to the plant changes due to variations in which bores are being used to supply the plant. The first treatment stage involves aeration for the oxidation and precipitation of reduced species of iron, manganese and sulfur. The water is then chlorinated, to further aid in oxidation of these reduced species. The chlorinated water is split into two streams. One stream is treated by MIEX® followed by "conventional" coagulation, flocculation and clarification. The other stream is treated by enhanced coagulation, then flocculation followed by clarification. Enhanced coagulation involves the use of increased amounts of aluminium sulfate and polyelectrolyte (typically double that used in the conventional process).

Biofilter operation

For the study, four pilot scale biofilters were used and consisted of 140 mm id. Perspex columns, each fed with

Table 1 | The types and depths of media used in each biofilter

Biofilter	Medium depth and type
1	0.3 m sand (ES[a], 0.65 mm; UC[b] 1.4) below 1.75 m anthracite (ES, 1.1 mm; UC 1.6)
2	0.3 m sand (ES, 0.65 mm; UC 1.4) below 0.65 m anthracite (ES, 1.1 mm; UC 1.6)
3	0.3 m sand (ES, 0.65 mm; UC 1.4) below 1.75 m GAC (ES, 1.3 mm; UC 1.4)
4	0.3 m sand (ES, 0.65 mm; UC 1.4) below 1.75 m GAC (ES, 0.7 mm; UC 1.7)

[a] = Effective size.
[b] = Uniformity coefficient.

either MIEX®-treated and clarified water or water from an enhanced coagulation clarifier. The types and depths of media used in each biofilter are given in Table 1.

The type of GAC used was steam activated coal-based carbon (Activated Carbon Technologies Pty Ltd, Eltham Victoria, Australia). The four biofilters were run in parallel so the effects of different filter media could be compared. At the commencement of the test program, on 12th January 2004, the biofilters were fed with MIEX®-treated clarified water with an empty bed contact time (EBCT) of 8 minutes. The biofilters were backwashed with non-chlorinated water three times a week (Monday, Wednesday and Friday) with air scouring and a backwash velocity that ensured a 30% bed expansion for 10 minutes. The biofilters were run under these conditions until 29th September 2004 (Stage 1). It was considered that the biofilters were in a "steady state" of operation from about July 2004, as the percentage of UV_{254} decrease over the GAC biofilters had stabilised from their initial 75 to 80% to values ranging between 20 and 30% for a number of months. To test the effect of EBCT on biofilter performance, the EBCT was increased to 16 minutes on 29th September 2004, and the biofilters were again sampled on 9th November 2004 (Stage 2). After this, the MIEX®-treated clarified influent water was replaced with water from the enhanced coagulation clarifier, and the EBCT was returned to 8 minutes. The biofilters were sampled again on the 1st December 2004 (Stage 3) after which MIEX®-treated clarified water was used again as influent water to the biofilters. To test the effect of the use of chlorinated backwash water on biofilter performance,

water from the clear water tank, which typically contained 1.3 mg L^{-1} free chlorine, was used to backwash the biofilters, which were again sampled on 12th January 2005 (Stage 4).

After the Stage 4 sampling, to test the efficacy of the biofilters in treating chlorinated influent water, water from the clear water tank at the treatment plant was used as influent to the biofilters. This water contained 1.3 ± 0.5 mg L^{-1} free chlorine. The filters were sampled on 9th February 2005 (Stage 5), and again on 27th May 2005 to examine the effect of prolonged treatment of chlorinated influent water.

For each stage, the biofilters were operated under the set conditions for at least three weeks after which a major analysis of biofilter performance was undertaken.

Monitoring

Routine, approximately weekly, monitoring of biofilter influents and effluents for UV_{254} and dissolved oxygen concentration was undertaken using probes and on-line monitors. From August 2004, two HACH SCI100 online dissolved oxygen sensors were available to measure oxygen concentrations in influent water and effluent water simultaneously, to and from one of the biofilters.

Several major sampling events were undertaken at the end of each stage. At these times the following parameters were measured in the influent water, and the effluent water from each of the biofilters: AOC, BDOC, DOC, UV_{254}, chlorine demand and trihalomethane formation potential (THMFP). Some of the determinations were carried out once only on single samples. Exceptions were AOC, which was determined in duplicate on a single sample, and DOC, which was determined up to five times on a single sample to attain a precision of $\pm 2.0\%$. BDOC measurements were usually carried out in duplicate but on occasion they were performed once only on a single sample. Tests for THMFP were done on a single sample, but the individual measurements for THMs were carried out in duplicate. Reproducibility in THM measures can be low, as the formation of THMs must account for error in the determination of 4 separate isomers. In addition to the major sampling events, UV_{254}, DOC, chlorine demand, and THMFP were also measured twice in Stage 5, after prolonged passage of chlorinated influent water through the biofilters. Briefly, the methodologies for the above determinations were as follows:

Dissolved organic carbon (DOC) measurements

DOC concentrations were measured routinely by an external laboratory. Prior to the collection of the water samples at the Wanneroo GWTP, sample bottles were prepared by heating overnight at 600°C. Water samples were pre-filtered through a pre-washed 0.45 μm Supor® membrane (IC Acrodisc). The filtered water was then injected into a Shimadzu TOC-Vwp Total Organic Analyzer that oxidises the DOC using persulfate, converting it to CO_2 which is measured using an infrared detector. The limit of quantitation of the Shimadzu TOC analyser was $0.02\,mg\,L^{-1}$.

Chlorine residual experiments

Each of the water samples was divided into four subsamples, each of which was dosed with a different concentration of chlorine, with the intent of retaining a chlorine residual of between 0 and $5\,mg\,L^{-1}$ in each subsample after 7 days. For each of the 4 water samples, the following procedure was undertaken: Approximately 450 ml of the water sample was transferred to a 500 ml volumetric flask and the predetermined amount of a concentrated chlorine solution was added. The volume of the solution was increased to 500 ml with the sample water and the sample was shaken vigorously for 2 minutes. Each solution was then used to fill a series of 20 ml screw cap vials. These vials were stored at room temperature (22°C) in the dark. At various time intervals up to 168 hours, a vial was opened and the residual chlorine concentrations (free and total) were measured.

