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## Urban wastewater reuse using a coupling between nanofiltration and ozonation: Techno-economic assessment

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25 **Highlights:**

26 Nanofiltration is a possible alternative to reverse osmosis for wastewater reuse

27 Nanofiltration combined to ozonation is a competitive process for wastewater reuse

28 Nanofiltration is a barrier for micropollutants with reasonable operating costs

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

30

31 **1. Introduction.**

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

46 After massive investment in energy-intensive desalination processes which increased the global  
47 production capacity of desalinated water by 67% to 52 million m<sup>3</sup>.d<sup>-1</sup> (GWI, 2009), governments are  
48 now looking at overall water efficiency and start to prioritize water reclamation and reuse as a solution  
49 to water scarcity since water reclamation requires only a third to a fourth of the energy needed for

50 reverse osmosis (RO) desalination. While the combined approach of seawater desalination and water  
51 reuse has proven to be successful in a number of coastal sites such as Barcelona, Singapore, land  
52 locked cities have to focus completely on water reuse for irrigation or aquifer recharge, sometimes  
53 even accepting direct potable reuse as practiced in Windhoek, Namibia (Bunani et al., 2015, Bellona et  
54 al., 2012). With a closed water loop, land locked cities require high-quality water reuse systems  
55 mostly based on membrane technology as affordable and efficient barrier.

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

77 desorb (Benstoem et al., 2017). To recover fresh reusable water from urban wastewater treatment,  
78 efficient additional treatment processes which are economically viable are required and thus must be  
79 developed.

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

95 However, retentate recirculation upstream of the biological treatment is not possible in the long term  
96 as salinity greater than 5 g.L<sup>-1</sup> may inhibit biological activity (Reid et al., 2006). In addition, the  
97 recirculation of NF concentrates within a MBR significantly increases its clogging (Kappel et al.,  
98 2014). Finally, direct discharge into surface or marine waters is not easy to encourage given the  
99 average composition of the urban retentates i.e. conductivities of 2 to 20 mS.cm<sup>-1</sup>, CODs ranging from  
100 20 to 55 mg.L<sup>-1</sup> and levels of organic micropollutants multiplied by 3 to 7 (Benner et al., 2008; Solley  
101 et al., 2010). Hence, treatment of membrane concentrates is recommended.

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

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

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

135

## 136 **2. Material and methods.**

### 137 *2.1. Cost of nanofiltration*

138 ROSA 9.0 software model was applied to predict water quality and flow rate for different membrane  
139 system configurations. ROSA software is a model designed by Nisan in 2005, applied to design and  
140 manipulate RO and NF systems taking into account effects of concentration polarization factor and  
141 hydrodynamic pressure drop in the concentrate-side on the membrane performance (Nisan et al.,  
142 2005). This software is often used in research work to simulate designs using reverse osmosis or  
143 nanofiltration plants (Yangali-Quintilla et al., 2010 ; Haryati et al., 2017).

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

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

155 (UF) membranes in polyvinylidene difluoride (PVDF) which have a pore size of 0.035 microns. The  
 156 total membrane surface area is about 7 605 m<sup>2</sup> and produces approximately 1 500 m<sup>3</sup> of permeate per  
 157 day while operating at a mixed liquor total suspended solids (TSS) concentration of between 4 and 7 g  
 158 L<sup>-1</sup>. Summary of major constituents of the secondary effluent is presented in table 1 with intervals  
 159 given with 95% confidence.

160  
 161

Table 1. Water quality of the WWTP effluent.

Parameters	Units	WWTP effluent
pH	-	7.69 ± 0.31
Conductivity	mS cm <sup>-1</sup>	1.30 ± 0.22
Alkalinity	mg eq CaCO <sub>3</sub> L <sup>-1</sup>	430 ± 80
COD	mg L <sup>-1</sup>	18 ± 3
DOC	mg L <sup>-1</sup>	6 ± 2
NO <sub>3</sub> <sup>-</sup> -N	mg L <sup>-1</sup>	2.0 ± 0.6
Total Nitrogen (TN)	mg L <sup>-1</sup>	4.1 ± 1.6
PO <sub>4</sub> <sup>3-</sup> -P	mg L <sup>-1</sup>	0.11 ± 0.08
Na <sup>+</sup>	mg L <sup>-1</sup>	152 ± 14
K <sup>+</sup>	mg L <sup>-1</sup>	22 ± 3
Ca <sup>2+</sup>	mg L <sup>-1</sup>	160 ± 20
Mg <sup>2+</sup>	mg L <sup>-1</sup>	6.6 ± 0.8
Cl <sup>-</sup>	mg L <sup>-1</sup>	267 ± 35
SO <sub>4</sub> <sup>2-</sup>	mg L <sup>-1</sup>	89 ± 13
Ionic strength	mM	22 ± 5

