

## Wastewater treatment using oxygenic photogranule-based process has lower environmental impact than conventional activated sludge process

Doris Brockmann, Yves Gérand, Chul Park, Kim Milferstedt, Arnaud Hélias,

Jérôme Hamelin

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1	Wastewater treatment using oxygenic photogranule-based
2	process has lower environmental impact than conventional
3	activated sludge process
4	Doris Brockmann <sup>1,2,§</sup> , Yves Gérand <sup>2,3, ‡</sup> , Chul Park <sup>4</sup> , Kim Milferstedt <sup>3</sup> , Arnaud
5	Hélias <sup>2,5,\$</sup> , Jérôme Hamelin <sup>3</sup>
6	<sup>1</sup> INRAE, Univ Montpellier, Bio2E, 102 avenue des Etangs, 11100, Narbonne, France
7	<sup>2</sup> ELSA Research Group, Montpellier, France
8	<sup>3</sup> INRAE, Univ Montpellier, LBE, Narbonne, France
9	<sup>4</sup> Department of Civil and Environmental Engineering, University of Massachusetts,
10	Amherst, MA 01003, USA
11	<sup>5</sup> INRAE, Univ Montpellier, LBE, Montpellier SupAgro, Montpellier, France
12	
13	<sup>‡</sup> current address: EVEA, Lyon, France
14	<sup>\$</sup> current address: INRAE, Univ Montpellier, ITAP, Montpellier, France
15	
16	<sup>§</sup> Corresponding author: doris.brockmann@inrae.fr
17	Abstract:
18	The Life Cycle Assessment (LCA) methodology was applied to assess the
19	environmental feasibility of a novel wastewater treatment technology based on oxygenic
20	photogranules (OPG) biomass in comparison to a conventional activated sludge (CAS)
21	system. LCA using laboratory scale experimental data allowed for eco-design of the
22	process during the early stage of process development at laboratory scale. Electricity
23	consumption related to artificial lighting, the fate of the generated biomass (renewable
24	energy and replacement of mineral fertilizer), and the nitrogen flows in the OPG system

were identified as major contributors to the potential environmental impact of the OPG
treatment system. These factors require optimization in order to reduce the
environmental impact of the overall OPG system. Nonetheless, the environmental
impact of a non-optimized OPG scenario was generally lower than for a CAS reference
system. With an optimization of the artificial lighting system, an energy neutral
treatment system may be within reach.

31

Key words: oxygenic photogranules, life cycle assessment, cyanobacteria, anaerobic
digestion, granular biomass

#### 34 1 Introduction

35 Recently, the development of oxygenic photogranules (OPG) from activated sludge has 36 been reported (e.g. Milferstedt et al., 2017b; Quijano et al., 2017). OPG are roughly 37 spherical biological aggregates with a diameter of up to 5 mm (Abouhend et al., 2018). 38 In OPG, the phototrophic biomass produces oxygen through photosynthesis and uses 39 carbon dioxide for growth. Conversely, the heterotrophic biomass produces carbon 40 dioxide and consumes the produced oxygen. The close vicinity of phototrophs and 41 heterotrophs engaged in this syntrophy makes mass transfer more efficient than between 42 dispersed cells found in high rate algal pond (Judd et al., 2015). It has been shown that 43 OPG can perform simultaneous oxidation of organic carbon and nitrogen removal 44 (Abouhend et al., 2018; Arcila & Buitrón, 2017; Arcila & Buitrón, 2016). A novel 45 OPG-based wastewater treatment technology was proposed, considering carbon and 46 nitrogen removal by OPG coupled to anaerobic digestion of harvested biomass for the 47 production of renewable energy (Milferstedt et al., 2017b; Park & Dolan, 2015). 48 Internally produced oxygen by OPG entirely replaces the energy intensive external 49 supply of oxygen for wastewater treatment. External aeration in conventional

50 wastewater treatment accounts for 45-75% of the overall electric energy consumption of 51 the wastewater treatment plant (WWTP; or water resource recovery facility) (Longo et 52 al., 2016). The overall biomass yield (per gram of removed organic carbon) of OPG 53 treating wastewater is up to three times higher than for activated sludge (Abouhend et 54 al., 2018). The biological methane potential (BMP) of the produced OPG is about 15-55 20% higher compared to activated sludge (Park et al., 2015), which is in line with other 56 works on digestion of an algal-sludge biomass (Shoener et al., 2014; Ward et al., 2014). 57 The differences in aeration requirements, sludge production and BMP, compared to 58 activated sludge systems, may result in significant net energy savings when using the 59 OPG process. However, a closer look at environmental performances of this innovative 60 process combining OPG production and anaerobic digestion has not yet been done and 61 therefore is required.

62 The aim of the present study was to evaluate the environmental performance of a 63 putative OPG process compared to conventional technology. For this, Life Cycle 64 Assessment (LCA) will be used, a standardized methodology (ISO 14040, 2006; ISO 65 14044, 2006) to assess potential environmental impacts considering the entire life cycle of products, processes or services based on the function they fulfill, in this case the 66 67 treatment of municipal wastewater. A product, process or service is modeled along its 68 life cycle (from raw material extraction to its end of life). Its impact is assessed with 69 regard to different impact categories (e.g., global warming, ozone depletion, 70 eutrophication, and acidification). LCA has been applied in the field of wastewater 71 treatment to assess the environmental impact of both conventional activated sludge 72 (CAS) wastewater treatment systems (e.g. Foley et al., 2010; Hospido et al., 2008), and 73 advanced wastewater treatment technologies (e.g. advanced oxidation (Muñoz et al., 74 2006) and high rate algal ponds (Colzi Lopes et al., 2018)). While the use of LCA for 75 assessing the environmental impact of existing wastewater treatment technologies

increases, its use for the eco-design of emerging treatment processes under developmentis rare.