The chlorine demand was defined as the initial chlorine concentration that would have produced a final concentration of $0\,mg\,L^{-1}$ after the 7-day period, and was calculated according to the following procedure (Warton *et al.* 2006): For each chlorine dose delivered to a water sample, the residual concentration was plotted against time. An exponential decay curve was fitted to each data set and an equation that defined the curve was fitted. The residual concentration after seven days in each sample was determined from its equation. These calculated residual concentrations were plotted against the initial chlorine doses. The X-intercept of the resultant straight line (i.e. the point at which the ideal residual concentration was zero) was taken as the seven day chlorine demand.

Trihalomethane potential formation experiments

Concentrated sodium hypochlorite solution (approx. $1000\,mg\,L^{-1}$ chlorine) was prepared by dilution of an aqueous sodium hypochlorite solution (12.5% w/v; APS Chemicals Ltd). Initial oxidant concentrations that would result in free chlorine residuals of $3–5\,mg\,L^{-1}$ after 7 days were calculated from the estimated chlorine demand. One litre amber bottles with the appropriate amount of concentrated sodium hypochlorite solution were filled with the sample water to eliminate headspace (approximately 1037 ml) and the bottles were placed in a water bath at 20°C for 168 hours. After a subsample was removed to measure the chlorine concentration, the residual chlorine was destroyed with aqueous sodium sulfite solution (100 μl; $100\,mg\,L^{-1}$). The water sample was then analysed for trihalomethanes (THMs) by headspace solid-phase micro-extraction (SPME) followed by gas chromatography – mass spectrometry (GC-MS). THM concentrations in the raw water samples without chlorine dosing were also separately determined by SPME-GC-MS. Briefly, for the SPME/GC-MS analysis, the water sample (10 g) was placed in a 20 ml vial fitted with a septum cap. Sodium sulfate (1.67 g) and 1,2-dibromopropane (standard) were added. The analytes were adsorbed onto a 100 μm PDMS SPME fibre for 10 minutes at 40°C in a Gerstel MPS apparatus interfaced to a Hewlett-Packard 6890 GC equipped with a 5973 MSD. The adsorbed analytes were desorbed in the GC injector for 15 minutes at 240°C for analysis on a DB5-MS capillary column.

AOC, BDOC and measures of biomass

AOC was measured by the method of van der Kooij *et al.* (1982), and BDOC was measured by the method of Joret & Levy (1986), although the biomass used to inoculate the flasks was grown on sintered glass Siran® beads in a biofilter fed with water from bore W257 in the Wanneroo borefield.

To determine the effect of different operational parameters on biomass in the biofilters, material from the top 10 cm of each biofilter was collected before and after backwash, and the biomass carried on each support material was determined by the lipid phosphate technique of Findlay *et al.* (1989). The complete backwash from each biofilter was captured in separate 210 L plastic drums, and the total

weights of the backwash waters were recorded. Biomass in a sub-sample of the homogeneously mixed drum contents was measured by counting with a fluorescence microscope after staining the cells with DAPI (4,6-diamidino-2-phenylindole). Lipid-P concentrations in the backwash were also determined by the method of Findlay *et al.* (1989).

RESULTS AND DISCUSSION

Effects of operational settings on removal of DOC, AOC and BDOC

The biofilters were operated for at least three weeks under each of the five different operational regimes with MIEX®-treated influent water unless otherwise stated (EBCT = 8 minutes, EBCT = 16 minutes, influent water from enhanced coagulation, use of chlorinated backwash water, and treatment of chlorinated influent water). After the three weeks' operation, at the time of major sampling events, waters influent to and effluent from the biofilters were sampled and measures were made of parameters considered important for assessment of the efficacy of the biofilters. The concentrations of DOC, AOC and BDOC in water influent to and effluent waters from the biofilters at the times of sampling are given in Table 2.

The waters from the MIEX®-clarifier were generally so low in AOC concentration that they would be considered biologically stable. BDOC concentrations were also close to concentrations considered biologically stable ($< 0.2 \, \text{mg L}^{-1}$; Piriou *et al.* 1998) except at the time when chlorinated backwash and influent water was used. The latter samples contained BDOC up to $0.31 \, \text{mg L}^{-1}$ and AOC concentrations up to $38 \pm 11 \, \mu\text{g L}^{-1}$. It is well known that chlorination can increase AOC and BDOC concentrations (Liu *et al.* 2001). Although the DOC concentrations in influent waters at the times of major sampling events were within a narrow range (1.66 to $1.96 \, \text{mg L}^{-1}$), the BDOC concentration in the influent water from enhanced coagulation ($0.08 \, \text{mg L}^{-1}$) was much lower than the BDOC in water from the MIEX®-clarifier (average = $0.23 \, \text{mg L}^{-1}$).

AOC concentrations were initially below the level of detection ($< 5 \, \mu\text{g L}^{-1}$) and lower than the previous measure of AOC in filtered MIEX®-clarified water ($62 \, \mu\text{g L}^{-1}$) in the summer of 2003 when BDOC was also at a much higher concentration ($1.42 \, \text{mg L}^{-1}$; Warton *et al.* 2005). The DOC content in the Wanneroo water after enhanced coagulation in this study ($1.78 \, \text{mg L}^{-1}$) was also lower than the DOC in this water during the summer of 2003 ($2.3 \, \text{mg L}^{-1}$), and may have resulted from the plant's use of better quality source water. AOC was detected in the water influent to the biofilters when enhanced coagulation water was used as feed ($21 \, \mu\text{g L}^{-1}$ on 1/12/2004), and when the feed water was chlorinated ($38 \, \mu\text{g L}^{-1}$ on 9/3/2005, $21 \, \mu\text{g L}^{-1}$ on 27/5/2005) and at the time when chlorinated backwash water was used ($45 \, \mu\text{g L}^{-1}$ on 14/1/2005). At these times, the AOC concentrations were still low although slightly greater than for water considered by some researchers as "biologically stable" (van der Kooij *et al.* 1999).

