



## Effectiveness of membrane bioreactor/reverse osmosis hybrid process for advanced purification of landfill leachate

Nawal Elfilali<sup>a</sup>, Nodar Essafi<sup>a</sup>, Mohamed Zait<sup>a</sup>, Mustapha Tahaikt<sup>a</sup>, Fatima Elazhar<sup>a,b</sup>, Azzedine Elmidaoui<sup>a</sup>, Mohamed Taky<sup>a,c,\*</sup>

<sup>a</sup>Laboratory of Advanced Materials and Process Engineering, Faculty of Sciences, Kenitra, BP 1246, Kenitra – Morocco, emails: Mohamed.taky@uit.ac.ma/takymohamed@gmail.com (M. Taky), nawal.elfilali@uit.ac.ma (N. Elfilali), nodar.essafi@uit.ac.ma (N. Essafi), mohamed.zait@uit.ac.ma (M. Zait), mustapha.tahaikt@uit.ac.ma (M. Tahaikt), fatima.elazhar@uit.ac.ma (F. Elazhar), elmidaoui@uit.ac.ma (A. Elmidaoui)

<sup>b</sup>National Higher School of Chemistry (NHSC), Ibn Tofail University, Kenitra – Morocco

<sup>c</sup>International Water Research Institute, Mohammed VI Polytechnic University, Lot 660, Hay Moulay Rachid Ben Guerir, 43150 – Morocco

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### ABSTRACT

The purpose of this article is to develop an effective hybrid method for treating Oum Azza landfill leachate (Rabat, Morocco). The treatment system studied is membrane bioreactor technology (MBR) which consists of biological treatment associated with a unit of ultrafiltration for the retention of the biomass followed by a reverse osmosis (RO) filtration step in order to achieve the Moroccan discharge limits. The performances of RO on the advanced treatment efficiency are investigated in tube pressure configuration mode, in order to achieve high quality of permeate with higher recovery rate and minimization of brine discharges. At the end of this study, it can be concluded that the performance of the MBR can achieve the reduction in 5-day biochemical oxygen demand and chemical oxygen demand of the order of 87% and 76% respectively. The permeate analysis at the outlet reverse osmosis shows that the controlled parameters are below legal standards Moroccan of direct discharge in nature, especially total dissolved solids content.

*Keywords:* Landfill leachate; Hybrid process (MBR/RO); Post-treatment; Water recovery; Brine management

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### 1. Introduction

Landfill leachate (LFL) is a potential source of pollution of water resources and may cause serious public health problems if they are discharged into water courses without any prior treatment [1–3]. Nowadays, the treatment of LFL has become every time a great challenge especially due to the fact that environmental regulations have become more stringent. LFL is highly variable and heterogeneous with a very complex composition [4–6]. The composition and concentration of contaminants are mainly influenced by

the age of the LFL [7,8]. The high concentration of refractory organic matter, ammonia, polluting/toxic organic and inorganic compounds, elevated pH levels and color, taken together with the high variation in composition and volume generated, make the treatment of LFL a real challenge. The most common systems used in the treatment of this effluent are based on biological processes. Biological processes are very effective when applied to young leachate, but their efficiency decreases with an increased leachate age [9,10]. In particular, conventional biological systems cannot

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\* Corresponding author.

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significantly treat old leachate, which contains recalcitrant contaminants resistant to biodegradation.

Moreover, with progressively more strict discharge standards being implemented in most countries, especially concerning total dissolved solids (TDS), membrane bioreactor (MBR) effluents may still require post-treatment [11,12]. A combination of membrane processes as a finishing step for MBR effluent allows for greater efficiency in pollutant removal [13].

Many published works have focused on hybrid processes for LFL treatment. When reverse osmosis (RO) is used downstream of a biological treatment, the elimination of chemical oxygen demand (COD) is of the order of 98% and that of  $\text{N-NH}_3$  is close to 100% [5–14]. Moreover, RO is widely recognized for its excellent capacity to remove pollutants that enables LFL treated biologically to meet strict discharge standards [5–15]. Results of preceding studies [16,17], of the MBR/RO combined process, show that the elimination of organic pollutants was higher than 97% and the reduction of inorganic pollutants ranged from 43% to 97% depending on treatment conditions. Likewise, Chen et al. [18] examined the transformation and removal of dissolved organic matters (DOM) in mature LFL by using MBR/RO process. The results showed that this hybrid method eliminates 99% of COD, 99.9% of  $\text{N-NH}_4^+$  and 99% of total nitrogen (TN). Elimination of UV254 and UV280 absorbance, which represent the degree of aromaticity of DOM in LFL and others wastewaters reaches 93%.

