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1           **Enhanced organic degradation and biogas production of**  
2           **domestic wastewater at psychrophilic temperature through**  
3           **submerged granular anaerobic membrane bioreactor for**  
4           **energy-positive treatment**

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10           **ABSTRACT**

11           This study deals with the conversion of organic matter into methane at ambient  
12           temperature, during anaerobic digestion of domestic wastewater combined with a submerged  
13           ultrafiltration membrane with no gas-sparging. A one-stage submerged granular anaerobic  
14           membrane bioreactor (G-AnMBR) and a control anaerobic digester (UASB type) were  
15           operated during four months, after 500 days of biomass acclimatization to psychrophilic and  
16           low loading rate conditions. Membrane barrier led to the retention of biomass, suspended  
17           solids and dissolved and colloidal organic matter which greatly enhanced total COD (tCOD)  
18           removal (92.3%) and COD to methane conversion (84.7% of tCOD converted into dissolved  
19           and gaseous CH<sub>4</sub>). G-AnMBR overcame the usual long start-up period and led to a higher  
20           sludge heterogeneity, without altering the granular biomass activity. The feasibility of the G-  
21           AnMBR without gas-sparging was also assessed and the net positive energy balance was  
22           estimated around +0.58 kWh.m<sup>-3</sup>.

23           **KEYWORDS**

24           ultrafiltration; anaerobic digestion; granular biomass; low-strength wastewater; energy  
25 recovery.

26           **1. Introduction**

27           For the last decade, large attention has been given to establish a balance between human  
28 well-being and environment preservation. In this way, intensive resource use is switching into  
29 a circular economy approach, based on the reuse and recycling of resources. For those  
30 reasons, wastewater is not only considered as an alternative source of water but also as a  
31 source of nutrients, minerals and energy (Batstone and Viridis, 2014). In this sustainable  
32 development context, anaerobic digestion (AD) presents many advantages over conventional  
33 activated sludge (CAS) processes for domestic wastewater treatment (DWWT) since (i) it  
34 does not require aeration, which decreases energy-demand and associated costs, (ii) it  
35 produces a smaller amount of sludge, begetting less sludge processing and disposal  
36 difficulties and costs, and (iii) it converts organic matter into energy in the form of methane  
37 (van Lier et al., 2008). Recent studies have evaluated a world domestic wastewater production  
38 of  $359.10^9 \text{ m}^3 \cdot \text{year}^{-1}$  (Jones et al., 2021) with a chemical oxygen demand (COD) concentration  
39 between 210 and 740  $\text{mgCOD} \cdot \text{L}^{-1}$  (Srinivasa Raghavan et al., 2017). Based on the theoretical  
40 conversion rate of  $0.35 \text{ m}^3 \cdot \text{CH}_4 \cdot \text{kgCOD}^{-1}$  and the calorific energy of methane of  $35.9 \text{ kJ} \cdot \text{L} \cdot$   
41  $\text{CH}_4^{-1}$ , it appears that 263-903  $\text{TWh} \cdot \text{year}^{-1}$  of energy could be recoverable from wastewater  
42 through AD, that means up to a third of the electricity consumed by the European Union. It  
43 highlights the high bioenergy potential of anaerobic domestic wastewater treatment as an  
44 alternative through fossil consumption and an ease on energy insecurity (Chen et al., 2016).

45           However, the feasibility of the dominant anaerobic technologies (i.e. upflow anaerobic  
46 sludge blanket (UASB) and expended granular sludge bed (EGSB)) for domestic wastewater  
47 is challenged by low influent substrate concentration, huge wastewater quantities and

48 psychrophilic temperature ( $\leq 25$  °C) (Maaz et al., 2019; Vinardell et al., 2020). Thus, even if  
49 the UASB are applied to domestic wastewater in some tropical countries (i.e. Brazil, India,  
50 Colombia), UASB treatment plants still exhibit substandard effluent quality discharge and  
51 poor biogas production (Chernicharo et al., 2015; Srinivasa Raghavan et al., 2017; Kong et  
52 al., 2021b). Anaerobic membrane bioreactor (AnMBR) has been found to overcome UASB  
53 weakness since it enables to operate at high sludge retention times (SRT) and short hydraulic  
54 retention times (HRT). Therefore, even at high volumetric flow rates, ultrafiltration  
55 membrane rejection ensures the growth of slow anaerobic communities and, as a result,  
56 improves the conversion of organic matter into methane energy content (Ji et al., 2021;  
57 Robles et al., 2018). Plus, the membrane unit performs pathogens rejection and could improve  
58 organic micropollutants removal (Robles et al., 2018). Since no nitrogen and phosphorus  
59 degradation is expected from AD, the nutrients-rich effluent could be viewed as a valuable  
60 product suitable for reuse applications (e.g. fertilizer, irrigation) (Maaz et al., 2019). Thus,  
61 AnMBR has the potential to produce a relevant effluent for water reuse while performing a  
62 positive net energy balance (NEB) (Robles et al., 2018; Vinardell et al., 2020). Nevertheless,  
63 dissolved methane and membrane fouling are key issues for full-scale implementation (Maaz  
64 et al., 2019). The loss of energy, in the form of dissolved methane within the effluent, is a  
65 well-known issue for anaerobic wastewater treatment at psychrophilic temperature, due to the  
66 methane solubility increase with a decrease in temperature and the entrapment of biogas  
67 inside the sludge bed. Typical dissolved methane concentration ranges between 10 and 25  
68 mg-CH<sub>4</sub>.L<sup>-1</sup> and up to 100% of the total produced methane could be lost in the effluent  
69 (Sohaib et al., 2022). Fortunately, developing processes, such as degassing membrane, air  
70 stripping oxidation and engineered methanotrophic community in photogranules, have proven  
71 their ability to remove most of the dissolved methane which is attractive for energy recovery

72 and/or to prevent greenhouses gas (GHG) emissions (Robles et al., 2018; Safitri et al., 2021;  
73 Sohaib et al., 2022).

74 As for now, the major constraint to achieve energy-positive AnMBR is membrane  
75 fouling (Maaz et al., 2019). Submerged membrane configuration presents the advantages to be  
76 less energy-consuming for suction and more compact (Liao et al., 2006), nonetheless, it  
77 implies that gas-sparging is the easiest way to mitigate membrane fouling. Studies based on  
78 AnMBR energy demand stated that more than 70% of the energy consumption owed to gas-  
79 sparging for fouling control (Batstone and Viridis, 2014; Smith et al., 2014). To cut the high  
80 operational cost, several research have been undertaken to understand, limit and control  
81 membrane fouling (Robles et al., 2018).

82 Granular anaerobic membrane bioreactor (G-AnMBR), a hybrid biotechnology that  
83 incorporates AnMBR and granular biomass, has raised attention as a sustainable AD process  
84 (Vinardell et al., 2020). Granular sludge presents a great advantage over flocculent sludge for  
85 its (i) high settling capacity, (ii) well-balanced bacteria consortia, (iii) compact and dense  
86 biomass structure and (iv) high-strength to loading rates and toxics shocks (van Lier et al.,  
87 2008). As a result, previous studies stated that granular biomass technology increases  
88 biological removal efficiencies, methane production yield and membrane mitigation (Deng et  
89 al., 2020; Iorhemen et al., 2017). Therefore, the G-AnMBR configuration could be a way to  
90 work without an energy-intensive gas-sparging fouling control strategy and could lead to a  
91 process where the energy recovery overcomes the energy consumption of the system (Smith  
92 et al., 2014).

