

Post treatment of groundwater denitrification fluidized bed reactor effluents to achieve drinking water quality

H Bach¹, S Tarre² and M Green²

¹Department of Molecular Microbiology and Biotechnology, Tel Aviv University, Ramat Aviv, Israel; ²Faculty of Agriculture Engineering, Technion, 32000 Haifa, Israel

Post treatment of effluents from heterotrophic groundwater denitrification fluidized bed reactors (FBR) designed to achieve drinking water quality has been investigated. The denitrification process adds to the dissolved organic compounds, biomass and bacteria in the effluent. They are also lacking dissolved oxygen. Effluents from the process were treated in combined post treatment processes based on either a trickling filter and sedimentation unit ('TF combination') or contact flocculation ('CF combination'). Both processes were followed by sand filtration, granular activated carbon (GAC) and chlorination. Results regarding total suspended solids (TSS) and turbidity removal showed an advantage to the 'CF combination', and the target turbidity (NTU <1) was always achieved when the alum dose was 10 or 20 mg L⁻¹. Backwash of the sand filter and GAC column was required after 27 h of operation (average value). An average total reduction in dissolved organic carbon (DOC) of 40% was observed with a final DOC of 3.5–4 mg L⁻¹. Most of the removal of the DOC occurred in the sand filter (28%), while the GAC contribution was smaller (18%). No regrowth potential was observed using the Werner method when a pure culture of *Pseudomonas fluorescens* P17 was used as inoculum in samples of chlorinated effluent (post chlorination). When a mixed culture of indigenous bacteria was used as inoculum, a high regrowth potential was observed. Installing an additional chlorination unit before the sand filter column (pre and post chlorination) resulted in effluent with no regrowth potential for both *Pseudomonas fluorescens* P17 and indigenous bacteria.

Keywords: fluidized bed; drinking water; post treatment; denitrification; bacterial regrowth

Introduction

In many areas of Israel, nitrate levels in groundwater have increased beyond the permissible 70 mg L⁻¹ (as NO₃⁻). Several technologies are applicable for nitrate removal; they may be classified as physico-chemical processes and biological processes. The physico-chemical processes, including ion exchange, electro dialysis and reverse osmosis, suffer from the principal disadvantage of brine production, the treatment of which is difficult from environmental and economic aspects. While the physico-chemical processes are generally unspecific and remove other constituents, biological denitrification has the advantage of selectively removing only the nitrates. However, the biological denitrification process adds to the DOC of the effluent, suspended solids and bacteria in the water. Additionally, they are lacking dissolved oxygen (DO). For these reasons, even though there has been great interest in biological denitrification of drinking water, the application of the technology has been limited. DOC and biodegradable dissolved organic carbon (BDOC) are important parameters in drinking water which indicate the potential for bacterial regrowth. Present drinking water standards do not include criteria for DOC and BDOC. This research was undertaken to evaluate post treatment of effluents from denitrification fluidized bed reactors (FBR).

Groundwater denitrification using fluidized bed (FB) reactors with ethanol as carbon source and electron donor,

including effluent post treatment has been extensively studied at the Technion, Haifa, Israel [5].

Materials and methods

Reactors

An experimental laboratory fluidized bed reactor was used whose internal diameter and height were 9 and 150 cm respectively. The bottom 10 cm of the reactor was filled with gravel to assure a uniform upflow in the reactor. Sand was used as carrier of the biomass. The basic technical information about the reactor is given in Table 1.

Table 1 Characteristics of the FBR

Parameter	Value
Total volume (L)	8.9
Internal diameter (cm)	9
Height (cm)	150
Fluidization height (m)	1.2
Recycling flow (cc min ⁻¹)	1500
Feed flow (cc min ⁻¹)	1000
Residence time (min)	9
Temperature (°C)	25
<i>Sand characteristics</i>	
Average diameter (mm)	0.86
Specific weight (g cc ⁻¹)	2.65
Bed porosity	0.65

Feeding solution

The reactor was fed with tap water enriched with nitrate (100 mg L^{-1} as NO_3^-), ethanol (60 mg L^{-1}) and phosphate (1 mg L^{-1} as PO_4^{3-}) pumped to the reactor by a peristaltic pump. A second peristaltic pump was used for recirculation. The ratio of ethanol to nitrate used (0.6) was slightly higher than the minimal value of 0.55 required to prevent the appearance of nitrites in the reactor effluents [5]. All results were obtained using a hydraulic residence time of 9 min. Nitrate removal was 97%.