Therefore, Naz and Lan [19] and Renou et al. [5] mentioned that RO, either as a main step in a LFL treatment chain or as a single post-treatment step has shown to be an indispensable means of achieving high purification, removal of hazardous metals and potential water recovery. In others study, Ribera-Pi et al. [20] investigated the combination of a MBR as pretreatment of LFL followed by RO. In this configuration, electro dialysis reversal (EDR) was used to treat RO brine stream. They indicated that MBR removed inorganic carbon concentration up to 92% and nitrogen and SS up to 85% and 99.9% respectively. Thereafter, RO rejected 95% of the most pollutants of MBR effluent and achieved 84% of the global recovery rate along with operation. The RO brine was further concentrated by the EDR unit achieving an average recovery rate of 67% throughout the operation. The overall average recovery rate of the pilot plant system was greater than 90%. However, although hybrid technologies have been developed trying to be economically attractive [21,22].

For the purpose of this study, the landfill of Oum Azza is chosen from many other landfills in Morocco. Currently, this landfill produces 480 m<sup>3</sup>/d of leachate, which is suspected of causing environmental pollution of groundwater and surface water as well as ambient air by the propagation of very bad toxic and allergenic odors [23]. The LFL treatment process adopted in Oum Azza is based on biological treatment using aerobic and anoxic basins, followed by RO. The main problem encountered is the inefficiency RO plant allows a recovery ranging from 40% to 50%, leading to the accumulation of large quantities of brine. The brine is stored in basins, and the capacity of these basins is rapidly exceeded.

The objective of this work is to develop an effective hybrid method for treating Oum Azza LFL while minimizing

the environmental impact of this method. The hybrid method adopted in this study is a combination of MBR and RO (MBR/RO). Firstly, the leachate will be treated by using MBR technology. Then, and in order to remove the high level of LFL TDS, RO membrane filtration will be employed as a post-treatment.

The performances of RO on the advanced treatment efficiency are investigated in pressure vessel configuration, in order to achieve high quality of permeate with a higher recovery rate and minimization of brine discharges.

## 2. Materials and methods

### 2.1. LFL site

LFL used in this study is collected from the landfill technical center of Oum Azza. The site of Oum Azza is located in the Rabat-Salé-Kénitra (RSK) region about 30 km in the southwest of Rabat City (capital of Morocco) and covers an area of 110 ha.

Fig. 1 gives the map localization of the Oum Azza landfill [24]. In 2007 Oum Azza was the only controlled landfill in the country. Nowadays, it receives almost 50,000 tons/y of household and similar waste (HSW) coming from 13 municipalities in the RSK region. These wastes are composed of more than 60% of very wet organic waste (50%–60% of water) and have a low calorific value of less than 900 kcal/kg [25]. It generates a large quantity of leachate and the estimated average is around 480 m<sup>3</sup>/d [26]. The chemical-physical characteristics of raw LFL are listed in Table 1.

### 2.2. Characterization of raw leachate

Physicochemical characteristics of Oum Azza LFL listed in Table 1 show that this effluent contains high levels of COD, 5-day biochemical oxygen demand ( $\text{BOD}_5$ ), and total suspended solids (TSS). In addition, LFL contains a higher concentration of other parameters, such as electric conductivity ( $E$ ), chloride, nitrogen, sodium and phosphorus. Distinctly, the  $\text{BOD}_5/\text{COD}$  ratio is around 0.17, so this leachate is classified as intermediate leachate with a medium biodegradability [27].

Samples of the incoming and treated leachate are taken periodically for each treatment cycle. Physical and chemical analyses are carried out for several parameters such as COD, TSS,  $\text{BOD}_5$ , electrical conductivity, total nitrogen, total phosphorous, inorganic compounds in accordance with standard methods [28,29]. These parameters are measured daily. While COD, TN and total phosphorous (TP) are measured with reagent kits (HACH DR4000, USA) twice per week [29]. Hydraulic retention time (HRT) is calculated on the basis of the influent flow rate and volume of the reactor tank. The aeration rate is measured using a flow meter.

