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Additional Information

1 **Navigating Environmental, Economic, and Technological**
2 **Trade-Offs in the Design and Operation of Submerged**
3 **Anaerobic Membrane Bioreactors (AnMBRs)**

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13
14 **Abstract**

15 Anaerobic membrane bioreactors (AnMBRs) enable energy recovery from
16 wastewater while simultaneously achieving high levels of treatment. The objective
17 of this study was to elucidate how detailed design and operational decisions of
18 submerged AnMBRs influence the technological, environmental, and economic
19 sustainability of the system across its life cycle. Specific design and operational
20 decisions evaluated included: solids retention time (*SRT*), mixed liquor suspended
21 solids (*MLSS*) concentration, sludge recycling ratio (*r*), flux (*J*), and specific gas
22 demand per membrane area (*SGD*). The possibility of methane recovery (both as
23 biogas and as soluble methane in reactor effluent) and bioenergy production,
24 nutrient recovery, and final destination of the sludge (land application, landfill, or

25 incineration) were also evaluated. The implications of these design and operational
26 decisions were characterized by leveraging a quantitative sustainable design (QSD)
27 framework which integrated steady-state performance modeling across seasonal
28 temperatures (using pilot-scale experimental data and the simulating software
29 DESASS), life cycle cost (LCC) analysis, and life cycle assessment (LCA).
30 Sensitivity and uncertainty analyses were used to characterize the relative
31 importance of individual design decisions, and to navigate trade-offs across
32 environmental, economic, and technological criteria. Based on this analysis, there
33 are design and operational conditions under which submerged AnMBRs could be
34 net energy positive and contribute to the pursuit of carbon negative wastewater
35 treatment.

36

37 **Keywords**

38 Anaerobic MBR; biomethane; global warming potential; life cycle analysis;
39 renewable energy; carbon neutral.

40

41 **1. Introduction**

42

43 Wastewater treatment plants (WWTPs) predominantly utilize aerobic bioprocesses, which rely
44 on the delivery of air (or oxygen) to achieve contaminant degradation to meet effluent standards.

45 This approach has been highly effective at achieving organic carbon removal from municipal
46 wastewaters, but has resulted in resource-intensive treatment that has broad environmental
47 consequences. Wastewater management in the United States, for example, is estimated to

48 represent roughly 3% of U.S. electricity demand (USEPA, 2006). With an estimated 0.3-0.6
49 kWh of electricity consumed per m³ of wastewater treated (Judd and Judd, 2011), this energy
50 demand equates to roughly 0.4-0.8 tonnes of CO₂ emitted per day by a WWTP treating 10 ML·d
51 ¹ (assuming the 2012 Spanish electricity mix). In addition to impeding progress toward carbon
52 neutral (or negative) WWTPs, these high levels of electricity consumption inflate operating costs
53 and incur a diverse set of life cycle environmental impacts stemming from electricity production
54 processes.

55

56 In recent years, there has been increasing interest in the development of mainstream (i.e., main
57 liquid stream) anaerobic treatment processes. In particular, submerged anaerobic membrane
58 bioreactors (AnMBRs) have gained attention for their ability to produce methane-rich biogas
59 during the treatment of urban wastewaters (Giménez et al., 2011; Robles et al., 2012; Raskin,
60 2012; Smith et al., 2013). AnMBRs circumvent several critical barriers to the environmental and
61 economic sustainability of wastewater treatment by eliminating aeration, reducing sludge
62 production, and generating methane (a usable form of energy) from organic contaminants in the
63 wastewater (Shoener et al., 2014). However, given the early stage of development and
64 uncertainties around AnMBR performance, it is unclear how detailed design and operational
65 decisions influence the environmental and economic impacts of AnMBR (Smith et al., 2014).

66

67 Recent studies (e.g., Smith et al., 2012) have identified the need to focus future research efforts
68 on achieving sustainable operation of AnMBRs treating urban wastewater. Although
69 environmental and economic criteria have been used to evaluate submerged AnMBRs relative to
70 alternative aerobic technologies (Smith et al., 2014), a critical barrier to advancing AnMBR

71 development has been the lack of understanding of how detailed design decisions influence
72 system sustainability; a barrier stemming from the lack of a calibrated and validated AnMBR
73 process model to predict system performance under various design and operational scenarios.
74 Ferrer et al. (2008) implemented a computational software called DESASS for designing,
75 simulating, and optimizing both aerobic and anaerobic technologies. The simulation software
76 incorporates a plant-wide model, biological nutrient removal model No. 2 (BNRM2) (Barat et
77 al., 2013), and has been calibrated and validated across a wide range of operating conditions in
78 an industrial-scale AnMBR system (Durán et al., 2013). By leveraging semi industrial-scale data
79 and modeling, Ferrer et al. (2015) and Pretel et al. (*Submitted*) have established an economic
80 basis for the minimum cost design of AnMBRs suitable for implementation in full-scale WWTPs
81 by considering the key parameters affecting membrane performance. However, the
82 environmental impacts of design and operational decisions, as well as the resulting trade-offs
83 across environmental and economic dimensions of sustainability, have not been characterized.

84
85 The aim of this study was to elucidate and navigate sustainability trade-offs in the detailed
86 design of submerged AnMBRs by evaluating the full range of feasible design alternatives using
87 technological, environmental, and economic criteria. To this end, the implications of AnMBR
88 design and operational decisions were characterized using a quantitative sustainable design
89 framework (QSD; Guest et al., 2009) integrating a calibrated and validated process performance
90 model with life cycle assessment (LCA) and life cycle costing (LCC) under uncertainty. By
91 integrating pilot-scale performance data into this QSD framework, our goal was to characterize
92 the relative importance of individual design and operational decisions of submerged AnMBR,
93 while also shedding light on key elements of the system that warrant further research and

94 development. Finally, QSD was used to optimize a submerged AnMBR system to demonstrate
95 how this methodology can be leveraged to navigate sustainability trade-offs in the design and
96 operation of treatment systems, including low energy and energy-producing wastewater
97 technologies.

