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Abstract

To decrease the use of fossil fuels and face the energetic demand, the integration of renewable energy is a necessary step. Part of this renewable energy can be supplied by the production of electricity from photovoltaic panels and windfarms. The massive use of these intermittent energies will lead to overproduction periods, and there is consequently a need to convert this surplus of electricity into a storable form of energy. Power-to-gas (PtG) technology consists in using electricity to convert water into hydrogen by electrolysis, and then to synthetize methane from carbon dioxide and hydrogen. Techno-economic and Life Cycle Assessment of methane production via the combination of anaerobic digestion and PtG technology have been applied to sewage sludge valorization. Process studies and equipment design have been addressed considering already available technologies. Sensitivity analyses have been done on biogas upgrading technologies, electricity prices, annual operation time and composition of the electricity mix with also a comparison between PtG and direct injection. It appears that the more the electricity is expensive, the longer the operation time of the methanation process must be to be competitive with injection of methane from biogas. Reduction of electricity consumption of the electrolysis step decreases production costs. Even if the current context does not feature adapted conditions to ensure an economically viable chain, the evolution of the energetic context in the next few years as well as the expected technological improvements will contribute to overall cost reduction. From an environmental point of view, continuous PtG generates more greenhouse gases than direct injection, but intermittent operation with use of renewable electricity can significantly reduce GHG emissions. From an endpoint impacts perspective, impact from continuous PtG are higher than biogas upgrading, but much lower than fossil energy. Future development of low electricity consumption of the electrolysis process, and integration of renewable credits from CO_2 valorization can increase the competitiveness of this technology.

Keywords: Life Cycle Assessment (LCA); Power-to-Gas; Methane; CO_2 valorization; Energy storage; Production costs

1 Introduction

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

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

To fulfill the needs of electricity demand with the integration of more renewables in the electricity mix, the installed capacity should be increased. This massive electricity production from renewable energies will lead to overproduction periods, where produced electricity would not be totally employed. Therefore there is a need to store this energy. Several electricity storage technologies are available like batteries, pumped hydropower plants, compressed air energy storage or hydrogen storage technologies, with different prices, run times and storage capacities (Hadjipaschalis et al. 2009). Pumped hydropower technology is the most massive storage technology available today, and is generally enough to balance the current electrical system. However it will be probably inadequate to store large amounts of overproduced electricity (ADEME 2014) and it has already been largely deployed. In this perspective, natural gas network offers a high storage capacity (for example 135 TWh in France, (DGEC (Direction générale de l'énergie et du climat) 2011)), and therefore conversion of electricity into gas would offer an interesting leverage to valorize overproduced electricity. In this perspective, production of methane (CH_4) by power-to-gas (PtG) technology can greatly increase the

total production of CH_4 from biogas by combining carbon dioxide (CO_2) contained in the biogas and converted to CH_4 via methanation with CH_4 already present in the biogas (Mohseni et al. 2012; Reiter & Lindorfer 2015; Zoss et al., 2016). PtG is defined here as using electricity to convert water into hydrogen (H_2) by electrolysis, and then to synthetize methane from carbon dioxide (CO_2) and H_2 through methanation (Sabatier reaction). H2 production from electrolysis plays a key role in integrated energy system (Yan, 2016), as fuel for transportation for instance (Nistor et al., 2016). Furthermore, the synthesis of methane through methanation can strongly contribute to large scale energy storage, as CH_4 injection is not limited in the gas grid, contrary to hydrogen which also faces process and safety management issues.

Both economic and environmental criteria are crucial to fully assess the relevance of a new technology. In this study we propose a model for CH_4 production from PtG that evaluates these two dimensions of sustainability, by taking into account intermittent operation functioning. Economic assessment is done by calculating capital expenditures (CAPEX) and operational expenditures (OPEX) for each analyzed configuration, as in de Boer et al., (2014). Economic assessment of methane production from PtG have been proposed recently (Götz et al., 2016), with sometimes the use of time dependent optimization approach (Rivarolo et al., 2014). Environmental evaluation is done with Life Cycle Assessment (LCA), a standardized tool allowing to assess the environmental impacts of the whole cycle of a process, from raw extraction to final wastes management (ISO 2006). It should be noticed that it is an attributional study (and not consequential), which means that changes in the techno-economic sphere induced by large-scale production of bioCH_4 from PtG are not considered (Ekvall & Weidema 2004). In particular, this implies that the use of long-term marginal data to characterize the electric mix is not in the scope of the study (Amor et al. 2014). Several LCAs of H_2 production have been done in the past years (Dufour et al. 2012; Dufour et al. 2009; Patyk et al. 2013; Cetinkaya et al. 2012), with sometimes a focus on the use of electricity from renewables (Mori et al. 2014). Nevertheless, only a few environmental studies have been conducted on CH_4 production by PtG: GHG emissions of used electricity for electrolysis in Jentsch et al., (2011) or direct carbon emissions (de Boer et al. 2014), and only one Life Cycle Approach to evaluate GHG emissions (Reiter & Lindorfer 2015). To our knowledge, this is the first time that both economic and environmental assessments are performed together to evaluate the production of bioCH_4 from PtG technology with continuous and intermittent operations modes.

2 Methodology

2.1 Overview of the system

PtG consists in the conversion of electricity into H_2 and further into CH_4, which can be stored without restriction in the natural gas network. CH_4 can then be further used for different purpose: storage of electricity, heat production, raw materials for chemical industry and transportation services via NGV production. In this paper, we will focus on the production of heat from CH_4, as it is the main use of natural gas, from two different sources: from biogas upgrading, with no valorization of the CO_2, or from biogas upgrading and CO_2 conversion into CH_4 via methanation (Figure 1). Different configurations are analyzed : CH4 from methanation after biogas upgrading, CH4 from methanation of the biogas without upgrading (direct methanation), and finally CH4 from biogas upgrading without methanation. A fossil reference system (natural gas) is also included in the LCA. The system assessed in the model includes all steps from biogas production to CH_4 combustion in a boiler: anaerobic digestion of sewage sludge, biogas upgrading and compression, electrolysis, methanation, completion, injection in the gas network, and combustion. The wastewater treatment plant of the study is designed for a population equivalent of 300 000 inhabitants. The daily quantity of sewage sludge processed is 16 440 kg of dry matter, which leads to a biogas production of 230 $m^3.h^{-1}$. The anaerobic digestion plant, as well as the electrolysis and the methanation installations are supposed to be near the wastewater treatment plant. The inventory is based on figures derived from academic resources, internal communications with industrials and processes described in the Ecoinvent database (Frischknecht et al. 2007). The location of the system is in France; as a consequence the electric mix is the French one, with a low carbon content. Infrastructures are included in the economic assessment but are not considered in the LCA, in line with the assessment of renewable energy production in The Renewable Energy Directive (Parliament 2009).