### 2.3. Experimental model

The advanced system used in this study is a pilot-scale hybrid MBR-RO system. It consisted of eight main components, including an inlet tank, an anoxic tank, an aerobic tank, an ultrafiltration (UF) membrane module, an intermediate tank, a RO unit, permeate and brine tanks. Fig. 2

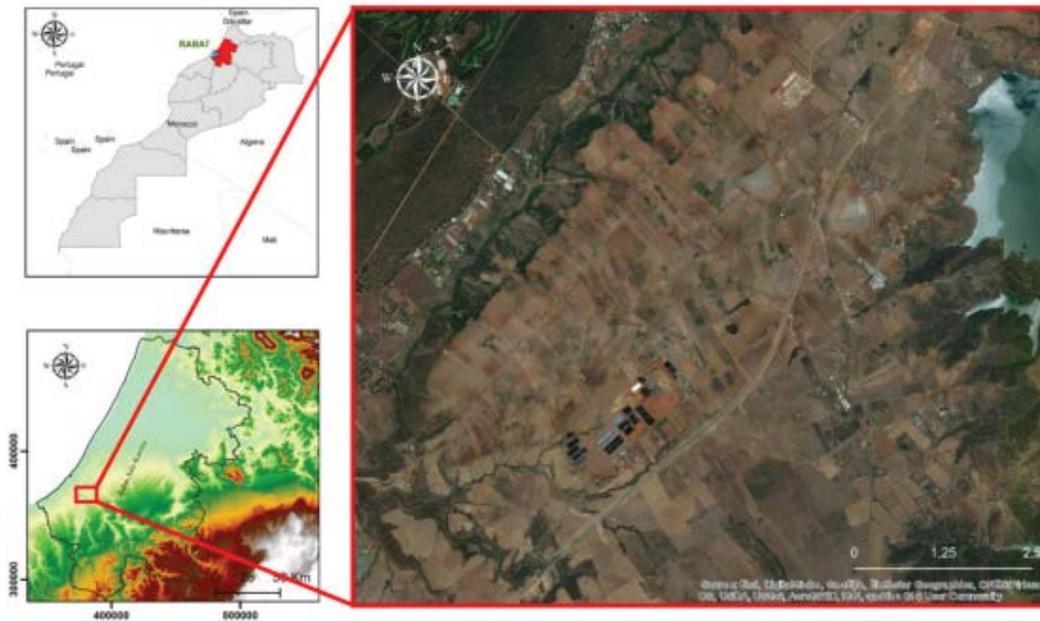


Fig. 1. Oum Azza landfill localization map [24].

Table 1  
Raw leachate characteristics

Parameters	Average value
Color	Dark brown
pH	8.14
Temperature (°C)	27
E (mS/cm)	20.9–30.85
COD (mg O <sub>2</sub> /L)	4,985–7,433
BOD <sub>5</sub> (mg O <sub>2</sub> /L)	895–1,250
TSS (mg/L)	387
BOD <sub>5</sub> /COD	0.17
TN (mg/L)	725–1,025
TP (mg/L)	28.97–53.65
Na <sup>+</sup> (mg/L)	3,605
Cl <sup>-</sup> (mg/L)	3,475

summarizes the schematic diagram of the whole process. The raw leachate is stored in the storage tank. Afterwards, the leachate is routed to MBR. This latter is composed of the anoxic tank as being the first stage and of the aerobic tank being the second stage and subsequently, the sludge is separated from the leachate treated by an ultrafiltration (UF) membrane. The MBR system removes organic compounds, and the treated leachate is stored in the intermediate tank (Fig. 2). To complete the purification of the treated leachate, removing salts and residual organics, the effluent is treated by a RO pilot. The permeate is then stored in a tank to be reused for different purposes and the brine is stored in another tank.