78 The present study aimed at evaluating the environmental performances of the novel 79 OPG treatment system using LCA in order to determine whether the OPG system is a 80 feasible technology from an environmental perspective in comparison to a CAS system. 81 The study was based on a hypothetical model of an average, midsize treatment plant and 82 data derived from laboratory scale process operation, having in mind that many 83 technological problems are still unsolved before considering industrialization. The goal 84 of the assessment at this early stage was to identify potential bottlenecks and critical 85 parameters with high environmental impact. Future research can then target these areas 86 to improve within an eco-design approach the process design prior to detailed process 87 engineering studies. In particular, it was considered how far artificial lighting can be 88 used for the OPG system without substantially deteriorating the environmental 89 performance.

90

#### 2 Materials and methods

91 2.1 LCA - Goal and scope

92 The goal of the LCA was to evaluate the environmental feasibility of the OPG system 93 with regard to a reference CAS system and to assess the impact of different operating 94 scenarios of the OPG system during early process development allowing the 95 identification of design considerations with highest environmental impact. Those will 96 then be targeted in future research to eco-design an up-scaled system. The system under 97 study included in addition to the operation phase, the construction of the infrastructure 98 and nutrient recycling from the sludge by agricultural use. Sludge utilization is 99 considered an integral part of wastewater treatment and the basis of a water resource 100 recovery facility.

101 The functional unit, based on which the processes are compared, is "the treatment of 1 102 m<sup>3</sup> of urban wastewater in an average, midsize WWTP". Environmental impacts were 103 computed in SimaPro (version 9.0.0.35, PRé Consultants) using the Ecoinvent database 104 version 3.5 (Wernet et al., 2016) and the Environmental Footprint life cycle impact 105 assessment method (Fazio et al., 2018). 106 A challenge when assessing the impact of a novel process is the availability of process 107 operation data. Here, the evaluation of the OPG system was based on experimental data 108 from the authors' laboratory scale OPG experiments (Abouhend et al., 2018; Milferstedt 109 et al., 2017b). These first experimental studies were mostly conducted to demonstrate a 110 proof of concept and were not dedicated towards optimization and intensification of the 111 process. A large margin of progress is to be expected in the future. Data from the 112 authors' own experimental studies were complemented with laboratory scale data from 113 another research group working on municipal wastewater treatment using microalgae-114 bacteria aggregates (Arcila & Buitrón, 2017; Arcila & Buitrón, 2016) that resemble the 115 OPGs used in the authors' works. Nonetheless, several assumptions were made, that are 116 laid out in detail later, regarding reactor configuration, operating conditions, and 117 treatment performance in a full-scale implementation as the process currently exists 118 only at the laboratory scale. Given the low technical readiness level of the innovative 119 OPG process, some variables remain unknown and need to be estimated from sparse 120 data. The present work, therefore, displays LCA results depending on the estimated 121 value of these variables, such as energy balance and nitrogen flow through the process.

122 **2.2 Life (** 

Life Cycle Inventory

The life cycle inventories for both OPG and CAS systems were estimated for an
hypothetical average midsize WWTP (10,000 to 50,000 people equivalent), which is
equivalent to a WWTP of the Swiss capacity class 3, inventoried in the Ecoinvent 3.5
database (Doka, 2009). A low loaded wastewater was considered here based on

127	experiments	carried out	t using	primary	effluent	from a	US V	<b>WWTP</b>	(Abouhend	et al.
	•	• • • • • • • • • •		printer j	••••••		00		(1.10.0.000000000	

128 2018). Its raw wastewater characteristics are presented in Table 1.

129	Table 1

130

Figure 1

#### 131 Reference system: Conventional activated sludge system

132 The reference scenario for this comparative LCA is a conventional midsize wastewater 133 treatment plant as described in Ecoinvent (Doka, 2009) and presented in Figure 1 a. Grit 134 removal is followed by primary settlers (not shown in the figure). Organic carbon and 135 nitrogen are removed in an activated sludge process. Phosphorus is removed by 136 precipitation using Fe(III)Cl<sub>3</sub>. The sludge is separated from the treated water in 137 secondary settlers. Primary and secondary sludge are treated together by anaerobic 138 digestion. The digester supernatant is returned to the influent of the WWTP. Digested 139 and dewatered sludge is assumed to be spread on land as organic fertilizer. 140 The Ecoinvent datasets for WWTPs of different sizes are based on average treatment 141 efficiencies of Swiss WWTPs dating from 1993-2002 (Doka, 2009) and do not reflect 142 current treatment efficiencies. Consequently, only infrastructure data for the sewer grid 143 and the WWTP were used from the dataset for the midsize treatment plant. Mass 144 balances for chemical oxygen demand (COD), nitrogen, phosphorus, total suspended 145 solids (TSS), and volatile suspended solids (VSS) for the CAS plant were calculated 146 following textbook mass balance equations for biological wastewater treatment (Henze 147 et al. 2008). A sludge retention time (SRT) of 15 days, a mixed liquor suspended solids 148 (MLSS) concentration of 4 g/L, and a design wastewater temperature of  $10^{\circ}$ C were 149 considered. N<sub>2</sub>O emissions from wastewater treatment were estimated using an 150 emission factor of 0.03 g N<sub>2</sub>O-N/g N<sub>denitrified</sub> (Foley et al., 2010; Kampschreur et al.,

151 2009). Treatment efficiencies for aerobic carbon oxidation, nitrification, and nitrogen

removal calculated from mass balance equations were 90%, 84% and 70%, respectively.

153 Phosphorus (P) not incorporated into biomass is precipitated, assuming that 2.5 g

154  $Fe(III)Cl_3/g P_{removed}$  are needed and that 4.9 g FePO<sub>4</sub>/g P<sub>removed</sub> are produced (Rittmann

155 & McCarty, 2001).