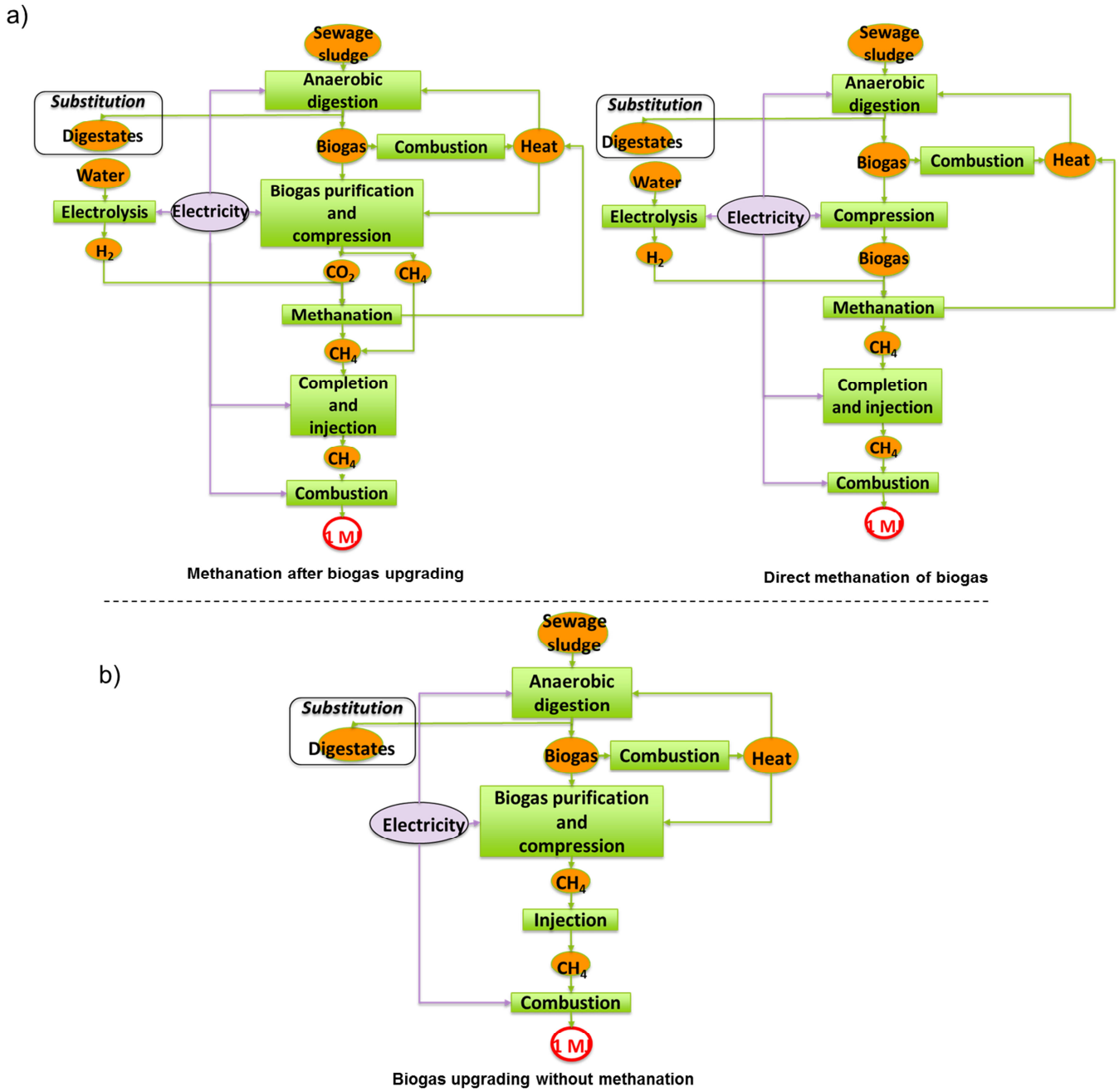

Figure 1: Overview of biomethane production with (a) and without (b) CH_4 synthesis via methanation

2.2 Anaerobic digestion step

The data used for the functioning of the anaerobic digestion plant are summed up in Table 1. The heat necessary for operating the anaerobic digestion plant is provided by burning part of the produced biogas in a boiler and by the recovery of wasted heat from methanation when it is functioning. Compositions of the biogas and the digestates come from literature survey and from Suez Environment internal data. Biogenic CH_4 emissions occur during biogas production and digestates storage, and NH_3 and N_2O emissions are linked with the spreading of the digestates on the

fields. Biogenic CO_2 fixation and emission are not taken into account, and it is supposed that the carbon of the digestates is entirely reemitted in the environment.

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 $m3.h^{-1}$ 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_2O emissions	808 $gN_2O.h^{-1}$	(Foley et al. 2010)
N-NH_3 emissions	7 809 gN-$NH_3.h^{-1}$	(Foley et al. 2010)

2.3 Biogas upgrading and compression steps

In the present study, the impurities contained in the biogas (especially sulphur and silicone derived compounds) are removed via activated carbon adsorption, this latter step being considered as integrated with the biogas production. Biogas is thus considered as composed of CH_4 and CO_2 and biogas upgrading focus on CO_2 removal. Four different technologies are considered for biogas upgrading, they are listed in Table 2 with their main characteristics. This table is based on different publications (Ryckebosch et al. 2011; Kaparaju 2013; IEA 2009; IEA 2014), and gives gross ranges for each of biogas upgrading technologies. The color code is the following: green is used for a characteristic that gives an specific advantage to the technology, especially in the context of downstream use of CO_2 in the methanation process, and on the contrary orange is used for drawbacks. For instance CO_2 outlet pressure after biogas upgrading can be around 1.8 bar (amines), which represents a relative benefit as there slightly less electricity is needed for CO2 compression before methanation compared to the other upgrading technologies. While water scrubbing technology is the favorite technology for biogas upgrading, one observes that the second preferred technology is the chemical absorption. For the latter process, the main drawback is the heat requirement at the regeneration step, a well-known drawback in the post-combustion carbon capture community (Steeneveldt et al. 2006). However, in the present case, where heat can be supplied by methanation, this drawback can easily be overcome, and even turned into an advantage. Similarly the methane loss of membrane technology is not anymore an issue since the CO_2 containing some CH_4 will be converted into CH_4 at the methanation step. Furthermore, membrane technology is considered as the easiest to operate which can be a major advantage for such small size units for 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

Amines scrubbing and membranes are thus the two different upgrading technologies that have been assessed in the present study. Chemical (amines) scrubbing corresponds to the formation of chemical bonds between the CO_2 and a solvent, in general an aqueous solution of alcanolamines. Regeneration of the solvent is endothermic, and required heat is provided by burning part of the biogas or, preferentially in the present case, by the recovery of wasted heat from methanation when it is functioning. Monoethanolamine (MEA) has been considered for solvent since it is a well-known industrial solvent characterized as a highly reactive high energy requirement solvent. The first characteristic is in favor of low capital expenditures (CAPEX) while the second is in favor of high operational expenditures (OPEX) (Raynal et al. 2011). In the present case, since heat is available at methanation step, CAPEX optimization was thought more important than OPEX optimization. Ammonia and acetaldehyde emissions due to oxidative and thermal degradation of the solvents have been taken into account. In membranes separation, pressurized biogas is passed through a membrane which is selective for CO_2. It is important to notice that depending on the chosen upgrading technology, CH_4 content in the CO_2 flow strongly varies. A scenario without biogas upgrading (direct methanation) is also assessed in this study. In all the cases previously described, a supplementary step of compression of the outputs of the upgrading process is needed before the methanation step. All the assumptions for biogas upgrading and compression steps are presented in Table 3.