#### 2.4. MBR system description

In this study, the laboratory pilot used is an external MBR supplied by Deltalab/Cossimi Co., France. Membrane

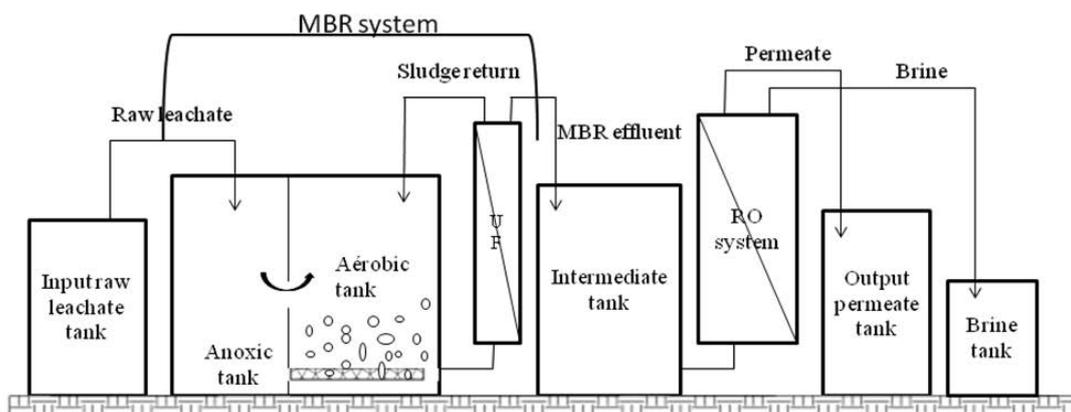


Fig. 2. Schematic diagram of MBR/RO process for LFL treatment.

filtration is done on an external UF membrane. The MBR setup is composed of an activated sludge tank with two components setting in succession. The first is an anoxic reactor and the second is an aerated one. After the biological treatment, the mixture feeds a membrane module, which is equipped with a tubular UF membrane. This membrane is used to separate solid sludge and liquid influent, the sludge and large size particles are rejected by the UF membrane, and water, salts and small size particles pass through the membrane. Then, the obtained MBR permeate is directed to the RO stage to complete the purification and to comply with discharge standards. The scheme of the MBR configuration is depicted in Fig. 3. Fig. 4 shows pictures of MBR and RO laboratory-scale pilots used for the treatment of Oum Azza LFL.

The reactor is continuously aerated using an air compressor to maintain dissolved oxygen concentration above 2 mg/L to supply oxygen for biomass. To adapt the activated sludge to the leachate, glucose, is provided as a substrate to help biomass to acclimatize easily to the complex leachate and to grow sufficient biomass for stable operation of the aerobic reactor. After that, low volumetric loading of diluted leachate is gradually introduced into the reactor, while aeration is maintained continuously for several days until the microorganisms could tolerate a high COD concentration of leachate. Being a second step, the internal recirculation of the mixed sludge liquor is carried out continuously from the aerobic tank to the anoxic tank in order to maintain a constant concentration of biomass throughout the biological treatment.

2.5. MBR start-up

2.5.1. Acclimatizing of the sludge

During the first step of operation, the aerobic reactor is operated and fed by an activated sludge taken from a wastewater treatment plant WWTP situated in the National Office of Electricity and Drinking Water (ONEE) in Rabat.

2.5.2. Operating conditions of the MBR

The MBR laboratory-scale pilot is composed of an anoxic bioreactor (20 L made of plexiglass) and an aerobic bioreactor (40 L also made of Plexiglass) and an ultra-filtration tubular membrane module. Table 2 gives the membrane characteristics. Ceramic UF membranes are by

