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Membrane bioreactor (MBR) performance in fish canning industrial wastewater treatment

H. Ayyoub[a], S. Kitanou[a,b,*], B. Bachiri[a], M. Tahaikt[a], M. Taky[a,c] and A. Elmidaoui[a]

[a] Laboratory of Advanced Materials and Process Engineering, Faculty of Sciences, Ibn Tofail University, P. O. Box: 1246, Kenitra, Morocco
[b] National High School of Chemistry, Ibn Tofail University, P. O. Box: 1246, Kenitra, Morocco
[c] International Water Research Institute, Mohammed VI Polytechnic University, Benguerir, Morocco
*Corresponding author. E-mail: sarra.kitanou@gmail.com

MT, 0000-0001-6658-4537

ABSTRACT

The fish canning industries produce a large amount of wastewater. The high-strength wastewater from fish processing industry is of great concern worldwide. This type of wastewater is rich in organic matter and has high total dissolved solids (TDS). The elimination of salts is usually expensive and, on the other hand, the high salinity and the seasonal variation of the effluent characteristics make the removal of organic matter difficult using only a biological process. For this purpose, the use of a membrane bioreactor (MBR) process combining both a biological treatment and ultrafiltration membrane separation could be a perfect option for fish canning wastewater treatment. In this work, the performance of a MBR process is investigated to treat the fish canning industry wastewater. The removal efficiency of important parameters such as chemical oxygen demand (COD), five day-biological oxygen demand in (BOD_5), total suspended solids (TSS), nitrate (NO_3^-), total Kjeldahl nitrogen (TKN), orthophosphate (PO_4^{3-}) and TDS are followed. Experimental results show that the overall removal efficiencies obtained are: 96.6% for COD, 98% for BOD, 99.5% for TSS, 90% for nitrate, 96% for TDS and greater than 97% for TKN and PO_4^{3-}. These results confirm that combination of both biological treatment and membrane process is regarded as an effective approach to reduce the contaminants in fish canning wastewater. Membrane bioreactor presents several advantages in terms of water resource protection because of the great quality of the treated water that can be reused.

Key words: fish canning industry wastewater, membrane bioreactor treatment, removal efficiencies

HIGHLIGHTS

- Membrane bioreactor process performances.
- Industrial wastewater rich in organic matter and have high total dissolved solids.
- Activated sludge treatment coupled to membrane filtration.
- Pollution removal efficiencies.

1. INTRODUCTION

One of the most common by-products of industrial activities is industrial wastewater, the water that was used to make commercial products across every industry in nearly all phases of production (Granger *et al.* 2019). Once this process water has been used, it is considered waste and needs to be treated before it is discharged (Ahumada *et al.* 2004). Industrial wastewaters have a far more complex composition than domestic or municipal ones, with a larger variety of pollutants of varying nature and properties, depending on the industrial process they originate from (Stefanakis 2018). With the increasing number of industrial applications, industrial wastewater treatment and reuse become a high technical challenge, considering the varying nature of the different pollutants found in industrial wastewater (Lim *et al.* 2003).

It can now be said that, although research on domestic/municipal wastewater is still ongoing (for example, to optimize the removal of pollutants), the current challenges lie in the effective treatment of industrial wastewaters. Different treatment technologies have successfully treated effluents from a range of industrial wastewaters, including dairy, food, beverage, cosmetics, pharmaceuticals, metal fabrication, textiles, tannery and paper manufacture (Chowdhury *et al.* 2010). Fish canning wastewater treatment has also been undertaken in many countries around the world (Stephenson *et al.* 2005). Among the different technologies that can be applied for the treatment of these wastewaters, the capability of membrane techniques highlights, aiming to produce dischargeable water, complying with the increasingly strict legislation on industrial pollution. Fish canning factories represent a significant portion of the food industry (Lema *et al.* 2002). This wastewater can present very high concentrations of biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS) and total nitrogen (TN) (Guyer 2013). These industries generate large quantities of wastewater with significant fluctuations of organic load and a high salt content (Garcia-Sanda *et al.* 2003). The production of saline effluents represents about 5% of the total effluents to be treated in the world (Muthukumaran & Baskaran 2013). The degradation of saline effluents is therefore a problem that affects both the industrialized environment in general and the urban environment of many developing countries including Morocco (Meskour *et al.* 2015). The research findings suggested that although the removal of carbon and biological nutrients has been demonstrated to be feasible at high salt concentrations, higher efficiencies are necessary to meet the regulatory constraints for public sewage discharging. For these reasons, a membrane bioreactor (MBR) could be used for treating such streams, due to the requirement of biological degradation of waste compounds and the physical separation of the biomass and treated water by employing membranes (Yang *et al.* 2017). MBR processes are increasingly used in the fields of chemical engineering, industrial effluent treatment processes and separation/purification of biotechnology products (Gil *et al.* 2011). Furthermore, MBRs exhibit good resistances to variations in hydraulic and organic loadings, have low head losses and low space requirements.