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

Description	Values	Reference
Amines scrubbing and CO_2 compression		
Inputs		
Biogas	230 m^3h^{-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_2outAmines	80 $m^3.h^{-1}$ CO_2: 97.27%mol / H_2O: 2.73%mol T: 150°C / P: 13.3 bar	Present data
CH_4outAmines	156 $m^3.h^{-1}$ CH_4: 96.79%mol / CO_2: 0.98%mol / H_2O: 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 CO_2 compression		
Inputs		
Biogas	230 m^3h^{-1}	
Electricity consumption	152 $MJ.h^{-1}$	Present data
Outputs		
CO_2outMb	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_4outMb	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^3h^{-1}	
Electricity consumption	108 $MJ.h^{-1}$	Present data
Outputs		
BiogasCompToMetha	225.5 m^3h^{-1} CH_4: 65.63%mol / CO_2: 34.37%mol T: 144°C / P: 15.3 bar	Present data
BiogasComp	4.5 m^3h^{-1} CH_4: 65.63%mol / CO_2: 34.37%mol T: 144°C / P: 15.3 bar	Present data

2.4 Electrolysis step

Water electrolysis from renewable energy is a sustainable method for H_2 production. It consists in splitting water into H_2 and oxygen (O_2). Inputs and outputs for this reaction are given in Table 4. Two main technologies are available on the market: alkaline or polymer electrolyte membrane (PEM) electrolysis. PEM water electrolysis is much more expensive, due to high capital cost of the cell stack compared to alkaline liquid electrolyte water electrolysis (Leng et al. 2012). On the other hand, intermittent operation of alkaline electrolysis is challenging (Reiter & Lindorfer 2015). Nevertheless in the latest alkaline electrolysis technologies, operating on intermittent renewable electricity was found to be durable (Graves et al. 2011). Consequently we chose alkaline electrolysis in this study.

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 Alphea, (2015)

Outputs		
H_2fromElectrolysis	312 $m^3.h^{-1}$ T: 40°C / P: 15 bar	Present data based on DNV, (2013), Wang et al., (2014) and Alphea, (2015)
O_2fromElectrolysis	156.5 $m^3.h^{-1}$ T: 40°C / P: 15 bar	Present data based on DNV, (2013), Wang et al., (2014) and Alphea, (2015)

2.5 Methanation step

Methanation consists in synthetizing CH_4 from CO_2 and H_2 (Sabatier reaction):

$4\ H_2 + CO_2 \rightarrow CH_4 + 2\ H_2O$.

In this study, H_2 comes from water electrolysis, and CO_2 is supplied by two ways: as almost pure CO_2 from upgrading step (by amine scrubbing or membrane separation), or mixed with CH_4 in the direct methanation scenario. Methanation can be done by chemical or biological catalysis (Burkhardt & Busch 2013). In this study, fixed-bed technology is used, with chemical catalysts mainly composed of nickel (Ocampo et al. 2009; Wang et al. 2011). Methanation reaction is exothermic, with a heat production of 0.165 $MJ.mol^{-1}$ of input CO_2 (Hoekman et al. 2010). Produced heat is used to regenerate the solvent in biogas upgrading by amines scrubbing and / or to heat the anaerobic digesters. Required data are presented in Table 5, and are in line with recent work of Reiter and Lindorfer (2015).

Table 5: Main assumptions at the methanation step

Description	Values	Reference
CO_2 from amines scrubbing		
Inputs		
CO_2outAmines	80 $m^3.h^{-1}$	
Electricity consumption	27.2 $MJ.h^{-1}$	Present data
H_2fromElectrolysis	310.2 $m^3.h^{-1}$	Present data
Outputs		
CH_4outMethaAmines	80 $m^3.h^{-1}$ CH_4: 96.56%mol / H_2: 1.69%mol / H_2O: 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
CO_2 from membranes		
Inputs		
CO_2outMb	95.1 $m^3.h^{-1}$	
Electricity consumption	33.3 $MJ.h^{-1}$	Present data
H_2fromElectrolysis	312 $m^3.h^{-1}$	Present data
Outputs		
CH_4outMethaMb	98 $m^3.h^{-1}$ CH4: 96.7%mol / H_2: 1.2%mol / H_2O: 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 $m3.h^{-1}$	
Electricity consumption	106 $MJ.h^{-1}$	Present data
H_2fromElectrolysis	310.2 $m^3.h^{-1}$	Present data
Outputs		
CH_4outMethaBiogas	230.9 m^3h^{-1} CH_4: 97.4%mol / H_2: 1.6%mol / H_2O: 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

2.6 Completion, injection and combustion steps

Before injecting the bioCH_4 in the natural gas network, a completion step is necessary in order to bring the dew point to existing specifications (here 40°C and 5 bar). Electricity consumptions of this operation for the three proposed scenarios are given in Table 6.

Table 6: Main assumptions at the completion step

Description	Values	Reference
CH_4 from CO_2 amines		
Inputs		
CH_4outMethaAmines + CH_4outAmines	80 m^{3}h^{-1} + 156 m^{3}h^{-1}	
Electricity consumption for completion	47.2 MJ.h^{-1}	Present data
Outputs		
CH_4CompletionAmines	232 m^{3}h^{-1} CH_4: 98.54%mol / H_2: 0.58%mol / CO_2: 0.80%mol / H_2O: 0.08%mol T: 40°C / P: 5 bar	Present data
CH_4 from CO_2 membranes		
Inputs		
CH_4outMethaMb + CH_4outMb	98 m^{3}h^{-1} + 135 m^{3}h^{-1}	
Electricity consumption for completion	37.3MJ.h^{-1}	Present data
Outputs		
CH_4CompletionMb	232 m^{3}h^{-1} CH_4: 98.58%mol / H_2: 0.68%mol / CO_2: 0.66%mol / H_2O: 0.08%mol T: 40°C / P: 5 bar	Present data
CH_4 from Biogas		
Inputs		
CH_4outMethaBiogas + CH_4outBiogas	230.9 m^{3}h^{-1} + 4.5 m^{3}h^{-1}	
Electricity consumption for completion	40 MJ.h^{-1}	Present data
Outputs		
CH_4CompletionBiogas	233 m^{3}h^{-1} CH_4: 97.8%mol / H_2: 1.15%mol / CO_2: 0.97%mol / H_2O: 0.08%mol T: 40°C / P: 5 bar	Present data

At last, data for injection and combustion have been directly taken from the Ecoinvent 2.2 database, with biogenic CO_2 emissions instead of fossil CO_2 emissions. .

2.7 Economic data

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

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

network allows an internal rate of return (IRR) equal to 10% on 15 years. Given the feed-in tariffs of bioCH4 - fixed by decree in France in 2014 (http://www.legifrance.gouv.fr/affichTexte.do?cidTexte=JORFTEXT000024833895) and taking into account the costs of the final steps - biogas upgrading and injection – this leads to a biogas production cost equal to 0.36 €.m-3.

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)

3 Results

3.1 Economic results

Continuous operation

Annualized production costs of can be seen on Figure 2, for an electricity price of 30 €.MWh^{-1} and with an IRR equal to 10% on 15 years. There is an important difference of total annualized production costs with and without methanation: approximatively 1650 k€.$year^{-1}$ for upgrading with PtG, and 940 k€.$year^{-1}$ for standard upgrading. The largest contribution to the production costs is the biogas production, (around 80% for upgrading technologies and 45% for PtG scenarios). This cost is the same in the five scenarios assessed (757 k€.$year^{-1}$), but as total production costs are much higher in the methanation scenarios, the contribution of biogas is lower. The higher costs of the methanation scenarios are mainly due to the electrolysis step. It represents more than 35% of the production costs, with a large share due to electricity consumption (70% of the electrolysis costs). At last for PtG production with CH_4 and CO_2 separation, upgrading and methanation costs are quite similar.

