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Evaluation of drinking water quality produced by ultrafiltration membranes in distribution systems

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ABSTRACT

The final quality of the drinking water in the consumers' tap depends on the purification treatment applied, but also on its route through the drinking water distribution system (DWDS). This study assesses the quality of water treated by ultrafiltration membranes in a DWDS at a pilot scale in order to determine how possible network fouling affects drinking water quality, with special interest in natural organic matter, trihalomethanes (THM) and biofilm. Two different configurations were tested: an initial stage with an ultrafiltration (UF) system and a subsequent stage with coagulation-hydraulic flocculation (CF) pretreatment coupled to the UF. Although CF pretreatment helped reduce the dissolved organic matter (DOM) of the effluent that passed through the UF system, the high concentration of DOM promoted the fouling of the DWDS by organic matter deposits. This, together with the increase in water temperature in DWDS, allowed the adhesion of bacteria on the inner pipeline surface, encouraging the formation of a biofilm. Finally, despite the high concentration of DOM, THM generation was negligible throughout the study.

Keywords: Drinking water; Distribution system; DWDS; Ultrafiltration; NOM; THM; Biofilm

1. Introduction

Membrane technology is being increasingly applied in water treatment as an alternative to conventional techniques due to advantages such as its ability to produce a constant quality of water [1] and to remove a wide range of substances, as well as to work without adding chemicals due to the membrane's capacity to physical retention of micro-organisms [1]. However, this technology also has some drawbacks such as fouling or clogging of the membrane [2] or low retention efficiency of dissolved organic matter (DOM) [2,3].

Ultrafiltration (UF) membranes are among the most widely used in water purification, because they

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provide high-quality water with a greater efficiency than microfiltration membranes [4] and have less fouling problems that nanofiltration membranes [5]. Spiral-wound UF have been applied successfully to produce drinking water from high-quality reservoirs [1], increasing its operational capacity by applying pretreatments such as coagulation-flocculation [3,6].

The final quality of drinking water is the result not only of the treatment used in its purification, but the drinking water distribution system (DWDS) also plays a key role because, from the drinking water treatment plant to the point of consumption, many complex factors may contaminate the water flowing through the network [7,8].

Corrosion and fouling of the DWDS, besides deteriorating the network itself [9], lowers the water quality due to the release of particles, either organic [10] or inorganic [9].

Another factor influencing drinking water quality and salubrity is the presence of micro-organisms [11,12]. Pathogenic viruses and bacteria may be free in the water of the DWDS, implying a microbiological risk to consumers [13]. On the other hand, the micro-organisms carried by the water can colonize the surface of DWDS, forming a biofilm [11,14], polluting the network [15,16].

Another detrimental component in drinking water is organic matter, because it can form deposits on the inner walls of pipelines [8,17], promoting bacterial regrowth [18] and biofilm development [12]. All types contribute to soiling of the DWDS.

Among the most important factors lowering drinking water quality are the by-products formed by the reaction of disinfectants with natural organic matter (NOM) in water [19]. The by-products of chlorination in drinking water are usually trihalomethanes (THM) [16], about which concern has grown since their identification, because some of them are potentially carcinogenic [20].

The aim of this work was to study the quality of the drinking water treated by UF membranes in the DWDS over time regarding physicochemical and microbiological parameters, as well as THM generation and the biofilm formation and development. In addition, these are studied under two different experimental configurations: water treated with the ultrafiltration system alone, and water treated with a coagulation-hydraulic flocculation (CF) pretreatment.

2. Materials and methods

2.1. Description of the pilot-scale plant and experimental procedure

The experimental facility (Fig. 1) was designed to purify surface water from the Genil river (Granada, Spain). The raw water was drawn into the experimental system by a centrifugal pump (1 m^3/h).

