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1 Techno-economic and Life Cycle Assessment of methane 2 production via biogas upgrading and power to gas technology

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13

14 **Abstract**

15 To decrease the use of fossil fuels and face the energetic demand, the integration of renewable
16 energy is a necessary step. Part of this renewable energy can be supplied by the production of
17 electricity from photovoltaic panels and windfarms. The massive use of these intermittent energies
18 will lead to overproduction periods, and there is consequently a need to convert this surplus of
19 electricity into a storable form of energy. Power-to-gas (PtG) technology consists in using electricity
20 to convert water into hydrogen by electrolysis, and then to synthesize methane from carbon dioxide
21 and hydrogen. Techno-economic and Life Cycle Assessment of methane production via the
22 combination of anaerobic digestion and PtG technology have been applied to sewage sludge
23 valorization. Process studies and equipment design have been addressed considering already
24 available technologies. Sensitivity analyses have been done on biogas upgrading technologies,
25 electricity prices, annual operation time and composition of the electricity mix with also a
26 comparison between PtG and direct injection. It appears that the more the electricity is expensive,
27 the longer the operation time of the methanation process must be to be competitive with injection
28 of methane from biogas. Reduction of electricity consumption of the electrolysis step decreases
29 production costs. Even if the current context does not feature adapted conditions to ensure an
30 economically viable chain, the evolution of the energetic context in the next few years as well as the
31 expected technological improvements will contribute to overall cost reduction. From an
32 environmental point of view, continuous PtG generates more greenhouse gases than direct injection,
33 but intermittent operation with use of renewable electricity can significantly reduce GHG emissions.
34 From an endpoint impacts perspective, impact from continuous PtG are higher than biogas
35 upgrading, but much lower than fossil energy. Future development of low electricity consumption of
36 the electrolysis process, and integration of renewable credits from CO₂ valorization can increase the
37 competitiveness of this technology.

38 **Keywords:** Life Cycle Assessment (LCA); Power-to-Gas; Methane; CO₂ valorization; Energy storage;
39 Production costs

40 **1 Introduction**

41 Even if fossils fuels accounted for 83% of total primary energy supply in 2008 (IPCC 2011), the use of
42 fossil energy faces many issues: decreasing reserves, emissions of greenhouses gas (GHG) and other
43 pollutants during their combustion, but also dependence to importations, with the associated risks. It
44 is consequently compulsory to develop alternative energy production technologies. Renewable
45 energy can play a central role in minimizing these issues, and can be partly supplied by electricity
46 from renewable sources, like photovoltaic panels and windfarms. This electricity emits less GHG
47 (Varun et al. 2009) and pollutants (Turconi et al. 2013; Turconi et al. 2014) than fossil fuels (except
48 nuclear electricity), and can be produced almost all over the planet (de Vries et al. 2007).
49 Nevertheless, renewable electricity production faces two major drawbacks: first its intermittency,
50 with a production which cannot be adjusted to electricity demand, and second its substitution to
51 high energy density fuels, especially in transport and heat (Sterner 2009).

52 While renewable electricity represents a small proportion of electricity production in Europe (24.7%)
53 and in France (16.5%) in 2014 ([http://ec.europa.eu/eurostat/statistics-
54 explained/index.php/Electricity_production_and_supply_statistics](http://ec.europa.eu/eurostat/statistics-explained/index.php/Electricity_production_and_supply_statistics)), recent publication of the
55 European Commission on energy production forecasts a large share of renewables (between 64%
56 and 97% depending on the scenarios) in the electricity mix in 2050 (European Parliament 2011), and
57 scenarios developed by the ADEME (ADEME 2015) go as far as to propose fully renewable electricity
58 mix.

59 To fulfill the needs of electricity demand with the integration of more renewables in the electricity
60 mix, the installed capacity should be increased. This massive electricity production from renewable
61 energies will lead to overproduction periods, where produced electricity would not be totally
62 employed. Therefore there is a need to store this energy. Several electricity storage technologies are
63 available like batteries, pumped hydropower plants, compressed air energy storage or hydrogen
64 storage technologies, with different prices, run times and storage capacities (Hadjipaschalis et al.
65 2009). Pumped hydropower technology is the most massive storage technology available today, and
66 is generally enough to balance the current electrical system. However it will be probably inadequate
67 to store large amounts of overproduced electricity (ADEME 2014) and it has already been largely
68 deployed. In this perspective, natural gas network offers a high storage capacity (for example 135
69 TWh in France, (DGEC (Direction générale de l'énergie et du climat) 2011)), and therefore conversion
70 of electricity into gas would offer an interesting leverage to valorize overproduced electricity. In this
71 perspective, production of methane (CH₄) by power-to-gas (PtG) technology can greatly increase the

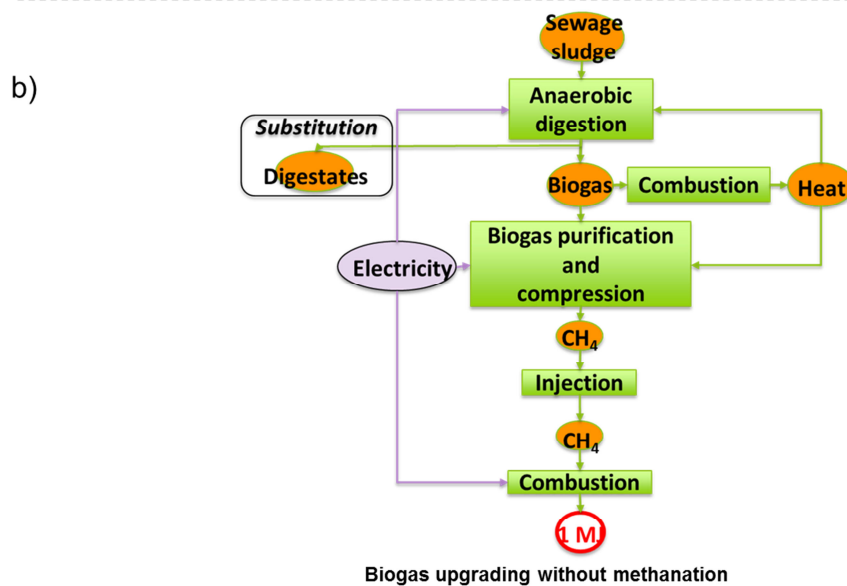
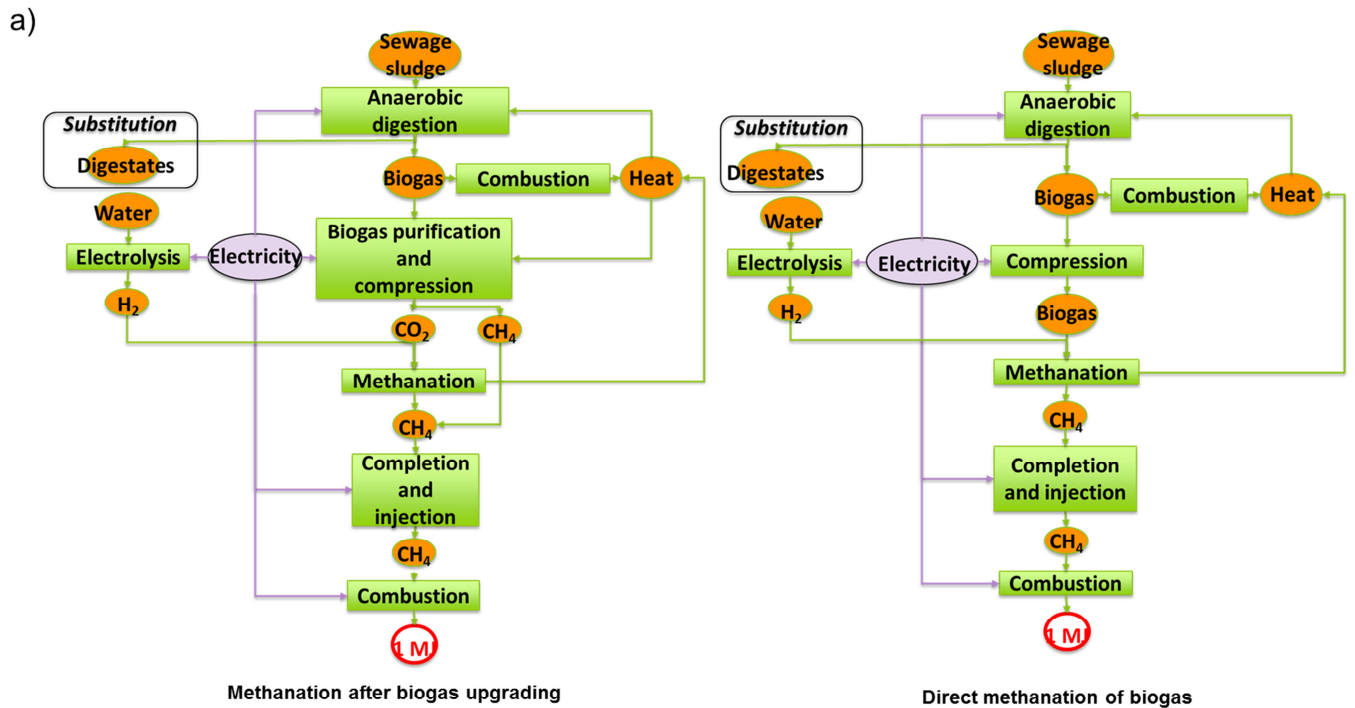
72 total production of CH₄ from biogas by combining carbon dioxide (CO₂) contained in the biogas and
73 converted to CH₄ via methanation with CH₄ already present in the biogas (Mohseni et al. 2012; Reiter
74 & Lindorfer 2015; Zoss et al., 2016). PtG is defined here as using electricity to convert water into
75 hydrogen (H₂) by electrolysis, and then to synthesize methane from carbon dioxide (CO₂) and H₂
76 through methanation (Sabatier reaction). H₂ production from electrolysis plays a key role in
77 integrated energy system (Yan, 2016), as fuel for transportation for instance (Nistor et al., 2016).
78 Furthermore, the synthesis of methane through methanation can strongly contribute to large scale
79 energy storage, as CH₄ injection is not limited in the gas grid, contrary to hydrogen which also faces
80 process and safety management issues.

