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Pilot-scale multi-stage reverse osmosis (DT-RO) for water recovery from landfill leachate

D. Cingolani [a,*], F. Fatone [a], N. Frison [b], M. Spinelli [a], A.L. Eusebi [a]

[a] Department of Science and Engineering of Materials, Environment and Urban Planning (SIMAU), Università Politecnica delle Marche, Via Brecce Bianche, 12, Ancona, Italy
[b] Department of Biotechnology, University of Verona, Via Le Grazie 15, 37134 Verona, Italy

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ABSTRACT

Recovery of high quality water from municipal landfill leachate was studied by three-stage disc tube reverse osmosis optimized in pilot-scale. Following UF-membrane-assisted activated sludge plant, overall 46.5 tons of leachate were post-treated in real environment and analyzed for conventional contaminants and hazardous compounds (e.g. heavy metals, boron, selenium) throughout operation of membrane system.

Operating pressure ranged from 21 to 76 bar, while permeate flux varied in the range 7.1–32.5 L m^{-2} h^{-1}. Rejection factors of specific ions were related to the pressure and global removals were assessed for each stage (e.g. $E\%_{COD}$ = 92.4–99.2%, $E\%_{NH4}$ = 46.2–95.8%, $E\%_{NOx}$ = 84.8–97.9%; $E\%_{TDS}$ = 88–95.5%). Boron removal was assessed in the range 34–48%, so as to require the third stage to reach standard for discharge or reuse. Two stages were sufficient to reach water recovery higher than 91%. Long-term operation and mathematical modeling demonstrated how the $\Delta\pi/\Delta P$ ratio can support the decisions for membrane cleaning and predictive maintenance: permeability decline was associated to the ratio increase from 0.72 to 0.73 to 1.13–1.21.

1. Introduction

Landfilling is still a major issue of the municipal solid waste management system in Europe and, even more, around the world. In 2015, 61 Mtons of municipal solid waste (MSW) have been landfilled in Europe while the generated leachate may be estimated between 12.2 and 61 Mtons (Brennan et al., 2016; Eurostat, 2015). This residual must be appropriately treated and managed, maximizing the recovery and minimizing the waste disposals. In particular, standalone on-site treatments are more and more attractive to cope with the changing and variable characteristics of leachate (Brennan et al., 2016). In this context, the use of membrane technologies allows stable quality of the permeate that can be locally reused or discharged in water bodies (Hasar et al., 2009). In particular, Reverse Osmosis (RO), either as a main step in a landfill leachate treatment chain or as single post-treatment step has shown to be an indispensable means of achieving high purification, removal of hazardous metals and potential water recovery (Ahmed and Lan, 2012; Renou et al., 2008). However, the specific energy consumption (SEC) of RO is much higher than other treatments. Judd (2017) highlighted that multi-stage RO improves water recovery and reduces SEC when less than five stages are used.

Feasibility and sustainability of RO system depends on the brine disposal. One of the most viable and practiced way is the reinjection (or recirculation) of the brine into the landfill. Generally, 30% of the volume of the raw leachate is returned to the landfill as concentrated stream (Li et al., 2009). Few scientific papers debate on this topic and opinions to such practices are conflicting (Calabrò et al., 2010; Peters, 1998). This operation will always return salinity and contaminants into the landfill resulting in increasing in osmotic pressure for leachate separation and higher SEC for the RO plant.

By avoiding the recirculation of the concentrate in the landfill, the viability of the on-site treatment of the leachate and discharge into the surface water bodies require a high energy consumption, up to an expensive of 30 €/m^3 of influent flow.

In desalination processes, costs related to thermal treatment and brine disposal may rise until 33% of the total cost of desalination (Pérez-González et al., 2012). Thus, the cost-related to the concentrate disposal in the separation of the landfill leachate might be much higher.

* Corresponding author.
E-mail address: d.cingolani@univpm.it (D. Cingolani).

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Nomenclature

μ	dynamic viscosity
A	membrane permeability
ABS	acrylonitrile butadiene styrene
COD	chemical oxygen demand
CP	concentration polarization
CV	coefficient of variation
DT-RO	disc tube reverse osmosis
E%	efficiency removal
ERS	energy recovery system
FP	booster pump
J_w	permeate flux
k	electrical conductivity
MBR	membrane biological reactor
MF	micro filter
MSW	municipal solid waste
MWWTP	municipal wastewater treatment plant
PA	polyamide
PLC	programmable logic controller
P_{max}	set-point of maximum pressure
PP	piston pump
Q_c	concentrate flow rate
Q_p	permeate flow rate
R	rejection factor
RO	reverse osmosis
RO1	RO stage one
RO2	RO stage two
RO3	RO stage three
RR%	recovery rate
SEC	specific energy consumption
SW	spiral wound
T	temperature
TDS	total dissolved solids
TFC	thin film composite
TKN	Kjeldahl nitrogen
UF	ultrafiltration
VC	concentrate spillage valve
WWTP	wastewater treatment plant
ΔP	operative pressure differential
$\Delta\pi$	osmotic pressure differential
π	osmotic pressure

Since the feasibility and sustainability of RO is usually limited by the disposal of the concentrate, its minimization is the first strategy to make the treatment sustainable.

The performances of RO can be optimized coupling two or more stages. This configuration minimizes the concentrate and maximizes the water recovery (Joo and Tansel, 2015; Subramani and Jacangelo, 2014), being able to achieve high removal of persistent anion like boron (Hilal et al., 2011).

However, although hybrid or conventional technologies have been developed trying to be economically attractive (Cingolani et al., 2017; Mukherjee et al., 2015), the technical and economic sustainability of RO multi-stage scheme is limited by membrane fouling (Bourgeous et al., 2001; Zhao et al., 2013). Disc tube RO (DT-RO) technology has widely been proposed for on-site landfill leachate, particularly for high suspended solid matrices (Gong et al., 2013; Hasar et al., 2009; Insel et al., 2013; Smol and Włodarczyk-Makuła, 2016; Zhang et al., 2013). Compared to the conventional spiral wound (SW), tubular or hollow fibre modules, the "plate-and-frame" configuration of the DT module and the shorter flow path (≈7.5 cm) guarantees higher turbulence and limits the concentration polarization (CP) effects along the surface of the membrane (Peters, 2001; Singh, 2015; Subramani and Jacangelo, 2014). Nevertheless, concentrate production and fouling rates in real environment are still gaps of knowledge.

