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Urban wastewater reuse using a coupling between nanofiltration and ozonation: techno-economic assessment.

Julie Mendret*, Antonin Azais, Thibault Favier, Stephan Brosillon

Institut Européen des Membranes, IEM – UMR 5635, ENSCM, CNRS, Univ Montpellier, Montpellier, France

Abstract: Combination of nanofiltration and ozonation was investigated for the treatment of urban wastewater. First objective was to demonstrate that nanofiltration can be used instead of reverse osmosis as it enables good rejection rates with reduced cost because of lower operating and maintenance costs. In this way, this paper presents an economic and technical evaluation of the proposed coupling where ozonation is used to treat retentates from nanofiltration. Reverse Osmosis System Analysis (ROSA) software was applied to simulate the filtration design. The effect of membrane choice on specific energy consumption, capital, operation and maintenance costs and scaling potential was investigated. It was demonstrated that using nanofiltration instead of reverse osmosis enable cost saving of 35 k$/year for 125 m^3/h. Second objective was to evaluate the impact of the treatment of retentates by ozonation on the global cost. It was highlighted that the coupling would be an acceptable solution from an economic point of view for wastewater reuse. The possible reuse of both permeate and concentrate enable an operating cost saving of 15.4 k$/year for 125 m^3/h. An optimum recovery rate of 80 % was found for which cost of membrane process is balanced by a decrease in the cost of ozonation.

Keywords: nanofiltration, ozonation, wastewater reuse, economic analysis.

Highlights:

Nanofiltration is a possible alternative to reverse osmosis for wastewater reuse

Nanofiltration combined to ozonation is a competitive process for wastewater reuse

Nanofiltration is a barrier for micropollutants with reasonable operating costs

Coupling nanofiltration and ozonation results in cost saving due to reuse possibility

1. Introduction.

Water resources management is a major challenge in economic development and public health. The demographic growth and associated economic development will further exacerbate the situation in the next twenty years by increasing water demand while discharging more and more polluted effluents. It is therefore necessary, in addition to the draft of very strict regulations, to protect the quality of the resource, to challenge the marine or brackish resources desalination by developing cost-effective systems, and also to define new and relevant treatment processes for urban wastewaters allowing direct wastewater reuse and improvement of water resource protection. This concept of wastewater treatment allowing direct recycling is an issue already tackled and resolved in part in the industrial environment and in some "early bird" countries (Singapore for drinking water and Tunisia, California and Florida for irrigation water) (Wintgens et al., 2005). However, it is still secondary in densely populated urban environments, where the major concern is to comply with standards imposed with regards to the fragility or constraints of the receiving environment. Nevertheless, this reuse becomes one of the essential points in optimal management of a freshwater resource, especially when considering its global cost, less than half the cost of desalination (Cote et al., 2005).

After massive investment in energy-intensive desalination processes which increased the global production capacity of desalinated water by 67% to 52 million $m^3.d^{-1}$ (GWI, 2009), governments are now looking at overall water efficiency and start to prioritize water reclamation and reuse as a solution to water scarcity since water reclamation requires only a third to a fourth of the energy needed for reverse osmosis (RO) desalination. While the combined approach of seawater desalination and water reuse has proven to be successful in a number of coastal sites such as Barcelona, Singapore, land locked cities have to focus completely on water reuse for irrigation or aquifer recharge, sometimes even accepting direct potable reuse as practiced in Windhoek, Namibia (Bunani et al., 2015, Bellona et al;, 2012). With a closed water loop, land locked cities require high-quality water reuse systems mostly based on membrane technology as affordable and efficient barrier.

Municipal wastewater is an alternative water source but increasingly contaminated with toxic organic pollutants (Schwarzenbach et al., 2006). The occurrence of organic micropollutants (OMPs) in the aquatic environment, often called 'emerging pollutants' as they became increasingly detectable since the last two decades, has been identified as a global issue. Research has shown the ubiquitous presence of a broad range of pharmaceuticals, personal care products and industrial chemicals in the water cycle, particularly in the effluent of wastewater treatment plants (WWTP) and surface waters in low concentrations from $ng.L^{-1}$ to $\mu g.L^{-1}$ (Jorgensen and Halling-Sorensen, 2000; Heberer, 2002, Wode et al., 2015, Hermes et al., 2018). Also, important concerns have been raised regarding that continuous discharge of antibiotics and anti-microbial products into aquatic environment may facilitate the development or proliferation of resistant strains of bacteria (Kim and Aga, 2007). Indeed, the emergence and spread of antibiotic resistant bacteria have been classified by the World Health Organization as one of the three biggest threats to public health in the 21st century. Moreover, chronic toxicity effects have been reported for aquatic organisms exposed to OMPs at trace concentrations. Current technologies in existing WWTPs struggle to eliminate OMPs or to limit their concentration and toxicity sufficiently to comply with the thresholds in new regulations. These evolutions are driving the urban wastewater treatment to come up with advanced technologies like RO or activated carbon (AC) are able to effectively remove OMPs without chemicals addition and limiting by-products formation (Gomez et al., 2012, Paredes et al., 2018). Nonetheless, RO is expensive because of high operating pressure and is very sensitive to fouling thus requires a high level of water pretreatment or works best on groundwater or low solids surface water. AC could be efficient but its performances are greatly affected by variations in pH, temperature, and flow rate as it could release or desorb (Benstoem et al., 2017). To recover fresh reusable water from urban wastewater treatment, efficient additional treatment processes which are economically viable are required and thus must be developed.

Nanofiltration (NF) has been proved to be efficient as a tertiary treatment as it enables high rejections of small organic molecules (around 200-300 Da) due to size exclusion and electrostatic or hydrophobic interactions. Moreover, this membrane technology requires operation pressure significantly lower than traditional RO processes and without the need of further permeate remineralization. Hence, NF provides an interesting alternative to RO for wastewater reuse with a number of advantages: lower energy consumption due to lower pressure requirement, less chemical additives, lower rejection of monovalent salts and thus less problematic membrane concentrates and finally less heavy post treatment of re-mineralization (Bellona and Drewes, 2007). However, two main limitations of NF application in WWTP are i) membrane fouling which affects directly process performances and costs; and ii) the disposal of retentates which contains a wide range of concentrated organic pollutants. Indeed, NF applied to a polluted water provides a large volume of practically clean water (permeate) that can be reused immediately and a concentrated stream that requires further treatment. The recovery ratio for NF is normally 50-85% which corresponds to a volume reduction factor (VRF) of 2 - 7 and results in a waste stream which could be returned to the biological treatment or could be discharged or treated depending on regulations and possibilities.