98

99 **2. Methodology**

100

101 *2.1 Experimental AnMBR Plant*

102

103 This study was carried out using five years of data from an AnMBR system featuring industrial-
104 scale, hollow-fiber (HF) membrane units. The influent to the pilot-scale system is the effluent
105 from the pre-treatment (screening, degritter, and grease removal) of the Carraixet WWTP
106 (Valencia, Spain), with wastewater at ambient temperature (T) from 15 to 30 °C. The AnMBR
107 consists of an anaerobic reactor with a liquid volume of 0.9 m³ (total volume of 1.3 m³)
108 connected to two membrane tanks each with a liquid volume of 0.6 m³ (total volume of 0.8 m³
109 each). Each membrane tank features an ultrafiltration HF membrane commercial system
110 (PURON[®], Koch Membrane Systems, 0.05 μm pore size, 30 m² total filtering area, and outside-
111 in filtration). One 0.5 mm rotofilter screen, one equalization tank (0.3 m³), and one clean-in-
112 place (CIP) tank (0.2 m³) are also included as main elements of the pilot plant. Further details of
113 this AnMBR system can be found in Giménez et al. (2011) and Robles et al. (2012).

114

115 *2.2 Design and Operational Decision-Making*

116

117 Recent work leveraging this pilot-scale system has identified that costs of the system are most
118 sensitive to the following parameters (Ferrer et al., 2015; Pretel et al., *Submitted*): sludge
119 retention time (*SRT*); mixed liquor suspended solids in the membrane tank (*MLSS*); sludge
120 recycling ratio (*r*; the ratio of recycled sludge to forward flow); 20 °C-standardized critical fluxes
121 (*J*); and specific gas demand per membrane area (*SGD*). These parameters influence both the
122 design (i.e., reactor/pump/membrane sizing and construction; Section 3.3.1) and operation
123 (Section 3.3.2) of submerged AnMBR. Based on extensive experimental data from the AnMBR
124 plant and DESASS modeling (Section 2.3), acceptable ranges of these critical parameters were
125 identified to be the following: *SRT* from 13 to 70 days (minimum *SRT* values were set based on
126 treatment efficacy, effluent standard, and sludge stabilization criteria); *r* from 0.5 to 8; *MLSS*
127 entering the membrane tank from 5 to 25 g·L⁻¹; *SGD* from 0.05 to 0.3 m³·m⁻²·h⁻¹; and *J* from 80
128 to 120% of the respective critical flux (*J_C*). To enable more detailed discussion of decision-
129 making, the evaluation of the AnMBR system is divided into its two sub-components: (i) the
130 *biological process*, which includes the anaerobic reactor and its hydraulic connection with the
131 membrane tank, and (ii) the *filtration process*, which includes the membranes and any related
132 maintenance or fouling mitigation.

133

134 Beyond these continuous decision variables, three discrete choices/options were also considered
135 in the design of the AnMBR system: the decision of whether to release or recover methane (both
136 biogas and soluble methane in the effluent) for energy production (via a microturbine); whether
137 or not treated effluent is used for fertigation (i.e., irrigation with nutrient-rich water) to offset
138 fertilizer needs; and the final fate of wasted sludge (land application to achieve fertilizer offsets,
139 incineration, or landfilling). The process flow diagram of the submerged AnMBR is shown in

140 Figure S1 of the Supplementary Data (SD), and the full range of design and operational decisions
141 can be found in Table S1 (also in the SD).

142

143 *2.3 Performance Modeling*

144

145 The simulated AnMBR system was designed to treat an influent flow of $50,000 \text{ m}^3 \cdot \text{d}^{-1}$, with a
146 chemical oxygen demand (COD) of $600 \text{ mg} \cdot \text{L}^{-1}$ and low sulfate content ($10 \text{ mg} \cdot \text{L}^{-1}$). The full
147 characterization of the sewage entering the AnMBR plant can be found in Ferrer et al. (2015).
148 The system was simulated using DESASS (Ferrer et al., 2008) with BNRM2 (Barat et al., 2013).
149 A total of 80 simulations were executed in DESASS and leveraged to characterize system
150 performance across 43,200 scenarios using an Excel-based model that also incorporated an
151 energy consumption tool, enabling the calculation of the overall energy balance (OEB) of the
152 different units at the WWTP. The methodology for the OEB followed the approach of Pretel et
153 al. (2013), which includes procedures for mechanistically calculating mechanical energy demand
154 and energy recovery from biogas.

155

156 *2.4 LCA Implementation*

157

158 Implementation of a LCA framework was conducted in accordance with ISO 14040 (2006) and
159 following industry best practices (Corominas et al., 2013). In order to define the goal and scope,
160 the environmental impacts of the AnMBR system associated with water line operations (i.e.,
161 primary and secondary wastewater treatment as well as final discharge of the treated effluent)
162 and sludge line treatment (i.e., stabilization to comply with discharge standards) were evaluated.
163 A functional unit of one m^3 of treated wastewater was used for the comparison of the different

164 design alternatives (i.e., the combinations of the SRT, MLSS, J, SGD, and r simulated under four
165 temperatures resulting in a total of 43,200 scenarios; Table S1). Figure 1 shows the system
166 boundary used for the LCA and LCC, including the inventory data of the individual materials
167 and processes in this study. As shown in Figure 1, the construction, operation, and demolition
168 phases of the WWTP as well as transportation of the materials, reagents, and sludge were all
169 included, but structural concrete and pipes were excluded from the demolition phase because
170 their useful life was greater than that of the project itself. A maximum useful membrane life of
171 20 years was assumed, with operational fluxes higher than J_C resulting in decreased membrane
172 life (for detailed discussion, see Ferrer et al., 2015). Briefly, membrane life was set from 8 years
173 (when $J = 120\%$ of J_C) to 20 years (when $J = 80\%$ of J_C), according to the maximum total
174 contact with chlorine permissible (500,000 ppm·hours cumulative) and the interval for
175 membrane chemical cleaning. Following the recommendations of Judd and Judd (2011), 9.5
176 months was set as the interval for membrane cleaning with chemicals when operating under
177 critical filtration conditions and with a SGD value of $0.1 \text{ m}^3 \cdot \text{m}^{-2} \cdot \text{h}^{-1}$. Cleaning frequency was
178 adjusted based on the flux (80-120% of the J_C) and SGD by leveraging experimental data
179 extracted in the semi-industrial AnMBR system (e.g., Robles et al., 2012; as described in Ferrer
180 et al., 2015). Pre-treatment processes (e.g., screening, grit removal, and grease removal),
181 rototilter use, equalization tanks, and CIP were not included in this study because their design
182 and operation (and thus, their costs and environmental impacts) were not influenced by the
183 design and operational decisions of the AnMBR process itself. As a result, these supporting
184 processes would not influence the comparative assessment of AnMBR design and operation, and
185 were subsequently placed outside the system boundary. Final effluent was either discharged to
186 natural surface waters or re-used for fertigation. Fugitive CH_4 emissions were accounted when

