

Water reclamation with hybrid coagulation–ceramic microfiltration: first part of a long-term pilot study in Portugal

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ABSTRACT

Sustainable water management must nowadays consider alternative water sources and the use of reclaimed water is a good candidate. Low-pressure ceramic membrane filtration is an emerging option for safe water reclamation given its high mechanical and chemical robustness with safety and operational advantages. One-year pilot studies of hybrid coagulation–ceramic filtration were developed in Portugal in two wastewater treatment plants in the Lisbon metropolitan area. The results obtained demonstrated the technology's effectiveness, reliability and efficiency towards water quality, with the hybrid process consistently producing water highly clarified (monthly median <0.1 NTU), bacteria-free and with reduced organic matter content, regardless of the strong and severe variations in its intake.

Key words | hybrid coagulation–ceramic membrane filtration, membrane pilot, microfiltration, treated wastewater, urban reuse, water reclamation

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INTRODUCTION

Portugal has abundant potential freshwater resources. However, there is a shortage of its availability given the irregularity of their occurrence, both along the territory and over the year. The climate variability along the Portuguese territory is the first reason for the pronounced spatial differences that are observed in freshwater availability. Moreover, the consequences of this climate diversity on freshwater availability are greatly amplified by the demographic characteristics and the pressure of some important economic activities to meet growing needs. It is, therefore, important to develop water sources alternative to the ground and surface waters largely explored (Viegas *et al.* 2011). The use of treated wastewater is a good candidate, particularly in urban areas where the production is close to its use, with a constant availability although with a variable quality (Viegas *et al.* 2011).

In the scope of FP7 project Transitions to the Urban water Systems of Tomorrow (TRUST), as part of

technologies and management options, the study of alternative water resources was envisaged, namely for urban water reuse. A pilot membrane plant was sought for tertiary treatment of effluent designated for reuse in Portugal.

Low pressure membrane technology is an adequate option for removing turbidity and micro-organisms (including biological forms chemically resistant, e.g. protozoan (o)cysts, microalgae and cyanobacteria) from water, and their efficiency and effectiveness for dissolved organic matter may be substantially improved by, e.g. the use of the right coagulant and/or adsorbent, namely powdered activated carbon (PAC) (Campinas & Rosa 2010).

Polymeric membranes currently dominate the market for public water treatment applications (Loi-Bruegger *et al.* 2006; Li *et al.* 2011). However, low pressure ceramic membrane filtration (CMF), namely with microfiltration (MF) membranes, is an emerging technology in water treatment worldwide (Loi-Bruegger *et al.* 2006; Meyn & Leikness

2010; Panglish *et al.* 2010; Freeman & Shorney-Darby 2011; Hofs *et al.* 2011; Li *et al.* 2011; Xu *et al.* 2011; Abeynayaka *et al.* 2012; Meyn *et al.* 2012; Lee *et al.* 2013), but not in Portugal. Owing to their high mechanical robustness and chemical resistance, they may offer safety and operational advantages over polymeric membranes (Loi-Bruegger *et al.* 2006; Lehman & Liu 2009; Meyn & Leiknes 2010; Hofs *et al.* 2011; Abeynayaka *et al.* 2012). The stability of the ceramic membranes yields high security regarding membrane integrity as well as a long service life. At the same time, the ceramic membranes' robustness allows for effective cleaning procedures, enabling reliable membrane fouling control and more demanding operation. It allows dead-end filtration, with higher fluxes, which yield high water recovery rates (>95%) along with low energy consumption (<0.1 kWh/m³) (Meyn & Leiknes 2010; Löwenberg *et al.* 2014).

As with polymeric membranes, a previous coagulation step may significantly improve the CMF performance with respect to fouling, and enhanced removals of viruses and organic matter may be also obtained with the hybrid coagulation–ceramic membrane filtration (HCMF) process (Meyn & Leiknes 2010; Li *et al.* 2011; Meyn *et al.* 2012). The organic matter removal, in turn, brings significant benefits to drinking water production (Abeynayaka *et al.* 2012) as well as to water reuse, namely by minimising disinfection by-products (DBPs) formation (after chlorination) and biological regrowth in the distribution networks. Despite its high potential, up to now mostly studied for drinking water production, new knowledge and practice are needed with HCMF, particularly for advanced wastewater treatment and water reclamation, an application that brings larger challenges related with the feedwater average quality and often sharp daily variations.

