1	Life Cycle Assessment of high rate algal ponds for wastewater
2	treatment and resource recovery
3	
4	Larissa Terumi Arashiro <sup>a,b</sup> , Neus Montero <sup>a</sup> , Ivet Ferrer <sup>a</sup> , Francisco Gabriel Acién <sup>c</sup> ,
5	Cintia Gómez <sup>c</sup> , Marianna Garfí <sup>a,*</sup>
6	
7	<sup>a</sup> GEMMA – Group of Environmental Engineering and Microbiology, Department of
8	Civil and Environmental Engineering, Universitat Politècnica de
9	Catalunya BarcelonaTech, c/ Jordi Girona 1-3, Building D1, E-08034 Barcelona, Spain.
10	
11	<sup>b</sup> Department of Industrial Biological Sciences, Ghent University, Graaf Karel de
12	Goedelaan 5, 8500 Kortrijk, Belgium
13	
14	<sup>c</sup> Department of Chemical Engineering, University of Almería, 04120 Almería, Spain
15	
16	* Corresponding author:
17	Tel: +34 9340 16412
18	Fax: +34 93 4017357
19	Email: marianna.garfi@upc.edu
20	
21	Arashiro, L.T., Montero, N., Ferrer, I., Acién, F.G., Gómez, C., Garfí, M.* (2018)
22	Life Cycle Assessment of high rate algal ponds for wastewater treatment and
23	resource recovery. Science of the Total Environment, 622–623 (1118–1130)
24	

### 25 Abstract

The aim of this study was to assess the potential environmental impacts associated with 26 27 high rate algal ponds (HRAP) systems for wastewater treatment and resource recovery in small communities. To this aim, a Life Cycle Assessment (LCA) was carried out 28 evaluating two alternatives: i) a HRAP system for wastewater treatment where 29 microalgal biomass is valorized for energy recovery (biogas production); ii) a HRAP 30 system for wastewater treatment where microalgal biomass is reused for nutrients 31 32 recovery (biofertilizer production). Additionally, both alternatives were compared to a typical small-sized activated sludge system. An economic assessment was also 33 performed. The results showed that HRAP system coupled with biogas production 34 35 appeared to be more environmentally friendly than HRAP system coupled with biofertilizer production in the climate change, ozone layer depletion, photochemical 36 oxidant formation, and fossil depletion impact categories. Different climatic conditions 37 have strongly influenced the results obtained in the eutrophication and metal depletion 38 impact categories. In fact, the HRAP system located where warm temperatures and high 39 40 solar radiation are predominant (HRAP system coupled with biofertilizer production) showed lower impact in those categories. Additionally, the characteristics (e.g. nutrients 41 42 and heavy metals concentration) of microalgal biomass recovered from wastewater 43 appeared to be crucial when assessing the potential environmental impacts in the terrestrial acidification, particulate matter formation and toxicity impact categories. In 44 terms of costs, HRAP systems seemed to be more economically feasible when 45 46 combined with biofertilizer production instead of biogas. On the whole, implementing HRAPs instead of activated sludge systems might increase sustainability and cost-47

- 48 effectiveness of wastewater treatment in small communities, especially if implemented
- 49 in warm climate regions and coupled with biofertilizer production.
- 50
- 51 Keywords: Biogas; Environmental impact assessment; Fertilizer; Life Cycle
- 52 Assessment; Microalgae; Resource recovery

# 53 **1. Introduction**

High rate algal ponds (HRAPs) for wastewater treatment were introduced around 50 54 55 years ago and used since then not only to grow microalgae biomass but also to treat a wide variety of municipal and industrial wastewaters (Cragg et al., 2014; Oswald and 56 Golueke, 1960). These systems are shallow, paddlewheel mixed, raceway ponds where 57 microalgae assimilate nutrients and produce oxygen, which is used by heterotrophic 58 bacteria to oxidise organic matter improving water quality (Craggs et al., 2014; Park et 59 60 al., 2011). Since mechanical aeration is not required, energy consumption in these systems is much lower compared to a conventional wastewater treatment plant (e.g. 61 activated sludge system) (around 0.02 kWh m<sup>-3</sup> of water vs. 1 kWh m<sup>-3</sup> of water, 62 63 respectively) (Garfí et al., 2017; Passos et al., 2017). Moreover, HRAPs are less expensive and require little maintenance compared to conventional systems (Cragg et 64 al., 2014; Garfí et al., 2017; Molinos-Senante et al., 2014). Due to their low cost and 65 low energy consumption, HRAP systems could have a wide range of applications in 66 Mediterranean regions, which present suitable climatic conditions for microalgae 67 growth (e.g. high solar radiation). However, to achieve a satisfactory performance, large 68 land area is required compared to conventional systems (around 6 m<sup>2</sup> p.e.<sup>-1</sup> vs. 0.5 m<sup>2</sup> 69 p.e.<sup>-1</sup> for HRAP and activated sludge systems, respectively), making them more suitable 70 71 for small communities (up to 10,000 p.e.).

Nowadays, there is an important need to shift the paradigm from wastewater treatment to resource recovery to alleviate negative effects associated with human activities, such as pollution of water bodies, greenhouse gas (GHG) emissions and scarcity of mineral resources. In this context, microalgae grown in HRAPs can be harvested and reused to produce biofuels or other non-food bioproducts. In particular, 77 intensive research has been developed during the last years to investigate the potential of microalgae to produce biofuels such as biogas. Indeed, the biogas produced from 78 79 microalgal biomass was found to contain high energy value, making microalgae anaerobic digestion an attractive alternative for biofuel production (Chew et al., 2017; 80 Jankowska et al., 2017; Montingelli et al., 2015; Uggetti et al., 2017). On the other 81 82 hand, microalgae also offer the potential to recover nutrients from wastewater and, subsequently, to be applied as a sustainable fertilizer. During the last decade, this 83 84 alternative has been described by several authors, considering the fact that microalgae contain high amounts of proteins rich in essential amino acids, as well as 85 phytohormones that stimulate plant growth (Coppens et al., 2016; García-Gonzalez and 86 87 Sommerfeld, 2016; Jäger et al., 2010; Uysal et al., 2015).

Recent studies have employed the Life Cycle Assessment (LCA) methodology 88 to assess the environmental impact of HRAP systems for wastewater treatment. They 89 demonstrated that HRAPs might help to reduce environmental impacts and costs 90 associated with wastewater treatment compared to conventional systems (e.g. activated 91 sludge system), especially in small communities (Garfí et al., 2017; Maga, 2016). These 92 93 studies also highlighted that the LCA methodology is an appropriate tool to support early-stage research and development of novel technologies and processes (Fang et al., 94 2016; Garfí et al., 2017). Indeed, LCA methodology takes into account and quantifies 95 all environmental exchanges (i.e. resources, energy, emissions, waste) occurring during 96 all stages of the technology life cycle (Ferreira et al., 2014; Ferreira et al., 2017; ISO, 97 98 2000).

99 Nevertheless, to the best of the authors' knowledge, there are no studies
100 assessing the environmental impacts of HRAP system for wastewater treatment
101 considering different configurations for resource and energy recovery.

102 The objective of this work was to evaluate the potential environmental impacts associated with HRAP systems for wastewater treatment taking into account two 103 resource recovery strategies. To this aim a LCA was carried out comparing the 104 105 following alternatives: (i) a HRAP system for wastewater treatment where microalgal biomass is valorised for energy recovery (biogas production); (ii) a HRAP system for 106 107 wastewater treatment where microalgal biomass is reused for nutrients recovery (biofertilizer production). For the sake of comparison, both scenarios were compared to 108 a typical small-sized activated sludge system. Additionally, an economic evaluation was 109 110 addressed in order to assess the feasibility of the HRAP alternatives based on the costs and benefits related to each of them. 111

This paper is organized as follows: Section 2 describes the wastewater treatment systems, as well as the methodology used for the LCA and the economic analysis; in Section 3 the results of the comparative LCA and the economic analysis are described; finally, in Section 4 the main conclusions are highlighted.

116

# 117 **2. Material and Methods**

# 118 2.1 Wastewater treatment systems description

119 The HRAP systems were hypothetical wastewater treatment plants based on 120 extrapolation from lab-scale and pilot-scale studies (up to  $100 \text{ m}^2$ ). The systems were 121 designed to serve a population equivalent of 10,000 p.e. and treat a flow rate of 1,950

 $m^3 d^{-1}$ . The HRAP system coupled with biogas production was considered to be 122 123 implemented in Catalonia (Barcelona, Spain), where the mean temperature and global solar radiation are 15.5°C and 4.56 kWh/m<sup>2</sup>d, respectively (AEMET, 2017). For this 124 case study, the design parameters were calculated taking into account the experimental 125 results obtained in lab-scale and pilot systems (up to 5  $m^2$ ) located at the Universitat 126 Politècnica de Catalunya-BarcelonaTech (UPC) (Barcelona, Spain) (García et al., 2000; 127 García et al., 2006; Gutiérrez et al., 2016; Passos and Ferrer, 2014, Solé-Bundó et al., 128 129 2015; Solé-Bundó et al., 2017). This system comprises a primary settler (Hydraulic Retention Time (HRT) = 2.5 h) followed by four HRAPs (Table 1). From these units, 130 131 wastewater goes through a secondary settler (HRT = 3 h) where microalgal biomass is 132 harvested and separated from wastewater. Treated water is then discharged into a surface water body. Part of the harvested microalgal biomass (2 and 10 % on a dry 133 weight basis in summer and winter, respectively) is recycled in order to enhance 134 spontaneous flocculation (bioflocculation) and increase microalgae harvesting 135 efficiency (Gutiérrez et al., 2016). The remaining harvested biomass is thickened (HRT 136 137 = 24 h), thermally pretreated (75 °C, 10 h) and co-digested with primary sludge (35 °C, 20 days). The biogas produced is then converted in a combined heat and power (CHP) 138 unit, while the digestate is transported and reused in agriculture. In this context, the 139 140 HRT of each HRAP has to be modified over the year (8, 6 and 4 days) in accordance with the weather conditions (i.e. solar radiation and temperature) in order to accomplish 141 wastewater treatment and meet effluent quality requirements for discharge (García et al., 142 143 2000; Gutiérrez et al., 2016). For this reason, it was considered that during summer months (from May to July) only two HRAPs work in parallel (HRT = 4 days), whereas 144 all of them are operated during winter months (from November to April) (HRT = 8145

146 days). During the rest of the year (from August to October), the HRT is 6 days (3147 HRAPs working in parallel).

148 The HRAP system coupled with biofertilizer production was considered to be implemented in Andalucía (Almeria, Spain), where the mean temperature and global 149 solar radiation are 19.1°C and 5.29 kWh/m<sup>2</sup>d, respectively (AEMET, 2017). For this 150 case study, the designed parameters were determined using the results obtained in a pilot 151 system located at the Las Palmerillas Expertimental Station (Almeria, Spain) (100 m<sup>2</sup>) 152 153 (Morales-Amaral et al., 2015a). This system consists of two HRAPs operating in parallel and followed by a settler (HRT = 3 h) where microalgal biomass is separated 154 using an organic flocculant (Table 2). From this unit, treated wastewater is discharged 155 156 into a surface water body, while harvested microalgae biomass is dewatered on-site using a centrifuge and later sold to a local company to produce a biofertilizer (NPK = 5-157 158 1-0.75). The biofertilizer produced from the dewatered biomass is then transported and reused in agriculture. In this case, due to the more favourable climatic conditions for 159 microalgae growth compared to Catalonia, the HRT was the same over the year (HRT = 160 161 3 days). It has to be noted that, for the same reason, the microalgal biomass production is considerably higher in the system implemented in Andalucía with respect to the one 162 located in Catalonia (3-26 g<sub>TSS</sub> m<sup>-2</sup> d<sup>-1</sup> vs. 15-30 g<sub>TSS</sub> m<sup>-2</sup> d<sup>-1</sup>, respectively) (Gutiérrez et 163 164 al., 2016; Morales-Amaral et al., 2015a).

For the sake of comparison, the potential environmental impacts of the HRAP systems were compared to those generated by a conventional small-sized wastewater treatment plant (10,000 p.e.). For that purpose, the design of a usual small-scale activated sludge system implemented in Spain was taken into account (Gallego et al., 2008; Garfí et al., 2017; Lorenzo-Toja et al., 2015). It comprises a primary settler,

170 followed by an activated sludge reactor with extended aeration and a secondary settler 171 (Table 3). Treated water is discharged into the environment and the sludge is 172 conditioned, thickened, centrifuged on-site and then transported to an incineration facility. 173

Figure 1 shows the flow diagrams of the treatment alternatives. Table 1, 2 and 3 174 show the characteristics and design parameters of the HRAP and activated sludge 175 systems. 176

**Please insert Table 3** 

- **Please insert Figure 1** 177 **Please insert Table 1** 178 **Please insert Table 2** 179
- 180
- 181

#### 182 2.2 Life Cycle Assessment

The LCA was conducted following the ISO standards (ISO, 2000; ISO, 2006) in order 183 to evaluate and quantify the potential environmental impact of the investigated 184 scenarios. It consisted of four main stages: i) goal and scope definition, ii) inventory 185 analysis, iii) impacts assessment and iv) interpretation of the results (ISO, 2006). The 186 following sections describe the specific content of each phase. 187

188

#### 2.2.1 Goal and scope definition 189

The goal of this study was to determine the potential environmental impact of HRAP 190 191 systems for wastewater treatment and resource recovery. In particular, two configurations were compared: 192

193

a) a HRAP system for wastewater treatment where microalgal biomass is

valorised for energy recovery (biogas production) (Scenario 1);

b) a HRAP system for wastewater treatment where microalgal biomass is reused
for nutrients recovery (biofertilizer production) (Scenario 2).

197 Moreover, both scenarios were compared to a typical small-sized activated sludge 198 system implemented in Spain (Scenario 3). The functional unit (FU) for this study was 199 set as  $1 \text{ m}^3$  of treated water, since the main function of the technologies proposed is to 200 treat wastewater.

201 The cradle-to-grave boundaries included systems construction, operation and maintenance over a 20-years period (Garfí et al., 2017; Pérez-López et al., 2017; 202 203 Rahman et al., 2016) (Figure 1). Input and output flows of materials (i.e. construction 204 materials and chemicals) and energy resources (heat and electricity) were systematically studied for all scenarios. Direct GHG emissions and  $NH_4^+$  volatilization associated with 205 wastewater treatment were also included in the boundaries. As treated water is 206 discharged into the environment, direct emissions to water were also taken into account. 207 Regarding digestate and biofertilizer reuse in agriculture in Scenarios 1 and 2, 208 transportation (20 km) (Hospido et al., 2004) and direct emissions to soil (heavy 209 metals), as well as direct GHG emissions, were accounted for. In the case of the 210 activated sludge system (Scenario 3), inputs and outputs associated with sludge disposal 211 212 (i.e. incineration) were also included in the boundaries. An average distance of 30 km 213 was considered for sludge transportation to incineration facilities, based on circumstances generally observed in our zone. The end-of-life of infrastructures and 214 215 equipment were neglected, since the impact would be marginal compared to the overall impact. 216

217

Since the studied scenarios would generate by-products (i.e. biogas,

218 biofertilizer), the system expansion method has been used following the ISO guidelines 219 (Guinée, 2002; ISO, 2006). In this method, by-products are supposed to avoid the 220 production of conventional products. Thus, the impact related to conventional products is withdrawn from the overall impact of the system (Collet et al., 2011; ISO, 2006; Sfez 221 et al., 2015). In this study, the digestate and the biofertilizer produced in HRAP systems 222 223 coupled with biogas and biofertilizer production (Scenarios 1 and 2, respectively) were 224 considered as substitutes to chemical fertilizer. Moreover, the avoided burdens of using 225 heat and electricity produced in Scenario 1 (HRAP systems coupled with biogas production), instead of heat from natural gas and electricity supplied through the grid, 226 227 were also considered.

228

### 229 2.2.2 Inventory analysis

230 Inventory data for the investigated scenarios are summarized in Table 4, 5 and 6. In the case of HRAP systems coupled with biogas and biofertilizer production (Scenarios 1 231 and 2), inventory data regarding construction materials and operation were based on the 232 233 detailed engineering designs performed in the frame of this study. Treated wastewater characteristics were estimated considering the removal efficiencies and experimental 234 results obtained in the pilot systems implemented at the Universitat Politècnica de 235 Catalunya-BarcelonaTech (UPC) (5 m<sup>2</sup>) (Gutiérrez et al., 2016) and at the Las 236 Palmerillas Experimental Station (100 m<sup>2</sup>) (Morales-Amaral et al., 2015a) for Scenarios 237 1 and 2, respectively.  $NH_4^+$  volatilization was estimated through nitrogen mass balance. 238 239 NH<sub>3</sub> and N<sub>2</sub>O emissions due to the application of digestate and biofertilizer on agricultural land were calculated using emissions factors from the literature (Hospido et 240 al., 2008; IPCC, 2006; Lundin et al., 2000). In this case, CH<sub>4</sub> emissions were not 241

242 considered since anaerobic decompositions do not occur if liquid fertilizer is used and 243 the climate is predominantly dry (Hobson, 2003; Lundin et al., 2000). Heavy metals and 244 nutrients (avoided Total Nitrogen (TN) and Total Phosphorous (TP)) content of the 245 digestate and biofertilizer were gathered from experimental results obtained in the above-mentioned pilot systems (Morales-Amaral et al., 2015a; Solé-Bundó, et al., 246 2017). In order to estimate electricity and heat production from biogas cogeneration in 247 Scenario 1 (HRAP systems coupled with biogas production), biogas production 248 249 obtained in lab-scale experiments was taken into account (Solé-Bundó et al., 2015; Passos et al., 2017). 250

As mentioned above, data regarding the typical small-sized activated sludge system implemented in Spain (Scenario 3) were gathered from the literature (Gallego et al., 2008; Garfí et al., 2017; Lorenzo-Toja et al., 2015).

Background data (i.e. data of construction materials, chemicals, energy production, avoided fertilizer, transportation and sludge incineration process) were obtained from the *Ecoinvent 3.1* database (Moreno-Ruiz et al., 2014; Weidema et al., 2013). The Spanish electricity mix was used for all electricity requirements (Red Eléctrica Española, 2016).

259

260 Please insert Table 4

- 261 Please insert Table 5
- 262

263

264 2.2.3 Impact assessment

265 The LCA was performed using the software  $SimaPro^{\text{®}}$  8 (Pre-sustainability, 2014).

**Please insert Table 6** 

266 Potential environmental impacts were calculated by the ReCiPe midpoint method (hierarchist approach) (Goedkoop et al., 2009). In this study, characterisation phase was 267 268 performed considering the following impact categories: Climate Change, Ozone Depletion, Terrestrial Acidification, Freshwater Eutrophication, Marine Eutrophication, 269 Photochemical Oxidant Formation, Particulate Matter Formation, Metal Depletion, 270 271 Fossil Depletion, Human Toxicity and Terrestrial Ecotoxicity. These impact categories 272 were selected according to the most relevant environmental issues related to wastewater 273 treatment and used in previous LCA studies (Corominas et al., 2013; Fang et al., 2016; Gallego et al., 2008; Garfí et al., 2017; Hospido et al., 2008). Normalisation was carried 274 275 out in order to compare all the environmental impacts at the same scale. This provides 276 information on the relative significance of the indicator results, allowing a fair comparison between the impacts estimated for each scenario (ISO, 2006). In this study, 277 278 the European normalisation factors have been used (Europe ReCiPe H) (Goedkoop et al., 2009). 279

280

# 281 2.3. Sensitivity analysis

In order to evaluate the influence of the most relevant assumptions have on the results, a sensitivity analysis was performed considering the following parameters:  $NH_3$ emissions due to the application of digestate and biofertilizer on agricultural land (Scenario 1 and 2); N<sub>2</sub>O emissions due to the application of digestate and biofertilizer on agricultural land (Scenario 1 and 2); digestate and biofertilizer transportation distance (Scenario 1 and 2). A variation of  $\pm$  10% was considered for all parameters and the sensitivity coefficient was calculated using Eq. (1) (Dixon et al., 2003):

Sensitivity Coefficient (S) = 
$$\frac{(\text{Output}_{high} - \text{Output}_{low})/\text{Output}_{default}}{(\text{Input}_{high} - \text{Input}_{low})/\text{Input}_{default}}$$
(1) 13

where Input is the value of the input variable (e.g.  $NH_3$  and  $N_2O$  emissions) and Output is the value of the environmental indicator (e.g. Climate Change).

293

# 294 *2.4 Seasonality*

Annual averages of potential environmental impacts from HRAPs scenarios (Scenario 1 295 296 and 2) were compared to those obtained considering the microalgal biomass production achieved in summer and winter months (highest and lowest production, respectively; 297 298 Table 1 and 2) to assess their fluctuations over the year. In particular, the microalgal biomass production considered for Scenario 1 (HRAP systems coupled with biogas 299 production) was 5 and 25  $g_{TSS}$  m<sup>-2</sup> d<sup>-1</sup> for winter and summer months, respectively. On 300 301 the other hand, for Scenario 2 (HRAP systems coupled with biofertilizer production) a microalgal biomass production of 15 and 30  $g_{TSS}$  m<sup>-2</sup> d<sup>-1</sup> was considered for winter and 302 303 summer months, respectively.

304

# 305 2.5 Economic assessment

The economic assessment was performed comparing the capital cost and the operation 306 and maintenance cost of Scenarios 1 and 2 (HRAP systems coupled with biogas and 307 biofertilizer production, respectively). The capital cost included the cost for 308 309 earthmoving and construction materials purchase. On the other hand, operation and 310 maintenance cost comprised costs associated with energy (electricity and heat) consumption and chemicals purchase. In both scenarios, prices were provided by local 311 312 companies. For Scenario 1 (HRAP systems coupled with biogas production), the surplus electricity generated from biogas cogeneration was supposed to be sold back to the grid. 313

Thus, the price of electricity sold to the grid was withdrawn from the overall operational and maintenance cost of the system. For Scenario 2 (HRAP systems coupled with biofertilizer production), the dewatered microalgae biomass is sold to a local company (BIORIZON BIOTECH S.L.) to produce the biofertilizer (Romero-García et al., 2012). Therefore, its price was withdrawn from the overall operational and maintenance cost of the system. Other costs (e.g. labour costs, transportation) were assumed to be similar in both scenarios and, thus, were not included in the analysis.

321

# 322 **3. Results and Discussion**

# 323 3.1 Life Cycle Assessment

# 324 3.1.1 Characterization

The potential environmental impacts associated with each alternative are shown in 325 Figure 2. Comparing HRAP scenarios (Scenarios 1 and 2), the results show that 326 Scenario 2 is the most environmentally friendly alternative in 7 out of 11 impact 327 categories. As far as Climate Change, Ozone Depletion, Photochemical Oxidant 328 329 Formation and Fossil Depletion Potentials are concerned, the potential environmental impact of Scenario 1 was lower than Scenario 2. This was mainly due to the offset 330 energy generated from biogas cogeneration and the avoided fertilizer (Figure 2). In 331 332 particular, the electricity generated by biogas cogeneration (avoided electricity) was 333 around 9 times higher than that consumed for system operation in Scenario 1 (Table 4). It means that the surplus electricity could be sold to the grid. This is in accordance with 334 335 previous studies that observed that, in a HRAP system for wastewater treatment, the energy balance is always positive when microalgal biomass is co-digested with primary 336 sludge and the biogas is used to cogenerate electricity and heat (Passos et al., 2017). 337

338 Moreover, it has to be noticed that the contribution of the avoided fertilizer to the overall impact was higher in Scenario 1 than Scenario 2 (Figure 2), since TN avoided 339 was higher in the former compared to the latter (25.9 vs. 5.77 g m<sup>-3</sup> of water; Table 4 340 and 5). This can be explained by the fact that, despite TN content was higher in the 341 biofertilizer (5 g<sub>TN</sub> kg<sub>biofertilizer</sub><sup>-1</sup>) than in the digestate (1.89 g<sub>TN</sub> kg<sub>digestate</sub><sup>-1</sup>), a lower 342 amount of biofertilizer is produced in Scenario 2 (1.15 kgbiofertilizer m<sup>-3</sup> of water) 343 compared to Scenario 1 (13.7 kg<sub>digestate</sub> m<sup>-3</sup> of water). Indeed, the total solids (TS) 344 345 content of the microalgal biomass obtained in Scenario 1 (2% TS) is lower compared to Scenario 2 (20%TS) due to its dewatering step (i.e. centrifugation). Nevertheless, it has 346 to be mentioned that the biofertilizer is a higher quality product compared to the 347 348 digestate, since it contains high amounts of proteins rich in essential amino acids, as well as phytohormones that stimulate plant growth and improve soil quality (Coppens et 349 al., 2016; García-Gonzalez and Sommerfeld, 2016; Jäger et al., 2010; Uysal et al., 350 2015). However, these benefits were not taken into account in this study. Regarding 351 Terrestrial Acidification and Particulate Matter Formation Potentials, Scenario 2 showed 352 353 lower risks to endanger the environment because this configuration causes fewer emissions to air (i.e. NH<sub>3</sub> emissions) derived from biofertilizer application to 354 agricultural soil compared to digestate from Scenario 1 (Table 4 and 5). With regards to 355 Freshwater and Marine Eutrophication Potentials, Scenario 1 showed higher 356 environmental impacts compared to Scenario 2. It is explained by the quality of treated 357 effluent (i.e. lower TN and TP removal efficiencies in Scenario 1 than in Scenario 2; 358 359 Table 4 and 5). The reason for this difference could be primarily due to the distinct climatic conditions, since the average temperature and global solar radiation in 360 Catalonia (Scenario 1), as previously mentioned, are lower than in Andalucía (Scenario 361

362 2). Indeed, previous studies reported that nutrient removal efficiencies are improved with higher temperature and solar radiation (Craggs et al., 2012; Mehrabadi et al., 363 364 2016). Concerning Metal Depletion Potential, Scenario 1 would impair abiotic resources more likely than Scenario 2. Since Metal Depletion Potential is mainly 365 influenced by construction materials, the lower environmental performance of Scenario 366 367 1 is owing to the larger surface area required for its implementation compared to Scenario 2 (4 m<sup>2</sup> p.e.<sup>-1</sup> vs. 3 m<sup>2</sup> p.e.<sup>-1</sup>, respectively). As mentioned above, in the system 368 369 implemented in Catalonia (Scenario 1), a higher HRT is needed (especially during 370 winter months) compared to that implemented in Andalucía (Scenario 2) in order to obtain a effluent quality suitable for discharge (García et al., 2000; Gutiérrez et al., 371 372 2016, Morales-Amaral et al. 2015a; Morales-Amaral et al. 2015b). The influence of the geographical location on the performance of HRAPs was also addressed in previous 373 374 studies, in which the use of this technology is not encouraged in northern regions, where the climatic conditions are not favourable to promote efficient wastewater treatment and 375 biomass productivity (Grönlund and Fröling, 2014; Pérez-López et al., 2017). 376 377 According to this, it is noteworthy to mention that, since in this study the two HRAP systems (Scenarios 1 and 2) were assumed to be implemented in locations with distinct 378 379 climatic conditions, it is not possible to define the best biomass valorisation strategy 380 (i.e. biogas vs. biofertilizer production). In fact, HRAP systems operating under similar conditions should be considered in order to enable a better comparison. In regard to 381 Human toxicity and Terrestrial Ecotoxicity Potentials, Scenario 1 showed higher 382 383 environmental impacts compared to Scenario 2 due to the higher concentration of heavy metals in the digestate than in the biofertilizer (Table 4 and 5). 384

385

According to the results presented in Figure 2, Scenarios 1 and 2 showed lower

386 environmental impacts in 6 out of 11 impact categories (i.e. Climate Change, Ozone Depletion, Freshwater and Marine Eutrophication, Photochemical Oxidant Formation, 387 Fossil Depletion) compared to Scenario 3. This was primarily due to the lower energy 388 consumption needed for system operation in HRAP scenarios (Scenario 1 and 2) than in 389 the activated sludge system (Scenario 3) (Table 4, 5 and 6). On the other hand, HRAP 390 scenarios (Scenario 1 and 2) showed lower environmental performance in Metal 391 392 Depletion category (Figure 2), since a higher amount of construction materials are 393 needed for their implementation compared to the activated sludge system (Scenario 3). Indeed, even if HRAP systems have low raw materials requirements for their operation, 394 a large amount of raw materials is needed for their construction. This fact could make 395 396 HRAP systems less favourable than conventional technologies (e.g. activated sludge systems) in the abiotic resources depletion impact categories. Nevertheless, this 397 drawback can be overcome by implementing HRAP systems in smaller agglomerations 398 than that considered in this study (e.g. around 2,000 p.e.) (Garfí et al., 2017). As far as 399 Terrestrial Acidification, Particulate Matter Formation, Human Toxicity and Terrestrial 400 401 Ecotoxicity Potentials are concerned, the potential environmental impacts of HRAPs scenarios (Scenario 1 and 2) were higher than that caused by the activated sludge 402 system (Scenario 3). It was mainly due to the  $NH_3$  air emissions derived from  $NH_4^+$ 403 404 volatilization in HRAPs and to the heavy metals content in the digestate/biofertilizer (emissions to soil). The results are consistent with previous studies that reported 405 increased toxicity in a comparative LCA by integrating a sidestream process into a 406 407 conventional wastewater treatment facility where microalgae are cultivated, harvested and then used for fertigation (Fang et al., 2016). Furthermore, it was observed that the 408 higher impacts on terrestrial environments are unavoidable in cases where sludge and 409

410 nutrients from wastewater are recycled and reused in agriculture (Tangsubkul et al., 2005). In order to address this issue, improved technologies to separate better heavy 411 412 metals from recycled sludge should be encouraged (Tangsubkul et al., 2005). In regard 413 to Freshwater Eutrophication Potential, the activated sludge system (Scenario 3) showed higher potential environmental impact compared to Scenario 2, but lower impact than 414 415 Scenario 1. This was because of the higher outlet Phosphorous concentration in Scenario 1 compared to the other scenarios, which might be related to the lower 416 417 nutrients removal efficiency caused by less favourable climatic conditions. Previous studies observed that eutrophication and toxicity impact categories were mainly affected 418 419 by water discharge emissions and sludge management, indicating that the best 420 alternatives seem to be the ones that provide lower nutrients and heavy metals emissions 421 (Corominas et al., 2013). This corroborates with the results obtained with this study, 422 where the configuration with higher nutrients concentration in the effluent and higher levels of heavy metals in the recycled biomass (Scenario 1) showed higher impacts in 423 those categories. 424

425 On the whole, HRAP systems coupled with biogas and biofertilizer production (Scenario 1 and 2) showed similar environmental performance if compared to the 426 427 activated sludge system (Scenario 3). In particular, HRAPs environmental performance 428 is better than the conventional system in the climate change, ozone layer depletion, 429 photochemical oxidant formation, and fossil depletion impact categories. It was in accordance with previous studies, which stated that, compared to a typical medium-430 431 sized conventional wastewater treatment plant, a HRAP system coupled with biogas production could offer clear benefits with regard to the protection of climate, protection 432 of fossil resources and ozone depletion (Maga, 2016). In order to reduce the 433

434	environmental impacts of HRAP systems for wastewater treatment and resource
435	recovery, the following improvements should be addressed and further assessed: i)
436	reducing $NH_4^+$ volatilization in HRAPs by controlling the pH through CO <sub>2</sub> injection; ii)
437	ensuring higher nutrients removal efficiencies by selecting a favourable geographical
438	location to implement the HRAP systems; iii) studying improved technologies to
439	separate heavy metals from recycled microalgal biomass; iv) improving HRAP design
440	in order to decrease the amount of construction materials used (e.g. excavation instead
441	of concrete structure).
442	
443	Please insert Figure 2
444	
445	3.1.2 Normalization
446	The normalised results show that Freshwater Eutrophication, Marine Eutrophication,
447	Terrestrial Acidification and Human Toxicity Potentials are the most significant impact
448	categories for all the scenarios considered (Figure 3). These results are in accordance
449	with previous LCAs on wastewater treatment (Fang et al., 2016; Gallego et al, 2008;
450	Hospido et al., 2004). In these impact categories, Scenario 2 showed to be the most
451	environmentally friendly alternative.
452	
453	Please insert Figure 3
454	
455	3.2 Sensitivity analysis
456	The results of the sensitivity analysis are shown in Table 7, where the most sensitive
457	inventory components are indicated by bold type.

The results showed that Terrestrial Acidification and Particulate Matter Formation Potentials are somewhat sensitive to  $NH_3$  emissions due to the application of digestate on agricultural land in Scenario 1 (sensitivity coefficient around 0.3 for both environmental indicators). Indeed, a 10% increase of this parameter would increase these indicators by around 3%.

463 Similarly, Climate Change Potential showed to be somewhat sensitive to  $N_2O$ 464 emissions due to the application of digestate on agricultural land in Scenario 1 465 (sensitivity coefficient = 0.36). This means that a 10% increase in  $N_2O$  direct emissions 466 would increase this environmental indicator by 3.6%.

Moreover, Photochemical Oxidant Formation Potential showed to be sensitive to digestate transportation distance in Scenario 1 (sensitivity coefficient = 2.7). Indeed, a 10% increase in digestate transportation distance would increase this environmental indicator by 27%. The transport of the sludge to agricultural applications is not a fixed parameter, as it depends on specific needs. However, the sludge is usually applied in soil relatively close to the plant location (Pasqualino et al., 2009).

In conclusion, the results were found to be sensitive to digestate transportation distance in Scenario 1. Nevertheless, since it mainly affect only one of the less significant impact categories considered (i.e. Photochemical Oxidant Formation Potential), it can be concluded that the main findings of this study are not strongly dependent on the assumptions considered.

478

479

**Please insert Table 7** 

480

# 481 3.3 Seasonality

The seasonal variation of the potential environmental impact for HRAPs scenarios 482 (Scenario 1 and 2) are shown in Figure 4. The potential environmental impacts of 483 Scenario 2 are fairly constant over the year. On the contrary, a strong seasonal variation 484 was observed in Scenario 1. It was due to the fact that the microalgal biomass 485 production range in Scenario 1 (5-25  $g_{TSS}$  m<sup>-2</sup> d<sup>-1</sup>) is lower than Scenario 2 (15-30  $g_{TSS}$ 486  $m^{-2} d^{-1}$ ) and represents a high variation due to the seasonal fluctuations. It was in 487 488 accordance with previous studies, which reported that meteorological conditions played a critical role in the LCA results of HRAPs for microalgal cultivation (Pérez-López et 489 490 al., 2017). The authors highlighted that HRAPs are more suitable for locations where 491 warm temperatures and high solar radiation are predominant (Pérez-López et al., 2017). Moreover, electricity and flocculants consumption, as well as water and biofertilizer 492 characteristics, are fairly constant over the year in Scenario 2, while the biogas 493 production and, consequently, the energy avoided, strongly depend on microalgal 494 biomass production. These facts have a great influence on the environmental impacts 495 496 seasonality in Scenario 1. As a result, Scenario 2 remained the most environmentally friendly alternative in 7 out of 11 impact categories compared to Scenario 1 over the 497 year. Similarly, HRAPs scenarios (Scenario 1 and 2) still showed lower potential 498 499 environmental impacts in 6 out of 11 impact categories compared to activated sludge system (Scenario 3) considering seasonal fluctuations. 500

- 501
- 502

### **Please insert Figure 4**

503

504 3.4 Economic assessment

505 Results of the economic analysis are shown in Table 8. With respect to capital costs, 506 Scenario 2 appeared as the less expensive alternative. It was due to its lower specific 507 area requirement and, thus, lower amount of purchased materials, compared to Scenario 1 (3 vs. 4 m<sup>2</sup> p.e.<sup>-1</sup>, respectively). Similar capital costs were found in previous studies 508 509 which carried out an economic analysis of HRAPs for wastewater treatment without any resource recovery strategies (Garfí et al., 2017, Molinos-Senante et al., 2014). In fact, in 510 this study the capital cost for ponds implementation was around 90% of the total capital 511 512 cost of the overall systems (i.e. primary settler, ponds, secondary settler, digesters). 513 Since the highest cost is due to ponds construction, implementing downstream units for 514 resource recovery strategies (e.g. digester) in a HRAP system for wastewater treatment 515 would slightly increase its capital costs. Regarding the operation costs, Scenario 2 showed to be the most expensive alternative, since this configuration requires higher 516 517 expenses for energy and flocculant purchase. Nevertheless, if the price of the coproducts (i.e. electricity sold back to the grid, microalgae biomass to produce the 518 biofertilizer) that the wastewater treatment plant could sell out are considered, Scenario 519 520 2 would be the most cost-effective alternative (Table 8). The results of the economic assessment are consistent with previous studies, which indicated that recycling valuable 521 522 compounds from microalgal biomass (such as nutrients and pigments) is likely to be 523 more economically feasible than producing biogas from it, due to the higher added value of the final products (Ruiz et al., 2016; Vulsteke et al., 2017). 524

525

526

### **Please insert Table 8**

527

528 **4.** Conclusions

529 In this study, the LCA methodology was a useful tool to identify the main 530 environmental bottlenecks to scale-up high rate algal pond (HRAP) systems for 531 wastewater treatment and resource recovery in small communities.

Results showed that HRAP system coupled with biogas production showed to be 532 more environmentally friendly than HRAP system coupled with biofertilizer production 533 in the climate change, ozone layer depletion, photochemical oxidant formation, and 534 535 fossil depletion impact categories. Different climatic conditions have strongly influenced the results obtained in the eutrophication and metal depletion impact 536 537 categories. In fact, the HRAP system located where warm temperatures and high solar 538 radiation are predominant (HRAP system coupled with biofertilizer production) showed lower impact in those categories due to its higher nutrients removal efficiencies and 539 540 lower hydraulic retention time (i.e. lower specific area requirement). The characteristics (e.g. total solids, nutrients and heavy metals concentration) of microalgal biomass 541 recovered from wastewater appeared to be crucial when assessing the potential 542 543 environmental impacts in the terrestrial acidification, particulate matter formation and toxicity impact categories. 544

Normalization identified Freshwater Eutrophication, Marine Eutrophication, Terrestrial Acidification and Human Toxicity as the most significant impact categories for all the scenarios considered. In these categories, HRAP system coupled with biofertilizer production and implemented in warm climate region showed to be the most environmentally friendly alternative.

550 Additionally, HRAP systems coupled with biogas and biofertilizer production 551 showed lower potential environmental impacts compared to an activated sludge system

in the climate change, ozone layer depletion, photochemical oxidant formation, andfossil depletion impact categories.

The environmental performance of HRAP technology for wastewater treatment and resource recovery in small communities might be improved by: i) reducing  $NH_4^+$ volatilization in HRAPs by controlling the pH through  $CO_2$  injection; ii) ensuring higher nutrients removal efficiencies by selecting a favourable geographical location to implement the HRAP systems; iii) studying improved technologies to separate heavy metals from recycled microalgal biomass; iv) improving HRAP design in order to decrease the amount of construction materials used.

In terms of costs, HRAP system coupled with biofertilizer production was the most cost-effective alternative, due to the higher added value of the biofertilizer compared to the energy obtained from biogas cogeneration.

In conclusion, HRAPs are sustainable and cost-effective technology for wastewater treatment in small communities, especially if implemented in warm climate regions and coupled with biofertilizer production. Their implementation and dissemination can help to support a shift towards resource recovery and a sustainable circular economy.

569

# 570 Acknowledgements

571 This research was funded by the Spanish Ministry of Economy and Competitiveness 572 (FOTOBIOGAS CTQ2014-57293-C3-3-R; EDARSOL CTQ2014-57293-C3-1-R) and 573 the European Union's Horizon 2020 research and innovation programme under the 574 Marie Skłodowska-Curie grant agreement No 676070. This communication reflects

- only the authors' view and the Research Executive Agency of the EU is not responsible
- 576 for any use that may be made of the information it contains.

### 579 **References**

- 580 AEMET (Agencia Estatal de Meteorología), 2017. Climatological data. Standard
- 581 values. Available from:
- 582 http://www.aemet.es/es/serviciosclimaticos/datosclimatologicos/valoresclimatol
  583 ogicos
- Chew, K.W., Yap, J.Y., Show, P.L., Suan, N.H., Juan, J.C. Ling, T.C., Lee, D.J.,
  Chang, J.S., 2017. Microalgae biorefinery: High value products perspectives.
  Bioresour. Technol. 229, 53–62.
- 587 Corominas, L.I., Foley, J., Guest, J.S., Hospido, A., Larsen, H.F., Morera, S., Shaw, A.,
- 588 2013. Life cycle assessment applied to wastewater treatment: State of the art.
  589 Water Res. 47(15), 5480-5492.
- Collet, P., Hélias, A., Lardon, L., Ras, M., Goy, R.A., Steyer, J.P., 2011. Life-cycle
  assessment of microalgae culture coupled to biogas production. Bioresour.
  Technol. 102, 207–214.
- Coppens, J., Grunert, O., Van Den Hende, S., Vanhoutte, I., Boon, N., Haesaert, G., De
  Gelder, L., 2016. The use of microalgae as a high-value organic slow-release
  fertilizer results in tomatoes with increased carotenoid and sugar levels. J. Appl.
  Phycol. 4, 2367–2377.
- 597 Craggs, R., Sutherland, D., Campbell, H., 2012. Hectare-scale demonstration of high
  598 rate algal ponds for enhanced wastewater treatment and biofuel production. J.
  599 Appl. Phycol. 24, 329-337.
- Craggs, R., Park, J., Heubeck, S., Sutherland, D., 2014. High rate algal pond systems
  for low-energy wastewater treatment, nutrient recovery and energy production.
  New Zealand J. Bot. 52(1), 60-73.

- Dixon, A., Simon, M., Burkitt, T., 2003. Assessing the environmental impact of two
  options for small-scale wastewater treatment: comparing a reedbed and an
  aerated biological filter using a life cycle approach. Ecol. Eng. 20(4), 297-308.
- Fang, L.L., Valverde-Pérez, B., Damgaard, A., Plósz, B.G., Rygaard, M., 2016. Life
  cycle assessment as development and decision support tool for wastewater
  resource recovery technology. Water Res. 88, 538-549.
- Ferreira, S., Cabral, M., da Cruz, N.F., Simões, P., Marques, R.C., 2014. Life cycle
  assessment of a packaging waste recycling system in Portugal. Waste
  Management. 34, 1725-1735.
- Ferreira, S., De Jaeger, S., Simões, P., Cabral, M., da Cruz, N., Marques, R.C., 2017.
  Life cycle assessment and valuation of the packaging waste recycling system in
  Belgium. J. Mater. Cycles Waste Manag. 19, 144-154.
- Gallego, A., Hospido, A., Moreira, M.T., Feijoo, G. 2008. Environmental performance
  of wastewater treatment plants for small populations. Resour. Conserv. Recycl.
  52(6), 931–940.
- García, J., Mujeriego, R., Hernandez-Marine, M., 2000. High rate algal pond operating
  stategies for urban wastewater nitrogen removal. J. Appl. Phycol. 12, 331–339.
- 620 García. J., Green, B.F.. Lundquist, T., Mujeriego, R., Hernández-Mariné, M., Oswald,
- W.J., 2006. Long term diurnal variations in contaminant removal in high rate
  ponds treating urban wastewater. Bioresour. Technol. 97, 1709–1715.
- 623 García-Gonzalez, J., Sommerfeld, M., 2016. Biofertilizer and biostimulant properties of
- the microalga Acutodesmus dimorphus. J. Appl. Phycol. 28, 1051–1061.

- Garfí, M., Flores, L., Ferrer, I., 2017. Life cycle assessment of wastewater treatment
  systems for small communities: activated sludge, constructed wetlands and high
  rate algal ponds. J. Clean. Prod. 161, 211-219.
- Goedkoop, M., Heijungs, R., Huijbregts, M., De Schryver, A., Struje, J., van Zelm, R.,
  2009. ReCiPe 2008. A life cycle impact assessment method which comprises
  harmonised category indicators at the midpoint and then endpoint level. Report
  I: Characterisation, first ed. Ministerie van Volkshuisvesting, Ruimtelijke
  Ordering en Milieubeheer, Netherlands.
- Grönlund, E., Fröling, M., 2014. Wastewater Treatment and Recycling with Microalgae
  in Cold Climate. In: Proceedings of the 20th International Sustainable
  Development Research Conference Trondheim 18-20 June 2014: Resilience –
  the new research frontier, 317-324. Tronheim: Norwegian University of Science
  and Technology, Department of Product Design.
- Guinée, J.B., 2002., Handbook on Life Cycle Assessment: Operational Guide to the
  ISO Standards, Springer, New York.
- Gutiérrez, R., Ferrer, I., González-Molina, A., Salvadó, H., García, J., Uggetti, E.,
  2016. Microalgae recycling improves biomass recovery from wastewater
  treatment high rate algal ponds. Water Res. 106, 539-549.
- Hobson, J., 2003. CH4 and N2O emissions from waste water handling. Good Practice
  Guidance and Uncertainty Management in National Greenhouse Gas
  Inventories. Available from: <u>http://www.ipcc-</u>
  nggip.iges.or.jp/public/gp/bgp/5\_2\_CH4\_N2O\_Waste\_Water.pdf

647	Hospido, A., Moreira, M.T., Fernández-Couto, M., Feijoo, G., 2004. Environmental
648	Performance of a Municipal Wastewater Treatment Plant. Int. J. LCA, 9(4),
649	261-271.

- Hospido, A., Moreira, M.T., Feijoo, G., 2008. A comparison of municipal wastewater
  treatment plants for big centres of population in Galicia (Spain). Int. J. LCA,
  13(1), 57-64.
- IPCC (Intergovernmental Panel on Climate Change) 2006., 2006 IPCC Guidelines for
  National Greenhouse Gas Inventories, Prepared by the National Greenhouse
  Gas Inventories Programme, Eggleston H.S., Buendia L., Miwa K., Ngara T.
  and Tanabe K. (eds). Published: IGES, Japan.
- ISO (International Organization for Standardization), 2000. Environmental
  management life cycle assessment life cycle impact assessment,
  International Standard ISO 14042, Geneva, Switzerland.
- ISO (International Organization for Standardization), 2006. Environmental
  management life cycle assessment principles and framework, International
  Standard ISO 14040, Geneva, Switzerland.
- Jäger, K., Bartók, T., Ördög, V., Barnabás, B., 2010. Improvement of maize (Zea mays
  L.) anther culture responses by algae-derived natural substances. South African
  J. Bot. 76, 511–516.

666 Jankowska, E., Sahu, A.K., Oleskowicz-Popiel, P., 2017. Biogas from microalgae:

Review on microalgae's cultivation, harvesting and pretreatment for anaerobic
digestion. Renew. Sust. En. Rev. 75, 692–709.

669	Lorenzo-Toja, Y., Vázquez-Rowe, I., Chenel, S., Marín-Navarro, D., Moreira, M.T.,
670	Feijoo, G., 2015. Eco-efficiency analysis of Spanish WWTPs using the LCA +
671	DEA method. Water Res. 68, 651-666.
672	Lundin, M., Bengtsson, M., Molander, S., 2000. Life Cycle Assessment of wastewater
673	systems: Influence of system boundaries and scale on calculated environmental
674	loads. Environ. Sci. Technol. 34(1), 180–186.
675	Maga, D., 2016. Life cycle assessment of biomethane produced from microalgae grown
676	in municipal waste water. Biomass Conv. Bioref. 7(1), 1-10.
677	Mehrabadi, A., Farid, M.M., Craggs, R., 2016. Variation of biomass energy yield in
678	wastewater treatment high rate algal ponds. Algal Res. 15, 143-151.
679	Molinos-Senante, M., Gómez, T., Garrido-Baserba, M., Caballero, R., Sala-Garrido, R.,
680	2014. Assessing the sustainability of small wastewater treatment systems: A
681	composite indicator approach. Sci. Total Environ. 497–498, 607–617.
682	Montingelli, M.E., Tedesco, S., Olabi, A.G., 2015. Biogas production from algal
683	biomass: A review. Renew. Sust. En. Rev. 43, 961-972.
684	Morales-Amaral, M.M., Gómez-Serrano, C., Acién, F.G., Fernández-Sevilla, J.M.,
685	Molina-Grima, E., 2015a. Outdoor production of Scenedesmus sp. in thin-layer
686	and raceway reactors using centrate from anaerobic digestion as the sole
687	nutrient source. Algal Res. 12, 99–108.
688	Morales-Amaral, M.M., Gómez-Serrano, C., Acién, F.G., Fernández-Sevilla J.M.,
689	2015b. Production of microalgae using centrate from anaerobic digestion as the
690	nutrient source. Algal Res. 9, 297–305.
691	Moreno-Ruiz, E., Lévová, T., Bourgault, G., Wernet, G., 2014. Documentation of
692	changes implemented in ecoinvent Data 3.1. Zurich: ecoinvent.

- 693 Oswald, W.J., Golueke, C.G., 1960. Biological transformation of solar energy., Adv.
  694 Appl. Microbiol. 2, 223–262.
- Pasqualino, J.C., Meneses, M., Abella, M., Castells, F. 2009. LCA and decision support
  tool for the environmental improvement of the operation of municipal
  wastewater treatment plant. Environ. Sci. Technol. 43, 3300-3307
- Passos, F., Ferrer, I., 2014. Microalgae conversion to biogas: Thermal pretreatment
  contribution on net energy production. Environ. Sci. Technol. 48(12), 7171700 7178.
- Passos, F., Gutiérrez, R., Uggetti, E., Garfí, M., García, J., Ferrer, I., 2017. Towards
  energy neutral microalgae-based wastewater treatment plants. Algal Res. 28,
  235–243.
- Park, J.B.K., Craggs, R.J., Shilton, A.N., 2011. Wastewater treatment high rate algal
  ponds for biofuel production. Bioresour. Technol. 102, 35–42.
- 706 Pérez-López, P., de Vree, J.H., Feijoo, G., Bosma, R., Barbosa, M., J., Moreira, M.T.,
- Wijffels, R.H., van Boxtel, A.J.B., Kleinegris, D.M.M., 2017. Comparative life
  cycle assessment of real pilot reactors for microalgae cultivation in different
  seasons. App. En. 205, 1151-1164.
- 710 PRé Sustainability, 2014. Available at: <u>https://www.pre-sustainability.com/simapro</u>
- 711 Rahman, S.M., Eckelman, M.J., Onnis-Hayden, A., Gu, A.Z., 2016. Life-Cycle
- 712 <u>Assessment of Advanced Nutrient Removal Technologies for Wastewater</u>
  713 Treatment. Environ. Sci. Technol. 50, 3020-3030.
- Red Eléctrica Española, 2016. The Spanish electricity system. Preliminary report 2016.
- 715 Available at: http://www.ree.es/en/statistical-data-of-spanish-electrical-
- 716 <u>system/annual-report</u>

717	Romero-García,	J.M.,	Acién-Fernández,	F.G.,	Fernández-Sevilla,	J.M.	2012
718	Developm	ent of a	a process for the pro-	duction	of l-amino-acids cond	centrate	es from
719	microalga	e by en	zymatic hydrolysis.	Bioreso	ur. Technol. 112,164-	-170.	
720	Ruiz, J., Olivieri,	G., De	Vree, J., Bosma, R.,	Willem	ns, P., Reith, J.H., Epp	pink, M	I.H.M.

- Kleinegris, D.M.M., Wijffels, R., Barbosa, M.J., 2016. Towards industrial
  products from microalgae. Energy Environ. Sci. 9, 3036-3043.
- Solé-Bundó, M., Passos, F., Garfí, M., Ferrer, I., 2015. Biogas potential from algalbased wastewater treatment systems: co-digestion of by-products, thermal
  pretreatment, energy balance and digestate characteristics. Proceedings of the
  14th World Congress on Anaerobic Digestion, AD2015, Viña del Mar, Chile
- Solé-Bundó, M., Cucina, M., Folch, M., Tapias, J., Gigliotti, G., Garfí, M., Ferrer, I.,
  2017. Assessing the agricultural reuse of the digestate from microalgae
  anaerobic digestion and co-digestion with sewage sludge. Sci. Total Environ.
  586, 1-9.
- Sfez, S., Van Den Hende, S., Taelman, S.E., De Meester, S., Dewulf, J., 2015.
  Environmental sustainability assessment of a microalgae raceway pond treating
  aquaculture wastewater: From up-scaling to system integration. Bioresour.
  Technol. 190, 321–331.
- Tangsubkul, N., Beavis, P., Moore, S.J., Lundie, S., Waite, T.D., 2005. Life cycle
  assessment of water recycling technology. Water Resour. Management 19, 521537.
- Uggetti, E., Passos, F., Solé, M., Garfí, M., Ferrer, I., 2017. Recent achievements in the
  production of biogas from microalgae. Waste Biomass Valorizat. 8(1), 129–139.

740	Uysal,	O.,	Uysal,	F.O.,	Ekinci,	К.,	2015.	Evaluation	of	Microalgae	as	Microbial
741	F	Fertil	lizer. Ei	uropea	n J. Sust	. De	velop.	4(2), 77-82.				

- Vulsteke, E., Van Den Hende, S., Bourez, L., Capoen, H., Rousseau, D.P.L., Albrecht,
  J., 2017. Economic feasibility of microalgal bacterial floc production for
  wastewater treatment and biomass valorization: A detailed up-to-date analysis
  of up-scaled pilot results. Bioresour, Technol. 224, 118-129.
- 746 Weidema B.P., Bauer C., Hischier R., Mutel C., Nemecek T., Reinhard J., Vadenbo C.
- 747 O., Wernet G., 2013. Overview and methodology. Data quality guideline for the
- ecoinvent database version 3. Ecoinvent Report 1 (v3). St. Gallen: Theecoinvent Centre.

# 751 Table 1. Characteristics and design parameters of the HRAP coupled with biogas production (Scenario

1)

System characteristics	Unit					
Inlet BOD <sub>5</sub> concentration	$mg_{BOD} L^{-1}$	300				
Outlet BOD <sub>5</sub> concentration	$mg_{BOD} L^{-1}$		<25			
Inlet TSS concentration	$mg_{TSS} L^{-1}$		150			
Outlet TSS concentration	$mg_{TSS}L^{-1}$		<35			
Inlet Total Nitrogen	$mg_{TN}L^{-1}$		39			
Outlet Total Nitrogen	$mg_{TN}L^{-1}$		9.38			
Inlet Total Phosphorous	$mg_{TP}L^{-1}$		5			
Outlet Total Phosphorous	$mg_{TP}L^{-1}$		3.69			
Flow rate	$m^3 d^{-1}$		1,950	)		
Population equivalent	<i>p.e.</i>	10,000				
Total surface area	$m^2$	40,000				
Specific area requirement	$m^2 p.e.^{-1}$	4				
HRAPs Design parameters	Unit	Summer         Winter         Rest of the y				
OLR	$g_{BOD} m^{-2} d^{-1}$		10			
HRT	d	4	8	6		
Number of ponds	-	2	4	3		
Channel width	т		12			
Channel length	т	812.5				
Water depth	т	0.4				
Microalgae biomass production	$g_{TSS} m^{-2} d^{-1}$	25.8 3.3 10.5				
Annual average microalgae biomass production	$g_{TSS} m^{-2} d^{-1}$		12	1		
Note: BOD: Biochemical oxygen demand; TSS:	Total suspende	ed solids; HF	RT: Hydrauli	ic Retention Time;		

OLR: Organic Loading Rate. Summer: from May to July; winter: from November to April.

#### Table 2. Characteristics and design parameters of the HRAP coupled with biofertilizer production

(Scenario 2)

System characteristics	Unit					
Inlet BOD <sub>5</sub> concentration	$mg_{BOD} L^{-1}$	300				
Outlet BOD <sub>5</sub> concentration	$mg_{BOD} L^{-1}$		<25			
Inlet TSS concentration	$mg_{TSS}L^{-1}$		200			
Outlet TSS concentration	$mg_{TSS}L^{-1}$		<35			
Inlet Total Nitrogen	$mg_{TN}L^{-1}$		50			
Outlet Total Nitrogen	$mg_{TN}L^{-1}$		2			
Inlet Total Phosphorous	$mg_{TP}L^{-1}$		10			
Outlet Total Phosphorous	$mg_{TP}L^{-1}$		1			
Flow rate	$m^3 d^{-1}$		1,950			
Population equivalent	<i>p.e.</i>	10,000				
Total surface area	$m^2$	30,000				
Specific area requirement	$m^2 p.e.^{-1}$	3				
HRAPs Design parameters	Unit	Summer Winter Rest of the y				
OLR	$g_{BOD} m^{-2} d^{-1}$	I	20			
HRT	d		3			
Number of ponds	-		2			
Channel width	т	12				
Channel length	m	1,219				
Water depth	m	0.2				
Microalgae biomass production	$g_{TSS} m^{-2} d^{-1}$	30 15 25				
Annual average microalgae biomass production	$g_{TSS} m^{-2} d^{-1}$	23				

Note: BOD: Biochemical oxygen demand; TSS: Total suspended solids; HRT: Hydraulic Retention Time;

OLR: Organic Loading Rate. Summer: from May to August; winter: from November to March

# **Table 3.** Characteristics and design parameters of the activated sludge system (Scenario 3)

System characteristics	Unit	
Inlet BOD <sub>5</sub> concentration	$mg_{BOD} L^{-1}$	300
Outlet BOD <sub>5</sub> concentration	$mg_{BOD} L^{-1}$	<25
Outlet TSS concentration	$mg_{TSS}L^{-1}$	<35
Flow rate	$m^3 d^{-1}$	1,950
Population equivalent	<i>p.e.</i>	10,000
Total surface area	$m^2$	900
Specific area requirement	$m^2 p.e.^{-1}$	0.6
Design parameters	Unit	
Primary settler HRT	h	2.5
Activated sludge reactor HRT	h	6
Secondary settler HRT	h	2

Note: BOD: Biochemical oxygen demand; TSS: Total suspended solids; HRT: Hydraulic Retention Time;

OLR: Organic Loading Rate.

771 Table 4. Summary of the inventory for Scenario 1: HRAP system for wastewater treatment where

772

773

functional unit (1 m<sup>3</sup> of water)

microalgal biomass is valorised for energy recovery (biogas production). Values are referred to the

Inputs	Scenario 1	Units
Construction materials		
Primary settler		
Concrete	2.55E-06	$m^3 m^{-3}$
Steel	2.04E-04	$kg m^{-3}$
HRAPs		
Concrete	5.94E-04	$m^3 m^{-3}$
Steel	4.76E-02	$kg m^{-3}$
Secondary settler		
Concrete	1.29E-05	$m^3 m^{-3}$
Steel	1.03E-03	$kg m^{-3}$
Thickener		
Concrete	1.78E-07	$m^3 m^{-3}$
Steel	1.42E-05	kg m <sup>-3</sup>
Thermal pretreatment		
Concrete	2.77E-07	$m^3 m^{-3}$
Steel	2.22E-05	$kg m^{-3}$
Digester		
Concrete	9.79E-06	$m^3 m^{-3}$
Steel	7.83E-04	$kg m^{-3}$
Operation		
Energy consumption*		
Primary settler	4.41E-03	$kWh m^{-3}$
HRAPs	1.13E-02	$kWh m^{-3}$
Secondary settler	2.52E-03	$kWh m^{-3}$
Thermal pretreatment	1.08E-04	$kWh m^{-3}$
Digester	4.17E-02	$kWh m^{-3}$
Total energy consumption	6.00E-02	$kWh m^{-3}$
Outputs		
Emissions to water*		
Total COD	7.63E+01	$g m^{-3}$
TSS	2.40E+01	$g m^{-3}$
TN	9.38E+00	$g m^{-3}$
TP	3.69E+00	$g m^{-3}$
Emissions to air*		-
<i>NH</i> <sub>4</sub> <sup>+</sup> volatilization in <i>HRAPs</i>		
NH <sub>3</sub>	3.80E+00	$g m^{-3}$
Digestate application as fertilizer		č
NH <sub>3</sub>	6.47E+00	$g m^{-3}$

N <sub>2</sub> O	2.59E-01	$g m^{-3}$
Emissions to soil*		U U
Digestate application as fertilizer		
Cd	3.53E-03	$g m^{-3}$
Cu	2.02E-01	$g m^{-3}$
Pb	9.08E-02	$g m^{-3}$
Zn	9.04E-01	$g m^{-3}$
Ni	4.15E-02	$g m^{-3}$
Cr	5.22E-02	$g m^{-3}$
Hg (value <)	4.52E-04	$g m^{-3}$
Avoided products*		
Electricity (from biogas cogeneration)	5.40E-01	$kWh m^{-3}$
Heat (from biogas cogeneration)	8.49E-01	$kWh m^{-3}$
N as Fertiliser (from digestate reuse)	2.59E+01	$g m^{-3}$
P as Fertiliser (from digestate reuse)	1.31E+00	$g m^{-3}$
nual averages		

778 Table 5. Summary of the inventory for Scenario 2: HRAP system for wastewater treatment where

microalgal biomass is reused for nutrients recovery (biofertilizer production). Values are referred to the

780

functional unit (1 m<sup>3</sup> of water)

Inputs	Scenario 2	Units
Construction materials		
HRAPs		
Concrete	4.32E-04	$m^3 m^{-3}$
Steel	3.45E-02	$kg m^{-3}$
Secondary settler		
Concrete	1.29E-05	$m^3 m^{-3}$
Steel	1.03E-03	$kg m^{-3}$
Centrifuge		
Steel	3.86E-05	$kg m^{-3}$
Operation		
Energy consumption*		
HRAPs	1.11E-02	$kWh m^{-3}$
Secondary settler	5.77E-03	$kWh m^{-3}$
Centrifuge	1.15E-02	$kWh m^{-3}$
Biofertilizer production	4.70E-02	$kWh m^{-3}$
Total energy consumption	7.54E-02	$kWh m^{-3}$
Chemicals*		
Organic flocculant	1.00E+01	$kg m^{-3}$
Outputs		
Emissions to water*		
Total COD	1.00E+02	$g m^{-3}$
TSS	5.00E+01	$g m^{-3}$
TN	2.00E+00	$g m^{-3}$
ТР	1.00E+00	$g m^{-3}$
Emissions to air*		
<i>NH</i> <sup>4+</sup> volatilization in <i>HRAPs</i>		
NH <sub>3</sub>	5.00E+00	$g m^{-3}$
Biofertilizer		
NH <sub>3</sub>	1.44E+00	$g m^{-3}$
N <sub>2</sub> O	5.77E-02	$g m^{-3}$
Emissions to soil*		
Biofertilizer		
Cd	3.46E-04	$g m^{-3}$
Cu	4.62E-02	$g m^{-3}$
Pb	2.31E-02	$g m^{-3}$
Zn	1.15E-02	$g m^{-3}$
Ni	1.15E-02	$g m^{-3}$

	Cr	3.46E-02	$g m^{-3}$
	Hg (value <)	2.31E-04	$g m^{-3}$
	Avoided products*		
	N as Fertiliser (from biofertilizer)	5.77E+00	$g m^{-3}$
	P as Fertiliser (from biofertilizer)	1.20E+00	$g m^{-3}$
781	* Annual averages		
782			

# Table 6. Summary of the inventory for Scenario 3: typical small-sized activated sludge system

implemented in Spain. Values are referred to the functional unit (1 m<sup>3</sup> of water)

Inputs	Scenario 3	Units
Construction materials		
Concrete	1.65E-05	$m^3 m^{-3}$
Steel	1.32E-03	$kg m^{-3}$
Operation		
Energy consumption		
Electricity	8.90E-01	$kWh m^{-3}$
Chemicals		
Polyelectrolyte	1.98E+00	$g m^{-3}$
Coagulant	3.18E+00	$g m^{-3}$
Outputs		
Emissions to water		
Total COD	1.25E+02	$g m^{-3}$
TSS	3.50E+01	$g m^{-3}$
TN	1.50E+01	$g m^{-3}$
TP	2.00E+00	$g m^{-3}$
Emissions to air		
$CO_2$	1.70E-01	$g m^{-3}$
N <sub>2</sub> O	1.10E-01	$g m^{-3}$
Waste to further treatment		
Sludge (incineration)	1.24E+00	kg m <sup>-3</sup>

# **Table 7**. Results of the sensitivity analysis for the considered parameters: NH<sub>3</sub> emissions due to the application of digestate and biofertilizer on agricultural land; N<sub>2</sub>O

7	q	n
	)	υ

emissions due to the application of digestate and biofertilizer on agricultural land; digestate and biofertilizer transportation distance.

	Parameters					
Impact categories	Scenario 1		Scenario 2			
	NH <sub>3</sub> emissions	N <sub>2</sub> O emissions	Digestate transportation	NH <sub>3</sub> emissions	N <sub>2</sub> O emissions	Biofertilizer transportation
Climate change	$\pm 0.000$	±0.367	±0.260	$\pm 0.000$	$\pm 0.068$	±0.015
Ozone Depletion	$\pm 0.000$	$\pm 0.000$	±0.204	$\pm 0.000$	$\pm 0.000$	±0.053
Terrestrial acidification	±0.337	±0.000	$\pm 0.008$	±0.213	$\pm 0.000$	±0.001
Freshwater eutrophication	±0.000	±0.000	±0.001	±0.000	±0.000	$\pm 0.000$
Marine eutrophication	±0.058	±0.000	±0.001	±0.052	±0.000	$\pm 0.000$
Photochemical oxidant formation	±0.000	±0.000	±2.713	±0.000	±0.000	±0.025
Particulate matter formation	±0.327	±0.000	±0.033	±0.179	±0.000	±0.003
Metal depletion	$\pm 0.000$	±0.000	±0.019	±0.000	$\pm 0.000$	$\pm 0.002$
Fossil depletion	±0.000	±0.000	±0.153	±0.000	±0.000	±0.027
Human toxicity	±0.000	±0.000	±0.021	±0.000	±0.000	±0.011
Terrestrial ecotoxicity	±0.000	±0.000	±0.019	±0.000	±0.000	±0.011

*N* 

Note: Scenario 1: HRAP system for wastewater treatment where microalgal biomass is valorized for energy recovery (biogas production); Scenario 2: HRAP system for wastewater treatment where microalgal biomass is reused for nutrients recovery (biofertilizer production)

# Table 8. Results of the economic analysis for the HRAPs scenarios.

	Unit	Scenario 1	Scenario 2
Capital cost	€ p.e. <sup>-1</sup>	192.55	139.34
Operation and maintenance cost (energy and flocculant consumption)	€ m <sup>3</sup> <sub>water</sub>	0.007	0.02
Price of electricity sold back to the grid	€ m <sup>3</sup> <sub>water</sub>	0.014	-
Price of microalgal biomass sold to a company to produce the biofertilizer	€ m <sup>3</sup> <sub>water</sub>	<u> </u>	8.08
Profit (calculated considering operation cost only)	€ m <sup>3</sup> <sub>water</sub>	0.007	8.06

Note: Scenario 1: HRAP system for wastewater treatment where microalgal biomass is valorised for energy recovery (biogas production); Scenario 2: HRAP system for wastewater treatment where microalgal biomass is reused for nutrients recovery (biofertilizer production)



**(b)** 



(c)



Figure 1. Flow diagrams and system boundaries of the wastewater treatment
alternatives: a) HRAP system for wastewater treatment where microalgal biomass is
valorised for energy recovery (biogas production) (Scenario 1); b) HRAP system for
wastewater treatment where microalgal biomass is reused for nutrients recovery
(biofertilizer production) (Scenario 2); c) activated sludge system (Scenario 3)

813



#### Terrestrial acidification





#### Freshwater eutrophication





#### Particulate matter formation





#### Metal depletion







## Terrestrial ecotoxicity



■ Construction materials	Biogas cogeneration and avoided energy
□ Digestate and biofertilizer application (including avoided fertilizer)	Emissions to water
⊠ Emissions to air (NH4+ volatilization in HRAP)	□ Energy consumption
□ Digestate, biofertilizer or sludge transportation	Chemicals
□ Sludge disposal	
<b>Figure 2.</b> Potential environmental impacts for the three scenarios: a) HRAP syster valorised for energy recovery (biogas production) (Scenario 1); b) HRAP syster reused for nutrients recovery (biofertilizer production) (Scenario 2); c) activate functional unit (1 m <sup>3</sup> of was	tem for wastewater treatment where microalgal biomass is em for wastewater treatment where microalgal biomass is ed sludge system (Scenario 3). Values are referred to the ater).



Figure 3. Normalised potential environmental impacts for the three scenarios: a) HRAP system for wastewater treatment where microalgal
 biomass is valorised for energy recovery (biogas production) (Scenario 1); b) HRAP system for wastewater treatment where microalgal
 biomass is reused for nutrients recovery (biofertiliser production) (Scenario 2); c) activated sludge system (Scenario 3).



0.00E+00



0.00E+00

Climate change

Ozone depletion

Summer

Summer



#### Photochemical oxidant formation





#### Particulate matter formation





Figure 4. Seasonal variation of the potential environmental impacts for the three scenarios: a) HRAP system for wastewater treatment
 where microalgal biomass is valorised for energy recovery (biogas production) (Scenario 1); b) HRAP system for wastewater treatment
 where microalgal biomass is reused for nutrients recovery (biofertilizer production) (Scenario 2); c) activated sludge system (Scenario 3).
 Values are referred to the functional unit (1 m<sup>3</sup> of water). Potential environmental impacts were calculated considering the microalgal
 biomass production achieved in summer and winter months (highest and lowest production, respectively).