FBR excess biomass removal

Two methods were evaluated for excess biomass removal. In the first method ('integrated stream'), excess biomass removal was carried out continuously within the reactor by allowing the biofilm-covered particles to flow through the recirculation pump. The shear forces of the pump effectively reduced the biofilm thickness. Post treatment experiments were carried out on effluents from the reactor which also included the suspended sheared biomass. In the second method ('separate stream'), excess biomass removal was carried out daily by draining from the reactor the portion of the biofilm-covered sand above a desired level. Stripping of the biomass from sand particles was performed by a high-speed blender. After stripping, the clean sand was returned to the reactor. In this case, post treatment experiments were carried out on effluents which did not include the waste biomass.

Post treatment process

Two alternatives for the post treatment of the denitrification reactor effluent to achieve drinking water quality were studied: (a) trickling filter followed by sedimentation unit, sand filtration, GAC column and chlorination ('TF combination'); (b) aeration unit followed by contact flocculation, sand filtration, GAC column and chlorination ('CF combination'). The following includes a description of the various elements of the post treatment processes.

Trickling filter and sedimentation: A trickling filter was built from three small bins (grooved plastic boxes), whose total dimensions were $900 \times 200 \times 200 \text{ mm}$. The bed used was recycled polypropylene strips (an average size of $8 \times 80 \text{ mm}$) and all the experiments were done with a recycling ratio of 0.5 : 1. The effluent then flowed into a sedimentation basin with a hydraulic retention time of 21 min.

Aeration and contact flocculation: A high density polyethylene tube whose empty volume was 4700 cm^3 and containing three holed partitions was used for aeration in the 'CF combination'. Alum was used for the contact flocculation and dosed directly with the FBR effluent at the beginning of the aeration unit. Aeration and contact-flocculated effluent was taken directly to the sand filter column.

Sand filter and GAC column: Two identical Plexi-glass columns were used with a diameter and height of 5.5 and 200 cm, respectively. A backwash system was installed in both columns. The sand filter was filled with a material whose characteristics are given in Table 1. The GAC column was filled with F-300 GAC (Calgon Carbon Corp).

The height of the media in both columns was 100 cm, with the remaining 100 cm provided to facilitate media expansion during backwash. A manometer was installed in the system to determine the headless in both columns. A filtration run was terminated when the headless reached 0.5 atm or when the turbidity was above 1 NTU.

Chlorination: Chlorination was carried out in a Plexi-glass tank whose residence time was 60 min and the chlorine final concentration was 1 mg L^{-1} . A mixer was installed to homogeneously distribute the chlorine.

Analyses

All analyses were carried out according to Standard Methods [1]. Nitrate (uv spectrophotometric screening method) and nitrite (sulfanilamide colorimetric method) concentrations in the reactor influent and effluent were regularly checked. The total and volatile suspended solids (TSS and VSS) concentrations were measured. The pH was measured by means of an El Hamma PBS 740 pH electrode. The DO concentration was determined by a Syland dissolved oxygen meter. The turbidity was determined by a Hach 2100P turbidimeter. The chlorine concentration was measured by the DPP technique with the color being measured at 515 nm.

Concentrations of dissolved organic carbon (DOC) were determined using a Dohrman 80 Total Carbon Analyser after sample filtration using 47 mm $0.22\text{-}\mu\text{m}$ pore size polycarbonate membrane filter (Poretics).

Regrowth potential of the post treatment effluent was determined using two types of inoculum: indigenous dominating denitrifying bacteria found in the FBR and *Pseudomonas fluorescens* P17. These two inocula were used to examine the regrowth potential according to the Werner method [9]. For this method, the samples were placed in quartz cells fitting Monitek model 251 turbidimeters attached to a HP41CX Hewlett-Packard minicomputer, which regulated the measuring time interval and recorded the turbidity.

Results and discussion

FBR effluent characterization

Characteristics of the 'separate stream' and 'integrated stream' effluents from the FBR are given in Tables 2 and 3 (TF unit). The TSS, turbidity and DOC were 55%, 59% and 28% lower in the 'separate stream' than in the 'integrated stream', respectively. The higher values observed in the 'integrated stream' were due to the grinding of the bioparticles by the peristaltic recycle pump.

