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Bioflocculation: Chemical free, pre-treatment technology for the desalination industry

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ABSTRACT

Rapid sand filtration (RSF), proceeded by chemical coagulation and flocculation, is a commonly used, effective pretreatment in the desalination industry. We designed and tested a novel, large pilot-scale, two-stage granular Rapid Bioflocculation Filter (RBF) based on a first-stage Bioflocculator (BF) unit followed by a mixed-media bed filter (MBF). The BF filter bed consisted of an extremely porous volcanic *Tuff* granular medium which provided an enlarged surface area for microbial development and biofilm proliferation. We compared the efficiency of the pilot RBF to that of a full-scale RSF, operating with upstream chemical coagulation, by measuring the removal from the same untreated seawater feed of key factors related to membrane clogging: SDI, turbidity, chlorophyll *a* (Chl *a*) and transparent exopolymer particles (TEP). After 2 weeks of operation, the *Tuff* grains were colonized extensively by coccoid bacteria that formed biofilm along the entire BF. With bacterial colonization and biofilm development, numerous aggregates of bacteria and some algal cells embedded in an amorphous organic matrix were formed on and within the *Tuff* grains. By 1–3 months, the biotic diversity within the *Tuff* filter bed had increased to include filamentous bacteria, cyanobacteria, fungi, protista and even crustaceans and marine worms. During and for ~24 h after each cleaning cycle (carried out every5 to 7 days by upward flushing with air and water), large numbers of floc-like particles, from ~15 μm to ~ 2 mm in size were observed in the filtrate of the BF unit. Microscopic examination of these flocs (stained with Alcian Blue and SYTO^{R} 9) showed that they were aggregates of many smaller particles with associated bacteria and algae within a polysaccharide gel-like matrix. These biogenic flocs (*bioflocs*) were observed to form during normal operation of the RBF, accumulating as aggregates of inorganic and organic material on the *Tuff* surfaces. With each flush cleaning cycle, these bioflocs were released into the BF effluent but were retained by the second phase MBF unit. No flocs were seen in the MBF filtrate. Over a year-long study, both the pilot RBF and the full-scale RSF showed similar filtration efficiencies, measured as the percentage removal of Chl *a*, TEP, turbidity and SDI from the same seawater feed. These results indicate the potential of the bioflocculation approach with no chemical additives as an alternative to conventional RSF pretreatment for large SWRO facilities.

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1. Introduction

The steep rise in world population coupled with rising demand for drinkable water without no foreseeable increase in natural freshwater supply, is predicted to cause a global shortage of potable water (Service, 2006). Extensive large-scale desalination will therefore serve as the main source of artificial freshwater supply, with desalinated water coming online at a rate of 40–50 million $m^3\ d^{-1}$ and predicted to increase up to 100 million $m^3\ d^{-1}$ in 20 years (Fritzmann et al., 2007).

Reverse osmosis (RO) desalination is a membrane filtration process where high salinity (up to ~40 ppt) intake water is forced at high pressure through specially structured membranes that reject salts and minerals while being highly permeable to H_2O molecules. Energy consumption needed to desalinize water (40–45% recovery), is around 3.6–4 kW h m^{-3} and depends on the osmolarity of the feedwater and the level of clogging due to membrane fouling. Membrane fouling is caused by inorganic scaling and by biofilm fouling by organic matter and growth of microbial communities. To minimize RO fouling two types of feedwater pretreatment techniques are commonly used in desalination facilities; granular medium gravity filtration and ultrafiltration (UF) membrane filtration.

In most large-scale seawater RO (SWRO) facilities, currently the main pretreatment procedure is based on granular media filters, also termed rapid sand filters, RSF. The widespread use of RSFs is due to their relative simplicity, low energy consumption, and overall low operational costs, despite the usual need to apply chemical coagulant such as ferric sulfate to the feedwater upstream (Bratby, 2006; Greenlee et al., 2009; Pearce, 2009). RSFs, with flow velocities of 5–30 m h^{-1} and water residence times of ~10–15 min are designed to remove suspended solids (>0.35 mm) from feedwater and reduce silt density index (SDI) levels to ~ 4 (arbitrary units, a.u.). Recent studies have shown that RSF can also reduce the amounts of particulate and dissolved organic carbon, chlorophyll *a* and transparent exopolymeric particles (TEP) in feedwater (Bar-Zeev et al., 2012a). In operation, when a RSF becomes overloaded with particles, a backwash procedure is applied to flush out the particulate matter that has accumulated in the filter bed (Drami et al., 2011).

For wastewater treatment, Slow Sand Filtration (SSF) with flow velocities ranging between 0.1 and 0.3 m h^{-1} and water residence times of 12–24 h, is a long-established, standard water cleansing method (Graham, 1999). In these SSF systems, biofiltration is the major process whereby a diverse microbial community that forms biofilm on the surfaces of the granular filter bed medium acts to decompose various organic pollutants to purify the wastewater (Bouwer and Zehnder, 1993).

As yet biofiltration has not been considered as a practical option in RSF operation due the high flow rates and extremely short water residence times compared to SSF. It was assumed that because of the short residence time of the feedwater within the filter bed no effective biodegradation processes would occur within the RSF. However, recent studies showed that a limited amount of biodegradation occurred in an operational RSF at a SWRO plant (Bar-Zeev et al., 2012a; Belkin et al., 2012). On this basis, it was suggested that some modification of RSF design could exploit this biofiltration potential to increase the overall pretreatment performance of these filters.

Here we describe an innovative, chemical-free, microbial-based pretreatment technology, utilizing a novel RSF configuration together with an extremely porous volcanic *Tuff* filtration medium which provides an enlarged surface area for microbial development and biofilm proliferation. We monitored the efficiency of a pilot-scale granular filter by measuring the removal from the feedwater (untreated seawater) of key factors related to membrane clogging; silt density index (SDI), turbidity, chlorophyll *a* (Chl *a*) and transparent exopolymer particles (TEP).[2]

The results from one year of operation of a large-scale pilot, dual-stage granular filter, designed to optimize microbiological activity within the filter bed, indicate that this pretreatment technology with no addition of coagulants or other chemicals gave results equivalent to a conventional RSF with prior chemical ($Fe_2[SO_4]_3$) treatment.