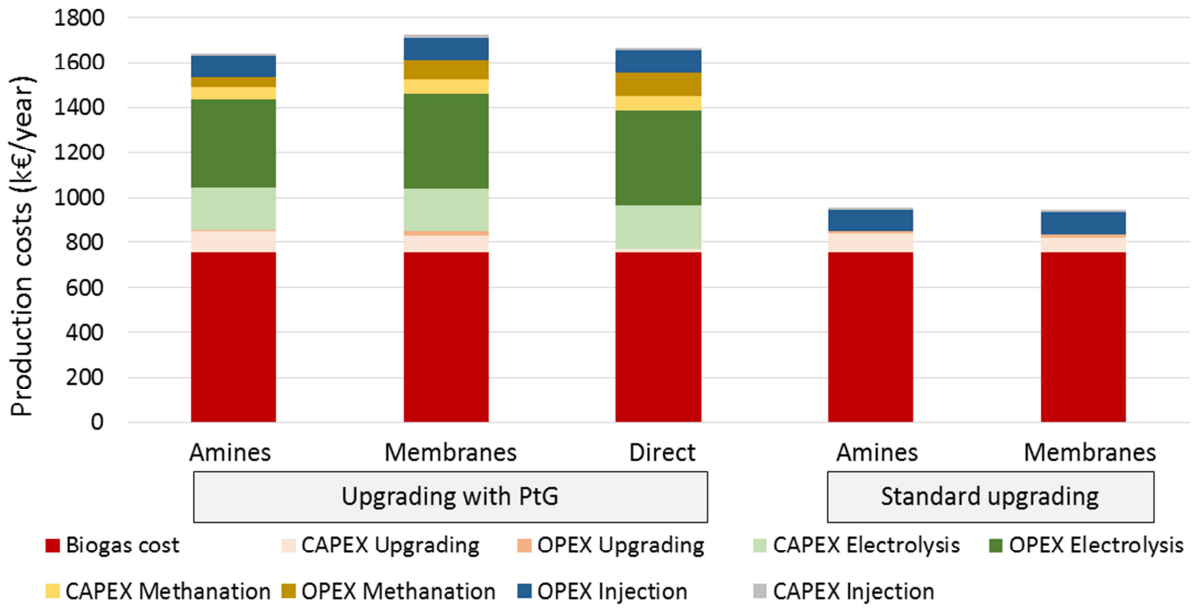

Figure 2: Contribution analysis of the annualized production costs

The contribution analysis of the production costs for 1 kWh of bioCH_4 from PtG or upgrading are shown on Figure 3, for the same electricity price and IRR rate than the Figure 2. It appears that production costs of the different options are quite the same: they range from a minimum of 0.096 €.kWh^{-1} for PtG from biogas with direct methanation and with membrane separation, and a maximum of 0.104 €.kWh^{-1} for biogas upgrading with membrane separation. In fact, the higher production cost of the PtG technologies are counterbalanced by a higher bioCH_4 injection in the gas network (41 m^3.h^{-1} on average for standard upgrading versus 138 m^3.h^{-1} on average for methanation scenarios). The respective contributions of the different steps are the same than for the annualized production costs.

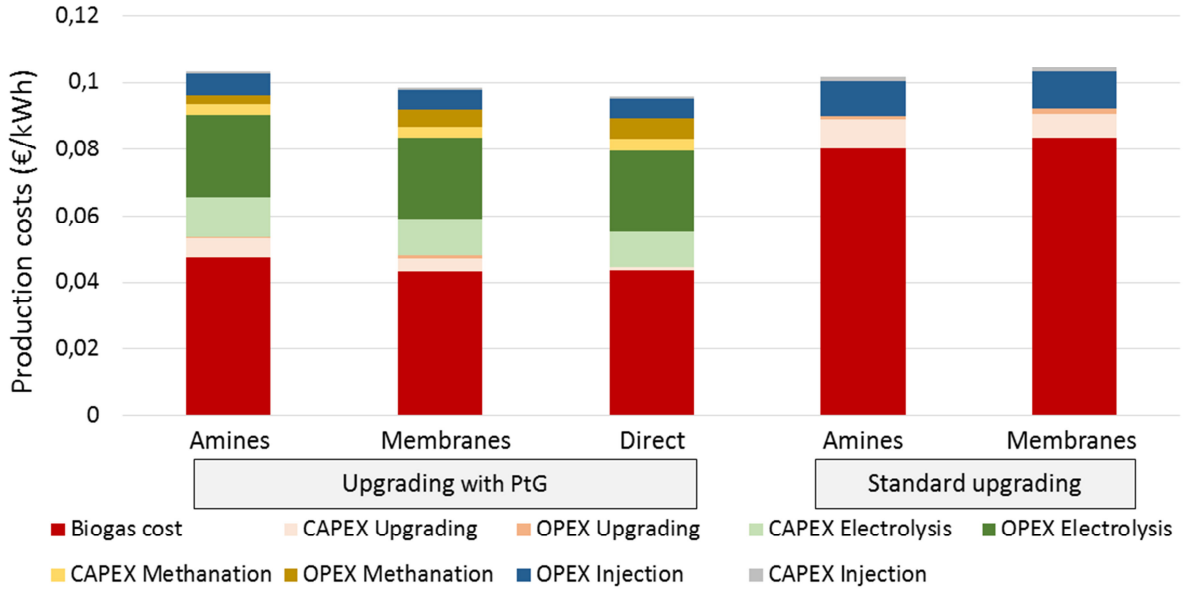

Figure 3: Contribution analysis of the production costs for 1 kWh injected in the gas network

In Figure 4, overall production costs of the different scenarios are presented for electricity prices ranging from 0 to 45 €.MWh^{-1}. Costs of CH_4 from upgrading technologies are almost insensitive to electricity price, as they are mainly driven by the fixed biogas cost, and because the only other relevant OPEX corresponds to the injection step which is not correlated with the electricity price (see Table 7). On the contrary, PtG scenarios are very sensitive to the electricity price, with an increase of approximatively 35% of the production costs, mainly because of the electricity consumption at the electrolysis step. Compared to production of CH_4 with the cheaper upgrading technology (amines scrubbing), CH_4 from PtG is competitive for electricity price below 27 €.MWh^{-1} for separation with amines scrubbing, below 34 €.MWh^{-1} with separation by membranes and below 38 €.MWh^{-1} for direct methanation.