Post treatment process combination including trickling filter, sedimentation unit, sand filter, GAC column and chlorination ('TF combination')

Table 2 summarizes the post treatment results for 'separate stream' by 'TF combination'. The results show total TSS reduction of about 80%, while the trickling filter/sedimentation unit removed 43% of the TSS. No significant differences in the removal of TSS were detected for both filtration velocities (9 and 18 m h^{-1}). Total tur-

Table 2 Results of post treatment (average values) for 'separate stream'—'TF combination'

Parameter	TF unit ^a		SF out ^b		GAC out ^c	
	In	out				
Filtration velocity (m h ⁻¹)			9	18	9	18
Time to backwash (h)			53	32		
TSS (mg L ⁻¹)	8.67 ± 7.42	4.97 ± 1.46	1.70 ± 0.35	1.67 ± 0.51	1.65 ± 0.30	1.42 ± 0.42
Turbidity (NTU)	3.67 ± 2.05	2.14 ± 0.58	0.93 ± 0.06	0.95 ± 0.21	0.57 ± 0.06	0.87 ± 0.16
DOC (mg L ⁻¹)	6.63 ± 1.39	6.64 ± 0.25	5.00 ± 0.74	ND	3.96 ± 0.16	ND
DO (mg L ⁻¹)	0.00	4.48 ± 0.91				

^aTrickling filter/sedimentation effluent.

^bSand filter effluent.

^cGAC column effluent.

ND, no data.

Table 3 Results of post treatment (average values) for 'integrated stream'—TF combination'

Parameter	TF unit ^a		SF out ^b		GAC out ^c	
	In	out				
Filtration velocity (m h ⁻¹)			9	18	9	18
Time to backwash (h)			64	21		
TSS (mg L ⁻¹)	19.23 ± 8.25	6.35 ± 1.67	1.77 ± 0.39	1.21 ± 0.19	2.28 ± 0.72	1.96 ± 0.42
Turbidity (NTU)	8.61 ± 2.73	2.86 ± 0.92	1.15 ± 0.08	1.42 ± 0.25	0.88 ± 0.23	0.88 ± 0.88
DOC (mg L ⁻¹)	9.14 ± 3.01	6.64 ± 0.25	4.88 ± 0.81	ND	4.08 ± 0.63	ND
DO (mg L ⁻¹)	0.00	4.48 ± 0.91				

^aTrickling filter/sedimentation effluent.

^bSand filter effluent.

^cGAC column effluent.

ND, no data.

bidity was reduced by 84% and 76% for filtration velocities of 9 and 18 m h⁻¹, respectively; while the trickling filter/sedimentation unit removed 42% of the turbidity. DOC concentration was not reduced at all by the trickling filter/sedimentation unit, while in the sand filter and GAC column a removal of 40% was observed. DO increased from 0.0 to 4.4 mg L⁻¹.

The average desired turbidity (<1 NTU) was achieved at both filtration velocities, but due to the large standard deviation in the GAC effluent, the results are acceptable only at a filtration velocity of 9 m h⁻¹.

Table 3 summarizes the post treatment results for 'integrated stream' by 'TF combination'. Total TSS was reduced by about 90%, while the trickling filter sedimentation unit removed 67% of the TSS. No significant differences in the removal of TSS were found for both filtration velocities. Total turbidity was reduced by 90%, while the trickling filter/sedimentation unit removed 66% of the turbidity. The contribution of the GAC to the removal of TSS and turbidity was either zero or very small. Here again, no significant differences were found for both filtration velocities. Total DOC concentration was reduced by 55%, 27% removal by the TF/sedimentor unit and 39% by the sand filter and GAC column. DOC concentration in the final effluent was similar in both the 'integrated' and 'separate stream': DO was increased from 0.0 to 4.4 mg L⁻¹. Results show that the average desired turbidity (<1 NTU) was

reached at both filtration velocities, but here again with a large standard deviation: an average value of 63% in the GAC effluent (between 26% and 100%). Although the 'integrated stream' originally had much higher turbidity and a higher SS concentration than those of the 'separate stream', after the post treatment both streams were very similar.

Post treatment process combination including aeration, contact flocculation, sand filter, GAC column and chlorination ('CF combination')

Results for the post treatments of both 'separate' and 'integrated' streams are given in Tables 4 and 5. The results indicate that for both 'separate' and 'integrated' stream processes, the desired turbidity was achieved, except for the case of an alum dose of 5 mg L⁻¹.

