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Techno-economic assessment of a membrane-based wastewater reclamation process

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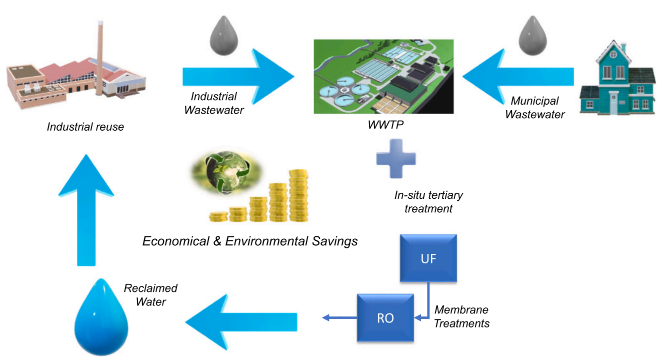
HIGHLIGHTS

- Results of UF-RO pilot plant on-site tests for wastewater reclamation are reported.
- 90% and 65% water recovery achieved for UF and RO stages, respectively
- Total removal of turbidity, TSS and microorganisms in UF treatment
- RO achieved high quality requirements for industrial reuse.
- Cost of reclaimed water (0.57 €/m^3) lower than tap water price (0.96 €/m^3)

GRAPHICAL ABSTRACT



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ABSTRACT

Reclaimed water plays a crucial role in the water cycle since it constitutes an effective way to improve the utilization of water resources and can help to cope with the water crisis. Membrane technologies for wastewater reclamation, especially ultrafiltration (UF) and reverse osmosis (RO), have received great attention in the past decades. In this work, an integrated prototype (2.5 m^3/h) based on the combination of UF and RO was operated in Vuelta Ostrera municipal wastewater treatment plant, located in the proximity of an industrial hub, to obtain water with the required quality for being industrially reused complying with Spanish law and the needs of industrial users. The influence of the process variables on water recovery was studied. Filtration time, backwash cycles duration and frequency, and the addition (or not) of coagulant, were the main variables studied during UF operation, while the recirculation rate of the concentrate stream and the UF permeate quality were the main variables for RO operation. Finally, the economic evaluation pointed to important savings in the OPEX of the process, when compared to the prize that industrial users are currently paying for water.

1. Introduction

The Sustainable Development Goals (SDGs) were adopted by all United Nation member states in 2015 as a universal call to action to end poverty, protect the planet and ensure that all people enjoy peace and prosperity by 2030 [1]. Among the 17 goals established, the number 6

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aims at ensuring universal availability and sustainable management of water and sanitation. However, nowadays more than 40% of the population lives in countries affected by water scarcity. By 2050, it has been projected that at least one in four people will suffer recurring water shortage [2]. For sustainable development to be achieve it is crucial to harmonize a secure supply of energy and fresh water within the environmental protection. The three elements are interconnected and are critical for the well-being of the general society. In this context, exploitation of new water sources is expected to contribute to satisfactorily comply with SDGs horizon. Research efforts have been conducted to develop new technologies and improve conventional treatments to provide quality water in a sustainable way [3,4]. Desalination and wastewater reclamation for further reuse have been highlighted as the main alternatives to procure water for different uses [5–7]. The large volumes of wastewater generated, because of the high consumption of water in households and industry, makes reclaimed water to appear as an important source of freshwater.

Identifying the most suitable treatment in each situation is a crucial step to achieve a cleaner and cost-effective production of reclaimed water, after integrating technical, economic and environmental aspects [8]. Pei et al. [9] identified several factors that should be generally considered before selecting the wastewater treatment technology: wastewater quality, wastewater quantity, construction and operating expenses, degree of difficulty in engineering construction, local natural and social conditions and whether there are new conflicts. The authors highlighted the treatment level required to reach the quality needed for the new use as the most important factor. Identifying wastewater composition within the intended reuse, in order to fulfill the quality criteria in each specific case, is also of utmost importance. As it was reported by Yang et al. [7] different reuse applications require different water quality specifications, so different treatment technologies must be applied.

With such perspective, membrane technology is widely applied as advanced technology for water treatment, because it has the ability to remove to very low concentration levels non-desirable compounds from wastewater and can offer new opportunities compared to conventional treatments [6,10,11]. Other advanced treatment technologies have also been applied, like electrochemistry-based technologies, such as electro-oxidation, electro-reduction, electro-coagulation or electrodialysis [12–14].

However, as it was reported by Liu et al. [15] although significant progresses have been made using these technologies, there is still a challenge to change their status of "promising technology" to "practical technology". Capacitive deionization (CDI), along with its variants, such as membrane capacitive deionization (MCDI) and flow-electrode capacitive deionization (FCDI) have also been tested, presenting promising results for both seawater desalination and wastewater reuse [16–18].

However, there are still many challenges to overcome to reach the same deployment level as membrane technologies have achieved.

Membranes are widely used due to their many advantages such as continuous and automatic operation, easy process realization, compact installation, selective separation and high rejection efficiency of contaminants [19]. Moreover, continuous improvements have been made over time, addressing among others, the use of improved membrane materials, better composite and multilayer membranes, novel coatings and coating modifications, and improved fabrication processes [3].

Pressure-driven membrane processes have been extensively used for wastewater treatment [20]. Among them, we can distinguish between low pressure processes, microfiltration (MF) and ultrafiltration (UF), and the medium-high pressure processes, nanofiltration (NF) and reverse osmosis (RO). Biological processes can also be combined with membranes in the form of membrane bioreactors (MBRs) a combination of a conventional activated sludge process (CAS) and a submerged or external MF/UF membrane filtration [21,22]. Among non-pressure driven membrane processes, the following technologies can be highlighted as for their interest in wastewater reclamation and desalination: forward osmosis (FO), membrane distillation (MD), and membrane crystallization (MC). Currently, RO has become the most mature membrane technology for seawater desalination and wastewater reclamation for industrial reuse [5,23,24]. However, membrane fouling adversely impacts the overall process efficiency becoming one of the main drawbacks of this technology [10,24].

In this sense, integrated membrane processes can offer the best solution in terms of efficiency of pollutants separation, performance, fouling control and cost [6]. Multiple-step processes or integrated processes combining biological, electrochemical and membrane based technologies are essential to improve the performance and purity of the water product [15].

A sustainable membrane-based wastewater treatment, which involves the implementation of an integrated system composed by UF - RO is proposed in this work. Pilot plant demonstration tests were performed on-site at a municipal wastewater treatment facility close to an industrial hub. In this work the technical and economic assessment of the integrated system proposed for industrial water reuse is performed. Moreover, a comparison between costs obtained in the present case study with those reported in the literature is also presented. Finally, a comparison of water costs currently paid by industries with the estimated cost values obtained for the treatment proposed in this work is also provided.