93 The novelty of this research lies in the unusual G-AnMBR configuration that combines  
94 a granular biomass and a membrane submerged directly inside the sludge bed, without gas-  
95 sparging for fouling control. Hence, the objective of this paper is two-fold. First, to evaluate

96 and understand the impact of a submerged membrane inside a granular anaerobic digester (i.e.  
97 one-stage reactor), focusing on the treatment performances, biogas production and granular  
98 biomass behavior. Second, to study the feasibility of an efficient and positive-energy DWWT  
99 through G-AnMBR at 25°C, without gas-sparging for fouling mitigation. Therefore, a  
100 submerged G-AnMBR and an UASB, as a control reactor, have been continuously operated  
101 during four months with the same operating conditions.

## 102 **2. Materials and methods**

### 103 **2.1 Inoculum and wastewater composition**

104 The seed granular sludge was taken from a full-scale UASB, treating the sewage from a  
105 manufacturer of recycled paper (Saica Paper Champblain-Laveyron, France), at mesophilic  
106 temperature (35-38°C) with an organic loading rate (OLR) of 18 kgCOD.m<sup>-3</sup>.d<sup>-1</sup>. The granular  
107 biomass was gradually acclimatized to ambient temperature (25°C) and low-strength synthetic  
108 wastewater during a period of 500 days (see supplementary material). The lab-scale reactors  
109 were then inoculated with the acclimatized anaerobic granular sludge at a concentration of 70  
110 ± 9.7 and 66 ± 9.2 gTS/L for the UASB and G-AnMBR respectively. The influent  
111 composition was adjusted from Layer et al. (2019) with a C:N:P ratio of 100:1:0.2 and a COD  
112 concentration corresponding to low-strength wastewater (WW). This complex synthetic WW  
113 was chosen for its capacity to lead to the development of granular sludge with the same  
114 characteristics to those fed with raw WW (Layer et al., 2019). The synthetic WW was  
115 prepared weekly and stored under mixing at 4°C. The influent was characterized by total  
116 COD (tCOD) of 274 ± 76 mgCOD.L<sup>-1</sup>, soluble COD (sCOD) of 224 ± 65 mgCOD.L<sup>-1</sup>,  
117 particulate COD (pCOD) of 50 ± 27 mgCOD.L<sup>-1</sup>, dissolved organic carbon (DOC) of 98 ± 30  
118 mgC.L<sup>-1</sup>, volatile fatty acids (VFA) of 130 ± 32 mgCOD.L<sup>-1</sup>, ammonia (NH<sub>4</sub><sup>+</sup>-N) of 3.8 ± 1.6  
119 mgN.L<sup>-1</sup>, nitrite (NO<sub>2</sub><sup>-</sup>-N) of 0.2 ± 0.1 mgN.L<sup>-1</sup>, orthophosphate (PO<sub>4</sub><sup>3-</sup>-P) of 0.5 ± 0.2 mgP.L<sup>-1</sup>

120 <sup>1</sup>, sulfate ( $\text{SO}_4^{2-}\text{S}$ ) of  $11.0 \pm 0.5 \text{ mgS.L}^{-1}$ . pCOD and VFA represent about 20% and 50%  
121 respectively of the influent tCOD.

## 122 **2.2 Experimental set-up and operating conditions**

123 Two granular anaerobic reactors, namely G-AnMBR and UASB (as a control reactor),  
124 were continuously operated in parallel during 120 days. The two experimental lab-scale  
125 reactors consisted of parallelepiped-shaped tanks with equal working volume of 6.2 L (see  
126 supplementary material). The liquid level was automatically controlled by a vibronic point  
127 level detector (Liquiphant FTL31, Endress+Hauser, Switzerland). The synthetic wastewater  
128 was introduced at the bottom of each reactors and flowed upwards the granular sludge bed. At  
129 the top of the two reactors, supernatant was pumped through a peristaltic pump (Watson  
130 Marlow (WMFTG), UK) and recirculated at the lower part to bring additional turbulences and  
131 homogenized the dissolved phase. The upflow liquid velocity (ULV) was set around  $2 \text{ m.h}^{-1}$ .  
132 A polyethersulfone (PES) flat sheet membrane (Microdyn-Nadir®, Germany) with a nominal  
133 pore size of  $0.04 \mu\text{m}$  and a surface area of  $0.34 \text{ m}^2$  was submerged at the center of the one-  
134 stage G-AnMBR reactor. The permeate was obtained through a peristaltic pump (LeadFluid®,  
135 China) and the net filtration flux was maintained at  $1.45 \pm 0.35 \text{ L.m}^{-2}.\text{h}^{-1}$  (LMH). Only an  
136 automatic intermittent suction cycle was performed to mitigate membrane fouling at a low  
137 energy-demand. The operation cycle was as follows: (i) 8 min 15 s of filtration, (ii) 30 s of  
138 initial relaxation, (iii) 45 s of backwash and (iv) 30 s of final relaxation. To get as close as  
139 possible to the G-AnMBR configuration and its hydrodynamic pathway, the same membrane  
140 module was immersed in the UASB tank but no permeate was suctioned through this  
141 membrane. The UASB effluent was pumped through supernatant with the same operation  
142 cycle as the G-AnMBR. As shown in Table 1, similar operating conditions were applied in  
143 both reactors. The reactors were equipped with temperature sensor, pH and oxidation  
144 reduction potential (ORP) probes (PCE Instruments, Deutschland). The physico-chemical

145 parameters were stable during the campaign and the very low redox (< -450 mV) values  
146 confirm that anaerobic conditions were fulfilled. The reactor temperature was kept at around  
147 25°C with a cryostat through a tubular heat exchanger. The hydraulic retention time (HRT)  
148 and the OLR were maintained at 13 h and 0.5 kgCOD.m<sup>-3</sup>.d<sup>-1</sup>. Pressure sensors were installed  
149 to monitor atmospheric pressure (Patm), reactors headspace pressures and permeate pressure  
150 to obtain the transmembrane pressure (ATM.ECO, Sensor Technik Sirmach (STS),  
151 Switzerland). No sludge was intentionally purged except for the need of sampling.

### 152 **2.3 Analytical methods**

153 DOC was analyzed two days a week on sample pre-filtered at 0.22 µm by TOC analyzer  
154 (TOC-V<sub>CSN</sub>, Shimadzu Corporation, Japan). Mixed liquor suspended solids (MLSS) and  
155 mixed liquor volatile suspended solids (MLVSS) were measured weekly in the UASB  
156 effluent and G-AnMBR permeate according to Standard Methods (APHA et al., 1998). COD  
157 concentrations were determined twice a week using pre-dosed photochemical test (Hach,  
158 Germany, LKC 500, 314, 1414, 514) and UV-Vis spectrophotometer (DR3900, Hach,  
159 Germany). sCOD was measured after sample filtration through 0.22 µm syringe filter. The  
160 denominated tCOD and sCOD removal rates (tCOD<sub>removal</sub> and sCOD<sub>removal</sub> respectively) were  
161 calculated as follow:

$$tCOD_{removal} = \frac{tCOD_{in} - tCOD_{out}}{tCOD_{in}} = \frac{COD_{removed}}{tCOD_{in}} \quad \text{Eq. 1}$$

$$sCOD_{removal} = \frac{tCOD_{in} - sCOD_{out}}{tCOD_{in}} \quad \text{Eq. 2}$$

162 where tCOD<sub>in</sub> is the tCOD measured in the influent, tCOD<sub>out</sub> is the tCOD of the effluent and  
163 sCOD<sub>out</sub> is the effluent sCOD concentration.



## 2.4 Three-dimensional fluorescence excitation-emission matrix (3DEEM)

Supernatant from G-AnMBR and effluents from both reactors were pre-filtered through 1.2  $\mu\text{m}$  filter (Grade GF/C, Whatman, UK) and diluted 25 times to limit overlapping signals. The three-dimensional fluorescence excitation-emission matrix (3DEEM) was divided into four areas according to their respective fluorophores and the volume of fluorescence beneath each region were obtained according to Jacquin et al. (2017). The volume of fluorescence (in arbitrary unit per  $\text{nm}^2$  (A.U/ $\text{nm}^2$ )) gives a semi-quantitative information about the amount of fluorophores for each region.