162

163 No nitrites were detected in the effluent so that the value of TN could be due to the presence of  
 164 ammonia because of incomplete nitrification. Regarding water pH and ionic composition, ROSA  
 165 estimates Langelier Saturation Index (LSI) which is a measure for the saturation of CaCO<sub>3</sub> in water.  
 166 The LSI should be used for low Total Dissolved Solids (TDS) ranges (<10 000 ppm). At the pH of  
 167 saturation (pH<sub>s</sub>), the water is in equilibrium with CaCO<sub>3</sub>. To avoid calcium carbonate scaling by acid  
 168 addition alone, the LSI in the concentrate stream must be negative. If a high quality scale inhibitor is



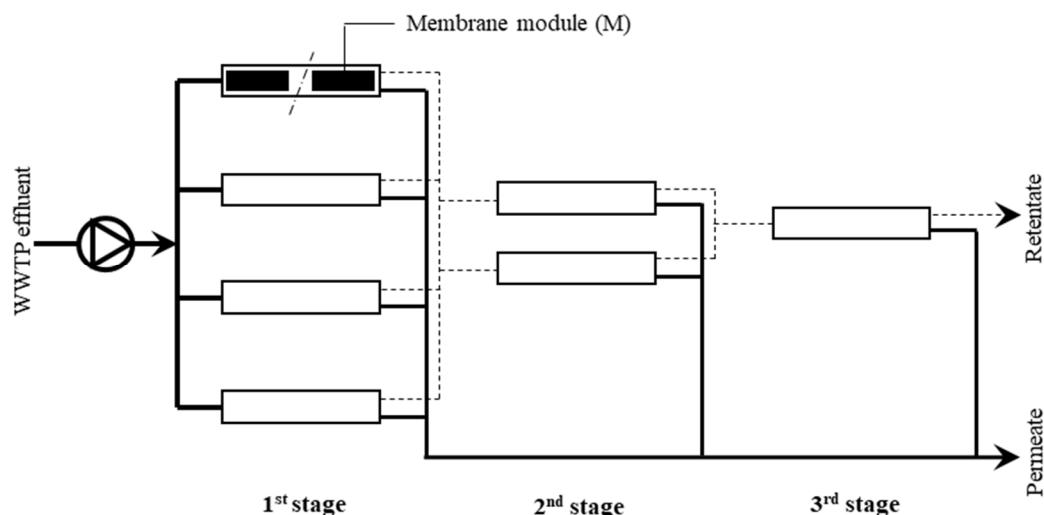
169 used, the LSI in the concentrate stream can be increase up to 1.5. This will reduce or eliminate the acid  
170 consumption. Many inhibitors allow operation up to an LSI of < 1.8 in the concentrate. In this study  
171 the water is hard, so acid ( $H_2SO_4$ ) was added. Finally, the low value of SDI will reduce fouling  
172 problems due to particulate matters which tends to accumulate at the membrane surface resulting in a  
173 membrane flux decline.