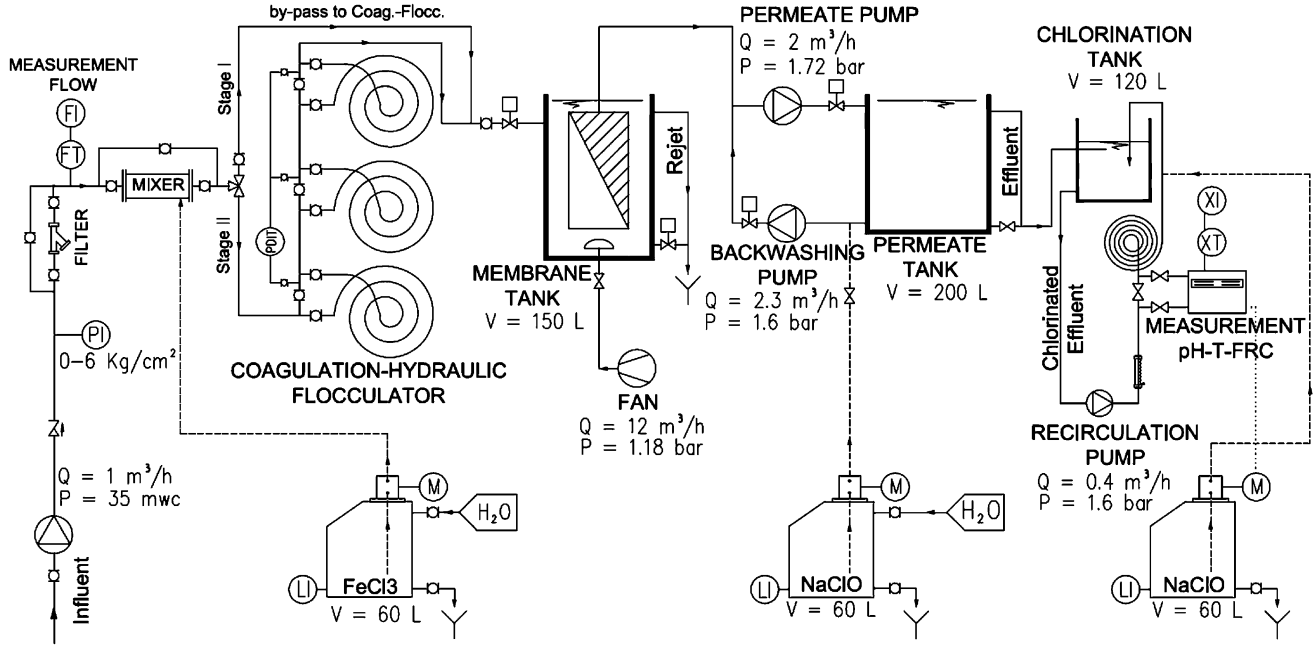

Fig. 1. Flow diagram of experimental pilot plant.

The ultrafiltration system was composed of a polyvinylidenfluoride spiral-wound membrane (SpiraSep 960, TriSep Corporation) with an effective pore size of 0.03 μm and 20.9 m^2 filtration area, which was submerged in the membrane tank. The water was filtered by a permeate pump (1 m^3/h) and was stored in the permeate tank. The membrane was continuously aerated by means of an air blower (15 Nm^3/h), and the permeate was periodically reversed to backwash the membrane (1.5 m^3/h, 30 s). Furthermore, chemical cleaning (i.e. soaking in a solution of NaClO 100 mg/l for 24 h, 20 min recirculation and rinse) was applied when the transmembrane pressure reached the limit (−0.7 bar).

The facility included a CF pretreatment, which could be coupled to the UF membrane. The hydraulic flocculator, consisting of three pipe rolls of 25 m each and 50 mm in diameter, was equipped with a set of valves to change the hydraulic retention time. $FeCl_3$ (10 mg/L) was dosed as coagulant.

The post-chlorination system was composed of a chlorination tank, a DWDS and a chlorine management system (Kontrol800, Seko). The DWDS was simulated with 300 m of polyethylene pipe of 16 mm diameter and a centrifugal pump providing a nominal flow rate of 0.5 m^3/h in order to achieve an approximate flow velocity of 1 m/s, which is the mean value used in real DWDS, according to CTE-BS-HS4 [21]. The chlorine management system consisted of a chlorine pH meter connected to a chlorine dosing pump which maintained a constant free residual chlorine (FRC) concentration according to the pH values. Temperature, pH and FRC were monitored each min and the data were recorded in a database.

The experimental facility was operated with two different configurations: configuration 1, the installation worked with the ultrafiltration system for 10 weeks (18 February to 28 April, 2013); and configuration 2, where the CF was coupled prior to the ultrafiltration module, which was tested for 18 weeks (29 February to 1 September, 2013). The DWDS was the same for both stages in order to track biofilm development over a longer time period.