Using the 'CF combination' and an alum dose of 10 or 20 mg L⁻¹ resulted in very low NTU values: about 0.5 NTU. However, time to backwash was much shorter in the 'CF combination': an average of 27 h in contrast to 60 h in the 'TF combination'.

An average total reduction in TSS concentration of 79% was observed. Results show that in most cases GAC column contribution to TSS and turbidity removal was negative.

Based on DOC results in the 'TF combination' which showed no significant differences between 'integrated' and

Table 4 Turbidity results (in NTU) of ‘CF combination’ (‘integrated’ and ‘separate’ streams) at filtration velocity of 9 m h⁻¹

Stream	Alum ^a	Backwash ^b	FBR		SF out ^c	GAC out ^d
			In	out		
‘Separate’	5	29	0.39 ± 0.09	2.92 ± 1.36	0.93 ± 0.24	1.04 ± 0.18
	10	25	0.39 ± 0.09	3.26 ± 0.51	0.77 ± 0.14	0.78 ± 0.09
	15	28	0.39 ± 0.09	3.68 ± 1.28	0.57 ± 0.18	0.68 ± 0.17
	20	26	0.39 ± 0.09	2.80 ± 0.98	0.44 ± 0.22	0.49 ± 0.19
‘Integrated’	5	12	0.39 ± 0.09	8.02 ± 0.24	1.41 ± 0.06	1.21 ± 0.08
	15	16	0.39 ± 0.09	8.50 ± 2.93	0.70 ± 0.20	0.77 ± 0.15
	20	26	0.39 ± 0.09	6.92 ± 2.64	0.28 ± 0.08	0.63 ± 0.14

^aAlum dose (mg L⁻¹).
^bTime to backwash (h).
^cSand filter effluent.
^dGAC column effluent.

Table 5 DOC and suspended solids results (mg L⁻¹) of ‘CF combination’ (‘separated’ stream) at filtration velocity of 9 m h⁻¹

	FBR		SF out ^a	GAC out ^b
	In	out		
DOC	3.10 ± 0.23	6.63 ± 1.39	4.81 ± 0.68	3.96 ± 0.67
Suspended solids	0.62 ± 0.23	8.67 ± 7.42	1.70 ± 0.49	1.85 ± 0.42

^aSand filter effluent.
^bGAC column effluent.

‘separate’ streams, DOC concentrations were measured only for the ‘separate’ stream. An average total reduction in DOC concentration of 40% was observed. Most of the degradation occurred in the sand filter (28%), while the GAC removed another 18%. Similar results were reported previously [3]; an average DOC removal by GAC of 15–20% was found when the feeding concentration was 3.90 mg L⁻¹.

Improving DOC removal by GAC

Since the final DOC concentration after the GAC column was relatively high, about 4 mg L⁻¹, additional experiments to improve DOC adsorbance on GAC were conducted:

1. Reducing the filtration velocity to 2.5 m h⁻¹ in continuous experiments: While in 9 m h⁻¹ velocity, the DOC concentration reduced from an average value of 5 ± 0.74 to 3.98 ± 0.80 mg L⁻¹, decreasing the filtration velocity to 2.5 m h⁻¹ resulted in effluent with DOC concentration of 2.96 ± 0.12 mg L⁻¹.

2. Increasing concentrations and exposure time in batch experiments: The effect of long exposure periods was studied in batch experiments with filtrated FBR effluents stirred with new GAC F-300 added to the effluent in excess. Samples were taken at different defined times and the DOC concentration was determined. A final DOC concentration of 2.8 mg L⁻¹ was measured after 180 min (Figure 1).

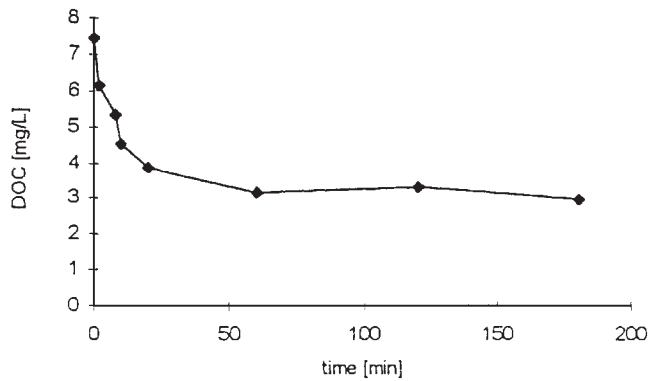


Figure 1 Reduction in DOC concentration as a function of time (GAC F-300).