81 Both economic and environmental criteria are crucial to fully assess the relevance of a new
82 technology. In this study we propose a model for CH₄ production from PtG that evaluates these two
83 dimensions of sustainability, by taking into account intermittent operation functioning. Economic
84 assessment is done by calculating capital expenditures (CAPEX) and operational expenditures (OPEX)
85 for each analyzed configuration, as in de Boer et al., (2014). Economic assessment of methane
86 production from PtG have been proposed recently (Götz et al., 2016), with sometimes the use of
87 time dependent optimization approach (Rivarolo et al., 2014). Environmental evaluation is done with
88 Life Cycle Assessment (LCA), a standardized tool allowing to assess the environmental impacts of the
89 whole cycle of a process, from raw extraction to final wastes management (ISO 2006). It should be
90 noticed that it is an attributional study (and not consequential), which means that changes in the
91 techno-economic sphere induced by large-scale production of bioCH₄ from PtG are not considered
92 (Ekvall & Weidema 2004). In particular, this implies that the use of long-term marginal data to
93 characterize the electric mix is not in the scope of the study (Amor et al. 2014). Several LCAs of H₂
94 production have been done in the past years (Dufour et al. 2012; Dufour et al. 2009; Patyk et al.
95 2013; Cetinkaya et al. 2012), with sometimes a focus on the use of electricity from renewables (Mori
96 et al. 2014). Nevertheless, only a few environmental studies have been conducted on CH₄ production
97 by PtG: GHG emissions of used electricity for electrolysis in Jentsch et al., (2011) or direct carbon
98 emissions (de Boer et al. 2014), and only one Life Cycle Approach to evaluate GHG emissions (Reiter
99 & Lindorfer 2015). To our knowledge, this is the first time that both economic and environmental
100 assessments are performed together to evaluate the production of bioCH₄ from PtG technology with
101 continuous and intermittent operations modes.

102 **2 Methodology**

103 **2.1 Overview of the system**

104 PtG consists in the conversion of electricity into H₂ and further into CH₄, which can be stored without
105 restriction in the natural gas network. CH₄ can then be further used for different purpose: storage of
106 electricity, heat production, raw materials for chemical industry and transportation services via NGV
107 production. In this paper, we will focus on the production of heat from CH₄, as it is the main use of
108 natural gas, from two different sources: from biogas upgrading, with no valorization of the CO₂, or
109 from biogas upgrading and CO₂ conversion into CH₄ via methanation (Figure 1). Different
110 configurations are analyzed : CH₄ from methanation after biogas upgrading, CH₄ from methanation
111 of the biogas without upgrading (direct methanation), and finally CH₄ from biogas upgrading without
112 methanation. A fossil reference system (natural gas) is also included in the LCA. The system assessed
113 in the model includes all steps from biogas production to CH₄ combustion in a boiler: anaerobic
114 digestion of sewage sludge, biogas upgrading and compression, electrolysis, methanation,
115 completion, injection in the gas network, and combustion. The wastewater treatment plant of the
116 study is designed for a population equivalent of 300 000 inhabitants. The daily quantity of sewage
117 sludge processed is 16 440 kg of dry matter, which leads to a biogas production of 230 m³.h⁻¹. The
118 anaerobic digestion plant, as well as the electrolysis and the methanation installations are supposed
119 to be near the wastewater treatment plant. The inventory is based on figures derived from academic
120 resources, internal communications with industrials and processes described in the Ecoinvent
121 database (Frischknecht et al. 2007). The location of the system is in France; as a consequence the
122 electric mix is the French one, with a low carbon content. Infrastructures are included in the
123 economic assessment but are not considered in the LCA, in line with the assessment of renewable
124 energy production in The Renewable Energy Directive (Parliament 2009).



125

126 **Figure 1: Overview of biomethane production with (a) and without (b) CH₄ synthesis via methanation**

127 **2.2 Anaerobic digestion step**

128 The data used for the functioning of the anaerobic digestion plant are summed up in Table 1. The
 129 heat necessary for operating the anaerobic digestion plant is provided by burning part of the
 130 produced biogas in a boiler and by the recovery of wasted heat from methanation when it is
 131 functioning. Compositions of the biogas and the digestates come from literature survey and from
 132 Suez Environment internal data. Biogenic CH₄ emissions occur during biogas production and
 133 digestates storage, and NH₃ and N₂O emissions are linked with the spreading of the digestates on the

134 fields. Biogenic CO₂ fixation and emission are not taken into account, and it is supposed that the
 135 carbon of the digestates is entirely reemitted in the environment.

136 **Table 1: Main assumptions at the anaerobic digestion step**

Description	Values	Reference
Inputs		
Sewage sludge	685 kg.h ⁻¹	
Electricity consumption	336 MJ.h ⁻¹	(Frank et al. 2012)
Heat consumption	1343 MJ.h ⁻¹	(Frank et al. 2012)
Outputs		
Biogas	230 m ³ .h ⁻¹ 0.220 L.g ⁻¹ SewSlu CH ₄ : 65.63%mol / CO ₂ : 34.37%mol T: 40°C / P: 1 bar	Present data
Digestates	685 kg.h ⁻¹ N:55 g.kg ⁻¹ Dig / P: 48.5 g.kg ⁻¹ Dig / K: 4.28 g.kg ⁻¹ Dig	(ADEME 2011)
Methane (biogenic) emissions	3 135 gCH ₄ .h ⁻¹	(Amon et al. 2006)
N ₂ O emissions	808 gN ₂ O.h ⁻¹	(Foley et al. 2010)
N-NH ₃ emissions	7 809 gN-NH ₃ .h ⁻¹	(Foley et al. 2010)

137 **2.3 Biogas upgrading and compression steps**

138 In the present study, the impurities contained in the biogas (especially sulphur and silicone derived
 139 compounds) are removed via activated carbon adsorption, this latter step being considered as
 140 integrated with the biogas production. Biogas is thus considered as composed of CH₄ and CO₂ and
 141 biogas upgrading focus on CO₂ removal. Four different technologies are considered for biogas
 142 upgrading, they are listed in Table 2 with their main characteristics. This table is based on different
 143 publications (Ryckebosch et al. 2011; Kaparaju 2013; IEA 2009; IEA 2014), and gives gross ranges for
 144 each of biogas upgrading technologies. The color code is the following: green is used for a
 145 characteristic that gives an specific advantage to the technology, especially in the context of
 146 downstream use of CO₂ in the methanation process, and on the contrary orange is used for
 147 drawbacks. For instance CO₂ outlet pressure after biogas upgrading can be around 1.8 bar (amines),
 148 which represents a relative benefit as there slightly less electricity is needed for CO₂ compression
 149 before methanation compared to the other upgrading technologies. While water scrubbing
 150 technology is the favorite technology for biogas upgrading, one observes that the second preferred
 151 technology is the chemical absorption. For the latter process, the main drawback is the heat
 152 requirement at the regeneration step, a well-known drawback in the post-combustion carbon
 153 capture community (Steenveldt et al. 2006). However, in the present case, where heat can be
 154 supplied by methanation, this drawback can easily be overcome, and even turned into an advantage.
 155 Similarly the methane loss of membrane technology is not anymore an issue since the CO₂ containing
 156 some CH₄ will be converted into CH₄ at the methanation step. Furthermore, membrane technology is
 157 considered as the easiest to operate which can be a major advantage for such small size units for
 158 which maintenance and labor operating cost must be minimum (Makaruk et al. 2010).

Table 2: Comparison of upgrading technologies.

	Water Scrubber	Pressure Swing Adsorption	Chemical absorption (amines)	Membrane
General characteristics				
Electricity demand (kWh.m ⁻³ _{BG})	0.2 -0.3	0.2 - 0.25	0.15	0.25
Heat demand (°C)	no	no	120-160	no
Operation pressure (bar)	5 - 10	4 - 7	0.1	5 - 10
Methane recovery (%)	98	98	99.96	80-99.5
Methane content in upgraded gas (%mol)	>97	>96	>99	>95
Exhaust gas treatment	yes	yes	no	yes
Water demand	yes (+)	no	yes	no
Demand on chemicals	no	no	yes	no
Ease of operation	complex	intermediate	complex	easy
Total costs (€/m ³ _{CH4})	0.13	0.25	0.17 - 0.28	0.12 - 0.22
Number of units				
number 2011	48	41	31	6
number 2013	113	58	61	27
Integration with methanation				
Heat integration	No	No	Yes (similar ΔH)	No
CO ₂ pressure	1	<1	1.8	1

160

161 Amines scrubbing and membranes are thus the two different upgrading technologies that have been
162 assessed in the present study. Chemical (amines) scrubbing corresponds to the formation of chemical
163 bonds between the CO₂ and a solvent, in general an aqueous solution of alkanolamines.
164 Regeneration of the solvent is endothermic, and required heat is provided by burning part of the
165 biogas or, preferentially in the present case, by the recovery of wasted heat from methanation when
166 it is functioning. Monoethanolamine (MEA) has been considered for solvent since it is a well-known
167 industrial solvent characterized as a highly reactive high energy requirement solvent. The first
168 characteristic is in favor of low capital expenditures (CAPEX) while the second is in favor of high
169 operational expenditures (OPEX) (Raynal et al. 2011). In the present case, since heat is available at
170 methanation step, CAPEX optimization was thought more important than OPEX optimization.
171 Ammonia and acetaldehyde emissions due to oxidative and thermal degradation of the solvents have
172 been taken into account. In membranes separation, pressurized biogas is passed through a
173 membrane which is selective for CO₂. It is important to notice that depending on the chosen
174 upgrading technology, CH₄ content in the CO₂ flow strongly varies. A scenario without biogas
175 upgrading (direct methanation) is also assessed in this study. In all the cases previously described, a
176 supplementary step of compression of the outputs of the upgrading process is needed before the
177 methanation step. All the assumptions for biogas upgrading and compression steps are presented in
178 Table 3.

