Nanofiltration and reverse osmosis biostability relative to alternative methods of water treatment

Suibing Liu, Michael LePuil, J. S. Taylor and A. A. Randall

ABSTRACT

Biostability of finished waters was assessed statistically using assimilable organic carbon, biodegradable dissolved organic carbon and heterotrophic plate counts. Biofilm growth on unlined cast iron, galvanized steel, lined cast iron and polyvinylchloride pipe material was assessed visually and statistically using potential exoproteolytic activity, which is a measure of biofilm cell density. Seven different treatment processes were used to simulate full scale treatment and distribution of ground, surface and saline sources to pilot distribution systems made from unlined cast iron, galvanized, lined cast iron and polyvinylchloride pipes taken from actual distribution systems. Bulk water biostability parameters as measured by assimilable organic carbon, biodegradable dissolved organic carbon and heterotrophic plate counts were lower in reverse osmosis finished water and higher in conventionally treated groundwater. Average finished water assimilable organic carbon indicated reverse osmosis and nanofiltration membrane processes reduced assimilable organic carbon relative to finished groundwater produced by conventional treatment or softening, and finished surface water produced by enhanced coagulation. This relationship was not observed clearly for biodegradable dissolved organic carbon or heterotrophic plate counts. Biofilm growth on coupons cut from the pipes used to build the pilot distribution systems typically decreased as the level of treatment increased with the exception of reverse osmosis finished water, which produced very high biofilm growth. However, the assessment of biostability indicated biostability generally increased as the level of treatment increased, and the general order of biostability of process finished waters was: membrane > precipitative > conventional; and the order of biofilm growth with respect to pipe material was unlined cast iron > galvanized > lined cast iron > polyvinylchloride. Hence, improved distribution system biological water quality, as measured by lower assimilable organic carbon, biodegradable dissolved organic carbon, heterotrophic plate counts and biofilm growth, was directly dependent on nonpurgeable organic carbon and improved as finished water nonpurgeable organic carbon decreased.

Key words | AOC, biostability, distribution, material, membrane, treatment

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INTRODUCTION

This work describes an assessment of biostability of finished waters produced from surface, ground and saline sources in distribution systems made from varying materials taken from actual distribution systems. Hence, it is a systems' assessment of biostability by source and pipe material in distribution systems. Assimilable organic carbon (AOC), biodegradable dissolved organic carbon (BDOC) and heterotrophic plate counts (HPC) were used to assess biostability in finished water and potential exoproteolytic activity (PEPA) was used to assess biofilm cell density on

doi: 10.2166/aqua.2007.049

different pipe materials. Finished water quality varied by source and associated (required) treatment processes. Finished waters produced from saline, surface and ground sources resulted in relatively high chlorides–low alkalinity, high sulfates–low alkalinity and high calcium–high alkalinity, respectively. Seven different treatment systems were used to produce seven different finished waters that were distributed to eighteen different pilot distribution systems for the entire project. The pilot distribution systems were made of polyvinylchloride, galvanized steel, lined cast iron and unlined cast iron pipes taken from existing distribution systems. Seven of those pilot distribution systems were identical, receiving unblended finished waters from the seven different treatment systems, respectively. Biological stability of those seven treatment systems and the associated seven pilot distribution systems are assessed in this work relative to treatment system and distribution system pipe material. The other eleven pilot distribution systems were used for project work, and are not described in this paper.

LITERATURE

Potable water in distribution systems contains extremely diversified microbial flora (Block *et al.* 1997) and complex organic matter (Croué *et al.* 2000). A relatively significant fraction of the organic matter is biodegradable (Block *et al.* 1992). Even when the organic fraction is considered to be small in so-called oligotrophic groundwaters (< 0.5 mg dissolved organic carbon (DOC)/L) or nanofiltered waters (< 0.2 mg DOC/L), bacterial growth is observed (Crissot-Laruade *et al.* 1999). Moreover, in most potable water distribution systems, the water–material interface is a favored site for cells and organic matter accumulation and for multiplication of bacteria (Block *et al.* 1993). This proliferation may be followed by the bacterial detachment (Ascon-Cabrera *et al.* 1995) or by displacement from the pipe surface (Rittman 1989) and transport to the bulk liquid (Taylor *et al.* 2005).

HPC was developed in 1881 by Robert Koch, which was the first tool for monitoring the microbial quality of drinking water (Frankland *et al.* 1894). HPC measurements can play an important role in validation and verification of treatment plant procedures (Yeh *et al.* 1998; Gauthier *et al.* 1999; Zhang *et al.* 2002; Camper *et al.* 2003; WHO 2003). Increased HPC in the distribution systems usually indicated bacterial regrowth. Bacterial regrowth refers to the proliferation of viable organisms, including the recovery and growth of organisms that were previously injured during the water treatment process (Escobar *et al.* 2001). HPC is not perfect and had the bias of selective media as only the bacteria culturable on that media are quantified. However, HPC on R2A agar is the most universally recognized method of enumerating bulk water heterotrophs in the drinking water industry and is even incorporated into regulatory requirements for meeting residual requirements. Molecular methods also have different biases. For example, DNA-based techniques rely on DNA extraction and thus only the microbial population with easily extracted DNA is detected or (for quantitative techniques) quantified. HPC does not determine the origin of bacteria, and it is true that it generally underestimates bacteria that are aggregated into biofilm or floc. However, it is reasonable to assume that the percent estimation of sloughed biofilm for a given system is reasonably constant and that HPC will be proportional to the amount of material sloughed off for a given disinfectant residual.

AOC and BDOC are used to determine the regrowth potential of treated water. AOC is the portion of the organic carbon that can be synthesized to cellular material by two bacterial strains (*Pseudomonas fluorescens strain P-17* and *Spirillum strain NOX*, as per *Standard Methods* 9217B (APHA, AWWA & WEF 1999)). The AOC assay can be used to determine the effect of treatment on potential bioactivity (van der Kooij & Hijnen 1984; Yeh *et al.* 1998; Charnock & Kjonno 2000; van der Kooij 2000; Sharp *et al.* 2001; Liu *et al.* 2002; Zhang *et al.* 2002; Li & Chu 2003; Jegatheesan *et al.* 2004; Hong *et al.* 2005; Polanska *et al.* 2005). The BDOC method was first introduced by Servais *et al.* (1987). BDOC is the fraction of DOC that can be metabolized by bacteria within a period of time. Pure strains are not used for the BDOC test. The BDOC assay is run using the fixed biofilm method which utilizes a heterogeneous biofilm cultivated using water similar to the waters to be tested with respect to alkalinity and dissolved organic carbon levels. The BDOC assay is run until a minimum DOC level is observed, which is only possible by incubating the sample until DOC begins to elevate after the minimum value is obtained. The time of