The present work intends to develop a real, effective and workable solution that avoids and reduces the rejection of wastewater from the fish canning industry, managing it in the most appropriate way to obtain a treated water with higher quality. This objective will be achieved through the optimization of a series of various treatment steps envisaging wastewater reuse in the industrial process, while reducing the discharge of effluent, limiting the water use and saving money. To our knowledge this is the first investigation of an MBR treatment of fish canning industrial wastewaters in Morocco.

2. MATERIALS AND METHODS

2.1. The MBR configuration

Experiments were performed in a laboratory-scale MBR pilot system using UF membrane. A schematic diagram of the experimental facility is shown in Figure 1, mainly consisting of three components: anoxic tank, aeration tank, UF membrane module and for chemical membrane cleaning, a cleaning tank.

The pilot is equipped by three pumps: the first one used to feed the anoxic tank (from the feed tank), the second one used to feed the aeration tank (from the anoxic tank) and also recycle the effluent in the anoxic tank, and the

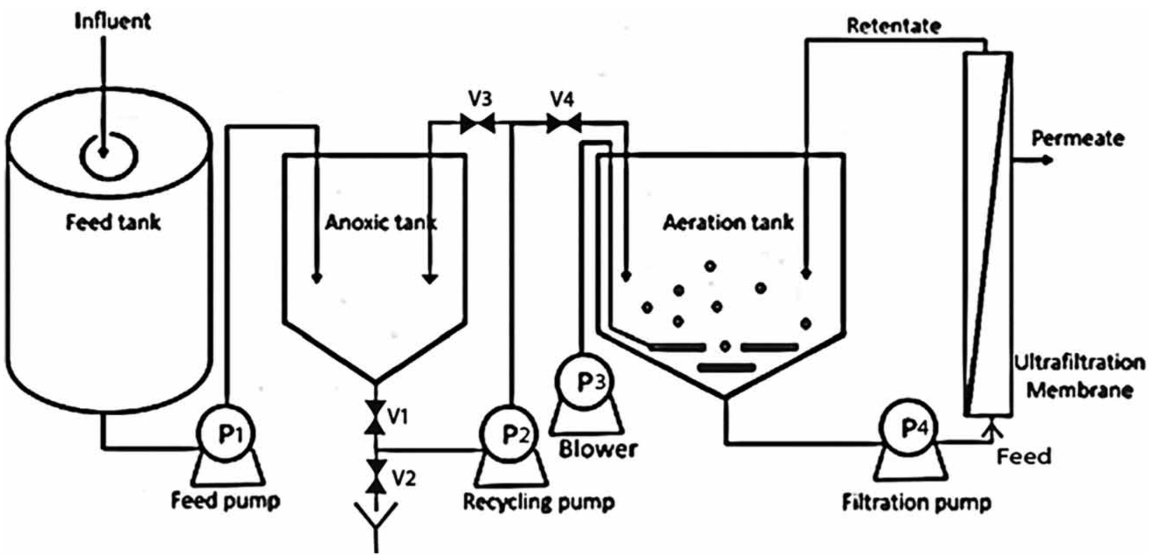

Figure 1 | Flow diagram of the experimental ultrafiltration MBR (Kitanou *et al.* 2021).

last one used for filtration (UF membrane). The treatment is carried out in a discontinuous way. The operation between the two compartments (biological and membrane) was discontinuous, i.e. filling of the aeration tank and the end of the biological treatment (depending on the operational conditions), followed by the beginning of the membrane filtration.

A constant average feed rate of 1 L/h was maintained in the system throughout the study. This is the flow rate applied by the recycle pump from the anoxic tank to the aeration tank.