179

Table 3: Main assumptions at the biogas upgrading and compression steps

Description	Values	Reference
Amines scrubbing and CO₂ compression		
Inputs		
Biogas	230 m ³ h ⁻¹	

Electricity consumption	94.3 MJ.h ⁻¹	Present data and (Starr et al. 2012)
Heat consumption	421 MJ.h ⁻¹	Present data
Water	126 kg.h ⁻¹	Present data
MonoEthanolAmines	11.5 g.h ⁻¹	Present data and (Reiter & Lindorfer 2015)
Outputs		
CO ₂ outAmines	80 m ³ .h ⁻¹ CO ₂ : 97.27%mol / H ₂ O: 2.73%mol T: 150°C / P: 13.3 bar	Present data
CH ₄ outAmines	156 m ³ .h ⁻¹ CH ₄ : 96.79%mol / CO ₂ : 0.98%mol / H ₂ O: 2.23%mol T: 50°C / P: 5.6 bar	Present data
Ammonia	16 g.h ⁻¹	(Reiter & Lindorfer 2015)
Acetaldehyde	423 g.h ⁻¹	(Reiter & Lindorfer 2015)
Membranes and CO₂ compression		
Inputs		
Biogas	230 m ³ .h ⁻¹	
Electricity consumption	152 MJ.h ⁻¹	Present data
Outputs		
CO ₂ outMb	95.1 m ³ .h ⁻¹ CO ₂ : 81.95%mol / CH ₄ : 18.05%mol T: 150°C / P: 13.3 bar	Present data
CH ₄ outMb	135 m ³ .h ⁻¹ CH ₄ : 99.15%mol / CO ₂ : 0.85%mol T: 50°C / P: 17 bar	Present data
Direct: Biogas compression		
Inputs		
Biogas	230 m ³ .h ⁻¹	
Electricity consumption	108 MJ.h ⁻¹	Present data
Outputs		
BiogasCompToMetha	225.5 m ³ .h ⁻¹ CH ₄ : 65.63%mol / CO ₂ : 34.37%mol T: 144°C / P: 15.3 bar	Present data
BiogasComp	4.5 m ³ .h ⁻¹ CH ₄ : 65.63%mol / CO ₂ : 34.37%mol T: 144°C / P: 15.3 bar	Present data

180 2.4 Electrolysis step

181 Water electrolysis from renewable energy is a sustainable method for H₂ production. It consists in
182 splitting water into H₂ and oxygen (O₂). Inputs and outputs for this reaction are given in Table 4. Two
183 main technologies are available on the market: alkaline or polymer electrolyte membrane (PEM)
184 electrolysis. PEM water electrolysis is much more expensive, due to high capital cost of the cell stack
185 compared to alkaline liquid electrolyte water electrolysis (Leng et al. 2012). On the other hand,
186 intermittent operation of alkaline electrolysis is challenging (Reiter & Lindorfer 2015). Nevertheless
187 in the latest alkaline electrolysis technologies, operating on intermittent renewable electricity was
188 found to be durable (Graves et al. 2011). Consequently we chose alkaline electrolysis in this study.

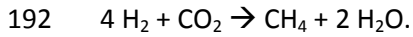
189 **Table 4: Main assumptions at the electrolysis step**

Description	Values	Reference
Inputs		
Tap water	252 kg.h ⁻¹ T: 15°C / P: 1 bar	
Electricity consumption	5634 MJ.h ⁻¹	Present data based on DNV, (2013), Wang et al., (2014) and Alpheia, (2015)

Outputs		
H ₂ fromElectrolysis	312 m ³ .h ⁻¹ T: 40°C / P: 15 bar	Present data based on DNV, (2013), Wang et al., (2014) and Alpheia, (2015)
O ₂ fromElectrolysis	156.5 m ³ .h ⁻¹ T: 40°C / P: 15 bar	Present data based on DNV, (2013), Wang et al., (2014) and Alpheia, (2015)

190 2.5 Methanation step

191 Methanation consists in synthetizing CH₄ from CO₂ and H₂ (Sabatier reaction):



193 In this study, H₂ comes from water electrolysis, and CO₂ is supplied by two ways: as almost pure CO₂
194 from upgrading step (by amine scrubbing or membrane separation), or mixed with CH₄ in the direct
195 methanation scenario. Methanation can be done by chemical or biological catalysis (Burkhardt &
196 Busch 2013). In this study, fixed-bed technology is used, with chemical catalysts mainly composed of
197 nickel (Ocampo et al. 2009; Wang et al. 2011). Methanation reaction is exothermic, with a heat
198 production of 0.165 MJ.mol⁻¹ of input CO₂ (Hoekman et al. 2010). Produced heat is used to
199 regenerate the solvent in biogas upgrading by amines scrubbing and / or to heat the anaerobic
200 digesters. Required data are presented in Table 5, and are in line with recent work of Reiter and
201 Lindorfer (2015).

202

Table 5: Main assumptions at the methanation step

Description	Values	Reference
CO₂ from amines scrubbing		
Inputs		
CO ₂ outAmines	80 m ³ .h ⁻¹	
Electricity consumption	27.2 MJ.h ⁻¹	Present data
H ₂ fromElectrolysis	310.2 m ³ .h ⁻¹	Present data
Outputs		
CH ₄ outMethaAmines	80 m ³ .h ⁻¹ CH ₄ : 96.56%mol / H ₂ : 1.69%mol / H ₂ O: 1.33%mol / CO ₂ : 0.80%mol T: 50°C / P: 9.5 bar	Present data
HeatoutMethaAmines	615.9 MJ.m ⁻³ CO ₂ outAmines	Present data
CO₂ from membranes		
Inputs		
CO ₂ outMb	95.1 m ³ .h ⁻¹	
Electricity consumption	33.3 MJ.h ⁻¹	Present data
H ₂ fromElectrolysis	312 m ³ .h ⁻¹	Present data
Outputs		
CH ₄ outMethaMb	98 m ³ .h ⁻¹ CH ₄ : 96.7%mol / H ₂ : 1.2%mol / H ₂ O: 1.1%mol / CO ₂ : 0.3%mol T: 50°C / P: 9.5 bar	Present data
HeatoutMethaMb	721 MJ.h ⁻¹	Present data
Biogas		
Inputs		
BiogasCompToMetha	225.5 m ³ .h ⁻¹	
Electricity consumption	106 MJ.h ⁻¹	Present data
H ₂ fromElectrolysis	310.2 m ³ .h ⁻¹	Present data
Outputs		
CH ₄ outMethaBiogas	230.9 m ³ .h ⁻¹ CH ₄ : 97.4%mol / H ₂ : 1.6%mol / H ₂ O: 1.3%mol / CO ₂ : 0.4%mol	Present data

	T: 50°C / P: 9.5 bar	
HeatoutMethaBiogas	586.3 MJ.m ³ Biogas	Present data

203 2.6 Completion, injection and combustion steps

204 Before injecting the bioCH₄ in the natural gas network, a completion step is necessary in order to
 205 bring the dew point to existing specifications (here 40°C and 5 bar). Electricity consumptions of this
 206 operation for the three proposed scenarios are given in Table 6.

207 **Table 6: Main assumptions at the completion step**

Description	Values	Reference
CH₄ from CO₂ amines		
Inputs		
CH ₄ outMethaAmines + CH ₄ outAmines	80 m ³ h ⁻¹ + 156 m ³ h ⁻¹	
Electricity consumption for completion	47.2 MJ.h ⁻¹	Present data
Outputs		
CH ₄ CompletionAmines	232 m ³ h ⁻¹ CH ₄ : 98.54%mol / H ₂ : 0.58%mol / CO ₂ : 0.80%mol / H ₂ O: 0.08%mol T: 40°C / P: 5 bar	Present data
CH₄ from CO₂ membranes		
Inputs		
CH ₄ outMethaMb + CH ₄ outMb	98 m ³ h ⁻¹ + 135 m ³ h ⁻¹	
Electricity consumption for completion	37.3MJ.h ⁻¹	Present data
Outputs		
CH ₄ CompletionMb	232 m ³ h ⁻¹ CH ₄ : 98.58%mol / H ₂ : 0.68%mol / CO ₂ : 0.66%mol / H ₂ O: 0.08%mol T: 40°C / P: 5 bar	Present data
CH₄ from Biogas		
Inputs		
CH ₄ outMethaBiogas + CH ₄ outBiogas	230.9 m ³ h ⁻¹ + 4.5 m ³ h ⁻¹	
Electricity consumption for completion	40 MJ.h ⁻¹	Present data
Outputs		
CH ₄ CompletionBiogas	233 m ³ h ⁻¹ CH ₄ : 97.8%mol / H ₂ : 1.15%mol / CO ₂ : 0.97%mol / H ₂ O: 0.08%mol T: 40°C / P: 5 bar	Present data

208
 209 At last, data for injection and combustion have been directly taken from the Ecoinvent 2.2 database,
 210 with biogenic CO₂ emissions instead of fossil CO₂ emissions. .