When non-chlorinated water was used as influent water to the biofilters, AOC values in water effluent from the biofilters were generally below $5 \, \mu\text{g L}^{-1}$ except for biofilter 2 when treating water from enhanced coagulation, when the AOC content was $8 \, \mu\text{g L}^{-1}$. AOC values in water effluent from all biofilters was also above $5 \, \mu\text{g L}^{-1}$ when chlorinated influent water was used on 9/3/2005 (Biofilter 1, $9 \pm 1 \, \mu\text{g L}^{-1}$; biofilter 2, $22 \pm 2 \, \mu\text{g L}^{-1}$; biofilter 3, $12 \pm 3 \, \mu\text{g L}^{-1}$; biofilter 4, $9 \pm 0.1 \, \mu\text{g L}^{-1}$) and on 27/5/2005 (Biofilter 2, $14 \pm 6 \, \mu\text{g L}^{-1}$). Use of chlorinated water as influent to the biofilters produced the poorest outcome for AOC concentrations in the biofilter effluents.

The following observations can be made from an evaluation of the data in Table 2.

GAC was superior to anthracite as a biofilter substrate. Enhanced coagulation followed by GAC biofiltration produced a water quality almost as good as the water quality from MIEX®-GAC biofiltration. However, this may be due to the enhanced coagulation process operating extremely well during the pilot trials, producing water with a BDOC content of $0.08 \, \text{mg L}^{-1}$, which would be considered "biologically stable water", before biofiltration, a state that is not normally achieved by enhanced coagulation treatment. Coarser GAC (1.3 mm diameter) was a marginally better substrate than finer GAC (0.7 mm diameter) as it produced waters with similar or slightly better BDOC and DOC concentrations. The longer EBCT of 16 minutes was superior to the shorter EBCT of 8 minutes. The use of chlorinated backwash water was the most detrimental to biofilter performance in terms of BDOC, and

Table 2 | Average[a] AOC, DOC and BDOC concentrations in influent waters to, and effluent waters from pilot scale biofilters, and their removal under different operational settings

Operational setting	AOC[b] µg L^{-1}	BDOC mg L^{-1}	ΔBDOC mg L^{-1}	% BDOC change	DOC mg L^{-1}	ΔDOC mg L^{-1}	%DOC change
Influent water							
EBCT = 8 min (29/9/2004)	<5	0.22 ± 0.00			1.66 ± 0.01		
EBCT = 16 min (9/11/2004)	<5	0.22 ± 0.00			1.78 ± 0.00		
Enhanced coagulation (1/12/2004)[c]	21 ± 0.2	0.08 ± 0.00			1.91 ± 0.01		
Chlorinated backwash (14/1/2005)	45 ± 0	0.18 ± 0.00			1.83 ± 0.00		
Chlorinated influent (9/3/2005)	38 ± 11	0.28 ± 0.03			1.96 ± 0.01		
Chlorinated in extended (27/5/2005)	21 ± 1	0.31			1.80 ± 0.00		
Filter 1: sand/1.7 m anthracite							
EBCT = 8 min	<5	0.21 ± 0.01	0.01	5	1.64 ± 0.00	0.02	1
EBCT = 16 min	<5	0.21 ± 0.01	0.01	5	1.72 ± 0.00	0.06	3
Enhanced coagulation	<5	0.08 ± 0.00	0	0	1.81 ± 0.00	0.10	5
Chlorinated backwash	<5	0.19 ± 0.00	−0.01	−6	1.81 ± 0.01	0.02	1
Chlorinated influent	9 ± 1	0.31 ± 0.00	−0.01	−3	1.96 ± 0.01	0.00	0
Chlorinated influent (extended)	<5	0.35	−0.04	−11	1.85 ± 0.01	0.04	2
Filter 2: sand/0.6 m anthracite							
EBCT = 8 min	<5	0.25 ± 0.01	−0.03	−14	1.68 ± 0.01	−0.02	−1
EBCT = 16 min	<5	0.19 ± 0.00	0.03	14	1.74 ± 0.00	0.04	2
Enhanced coagulation	8 ± 0	0.06 ± 0.01	0.02	25	1.81 ± 0.00	0.10	5
Chlorinated backwash	<5	0.2 ± 0.01	−0.02	−11	1.79 ± 0.05	0.04	2
Chlorinated influent	22 ± 2	0.28 ± 0.02	0	0	1.90 ± 0.06	0.06	3
Chlorinated influent (extended)	14 ± 6	0.38	−0.06	−22	1.85 ± 0.01	0.04	2
Filter 3: sand/1.7 m GAC (1.3 mm)							

Table 2 | (continued)