2. Description of the case-study: UF + RO integrated system

In this work, we evaluate a case-study that considers an integrated system combining UF and RO at pilot plant scale. This integrated system serves as *in-situ* tertiary treatment for the secondary effluent of a municipal wastewater treatment plant (WWTP) located in Vuelta Ostrera (Cantabria, northern coast of Spain). During the studied period, the WWTP was treating an average influent flowrate of 52,538 m^3/day, with a population equivalent (PE) of 310,000 inhabitants. The secondary effluent of the WWTP presents high variability in the main parameters during the timeline of a 20 months sampling period, as it can be observed in Table 1, that presents minimum, maximum and average values for each parameter, accompanied by the analytical procedure followed for their determination. The arithmetic averages have been calculated taking into account all data available for each parameter.

UF has been demonstrated to be able to remove suspended solids, turbidity, organic matter, sediments, colloidal substances, microorganisms and certain macromolecules, providing a suitable feed to the RO unit [25–28]. The aim of the RO unit is to achieve low conductivity permeate water with adequate properties to feed low and medium pressure boilers, and fulfilling the standards established in the Royal Decree 1620/2007 [29] which sets the legal framework for the reuse of reclaimed water in Spain. A process diagram of the integrated system is shown in Fig. 1. Both pilot plants, UF and RO, were designed and manufactured by the Spanish company APRIA Systems S.L. (Cantabria, Spain), in collaboration with the company Hidroglobal S.A. (Barcelona, Spain).

The UF unit is composed of: i) a pre-filter (Arkal, Spin Kin), which is a ring filter made of propylene O-rings, able to retain particles larger than 130 μm; the pre-filter consists of two modules, one of them for filtration and the other for providing the air needed during BW, and ii) two hollow fiber (HF) membrane modules, placed in parallel. The UF unit operates in cyclic mode. During the filtration stage, the secondary effluent, after retention of coarse particles in the ring filter, is fed through the bore of the HF in dead-end filtration mode, and a cake layer of solids is progressively formed on the inner side of the porous HF membrane. After a certain filtration time, the backwash (BW) cycle is started, aimed at removing the cake layer of retained suspended solids and the reversible membrane fouling. Then, the next filtration cycle is started. The chemical cleaning (CC) stage is applied only when the BW is not sufficiently effective to return the transmembrane pressure to the initial value. CC

Table 1
Quality of the secondary effluent of the WWTP used as feed water to the advanced membrane treatment in the present case-study.

Parameter	Units	Range	Average	Analytical method
Turbidity	NTU	1.12–26.3	6.62	Portable turbidimeter HANNA HI-731321
pH		6.34–8.86	7.19	Portable pH-meter HANNA pH 500
Total Suspended Solids (TSS)	mg/L	0.0–30.0	10.7	Filtration through glass fiber filters
Total Dissolved Solids (TDS)	mg/L	267–3320	639	Portable conductimeter HACH Sension 5
Conductivity	μS/cm	571–6340	1325	
Bicarbonate	mg/L	61.0–415	300	Titrimetric method with HCl and methyl orange indicator
E. coli	cfu/100 mL	$3.1 \cdot 10^3$–$7.7 \cdot 10^5$	$1.6 \cdot 10^5$	Enzymatic method – Colilert – 18 IDEXX
Total coliforms	cfu/100 mL	$6.3 \cdot 10^3$–$5.2 \cdot 10^6$	$8.7 \cdot 10^5$	
$N\text{-}NH_4^+$	mg/L	17.3–60.8	35.7	Distillation/titration, Standard Methods 4500-NH3
Total Organic Carbon (TOC)	mg/L	4.56–50.8	22.0	TOC Analyzer – TOC-VCPH Shimadzu
Polysaccharides	mg glucose/L	1.52–12.4	5.63	Dubois colorimetric method
Proteins (mg/L)	mg/L	3.25–15.4	9.47	Lowry Peterson Method – TP0300 Sigma Aldrich
Chlorine	mg/L	0.00–0.23	0.04	Portable colorimeter HACH DR/890, kit DPD method.
SiO_2	mg/L	6.10–10.3	8.17	Portable colorimeter HACH DR/890, kits: - Heteropoly Blue, 0–1.6 mg/L - Silicomolybdate method, 0–75 mg/L
F^-	mg/L	2.45–4.77	3.22	Ion Chromatography (Dionex)
Cl^-	mg/L	35.4–366	122	
NO_3^-	mg/L	1.34–5.59	3.47	Anion analysis: AS9-HC column, eluent Na_2CO_3 9 mM
PO_4^{3-}	mg/L	2.10–5.53	3.48	
SO_4^{2-}	mg/L	55.6–103	72.1	
K^+	mg/L	4.44–19.5	12.0	Cation analysis: AG9-HC column and methanesulfonic acid 9 mM
Ca^{2+}	mg/L	65.3–121	82.9	
Mg^{2+}	mg/L	12.0–45.5	18.9	

operation is similar to BW but with the addition of chemical reagents (NaOH, HCl, NaOCl). The detailed procedures for performing BW and CC are included as Supplementary Material. The UF permeate water is collected in a dumping tank to feed the RO unit or use in the BW and CC cycles. The RO unit is formed by two spiral wound modules placed in series, with partial recirculation of the second retentate to the feed of the RO unit. As a rule, the RO flushing was applied when the pressure drop (difference between the feed pressure and the pressure of the concentrate stream) increased by 10% of the initial value. Table 2 summarizes the characteristics of the UF and RO membrane modules implemented in the pilot units.

The secondary effluent was fed into the dead-end UF unit at an average flowrate of 2.5 m^3/h. The RO pilot plant was operated at a low-pressure gradient ($P = 11$ bar) and feed flowrate of 2.25 m^3/h. A dose of 8 mL/m^3 of commercial antiscalant was added in order to prevent scaling during operation. The dosage point is indicated in Fig. 1. The UF unit implemented on-line monitoring of the feed flowrate, turbidity and inlet and outlet pressures, while the RO unit registered the permeate flowrate and conductivity and the pressures of the feed and concentrate streams. Every working day, we sampled the UF feed, permeate and BW streams and the RO permeate and concentrate streams. Regular analysis of TSS, turbidity, pH, conductivity, TDS, *Total Coliform, E. coli*, bicarbonate concentration, $N\text{-}NH_4^+$, free chlorine, proteins, polysaccharides, anion and cation concentrations were performed, following the procedures compiled in Table 1.

The following parameters were calculated to characterize the performance of the water treatment process:

UF unit:

- Transmembrane pressure (*TMP*): is the difference between the feed pressure and the permeate pressure, measured at the inlet and outlet ports of the UF module operated in dead-end filtration mode,

$$TMP = P_{in} - P_{out} \tag{1}$$

Table 2
Characteristics of the UF and RO membrane units.