156 Mass balances for COD, nitrogen, TSS, VSS, biogas and flow rates for the anaerobic

157 digester were calculated using the steady state model for anaerobic digestion developed

158 by Sötemann (2005), considering a hydraulic retention time of 10 days based on Ekama

159 (2009). Kinetic parameters for anaerobic digestion of primary and secondary sludge

160 were taken from Ekama (2009) and Sötemann (2005). BMPs of primary and secondary

161 sludge vary widely (e.g. Mottet et al., 2010). For primary sludge, a methane production

162 of 230 mL CH<sub>4</sub>/g VSS was assumed based on a hydrolysis rate of 2 g COD/L/d

163 (Sötemann, 2005). This is in the range of methane productions of primary sludge

164 reported by Elbeshbishy et al. (2012). For secondary sludge, a methane production of

165 195 mL CH<sub>4</sub>/g VSS was assumed (Park et al., 2015). A lower methane production of

166 secondary sludge compared to primary sludge is in accordance with literature (e.g.

167 Mahdy et al., 2015). A biogas composition of 65% CH<sub>4</sub> and 35% CO<sub>2</sub> was considered.

168 All CH<sub>4</sub> was assumed to be oxidized to CO<sub>2</sub> during biogas combustion, producing 3

169 kWh electricity per m<sup>3</sup> CH<sub>4</sub> (Nowak, 2003). The solids concentration the digested and

170 dewatered sludge was assumed to be 200 kg TSS/m<sup>3</sup>.

171 A mean electricity consumption of the WWTP of 0.4 kWh/m<sup>3</sup> was considered based on

average data for European WWTPs (Jonasson, 2007). Half of the overall electricity

173 consumption was assumed to be spent for aeration (Longo et al., 2016). It was

174 considered that electricity produced from biogas combustion is used onsite, covering a

175 part of the needed electricity. For the remainder of needed electricity, an average

176 European electricity mix was considered produced from fossil fuels (50%), nuclear

power (27%), hydropower (17%), renewable sources and others (6%) (Itten et al.,
2014).

179 Digested and dewatered sludge was assumed to be applied to farmland as organic 180 fertilizer, replacing ammonium nitrate and triple superphosphate application at 181 equivalent nutrient rates. The long-term nitrogen mineral fertilizer equivalent (MFE) of 182 the sludge, nitrogen field emissions from sludge spreading, and avoided field emissions 183 from mineral fertilizers were calculated following Brockmann et al. (2018), considering 184 a nitrogen content of 9.7 kg N per m<sup>3</sup> sludge (6.4% total ammonia nitrogen (TAN) and 185 93.6% organic nitrogen (Norg)) as calculated from the mass balance equations. For 186 phosphorus, a MFE value of 0.95 was assumed. Phosphorus field emissions were 187 estimated following Brockmann et al. (2014). Sludge application by broadcaster without 188 incorporation and mean French soil and climate conditions were assumed. Tables 2 and 189 3 summarize characteristics of the CAS system and calculated emissions from 190 wastewater treatment and sludge spreading. Parameters and emission factors used for 191 calculating field emissions were taken from Brockmann et al. (2014) and Brockmann et 192 al. (2018).

#### 193 Differences in the oxygenic photogranule (OPG) system

194 The only difference between the considered reference CAS and the OPG plant layouts is 195 the basin for secondary treatment and the absence of secondary settlers (Figure 1 b). In 196 the OPG system, secondary treatment took place in sequencing batch reactors (SBR). 197 As settling of the OPG was carried out in the SBRs during a settling phase, secondary 198 settlers were not needed. The model of the full-scale OPG system was based on data 199 from the authors' laboratory scale OPG studies (Abouhend et al., 2018; Milferstedt et 200 al., 2017b) complemented with data from laboratory scale experiments with microalgae-201 bacteria aggregates treating municipal wastewater (Arcila & Buitrón, 2017; Arcila & 202 Buitrón, 2016). Based on the authors' laboratory scale OPG experiments, operation in

SBRs at a hydraulic retention time (HRT) of 0.5 days and a MLSS concentration of 4
g/L was assumed. For these operating conditions, the overall SBR reactor volume
needed is similar to that of the activated sludge reactor plus the secondary settlers.
Therefore, the same environmental impact for the required infrastructure was assumed
for both systems.

208 Sludge production was estimated based on a biomass yield of 0.7 g VSS/g COD<sub>consumed</sub> 209 (1.26 g COD/g COD<sub>consumed</sub>) and a COD/VSS ratio of the OPG of 1.8 g COD/g VSS 210 (Abouhend et al., 2018). An average organic carbon removal efficiency of 85% was 211 considered (Abouhend et al., 2018; Arcila & Buitrón, 2016), assuming that 50% of the 212 removed organic carbon was incorporated into the heterotrophic part of OPG biomass. 213 The remaining 50% of the removed organic carbon were assumed to be oxidized and 214 then available for phototrophic uptake. An average ammonia transformation efficiency 215 of 90% was considered (Abouhend et al., 2018), including nitrogen incorporation into 216 biomass and nitrification. Nitrogen incorporation into biomass was estimated using a 217 nitrogen content of the OPG of 10% (as for CAS), resulting in 54% of the organic and 218 ammonia nitrogen being incorporated into OPG (vs. 22% calculated for CAS). The 219 remaining unaccounted transformed ammonia was assumed to be nitrified. Based on 220 this assumption, 33% of the NH<sub>4</sub>-N transformed was converted to NO<sub>3</sub>-N, which was 221 below an observed NO<sub>3</sub>-N production rate of 50% (Abouhend et al., 2018; Arcila & 222 Buitrón, 2016), but of the same order of magnitude as reported by Arcila and Buitrón 223 (2017). Denitrification by OPG has, so far, not been reported, but the presence of genes 224 (Stauch-White et al., 2017) and of 16S rRNA bacterial sequences (Milferstedt et al., 225 2017b) associated with denitrification was revealed. It was assumed that the OPG 226 system includes a denitrification step with a denitrification efficiency similar to the 227 reference system (80%). Overall, a total nitrogen removal efficiency of 71% was 228 obtained based on mass balances, which was equivalent to the CAS system. A methane

production of 290 mL CH<sub>4</sub>/g VSS for OPG (Arcila & Buitrón, 2016) and a biogas

composition of 65% CH<sub>4</sub> and 35% CO<sub>2</sub> were considered.