Therefore, the paper demonstrates how the optimization of water recovery and removal of nitrate, boron and selenium from pre-treated landfill leachate, up to reuse or discharge quality standard, need a triple-stage DT-RO scheme.

The plant was then studied in terms of operational viability and to compare performances with single stage RO, that was able to achieve the required effluent standard.

Three RO stages were studied to maximize the water recovery in the first two (Alghoul et al., 2009), while the third was investigated to achieve high quality permeate. As the pilot plant was installed in full-scale field, attention was paid to the relevant, unpredictable and sudden variability of influent that can drastically influence permeability, recovery rates and permeate quality.

Finally, removal of persistent ions such as boron and selenium was investigated to define suitable configuration to achieve standard for reuse or discharge in sensitive water bodies.

2. Material and methods

The multi-stage RO was operated for three relevant months to treat the effluent of a full-scale plant (Marche Region, Italy) that is treating municipal landfill leachate with treatment capacity of 300 m^3/d. Before the RO, hereby focused, the leachate is pretreated by clari-flocculation, activated sludge with intermittent aeration and tertiary membrane ultrafiltration (Eusebi et al., 2009). The plant has already been monitored for one year by defining the characteristics of influent and effluent: results have been published in a previous work (Cingolani et al., 2017).

The raw leachate is originated from two nearby MSW landfills serving a basin of 460,000 inhabitants in Marche Region (central Italy). The overall treatment capacity of 115,800 ton of MSW per year is divided between a 44-ha site operating since 1989 and a 11.4-ha operating since 1999 (ARPAM, 2016).

2.1. The DT-RO pilot plant

The RO pilot-scale plant was equipped with DT modules (Fig. 1a). The membranes (Gel GPT-BW 30) were installed into the stainless-steel vessel of 1.2 m length (Fig. 1a). Supporting discs were made in acrylonitrile butadiene styrene (ABS) with an outer diameter of 197 mm. Every overlapped disc (Fig. 1a) contains two films of polyamide (PA) thin-film-composite (TFC) with the following specifications: NaCl rejection >98%, max pressure 120 bar, max temperature 40 °C, pH operating 3–11, free chlorine tolerance <0.1 ppm. The whole membrane area was of 7.7 m^2.

Inlet feeding was provided through one booster (FP) and one piston (PP) pump (Fig. 1b).

The feeding inter-crosses the disc package from the bottom to the top. The pressure-driven process directs the permeate towards the central channel of the vessel (Fig. 1a), besides the concentrate continuously crosses the package to be sent to the feed outlet pipe.

Chemical conditioning of the feeding was provided by the dosage of sulfuric acid (30% w/w) through the control of pH-meter (Georg Fischer, electrode model 3-2724-01).

Electrical conductivity (k) measurements were performed both for feeding and permeate streams (EMEC, models ECDC/1 and ECDCC/10), the temperature was monitored in the feeding pipe

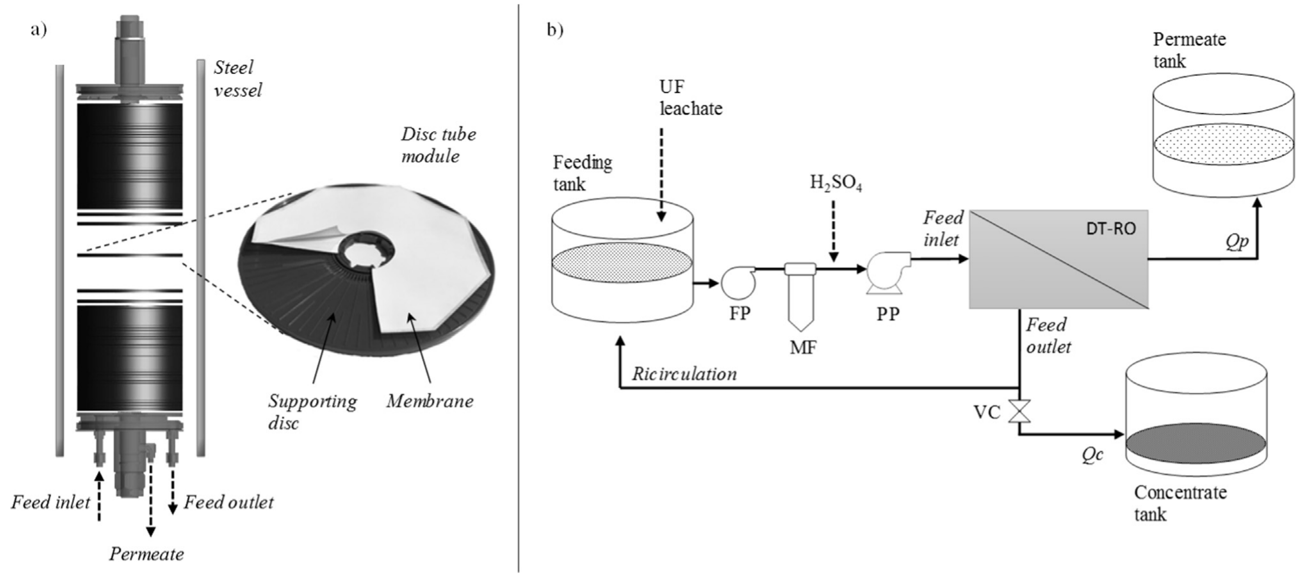

Fig. 1. DT-RO system (a) and pilot plant flow scheme in RO1 configuration (b).

(IFM model TA2435). Inlet and outlet pressures were continuously monitored (Siemens SITRANS P220) and flow rates were measured for the permeate (Q_p) and the concentrate (Q_c) (IFM model SM6100).

2.2. Reverse osmosis plant and operative process parameters

The RO was organized in consecutive phases (Fig. 2). RO1 treated the effluent from the full-scale membrane bioreactor. The concentrate from RO1 was reduced by RO2 stage at high pressure and the last refinement stage RO3 treated the mixed permeate from RO1 and RO2. Table 1 shows the operating values for the permeate flow rate (Q_p) in each stage.