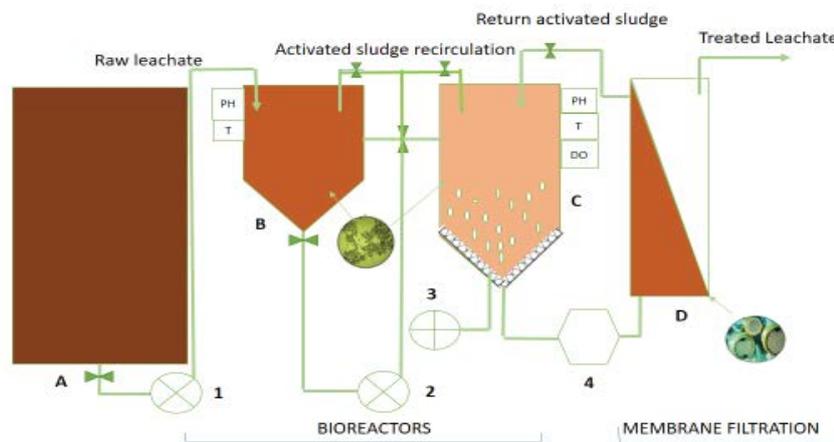


Fig. 3. Schematic diagram of external MBR. (A) Feed tank, (B) Anoxic tank, (C) Aerobic tank, (D) ultrafiltration module, (1) Peristaltic pump, (2) Recirculation pump, (3) Air compressor, (4) Filtration pump.



Fig. 4. Picture of (A) MBR pilot and (B) RO pilot.

Table 2  
Characteristics of the UF membrane

Membrane material	Ceramic
Module	Tubular
Provider Pall	Exekia
Membrane area	0.45 m <sup>2</sup>
Cut-off	15 kDa
Membrane length	1,178 cm
Diameter of the channels	6 mm
TMP	0.05–1.35 bar

far widely used through the physical removal of particles from the liquid in the size range of 0.01–0.1  $\mu\text{m}$ , because of their potential advantages including physical strength, chemical and thermal stability. Pressure sensors and pressure gauges are placed at the recirculation pump outlet just before the membrane module inlet, at the outlet of the membrane module and in the permeate collection circuit. The operating transmembrane pressure (TMP) is 1.30 bar.

The raw leachate is fed from a storage tank to an anoxic reactor by a peristaltic pump, the feed flow is regulated with two-level sensors to maintain a constant working volume of liquid in the reactor. Afterward, the effluent is pumped to the aerobic reactor. Sequenced aeration is done by four diffusers placed at the bottom of the aerated reactor, providing the necessary oxygen for good treatment. The aeration cycles are fixed by the oxygen transmitters to control the air blowing. Furthermore, an internal recirculation of the mixed liquor sludge is continuously done from the aerobic tank into the anoxic tank. Table 3 gives the operating conditions of MBR. The UF membrane is cleaned after each use following the manufacturer's recommendation. However, the membrane filtration unit is disassembled from the setup before starting the chemical cleaning of the membranes. Prior to the cleaning exercise, the membrane module is rinsed two to three times with tap water for removing the sludge layer and solid particles deposited on the membrane surface. Then, citric acid solution and alkali solution are prepared and put in the cleaning tank, each solution recirculated through the membrane for 20 min [30].

### 2.6. Experimental set-up of RO

The effluent from the MBR is post-treated by a RO unit. RO experiments are performed on an industrial pilot

Table 3  
Operating conditions of MBR

Parameters	Values
pH	7
T (°C)	20–35
Dissolved oxygen (mg O <sub>2</sub> /L)	2–5
SRT (d)	30
MLSS (g/L)	16
HRT (h)	72

NF/RO provided by the company TIA (Applied Industrial Technologies, France). This pilot was described in detail in previous papers [31,32]. The pilot is equipped with two identical modules in series. The experiments are conducted using RO membrane (SW TM810). The main characteristics of this membrane are shown in Table 4. After the run, the membrane is cleaned with alkaline and acidic cleaning solutions according to the manufacturer's recommendation.

## 3. Results and discussion

### 3.1. MBR efficiency

The LFL originated from the Oum Azza is treated by a MBR. Its efficiency in the treatment of Oum Azza LFL is presented in Table 5.

As shown in Table 5, MBR may provide an excellent pretreatment for subsequent RO stages. Thus, the rejection of tested pollution indicators varies significantly during the MBR step. It is generally higher than 85% for TSS and BOD<sub>5</sub>, for phosphorus and nitrogen rejection is approximately equal to 60%. In the case of TN, this low rejection is a result of limited nitrification and denitrification efficiency. The high rejection obtained for COD (76%) shows high efficacy of the MBR process in the treatment of leachate heavily loaded with organic matter in comparison to other systems. Moreover, a slight reduction of conductivity, chloride and sodium content after biological treatment, 26%, 20%, 15% respectively is observed. A part