2. Materials and methods

2.1. Sampling site description

The pilot Rapid Bioflocculation Filter (RBF) was constructed with two fiberglass columns (each 6 m high and 1 m diameter); an upward flowing Bioflocculator (BF) unit, packed with 3 m natural, porous volcanic *Tuff* medium (*Tuff* Merom Golan Co.) and a downward flowing, mixed-media bed filter (MBF) consisting of 80 cm Filtralite® (http://www.filtralite.com/) over 80 cm sand (Fig. 1). Note, the configuration of the filter bed media in the MBF unit corresponded to that of a standard RSF. During the entire year of operation, neither *Tuff* medium in the pilot RBF nor the media in the MBF were replaced.

Flow conditions were maintained by pumping untreated seawater upward through the BF at 50 m h^{-1}. The flow then passed by gravity down through the MBF at 10 m h^{-1} (Fig. 1). Excess filtrate from the BF (~40 m h^{-1}) was discarded via the overflow. During the entire test period, the filter bed media of both the BF and MBF were routinely cleaned by upward flushing with air (10 min), air and water (20 min) and water (20 min). Preliminary testing of the RBF indicated that when flush cleaning procedures were carried out after 21–30 days severe clogging (characterized by large, 1–10 cm, mud balls) occurred, resulting in a filter breach. Therefore during this study the RBF was flush cleaned at 5–7 day intervals. The volumetric flow rate during normal operation between filter flushing was ~1190 $m^3\ d^{-1}$ with a loading rate of approximately 6–8 m h^{-1}. Note, no chlorination was ever carried out and no coagulants were added to the feedwater at any stage or time of the RBF operation.

In order to evaluate the pilot RBF performance, we compared RBF filtration efficiency in respect to the same

[2] Chlorophyll *a* is an easily measured proxy for algal concentrations. TEP are ubiquitous, microscopic (~1–200 μ m), organic gelatinous particles that have been shown to be involved in biofilm formation and cause membrane clogging (Bar-Zeev et al., 2012b; Berman et al., 2011).

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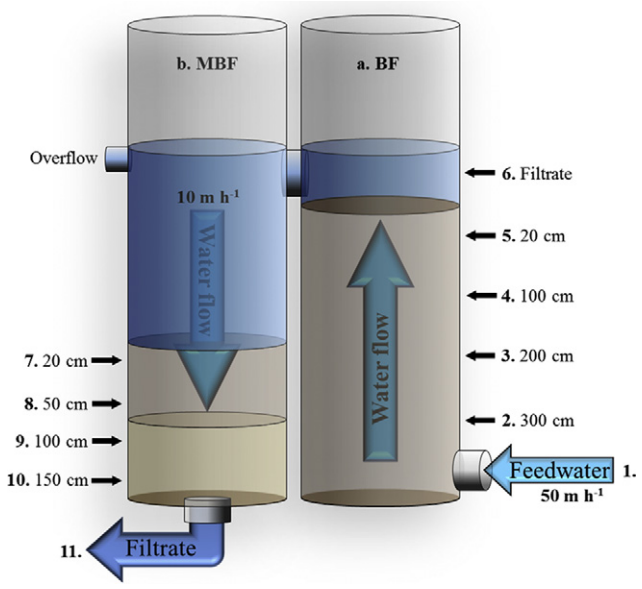

Fig. 1 – Schematic overview of the pilot, two-stage Rapid Bioflocculation Filter (RBF). a. Bioflocculator (BF) unit with volcanic *Tuff* granular medium b. Mixed Bed filter (MBF) with Filtralite® above sand media. The numbers correspond to sampling locations as indicated (see text for details).

water quality parameters of a full-scale, operating RSF that received the identical feedwater. However, in this case, throughout the year, the seawater was treated upstream of the RSF with coagulants ($Fe_2[SO_4]_3$) ranging in concentration between 0.2 and 1 ppm. The RSF was routinely backwashed after no more than 72 operating hours. RSF sampling was performed approximately 15 h after the backwash.

2.2. Sampling strategies and methods

Sampling locations for the RBF were: (**1**) Plant intake (seawater feed reference); (**2** to **5**) four depths within the BF (~300, 200, 100, 20 cm), (**6**) BF filtrate; (**7** to **10**) four depths within the MBF (~20, 50, 100, 150 cm); (**11**) RBF filtrate (see Fig. 1). In parallel, samples were taken from the filtrate of an operating, full size RSF (**12**) that received the same feedwater. Interstitial water and granular samples from the filter bed were collected on 9 sampling dates from August 2010 to July 2011.

A specially constructed sampler/corer (Belkin et al., 2012) was used to sample interstitial water and particulate media from different depths within the filter bed. Particulate granular samples were stored in 15 ml polycarbonate centrifuge tubes (BD Falcon Labware) at 4 °C until analysis in the laboratory. Water (3 L) was pumped directly from each sampling depth into clean flasks and analyzed within several hours at the laboratory.

At each sampling date, the following water quality parameters were determined: chlorophyll *a* (Chl *a*), transparent exopolymer particles (TEP) and turbidity (as nephelometric turbidity units, NTU) in the feedwater and from the interstitial water and filtrates of the BF and MBF, as well as from a full-scale, operating RSF (see above for sampling locations). SDI was measured only in the RBF and RSF filtrates. The establishment and development of microbial communities and biofilm on the *Tuff*, Filtralite® and sand media grains was followed by scanning electron microscopy (SEM).

2.2.1. Chlorophyll a (Chl a)

Samples (150 ml) were collected on Whatman GF/F filters and Chl *a* was extracted in cold acetone (90%) overnight in the dark. Chl *a* concentrations ($\mu g\,L^{-1}$) were determined fluorometrically in duplicate samples according to Holm-Hansen et al. (1965).

2.2.2. Transparent exopolymer particles (TEP)

For TEP determinations, water samples (100 ml) were filtered gently (<150 mbar) onto 0.4 μm polycarbonate filters and TEP concentrations were quantified as μg Gum Xanthan (GX) equivalents L^{-1} by the method of Passow and Alldredge (1995).

2.2.3. Turbidity (NTU)

Water samples (10 ml) were measured on site using a portable Turbidimeter (2100P, HACH Company, USA).