	UF	RO
Membrane ID	X-Flow Aquaflex (Norit)	LFC1 4040 (Hydranautics)
Module configuration	Hollow fiber	Spiral wound
Membrane material	Polyethersulfone (PES)	Polyamide (PA)
Average pore size	0.02 μm	Dense layer
Diameter of capillaries	Inner diameter: 8 mm	–
Number of units	2 in parallel	2 in series
Membrane area per module (m^2/unit)	40	7.9
Total membrane area (m^2)	80	15.8
Operational mode	Dead-end filtration	Tangential flow
Other parameters	Feed pressure varied between 0 and 2 bar	Feed pressure P = 11 bar

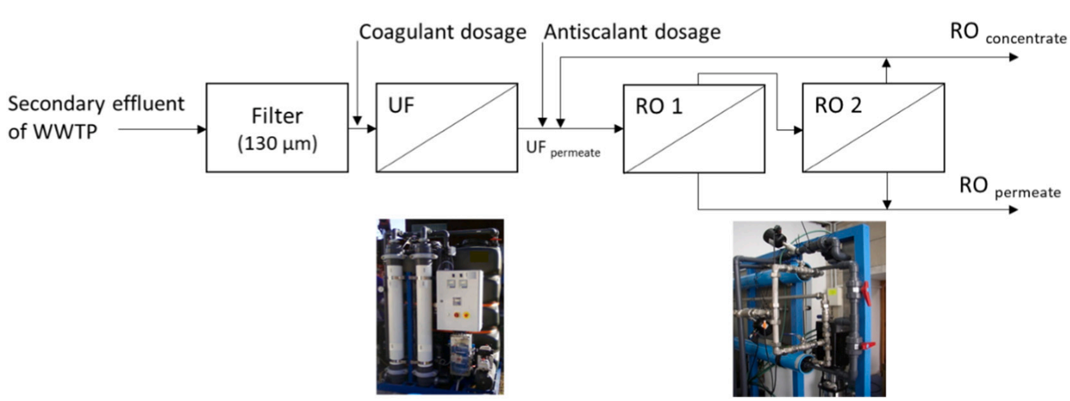

Fig. 1. Scheme of the advanced UF-RO treatment applied to the secondary effluent of the municipal WWTP.

- Permeate flux (J): is the permeate flow rate per unit membrane area. It varies along the duration of the filtration cycle,

$$J = \frac{1}{A}\cdot\frac{dV}{dt} \tag{2}$$

where A is the membrane surface area (m^2), and V is the permeate volume (m^3).

- Permeation Resistance (R_T): is the sum of the membrane resistance (R_M) and the resistance caused by fouling (R_F). Darcy's law correlates R_T with J (m^3/m^2 h), the fluid viscosity, η (Pa·s) and the TMP (Pa):

$$R_T = R_M + R_F = \frac{TMP}{\eta\cdot\boldsymbol{J}} \tag{3}$$

- Eq. (4) is used to correct the relative viscosity as a function of temperature (T), where T is in °C,

$$\eta = \frac{497\cdot10^{-3}}{(T+42.5)^{1.5}} \tag{4}$$

- Water recovery: is the percentage of feed water obtained as permeate (V_P) after subtracting the amount of water consumed in the BW (V_b), relative to the total permeate water,

$$\%water\ recovery = \frac{J\cdot A\cdot t_f - V_b}{J\cdot A\cdot t_f}\cdot100 = \frac{(V_P - V_b)}{V_P}\cdot100 \tag{5}$$

where t_f is the filtration time (h) and V_b is the volume of permeate water used for the BW.

RO unit:

- Recirculation rate: is the percentage of concentrate that is recirculated back to the feed stream,

$$Recirculation\ rate = \frac{Q_{CR}}{Q_P + Q_{CD}}\cdot100 \tag{6}$$

where Q_{CR} is the flowrate of concentrate that is recirculated, Q_P is the RO permeate flowrate and Q_{CD} is the flowrate of concentrate that leaves the pilot unit.

- Productivity: is the RO efficiency calculated as the % of the feed stream (Q_{UF}) transformed into permeate water (Q_P),

$$\%Productivity = \frac{Q_P}{Q_{UF}}\cdot100 \tag{7}$$

where Q_{UF} is the inlet feed flowrate to the RO unit, with no recycling.

The RO permeate flux is normalized to a reference temperature of 25 °C using a correction factor (TFC). The TCF is an empirically based, exponential function that is inversely proportional to temperature [30] and is calculated as follows:

$$J_{corr} = J\cdot TCF = J\cdot exp\left(k\cdot\left(\frac{1}{273+T} - \frac{1}{298}\right)\right) \tag{8}$$

here T is expressed in °C, J in L/m^2h and k is a factor related to the membrane material, that for polyamide membranes used in this case-study is 2700.

- The permeability coefficient ($L_{P,RO}$) of the RO membrane is calculated at 25 °C as the ratio between the normalized RO permeate flux over the pressure difference between the feed side and permeate side of the RO membrane:

$$L_{P,RO} = \frac{J_{corr}}{\left(P_{RO,feed} - P_{RO,permeate}\right)} \tag{9}$$

3. Technical assessment of the case-study

In a first step the UF system was operated independently in daily periods of 6–8 h. Afterwards, the RO unit was also installed, and both systems (UF - RO) operated 6–8 h/day. Once the system was working properly, the prototype was operated in continuous mode 24 h/day. In the last period, the UF unit operated automatically while the RO system operated in a semi-automatic mode, as the flushing of the membrane modules were activated manually.

3.1. UF pilot plant

Fig. 2 presents an example of the daily operation of the UF unit in terms of TMP and R_T along five consecutive filtration + BW cycles that were performed in one working day. During each filtration cycle the TMP increased with time, because a cake layer appeared on the membrane surface owing to the accumulation of the retained solids. After the filtration cycle, the BW was activated, and initial TMP values were very much recovered. The same observation was previously reported by Sangrola et al. [31], who established that BW is one of the most widely used membrane regeneration techniques in large-scale water and wastewater treatment applications. However, after each filtration cycle, the initial TMP value is a bit higher than in the previous cycle, due to internal pore fouling issues. A point is reached where BW is no longer sufficient and CC is required to recover the initial TMP [25]. In this work, the chemical cleaning was activated after 21 filtration cycles, in order to recover the initial TMP value.

Attending to Eq. (5), the water recovery can be modified by varying the duration of the filtration (t_f) and the volume of permeate water (V_b) used in the BW cycles. In Fig. 3 the relation between water recovery and TMP is presented. These data were obtained in experiments with coagulant addition. In this work, 90% water recovery was achieved by implementing filtration cycles of 60 min and BW time of 50 s. By increasing the water recovery from 78 to 83%, the maximum TMP achieved during the filtration cycles observed a significant increase. However, further increase of water recovery did not lead to a notable change on TMP, which remained around 250 mbar.