231 In contrast to the CAS system, the OPG system does not require aeration since OPG 232 produces oxygen under light. Based on laboratory scale experiments with 3.5 hours 233 illumination per 6 hours cycle (Milferstedt et al., 2017b), illumination during 14 hours 234 for the four daily cycles is considered. With an oxygen production rate of the OPG 235 under light of 12.6 mg  $O_2/(g \text{ VSS} \cdot h)$  (Abouhend et al., 2018), oxygen production by 236 OPG with 14 h/day illumination exceeds the oxygen requirements for organic carbon 237 oxidation and nitrification. To ensure adequate suspension of OPG, mechanical mixing 238 is needed. Electricity consumption for mixing of OPG was estimated following Walas 239 (1990) considering a turbulent regime and a reactor geometry similar to that of activated 240 sludge reactors. Considering an anchor or gate paddle per reactor and a mixing time of 241 22 h/day, mixing of OPG consumes 26 Wh/m<sup>3</sup> electricity. Electricity consumption for 242 lighting was estimated based on a photosynthetically active radiation (PAR) of 150 243 µmol/m<sup>2</sup>/s (Abouhend et al., 2018) and a PAR efficiency of 2 µmol/s/W (Blanken et al., 2013), yielding an electricity consumption of 75  $W/m^2$ . The enlightened surface area 244 was assumed twice the footprint of the reactors without further detailed design of the 245 246 artificial lighting system, resulting in an electricity consumption of 134 Wh/m<sup>3</sup> 247 wastewater. This way of operation represents a generic, non-optimized solution. An electricity consumption for pumping, valves, sensors etc. of 200 Wh/m<sup>3</sup> was estimated 248 249 based on the electricity consumption of the CAS system without aeration. Thus, the 250 overall electricity consumption of the OPG system consists of 200 Wh/m<sup>3</sup> for pumping, 251 valves etc., 26 Wh/m<sup>3</sup> for 22 hours of mixing, and 134 Wh/m<sup>3</sup> for lighting. 252 For land application of the OPG, a long-term MFE of 0.562 was calculated, based on a 253 nitrogen content of 13.4 kg N/m<sup>3</sup> OPG (15.4% TAN and 84.6% Norg), as calculated 254 from mass balance equations.

255	Table 2
256	Table 3
257	
258	Operating scenarios
259	OPG is exposed to sunlight during the day, enabling oxygen production by
260	photosynthesis. For providing oxygen during night-time, when OPG is not exposed to
261	sunlight and thus not photosynthetically active, different operating scenarios were
262	considered:
263	• Scenario 1: Artificial lighting is provided during the night. Considering overall
264	14 h/day of illumination and assuming that natural light covers on average 7
265	h/day, the same duration of artificial lighting is needed.
266	• Scenario 2: Aeration replaces the 7 h/day of artificial lighting. Thus, aeration is
267	needed for 7 h/day and mixing for 15 h/day.
268	• Scenario 3: OPG is not exposed to sunlight at all and artificial lighting is
269	provided 14 h/day.
270	• Scenario 4: As scenario 1 but with a 50% reduction in light energy provided.
271	Scenario 1 was used as the default scenario for the OPG system. The impact of the other
272	operating scenarios was assessed in a sensitivity analysis.
273	
274	3 Results and discussion
275	<b>3.1</b> Comparison of the two treatment systems
276	The environmental impact of treating 1 m <sup>3</sup> municipal wastewater and applying the
277	resulting sludge on farm land was compared using a CAS system and the OPG system

278 (scenario 1). A graphical representation of this comparison is shown as radar plot in

21)	rigure 2 for an evaluated impact categories. The black line signifies the erro system as
280	reference, and the blue line the OPG system. For most impact categories, the OPG
281	system's environmental impact is inferior of the CAS impact ranging from a 4%
282	difference for freshwater eutrophication to 61% for ionizing radiation. The two notable
283	exceptions are for the impact categories terrestrial eutrophication and acidification being
284	2 and 3 times higher, respectively, compared to the CAS system.

Figure 2 for all evaluated impact categories. The black line signifies the CAS system as

285

279

#### Figure 2

286

287 In Figure 3, the environmental impacts in the various impact categories are related to 288 where they are generated, i.e., treatment infrastructure, emissions from treatment, 289 electricity consumption, Fe(III)Cl<sub>3</sub> use for precipitation, sludge spreading and the 290 substitution of mineral fertilizer. Contributions that are identical for both systems are 291 not displayed even though considered in the overall calculations (e.g., sewer grid 292 infrastructure, emissions from overload discharge, grit removal, and treatment plant 293 infrastructure). It should be noted that sewer grid and treatment plant infrastructure are 294 major contributors to the environmental impact. But in this comparison, this impact is 295 not considered relevant as it is assumed to be independent of the biological 296 configuration of the treatment plant. Figure 3 reveals that the significantly higher 297 impacts of the OPG system in the impact categories acidification, terrestrial 298 eutrophication, and respiratory inorganics can be considered a mass effect, as 299 significantly larger quantities of digested sludge are land-applied. The impact is not 300 specific to OPG. In particular, most of the increased impact on terrestrial eutrophication, 301 acidification, and respiratory inorganics from sludge spreading is caused by higher 302 ammonia emissions from land application. Ammonia emissions from sludge spreading 303 contributed 114%, 107%, and 64%, respectively, to the mentioned impact categories. 304 Twice as much nitrogen was recovered in the digested OPG and spread on land than for