2.2.4. Dissolved oxygen (DO) measurements within the filter bed

To measure dissolved oxygen DO concentrations at various depths in the filter media, we used a specially designed corer that contained a pre-calibrated combined optode and temperature sensor connected to a Microx TX3 oxygen meter (PreSens Precision Sensing GmbH) and PC (Belkin et al., 2012).

2.2.5. Silt density index (SDI)

Samples (20–40 L) were taken from filtrates of the RBF and an operating RSF at the Hadera Desalination Plant, Israel. SDI was measured with a commercial multi-channel on-line Quality Water Analyzer (SDI 2200, MABAT Chemical Systems Ltd).

2.2.6. Scanning electron microscopy (SEM) of filter bed grains

Precise depth sampling of media grains within the filter beds was carried out with a specially designed corer (Belkin et al., 2012). The granular samples were immediately fixed using 0.7 ml of Karnovsky solution (2% paraformaldehyde and 2.5% glutaraldehyde in 0.06 M Sorenson's phosphate buffer, pH 7.2–7.4) and kept at 4 °C prior to preparation for SEM imaging according to Gamliel (1985). When dry, samples were carbon coated for 60 s in a Desk IV Denton Vacuum. Three to 5 grains from each sample were analyzed using a JEOL 840 scanning electron microscope at 4000X magnification.

2.2.7. Floc counting and characterization

At the beginning of each flush cleaning cycle, the effluent from the BF contained numerous microscopic flocs. Particle counts of flocs in the BF filtrate were made in triplicate with a laser particle counter (Hach, USA). Samples were run at concentration mode (particles ml^{-1}). Double distilled water was taken as a blank and subtracted as background. This instrument measured only particles >2 μm.

In order to examine the composition of the flocs, BF filtrate samples (10 mL) were stained in a 15 mL centrifuge tube with 500 μl Alcian Blue (Sigma; 0.4% w/w at pH 2.5) and 7 μl of 5 mM SYTO® 9 (Invitrogen) for TEP and bacteria detection,

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respectively. Samples were rinsed three times after each staining procedure with 0.2 μm filtered seawater to remove any excess stain. The flocs were resuspended in 10 mL of 0.2 μm filtered seawater, placed in a petri-dish and visualized with a Nikon Eclipse 80i microscope equipped with 10 × /0.45plan fluor lens using epifluorescence microscopy for bacteria (green) and bright-field microscopy for TEP (blue). Algal cells were identified by their chlorophyll *a* auto-fluorescence (red).

For morphological examination, 10 mL of BF filtrate samples were placed in a petri-dish. Several hundred micrographs of each sample were taken using a binocular microscope (Nikon AZ_{100}-STD with AZ Plan Fluor X2 lens). Floc sizes were quantified using Image J software (http://rsbweb.nih.gov).

To quantify the percentage of organic matter in the flocs, 10 mL of the BF filtrate were filtered on pre-combusted GF/F filters (Whatman). The samples were dried overnight at 60 °C and weighed. Then samples were combusted overnight at 450 °C and the organic content was determined as the weight loss on combustion.

2.2.8. *Flush cleaning experiments*

Two experiments were run to examine the effects of flushing on the functioning of the BF unit of the Pilot RBF. In each case, the RBF was operated normally for 5–7 days prior to flushing in order to provide a build-up of material within the filter bed.

The first experiment was designed to follow in detail the particulate material removed at the onset of flushing from the *Tuff* filter bed of the BF. A constant air–water flush was carried out for 100 min during which samples were taken from the BF filtrate at 1, 2, 5, 7, 10, and 100 min. These samples were analysed for floc size distribution and turbidity.

The second flushing experiment examined the impact of an extended flushing procedure (4 h) on material that had accumulated within the BF filter bed as well as the ability of the system to resume desired filtration efficiencies following flushing. Turbidity, particle count, TEP, Chl *a* and floc sizes were chosen as indicators of water quality to evaluate the filtration efficiency. In this case, all parameters were normalized to feedwater concentrations (taken as 100%) and measured after several days of normal operation immediately prior to the start of flushing. A constant air–water flush was carried out for 4 h during which interstitial water and filtrate samples were collected at 1–5, 60 and 240 min. Then flushing was stopped and the RBF resumed normal operation with a feedwater volumetric flow rate of 50 $m^3\ h^{-1}$ for a further 15 h when interstitial water and filtrate samples were again taken (1140 min after start of the experiment). Subsequently, a second, short (several minutes only) cleaning flush was activated, immediately after which filtrate samples were taken for floc size measurement. Note, floc sizes were determined in the BF filtrate prior to the flush procedure (PF), 1, 60, 240, 1140 min and after the second flush (Table 1).

Table 1 – Biofloc size distributions in the BF effluent during the second, long-term flush cleaning experiment.[a]

	Pre-flush	Time (min.)			
		1–5	60	240	1140
Floc size range (μm)	No flocs	17–1575	12–264	8–220	9–334
Floc av. size (μm)	No flocs	137 ± 184	58 ± 36	44 ± 35	58 ± 50
		$n = 364$	$n = 217$	$n = 93$	$n = 107$
Floc size distribution (%)					
0–10 (μm)	No flocs	0	0	1	1
10–50 (μm)	No flocs	27	52	77	52
50–100 (μm)	No flocs	31	38	17	38
100–200 (μm)	No flocs	27	9	5	7
200–500 (μm)	No flocs	12	1	0	2
500–1000 (μm)	No flocs	2	0	0	0
>1000 (μm)	No flocs	1	0	0	0

a see text for details.

3. Results and discussion

3.1. *Bioflocculator (BF) medium; porous volcanic* Tuff

The RBF technology depended on the properties of natural, crushed volcanic *Tuff* used as the BF filter bed medium. This highly porous granular material provided a significantly greater surface area available for microbial colonization during filter operation than other conventionally used media. Additionally the biofilm that developed within the *Tuff* pores was less susceptible to removal during cleaning flushes and provided a substrate for subsequent rapid bacterial proliferation and biofloc formation (see section 3.3).