Another important variable analyzed during UF operation was the addition of $FeCl_3$ for conditioning the UF feed water. Some researchers recommend the use of a coagulant to promote the formation of a cake layer that can be easily removed during BW [15,32,33]. Fig. 4a compares the TMP values in the UF unit, obtained with and without $FeCl_3$ addition to the feed water. The coagulant dose used in these experiments was 4.5 mL/m^3 of $FeCl_3$ (40%). As can be observed, dosing $FeCl_3$ resulted in lower TMP values, proving that the coagulant was efficient in modulating the porosity of the cake layer, resulting in more permeable cake layers that were easier to clean off in the BW step. This behavior could be directly correlated to the turbidity of the UF feed. Fig. 4b shows that lower turbidities were observed when $FeCl_3$ was added to the UF feed, demonstrating that $FeCl_3$ was efficient in creating aggregates. Moreover, in Fig. 4c the relationship between feed turbidity and TMP is presented, showing that higher turbidity values are translated into higher TMP average values.

During the UF treatment, the operating conditions, defined by the duration of the filtration cycles and the frequency of the BW and chemical cleaning, influenced the productivity rate. However, the operating conditions did not affect notably the quality of permeate water. Table 3 shows the characteristics of the UF permeate stream with the removal percentage obtained during UF for each parameter, calculated from the average of the UF permeate and the WWTP effluent average value (Table 1). Note that the permeate sample did not come from the influent sample, as the hydraulic retention time in the UF unit was not considered. It can be noted that turbidity, TSS and

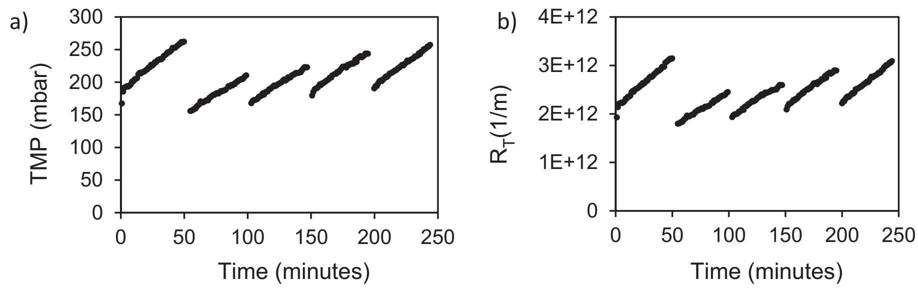

Fig. 2. Cyclic behavior of a) *TMP* and b) R_T during daily operation of the UF unit.

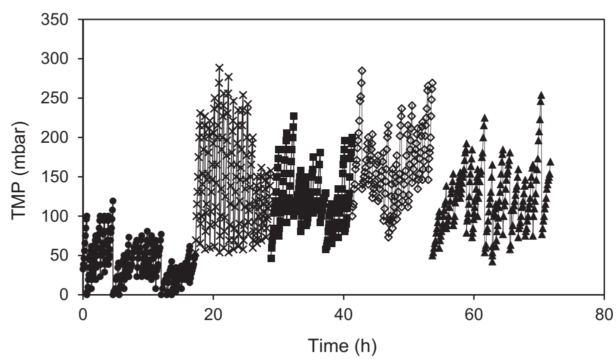

Fig. 3. TMP values at different % water recovery of the UF unit: (●) 78% water recovery (filtration cycle 45 min. and BW time 70 s.); (×) 83% water recovery (filtration cycle 45 min. and BW time 60 s.); (■) 87% water recovery (filtration cycle 60 min. and BW time 60 s.); (◊) 89% water recovery (filtration cycle 45 min. and BW time 40 s.); (▲) 90% water recovery (filtration cycle 60 min. and BW time 50 s.).

microorganisms, expressed as *Total Coliform* and *E. coli*, were virtually eliminated in their entirety. Moreover, high removal percentages were reached for polysaccharides, proteins and organic matter determined as total organic carbon (TOC). On the other hand, conductivity, bicarbonate, silica, ammonium and ions concentration remained practically the same as in the feed water.

3.2. RO pilot plant

The RO unit was fed with the UF permeate water, obtained in UF experiments in which iron chloride coagulant was applied. The input variables analyzed during the RO treatment were: i) the recirculation rate of the concentrate stream (Eq. (6)), and ii) the conductivity of the UF permeate used as the RO feed. Both variables will determine the characteristics of the permeate and concentrate streams of the RO unit. So, in this work different RO productivities, calculated as given by Eq. (7), were obtained by varying the recirculation rate of the RO concentrate stream, the quality of the permeate being influenced by the variation in the feed water composition.

Fig. 5 presents the RO permeate flux obtained when the unit was operated at different productivities in the range 40–80%. Pilot scale experiments were performed along a period of 9 months, from February to October, in which the feed water temperature varied substantially.

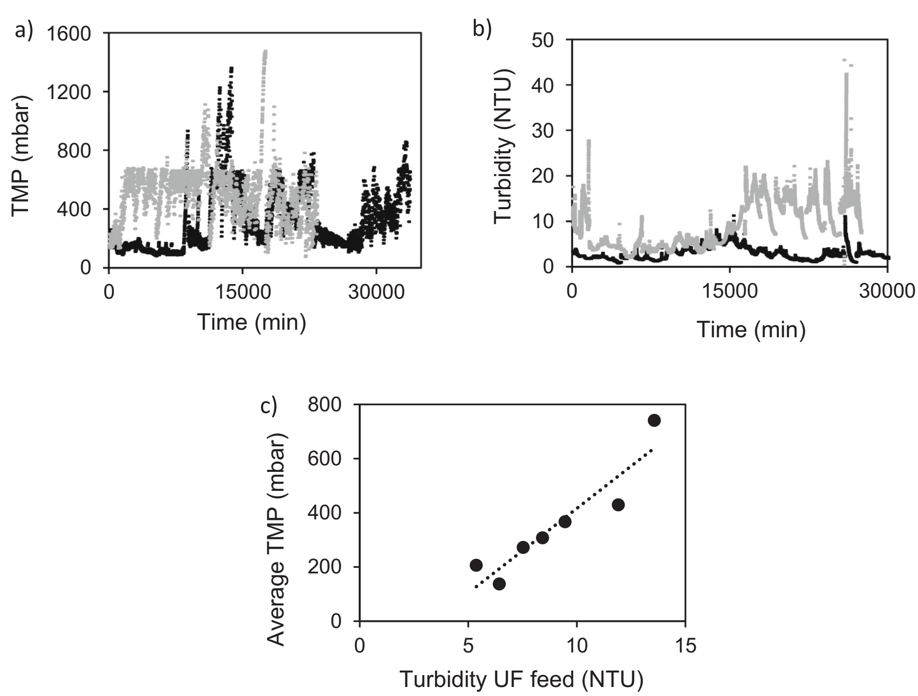

Fig. 4. Comparison of UF performance (●) when $FeCl_3$ was added as coagulant ($FeCl_3$ dose: 4.5 mL/m^3), and (●) when there is no addition of any chemicals. a) TMP; b) turbidity of the feed water before its entrance in the UF unit, and c) relationship between the turbidity of the UF feed water and the average TMP developed during UF operation. Linear fitting: $TMP = 62.4 \times Turbidity - 207.6$. $R^2 = 0.86$.