The membrane is placed outside the bioreactor where recirculation of the mixed liquor is provided by a volumetric pump without intermittence. The flow rate applied to the inlet of the ultrafiltration membrane is around 138 L/h with a conversion rate of 10%.

The detailed design and operating parameters are shown in Table 1. The detailed operation of the MBR setup has been explained elsewhere (Kitanou *et al.* 2017).

Table 1 | Characteristics of the membranes used (Kitanou *et al.* 2018)

Membrane material	Ceramic
Module	Tubular type P10
Membrane area	0.45 m^2
Cutoff	15 kD/10–20 nm
Membrane length	1.178 m
Diameter of the channels	6 mm
Permeate flow (average operation)	33 $L/m^2/h$
Retentate flow	124 L/h
Transmembrane pressure (TMP)	0.35 bar

2.2. Inoculation, operational parameters and wastewater pretreatment

The seed sludge was obtained from an activated sludge (AS) taken from a wastewater treatment plant (WWTP) situated in Kenitra, Morocco. The sludge undergoes an acclimatization step, which lasts about 10 days. The aerobic tank is maintained in alternating aeration between aerobic and anoxic conditions, The aeration cycles were fixed by the oxygen transmitters that controlled the air blowing in, and the aeration range was 1,300 NL/h alternating between aerobic and anoxic conditions. A constant 24-hour hydraulic retention time (HRT) including only the aeration tank was maintained in the system throughout the study (Table 2). Sedimentation and flotation are the first steps in the wastewater treatment process to remove suspended particles from the wastewater. Indeed, the effluent is left in a tank with a capacity of 20 liters for 2 hours, which is the optimal time after several of the tests. At the back, the settled particles are extracted and the effluent is collected. These samples are then analyzed according to physico-chemical analysis protocols (Cristóvão *et al.* 2015a).

Table 2 | Operational parameters

Parameter	Value
Aeration rate	1,300 NL/h
Hydraulic retention time (HRT)	24 h
Solids retention time (SRT)	12 days
Ratio of feed to microorganisms (F/M)	0.24 kg BOD_5/kg VSS. day
Volumetric loading rate	2.23 kg BOD_5/ m^3. day
Organic load rate (OLR)	4.27 kg COD/m^3.day
Biomass concentration in the mixed liquor	10 g/L TSS and 8.5 g/L VSS
Flow rate of the retentate	124 L/h
Sludge generated	1.46 g/h of TSS

2.3. Analytical methods

Samples of wastewater were taken before and at the end of each treatment cycle. They were collected periodically and analyzed for various physical, chemical parameters in accordance with the Standard Methods (APHA 2005; Rodier *et al.* 2009). The pH was measured using a pH meter (JENWAY). Conductivity, salinity and TDS were measured using a multi-parameter conductivity meter (inoLab) with an electrode consisting of two 1 cm^2 square platinum strips. Quality parameters such as COD (Hach DR2800 Spectrophotometer), TSS and volatile suspended solids were determined following sample filtration through 0.45 μm. BOD_5 is measured after 5 days (OxiTop WTW), the nitrate content was measured using an electrode (Sension MM 340). The determination of orthophosphates is carried out by the colorimetric method based on the formation of a complex with ammonium molybdate and antimony-potassium double tartrate in an acidic medium (Bliefert & Perraud 2009). TKN was determined using the Kjeldahl methods (VELP 2016). Oil and grease were quantify using the separation liquid-liquid extraction (Dunn 2013). The HRT was calculated according to the flow rate of the influent and the size of the aeration basin. The aeration rate was measured using a flow meter.

3. RESULTS AND DISCUSSION

3.1. Analysis of wastewaters from fish canning industries

The quality of fish canning wastewaters varies according to the overall production of the fish canning industry. To obtain a representative set of data on effluent properties, several samples were collected at different times and analyzed.

Table 3 represents the raw effluent characteristics brought back from the fish canning industry after pretreatment, with minimum, maximum and average values. COD concentration values indicate a high contamination by organic matter. Also, high contents of O&G and salts are present, as indicated by the Cl^- and Na^+ concentrations and conductivity (corresponding to salinity between 2.5 and 5 ppt).