187 methane was not captured and recovered for energy production. The CML characterization factor
188 of 23 kg CO₂ eq. per kg of CH₄ was used for evaluating the climate implications of fugitive
189 methane. Direct CO₂ releases (i.e., fugitive CO₂ emissions) during sludge dewatering and biogas
190 capture were not quantified because the released CO₂ is classified as biogenic according to IPCC
191 guidelines (Hobson, 2000). Direct emissions to air (e.g., CO, SO₂, NO₂, non-methane volatile
192 organic compounds) resulting from methane combustion through a microturbine-based CHP
193 system were excluded because of a lack of information.

194

195 The life cycle inventories (LCI) of individual materials and processes were compiled using the
196 Ecoinvent Database v.3 accessed via SimaPro 8.01 (PRé Consultants; The Netherlands). The
197 Centre of Environmental Science (CML) 2 baseline 2000 methodology was used to conduct the
198 impact assessment. The impact categories considered in this study were as follows:
199 eutrophication (kg PO₄ eq.), global warming potential with a 100-year time horizon (GWP₁₀₀; kg
200 CO₂ eq.), abiotic depletion (AD, kg Sb eq.), and marine aquatic ecotoxicity (kg 1,4-DB eq.). No
201 grouping, weighting, or aggregation of impact categories was used.

202

203 *2.5 LCC Implementation*

204

205 In order to determine the LCC of the system, all costs were converted to uniform annual cost.
206 Capital costs were annualized assuming a discount rate of 10% and a project lifetime of 20 years.
207 Annual operating and maintenance (O&M) costs were estimated based on energy and reagent
208 consumption, sludge handling and disposal, as well as the replacement of the equipment
209 required. Unit costs and further details about the LCC methodology can be found in Table S2 as
210 well as Ferrer et al. (2015) and Pretel et al. (*Submitted*).

211

212 *2.6 Characterization of the Relative Importance of Design and Operational Decisions*

213

214 In order to elucidate the relative importance of individual design and operational decisions on

215 AnMBR system sustainability, a sensitivity analysis was conducted in two stages (Figure 2):

216 Stage 1 evaluated the full decision space, and Stage 2 focused only on the designs that were

217 likely to be chosen by decision-makers based on economic and environmental criteria (i.e.,

218 design and operational decisions resulting in costs below the 15th percentile; see Figure 2, left-

219 center panel). The uncertainty around absolute values of cost and LCA results, as well as the

220 relative sensitivity of results to key assumptions (including discount rate, membrane cost,

221 electricity cost, concrete cost, energy for stirring, microturbine efficiency, transportation

222 distance, and percent of produced methane dissolved in the effluent), were also evaluated, with

223 details in the SD (Table S3).

224

225 To setup the sensitivity analysis, continuous (*MLSS*, *SRT*, *r*, *SGD*, and *J*) and discrete (fate of

226 methane, fate of effluent, and fate of sludge) decisions were sampled from across the decision

227 space, resulting in a total of 10,800 scenarios – where a *scenario* is a single, unique combination

228 of design and operational decisions – at each of four temperatures (totaling 43,200 total

229 simulations; see Table S1 in the SD for the values sampled from each continuous decision). The

230 costs and GWP₁₀₀ stemming from capital, O&M₁₅ (O&M at 15 °C), and O&M₃₀ (O&M at 30 °C)

231 were then quantified for each scenario. To quantify the effect that individual decisions had on

232 environmental and economic criteria, the results were segregated across the decision space for

233 each individual parameter. For Stage 1 of the sensitivity analysis, the median, 5th, 25th, 75th, and

234 95th percentiles were then calculated for a given parameter value or discrete decision, as was the

235 global median (i.e., the median of all the results). The range between the maximum and
236 minimum value for each percentile was then normalized to the global median in order to quantify
237 how much the range and absolute value of output metrics change across the full decision space
238 for each individual parameter (see the top panel of Figure 2 for a visual representation of this
239 methodology).

240

241 Recognizing that design and operational decisions resulting in costs below the 15th percentile are
242 most likely to be chosen (so long as they meet treatment objectives) by WWTP designers and
243 decision-makers, these scenarios were the focus of Stage 2 of the sensitivity analysis. Once this
244 subset of scenarios was identified (consisting of the “practical” scenarios most likely to be
245 chosen for implementation), the *practical* average and standard deviation of cost and GWP₁₀₀
246 across all continuous decisions were determined. Next, the *local* average and standard deviation
247 were calculated for each simulated value across the range of an individual design or operational
248 decision (e.g., *MLSS* = 5, 10, 15, 20, and 25 kg·m⁻³). For a given decision, the greatest difference
249 between a *local* and the *practical* average was then used to calculate the maximum percent shift
250 from the practical average stemming from that decision (this calculation of the maximum percent
251 shift was repeated for the *practical* standard deviation using *local* standard deviations; bottom-
252 left graph in Figure 2). The relative importance of each continuous decision variable on a given
253 metric (costs and GWP₁₀₀ stemming from capital and average O&M) was determined by taking
254 the sum of the percent change in average and percent change in standard deviation and ranking
255 those sums in descending order (bottom-right graph in Figure 2), similar to the ranking process
256 of Morris’ one-at-a-time method (Saltelli *et al.*, 2004). As a final step in the Stage 2 sensitivity

257 analysis, Monte Carlo simulation was conducted with 10,000 trials to examine the change in rank
258 of the five continuous decision variables in order to characterize the robustness of these rankings.

259

260 **3. Results and Discussion**

261

262 Four main sections have been established in order to elucidate and navigate sustainability trade-
263 offs stemming from detailed decision-making for submerged AnMBR: the relative importance of
264 individual design and operational decisions (Section 3.1), navigating trade-offs across
265 dimensions of sustainability (Section 3.2), optimization of the AnMBR process (Section 3.3),
266 and uncovering how and why individual design/operational decisions impact AnMBR
267 sustainability (Section 3.4). Taken altogether, these sections demonstrate how QSD can be used
268 to optimize wastewater treatment technologies, including those targeting energy and broader
269 resource recovery from wastewater. Results and discussion presented here are centered on
270 linking design decisions to costs and life cycle environmental impacts, with a focus on global
271 warming potential with a 100 year time horizon (GWP_{100}) as a representative example of broader
272 environmental impacts. It should be noted, however, that most environmental impact categories
273 followed similar trends as those of GWP_{100} .