211 2.7 Economic data

212 In Table 7, operating expenditure (OPEX) and capital expenditure (CAPEX) for all the processes are
 213 presented. OPEX corresponding here to electricity consumption can be calculated from previous
 214 tables, and for different electricity prices. CAPEX of each process (upgrading, electrolysis,
 215 methanation and injection) have been evaluated by IFPEN and are specific results of this study. The
 216 capital cost estimations have been assumed to be in the range of -30% / + 50%.

217 Since the biogas production process has not been modeled from an economic point of view, the
 218 biogas production price has been estimated. We have supposed that biomethane injection in the

219 network allows an internal rate of return (IRR) equal to 10% on 15 years. Given the feed-in tariffs of
 220 bioCH₄ - fixed by decree in France in 2014
 221 (<http://www.legifrance.gouv.fr/affichTexte.do?cidTexte=JORFTEXT000024833895>) and taking into
 222 account the costs of the final steps - biogas upgrading and injection – this leads to a biogas
 223 production cost equal to 0.36 €/m³.

224 **Table 7: Main assumptions for the CAPEX and the OPEX**

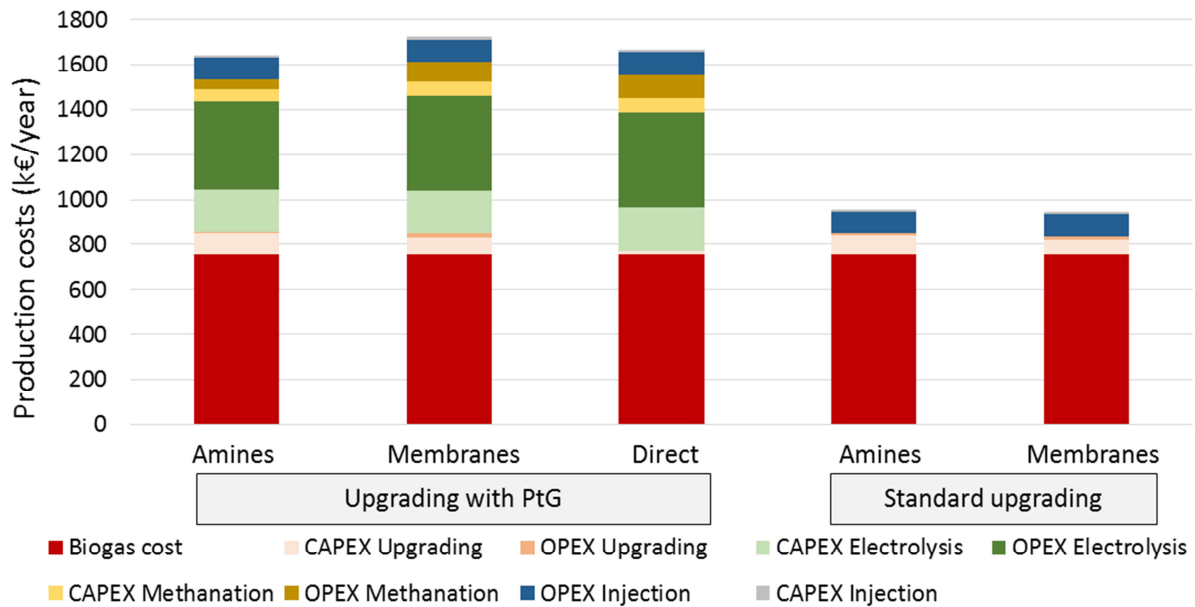
Process		OPEX	CAPEX	References
Biogas		0.36 €/m ³ Biogas	-	
Electrolysis		Electricity consumption + 100 k€/year ⁻¹ for maintenance	1000 €/kW ⁻¹	Present data based on DNV, (2013), ADEME, (2014), IEA, (2015) and Götz et al. (2016)
Amines	Separation	Electricity consumption + 0.8 €/hour ⁻¹	1050 k€, i.e. 4570 €/m ³ .h ⁻¹	Present data, in line with 5000 €/m ³ .h ⁻¹ (Bauer et al., 2013 and TUV, 2012) for a capacity of 250 m ³ .h ⁻¹
	Methanation	Electricity consumption + 6.2 €/hour ⁻¹	400 k€	Present data, in line with Götz et al. (2016)
Membranes	Separation	Electricity consumption + 1.7 €/hour ⁻¹	750 k€, i.e. 3260 €/m ³ .h ⁻¹	Present data, in line with 3200 €/m ³ .h ⁻¹ (Bauer et al., 2013) for a capacity of 250 m ³ .h ⁻¹
	Methanation	Electricity consumption + 12.4 €/hour ⁻¹	650 k€	Present data, in line with Götz et al. (2016)
Direct methanation		Electricity consumption + 14.1 €/hour ⁻¹	650 k€	Present data, in line with Götz et al. (2016)
Injection		100 k€/year ⁻¹	130 k€	Present data based on DNV, (2013), ADEME, (2014) and IEA, (2015)

225 3 Results

226 3.1 Economic results

227 *Continuous operation*

228 Annualized production costs of can be seen on Figure 2, for an electricity price of 30 €/MWh⁻¹ and
 229 with an IRR equal to 10% on 15 years. There is an important difference of total annualized production
 230 costs with and without methanation: approximatively 1650 k€/year⁻¹ for upgrading with PtG, and 940
 231 k€/year⁻¹ for standard upgrading. The largest contribution to the production costs is the biogas
 232 production, (around 80% for upgrading technologies and 45% for PtG scenarios). This cost is the
 233 same in the five scenarios assessed (757 k€/year⁻¹), but as total production costs are much higher in
 234 the methanation scenarios, the contribution of biogas is lower. The higher costs of the methanation
 235 scenarios are mainly due to the electrolysis step. It represents more than 35% of the production
 236 costs, with a large share due to electricity consumption (70% of the electrolysis costs). At last for PtG
 237 production with CH₄ and CO₂ separation, upgrading and methanation costs are quite similar.



238

239

Figure 2: Contribution analysis of the annualized production costs

240

The contribution analysis of the production costs for 1 kWh of bioCH₄ from PtG or upgrading are

241

shown on Figure 3, for the same electricity price and IRR rate than the Figure 2. It appears that

242

production costs of the different options are quite the same: they range from a minimum of 0.096

243

€·kWh⁻¹ for PtG from biogas with direct methanation and with membrane separation, and a

244

maximum of 0.104 €·kWh⁻¹ for biogas upgrading with membrane separation. In fact, the higher

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production cost of the PtG technologies are counterbalanced by a higher bioCH₄ injection in the gas

246

network (41 m³·h⁻¹ on average for standard upgrading versus 138 m³·h⁻¹ on average for methanation

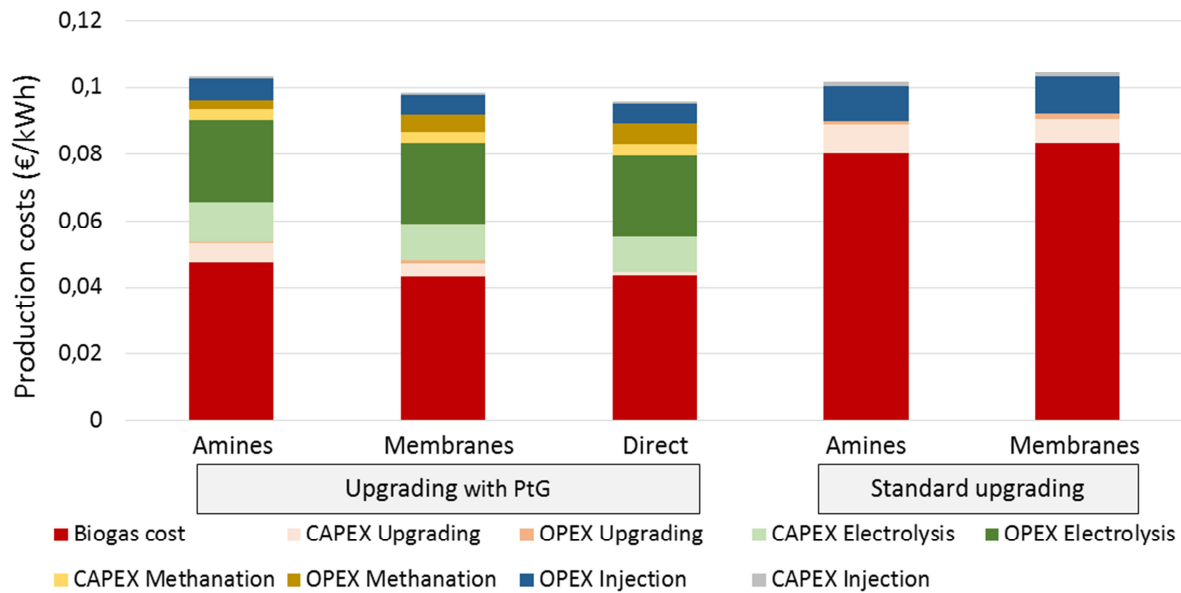
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scenarios). The respective contributions of the different steps are the same than for the annualized

248

production costs.