The aim of this paper is therefore to demonstrate at pilot scale the long-term effectiveness, reliability and efficiency of the HCMF process towards water quality, especially regarding bacteriological content and turbidity (enhanced removal of organics is to be addressed in a study to follow).

To assess the membrane performance under different intake water qualities, three reclamation schemes (RSs) were studied in two test sites, Frielas and Beirolas wastewater treatment plants (WWTPs). These are two of the biggest WWTPs in Lisbon metropolitan area, with daily average flowrates of 54,500 m³/day and 70,000 m³/day,

respectively, and with on-going and planned treated wastewater uses (Table 1).

MATERIAL AND METHODS

Three RSs were studied (Table 1). The HCMF pilot is depicted in Figure 1 and the MF membrane (Metawater, Tokyo, Japan) characteristics are shown in Table 2. The wide range of operating conditions tested is summarised in Table 3.

The pilot was fully automated, remote controlled and had in-line monitoring of pressure, flow rate, temperature, pH and turbidity. It was operated at constant flux, in dead-end filtration mode and with in-line dosing of coagulant, in this case, ferric chloride. Regular cleaning was performed at the end of a filtration cycle by an initial backwash with pressurised permeate and then, after some seconds, by simultaneous air blown into the feed channels. The backwash pressure expanded from an initial value of 5 bar to about 2 bar at the end of the regular cleaning. The pressure of the air blow was also 2 bar. In total, the regular cleaning cycle lasted about 20 seconds. Chemically enhanced backwash (CEB) cycles were performed by backflushing and soaking the membranes for 10–15 minutes, either with sulphuric acid or sodium hypochlorite. The CEB procedure was conducted no more than four times per day.

Table 1 | Test sites and RSs studied

WWTP	Flowrate (m ³ /day)	On-going and planned treated wastewater use	RS
Frielas	70,000	<ul style="list-style-type: none"> • Agricultural and landscape irrigation • Air conditioning cooling system 	RS 1: Secondary effluent + Biofiltration + HCMF RS 2: Secondary effluent + Biofiltration + 100 µm Disc filter + HCMF
Beirolas	54,500	<ul style="list-style-type: none"> • Non-potable urban uses • Irrigation of public and private gardens 	RS 3: Secondary effluent + Sand filtration + HCMF

RS, reclamation scheme.

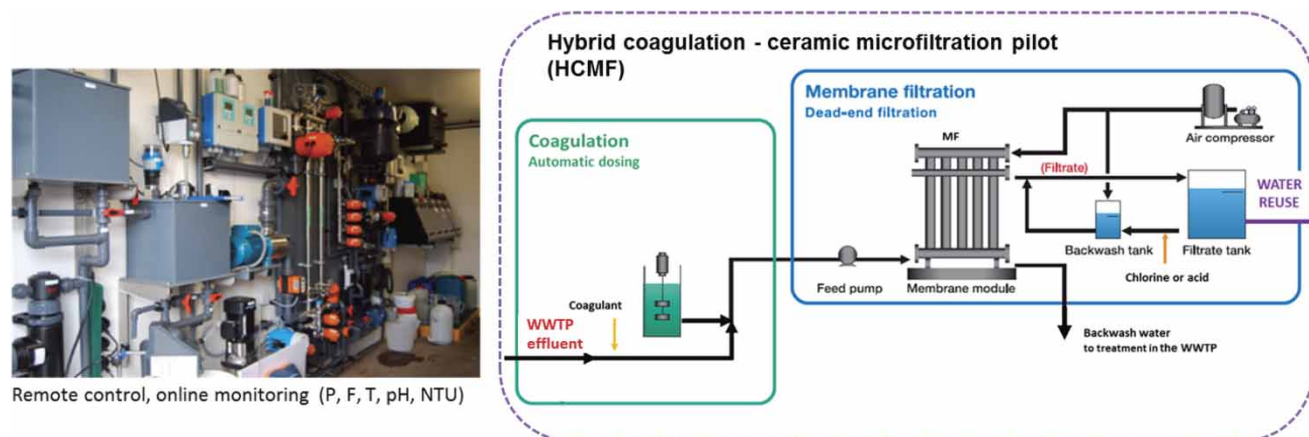


Figure 1 | HCMF pilot overview (on the left) and scheme (on the right, adapted from Metawater).