Table 3
Characteristics of the UF permeate stream and removal percentage obtained for each parameter during UF.

Parameter	UF permeate		% Removal
	Range	Average	
High removal			
Total coliform (cfu/100 mL)	0–921	70	99.9
E. coli (cfu/100 mL)	0–649	17	99.9
Turbidity (NTU)	0.00–2.98	0.25	96.2
TSS (mg/L)	0.00–6.00	1.16	89.2
Partial removal			
TOC (mg/L)	0.00–12.9	5.80	73.6
Polysaccharides (mg/L)	0.23–6.99	1.99	64.7
Proteins (mg/L)	1.40–9.89	5.92	37.5
Low removal			
TDS (mg/L)	293–2030	564	11.7
Conductivity (μS/cm)	602–3920	1187	10.4
Bicarbonate (mg/L)	61–415	285	5.0
SiO_2 (mg/L)	6.00–8.60	7.5	8.2
$N\text{-}NH_4^+$ (mg/L)	16.9–72.0	35.6	0.28

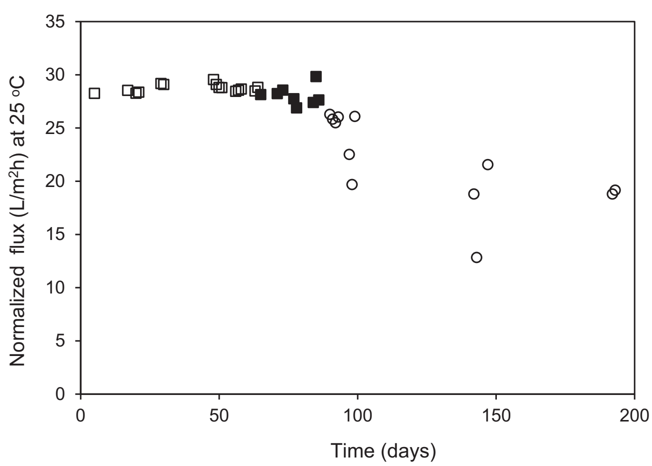

Fig. 5. RO permeate flux corrected at 25 °C for the productivity ranges assessed: □ 40–50% productivity ■ 50–60% productivity ○ 70–80% productivity. Feed pressure: 10.9 ± 0.7 bar.

Therefore, permeate water fluxes were normalized to 25 °C, using Eq. (8). The normalized permeate flux was maintained at 28.5 ± 0.68 L/m^2h, when the RO unit was operated in the RO productivity range from 40 to 60%. The membrane permeability, defined in Eq (9), results in a value of $L_{p,RO} = 2.6$ L/h m^2 bar. The volumetric water flux data achieved in the present study are in good agreement with data reported by Bartels [34]. The membrane permeability is similar to other water permeability values reported in literature for RO membranes used in brackish water desalination, *e.g.*: the BW30 Filmtec membrane water permeability is 2.9 L/h m^2 bar, according to the manufacturer [35]. The differences could be assigned to the proprietary characteristics of the commercial membrane module used in the present study, which are fitted to the treatment of low salinity brackish water.

Increasing the productivity of the RO unit at 70–80% resulted in a significant reduction of the permeate flux. This behavior can be attributed to the increase in the recirculation rate which is translated into a higher salinity of the feed solution and in consequence an increase in the osmotic pressure, and a decrease in the effective pressure gradient across the membrane. In fact, conductivity values in the concentrate stream for 40, 50 and 70% recirculation rate were 1767 ± 155 μS/cm, 2185 ± 1216 μS/cm and 4043 ± 1987 μS/cm, respectively. The increase of the concentrate conductivity generates a slight increase of the osmotic pressure, that does not justify the drastic change of the permeate flux. Therefore, the steep permeate flux decay is mostly assigned to membrane fouling phenomena occurring at the high salts concentrations that are achieved in the concentrate stream, even though a broad spectrum antiscalant was added to prevent the formation of calcium carbonate and other common scaling species.

Conductivity values for the UF and RO permeates were registered during the operation, showing that the quality of the RO permeate is directly influenced by the quality of the feed stream, that in this case corresponds to the UF permeate. In parallel, the UF permeate conductivity is practically the same as of the secondary effluent of the WWTP.

Fig. 6 presents the conductivity achieved in the RO permeate as a function of the conductivity of the UF permeate, showing a linear response for a wide range of the secondary effluent conductivity values between 600 and 4200 μS/cm for the feasible productivity range of 30–60%.

The characteristics of the permeate and concentrate streams of the RO unit and the limits established by Spanish legislation [29] and by EPA Guidelines for Water Reuse [36], are summarized in Table 4, it can be seen that the most demanding water quality requirements for industrial reuse can be reached in the RO permeate, except the ultrapure water that is demanded by high pressure boilers in thermal power generation. Salt rejection was higher than 99%, as calculated from conductivity data. Similar rejection values were observed for TDS and silica. Removal values greater than 90 and 80% were obtained for TOC and ammonium, respectively. These results are in agreement with those observed by Ozbey-Unal et al. [37] who obtained reclaimed water with acceptable limits for reuse as industrial cooling and boiler water systems by applying an integrated treatment of MF and RO.

So, it can be concluded that the proposed treatment composed by UF and RO pilot plants is suitable for wastewater reclamation for industrial reuse. The main disturbance to the operation of the tertiary treatment is the high variability of the secondary effluent quality. Ultrafiltration is aimed at the removal of suspended solids mostly formed by bacteria, natural and synthetic macromolecules. The productivity of the UF unit is affected by the content of total suspended solids, linearly related to its turbidity. Using the transmembrane pressure as indicator, the process control should modify the filtration cycle time and the frequency of BW and chemical cleaning. Both variables determine the productivity rate, while only the feed quality has a relevant influence on the permeate quality. RO is aimed at the production of a purified low salinity permeate, although retention of organic compounds, micropollutants, silica and ammonia is also needed. In wastewater reclamation facilities, given the mild salinity of the WWTP secondary effluent, the RO productivity is mostly determined by the feed pressure, whenever the productivity range is maintained below 60%. Further productivity

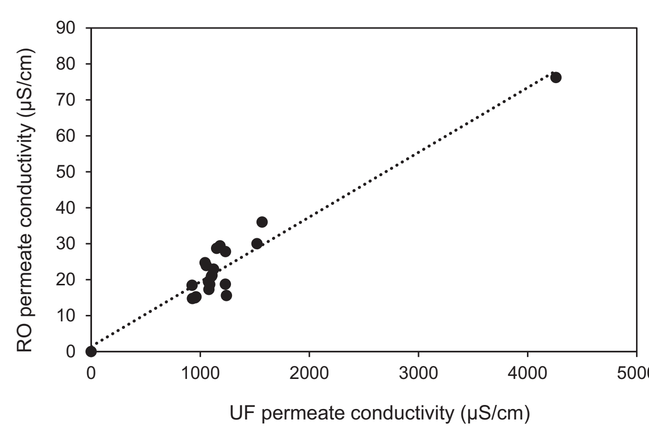

Fig. 6. Relationship between the conductivity of the UF permeate and RO permeate streams for 30–60% productivity. The linear relationship can be established by the equation: $Cond._{RO,\ permeate} = 0.019 \times Cond._{UF,\ permeate}$, $R^2 = 0.914$.