Table 4  
RO membrane characteristics

Membrane	SW TM810
Type	Spiral
Area (m <sup>2</sup> )	7
P max (bar)	69
pH	1–11
Max. temp. (°C)	45
Material	Polyamide
Salt rejection (%)	99.75

Table 5  
Effectiveness of MBR treatment

	MBR/UF effluent	Rejection (%)	MLDS <sup>a</sup>
pH	7–8	–	–
Color	Brown	–	–
E (mS/cm)	15.48–22.8	26	2.7
COD (mg/L)	1,196–1,784	76	120
BOD <sub>5</sub> (mg O <sub>2</sub> /L)	116.5–162.5	87	40
TSS (mg/L)	38	89	30
TN (mg/L)	292.2–413	59.7	40
TP (mg/L)	11.70–21.7	59.6	15
Na <sup>+</sup> (mg/L)	2,884	20	–
Cl <sup>-</sup> (mg/L)	2,953.75	15	–

<sup>a</sup>Moroccan Liquid Discharges Standards (MLDS, 2018).

of this decline may be due to the elimination of ammonium and the racking of excess sludge, which are regularly withdrawn from the processing system. In addition, improvements in the color and conductivity of treated water by MBR are not obvious. Therefore, the leachate still did not meet the effluent discharge of Moroccan standards and the effluent at the outlet of MBR treating LFL is characterized by a low BOD<sub>5</sub>/COD ratio (lower than 0.1) indicating the presence of refractory organic matter. Hence, additional treatment is required. An interesting idea would be to use the RO unit stage to complete the treatment started by MBR to achieve the direct discharge standards requirements. It should be noted that the main advantage of the MBR process is that it reduces the importance of biomass sedimentation, thus allowing a significantly smaller tank to be used for the bio-treatment process. The second main advantage of MBR is that the treated water quality is better than from a conventional process since the membrane barrier removes essentially all particles above the pore size rating of the membrane [33]. However, the propensity of the UF membrane to fouling is the major constraint of this filtration step.

### 3.2. UF permeability and membrane cleaning

The variation of permeability with operating time is presented to assess the ultrafiltration membrane fouling behavior in MBR. The characteristics of the used membrane are listed in Table 2. The operating TMP has maintained at 1.30 bar thanks to the periodical cleanings performed. Thus, the UF stage of MBR is held under appropriate filtration conditions and no major issues are detected. Fig. 5 gives the variation of monitoring permeability and the TMP as a function of time. The permeability ( $L_p$ ) is calculated from Eq. (1) as follows:

$$L_p = \frac{J_p}{\Delta P \text{ TM}} \tag{1}$$

where  $J_p$  is the permeate flux (L/m<sup>2</sup> h), and  $\Delta P$  is the TMP (bar).

As indicated in Fig. 5, the membrane permeability varies through the process between 8.4 and 19 L/m<sup>2</sup> h bar. The flux remains constant and the UF membrane permeability is stable. The decline in the permeate flux is particularly caused by the degree of membrane fouling during the continuous operation. Wisniewski and Grasmick [34] and Defrance et al. [35] indicate that such behavior in MBR is attributed primarily to suspended materials (biological flocs), while Bouhabila et al. [36] attribute UF membrane fouling in MBR to colloidal materials. However, an analysis of those studies enables one to extend the hypothesis that fouling is a result of the contribution of the various sludge fractions depending on membrane characteristics, hydrodynamic conditions, and biomass characteristics. The maintenance chemical cleaning is performed after 50 h of operation for fouling control as indicated in Fig. 5. The downtime for effective membrane cleaning took 1 h in this study. After cleaning the membrane, higher permeability values are observed, indicating when chemical cleaning is carried out successfully.

### 3.3. RO Treatment of the MBR permeate

The permeate obtained by the MBR process is post-treated by the RO unit in order to improve MBR permeate quality and to achieve the required Moroccan discharge limits. In this configuration treatment, the multistage design of the SWTM810 membrane processes is considered. Fig. 6 shows a cascade design of membrane modules arranged in series, for the purpose of illustrations. The brine of the first module serves as the feed to the second module, and all modules are arranged in the same mode. Each module consists of two membranes. All permeates are collected as product water.