However, retentate recirculation upstream of the biological treatment is not possible in the long term as salinity greater than 5 $g.L^{-1}$ may inhibit biological activity (Reid et al., 2006). In addition, the recirculation of NF concentrates within a MBR significantly increases its clogging (Kappel et al., 2014). Finally, direct discharge into surface or marine waters is not easy to encourage given the average composition of the urban retentates i.e. conductivities of 2 to 20 $mS.cm^{-1}$, CODs ranging from 20 to 55 $mg.L^{-1}$ and levels of organic micropollutants multiplied by 3 to 7 (Benner et al., 2008; Solley et al., 2010). Hence, treatment of membrane concentrates is recommended.

With this view in mind, ozonation was found to degrade efficiently most OMPs present in treated wastewater through the strong oxidative properties of ozone (applied dose: 3–8 mg $O_3.L^{-1}$) and of the hydroxyl radicals produced spontaneously during its decomposition. AOP based on ozonation could thus be used to treat membranes rejection streams (retentate) containing high concentration of harmful organics. The general advantages of the combined process NF/AOP or AOP/NF with recirculation of the concentrate over direct treatment would be, depending on the configuration: (i) lower ozonation treatment time, (ii) more efficient reagent consumption, (iii) treatment of lower flow rate, (iv) higher permeate flow (Van Geluwe et al., 2011; Park et al., 2017). Concentration of OMPs can be reduced to a desired level by NF or ozonation separately, but only few researches took advantage of these two wastewater treatment process to produce high quality effluent.

Over past years, objective of our researches is to prove that the combination of NF and ozonation could be a promising, efficient and affordable solution for OMPs and global toxicity removal from urban wastewater while reducing costs and limitations occurring when the two processes are carried out separately, making wastewater reuse possible and safe for various applications (Azais et al., 2014; Azais et al., 2016; Azaïs et al., 2017). The final objective is to design novel integrated process schemes combining NF and ozonation with structural (flow-sheet) and parameters optimization of these processes. One of the design objectives of the integrated processes should be the minimization of liquid wastes, thus reducing the load of the following waste treatment stage. Any new NF membrane plant needs an evaluation of the membrane performance in order to evaluate the number of NF elements and type of membrane, this is essential in the prediction of the initial cost of the plant. In addition, costs of emerging processes have to be evaluated. To our knowledge, this has never been done for such coupling applied to wastewater reuse. In this way, the specific objective of this paper is to present an economic and technical evaluation of the proposed coupling between NF and ozonation where ozonation is used to treat retentate from nanofiltration. Reverse Osmosis System Analysis (ROSA) software developed by Filmtec (Dow Water and Process Solutions, 2011) was applied in this study to simulate the proposed filtration design. The effect of membrane choice on power consumption in terms of specific energy consumption ($kWh.m^{-3}$), capital, operation and maintenance

(O&M) costs and scale potential was investigated. Interest of the use of NF instead of RO was analyzed by comparing both processes. A process analysis and a comparative economic analysis of capital and operating costs for both technologies were performed. Cost of ozonation was estimated based on experimental results concerning ozonation of real wastewater matrice concentrated by nanofiltration (Azais et al., 2016a). Thus, ozone doses are known for this effluent. Finally, impact of the treatment of concentrates by ozonation on the treatment costs was considered.

2. Material and methods.

2.1. Cost of nanofiltration

ROSA 9.0 software model was applied to predict water quality and flow rate for different membrane system configurations. ROSA software is a model designed by Nisan in 2005, applied to design and manipulate RO and NF systems taking into account effects of concentration polarization factor and hydrodynamic pressure drop in the concentrate-side on the membrane performance (Nisan et al., 2005). This software is often used in research work to simulate designs using reverse osmosis or nanofiltration plants (Yangali-Quintilla et al., 2010 ; Haryati et al., 2017).

The main inputs of the model include the amount of feed water and its chemical characteristics including the SDI range, feed water and concentrate feed pressures, temperature and pH. Then, setting up of the number of membranes, pressure vessels, type of membrane, and if needed booster pumps, is performed by ROSA. Depending upon the system, the goal is to minimize feed pressure and membrane costs whilst maximizing permeate quality and recovery. After performing calculations, ROSA produces a report predicting among others the water quality and flow rate.

Feed water whose characteristics were implemented in ROSA was the one used in previous studies (Azais et al., 2014; Azais et al. 2016 a), 2016 b), Azais et al., 2017). It is a real secondary effluent from a municipal WWTP located in the south of France (Baillargues) with a SDI=2.5 which is inferior to 3 as recommended for a direct use of the effluent as feed water for NF, without pretreatment. The membrane bioreactor (MBR) system incorporates immerged ZeeWeed 500 (GE Zenon) ultrafiltration

(UF) membranes in polyvinylidene difluoride (PVDF) which have a pore size of 0.035 microns. The total membrane surface area is about 7 605 m² and produces approximately 1 500 m^3 of permeate per day while operating at a mixed liquor total suspended solids (TSS) concentration of between 4 and 7 g L^{-1}. Summary of major constituents of the secondary effluent is presented in table 1 with intervals given with 95% confidence.

Table 1. Water quality of the WWTP effluent.

Parameters	Units	WWTP effluent
pH	-	7.69 ± 0.31
Conductivity	mS cm^{-1}	1.30 ± 0.22
Alkalinity	mg eq $CaCO_3$ L^{-1}	430 ± 80
COD	mg L^{-1}	18 ± 3
DOC	mg L^{-1}	6 ± 2
NO_3^--N	mg L^{-1}	2.0 ± 0.6
Total Nitrogen (TN)	mg L^{-1}	4.1 ± 1.6
PO_4^{3-}-P	mg L^{-1}	0.11 ± 0.08
Na^+	mg L^{-1}	152 ± 14
K^+	mg L^{-1}	22 ± 3
Ca^{2+}	mg L^{-1}	160 ± 20
Mg^{2+}	mg L^{-1}	6.6 ± 0.8
Cl^-	mg L^{-1}	267 ± 35
SO_4^{2-}	mg L^{-1}	89 ± 13
Ionic strength	mM	22 ± 5

No nitrites were detected in the effluent so that the value of TN could be due to the presence of ammonia because of incomplete nitrification. Regarding water pH and ionic composition, ROSA estimates Langelier Saturation Index (LSI) which is a measure for the saturation of $CaCO_3$ in water. The LSI should be used for low Total Dissolved Solids (TDS) ranges (<10 000 ppm). At the pH of saturation (pH_S), the water is in equilibrium with $CaCO_3$. To avoid calcium carbonate scaling by acid addition alone, the LSI in the concentrate stream must be negative. If a high quality scale inhibitor is used, the LSI in the concentrate stream can be increase up to 1.5. This will reduce or eliminate the acid consumption. Many inhibitors allow operation up to an LSI of < 1.8 in the concentrate. In this study the water is hard, so acid (H_2SO_4) was added. Finally, the low value of SDI will reduce fouling problems due to particulate matters which tends to accumulate at the membrane surface resulting in a membrane flux decline.