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

179 A two or three stage configuration with a decreasing number of pressure vessels in parallel allows  
180 respecting the minimum concentrate flow rate constraint and other required hydraulic conditions in  
181 nanofiltration process. Consequently, this kind of configuration was chosen in this study in order to  
182 reach high conversion rates (> 80 %), obtain a reliable permeate quality without a severe fouling risk  
183 (figure 1). First stage is composed of N pressure vessels each including M elements in series. Next  
184 stage has N/2 pressure vessels and last stage N/4 pressure vessels. In the experimental study, three  
185 Volume Reduction Factor (VRF) were tested: 2, 5 and 10 which correspond to recovery rate (Y,  
186 defined as the ratio of permeate flow by feed flow) of about 50, 80 and 90 % respectively (Azais et al.,  
187 2016 a). To reach these recovery rates, number of stage has to be adapted. Y of 50 % was applied to  
188 the first stage and Y of 60% was applied to second stage thus resulting in global Y of 80 % for two  
189 stages. Y of 50% was applied on last stage resulting in final Y of 90 % close to VRF 10 which was  
190 obtained at pilot scale (Azais et al., 2016b). To reach these conversion rates, ROSA adjusted pressure  
191 at the entrance of each stage according to the number of stage and pressure vessels and type of  
192 membrane.

193

194



195

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

198

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

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

212 Cost of NF 90-400 element and pressure vessel was estimated to respectively 850 and 1 500 \$. Price  
 213 of kilowatt-hour was based on current rate in France which is about  $0.09 \text{ \$} \cdot \text{kWh}^{-1}$  in accordance with  
 214 Plumlee et al. (2014) (Plumlee et al., 2014). Moreover, annual membrane replacement rate was taken

215 equal to 10% as estimated by Yangali-Quintanila et al. (2010) (Yangali-Quintanila et al., 2010). To  
216 calculate the annual capital costs, project lifetime was taken equal to 10 years (life time of membranes)  
217 with an interest rate of 7%.

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

222

## 223 *2.2. Cost of ozonation*

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

229 As only little ozone doses are required to treat retentates of this study, on site oxygen generation using  
230 Pressure Swing Absorption (PSA) was chosen. This is ideally suited to supply an ozone generator with  
231 medium frequency (60 to 1 000 Hz). Specific energy for ozone production was estimated to 10  
232 kWh.kgO<sub>3</sub><sup>-1</sup> (Rosenfeldt et al., 2006). According to Margot et al. (2013), the transfer efficiency of  
233 ozone into the dissolved phase in ozonation pilot plant is between 70 to over 90% depending on the  
234 gas flow (Margot et al., 2013). In this study, it was fixed to 95%. Investment cost was taken equal to  
235 100 \$.gO<sub>3</sub><sup>-1</sup>.h<sup>-1</sup> as suggested by ozone water treatment plants manufacturers and suppliers. The  
236 operational cost was then evaluated in this study. Finally, as cooling water is used in a close loop  
237 system, this item of expenditure was neglected.

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

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

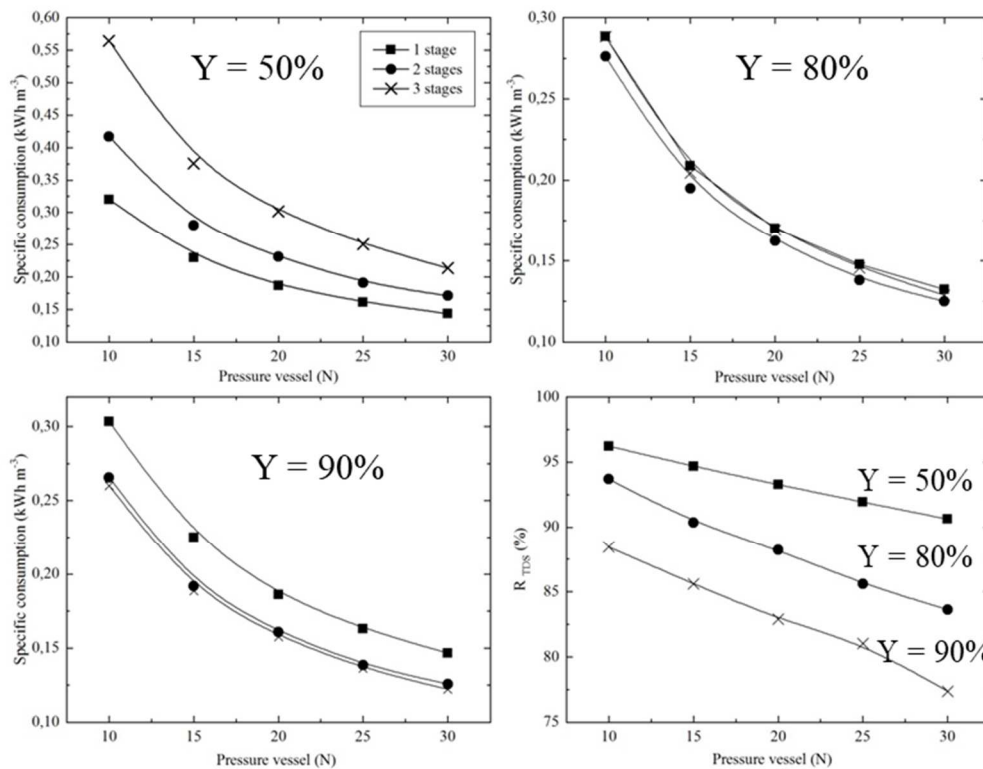
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### 246 **3. Results and discussion**