3. Testing other types of GACs including F-100 and F-400 (Calgon Co) and Merck GAC in continuous experiments: The performance of the different GACs was studied in continuous experiments using a ‘CF combination’ with an alum dose of 20 mg L⁻¹ and a filtration velocity of 2.5 m h⁻¹. No significant improvement in DOC removal was observed (Table 6). The results show that using other types of GAC did not reduce DOC concentration. The final DOC concentration was again 2.8 mg L⁻¹. The results show that the DOC remaining in the effluent after sand filtration had little affinity to the GAC. Similar results were obtained by others [2,6]. Other approaches that reduce DOC concentrations below the values obtained in this research were applied on reservoir water which may be characteristically different than FBR effluents [8]. DOC

Table 6 Comparison of DOC removal in different GAC types

GAC type	DOC (mg L ⁻¹)	
	FBR effluent	GAC effluent
F-100	6.72	2.79
F-300	7.45	2.81
F-400	6.28	3.37
Merck	6.35	5.45

constituents from the FBR are probably the result of bacterial activity including complex and simple sugars, proteins, fatty acids, etc that are known to have little affinity to GAC [4,7]. There are no specific data available in the literature on the composition of the DOC constituents.

Regrowth potential

Regrowth potential was studied only on the effluent of the 'CF combination' since only this combination was found to give low enough NTU values, while DOC removal was similar in both combinations. All experiments were done using an alum dose of 20 mg L⁻¹ and a filtration velocity of 9 m h⁻¹. Regrowth potential of the final chlorinated effluents, ie, after GAC column effluent chlorination (post chlorination) was studied using the Werner method [9]. Results are shown in Figure 2.

According to the Werner method, a regrowth exists when the maximum growth rate (MGR) is above 0.15 h⁻¹. In this experiment *Pseudomonas fluorescens* P17 was used as inoculum in samples taken from the chlorinated effluent. The MGR values observed were 0.0863, ie no regrowth was observed (<0.15 h⁻¹). Since pure cultures may sometimes fail to show the maximum regrowth potential, a second experiment was performed using indigenous bacteria originating from the FBR. Results showed a very high MGR of 0.3009 h⁻¹ (Figure 2).

To lower the regrowth potential, an additional unit for prechlorination (final chlorine concentration of 1 mg L⁻¹ and contact time of 1 h) was installed before the sand filter column to check whether oxidation of the organic matter will affect its biodegradability and/or improve organics adsorption in the GAC column.

The results from the experiments carried out in the 'CF combination' system, including both 'pre' and 'post chlorination', showed MGR <0.08 h⁻¹ (no regrowth) when *Pseudomonas fluorescens* P17 was used as an inoculum. Similar results were observed when indigenous bacteria from the FBR were used as inoculum (Figure 3). Although the regrowth potential decreased dramatically when prechlorination was practised, no significant decrease in the DOC concentration was observed: from 4.00 to 3.80 mg L⁻¹. These results indicate that the prechlorination

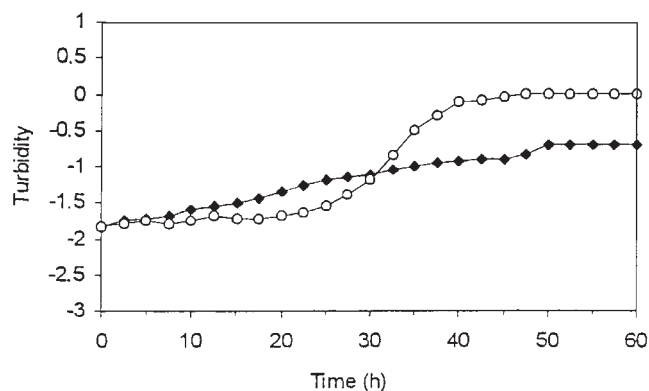


Figure 2 Results of batch experiment to determine regrowth potential (Werner method) of chlorinated effluent (post chlorination) using indigenous bacteria (—○—) from the denitrification reactor and *Pseudomonas fluorescens* P17 (—◆—) as inocula.