Capital cost of a NF plant is directly impacted by the design of the system. This issue should be taken into account into the system design as most companies try to reduce the capital cost. Moreover, maintenance cost of NF system can be reduced because of lower propensity to fouling of elements when correctly chosen. Thus, balance has to be found between design and maintenance costs to meet both economic and technical requirements.

A two or three stage configuration with a decreasing number of pressure vessels in parallel allows respecting the minimum concentrate flow rate constraint and other required hydraulic conditions in nanofiltration process. Consequently, this kind of configuration was chosen in this study in order to reach high conversion rates (> 80 %), obtain a reliable permeate quality without a severe fouling risk (figure 1). First stage is composed of N pressure vessels each including M elements in series. Next stage has N/2 pressure vessels and last stage N/4 pressure vessels. In the experimental study, three Volume Reduction Factor (VRF) were tested: 2, 5 and 10 which correspond to recovery rate (Y, defined as the ratio of permeate flow by feed flow) of about 50, 80 and 90 % respectively (Azais et al., 2016 a). To reach these recovery rates, number of stage has to be adapted. Y of 50 % was applied to the first stage and Y of 60% was applied to second stage thus resulting in global Y of 80 % for two stages. Y of 50% was applied on last stage resulting in final Y of 90 % close to VRF 10 which was obtained at pilot scale (Azais et al., 2016b). To reach these conversion rates, ROSA adjusted pressure at the entrance of each stage according to the number of stage and pressure vessels and type of membrane.

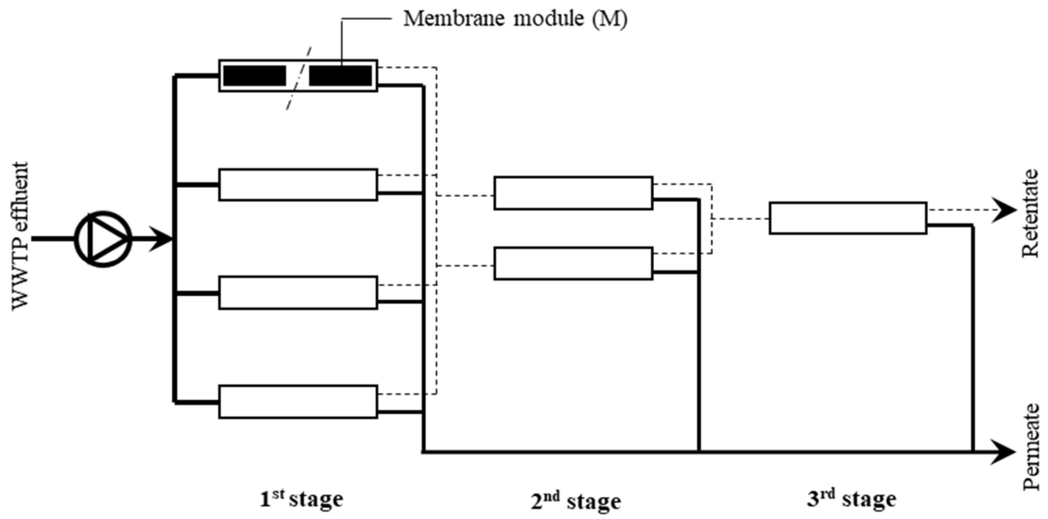

Figure 1. Configuration of the nanofiltration unit (number of pressure vessels: first stage N=4, second stage N/2, third stage N/4).

The average flux of the entire system, i.e. the system permeate flow rate related to the total active membrane area of the system, is a characteristic parameter of a given design. The system flux is a useful number to quickly estimate the required number of elements for a new project. In this study, the feed water flow of Baillargues wastewater treatment plant (125 $m^3.h^{-1}$) was chosen as inlet feed flow in ROSA. Tested recovery rates (Y) were chosen as they correspond to VRF reached in a previous study (Azais et al., 2016 a)).

Among elements proposed by ROSA, NF 90-400 was first chosen. NF90-400 element is a high area, high productivity element designed to remove a high percentage of salts, nitrate, iron and organic compounds. The high active area membrane combined with high permeability of the membrane allows the removal of these compounds at low operating pressure. The membrane active area is 400 square feet; the diameter and length are respectively 7.9 and 40 inch. Then, same calculation were performed for NF 270-400 (same dimensions as NF 90-400) and for BW 30-400 (brackish water reverse osmosis membrane, same dimensions as NF 90-400).

Cost of NF 90-400 element and pressure vessel was estimated to respectively 850 and 1 500 $. Price of kilowatt-hour was based on current rate in France which is about 0.09$.$kWh^{-1}$ in accordance with Plumlee et al. (2014) (Plumlee et al., 2014). Moreover, annual membrane replacement rate was taken equal to 10% as estimated by Yangali-Quintanila et al. (2010) (Yangali-Quintanila et al., 2010). To calculate the annual capital costs, project lifetime was taken equal to 10 years (life time of membranes) with an interest rate of 7%.

Finally, two uses of the treated water (permeate) were considered: irrigation according to the FAO recommendations and the aquifer recharge in agreement with the Groundwater Recharge Requirements proposed by the California Department of Public Health (CDPH). The standard values are presented in a previous study (Azais et al. 2014).

2.2. Cost of ozonation

Economic evaluation of NF retentate ozonation is necessary so as to define an optimal conversion rate which assures a compromise between permeate flux and retentate treatability. This economic evaluation was done on the basis of transferred ozone dose in concentrated real wastewater which guarantees a 90% elimination of three pollutants among the four chosen in our previous studies (see Azais et al., 2016 a)).

As only little ozone doses are required to treat retentates of this study, on site oxygen generation using Pressure Swing Absorption (PSA) was chosen. This is ideally suited to supply an ozone generator with medium frequency (60 to 1 000 Hz). Specific energy for ozone production was estimated to 10 $kWh.kgO_3^{-1}$ (Rosenfeldt et al., 2006). According to Margot et al. (2013), the transfer efficiency of ozone into the dissolved phase in ozonation pilot plant is between 70 to over 90% depending on the gas flow (Margot et al., 2013). In this study, it was fixed to 95%. Investment cost was taken equal to 100 $\$.gO_3^{-1}.h^{-1}$ as suggested by ozone water treatment plants manufacturers and suppliers. The operational cost was then evaluated in this study. Finally, as cooling water is used in a close loop system, this item of expenditure was neglected.

Considering the different input parameters (economic as well as process design and performance data) and their variability in the cost assessment, the maturity level is fixed to 5 (corresponding to 10 % of complete definition) with an accuracy range of – 50 % to 50 % according to the AACE (American

Association of Cost Engineering) cost estimate classification matrix. A narrow accuracy range was defined since the approach developed in this study could easily lead to a feasibility study or even to a pre-budget authorization (maturity level 4 or 3) as the membrane and ozonation technologies are mature and controlled.