Volcanic *Tuff* grain sizes ranged between 3 and 5 mm in diameter, with a bulk density of 2110 kg m^{-3} and porosity relative to volume of 26.7%. The total pore area was 20 $m^2\ g^{-1}$, with extremely wide pore size ranges (0.05 to >10 μm); median pore diameter was 0.75 μm with a characteristic pore length of 62 μm. The large range of pore sizes enabled a wide diversity of microorganisms to colonize the medium pores as a result of reduced shear forces (Characklis and Marshall, 1990). Although detailed chemical characteristics of *Tuff* were not analyzed, it is likely that iron is a significant constituent, given the volcanic origin of this material (Silber et al., 1994). We speculate, similarly to others (Mills et al., 1994) that iron oxides within the *Tuff* grains might endow the inner pore surfaces with a positive charge, which would enhance the interaction between negatively surface charged particles such as bacteria,[3] TEP and organic colloids (Stevik et al., 2004; Van Loosdrecht et al., 1989).

3.2. *Development of microorganisms in the BF* Tuff *filter bed*

At the start of RBF operation, the *Tuff* grains were devoid of any biological coverage. After 2 weeks of operation, the *Tuff* grains were colonized extensively by coccoid bacteria forming a biofilm covering along the entire BF column especially in cracks, crevices and tunnels of the medium (Figs. 2 and 3). By 1–3 months of RBF operation, the diversity of the biota had increased to include filamentous bacteria, cyanobacteria, fungi ciliates and even individual crustaceans (isopods) and

[3] Note, in this paper the term "bacteria" includes both bacteria and archaea

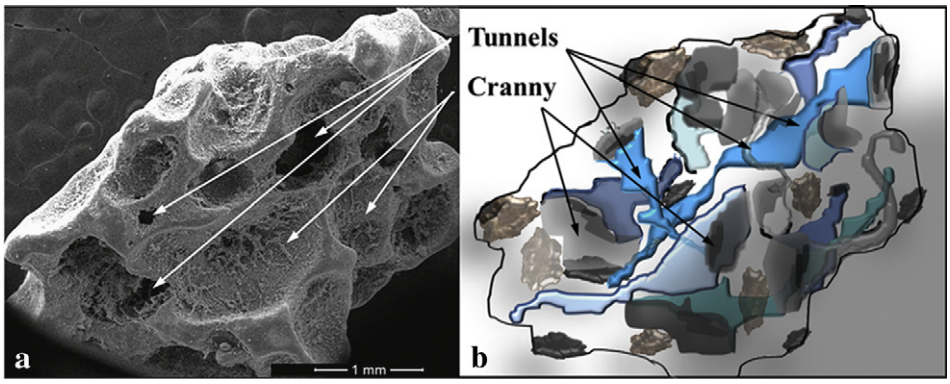

Fig. 2 – (a) Scanning Electron Microscopy (SEM) image of a representative *Tuff* grain showing the complex architectural surface (b). Corresponding 3D schematic diagram of the same *Tuff* grain illustrating the maze of tunnels (shades of blue) and crannies (shades of gray) that shelter the microbiota. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Sipuncula (Peanut worms), which entirely covered the *Tuff* grains throughout the BF (Fig. 3; Movie 1). This massive biological cover was then maintained throughout the entire year of this study (Fig. 3).

Supplementary video related to this article can be found at http://dx.doi.org/10.1016/j.watres.2013.03.013.

We assume that despite the high flow rate of seawater through the BF, most of the observed multicellular organisms mentioned above were able to subsist by "sheltering" within the numerous crannies and pores of the *Tuff* grains where they could filter particulate organic matter in the interstitial water and also graze on biofilm attached to the grain surfaces (Fig. 3).

During the entire 12 months of our study, only sporadic microbial colonization was observed on the sand or Filtralite® grains of the MBF column. The reason/s for the sparse microbial colonization on either of the MBF media is unclear.

Recently Bar-Zeev et al. (2012a) reported the dynamic formation of a mature microbial community in the filter bed of a standard operating RSF. A fairly extensive, complex microbial coverage developed after two months of operation within this

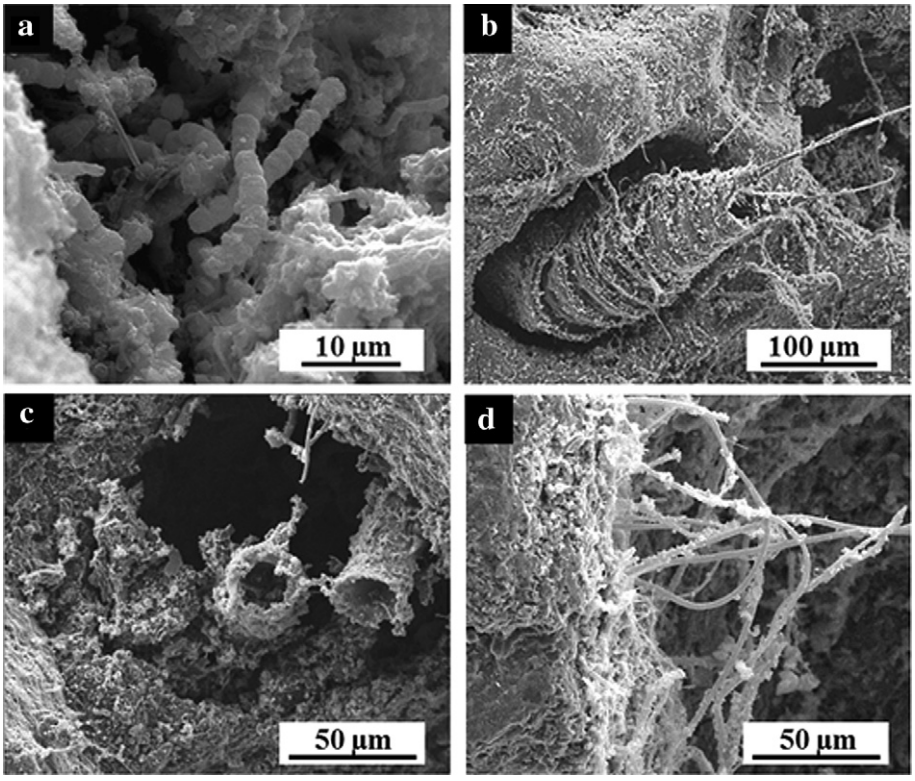

Fig. 3 – SEM micrographs of various microbiota on and within *Tuff* grains; (a) filamentous bacteria, (b) and (c) isopods and marine worms in *Tuff* tunnels, (d) filamentous cyanobacteria within *Tuff* pores.