incubation depends on the kinetics of the biodegradable DOC in the sample, and for this study the minimum was typically observed between 3 and 5 days, with incubations taking just under one week on average. BDOC was first used for raw water quality tests and biological activated carbon systems, but it became a widely used parameter in the field of treatment processes and the assessment of microbial proliferation in distribution systems (Escobar & Randall 1999; Sharp *et al.* 2001; Volk & LeChevallier 2002; Jegatheesan *et al.* 2004). BDOC is higher than AOC for the same type of water, because the BDOC test uses a relatively high concentration of biomass of an adapted microbial community, whereas low numbers of two pure cultures are used in the AOC test. The AOC test is considered an index which is related to the increase in culturable bacteria in distribution systems while BDOC attempts to quantify as much of the biodegradable organic material (BOM) as possible (Prévost *et al.* 2005). The AOC test results in growth proportional to the most readily degradable fraction of the BOM, while BDOC also includes less readily degradable BOM fractions.

Technical limitations make it difficult to obtain a precise idea of the diversity and activity of the biofilm (Byrd *et al.* 1991). In previous studies, detachment of the biofilm followed by enumeration using HPC was used to quantify the biofilm (Sharp *et al.* 2001; Okabe *et al.* 2002; Camper *et al.* 2003). A non-destructive technique that quantifies the intact biofilm is called the Protein ExoProteolytic Activity (PEPA) assay (Laurent & Servais 1995) which directly determines the fixed biomass in distribution systems by relating hydrolytic activity to quantity of biomass (Batte *et al.* 2003; Servais *et al.* 2004; Taylor *et al.* 2005). The assay was shown to correlate well with HPC counts from detached drinking water biofilm and also to be sensitive enough to detect low quantities of biofilm in the presence of disinfectant residual (Butterfield *et al.* 2002). While the HPC test can be adapted to biofilm quantification by scraping and homogenizing the biofilm at very high torque, biofilm cell density is measured using the PEPA assay, which allowed measurement of cell density in an intact biofilm.

The biofilm structure also depends on the nature of the pipe materials used for potable water distribution systems (Camper *et al.* 2003; Taylor *et al.* 2005). Biofilms form on the interior surfaces of all pipe materials in distribution systems and eventually become heavily colonized (10^6 cells/cm^2) by microorganisms (Niquette *et al.* 2000). However, the type of pipe material in the distribution system significantly affects the structure and specific microorganism in the biofilm (Kerr *et al.* 1999). In a laboratory study using chlorine or monochloramine with or without humic substances, and in the field test of an existing system at different locations, Camper *et al.* (2003) found that iron pipe had the highest bacterial biofilm cell densities. The hierarchy of biofilm cell densities by material was polyvinylchloride < lined cast iron < unlined cast iron and galvanized steel. Pipe material determines the adsorption efficiency of the initial biofilms and can be a source of nutrients or growth factors. Morton *et al.* (2005) found that unlined cast iron leached nutrients such as phosphorus, and to a lesser extent nitrogen and carbon, which resulted in high biofilm cell densities. Finally, in the case of materials susceptible to corrosion, it was clearly demonstrated that the presence of iron corrosion products enhances the activity and the production of heterotrophic biomass (Appenzeller *et al.* 2001).

The effects of treatment processes on biostability were investigated. Sharp *et al.* (2001) found that coagulation prior to filtration had little effect on biostability, granular active carbon (GAC) and anthracite filter media produced essentially biostable water with low regrowth potential, and dissolved air flotation followed by either GAC or anthracite filtration produced the most biologically stable water. Escobar & Randall (1999) observed that nanofiltration decreased the BDOC concentration of the treated water, while nanofiltration did not reject the major fraction of AOC. Hong *et al.* (2005) indicated that a reverse osmosis system did not remove the majority of AOC.

Okabe *et al.* (2002) revealed that biological treatments significantly improved the biostability of finished drinking water. A rotating biofilm membrane reactor and biological activated carbon filter removed 50% more AOC than conventional multi-media filtration and ultrafiltration.

This work considered the effects of different treatment processes and pipe materials on biofilm cell density using identical materials and operating conditions. No previous study assessed biostability by comparing AOC, BDOC and HPCs for finished waters produced using membrane and commonly used non-membrane processes. Nor did any previous study assess biofilm cell density using PEPA on

different pipe materials commonly used in drinking water distribution systems in a controlled field study using identical pilot distribution systems. Studies addressing the effect of treatment processes on biostability were often confined to bulk liquid parameters (Escobar & Randall 1999; Yeh *et al.* 2000; Polanska *et al.* 2005), or represented information generated in lab scale annular reactors using pristine pipe material coupons (Camper *et al.* 1996, 2003), or did not include aged (corroded) pipe material as it is found *in situ* in distribution systems (Zacheus *et al.* 2000; Camper *et al.* 2003; Morton *et al.* 2005). In addition this study included ground, surface and reverse osmosis finished and blends of these finished waters. It should be noted that the word "blend" here refers to the blending of the three ground, surface and reverse osmosis finished waters as source water, which was used to produce one nanofiltration finished water and one softening finished water. The seven finished waters were not blended in the seven distribution systems in this study. Previous studies of blended water on biostability were extremely limited and the few references that may be found usually pertain to taste and odor concerns rather than regrowth potential directly (Fabrellas *et al.* 2004).

METHODS AND MATERIALS

Methods and materials include the construction of the pilot treatment facility and the pilot distribution systems as well as the chemical and biological methods used for assessment of source biostability and biofilm cell density.

Pilot facilities

The Tampa Bay Water (TBW) field research facility was designed and built by the University of Central Florida (UCF) personnel with significant assistance from TBW and member government personnel. These facilities were located at the TBW Cypress Creek Wellfield. The pilot facilities consisted of seven different treatment systems and eighteen pilot distribution systems. The data for this article was generated from discharging finished waters from the seven different treatment systems to seven identical pilot distribution systems. The covered facilities for producing finished waters and the pilot distribution systems are shown in Figure 1 and Figure 2, respectively. As shown in Figure 2, the coupons were cut from actual distribution system pipes, and inserted in polyvinylchloride cradles that succeeded the pilot distribution systems. The cradles as shown in Figure 2 (left) were used for the biofilm cell density study. These cradles operated as a closed system and provided an aqueous environment for biofilm growth on coupons as shown in Figure 2 (right). These coupons were obtained from the same pipes that were used to build the pilot distribution systems. These treatment systems and pilot distribution systems are detailed in other publications (Taylor *et al.* 2005; Liu *et al.* 2005). All finished waters were stabilized with respect to calcium carbonate,

Figure 1 | Pilot treatment facilities and distribution systems.