247

#### 248 *3.1. Optimization of nanofiltration design for NF-90*

249 To verify that the chosen configuration (figure 1) is well adapted to the targeted conversion rates,  
250 simulations were done by imposing the three recovery rates for different configuration (one, two or  
251 three stages). For this, value of the number of pressure vessel in first stage (N) was modified between  
252 10 and 30. At first approximation, number of elements in a pressure vessel was fixed to 7 as usually  
253 encountered at industrial scale (Van der Meer et al., 1998). For each simulation, total power to be  
254 supplied by the pumps was noticed so as to determine specific energy consumption (kWh/m<sup>3</sup>) of each  
255 configuration. In addition, the rejection of total dissolved solids (TDS) was estimated as a function of  
256 N. Figure 2 presents the results of these simulations.



257

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

259

260 Regarding specific energy consumption, it appears that each recovery rate is adapted to a certain

261 number of stages. A single stage system is more energy efficient when a conversion rate of 50% is

262 applied. Two stages are more energy efficient to reach Y=80% and three stages to reach Y=90%.

263 Indeed, at low conversion rate, the concentrate flow rate is high thus when using only one stage

264 pressure drop along the unit is limited. On the contrary, for higher conversion rate (lower concentrate

265 flow rate), the decrease of hydraulic pressure due to the increase of stages is more interesting from an

266 energetic point of view. In the next part of the article, a conversion rate of 50%, 80% and 90% was

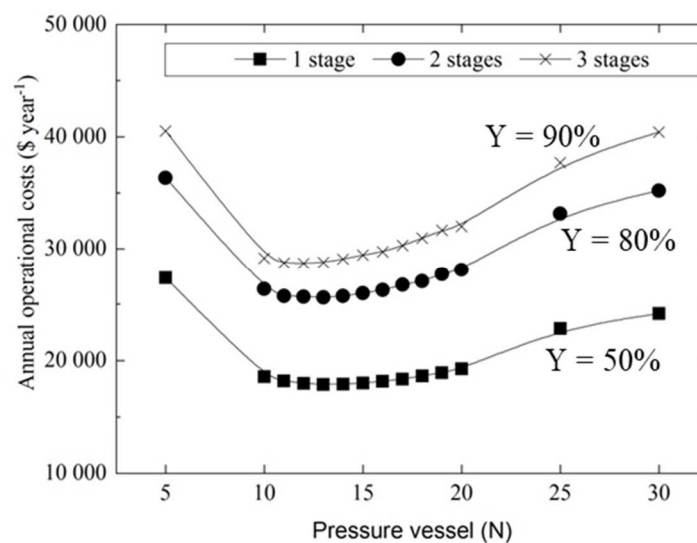
267 thus reached with respectively one, two and three stages. On figure 2, it appears that specific energy

268 consumption decreases with increasing number of pressure vessels in the first stage. For example, feed

269 pressure decreases from 4.6 to 2.1 bars when N increases from 10 to 30. Nonetheless, an increasing

270 number of pressure vessels will have a direct consequence on capital and maintenance cost. Moreover,  
 271 the decrease of inlet pressure leads to a decrease in TDS rejection because of high diffusive transfer.