## 2.5 Granular sludge characteristics

Total solids (TS) and Volatile solids (VS) were measured during reactor seeding according to Standard Methods (APHA et al., 1998). Sludge volume index after 10 minutes ( $\text{SVI}_{10}$ ) and 30 minutes ( $\text{SVI}_{30}$ ) were evaluated by dividing the volume of sludge bed (mL) by the initial sample volume (mL) and the TS concentration of the sample ( $\text{gTS}\cdot\text{L}^{-1}$ ). The zeta potential of the sludge was measured by Litesizer 500 (Anton Paar, Spaar). The particle size distribution (PSD) of the granular sludge was performed by wet sieving following the method of Derlon et al. (2016) with standard sieves of 0.63 and 0.125 mm mesh sizes. Hence, the sludge was separated into three fractions based on the particle diameter ( $d_p$ ) such as: (i) large granules with a  $d_p$  higher than 0.63 mm ( $d_{p \geq 0.63}$ ), (ii) medium granules mixed with small granules with a  $d_p$  between 0.63 and 0.125 mm ( $d_{p 0.125-0.63}$ ) and (iii) flocs for the sludge with a  $d_p$  less than 0.125 mm ( $d_{p \leq 0.125}$ ). TS and VS of each fraction were measured. The proportion of each class of sludge was expressed as percentage of total sludge mass (Eq. 3). VS/TS ratio was calculated within the fractions.

$$\text{Mass fraction}_i (\%) = \frac{m_i}{\sum_1^3 m_i} \times 100 \quad \text{Eq. 3}$$

## 186                    **2.6 Methane production**

187            Produced methane was measured in the gas and liquid phases. The gas phase flow rate  
188 was continuously recorded by a volumetric flowmeter (MilliGas Counter, Ritter, Germany).  
189 Biogas composition of the headspace was regularly analyzed by a gas chromatography system  
190 (Clarus 400, Perkin Elmer, USA) coupled with a thermal conductivity detector at 150°C (GC-  
191 TCD). Gas phase was collected through a quick gas connection (Swagelok, USA) into an  
192 airtight system. 300 µL of the biogas was then sampled through a septum using a gastight  
193 syringe (Hamilton Company, USA) and injected into GC-TCD. Methane concentration was  
194 defined as the average of all measurements performed during the stable phase of operation.  
195 Methane flow rate was converted in NL-CH<sub>4</sub>.day<sup>-1</sup> by using the average temperature and the  
196 atmospheric pressure (*P*<sub>atm</sub>) of the day. Dissolved methane was quantified based on the  
197 headspace method (Giménez et al., 2012; Souza et al., 2011). Vials equipped with a stirrer  
198 (total volume (*V*<sub>T</sub>) of 11.6 mL) were sealed with a septum cap and drained with helium during  
199 10 min to avoid any presence of air. A known volume of effluent sample (*V*<sub>L</sub>) was collected  
200 using a gastight syringe (Hamilton Company, USA) and injected in sealed vials. Then, vials  
201 were kept for stirring at 700 rpm and 25°C for 2h in order to reach gas/liquid equilibrium.  
202 Once the equilibrium reached, the total pressure of the headspace (*P*<sub>T</sub>) was recorded and the  
203 biogas was analyzed through GC-TCD to determine the methane molar fraction (*y*<sub>CH<sub>4</sub></sub>). The  
204 dissolved methane concentration (*C*<sub>CH<sub>4</sub></sub><sup>L</sup>) in the effluent was then calculated according to the  
205 following expressions:

$$C_{CH_4}^L = \frac{C_{CH_4}^G \times (V_G + \frac{V_L}{H(T)})}{V_L} \quad \text{Eq. 4}$$

206 with

$$C_{CH_4}^G = \frac{y_{CH_4} \times P_T \times MM(CH_4)}{R \times T} \quad \text{Eq. 5}$$

207 where  $C_{CH_4}^G$  is the CH<sub>4</sub> concentration in the gas phase (mg.L<sup>-1</sup>),  $V_G$  is the volume of the gas  
 208 phase (L),  $MM(CH_4)$  is the molecular weight of methane (16g.mol<sup>-1</sup>),  $R$  is the universal  
 209 constant of gases (0.082 atm.L.mol<sup>-1</sup>.K<sup>-1</sup>),  $T$  is the temperature (K) and  $H(T)$  is the  
 210 dimensionless temperature-dependent Henry's constant for methane (Giménez et al., 2012).  
 211 The saturation degree was calculated based on the theoretical value of methane dissolved in  
 212 the liquid phase ( $C_{CH_4}^{L*}$ , in mg.L<sup>-1</sup>) calculated according to the Henry's law thermodynamic in  
 213 equilibrium with the gas phase as described in Eq. 6.

$$Saturation\ degree = \frac{C_{CH_4}^L}{C_{CH_4}^{L*}} = \frac{C_{CH_4}^L}{H(T) \cdot C_{CH_4}^G} \quad Eq. 6$$

## 214 **2.7 COD mass balance**

215 The COD mass balance was determined based on Eq. 7:

$$tCOD_{in} = tCOD_{out} + COD_{CH_4}^G + COD_{CH_4}^L + COD_{SO_4} + \Delta COD_{biomass} \quad Eq. 7$$

216 where  $tCOD_{in}$  and  $tCOD_{out}$  are the tCOD measured in the influent and effluent respectively,  
 217  $COD_{CH_4}^G$  and  $COD_{CH_4}^L$  correspond to the methane quantified in the gas and liquid phase  
 218 respectively and converted in equivalent COD using the empirical relationship of 0,38 L-  
 219 CH<sub>4</sub>.gCOD<sup>-1</sup> at 25°C.  $COD_{SO_4}$  is the theoretical COD used for sulfate reduction by sulfate-  
 220 reducing bacteria (SRB) based on the experimental amount of sulfate removal measured and  
 221 the theoretical value of 0.67 gCOD.gSO<sub>4</sub><sup>-1</sup>. The residual COD was considered as biomass  
 222 conversion ( $\Delta COD_{biomass}$ ) and used to calculate the sludge yield ( $Y_H$ ) of granular biomass as  
 223 follows :

$$Y_H = \frac{\Delta COD_{biomass}}{1.42 \cdot COD_{removed}}$$

224 where  $Y_H$  is in  $\text{gVSS} \cdot \text{gCOD}_{\text{removed}}^{-1}$ ,  $\Delta COD_{biomass}$  is in  $\text{gCOD} \cdot \text{day}^{-1}$ ,  $COD_{removed}$  is in  
 225  $\text{gCOD}_{\text{removed}} \cdot \text{day}^{-1}$  and 1.42 is the COD to VSS ratio based on the biomass stoichiometry  
 226 ( $\text{C}_5\text{H}_7\text{O}_2\text{N}$ ) (van Lier et al., 2008).