According to Hasar et al. (2009) and Linde et al. (1995), permeate flux (J_w) was set to 13 L m^{-2} h^{-1} in the first RO1. Due to the concentration of the inlet, J_w was decreased up to 7.1 L m^{-2} h^{-1} in the RO2. Finally, J_w was set to 32.5 L m^{-2} h^{-1} in the refinement stage RO3.

With the purpose to maintain the recovery rate (RR%) stable, concentrate production (Q_c) was consequently managed by recirculating in the feeding tank or spilling (Fig. 1b).

The programmable logic controller (PLC) automatically adjusted the operating pressure to keep the permeate flux constant until the maximum set pressure value (P_{max} – Table 1). J_w of 2 L m^{-2} h^{-1} was chosen as setpoint to stop the test.

Sulfuric acid was dosed to maintain the pH of 6.5 in the RO1 trials. All set of runs were performed in the range 29.1–37.9 °C.

2.3. Analytical methods and process model

Membrane fouling phenomena can be investigated by monitoring the water permeability (Kim and Hoek, 2005). To monitor the decline of membrane permeability the following assumptions were made: (1) the membrane package was considered as a single layer, (2) ΔP is averaged between inlet and outlet, (3) $\Delta\pi$ is assumed as bulk osmotic pressure differential.

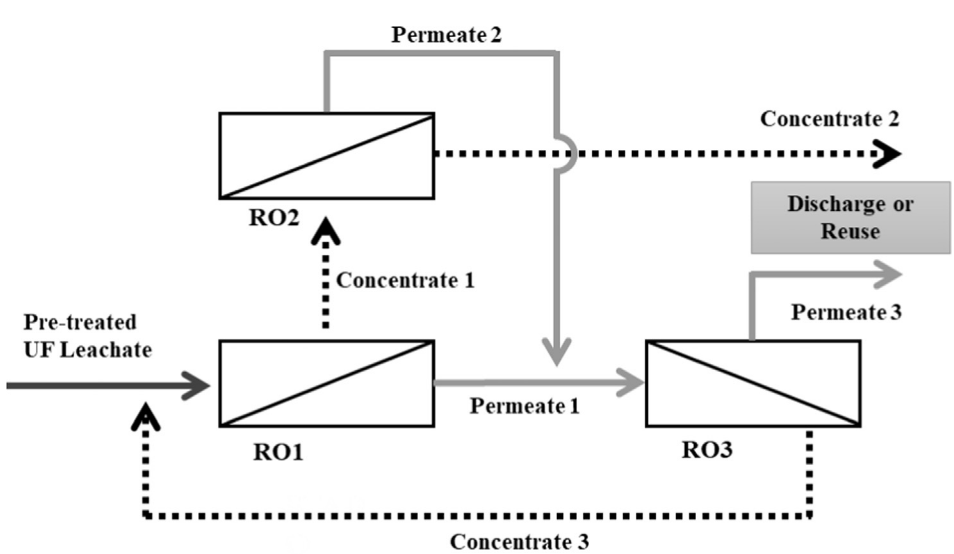

Fig. 2. Scheme of the multi-stage treatment by RO membranes.

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Table 1
Operative parameters of permeate flow rate (Q_p) and flux (J_w), maximum pressure (P_{max}) and temperature.

RO stage	T (°C)	Q_p ($L\,h^{-1}$)	J_w ($L\,m^{-2}\,h^{-1}$)	P_{max} (bar)	Run
RO1	34–36	100	13.0	60–67	A, B, C
RO2	29–37	55	7.1	97	A, B, C
RO3	29–31	250	32.5	60	A, B

The permeability was normalized at 25 °C according to Eq. (1) (Sassi and Mujtaba, 2012), where μ is the dynamic viscosity of feeding:

$$A^T = A^{T_0}\frac{\mu^{T_0}}{\mu^T} \quad (1)$$

During the experimental phase the pilot plant was operated 24 h per day. Samples of pre-treated ultra-filtered leachate (UF leachate) were daily collected. Differently, samples of the ROs streams (feeding, permeate and concentrate) were collected 3 times per day every 4 h separately for RO1, RO2 and RO3, then stored at 4 °C and analysed within 12–24 h. Analytical study was executed for the main conventional pollutants (COD, ammonia and TKN) according to standard methods (APHA, 2005). pH was measured using a Metrohm 848 titrator and alkalinity was determined through titration via chloridric acid. Electrical conductivity (k) was performed by an XS Cond 70. Ion concentrations (NO_2^-, NO_3^-, $PO_4^=$, Cl^-, $SO_4^=$, Na^+, K^+, Mg^{++}, and Ca^{++}) were measured using ion chromatograph (IC) (Dionex, DX-120 equipped with IonPac AS9-HC column, ICS-1000 equipped with IonPac CS12A column).

Total metals and non-metals concentrations were determined by inductively coupled plasma mass spectrometry (ICP-MS) (Perkin Elmer model Optima 8300). ICP-MS analysis was performed on composite samples only on the streams of UF leachate, permeate of RO1, permeate of RO3 and residual concentrate from RO2 previously acidified to a pH of less than 2 with nitric acid.

The osmotic pressure was monitored by relating k and π: online values of π and temperature were used to predict the permeability of the membrane package in each stage.

Leachate strength and process performances were mainly evaluated by the removal of salt content. Rejection factors (R) were calculated according to Eq. (2).

$$R(\%) = \left(1 - \frac{C_p}{Ci}\right) \times 100 \quad (2)$$

where C_p (mg/L) and C_i (mg/L) are the ion concentrations in the permeate and the corresponding feeding streams, respectively.

On the other hand, the removal efficiency (E) was calculated on mass balances basis.

Recovery rate (RR%) was calculated as the ratio of the volume permeated with respect to the volume treated. Membrane cleaning was carried out at the beginning of each stage by alternating water flushing and chemical washings. Acid (citric acid 2% w/w, lactic acid 0.5% w/w, dodecylbenzenesulfonic acid 0.5% w/w) and basic cleaners (KOH 4% w/w and Na_4EDTA 0.5% w/w) were used.