Table 2 | CMF membrane characteristics

Pore size	Material	No. of channels	Channel diameter	Module length/diameter	Membrane area per module	Total membrane area
0.1 μm	Al_2O_3	55	2.5 mm	1 m/30 mm	0.4 m^2	0.8 m^2

Table 3 | Range of operating conditions tested

RS	Fe dosing (mg/L)	Flux (lmh)	Filtration time (min)	CEB (acid + chlorine) /day
RS 1	3–7	80–100	30–60	≤ 4
RS 2	5–10	80–120	60–110	≤ 4
RS 3	7–10	100	60–110	≤ 4

Before and after the HCMF process, turbidity and pH were monitored in-line and grab samples were analysed in the laboratory for fecal coliforms (FC), total heterotrophs, transmittance at 254 nm (UVT254), colour (Abs436), dissolved organic carbon (DOC), UV absorbance at 254 nm (UVA254), chemical and biochemical oxygen demands, total suspended solids and electrical conductivity (data not shown herein for the latter four). Standard methods of analysis were used (Clesceri *et al.* 1998). Abs436, DOC and UVA254 were analysed onto samples filtered through 0.45 μm polypropylene filters. DOC expresses the dissolved organic matter concentration, whereas UVA254 expresses its character: the higher content in aromatic rings, conjugated double bonds and molecular

weight, the higher absorbance at 254 nm. UVT254 started being monitored (on non-filtered samples) at the end of RS1, as an indicator of water quality required to guarantee the effectiveness of UV disinfection in place in the studied WWTPs.

DOC was measured by the UV/persulphate chemical oxidation method using a Teledyne, TOC Fusion total organic carbon analyser. UVT254, UVA254 and Abs436 were measured using an UV/Vis Jasco V-630 spectrophotometer. pH and electrical conductivity were measured on a Consort C863T multiparametric potentiometer.

For in-line measured data (turbidity and pH), monthly box-plots were calculated, showing the monthly average, median, percentiles 5 and 95 (P5 and P95, respectively), maximum and minimum values.

The statistical significance of differences was assessed through one-way analysis of variance (ANOVA) *p*-value. Using this method, for a *p*-value greater than 0.05 (corresponding to 95% confidence), there is no statistically significant difference between the group means being compared, while for *p*-values below 0.05 a significant difference exists.

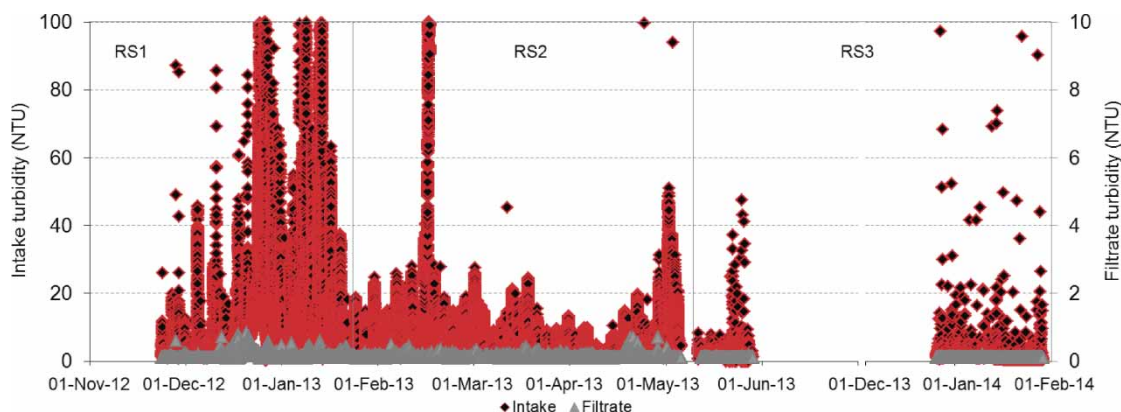


Figure 2 | In-line records of water turbidity in HCMF intake (left axis) and filtrate (right axis).