274

275 *3.1 Relative Importance of Design and Operational Decisions to AnMBR Sustainability*

276

277 Figure 3 shows the effect of the continuous ($MLSS$, SRT , r , J , and SGD) and discrete (methane
278 fate, effluent fate, and sludge fate) decisions on costs and environmental impacts across capital,
279 $O\&M_{15}$, and $O\&M_{30}$. Considering continuous variables, all five influenced costs to a similar
280 degree, although $MLSS$ and J were most responsible for the variance in the LCC results

281 stemming from capital and O&M costs, respectively (Figure 3A). The variables r , $MLSS$, and
282 SGD were the most significant contributors to the variance in LCA results, mostly due to O&M
283 (Figure 3C). For almost all parameters, the largest variance in economic and environmental
284 performance was observed at the 95th percentile and the lowest variance at the 5th percentile.
285 Discrete variables had similar cost implications as the design and operational parameters
286 (Figures 3A and 3B), but disproportionately high GWP₁₀₀ consequences (one to two orders of
287 magnitude higher; see y-axis scales in Figures 3C and 3D). This observation stemmed from the
288 climate implications of fugitive methane (23 kg of CO₂ eq. per kg of fugitive CH₄), energy
289 offsets (0.13 kg of CO₂ per kWh produced), and fertilizer offsets (2.68 kg of CO₂ equivalents per
290 kg of N). In comparison to the baseline set of discrete decisions (recovery of biogas and soluble
291 methane for electricity production, effluent reuse, and land application of biosolids), allowing
292 fugitive methane emissions and managing sludge through incineration were the least preferable
293 options in terms of cost (Figure 3B). Regarding LCA results, eliminating energy recovery from
294 methane and final disposal of the sludge into landfill were the least preferable options (Figure
295 3D).

296

297 In order to provide insight into the role of individual design and operational decisions on the
298 relative sustainability of practical designs (i.e., the final set of designs likely to be considered by
299 decision-makers), Stage 2 of the sensitivity analysis focused on the scenarios below the 15th
300 percentile for costs (as shown in Figure 2). The relative importance of the five continuous
301 decision variables was evaluated across four categories: influence on costs and GWP₁₀₀
302 stemming from capital and average O&M (i.e., average of O&M at 15 and 30°C; Figure 4). The
303 results of the Monte Carlo simulation (Figure 4 and Table S4) show that $MLSS$ consistently (71-

304 100%) had the largest impact on capital costs and both LCA categories, and was ranked second
305 for its impact on LCC O&M across all simulations. *SRT* only had a high impact on LCA Capital
306 (ranked second), which is a result of its effect on tank volume, which in turn determines
307 construction material requirements. *r* was most often ranked second for LCC Capital, which was
308 due mainly to its effect on tank volume when building the plant. *SGD* consistently impacted
309 LCA O&M (ranked second) because of electricity demand from blower operation. *J* was ranked
310 first for LCC O&M (across all simulations) because of its effect on membrane operation and
311 replacement cost. Thus, the factors driving environmental impacts were tankage and electricity
312 for gas sparging, while costs were driven by tankage and membranes. In comparison to Figure 3
313 and the analysis of the full decision space, the results presented in Figure 4 provide much more
314 meaningful insight for decision-makers by focusing on the scenarios most likely to be chosen.
315 This analysis eliminates observations that are irrelevant (e.g., stemming from scenarios that
316 would never be chosen), and also allows decision-makers to prioritize individual design and
317 operational decisions as part of a participatory planning process incorporating locality-specific
318 factors (Guest et al., 2010).

319

320 *3.2 Navigating Trade-Offs Across Dimensions of Sustainability*

321

322 In order to develop a final set of parameters, it becomes necessary to characterize the interactions
323 among design and operational decisions. To this end, we evaluated relationships among decision
324 variables to identify trade-offs and synergies, where *trade-offs* exist when adjusting a decision
325 variable produces tension between sustainability metrics (i.e., to get better in one, you must get
326 worse in the other), and *synergies* occur when changing a given decision variable moves

327 sustainability metrics in the same direction (either both become more desirable, or both become
328 less desirable).

329

330 When synergies exist between LCC and LCA results, it can be expected that designers would
331 seek to simultaneously improve both costs and environmental impacts by adjusting the decision
332 variable. If the LCC and LCA results follow opposing trends, trade-offs can be considered by
333 comparing the ratio of additional costs (€) to the tonnes of CO₂ equivalents that are saved (i.e.,
334 not released to the environment). This approach to quantifying the tension between sustainability
335 metrics enables the comparison of a given decision to an external benchmark – the carbon
336 emissions trading system – which enables the purchase of carbon offsets (€·t CO₂⁻¹). In general,
337 emissions trading seeks to reduce pollution by providing economic incentives for companies to
338 limit their emissions (Stavins, 2003). The largest international framework for greenhouse gas
339 emissions is the European Union Emission Trading Scheme, which currently spans power plants
340 and industrial plants across 31 countries (EU, 2013). By using market prices for carbon offsets as
341 a benchmark (e.g., in Spain the emissions trading system is currently around 6 €·tonne CO₂
342 saved⁻¹ (REE, 2012)), the rationality of having a WWTP incur additional costs to reduce carbon
343 emissions can be evaluated.

