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1 **Novel housing designs for nanofiltration and ultrafiltration gravity-driven recycled**
2 **membrane-based systems**

3 Raquel García-Pacheco^{1,2,3*}, Qiyuan Li^{2,4}, Joaquim Comas^{1,5}, Robert A Taylor⁴,
4 Pierre Le-Clech²

5 ¹*LEQUIA. Institute of the Environment. University of Girona Campus Montilivi, carrer Maria Aurèlia*
6 *Capmany, 69, E-17003 Girona. Catalonia. Spain.*

7 ²*UNESCO Centre for Membrane Science and Technology, School of Chemical Engineering, The University of*
8 *New South Wales (UNSW), Kensington, New South Wales 2052, Australia*

9 ³*IMDEA Water Institute, Avenida Punto Com. n.º2. 28805, Alcalá de Henares, Madrid, Spain*

10 ⁴*School of Mechanical and Manufacturing Engineering, The University of New South Wales (UNSW),*
11 *Kensington, New South Wales 2052, Australia*

12 ⁵*Catalan Institute for Water Research (ICRA), 17003 Girona, Spain*

13
14 **Abstract:**

15 Ultra-low pressure gravity-driven membrane (GDM) systems have the potential to be
16 significantly less costly and complex than conventional membranes for water treatment
17 applications. To build upon this inherent advantage, this study assesses the reuse of recycled
18 membranes in GDM systems for producing drinking water. Two reverse osmosis spiral-
19 wound modules were recycled into nanofiltration (NF)-like and ultrafiltration (UF)-like
20 membranes via controlled exposure to free chlorine. To operate the recycled membranes, two
21 housing devices, based on a simple fitting and an advanced
22 end-caps design, were developed. The recycled membrane systems were tested under a range
23 of conditions (submerged vs. external system configuration and continuous vs. intermittent
24 filtration mode). Synthetic river water feed solutions were used in the tests where
25 performance, fouling, and clogging were measured. NF-like recycled membranes resulted in
26 poor salt rejection and low permeability ($\sim 1.7 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$), but also in high rejection
27 ($>81\%$) of dissolved organic carbon. UF-like recycled membranes maintained their capacity
28 to reject biopolymers (BP) ($>74\%$) and featured up to 18-fold higher permeate rate than NF-
29 like recycled membranes. The optimized operating conditions were found when the recycled
30 membranes were housed in the end-caps device and operated intermittently (relaxation time

31 plus forward flushing). Flushing reduced the fouling accumulation inside the membrane (only
 32 12% and 40% of BP accumulation was observed in the NF-like and UF-like, respectively).
 33 However, the end-caps-based device was estimated to be more expensive during the
 34 economic analysis. To address this techno-economic trade-off, a decision-making tree was
 35 developed to select the appropriate configuration based upon the implementation context.
 36 Overall, this study concludes that these designs can serve as robust, low-cost (water
 37 production cost <1 USD ct. yr. L⁻¹), and light-weight GDM alternatives. This study is
 38 beneficial for developing compact GDM systems based on recycled spiral-wound membranes
 39 for both rural areas and emergency response.

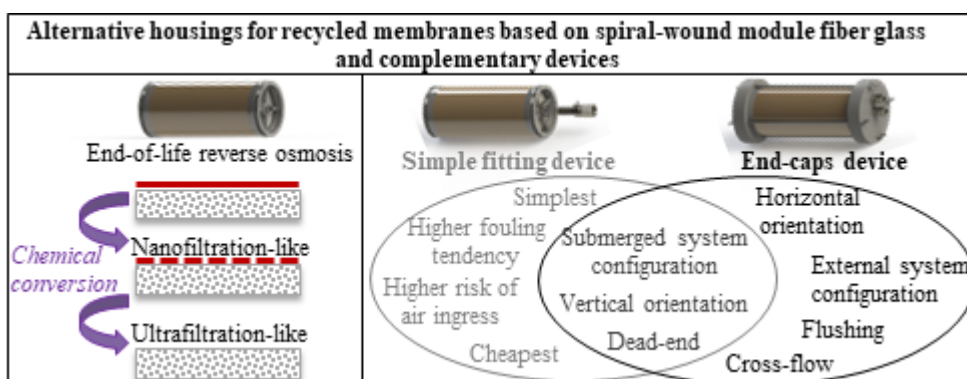
40 **Keywords:** nanofiltration, ultrafiltration, recycled membranes, membrane relaxation,
 41 flushing, gravity-driven membrane system, household water treatment; spiral-wound.

42

43 ***Corresponding author.** Tel.: +34 972 41 98 59. E-mail addresses: raquel.garcia@udg.edu

44

45 **Graphical abstract:**



47

48 **Highlights**

- 49 • Recycled spiral-wound membranes modules tested in gravity-driven membrane
50 processes
- 51 • Novel membrane housing devices were developed for compact water treatment
52 systems
- 53 • Membranes achieved permeability and rejection of organics for household treatment
- 54 • Intermittent operation (relaxation time and flushing) improved membrane
55 performance
- 56 • A decision-making tree help to select the housings and the operation conditions

57 **Abbreviations**

- | | | | |
|----|---|----|------------------------------------|
| 58 | BB: building blocks | 75 | LMW: low molecular weight |
| 59 | BP: biopolymers | 76 | MF: microfiltration |
| 60 | BSA: bovine serum albumin | 77 | N.A.: No data available |
| 61 | CapEx: capital expenditures or investment | 78 | NF: nanofiltration |
| 62 | CP: concentration polarization | 79 | OpEx: operational expenditures |
| 63 | DI: deionized water | 80 | PA: polyamide |
| 64 | DOC: dissolved organic carbon | 81 | PES: poly-ethersulfone |
| 65 | EoL: end-of-life | 82 | PSF: polysulfone |
| 66 | GDM: gravity-driven membrane | 83 | PVC: polyvinyl chloride |
| 67 | GDRM: gravity-driven recycled | 84 | RO: reverse osmosis |
| 68 | membrane-based | 85 | TMP: transmembrane pressure |
| 69 | HA: humic acids | 86 | TAC: total annual cost |
| 70 | HS: humic substances | 87 | SRW: synthetic river water |
| 71 | HWTS: household water treatment and | 88 | SS: suspended solids |
| 72 | safe storage | 89 | UF: ultrafiltration |
| 73 | LC-OCD: liquid chromatography organic | 90 | WASH: water sanitation and hygiene |
| 74 | carbon detection | 91 | promotion |

93 **1. Introduction**

94 A lack of access to safe water resources is frequent following natural disasters and in rural
95 locations [1]. In the last few decades, household water treatment and safe storage (HWTS)
96 systems, have been gaining interest to support resilience and for water sanitation and hygiene
97 promotion (WASH) actions for disease prevention [2]. These modular devices can be applied
98 across the spectrum of short (emergency response), medium (early recovery), and long-term
99 (international cooperation) programs [2]. Most HWTS systems are designed for point-of-use
100 applications and are sized to deliver just enough water to meet the survival requirements
101 (3-15 L day⁻¹ per person[1]) [2].

102

103 Several reviews have compared HWTS systems for their use in potable water for rural areas
104 and for emergency water supply [3–5]. An interesting subset of the HWTS field is gravity-
105 driven membrane (GDM) systems, which are mainly based on ultrafiltration (UF) and
106 microfiltration (MF) membranes. These offer high potential for situations where the electrical
107 supply is intermittent, as GDM systems require no pumps and are easy to use, operate, and
108 maintain. GDM systems are driven by the height difference between the feed tank and the
109 membrane unit (i.e. typically requiring 40-200 mbar of pressure), enabling reliable, stable
110 operation without backwashing, cross-flow or chemical cleaning, due to the phenomenon of
111 low flux stabilization (in the range of 1-20 L m⁻² h⁻¹) [6,7]. However, GDM systems using
112 spiral-wound nanofiltration (NF) or UF membranes have not yet been investigated.

113

114 Although GDM processes appear attractive for HWTS systems, the following challenges
115 have limited their application: i) low flux, ii) membrane price, iii) limited removal of small-
116 sized soluble organics, such as humic substances (HS), building blocks (BB), and low

117 molecular weight (LMW)-neutrals organics, and, iv) membrane fouling. Several of these
118 challenges have been addressed (to some degree) in the literature.

119

120 The ultra-low pressure used in GDM systems is the main cause for their lower flux as
121 compare to other pressure-driven processes [8,9] (i.e. challenge (i)). Due to the low flux,
122 GDM systems require a relatively higher specific membrane filtration area per unit module
123 volume compared with conventional systems [8,10], which impacts the total cost (estimated
124 membrane cost at around 25 USD m⁻² [11]) (i.e. challenge (ii)). The limited removal of small-
125 sized soluble organics (i.e. challenge (iii)) is due to the physical pore size of the membranes
126 but also because of biofilm activity [9]. Lastly, although it is generally known that membrane
127 fouling (i.e. challenge (iv)) results from a biofilm layer of the rejected substances (e.g.
128 particulate organic and inorganic material, organic aggregated colloidal material, and
129 microorganisms) on the GDM surface [12], studies have indicated that the *rate* of membrane
130 fouling is highly dependent on factors such as different types of feed water [6,7,13],
131 membrane geometry (flat sheet or hollow fibers) [14,15], hydrodynamics [16], and operation
132 mode (continuous or intermittent) [7,17].