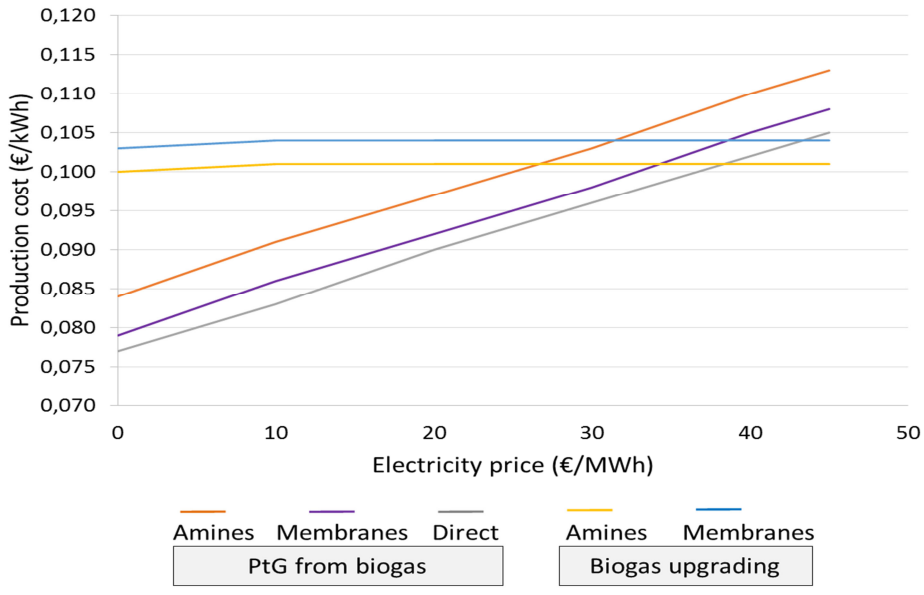

Figure 4: Productions costs for 1 kWh injected in the gas network depending on the price of electricity

Intermittent operation

As previously pointed out, PtG can be seen as a relevant way to store overproduced electricity, and then intermittent operation should also be analyzed. According to Gahleitner (2013) the use of such fluctuating power sources is satisfying, and is currently in use in a lot of PtG pilot plants. Economic results for operation time varying between 2000 and 8760 hours (continuous operation) have been assessed for PtG scenarios and CH_4 from amines scrubbing (Figure 5), with an electricity price equal to 20 €.MWh^{-1}. Biogas upgrading is insensitive to the operation time. This can be explained by the important contribution of the biogas production, and because others OPEX are almost exclusively driven by the fixed cost of the rental of the injection station. On the opposite, costs of direct methanation are very strongly connected with the operation time, with costs between 0.330 €.kWh^{-1} for 2000 hours of operating time, and 0.090€.kWh^{-1} for continuous operation. In fact in this configuration CH_4 contained in the biogas is not recovered and injected in the gas grid when methanation is not in operation. Consequently, the volume of energy produced is drastically reduced with the increase of the intermittency. On the other hand, fixed costs (biogas production, all the CAPEX and rental of the injection unit) are unchanged. Therefore the direct methanation leads to a dramatic increase of the production costs with an intermittent functioning, and so this technology is not suitable for non-continuous operation mode. Finally, like in the direct methanation, production costs decrease for membranes and amines technologies of PtG with the increase of the operation, but to a much lesser degree. Depending of the duration of the operation time, and for a given price of electricity, production costs can be higher or lower than CH_4 from biogas upgrading. For instance, for a electricity at 20 €.MWh^{-1}, operation time should be at least equal to 6160 hours for membranes separation and to 7240 hours for amines separation to be competitive with biogas upgrading.

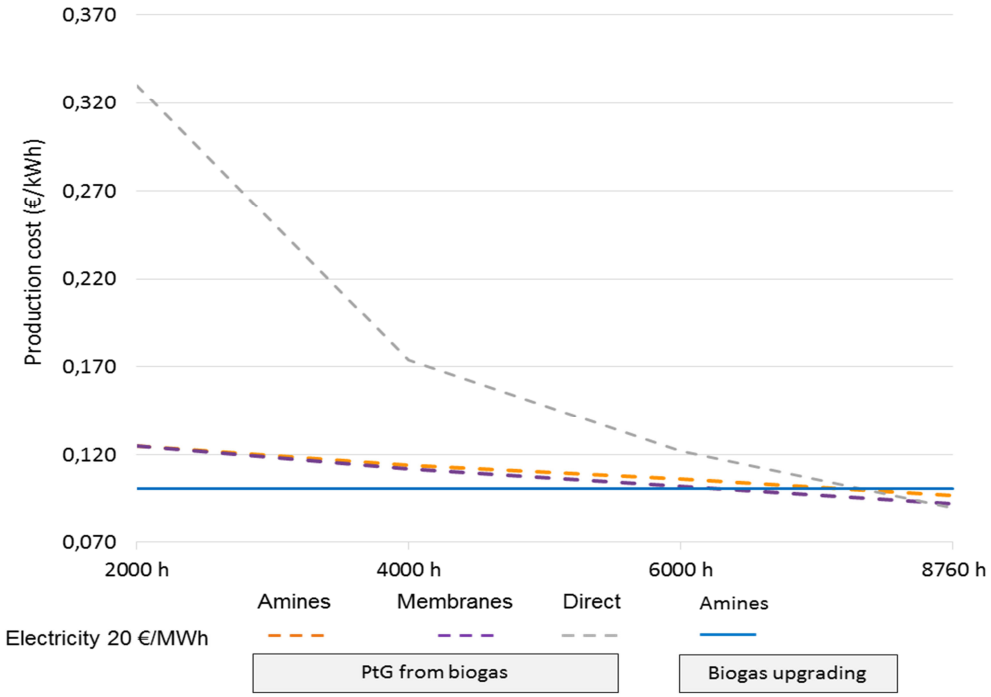

Figure 5: Productions costs for 1 kWh injected in the gas network depending on the technology and the operating time

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

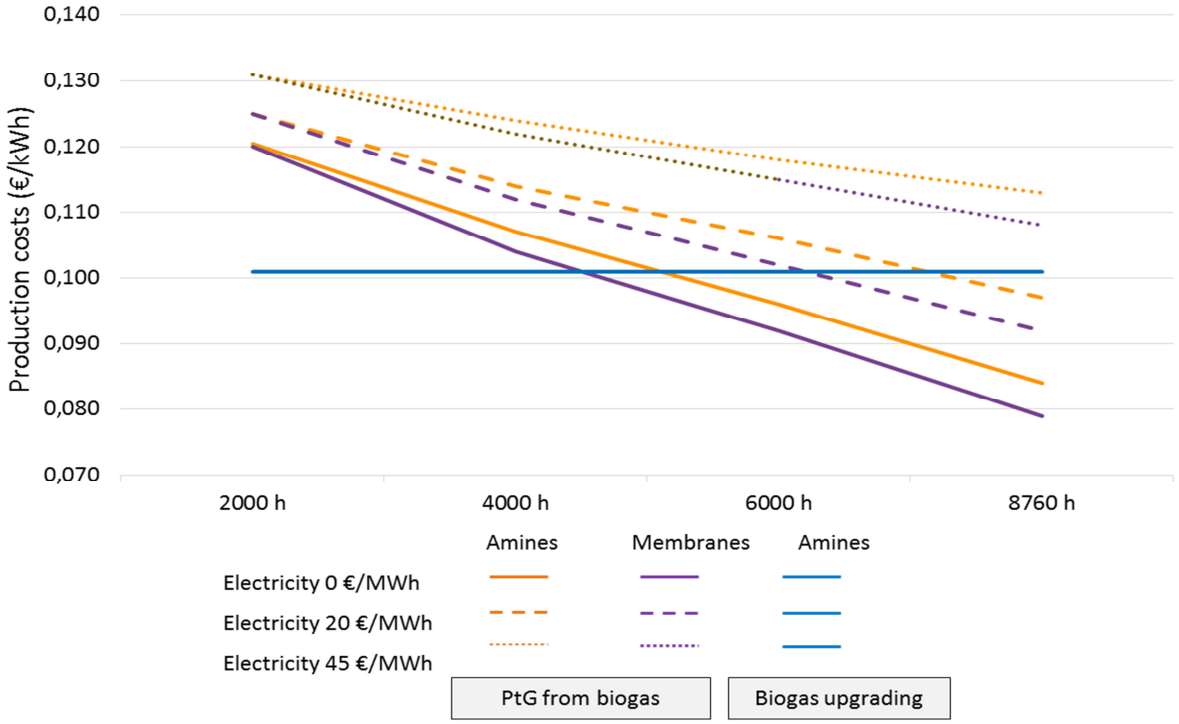

Figure 6: Productions costs for 1 kWh injected in the gas network depending on the price of electricity and the operating time