344

345 Figure 5 shows the effect of *MLSS* (Figure 5A), *J* (Figure 5B), *SRT* (Figure 5C), and *SGD*
346 (Figure 5D) in order to illustrate the potential for trade-offs and synergies between costs and
347 environmental impacts. Although simulations were performed across the full range for all
348 continuous decision variables, four illustrative examples (the min-max combinations of two other
349 decision variables) are plotted in each figure. In Figure 5A, *MLSS* was varied from 5-25 g·L⁻¹ for

350 four possible design/operational scenarios at the min-max of J (80 and 120% of J_C) and r (0.5-8).
351 For these example scenarios, costs and GWP_{100} were synergistic below $MLSS$ values of $15 \text{ g}\cdot\text{L}^{-1}$.
352 In Figure 5B, flux was varied from 80-120% of J_C for four possible design/operational scenarios
353 at the min-max $MLSS$ (5 and $25 \text{ g}\cdot\text{L}^{-1}$) and SGD (0.05 and $0.30 \text{ m}^3\cdot\text{m}^{-2}\cdot\text{h}^{-1}$). At a flux below 97%
354 and above 112%, synergy occurs between the LCA and LCC results, which indicates that both
355 impacts can be lessened by increasing or decreasing the flux in the direction of the synergy
356 arrows shown in Figure 5B. However, between 97-112% of J_C , tension exists between economic
357 and environmental impacts, thus requiring the navigation of trade-offs. In Figure 5C, SRT was
358 varied from 13 to 70 days across combinations of $MLSS$ (5 and $25 \text{ g}\cdot\text{L}^{-1}$) and r (0.5-8) (when
359 methane is not recovered), and was shown to be synergistic at all values examined, which
360 indicates that LCC and GWP can be lessened by minimizing SRT across the entire decision
361 space. In contrast, Figure 5D demonstrates that SGD often results in trade-offs across the full
362 range of values considered (0.05 to $0.30 \text{ m}^3\cdot\text{m}^{-2}\cdot\text{h}^{-1}$), shown with combinations of $MLSS$ (5 and
363 $25 \text{ g}\cdot\text{L}^{-1}$) and J (80 and 120% of J_C).

364

365 As one proposed approach to identify an optimal design, Figure 6 benchmarks the ratio of
366 $\text{€}\cdot\text{tonne}\cdot\text{CO}_2 \text{ saved}^{-1}$ for the WWTP against the Spanish emissions trading system across the
367 feasible range of J values (for this analysis, $SGD = 0.30 \text{ m}^3\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ and $MLSS = 25 \text{ g}\cdot\text{L}^{-1}$).
368 Across the bulk of the design space where trade-offs exist, the cost of mitigating carbon
369 emissions at the WWTP was drastically higher than the market-based benchmark, with costs of
370 up to $30,000 \text{ €}\cdot\text{tonne}\cdot\text{CO}_2 \text{ saved}^{-1}$ at the treatment plant. In this particular case, therefore,
371 treatment plants seeking to lower their carbon footprint beyond leveraging synergies with cost
372 may achieve a more meaningful environmental benefit at much less cost if they were to purchase

373 credits on the trading market (if such an action is possible). In the future, however, this QSD
374 framework may provide additional support for the creation of carbon crediting systems for the
375 wastewater sector (proposed by Wang et al., 2011 in the context of reducing nitrogenous
376 greenhouse gas emissions); such a transition could enable utilities to take a more proactive
377 posture and secure additional financial resources for the installation of low-energy and energy
378 positive treatment technologies.

379

380 *3.3 Optimization of Submerged AnMBR*

381

382 We propose that the optimization of AnMBR design should minimize costs subject to effluent
383 water quality constraints, and only consider further reducing greenhouse gas emissions when (i)
384 there are no readily available, less expensive alternatives for GHG reduction, and (ii) it is part of
385 a transparent, inclusive planning and design process that addresses locality-specific factors in
386 decision-making (Guest et al, 2009, 2010). For the submerged AnMBR system evaluated here,
387 costs and GHG emissions were largely in synergy (reducing one reduced the other), and design
388 conditions that resulted in trade-offs between costs and GWP₁₀₀ had incurred costs for CO₂
389 mitigation that far exceeded the European Union Emission Trading Scheme (except for a very
390 narrow band in which carbon could be offset at an expense to the utility of < 6 €·tonne-CO₂
391 saved⁻¹). Thus, the optimization of the submerged AnMBR system (detailed below) focused on
392 cost minimization, with all potential designs subject to year-round treatment requirements with
393 treatment efficacy confirmed through DESASS modeling under summer and winter conditions.
394 It should be noted that this methodology leveraged pilot-scale experimental data for the design

395 and simulation of a full-scale treatment process, and that additional scale-up challenges –
396 although outside the scope of this study – may influence system sustainability.

397

398 *3.3.1 Optimizing the Construction of the Submerged AnMBR System*

399

400 Capital costs represented a meaningful fraction of life cycle costs across the full range of
401 AnMBR design alternatives (with typical values of $45 \pm 8\%$; average \pm standard deviation),
402 whereas life cycle environmental impacts were largely dominated by the O&M phase (e.g., $74 \pm$
403 14% for GWP_{100} when total methane is recovered, 99% if methane is released as fugitive
404 emissions). Following the approach to optimization outlined immediately above (Section 3.3),
405 the anaerobic reactor and membrane area were sized by selecting the configuration (based on
406 10,800 evaluated combinations of MLSS, SRT, r , J , and SGD) that resulted in the minimum LCC
407 while enabling the plant to meet treatment requirements across all simulated temperatures (from
408 $15\text{ }^{\circ}\text{C}$ to $30\text{ }^{\circ}\text{C}$). In this respect, winter conditions ($15\text{ }^{\circ}\text{C}$) governed the sizing of the constructed
409 system. J was set slightly above the critical flux (105% of J_C , based on the least favorable SGD
410 and MLSS values), r was set to 3 and the anaerobic reactor volume was set in $35,190\text{ m}^3$. By
411 selecting the minimum cost values for these parameters as opposed to the minimum or maximum
412 ($17,800\text{ m}^3$ or $373,440\text{ m}^3$ for volume, 80% or 120% for J , and 0.5 or 8 for r , respectively), the
413 overall LCC reduced by $35/70\%$ (minimum/maximum) for volume, $17/47\%$ for J , and $22/4\%$ for
414 r . When considering the LCA, there was no obvious benefit to selecting the optimum values for
415 construction-phase elements because their impact on the life cycle environmental impacts was
416 minimal.

417

418 3.3.2 Optimizing the Operating Submerged AnMBR

419
420 In the O&M phase, an operational volume (calculated from r and required to be below the
421 constructed volume), an operational membrane area (calculated from the operating J for each
422 SGD and $MLSS$ value at a flux of 105% J_C , and required to be smaller than constructed area), and
423 an operating r value (at or below the constructed r capacity) have been considered for the full
424 range of feasible design alternatives in order to assess the overall LCC and LCA results for the
425 AnMBR system. Further details on the interactions among the detailed design calculations with
426 decision variables can be found in Ferrer et al. (2015). Based on economic and environmental
427 criteria, the optimum operating parameters of the AnMBR design ($MLSS$, r , SRT , SGD , and J)
428 were determined at different temperatures (see Table 1). Details of the mechanisms governing
429 the selection of individual parameters is discussed in more detail in Section 3.4.