3. Results and discussion

3.1. Operation of RO1

The chemical and physical characterization of the UF leachate is reported in Table 2. Feeding in the RO1 stages had a considerable amount of non-biodegradable organic carbon and high salinity, mainly related to chlorides and sodium. Ammonia was in the range 35 ± 46 mgNH_4-N/L thanks to the secondary biological treatment. Differently, NO_x-N in the range 398 ± 282 mgN/L were related to scarce biological denitrification.

Relevant variability of the UF leachate was observed along the experimental period that included both dry and wet seasons.

The management of recycles and retentate in a multiple stage RO is a key strategy often neglected in lab scale experiments. In this full-scale field study, the monitored parameters of $RO1_C$ run are shown in Fig. 3 where Q_c was null at the beginning of the test. Pressure-driven process was continuously concentrating the feeding due to the recirculation stream (Fig. 1b), so the electrical conductivity of the inlet increased from 12 mS/cm up to 45 mS/cm. The concentrate flow rate (Q_c) was pumped out from hour 45 onwards in order to stabilize electrical conductivity in the influent. Therefore, k was manually adjusted at 35 mS/cm.

When the pressure set point (P_{max}) was reached, Q_p began to fluctuate from 80 to 100 $L\,h^{-1}$ due to the concentrate flow adjustments and the changes of k in the feed.

After 100 h, fouling led the permeate flux decline from 13 to 6.8 $L\,m^{-2}\,h^{-1}$ (corresponding to Q_p decline from 100 to 52.5 $L\,h^{-1}$,

Table 2
Results of characterization from 20 samples of UF leachate (RO1 feeding) over three months of monitoring and quality of the permeate of each RO stage.

Parameters	u.m.	UF leachate		RO1 permeate	RO2 permeate	RO3 permeate
			CV			
k (25 °C)	mS/cm	8.67 ± 1.97	23%	1.1 ± 0.4	4.4 ± 1.0	0.058 ± 0.033
pH	–	7.2 ± 0.4	6%	5.8 ± 0.1	6.7 ± 0.6	6.0 ± 0.5
Alk	mg$CaCO_3$/L	699 ± 437	63%	44.1 ± 4.0	88 ± 30	11.3 ± 7.9
COD	mg/L	1368 ± 422	31%	82 ± 65	155 ± 139	11.5 ± 6.2
NH_4-N	mg/L	35 ± 46	131%	13.0 ± 9.4	31 ± 26	5.5 ± 0.5
TKN	mg/L	104 ± 85	82%	35 ± 25	42 ± 51	5.9 ± 1.0
NO_2-N	mg/L	206 ± 147	71%	39 ± 27	91 ± 79	2.0 ± 1.5
NO_3-N	mg/L	192 ± 135	70%	90 ± 112	115 ± 93	2.8 ± 2.7
PO_4-P	mg/L	7.5 ± 3.3	44%	0.6 ± 0.7	0.6 ± 0.7	0.1 ± 0.2
Cl	mg/L	1925 ± 426	22%	340 ± 260	631 ± 36	5.3 ± 0.4
SO_4	mg/L	133 ± 46	35%	27 ± 27	47 ± 17	0.3 ± 0.1
Na	mg/L	1562 ± 254	16%	266 ± 178	515 ± 100	9.9 ± 2.3
K	mg/L	556 ± 76	14%	118 ± 82	214 ± 31	4.4 ± 2.4
Mg	mg/L	96 ± 20	21%	6.1 ± 5.3	14.7 ± 2.5	0.5 ± 0.3
Ca	mg/L	161 ± 25	16%	13.5 ± 14.8	25.8 ± 4.2	5.7 ± 0.4

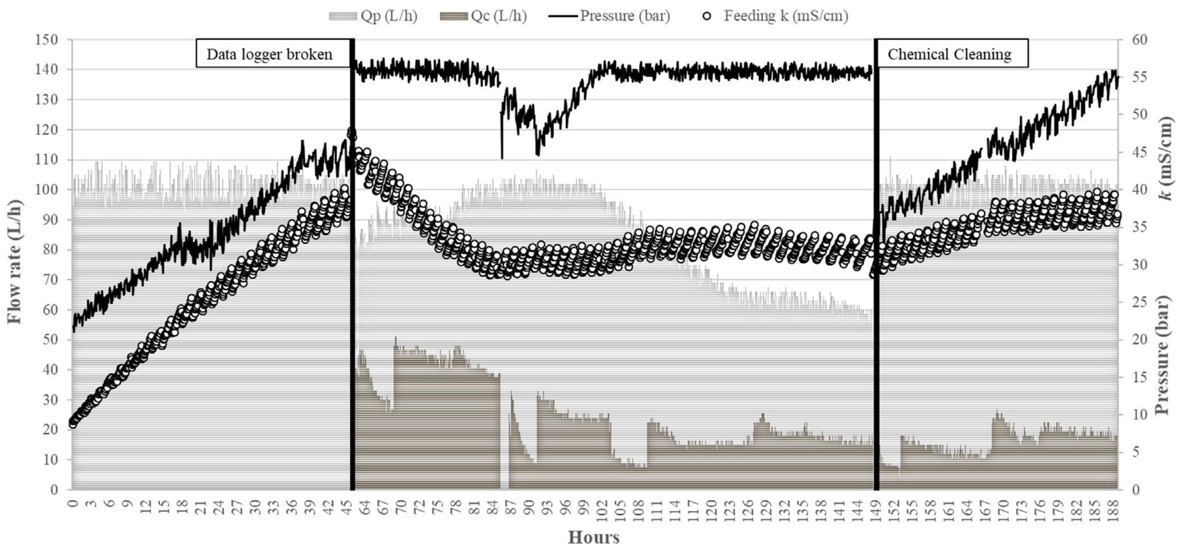


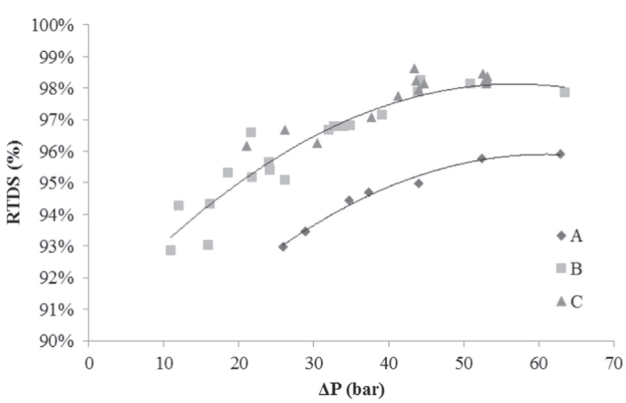
Fig. 3. $RO1_C$ run: flow rates of permeate (Qp) and concentrate (Qc), operative pressure and electrical conductivity (k) of the feeding.