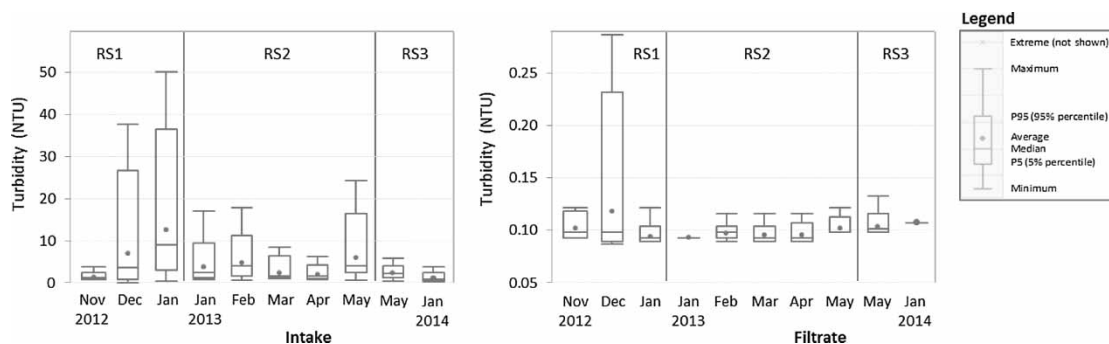


Figure 3 | Monthly box plots of water turbidity in HCMF intake (left) and filtrate (right).

RESULTS AND DISCUSSION

Representative parameters of HCMF intake and filtrate water quality are depicted in Figures 2–4.

In RS1 (Table 1), the biofilter effluent presented severe fluctuations of turbidity (Figure 2) from 1 NTU (percentile 5 in November 2012) to 37 NTU (P95 January 2013) (Figure 3). These fluctuations may limit the efficiency and compromise the effectiveness of UV disinfection, the option available in the WWTP for disinfecting the treated wastewater intended for reuse, and constituted a challenge for the HCMF process. Nevertheless, the HCMF pilot was able to cope with such water fluctuations and the treated water presented a very good and constant quality, with a monthly median of turbidity equal or lower than 0.10 NTU (Figure 3) and zero FC (intake FC was 1×10^4 – 5×10^5 CFU/100 mL) (Figure 4). The water pH was not substantially affected by HCMF, with 95% of the intake

values between 6.8 and 7.3 and the filtered water 0.3 units below (Figure 4), since the coagulant (ferric chloride) addition required for turbidity and FC removal was kept to 3–7 mg Fe/L.

In RS2, the biofilter effluent was filtered through a 100 μ m disk filter before feeding the HCMF (Table 1). This additional operation produced an HCMF intake of more steady quality, with turbidity values mostly between 1 and 10 NTU (Figures 2 and 3). In this period, the filtrate turbidity was mostly around 0.1 NTU (Figures 2 and 3). The intake water showed UVT₂₅₄ values between 44 and 70% (the lower values recorded may compromise UV disinfection effectiveness, depending on the UV lamp design) and the filtered water between 61 and 79% (Figure 4). The intake water showed up to 2×10^5 CFU/100 mL of FC, which were totally removed by the HCMF regardless of the water transmittance. During this longer period, with more frequent water analysis, intake DOC was 6–12 mg C/L and