Figure 2 | Cradles of pilot plant (left) and coupons in the cradles (right).

disinfected to meet USEPA criteria, and utilized chloramines for residual maintenance.

Treatment systems

The seven treatment systems are described in the following text and are separated into groups of membrane treatment systems and non-membrane treatment systems. Post-treatment for the seven treatment systems included stabilization and disinfection. Stabilization was done by aeration, which stabilized finished waters relative to $CaCO_3$. Six of the seven treatment systems consisted of disinfection using free chlorine to maintain a 4 mg/L residual after a 5 min contact time to meet *CT* (chlorine concentration times time) requirement, and then ammonia was added to produce 4 mg/L combined chlorine residual. This method of post-treatment is described as common chloramination. Post-treatment in the enhanced coagulation system consisted of ozonation, aeration and chloramination to produce 4 mg/L chlorine residual. These treatment systems simulated the member government treatment systems as closely as possible.

- Reverse Osmosis finished water was produced by conventional Osmonics reverse osmosis membrane filtration of groundwater and augmentation of the permeate with sea salt to match the finished water quality of the TBW Regional Desalination Facility, which used the Gulf of Mexico as a source. Common chloramination was used for post-treatment.
- Integrated membrane system (IMS) finished water was produced by a conventional integrated membrane system in that surface water was pretreated by ferric sulfate coagulation–sedimentation–filtration (CSF), scaling control and cartridge filtration, nanofiltered using an Osmonics membrane and post-treated by common chloramination.
- Blended nanofiltered finished water was produced by nanofiltering a blend of 62% conventionally treated ground water, 27% enhanced coagulated surface water and 11% reverse osmosis treated groundwater. The same nanofilter as used in the integrated membrane system was used in the nanofiltration treatment of blend water. This treatment system was piloted because some member governments were interested in the additional treatment of all waters received from TBW, which supplied member governments with blended water that was produced from saline, surface and groundwater sources. The rationalization for additional treatment of essentially finished waters may not be apparent until the resistance to surface water utilization by utilities that had used only groundwaters for decades is considered. Common chloramination was used for post-treatment.

The four remaining treatment systems did not utilize membranes and are described as:

- Enhanced coagulation of surface water was accomplished by ferric sulfate coagulation–sedimentation–filtration, ozonation, biologically activated carbon (BAC) filtration and cartridge filtration to simulate mixed media filtration, which represented the most

advanced coagulation treatment system commonly used for surface water treatment and closely simulated the TBW Regional Surface Water Treatment Facility.
- Conventionally treated finished groundwater was produced in the sequence of chlorination, chloramine formation, aeration and storage prior to distribution. Common chloramination was used for post-treatment.
- Softened finished water was produced by a conventional lime softening process, which consisted of $Ca(OH)_2$ addition, sedimentation, filtration and aeration. Common chloramination was used for post-treatment.
- Blended softened finished water was produced by softening a blend of 62% conventionally treated ground water, 27% enhanced coagulated surface water and 11% reverse osmosis treated groundwater. This treatment system also was considered by a member government that wished to evaluate further treatment of treated surface water. Common chloramination was used for post treatment.

Pilot distribution systems

Each pilot distribution system was made from polyvinylchloride, lined cast iron, unlined cast iron and galvanized steel pipes that were excavated from actual distribution systems for use in this project. A picture of the pilot distribution systems is shown in Figure 1. Flow in and out of each pilot distribution system was controlled by entrance and exit standpipes and valves. Each standpipe maintained a constant head through the pilot distribution systems and ensured the flow through the standpipes. Cradles as shown in Figure 2 housed the coupons for assessment of biofilm cell density. Flow to the cradles was controlled in a like manner as to the pilot distribution systems. The pilot distribution systems and cradles were constructed as paired systems, such that each pilot distribution system – cradle pair was operated as a closed system, were always wet and received identical influent water quality. Each cradle housed polyvinylchloride, lined cast iron, unlined cast iron and galvanized steel coupons. The overall hydraulic residence time (HRT) in each pilot distribution system was 5 d. All pilot distribution system pipe diameters were 150 mm except for galvanized steel, which was 50 mm. The pilot distribution systems were approximately 30.5 m long and the retention times in each section of the hybrid pilot distribution systems were equal. The pilot distribution systems were flushed weekly with five pilot distribution system volumes of finished water at a velocity of 0.3 m/s to simulate actual operation of a full scale distribution system (Liu *et al.* 2005).

Bulk and biofilm parameters

General water quality

The general water quality parameters are listed in Table 1, and include alkalinity, ammonia-N, total chlorine, total and calcium hardness, nonpurgeable dissolved organic carbon (NPDOC), dissolved oxygen (DO), pH, total dissolved solid (TDS), turbidity and temperature. The parameters are utilized to demonstrate differences in finished water quality by source and associated treatment system.

PEPA

The Potential ExoProteolytic Activity method was used to measure the cell density of biofilms. This method measures the global activity of the biofilm by estimating the potential of bacteria to hydrolyze proteins via a protein non-fluorescent artificial substrate (here L-Leucine β-Naphtylamide, LLβN). The enzymatic hydrolysis of this substrate leads to a fluorescent product (here β-Naphtylamine, βN), which can be detected by spectrofluorimetry. Fluorescence was plotted as a function of time and the rate of degradation gave an estimate of biological activity in the sample (Laurent & Servais 1995). The same four pipe materials (polyvinylchloride, lined cast iron, unlined cast iron, and galvanized steel) were used for PEPA testing as were used in the pilot distribution system. The pilot distribution systems and the cradles housing the coupons used for measurement of the biofilm cell density received identical finished waters. Coupons were put into the cradles at the start of each phase and removed for measurement of biofilm cell density at the end of each phase.