2.2. Analytical methods

Throughout the study period, samples of water treated after the membrane (Eff_T) and distributed water (Eff_D) were taken daily to analyse both physicochemical and microbiological parameters. In addition, a segment of the pipeline from the DWDS was taken monthly in order to study the biofilm.

For the physicochemical analysis, water samples were collected in thoroughly cleansed plastic bottles and analysed immediately. All water samples were analysed for turbidity, UV absorbance at 254 nm wavelength (UV_{254}), total and dissolved organic carbon (TOC and DOC), specific UV absorbance (SUVA) [22] and THM.

Turbidity was measured by diffused radiation (DINKO D-112). For the determination of UV_{254}, water samples were passed through a filter of 0.45 μm and

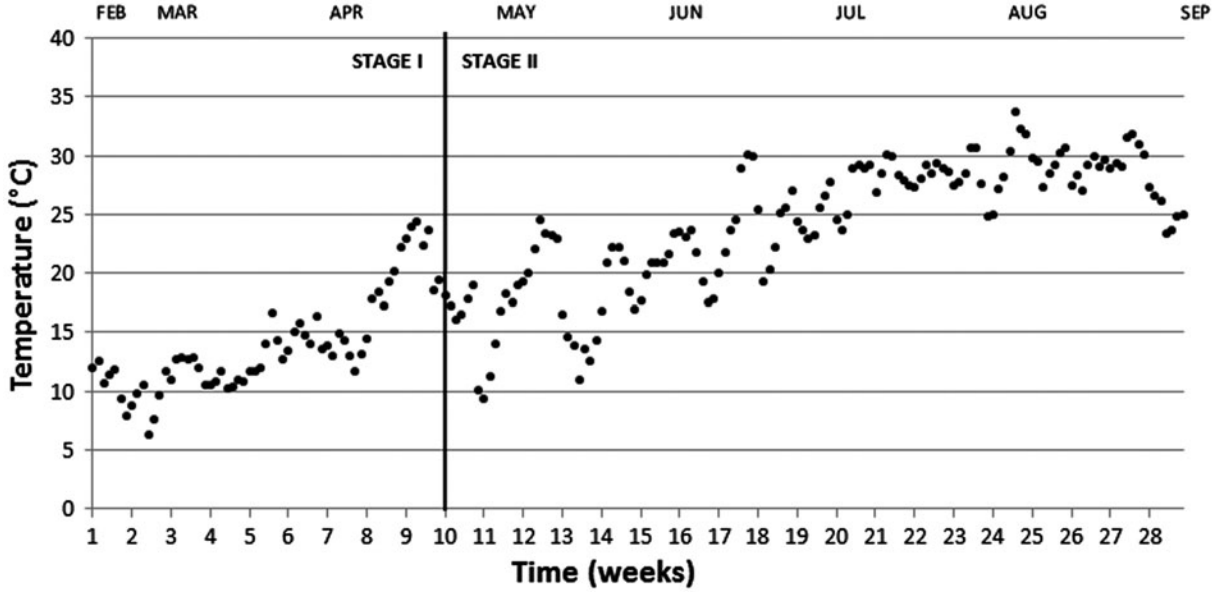

Fig. 2. Time course of the average daily water temperature in the experimental DWDS.

then a UV-visible spectrophotometer was used (Helios γ) with a 1 cm quartz cell. TOC and DOC were measured using a combustion TOC Analyser (FormacsTH, SKALAR). THM were analysed by gas chromatography with electron-capture detection coupled with mass spectrometry (GC–MS) and quantified according to analytical standards (Sigma).

For microbiological analyses, water samples were collected in 100 mL sterile glass bottles, which contained 1 mL of sodium thiosulfate ($Na_2S_2O_3$, 3%) to neutralize the chlorine effect, and they were analysed immediately after their collection. The total aerobic bacteria count (TAC_{22}) was carried out at 22˚C using the method specified in UNE-EN ISO 6222:1999. The presence of *E. coli* was studied using the membrane filtration procedure, according to UNE-EN ISO 9308–1:2001. In addition, with the purpose of discarding the faecal origin of the bacteria present in the drinking water, catalase, oxidase and API tests (BioMérieux) were carried out.