Operational setting	AOC[b] μg L^{-1}	BDOC mg L^{-1}	ΔBDOC mg L^{-1}	% BDOC change	DOC mg L^{-1}	ΔDOC mg L^{-1}	%DOC change
EBCT = 8 min	<5	0.18 ± 0.01	0.04	18	1.54 ± 0.01	0.12	7
EBCT = 16 min	<5	0.17 ± 0.00	0.05	23	1.45 ± 0.00	0.33	19
Enhanced coagulation	<5	0.06 ± 0.01	0.02	25	1.70 ± 0.00	0.21	11
Chlorinated backwash	<5	0.16 ± 0.01	0.02	11	1.66 ± 0.02	0.17	9
Chlorinated influent	12 ± 3	0.24 ± 0.02	0.06	20	1.78 ± 0.02	0.18	9
Chlorinated influent (extended)	<5	0.27	0.04	14	1.66 ± 0.00	0.14	8
Filter 4: sand/1.7 m GAC (0.7 mm)							
EBCT = 8 min	<5	0.21 ± 0.02	0.01	5	1.57 ± 0.01	0.09	5
EBCT = 16 min	<5	0.17 ± 0.01	0.05	23	1.57 ± 0.01	0.21	12
Enhanced coagulation	<5	0.06 ± 0.03	0.02	25	1.73 ± 0.02	0.18	9
Chlorinated backwash	<5	0.18 ± 0.01	0	0	1.72 ± 0.00	0.11	6
Chlorinated influent	9 ± 0.1	0.23 ± 0.01	0.07	23	1.81 ± 0.01	0.15	8
Chlorinated influent (extended)	<5	0.28	0.04	12	1.68 ± 0.00	0.12	7

[a]Average and standard deviation of duplicate measures are given unless a single value with no error is given, in which case the concentration was measured once.
[b]AOC units are in acetate-C equivalents. Unless a single value is given, the value.
[c]Influent water was from the MIEX® clarifier on all occasions except on 1st December 2004.
Δ = average influent concentration − average effluent concentration.

the use of chlorinated influent water was most detrimental to biofilter performance in terms of AOC removal. The use of chlorinated influent water or chlorinated backwash water was detrimental to the effectiveness of the biofilters when anthracite was used as the biofilter substrate. In some cases, and especially when chlorinated influent and backwash water was used with the anthracite biofilters, the effluent from these biofilters contained more BDOC than the influent (a negative value for % BDOC removal in Table 2), which may have resulted from release of BDOC from the biomass in response to the stress from exposure to the disinfectant.

With an 8 minute EBCT, GAC biofiltration removed a greater percentage of the BDOC and DOC from water treated by enhanced coagulation than from combined MIEX®-clarified water, although influent BDOC in water after enhanced coagulation was much less (Table 2). BDOC removal was compromised with the use of chlorinated backwash water when using GAC as the substrate in the biofilters, but DOC removal was little affected. Less BDOC and DOC was removed by the GAC biofilters after extended use of chlorinated influent water for 132 days, when compared with use of chlorinated influent water over 53 days. The use of chlorinated influent water initially did not influence substantially the efficacy of the GAC biofilters in removing BDOC, but produced a poorer outcome for DOC removal. The anthracite biofilters (biofilters 1 and 2) were net BDOC producers when treating chlorinated influent water over an extended period, which probably resulted from reaction of the biomass in the biofilter with the influent disinfectant. The total mass of DOC removal across each of the biofilters was usually greater than the total mass of BDOC removal under each of the operational settings and this was more pronounced for the GAC biofilters (Table 2). This means that adsorption was playing a role in DOC and BDOC removal in addition to biodegradation of BDOC through biological activity.

The outcomes from the examination of the effects of the operating conditions on AOC, DOC and BDOC removal generally agree with the outcomes observed by others. GAC-sand biofilters generally out-perform anthracite-sand biofilters for the removal of total organic carbon (TOC) and AOC (Urfer 1998). Increases in the EBCT in GAC biofilters improve DOC and BDOC removal, although the removal was not directly proportional to the EBCT increase (Urfer *et al.* 1997).

Backwash with chlorinated water has been shown by others to severely inhibit biomass development on anthracite but not on GAC biofilters, although removal of easily degradable NOM (e.g. acetate) is not greatly affected by the use of chlorinated backwash water for either anthracite or GAC biofilters (Liu *et al.* 2001). However Urfer (1998) reported a substantial negative effect of chlorinated backwash water on biodegradable organic matter removal on both anthracite/sand and GAC/sand biofilters when chlorine levels reached $1 \, mg \, L^{-1}$, the level targeted for the chlorinated backwash test in this study. Liu *et al* (2001) recommended that the use of chlorinated backwash water in biofilters should be avoided unless occasional control of biomass build-up is required. Biofiltration with GAC has been successfully used with $3 \, mg \, L^{-1}$ free chlorine in the influent water as Cl_2 decomposes rapidly at the surface of GAC (Urfer *et al.* 1997).

NOM removal in biofilters has been shown previously to be favoured by the greater surface area afforded by smaller rather than larger diameter GAC (Wang *et al.* 1995). In our experiments, the larger diameter GAC was marginally more effective as a biofilter medium for the removal of BDOC and DOC, although the difference in the efficacy of the two substrates was probably not significant.

Effects of operational settings on removal of oxygen

Online monitoring of oxygen consumption across the biofilters at different times during each filter run, between the hours of midnight and 6:00 am, was used to determine if oxygen consumption could serve as a surrogate measure for the more laborious measures of BDOC consumption and DOC consumption. The change in DOC or BDOC was measured across each of the filters and was compared with the change in oxygen concentrations during the major sampling events. The outcomes are shown in Figures 1 and 2.

Oxygen consumption was greatest across the GAC biofilters. There were correlations, although relatively poor, between both BDOC and oxygen consumption ($r^2 = 0.45$, P = 0.006; Figure 1) and DOC and oxygen consumption ($r^2 = 0.58$, P = 0.0009; Figure 2). Oxygen consumption across the biofilters could only be used as a rough operational estimate of DOC or BDOC removal or biofilter performance. The poor correlation between BDOC and
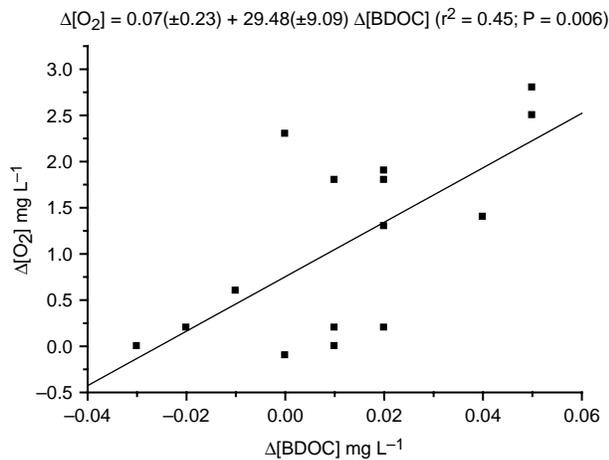
$\Delta[O_2] = 0.07(\pm 0.23) + 29.48(\pm 9.09) \Delta[BDOC] \ (r^2 = 0.45; P = 0.006)$

Figure 1 | Change in BDOC concentration versus change in oxygen concentration in the biofilters.