AOC

Assimilable organic carbon (AOC) is DOC that can be easily assimilated by bacteria and converted to cell mass (i.e. the most labile portion of the BOM (Prévost *et al.* 2005)). AOC concentration corresponds directly to bacterial

Table 1 | General water quality methods for distribution systems, treatment process raw and finished water analysis

Parameter	Method	Reference
Alkalinity	Titration	SM 2320 B
Ammonia-N	Ion Select Electrode Probe	SM 4500-NH_3 D
Chlorine, total	DPD colorimetric	Hach 8167
Chlorides	Ion Chromatography	SM 4500-CLF
Hardness (total, calcium)	EDTA Titration	SM 2340 C
Nonpurgeable dissolved organic carbon (NPDOC)	Persulfate/UV Oxidation	SM 5310 C
Oxygen, dissolved (DO)	Membrane probe	SM 4500-O G
pH	Electrometric	SM 4500-H^+B
Solids, Total Dissolved	Gravimetric (also used ion summation)	SM 2540
Sulfates	Ion Chromatography	SM 4500-SO_4B
Temperature	Direct reading	SM 4500-H^+B
Turbidity	Nephelometric	SM 2130 B

SM = Standard Methods of Water and Wastewater Analysis, 20th ed (1999), Hach = DR4000 Spectrometer Method Handbook, DPD = N,N-Diethyl-P-Phenylenediamine, EDTA = Ethyienediaminetetraacetic Acid.

regrowth potential. The AOC method was first published in Europe (van der Kooij *et al.* 1982). A modified version of van der Kooij's method was published by LeChevallier *et al.* (1993). *Pseudomonas fluorescens strain P17* and *Spirillum strain NOX* indigenous in drinking water were the measuring strains. It was assumed that cell numbers in the stationary phase for the two bacteria were linearly correlated to AOC concentration in water. AOC concentration can be calculated by comparing the cell number by incubating a biodegradable organic compound (sodium acetate) at a standard concentration versus the cell number obtained by incubating *P17* and *NOX* in water samples (*Standard Methods* 9217B (APHA, AWWA & WEF 1999)).

BDOC

Biodegradable organic carbon was determined by bacteria attached to sand that was provided from an unchlorinated sand filter in a full scale water treatment plant in Quebec, Canada. Three hundred mL water samples were inoculated with 100 g of biologically active sand from a biological filter and incubated at 20°C under 4 L/h aeration. Sample fractions were collected daily and filtered through a 0.45 μm membrane. DOC was measured until minimum DOC values were reached. The BDOC concentrations were derived from the difference between the initial and minimum DOC levels (Volk & LeChevallier 2002).

HPC

HPC was performed by spread plating on R2A agar and incubating at 25°C for seven days as outlined in *Standard Method* 9215C (APHA, AWWA & WEF 1999). In this paper, only bulk water HPC values were used.

Data analysis and comparison

Data for this paper were generated during a nine-month period during which pilot distribution system influent water quality was changed every three months, which were

described as phases. Phase I was from 12/06/2001 to 3/14/2002, phase II from 3/15/2002 to 6/13/2002, and phase III from 6/14/2002 to 9/12/2002. The average and standard deviation (SD) of the general finished water quality of the seven treatment systems are listed in Table 2. Influent and effluent AOC, BDOC and HPC were measured at the start and end of each phase. PEPAs were measured at the end of each phase. The general finished water quality data were taken at the influent of the pilot distribution systems weekly. Data analyses consisted of visual bar charts and limited development of linear regression models. Paired *t*-tests were used for statistical analysis and were conducted using Microsoft Excel and Sigma Plot regression software.

RESULTS AND DISCUSSIONS

The project scheme allows the assessment of different treatment processes' biostability, which is timely information for the water community. As previously noted, process biostability was assessed by comparing finished water AOC, BDOC and HPC, which were bulk water measures of biostability. PEPA was used to assess biofilm cell density as a function of both finished water and pipe material.

General finished water quality

The general finished water quality produced by the seven different water treatment systems is shown in Table 2. Finished water pH was relatively constant for these waters, but did increase as calcium and alkalinity decreased due to stabilization with respect to $CaCO_3$. Hence pH of the conventionally treated finished water was 7.9 and slightly lower than other finished water pHs because of stabilization at a slightly higher calcium and alkalinity. However, the concentrations of NPDOC, chlorides, sulfates, calcium, alkalinity and TDS constituted the primary water quality differences among these finished waters. The NPDOC, calcium and alkalinity were greater for the conventionally treated finished water than for all others, which was not surprising for a conventionally treated Florida groundwater.

There were few differences between the finished water quality for the softened groundwater and blended softened finished waters except that alkalinity was lower and TDS

Table 2 | Water quality averages/standard deviations for the water pilot treatment system

System	T Cl_2 mg/L Cl_2	pH	Alkalinity mg/L $CaCO_3$	Ca^{2+} mg/L $CaCO_3$	NPDOC mg/L	SO_4^{2-} mg/L	Cl^- mg/L	Turbidity NTU	D.O. mg/L	TDS mg/L	Temp. °C
Conv-GW	4.7/0.6	7.9/0.2	208/20	212/10	3.3/1.4	27/2	28/3	0.5/0.2	7.5/0.6	346/39	23/3
Soften-GW	4.8/0.4	8.0/0.2	93/12	88/10	2.2 /.0	26/3	23/3	0.2/0.2	7.8/0.7	194/18	23/3
Soften-BL	4.6/0.7	8.1/0.2	64/17	99/11	1.5/0.6	96/16	49/11	0.3/0.2	8.6/0.9	285/25	23/3
NF-BL	4.6/0.7	8.2/0.1	88/19	77/16	0.8/0.4	5/1	47/11	0.1/0.1	8.4/0.8	207/21	23/3
RO-GW	4.8/0.6	8.3/0.2	68/4	66/4	0.3/0.1	11/1	80/8	0.2/0.1	8.2/0.6	274/18	23/3
ENCoag-SW	4.5/0.4	8.2/0.2	59/5	134/54	1.2/0.5	235/26	44/6	0.3/0.2	9.5/1.8	394/58	23/4
NF-SW	5.0/0.7	8.3/0.2	68/6	62/5	0.4/0.2	12/3	64/10	0.2/0.1	8.2/0.9	238/32	23/5

RO = Reverse Osmosis, GW = Groundwater, NF = Nanofiltration, SW = Surface Water. EnCoag = Enhanced Coagulation, Bl = Blend, Conv = Conventional Treatment, Soften = Softening,T = Total, NPDOC = Nonpurgeable Dissolved Organic Carbon, D.O. = Dissolved Oxygen, TDS = Total Dissolved Solid, Temp. = Temperature.

was higher in the blended softened finished water. These differences were due to the presence of 27% surface water in the blend that was softened. The finished surface water had relatively higher sulfate and lower alkalinity. NPDOC in the finished water was lower in the softened water produced from the blend because of the lower NPDOC in the portions of the blend that were treated by enhanced coagulation and reverse osmosis filtration.