430
431 The uncertainty analysis was conducted on LCC and LCA results at 15 °C (taking the scenario
432 with the optimum operating parameters from Table 1), and an additional sensitivity analysis was
433 performed to better understand the influence of individual assumptions. Based on the LCC
434 considering fugitive methane emissions, the input parameters affecting the output were (in
435 descending order): membrane cost, discount rate, energy for stirring and electricity cost. When
436 methane was recovered, the microturbine efficiency became more important than the stirring
437 energy and the electricity cost. Based on the LCA, when methane was not recovered, the only
438 input parameter affecting the output was the percentage of dissolved methane emitted to air
439 (where the balance of dissolved methane is assumed to be degraded to CO_2). When total methane
440 recovery was considered, the efficiency of the microturbine became the most important
441 (approximately 50%), followed by transportation distance (35%), and stirring energy (15%). The

442 results showed that although there was uncertainty surrounding model outputs (Figure S2 in the
443 SD), alternative values for these assumed parameters would not have changed the observed
444 trends and narrative surrounding the sustainable design of submerged AnMBR.

445

446 *3.4 Connecting Design and Operational Decisions to Sustainability Metrics*

447

448 *3.4.1 The Impact of SGD and MLSS on Membrane Filtration*

449

450 In order to better understand the mechanisms governing the impact of *SGD* and *MLSS* on LCA
451 and LCC results, these values were varied across the decision space at a temperature of 15 °C.

452 Other parameters corresponding to biological processes (i.e., *r* and *SRT*) were fixed at 3 and 41

453 days, respectively. Total methane recovery, nutrient recovery from effluent, and agricultural

454 application of sludge were considered for the discrete decisions. Based on the LCC results, gas

455 sparging was the most significant process at high *MLSS* and *SGD*, contributing nearly 62% of the

456 total operating cost. However, reagent consumption had an increased impact when operating at

457 high *MLSS* and low *SGD* values – representing up to 41% of the total operating cost – due to the

458 increased membrane cleaning requirement.

459

460 When *MLSS* was held constant, *SGD* had a positive correlation with filtration costs, increasing

461 the filtration operating cost by up to 0.063 €·m⁻³ (representing a 19% increase), but had no effect

462 on biological costs. Similarly at a given *SGD* value, increasing *MLSS* increased filtration costs,

463 but it also decreased costs associated with biological processes. Based on this, the optimum

464 parameters for this study were the optimum value for *SGD* (0.10 m³·m⁻²·h⁻¹) and *MLSS* = 10-15

465 g·L⁻¹ (Table 1). This value was chosen for *MLSS* because at larger values, the increase in

466 filtration costs was not offset by a decrease in biological costs. Similarly, lower *MLSS* values
467 increased biological costs much more than the filtration costs decreased (up to 85% of the total
468 operating costs).

469

470 Methane recovery was not affected by changes in *MLSS* and *SGD*. Based on the LCA results,
471 reagent consumption did not have a significant environmental impact ($\text{GWP}_{100} = 0.003 \text{ kg}$
472 $\text{CO}_2 \cdot \text{m}^{-3}$ and marine ecotoxicity = $0.428 \text{ kg } 1,4\text{-DB} \cdot \text{m}^{-3}$). Gas sparging presented the greatest
473 environmental impact based on GWP at high *MLSS* and *SGD*, increasing GWP to 0.051 kg
474 $\text{CO}_2 \cdot \text{m}^{-3}$ and marine ecotoxicity to $21.479 \text{ kg } 1,4\text{-DB} \cdot \text{m}^{-3}$. Biological processes had a beneficial
475 impact on reducing GWP_{100} because of the decreased emissions from methane and by enabling
476 nutrient recovery, achieving values as low as $-0.039 \text{ kg } \text{CO}_2 \cdot \text{m}^{-3}$ for GWP_{100} and $-19.0 \text{ kg } 1,4\text{-}$
477 $\text{DB} \cdot \text{m}^{-3}$ for marine ecotoxicity.

478

479 *3.4.2 The Impact of r and *MLSS* on the Bioprocess*

480

481 To better understand the underlying relationships among r , *MLSS*, and LCA and LCC outputs,
482 these values were varied across the decision space while *SGD* and J were fixed at their optimum
483 values ($0.10 \text{ m}^3 \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ and 105% of J_C , respectively). Based on the LCC results, mixer
484 operation was the most significant cost – comprising up to 80% of the total operating cost of 0.11
485 $\text{€} \cdot \text{m}^{-3}$ – and was highest at low *MLSS* and r values. The sludge recycling pump accounted for a
486 small fraction of the operating cost (approximately 8%).

487

488 At a given *MLSS* value, decreasing r increased the cost of biological processes; at lower *MLSS*
489 values, this increase was even more pronounced, raising the biological operating cost up to 0.091

490 $\text{€}\cdot\text{m}^{-3}$ (representing a 48% increase). Conversely, the filtration process costs was not affected by
491 r . When r was fixed, $MLSS$ had a similar trend in terms of biological process cost, but filtration
492 costs also decreased. Therefore, the lowest total cost occurred at $r = 2-3$ and $MLSS = 10-15 \text{ g}\cdot\text{L}^{-1}$
493 (Table 1).

494

495 Changes in r and $MLSS$ had no effect on methane recovery. Based on the LCA results, the sludge
496 recycling pump contributed very little to overall environmental impact (i.e., increases in GWP_{100}
497 by $0.002 \text{ kg CO}_2\cdot\text{m}^{-3}$ and marine ecotoxicity by $1.14 \text{ kg 1,4-DB}\cdot\text{m}^{-3}$). Mixer operation had a
498 much greater impact overall. At low $MLSS$ and r values, GWP_{100} increased by $0.077 \text{ kg CO}_2\cdot\text{m}^{-3}$
499 and marine ecotoxicity increased by $40 \text{ kg 1,4-DB}\cdot\text{m}^{-3}$. When considering methane and nutrient
500 recovery, however, GWP_{100} decreased to $-0.039 \text{ kg CO}_2\cdot\text{m}^{-3}$ and marine ecotoxicity to -17.9 kg
501 $1,4\text{-DB}\cdot\text{m}^{-3}$.