Sensitivity analysis

As it has been underlined in the contribution analysis, electrolysis and more precisely the electricity consumption of this step, is one of the main contributor to the production costs of CH_4 from PtG. Thus, we conduct a sensitivity analysis on the electricity consumption for H_2 production. We discuss a favorable scenario, with a reduction of 25% of the electricity consumption (*i.e* 3.75 $kWh.m^{-3}$ H_2). This value is in line with predicted values of future electricity consumptions around 3 $kWh.m^{-3}$ H_2 (Udagawa et al. 2007; Zeng & Zhang 2010). The evolution of production costs with low electricity consumption scenarios and for different electricity prices is shown on Figure 7. As expected, the more electricity is expensive, the more electricity consumption reduction has an important effect on the diminution of the production costs. For a price of electricity of 10 $€.MWh^{-1}$, the decrease of the production cost is only equal to 0.001 $€.KWh^{-1}$, but with an electricity at 45 $€.MWh^{-1}$, the reduction of the production cost is equal to 0.007 $€.kWh^{-1}$. Consequently, the minimum price of electricity so that CH_4 from PtG can be competitive with CH_4 from upgrading increases: it reaches 35 $€.MWh^{-1}$ for amines scrubbing and is around 45 $€.MWh^{-1}$ for membranes separation.

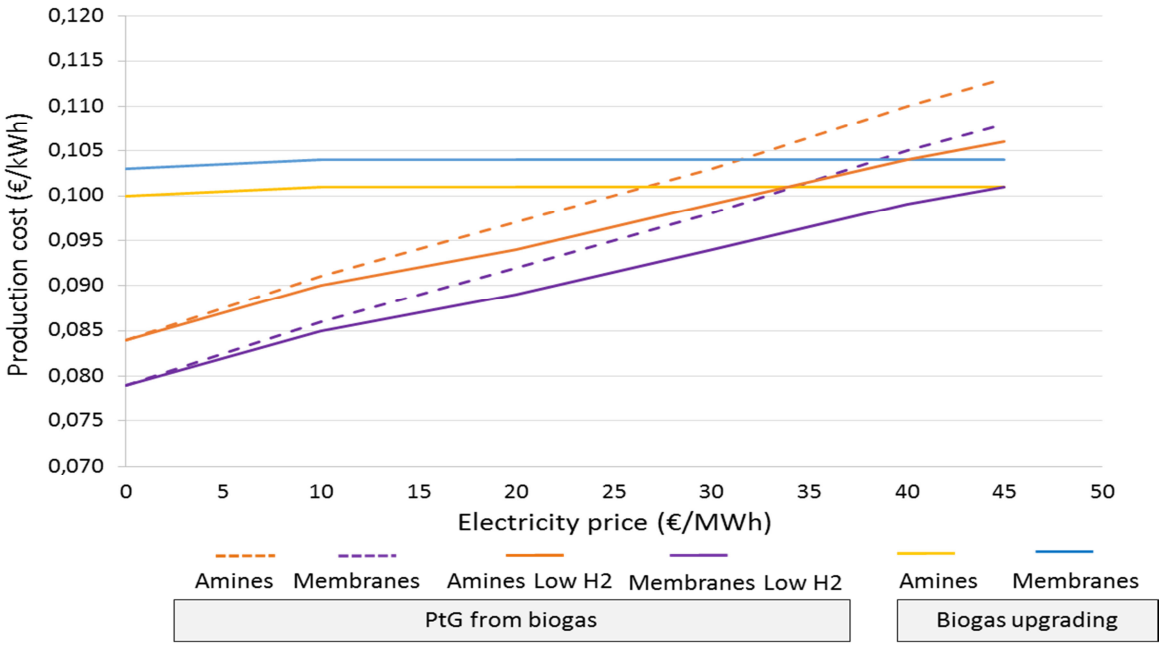

Figure 7: Productions costs for 1 kWh injected in the gas network depending on the price of electricity and the electricity consumption for H_2 production

3.2 Environmental impacts

Environmental impacts of the study have been assessed within the methodological framework of the LCA based on a ‘‘cradle to grave” inventory of emissions and resources consumption. The considered functional unit in the study is one MJ produced by combustion of CH_4 in a boiler, based on the lower heating value of CH_4 (50.0 $MJ.kg^{-1}$). As one of the main objective is to reduce GHG emissions in the atmosphere with respect to fossil fuels, we therefore consider the climate change impact. It has been assessed with the characterization factors provided by the fourth IPCC report for a temporal horizon of 100 years (IPCC 2007) . It should be underlined that the characterization factor of CH_4 in case of methane loss has to be adapted. Since the carbon of the CH_4 comes from the sewage sludge (and previously from the atmosphere), the characterization factor must be the one of biogenic CH_4, which is slightly below the one of the fossil CH_4 (22.3 kg CO_2eq.kg^{-1} vs. 25 kg CO_2eq.kg^{-1}).

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

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

Continuous operation

Principal contributors to GHG emissions for the different assessed technologies are shown on Figure 8. From a global perspective, GHG emissions from PtG are almost equivalent for the three technologies, and higher than biogas upgrading. Nevertheless, they are still lower than GHG emissions from natural gas combustion. For the fossil scenario (natural gas burned in a boiler), most of the emissions correspond to fossil CO_2 from gas combustion. The remaining emissions are linked to resource extraction and transportation of the gas in the pipelines. In the scenarios with production of energy from sewage sludge, main GHG emissions come from the management of the digestates. Environmental burdens are associated with loss of CH_4 (40%) and emissions of N_2O (60%). On the other hand, negative impacts are due to the substitution of the digestates to the production and the use of mineral fertilizers. For PtG technologies, electricity consumption is also an important contributor to climate change. As the carbon content of the French electricity is particularly low due to the large part of the nuclear in the mix composition, a sensitivity analysis on the source of electricity (and as a consequence on its carbon content) is done in a following paragraph.