The enhanced coagulated finished water had higher sulfates and TDS than other finished waters because of sulfate addition during coagulation. Although the raw surface water NPDOC averaged 18 mg/L, the enhanced coagulated finished water NPDOC was lower than the NPDOC in the conventionally treated, softened or blended softened finished waters, even though the raw water groundwater NPDOC was 3.3 mg/L. Conventional treatment or softening removed little or no NPDOC relative to enhanced coagulation; however, the finished waters from all membrane processes had lower NPDOC than the enhanced coagulated finished water because of NPDOC rejection by nanofiltration or reverse osmosis.

The membrane-finished waters differed by chloride concentrations, as reverse osmosis processes were used to treat highly saline sources, which had very high chloride and sodium concentrations. The energy required to push saline waters through reverse osmosis membranes was relatively high because they were tight so as to reject chloride and sodium ions and produced a permeate stream that did not exceed goals or regulations. Reverse osmosis membranes typically use enough energy to just meet sodium or chloride goals and regulations. Hence, the finished water from the TBW Regional Desalination Facility has higher chloride concentrations than other finished waters, which is the case for almost all desalination systems.

The data in Table 2 clearly show significant differences for water quality among the finished waters produced by these treatment systems. Such water quality was representative of finished waters that represented required treatment processes to treat ground, surface or saline sources.

Biostability and NPDOC

Bulk water biostability

NPDOC is the sum of organic solutes in water and has subsets of AOC and BDOC as well as disinfection by-product (DBP) precursors. NPDOC can be viewed as a pseudo-master organic water quality parameter. Several models were developed that related NPDOC to DBP formation (Speitel *et al.* 2005) and to biological parameters such as BDOC in drinking water. Hence attempts were made to relate NPDOC, AOC, BDOC, HPC and PEPA (dependent variables) to source and treatment (independent variables), and to relate NPDOC (now independent variable) to AOC, BDOC, HPC and PEPA (dependent variables). The finished water NPDOC of each treatment system is shown in Figure 3. The raw water NPDOC for the groundwater averaged 3.3 mg/L, the raw water source for the surface water averaged 18 mg/L, which was reduced to 4 mg/L on average by CSF, which in turn was used for the feed stream to the nanofilter in the integrated-membrane system. The finished water NPDOC decreased by the degree of treatment as shown in Figure 3.

Conventional groundwater treatment removed no NPDOC and hence conventionally treated finished water had the highest finished water NPDOC of all treatment systems. Removal of calcium hardness by lime softening did not remove a large fraction of raw water NPDOC. Hence, the softened finished water had the second highest finished water NPDOC (2.2 mg/L) of all treatment systems, which was about one-third less than the raw groundwater NPDOC. The blended softened finished water NPDOC was 2.3 mg/L and was greater than raw groundwater because 27% of it was treated surface water and 11% of the blend was groundwater treated by reverse osmosis. Blended softened finished water removed approximately one-third of the NPDOC, which produced 1.5 mg/L NPDOC in the finished water. Such NPDOC removal observations during lime softening was reported in the literature (Taylor *et al.* 1986; Benefield & Morgan 1999).

The enhanced coagulation system reduced raw surface water NPDOC from 18 mg/L to 1.2 mg/L or 93% NPDOC removal on average. The CSF portion of the enhanced coagulation treatment system reduced NPDOC to 4 mg/L on average (78% reduction), hence the ozone-BAC process reduced the NPDOC from 4 to 1.2 mg/L. Only CSF was utilized for nanofiltration pretreatment, which meant the NPDOC concentration to the nanofilter in the integrated membrane system was 4 mg/L. The average NPDOC reduction by the nanofilter was from 4.0 to 0.8 mg/L or

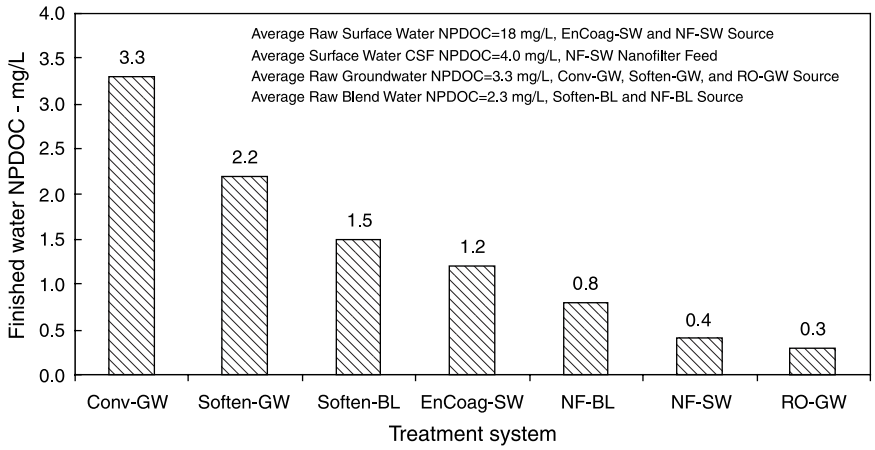

Figure 3 | Finished water NPDOC by treatment system. RO = Reverse Osmosis, GW = Groundwater, NF = Nanofiltration, SW = Surface Water. EnCoag = Enhanced Coagulation, Bl = Blend, Conv = Conventional Treatment, Soften = Softening, NPDOC = nonpurgeable dissolved organic carbon, CSF = Coagulation-Sedimentation-Filtration.

80%, which was expected by a nanofiltration system (Taylor *et al.* 1986; Taylor & Wiesner 1999). Even though CSF pretreatment significantly reduced the NPDOC concentration in the nanofilter feed stream for surface water treatment, the finished water NPDOC concentration in the permeate is typically not decreased as shown by field studies. However, fouling of the nanofiltration is significantly reduced by CSF pretreatment, which is necessary to render nanofiltration of highly organic surface waters feasible (Taylor *et al.* 1986). Although it is possible to remove more NPDOC using BAC than can be achieved by nanofiltration, such systems are not likely to be built because of cost. Hence surface water nanofiltration would typically be expected to remove more NPDOC from the same raw surface water than an enhanced coagulation system using ozone and BAC. The reverse osmosis treatment system produced the lowest average finished water NPDOC at 0.3 mg/L.