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dual media (anthracite over sand) RSF indicating the potential for biofiltration. However, in contrast to the BF unit in our present study, no multicellular organisms were observed in the RSF filter media and microbial coverage was much sparser and less diverse.

3.3. *Formation of bio-aggregates and bioflocs*

Two to three weeks after the RBF began operation, concomitantly with bacterial colonization and biofilm formation; we observed the formation of numerous aggregates of bacteria and some algal cells embedded in an amorphous organic matrix on and within the *Tuff* grains of the BF (Fig. 4). The density of these bio-aggregates increased with time until a cleaning cycle was started. During flushing, some, but not all of this layer was removed and appeared as floc-like particles in the BF filtrate (see 3.4). When normal operation resumed, bacteria in the remaining biofilm layer multiplied rapidly forming new bioflocs, until the next cleaning cycle was activated within 5–7 days.

The development of these contiguous, biologically active bio-aggregates and biofilm layers followed recent descriptions of aquatic biofilm formation that highlighted the role of transparent exopolymer particles (TEP) and protobiofilm microgels in biofilm development on submerged surfaces (Bar-Zeev et al., 2012b). Previous studies have also shown that attached bacteria generate extracellular polymeric substances (EPS) which enhance further bacterial adhesion to surfaces (Stevik et al., 2004; Stoodley et al., 2002). The highly porous *Tuff* medium provides a large surface area for the initial adhesion of TEP microgels and bacteria that subsequently secrete EPS, stimulating the aggregation of organic colloids and microgels, bacteria and other microorganisms (Fig. 4). These attached layers of biological complexes act as "hot spots" of intense microbial metabolism that are sites of organic matter degradation (Azam and Malfatti, 2007; Liu and Tay, 2004).

Of immediate consequence to the present study was our observation that when the BF was cleaned by flushing (see 3.4) large numbers of floc-like particles, from ~15 μm to ~ 2 mm in size were observed in the BF filtrate during and immediately after cleaning cycles (Fig. 5, Movie 1). Microscopic examination of these flocs which stained with both Alcian Blue (polysaccharides) and SYTO^{R} 9 (nucleic acids and bacterial cells), showed that they consisted of smaller inorganic particles (silt and sediment) with numerous associated bacteria and algae enclosed within a polysaccharide gel-like matrix that held together the aggregate structure (Fig. 5). On average, about 18% of these flocs were organic, as indicated by weight loss after combustion at 450 °C. To emphasize the biogenic origin of these particles we have dubbed them, "*bioflocs*".

Note that these bioflocs were not observed in the BF filtrate during routine operation of the RBF, only during and for some time after a cleaning cycle (see 3.4). Also no bioflocs were seen at any time in the MBF filtrate. We suggest that the source of the bioflocs was material adsorbed to, or formed by, the biologically active layer growing on the *Tuff* grain surfaces. Under normal operation, the sheer forces acting on this layer were insufficient to cause much detachment. However, during flushing cycles, when water and air flows occurred simultaneously the entire BF filter bed was resuspended and mixed,

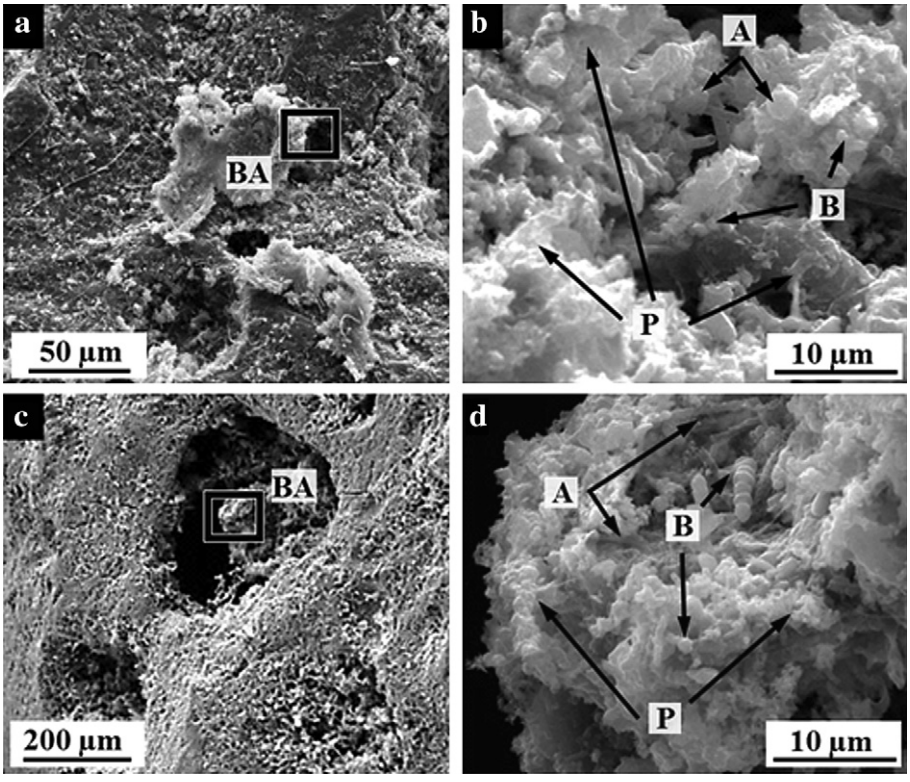

Fig. 4 – SEM micrographs showing attached bio-aggregates, [(a) and (c)]; Details of the attached bio-aggregates [(b) and (d)] showing bacteria (B) and algal cells (A) held together in an amorphous gel like matrix.