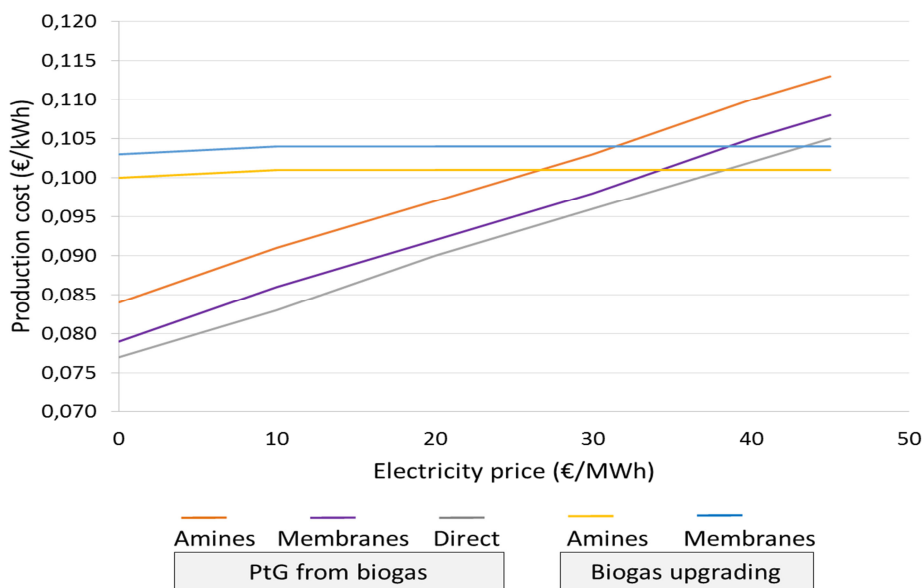
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250

251 **Figure 3: Contribution analysis of the production costs for 1 kWh injected in the gas network**

252 In Figure 4, overall production costs of the different scenarios are presented for electricity prices
 253 ranging from 0 to 45 €/MWh⁻¹. Costs of CH₄ from upgrading technologies are almost insensitive to
 254 electricity price, as they are mainly driven by the fixed biogas cost, and because the only other
 255 relevant OPEX corresponds to the injection step which is not correlated with the electricity price (see
 256 Table 7). On the contrary, PtG scenarios are very sensitive to the electricity price, with an increase of
 257 approximately 35% of the production costs, mainly because of the electricity consumption at the
 258 electrolysis step. Compared to production of CH₄ with the cheaper upgrading technology (amines
 259 scrubbing), CH₄ from PtG is competitive for electricity price below 27 €/MWh⁻¹ for separation with
 260 amines scrubbing, below 34 €/MWh⁻¹ with separation by membranes and below 38 €/MWh⁻¹ for
 261 direct methanation.

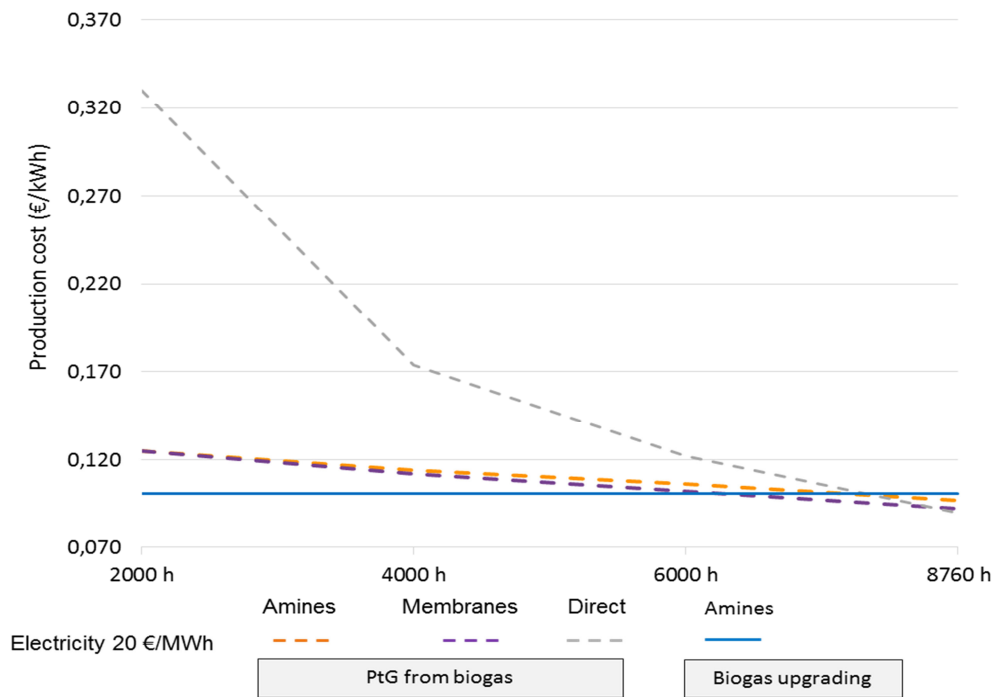


262

263 **Figure 4: Productions costs for 1 kWh injected in the gas network depending on the price of electricity**

264 *Intermittent operation*

265 As previously pointed out, PtG can be seen as a relevant way to store overproduced electricity, and
266 then intermittent operation should also be analyzed. According to Gahleitner (2013) the use of such
267 fluctuating power sources is satisfying, and is currently in use in a lot of PtG pilot plants. Economic
268 results for operation time varying between 2000 and 8760 hours (continuous operation) have been
269 assessed for PtG scenarios and CH₄ from amines scrubbing (Figure 5), with an electricity price equal
270 to 20 €.MWh⁻¹. Biogas upgrading is insensitive to the operation time. This can be explained by the
271 important contribution of the biogas production, and because others OPEX are almost exclusively
272 driven by the fixed cost of the rental of the injection station. On the opposite, costs of direct
273 methanation are very strongly connected with the operation time, with costs between 0.330 €.kWh⁻¹
274 for 2000 hours of operating time, and 0.090€.kWh⁻¹ for continuous operation. In fact in this
275 configuration CH₄ contained in the biogas is not recovered and injected in the gas grid when
276 methanation is not in operation. Consequently, the volume of energy produced is drastically reduced
277 with the increase of the intermittency. On the other hand, fixed costs (biogas production, all the
278 CAPEX and rental of the injection unit) are unchanged. Therefore the direct methanation leads to a
279 dramatic increase of the production costs with an intermittent functioning, and so this technology is
280 not suitable for non-continuous operation mode. Finally, like in the direct methanation, production
281 costs decrease for membranes and amines technologies of PtG with the increase of the operation,
282 but to a much lesser degree. Depending of the duration of the operation time, and for a given price
283 of electricity, production costs can be higher or lower than CH₄ from biogas upgrading. For instance,
284 for a electricity at 20 €.MWh⁻¹, operation time should be at least equal to 6160 hours for membranes
285 separation and to 7240 hours for amines separation to be competitive with biogas upgrading.

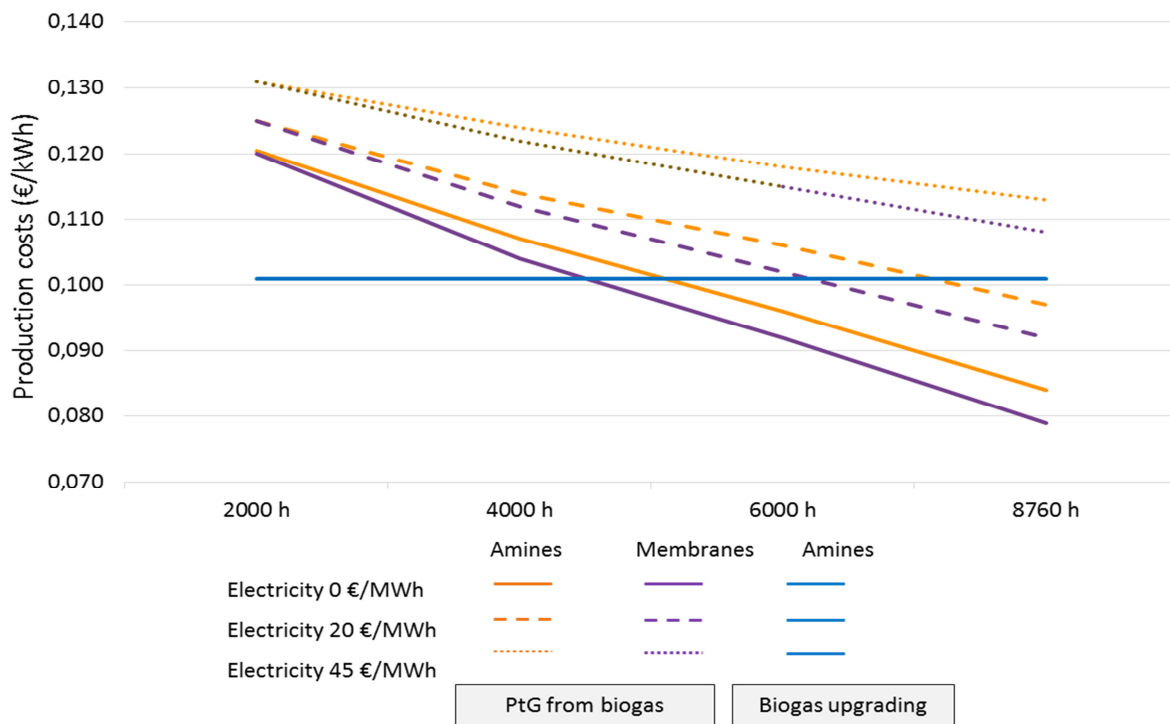


286

287 **Figure 5: Productions costs for 1 kWh injected in the gas network depending on the technology and the**
 288 **operating time**

289

290 Combined effect of electricity price and operation time have also been assessed (Figure 6).
 291 Depending on the price of electricity, the required operation time to be competitive with biogas
 292 upgrading can strongly vary. For instance, for a free electricity, operation time should be at least
 293 equal to 4445 hours for membranes separation and to 5232 hours for amines separation to be
 294 competitive with biogas upgrading. It means that the methanation unit should be in operation at
 295 least almost 4500 hours to counterbalance the fixed costs and the CAPEX. At last, with an electricity
 296 price equal to 45 €.MWh⁻¹, PtG technologies cannot compete with biogas upgrading. We also clearly
 297 see on Figure 6 that the operation cost difference between PtG from amines and from membranes
 298 increase with the operation time.