Fig. 3). Then, the permeability A^{T0} was recovered at 1.13 L m^{-2} h^{-1} bar^{-1} thanks to chemical cleaning.

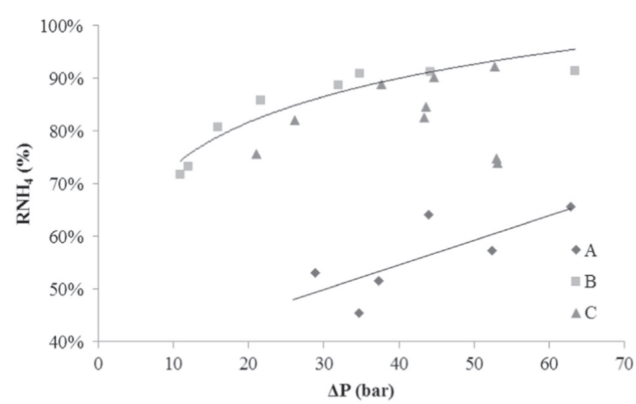
TFC-PA membranes from the biggest manufacturers in the world have a normalized water permeability from 0.8 to 8 (Lee et al., 2011) depending on salt rejections (NaCl) between 98% and 99.8%. The actual permeability makes the membrane falling within the category of high rejections and less production of permeate.

When the inlet pressure in RO1 runs ranged between 20 and 67 bar, the mean working conditions were of 50.5, 33.4 and 47.7 bar for $RO1_A$, $RO1_B$ and $RO1_C$, respectively. Major effect on the overall pressure drop was found by the hydraulics of the membrane module. On the other hand, membrane fouling did influence the permeate flowrate.

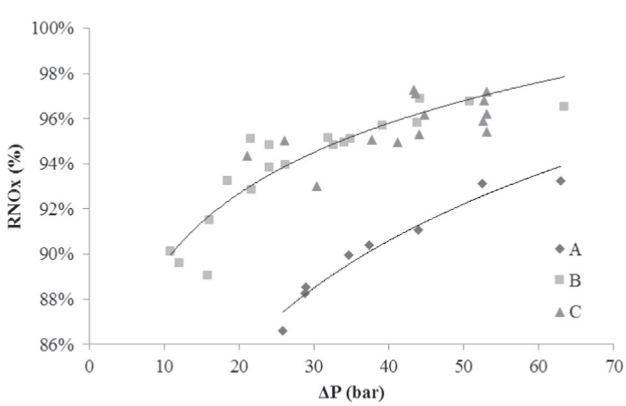
Fig. 4 reports rejection of conventional pollutants in RO1: remarkably, ΔP influenced mainly the rejection of ammonia and TDS. In run $RO1_A$, rejections of TDS (93–96%, Fig. 4a) and ammonia (27.4–65.5%, Fig. 4b) increased from ΔP of 26 to 63 bar. Better rejection trends were found for the B and C runs, where R_{NH4}



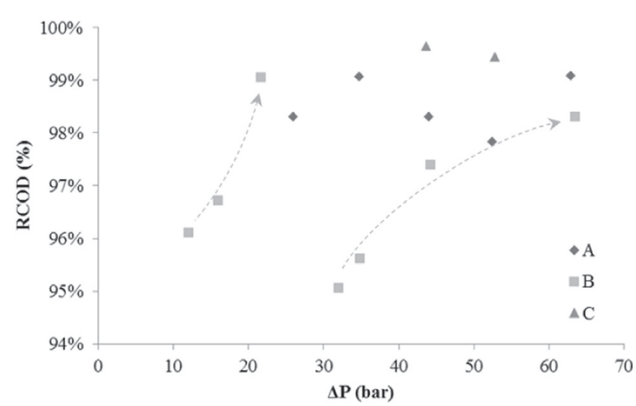
-a Rejection factor for TDS



-b Rejection factor for NH_4



-c Rejection factor for NO_x



-d Rejection factor for COD

Fig. 4. Rejection factors for TDS, NH_4, NOx and COD observed in RO1 runs (A, B and C).

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was between 73% and 91% and ΔP ranged from 10.9 to 63.4 bar. Similar trend was observed for NO_x-N (Fig. 4c).

No clear correlation between ΔP and COD rejection was found (Fig. 4d), but two tendencies can be clearly seen in the run B: both increase COD rejection as a function of pressure. Since the temperature was of 34.3 ± 2.4 °C, 35.6 ± 1.8 °C, 35.5 ± 2.6 °C respectively for A, B and C runs, its effect on COD rejection has been excluded. The behavior has been related to an intermediate stop and restart of the run, for approximately 2 days of break, where a rinsing with water was executed.

COD removal between 95% and 99% was observed as reported also by other authors (Kuusik et al., 2014; Talalaj, 2015) as well as rejection factors related to pressure drops (Singh, 2015). However, increase of rejection rates were observed from run A to run B and C and were associated to the irreversible fouling and water permeability decline (Bellona et al., 2004).

3.2. Operation of RO2 and RO3

The conductivity in the feed of RO2 stages was 18.6 mS/cm in run A, 60.3 mS/cm in B and 36.9 mS/cm in C. These values are indirect indicators of COD and TDS that were: 2335 mgCOD/L and 15.6 gTDS/L for $RO2_A$; 13,512 mgCOD/L and 57.3 gTDS/L for $RO2_B$; 7352 mg/L mgCOD/L and 34.5 gTDS/L for $RO2_C$.

Contrary to the RO1 stages, the high pressure (RO2) and refinement (RO3) ones were executed without withdrawing the concentrate (Fig. 1b). This approach led to the continuous increase of feeding π and operative pressure that influenced the fouling rate.