The development of biofilms on the surface of network was controlled by analysing the presence of bacteria on the wall of the pipeline. A segment of pipeline was analysed monthly and preserved with glutaraldehyde (3%) in a phosphate buffered saline solution (130 mM NaCl and 10 mM Na_2HPO_4/NaH_2PO_4, pH 7) in order to fix the possible biofilm in the segment. Samples were treated with methods described by Rúa et al. [23] and viewed by scanning electron microscopy (SEM) using a Zeiss DSM950 SEM operating at 5–30 kV, equipped with an energy-dispersive spectrometer [EDS Link Analytical Pentafet Si(Li)]. The single bacteria were counted in each micrograph (45 micrographs per sample) and their average value was calculated in bacteria per mm^2.

2.3. Statistical analysis

The data were analysed using the software IBM® SPSS® Statistics (v.21) for Windows. An analysis of variance (ANOVA) was run to determine the existence of statistically significant differences between the two stages with a significance level of 5% (p value < 0.05). Likewise, principal components analysis (PCA) was performed in order to determine which set of

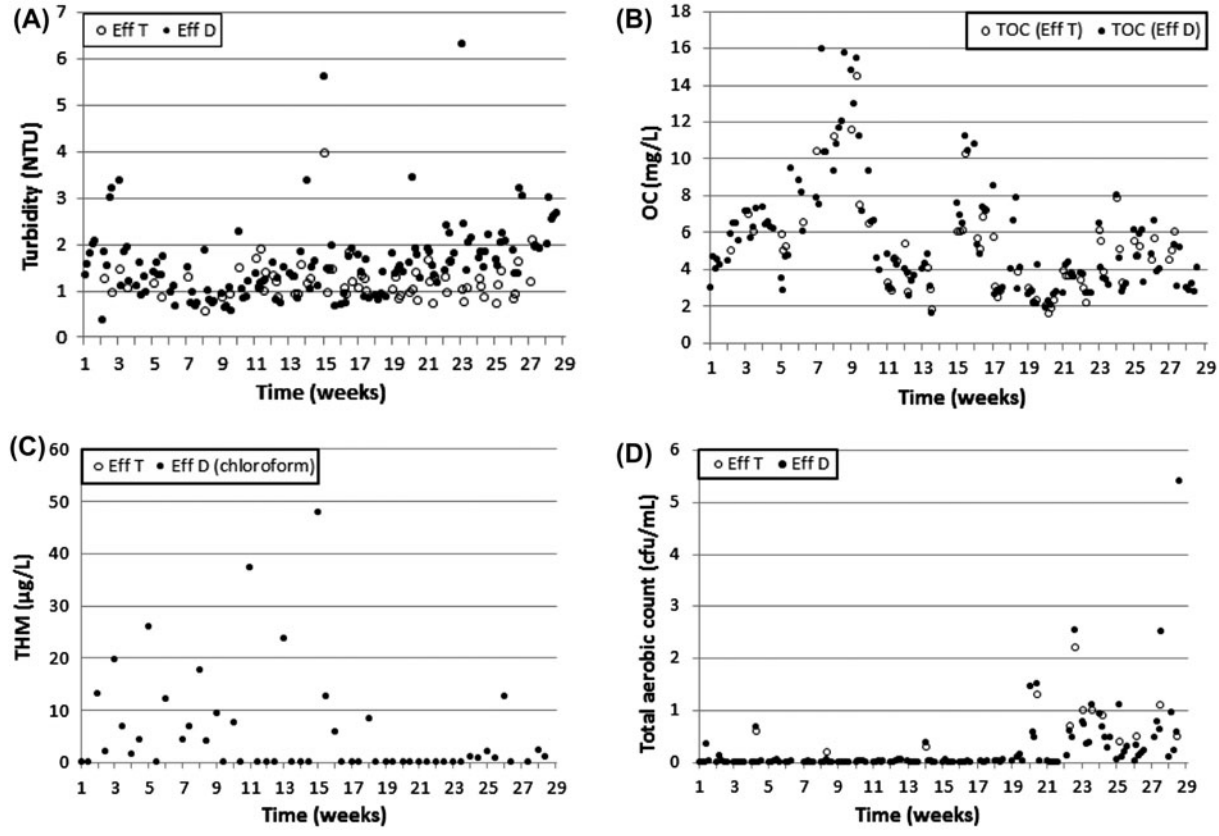

Fig. 3. Time course of the turbidity (A), TOC, and DOC (B), THM (C) and total aerobic count to 22˚C (D) of water after treatment (Eff_T) and water from the DWDS (Eff_D).

variables could reduce the dimensionality of the data bank.