305 the CAS system (203 kg N/d vs. 110 kg N/d). The amount of nitrogen emissions from 306 land application does not only depend on the amount of nitrogen applied, but also on the 307 method of sludge application. Surface application of sludge without subsequent 308 incorporation into the soil is the worst case with respect to ammonia emissions. The 309 significantly lower impact of the OPG system on ionizing radiation is due to a much 310 lower electricity consumption from the grid compared to the CAS system. The OPG 311 system has an overall electricity consumption of 359 Wh/m<sup>3</sup>, of which 269 Wh/m<sup>3</sup> are 312 covered by combustion of the produced biogas and 90 Wh/m<sup>3</sup> by electricity from the 313 grid. In contrast, to cover the overall electricity consumption of the CAS system of 400 314 Wh/m<sup>3</sup>, 263 Wh/m<sup>3</sup> are needed from the grid as only 137 Wh/m<sup>3</sup> are produced from 315 biogas combustion. Lower impacts on ozone depletion, photochemical ozone formation, 316 human health effects, freshwater ecotoxicity, and resource use are due to lower 317 electricity consumption and higher amounts of mineral fertilizers replaced by land-318 applied digested biomass. The reduced impact on climate change, compared to the CAS 319 system, result from lower N<sub>2</sub>O emissions from wastewater treatment and lower 320 electricity consumption of the OPG system. As for the CAS system, N<sub>2</sub>O emissions 321 from wastewater treatment with OPG were estimated based on the amount of nitrogen 322 denitrified and may change using real data. Nonetheless, climate change impacts of the 323 OPG system remain inferior to the ones of the CAS system even when the contribution 324 of N<sub>2</sub>O emissions is excluded.

325

#### Figure 3

326

# 327 3.2 Effects of alternative illumination scenarios on the environmental impact of 328 an OPG treatment plant

329 The default OPG system (scenario 1) operated with 7 h/day artificial lighting, even in its
330 immature design state, outperforms the CAS system. In the following, the sensitivity of

331 the LCA results is assessed using three envisioned operating scenarios, which affect the 332 electricity consumption of the OPG treatment plant: 7 h/day aeration (scenario 2), 14 333 h/day artificial lighting (scenario 3) and a modified scenario 1 with a 50% cut-off in 334 light energy provided to the system (scenario 4). It is acknowledged that scenarios 2 and 335 4 could affect the stability and the performance of the OPG system. Despite these 336 potentially important unknowns, the suggested scenarios allow to assess whether these 337 or similar approaches are environmentally feasible, before even starting the appropriate 338 experiments. The LCA methodology should be considered here a modeling approach 339 that evaluates the environmental impact of putative potential alternatives. For this 340 evaluation, it was assumed that operating conditions for scenarios 3 and 4 impact only 341 electricity consumption, while biochemical conversion rates and overall treatment plant 342 performance remained unchanged. For scenario 2 (7 h/day aeration), the phototrophic 343 biomass yield was reduced, assuming that with only half of the light, half of the 344 phototrophic growth will occur. The reduced phototrophic biomass yield entails lower 345 biomass and, thus, biogas and electricity production. Replacing artificial lighting during 346 night by aeration reduced electricity consumption by about 25% and electricity production by about 15%, requiring only 48 Wh/m<sup>3</sup> from the grid. It may be surprising 347 348 that the partially aerated system is less energy consuming than the fully photosynthetic 349 system. This is possibly because of the substantial increase of energy recovered from 350 the digested phototrophic sludge. A lower biomass production also implicates lower 351 ammonia emissions from field application of the digested biomass, and lower amounts 352 of mineral fertilizers replaced. Figure 4 shows that the reduced electricity consumption 353 from the grid decreased environmental impacts of the OPG system on ionizing radiation 354 by 35% and on resource use of energy carriers by 11%, compared to scenario 1. 355 Furthermore, the reduced amount of land-applied digested OPG reduced environmental 356 impacts on acidification and terrestrial eutrophication by 24% and on respiratory

357 inorganics by 13%, but increased the environmental impact on resource use of minerals 358 and metals by 14%, compared to scenario 1. Impacts in other impact categories changed 359 by less than 7%. For scenario 3, where artificial lighting needed to be provided 14 360 h/day, electricity consumption from the grid more than doubled compared to scenario 1, 361 but remained below the electricity consumption from the grid of the CAS system. Due 362 to increased electricity consumption from the grid, environmental impacts on ionizing 363 radiation doubled and increased by 46% for resource use of energy carriers and land 364 use, and by 31% ozone depletion (Figure 4, orange line). Impacts on other impact 365 categories increased by 1 to 17%.

366 The artificial lighting system considered here represents a largely non-optimized 367 solution. The efficiency of LED lighting is drastically developing and will likely 368 improve significantly over the next years (Zhang et al., 2018), reducing the energy 369 requirements to yield a given photoactive radiation. This kind of development is not 370 expected for the aeration in the CAS system, which is already a mature technology. In 371 addition, the use of white light LEDs was assumed here of which a considerable 372 bandwidth is not suitable for photosynthesis. The use of LEDs with adapted spectra for 373 phototrophic light alone could yield significant energy savings at a similar biological 374 activity (Abomohra et al., 2019). In addition, the use of suspended, free-moving LED 375 (Murray et al., 2017) may further reduce energy consumption for artificial lighting. 376 Therefore, an optimized artificial lighting system with an assumed improved energy 377 efficiency of LED lighting of 50% (scenario 4) was evaluated here and compared to 378 scenario 1. All other parameters of scenario 1 were kept. With optimized artificial 379 lighting, the electricity consumption of scenario 1 decreased to 293 Wh/m<sup>3</sup>, requiring 380 only 23 Wh/m<sup>3</sup> from the grid. Thus, with an optimized artificial lighting system, an 381 energy neutral treatment system may be within reach.