In the $RO2_A$, inlet pressure increased from 15 to 35 bar and the permeate flux was kept to 6.4 L m^{-2} h^{-1}. Differently, in the cases of $RO2_{B-C}$ the starting pressures were of 60 and 40 bar respectively, due to higher salinity of feeding. After that pressure raised up to P_{max}, J_w was rapidly decreasing from 6.4 to 2 L m^{-2} h^{-1} until the runs were stopped. Average values of inlet pressure are reported in Table 3. The lower pressure in $RO2_A$ than $RO2_{B-C}$ was linked both to the low salinity and to the set operative flux of permeate (half to the RO1 one).

Rejection factors into the RO2 stages were found as function of pressure likely the RO1 ones, R_{TDS} changed from 88% to 94% at 16 and 30 bar respectively in the $RO2_A$ case, while it was from 95% to 98% at 40 bar and 97 bar in the others $RO2_{B-C}$ runs.

Besides, worsening rejection of TDS was noticed when the permeate flux was decreasing. R_{TDS} of 92% were recorded at the end of $RO2_C$ due to highest $\Delta\pi$. Rise of salt passage was related to an increase of CP to the permeate side due to flux decline (Agenson and Urase, 2007; Wijmans and Baker, 1995).

Combination of permeates originated from RO1 and RO2 stages were characterized by 53 ± 26 mg COD/L, 13.2 ± 6.7 mg NH_4-N/L, 94 ± 60 mg NO_x-N/L, 203 ± 81 mgCl^-/L and 206 ± 49 mgNa^+/L. Accordingly, the electrical conductivity of feeding in RO3 runs was of 1.28 and 1.69 mS/cm respectively for A and B series.

During the RO3 stages, the assigned permeate flow rate of 250 L h^{-1} was kept constant over all the testing period. $RO3_A$ continuously worked at 23.4 ± 1.2 bar of inlet pressure, while $RO3_B$ started from 40 bar up to reach 63 bar (average pressure of 41.8 ± 4.5 bar). Both RO3 runs were performed until achieving the feeding concentration factor up to 10 times.

Inconsistently operative pressures among RO3 stages have been linked to membrane condition, as stated by permeability that decreased from 1.5 to 0.8 L m^{-2} h^{-1} bar^{-1}.

3.3. Recovery rates and permeate quality

RO1 and RO2 played a major role in the water recovery (Fig. 2). However, the recovery was relatively high (>90%) even in the third stage.

Depending on the quality of the UF leachate, recovery rates (RR %) ranged from 66% to 87% in the three RO1 runs, where the feeding k varied between 11.1 ± 0.7 mS/cm and 6.2 ± 0.9 mS/cm.

Differently, the RR% in RO2 increased from 34% (Feeding k = 60.3 mS/cm) up to 72% (Feeding k = 18.6 mS/cm), confirming the relation between the recovery rate and the initial electrical conductivity. Therefore, global RR% of the multi-stage filtration was observed in a range of 91–95%.

Table 3
Removal efficiencies of the main macropollutants and relative pressure for each RO stage.

Run	Average P (bar)	E_{COD} (%)	E_{NH4-N} (%)	E_{TKN} (%)	E_{NOx} (%)	E_{TDS} (%)
$RO1_{A-B-C}$	33–50	95.9 ± 3.0	60.9 ± 13.4	69.4 ± 5.8	86.4 ± 1.9	91.1 ± 1.5
$RO2_A$	21	98.7	86.7 ± 12.9	88.4	84.7	88.9
$RO2_{B-C}$	67–76			94.8	95.4 ± 3.7	97.7 ± 1.1

Table 4
Hazardous compounds concentration in feeding and permeate of RO1, permeate of RO3 and concentrate of RO2.

	UF leachate		RO1 permeate		RO3 permeate		RO2 concentrate	
(mg/L)	Ave.	St.Dev.	Ave.	St.Dev.	Ave.	St.Dev.	Ave.	St.Dev.
Al	**0.319**	*0.326*	**0.005**	*0.002*	**0.008**	*0.009*	**0.925**	*0.307*
Sb	**0.017**	*0.006*	**0.000**	*0.000*	**0.000**	*0.000*	**0.098**	*0.025*
As	**0.074**	*0.023*	**0.002**	*0.001*	**0.001**	*0.000*	**0.521**	*0.166*
Ba	**0.102**	*0.020*	**0.017**	*0.005*	**0.008**	*0.001*	**0.566**	*0.151*
B	**5.688**	*2.549*	**3.748**	*0.657*	**1.950**	*0.841*	**16.256**	*6.524*
Cd	**0.001**	*0.000*	**0.000**	*0.000*	**0.002**	*0.003*	**0.003**	*0.001*
Cr	**0.514**	*0.175*	**0.004**	*0.001*	**0.002**	*0.000*	**6.129**	*2.906*
Fe	**2.272**	*1.265*	**0.150**	*0.058*	**0.169**	*0.059*	**13.610**	*5.675*
Mn	**0.230**	*0.175*	**0.019**	*0.016*	**0.004**	*0.001*	**1.987**	*1.379*
Hg	**0.002**	*0.001*	**0.001**	*0.002*	**0.000**	*0.000*	**0.003**	*0.001*
Ni	**0.420**	*0.164*	**0.002**	*0.000*	**0.001**	*0.001*	**3.422**	*1.115*
Pb	**0.012**	*0.005*	**0.001**	*0.000*	**0.000**	*0.000*	**0.044**	*0.004*
Cu	**0.119**	*0.046*	**0.007**	*0.003*	**0.003**	*0.001*	**0.908**	*0.189*
Se	**0.063**	*0.036*	**0.004**	*0.002*	**0.000**	*0.000*	**0.471**	*0.204*
Sn	**0.026**	*0.007*	**0.001**	*0.000*	**0.001**	*0.000*	**0.125**	*0.040*
V	**0.131**	*0.046*	**0.003**	*0.001*	**0.001**	*0.000*	**1.004**	*0.388*
Zn	**0.680**	*0.183*	**0.199**	*0.066*	**0.164**	*0.044*	**3.214**	*0.189*

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Few literature papers report water recovery data for similar applications on landfill leachate, as they commonly refer to the brackish water (BW) desalination (with TDS concentration between 1000 and 15,000 mg/L). The design practice usually leads to a global recovery of 82%: 64% in the first stage and 50% in the second one (Alghoul et al., 2009).