3. Results and discussion

3.1. Continuous control parameters

FRC in the DWDS remained fairly constant at around the setpoint (0.4 mg/L) throughout the study period, indicating that the chlorine management system worked properly. Only on specific occasions, FRC values were recorded over the setpoint after a phase of membrane soaked in chlorine.

The pH of the water in the network remained consistently between 6.5 and 8.6, with mean values of 8.3 during stage 1, and 7.3 during stage 2 due to the acidifying effect of the coagulant. Changing of the setpoint of FRC due to pH was not necessary.

Regarding the water temperature in the DWDS, a seasonal variation (Fig. 2) was detected with higher values during summer (stage 2) due to the incidence of the solar radiation on the pipeline. This variation from 10 to 30˚C could affect the quality parameters such as the presence of bacteria in the water or further development of biofilms [23].

3.2. Drinking water quality in the DWDS

The turbidity registered values consistently below the legal limit (C.D. 98/83 EC) [24], except on certain occasions due to technical failures (Fig. 3(A)). During stage 1, the turbidity medium value of the Eff_D was around 1.4 NTU, while the turbidity mean value of Eff_T was 1.1 NTU without statistically significant differences (Table 1). During stage 2, Eff_D the mean turbidity value was 1.8 NTU while Eff_T was 0.9 NTU with statistically significant differences. These results indicate a deterioration in the drinking water quality during distribution, as previously reported by Rojas et al. [25], which was more prominent during the application of CF (stage 2), despite that the best water quality resulted after treatment. Turbidity values in DWDS followed an upward trend during stage 2 (Fig. 3(A)), coinciding with the increase in water temperature in DWDS (Fig. 1). Thus, a negative effect of temperature on drinking water quality can be expected in DWDS, according to the trend in turbidity values. Temperature and turbidity values showed a statistical correlation (Fig. 6).

The mean values found for TOC and DOC of Eff_T as well as Eff_D in both stages indicate that the water quality was low (Fig. 3(B)), which might be expected due to low efficiency in retention of DOC by the UF membrane [1,25]. Most of the TOC (over 95%) corresponded to DOC, confirming their influent origin [17]. Although the differences between Eff_T and Eff_D in organic carbon content was not significant, the decrease in more than 40% (Table 1) of the DOC in the Eff_D during stage 2 relative to stage 1 highlights the greater efficiency of the UF system with pretreatment of CF [6]. The results revealed neither a higher DOC concentration in the drinking water as it passed through the DWDS nor a direct effect of temperature.

The UV_{254} showed a behaviour similar to that of DOC, but with a less dispersion of the values and sharper differences between stages. These results reflect the greater efficiency of UF membrane associated with CF to remove humic compounds [6] given that stage 2 shows a lower rate more clearly than in stage 1.

Table 1
Summary of the physicochemical and microbiological parameters of water after treatment (Eff_T) and water from the DWDS (Eff_D)

	Stage I		Stage II	
Parameter	Eff_T $\bar{x} \pm SD$	Eff_D $\bar{x} \pm SD$	Eff_T $\bar{x} \pm SD$	Eff_D $\bar{x} \pm SD$
Turbidity (NTU)*	1.1 ± 0.4	1.4 ± 0.7	0.9 ± 0.1	1.8 ± 0.9
UV_{254} (m^{-1})*	11.3 ± 9.1	9.9 ± 7.6	2.9 ± 1.7	3.2 ± 2.7
TOC (mg/L)*	7.8 ± 2.6	7.6 ± 3.4	4.2 ± 1.7	4.3 ± 2.0
DOC (mg/L)*	7.3 ± 2.3	7.3 ± 3.1	4.2 ± 1.8	4.2 ± 2.0
SUVA (L/mg m)*	1.2 ± 0.7	1.2 ± 0.8	0.7 ± 0.2	0.7 ± 0.3
THM (μg/L)	nd	7.1 ± 7.5	nd	4.4 ± 10.8
TAC_{22} (cfu/mL)*	0.0 ± 0.1	0.0 ± 0.1	0.3 ± 0.6	0.3 ± 0.7
E. coli (cfu/100 mL)	0	0	0	0

Note: nd: not detected.