NPDOC removal by treatment system was ranked using statistical analysis. Pairwise *t*-test *p*-values for comparison of average finished water NPDOCs from the various treatment systems are shown in Table 3. For example, the *p* value for the comparison of the average 0.3 mg/L NPDOC in reverse osmosis produced finished water to the average 3.3 mg/L NPDOC produced by conventional groundwater treatment is 1.22×10^{-47}, which indicated that NPDOC removal by reverse osmosis was nearly always more than NPDOC removal by conventionally treated groundwater. The confidence level (CL) associated with this *p* value would be 1 minus the *p* value or 1. The CL typically used in engineering assessment is 95%, which meant any *p* value of 0.05 or less is statistically significant and that an error of rejecting observations with *p* values more than 0.05 will be correct 19/20 times and wrong 1/20 times. Lower CLs such as 90%, 80% and 70% are useful for interpreting experimental results and will result in making correct choices 9/10, 8/10 and 7/10 times, respectively, and wrong choices 1/10, 2/10 and 3/10 times, respectively. Hence the *p* values presented in Table 3, Table 4 and Table 5 can be compared for exact levels of statistical significance.

Inspection of Table 3 showed the ranking of NPDOC removal by treatment systems implied in Figure 3 was also statistically significant, with the exception of NPDOC in finished water produced by nanofiltration of surface water and nanofiltration of the blend. A line of ones run diagonally across Table 3 because the different treatment systems were identically listed in the first row and first column. The lack of statistical difference between the nanofiltered blend and nanofiltered surface water was not surprising since both systems used the same nanofilter, and the feed stream NPDOC into both nanofilters was similar (2.2 mg/L for blended nanofiltration system and 4 mg/L for surface water nanofiltration system).

NPDOC removal can be compared for common sources, which was perhaps a more rational basis for comparison. Hence, the data set was subdivided into common source waters, which generated three groups of source waters and treatment systems. Groundwater was

Table 3 | *p* values for treatment system NPDOC comparison

	RO-GW	NF-SW	NF-BL	ENCoag	Soften-BL	Soften-GW	Conv-GW
RO-GW	1.00	4.4E-07	2.40E-05	2.0E-16	3.0E-34	7.5E-47	1.2E-47
NF-SW	4.4E-07	1.00	0.34	1.1E-06	1.8E-15	7.4E-23	5.9E-26
NF-BL	2.4E-05	0.3	1.00	9.6E-07	5.6E-15	7.0E-22	3.3E-25
ENCoag-SW	2.0E-16	1.1E-06	9.6E-07	1.00	0.01	1.1E-08	2.8E-14
Soften-BL	3.0E-34	1.8E-15	5.6E-15	0.01	1.00	1.1E-08	7.4E-12
Soften-GW	7.5E-47	7.5E-23	7.0E-22	1.2E-08	1.1E-08	1.00	0.01
Conv-GW	1.2E-47	6.0E-26	3.3E-25	2.8E-14	7.4E-12	0.01	1.00

RO = Reverse Osmosis, GW = Groundwater, NF = Nanofiltration, SW = Surface Water. EnCoag = Enhanced Coagulation, Bl = Blend, Conv = Conventional Treatment, Soften = Softening, NPDOC = Nonpurgeable Dissolved Organic Carbon,underlined values indicate the differences are not significant to indicated level of significance.

treated by conventional, softening and reverse osmosis treatment systems. The reverse osmosis treatment system was used to simulate desalination but used groundwater as the source. Surface water was treated by an enhanced coagulation system that utilized ozone and BAC filtration and an nanofiltration using an integrated membrane system. Blended waters consisting of 62% GW, 27% SW and 11% reverse osmosis were treated by softening and nanofiltration treatment systems.

NPDOC, AOC, BDOC, log HPC and log PEPA in finished groundwater produced by conventional, softening and reverse osmosis treatment systems are shown in Figure 4. The same degree of statistical significance as observed for the comparison NPDOC was not observed for AOC, BDOC, log HPC and log PEPA. Inspection of Figure 4 revealed that the average finished water AOC, BDOC and log HPC decreased with increasing treatment (conventional to softening to reverse osmosis). In fact, there was little difference among biofilm cell density for any of the three groundwater treatment systems. Taking the finished water produced by the conventionally treated groundwater as a base, softening reduced AOC, BDOC and HPC by 19%, 35% and 74%, respectively, and desalination reduced AOC, BDOC and HPC by 61%, 72% and 97%, respectively. By these measures, reverse osmosis membrane treatment produced a more biostable finished groundwater than did either conventional or softening groundwater treatment.

NPDOC, AOC, BDOC, log HPC and log PEPA in finished surface water produced by enhanced coagulation with ozone and BAC and a CSF integrated membrane system using nanofiltration are shown in Figure 5. AOC, log HPC and log PEPA of finished surface water produced using the membrane system were lower, but BDOC was higher than in enhanced coagulation system with ozone and BAC as shown in Figure 5. Taking the finished water produced from the enhanced coagulation system as a base, the surface water membrane treatment system reduced the average AOC, HPC and PEPA by 42%, 40% and 24% but increased BDOC by 31%, relative to the enhanced coagulation system. The membrane treatment of the surface source appeared to have produced a more biostable finished surface water than did enhanced coagulation.

NPDOC, AOC, BDOC, log HPC and log PEPA of finished water produced from softening a blend of conventional, enhanced coagulation and reverse osmosis finished waters and from nanofiltration of the same blend are shown in Figure 6. AOC, BDOC and log PEPA of finished water produced by nanofiltration of the blend were less than the AOC, BDOC and log PEPA produced by softening of the same blend. However, log HPC was greater in the finished water produced by nanofiltration of the blend. The higher pH during softening might cause less HPC growth in blended softened finished water. Taking the blended softened finished water as a base, the nanofiltration system

reduced AOC, BDOC and log PEPA by 50%, 15% and 29%, respectively, but increased HPC by 182% relative to the blended softened finished water. The membrane treatment appeared to have produced a more biostable finished water from the blended source than did softening the same blended source.