3. Results and discussion

3.1. Optimization of nanofiltration design for NF-90

To verify that the chosen configuration (figure 1) is well adapted to the targeted conversion rates, simulations were done by imposing the three recovery rates for different configuration (one, two or three stages). For this, value of the number of pressure vessel in first stage (N) was modified between 10 and 30. At first approximation, number of elements in a pressure vessel was fixed to 7 as usually encountered at industrial scale (Van der Meer et al., 1998). For each simulation, total power to be supplied by the pumps was noticed so as to determine specific energy consumption (kWh/m^3) of each configuration. In addition, the rejection of total dissolved solids (TDS) was estimated as a function of N. Figure 2 presents the results of these simulations.

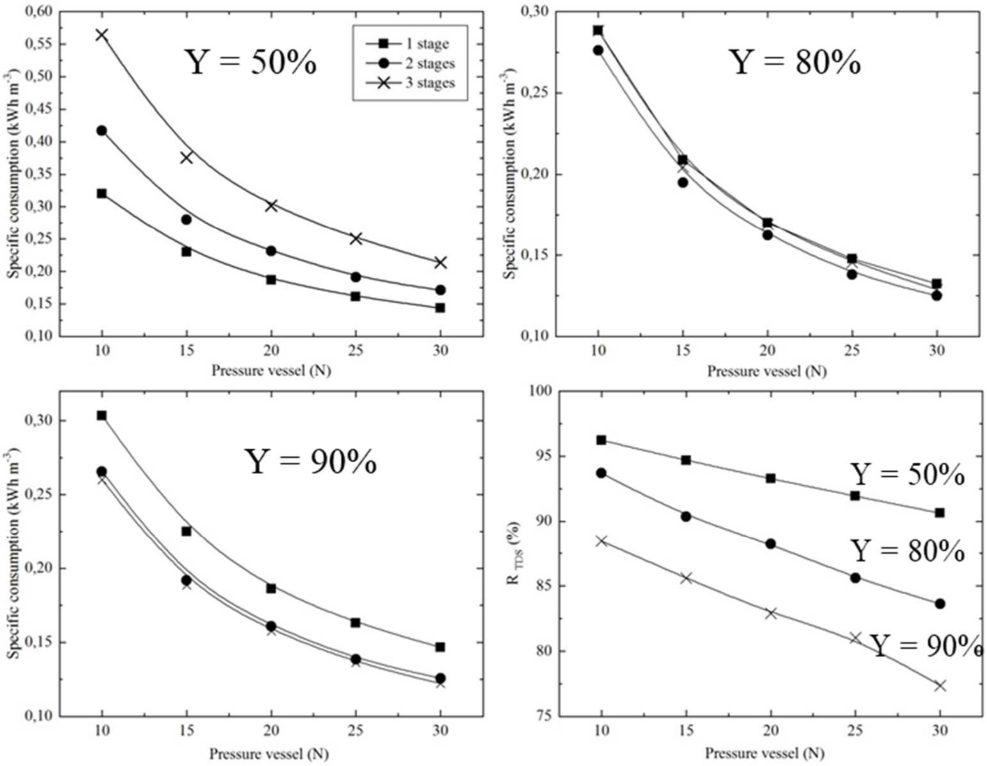

Figure 2. Influence of pressure vessel number in the first stage (N) (number of elements M=7).

Regarding specific energy consumption, it appears that each recovery rate is adapted to a certain number of stages. A single stage system is more energy efficient when a conversion rate of 50% is applied. Two stages are more energy efficient to reach Y=80% and three stages to reach Y=90%. Indeed, at low conversion rate, the concentrate flow rate is high thus when using only one stage pressure drop along the unit is limited. On the contrary, for higher conversion rate (lower concentrate flow rate), the decrease of hydraulic pressure due to the increase of stages is more interesting from an energetic point of view. In the next part of the article, a conversion rate of 50%, 80% and 90% was thus reached with respectively one, two and three stages. On figure 2, it appears that specific energy consumption decreases with increasing number of pressure vessels in the first stage. For example, feed pressure decreases from 4.6 to 2.1 bars when N increases from 10 to 30. Nonetheless, an increasing number of pressure vessels will have a direct consequence on capital and maintenance cost. Moreover, the decrease of inlet pressure leads to a decrease in TDS rejection because of high diffusive transfer.

Number of pressure vessels influences both TDS rejection and energy consumption. Thus, it has to be optimized so as to minimize operating costs. Annual power cost ($ year^{-1}) was obtained by multiplying specific energy consumption (kWh m^{-3}) by the price per kilowatt hour ($ kWh^{-1}) and by annual permeate flow (m^3 year^{-1}). Maintenance costs were obtained by multiplying total number of elements (M) by replacement rate of 10 % and by the price of one element. These maintenance costs were calculated for each conversion rate (50, 80 and 90 %) by varying the number of pressure vessels in the first stage, N, thus varying the total number of pressure vessels. Figure 3 presents operating costs as a function of N and number of stages. Number of elements (M) in a pressure vessel was fixed to 7.

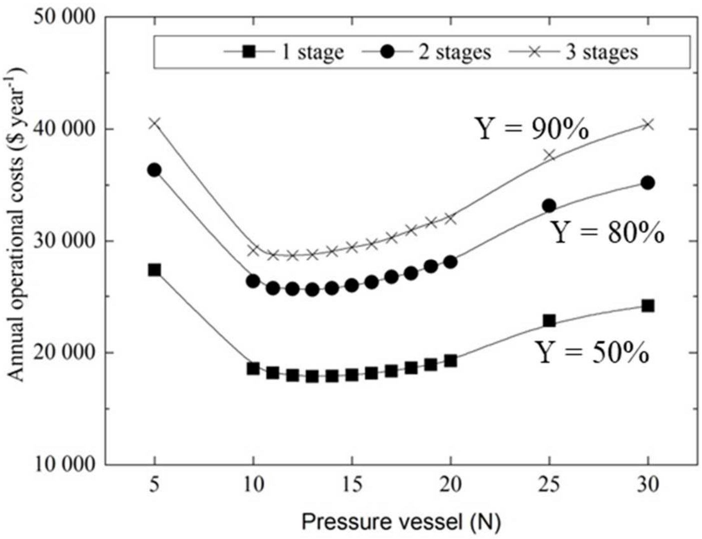

Figure 3. Influence of number of pressure vessel in the first stage (N) and number of stages on annual operating costs (number of elements M=7).

For each case, annual operating costs show a minimum for an optimum number of pressure vessel which is equal to 13 for Y=50% and Y=80% and 12 for Y=90%. In the rest of the study, N was taken equal to 12 so has to have 6 and 3 pressure vessels respectively in the second and third stage.

Concerning the number of elements per pressure vessel (M), van der Meer et al. (1998) showed that even though the classical system configuration of pressure vessels in RO and NF systems is a multistage design with a number of membrane modules per pressure vessel of 6 up to 8, this design trend results in high pressure losses because of high feed velocities and long feed/brine channel lengths (van der Meer et al., 1998). The authors showed that an increase in permeate productivity of 20% can be achieved by lowering the number of membrane modules per pressure vessel. In contrast to the hydraulic optimum, they found an economic optimum between 3 to 4 elements per pressure vessel. Research of optimal number of elements M was conducted using same approach as for N. The pressure vessel number was 12 in the first stage, 6 in the second stage and 3 in the last stage. M was varied between 2 and 8 (maximum authorized by ROSA). Then, operating costs were evaluated for each configuration. Results are presented in figure 4.

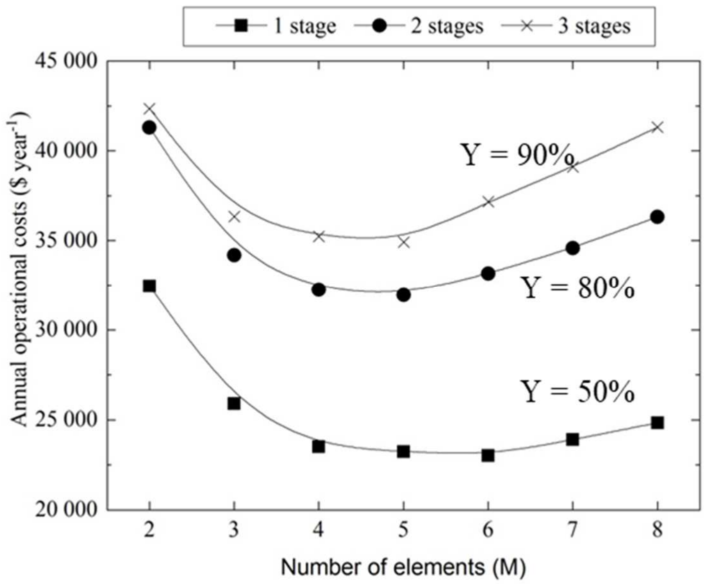

Figure 4. Influence of number of elements (M) and stages on annual operating costs (total number of pressure vessels N=12).

The economic optimum is M=6 elements for a single stage and M=5 for two and three stages which is in accordance with van der Meer et al. (1998) (van der Meer et al., 1998). At low number of membrane modules, the permeate flow rate decreases as a result of the increasing concentration polarization in this region. This effect becomes more pronounced at higher recoveries, caused by increasing osmotic pressure differences across the membrane surface, as a result of the increasing salt concentrations at the feed/brine side of the membranes at high recoveries. In the rest of the paper, number of elements (M) was taken equal to 6 for one stage and to 5 for two or three stages.

3.2. Economic analysis of filtration

Table 2 presents the economic analysis of the process in term of final membrane capital cost (pressure vessel and membrane module) with one, two or three stages as shown on figure 1.

Table 2. Economic analysis of membrane capital cost for nanofiltration operation with NF90-400 elements.

Targeted conversion rate	Y = 50 %	Y = 80 %		Y = 90 %		
Stage	**1**	**1**	**2**	**1**	**2**	**3**
System details						
Feed flow ($m^3.h^{-1}$)	125	125	62.5	125	62.5	25
Permeate flow ($m^3.h^{-1}$)	62.5	62.5	37.5	62.5	37.5	12.5
Retentate flow ($m^3.h^{-1}$)	62.5	62.5	25	62.5	25	12.5
Recovery rate (%)	50	50	60	50	60	50
Total membrane area (m^2)	2 676	3 344		3 902		
Average permeate flux ($L.h^{-1}.m^{-2}$)	23.4	29.9		28.8		
Feed pressure (bar)	5.04	5.55	7.52	5.55	7.52	6.75
Economic analysis						
Pressure vessel (quantity)	12	12	6	12	6	3
Pressure vessel cost ($)	18 000	27 000		31 500		
Membrane module (quantity)	72	60	30	60	30	15
Membrane module cost ($)	61 200	76 500		89 250		
Membrane capital cost ($)	**79 200**	**103 500**		**120 750**		
Annual membrane capital cost ($/year)	**11 276**	**14 736**		**17 192**		
Annual membrane capital cost ($/m³)	**0.020**	**0.016**		**0.017**		

The economic analysis of the process shows that an optimum conversion rate of 80% leads to a membrane capital cost of 0.016 $\$/m^3$. The increase in recovery rate enables a decrease in operating cost per cubic meter of treated water. Nonetheless, this increase in conversion rate would result in an increase in polarization concentration at membrane surface resulting in severe risk of scaling and a lower salt rejection. Indeed, TDS rejection decreases from 94.2% to 89.0% when increasing Y from 50 to 90%. However, this strategy avoids a too high demineralization of permeate water. In addition, nitrate concentration in permeate for Y=90% is still inferior to the recommendation for groundwater recharge (Bellona et al. 2012).

As mean value of LSI of feed water was 1.3 which is superior to the recommended value of 1, use of acid was included in the economic evaluation of the process. ROSA proposes two agents (HCl, H_2SO_4) which enable the LSI to be lowered close to zero value by modifying pH. In this study, H_2SO_4 was chosen as sulfate ions are retained by more than 90% by NF-90 membrane and thus did not impact the permeate quality. The targeted LSI was 0.2 which required 140.5 mg of H_2SO_4 per liter of inlet effluent. This represented about 84 m^3 of H_2SO_4 (96%) per year. As the cost of a cubic meter of H_2SO_4 is about 730 \$ thus annual expense for scaling control by acidification was about 61 000 \$ (Table 3). Finally, acid cost was about four times operation and maintenance expenses. Proportion of electrical cost slightly increases when increasing conversion rate (figure 5).

Table 3. Economic analysis of operating and maintenance cost for nanofiltration operation with NF90-400 elements at several conversion rates.