DOC removal and oxygen consumption across the biofilters may in some part be due to the adsorption of some DOC and BDOC on the filter matrix as a removal process rather than its removal by bacterial oxidative reactions alone.

Effects of operational settings on trihalomethane formation potential (THMFP) and chlorine demand

The effects of the different biofiltration operational settings on chlorine demand and THMFP are shown in Table 3. In general, passage of the water through the biofilters reduced the chlorine demand, with the chlorine demands in the
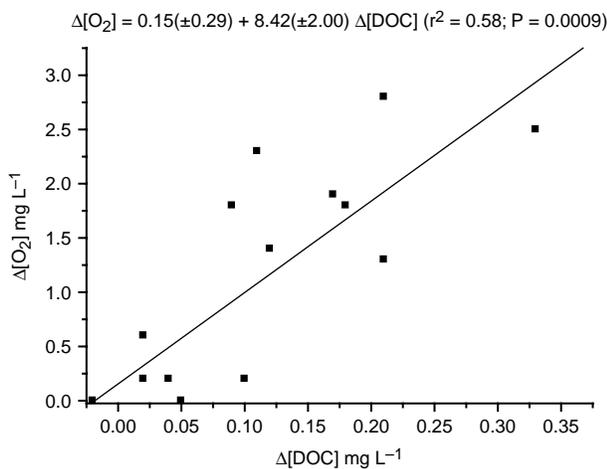
$\Delta[O_2] = 0.15(\pm 0.29) + 8.42(\pm 2.00) \Delta[DOC] \ (r^2 = 0.58; P = 0.0009)$

Figure 2 | Change in DOC concentration versus change in oxygen concentration in the biofilters.

water from GAC Biofilters 3 and 4 being reduced more than in water from the anthracite biofilters. In most instances, chlorine demand showed a greater reduction in the water passed through the finer grain GAC (Biofilter 4) than the coarser GAC (Biofilter 3), even though there was no pronounced difference in the performance of these biofilters for the removal of BDOC and DOC.

With respect to GAC biofilters, the largest reduction in chlorine demand occurred when treating non-chlorinated MIEX®-treated clarified water, and when non-chlorinated backwash water was used. Increasing the empty bed contact time from 8 to 16 minutes increased the removal of chlorine demand in water from Biofilter 3, but had a marginal effect on chlorine demand in water from Biofilter 4. When treating water from enhanced coagulation, the reduction in chlorine demand in the effluent from Biofilter 3 was much less than in the effluent from Biofilter 4.

The presence of chlorine in the influent water appeared to increase the chlorine demand in the effluent from the GAC biofilters, but paradoxically, decreased the chlorine demand in the effluent from the anthracite biofilters. One possible explanation is that the chlorine would have reacted with DOC in the water until all the free chlorine was consumed in the water passing through the anthracite filters which contained free chlorine (up to 2.80 mg/L in the influent to Biofilter 2). This reaction would have consumed some of the DOC that causes chlorine demand and would have resulted in a decrease in the chlorine demand in the effluent. In contrast, in the GAC filters, free chlorine would have been removed rapidly, either through reaction with GAC or with the adsorbed biomass, or with both. The water passing through the filters would thus not have contained any residual chlorine that could remove any of the DOC that causes chlorine demand.

The values for THM formation potential (THMFP) in the influent waters to the biofilters were quite variable during the study, ranging from $134 \ \mu g \ L^{-1}$ in the water from enhanced coagulation to $252 \ \mu g \ L^{-1}$ when chlorinated influent water was used (Table 3). It is important to note that THMFP is not a measure of the expected THMs in water upon chlorination under normal operational conditions, but rather a measure of the propensity of waters to form THMs after a 7 day reaction time with high concentrations of chlorine, such that a free chorine residual of 3 to $5 \ mg \ L^{-1}$ persists (i.e. a "worst case scenario" that would not exist in practice).

Table 3 | Chlorine demand and THMFP concentrations and changes between influent and effluent in pilot scale biofilters under different operational settings

Operational setting	Cl demand mg L^{-1}	ΔCl demand mg L^{-1}	%Cl demand change	THMFP μg L^{-1}	ΔTHMFP μg L^{-1}	%THMFP change
Influent water characteristics						
EBCT $= 8$ min[a]	3.57			144		
EBCT $= 16$ min	3.98			167 ± 109[b]		
Enhanced coagulation	4.26			134 ± 44		
Chlorinated backwash	3.87			215 ± 166		
Chlorinated influent	2.06			252 ± 338		
Chlorinated influent extended				180 ± 19		
Filter 1: sand/1.7 m anthracite						
EBCT $= 8$ min[a]	3.38	0.19	6	146	−1	0
EBCT $= 16$ min	4.54	−0.56	−14	126 ± 70	41	24
Enhanced coagulation	4.24	0.02	1	117 ± 33	16	12
Chlorinated backwash	4.05	−0.18	−5	221 ± 20	−6	−3
Chlorinated influent	1.86	0.20	10	227 ± 35	26	10
Chlorinated influent (extended)				118 ± 10	62	35
Filter 2: sand/0.6 m anthracite						
EBCT $= 8$ min[a]	3.80	−0.23	−7	140	4	−4
EBCT $= 16$ min	3.71	0.27	7	352 ± 93	−185	−111
Enhanced coagulation	4.32	−0.06	−1	151 ± 76	−17	−13
Chlorinated backwash	3.90	−0.03	−1	197 ± 80	18	9
Chlorinated influent	1.74	0.32	16	221 ± 127	31	12
Chlorinated influent (extended)				116 ± 16	63	35
Filter 3: sand/1.7 m GAC (1.3 mm)						
EBCT $= 8$ min[a]	2.24	1.33	39	85	60	28
EBCT $= 16$ min	1.94	2.04	51	84 ± 41	83	50
Enhanced coagulation	3.33	0.93	22	116 ± 11	18	14
Chlorinated backwash	2.91	0.96	25	196 ± 10	18	9