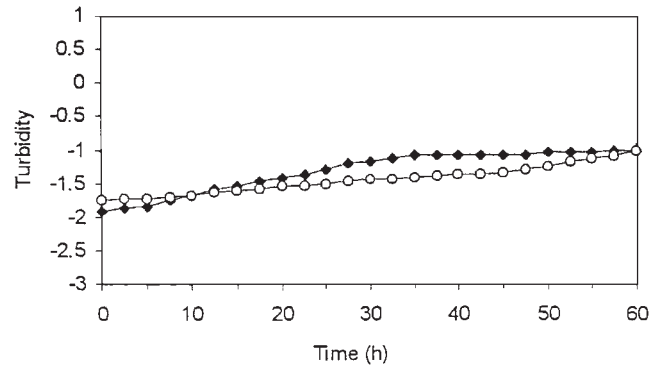


Figure 3 Results of batch experiment to determine regrowth potential (Werner method) using *Pseudomonas fluorescens* P17 (—◆—) and indigenous bacteria (—○—) from the denitrification reactor as inocula on effluent after pre and post chlorination units.

caused a decrease in the biodegradability of the organic matter but did not change its affinity to GAC.

Conclusions

This research project concentrated on the post treatment of heterotrophic denitrification FBR effluents to achieve drinking water quality. Although biological denitrification FBR effluents contain very low nitrate concentrations, the process adds to the dissolved organic compounds, suspended solids and bacteria in the effluent. Additionally the effluents are lacking dissolved oxygen. Therefore, post treatment of the effluent water is necessary to achieve drinking water quality. The following conclusions were extracted from this research.

Two methods for the removal of excess of biomass were used:

- The 'integrated stream' had originally much higher SS concentration and higher turbidity than those of the 'separate stream'; however, the results showed that the effluents after the post treatment from both streams were very similar.
- Results regarding TSS and turbidity removal showed an advantage to the 'CF combination' and the target turbidity (NTU<1) was always achieved on both 'separate' and 'integrated' streams when the alum dose was 10 or 20 mg L⁻¹. Backwash of the sand filter and the GAC column was required after 27 h of operation (average value).
- An average total reduction in DOC concentration of 40% was observed with a final DOC concentration of about 4 mg L⁻¹. Most of the degradation of the DOC occurred in the sand filter (28%), while the GAC contribution was small (18%). Experiments conducted to improve the DOC removal by GAC (reducing the filtration velocity, using different kinds of GAC, long exposure time) showed that the minimal DOC concentration that can be achieved with the GAC is 2.8 mg L⁻¹. This high DOC value indicates low affinity of the DOC constituents (mainly from bacterial origin) to the GAC.
- When a pure culture of *Pseudomonas fluorescens* P17 was used as inoculum in samples of chlorinated effluent

(post chlorination), no regrowth potential was observed. When a mixed culture of indigenous bacteria was used as inoculum, a high regrowth potential was observed.

- Installing an additional chlorination unit before the sand filter column (pre and post chlorination) resulted in an effluent with no regrowth potential. This result obtained for both *Pseudomonas fluorescens* P17 and indigenous bacteria as the inocula.

References

- 1 APHA. 1985. Standard Methods for the Examination of Water and Wastewater, 16th edn. American Public Health Association, Washington, DC.
- 2 Bonnet MC, B Welte and A Montiel. 1992. Removal of biodegradable dissolved organic carbon in a water treatment plant. *Wat Res* 26: 1673–1680.
- 3 Carlsson M, K Heffernan, C Ziesemer and E Snyder. 1994. Comparing two GACs for adsorption and biostabilization. *AWWA* 3: 91–102.
- 4 Ford DL. 1981. Wastewater characteristics and treatment. In: Activated Carbon Adsorption for Wastewater Treatment (Perrich J, ed), pp 1–27. CRC Press, Boca Raton, FL.
- 5 Green M, M Shnitzer and S Tarre. 1994. Fluidized bed reactor operation for groundwater denitrification. *Wat Sci Tech* 29: 509–515.5.
- 6 Malley J, T Taylor, M Collins, J Royce and D Morgan. 1993. The performance and microbiology of ozone-enhanced biological filtration. *AWWA* 12: 47–57.
- 7 Rice R and M Robson. 1982. Biological processes in the treatment of drinking water. In: Biological Activated Carbon, pp 145–196, Ann Arbor Science Publishers, Ann Arbor, MI.
- 8 Schmidt P, J Tobiason, J Edzwald and H Dunn. 1995. DAF treatment of a reservoir water supply. *Wat Sci Tech* 31: 103–111.
- 9 Werner P and B Hamsch. 1986. Investigations on the growth of bacteria in drinking water. *Wat Supply* 4: 227–232.