Table 4
Characteristics of the RO permeate and concentrate streams.

Parameters	Productivity								Spanish legilstation	USEPA Guidelines
	30–40%		40–50%		50–60%		70–80%			
	RO perm.	RO conc.	RO perm.	RO conc.	RO perm.	RO conc.	RO perm.	RO conc.		
Turbidity (NTU)	0	0–0.9	0	0–22.4	0	0–0.63	0–1.22	0–9.4	15[a] 1[c]	
Conductivity (μS/cm)	6.23–36.8	1004–5240	8.92–18.9	1446–2550	9.8–26	1650–3560	27.2–189	1758–5370		80–5400[d] 0.15–0.25[e]
TOC (mg/L)	<LOQ[f]	<LOQ[f]	<LOQ – 2.7[f]	8.4–28	<LOQ –1.7[f]	14.4–25.4	0–2	0.5–32.4		≤30 mg/L BOD[e]
SiO_2 (mg/L)	<LOQ[f]	<LOQ[f]	0.05–0.19	11.7–15.1	0.04–0.17	15.2–18.2	0.13–0.34	33.1–46.7		1–150[d]
E. coli (fcu/100 mL)	<1	<1	<1	<1	<1	<1	<1	<1–6	10,000[a] 1000[b] <1[c]	≤200 fcu fecal coliform/100 mL[e]
TSS (mg/L)	0	0–5	0	0–10	0	0–4	0–3	4–34	35[a] 35[b] 5[c]	1–15[d] ≤30[e]
$N\text{-}NH_4^+$ (mg/L)	<LOQ[f]	n.m.[g]	<LOQ[f]	32–56	<LOQ[f]	18	12–20	63–300		
TDS (mg/L)	2.5–17	493–2750	3.7–8.7	714–1294	4.1–12.3	823–1836	12.6–90.3	879–2830		
Bicarbonate (mg/L)	<LOQ[f]	268–415	<LOQ[f]	323–647	<LOQ[f]	543–836	<LOQ[f]	580–2044		
Proteins (mg/L)	n.m.[g]	n.m.[g]	<LOQ[f]	15.0–16.8	<LOQ[f]	6	<LOQ[f]	28–280		
Polysaccharides (mg/L)	n.m.[g]	n.m.[g]	<LOQ[f]	2.8–9.35	<LOQ[f]	4.5	<LOQ[f]	12–310		

[a] Process and cleaning water, except in the food industry.
[b] Process and cleaning waters for use in the food industry.
[c] Cooling towers & evaporative condensers.
[d] Boiler water, range is based on boiler operating pressure 0–140 bar.
[e] Once-through cooling and recirculating cooling towers.
[f] LOQ: Limit of quantification.
[g] n.m.: not measured.

enhancement will require higher recirculation rates with the penalty of lower productivities. Finally, the quality of the permeate product water is directly related to the salinity of the secondary effluent of the WWTP.

4. Costs analysis of the case-study

Next, we present the economic evaluation of the system designed to obtain reclaimed water. Capital Expenditures (CAPEX) and Operational Expenditures (OPEX) were estimated for this case-study formed by an UF and a RO units with a feed flowrate treatment capacity of 2.5 m^3/h (scenario 1). Two additional scenarios were also evaluated, considering feed flowrates of 5 m^3/h (Scenario 2) and 20 m^3/h (scenario 3). Costs have been estimated considering the total volume of the final product obtained in each scenario. These data are compiled in Table 5 for each scenario and each step, UF and RO, considering 8760 h/year of operation. Attending to the resulting product, two different water qualities can be achieved: *quality 1*, which corresponds to the UF permeate, that it is characterized as a water stream with turbidity values lower than 1 NTU and free of pathogens, but still with conductivity values similar to the effluent of the WWTP; and *quality 2*, which corresponds to the RO permeate, that it is characterized as a stream with low conductivity and very low organic load. Each stream meets the requirements for being industrially reused in cooling towers (*quality 1*) and in the production of steam in low and medium pressure boilers (*quality 2*) [29]. The operating conditions were taken from the case study, which considered RO feed pressure of 11 bar, and limited the UF and RO productivity to the viable ranges described in the previous section, 90% water recovery during UF and 60% water recovery during RO. CAPEX refers to the cost of equipment acquisition and installation until the plant starts operating, while OPEX refers to the operation cost, and it includes variable and fixed costs. The variable costs are dependent on the flowrate of reclaimed water obtained during the process [38,39]. Pumping energy and cost of reactants used for water conditioning and membrane cleaning are considered for the calculation of UF and RO variable costs. Fixed costs comprise personnel and maintenance costs.

Related to CAPEX, apart from the acquisition of membrane modules (UF modules: 2106 €/each; and RO: 205 €/each), the cost of instrumentation and automation systems, reagent dosing pumps and pre-filters must also be considered. Thus in Fig. 7, the evolution of the investment costs for an UF and a RO installation as a function of the membrane surface installed are presented. These functions plotted in Fig. 7 have been obtained by adjusting real costs of implementing membrane systems, provided by an engineering and consultancy company in Cantabria. These data are available as Supplementary Material. In both cases, the greater the installed membrane area, the greatest its contribution to the total cost of the installation. Eq. (10) and Eq. (11) define the calculation of installation cost of an UF and a RO plant, as a function of the membrane area (A), respectively. The obtained fittings are valid for the range of membranes applied, for UF membranes the validity range covers from 40 to 640 m^2 and for RO from 8 to 1113 m^2.

As it is shown in Fig. 7, UF installation costs are higher than those related to RO when compared for a given membrane area. Moreover, UF has also higher CAPEX in the three different scenarios detailed in Table 5. Membrane pretreatment requirements and lower productivity per membrane area due to higher fouling restrictions are the underlying reasons behind these results.

$$\textit{Installation Cost}, UF\ (€) = 5.175 \times A^{0.49}; R2 : 1.000 \tag{10}$$

$$\textit{Installation Cost}, RO\ (€) = 2.351 \times A^{0.52}; R2 : 0.973 \tag{11}$$

The estimation of pumping energy demands can be calculated as a function of the pressure and the feed flowrate, attending to data supplied by the pump manufacturers. In this case, the energy consumption of the UF pump is estimated to be around 0.44 kWh, for a feed flowrate of 2.5 m^3/h and a pressure of 2 bars, while an energy consumption of 1.7 kWh was considered for the RO operating at a pressure of 11 bar and a feed flowrate of 2.25 m^3/h.