Table 3 | (continued)

Operational setting	Cl demand mg L^{-1}	ΔCl demand mg L^{-1}	%Cl demand change	THMFP μg L^{-1}	ΔTHMFP μg L^{-1}	%THMFP change
Chlorinated influent	2.03	0.03	1	239 ± 103	13	5
Chlorinated influent (extended)				89 ± 4	90	50
Filter 4: sand/1.7 m GAC (0.7 mm)						
EBCT = 8 min[a]	1.51	2.06	59	87	57	28
EBCT = 16 min	1.78	2.20	55	109 ± 14	58	35
Enhanced coagulation	2.22	2.04	48	75 ± 25	59	44
Chlorinated backwash	2.51	1.36	35	179 ± 7	36	17
Chlorinated influent	2.12	− 0.06	− 3	224 ± 11	29	11
Chlorinated influent (extended)				114 ± 8	65	36

[a] = average of measures made on four separate occasions.

[b] = where errors are given the value reports the average and standard deviation of concentrations of THMs from two measurements made on a single sample.

Δ = average influent concentration – average effluent concentration.

The two GAC biofilters consistently reduced THMFP more than the anthracite filters, although the anthracite filters still produced a significant reduction of THMFP in some circumstances (e.g. a 35% reduction when chlorinated influent water was used). Biofilter 2 generally did not perform as well as the longer anthracite biofilter (Biofilter 1) in reducing the THMFP of the water. The greatest THMFP reduction was from the GAC Biofilter 3 (50%) at an EBCT of 16 minutes, or when chlorinated influent water was used. Increasing the EBCT for GAC biofilters increased removal of THMFP from 28% to 35–50%. The effect of GAC particle size on THMFP was mixed, with larger particle size GAC removing more THMFP from MIEX®-treated clarified water, and lower particle size GAC removing more THMFP from water treated by enhanced coagulation.

Effects of operational setting on biomass in biofilter backwash waters

On each major sampling occasion, the biofilters were backwashed, and the total volume of backwash was collected in drums, returned to the laboratory, fully mixed, and the biomass in the backwash was determined by analysis of lipid-phosphate (Findlay et al. 1989) and direct microscopic counting. Biomass estimates in biofilter backwash waters for each of the filters under each of its operational settings are given in Table 4.

The total numbers of cells in the backwash ranged from 10^{12} to 10^{13} cells. However, the biomass of cells in biofilter backwashes is only a coarse indicator of biomass production in a biofilter, as the characteristics of the backwash varies each time it is done (e.g. the volume of each backwash is different on each occasion, Table 4), and although the times between backwashes were similar, they were not constant. No protozoa were observed in any of the backwash waters. The lack of grazing protozoa means that backwash is the only method of biomass removal. It has been suggested that a "strong population" of protozoa in the liquid phase is optimal for the performance of biofilters (Scholz & Martin 1997). The lack of protozoans in the biofilters may suggest that there is something specific about MIEX®-treated water that inhibits protozoa.

Lipid-P is an indirect chemical measure of the viable biomass in a sample (Findlay et al. 1989). Lipid-P in the

Table 4 | Amounts of lipid-P and numbers of cells in the backwash waters from the four pilot scale biofilters under different operational settings

Operational setting	Backwash volume (L)	Lipid-P nmol L^{-1}	Cell number ml^{-1}	Cells in total backwash
Filter 1				
EBCT = 8 min	71.2	877 ± 323	5.7×10^7	4.1×10^{12}
EBCT = 16 min	73.6	5159 ± 691	8.4×10^7	6.2×10^{12}
Enhanced coagulation	74.2	600 ± 54	6.2×10^7	4.6×10^{12}
Chlorinated backwash	77.4	318 ± 33	0	0
Chlorinated influent	89.6	16 ± 1	0	0
Filter 2				
EBCT = 8 min	117	1247 ± 117	4.7×10^7	5.5×10^{12}
EBCT = 16 min	98.2	4942 ± 302	6.9×10^7	6.8×10^{12}
Enhanced coagulation	98.8	1226 ± 212	3.2×10^7	3.2×10^{12}
Chlorinated backwash	90.6	65 ± 12	0	0
Chlorinated influent	101.2	0 ± 0	0	0
Filter 3				
EBCT = 8 min	90.2	1623 ± 888	7.1×10^7	6.4×10^{12}
EBCT = 16 min	79.8	5229 ± 8	1.5×10^8	1.2×10^{13}
Enhanced coagulation	115	3422 ± 254	1.1×10^8	1.3×10^{13}
Chlorinated backwash	100.5	361 ± 41	0	0
Chlorinated influent	114.9	43 ± 14	0	0
Filter 4				
EBCT = 8 min	114.5	3207 ± 194	8.8×10^7	1.0×10^{13}
EBCT = 16 min	99.2	4739 ± 744	1.3×10^8	1.3×10^{13}
Enhanced coagulation	77.6	2375 ± 77	9.8×10^7	7.6×10^{12}
Chlorinated backwash	61.6	445 ± 51	0	0
Chlorinated influent	68	241 ± 39	0	0

backwash was greatly reduced for all biofilters when chlorinated backwash water or chlorinated influent water was used (Table 4). No cells were observed in the backwashes from the biofilters when chlorinated influent or backwash water was used, although some lipid-P was measured under these conditions for most of the biofilters, and indicates that viable biomass did occur in the backwash. The lack of microscopically observable cells in these backwash waters probably