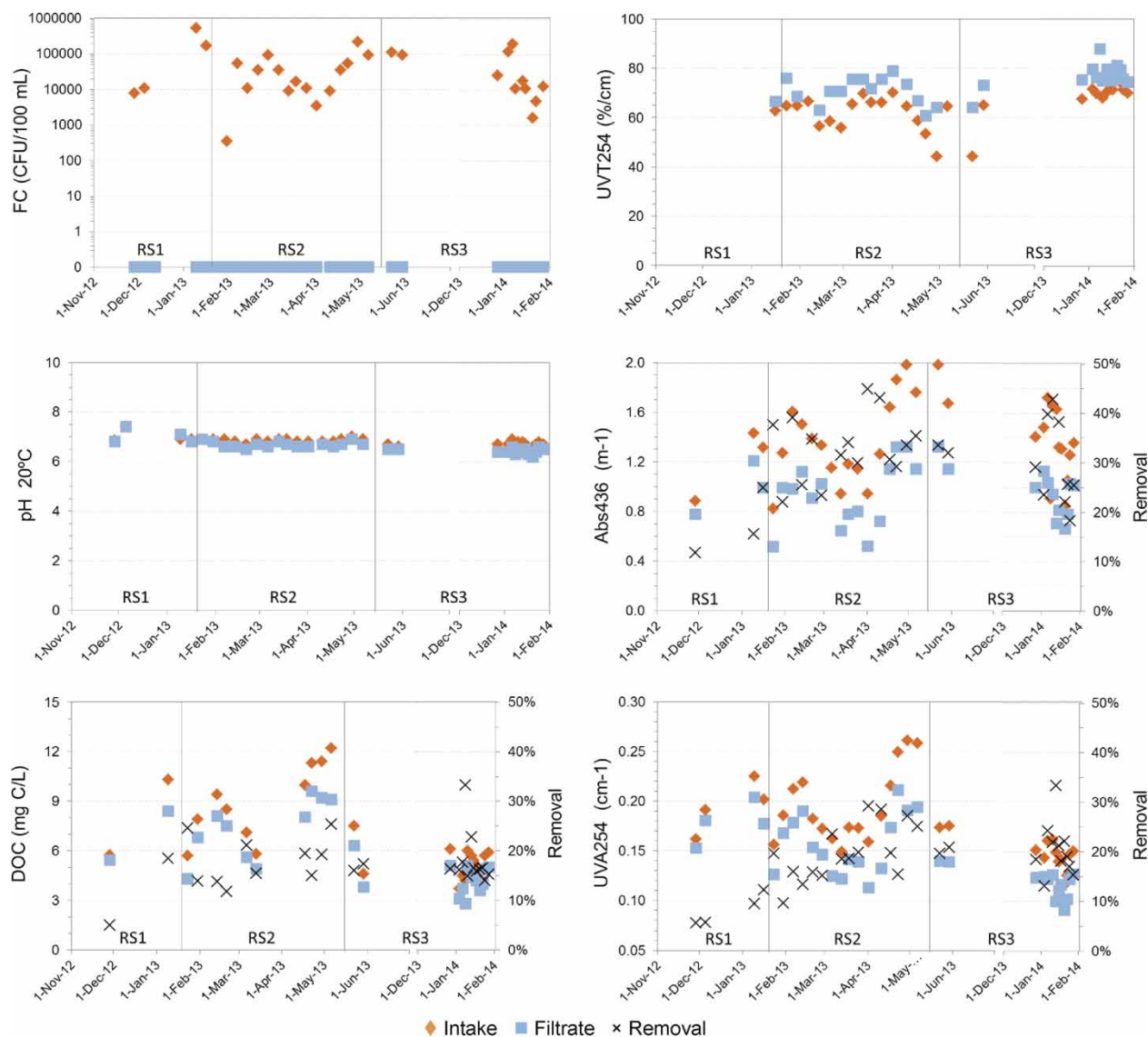


Figure 4 | HCMF intake and filtrate water quality.

UVA254 $0.15\text{--}0.26\text{ cm}^{-1}$ (Figure 4). The removals were up to 25% for DOC (12–25%) and 29% for UVA254 (9–29%). There were no statistically significant differences between DOC and UVA254 removal means as determined by one-way ANOVA (p -value was 0.48), so there was no significant preferential removal of UVA254 absorbing substances, i.e. of aromatic high molecular weight organics, over DOC. The colour (Abs436) removal varied between 22 and 45%, often exceeding 30% (Figure 4) and was significantly higher than DOC or UVA254 removals, as evidenced by the one factor ANOVA p -value of 0.0006 obtained. As in

RS1, the water pH was quite steady, again P5–P95 of 6.8–7.3 in the intake and 6.6–7.1 in the treated water, values obtained using a coagulant dose of 5–10 mg Fe/L.

RS3 was tested in a different test site, the Beirolas WWTP, where the secondary effluent is sand filtered prior to feeding the HCMF pilot (Table 1). The HCMF intake water was also of steadier quality than in RS1, showing turbidity values in a similar range as in RS2 (P95 of 4 NTU and 2.5 NTU in May 2013 and January 2014, respectively) (Figures 2 and 3). Again, the filtrate presented a very good and constant quality, with turbidity mostly around 0.10

NTU. The FC found in the HCMF intake water (with 44–76% UVT₂₅₄) were up to 2×10^5 CFU/100 mL (Figure 4) and were totally removed by the HCMF. The DOC values of the HCMF intake water were lower than in RS2, 4–8 mg C/L, as well as the UVA₂₅₄ values, 0.12–0.18 cm⁻¹ (Figure 4). The removals varied between 14 and 33% for DOC and 13–33% for UVA₂₅₄, and no significant preferential removal of one over the other was observed (*p*-value was 0.20). The colour removal varied between 18 and 57% and was significantly greater than DOC or UVA₂₅₄ removals, as determined by the one-way ANOVA *p*-value of 0.001 (less than 0.05). The HCMF intake water had a slightly lower pH in this test site, with 95% of the values between 6.5 and 7.2 and the HCMF treated water was 0.1 units below, with 95% of the values between 6.4 and 7.2, when dosing 7–10 mg Fe/L.