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

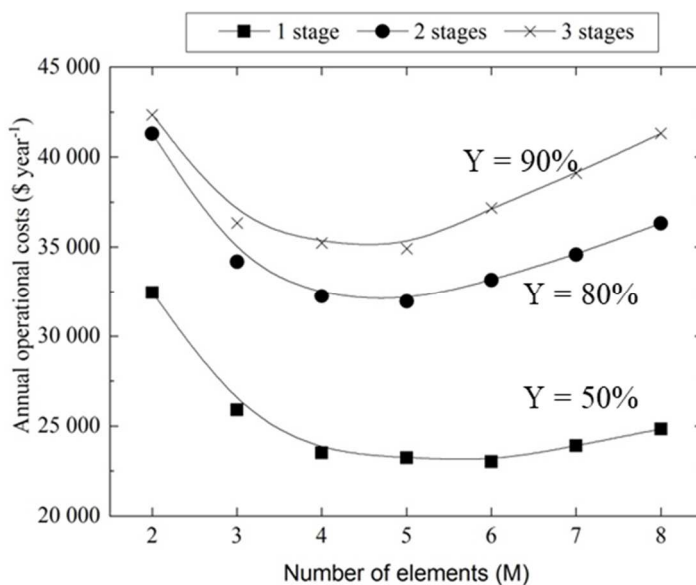


280  
 281 Figure 3. Influence of number of pressure vessel in the first stage (N) and number of stages on annual  
 282 operating costs (number of elements M=7).

283

284 For each case, annual operating costs show a minimum for an optimum number of pressure vessel  
 285 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  
 286 equal to 12 so has to have 6 and 3 pressure vessels respectively in the second and third stage.

287 Concerning the number of elements per pressure vessel ( $M$ ), van der Meer et al. (1998) showed that  
 288 even though the classical system configuration of pressure vessels in RO and NF systems is a  
 289 multistage design with a number of membrane modules per pressure vessel of 6 up to 8, this design  
 290 trend results in high pressure losses because of high feed velocities and long feed/brine channel  
 291 lengths (van der Meer et al., 1998). The authors showed that an increase in permeate productivity of  
 292 20% can be achieved by lowering the number of membrane modules per pressure vessel. In contrast to  
 293 the hydraulic optimum, they found an economic optimum between 3 to 4 elements per pressure vessel.  
 294 Research of optimal number of elements  $M$  was conducted using same approach as for  $N$ . The  
 295 pressure vessel number was 12 in the first stage, 6 in the second stage and 3 in the last stage.  $M$  was  
 296 varied between 2 and 8 (maximum authorized by ROSA). Then, operating costs were evaluated for  
 297 each configuration. Results are presented in figure 4.



298  
 299 Figure 4. Influence of number of elements ( $M$ ) and stages on annual operating costs (total number of  
 300 pressure vessels  $N=12$ ).

301  
 302 The economic optimum is  $M=6$  elements for a single stage and  $M=5$  for two and three stages which is  
 303 in accordance with van der Meer et al. (1998) (van der Meer et al., 1998). At low number of  
 304 membrane modules, the permeate flow rate decreases as a result of the increasing concentration

305 polarization in this region. This effect becomes more pronounced at higher recoveries, caused by  
 306 increasing osmotic pressure differences across the membrane surface, as a result of the increasing salt  
 307 concentrations at the feed/brine side of the membranes at high recoveries. In the rest of the paper,  
 308 number of elements (M) was taken equal to 6 for one stage and to 5 for two or three stages.

309 *3.2. Economic analysis of filtration*

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

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

Targeted conversion rate	Y = 50 %		Y = 80 %		Y = 90 %	
Stage	1	1	2	1	2	3
<b>System details</b>						
Feed flow (m <sup>3</sup> .h <sup>-1</sup> )	125	125	62.5	125	62.5	25
Permeate flow (m <sup>3</sup> .h <sup>-1</sup> )	62.5	62.5	37.5	62.5	37.5	12.5
Retentate flow (m <sup>3</sup> .h <sup>-1</sup> )	62.5	62.5	25	62.5	25	12.5
Recovery rate (%)	50	50	60	50	60	50
Total membrane area (m <sup>2</sup> )	2 676	3 344		3 902		
Average permeate flux (L.h <sup>-1</sup> .m <sup>-2</sup> )	23.4	29.9		28.8		
Feed pressure (bar)	5.04	5.55	7.52	5.55	7.52	6.75
<b>Economic analysis</b>						
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		
<b>Membrane capital cost (\$)</b>	<b>79 200</b>	<b>103 500</b>		<b>120 750</b>		
<b>Annual membrane capital cost (\$/year)</b>	<b>11 276</b>	<b>14 736</b>		<b>17 192</b>		
<b>Annual membrane capital cost (\$/m<sup>3</sup>)</b>	<b>0.020</b>	<b>0.016</b>		<b>0.017</b>		