Table 5 | Amounts of lipid-P and biomass on the surface of the four pilot biofilters under different operational settings before and after backwashing

Operational setting	Lipid-P nmol (g dry wt.)$^{-1}$		Biomass[a] cells (g dry wt)$^{-1}$	%biomass change[b]
	before[b]	after[b]		
Filter 1				
EBCT = 8 min	7 ± 3	2 ± 1	2.7×10^8	71
EBCT = 16 min	24 ± 2	1 ± 0	9.4×10^8	96
Enhanced coagulation	107 ± 6	12 ± 5	4.2×10^9	89
Chlorinated backwash	1 ± 0	1 ± 1	3.9×10^7	0
Chlorinated influent	0 ± 0	1 ± 0	0	
Filter 2				
EBCT = 8 min	9 ± 2	6 ± 1	3.5×10^8	33
EBCT = 16 min	4 ± 2	2 ± 0	1.6×10^8	50
Enhanced coagulation	37 ± 10	18 ± 7	1.4×10^9	51
Chlorinated backwash	4 ± 2	2 ± 0	1.6×10^8	50
Chlorinated influent	2 ± 1	1 ± 1	7.8×10^7	50
Filter 3				
EBCT = 8 min	58 ± 4	26 ± 9	2.3×10^9	55
EBCT = 16 min	82 ± 5	8 ± 3	3.2×10^9	90
Enhanced coagulation	213 ± 60	90 ± 47	8.3×10^9	58
Chlorinated backwash	17 ± 4	6 ± 2	6.6×10^8	65
Chlorinated influent	2 ± 0.1	2 ± 0	7.8×10^7	0
Filter 4				
EBCT = 8 min	72 ± 5	37 ± 9	2.8×10^9	49
EBCT = 16 min	54 ± 8	1 ± 1	2.1×10^9	98
Enhanced coagulation	272 ± 3	203 ± 23	1.1×10^{10}	25
Chlorinated backwash	17 ± 4	4 ± 0	6.6×10^8	76
Chlorinated influent	6 ± 2	5 ± 1	2.3×10^8	17

[a] = determined with the conversion factor 1 nmol lipid-P = 3.9×10^7 cells. Biomass before backwash.
[b] = at the surface.

resulted from the time delay between collection of these samples and cell counting, which meant that the planktonic cells in the backwash were exposed to high concentrations of chlorine (*ca.* 1 mg L^{-1}) for some hours.

By using the cell count and lipid-P concentration data collected for the different backwash waters, with the exception of the chlorinated backwash waters, the ratio of lipid-P to cell number in the biofilter systems was calculated. For cells in this system, 1 nmol lipid-P equated to $3.9 \pm 2.4 \times 10^7$ cells (n = 12). This conversion factor is used to determine the biomass on the biofilters from the amount of lipid-P extracted from the biofilters and is similar to the conversion value determined for bacterial populations in marine sediments of 3.4×10^7 cells (nmol lipid-P)$^{-1}$ (Findlay *et al.* 1989), or 4.0×10^7 cells (nmol lipid-P)$^{-1}$ in subsurface aquifers (Balkwill *et al.* 1988).

Biomass on the biofilters

GAC or anthracite was collected for up to 10 cm depth from the surface of each biofilter before and after backwash on each of the major sampling events, and the amount of lipid-P attached to the collected material was determined (Table 5). Using the conversion value determined from the relationship of cell number to lipid-P concentration in the backwash waters, the lipid-P amounts in the biofilter surface materials were converted to a cell number, and the % change of biomass at the surface of each biofilter as a result of backwashing was calculated (Table 5). The % biomass change on the surface does not result only from biomass physically removed through backwash, but also relates to a redistribution of support material in the biofilters during backwash, as support material deep in biofilters generally carries less biomass than the surface support material (Wang *et al.* 1995).

In all biofilters, surface biomass was 3 to 9 times greater when the filters received water from enhanced coagulation than under any other operational setting. This is not surprising as the AOC content of the water from enhanced coagulation was greater than in the MIEX®-treated water unless chlorine was also being used in backwash or influent water which would restrict biofilm development through disinfection. GAC supported more biomass at the surface of the biofilter than did anthracite, which is consistent with
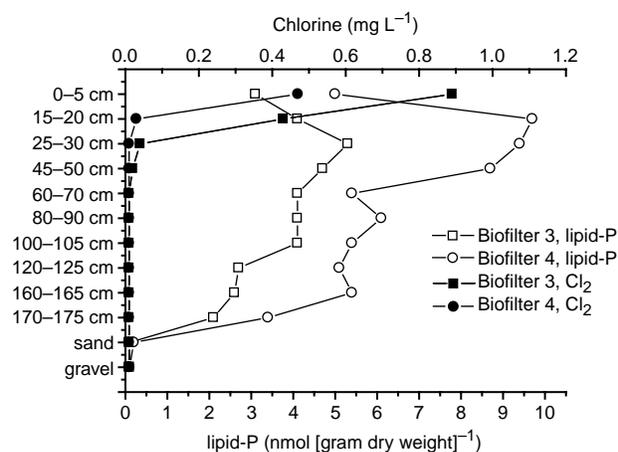

Figure 3 | Biomass (as lipid-P) in the two GAC biofilters after extended exposure to chlorinated influent water, and chlorine concentration in the pore water, at different depths within the biofilters.