*There are statistically significant differences among the Eff_D from both stages ($p < 0.05$).

Almost all the values of SUVA for the Eff_T and Eff_D were lower than 2 L/mg m, indicating that the organic matter present in the water was mostly non-humic material, of low molecular weight and low hydrophobicity [26]. This would imply that the risk of THM generation should be low [26].

The chromatograms plotted from the analysis of THM indicate that these were not detected in the water after the membrane and that chloroform was the only one of the four THM analysed present in the water of the DWDS. Chloroform was detected in very low concentrations (C) as the SUVA values

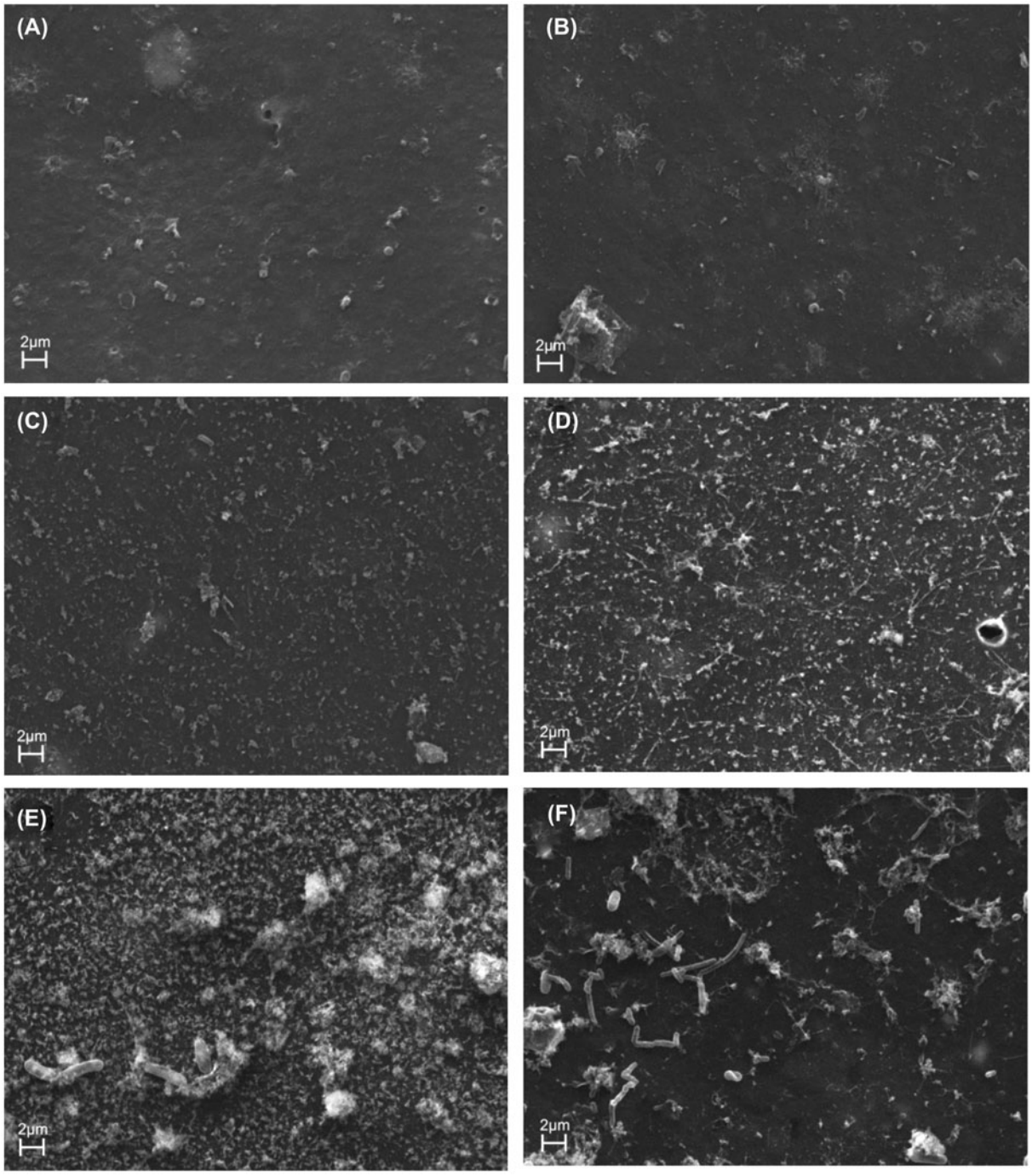

Fig. 4. Micrographs made with SEM of each sample of pipeline segment taken from the DWDS during the study. (A) March, (B) April, (C) May, (D) June, (E) July, (F) August (A and B from stage I, C, D, E and F from stage II).