## 227 **3. Results and discussion**

### 228 **3.1 AnMBR and UASB organic conversion and effluent quality**

#### 229 **3.1.1 Organic matter removal**

230 Fig. 1 presents the tCOD and sCOD removal rate in the G-AnMBR and UASB reactors  
 231 during the overall operation period. Table 2 gives complementary results of the average  
 232 concentrations of tCOD, sCOD, DOC, MLSS and MLVSS measured in the outlets of both  
 233 reactors. During the first month of the experiment, the UASB reactor had a very poor tCOD  
 234 removal efficiency compared to the G-AnMBR reactor which was almost directly stable and  
 235 efficient ( $35.9 \pm 12.6\%$  and  $81.6 \pm 3.8\%$  respectively (Table 2)). This difference is clearly due to  
 236 the UF membrane separation that retains all particles and most of the dissolved and colloidal  
 237 organic matter (DCOM) in the G-AnMBR whereas small granules, flocs and free-bacteria,  
 238 just as some DCOM and intermediate degradation products, were washed-out within the  
 239 supernatant of the UASB (Chen et al., 2017; Ozgun et al., 2015). This can be confirmed by  
 240 the high amount of MLSS measured in the UASB effluent during the transient period (Table  
 241 2) and the same concentration profiles followed by total COD and TSS. As a result, the  
 242 removal rate for tCOD in the UASB showed an increasing trend until it achieved a steady-  
 243 state after 31 days of operation. After the transient period, high organic removal efficiencies  
 244 were carried out in both reactors, as describes in Table 2. Most notably, the G-AnMBR  
 245 performed higher organic compounds removal ( $92.3 \pm 4.1\%$  of tCOD,  $90.0 \pm 5.4\%$  of sCOD

246 and  $97.8 \pm 1.0\%$  of DOC) in comparison to the UASB ( $79.2 \pm 8.5\%$ ,  $80.7 \pm 7.3\%$  and  
247  $86.5 \pm 9.1\%$  of tCOD, sCOD and DOC respectively). This result is well in line with previous  
248 studies wherein 40-80% of the tCOD was removed by UASB reactors (Chernicharo et al.,  
249 2015) and 71-98% by AnMBR at psychrophilic temperature (see supplementary material).  
250 Hence, it appears that the G-AnMBR configuration without gas sparging permits the  
251 degradation of organic compounds as high as gas-sparging ones.

252 Relevant differences in sCOD were observed between G-AnMBR supernatant  
253 ( $55.3 \pm 16.3 \text{ mgCOD.L}^{-1}$ ) and sCOD permeate ( $17.4 \pm 4.3 \text{ mgCOD.L}^{-1}$ ) indicating that a part  
254 of soluble compounds does not pass across the membrane. Hence,  $16.4 \pm 4.7\%$  of the entering  
255 sCOD is retained by the membrane barrier and/or removed by the biofilm. The average pore  
256 size of the flat sheet membrane is  $0.04 \mu\text{m}$  therefore a part of DCOM (i.e. proteins,  
257 polysaccharides, humic aggregates) was expected to be retained by the membrane due to their  
258 size. Mechanisms responsible of the retention of the soluble and colloidal matter can be either  
259 related to physical phenomenon (i.e. size exclusion, adsorption, charge exclusion) as well as  
260 microbial biofilm activity (Smith et al., 2013). Since no COD accumulation was observed in  
261 the G-AnMBR supernatant, it can be reasonably assumed that the membrane provides a  
262 physical barrier to slowly-biodegradable and non-settleable compounds (e.g. flocs, particles)  
263 or by-products (e.g. products from polymers hydrolysis, proteins, SMP) which are later  
264 biologically transformed thanks to a longer contact time between bulk sludge and organic  
265 material (Gouveia et al., 2015; Ozgun et al., 2015) and thus, explains the enhanced G-  
266 AnMBR performances on organic matter removal.

267 Table 3 and Fig. 2 provides complementary information about DCOM behavior inside  
268 the anaerobic reactors. Comparing the total volume of fluorescence of the two effluents, it is  
269 noticeable that more organics were removed in the G-AnMBR. On average 42.3% of the  
270 fluorescent compounds present in the supernatant of the G-AnMBR did not pass through the

271 membrane. It can be seen from 3DEEM spectra (Fig. 2) and membrane retention rate (Table  
272 3) that protein- and SMP-like molecules (i.e. regions I+II and IV) are the main compounds  
273 retained by the submerged membrane. Conversely, humic-like substances (combining fulvic-  
274 (III) and humic- (V) acid-like molecules) had the smallest rejection capacity due to their  
275 lower molecular weight. No notable increase in total volume of fluorescence was observed in  
276 the supernatant of the G-AnMBR over the time. Interestingly, Jacquin et al. (2017) concludes  
277 that the protein-like regions are more associated with colloidal proteins whereas the SMP-like  
278 are supposed to be macromolecular proteins present in the dissolved phase. These 3DEEM  
279 fluorescence results support the substantial benefit of the membrane incorporation inside the  
280 anaerobic reactor over the retention and bio-conversion of the macromolecules and colloids.  
281 Instead of being taken away with the effluent, like in the UASB, these compounds are kept  
282 into the supernatant and could be used as additional organic matter for biogas production.

### 283 **3.1.2 Suspended solids removal**

284 Concerning the suspended solids removal, nearly all suspended solids (SS) were  
285 removed by the membrane filtration (Table 2). In comparison, an average of 10.8 mg/L was  
286 found in the UASB effluent during the stable period which is much higher than the G-  
287 AnMBR, even if it is under the 35 mgSS.L<sup>-1</sup> standard regulation discharge (Directive  
288 91/271/EEC). Thus, the results emphasized that UASB combined with UF filtration for  
289 DWWT at ambient temperature can achieve an excellent water quality (physical disinfection  
290 and suspended matter retention).

### 291 **3.2 Biogas production**

292 Table 4 presents the results of methane quantification in the gas and the liquid phases.  
293 Methane to carbon dioxide ratio of the biogas was around 80/20. This high methane content is  
294 a benefit for the energy mass balance and shows a well-functioning of methanogens  
295 populations (Chen et al., 2020). The methane flow rate achieved during the steady-state was

296 significantly upper in the G-AnMBR than in the UASB ( $0.85 \pm 0.06$  and  $0.57 \pm 0.05$  NL-  
297  $\text{CH}_4 \cdot \text{day}^{-1}$  respectively). Interestingly, only a slight difference was observed between methane  
298 yields of  $0.27 \pm 0.03$  for the G-AnMBR and  $0.22 \pm 0.04$   $\text{L-CH}_4 \cdot \text{gCOD}_{\text{removed}}^{-1}$  for the UASB.  
299 This result indicates that no significant improvement in sludge methanogenic activity was  
300 observed with the incorporation of a membrane in spite of an increase in global carbon  
301 conversion performances (Ozgun et al., 2015). It supports that membrane filtration enhances  
302 the net production of methane by retaining particulate matter and DCOM (including VFA).  
303 Longer contact time between organics and active biomass allows the conversion of slowly-  
304 biodegradable matter and by-products into additional methane.

305 The concentration of methane inside the aqueous phase is about  $12.8 \text{ mg-CH}_4 \cdot \text{L}^{-1}$  for the  
306 G-AnMBR and  $11.8 \text{ mg-CH}_4 \cdot \text{L}^{-1}$  in the UASB, which corresponds approximately to 22% and  
307 27% respectively of the total methane produced. Some studies have demonstrated a saturation  
308 degree lower in AnMBR than in UASB effluents (Gouveia et al., 2015a). This phenomenon is  
309 mainly due to turbulences caused by membrane filtration operating conditions and fouling  
310 mitigation technics used that provide a better mixing and so, a better gas-liquid transfer. In  
311 this study, the results showed a saturation degree in the G-AnMBR of  $2.00 \pm 0.10$  which is  
312 slightly higher than the UASB where the value is  $1.80 \pm 0.07$ . Saturation degrees obtained are  
313 consistent with Smith et al. (2015) and Souza et al. (2011) results which found oversaturation  
314 degrees about 2.2 at  $15^\circ\text{C}$  and 1.4-1.7 at  $25^\circ\text{C}$  respectively in a UASB treating domestic  
315 wastewater. The closeness of the G-AnMBR and UASB values could be explained by the  
316 same setup configurations and no fouling control method used aside intermittent filtration.  
317 Therefore, no obvious hydrodynamic difference should occur between reactors.