314



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

323 As mean value of LSI of feed water was 1.3 which is superior to the recommended value of 1, use of  
324 acid was included in the economic evaluation of the process. ROSA proposes two agents (HCl,  
325 H<sub>2</sub>SO<sub>4</sub>) which enable the LSI to be lowered close to zero value by modifying pH. In this study, H<sub>2</sub>SO<sub>4</sub>  
326 was chosen as sulfate ions are retained by more than 90% by NF-90 membrane and thus did not  
327 impact the permeate quality. The targeted LSI was 0.2 which required 140.5 mg of H<sub>2</sub>SO<sub>4</sub> per liter of  
328 inlet effluent. This represented about 84 m<sup>3</sup> of H<sub>2</sub>SO<sub>4</sub> (96%) per year. As the cost of a cubic meter of  
329 H<sub>2</sub>SO<sub>4</sub> is about 730 \$ thus annual expense for scaling control by acidification was about 61 000 \$  
330 (Table 3). Finally, acid cost was about four times operation and maintenance expenses. Proportion of  
331 electrical cost slightly increases when increasing conversion rate (figure 5).

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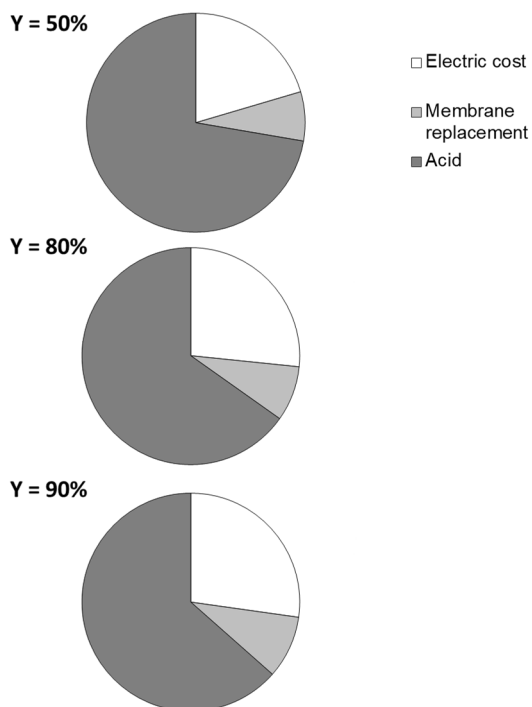
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339 Table 3. Economic analysis of operating and maintenance cost for nanofiltration operation with NF90-  
 340 400 elements at several conversion rates.

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



341

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

343 *3.3. Comparison with loose-nanofiltration and reverse osmosis process*

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

349 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 <sup>3</sup> )
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

350

351 Table 5 presents the comparison of operating and maintenance cost of nanofiltration and reverse  
 352 osmosis processes. For high conversion rates, booster pumping may be required between stages.  
 353 Operating and maintenance cost varies between 0.09 and 0.19 \$/m<sup>3</sup> depending on the conversion rate  
 354 and membrane type. These costs determined by ROSA are in accordance with literature concerning  
 355 NF/RO systems (Cote et al., 2005; Englehardt et al., 2013). Globally the cost decreases when  
 356 increasing membrane permeability ( $L_p$ ) which is as follow in l/h/m<sup>2</sup>/bar (LMHB):  $L_p$  BW30-400 = 3.2  
 357 LMHB < NF90-400 = 10.8 LMHB < NF270-400 = 13.6 LMHB. For  $Y=80\%$  and an inlet flow rate of  
 358 125 m<sup>3</sup>/h, 29.6 k\$/year and 35.2 k\$/year could be saved using respectively NF90-400 and NF270-400  
 359 instead of BW30-400. This is in accordance with other authors (Yangali-Quintanilla et al., 2011;  
 360 Bellona et al., 2012). Indeed, even if RO elements are less expensive than NF elements, energy  
 361 requirement of the pumps was twice or third the one needed with NF processes which make operating  
 362 and maintenance costs of RO process higher. Finally, type of NF (loose or tight NF) does not seem to  
 363 influence considerably the process cost. Nonetheless, this influences performances and rejection rates  
 364 of the process as it can be seen in table 6.