Altaee and Hilal (2015) proposed a hybrid multi-stage membrane treatment (NF-FO/BWRO) to produce fresh water for humans and agriculture. NF recovery was from 50% to 75% at feed salinity from 1 g/L to 2.4 g/L, while FO-BWRO could recover only 18%. Linde et al. (1995) reported recovery between 51 and 71% for the first RO stage in landfill leachate desalination. Where feeding k was in the range 1.5–2.5 mS/cm, pressure was assessed of 30–40 bar.

The quality of permeate from each RO stage is shown in Table 2, while heavy metal concentrations of the main streams are reported in Table 4. Except for NO_2-N and NO_3-N, RO1 allowed to reach the standard for discharge in sensitive water bodies. However, water recovery can be optimized to more than 90% only by following RO2 and RO3 stages.

Rejection factors are influenced by the inlet pressure as well as removal efficiencies (Table 3).

Metals concentrations reached standard for discharge already after RO1. Selenium removal was higher than 94% while boron removal was assessed in the range 34–48%. As result, the full three-stage configuration was necessary to decrease boron in the permeate below 2 mg/L and achieve the standard for reuse. However, it must be noticed the high B concentration in the concentrate that must be incinerated or crystallized, leading to high overall treatment costs.

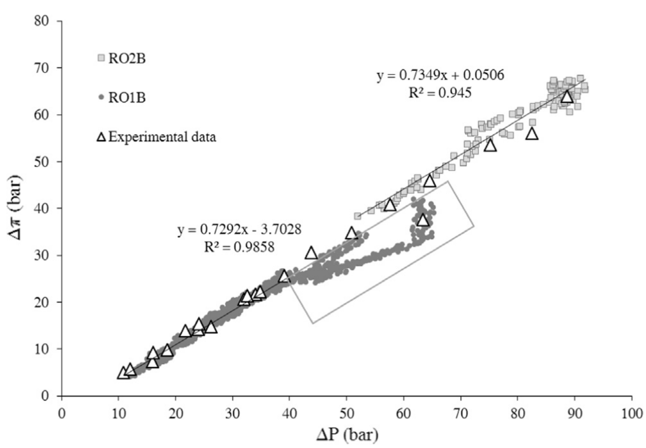

Fig. 5. Osmotic pressure differential ($\Delta\pi$) and operative pressure (ΔP) relationship: experimental and modeling data in $RO1_B$ and $RO2_B$ runs.

Al, Fe and Cd concentrations in the permeate of RO3 were higher than RO1. As the refinement RO3 treated also the permeate of RO2 (Fig. 2), the increase in RO3 effluent could be related to higher influent loading. However, no analytical evidence can support this comment.

These results are comparable with Smol and Włodarczyk-Makuła (2016) that studied an integrated system of coagulation-NF/RO. Lower performance on COD removal (59%) was reported by Ahn et al. (2002) by using SWRO PA membranes (Filmtec) to study an MBR-RO configuration to treat landfill leachate in full-scale.

3.4. Modeling membrane permeability and fouling rates

The model was calibrated to obtain π values and was used to evaluate Δπ that were determined according to the following quadratic equation.

$$\pi(T) = (a(T) \cdot k^T + b(T))^2 \tag{3}$$

where k^T is the electrical conductivity at the operating temperature, $a(T)$ and $b(T)$ are the experimental coefficients corresponding to the specific temperature. The calibration of the model is reported in Fig. 5. The fouling effect on permeability in the RO1 trials can be recognized in Fig. 6.

In run $RO1_B$, the initial permeability was 1.4 L m^{-2} h^{-1} bar^{-1}, after 150 h without intermediate cleanings it dropped down to 0.39 L m^{-2} h^{-1} bar^{-1}. At the same test conditions, in the $RO1_C$ the permeability of 0.4 L m^{-2} h^{-1} bar^{-1} was reached in about 75 h from the start of the test and the initial permeability was lower than 1 L m^{-2} h^{-1} bar^{-1}. Notwithstanding the manufacturer's recommended chemical cleanings, the initial permeability (A) was not recovered and the fouling rate was higher (15.6×10^{-3} vs 6.5×10^{-3} L m^{-2} h^{-2} bar^{-1}) (Fig. 6). This demonstrates the need of stronger and intermediate chemical cleaning.

In a full-scale DT-RO application treating raw leachate (Liu et al., 2008), alternating alkaline cleaning every 100 h and acidic cleaning every 500 h Authors were able to recover 99% of the initial permeability. Gong et al. (2013) in application of DT-RO for concentrating anaerobic digestate (75% recovery and feeding k = 22 mS/cm) stated the same result. Although they did not report information about permeability decline, the fouling rate has been estimated to be 6×10^{-4} L m^{-2} h^{-2} bar^{-1}, whereas the permeate flux was 12 L m^{-2} h^{-1}. As result, the accurate control on chemical cleaning ensured the longevity of the membrane.

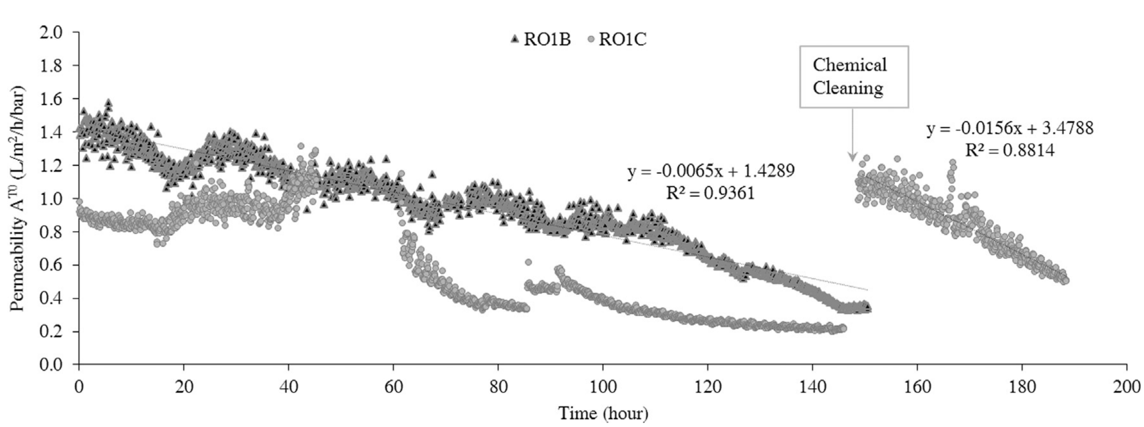

Fig. 6. Membrane permeability (A^{T0}) decline in $RO1_{B\text{-}C}$ runs.