Targeted conversion rate	**Y = 50 %**	**Y = 80 %**	**Y = 90 %**
Stage	**1**	**2**	**3**
Specific consumption (kWh.m^{-3})	**0.35**	**0.316**	**0.295**
Electric cost ($.year^{-1})	**17 246**	**24 913**	**26 165**
Membrane replacement ($.year^{-1})	**6 120**	**7 650**	**8 925**
Acid ($.year^{-1})	**61 000**	**61 000**	**61 000**
O&M cost ($/year)	**84 366**	**93 563**	**96 090**
O&M cost ($/m^{3}permeate)	**0.15**	**0.11**	**0.10**

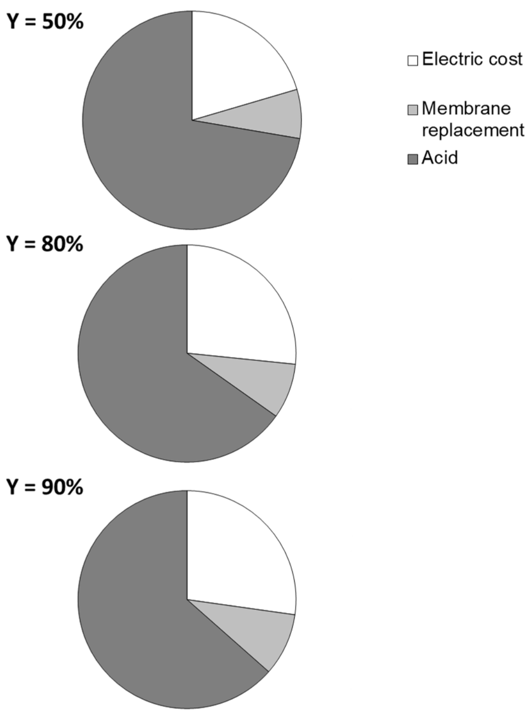

Figure 5. Part of operating and maintenance costs for several conversion rates.

3.3. Comparison with loose-nanofiltration and reverse osmosis process

To highlight the interest of using nanofiltration instead of reverse osmosis, same simulations were performed with NF-270, a loose NF membrane, and BW-30 a RO membrane (Dow Filmtec). Price of NF-270 and BW-30 elements were respectively 900 \$ and 680 \$ (Yangali-Quintanilla et al., 2010). Table 4 presents the comparison of these processes for Y=80%. As expected, use of RO enables capital cost saving of about 2-3 k\$ per year because of lower prices of RO equipment.

Table 4. Comparison of the membrane capital cost of nanofiltration and reverse osmosis for Y=80%.

Membrane	Membrane capital cost (\$)	Annual membrane capital cost (\$/year)	Annual membrane capital cost (\$/m^3)
NF90 - 400	103 500	14 736	0.016
NF 270 - 400	108 000	15 377	0.017
BW30 - 400	88 200	12 558	0.014

Table 5 presents the comparison of operating and maintenance cost of nanofiltration and reverse osmosis processes. For high conversion rates, booster pumping may be required between stages. Operating and maintenance cost varies between 0.09 and 0.19 \$/m^3 depending on the conversion rate and membrane type. These costs determined by ROSA are in accordance with literature concerning NF/RO systems (Cote et al., 2005; Englehardt et al., 2013). Globally the cost decreases when increasing membrane permeability (L_p) which is as follow in l/h/m²/bar (LMHB): Lp BW30-400 = 3.2 LMHB < NF90-400 = 10.8 LMHB < NF270-400 = 13.6 LMHB. For Y=80% and an inlet flow rate of 125 m^3/h, 29.6 k\$/year and 35.2 k\$/year could be saved using respectively NF90-400 and NF270-400 instead of BW30-400. This is in accordance with other authors (Yangali-Quintanilla et al., 2011; Bellona et al., 2012). Indeed, even if RO elements are less expensive than NF elements, energy requirement of the pumps was twice or third the one needed with NF processes which make operating and maintenance costs of RO process higher. Finally, type of NF (loose or tight NF) does not seem to influence considerably the process cost. Nonetheless, this influences performances and rejection rates of the process as it can be seen in table 6.

Table 5. Comparison of operating and maintenance cost of nanofiltration and reverse osmosis processes.

Targeted conversion rate	Y = 50 %	Y = 80 %		Y = 90 %		
NF270-400						
Feed pressure (bar)	3.57	3.91	4.77	3.91	4.77	3.89
Pumping power (kW)	16.97	23.87		24.41		
Specific consumption ($kWh.m^{-3}$)	0.27	0.24		0.22		
Electric cost ($\$.year^{-1}$)	13 379	18 819		19 245		
Membrane replacement ($\$.year^{-1}$)	6 480	8 100		9 450		
Acid ($\$.year^{-1}$)	61 000	61 000		61 000		
O&M cost ($/year)	**80 859**	**87 919**		**89 695**		
O&M cost ($\$.m^{-3}$)	**0.12**	**0.10**		**0.09**		
NF90-400						
Feed pressure (bar)	5.04	5.55	7.52	5.55	7.52	6.75
Specific consumption ($kWh.m^{-3}$)	0.35	0.32		0.29		
Electric cost ($\$.year^{-1}$)	17 246	24 913		26 165		
Membrane replacement ($\$.year^{-1}$)	6 120	7 650		8 925		
Acid ($\$.year^{-1}$)	61 000	61 000		61 000		
O&M cost ($/year)	**84 366**	**93 563**		**96 090**		
O&M cost ($\$.m^{-3}$)	**0.15**	**0.11**		**0.10**		
BW30-400						
Feed pressure (bar)	11.18	13.2	17.22	13.2	17.22	15.68
Specific consumption ($kWh.m^{-3}$)	0.80	0.71		0.63		
Electric cost ($\$.year^{-1}$)	39 428	56 071		56 071		
Membrane replacement ($\$.year^{-1}$)	4 896	6 120		7 140		
Acid ($\$.year^{-1}$)	61 000	61 000		61 000		
O&M cost ($/year)	**105 324**	**123 191**		**124 211**		
O&M cost ($\$.m^{-3}$)	**0.19**	**0.14**		**0.13**		

Table 6. Comparison of performances and rejection rates of nanofiltration and reverse osmosis process.

Targeted conversion rate	Y = 50 %	Y =80 %		Y = 90 %		
Stage	**1**	**1**	**2**	**1**	**2**	**3**
NF-270						
Feed pressure (bar)	**3.57**	**3.91**	**4.77**	**3.91**	**4.77**	**3.89**
Osmotic pressure (bar)	0.66	0.95		1.27		
Average permeate flux ($L.h^{-1}.m^{-2}$)	23.4	29.92		28.84		
TDS rejection (%)	**62.5 ± 2.0**	**64.0 ± 1.5**	**70.0 ± 1.2**	**64.0 ± 1.5**	**70.0 ± 1.2**	**70.2 ± 0.1**
Average permeate conductivity ($\mu S.cm^{-1}$)	868.48	927.77		1037.4		
Permeate nitrate concentration ($mg.L^{-1}$)	4.93	4.78	5.2	4.78	5.2	6.36
pH of permeate	6.15	6.2		6.28		
NF-90						
Feed pressure (bar)	**5.04**	**5.55**	**7.52**	**5.55**	**7.52**	**6.75**
Osmotic pressure (bar)	0.78	1.51		2.52		
Average permeate flux ($L.h^{-1}.m^{-2}$)	23.4	29.9		22.44		
TDS rejection (%)	**91.8 ± 1.9**	**95.5 ± 0.4**	**96.3 ± 0.1**	**91.8 ± 1.8**	**92.0 ± 2.3**	**90.3 ± 6.6**
Average permeate conductivity ($\mu S.cm^{-1}$)	98.2	113		152.5		
Permeate nitrate concentration ($mg.L^{-1}$)	3.40	3.23	3.52	3.13	3.52	4.01
pH of permeate	5.22	5.28		5.41		
BW30						
Feed pressure (bar)	**11.18**	**13.2**	**17.22**	**13.2**	**17.22**	**15.68**
Osmotic pressure (bar)	0.8	1.53		3.02		
Average permeate flux ($L.h^{-1}.m^{-2}$)	23.37	29.9		29.21		
TDS rejection (%)	**> 99**	**> 99**		**> 99**		
Average permeate conductivity ($\mu S.cm^{-1}$)	12.94	15.97		22.93		
Permeate nitrate concentration ($mg.L^{-1}$)	0.76	0.66	1.38	0.66	1.38	3.94
pH of permeate	4.71	4.77		4.8		