133

134 To date, most of the investigations on GDM systems have focused on the submerged
135 configuration [7–9,17–20], although some commercially available HWTS systems are
136 designed to be used in an external configuration (e.g. Squirt [21], SkySpod [22], LifeStraw
137 community [23] and ZeroTWO™ [24]). For all of these systems membrane fouling remains a
138 core issue, although several fouling reduction techniques have been proposed, including
139 membrane surface relaxation [11,17,25,26] and periodic application of shear stress (e.g.
140 flushing [11,17,25] and backwashing [27]).

141

142 The use of recycled membranes in those systems has the potential to overcome some of the
143 aforementioned barriers, such as the availability of membranes (i.e. availability of membrane
144 surface to face challenge (i)), cost (i.e. challenge (ii)), removal of LMW or small-sized
145 neutrals organics (i.e. challenge (iii)), and versatility in terms of the system configurations.
146 The production of recycled membranes using free chlorine solutions is emerging as an
147 alternative management to end-of-life (EoL) reverse osmosis (RO) membrane disposal
148 [28,29]. Senán-Salinas et al. recently estimated that around 72% of EoL RO membranes
149 could be recycled in that way [30]. UF-like and NF-like recycled membranes from EoL RO
150 membranes used in desalination plants have demonstrated to be comparable to their
151 commercially available homologues for cross-flow pressure-driven processes [31,32].
152 Amongst recycled membrane applications, only two cases assessed their potential use as
153 HWTS system (submerged GDM system configuration) [28,33]. Indeed, the production cost
154 of the second-hand membranes has been estimated to be between 1 USD m⁻² [34] and 6 USD
155 m⁻² [30]), which could drop the cost of HWTS systems significantly. Since the NF-like and
156 UF-like recycled membranes originate from standard RO spiral-wound membranes, the
157 conventional pressure vessels used in desalination facilities, normally manufactured to host 6-
158 8 membrane modules, remain the most used type of membrane housing available. Alternative
159 membrane housings for single RO membrane modules are usually used for characterization
160 and punctual tests [31], and other alternatives have been patented [35–38]. However, all of
161 them feature complex arrangements, mostly focused on high-pressure resistance and most
162 include an external housing (vessel) to host the whole membrane.

163

164 Less complex arrangements may enable low-cost and compact HWTS systems which utilize
165 recycled spiral-wound membranes. However, appropriate membrane housings for this very
166 specific application are still needed. At present, there is no simple, low-cost and design

167 option or system configuration designed for GDM processes. It is also not well known how
168 recycled membranes for producing drinking water can handle issues of fouling and clogging.
169 Finally, strict validation methods are needed to limit the risk pathogen presence in the
170 permeate [28].

171

172 In the present study, we investigate the development of two alternative membrane housings to
173 the standard spiral-wound pressure vessels for recycled membranes. The focus is on a
174 systematic approach at comparing membrane housings, system configurations (submerged vs.
175 external) and operation modes (continuous vs. intermittent) in terms of their effect on NF-like
176 and UF-like recycled membranes from a pristine RO spiral-wound membrane. Membrane
177 performance, fouling, and feed spacer clogging were assessed. In addition, selected results
178 were validated by using a recycled membrane from an EoL RO spiral-wound module. Since
179 these key factors have not been investigated in any previous study, this will provide a
180 systematic new data set for the GDM field. Critically, this study also used these results to
181 investigate the technical and economic limitations of gravity-driven recycled membrane-
182 based (GDRM) systems to identify promising HWTS designs for rural areas and emergency
183 response.

184

185 **2. Methods**

186 In this study, a wide range of design and operation parameters such as membrane housing
187 design, system configuration, membrane orientation, filtration mode and type of feed water
188 were assessed to identify the optimal design and operating conditions for the NF-like and UF-
189 like recycled membranes in dead-end GDM systems (see Table 1). A total of 51 short-term
190 experiments using deionized water (DI) were carried out. These experiments (using the

191 constant transmembrane pressure (TMP) of 0.16 bar) were repeated between 2-4 times,
 192 determining an experimental error of less than 12%. Moreover, 14 experiments using
 193 synthetic river water (SRW) solutions were carried out.

194

195 Table 1. Summary of studied parameters

GDRM system	Advanced	Simplest	
RO membrane origin	Pristine module	Pristine module EoL	Section 2.1
Spiral-wound recycled membrane type	NF-like UF-like	NF-like UF-like	Section 2.1
Membrane housing: spiral-wound module fiber glass with a complementary device	End-caps design	Simple fitting	Section 2.2
System configuration	Submerged External	Submerged	Section 2.3
Membrane orientation	Vertical Horizontal	Vertical	Section 2.3
Feed water	DI water SRW solutions	DI water SRW solutions	Section 2.4
Filtering modes	Continuous Intermittent: relaxation time plus forward flushing	Continuous Intermittent: relaxation time	Section 2.5.1

196

197 2.1 Membranes types and recycling process

198 A new and an EoL RO spiral-wound module (e.g. TM730HP-4611 from Dow Filmtec; with
 199 a 0.29 m length, a 0.12 m diameter and a 2.23 m² surface area) were used. According to the
 200 manufacture's specification datasheet, this membrane model is often applied in municipal and
 201 industrial systems. The main study was conducted based on the recycling (or conversion) of a
 202 pristine RO membrane. The EoL RO membrane was stored dry for 3 years (no information
 203 related to its operation life was available). The EoL RO membrane was rehydrated before
 204 being recycled, using a 50% w/w ethanol solution for 15 minutes—with full procedure
 205 described in [33]. This membrane presented external damage, which made it not possible to
 206 use the advance membrane housing described in Section 2.2. Thus, the recycled EoL
 207 membrane was used to confirm that the pristine recycled membranes have similar
 208 performance trends to the true EoL membrane.

209

210 To produce NF-like and UF-like recycled membranes, both pristine and EoL RO membranes
211 were passively immersed in a solution containing 6,000 ppm of free chlorine, following an
212 existing protocol from the literature [31]. In this process, the polyamide (PA) layer of the RO
213 membrane was degraded to NF-like and UF-like levels by controlling the exposure time. The
214 standard exposure dose of 6,000 ppm·h of free chlorine was enough to achieve the NF-like
215 performance when using the EoL RO membrane, while a higher exposure dose was required
216 for the pristine model (26,500 ppm·h). To achieve UF-like performance both membranes
217 were overexposed to 300,000 ppm·h. Afterward, the membrane was thoroughly rinsed with
218 DI water. Although the PA layer was chemically attacked, the membrane module maintained
219 its original spiral-wound structure.

220

221 The success of the recycling process was confirmed by filtering DI water in gravity-driven
222 conditions (Section 2.3). The resulting permeability values were compared to the
223 permeability ranges found in literature (i.e. 1.5-15 and 10-50 L m⁻² h⁻¹ bar⁻¹ for NF and UF
224 performance, respectively [39]). Additionally, NF-like performance achieved by recycling the
225 pristine RO module was further validated by using a cross-flow pressure-driven test (Section
226 2.7.1). The total degradation of the PA layer was not confirmed in this work. However, it was
227 earlier demonstrated by the authors that UF-like performance was always achieved when RO
228 membranes were overexposed at the same conditions [29,40].

229

230 **2.2 Membrane housing devices**

231 In this study the fiber glass of the spiral-wound module was directly used as the main part of
232 the membrane housing of the GDRM system. Two additional devices were designed to
233 complement the fiber glass housing, based on light-weight and low-cost materials appropriate
234 for GDM systems. Taken together this represents an alternative solution to the conventional

235 fiber glass or stainless steel membrane pressure vessels which are typically used for cross-
236 flow pressure-driven systems (e.g. membrane testing [31]).

237

238 The lowest-cost device, named from here as the simple fitting, consisted of implementing a
239 simple fitting in the permeate tube at the tail end side. Figure A1-1a (which can be found in
240 the Supplementary data in Annex 1) shows that the simple fitting reduced the 0.019 m
241 diameter of the permeate tube to a 0.003 m diameter for the flexible line. Both ends of the
242 membrane module remained open, so the membrane only operated in a dead-end submerged
243 system configuration (see Section 2.3, Figure 1-Scenario A). The feed water filled the module
244 using both open ends of the membranes, while the treated water was collected using the
245 permeate tube.

246

247 An advanced membrane housing design, named from here as the end-caps design, was also
248 developed to close the open areas of the membrane end sides. This end-caps design was
249 based on two recyclable polyvinyl chloride (PVC) end parts, matching with the diameter of
250 the commercial RO module (as shown in Figure A1-1b in Supplementary data in Annex 1).
251 The end-caps design included the mechanical fixing of these two end-caps to the membrane
252 to prevent leakage from the feed and the permeate side of the module. While the liquid feed
253 flow in and out of the shell side of the module was directed via feed inlet and outlet fittings,
254 the treated water was directed out from the permeate collecting duct through the permeate
255 outlet (see Section 2.3, Figure 1). In the present study, the system was set up to filter in a
256 dead-end mode by blocking the concentrate stream with a valve. However, the concentrate
257 line was used to flush feed water after a set relaxation time (see Section 2.5). The feed water
258 was pumping in following the flow direction indicated by the membrane manufacture and the

259 treated water was collected using the original permeate tube. Detailed figures of the end-caps
260 design are shown in the Supplementary data (Annex 1) and in a recent patent [41].