Regarding the chemical reagents used for the chemically enhanced backwash cleaning of the membrane (CC), HCl was added for removing inorganic fouling, NaOH was needed for removing organic fouling and

Table 5
Detailed cost analysis for the 3 scenarios: feed flowrates 2.5; 5 and 20 m^3/h.

Process step	Costs			
	Cost type	Scenario I	Scenario II	Scenario III
UF	*Total membrane area* (m^2)	80	160	640
	CAPEX (€)	44,302	62,220	122,727
	OPEX (€/m^3)			
	Variable costs			
	Pumping energy consumption	0.022	0.022	0.022
	Chemicals	0.110	0.110	0.110
	Total variable costs **(€/m^3)**	**0.131**	**0.131**	**0.131**
	Fixed costs			
	Personnel	0.090	0.045	0.011
	Maintenance	0.034	0.024	0.012
	Total fixed costs **(€/m^3)**	**0.124**	**0.069**	**0.023**
	Amortization and financing			
	Fixed assets and interest	0.306	0.215	0.106
	Membranes (lifespan 4 years)	0.053	0.053	0.053
	Prefilter (lifespan 1 year)	0.015	0.008	0.002
	Total water produced (m^3/h)	2.25	4.5	18
	Total UF costs **(€/m^3)**	***0.630***	***0.476***	***0.316***
RO	*Total membrane area* (m^2)	15.8	31.6	126.4
	CAPEX (€)	9875	14,161	29,118
	OPEX (€/m^3)			
	Variable Costs			
	Pumping energy consumption	0.139	0.139	0.139
	Chemicals	0.040	0.040	0.040
	Total variable costs **(€/m^3)**	**0.178**	**0.178**	**0.178**
	Fixed Costs			
	Personnel	0.150	0.075	0.019
	Maintenance	0.013	0.009	0.005
	Total fixed costs **(€/m^3)**	**0.163**	**0.084**	**0.023**
	Amortization and financing			
	Fixed assets and interest	0.114	0.082	0.042
	Membranes (lifespan 4 years)	0.011	0.011	0.011
	Total water produced (m^3/h)	1.35	2.7	10.8
	Total RO costs (€/m^3)	0.470	0.358	0.256
	Total treatment cost (€/m^3)	1.096	0.831	0.570

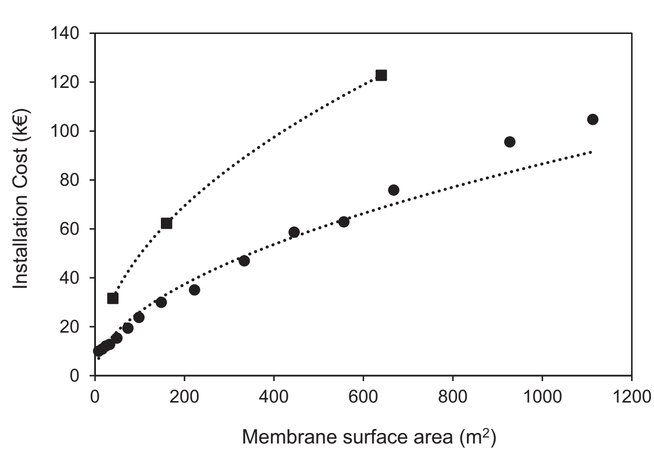

Fig. 7. CAPEX for the ● UF unit and ■ RO unit as a function of the installed membrane area.

NaOCl for disinfection purposes. The cleaning in place (CIP) is a more intensive cleaning and normally uses citric acid for both systems, UF and RO [26], although CIP was applied in much less frequency than CC.

Table 6 summarizes the chemical products and quantities required for the operation and cleaning steps of the UF and RO units, with their cost.

For the cost estimation, the next assumptions were made: i) CC frequency in the UF unit: 1 each 21 filtration cycles with a duration of 25 min; ii) CIP cleaning frequency of the UF unit: 1/month; iii) CIP cleaning frequency of the RO unit: 1/month.

Furthermore, fixed costs must be considered. For installations with a high level of automation and low maintenance costs, a personnel cost of 0.1 man/year is usually estimated. This cost has been considered for the simultaneous operation of UF and RO units, at a cost of 21 €/h, as the average manpower cost. The maintenance costs are calculated as 1.5% of investment costs, following a general rule of considering a percentage of the investment costs and used by other authors like Chen et al. [39]. A 12 year-period with an interest rate of 5.288% has been selected as fixed assets and interest, to calculate the amortization and financing costs. Finally, the lifespan of UF and RO membranes is assumed to be 4 years, and of 1 year for the filter.

A detailed cost estimation for each scenario is given in Table 5. An average electricity price of 0.11 €/kWh has been used considering the prices in Spain as recently reported [38]. Attending to the data presented in this table, OPEX contribution of UF is higher than RO for all scenarios proposed. The total cost for obtaining water with *quality 1* (UF permeate) can vary from 0.630 to 0.316 €/m^3, as the water produced increases from 2.25 to 18 m^3/h. The total cost of water with *quality 2* (RO permeate) can be reduced from 1.1 to 0.57 €/m^3, for water production capacities ranging from 1.35 to 10.8 m^3/h. Distance of the OPEX values between UF and RO becomes lower with scaled-up capacities, but an extra-cost close to 80% needs to be charged from *quality 1* (UF) to *quality 2* (RO) in all scenarios. As expected, pumping energy consumption is especially relevant in the cost of *quality 2* water. Since variable costs are not scale dependent, fixed cost influence on total cost is significantly reduced from scenario I to scenario III, especially due to personnel contribution. The automatization level of these plants allows to operate larger plants with similar manpower. From Table 5, it can be seen that fixed assets and interest is the most important component of CAPEX for small scale scenarios (38–35% of total treatment costs for scenarios I and II) while its scale-up dependence allows to reduce its contribution in larger scales (26% of total treatment costs in scenario III). Thus, this component of OPEX needs to get attention when costs calculation is performed.

These costs could vary notably depending on the place in which the treatment prototype is installed, because electricity, personnel and maintenance costs, among others, can vary significantly. For instance, Del Villar et al. [40] presented costs for reclaimed water obtained by using membrane filtration and RO of around 0.46 €/m^3. Corzo et al. [41] proposed the implementation of an FO-NF demonstration plant as a promising technology for wastewater reuse, finding a treatment cost of 0.96 €/m^3. Iglesias et al. [42] estimated the cost of the large scale UF-RO process at 0.35–0.45 €/m^3, which are values lower than those reported in this work, although the cited studies did not present an exhaustive costs breakdown. As Iglesias et al. [42] mentioned, costs were calculated from tenders, operation and maintenance costs only included personnel and routine analysis, while taxes and amortization were not considered. A cost estimation for an UF treatment after coagulation for the treatment

Table 6
Cost of chemical products and quantities required for the operation and cleaning steps of the UF and RO units.