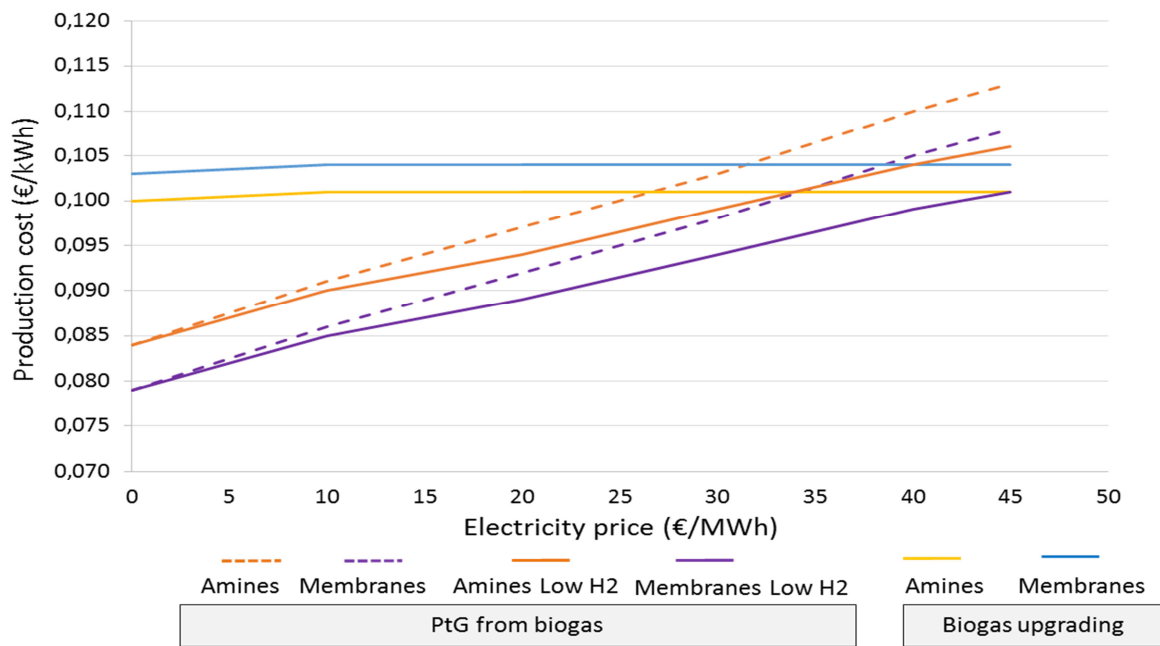
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300 **Figure 6: Productions costs for 1 kWh injected in the gas network depending on the price of electricity and**
 301 **the operating time**

302 *Sensitivity analysis*

303 As it has been underlined in the contribution analysis, electrolysis and more precisely the electricity
 304 consumption of this step, is one of the main contributor to the production costs of CH₄ from PtG.
 305 Thus, we conduct a sensitivity analysis on the electricity consumption for H₂ production. We discuss a
 306 favorable scenario, with a reduction of 25% of the electricity consumption (*i.e* 3.75 kWh.m⁻³ H₂). This
 307 value is in line with predicted values of future electricity consumptions around 3 kWh.m⁻³ H₂
 308 (Udagawa et al. 2007; Zeng & Zhang 2010). The evolution of production costs with low electricity
 309 consumption scenarios and for different electricity prices is shown on Figure 7. As expected, the
 310 more electricity is expensive, the more electricity consumption reduction has an important effect on
 311 the diminution of the production costs. For a price of electricity of 10 €.MWh⁻¹, the decrease of the
 312 production cost is only equal to 0.001 €.kWh⁻¹, but with an electricity at 45 €.MWh⁻¹, the reduction of
 313 the production cost is equal to 0.007 €.kWh⁻¹. Consequently, the minimum price of electricity so that
 314 CH₄ from PtG can be competitive with CH₄ from upgrading increases: it reaches 35 €.MWh⁻¹ for
 315 amines scrubbing and is around 45 €.MWh⁻¹ for membranes separation.

316



317

318 **Figure 7: Productions costs for 1 kWh injected in the gas network depending on the price of electricity and**
 319 **the electricity consumption for H₂ production**

320

321 **3.2 Environmental impacts**

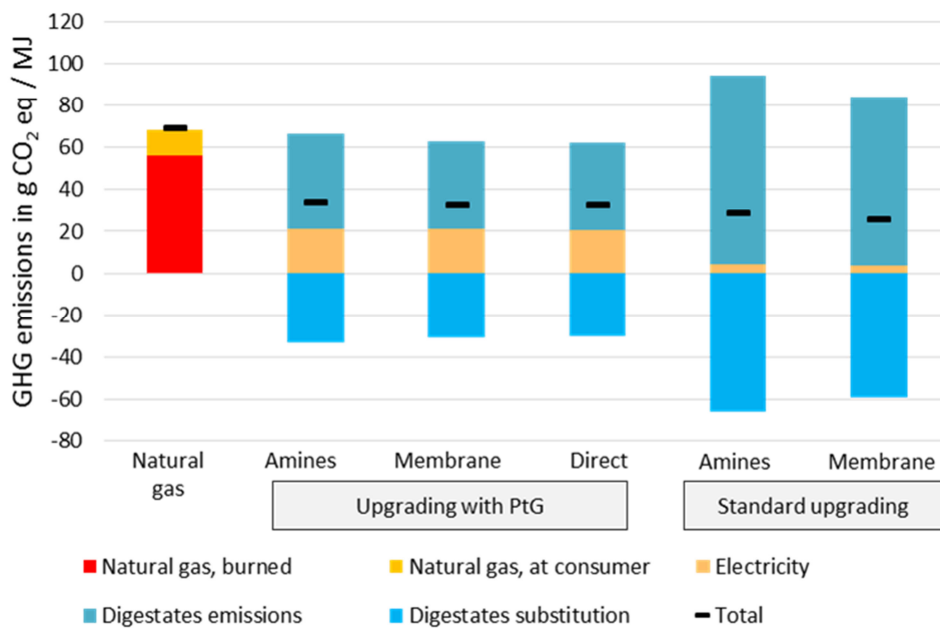
322 Environmental impacts of the study have been assessed within the methodological framework of the
 323 LCA based on a “cradle to grave” inventory of emissions and resources consumption. The considered
 324 functional unit in the study is one MJ produced by combustion of CH₄ in a boiler, based on the lower
 325 heating value of CH₄ (50.0 MJ.kg⁻¹). As one of the main objective is to reduce GHG emissions in the
 326 atmosphere with respect to fossil fuels, we therefore consider the climate change impact. It has been
 327 assessed with the characterization factors provided by the fourth IPCC report for a temporal horizon
 328 of 100 years (IPCC 2007) . It should be underlined that the characterization factor of CH₄ in case of
 329 methane loss has to be adapted. Since the carbon of the CH₄ comes from the sewage sludge (and
 330 previously from the atmosphere), the characterization factor must be the one of biogenic CH₄, which
 331 is slightly below the one of the fossil CH₄ (22.3 kg CO₂eq.kg⁻¹ vs. 25 kg CO₂eq.kg⁻¹).

332 As recommended in The Renewable Energy Directive (Parliament 2009), upstream impacts of sewage
 333 sludge are not taken into account . The same approach has been applied on LCA of sewage sludge
 334 valorization (Sadhukhan 2014; Yoshida et al. 2013). The substitution method has been used for
 335 digestates accounting, in accordance with the ISO guidelines, which suggest to choose the
 336 substitution instead of the allocation when it is possible. It corresponds to an expansion of the
 337 system boundaries in order to consider the impacts engendered by the by-products. Therefore, the
 338 use of digestates as fertilizers are supposed to save the equivalent amount of mineral fertilizers for
 339 N, P and K. In Figure 8, Figure 9 and Figure 11, the legend “digestates substitution” corresponds to

340 this environmental credit, and “digestates emissions” to the emissions at the biogas plant and at the
 341 field.