suggested. The chloroform concentrations in the second stage declined with respect to first stage, perhaps due to greater DOC removal by the CF pretreatment, despite that this stage registered higher temperatures.

In terms of the microbiological parameters analysed, the effluent quality on DWDS was excellent since *E.coli* was completely absent and the TAC_{22} gave practically null values (Fig. 3(D)). This demonstrates the high efficiency of membrane technology as a physical disinfection method [4]. Several authors [1,4] have reported the presence of bacteria after a UF membrane was applied to water treatment, mainly in systems without sterile conditions. This justifies the need to maintain a residual chlorine concentration in the water distributed. Nonetheless, a slight increase was observed in the recounts of TAC_{22} at the end of the period corresponding to the summer months, but invariably with values below 6 cfu/mL. Again, an upward trend was found, in this case in a microbiological parameter, which cannot be attributed to failures in the membrane or to the application of CF as a pretreatment, in view of the values of Eff_T.

From all the colonies which were grown on culture plates for TAC_{22} and which came from the distribution system water, six different strains were isolated for quick identification. The API biochemical test revealed that all isolated strains were typical environmental proteobacteria, corresponding to Gram-negative aerobic bacilli, not enterobacteria, so its origin was not faecal. None of the isolated strain was pathogenic bacteria.

The ANOVA results gave statistically significant differences ($p < 0.05$) between all parameters related to effluent quality of the DWDS from the two compared stages except for THM (Table 1). The differences found for turbidity and TAC_{22} were opposite to those on the other parameters (Table 1). Both worsened over time with the rise in water temperature.

3.3. Development of the biofilm

The distribution system maintained a constant concentration of FRC, which should have prevented the proliferation of biological developments [27]. However, there was a significant risk of biofilm generation due to three factors: the presence of organic matter in significant concentrations due to the use of UF membranes in the purification process [6,25,28], the increase in water temperature in the DWDS that could have affected to the biofilm development [29] and the presence of bacteria in the permeate.

Fig. 4 shows a series of representative SEM micrographs of each sample of the pipeline segment. Over time, a significant accumulation of deposits was noted on the inside surface of the pipe. The chemical analysis of the deposits showed that the main component was carbon, revealing its organic origin. Although the experimental plant reduced the organic carbon content of the water mainly in the second stage, the surface of the pipeline was gradually covered by an accumulation of deposits (Fig. 4) despite that other studies conclude that the accumulation of particles in the DWDS can be controlled by reducing particles in the water treatment plant [30]. However, the accumulations of these deposits was expected due to the significant presence of organic matter in the treated water, which may be the first step in the subsequent colonization of the pipeline surface by bacteria [12].

Some bacteria begin to deposit on the inside surface of the pipeline, as be appreciated in the Fig. 4(C). The number of attached bacteria to the pipeline increased with time (Fig. 4(C–F)), which may have led to biofilm development [12]. In fact, bacterial division and development of structures such as fimbriae or pili became evident (Fig. 4(F)), and this could allow a reversible phase to become irreversible in the biofilm generation [29]. However, a real biofilm was not visible inside the pipeline after 7 months of operation, which can be considered an early stage of formation according to other studies [27,29].