RO experiments are performed at operating pressure varying from 58 to 54 bar. The global recovery rate of the RO

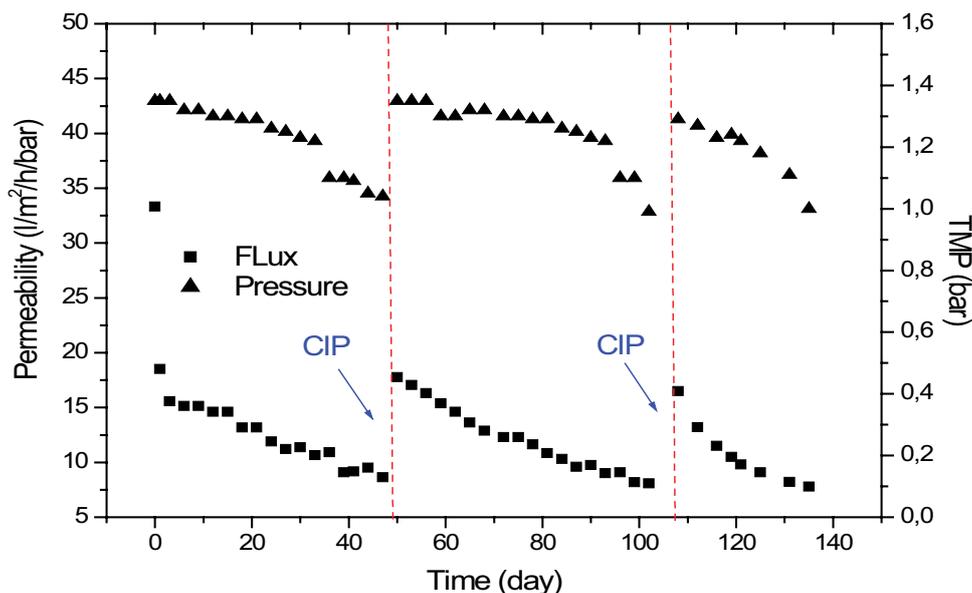


Fig. 5. Membrane permeability vs. TMP.

unit is around 84% generating a total volume of brine stream of 46.9 L from the 290 L of MBR leachate permeate treated in the RO step. However, TPM increase is mainly due to the increase of TDS in RO feed and thus to the increase of osmotic pressure. This phenomenon accentuates the tendency of membrane fouling although it did not hamper the preservation of the permeate flow. Alternatively, a pressure vessel configuration is considered adequate to minimize the brine discharge volume and to increase the overall recovery rate of the hybrid system. It should also be noted that the quality of total permeate water is improved to satisfy the water quality regulations for (irrigation or industrial process). Three tests are performed on MBR leachates permeate by RO. The main

operating conditions and electric conductivity rejections are collected in Table 6.

Fig. 7 shows the performances of RO membrane on the rejection of several compounds of LFL and it indicates that the permeate quality of RO unit, is largely improved, rejection of the most pollution indicators is greater than 95%. Indeed, abatement of electric conductivity reaches 98% with a recovery above 84%. The RO permeate quality obtained with a pressure vessel of three membrane modules meets all discharge Moroccan standards. Thus, considering the following industrial reclaimed water uses: process and cleaning water, and cooling towers and condensers, the obtained permeate could be reused within landfill facilities decreasing

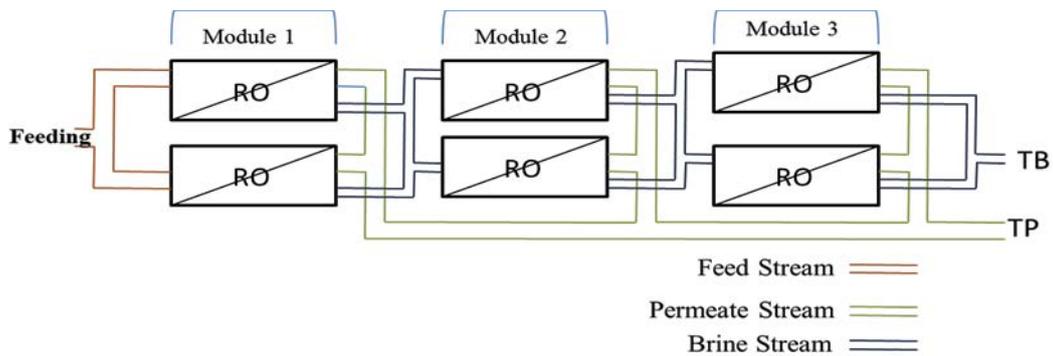


Fig. 6. Cascade design of membrane modules arranged in series. TP: Total permeates, TB: Total brine.