Table 3 | Characteristic of the fish canning wastewater

Parameter	Min.	Max.	Average Value
pH	5	6	5.5
Temperature °C	17	24	21
Conductivity (μS/cm)	3,600	5,100	4,350
TSS (mg/L)	1,000	2,700	1,850
COD (mg/L)	3,750	4,800	4,275
BOD_5 (mg/L)	1,000	3,100	2,050
TKN (mg/L)	11.5	32	21.5
TP (mg/L)	6	13	9.5
PO_4^{3-} (mg/L)	9	13.5	11.25
NO_3^- (mg/L)	110	132	121
Cl^- (mg/L)	1,240	3,550	2,395

3.2. Primary treatment by sedimentation/flotation

Table 4 shows the concentration value and the efficiency removal obtained by the primary treatment. The sedimentation/flotation phase lasts 2 hours and gives effective results with a TSS removal of 65% but the removal of COD does not exceed 5%, similarly BOD5 does not exceed 1% which means that sedimentation and flotation are not effective for organic reduction. For oil and grease, the removal is up to 57%. The oils inhibit the supply of oxygen and thus promote the development of anaerobic conditions, which is not beneficial for biological treatment. Raquel O. Cristovao (Cristóvaõ *et al.* 2015b) obtained results by sedimentation/flotation that showed it is effective with oil and grease (48%), TSS (75%) and COD (4%). The majority of suspended solids that contribute to the organic load would be difficult to remove using a conventional sedimentation system, but dissolved air

Table 4 | Concentration value after sedimentation/flotation treatment

Parameter	Raw effluent	After sedimentation/flotation	Abatement
BOD (mg/L)	2,050	2,030	1%
TSS (mg/L)	1,850	648	65%
COD (mg/L)	4,275	4,061	5%
Oil and grease	942	399	57%

flotation has the potential to effectively remove oil, grease and other highly settleable materials (Muthukumaran & Baskaran 2013). The oils inhibit the supply of oxygen and thus promote the development of anaerobic conditions.

The previous step, sedimentation/flotation, was not very effective in DCO removal, because soluble organics contribute significantly to the overall organic matter content of the fish canning wastewater. The pretreated wastewater requires removal of organic components by a biological treatment process and a membrane filtration. In this case an aerobic biological treatment by AS was employed. The biological aerobic reactor operated at HRT of 12, 15, 20 and 24 h. The obtained results in terms of pollutants removal efficiency are presented in Figures 2–4.