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

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

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

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373 Inlet pressure required to ensure the permeate flux strongly increases when using RO elements. BW30  
374 elements enable near-total rejection of ionic species except nitrate ion whose rejection is 91%. These

375 high rejection rates imply high hydraulic pressure almost twice the hydraulic pressure in NF. Thus,  
376 excepted for direct potable reuse, this type of membrane is not recommended for water reuse for  
377 groundwater recharge of other use than direct consumption. Rejection of monovalent ionic species with  
378 NF-270 does not exceed 70 % which makes it unsuitable for water reuse projects. In fact, as  
379 demonstrated in a previous paper (Azais et al. 2014), the NF-270 membrane could produce a good  
380 water quality according to the FAO irrigation standards for all of the analyzed parameters except for  
381 the potassium. Concerning the aquifer recharge, the CDPH specifies that for replenishing a  
382 groundwater basin, the effluent must not exceed 5 mg.L<sup>-1</sup> of total nitrogen and 0.5 mg.L<sup>-1</sup> of TOC for  
383 projects where no dilution in the subsurface is occurring (reclaimed water contribution of 100%). As  
384 presented in the Table 6, the major limitation of the NF-270 for water reuse is the poor rejection of  
385 nitrogenous species for groundwater recharge. The nitrate concentration in the NF-270 permeate is too  
386 close of the accepted limit to preconize this membrane for indirect potable water production. Besides,  
387 from previous laboratory tests, the NF-270 TOC permeate concentration marginally lower than the  
388 standard of the CDPH could limit its application for water reclamation.

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

### 393 *3.4 Sensivity analysis of the cost of nanofiltration*

394 A sensibility analysis was performed in order to evaluate the impact on the total cost of slight cost  
395 variations of membrane equipment. Basic case was defined as the one previously presented with NF-  
396 90 and Y=80%. Several cases were tested: a 10% decrease of both elements and pressure vessels, a  
397 10% increase of both elements and pressure vessels, a membrane replacement rate of 5% instead of  
398 10%, a membrane replacement rate of 15% instead of 10%, and a mix of these conditions (table 7).

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400

401 Table 7. Effect of slight variations of membrane cost and replacement rates on the total cost of  
 402 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

403  
 404 As it can be seen in table 7, variations of membrane equipment and/or membrane replacement rate can  
 405 result in a modification of annual cost from -5 682 to 6 446 \$/year in comparison with the basic case.  
 406 Thus, considering the project lifetime of ten years, total amount of the project can be modified by  
 407 around 60 000 \$ depending of current costs which is not negligible.

408 *3.5. Economic evaluation of ozonation of NF retentates*

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

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

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

420

421 Cost of ozonation of secondary effluent in this study is in accordance with literature (Margot et al.,  
 422 2013; Ni et al., 2013). Ozonation treatment would result in an increase of 30 % of the WWTP cost if  
 423 considering that mean cost of WWTP with MBR is 0.49 \$/ m<sup>3</sup> (Guo et al., 2014). Increase in  
 424 conversion rate lead to a decrease in operating and maintenance cost because of the reduction of  
 425 treated volume and thus of the power of ozone generator. Finally, Table 9 presents a cost analysis of  
 426 the coupling between ozonation and nanofiltration (NF90-400) for several conversion rates and  
 427 considering a reuse of both permeate and retentate.