261

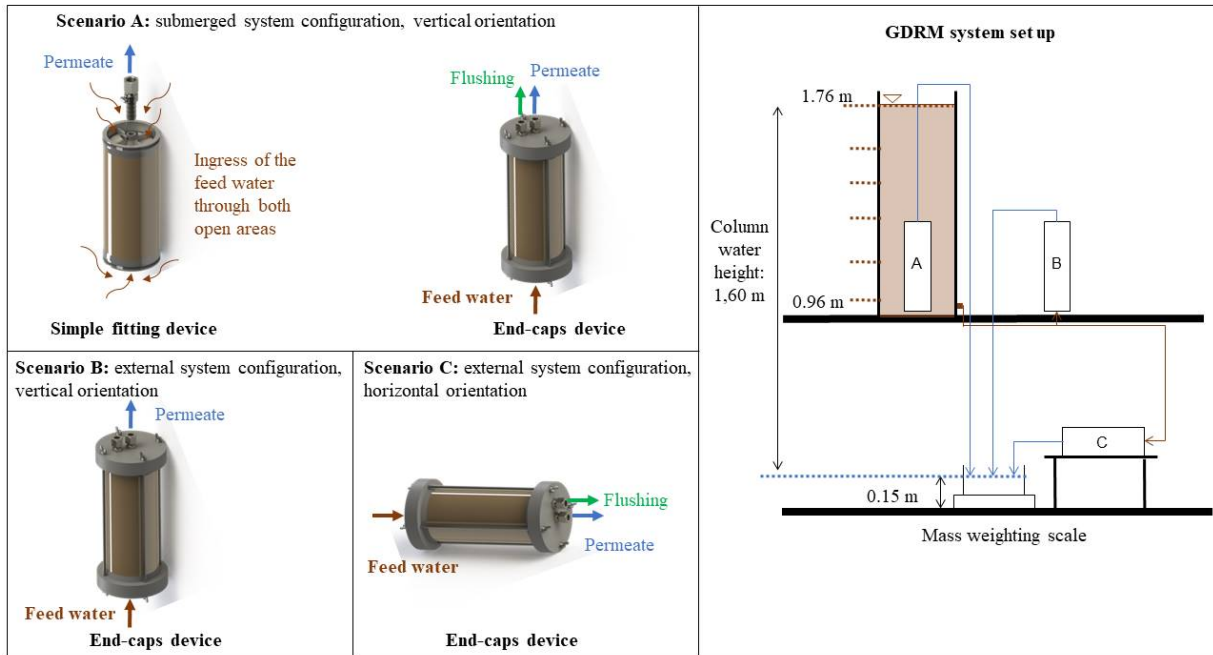
262 **2.3 System configurations**

263 A pilot-scale GDRM system was set up for two distinct membrane system configurations:
264 submerged and external, as shown in Figure 1. In the submerged configuration (Figure 1-
265 Scenario A), the membrane was placed vertically inside the feed tank (20 L). In the external
266 configuration, two membrane orientations were tested: vertical (Figure 1-Scenario B), as
267 usually operates gravity-driven systems for HWTS and horizontal (Figure 1-Scenario C), as
268 is commonly found for cross-flow pressure-driven spiral-wound membranes. In Scenario C,
269 the membrane was placed close to the floor aiming at reproducing the most likely scenario in
270 a potential HWTS application using the standard spiral-wound membranes (with 37 m²
271 surface area and around 15 kg drained weight [42]). During the test, the room temperature
272 was maintained at 20 ± 1 °C. In addition, the permeate line was filled with water and kept
273 filled during the experiment.

274

275 The permeate collected during a given period of filtration was weighed in a balance and then
276 recirculated to the feed tank (i.e. a batch operation). The permeate volume was calculated
277 considering the water density as pure water. Therefore, the membrane permeability
278 (L m⁻² h⁻¹ bar⁻¹) was obtained by dividing the permeate flow rate to the membrane surface
279 area and the TMP. The TMP was estimated considering the effective water head, which
280 includes the height of the water level of the tank and the height of the end of the permeate
281 tube [17,43].

282



283

284 Fig 1. Set-up of the GDRM system using submerged (Scenario A) and external (Scenario B and Scenario C)
 285 system configurations. In Scenario B, the membrane was placed vertically at the same height than in Scenario
 286 A. In Scenario C the membrane was placed horizontally. Note that the blue permeate lines were always filled
 287 with water, while brown lines represent the feed line.

288

289 2.4 Feed water solutions

290 2.4.1 Deionized water tests

291 DI water tests were conducted for comparison of membrane housing devices at various TMP
 292 levels. The water level of the tank was kept constant for all the selected TMP tests, during
 293 30 minutes of each test. The TMP varied from 0.16 to 0.08 bar, so these values were used to
 294 determine the membrane permeability. Both membrane housing designs and the three system
 295 configurations were studied for both NF-like and UF-like recycled membranes.

296

297 DI water was also filtered before and after conducting the fouling test (see Section 2.5). After
 298 each set of experiments using SWR feeds, the NF-Like and UF-like recycled membranes
 299 were backwashed ($0.43 \text{ L m}^{-2} \text{ h}^{-1}$) and DI water was tested again for comparison with the
 300 initial values.

301

302 **2.4.2 Synthetic river water tests**

303 Fouling tests were conducted with SRW solutions (with $900 \mu\text{S cm}^{-1}$ of conductivity, neutral
304 pH and $2\text{-}6 \text{ mg L}^{-1}$ of dissolved organic carbon (DOC), given by bovine serum albumin
305 (BSA), humic acids (HA) and alginate. The SRW solutions were renovated in each
306 experiment. The SRW solutions were used to assess both membrane housing devices for NF-
307 like and UF-like recycled membranes configured in Scenario A and Scenario C, at a constant
308 pressure of 0.16 bar.

309 Two SWR feed solutions were used. Feed A simulates pre-filtered river water (i.e. 80 mg L^{-1}
310 MgSO_4 , 400 mg L^{-1} NaCl, 12 mg L^{-1} BSA, 12 mg L^{-1} HA) which was employed when using
311 the NF-like recycled membranes and the UF-like recycled EoL membrane. The second
312 solution, Feed B (i.e. 80 mg L^{-1} MgSO_4 , 400 mg L^{-1} NaCl, 30 mg L^{-1} BSA, 30 mg L^{-1} HA,
313 6 mg L^{-1} alginate acid and 300 mg L^{-1} diatomaceous earth), was used with the UF-like
314 recycled pristine membrane to accelerate fouling on the membrane surface (due to increased
315 organic content and suspended solids compared to Feed A). All the chemicals were analytical
316 grade reagents. The concentration of the main organic fraction of Feeds A and B are shown in
317 the supplementary data (Table A2 in Annex 2).

318

319 **2.5 Assessment of continuous and intermittent operation**

320 **2.5.1 Filtering modes**

321 The impact of two filtration modes (continuous and intermittent) on the NF-like and UF-like
322 recycled membrane performance and feed spacer clogging were evaluated, for both GDRM
323 systems (simple and advanced) and two system configurations (i.e. Scenario A and
324 Scenario C). The total filtration time was 6 h. The intermittent condition was tested because it
325 simulates a more realistic operation mode for HWTS systems. Thus, in this work, the

326 operation mode consisted of cycles of 1 h of filtration, followed by a relaxation time of 40
327 minutes (or 12 h in the case of the last cycle). For the advanced GDRM system, relaxation
328 time was followed by a thorough flushing of 10L of the feed water (at $\sim 140 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}$ for
329 2 minutes at 0.16 bar), while for the simple GDRM system flushing was not possible.
330 Flushing was conducted by opening the valve of the concentrate line, which was already
331 filled with feed water at the beginning of each experiment (no additional components or input
332 energy was required for this step).

333

334 **2.5.2 Membrane rejection and fouling**

335 The feed and permeate water samples were collected initially and after 1 h and 6 h of
336 filtration for further analysis. The major component rejection (% R) was calculated following
337 Equation 1. Where, C_p and C_f are the specific concentration measured at permeate and feed,
338 respectively.

$$339 \quad \% R = \left(1 - \frac{C_p}{C_f}\right) \cdot 100 \quad \text{Eq. 1}$$

340 To assess the percentage of organic fouling accumulation (% FA) inside the module
341 (i.e. DOC and the organic fractions), the feed concentration reduction was calculated
342 following Equation 2. Where C_{f-0} and C_{f-6} are the concentration of feed components at the
343 beginning and after 6 h of duration, respectively.

$$344 \quad \% FA = \left(\frac{C_{f-0} - C_{f-6}}{C_{f-0}}\right) \cdot 100 \quad \text{Eq. 2}$$

345 The accumulation of solid inside the membrane was calculated with a mass balance by
346 considering the initial SS added to the tank (6 g) and the SS contained in the water samples
347 collected after applying the following cleaning process: drainage after filtering SRW model
348 solution, drainage after filtering DI water, and the 10 L backwashing process.

349

350 **2.6 Water quality analysis**

351 Conductivity, pH, and turbidity were tested with a multiparameter sensor (Hanna Instruments
352 HI9829). The SS were determined following the APHA 2540 D method. DOC and the
353 organics fractions were analyzed based on size exclusion (Toyopearl TSK HW-50S, diameter
354 2 cm, length 25 cm) using a dual column liquid chromatography organic carbon detection
355 (LC-OCD), system model 8, based on the Gräntzel thin film reactor developed by DOC
356 Labor, Germany [44]. Chromatographs were interpreted using the DOC Labor ChromCALC
357 2013 software. Thus, the organic components used such as BSA, HA and alginate represented
358 in the chromatographs as biopolymers (BP), HS and polysaccharides (which elute at similar
359 retention time than BP), respectively [45]. Other organic fractions studied in this work are the
360 BB (which reflects breakdown products of HS with lower molecular weight) and LMW-
361 neutrals (which reflect organic fractions with even lower molecular weight) [44].