382

Figure 4

383

#### **384 3.3 Operation of the OPG system at higher biomass concentrations**

385 The presented impact assessment was computed for operating the OPG system at the 386 same biomass concentration as the laboratory scale reactor (4 g/L). This is a typical 387 biomass concentration for CAS systems and a relatively low MLSS concentration 388 compared to other granular biomass systems (Milferstedt et al., 2017a). For example, 389 aerobic granular sludge systems can be operated at biomass concentrations up to 10 g/L 390 (Keller & Giesen, 2010). It is worthwhile investigating the OPG treatment performance 391 at higher biomass concentrations and lower HRT, while maintaining the SRT in the 392 system, to reduce the overall reactor volume of the treatment system. This will decrease 393 treatment plant infrastructure needs, which significantly contribute to environmental 394 impacts on land use, human health effects, freshwater ecotoxicity, resource use, 395 photochemical ozone formation, and respiratory inorganics. Assuming operation of the 396 OPG system at a biomass concentration of 6 g/L and keeping the sludge retention time 397 unchanged, reduces the HRT to 0.33 days and the required reactor volume by 30%. 398 Energy requirements for mixing need to be considered in a putative OPG system. 399 Several alternative modes of mixing could be envisioned, e.g., intermittent gas sparging 400 or mixing by pulse-like waves (e.g., oloid.ch). Here, a traditional mixing approach using 401 constantly turning impellers was considered. Energy needs for mixing will decrease 402 with reduced reactor diameter, as the impeller diameter decreases, but energy 403 requirements for artificial light supply may increase with a higher biomass 404 concentration. It is assumed here that the potentially higher energy requirements for 405 artificial light supply compensate the energy savings from reduced energy needs for 406 mixing. This means that the reduction of the reactor volume does not significantly affect 407 the overall electricity consumption of the OPG system. It is further considered that 408 operation at higher biomass concentration and shorter HRT does not result in a

409 considerable loss of treatment performance. Based on the aforementioned assumptions, 410 the reduction of the required reactor volume decreases the environmental footprint with 411 regard to resource use of minerals and metals by 50%, to land use by 37%, to ozone 412 depletion and non-cancer human health effects by 17%, and to photochemical ozone 413 formation by 16%. Reductions on other impact categories range from 0.7 to 13%. These 414 results are based on assumptions that still need to be validated experimentally, but show 415 the interest (from an environmental perspective) of a more compact treatment system.

416 **3.4** 

#### Nitrogen flows in the OPG system

417 In the LCA model, nitrogen emissions in the forms of ammonia, nitrite/nitrate, and 418 organic nitrogen from wastewater treatment with OPG were on the same order of 419 magnitude as for the reference CAS system. N<sub>2</sub>O emissions from biological treatment, 420 estimated based on the amount of nitrogen denitrified, were lower for the OPG system, 421 because more nitrogen was incorporated into biomass, requiring less denitrification. The 422 same emission factor for estimating N<sub>2</sub>O emissions from a wastewater treatment with 423 OPG and CAS system was used. The factor is based on Foley et al. (2010) and 424 Kampschreur et al. (2009) and remains to be estimated using experimental data from the 425 OPG system. It was demonstrated that major nitrogen emissions occurred also 426 downstream of the treatment plant, at the stage of land application of the digested 427 biomass (Figure 3), affecting largely the impact categories terrestrial eutrophication, 428 acidification, and respiratory inorganics. This was caused by significantly higher 429 biomass produced by the OPG system, resulting in twice as much nitrogen spread on 430 land with the digested OPG than with the activated sludge produced by the CAS system. 431 Consequently, significantly higher nitrogen field emissions for the same volume of 432 wastewater treated can be expected (23 kg NH<sub>3</sub>/d for OPG system vs. 5.1 kg NH<sub>3</sub>/d for 433 CAS system). Optimization of the environmental impact of the assessed OPG system 434 should, therefore, also consider the fate of the generated biomass in applications

435 downstream of the treatment plant. The observed large contribution of land application 436 of the digested biomass to the environmental burdens of the treatment systems is in line 437 with other works considering land application of sludge and replacement of mineral 438 fertilizers (e.g. Brockmann et al., 2014; Pasqualino et al., 2009). 439 In the default scenario, land application of digested sludge by a broadcaster, without 440 incorporation into the soil was assumed. This is common agricultural practice (Loyon, 441 2018) and known to be the worst case scenario with regard to ammonia field emissions 442 (Bittman et al., 2014). Land application of digested biomass by deep injection, 443 considered the best case scenario with regard to ammonia field emissions (Bittman et 444 al., 2014), significantly reduced ammonia field emissions in the LCA model from 1504 g NH<sub>3</sub>/m<sup>3</sup> sludge to 301 g NH<sub>3</sub>/m<sup>3</sup> sludge for OPG, and from 452 g NH<sub>3</sub>/m<sup>3</sup> sludge to 445 446 91 g NH<sub>3</sub>/m<sup>3</sup> sludge for activated sludge. The decreased ammonia field emissions 447 vielded in larger amounts of ammonia nitrogen available to the plants, increasing the 448 nitrogen MFE values for OPG (0.64 vs. 0.56) and CAS (0.62 vs. 0.59). Consequently, more mineral fertilizers could be replaced by the digested biomass (10.2 kg N/m<sup>3</sup> sludge 449 vs. 9 kg N/m<sup>3</sup> sludge from OPG, 7.2 kg N/m<sup>3</sup> sludge vs. 6.9 kg N/m<sup>3</sup> sludge from CAS). 450 451 The effect of agricultural practice is illustrated in Figure 5. Optimizing the land 452 application of digested biomass from broadcaster application to deep injection 453 significantly decreased the environmental impact in three categories: terrestrial 454 eutrophication, acidification, and respiratory inorganics. The double effect of reducing 455 ammonia emissions from land application of digested biomass and increasing the 456 amount of replaced mineral fertilizers and associated emissions yields in a lower 457 environmental footprint of the OPG system for all 16 impact categories. Thus, in 458 addition to modifications at the treatment plant, the fate of the excess sludge is a 459 determining factor for the environmental impact of the OPG process, and as well for

460 CAS. Particularly, changes of the sludge disposal method can considerably drive the

461 environmental impact and must be considered already in the process design.