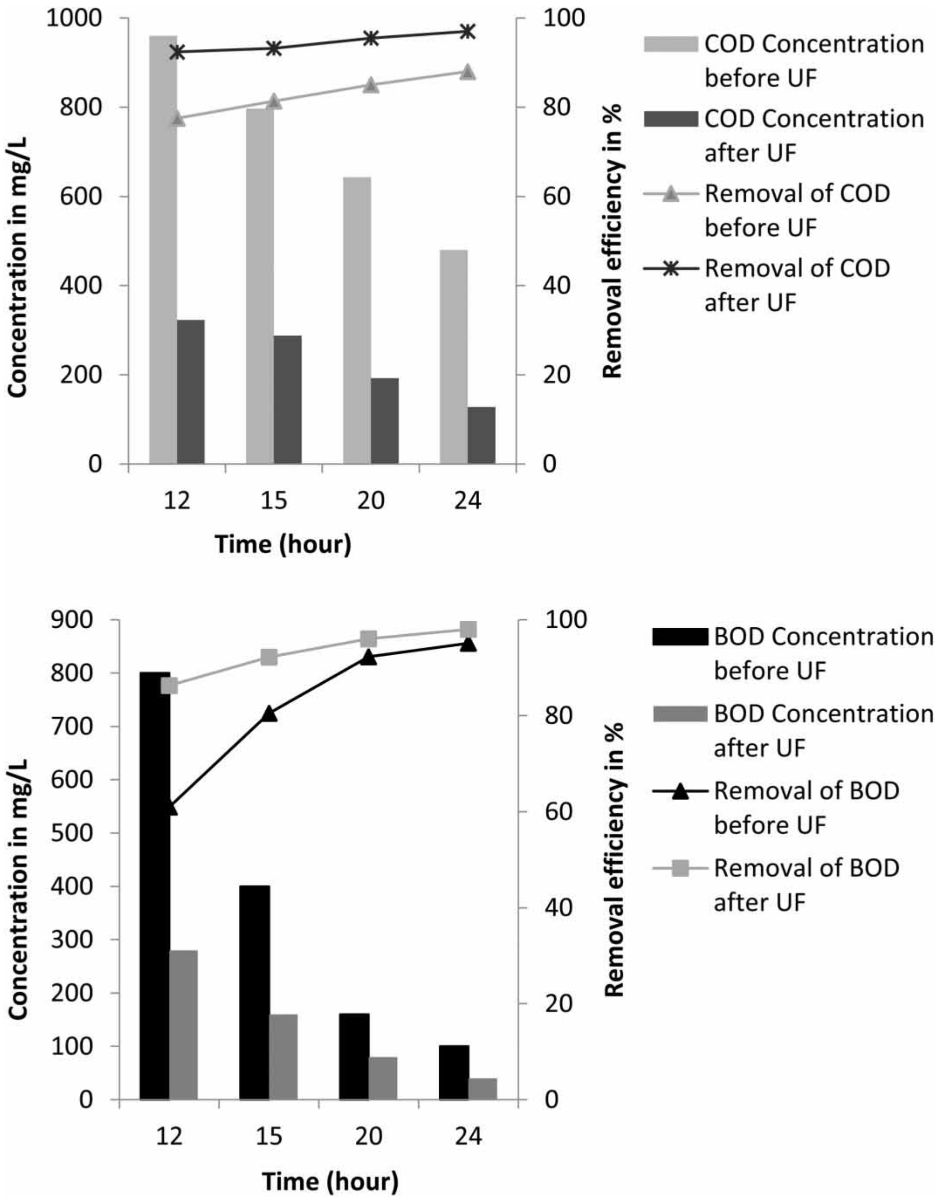

Figure 2 | Concentrations and removal efficiencies rate of COD and BOD as a function of HRT.