362

363 **2.7 Further testing and economical assessment for decision-making**

364 **2.7.1 Cross-flow pressure-driven test**

365 Cross-flow pressure-driven assays (up to 4.4 bar) using the end-caps device were conducted
366 to assess the membrane performance after being exposed to free chlorine, to confirm the
367 partial degradation of the RO PA layer. Tests were run for 30 minutes for filtering different
368 saline concentration solutions, up to 3750 $\mu\text{S cm}^{-1}$ (e.g. 2000 mg L^{-1} NaCl or 2000 mg L^{-1}
369 MgSO_4). The feed flow was maintained constant at 3 L per minute and the room temperature
370 was maintained at 22 °C. Another goal of this study was to demonstrate the mechanical
371 resistance of the advanced membrane housing device (end-caps) for using the NF-like
372 recycled membrane in standard cross-flow filtration.

373

374

375 2.7.2 Economic assessment

376 After determining the technical performance, the next step in the present study considered the
377 economic performance of the proposed designs. The first step in this analysis was to
378 determine the capital expenditures or investment cost (CapEx) of the GDRM system. This
379 included the novel membrane housing devices and one recycled membrane, which was
380 calculated on an annual equivalent cost (AEC; USD/year) using Equation 3.

$$381 \quad AEC = C_0 \left[\frac{r(1+r)^n}{(1+r)^n - 1} \right] \quad \text{Eq. 3}$$

382 In Eq. 3, C_0 represents the cost of each item at year 0 (USD), r is the discount (interest) rate
383 per period (e.g. assumed as 2 % per year), and n is the number of years (e.g. the lifespan). A
384 conservative lifespan of only one year was selected aiming at the application of the GDRM
385 system in an emergency response context or early recovery projects. The operational
386 expenditures (OpEx) were considered negligible because of the simplicity of the system, with
387 no need for electrical inputs or cleaning products (few cleanings are expected in long-term
388 operation due to the phenomenon of flux stabilization [7]). Therefore, the total annual cost
389 (TAC), which is the sum of CapEx and OpEx was assumed to be equal to CapEx.

390

391 The cost of producing NF-like and UF-like membranes can be lumped as a cost for the EoL
392 RO recycling process. According to the literature [30], the production cost of 4500
393 membranes using a passive recycling system was around 6 USD m⁻², including membrane
394 characterization and transport costs. In this work, the selling cost of the recycled membranes
395 (NF-like and UF-like membranes) was assumed to be 7 USD m⁻² (data used in this study),
396 which would include the company profits.

397

398 The cost of the simple fitting device and the rest of the parts of the GDRM system (e.g. those
399 described in Sections 2.2 (Figure A1-1 in Annex 1 in Supplementary data) and Section 2.3
400 (Figure 1)), were found from online suppliers (in Australia), where most of the materials were
401 purchased. The end-caps device cost was estimated by knowing the quantity of PVC material
402 used and the labor (e.g. person-h) needed to manufacture it (excluding design time).

403

404 **3. Results and discussion**

405 **3.1 Initial DI testing**

406 The main objective of the initial DI water tests was to assess the relative hydraulic
407 advantages of the end-caps device compared to the simple fitting, the impact of the system
408 configuration (submerged vs. external) and the membrane orientation. For that purpose, 20 L
409 of DI water was filtered with no water recirculation, until the tank was empty.

410

411 Table 2 shows the average permeability values of the NF-like and UF-like recycled pristine
412 membranes for the distinct TMP. Similar permeability values were obtained when using both
413 membrane housings in the submerged system configuration and membranes were fully
414 covered with water (TMP 0.16-0.10 bar). Therefore, the use of the end-caps device does not
415 seem to have a significant impact on hydraulic membrane performance. Membrane
416 orientation, vertical or horizontal, does not seem to have any impact neither (during external
417 system configuration testing).

418

419 Table 2. NF-like and UF-like recycled pristine membrane permeability ($L m^{-2} h^{-1} bar^{-1}$) values filtering DI water
 420 in continuous mode. The simple fitting and end-caps devices are compared using the submerged configuration
 421 system and all Scenarios. It is shown the average of data achieved varying TMP from 0.16 to 0.10 bar (using
 422 data of 4 experiments, n=4), and at 0.08 bar of TMP (one experiment, n=1).

Parameter			Permeability ($L m^{-2} h^{-1} bar^{-1}$)			
System configuration			Submerged		External	
Scenario - Membrane orientation			A-Vertical	A-Vertical	B-Vertical	C-Horizontal
Membrane housing			Simple fitting	End-caps	End-caps	End-caps
RO membrane origin	Recycled membrane type	TMP (bar)				
Pristine	NF-like	0.16-0.10, n=4	7.5 ± 0.1	7.9 ± 0.1	7.6 ± 0.0	8.0 ± 0.1
		0.08, n=1	3.5 ± 0.3	7.8 ± 0.3	7.5 ± 0.2	7.9 ± 0.1
	UF-like	0.16-0.10, n=4	18.2 ± 0.0	17.8 ± 0.2	19.2 ± 0.1	19.3 ± 0.3
		0.08, n=1	6.6 ± 0.3	17.2 ± 0.7	19.3 ± 0.2	18.8 ± 0.1

423 N.A.: No data available

424 However, differences in permeability were observed in the submerged configuration system,
 425 when the membrane was not fully covered by water. When using the simple fitting, the
 426 permeability values decreased up to around 3-fold (highlighted in Table 2). Air entered inside
 427 the membrane module when the water level was lower than the membrane height. The
 428 negative impact on the water permeability was due to the reduction of the active membrane
 429 surface. These results are in concordance with the previous publication, where a totally or
 430 partially submerged flat sheet gravity-driven UF membrane performance was assessed [8].
 431 The end-caps device prevented this effect, keeping the membrane surface fully covered by
 432 water. Therefore, the permeability was maintained constant at any hydraulic pressure.

433
 434 Considering the application of the system using a single membrane module for HWTS and
 435 aiming at preventing the risk of air ingress in modules, the end-caps device seems to be a
 436 better option than the simple fitting. Nevertheless, further study was conducted to assess the
 437 impact of those configurations on fouling. The external system configuration using the
 438 membrane vertically (Figure 1-Scenario B) was not tested further due to the complexity of
 439 the set up.

440

441 **3.2 Synthetic river water tests**

442 The following sub-sections evaluate the NF-like and UF-like recycled membrane
443 performance as a function of the following variables: i) the filtration mode (continuous vs.
444 intermittent), ii) the system configuration (vertically-submerged vs. horizontally-external)
445 and iii) the accumulation/sorption of the organic compound and SS on the membrane surface.
446 To assess the potential risk of clogging of feed water, the membrane permeate was
447 continuously circulated to the feed tank. For instance, for this series of experiments, during
448 the 6 h of filtration, the NF-like recycled pristine membrane filtered a total of 3.5 L of feed
449 water, while the UF-like recycled pristine membrane filtered 34.2 L.

450

451 **3.2.1 Permeability**

452 **3.2.1.1 Assessment of NF-like recycled membrane performance**

453 Figure 2 shows the permeability and salt rejection when operating in continuous and
454 intermittent filtration modes for the NF-like recycled membrane submerged in the feed tank
455 (Scenario A) and externally configured (Scenario C). The recycled pristine membrane was
456 housed in both the simple fitting (Figure 2-a) and the end-caps devices (Figure 2-b and
457 Figure 2-c). Continuous filtration results show that the initial permeability (around
458 $2.1 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) decreased over time until reaching a plateau (around $1.7 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$),
459 where the rate of deposition and accumulation of foulants were balanced by back diffusion
460 and re-dissolution [46]. The recycled EoL membrane (Figure 2-d) showed a similar
461 permeability.

462

463 During the intermittent filtration, in each filtering cycle, permeability temporarily increased,
464 followed by a decrease towards the stable value. A slight improvement was observed when
465 flushing was applied after the relaxation time (membrane hosted in the end-caps device).

466 Indeed, no significant membrane performance differences were observed between cycles 2
467 and 4. In all cases, a longer relaxation time (12 h between cycles 5 and 6) improved the
468 average membrane permeability.