318 By taking into consideration the whole methane generated ( $C_{\text{CH}_4}^L + C_{\text{CH}_4}^G$ ), the methane  
319 yields obtained were  $0.33 \pm 0.03$  and  $0.28 \pm 0.04$   $\text{NL-CH}_4 \cdot \text{gCOD}_{\text{removed}}^{-1}$  for the G-AnMBR  
320 and UASB respectively. The closeness of the average methane yields measured to the

321 theoretical maximal value ( $0.38 \text{ NL-CH}_4.\text{gCOD}_{\text{removed}}^{-1}$ ) demonstrates that an acclimatized  
322 biomass could well-convert COD into methane at sub-optimal temperature. Methane yields  
323 obtained in this study are in the high-range of the ones conventionally found in anaerobic  
324 digestion studies that are mostly between  $0.12\text{-}0.25 \text{ L-CH}_4.\text{gCOD}_{\text{removed}}^{-1}$ . This high  
325 conversion rate could be attributed to the long one-year acclimatization of the anaerobic  
326 granular sludge at the operating conditions and to the relatively high VFA concentration in the  
327 synthetic wastewater that promotes acetogenesis and methanogenesis pathways. Moreover, in  
328 this study, the conversion rate takes into account the measured dissolved methane which is  
329 generally neglected or estimated through Henry's law coefficient.

### 330 **3.3 COD mass balance**

331 COD mass balance for the G-AnMBR and UASB are presented in Fig. 3. It is apparent  
332 from those results that the G-AnMBR allowed a higher removal rate of the COD since the  
333 COD remaining in the UASB effluent is almost three-times higher. The total methane  
334 produced by the anaerobic digestion process for the UASB and G-AMBR were 61.1% and  
335 84.7% respectively but only 44.6% and 66.3% were in the gaseous phase and can be directly  
336 valuable. This shows the importance to recover dissolved biogas from effluent to avoid  
337 environmental issues but also to improve the potential energy recovery. These results are  
338 consistent with those obtained by Ji et al. (2021) for an AnMBR pilot-scale treating raw  
339 domestic wastewater with close operating conditions ( $\text{OLR} = 0.72 \text{ kgCOD}.\text{m}^{-3}.\text{d}^{-1}$  ;  $\text{HRT} =$   
340  $12\text{h}$  ;  $25^\circ\text{C}$ ) where 10.7% of the COD was remaining in the effluent and 75% of the COD of  
341 the influent was converted into methane of which 63% in gaseous phase. It can be seen that  
342 the amounts of  $\text{COD}_{\text{in}}$  used for sulfate reduction and the one transformed into dissolved  
343 biogas are nearly the same for both anaerobic reactors. This implies that the difference  
344 between both methane conversion rate is due to the COD removal capacity and the use of  
345  $\text{COD}_{\text{in}}$  for biomass synthesis.



346 From the COD entering in the systems, 6.3% (G-AnMBR) and 14.7% (UASB) of the  
347 COD were used for biomass production (Fig. 3). This suggests that a higher part of the COD<sub>in</sub>  
348 was consumed for biomass synthesis in the UASB and was therefore not available for  
349 methane production. As a result, the calculated sludge yield was about 0.05  
350 gVSS.gCOD<sub>removed</sub><sup>-1</sup> in the G-AnMBR against 0.11 gVSS.gCOD<sub>removed</sub><sup>-1</sup> in the UASB. The  
351 sludge yield of the granular biomass corresponds to the typical low anaerobic sludge yield and  
352 matches with previous AnMBR studies for DWWT at ambient temperature (see  
353 supplementary material). The sludge yield in the UASB was twice higher than the one in the  
354 G-AnMBR system. This phenomenon can be explained by the biomass washout occurring in  
355 the UASB at the early stage because of the selective hydraulic pressure. According to the  
356 Monod equation, the increase of the specific organic loading rate, caused by the loss of  
357 flocculated sludge and small granules, stimulates the biomass growth (Chen et al., 2017;  
358 Ozgun et al., 2015).

359 It should be noticed that in the case of AnMBR with external submerged membrane  
360 configuration, the selective pressure is not overcome, though, there is an increase in the  
361 biomass yield (Chen et al., 2017). Those findings suggest that the G-AnMBR configuration  
362 with the membrane submerged inside the mixed granular anaerobic digester enhances the  
363 conversion of the COD into methane while maintaining a low sludge production rate. This is  
364 of great interest compared to conventional activated sludge processes since the energy  
365 requirements as well as the treatment and disposal costs for sludge management could be  
366 minimized.

### 367 **3.4 Granular sludge properties**

368 The settling ability and granulation behavior of the biomass were evaluated through the  
369 SVI<sub>30</sub> and SVI<sub>10</sub> to SVI<sub>30</sub> ratio (see supplementary material). Both reactors were inoculated  
370 with similar granular sludge SVI of 31.1 and 28.9 mL.gTS<sup>-1</sup> for UASB and G-AnMBR. Also,

371 the  $SVI_{10}/SVI_{30}$  ratio was around 1.3-1.4 meaning that the sludge is a mixture of readily  
372 settleable granules and small granules and flocs with a lower capacity of settling. At the end  
373 of the operation,  $SVI_{30}$  value for the UASB granular sludge decreased sharply ( $14.7 \text{ mL.gTS}^{-1}$ )  
374 <sup>1)</sup> and exhibited a  $SVI_{10}/SVI_{30}$  ratio of 1. These results confirm the selective pressure that  
375 occurs in the UASB reactor where sole well-settleable granular biomass is kept into the  
376 UASB. In contrast, the G-AnMBR configuration does not allow free and flocculated biomass  
377 to run-off from the reactor and, as a result, higher  $SVI_{30}$  ( $22.9 \text{ mL.gTS}^{-1}$ ) and  $SVI_{10}/SVI_{30}$   
378 ratio (1.3) were measured in comparison to the UASB values.

379 Zeta potential was also measured as it gives an indication about the aptitude of sludge  
380 biomass to aggregate among themselves. As suggested by others studies, a higher zeta  
381 potential meaning a decline of the negative surface charge and, therefore, electrostatic  
382 repulsion could be easily neutralized (Chen et al., 2017). Zeta potential values (see  
383 supplementary material) were found higher at the end of the experiment than that at the  
384 seeding for the G-AnMBR and UASB reactors suggesting that the biomass attachment has  
385 been promoted during process operation.

386 Fig. 4 provides the changing in particle size distribution (PSD) of the granular sludge  
387 for both reactors. Seed sludge for UASB and G-AnMBR showed almost identical profiles of  
388 PSD corroborating the same inoculation. At the initial stage, large granules ( $d_p \geq 0.63$ )  
389 accounted for  $\approx 70\%$  of the total TS mass of the granular sludge whereas the main part of the  
390 VS, commonly assimilated to active biomass, belonged to the medium fraction ( $d_p 0.125-0.63$ )  
391 with  $65 \pm 3 \%$  of the total VS mass. As expected, the flocs fraction represented a very low  
392 amount of the total biomass since it has a small density compared to granular sludge. After  
393 115 days of operation, a significant difference was observed for the granular sludge PSD of  
394 the UASB reactor. As expected, shift to larger granules was observed in the UASB reactor  
395 with 87.2% and 74.6% of the total TS and VS mass respectively measured in the biggest

396 fraction ( $d_{p \geq 0.63}$ ). This hypothesizes that, besides the sludge washout of poor-settleable  
397 particles, growth of larger-sized granules was promoted which is in accordance with the zeta  
398 potential values. On the other hand, the granular sludge PSD of the G-AnMBR showed a  
399 broader repartition because of the physical barrier retention that permitted the development of  
400 various sludge type including free-bacteria and light flocs and granules. As reported in  
401 previous studies, a wide variety of biomass (i.e. free-bacteria, fines flocs, granules, cake layer,  
402 etc.) results in a large microbial diversity (Lin et al., 2011; Zhou et al., 2019). These results  
403 clearly indicate that membrane incorporation enables to maintain the PSD tendency of the  
404 granular sludge and leads to a more diversified anaerobic population by allowing the slow  
405 growth of anaerobic methanogens (Lin et al., 2013). Moreover, frequent microbial samples  
406 were taken during this experiment and will be later analyzed to characterize and confirm the  
407 change in diversity and density of the microbial communities following the membrane  
408 incorporation. Plus, further experiments should be conducted to evaluate the structural  
409 stability of the granules with varying operating conditions to see the range of applicability of  
410 the granular biomass.

411 VS/TS ratio depicts in Fig. 4 is an indicator to evaluate the amount of biomass within  
412 the granular sludge. A global decrease in VS/TS ratio occurred in UASB intermediate and  
413 smaller fractions while the decrease concerned particularly the intermediate granule fraction  
414 in the G-AnMBR. This translates an increase in mineral content inside the granular fraction  
415 that is exacerbated by biomass loss in the UASB. SEM-EDX analysis provided in  
416 supplementary material indicates that the mineral part of the granular sludge was mainly  
417 composed of calcium carbonate  $\text{CaCO}_3$ . Prior studies stated that calcium concentration in  
418 granules is negatively correlated to VS/TS ratio and bacterial specific activity. Core  
419 calcification of granules is a harmful phenomenon as the calcium mineral precipitate and  
420 calcium salts deposited in the outer layer limit the diffusion of molecules in granules

421 interstitial spaces (Zhang et al., 2021). It is likely that membrane attenuates the impact of  
422 calcification by retaining the flocculated biomass within the reactor which further promote the  
423 active biomass inside the granules (i.e. VS/TS ratio).

### 424 **3.5 G-AnMBR energy recovery evaluation and competitiveness**

425 Based on the methane produced from COD conversion, the energy potentially recovered  
426 from DWWT was calculated. Table 5 provides an overview of net energy balance (NEB) and  
427 effluent quality for the most common aerobic and anaerobic processes for DWWT at ambient  
428 temperature. Anaerobic treatments have an evident energetic advantage comparing to the  
429 aerobic DWWT that exhibits a negative NEB from -0.2 to -2 kWh.m<sup>-3</sup>. Although UASB has a  
430 low energy demand and high energy recovery (NEB of 0.35-0.5 kWh.m<sup>-3</sup>), the effluent quality  
431 obtained is not enough to meet discharge standards and to outcompete CAS or AeMBR  
432 reactors (Chernicharo et al., 2015; Ozgun et al., 2013). Conversely, conventional AnMBR  
433 reach high organics and suspended solids removal efficiencies but achieve a NEB close to  
434 zero due to the gas-sparging energy-demand for membrane fouling control that accounts for  
435 more than 70% of the process energy consumption (Smith et al., 2014). Therefore, it is  
436 apparent from Table 5 that the G-AnMBR operated in this study is the only system which  
437 allows both excellent effluent quality and strong positive net energy balance, even when the  
438 dissolved methane is not recovered (0.58 kWh.m<sup>-3</sup>). This finding highlights the interest of the  
439 G-AnMBR submerged configuration with no gas-sparging fouling control which optimizes at  
440 the same time COD to methane conversion rate, energy-requirement, effluent quality and  
441 reactor compactness. Hence, G-AnMBR technologies could be useful in poor-energy, water-  
442 shortage and scarce-space areas (Kong et al., 2021a; Robles et al., 2018). In addition,  
443 considering the complete recovery of dissolved biogas, nearly 0.20 kWh.m<sup>-3</sup> of additional  
444 energy could be generated from the domestic WW by G-AnMBR treatment. As mentioned  
445 above, it points out that the dissolved methane recovery needs further investigation as it

446 accounts for more than 25% of the total produced methane and because methane is an  
447 important greenhouse concern (Gouveia et al., 2015; Ji et al., 2021; Smith et al., 2013).

## 448 **Conclusion**

449 It has been demonstrated that the submerged G-AnMBR configuration, without gas  
450 sparging, represents a sustainable biotechnology for DWWT at ambient temperatures over  
451 conventional processes with regard to effluent quality and energy requirements. The  
452 membrane barrier helped to maintain the process stability, led to a broader sludge diversity  
453 with no granular sludge activity alteration. The G-AnMBR reached organic removal rate as  
454 high as aerobic process with a tCOD removal of ~92.3%. The NEB was maximized to +0.58  
455 kWh.m<sup>-3</sup> with the G-AnMBR setup without gas-sparging, bringing the G-AnMBR as a  
456 promising process which may be part of the circular economy strategies.

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593 Table 1 - G-AnMBR and UASB operating conditions (mean values  $\pm$  SD).

Parameter	G-AnMBR	UASB
Temperature ( $^{\circ}$ C)	$25.0 \pm 0.8$	$25.8 \pm 1.8$
pH (-)	$7.0 \pm 0.2$	$7.1 \pm 0.2$
Redox (mV)	$-462 \pm 37$	$-456 \pm 32$
Jp <sub>20,NET</sub> (LMH)	$1.34 \pm 0.14$	-
Jp <sub>20,INST</sub>	$2.48 \pm 0.22$	-
HRT (h)	$13.1 \pm 2.0$	$13.2 \pm 3.0$
OLR (kgCOD.m <sup>-3</sup> .d <sup>-1</sup> )	$0.50 \pm 0.14$	$0.48 \pm 0.15$
ULV (m.h <sup>-1</sup> )	2.25	2.04

594

595 Table 2 - Effluent composition and removal efficiencies of the G-AnMBR and UASB (mean values  $\pm$  SD ;  
 596  $n \geq 20$ ).

Phase		G-AnMBR permeate		UASB effluent	
		Transient	Steady	Transient	Steady
tCOD <sub>eff</sub>	(mg/L)	43.3 $\pm$ 13.1	22.0 $\pm$ 14.5	150.2 $\pm$ 36.0	59.0 $\pm$ 28.5
tCOD <sub>removal</sub>	(%)	81.6 $\pm$ 3.8	92.3 $\pm$ 4.1	35.9 $\pm$ 12.6	79.2 $\pm$ 8.5
sCOD <sub>eff</sub>	(mg/L)	43.3 $\pm$ 13.1	22.0 $\pm$ 14.5	69.0 $\pm$ 8.1	44.1 $\pm$ 18.9
sCOD <sub>removal</sub>	(%)	76.7 $\pm$ 3.6	90.0 $\pm$ 5.4	63.9 $\pm$ 7.8	80.7 $\pm$ 7.3
DOC <sub>eff</sub>	(mg/L)	7.5 $\pm$ 0.0	2.1 $\pm$ 0.6	23.6 $\pm$ 0.0	13.8 $\pm$ 9.4
DOC <sub>removal</sub>	(%)	85.7 $\pm$ 0.0	97.8 $\pm$ 1.0	60.0 $\pm$ 0.0	86.5 $\pm$ 9.1
MLSS <sub>eff</sub>	(mg/L)	ND	ND	113.1 $\pm$ 87.7	10.8 $\pm$ 6.0
MLVSS <sub>eff</sub>	(mg/L)	ND	ND	60.4 $\pm$ 37.4	9.3 $\pm$ 5.4

597 ND : non-detectable

598

599 Table 3 - Average normalized volume of fluorescence for the different regions of the 3DEEM spectra for G-  
 600 AnMBR supernatant and permeate and UASB effluent and the membrane rejection during the steady-state  
 601 period. Errors represent standard deviation (n=7).

		3DEEM Region			
		I+II	IV	III+V	Total
G-AnMBR	Supernatant ( $\times 10^8$ A.U/nm <sup>2</sup> )	2.74 $\pm$ 1.04	0.12 $\pm$ 0.03	0.52 $\pm$ 0.09	3.38 $\pm$ 1.10
	Permeate ( $\times 10^8$ A.U/nm <sup>2</sup> )	1.46 $\pm$ 0.68	0.05 $\pm$ 0.01	0.40 $\pm$ 0.07	1.90 $\pm$ 0.69
	Membrane rejection (%)	45.9 $\pm$ 20.0	59.8 $\pm$ 18.3	24.0 $\pm$ 14.6	42.3 $\pm$ 17.6
UASB	Effluent ( $\times 10^8$ A.U/nm <sup>2</sup> )	2.45 $\pm$ 1.40	0.12 $\pm$ 0.08	0.45 $\pm$ 0.14	3.02 $\pm$ 1.59

602

603 Table 4 - Methane production in the gas phase and liquid phase (mean values  $\pm$  SD ;  $n \geq 15$ ).

	Unit	G-AnMBR	UASB
CH <sub>4</sub> flow rate	NL-CH <sub>4</sub> .day <sup>-1</sup>	0.85 $\pm$ 0.06	0.57 $\pm$ 0.05
CH <sub>4</sub> /CO <sub>2</sub> ratio	% / %	79.9/20.1 $\pm$ 3.5	81.9/18.1 $\pm$ 6.8
Gaseous methane yield	NL-CH <sub>4</sub> /g-COD <sub>removed</sub>	0.27 $\pm$ 0.03	0.22 $\pm$ 0.04
Dissolved methane	mg-CH <sub>4</sub> .L <sup>-1</sup>	12.8 $\pm$ 0.7	11.7 $\pm$ 1.1
Degree of saturation	-	2.00 $\pm$ 0.10	1.80 $\pm$ 0.07
Dissolved CH <sub>4</sub> flow rate	NL-CH <sub>4</sub> .day <sup>-1</sup>	0.24 $\pm$ 0.01	0.22 $\pm$ 0.02
Total methane yield	NL-CH <sub>4</sub> /g-COD <sub>removed</sub>	0.33 $\pm$ 0.03	0.28 $\pm$ 0.04

604

605 Table 5 - Comparison of net energy balance and effluent quality between aerobic and anaerobic technologies for  
 606 low-strenght domestic wastewater treatment at ambient temperature ( $\leq 25^{\circ}\text{C}$ ).

	Energy consumption ( $\text{kWh.m}^{-3}$ )	Energy from $\text{CH}_4$ ( $\text{kWh.m}^{-3}$ )	Net Energy Balance ( $\text{kWh.m}^{-3}$ )	Effluent quality	
				COD removal (%)	Effluent TSS ( $\text{mg.L}^{-1}$ )
CAS	0.2 – 0.8 <sup>a,b</sup>	-	-0.2 – -0.8	85-93 <sup>a</sup>	<30
AeMBR	0.6 – 2 <sup>b,c</sup>	-	-0.6 – -2	>95 <sup>a</sup>	<1 <sup>c</sup>
UASB	0.11 <sup>d</sup>	0.6 <sup>d</sup>	0.5	40-80 <sup>g,h</sup>	60-200 <sup>h</sup>
		0.46 (0.64*) <sup>j</sup>	0.35 (0.53*)	79 <sup>j</sup>	$\cong$ 10 <sup>j</sup>
AnMBR	0.3 – 0.5 <sup>e,f</sup>	0.3 – 0.4 <sup>e,f</sup>	-0.1 – 0.1	>80-90 <sup>i</sup>	<1 <sup>h,i</sup>
G-AnMBR	0.12 <sup>j</sup>	0.70 (0.89*) <sup>j</sup>	0.58 (0.77*)	92 <sup>j</sup>	<1 <sup>j</sup>
				>90 <sup>i</sup>	

607 a. (Su et al., 2019) ; b. (Roccaro and Vagliasindi, 2020) ; c. (Liao et al., 2006) ; d. (Lobato, Chernicharo. et  
 608 Souza 2012) ; e. (Kong et al., 2021a) ; f. (Smith et al., 2014) ; g. (van Lier et al., 2008) ; h. (Chernicharo et al.,  
 609 2015) ; i. (Chen et al., 2020) ; j. This study.

610 (\_\*) Those values in brackets take into account the complete dissolved methane recovery.  
 611 MBR values are for submerged membrane configuration.



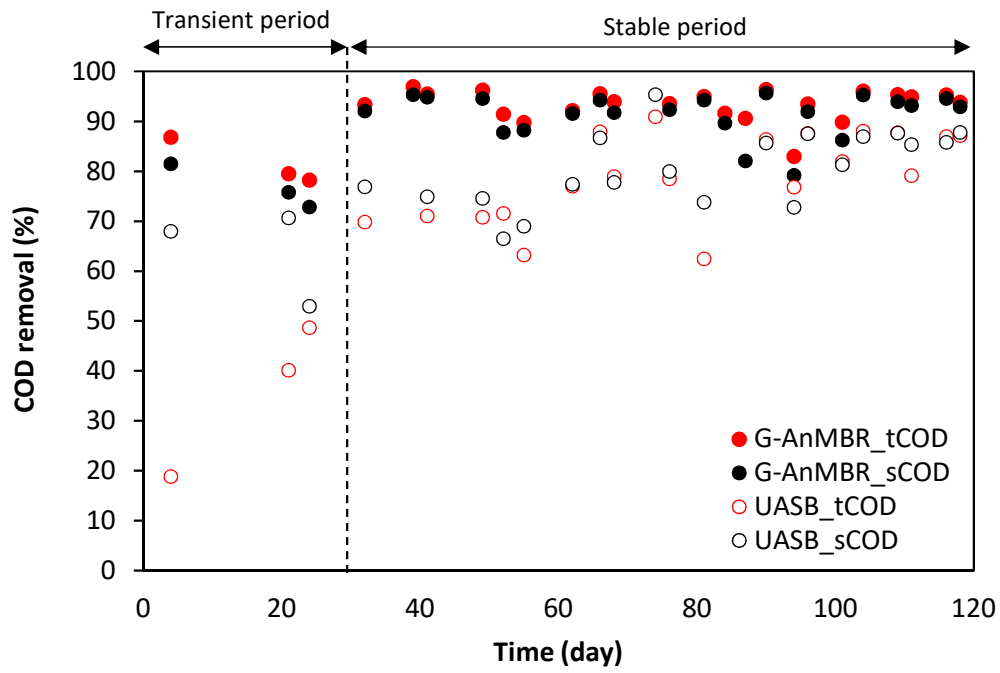


Fig. 1 - Total and soluble COD removal efficiencies in the UASB effluent and AnMBR permeate during the 120 days of operation.

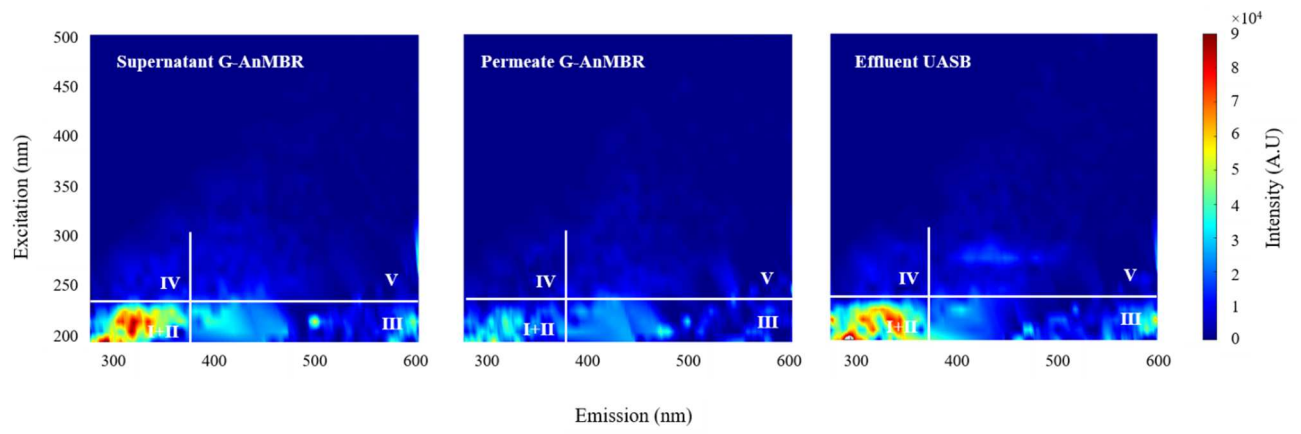


Fig. 2 - 3DEEM fluorescence spectra of G-AnMBR supernatant and permeate and UASB effluent on day 94