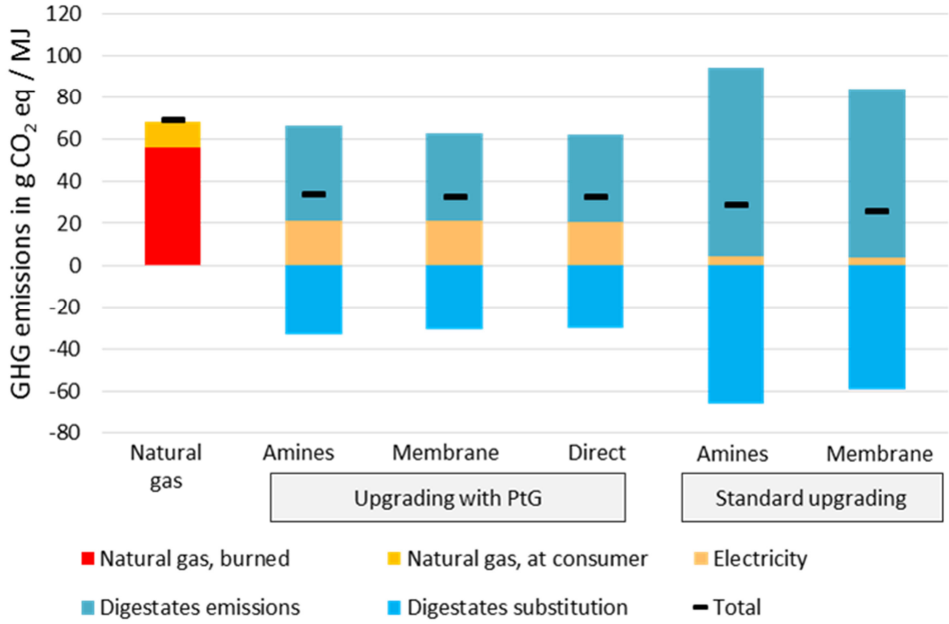

Figure 8: Contribution analysis of GHG emissions for 1 MJ injected in the gas network

Intermittent operation

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

electricity surplus is 80% renewable and 20% nuclear by 2050. Figure 9 presents results for intermittent methanation during overproduction period. It is supposed that when methanation is not functioning, the non-injected gas (CO_2 with 18% of CH_4 in membrane separation and biogas with 65% of CH_4 in direct methanation) is flared, in order to reduce direct CH_4 emissions to the atmosphere. In this scenario, the results for membrane and amines separation are almost the same, so only amine technology will be represented for the next environmental impact assessment results.

In direct technology configuration, biogas is not recovered and injected in the gas grid when methanation is not in operation, like in the economic assessment. This leads to GHG emissions which are much higher than continuous operation (and even higher than natural gas combustion) for the three time horizons analyzed. On the contrary, intermittent operation with electricity from renewable resources and amines separation leads to an important decrease compared to continuous operation with French average electricity mix (-37% in 2020 and -40% in 2050). For the three scenarios assessed, climate change impact of PtG is even lower than biogas upgrading.

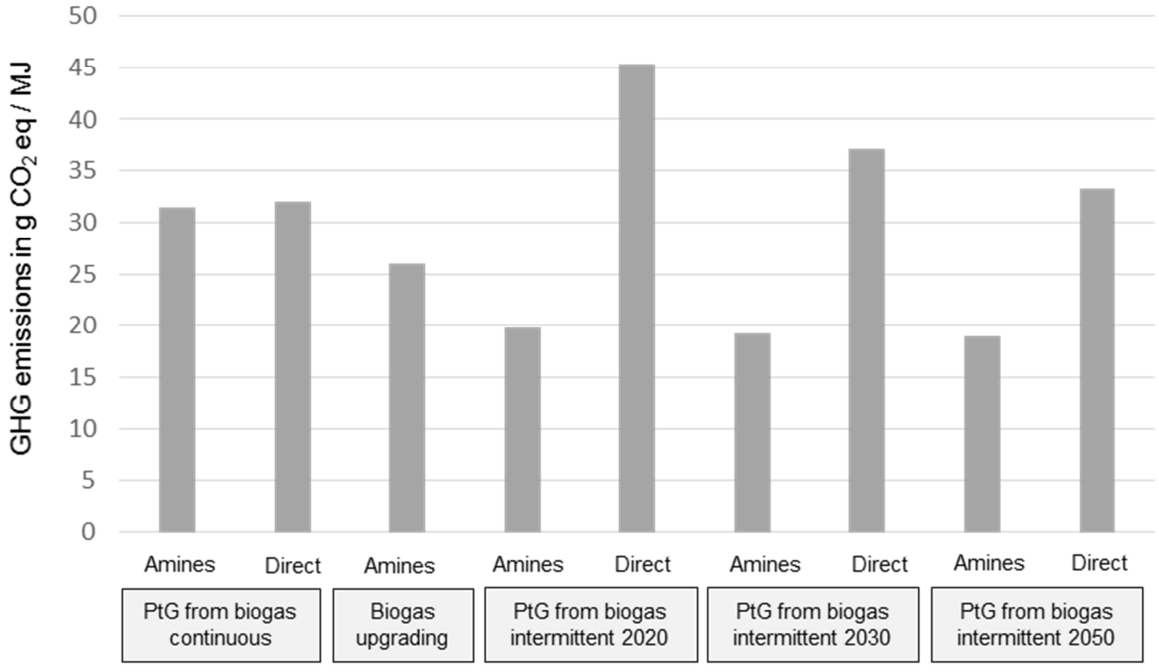

Figure 9: GHG emissions for 1 MJ injected in the gas network depending on the price of electricity and the operating time

Sensitivity analysis

Change in the electricity mix composition

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

only 70% of the emissions from biogas upgrading. This is due to the fact that PtG technologies consume much more electricity than upgrading technologies, combined with the different carbon contents of the electricity mix. In fact GHG emissions of 1 MJ of European electricity is 0.156 CO_2eq, and only 0.029 CO_2eq for French electricity, because French mix is mainly composed of nuclear electricity (78% according to the Ecoinvent database), with very low GHG emissions (0.00172 CO_2eq.MJ^{-1}). Consequently, with the use of a European electricity mix (or with another average electricity mix with high GHG emissions, like China or United Kingdom), climate change impact of PtG technology becomes higher than natural gas, with the highest contribution coming from the electricity consumption. Thus electricity with low carbon content is a mandatory prerequisite to achieve a sustainable production of CH_4 from CO_2.