469

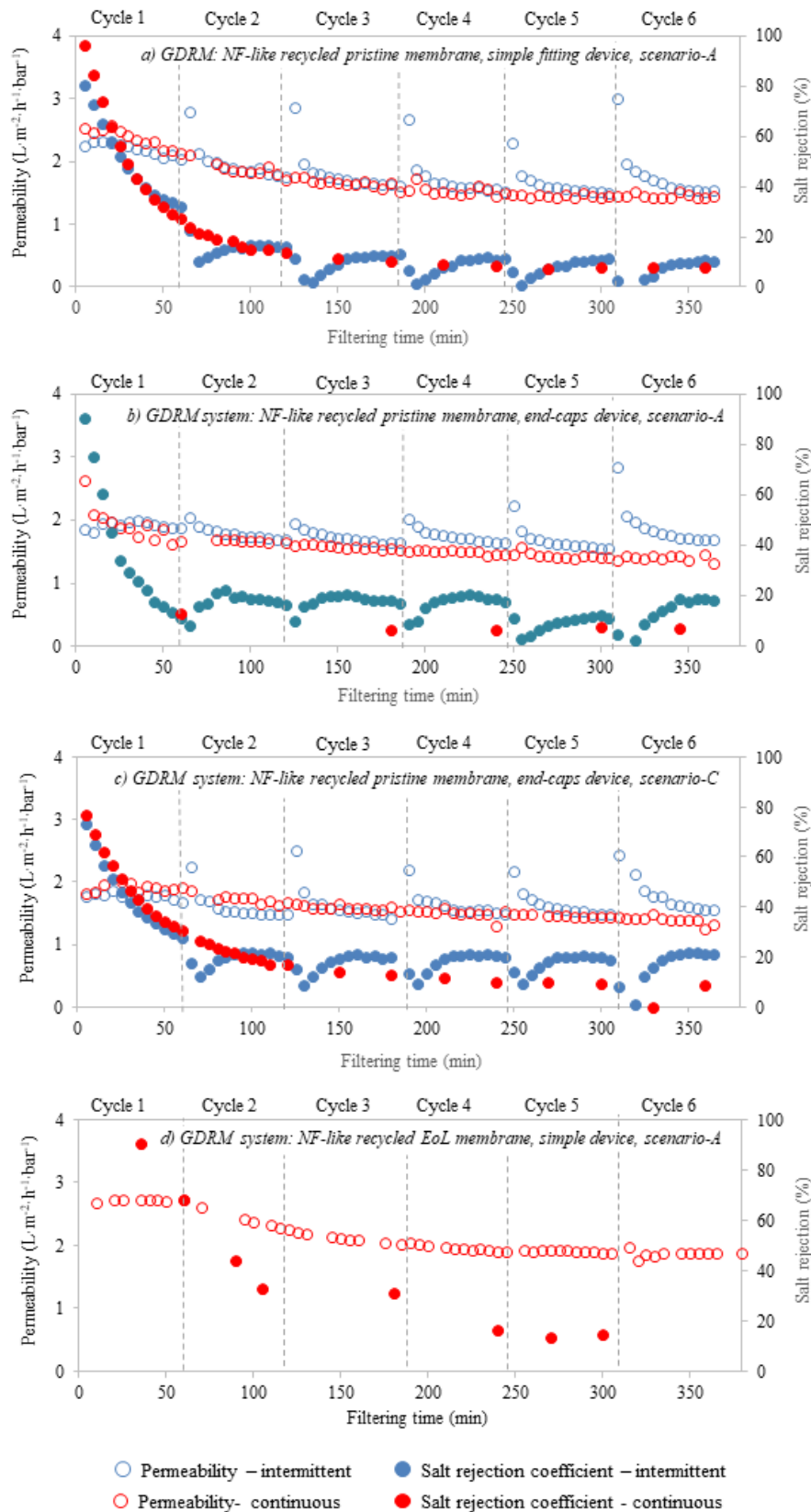
470 In contrast to what typically occurs in cross-flow pressure-driven systems with NF
471 membranes, a sharp decline in salt rejection was observed in the first h of filtration to fall
472 under 16% after 80 minutes. This is likely caused by the formation of a boundary layer near
473 the membrane surface, which appears when salts are rejected, also referred to concentration
474 polarization (CP). In the CP layer, the salt concentration is higher than the salt concentration
475 in the bulk solution, which can explain the low rejection values and the permeability
476 reduction [47]. The decrease of the salt rejection at the beginning of the following cycle
477 compared to the last cycle was, therefore, linked to the formation of the CP layer. There was
478 also a salt diffusion effect when the filtration stopped and it took around 30 minutes to
479 stabilize again to the plateau. Salt diffusion might occur due to the high concentration of salt
480 over the membrane surface. The combination of membrane relaxation and flushing (Figure
481 2-b and Figure 2-c) has the greatest impact to improve the salt rejection compared to apply
482 only the relaxation time (Figure 2-a). It was hypothesized that water flushing was responsible
483 for the decrease of the thickness of the boundary layer near the membrane surface [48]. Low
484 salt rejection will limit the application of NF membranes to only freshwater feeds, but they
485 could still serve in HWTS, much like UF membranes (with null salt rejection capability).

486

487 Continuous filtration results also confirmed that the usage of the end-caps device does not
488 have a significant impact on the membrane performance. In all the three tested Scenarios,
489 fouling was mostly reversible, since the permeability recovered generally well after applying
490 backwashing (data are shown in Supplementary data in Table A3-3 in Annex 3). A

491 comparison with other similar studies was not possible, since NF membranes have never been
492 used in GDM systems for producing drinking water before. However, the impact of
493 combining relaxation time and flushing has been earlier reported using UF membranes (as
494 will be discussed in the comparison below).

495



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502

Figure 2. Permeability and salt rejection of the NF-like recycled pristine membrane: a) Submerged configured using the simple fitting (Scenario A), b) Submerged configured using the end-caps device (Scenario A), c) External configured using the end-caps device (Scenario C) and d): Permeability and salt rejection of the NF-like recycled EoL membrane submerged configured using the simple fitting (Scenario A). Table A3-1 (Annex 3 in Supplementary data) shows the average values and the standard deviation of all the cases. [Note: In figure b), between cycle 4 and 5, no flushing was applied].

503 3.2.1.2 Assessment of UF-like recycled membrane performance

504 Figure 3 shows the permeability when operating in continuous and intermittent filtration
505 modes for the UF-like recycled membranes submerged in the feed tank (Scenario A) and
506 externally configured (Scenario C). The recycled pristine membrane was housed in both the
507 simple fitting (Figure 3-a) and the end-caps devices (Figure 3-b and Figure 3-c). The UF-like
508 recycled EoL membrane was housed in the simple fitting (Figure 3-d) and the salt rejection
509 coefficients were below 5% in all the cases (not shown in the figure).

510

511 Despite the accumulation of SS (Section 3.2.4) and of protein (see Section 3.2.3 Table 4),
512 which is known to cause severe fouling in low-pressure UF process [49,50], the permeability
513 of the recycled pristine membrane slightly declined during the first h and then it remained
514 constant over time in both filtration modes. The low average permeability indicated a
515 permeate flux rate around $2.6 \text{ L m}^{-2} \text{ h}^{-1}$, which was lower than the typical reported standard
516 stabilized flux of $4\text{-}10 \text{ L m}^{-2} \text{ h}^{-1}$ for UF GDM systems operated long-term [6]. However, it
517 was still similar to studies that treated diluted wastewater ($15 \text{ mg L}^{-1} \text{ DOC}$) [18] and grey
518 water [51]. This can be mainly attributed to several factors: i) the nature of the PSF layer of
519 the recycled membranes, which are designed to support the PA layer of the RO membranes;
520 ii) the feed water quality, a SRW solution rather than natural water, and iii) the membrane
521 modules type, which in our study was spiral-wound rather than hollow fiber. However, higher
522 initial permeate flux for spiral-wound UF recycled modules also resulted in higher flux
523 decline, even though the feed was SS-free. The initial flux of the UF-like recycled EoL
524 membrane ($7.4 \text{ L m}^{-2} \text{ h}^{-1}$) is expected to be above its critical value, while the UF-like recycled
525 pristine membrane operated below it (around the aforementioned flux of $2.6 \text{ L m}^{-2} \text{ h}^{-1}$),
526 without showing any reduction in permeability [52]. The spiral-wound membrane
527 configuration has an impact on the critical flux compared to other configurations used in

528 GDM systems (i.e. hollow fiber), due to lower shear rates [53]. Therefore, the critical flux for
529 the recycled spiral-wound UF membrane is expected to be within the standard range of the
530 stabilized flux reported in the literature for GDM systems (i.e. 4-10 L m⁻² h⁻¹ [6]).

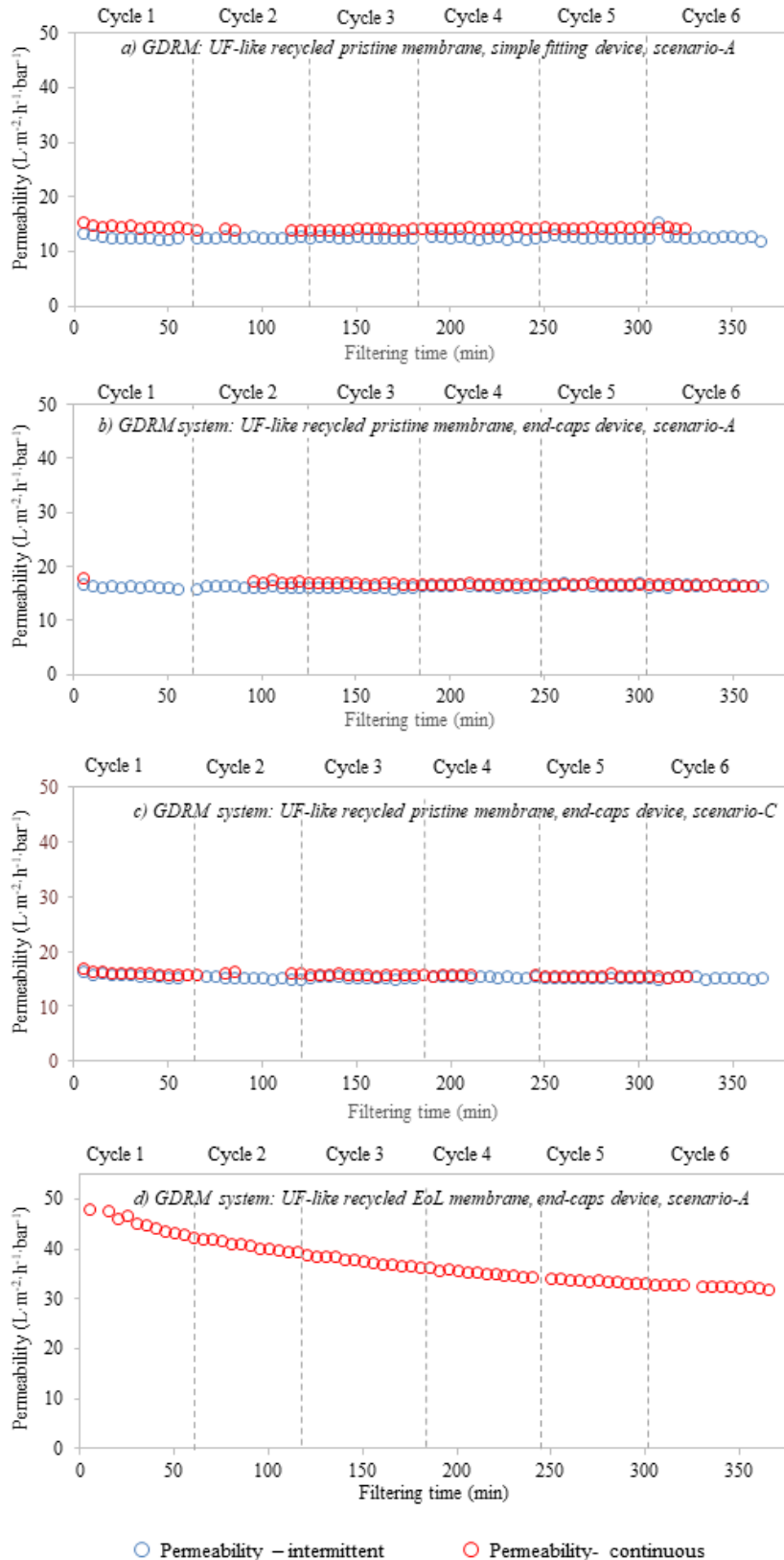
531

532 When operating with the UF-like recycled pristine membrane housed in the end-caps device,
533 an improvement in the permeability was not observed when applying flushing after the
534 relaxation time (when using the end-caps device). However, previous studies using UF GDM
535 systems to treat surface water reported that intermittent operation, when combined with
536 forward flushing, further enhanced the membrane performance [11,17,25]. Shi et al. [17]
537 demonstrated that only applying membrane relaxation or shear stress (flushing cross-flow
538 velocity at 360 m h⁻¹) was ineffective in improving the stable flux in long-term filtration,
539 while their combination enhanced the stable flux by 70%.

540

541 As occurred with NF-like recycled membranes, the permeability recovered after applying
542 backwashing indicating the reversibility of fouling (values are shown in Supplementary data
543 (Table A3-3 in Annex 3).

544



545
 546 Figure 3. Permeability of the UF-like recycled pristine membrane: a) Submerged configured using the simple
 547 fitting (Scenario A), b) Submerged configured using the end-caps device (Scenario A), c) External configured
 548 using the end-caps device (Scenario C) and d) Permeability of the UF-like recycled EoL membrane submerged
 549 configured using the simple fitting (Scenario A). Gaps in the figures are due to experimental issues of the data
 550 acquisition device. Table A3-2 (Annex 3 in Supplementary data) shows the average values and the standard
 551 deviation of all the cases.

552 3.2.2 Organic rejection

553 Table 3 shows the rejection coefficients (average values for the observed rejection and the
554 standard deviation) of DOC and four fractions: BP, HS, BB and LMW neutrals, with no
555 distinction of the system configuration and filtering mode. The NF-like membrane showed a
556 high ability to separate organic compounds, reaching rejection coefficients above 77% in
557 both recycled pristine and recycled EoL membranes. The UF-like recycled pristine membrane
558 still rejected protein (at above 99% BP) but only 36% and 61% rejection was observed for
559 BB and LMW-neutrals, respectively. The UF-like recycled EoL membranes showed lower
560 protein rejection (around 74%), being in concordance with the high permeability results.

561

562 Although HS is potentially larger compounds than BB or LMW-neutrals, similar
563 concentration was observed in the feed and the permeate streams when using recycled UF
564 membranes. It was hypothesised that HS were not able to form aggregates with bigger size as
565 occurred in long-term GDM filtration investigation, and therefore the HS easily permeated
566 [46]. Very low rejection of HS has been reported in the literature for UF membranes [27],
567 where rainwater was treated by submerged hollow fiber GDM system. However, the long-
568 term operation of other UF GDM systems showed above 20% rejection [46,54]. On one hand,
569 such a difference compared to the HS rejection observed in our work could be explained due
570 to the molecular size. Ding et al. [54] used bigger HS molecules, in the range of 1000-3500
571 Da, while in our work the average size was defined around 600 Da. On the other hand, in
572 long-term experiments (operating during several weeks), the bio-fouling layer developing on
573 the membrane surface is expected to act as a secondary barrier, enhancing the rejection of
574 organics such as BP, HS [54], BB and LMW compounds [27].

575 Table 3. Rejection (in %) observed filtering SRW solutions using recycled membranes. Average and standard
 576 deviation with no distinction of the system configuration and filtering mode (A total of six experiments (n=6)
 577 for each membrane type was used when using the recycled pristine membrane data, while only one experiment
 578 (n=1) represents the case of using the recycled EoL membranes).
 579

Parameter	Organic rejection (%)			
	Submerged and external		Submerged	
System configuration	Submerged and external		Submerged	
Scenario - Membrane orientation	A-Vertical and C-Horizontal		A-Vertical	
Membrane housing	Simple fitting and End-caps		Simple fitting	
Feed solution	A	B	A	A
Filtering mode	Continuous and Intermittent		Continuous	
Number of experiments, n	n=6	n=6	n=1	n=1
RO membrane origin	Pristine		EoL	
Recycled membrane type	NF-like	UF-like	NF-like	UF-like
DOC	88.6 ± 3.2	48.0 ± 14.6	81.5	37.8
BP	99.4 ± 0.5	99.7 ± 0.4	93.1	74.2
HS	97.2 ± 3.0	12.5 ± 4.4	81.7	22.0
BB	84.8 ± 11.0	35.7 ± 10.6	82.6	35.0
LMW-neutrals	77.9 ± 5.8	61.2 ± 14.0	78.7	30.7

580

581 3.2.3 Fouling accumulation

582 Due to the mode of filtration (recycling of permeate into feed tank), the accumulation of
 583 foulants on the membrane was expected. It was assumed that in short-time experiments, the
 584 degradation of organic substances in the feed was negligible. Therefore, the reduction of the
 585 organic fraction concentrations at the feed after 6 h of filtration compared to the initial values
 586 represented the percentage of fouling accumulation (FA) inside the membrane. Table 4 shows
 587 the FA values of BP and HS for the NF-like and UF-like recycled membranes operating in
 588 the submerged and external system configurations, in the continuous and intermittent filtering
 589 mode. Similar FA was observed during continuous and intermittent filtration when the
 590 recycled pristine membrane was housed in the simple fitting. The simple fitting device does
 591 not enable water flushing after the relaxation time. Thus, applying only membrane relaxation

592 did not have a significant impact on preventing fouling accumulation inside the module. In
593 this case, the lowest FA was observed when the recycled pristine membrane was housed in
594 the end-caps device and operated intermittently (i.e. lower reduction of the feed organic
595 fraction concentrations over time). Therefore, less fouling accumulated inside the module.
596 This is again a consequence of the combination of the filtration relaxation and water flushing.

597

598 The combination of relaxation time and flushing is expected to reduce the fouling tendency
599 over the membrane. For instance, in the case of the NF-like recycled pristine membrane, 23%
600 and 16% FA of BP and HS, respectively, were observed when filtering in continuous mode
601 (using both housings devices). However, less than 12% FA of both compounds was observed
602 when flushing was applied after the relaxation time. These results also proved that the BP and
603 HS accumulated or deposited inside the membrane module, but there was no significant
604 adsorption or aggregate deposition over the membrane. Deposition and adsorption of BP and
605 HA have been earlier reported as potential fouling mechanism in GDM systems [26,46]. For
606 UF-like recycled pristine membrane, around 40% FA of the BP concentration in the feed was
607 observed when flushing was applied, while more than 87% FA was detected when membrane
608 operated continuously. Chawla et al. [11] observed that the foulant load delivered per unit
609 membrane area was lower (around half) when hollow fiber UF membranes operated
610 intermittently comparing with a 24 h continuous filtration. However, this could be attributed
611 to the fact that the total filtering time in both operation modes were different. In addition,
612 other investigation also highlighted that applying high cross-flow conditions after the
613 membrane relaxation caused a higher reduction of foulants accumulation [17].

614

615 When using the recycled EoL membranes (i.e. with the simple fitting device and the
616 continuous filtering mode), a higher FA was observed. The NF-like recycled EoL membrane

617 showed 51% and 47% FA of BP and HS, respectively. The UF-like EoL membrane showed
 618 79% and 45% FA of BP and HS, respectively. This could be due to the residual fouling
 619 expected in the EoL RO membrane, even after applying the recycling protocol.