342 *Continuous operation*

343 Principal contributors to GHG emissions for the different assessed technologies are shown on Figure
 344 8. From a global perspective, GHG emissions from PtG are almost equivalent for the three
 345 technologies, and higher than biogas upgrading. Nevertheless, they are still lower than GHG
 346 emissions from natural gas combustion. For the fossil scenario (natural gas burned in a boiler), most
 347 of the emissions correspond to fossil CO₂ from gas combustion. The remaining emissions are linked
 348 to resource extraction and transportation of the gas in the pipelines. In the scenarios with production
 349 of energy from sewage sludge, main GHG emissions come from the management of the digestates.
 350 Environmental burdens are associated with loss of CH₄ (40%) and emissions of N₂O (60%). On the
 351 other hand, negative impacts are due to the substitution of the digestates to the production and the
 352 use of mineral fertilizers. For PtG technologies, electricity consumption is also an important
 353 contributor to climate change. As the carbon content of the French electricity is particularly low due
 354 to the large part of the nuclear in the mix composition, a sensitivity analysis on the source of
 355 electricity (and as a consequence on its carbon content) is done in a following paragraph.



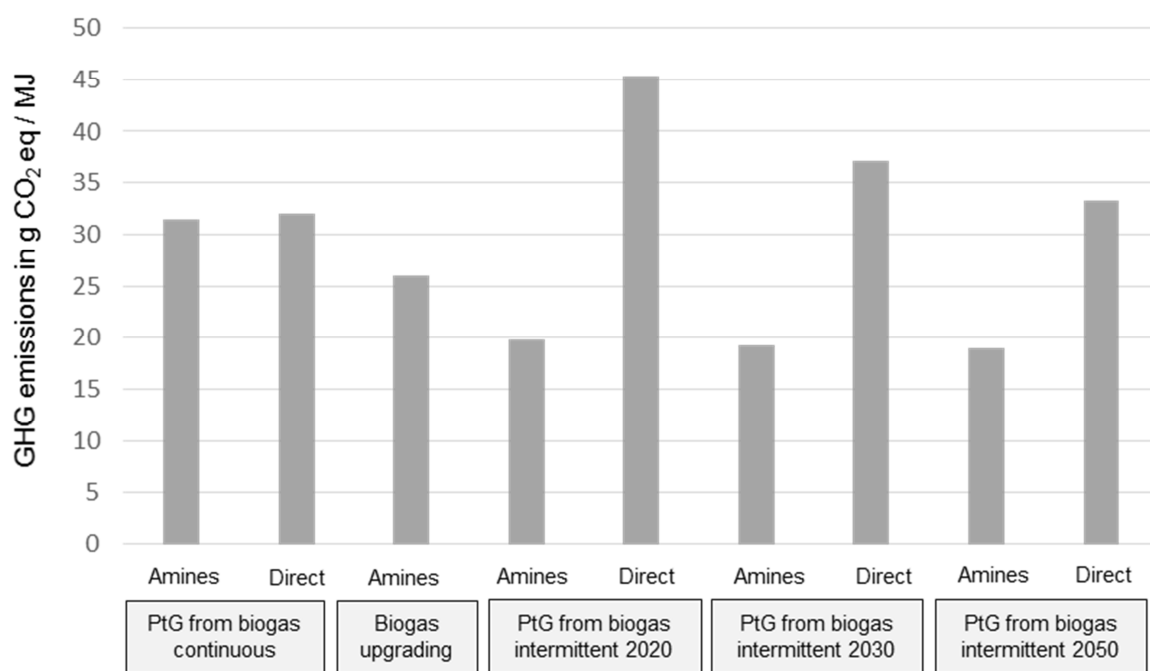
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357 **Figure 8: Contribution analysis of GHG emissions for 1 MJ injected in the gas network**

358 *Intermittent operation*

359 According to simulations based on ADEME 2030 – 2050 scenarios (ADEME 2012), hours of electricity
 360 overproduction occurrence was estimated at 28% of the year for 2020, 36% in 2030 and 41% in 2050,
 361 which is consistent with the ADEME’s study on methanation (ADEME 2014). The composition of

362 electricity surplus is 80% renewable and 20% nuclear by 2050. Figure 9 presents results for
 363 intermittent methanation during overproduction period. It is supposed that when methanation is not
 364 functioning, the non-injected gas (CO₂ with 18% of CH₄ in membrane separation and biogas with 65%
 365 of CH₄ in direct methanation) is flared, in order to reduce direct CH₄ emissions to the atmosphere. In
 366 this scenario, the results for membrane and amines separation are almost the same, so only amine
 367 technology will be represented for the next environmental impact assessment results.
 368 In direct technology configuration, biogas is not recovered and injected in the gas grid when
 369 methanation is not in operation, like in the economic assessment. This leads to GHG emissions which
 370 are much higher than continuous operation (and even higher than natural gas combustion) for the
 371 three time horizons analyzed. On the contrary, intermittent operation with electricity from
 372 renewable resources and amines separation leads to an important decrease compared to continuous
 373 operation with French average electricity mix (-37% in 2020 and -40% in 2050). For the three
 374 scenarios assessed, climate change impact of PtG is even lower than biogas upgrading.



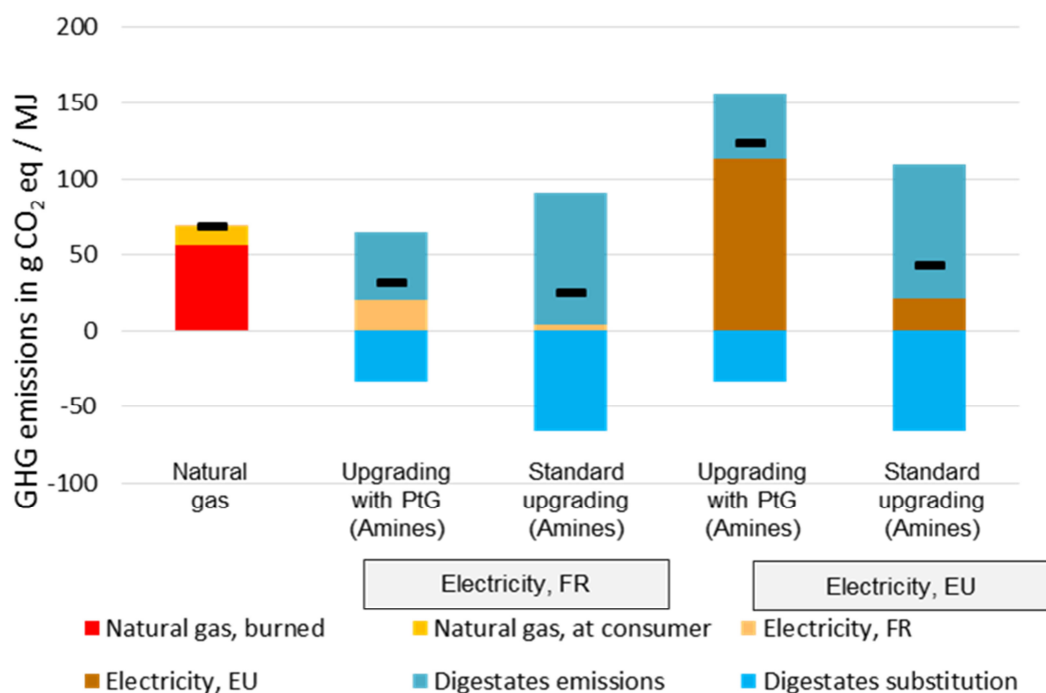
375
 376 **Figure 9: GHG emissions for 1 MJ injected in the gas network depending on the price of electricity and the**
 377 **operating time**

378 *Sensitivity analysis*

379 *Change in the electricity mix composition*

380 Results of using two different types of electricity are presented on Figure 10 (French electricity mix
 381 and European electricity mix). The results indicate that amines PtG technology are more sensitive to
 382 the composition of the electricity than biogas upgrading. The change from French electricity mix to
 383 European electricity mix induces an increase of 290% of the GHG emissions of PtG technology, and

384 only 70% of the emissions from biogas upgrading. This is due to the fact that PtG technologies
 385 consume much more electricity than upgrading technologies, combined with the different carbon
 386 contents of the electricity mix. In fact GHG emissions of 1 MJ of European electricity is 0.156 CO₂eq,
 387 and only 0.029 CO₂eq for French electricity, because French mix is mainly composed of nuclear
 388 electricity (78% according to the Ecoinvent database), with very low GHG emissions (0.00172
 389 CO₂eq.MJ⁻¹). Consequently, with the use of a European electricity mix (or with another average
 390 electricity mix with high GHG emissions, like China or United Kingdom), climate change impact of PtG
 391 technology becomes higher than natural gas, with the highest contribution coming from the
 392 electricity consumption. Thus electricity with low carbon content is a mandatory prerequisite to
 393 achieve a sustainable production of CH₄ from CO₂.