the observation that the GAC biofilters removed more DOC and BDOC than the anthracite biofilters (Table 2). The amount of biomass on the surface of GAC biofilters operated by Wang *et al.* (1995) for over 100 days ranged from 269 to 750 nmol lipid-P (g medium)$^{-1}$. This range is higher than that measured in the Wanneroo biofilters (2 to 272 nmol lipid-P (g medium)$^{-1}$ (Table 5). The MIEX®-clarified water that was used as influent to the Wanneroo biofilters did contain low concentrations of biologically available organic matter (BDOC and AOC) whereas water influent to biofilters of Wang *et al.* (1995) originated from surface waters from the Ohio River and had, on occasion, undergone ozonation. Ozonation makes DOC more biodegradable and hence would be expected to promote more
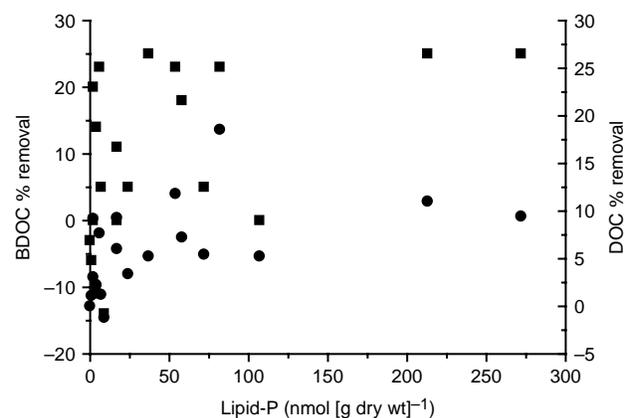

Figure 4 | Percentage BDOC (■) and DOC (●) removal as a function of amount of viable biomass (lipid-P) on the surface of four pilot biofilters.

Table 6 | Average and standard deviation of the percentage removal of UV$_{254}$ absorbance across the pilot biofilters for different operational settings

Operational setting	Biofilter 1	Biofilter 2	Biofilter 3	Biofilter 4
EBCT = 8 min	2.4 ± 5.5	3.8 ± 6.6	20.6 ± 6.9	20.6 ± 6.9
EBCT = 16 min	3.3 ± 3.5	4.1 ± 4.2	23.4 ± 7.6	22.1 ± 3.7
Enhanced coagulation	4.7 ± 2.1	4.8 ± 0.4	22.3 ± 1.7	20.8 ± 2.8
Chlorinated backwash	1.5 ± 4.2	4.5 ± 3.1	17.0 ± 7.6	17.1 ± 4.7
Chlorinated influent	3.7 ± 4.4	2.8 ± 7.6	20.2 ± 8.6	20.1 ± 10.5

biomass growth (Glaze 1987). DOC in the Ohio River water ranged from 1.1 to 2.2 mg L^{-1}, with a slightly higher maximum DOC concentration than in the MIEX-treated water influent to the Wanneroo biofilters.

The decrease in biomass at the surface of the biofilters with backwashing ranged from 0 to 98% and averaged 54%, which is large when compared with data from Miltner *et al.* (1995), who showed that for their biofilters, 22% of the biomass was removed when chlorinated water was used for backwashing, and minimal biomass change occurred when non-chlorinated backwash water was used.

The total amount of lipid-P in the surface of each biofilter was least when the biofilters received chlorinated influent water. This was expected as the influent chlorine would inhibit biomass development at and near the surface of the biofilters until the chlorine was consumed within the biofilter. In some cases the amount of biomass at the surface of the biofilter increased after backwash when chlorinated water was used as the influent water. This increase probably resulted from a redistribution of biomass carrying substrate from deeper in the biofilter to the surface of the biofilter during backwashing.

A graph showing the distribution of biomass and chlorine in the GAC biofilters while they received chlorinated influent water is shown in Figure 3. Although biomass at the surface of the two GAC biofilters was low when chlorinated influent water was used (2 to 6 nmol lipid-P (g medium)$^{-1}$), the chlorine was destroyed by the GAC to insignificant concentrations at depths 20 to 30 cm in the biofilters (see Figure 3). This allowed a recovery in the biomass within the filters at depth to 5 to 10 nmol lipid-P (g medium)$^{-1}$, but this was, at most, one fifth the biomass at

the surface of the GAC biofilters that received water which was not chlorinated (50 to 272 nmol lipid-P (g medium)$^{-1}$).

There was no relationship between the amount of biomass on the surface of the biofilters and the amount of either BDOC or DOC removed (Figure 4), as was observed by Wang *et al.* (1995).

CONCLUSIONS

Although MIEX® treatment of groundwater produced water that was low in its content of biologically available organic carbon (AOC and BDOC), biofilters fed with this water developed reasonable biomass, although somewhat less than biofilters which treat surface waters, and the biofilter treatment of the MIEX®-clarified water further improved a number of water quality parameters.

Experimentation with these pilot scale biofilters showed that for optimal performance of biofilters used for the treatment of MIEX®-clarified water, GAC outperformed anthracite as a biological support medium and two different effective sizes (1.3 mm or 0.7 mm diameter) had marginal effect on the outcome. A longer EBCT improved performance of the GAC biofilters with respect to DOC, BDOC, AOC, THMFP, chlorine demand and UV$_{254}$ absorbance removal, but the improvement in performance (e.g. DOC or BDOC removal) was not linear with respect to EBCT. Provided the MIEX®-clarified water was not chlorinated prior to its passage through the biofilters, most water quality parameters (AOC, BDOC, THMFP, chlorine demand) improved despite the high quality of the water produced via MIEX® treatment. Despite the good quality water produced by MIEX® treatment, under optimal

operating conditions, biofiltration still removed close to a further 20% DOC, 25% BDOC and 20% UV$_{254}$ absorbance (Table 6), and reduced the chlorine demand and trihalomethane formation potential by 50%. What was unusual in this case was that the biofilters fed with MIEX®-clarified water did not develop a discernable population of protozoa, which are normally considered important for optimal biofilter performance.

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