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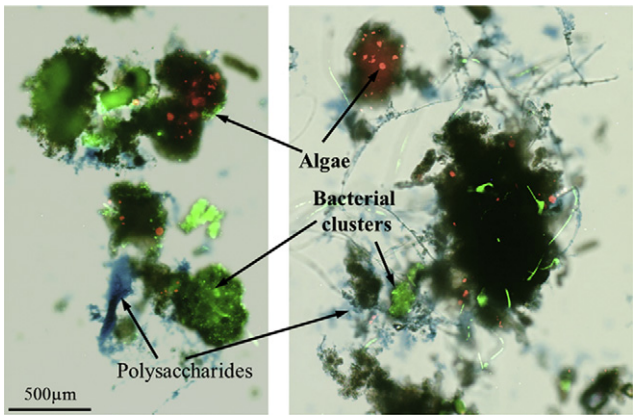

Fig. 5 – Bright-field and epifluorescence overlay images of bioflocs collected from the BF filtrate after flush cleaning procedure. Bacteria (green) were stained with SYTO^{R} 9 and TEP (blue) with Alcian Blue. Algae (red) were identified by Chl *a* auto-fluorescence. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

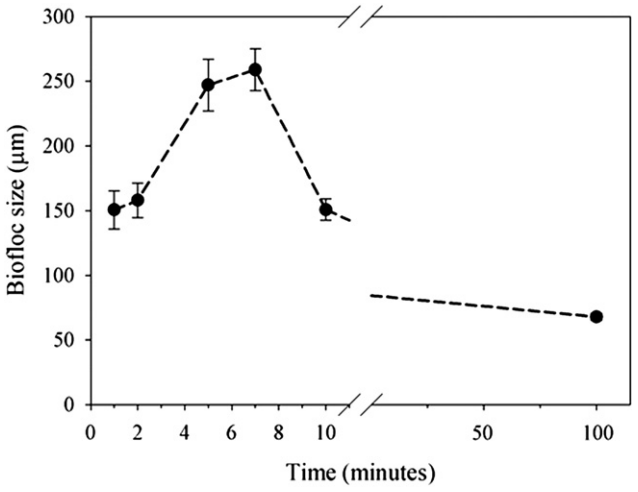

Fig. 6 – Time course of change in biofloc size during constant flush cleaning over 100 min (first experiment, see text for details).

then considerable amounts of material that had collected in the microbial layer were released as bioflocs into the BF filtrate.

Whatever the exact mechanism of their production, these biogenic flocs were large and stable enough to subsequently filter out by the MBF; as noted, no bioflocs were ever observed in the MBF filtrate. Therefore, we propose that the main function of the BF unit of the RBF was to act as a Bioflocculator, replacing the need for chemical coagulation that is currently required in standard SWRO pretreatment.

3.4. *Flush cleaning: impact on bioflocs and water quality parameters*

Conventional pretreatment procedures in SWRO rely mainly on RSF that mechanically removes suspended solids (>0.35 μm) formed upstream after the addition of chemicals in a coagulation and flocculation step (Bratby, 2006; Pearce, 2009; Prihasto et al., 2009). When the RSF is overloaded with particles as indicated by high differential pressure (ΔP) across the filter, a backwashing procedure is carried out to flush and clean the filter bed (Drami et al., 2011).

Two experiments were carried out in order to clarify details of the release of biogenic flocs from the *Tuff* material in the BF as a result of the flushing procedure (see 2.2.9). A short term experiment examined the levels of biofloc release over a period of 100 min from the start of the cleaning cycle. Immediately after flushing began (1–2 min) small size flocs were released into the BF filtrate (Fig. 6) and NTU increased from 0.6 to 85. The largest bioflocs (up to 1.5 mm) and highest turbidity (345 NTU) were observed in the BF filtrate, 5–7 min after the start of the flush. By 100 min, biofloc size had decreased significantly (Fig. 6); nevertheless these particles were completely retained by the second stage MBF.

In the second, long term experiment (19 h, see 2.2.8), prior to the flushing procedure, no bioflocs were apparent in the BF filtrate. The largest bioflocs were observed in the BF filtrate shortly (1–5 min) after the first flush cycle began (Table 1.). By 1 h and 4 h of continuous flushing, biofloc sizes had declined by about 50%. When, after a further 15 h of normal operation during which there were no bioflocs in the BF filtrate, a second cleaning cycle was activated, bioflocs appeared again in the BF filtrate (Table 1). As in the previous experiment, no bioflocs were detected in the RBF effluent at any time.

In order to follow the impact of the cleaning cycles on some key parameters of water quality, we measured changes in turbidity, total particle count, Chl *a* and TEP at various depths in the BF column and in the BF filtrate during the course of the second experiment (Fig. 7). Prior to the start of the flushing procedure, all four parameters were at their lowest measured levels at all sampling points. The most drastic (50–80%) removal of turbidity, total particle count and Chl *a* occurred in the first 200 cm of *Tuff* medium. This significant reduction was probably due to the adhesion of relatively large, organic particles (such as algae and microgels) forming bio-aggregates on the surfaces of the *Tuff* grains (Fig.7). After 1 h of continuous flushing, maximum levels were observed for all parameters, with increases relative to the feedwater of 28-fold for Turbidity, 4.3-fold for Chl *a*, 4.5-fold for Particle counts and 1.3-fold for TEP in the BF filtrate. These results show that the flushing procedure was releasing considerable amounts of material from the BF filter medium into the BF effluent. With time during the flush procedure, less material was released from the BF medium; by 4 h, Turbidity in the filtrate was 10-fold relative to the intake, Chl *a* 1.5-fold, Particle count 3.5-fold and TEP 0.9-fold.

After 4 h the air–water flush was stopped and the RBF commenced normal operation. When sampled again after 15 h of normal operation, turbidity, particle counts and Chl *a* showed removal levels (88, 76 and 55% removal, respectively) very similar to those measured before at the beginning of the experiment, i.e. before the first flush cycle (Fig.7). TEP concentrations in most of the BF column remained high, but nevertheless decreased to 70% of feedwater levels in the BF filtrate (Fig.7).