The average bacterial count of each SEM micrograph of the pipeline surface showed a progressive increase, whose growth was adjusted to an exponential regression (Fig. 5). This count increase in the last few months was conditioned by the rise in water temperature and could accelerate the biofilm formation in the subsequent months. This could lower the drinking

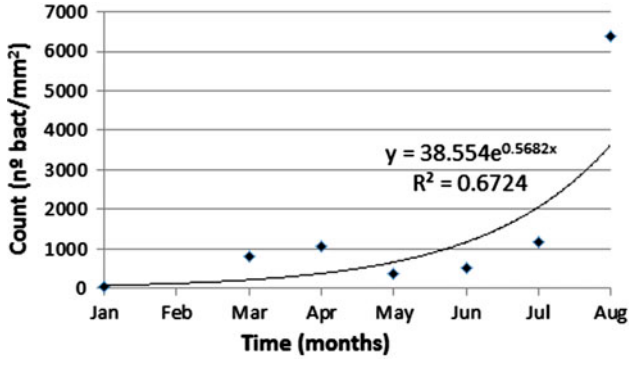

Fig. 5. Monthly progression of bacterial growth on the pipeline walls from DWDS.

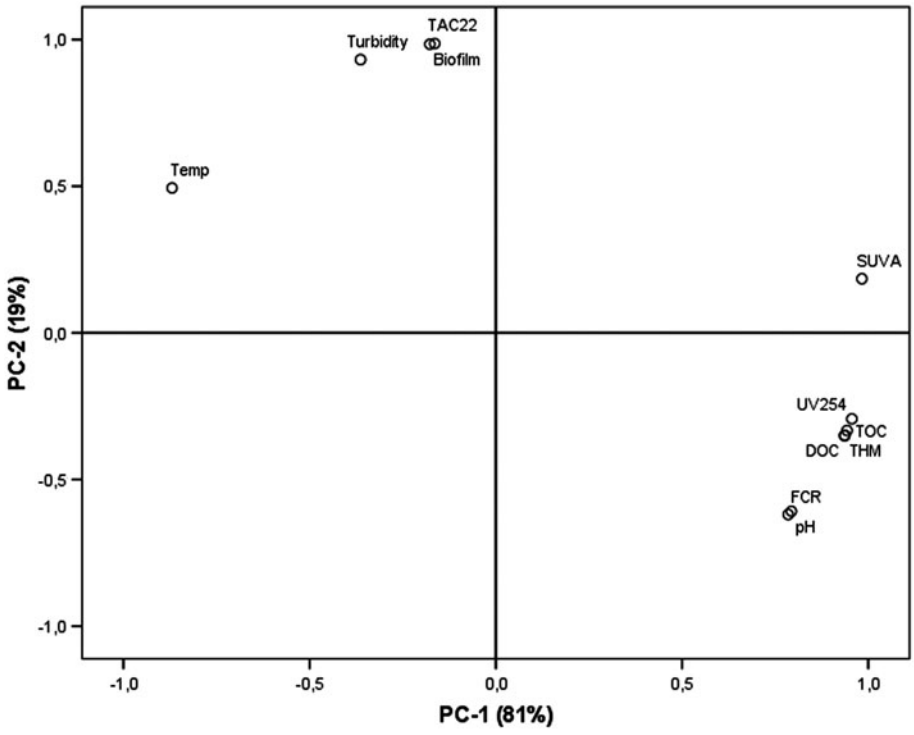

Fig. 6. Principal analysis component (PCA) of the DWDS data-set.

water quality by promoting bacterial proliferation, turbidity and NOM, as well as heightening the risk of THM generation.

The PCA showed that the system variables can be explained to 100%, reducing it to two principal components. As shown in Fig. 6, there was a strong correlation between the variables related to the NOM and the THM, since these variables are plotted in the same region. Likewise, turbidity, TAC_{22} and biofilm were positively related to the temperature. These two subsets of variables were independent of each other, because they were plotted orthogonally from one another.

4. Conclusions

Although the CF pretreatment helped decrease the DOM in the effluent produced by ultrafiltration system, its high concentration promoted the accumulation of deposits of organic matter on the pipeline surface of the DWDS. Despite the high concentration of DOC in the treated drinking water, the generation of THM in DWDS was negligible, reflecting low concentrations of chloroform.

The rise in temperature conditioned the water quality in the DWDS due to of the proliferation of bacteria attached to the inside surface of pipeline, with significantly greater turbidity and higher TAC_{22} values, regardless of purification treatment applied.

The operating conditions of the DWDS, mainly the high water temperatures reached and the characteristics of the treated water, resulted in rapid accumulations of deposits on the inner surface of the pipeline, enabling the attachment of bacteria. All this encouraged the formation of a biofilm. However, after 7 months of operation, the biofilm formation was hardly developed.

Acknowledgements

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