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

Membrane	Y=50%	Y=80%	Y=90%
<b>Membrane CAPEX</b> (\$/m <sup>3</sup> permeate)	<b>0.010</b>	<b>0.013</b>	<b>0.016</b>
<b>Ozonation CAPEX</b> (\$/m <sup>3</sup> permeate)	<b>0.014</b>	<b>0.008</b>	<b>0.006</b>
<b>Total CAPEX (\$/m<sup>3</sup>permeate)</b>	<b>0.024</b>	<b>0.021</b>	<b>0.022</b>
<b>Membrane OPEX (\$/m<sup>3</sup>permeate)</b>	<b>0.077</b>	<b>0.085</b>	<b>0.088</b>
<b>Ozonation OPEX (\$/m<sup>3</sup>permeate)</b>	<b>0.007</b>	<b>0.006</b>	<b>0.003</b>
<b>Total OPEX (\$/m<sup>3</sup>permeate)</b>	<b>0.084</b>	<b>0.091</b>	<b>0.092</b>

433

434 NF/O<sub>3</sub> treatment of the effluent (without considering the benefit of the reuse) resulted in an increase of  
 435 almost 80% of the ozone process cost as demonstrated by Plumlee et al. (2014) (Plumlee et al., 2014).  
 436 Nonetheless, quality of the treated water is much more satisfying than for ozonation alone and enables  
 437 its reuse (Azais et al., 2016 b)). Indeed, ozonation alone does not enable elimination of ionic species  
 438 and a complete mineralization and thus does not satisfy actual water reuse standards.

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

445 If considering annual treated volumes with possible retentate recovery (thus increase of the annual  
446 treated volume), NF/O<sub>3</sub> process would thus be more economical than nanofiltration alone. It has  
447 indeed the potential to be cost-competitive in some situations, especially when the recovery of  
448 otherwise wasted water is considered. This is in accordance with other authors who concluded that  
449 costs associated with zero liquid discharge (concentrate treatment) may be partially offset by the  
450 recovery (Khan et al., 2009; Umar et al., 2015). Finally, by taking into account economy of scale,  
451 which is found for both capital, operating and maintenance costs (Guo et al., 2014), cost predicted in  
452 the present study could be considerably decreased which contributed to make this process attractive  
453 for wastewater reuse.

#### 454 **4. Conclusion**

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

470

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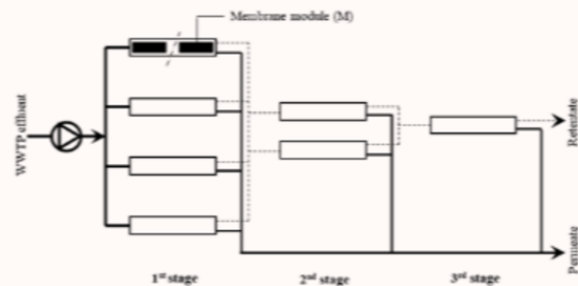
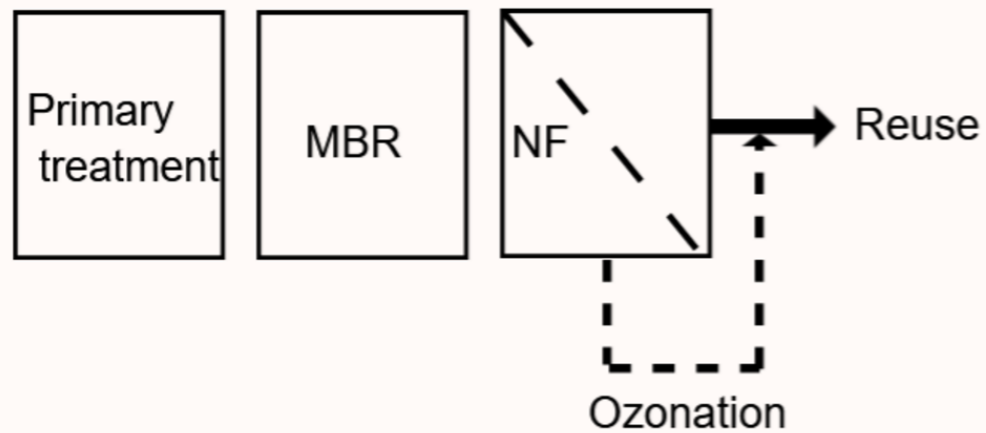
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# Zero-liquid discharge process



Process optimization

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Economic analysis