502

503 *3.4.3 The Impact of SRT and T on the Bioprocess*

504

505 For the LCA, the effects of sludge disposal (agriculture), methane production, and effluent
506 discharge were also evaluated by varying SRT across temperatures (T). At high SRT and T ,
507 biogas production and nutrient solubility were large. Sludge disposal, stirring, and sludge recycle
508 pumping all contributed significantly to marine ecotoxicity (up to $13.1 \text{ kg 1,4-DB}\cdot\text{m}^{-3}$ for sludge
509 disposal, which corresponded with the lowest value of SRT and up to $10.6 \text{ kg 1,4-DB}\cdot\text{m}^{-3}$ for the
510 latter two, which corresponded with the highest value of SRT). When neither nutrients nor
511 methane are recovered, emitted methane represented almost 100% of the GWP (increasing it up
512 to $1.61 \text{ kg CO}_2\cdot\text{m}^{-3}$) and discharged nutrients increased eutrophication up to $0.042 \text{ kg PO}_4^{-3}\cdot\text{m}^{-3}$.
513 However, if nutrients, biogas, and soluble methane are all recovered, this system achieved

514 carbon offsets through resource recovery (up to $-0.059 \text{ kg CO}_2 \cdot \text{m}^{-3}$ for methane recovery and up
515 to $-0.067 \text{ kg CO}_2 \cdot \text{m}^{-3}$ for nutrient recovery) as well as reductions in marine ecotoxicity (up to -
516 $18.6 \text{ kg 1,4-DB} \cdot \text{m}^{-3}$ for methane recovery and up to $-37.3 \text{ kg 1,4-DB} \cdot \text{m}^{-3}$ for nutrient recovery).
517 In terms of eutrophication, a reduction of around 50% can be achieved as a result of recovering
518 nutrients in the effluent.

519

520 *3.4.4 Energy, Nutrient, and Residuals Management*

521

522 Regarding methane recovery, three options were considered: no recovery, only recovering
523 biogas, or total methane recovery (recovery of both biogas and methane dissolved in the
524 effluent). The LCC results show that cost savings of up to 16 and 36% (at 15°C and 30°C ,
525 respectively) are possible. By accounting for the energy offsets through on-site production,
526 greenhouse gas savings up to 76-104% (at 15°C and 30°C , respectively) can be achieved. These
527 calculations were made assuming both biogas and dissolved methane in the effluent stream were
528 recovered and utilized for energy generation. The total cost of the technologies needed for these
529 processes (degassing membrane for dissolved methane and microturbine-based CHP for energy
530 generation) were also considered. Based on this analysis, there may exist submerged AnMBR
531 design/operational scenarios that have the potential to generate energy in excess of what is
532 required to run the AnMBR system, making them net energy positive.

533

534 The framework in this study examined whether or not treated effluent is used for fertigation (i.e.,
535 irrigation with nutrient-rich water) to offset fertilizer needs. Note that calculations of fertilizer
536 offsets from fertigation included assumptions of nitrogen and phosphorus bioavailability (50%
537 and 70%, respectively), consistent with other studies (Gallego et al., 2008; Rodriguez-Garcia et

538 al., 2011; Garrido-Baserba et al., 2013; Pretel et al., 2013). Based on the LCA data, nutrient
539 recovery reduced eutrophication by approximately 50% and significantly reduced marine toxicity
540 (around $-37 \text{ kg } 1,4\text{-DB}\cdot\text{m}^{-3}$), GWP ($-0.07 \text{ kg CO}_2\cdot\text{m}^{-3}$) and AD (-0.0005 kg Sb eq) due to the
541 fertilizer avoided. For sludge disposal, three options were considered in this study: agricultural
542 application, incineration, or landfilling. Based on the LCC results, there were savings of 50% or
543 90% using agricultural application over landfilling or incineration, respectively. Based on the
544 LCA results, incineration could be a better option over agriculture in terms of GWP_{100} and
545 eutrophication, because while agricultural application offsets fertilizer use, it still results in direct
546 emissions to air (e.g., N_2O , NH_3), water (e.g., PO_4), and soil (heavy metals). Although the
547 approach used to estimate emissions from land application and fertilizer offsets were consistent
548 with other studies (Gallego et al., 2008; Rodriguez-Garcia et al., 2011; Garrido-Baserba et al.,
549 2013; Pretel et al., 2013), this approach does not account for direct fugitive emissions to air and
550 water that stem from synthetic fertilizers. The negative consequences of land application in terms
551 of GWP_{100} and eutrophication, therefore, would be reduced if direct emissions from synthetic
552 fertilizers were included in the system boundary, since a portion of these emissions would be
553 offset. Beyond GHG and nutrient emissions, agriculture also had the fewest negative impacts in
554 AD and marine toxicity.

555

556 **4. The Role of AnMBR in Carbon Neutral Wastewater Treatment**

557

558 The main challenge of AnMBR technology is optimizing design and operation of the process in
559 order to improve the sustainability of the technology to treat wastewaters. The AnMBR system
560 may be suitable to treat most municipal wastewater streams, since it can achieve high quality

561 effluent (Smith et al., 2012; Lin et al., 2013) while also achieving meaningful steps toward
562 sustainable wastewater treatment: lower inherent energy demand stemming from no aeration and
563 energy recovery through methane production. Although conventional activated sludge treatment
564 plants consume roughly 0.2-0.6 kWh·m⁻³ (McCarty et al., 2011; Judd and Judd, 2011; Fenu et
565 al., 2010), a sub-set of design scenarios here achieved on-site energy production in excess of
566 estimated on-site energy demands. However, consistent with findings from other energy
567 assessments of AnMBRs (Smith et al., 2014; Shoener et al., 2014; Martin et al., 2011), sparging
568 remains a critical challenge as it accounts for the majority of AnMBR energy demand (with
569 typical values of 52 ± 21%; average ± standard deviation), in this study). Fouling mitigation
570 (during operation) and membrane capital costs – as well as anaerobic reactor construction and
571 mixing – remain the dominant sources of costs, which are critical challenges to enable AnMBR
572 to overtake activated sludge in practice (Lin et al., 2011; Ferrer et al., 2015). Additionally,
573 maximizing the capture of methane is another key component of AnMBR technology for
574 achieving energy savings and reducing the overall WWTP carbon footprint in a way that is
575 financially viable. Particularly in this study, greenhouse gas savings up to 76-104% (at ambient
576 temperature of 15 °C and 30 °C, respectively) were achieved by accounting for energy offsets
577 through on-site production when methane (from both biogas and effluent streams) is captured
578 and utilized for energy generation.