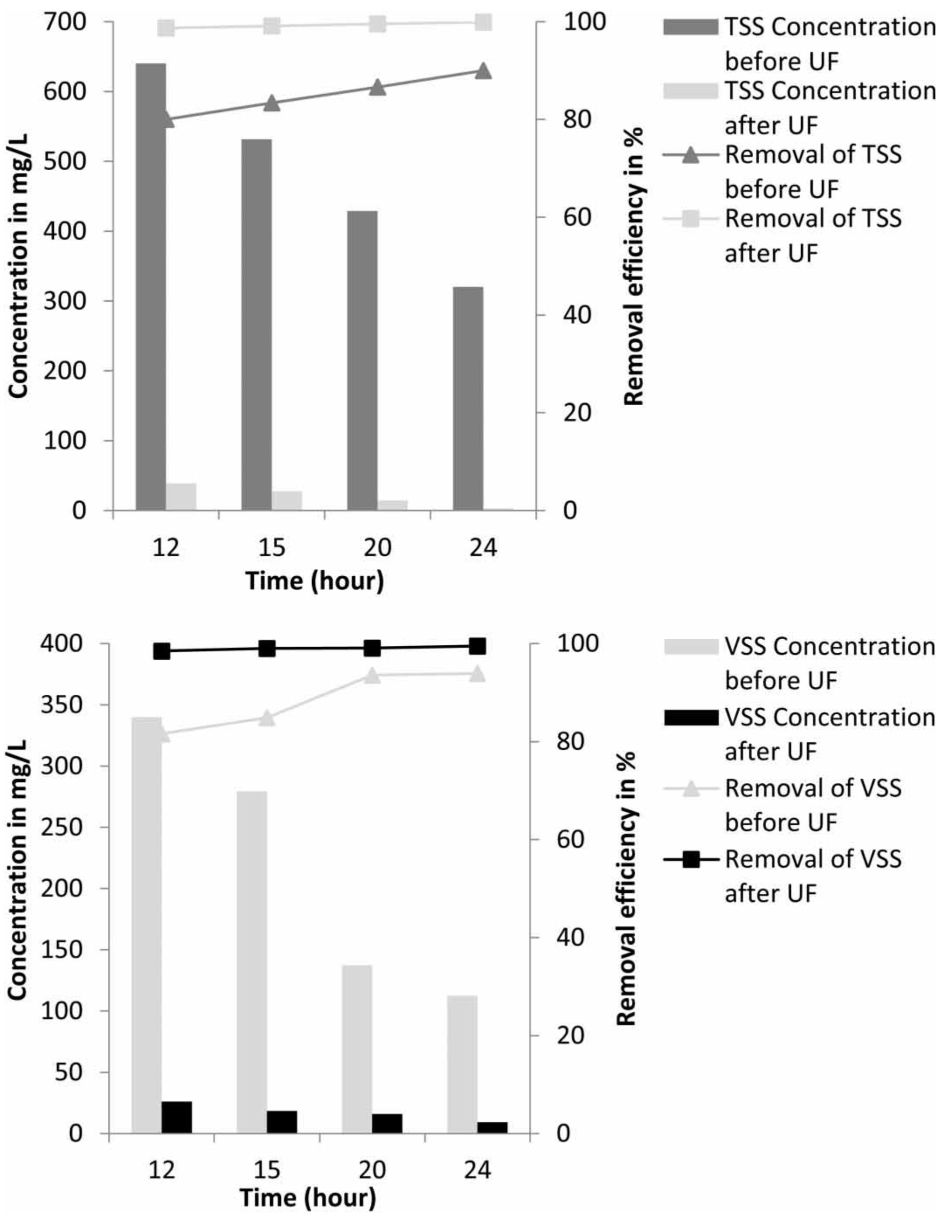

Figure 3 | Concentrations and removal efficiencies rate of TSS and VSS as a function of HRT.

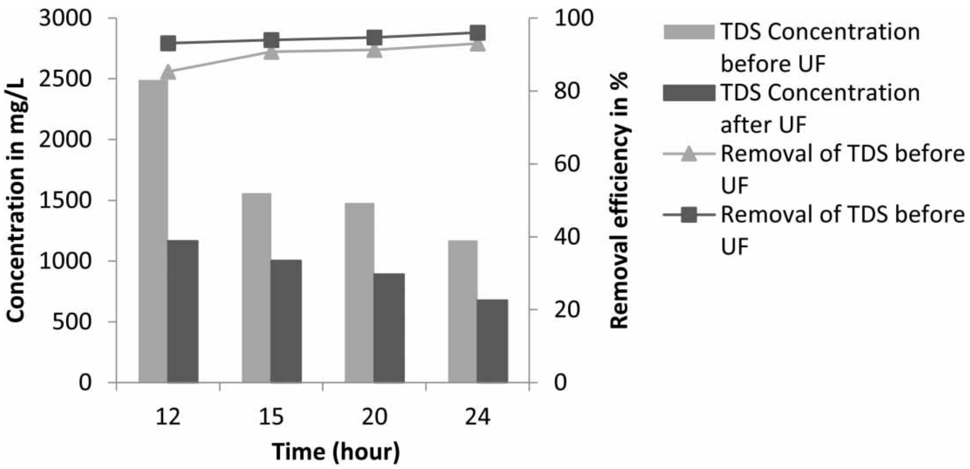

Figure 4 | Concentration and removal efficiencies rate of TDS as a function of HRT.

3.3. MBR treatment

3.3.1. Organic carbon removal

To determine the MBR performance, the organic carbon removal efficiency was monitored at different HRT. Therefore, organic carbon was measured by means of BOD_5 and COD. In Figure 2, the concentration and the removal efficiencies related to the COD and BOD_5 are reported. The results showed that the concentration of COD decreases in the permeate according to the HRT; achieve 128 mg/L during 24 h of the HRT. It should be noted that more than 95% of COD was removed by the MBR process. The relatively high effluent concentrations of COD can be attributed to the presence of slowly biodegradable and non-biodegradable compounds in the fish caning wastewater. In the same way, the concentration of BOD_5 decreases to 280, 160, 80 and 40 mg/L respectively for HRT of 12, 15, 20 and 24 h. The elimination rate of BOD_5 was 98% at the higher HRT (24 h). The results show that, the highest COD and BOD removal efficiency (up to 95%) corresponds to the highest HRT, which means that, if necessary, higher hydraulic retention times could be used, but greater aeration tanks would be required. These results are consistent with the results reported by Capodici *et al.* (Capodici *et al.* 2018) in their study, they confirmed that a sufficient HRT is required to ensure the process effectiveness. In this study, it needed to be more than 24 h.