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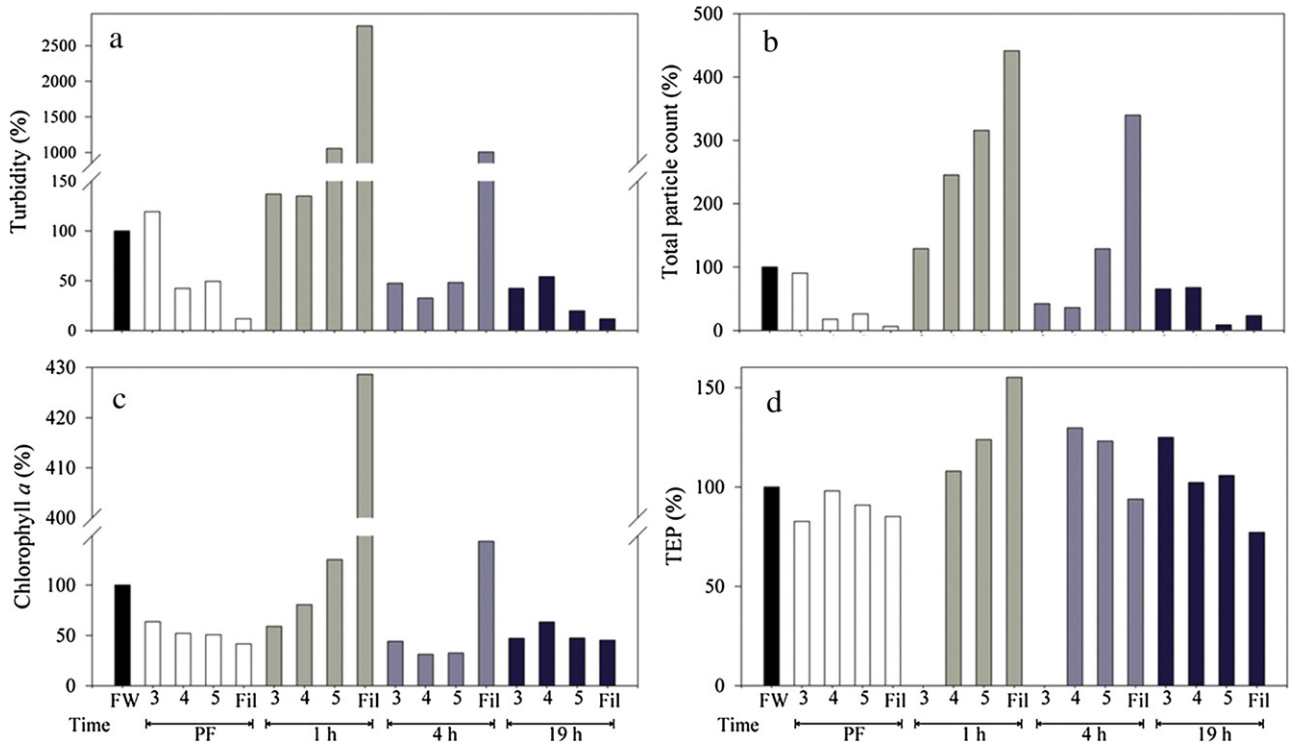

Fig. 7 – Changes in water quality parameters in the BF column and filtrate (Fil.) during the second flushing experiment; (a) Turbidity, (b)total particle count, (c) Chl *a* and (d), TEP. All parameter values given as percentages normalized to feedwater (FW) taken as 100%. Average concentrations in the feedwater were as follows: turbidity 1.9 ± 0.1 (NTU), total particles $1.2 \times 10^7 \pm 2.2 \times 10^5$ (L^{-1}), Chl *a* 0.228 ± 0.004 ($\mu g\ L^{-1}$) and TEP 622 ± 98 (μg GX L^{-1}).

These results show that during normal operation considerable amounts of particulate organic material resulting from microbial processes accumulate within the *Tuff* medium. When a flush cleaning cycle is activated, some of this accumulated material is released as biofloc into the BF effluent. Although the size of the biofloc diminishes over the course of the flush cycle, nevertheless these particles remain large enough to be effectively retained by the MBF and do not appear in the RBF filtrate.

3.5. *Comparison of RBF and RSF filtration efficiencies*

The seawater feed for the Hadera SWRO Plant that was used for both the RBF and RSF is located at ~3 m depth, ~1 km offshore. The average concentrations of Chl *a*, TEP, DO and turbidity measured in the feedwater during this study are shown in Table 2 (for more details see Bar-Zeev et al., 2012a). The filtration efficiency (Δ_T) for each of these water quality parameters was determined as the percentage difference measured between their levels in the seawater feed (taken as 100%) and in BF, MBF, RBF or RSF effluents respectively. Also presented in Table 2 are the average SDI values measured in both RBF and RSF.

3.5.1. *Chlorophyll a (Chl a)*

Chl *a* is becoming recognized belatedly as an important water quality parameter for the SWRO desalination industry. Coastal algal blooms have been shown to cause problems of clogging and may release large amounts of microgels such as TEP and other organic foulants. There is also increasing concern that some species of harmful algal blooms may release toxins into intake water.

Chl *a* concentrations were low in the feedwater, ranging from 0.1 to 0.3 $\mu g\ L^{-1}$ during this study. On average, Chl *a* in the BF filtrate was reduced by 63% in comparison to feedwater while the MBF reduced Chl *a* by a further 19%. Overall, the RBF and RSF were equally effective in removing Chl *a* from the seawater feed (Table 2).

Detailed examination of the BF column profile showed that most of the algal cells from the seawater feed were retained within the first meter of the column forming large aggregates adhering to *Tuff* granules (Fig. 4). Confirmation of this was shown in samples taken further up the BF column, (sampling ports 4 and 5 in Fig. 1) in which Chl *a* concentrations in the interstitial water were ~40–45% of those measured at sampling ports 2 and 3.

3.5.2. *Transparent exopolymer particles (TEP)*

TEP are nano- and microgels that are in a state of constant dynamic change (Verdugo et al., 2004) and therefore are extremely difficult to remove effectively from seawater. To date there is no published data on any biofiltration technology for removing TEP and there are only a few reports on removal of TEP from feedwater by membrane or other filtration (Villacorte et al., 2009; Berman, 2012; Eshel et al., 2012; Komlenic et al., 2012).