579

580 As we pursue improved designs of submerged AnMBR systems, the greatest opportunities for
581 simultaneously improving economic and environmental performance will be through reduced
582 energy consumption. Based on the QSD results presented here, it is also worth highlighting the
583 importance of (i) reducing energy-intensive sparging, (ii) increasing flux to decrease required

584 membrane area, and (iii) developing efficient dissolved methane recovery processes in order to
585 maximize energy recovery and avoid direct greenhouse gas emissions. In any case, these pursuits
586 to reduce life cycle environmental impacts should not jeopardize effluent quality – the primary
587 responsibility of WWTPs. The high quality effluent provided by AnMBRs is one of the
588 technology’s greatest strengths. The membranes help ensure robust treatment and can enable safe
589 nutrient recovery through fertigation, the latter of which can have significant economic and
590 environmental benefits through fertilizer and freshwater offsets.

591

592 **5. Conclusions**

593

594 A quantitative sustainable design process has been leveraged to develop a detailed design of
595 submerged AnMBR by evaluating the full range of feasible design alternatives using
596 technological, environmental, and economic criteria. Results showed that J , SGD , $MLSS$, and r
597 required the navigation of sustainability trade-offs, but minimizing SRT simultaneously
598 improved environmental/economic performance. Moreover, $MLSS$ and J had the strongest
599 influence over LCA results and capital costs, with J governing O&M costs. Based on this
600 analysis, there are design and operational conditions under which submerged AnMBRs could be
601 net energy positive at higher operating temperatures and contribute to the pursuit of carbon
602 negative wastewater treatment. More broadly, this work demonstrates the use of QSD, which can
603 be leveraged to quantify and navigate sustainability trade-offs in the optimization of wastewater
604 treatment and resource recovery systems.

605

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607

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615

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720 **Tables**

721

722 **Table 1.** Optimum operating parameters at different ambient temperatures of the AnMBR design
 723 and total cost.

Optimum Operational Parameters							Scenarios for Sludge Disposal			Scenarios for Methane Recovery	
T, °C	MLSS, g·L ⁻¹	SRT, days	r	SGD, m ³ ·m ⁻² ·h ⁻¹	J, % of J _c (in LMH)	Total cost [†] , €·m ⁻³	Land application [‡] , €·m ⁻³	Incineration [‡] , €·m ⁻³	Landfilling [‡] , €·m ⁻³	Biogas recovery [*] , €·m ⁻³	Total CH ₄ recovery [*] , €·m ⁻³
15	15	41	3	0.10	105 (16)	0.130	0.001	0.049	0.006	- 0.021	- 0.005
20	15	28	2.5	0.10	105 (16)	0.125	0.001	0.049	0.006	- 0.022	- 0.004
25	10	19	2.5	0.10	105 (23)	0.094	0.001	0.049	0.006	- 0.024	- 0.004
30	10	13	2	0.10	105 (23)	0.079	0.001	0.050	0.006	- 0.026	- 0.002

724

725 [†] Cost of the AnMBR system, excluding sludge disposal and methane recovery.

726 [‡] Cost of sludge management and disposal assuming 100% of sludge is managed with a single method.

727 ^{*} Cost of 100% biogas or total methane (biogas and soluble methane) recovery (capital and operating cost of the
 728 technology are included). Negative values represent net profit.

729 **Figure captions**

730

731 **Figure 1.** System boundary for the LCA and LCC of the submerged AnMBR.

732 **Figure 2.** Sensitivity analysis methodology used to characterize the relative importance of individual design
733 decisions on (Stage 1) the full range of possible designs or (Stage 2) the range of practical designs (where *practical*
734 designs are those combinations of design and operational parameters that resulted in the lowest 15th percentile of
735 cost or environmental impact, believed to be the most likely to be chosen by decision-makers for implementation).
736 Results of this sensitivity analysis can be found in Figures 3 and 4.

737 **Figure 3.** Effect of the continuous (*MLSS*, *SRT*, *r*, *J*, and *SGD*) and discrete (methane recovery, nutrient recovery,
738 and sludge disposal) decisions on the outputs (LCC and LCA) stemming from capital, O&M₁₅, and O&M₃₀ and
739 considering 5th, 25th, 75th, and 95th percentiles. The range between the maximum and minimum for each percentile
740 was normalized by the global median. Discrete selections are listed as †methane fate (*fugitive* emission, *biogas*
741 recovery for electricity production, total *methane* recovery – including biogas and soluble methane – for electricity
742 production), ‡effluent fate (*reuse* for fertigation, direct *discharge*), and *residuals fate (*land* application, *landfill*,
743 *incineration*).

744 **Figure 4.** Radar plot showing the average relative importance of five continuous decisions (*MLSS*, *SRT*, *r*, *J*, *SGD*)
745 on four outputs (LCC Capital and O&M, LCA Capital and O&M). The influence of each decision on LCC and LCA
746 outputs was ranked from 1–5 – with one having the highest impact on each result – across 10,000 trials. The size of
747 the green area represents the magnitude of the decision’s impact. Average ranks and standard deviations (from the
748 10,000 trials) can be found in Table S4 in the SD.

749 **Figure 5.** LCC (€·m⁻³) and GWP (kg-CO₂ eq·m⁻³) results of a subset of AnMBR scenarios showing trade-offs and
750 synergies between economic and environmental criteria across the decision space for: (A) *MLSS* (g·L⁻¹); (B) *J* (%
751 *J_c*); (C) *SRT* (days); and (D) *SGD* (m³·m⁻²·h⁻¹).

752 **Figure 6.** Evaluation of the ratio of €·tonne-CO₂ saved⁻¹ in the selection *J* values (when *SGD* = 0.30 m³·m⁻²·h⁻¹ and
753 *MLSS* = 25 g·L⁻¹) and comparison with the Spanish emissions trading system (6 €·t CO₂⁻¹) as a benchmark.