As shown in Table 4, the biostability of water treatment systems was assessed statistically by comparing the means of bulk water AOC, BDOC, log HPC and biofilm log PEPA using a pairwise *t*-test. Four of the five AOC observations were significant. Based on the *p* values shown in Table 4, reverse osmosis finished water AOC was less than AOC in finished water produced by softening or conventional treatment at CL of 97%, 94% and 88%, respectively. Nanofiltered blended finished water AOC was also less than AOC in the blended softened finished water. Finished surface water AOC produced by the integrated membrane system was less than AOC in finished water produced by enhanced coagulation, ozone and BAC filtration. Conventional groundwater treatment AOC was higher than nanofiltration of surface water or blend water at 93% and 92% CL, respectively. BDOC in reverse

Table 4 | *p* values following a *t*-test comparison of AOC, BDOC, log HPC and log PEPA averages by source and treatment system

			GW		Blend	SW
Parameter	**Source**	**System**	**RO**	**Conv**	**Soften**	**NF**
AOC (ug/L)	GW	Conv	0.03			0.07
		Soften	0.12	0.49		
	Blend	NF		0.08	0.06	
	SW	EnCoag				0.17
BDOC (mg/L)	GW	Conv	0.04			
		Soften	0.27	0.30		
	Blend	NF			0.87	
	SW	EnCoag				0.71
Log HPC (cfu/mL)	GW	Conv	0.01			
		Soften	0.52	0.05		
	Blend	NF			0.52	
	SW	EnCoag				0.74
Log PEPA (cfu/mL)	GW	Conv	0.90			
		Soften	0.72			
	Blend	NF		0.63	0.64	
	SW	EnCoag				0.68

RO = Reverse Osmosis, GW = Groundwater, NF = Nanofiltration, SW = Surface Water. EnCoag = Enhanced Coagulation, Conv = Conventional Treatment, Soften = Softening, NPDOC = Nonpurgeable Dissolved Organic Carbon, AOC = Assimilable Organic Carbon, BDOC = Biodegradable Organic Carbon, HPC = Heterotrophic Plate Counts, PEPA = potential exoproteolytic activity, underlined values indicate the differences are significant to indicated level of significance.

Table 5 | *t*-test *p*-values of a comparison of average polyvinylchloride (PVC), lined cast iron(LCI), unlined cast iron (UCI) and galvanized steel(G) coupon biofilm densities (log PEPAs)

	PEPA			
	PVC	**LCI**	**G**	**UCI**
PVC	–	0.09	0.00	0.00
LCI	0.09	–	0.21	0.00
G	0.00	0.21	–	0.11
UCI	0.00	0.00	0.11	–

Based on average of biofilm densities of all treatment systems, PEPA = potential exoproteolytic activity,,underlined values indicate the differences are significant.

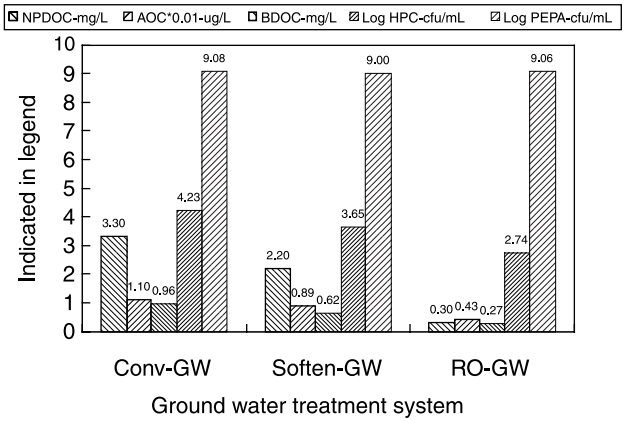

Figure 4 | Biostability finished water parameters and biofilm density for groundwater treatment systems. Conv = Conventional Treatment, GW = Groundwater, RO = Reverse Osmosis, Soften = Softening, NPDOC = Nonpurgeable Dissolved Organic Carbon, AOC = Assimilable Organic Carbon, BDOC = Biodegradable Organic Carbon, HPC = Heterotrophic Plate Counts, PEPA = potential exoproteolytic activity.

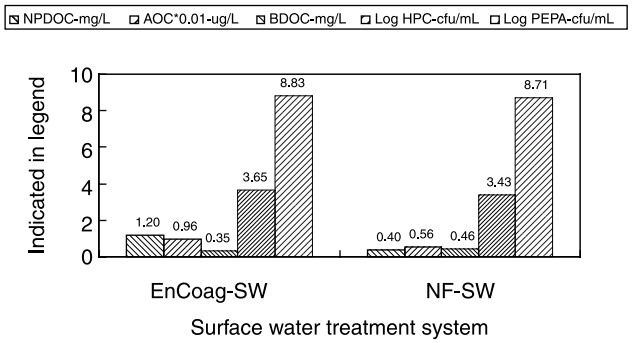

Figure 5 | Biostability finished water parameters and biofilm density for surface water treatment systems. EnCoag = Enhanced Coagulation, SW = Surface Water, NF = Nanofiltration, NPDOC = Nonpurgeable Dissolved Organic Carbon, AOC = Assimilable Organic Carbon, BDOC = Biodegradable Organic Carbon, HPC = Heterotrophic Plate Counts, PEPA = potential exoproteolytic activity.

osmosis finished water was less than BDOC in finished water produced by softening or conventional treatment. However, the CL of the comparison of reverse osmosis to conventional groundwater treatment was 96%. Softened groundwater BDOC was less than conventional treatment BDOC at a 70% CL. Finished groundwater produced by reverse osmosis and softening log HPC was less than conventionally treated groundwater log HPC at a 95% CL, which indicated both membrane filtration and softening effectively reduced HPCs relative to conventional treatment. Log PEPA did not vary among treatment systems, which indicated biofilm cell density was unaffected by treatment system in this work, which may due to the fact that no nutrient was limiting biofilm growth.

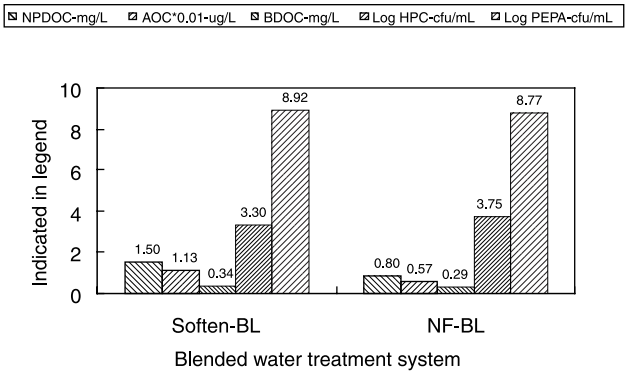

Figure 6 | Biostability finished water parameters and biofilm density for blended water treatment systems. Soften = Softening, BL = Blend, NF = Nanofiltration, NPDOC = Nonpurgeable Dissolved Organic Carbon, AOC = Assimilable Organic Carbon, BDOC = Biodegradable Organic Carbon, HPC = Heterotrophic Plate Counts, PEPA = potential exoproteolytic activity.