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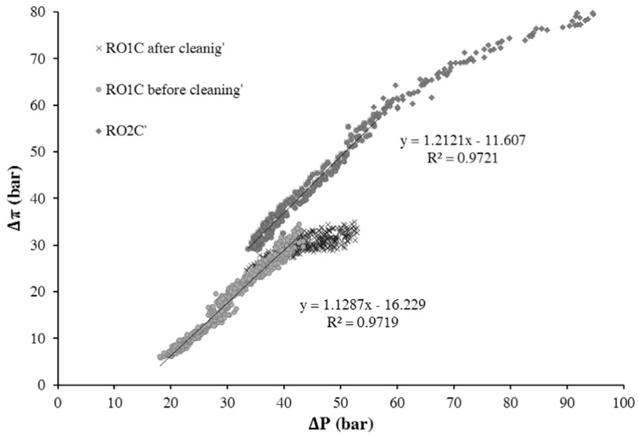

Fig. 7. Osmotic pressure differential ($\Delta\pi$) and operative pressure (ΔP) relationship in $RO1_C$ and $RO2_C$ runs (modeling data).

Furthermore, the linear relationship among modeled data of $\Delta\pi$ (Eq. (3)) with the pressure (ΔP) was found (Figs. 5 and 7) and is expressed by the following equation.

$$\Delta\pi = c\Delta P - d \quad (4)$$

The slope value (c) is the fouling rate while the intercept with the y-axis (d) is the initial operative pressure. Both parameters show preliminary information about the initial condition of the membrane system. Therefore, the whole permeability could be assumed as function of the pressure drop (ΔP), according to the Eq. (5):

$$A = \frac{J_w}{\Delta P + (1 - c) + d} \quad (5)$$

Ideally, c and d must be the same for similar hydrodynamics conditions, cleaning state and same effects of internal and external concentration of polarization. Practically, both the coefficients are influenced by piping configuration and membrane modules.

As stated by Eq. (5), the permeability (A) decreases by raising d or decreasing c when the other values are assumed constant.

When pressure increases the slope of the curve decreases (c parameter decreases - highlighted zone in Fig. 5). In particular, from hour 130 on in run $RO1_B$ the correlation between ΔP and $\Delta\pi$ is not linear, although the permeate flux remains at 13 $L\,m^{-2}\,h^{-1}$, while ΔP increases from 40 to 67 bar. In the highlighted zone, the performances of the membrane decreased both for permeability (Fig. 6) and for the concentrations of the permeate. As consequence, $\Delta\pi$ decreases together with the rejection performances. In general, the fouling effects could be linked to long filtration time, to cake layer formation, to extension and more compact of cake layer impact and to internal and external polarization phenomena (Le and Nunes, 2016). However, the contribution of each single phenomenon cannot be distinguished. Similar linear trends have been observed also on the run C (Fig. 7).

Therefore, the analysis of $\Delta\pi/\Delta P$ could be used as indicator of the fouling rate and can be related to the rejection parameters. In addition, the $\Delta\pi/\Delta P$ ratio can support the decisions for membrane cleaning and predictive maintenance.

3.5. DT-RO applicability and energy consumptions

Comparing and assessing different RO membranes and modules geometry should consider both performances and costs. Those include mainly energy consumption, chemicals, replacement and initial investment.

In RO purification of landfill leachate with a conductivity of 15–16 mS/cm, Peters (1998) has stated the time to replace the DT membranes as more than three years. Recovery rate around 80% and reinjection of concentrate into the landfill were adopted in that case.

Performances on concentrating landfill leachate by alternative RO systems are widely described in literature papers. However, there is a gap of knowledge in assessing the optimal operating parameters. Li et al. (2009) presented the RO treatment of landfill leachate (k = 16 mS/cm) in tertiary treatment by using SW modules equipped by TFC membranes. Operating at average flux of 6.5 $L\,m^{-2}\,h^{-1}$ with a recovery of 53.4%, fouling rate was assessed to $1.8 \times 10^{-3}\,L\,m^{-2}\,h^{-2}\,bar^{-1}$ in a 90 h cycle. Nonetheless, after 2 weeks of operation permeability was permanently loss. In RO seawater desalination, SW membranes have an estimated lifespan of 2–5 years (Avlonitis et al., 2003), in high salinity brackish water with high recovery rates the duration is presumably shortened.

Among the available RO technologies, the cost for chemicals is almost uniform (0.28–0.33 €/m^3). With regard to the capital cost, Vibratory Shear Enhanced Process (plate and frame RO modules) has been considered representative for DT-RO CAPEX, equal to 34,900 €/m^3/h (75–85% recovery) (Subramani and Jacangelo, 2014). Capital cost of the cheaper SWRO plant ranges in 5,000–20,000 €/m^3/h.

Lastly, the specific energy consumption of the pilot plant was in the range 15.8–20.9 kW h per ton of treated leachate. However, the scale of the plant overestimate the SEC that has been reported to be as high as 8.5 kW h/m^3 in full scale DT-RO treatment plants (Rautenbach and Linn, 1996) with 97% water recovery.

4. Conclusion

The triple-stage RO with DT technology has been studied as tertiary treatment for landfill leachate in order to maximize water recovery and achieve the quality standard for discharge in sensitive water bodies or water reuse.

The study has defined the membrane performances in terms of ions and metals rejection and permeate quality of each single RO stage.

Rejection of the COD was higher than 95% and TDS removal was in the range 91.1% to 97.7% when mean operating pressure was 33–76 bar. Rejection of ammonia ranged between 57.4% and 77.3%.

By combining RO1 and RO2 the achieved recovery rate (RR%) was higher than 90%, but RO3 was necessary to achieve nitrogen and boron standards for discharge or reuse.

Drastic membrane and irreversible fouling was observed after 150 h of continuous filtration, and the $\Delta\pi/\Delta P$ ratio can support the decisions for membrane cleaning and predictive maintenance. Prediction of fouling rates is feasible by measures of electrical conductivity and ions concentrations.

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