Inlet pressure required to ensure the permeate flux strongly increases when using RO elements. BW30 elements enable near-total rejection of ionic species except nitrate ion whose rejection is 91%. These high rejection rates imply high hydraulic pressure almost twice the hydraulic pressure in NF. Thus, excepted for direct potable reuse, this type of membrane is not recommended for water reuse for groundwater recharge of other use that direct consumption. Rejection of monovalent ionic species with NF-270 does not exceed 70 % which makes it unsuitable for water reuse projects. In fact, as demonstrated in a previous paper (Azais et al. 2014), the NF-270 membrane could produce a good water quality according to the FAO irrigation standards for all of the analyzed parameters except for the potassium. Concerning the aquifer recharge, the CDPH specifies that for replenishing a groundwater basin, the effluent must not exceed 5 $mg.L^{-1}$ of total nitrogen and 0.5 $mg.L^{-1}$ of TOC for projects where no dilution in the subsurface is occurring (reclaimed water contribution of 100%). As presented in the Table 6, the major limitation of the NF-270 for water reuse is the poor rejection of nitrogenous species for groundwater recharge. The nitrate concentration in the NF-270 permeate is too close of the accepted limit to preconize this membrane for indirect potable water production. Besides, from previous laboratory tests, the NF-270 TOC permeate concentration marginally lower than the standard of the CDPH could limit its application for water reclamation.

NF-90 permits high TDS rejection which is not impacted per recovery rates. Moreover, it was shown that nitrate concentration in the NF-90 permeate would enable its reuse in direct consumption or ground recharge. Thus, tight NF membranes seem to be well-suited for reuse water projects with strict usages which could be an alternative to the current RO schemes.

3.4 Sensivity analysis of the cost of nanofiltration

A sensibility analysis was performed in order to evaluate the impact on the total cost of slight cost variations of membrane equipment. Basic case was defined as the one previously presented with NF-90 and Y=80%. Several cases were tested: a 10% decrease of both elements and pressure vessels, a 10% increase of both elements and pressure vessels, a membrane replacement rate of 5% instead of 10%, a membrane replacement rate of 15% instead of 10%, and a mix of these conditions (table 7).

Table 7. Effect of slight variations of membrane cost and replacement rates on the total cost of membrane operation.

Membrane capital cost ($)	Annual membrane capital cost ($/year)	Annual membrane replacement ($/year)	Δ$/year
Basic case NF-90 Y=80%	14 736	7 650	0
Pressure vessels and elements - 10%	13 262	6 885	-2 239
Pressure vessels and elements + 10%	16 210	8 415	2 239
Annual membrane replacement rate 5%	14 736	3 825	-3 825
Annual membrane replacement rate 15%	14 736	11 475	3 825
Pressure vessels and elements + 10% and Annual membrane replacement rate 15%	16 210	12 622	6 446
Pressure vessels and elements - 10% and Annual membrane replacement rate 5%	13 262	3 443	-5 682

As it can be seen in table 7, variations of membrane equipment and/or membrane replacement rate can result in a modification of annual cost from -5 682 to 6 446 $/year in comparison with the basic case. Thus, considering the project lifetime of ten years, total amount of the project can be modified by around 60 000 $ depending of current costs which is not negligible.

3.5. Economic evaluation of ozonation of NF retentates

Economic evaluation of ozonation step is necessary so as to define the optimal conversion rate which ensures a compromise between permeate flux and treatability of the NF retentate. Cost of retentate ozonation was estimated for the three conversion rates of the study and compared to direct ozonation of the effluent. Results are presented in table 8.

Table 8. Economic evaluation of ozonation step on secondary effluent for several conversion rates.

	Secondary effluent	Y = 50 %	Y = 80 %	Y = 90 %
Inlet flowrate ($m^3.h^{-1}$)	**125**	**62.5**	**25**	**12.5**
Required transfered ozone dose ($mgO_3.L^{-1}$)	**5.5 – 12.5**	**5.5 - 22.5**	**10.0 - 30.0**	**20.0 - 40.0**
Required ozone production ($kgO_3.h^{-1}$)	**0.7 – 1.6**	**0.4 - 1.5**	**0.3 - 0.8**	**0.3 - 0.5**
Required power (kW)	**11 - 16**	**5 - 15**	**4 - 8**	**3.9 – 5.3**
Specific consumption ($kWh.m^{-3}$)	**0.09 - 0.13**	**0.09 - 0.24**	**0.16 - 0.32**	**0.32 – 0.42**
O&M cost ($\$.year^{-1}$)	**8 558 – 12 967**	**4 273 – 11 670**	**3 112 – 9 336**	**3 112 – 4 150**
Average O&M cost ($\$.m^{-3}$)	**0.010**	**0.014**	**0.028**	**0.033**
Average capital cost ($)	**115 000**	**95 000**	**55 000**	**40 000**
Average capital cost ($\$.year^{-1}$)	**19 600**	**16 200**	**9 400**	**6 800**
Average capital cost ($\$.m^{-3}$)	**0.018**	**0.029**	**0.043**	**0.062**

Cost of ozonation of secondary effluent in this study is in accordance with literature (Margot et al., 2013; Ni et al., 2013). Ozonation treatment would result in an increase of 30 % of the WWTP cost if considering that mean cost of WWTP with MBR is 0.49 \$/ m^3 (Guo et al., 2014). Increase in conversion rate lead to a decrease in operating and maintenance cost because of the reduction of treated volume and thus of the power of ozone generator. Finally, Table 9 presents a cost analysis of the coupling between ozonation and nanofiltration (NF90-400) for several conversion rates and considering a reuse of both permeate and retentate.

Table 9. Cost analysis of coupling between ozonation and nanofiltration using NF90 - 400 with reuse of permeate and retentate.