Large variations in TEP concentrations (from 153 to 492 μg GX L^{-1}) were observed in the feedwater over the entire

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Table 2 – Comparison of Filtration Efficiencies of a pilot Rapid Bioflocculation Filter (*without any addition of chemical coagulants*) and an operating, large-scale Rapid Sand Filter (*with $Fe_3[SO4]_2$ added upstream*).[a]

	Months sampled (n)	Feedwater avg. conc.	BF Δ_T (%)	MBF Δ_T (%) contribution	RBF Δ_T (%)	RSF Δ_T (%)
Chl *a* ($\mu g\ L^{-1}$)	8	0.18 ± 0.06	63 ± 13	19 ± 14	75 ± 9	79 ± 8
TEP ($\mu g\ GX\ L^{-1}$)	9	350 ± 137	14 ± 29	9 ± 10	26 ± 25	17 ± 28
Turbidity (NTU)	8	1.2 ± 0.7	53 ± 20	13 ± 7	65 ± 14	61 ± 25
DO ($mg\ L^{-1}$)	4	9 ± 1.3	3 ± 2	23 ± 11	26 ± 14	ND
			BF	MBF filtrate	RBF filtrate	RSF filtrate
SDI (a.u.)	8	>5	4.7 ± 0.3	1.15 ± 0.5	3.5 ± 0.4	3.5 ± 0.3

a Table shows the average filtration efficiencies (Δ_T) as % change in Chl *a*, TEP, turbidity and DO (normalized to untreated seawater feed concentrations taken as 100%) measured in the BF, RBF and RSF filtrates. The additional contribution of the MBF unit of the RBF to % filtration efficiency is also shown. SDI values in these filtrates and average concentrations of Chl *a*, TEP, DO and turbidity in the seawater feed are also given. Data were collected over a 12-month period.

sampling period, with maxima in late summer and early spring. Both the RBF and the RSF showed highly variable efficiency in removing TEP from the feedwater. The removal efficiency for TEP ranged from 0 to 70% and from 0 to 65% for the RBF and MBF, respectively. The average reduction of TEP by the BF unit was 14%; the MBF gave a further 9% reduction. In total, the RBF reduced TEP by an average 26% compared to 17% by the standard RSF (Table 2). Thus, the overall, yearly average TEP removal by the RBF was comparable and even slightly better than that of the standard RSF.

During the last three summer months of RBF operation, when the coastal water was severely impacted by swarms of *Scyphomedusae* jellyfish that release large quantities of slimy substances, the average TEP removal was only 14%, but at the same time an average 4% increase of TEP was measured in the RSF effluent. At present, we have no clear explanation why TEP filtration efficiencies were so low during this period.

On the basis of our present data, we cannot evaluate the extent of direct removal, possible degradation, grazing, or for that matter, regeneration of TEP within the filter media; our results show only the net outcome of all those processes. Although, removal of some planktonic TEP from inflowing feedwater occurs by continuous adhesion to large surface areas of developing biofilm within the *Tuff* grains (Fig. 4). Some of the polysaccharides and other organic TEP constituents were probably removed by microbial degradation as well as by microplankton grazing on the biofilm communities (Fig. 4; Mov. 1). In contrast, release of TEP could occur when portions of the EPS matrix detached from maturing biofilm on the *Tuff* grains and passed into the BF filtrate. The scouring of EPS from the filter bed medium which then appeared as TEP in the effluent was clearly seen as a 155% increase in TEP concentration in the BF filtrate after 1 h of flushing (Fig. 7).

3.5.3. *Turbidity*

In respect to Turbidity, no significant differences in filtration efficiency were measured between the RBF and the operational standard RSF (Table 2). The greatest reduction in turbidity was observed in the BF filtrate (53%), while the MBF contributed a further (13%).

3.5.4. *Dissolved oxygen (DO)*

Generally, biodegradation of organic matter is accelerated under aerobic conditions relative to anaerobic conditions (Erses et al., 2008). To ensure oxidation of the *Tuff* filter bed, the flow of feedwater was upward from the base of the BF column (Fig. 1). The upward water flow resulted in a loose packing and constant motion of the *Tuff* medium within the column. Consequently, we observed no significant reduction in dissolved oxygen (DO) concentrations within the BF column (Table 2). In contrast, there was a slight reduction (26%) of DO in the MBF which occurred beneath a cake layer, comprised of tightly packed organic matter. This layer always appeared at the boundary between the Filtralite® and sand media of the MBF, at the depth of 80 cm.

3.5.5. *Silt density index (SDI)*

The silt density index (SDI) is the major water quality parameter used in the desalination industry. During most of the study, feedwater SDI levels exceeded the instrument's accurate measuring maximum (>5 a.u.). Therefore, in Table 2 we have only given the actual SDI numbers as measured in the filtrates of the BF and MBF components of the RBF and in the RSF filtrate. As previously, both RBF and RSF gave comparable results. Importantly, the SDI levels in the filtrates of both filters were constantly below the operational range (<4) required by RO membrane manufacturers.

4. Conclusions

This study has demonstrated the potential of biologically based pretreatment for SWRO desalination. Our results, based on a year-long study, show comparable performance by a large pilot, two-stage, granular Rapid Bioflocculation Filter consisting of a Bioflocculator unit with volcanic *Tuff* medium followed by a Mixed Bed Filter with no prior chemical additives and a standard RSF operating with addition of chemical flocculant [$Fe_2(SO_4)_3$] upstream at the Hadera SWRO facility (Table 2).

Much of the effective performance of the RBF is due to the bioflocculation process which occurs within the *Tuff* medium. Some biodegradation may also take place but, because of the rapid flow rates through the RBF, this is unlikely to be a major factor in the filtration process. We have shown that during normal operation there is continuous microbial growth and development of a biofilm layer of organisms within an organic matrix that effectively retains different types of colloidal and particulate matter. When shear forces increase during flush cleaning cycles, some of this bio-aggregated material is released into the BF filtrate as bioflocs. These bioflocs are large

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enough to be mechanically retained with high efficiency by the MBF. The bioflocculation process that occurs in the first stage of the RBF depends on the metabolism of an extensive, biological food web comprising of diverse populations of bacteria, archaea, cyanobacteria, protozoa, and even crustaceans and marine worms. It is noteworthy that this kind of microbial environment only developed on the highly porous *Tuff* grains of the BF and not on the MBF media.

In conclusion, we have demonstrated that with suitable filter bed media and design modification, it is possible to construct a rapid granular filter that achieves effective large-scale pretreatment filtration equivalent to that of currently operating RSF but without the need for prior chemical coagulation. We suggest that our approach of using bioflocculation without chemical additives has considerable potential as an alternative to conventional RSF pretreatment for large SWRO facilities.

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