620
 621 Table 4. FA (in %) of BP and HS inside the membrane module after 6 h of filtration when using recycled NF-
 622 like and UF-like recycled pristine membranes housed in the designed devices and operated in the submerged
 623 and external system configurations in continuous and intermittent filtration modes.

Parameter				FA (in %) of BP and HS after 6 h of filtration					
System configuration				Submerged				External	
Scenario - Membrane orientation				A-Vertical		A-Vertical		C-Horizontal	
Membrane housing				Simple fitting		End-caps		End-caps	
Filtration mode				Continuous	Intermittent (relaxation)	Continuous	Intermittent (relaxation +flushing)	Continuous	Intermittent (relaxation + flushing)
Recycled Membrane Type	NF-like	Organic fraction	BP	26.8	30.6	23.5	4.6	19.1	12.0
			HS	21.6	22.9	15.2	0.0	10.5	6.5
	UF-like	Organic fraction	BP	82.7	83.3	94.6	45.7	78.3	33.8
			HS	9.0	3.3	12.8	23.4	10.1	11.9

624

625 3.2.4 Suspended solids

626 The fate of the SS was assessed during filtrating by the UF-like recycled pristine membrane.
 627 During the experiments, the systematic reduction of turbidity at the feed was observed over
 628 time. On average, feed turbidity dropped from an initial value of 75 to 14 NTU after 6 h of
 629 filtration. As a result, feed turbidity was used as an in-situ indicator of the accumulation of
 630 the SS inside the module. Although the tank was aerated to mix the solution, a settlement was
 631 observed in the feed tank. At the end of the filtration, it was estimated that around $52 \pm 11\%$
 632 of the original SS loading (6 ± 0 g) settled. In addition, no particles were detected neither in
 633 suspension nor in the permeate stream (after 6 h). Therefore, around $39 \pm 8\%$ of the original
 634 SS loading was trapped inside the membrane. Although the positive effect of cross-flow
 635 velocity on fouling limitation has been demonstrated well in literature [55,56], no clear
 636 correlation between the filtering modes was detected. Therefore, we were unable to determine
 637 the positive effect of flushing during the intermittent filtration to prevent the accumulation of

638 SS. This was due to the negligible cross-flow velocity applied ($8.5 \cdot 10^{-5} \text{ m s}^{-1}$), which was
639 estimated according to the literature [56]. Consequently, such a low cross-flow velocity
640 limited the shear rate necessary to flush SS particles away from the feed spacers. Indeed, it
641 might be even lower when using bigger recycled spiral-wound modules (standard size of 8
642 inches diameter and 37 m^2 membrane surface), due to the feed spacer channel pressure drop
643 [56]. The usage of a standard spiral-wound UF-recycled membrane at the same experimental
644 conditions of the present work (GDM and flushing of $138 \text{ L m}^{-2} \text{ h}^{-1}$ feed water) might drop
645 the flushing cross-flow velocity to $3.8 \cdot 10^{-5} \text{ m s}^{-1}$.

646

647 Between 23 and 78% of the solid trapped inside the membrane were recovered in the flushing
648 stage. However, neither the flushing nor the backwash fully recovered the SS. Although the
649 membrane accumulated particles after each filtering cycle, no significant changes in
650 permeability were observed in the subsequent experiments. This was also observed by Peter-
651 Varbanets [46], which demonstrated that adding particles in the feed water (30 and
652 300 mg L^{-1} kaolin) did not influence the level of flux stabilization in long-term UF GDM
653 filtration. However, in longer operation the accumulation of SS inside the module will clog
654 the membrane. Therefore, further tests are encouraged to determine which concentration of
655 SS limits the membrane performance compromising its lifetime.

656

657 **3.3. Further testing and economic assessment for decision-making**

658 Additional experiments were carried out to complement the main research. The limitation of
659 separating salts using the NF-like recycled membrane, how to reduce the SS ingress into the
660 membrane module and an economic analysis were assessed. All experiments together were
661 used to develop a decision-making tree to select the appropriate membrane housing devices
662 and the operating conditions to apply the recycled membranes for HWTS.

663 **3.3.1 Pressure-driven test**

664 The NF-like recycled pristine membrane was characterized in a pressure-driven cross-flow
665 system (up to 4.4 bar) using the end-caps device and saline solutions. The resistance of the
666 end-caps device was tested during 12 h of operation. Contrary to the observation operating
667 with the GDM system, the membrane was able to reject salts up to 89%. Results shown in
668 Supplementary data (Table A4 in Annex 4) are similar to earlier studies focusing on several
669 kinds of NF-like membranes [31,57]. Therefore, NF-like recycled membranes hosted in the
670 end-caps device could be suitable for cross-flow pressure-driven processes at a low feed
671 salinity (below 4000 $\mu\text{S cm}^{-1}$).

672

673 **3.3.2 Preliminary economic assessment**

674 An economic assessment was carried out in this study which conservatively assumes only
675 one year lifespan for the GDRM systems (aiming at its application in emergency response
676 and early recovery WASH projects). The operational cost was assumed to be negligible over
677 one year for this type of GDRM (with no electrical or chemical inputs required). When using
678 the simple fitting, the TAC was 46 USD.yr.⁻¹. As shown in Supplementary data (Table A5-1
679 in Annex 5), the main cost of the system was attributed to the water containers and the
680 recycled membrane (39% and 34% out of the total, respectively). The fitting associated with
681 the membrane housing represented only 21% of the total investment. However, when the
682 end-caps membrane housing was employed (Table A5-2 and Table A5-3 in Annex 5), the
683 manufacturing of the device was the most important contributor, with 74% of the TAC (on
684 average 176 USD.yr.⁻¹ TAC). Despite the fact that the end-caps device showed a marginal
685 technical advantage, it cannot be manufactured by standard and existing items like the simple
686 fitting device. Thus, while the cost of the first end-caps prototype was USD 130, it would
687 drop significantly if massive production and/or by manufacturing in local workshops.

688 These results show that the GDRM system is a low-cost solution (defined by
689 0.01-0.10 USD L⁻¹ [4]) that could compete with the commercially available HWTS systems
690 based on UF membranes for humanitarian context (Table A5-4 in Annex 5 in Supplementary
691 data). Although many commercial HWTS systems claim to have a lifespan up to 10 years
692 (same as membranes), the lifespan of those HWTS systems in an emergency response context
693 depends of many factors (e.g. the treated water quality and membrane surface [3], the risk of
694 the membrane getting dry, replicates of the natural disaster etc.). Therefore, a relative cost
695 comparison was done considering one year lifespan, operating intermittently 8 h per day 7
696 days a week. DI water production for the recycled pristine membranes and the calculated
697 TAC were used for the present case, while the clean water production and the selling price
698 were employed in the case of the commercial systems. The water production cost of 4 out of
699 5 commercial HWTS based on hollow fiber UF membranes was lower or equal to
700 0.2 USD ct. yr. L⁻¹ (Squirt, Sawyer Point, Orisa and SkyPod systems). The GDRM system
701 using UF-like recycled membranes and the simple fitting showed a potential water production
702 cost of 0.2 USD ct. yr. L⁻¹, therefore it would be the most approachable solution to the
703 market. The GDRM system based on the end-caps device with a water production cost of
704 0.9 ct. USD yr. L⁻¹, would be closer to the Safir EAWAG (0.7 USD ct. yr. L⁻¹, flat sheet
705 membranes), the familiar LifeStraw HWTS (1.1 USD ct. yr. L⁻¹, hollow fiber membranes)
706 and the Paul (1.3 USD ct. yr. L⁻¹, flat sheet fiber membranes) systems. Operating with the
707 NF-like recycled membrane would increase up to 2.5-fold the water production cost, since its
708 water production is remarkably lower than the UF-like recycled membrane.

709

710 **3.3.4 Decision-making tree**

711 In this study, the optimized operating conditions using the GDM system were found when the
712 recycled membranes were housed in the end-caps device and operated intermittently

713 (relaxation time plus forward flushing). The NF-like recycled pristine membrane
714 performance at the optimized condition was $1.7 \pm 0.1 \text{ L m}^2 \text{ h}^{-1} \text{ bar}^{-1}$, around 20% salt
715 rejection and close to 89% DOC rejection. At this condition, less than 12% FA of BP and HS
716 were observed when flushing was applied after the relaxation time. The UF-like recycled
717 pristine membrane performance at the optimized condition was $16.0 \pm 0.5 \text{ L m}^2 \text{ h}^{-1} \text{ bar}^{-1}$ and
718 48% DOC rejection. At this condition, around 40% of FA of BP were observed when
719 flushing was applied after the relaxation time. However, a wide range of contextual factors
720 might condition the feasibility of applying the optimized conditions.

721

722 Based on this present work and complementary literature, a decision-making tree (Figure 4)
723 was developed to select the most appropriate type of the spiral-wound recycled membrane,
724 the appropriate membrane housing device, and the setup system configuration. As shown in
725 Figure 4, the water quality (pH, conductivity and turbidity) determines whether using GDM
726 systems or not and the need for a pre-treatment. For drinking water use, desalination
727 technology is recommended when feed conductivity is higher than $2500 \mu\text{S cm}^{-1}$ [58].
728 NF-like recycled membranes could be housed in the end-caps device in a low-pressure (up to
729 4 bar) cross-flow driven system when water has low salinity [$2500\text{-}4000 \mu\text{S cm}^{-1}$] (See Table
730 A4 in Annex 4 in Supplementary data). Otherwise, for water with lower conductivity, both
731 NF-like and UF-like recycled membranes could be used in a GDM system. The height
732 between the water level of the tank and the end of the permeate tube should be enough to
733 achieve the necessary TMP. Ultra-low pressure can be achieved from 0.4 m [7] up to 3 m
734 [21] (1.6 m was used in the present work).