Membrane	Y=50%	Y=80%	Y=90%
Membrane CAPEX ($/m^{3}permeate)	**0.010**	**0.013**	**0.016**
Ozonation CAPEX ($/m^{3}permeate)	**0.014**	**0.008**	**0.006**
Total CAPEX ($/m^{3}permeate)	**0.024**	**0.021**	**0.022**
Membrane OPEX ($/m^{3}permeate)	**0.077**	**0.085**	**0.088**
Ozonation OPEX ($/m^{3}permeate)	**0.007**	**0.006**	**0.003**
Total OPEX ($/m^{3}permeate)	**0.084**	**0.091**	**0.092**

NF/O_3 treatment of the effluent (without considering the benefit of the reuse) resulted in an increase of almost 80% of the ozone process cost as demonstrated by Plumlee et al. (2014) (Plumlee et al., 2014). Nonetheless, quality of the treated water is much more satisfying than for ozonation alone and enables its reuse (Azais et al., 2016 b)). Indeed, ozonation alone does not enable elimination of ionic species and a complete mineralization and thus does not satisfy actual water reuse standards.

In comparison with NF alone, coupling of NF and ozonation results in approximately same CAPEX (table 3). If considering the optimum conversion rate of 80%, which is usually encountered at industrial scale (Van der bruggen et al., 2013), in comparison with NF alone the coupling results in an increase capital costs of 0.005 $/m^3/year and a decrease of operating cost of 0.019 $/m^3/year. The cost saving due to the coupling between NF and ozonation is thus 0.014 $/m^3/year which means 15 330 $/year for a flow rate of 125 m^3/h.

If considering annual treated volumes with possible retentate recovery (thus increase of the annual treated volume), NF/O_3 process would thus be more economical than nanofiltration alone. It has indeed the potential to be cost-competitive in some situations, especially when the recovery of otherwise wasted water is considered. This is in accordance with other authors who concluded that costs associated with zero liquid discharge (concentrate treatment) may be partially offset by the recovery (Khan et al., 2009; Umar et al., 2015). Finally, by taking into account economy of scale, which is found for both capital, operating and maintenance costs (Guo et al., 2014), cost predicted in the present study could be considerably decreased which contributed to make this process attractive for wastewater reuse.

4. Conclusion

This study has put in evidence that ozonation of nanofiltration retentates would be an acceptable alternative solution from an economic point of view for wastewater reuse. It was shown that NF could be used in lieu of RO as advanced treatment. NF is an effective barrier for organic contaminants which is less expensive than RO because of lower O&M costs. Cost saving for a flow rate of 125 and Y=8 % could be 35 k£/year when using nanofiltration instead of reverse osmosis. This may be enhanced in the future if more demand of NF develops the market, meaning less capital cost when selecting NF membranes instead of RO. Concerning the filtration step of the proposed advanced process, a three stages configuration with a decreasing number of pressure vessels (N) in parallel - 12, 6 and 3 pressures vessels with 6 elements (M) on first stage and 5 elements on second and third stages - is well-adapted. An optimum conversion rate of 80 % was found. Ozonation of nanofiltration retentates enable a possible reuse of both permeate and retentate and thus can result in lower production costs. For instance, an economy of 15.4 k$/year of operating costs was obtained for a flow rate of 125 m^3/h. One can expect considerable cost saving for higher flow rates as usually encountered. Considering such a process will result in a reduction of greenhouse gas emission compared to conventional high-energy reuse processes based on the use of reverse osmosis.

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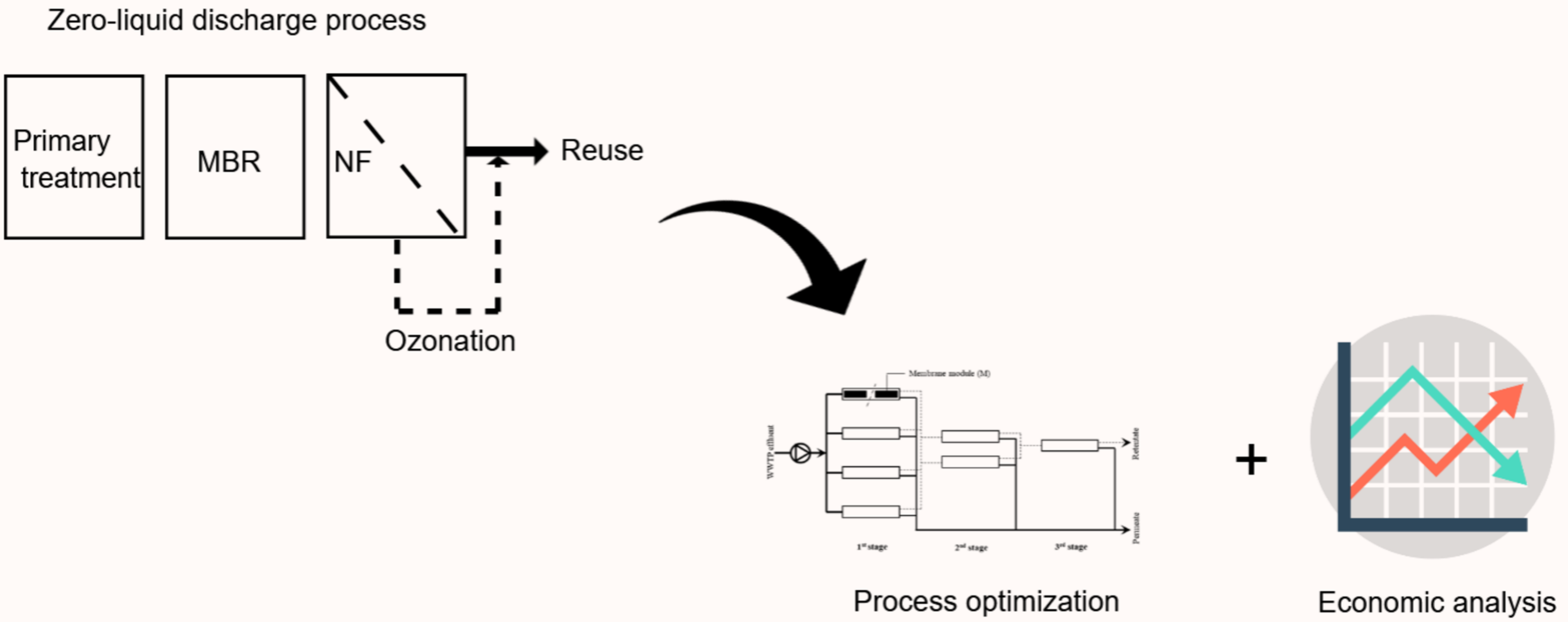
Zero-liquid discharge process
Primary treatment
MBR
NF
Reuse
Ozonation
Membrane module (M)
WWTP effluent
Retentate
Permeate
1st stage
2nd stage
3rd stage
Process optimization
+
Economic analysis