3.3.2. TSS and VSS elimination

The TSS and VSS parameters represent the suspended fraction of organic pollution. The concentration and the removal efficiencies related to the TSS and VSS are reported in Figure 3. The results showed a reduction in the concentration of TSS and VSS in the permeate, reaching respectively 39, 28, 19 and 3 mg/L for the HRT of 12, 15, 20 and 24 h for TSS. In the same way, the concentration of VSS decreases to 26, 18.4, 16 and 9.3 mg/L respectively for HRT of 12, 15, 20 and 24 h. Therefore, the suspended solids concentrations in the filtration permeate was 99.5% for TSS and 100% for VSS. These two parameters therefore behave the same regardless of where the pollution is taken from within the production chain, and they probably have no direct influence on the other parameters. However, the suspended biomass fraction grew as dispersed biomass that did not settle (Muthukumaran & Baskaran 2013). Despite that biomass in the MBR was not purged throughout this period, suspended biomass concentration in the reactor attained low values, up to 10 g TSS/L.

3.3.3. Nutriments removal

To evaluate the nitrogen removal efficiency of each compartment of MBR process, nitrate (NO_3^-) and total Kjeldahl nitrogen (TKN) concentrations were analyzed after each steps treatment and in deferent HRT (Table 5). The results showed that the concentration of NO_3^- decreases in the permeate according to the HRT; achieve 12 mg/L during 24 h of the HRT, with a removal efficiency of 95%. Nitrate is usually the final product of the nitrification process, although nitrite accumulation occurs if operational conditions, especially pH and ammonia concentration, are high enough to result in the inhibition of *Nitrobacter* by free ammonia (Mao *et al.* 2020). The TKN values in the AS effluent and filtration permeat are shown in Table 5. The TKN in the AS effluent was progressively decreased from 1.3 to 0.7 mg/L and from 0.7 to 0.4 mg/L as a function of HRT. The fraction of TKN as ammonia changed from 93% in the AS effluent to 96% in the permeat, showing that almost the entire organic fraction of in the aerobic reactor was hydrolysed to ammonia nitrogen. TKN of the permeat is lower due to

Table 5 | Concentrations and removal efficiencies rate of nutriments (nitrogen and phosphorus)

		Activated Sludge Effluent				MBR Permeate			
HRT (hour)		12 h	15 h	20 h	24 h	12 h	15 h	20 h	24 h
NO_3^-	Concentration (mg/L)	50	32	24	14.6	20.3	15.6	13.2	12
	Removal efficiency (%)	59	74	80	88	83	87	89	90
TKN	Concentration (mg/L)	1.3	1.1	1	0.7	0.7	0.6	0.5	0.4
	Removal efficiency (%)	93	94	95	96	96	97	97	98
PO_4^{3-}	Concentration (mg/L)	8	6.8	2.3	1.5	1.4	1.2	1.18	0.4
	Removal efficiency (%)	28.8	40	79.5	87.1	87.5	89.3	89.5	97

the nitrogen requirement for biomass synthesis and the nitrification process (Perez de Ortiz 2008). These results are consistent with the results reported by Jemli *et al.* (2015) in their study, they found that the TKN concentration in the permeate decreased reaching an average removal efficiency of 92%.

The orthophosphate (PO_4^{3-}) in the AS effluent decreased progressively from 8 to 1.5 mg/L and from 1.4 to 0.4 mg/L in the filtration permeate during 24 h of the HRT, with removal efficiency of 97% at the end of the MBR treatment. Phosphorus (P) is an essential nutrient to the growth of microrganisms. The usual forms of phosphorus found in aqueous solutions include orthophosphate, polyphosphate and organic phosphate (Alves 2014). The orthophosphate is available for biological metabolism without further breakdown. Polyphosphates and organic phosphate undergo hydrolysis in aqueous solutions and revert to orthophosphate forms; however, this hydrolysis is usually quite slow (Lin 2007).

3.3.4. TDS reduction

TDS describes all solids (including mineral salts) dissolved in wastewater (Choo 2019). TDS is also a measure of all dissolved particles smaller than 2 microns in wastewater samples tested. This includes all inorganic and organic substances in the ionic molecule. The concentration and the removal efficiencies related to the TDS are reported in Figure 4. The results showed that the concentration of TDS decreases in the permeate according to the HRT; achieve 678 mg/L during 24 h of the HRT. It should be noted that more than 95% of TDS was removed by the MBR process.

According to the EPA (2012), removal of these solids is accomplished by discharging a portion of the cooling water, referred to as blow-down water, which is usually treated by a chemical process and/or a filtration process before disposal for wastewater treatment (Guimarães *et al.* 2018).