These results indicate that membrane filtration by reverse osmosis or nanofiltration reduced finished water AOC relative to conventional, enhanced coagulation or softening treatment systems. Reverse osmosis did reduce BDOC relative to conventional treatment and softening; however, nanofiltration did not reduce finished water BDOC relative to enhanced coagulation or softening. Reverse osmosis did reduce finished water log HPC relative to conventional treatment but not to softening. Softening effectively reduced finished water BDOC and log HPC relative to conventional treatment. Reverse osmosis treatment effectively reduced all bulk water measures of finished water biostability. Nanofiltration reduced finished water AOC, but did not reduce finished water BDOC and log HPC relative to softening and enhanced coagulation. There was little doubt that NPDOC removal increased by level of treatment; however, the same trend for AOC, BDOC, log HPC and log PEPA removal was not realized by the level of treatment with the possible exception of reverse osmosis.

Biofilm density

The average biofilm densities (log PEPAs) for varying pipe materials are shown in Figure 7. The results of a comparison of log PEPA means for polyvinylchloride, lined cast iron, unlined cast iron and galvanized steel pipe materials by a pairwise *t*-test are shown in Table 5. The visual presentation shown in Figure 7 gave the log PEPA order, and the statistical analysis shown in Table 5 gave the significance of the differences. That is, the order of biofilm cell density on pipe material was polyvinyl chloride < lined cast iron < galvanized steel < unlined cast iron. The range of

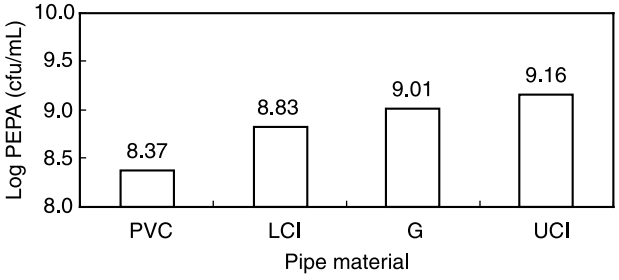

Figure 7 | Biofilm density (log PEPA) versus pipe material. PVC = Polyvinylchloride, LCI = Lined Cast Iron, UCI = Unlined Cast Iron, G = Galvanized Steel, PEPA = potential exoproteolytic activity.

CLs in Table 5 varied from greater than 99% to 79%. The lower CLs were associated with comparisons of biofilm cell densities on galvanized steel coupons to unlined and lined cast iron coupons, which were 89% and 79%, respectively. The polyvinylchloride and lined cast iron coupons had smoother interior surfaces than the galvanized steel and unlined cast iron pipe materials based on visual inspection. Additionally, the polyvinylchloride coupons appeared visually smoother than the lined cast iron coupons as the polymer pipe interior appeared smoother to the eye than the cement pipe interior. The unlined cast iron coupons appeared very rough as they were laden with iron tubercles. The galvanized steel coupons also appeared laden with corrosion scales, but slightly less so than the unlined cast iron coupons. Hence the order of biofilm growth did coincide with the order of roughness as determined by visual observation. The results coupled with the bulk water biostability parameters indicated that biofilm growth was controlled by material as opposed to water quality for the water quality and material conditions utilized in this work.

Linear regression

The results of linear regression of the averages AOC, BDOC, log HPC and log PEPA as dependent variables against the average NPDOC as the independent variable for a combined data set of all source waters are shown in Table 6. The coefficients of determination range from 0.33 to 0.76, which are not high for R^2; however, the p values range from 0.07 to near 0, which indicates a generally increasing trend of AOC, BDOC, log HPC and log PEPA with increasing NPDOC is correct. This was somewhat surprising that BDOC and log HPC did not decrease as NPDOC decreased in membrane treated surface waters. However, AOC, BDOC, log HPC and NPDOC were lower in membrane treated groundwater than in conventionally treated groundwater. Hence, the bulk water biostability parameters and the biofilm parameter were indicated to be dependent on source water NPDOC characteristics as well as NPDOC concentration, as shown in Figure 3 to Figure 6.

Table 6 | Linear regression of biostability parameters to NPDOC

Linear regression	R^2	p value
AOC $= 20.8 * \text{NPDOC} + 51.8$	0.62	0.04
BDOC $= 0.20 * \text{NPDOC} + 0.19$	0.78	0.07
Log HPC $= 0.32 * \text{NPDOC} + 3.09$	0.57	2.84E-05
Log PEPA $= 0.08 * \text{NPDOC} + 9.80$	0.33	1.55E-09

NPDOC = Nonpurgeable Dissolved Organic Carbon, AOC = Assimilable Organic Carbon, BDOC = Biodegradable Organic Carbon, HPC = Heterotrophic Plate Counts, PEPA = potential exoproteolytic activity.

CONCLUSIONS

Finished water quality was dependent on the source water quality and associated treatment system. Finished water produced by reverse osmosis and nanofiltration membrane treatment systems produced higher inorganic and natural organic water quality than enhanced coagulation, softening or conventional groundwater treatment systems.

Bulk water biostability parameters (AOC, BDOC and log HPC) were lower in reverse osmosis finished water and higher in finished water produced from a conventional groundwater treatment system. Average AOC in finished waters produced by reverse osmosis and nanofiltration membrane treatment systems was less than AOC in finished conventionally treated groundwater, softened groundwater and finished surface water produced by enhanced coagulation. This relationship was not clearly observed for BDOC or log HPC.

Biofilm growth on coupons cut from the pipes used to build the pilot distribution systems typically decreased as the level of treatment increased, with the exception of reverse osmosis finished water, which produced very high biofilm growth.

Biostability generally increased as the level of treatment increased, and the general order of biostability of process finished waters was: membrane > precipitative > conventional; and the order of biofilm growth with respect to pipe material was: unlined cast iron > galvanized > lined cast iron > polyvinylchloride.

Improved distribution system biological water quality as measured by lower AOC, BDOC, HPC and biofilm growth was directly dependent on nonpurgeable organic carbon and improved as the decrease of finished water nonpurgeable organic carbon, which decreased as the level of treatment increased.

ACKNOWLEDGEMENTS

Support for this research was provided by Tampa Bay Water (TBW), and AWWA Research Foundation (AwwaRF). The authors specially acknowledge Roy Martinez, AwwaRF Senior Account Officer, who was the Project Officer, and Chris Owen, TBW Quality Assurance Officer. The TBW Member Governments: Pinellas County, Hillsborough County, Pasco County, Tampa, St. Petersburg and New Port Richey; and the AwwaRF Project Advisory Committee are recognized for their review and recommendations. Pick Talley, Robert Powell, Dennis Marshall and Oz Wiesner from Pinellas County, and Dr. Luke Mulford from Hillsborough County are also specifically recognized for their contributions. UCF Environmental Engineering graduate students, especially Jorge Arevalo and faculty who worked on this project, are recognized for their efforts.

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First received 17 May 2006; accepted in revised form 9 July 2006