In general, the organic matter removals obtained were in the range expected. In other long-term studies involving ceramic MF of wastewater, Lehman & Liu (2009) observed DOC removals of 18–38%, quite similar to those found in the present study. For drinking water, similar studies (Loi-Bruegger *et al.* 2006; Meyn & Leiknes, 2010; Abeynayaka *et al.* 2012) reported removals from 20 up to 80%. As previously stated, in a study to follow, the enhanced removal of organics is to be addressed, namely by PAC dosing.

For the three RSs studied, the electrical conductivity was observed to be constant and around 1 mS/cm, and did not change after HCMF.

From the operational point of view, stable operation of the HCMF was obtained for each RS. As expected, higher quality intake water allowed a more demanding operation in RS2 and RS3, while for RS1 milder operation conditions behaved better.

In RS1, steady operation was achieved with a flux of 80 lmh, 60 minutes filtration time, 4 CEB/day and Fe dosing of 7 mg/L, with a transmembrane pressure (TMP) of 0.33 bar. With this set of mild operating conditions a water recovery rate of 96.6% and a treatment capacity of 1.71 m³/(m² d) were achieved.

In RS2, a more demanding set of operating conditions was attained, i.e. a flux of 100 lmh, 110 minutes filtration time, 2 CEB/day and Fe dosing of 7 mg/L, with 0.45 bar TMP. These conditions corresponded to 98.6% water recovery and a treatment capacity of 2.28 m³/(m² d).

In RS3, two sets of stable operating conditions were found, one corresponding to higher productivity and the other to lower energy consumption. The first set corresponded to a 100 lmh flux, 110 minutes filtration time, 2 CEB/day and 7 mg Fe/L, which yielded 98.6% water recovery, a treatment capacity of 2.28 m³/(m² d) and 0.45 bar TMP. The second set of operating conditions corresponded to equal flux (100 lmh) and cleaning practices (2 CEB/day) but to a lower filtration time, 60 minutes, and a higher Fe dosing, 10 mg Fe/L. With this second set of operating conditions the corresponding water recovery and treatment capacity were slightly lower, 97.7% and 2.20 m³/(m² d), respectively, but TMP was 0.36 bar, a value which is 20% lower than the value found with the previous set. It should be stressed that with these milder operating conditions the pilot was operated non-stop for 4 weeks, with only minor maintenance for replenishment of reagents.

In comparison with other long-term studies involving ceramic MF of secondary wastewater effluent, Lehman & Liu (2009) reported higher fluxes, 170 lmh, than those achieved in this study, although by the use of an ozonation pre-treatment step. A wide range of fluxes, from 60 to 250 lmh, is reported in the literature for drinking water treatment (Loi-Bruegger *et al.* 2006; Meyn & Leiknes 2010; Panglisch *et al.* 2010; Abeynayaka *et al.* 2012). As expected, similar studies with polymeric membranes for either wastewater treatment (Löwenberg *et al.* 2014) or drinking water production (Panglisch *et al.* 2010) achieved lower fluxes, up to 80 lmh.

CONCLUSIONS

The results obtained in this long-term testing of three RSs with significant differences in the intake water quality demonstrated the effectiveness, reliability and efficiency of the hybrid coagulation–ceramic MF pilot towards water quality. Regardless of the strong and severe variations in the HCMF pilot intake, particularly observed in RS1, the pilot consistently produced highly clarified water (monthly median <0.1 NTU), bacteria-free. In addition, it was able to reduce the organic matter content of the intake water, therefore reducing the DBP formation potential and producing high-quality reclaimed water for a wide range of uses.

From the operational point of view, stable operation was achieved in all RSs, using mostly 7–10 mg Fe/L, with fluxes between 80 and 100 lmh and with high water recovery rates, ranging from 96.6 to 98.6%.

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