Unit	Operating mode/Step	Reactant	Price	Consumption
UF	Filtration	$FeCl_3$ (40 wt%)	0.415 €/L	4.5 mL/m^3
	CIP	Citric acid	2.126 €/kg	2 kg/module
	CC	HCl (33%)	0.325 €/L	2.1 L/module
		NaOH (50%)	0.335 €/L	2.6 L/module
		NaOCl (160 g/L)	0.290 €/L	4.2 L/module
RO	Filtration/production	Anti-scaling	2,50 €/L	8 mL/m^3
	CIP	Citric acid	2.126 €/Kg	1 kg/module

of an old landfill leachate was performed by Nazia et al. [43], establishing a cost of 1.22 €/m^3. AzadiAghdam et al. [44] showed that fluidized bed crystallization combined with coagulation/flocculation with $FeCl_3$ was able to treat RO concentrates, in order to obtain a suitable stream for a second RO stage, in which they were able to produced reclaimed water with an estimated energy consumption of 5.5 kWh/m^3, including also an electrodialysis stage with bipolar membranes to produce acid and base streams. They also reported higher treatment costs for other technologies like electrodialysis, membrane distillation or brine concentrator. Sun et al. [3] compared the cost of reusing recycled irrigation runoff water with the cost of using municipally supplied water, observing lower values when reclaimed water was used. The same observation can be noted in this study if we take into account the average price of drinking water supply in Spain of 1.77 €/m^3 [45]. Therefore, water reclamation is competitive compared to the use of municipal water, leading to significant economic savings and, of course, with outstanding environmental benefits.

We have collected water consumption and cost data from nine companies dedicated to diverse industrial activities. A summary of water costs together with their annual consumption, the kind of water used and the main activity of each company is presented in Table 7. These data, as supplied by each company, correspond to current water prices (2020 and 2021 years) and consumptions. In general terms, when tap water is directly used as process water, the same rates are applied for all companies, and the supply cost depends on the volume consumed: 0.963 €/m^3 for water consumption lower than 100 m^3/year; 1.272 €/m^3 for water consumption between 101 and 1000 m^3/year, and 1.342 €/m^3 for water consumption higher than 1000 m^3/year. If water comes from other sources the prices vary widely.

Company 1, which presents the highest water consumption, uses two water supplies, tap water and clarified surface water. 260,000 m^3/year out of the total consumption are dedicated to feed the cooling towers, where clarified water is needed. There is no information available about the specific treatments applied for obtaining that clarified water, however, its cost rises to 7.5 €/m^3. *Company 1* needs of very high quality water to generate steam for turbine drive in thermoelectric power processes, which cannot be achieved with the proposed treatment. However, the company could also achieve important savings by combining UF/RO with the ion exchange process, as the proposed treatment would enlarge the capacity of the ultrapure water production system, at the same time the cost of regenerants and waste management would be largely reduced. A similar situation appears in *Companies 2 and 3*, that use clarified process water in boilers and condensers, with a cost around 3.1 €/m^3 and the treatments comprise sand filters and ion exchange treatments. For other processes in the factory, they also use demineralized water, at a cost of 1.0 €/m^3. Tap water is also used at the rates indicated above depending on the volumes consumed. Since the water quality obtained in this work achieves the most demanding quality levels required for being reused in boilers and in cooling towers, significant savings could be achieved, especially in view of the high volumes of water needed by these companies. Apart from tap water, *Company 4* produces 18 m^3/day of its own demineralized water by applying a RO treatment at a cost of 1.9 €/m^3, more than double the cost of producing *quality 2* water in scenario 3.

On the other hand, *Company 5* uses tap water for all their processes; however, for feeding the boilers they used preheated water coming from the condenser, with the aim of reducing the heating cost. *Company 6* also uses tap water in all their processes, including the water used in the cooling tower, with a minor 1.5% contribution to the total water consumption of the company. Although this company has the lowest water cost, the use of reclaimed water would be a better cost-effective choice. A comparable situation is observed for *Companies 7 and 8* where tap water is used at a higher cost than that of reclaimed water obtained in the present study.

Table 7
Cost of water paid by private companies with activity in chemical production, metallurgy and waste treatment.

Company	Main activity	Type of water	Total water consumption (m^3/year)	Water Cost (€/m^3)
1	Production of inorganic chemical products and thermoelectric power	Clarified water Tap water	24,267,164	7.5 1.34
2	Production of organic chemical products	Clarified water Tap water	893,454	3.1 1.34
3	Production of inorganic chemical products	Clarified water Demineralized water Tap water	320,076	3.1 1.0 1.3
4	Production of pharmaceutical ingredients	Demineralized water Tap water	151,800	1.9 1.34
5	Hazardous waste treatment facility	Tap water	6218	1.34
6	Coil rolling mill with chemical surface treatment	Tap water	6000	1.34
7	Manufacture of cooking appliances	Tap water	6000	2.0
8	Engineering and technology services	Tap water	100	0.963
9	Metallurgy investment casting	Tap water	14,206	1.33

5. Conclusions

A techno-economic assessment of an integrated system formed by UF and RO treatments for wastewater reclamation has been performed. The secondary effluent of a real WWTP was used as the feed of the UF-RO in the demonstration activities performed on site. During UF operation the main variables evaluated were the time of filtration and backwash cycles and the addition (or not) of coagulant, and their influence on the water recovery. During RO operation, the recirculation rate of the concentrate stream and the quality of the UF permeate appeared as the main process variables affecting the quality of the permeate. For all variables studied, produced water met the most demanding quality levels for being industrially reused, including the absence of bacteria needed for feeding cooling towers. In addition, an economic estimation for three scenarios corresponding to three different feed flowrates (2.5; 5 and 20 m^3/h) was developed. Higher CAPEX and OPEX were obtained for UF operation than for RO operation. This fact can be attributed to the membrane pretreatment requirements and fouling issues. However, as expected, lower prices were obtained when higher flowrates were applied. Comparing the estimated cost values obtained from the economic assessment with the prices that several industrial companies are currently paying for water, it can be established that important savings could be achieved if companies used their own wastewaters after an adequate treatment, like the proposed in the present work where water of a high quality can be obtained; besides it is worth highlighting the significant environmental benefits that could also be achieved substituting the use of natural water sources by properly reclaimed water.

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CRediT authorship contribution statement

Gema Pérez: Writing – original draft. **Pedro Gómez:** Methodology, Resources. **Inmaculada Ortiz:** Data curation, Writing – review & editing, Supervision, Funding acquisition. **Ane Urtiaga:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.desal.2021.115409.

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