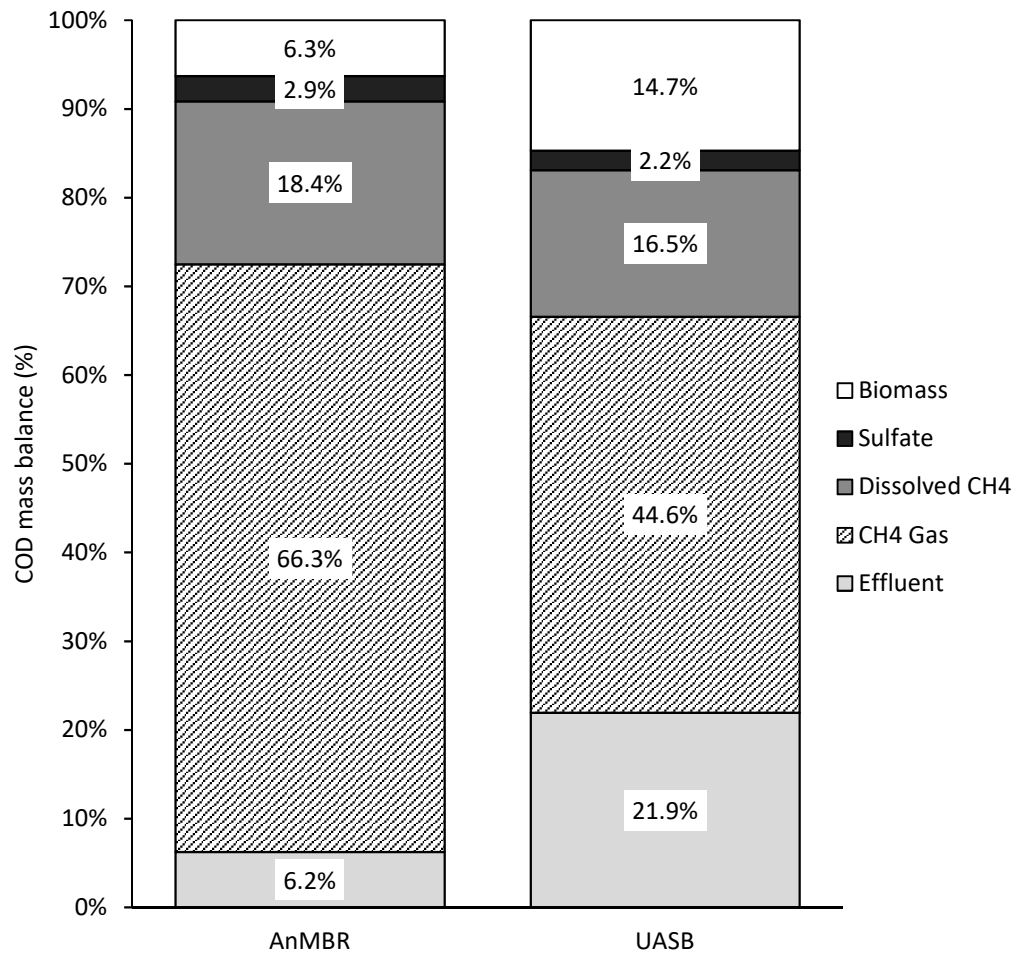
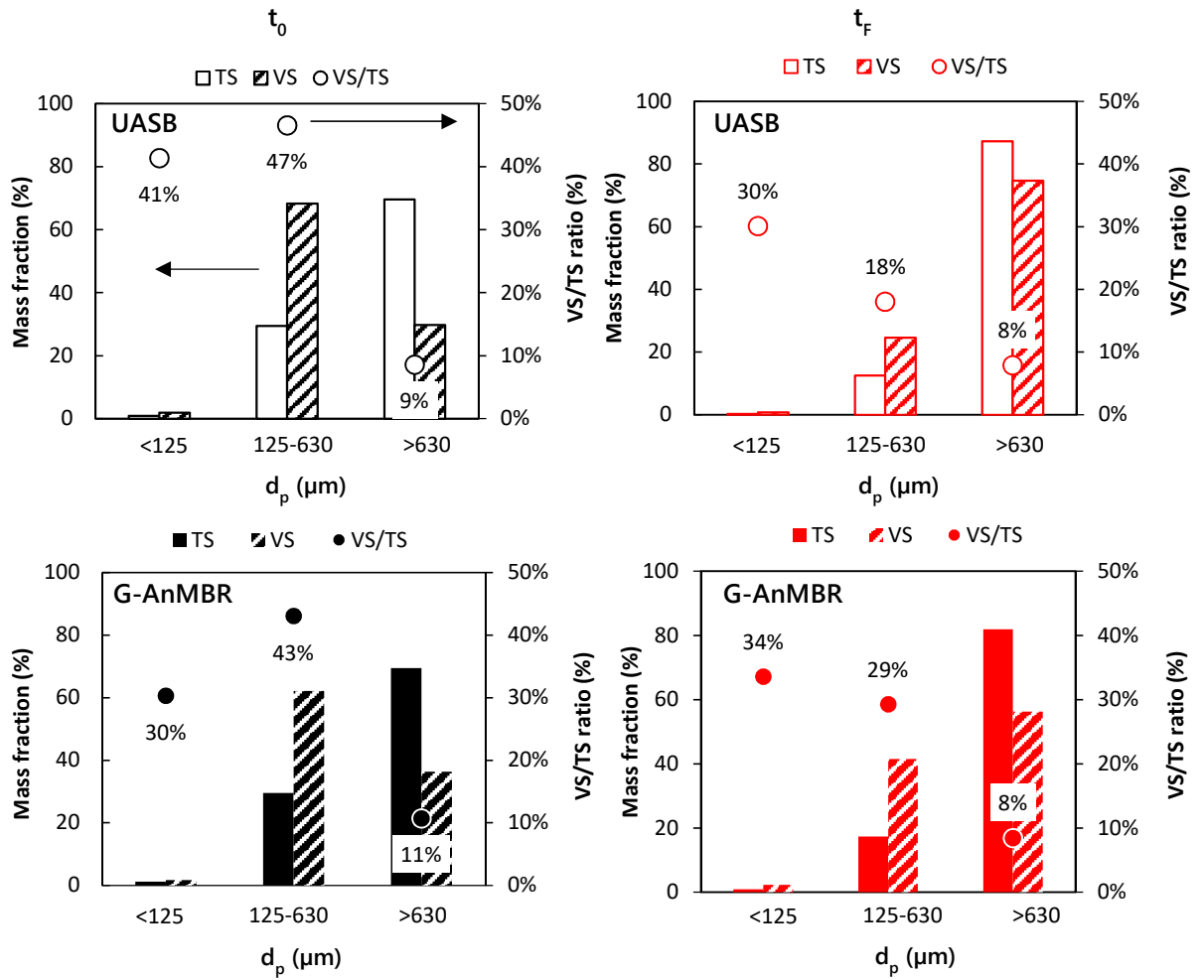


Fig. 3 - Average COD mass balance in the UASB and G-AnMBR reactors during the steady-state period (from day 30 to 120).



1

2 Fig. 4- Particle size distribution of the UASB and G-AnMBR granular sludge at the beginning (t<sub>0</sub> = day 1)  
 3 and at the end (t<sub>F</sub> = day 115) of the experiment.