735

736 According to our study, water with turbidity up to 75 NTU could be directly treated by the
737 GDM system. However, during long-term use, membranes could rapidly clog. Therefore, it is

738 preferable to install a pre-treatment to preserve the membrane lifespan. A relative low-cost
739 simple cartridge device with popular filtering materials (e.g. 100% cotton T-shirt) could be
740 implemented to reduce SS (see Table 6 in Annex 6 in Supplementary data). However, pre-
741 treatment might limit the feed flow to the membrane unit. For conventional cross-flow
742 pressure-driven processes using NF membranes, pre-treatment must be implemented with
743 feed turbidity higher than 0.2 NTU [48].

744

745 The selection of the membrane type will rely on the compromise between the treated water
746 production and the level of purification needed. If LMW-neutrals organic compounds, BB,
747 HS or other substances with similar size are expected to be present in the water (typical river
748 and lake waters with these compounds has around 4 mg L⁻¹ DOC [7]), then NF-like recycled
749 membranes should be selected. For treating bigger compounds such as BP, UF-like recycled
750 membranes are more efficient, since they produce a higher volume of water.

751

752 To decide which system configuration is better suited to the application, membrane users are
753 invited to consider some additional variables, such as tank dimension, location, safety, and
754 (of course) the cost of the system. When the conditions are appropriate to use the submerged
755 configuration, both membrane housing devices could be selected. However, it is more
756 appropriate to use the end-caps device, if it is feasible to afford its cost (around
757 0.9 USD ct. L⁻¹ yr.⁻¹). The end-caps device enables flushing after relaxation time, which is a
758 way to induce shear stress on the membrane surface to mitigate fouling and potentially
759 substantially improve the GDM system performance over its life (see Section 3.2). In this
760 work, no significance differences were observed related to the membrane orientation.
761 However, when operating with standard spiral-wound size (8 inches diameter and 37 m²

762 surface), the easiest setup was found to be the external configuration, placing the membrane
763 horizontally and close to the floor, while vertical orientation is more practical when
764 membranes are submerged. As it was also demonstrated in this work, the intermittent mode
765 of operation will be the optimal filtering mode. However, membrane users will have to pay
766 attention on storing the membrane module wet for long-term preservation [33]. In that sense,
767 the end-caps device could prevent air ingress better than the single fitting device.

768

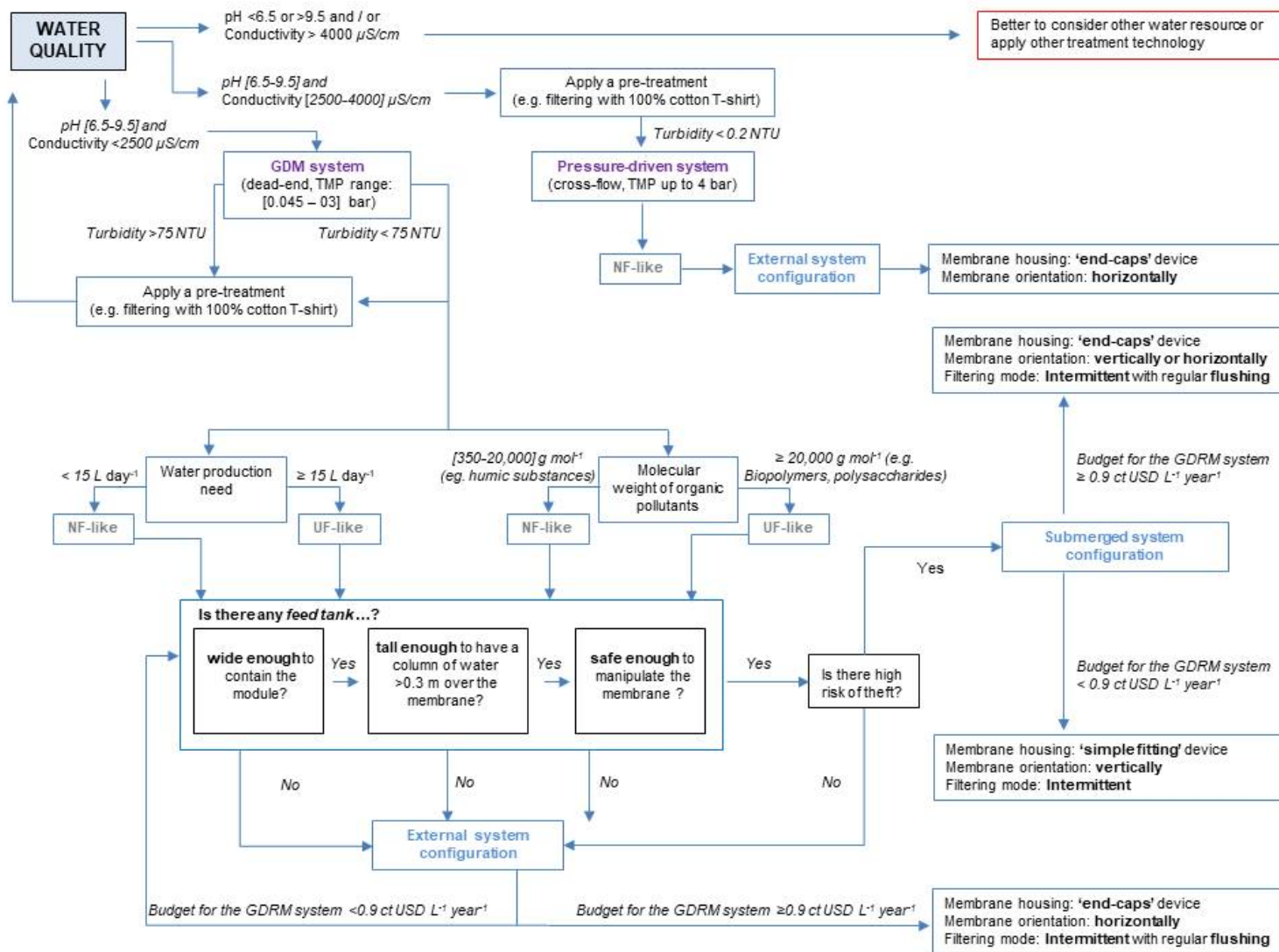


Figure 4. Decision-making tree for the selection of the most appropriate membrane housing device and operating conditions. The starting point is the water quality.

757 **4. Conclusions**

758 Two million EoL RO membranes per year are expected to be discarded from the desalination
759 industry and disposed of in landfills, by 2025. Membrane recycling is a potential alternative
760 solution to prolong the lifespan of the filter, which can be reused in other water treatment
761 sectors.

762

763 This study focused on the development of two alternative membrane housings to the standard
764 pressure vessels that are based on profiting the fiber glass of the spiral-wound recycled
765 membranes with additional devices (simple fitting or end-caps). The new alternatives are
766 suitable to apply a single recycled membrane module for HWTS in rural, decentralized areas
767 and emergency response. Both membrane housings are simple, robust, cost-effective and
768 light-weight, easily transportable and easy to set up and disassemble, which make them
769 adequate for housing these types of membranes in GDM systems. It was demonstrated that
770 the end-caps device was the most versatile solution. It enabled the use of NF-like and UF-like
771 spiral-wound recycled membranes in a GDM system with external configuration (first tested
772 worldwide) and showed other relative advantages (air ingress prevention and flushing)
773 compared to the membrane housing with the simple fitting.

774

775 From the experiments, it can be concluded that recycled membrane-based gravity-driven
776 systems could achieve an acceptable water production for emergency contexts, in terms of
777 quantity (NF-like up to $1.7 \text{ L h}^{-1} \text{ m}^{-2} \text{ bar}^{-1}$ and UF-like up to $37.2 \text{ L h}^{-1} \text{ m}^{-2} \text{ bar}^{-1}$) and quality
778 (up to 89% DOC rejection). The affordable membrane surface cost per area (around 7 UDS
779 m^{-2} , i.e. challenge (ii)) could help addressing the limited flux of GDM systems (i.e. challenge
780 (i)) by using higher membrane surface than usual. Indeed, the high rejection values of:
781 protein (up to 99%), HS (up to 97%), BB (up to 85%) and LMW-neutrals (up to 78%)

782 observed for the NF-like recycled membranes are particularly interesting. This could shed
783 light on increasing the limited rejection capability of small sized organics of standard GDM
784 systems (i.e. challenge (iii)). Additionally, it was demonstrated that the combination of
785 membrane relaxation and flushing of the feed water could be a beneficial and easy practice to
786 be implemented at household level to reduce membrane fouling (i.e. challenge (iv)).

787

788 The economic study showed that compact systems using the novel membrane housing with
789 recycled membranes could produce water with less than 1 USD ct. yr. L⁻¹, being competitive
790 with other commercial HWTS systems. Further scaled-up implementation to create GDM
791 systems using the standard spiral-wound recycled membranes is encouraged, including long-
792 term operation tests to assess bacteria separation and the flux stabilization. Water
793 potabilization and urban waste reclamation are potential application fields.

794

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802

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