Table 6  
RO unit operating conditions and performances

	TMP (bar)	Flux (L/h)	Recovery (%)	Feed conductivity ( $\mu\text{S/cm}$ )	Permeate conductivity ( $\mu\text{S/cm}$ )	Rejection (%)
RO1	58	3,698.12	44.15	22,000	476	97.83
RO2	56	2,445.85	38.15	35,000	642.23	98.78
RO3	54	1,754.58	27.87	57,000	1,483.56	98.12

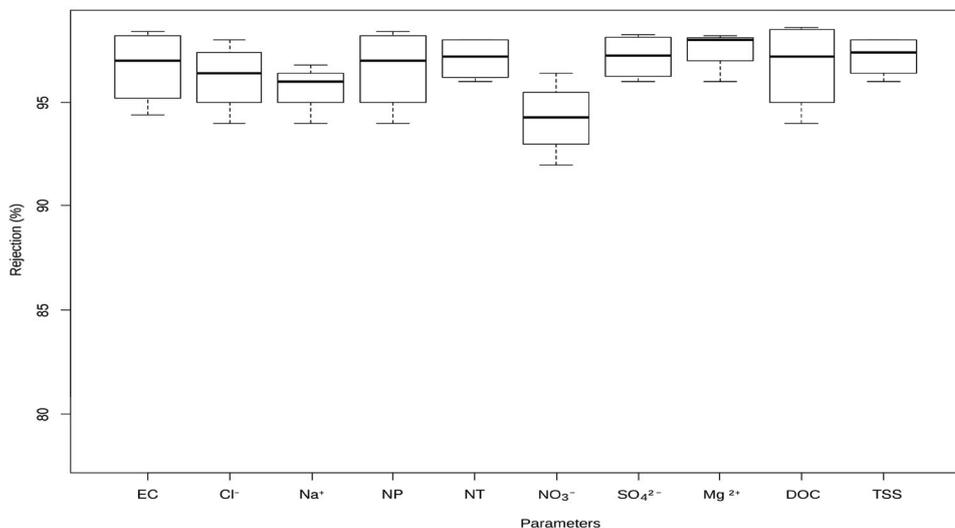


Fig. 7. Performances of RO membrane on the rejection of several compounds of LFL.

water consumption of the landfill and contributing to close the water circular economy loop.

#### 4. Conclusion

The performances of an integrated method of MBR/RO have been investigated to treat Oum Azza LFL. The previous study has highlighted that Oum Azza leachate is characterized by a high concentration of indicators pollutants. Distinctly, The BOD<sub>5</sub>/COD ratio is around 0.17, so this leachate is classified as intermediate leachate with a medium biodegradability. The treatment of LFL by MBR shows that it is enabled to reduce biodegradable organic matter. On the other hand, it diminishes the fouling phenomenon of UF which is still the main limitation of the MBR performance. On the contrary, MBR offers a poor reduction of TDS. RO placed downstream of MBR completes and perfects the purification started by MBR. Optimization of RO, in terms of water recovery and TDS rejection, provides significant performances of recovery rate up to 84% and TDS rejection up to 98%. Lastly, the water quality at the outlet of the RO step is significantly improved to be reused in the landfill Technical center Oum Azza for irrigation or for industrial purposes to reducing water consumption and contributing to close the water circular economy loop. The advanced hybrid treatment MBR/RO of LFL seems to be a viable solution for lessening environmental risks and obtaining a higher recovery value, but it is limited by its operating cost. Previous studies have estimated the operating cost of MBR/RO hybrid systems [37] and MBR/NF/RO systems [38] for the treatment of LFL at 3.86 USD/m<sup>3</sup> and 4.55 USD/m<sup>3</sup>, respectively. A detailed technical economic study of this MBR/RO hybrid process and its comparison with the current LFL treatment process at the landfill technical center of Oum Azza will be the subject of the next paper.

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