In addition, the TDS represent an aggregate measure of all dissolved cations and anions in water and is mainly related to electrical conductivity (EC). Van Niekerk *et al.* reported that there is a relationship between TDS and EC (Van Niekerk *et al.* 2014). Therefore, EC is a useful surrogate for TDS. EC is more rapidly and easily measurable with reasonably-priced equipment. The CE results show that the reduction is high (up to 76%) for 24 h of HRT and the EC and TDS were related at a very high level (Table 6). EC depends on the presence of soluble ions in the solution and organic matter. However, when the organic load decreases significantly, the conductivity decreases.

Table 6 | Electrical conductivity reduction as a function of HRT

HRT (hour)	Conductivity in supernatant of aeration tank (μS/cm)	Conductivity in permeate (μS/cm)	Total reduction rate (%)
12	3,890	1,822	58,1
15	2,434	1,565	64,0
20	2,311	1,392	68
24	1,828	1,059	76

3.3.5. Analysis of MBR effluent quality

In regulating the discharge and the reuse of treated wastewater, chemi-cal parameters must be considered alongside the biological parameters. These settings are also related to the protection of health and the environment (soil, water, etc.). The effluent quality of the MBR effluent is listed in Table 7. The important chemical parameters to consider are the following: biodegradable organic compounds (COD and BOD_5), TSS, TKN, pH and conductivity. As shown, the value of these parameters in the MBR permeate were, 128, 40, 9.3 and 0.34 mg/L, respectively. The pH was 8.6 and the conductivity was 1,059 μS/cm. The concentrations of these parameters are less than the value limit of Moroccan effluent discharge (MDCE 2014). The chlorine, orthophosphate and oil and grease parameters are also pollution indicators, they comply with Moroccan rejection standards. In water, chlorine forms with organic nitrogen toxic chloramines and chlorophenols. In addition, in a small concentration, this compound is very effective in reducing the number of filamentous organisms present in the mixed liquor (Gerardi 2003).

Moreover, the results were compared to water standards reuse according to their intended use (Table 5). In this case, only the conductivity, TSS and NTK concentrations were complied with Moroccan reuse standards (SEEE

Table 7 | Water standards for effluent discharge and reuse

Parameter	Unit	MBR effluent	Discharge limit values[a]	Reuse limit values[b]
pH value	–	8.6	5.5–9.5	6.5–8.4
Temperature	°C	17.5	30	35
Conductivity	μS/cm	1,059	2,700	1,200
COD	mg/L	128	500	100
Five-day BOD	mg/L	40	100	20
TSS	mg/L	9.3	100	<50
TKN	mg/L	0.34	40	<5
Oil and grease	mg/L	41	30	n.e.
Chlorine (Cl^-)	mg/L	0,29	0.2	n.e.
Orthophosphate	mg/L	0.34	15	n.e.

n.e., not established.
[a]These are the maximum admissible values for discharge into the natural environment, Morocco.
[b]This is the maximum permissible values according to Directive FAO and Water reuse standard for irrigation, land watering etc., Morocco.

2007). However, the pollution of fish canning wastewater appears mainly linked to the heavy load of inorganic, suspended matter and salt used in the process for preserving fish. The high sodium chloride load of the final waste seems to be the main environmental problem.

4. CONCLUSION

An MBR was used to treat wastewater generated in fish canning industry. The study was investigated in order to obtain water with quality requirements to discharge value and standards reuse in the industrial process. The proposed treatment, combining sedimentation/flotation, aerobic biological degradation by activated sludge, and ultrafiltration, proved to be very effective. In the sedimentation/flotation stage, removals of 65% for TSS, 1 and 5% for BOD and COD and 57% for oil and grease were observed. In the AS effluent, the removal was in the range of 87–95% for the majority of parameters, whereas membrane ultrafiltration removed 99% of suspended solids, about 97% of organic carbon, 96% of TDS (salinity) and above 96% of the analyzed ions. However, the implementation of this process at large scale will bring both environmental and economic benefits, since it allows the reduction of the effluent to be discharged and the water consumption, thus decreasing the associated costs. Furthermore, since the fish canning industry consumes large amounts of water and the demand for manufactured fish goods has increased, the implementation of water management and wastewater reuse techniques must be recognized and stimulated, as industrial wastewater reuse is an excellent alternative to the preservation of fresh drinking water.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICTS OF INTEREST STATEMENT

The authors declare there is no conflict.

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