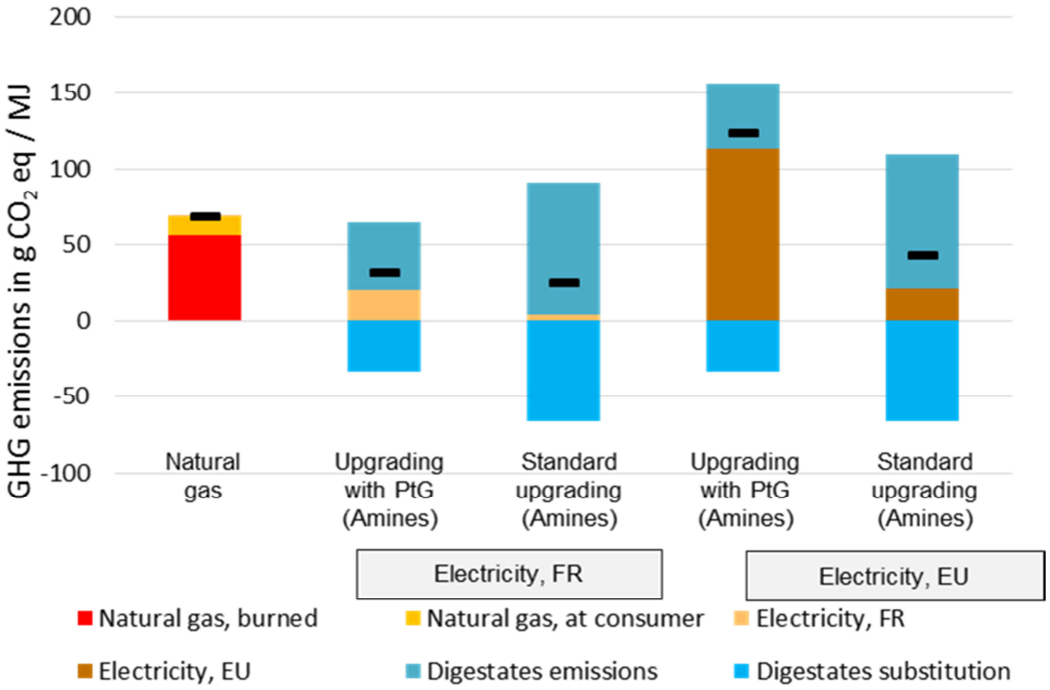

Figure 10: Contribution analysis of GHG emissions for 1 MJ injected in the gas network depending on the electricity mix

Endpoints impacts

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

- Human health, defined as the sum of years of life lost and years of life disabled, and 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.

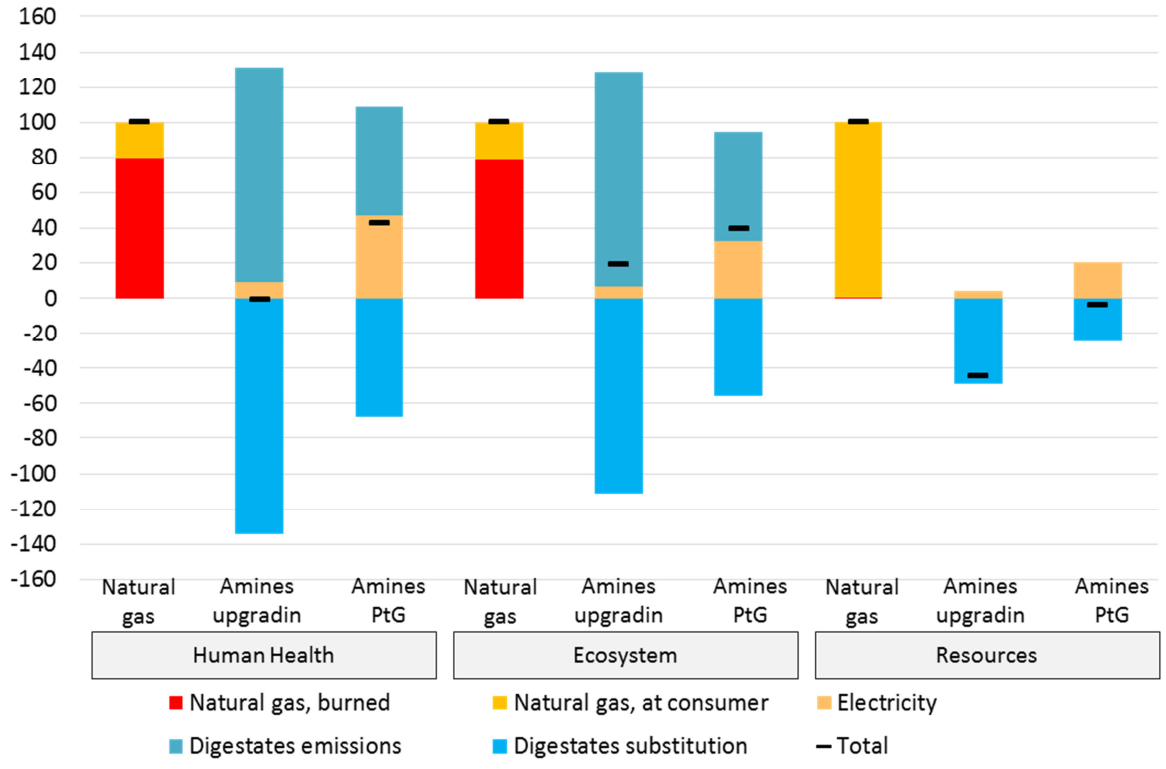

Figure 11: Contribution analysis of endpoints impacts (Human Health, Ecosystem and Resources) for 1 MJ injected in the gas network

4 Discussion

It must be underlined that this is a prospective LCA of a process that does not yet exist. Consequently the systems described in this study can be subjected to important modifications. From a technical point of view, some changes could be proposed to improve the economic and environmental balance of this process. As biogas is used to heat the anaerobic digesters, the CO_2 resulting from the combustion of this biogas can also be converted into bioCH_4 by methanation. The auto-consumption of the biogas ranges from 21% (membrane upgrading) to 28% (amines upgrading), so the added amount of bioCH_4 that could be injected in the gas network is far from being marginal, and could potentially improve the overall performances of the process. About economic assumptions, biogas production costs are based in this paper on French feed-in tariffs and an expected IRR. These values may seem conservative, compared to data from literature (between 0.24 and 0.30 €.m^{-3} of biogas in Patterson et al. (2011)). In the future, a more detailed approach could be investigated to consolidate biogas production costs, for instance by including the biogas production process in the economic assessment. However, as the same biogas production costs have been used for upgrading and PtG technologies, it should be underlined that this inclusion would not change the general conclusion of the economic study. It also has to be noticed that electricity price is strongly correlated with the annual operation hour: average electricity price increases with the load factor (ENEA, 2016). In our study, different electricity prices are proposed for different operation time. This has been done to assess how the analyzed system responds to these two key parameters independently. Further work could therefore integrate price duration curves for different locations and different time horizons in order to map out more clearly potential prospective scenarios. To go deeper into details about the economic feasibility of the combination of anaerobic digestion and PtG technology, the effect of the introduction of an incentive for "green gas" production could be assessed. As underlined by Guandalini et al. (2015), there is a need of specific incentive for PtG technologies in order to push the competitiveness of this energy production system. They propose a "green gas" incentive between 0 and 20 €.MWh^{-1}. As this incentive is only related to the CH_4 from CO_2 conversion by methanation, its amount is function of the proportion of bioCH_4 from methanation. In our system, CO_2 conversion into bioCH_4 represent approximatively 50% of the total injected bioCH_4 for the amines scrubbing and the membrane upgrading technologies. Applied to the case study described in section 3.1 (continuous operation and an electricity price of 30 €.MWh^{-1}), an incentive of 20 €.MWh^{-1} leads to a reduction of the production costs of almost 10%. At last learning curves (or experience curves) could be used to quantify the future reduction of investment costs for the different assessed technologies. Progress ratios (PR, defined as the rates of unit cost decline with each doubling of cumulative production) are expected to be around 0.88 (Junginger et al., 2006) for biogas plant and 0.82 for H_2 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 bioCH_4, using methanation to valorize
463 the CO_2 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 bioCH_4 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 bioCH_4. 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 €.MWh^{-1}
476 for direct methanation and separation by membranes. The lower competitiveness of amines
477 scrubbing is due to a lower volume of bioCH_4 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 CO_2 from biogas was converted into BioCH_4, but the optimum

491 from both environmental and economic points of view may consist in a more complex process where
492 CO_2 from internal biogas combustion should also be turned into bioCH_4.
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