394

395 **Figure 10: Contribution analysis of GHG emissions for 1 MJ injected in the gas network depending on the**
 396 **electricity mix**

397 *Endpoints impacts*

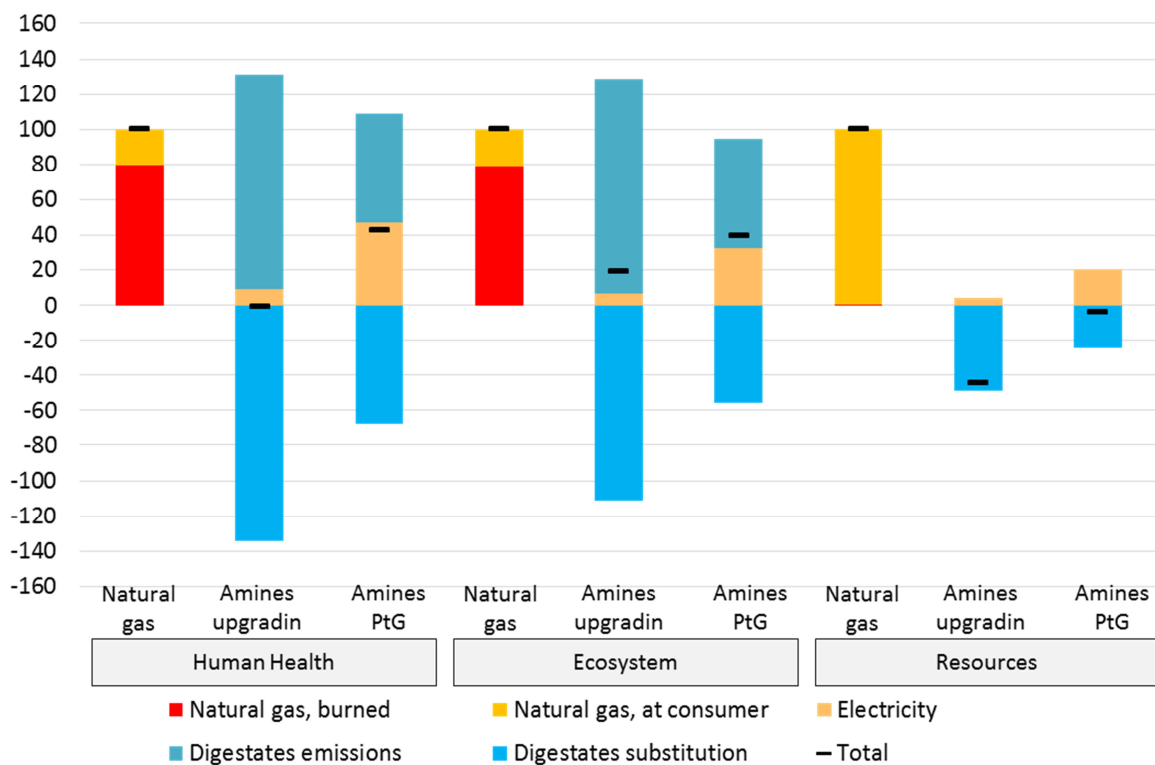
398 As pointed out by Reiter and Lindorfer, (2015), other impacts than climate change have never been
 399 assessed for biomethane produced from PtG. In this article, endpoint impacts have been assessed
 400 with the ReCiPe method with normalization values for Europe and the Hierarchist perspective
 401 (Goedkoop et al. 2009). This perspective is based on the most common policy principles with regard
 402 to time frame and other issues. The time horizon of the selected impact is in this case 100 years. The
 403 three assessed endpoint impacts are:

- 404 - Human health, defined as the sum of years of life lost and years of life disabled, and
 405 expressed in years.

- Ecosystem diversity, based on the loss of species during a certain time in a certain area as, and expressed as the potentially disappeared fraction of species (PDF) integrated over area (PDF x year).
- Resources availability, corresponding to the marginal increase in costs due to the extraction of a resource, and expressed in \$.

This method is easy to interpret because endpoints are directly associated with an area of protection, unlike midpoints impacts (Reap et al. 2008). On the other hand, it should be noticed that their level of uncertainty is higher than midpoints indicators (Bare et al. 2000). Results are shown on Figure 11, and each impact is standardized with the value of the worst scenario for this impact.

As for climate change, impacts of PtG are lower than the ones of natural gas, but higher than biogas upgrading for continuous operation. Contributors of human health and ecosystem impacts are quite the same than for climate change. For resources impact, contributors associated with pollutants emissions are not in the scope of the impact calculation. This leads to negative impact for bioenergy production, as only resource consumption for electricity generation and positive effect of mineral substitution of the mineral fertilizers by the digestates are taken into account.



421
422 **Figure 11: Contribution analysis of endpoints impacts (Human Health, Ecosystem and Resources) for 1 MJ**
423 **injected in the gas network**

424 4 Discussion

425 It must be underlined that this is a prospective LCA of a process that does not yet exist. Consequently
426 the systems described in this study can be subjected to important modifications. From a technical
427 point of view, some changes could be proposed to improve the economic and environmental balance
428 of this process. As biogas is used to heat the anaerobic digesters, the CO₂ resulting from the
429 combustion of this biogas can also be converted into bioCH₄ by methanation. The auto-consumption
430 of the biogas ranges from 21% (membrane upgrading) to 28% (amines upgrading), so the added
431 amount of bioCH₄ that could be injected in the gas network is far from being marginal, and could
432 potentially improve the overall performances of the process. About economic assumptions, biogas
433 production costs are based in this paper on French feed-in tariffs and an expected IRR. These values
434 may seem conservative, compared to data from literature (between 0.24 and 0.30 €.m⁻³ of biogas in
435 Patterson et al. (2011)). In the future, a more detailed approach could be investigated to consolidate
436 biogas production costs, for instance by including the biogas production process in the economic
437 assessment. However, as the same biogas production costs have been used for upgrading and PtG
438 technologies, it should be underlined that this inclusion would not change the general conclusion of
439 the economic study. It also has to be noticed that electricity price is strongly correlated with the
440 annual operation hour: average electricity price increases with the load factor (ENEA, 2016). In our
441 study, different electricity prices are proposed for different operation time. This has been done to
442 assess how the analyzed system responds to these two key parameters independently. Further work
443 could therefore integrate price duration curves for different locations and different time horizons in
444 order to map out more clearly potential prospective scenarios. To go deeper into details about the
445 economic feasibility of the combination of anaerobic digestion and PtG technology, the effect of the
446 introduction of an incentive for “green gas” production could be assessed. As underlined by
447 Guandalini et al. (2015), there is a need of specific incentive for PtG technologies in order to push the
448 competitiveness of this energy production system. They propose a “green gas” incentive between 0
449 and 20 €.MWh⁻¹. As this incentive is only related to the CH₄ from CO₂ conversion by methanation, its
450 amount is function of the proportion of bioCH₄ from methanation. In our system, CO₂ conversion into
451 bioCH₄ represent approximatively 50% of the total injected bioCH₄ for the amines scrubbing and the
452 membrane upgrading technologies. Applied to the case study described in section 3.1 (continuous
453 operation and an electricity price of 30 €.MWh⁻¹), an incentive of 20 €.MWh⁻¹ leads to a reduction of
454 the production costs of almost 10%. At last learning curves (or experience curves) could be used to
455 quantify the future reduction of investment costs for the different assessed technologies. Progress
456 ratios (PR, defined as the rates of unit cost decline with each doubling of cumulative production) are
457 expected to be around 0.88 (Junginger et al., 2006) for biogas plant and 0.82 for H₂ production by

458 water electrolysis (Schoots et al., 2008). It is in line with Anandarajah et al. (2013) who report PR
459 around 15-20% for new technologies. Other authors suggest more globally that investments costs
460 could be divided by 2 in 20 years for thermochemical processes and in 30 years for biochemical
461 processes (due to more important sources of process locks for biological manipulations). From the
462 environmental point of view, conversion of sewage sludge into bioCH₄, using methanation to valorize
463 the CO₂ of the biogas, can also be seen as a waste treatment process. Many LCAs have been
464 conducted in the past years to assess the environmental impacts of sewage sludge management
465 (Hospido et al. 2010; Sadhukhan 2014; Yoshida et al. 2013). A comparison with the most used
466 technology of sewage sludge valorization in France, i.e the production of electricity and heat by
467 cogeneration, could therefore be an interesting perspective.

468 **5 Conclusion**

469 In this study a techno economic and environmental assessment of bioCH₄ production via biogas
470 upgrading and PtG technology has been performed. Five different scenarios have been analyzed: two
471 biogas upgrading scenarios (with amines and membrane technologies), and three scenarios with the
472 use of PtG technologies to produce bioCH₄. These scenarios have been compared to a fossil
473 reference scenario of methane production from natural gas. Sensitivity analyzes have been done,
474 both on the economic and environmental assessments. From the economic point of view, PtG
475 technologies are competitive with upgrading ones for an average electricity price equal to 38 €/MWh¹
476 for direct methanation and separation by membranes. The lower competitiveness of amines
477 scrubbing is due to a lower volume of bioCH₄ produced, mainly because of the biogas auto-
478 consumption to produce heat for the amines regeneration. For intermittent operation, competitive
479 prices of electricity for methanation can be higher than the upgrading scenarios, depending on the
480 operation time. It should be pointed out that direct methanation cannot be used in intermittent
481 operation, as the produced energy is drastically reduced because of the biogas lost when
482 methanation is not functioning. A reduction of 25% of the electricity consumption at the electrolysis
483 step causes an important decrease of the production costs, especially for scenarios with high
484 average electricity prices. From an environmental perspective, impacts of PtG are higher than biogas
485 upgrading for continuous operation. Intermittent operation can severely lower the climate change
486 impact for non-direct methanation scenarios, as the consumed electricity is mainly based on
487 renewable sources. The change in the electricity mix composition can also have a strong influence on
488 GHG emissions. This work is a first attempt to assess both the economic and the environmental
489 impacts of PtG technologies, and highlights the main bottlenecks in this production. Here we focus
490 on a simplified process where only CO₂ from biogas was converted into BioCH₄, but the optimum

491 from both environmental and economic points of view may consist in a more complex process where
492 CO₂ from internal biogas combustion should also be turned into bioCH₄.

493

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