462

Figure 5

463

464 **3.5 Use of produced OPG biomass** 

465 For a water resource recovery facility, the produced biomass is a resource that can be 466 transformed into bioenergy and other bioproducts. Even though the high yield in an 467 OPG system and the high degree of uniform phototrophic biomass may make other 468 transformations and valorization ways possible, a 'classical' transformation of the 469 biomass into energy and organic fertilizer by anaerobic digestion was considered in this 470 study. Considering the same biomass transformation for the OPG and the reference 471 CAS system made it possible to show that wastewater treatment using the evaluated 472 OPG system allows for higher energy and nitrogen recovery than the CAS system. The 473 OPG system produced twice as much methane per m<sup>3</sup> of treated wastewater as the CAS system (0.090 vs. 0.046 Nm<sup>3</sup> CH<sub>4</sub>/m<sup>3</sup> wastewater) and replaced 75% more mineral 474 475 nitrogen fertilizer (per m<sup>3</sup> wastewater treated) through agricultural use of the digested 476 biomass. This 'base' case of agricultural use was considered because it is current 477 common agricultural practice in France, Spain, and Ireland (European Commission, 478 2019a) and commonly used when assessing the environmental impact of wastewater 479 treatment (Foley et al., 2010; Hospido et al., 2008). The land application of digested 480 sludge is probably the worst case with regard to the environmental impact/benefit of the 481 biomass use. In addition, land application of digested sewage sludge will be more 482 restricted in the EU due to more stringent limits for contaminants following the new 483 Fertilizing Products Regulation of the EU (European Commission, 2019b). Electricity 484 generation from biogas is only one potential use of the biogas. Other uses such as

485 upgrading to biomethane as a substitute for natural gas or as a raw material to produce 486 platform chemicals (Tsui & Wong, 2019) should be considered as well in the future. 487 The objective of this LCA was to guide research efforts to eco-design and improve the 488 sustainability of the OPG process. It is therefore wise to envision alternative uses of the 489 produced OPG, possibly even resulting in larger environmental benefits. One area of 490 active research is the downstream use of extracellular polymeric substances from 491 granular sludge (Quijano et al., 2017; van Loosdrecht & Brdjanovic, 2014). OPG may 492 be a suitable candidate for this approach, as these granules are composed of a mat-like 493 outer layer densely populated by filamentous cyanobacteria (Milferstedt et al., 2017b). 494 This layer contains large amounts of extractible extracellular polymeric substance (EPS) 495 produced by cyanobacteria (e.g. Ansari et al., 2019; Milferstedt et al., 2017b). The 496 extracted EPS may serve, depending on the physico-chemical properties (e.g., molecular 497 weight, charge, hydrophobicity) as hydrogels, biosurfactants or bioflocculants, as 498 demonstrated in part already for aerobic granules (Lin et al., 2015). It is of great 499 advantage that through the syntrophy between heterotrophs and phototrophs, 500 heterotrophically produced CO<sub>2</sub> is apparently immediately fixed in OPG by the growing 501 phototrophic biomass. Resource recovery and conservation of fixed nitrogen are 502 intrinsic feature of this biomass and must be taken advantage of.

503 **3.6 Research perspectives** 

Two major groups of bottlenecks in the development of a potential OPG-based
bioprocess have been identified in the analysis: reducing the environmental costs of
providing light to OPGs and increasing the environmental benefit of generating OPG
biomass.

508 Light dependencies touch reactor configuration as much as they potentially affect the 509 biological activity of the phototrophic biomass. Research on the activity of OPGs as a 510 function of lighting, possibly even at the scale of individual photogranules, must be

511 coupled to identifying actual lighting conditions in current or prospective bioreactor

designs. This research must also consider effects of biomass density and mixing tosuccessfully narrow down process engineering constraints.

514 While coupling of microbiological and engineering research has been successfully done 515 for years, biomass valorization requires the establishment of new collaborations. Novel 516 valorization approaches will be a compromise between constraints from wastewater 517 treatment (e.g., treatment efficiency) and the target quality and quantity of potential bio-518 products. Developing a value chain that meets these requirements can only be done 519 through the commitment of public and private entities across sectoral borders and 520 disciplines.

#### 521 **4** Conclusions

522 The environmental impact of a novel OPG-based treatment process was evaluated using 523 LCA and compared with the well-established conventional activated sludge (CAS) 524 process. The environmental impact of a non-optimized OPG scenario was generally 525 lower than for the reference CAS system. Electricity consumption related to artificial 526 lighting, the fate of the generated biomass (renewable energy and replacement of mineral fertilizer), and the nitrogen flows in the OPG system were identified as the 527 528 major contributors to the potential environmental impact of the OPG treatment system. 529 With an optimized artificial lighting system, an energy neutral treatment system is 530 within reach.

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	690	7 Figure captions

Figure 1: Process schemes of the two compared wastewater treatment systems. a)
Conventional activated sludge system (CAS), and b) oxygenic photogranules
(OPG) system. In b), only changes with respect to a) are highlighted. Upstream
unit processes identical for both processes (overload discharge, grit removal, and
primary settling) are omitted from the figure for clarity. Line colors correspond
to sludge (brown), gas and energy (green) and liquid phase (blue).

697

# Figure 2: Comparison of environmental impacts of the treatment of 1 m<sup>3</sup> urban wastewater by the OPG system (scenario 1) and the CAS system as reference. Black: CAS system, blue: OPG system. All calculated environmental impacts

- 701 were normalized by the impact obtained for the reference CAS system.
- 702 Distances between circles are log2 scaled.

703

Figure 3: Environmental impacts by impact category and differentiated by their origin
(colored stacks) for the treatment of 1 m<sup>3</sup> municipal wastewater in the OPG
system (scenario 1) and the reference CAS system. The units of the different
impact categories are given in the panel headers.

709	Figure 4: Environmental impact of different operating scenarios for the OPG system.
710	Scenario 1 (as reference): 7 h/day artificial light (solid blue line). Scenario 2: 7
711	h/day aeration instead of lighting (short yellow dashes). Scenario 3: 14 h/day
712	artificial lighting (orange dots). Scenario 4 (light-optimized scenario 1): 7
713	h/day artificial lighting with twice more efficient lighting system (dash-dots).
714	
715	Figure 5: Effects of agricultural practice on the environmental impact of the OPG
716	(scenario 1) and the reference CAS systems. Black, solid: default CAS system,
717	including surface spreading of sludge; blue, solid: OPG system (scenario 1),
718	including surface spreading of sludge; Black, dashed: CAS system with deep
719	injection of digested biomass; blue, dash-dots: OPG system with deep injection
720	of digested biomass. For each impact category, calculated environmental
721	impacts were normalized by the impact obtained for the reference CAS system.
722	The distance between circles is log2-scaled.

#### **8 Tables**

720 Table 1. Kaw municipal wastewater characteristics (05	726	Table 1: Raw	municipal	wastewater	characteristics (	(US)
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Parameter	Symbol	Unit	Values
Flow rate	Qin	m <sup>3</sup> /d	15000
Total chemical oxygen demand	Total COD	g COD/m <sup>3</sup>	500
Soluble chemical oxygen demand	Soluble COD	g COD/m <sup>3</sup>	200
Total Kjeldahl Nitrogen (N <sub>org.</sub> + NH <sub>4</sub> -N)	TKN	g N/m <sup>3</sup>	30
Ammonia nitrogen	NH <sub>4</sub> -N	g N/m <sup>3</sup>	20
Nitrate nitrogen	NO <sub>3</sub> -N	g N/m <sup>3</sup>	0
Total phosphorus	Total P	g P/m <sup>3</sup>	6
Orthophosphate	Ortho-P	g P/m <sup>3</sup>	4
Total suspended solids	TSS	g TSS/m <sup>3</sup>	250
Volatile suspended solids	VSS	g TSS/m <sup>3</sup>	-

- Table 2: Characteristics of the reference CAS system and the OPG system:
- 730 Consumptions and productions, digested sludge characteristics, and avoided
- 731 mineral fertilizer as calculated from mass balances. Units are per m<sup>3</sup>

732 wastewater unless otherwise stated

	Unit	CAS	OPG		
Consumptions					
Electricity	kWh/m <sup>3</sup>	0.4	0.2+mixing+ligh t <sup>(*)</sup>		
Iron(III)chloride	g/m <sup>3</sup>	19.4	8.7		
Productions					
Primary sludge	kg VSS/d	2184	2184		
Secondary sludge	kg VSS/d	962	2916		
Methane	Nm <sup>3</sup> CH <sub>4</sub> /m <sup>3</sup>	0.046	0.090		
Electricity from CH <sub>4</sub> combustion	kWh/m <sup>3</sup>	0.137	0.269		
Digested sludge characteristics					
Dewatered sludge	m <sup>3</sup> /d	11.3	15.2		
TSS content of sludge	kg TSS/m <sup>3</sup> sludge	200	200		
N content of sludge	kg N/m <sup>3</sup> sludge	9.7	13.4		
TAN/N <sub>tot</sub>	%	6.4	15.4		
Norg/Ntot	%	93.6	84.6		
P content of sludge	kg P/m <sup>3</sup> sludge	5.6	4.2		
Nitrogen MFE (long-	-	0 594	0 562		
term)		0.571	0.502		
Phosphorus MFE	-	0.95	0.95		
Avoided mineral fertilizers					
Ammonium nitrate	kg N/m <sup>3</sup> sludge	6.9	9.0		
Triple superphosphate	kg $P_2O_5/m_{sludge}^3$	24.2	17.1		

<sup>733 &</sup>lt;sup>(\*)</sup> Mixing (22h/d): 0.026 kWh/m<sup>3</sup>; light (7h/d): 0.134 kWh/m<sup>3</sup>

Table 3: Emissions from wastewater treatment and sludge spreading, and avoided

emissions from mineral fertilizers as calculated from mass balances. Units are

per m<sup>3</sup> wastewater for wastewater treatment and per m<sup>3</sup> sludge for sludge

738 spreading.

	Unit	CAS	OPG				
Effluent characteristics							
COD	g COD/m <sup>3</sup>	31.3	48.3				
TKN	g N/m <sup>3</sup>	3.8	4.6				
NH4-N	g N/m <sup>3</sup>	1.2	2.0				
NO <sub>3</sub> -N	g N/m <sup>3</sup>	3.7	2.3				
Total P	g P/m <sup>3</sup>	1.8	1.8				
Emissions from wastewater treatment							
N <sub>2</sub> O-N	g N/m <sup>3</sup>	0.44	0.29				
Biogenic CO <sub>2</sub> from carbon removal	g CO <sub>2</sub> /m <sup>3</sup>	283	202				
Biogenic CO <sub>2</sub> from AD	g CO <sub>2</sub> /m <sup>3</sup>	45	89				
Biogenic CO <sub>2</sub> from CH <sub>4</sub> combustion	g CO <sub>2</sub> /m <sup>3</sup>	84	165				
Emissions from sludge spreading							
NH <sub>3</sub>	g/m <sup>3</sup> sludge	452	1504				
N <sub>2</sub> O	g/m <sup>3</sup> sludge	173	250				
NO <sub>x</sub>	g/m <sup>3</sup> sludge	243	316				
NO <sub>3</sub> -	g/m <sup>3</sup> sludge	4764	6581				
Р	g/m <sup>3</sup> sludge	47	33				
Avoided emissions from mineral fertilizers							
NH <sub>3</sub>	g/m <sup>3</sup> sludge	255	333				
$N_2O$	g/m <sup>3</sup> sludge	122	159				
NO <sub>x</sub>	g/m <sup>3</sup> sludge	174	227				
NO <sub>3</sub> <sup>-</sup>	g/m <sup>3</sup> sludge	3383.6	4424.8				
Р	g/m <sup>3</sup> sludge	11.6	8.2				

739

#### **9 Figures**

#### 742 Figure 1:



#### 745 Figure 2:







#### 751 Figure 4:



