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Sustainable Production and Consumption 49 (2024) 625–640

Contents lists available at ScienceDirect

Sustainable Production and Consumption

journal homepage: www.elsevier.com/locate/spc

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ARTICLE INFO

Editor: Dr. Carlos Pozo

Keywords:
Chemical absorption
Membrane separation
Cryogenic separation
Water scrubbing
Life cycle assessment

ABSTRACT

The conversion of biogas into biomethane and CO_2 offers a potential solution for climate mitigation, avoiding the use of fossil carbon. While previous studies compared various carbon capture (CC) technologies for biogas upgrading, this research specifically addresses the co-production of biomethane and CO_2 as high purity co-products. This work simulates, validates with literature and onsite data, and compares four CC technologies (Chemical Absorption (CA), Membrane Separation (MS), Cryogenic Separation (CS), and High-Pressure Water Scrubbing (HPWS)) at different scales, considering both technological and environmental impacts evaluated through the Life Cycle Assessment (LCA) method. ProSimPlus® software was employed to conduct process simulations, having as target biomethane for grid injection (> 96 % mol) and liquid CO_2 (purity as high as possible). The analysis used data from a medium-scale biogas site (365.24 Nm^3/h). The considered criteria were biomethane and CO_2 purity and recovery rates, energy consumption, and environmental impacts. The comparison showed that all studied technologies met these targets, with CA technology demonstrating the highest purity and recovery rate (higher than 98 %), but with the highest energy consumption (electricity 0.8 kWh/Nm^3 biogas and heat 0.5 kWh/Nm^3 biogas). MS technology exhibited similar purity and recovery rates with significantly lower energy consumption (electricity about 0.2 kWh/Nm^3 biogas) than the other alternatives. Moreover, MS technology demonstrated the best environmental performance, with a global warming impact (GWP100) of 1.09 kg CO_2-eq, outperforming CA, CS, and HPWS, which recorded values of 1.42 kg CO_2-eq, 1.13 kg CO_2-eq, and 1.12 kg CO_2-eq respectively, impacts expressed per 1 kg liquid CO_2. Furthermore, MS outperformed the other technologies for all 18 impact categories calculated with ReCiPe midpoint method. For example, MS demonstrated the lowest water consumption potential, freshwater ecotoxicity potential or photochemical oxidant formation potential-humans (1.94 10^{-3} m^3, 3.15 10^{-2} kg 1,4-DCB-eq, 6.64 10^{-5} kg NO_x-eq respectively) while CA the highest values for the same impact categories (6.27 10^{-3} m^3, 7.62 10^{-2} kg 1,4-DCB-eq, 5.4 10^{-4} kg NO_x-eq, respectively). The difference between the impact results of the four technologies is highly dependent on the used electricity mix: more carbonated is the electricity mix, more the difference between the four technologies is enhanced.

1. Introduction

Over the past century and up to now, the emissions of greenhouse gases, including carbon dioxide, nitrous oxide, and methane have increased significantly (Abu-Zahra et al., 2007). Different emission sources such as energy production, cement production or chemical plants are responsible for this phenomenon. In order to mitigate their impact on the environment, governmental policies have been put in place. For instance, several European directives such as (EU) 2018/2001 "RED II" (Renewable Energy Directive) (Ardolino et al., 2021) has been issued to reduce fossil fuel consumption and greenhouse gas (GHG) emissions. Additionally, the Intergovernmental Panel on Climate Changes (IPCC) has assessed that about 50 % of the increase in greenhouse gas emissions is due to carbon dioxide emissions, which poses a significant challenge in either avoiding CO_2 emissions or capturing CO_2 from the atmosphere (Abu-Zahra et al., 2007).

A reduction in greenhouse gas (GHG) emissions by over 80 % compared to emissions resulting from the use of traditional fossil fuels can be obtained by the production and utilization of biomethane that is generated through the biogas upgrading process. The implementation of anaerobic digestion sites using waste (e.g. manure, biowaste, agriculture residues, etc.) to produce biogas (about 60 % methane and 40 % CO_2)

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https://doi.org/10.1016/j.spc.2024.07.010
Received 14 March 2024; Received in revised form 5 July 2024; Accepted 11 July 2024
Available online 18 July 2024

has increased in the last years (Holm-Nielsen et al., 2009). Biogas could be used as a renewable energy source to produce heat and electricity or be upgraded into biomethane and be used as a replacement of natural gas. Biomethane can be injected directly into the natural gas grid by respecting the natural gas standard conditions. In 2017, Europe witnessed the generation of approximately 20,000 GWh of biomethane from >500 biogas plants (EBA_Statistical-Report-2018, 2024). Furthermore, 15 billion cubic meters of biogas, which is equivalent to 9 billion cubic meters of biomethane, were produced in 2018 by 100,000 modern biogas plants situated in China (Scarlat et al., 2018).

Besides biomethane, the captured biogenic CO_2, which is normally released to the atmosphere, can be used for different industrial applications in replacement of fossil carbon sources. This carbon capture and utilization (CCU) approach could represent a mitigation potential for global warming and also a revenue source for the biogas plants. However, in order to ensure the expected mitigation results, the environmental performances must be evaluated before the technologies are implemented at real scale.

The most widely utilized and studied carbon capture (CC) technologies are chemical absorption (CA), membrane separation (MS), water scrubbing (WS), and cryogenic separation (CS) (Bauer et al., 2013). However, comparisons of these technologies in current literature are neither fair nor equitable, as they often focus on biomethane production rather than on CO_2 capture. Additionally, these technologies are implemented at various scales under different inlet conditions, or are still in the pilot stage, making thorough analysis difficult. Another challenge in achieving a fair comparison is that these technologies are sometimes evaluated at different scales, which is not ideal for a good comparison. This work presents a fair comparison of these four CC technologies for the co-production of biomethane and liquefied CO_2 considering their technical and environmental performances. This comparison was conducted via simulations under identical inlet conditions and at the same scale.

2. Literature review

The most mature CC technology is chemical absorption (Kapoor et al., 2019). This method has been in use for a long time for removing CO_2 at industrial scale. For instance, this technology has been used in the Econoamine FG Plus™ process (a technology used for CO_2 capture from post combustion sources using amines as solvent), licensed by Fluor Co (Carbon Capture – Fluor's Patented Econamine Technology, 2024). A study conducted by Naeiji et al. (Naeiji et al., 2022), compared the simulation results for biogas upgrading and hydrogen production from syngas (from methane steam reforming) with two technologies, i.e. CA and CS. High biomethane purity was obtained with both processes (CS 98.74 % mol; CA 98.73 % mol) from a biogas containing 62 % mol of methane. An economic analysis indicated that the operational cost including the annual and investment cost, for CS were higher than CA by 2.2 and 2.3 times, respectively. On the other hand, CA was the most effective process to remove CO_2 from syngas. The quite novel CS process was proven to be a competitive technology compared to CA because of the lower energy consumption. However, it currently fails in terms of cost and overall performance in case of some applications. CS processes exhibit clear superiority in achieving high methane purity with minimal methane loss ($\leq$1 %) when compared to conventional methods such as HPWS or PSA (Song et al., 2019). This high purity methane stream could be directly injected into the gas grid or used as a vehicle fuel, as noted by Savickis et al. (Savickis et al., 2020). Additionally, CS offers the unique advantage of producing high-purity liquid CO_2 without additional liquefaction operation, often reaching purity levels of up to 98 % (Yousef et al., 2018).

Regarding water scrubbing technology, the study of T. Gantina et al. (Gantina et al., 2020) suggested that it involves a high electricity cost, which depended on the operating pressure: 0.24 kWh/m^3 of raw biogas were needed at atmospheric pressure versus 0.34 kWh/m^3 raw biogas at 10 bar.

Physical absorption (PA) is an upgrading technology for removing the undesired component of the gas stream by a non-reactive fluid such as Genosorb 1753. Patterson et al. (Patterson et al., 2011a) conducted a literature-based comparison of six carbon capture technologies: PSA, WS, PA, CA, MS, and CS focusing on energy requirements. The findings indicated that upgrading biogas through PSA demands less energy compared to other technologies. More specifically, the study showed that PSA technology demands 128 %, 190 %, 145 %, 230 %, and 125 % more energy than the upgrading of an equivalent quantity of biogas using WS, CA, PA, MS, and CS, respectively. In a different study (Barbera et al., 2019), the performances of HPWS, hot potassium carbonate, and PSA were compared in terms of products purities, recovery rate, and energy requirement, where the results demonstrated that PSA technology provided the highest carbon dioxide purity.

Another study (Khan et al., 2021) compared biogas upgrading technologies for biomethane production, including MS, PSA, WS, CA, and organic solvent scrubbing based on the energy requirement per cubic meter of biomethane produced. This study reported that MS needed the lowest energy demand. Additionally, MS had a small biomethane recovery difference (2.5 %) compared to the most performant technology CA in terms of purities and recovery rates. Notably, MS did not require water or chemicals, making it a recommended choice in the biogas upgrading field.

Concerning the environmental impacts, some of these technologies have been compared via the Life Cycle Assessment (LCA) method. The studies on LCA of carbon capture technologies for biogas upgrading have taken an attributional LCA approach using ReCiPe method (Florio et al., 2019), CML (Lombardi and Francini, 2020), and IMPACT 2002 (Ardolino et al., 2021) as impact assessment methods. Those studies have focused on midpoint impacts such as, global warming potential, acidification, eutrophication, and ozone depletion. For instance, a comparison was performed (Zhang et al., 2020) between MS, PSA and CA, and the conventional natural gas production. Biomethane produced by MS and CA had the lowest greenhouse gas (GHG) emissions.

Another LCA of five upgrading technologies (HPWS, CA, potassium carbonate scrubbing, PSA and membrane permeation) was performed by Lombardi et al. (Lombardi and Francini, 2020). The material/energy inventories for all processes were taken from different literature sources. CA technology showed the best environmental performances in terms of global warming due to its minimal CH_4 losses and the low electricity consumption, while PSA technology had the lowest human toxicity indicator.

Most of these studies ((Song et al., 2019; Savickis et al., 2020), (Yousef et al., 2018)) investigated the CO_2 removal from biogas for biomethane production based on different process configurations and operation conditions. The CO_2 was not treated as a product, but as a waste gas. Consequently, the reported energy efficiencies must be understood in relation to their specific context (input gas, purity of biomethane, and no CO_2 production). While (Khan et al., 2021) focused on CO_2 production by MS, CA and TSA, this work was applied to coal power plant flue-gas and not to biogas. Only one study (Patterson et al., 2011a) investigated the joint production of CH_4 and CO_2 but only by CS process. Another aspect to be mentioned is that all the studies cited above are based on simulations only, without any experimental validation.

Concerning the environmental performances, LCA was applied to biomethane upgrading without CO_2 production in several studies ((Lombardi and Francini, 2020; Zhang et al., 2020), (Pavičić et al., 2022)). Moreover, only (Pavičić et al., 2022) used simulations on a common basis to compare MS, WS and CA, but not for biomethane/CO_2 co-production, while (Lombardi and Francini, 2020; Zhang et al., 2020) used inventory data from heterogenous sources, and consequently the LCA results are hardly comparable.

An analysis and comparison of the technical, energy and environmental performances of technologies for the joint production of biomethane and CO_2 should be conducted under the same conditions, i.e. inlet gas mixture and output products in each scenario to ensure an

equitable evaluation of these technologies. At our best knowledge, such analysis is missing in the literature and is what we propose to investigate in the following sections.

This work presents a simulation-based analysis with the aim to propose a fair comparison of four CC technologies for the co-production of biomethane and liquefied CO_2, i.e. MS, CA, CS and HPWS, considering their technical and environmental performances. Our study will primarily focus on comparing these technologies at different scales based on common criteria such as energy consumption, CO_2 capture recovery, CO_2 purity, CH_4 capture recovery and CH_4 purity. A comparison with available data in the literature and from biogas production sites is also discussed. Finally, the LCA method is applied to these processes in order to uncover their environmental performance and ranking.

3. Methods

This work was articulated in three parts: 1) process model implementation in ProSimPlus® (process simulation software) and validation based on literature data; 2) simulation of the four CC technologies based on real biogas production sites, sensitivity analysis and technical performance; 3) Life cycle assessment.

Biogas contains between 40 % to 75 % of CH_4, 60 % to 25 % of CO_2, and impurities such as H_2S, N_2, H_2O (vapor) and VOCs (volatile organic compounds) (Pavičić et al., 2022). In this study, we assumed that biogas was dried in a condenser and then passed through a desulfurization unit to remove H_2S which can be harmful for the CC technologies. These well-known pretreatments were common to the four CC technologies. Therefore, biogas containing only CO_2 and CH_4 was used as input of the CC units. The purified biomethane stream (96 % mol or higher) was compressed to 16 bar, which is required pressure for injection into the natural gas network (Scholz et al., 2015). The purified CO_2 stream was compressed to 69 bar and stored in liquid phase with the highest achievable purity destined to further applications.

The models for the CC units were built upon literature data (section 3.1), where these data were used solely to design the simulation models and validate their accuracy in representing the carbon capture processes. The details about the biogas characteristics for each simulation are reported in supplementary information (Table S5).

Once validated, these models were used under identical inlet conditions to compare their performances fairly. The data used for this fair comparison come from three existing biogas sites at different scales: small (62.5–125 Nm3/h), medium (125–625 Nm3/h) and large (> 625 Nm3/h) (Mühl and de Oliveira, 2022) (section 3.2). These similar data sets ensure an equitable comparison across different scales.

3.1. Modeling of the carbon capture technologies

3.1.1. Chemical absorption technology

CA is based on a countercurrent flow configuration using a chemical solvent where high gas product purity and recovery (99.5 mol% - 99.9 mol%) can be achieved. The process implies several chemical reactions between CO_2 in the gas phase and an amine solution This process consists in an absorption column, a heat exchanger and a stripper column with condenser and reboiler.

Gamba et al. (Gamba and Pellegrini, 2014) compared the energy consumption of WS and CA used with methanol-amine (MEA) and methyl-diethanol-amine (MDEA), applied to the biogas from a wastewater treatment plant producing 51.9 kmol/h of raw biogas. They have explained that low percentages of amine result in low purity of biomethane. For instance, they showed that a 15 % MEA solution led to a purity of methane of 91.73 mol%, lower than those obtained with MDEA which achieved 96.32 mol% methane. This is in line with the results of Kapoor et al. (Kapoor et al., 2019), which suggest the use of a 30 % amine solution as a compromise between efficiency and avoidance of corrosion. In terms of heat duty, CA using MEA as solvent consumes more than CA with MDEA. Another comparative study between different chemical solvents was done by Sepúlveda et al. (Sepúlveda et al., 2018) who investigated diethanol-amine (DEA) besides MEA and MDEA, to increase the methane concentration from 57.3 % to 90 %. Results indicated that MEA had the highest performance, achieving a CH_4 concentration of 90.37 mol%.

By using this technology with a MEA blend as solvent, 30 % of the energy consumption was reduced comparing to the conventional MEA. This mature technology has been applied by Mitsubishi Heavy Industries licensed KM-CDR™ process as it is a very good option to decrease the CO_2 emissions in coal-fired plants (Stéphenne, 2014).

The process model was implemented in ProSimPlus® (Fig. 1) based on the work of Gamba et al. (Gamba and Pellegrini, 2014). The model calibration was performed under the same conditions as in (Gamba and Pellegrini, 2014). The configuration and the results were cross-validated with the works of Sepúlveda et al. (Sepúlveda et al., 2018) and Hassan (Hassan, n.d.). The thermodynamic model used in the simulation was the "Amines and acid gases" model in ProSimPlus® which uses the Peng-Robinson equation of state. The design parameters are reported in Table S1.

Process layout for CA. Water in biogas is removed through a condenser (C-101) that operates at 30 °C. The dried biogas is compressed (B-101) to 1.2 bar and cooled down to 25 °C (C-102) before entering the absorber in counter-current with the solution of MEA, at 25 °C and 1 bar. In this work, the used percentage of MEA in water was 30% *w*/w, and for the rich loading stream leaving at the bottom of the absorption column (Absorber), the MEA content was limited to 0.35. The lean loading, i.e. the loading of the amine fed to the absorption column, was set to 0.23 in order to minimize the heat duty required by the regeneration (Gamba and Pellegrini, 2014). The absorber operates at a pressure (P) and temperature (T) of 1.2 bar and 25 °C. Biomethane exits the column at the top with a mole fraction >0.96 mol%.

A solution of amine rich in CO_2 (rich-amine) exits the column from the bottom and is sent to a heat exchanger (HE-101) to be heated up to 110–120 °C before entering the stripper column where CO_2 will be desorbed. It is important to note that the regeneration of MEA must be performed at temperatures lower than 137 °C to avoid amine degradation (Cavaignac et al., 2021). Generally, the pressure of the regeneration column is higher than the atmospheric pressure, i.e. 1.8 bar to 2 bar (Madeddu et al., 2018). The regenerated amine solution, containing traces of CO_2 (lean-amine), is sent to the heat exchanger (HE-101) to be cooled down while it heats up the rich-amine solution. The lean-amine is then mixed with a make-of amine solution in order to keep a constant rate of 0.3 mol CO_2/mol MEA. Finally, the solution is conditioned to 25 °C before being recirculated to the absorber.

Water streams going out of the condensers are recycled and used as make-up water stream. Operating conditions of the process are listed in the supplementary information (SI). The recycling loop is designed to reduce the amount of solvent needed, but it requires a lot of energy. The steam coming from the condenser (top of the regeneration column) can be used to supply a part of the energy required for solvent regeneration in the reboiler at the bottom of the regeneration column.

The desorbed CO_2 leaves the stripper from the top where it is dried in a flash separator in order to remove the maximum of water at the lower dewpoint (0.5 °C) (Ryckebosch et al., 2011), chosen to avoid the freezing of the heat exchanger surface. Then, CO_2 is compressed to 69 bar at 18 °C to be stored in liquid phase (IEA, 2024). This compression section consists of two compressors, each with an efficiency of 80 %. Similarly, biomethane undergoes a dehydration process in a flash separator, then it is compressed using two compressors with 80 % efficiency each and leaves the process at 16 bar and 25 °C. The simulation results are shown in Table 1 and compared with the literature data (references indicated in the column head).

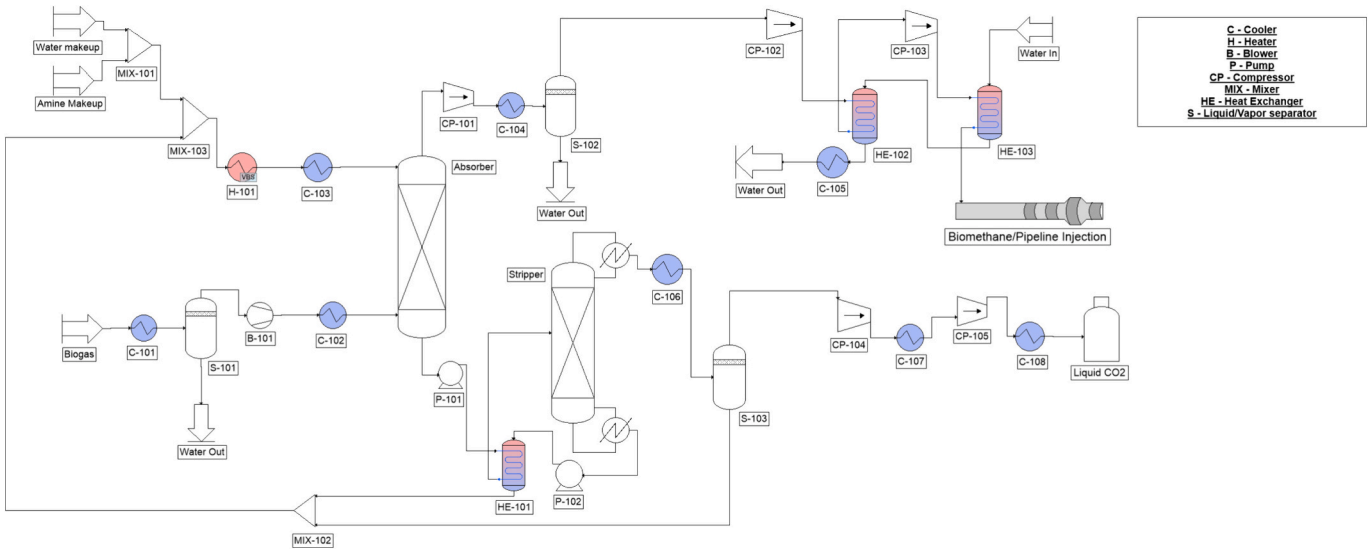

Fig. 1. Flow sheet diagram of the chemical absorption process.

Table 1
Model validation results for the four CC technologies.

	CA			MS			CS			HPWS		
	Results	Ref (Gamba and Pellegrini, 2014)	Error %	Results	Ref (Scholz et al., 2015)	Error %	Results	Ref (Yousef et al., 2018)	Error %	Results	Ref (Wantz et al., 2023)	Error %
$P_{CH4\%}$	98.7	98.7	0.0	98.5	96.0	2.0	96.5	97.1	0.5	96.6	95.0	1.6
$R_{CH4\%}$	99.93	99.90	0.03	99.30	99.60	0.30	99.57	99.95	0.38	98.00	98.00	0.00
$P_{CO_2\%}$	99.5	–	–	98.9	99.4	0.5	99.3	99.9	0.6	95.5	–	–
$R_{CO_2\%}$	98.2	–	–	97.8	93.7	4.0	94.6	95.5	0.9	94.0	–	–
Energy Consumption (Capture system)												
$CE_{(kW)}$	1038.3	–	–	4.2	–	–	469.5	475.2	1.2	3.5	5.0	30.0
RHD/ $HE_{(kW)}$	948.9	–	–	–	–	–	–	–	–	–	–	–
$EE_{(kW)}$	36.6	–	–	27.5	26.7	2.9	365.1	359.7	1.5	16.0	17.0	5.8

P: purity, R: recovery rate, CE: cooling energy, RHD: reboiler heat duty, HE: heating energy, EE: electrical energy. The reboiler heat duty (RHD) is indicative of the heating energy consumption in CA, whereas the term heating energy (HE) is applicable to the other technologies.

3.1.2. *Membrane separation technology*

MS employs modules of hollow fibers composed of polymeric materials as polysulfone, polyimide, or polydimethylsiloxane, which are integrated within a stainless-steel tube (Ardolino et al., 2021). These materials are very permeable to CO_2, H_2O and NH_3, less permeable to H_2S and O_2, and very low permeable to CH_4 and N_2. These materials exhibit a high degree of selectivity in separating methane from carbon dioxide (Chemical Society Reviews, 2023), for example selectivity factors up to 1000/1 were reported for CO_2/CH_4 (Ryckebosch et al., 2011). Two streams leave the membrane unit, permeate stream and retentate stream. Using these different materials, the permeate stream will be composed mainly of CO_2, H_2O and NH_3 while the retentate stream will be composed mainly of CH_4.

This separation operation is based on the permeability and the selectivity of the membrane used (Deng and Hägg, 2010). Permeability G_i and selectivity $\alpha_{i/j}$ of a constituent i are defined in eqs. (1) and (2), in function of the volumetric flow rate (q_i) of the permeating gas (m^3(STP)/h). The required membrane permeation surface area A (m^2) and the pressure on the feed and permeate sides (ΔP_i).

$$G_i = q_i/\Delta P_i \times A \quad (1)$$

$$\alpha_{i/j} = G_i/G_j \quad (2)$$

High surface area is required to achieve a good separation, that is why multiple membrane modules are commonly employed (Chen et al., 2015a).

Deng et al. (Deng and Hägg, 2010) conducted a study using process simulation to upgrade biogas at a farm scale. They explored various membrane module configurations, and a two-stage membrane configuration was identified as the optimal choice among the four considered configurations. This configuration demonstrated high purity and recovery rates for the products. Typically, a single-stage membrane is not a suitable option for biogas upgrading as it fails to meet the desired quality standards. On the other hand, configurations with more than three stages result in excessive energy consumption or economic impracticality (Hoorfar et al., 2018). In addition, removing H_2S and other impurities, still remains a critical point.

The MS simulation of this work (Fig. 2) was implement based on the process outlined by Scholz et al. (Scholz et al., 2015) and with the same operating conditions (presented in Table S2). A commercially available membrane in polyimide was used due to its exceptional selectivity in separating methane from carbon dioxide (CO_2/CH_4 selectivity 60, CO_2 permeance 60 GPU). A three membrane stages process using a single compressor was used to produce grid-compatible biomethane from a binary mixture of CH_4 and CO_2. This configuration allowed a CH_4 purity of 96 mol% and a high CH_4 recovery of 99.5 mol%.

Process layout for MS. Firstly, the raw biogas is compressed in (CP-101) directly to the desired pressure 16 bar. Then it is cooled down in (C-101) to 25 °C before feeding the first membrane stage (M-101) where a bulk separation of CO_2 and CH4 takes place the permeate stream of the first stage contains a significant amount of CH4. This stream, after being compressed to 3.3 bar by compressor (CP-102) will feed the third

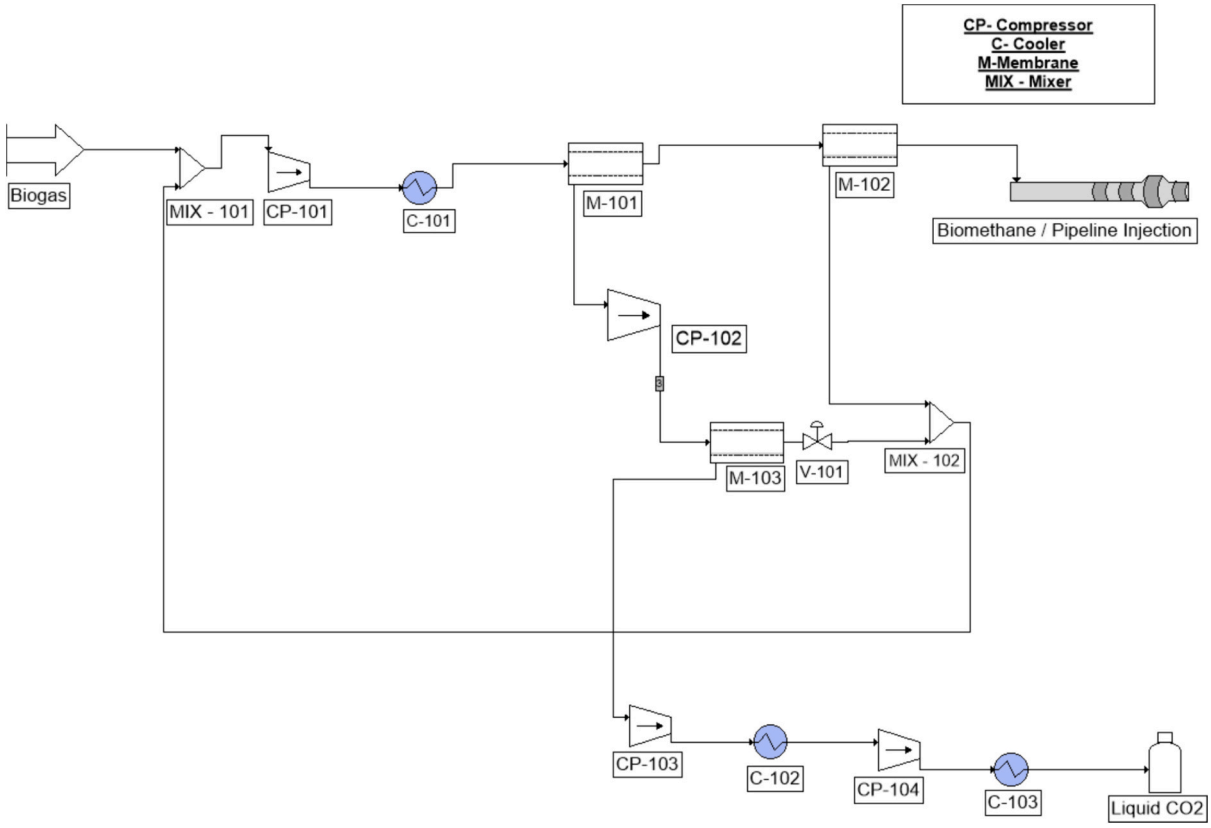

Fig. 2. Flow sheet diagram of the membrane separation process.

membrane stage (M-103) to be purified while the retentate stream will feed the second stage (M-102). In this way, high CH4 purity is obtained in the retentate stream of the second stage. The permeate stream of the second stage is mixed with the retentate stream of the third stage to be recycled and to enhance the CH_4 recovery. The pressure valve (V-101) located at the retentate outlet of the third stage is responsible for regulating the feed pressure within the third stage, thereby creating the driving force required for permeation. Since the increased pressure on the permeate side of the initial stage decreases the driving force within that stage, the ideal feed pressure for the third membrane stage is determined to be 3.3 bar (Scholz et al., 2015).

In order to achieve the required characteristics of CO_2 to be used later (liquid, 69 bar, 18 °C (IEA, 2024).), CO_2 stream is compressed two stages (compressors efficiency is 80 %). The thermodynamic model for the simulations used the Peng-Robinson equation of state.

Table S5 (SI) and Table 1 provide the input biogas composition used in simulations and the results of the model validation with respect to the reference study (Scholz et al., 2015). The configuration and the results were also cross validated with the work of Deng et al. (Deng and Hägg, 2010) and Hoorfar et al. (Hoorfar et al., 2018) where the computed errors were around 3 % (Table S6).

3.1.3. Cryogenic separation technology

CS involves the progressive cooling and condensation of the gas mixture, enabling the selective separation of CO_2, CH_4, and H_2O, etc. based on their distinct dew and sublimation points (Song et al., 2019).

One of the advantages of CS is that the produced liquid CO_2 stream can be used directly in other applications where there is no need to modify its properties (Zhang and Lior, 2006). The incoming biogas must undergo pretreatment and dehydration to achieve a water content of <1 ppm (Yousef et al., 2018). Herein, the raw biogas is assumed to contain only CH_4 and CO_2 before entering the low temperature upgrading process. The CS process layout presented by Yousef et al. (Yousef et al., 2018) was implemented in ProSimPlus® as displayed in Fig. 3 using the parameters in Table S3. The simulations were built with the same operating conditions. The thermodynamic model employed throughout the simulation was the “Peng-Robinson” equation of state. Finally, the configuration and the results were cross-validated with the work of Naeiji et al. (Naeiji et al., 2022).

Process layout for CS process. The raw biogas is compressed by a four-stage compression (CP-101 / CP-104) to increase the gas pressure from 120 to 4983 kPa (distillation column pressure). The pressure ratio used in the compressors (compression section) was 2.54 with 80 % compression efficiency. Pressure drops within simulated components such as piping, distillation columns, and heat exchangers were neglected within the simulation. These compression stages are combined with an inter-cooler at 35 °C, utilizing water cooling (C-101 / C-104). Then, the biogas is cooled down from 35 °C (temperature of the last cooling water heat exchanger) to −65 °C (RF-101) by an auxiliary refrigeration cycle. The refrigeration cycle located before the first distillation column (ST-101) was not included in the simulation, treating it instead as a simple heat exchanger to quantify the energy needed for the system. This refrigeration system could be modeled as a vapor compression cycle with a cascade configuration, employing pure propane in the upper cycle and ethane in the lower cycle as refrigerants. This refrigeration cycle will provide the necessary energy for cooling duties in both the first (ST-101) and second (ST-102) distillation column condensers.

After reaching the required pressure and temperature, biogas is separated in the first (ST-101) and the second distillation columns (ST-102) (details in SI). The purity of the methane stream generated at the top of the first column increased from 60 mol% to 93.75 mol%. Bottom product (liquid CO_2) exits the reboiler with a high purity of 97.45 mol%. Both products are maintained at the same pressure of 4983 kPa and a temperature of 8.73 °C for the liquid CO_2. The methane stream (top stream of the first column) is sent to a second distillation column where its purity increased from 93.75 mol% to 96.55 mol%. These results are

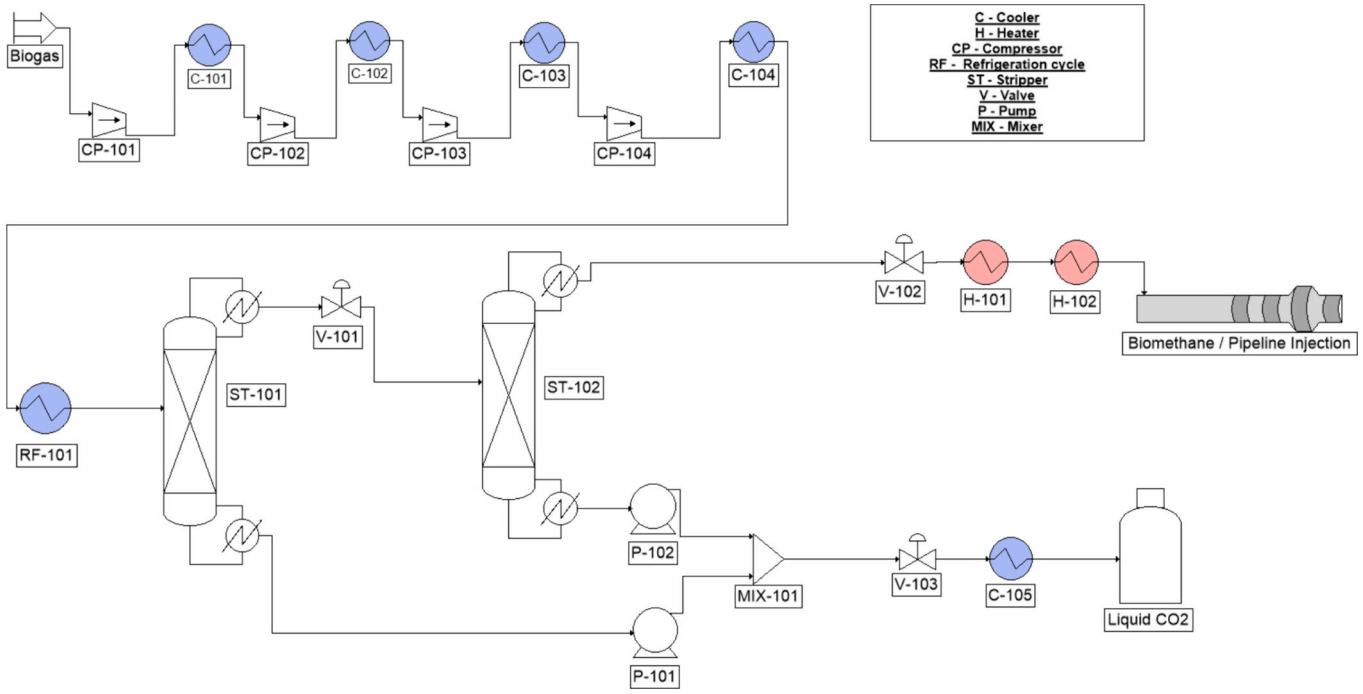

Fig. 3. Flow sheet diagram of the cryogenic separation process.

achieved by reducing the pressure to 4763 kPa by using a valve. This valve ensures the optimal operating pressure for the second column and the highest attainable purity without the risk of frosting. The biomethane is brought to the required parameters (25 °C, 16 bar) through two heaters (H-101 / H-102). Additionally, a second liquid CO_2 stream is generated from the second column, also operating at 4763 kPa. By merging the first and the second CO_2 streams, and after using a valve (V-103) and a cooler (C-105), the liquid CO_2 product attains 69 bar and 18 °C, and a purity of 99.33 mol%. To lower the energy consumption, the condensation steam generated by both condensers can be used directly in the reboilers located at the bottom of each distillation column. This approach leads to an overall reduction in the process's energy consumption, thereby enhancing its competitiveness compared to other technologies.

In this case, the total energy consumed by the cryogenic technology (capture system) will be 834.63 kW (469.53 for cooling +365.1 other electricity consumption), which leads to 0.373 kWh/Nm^3 raw biogas. The specific biogas input composition is reported in Table S5 (SI), while the validation results are presented in Table 1 (errors <1 %).

3.1.4. *High pressure water scrubbing technology*

HPWS stands out as one of the most commercially viable technology due to it is simple operation (Ghaib and Ben-Fares, 2018). This technology offers the advantage of effectively removing both CO_2 and H_2S from the biogas while enriching the product gas with CH_4, taking advantage of the differences in the gases' solubilities in water, as expressed by the Henry's Law (Petronela et al., 2013).

Cozma et al. (Cozma et al., 2015) upgraded the biogas (CH_4, CO_2 and small fractions of impurities like H2S, H2, N2, and O2) by absorbing the CO_2 in water, operating at 8 bar and 20 °C in a set-up with a capacity of 500 Nm3/h. The biomethane purity in the upgraded biogas reached 96.5 mol%, coupled with a high carbon dioxide recovery rate of 99.12 mol% and very low methane loss (0.326 mol%).

Model calibration and validation of the HPWS process was carried out based on the experimental data published by Wantz et al. (Wantz et al., 2023). The thermodynamic model proposed by ProSimPlus called "Amines and acid gases" was used in simulations. This model uses the Peng-Robinson equation of state to model the gas behavior and Henry's law with Poynting correction to model the fugacity of pure liquid. The design parameters are reported in Table S4 and the process configuration is presented in Fig. 4.

Process layout of HPWS technology. The process consists of a theoretical 10-stages packed column working in a closed loop with water as the liquid phase. Raw biogas leaves the digester at a temperature of around 43 °C. This raw biogas is compressed to 9.7 bar (CP-101) and crosses the column through the random packing, in countercurrent with the liquid flow. Water is injected by a pump at the top of the column at 9.7 bar and 25 °C. The biomethane is recovered directly at the top of the column with high purity 96.6 mol%. Then, the gas phase (rich in CO_2) is desorbed from the liquid by passing through a "gas liquid cylindrical cyclone (GLCC)". This separator recovers a gas mixture which is then mixed with the inlet biogas. The aqueous stream containing absorbed CO_2 passes through a vacuum separator to desorb the CO_2 from the water.

The water leaving the separator is sent directly to the column after passing through an exchanger that sets its temperature at 25 °C. In this simulation, the pressure drop inside the column is 20 kPa and the compressors (CP-101 / CP-105) employed exhibit isentropic efficiency of 80 %. The pump (P-101) achieves 98 % efficiency in both mechanical and electrical aspects, coupled with a 65 % volumetric efficiency. The biomethane stream with 96.6 mol% purity is compressed in (CP-102) to 16 bar and cooled down to 25 °C. CO_2 purity of 95.5 mol% can be improved through an optimization step, which involves removing any remaining water in a liquid/vapor separator (GLCC). After the compression through three compressors (CP-103 / CP-105) to 69 bar followed by three coolers (C-105 / C-107) to lower the temperature to 18 °C, a high purity CO_2 stream will be produced. Input gas composition and the simulation results compared with the reference study is presented in Table S5 (SI) and Table 1. In the reference work (Wantz et al., 2023) the CO_2 was not recovered as product but released to the atmosphere.

3.2. *Performance analysis for different treatment capacities*

After the validation of the process models, simulations were performed for the four CC technologies at different treatment capacities

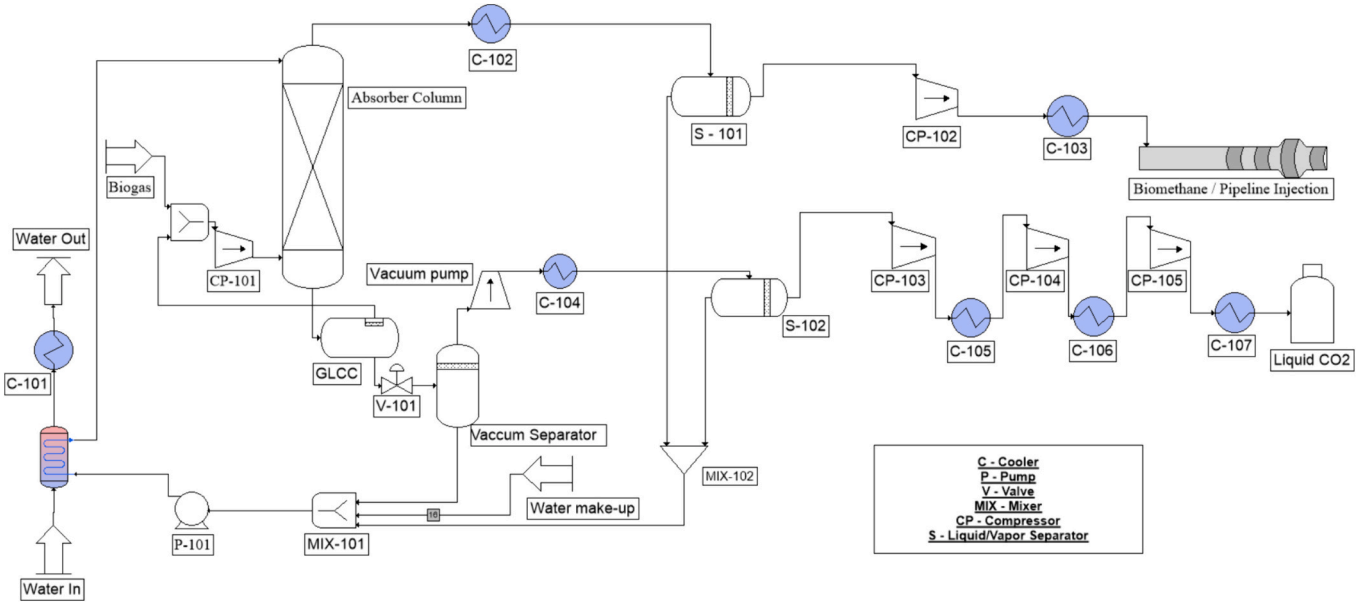

Fig. 4. Flow sheet diagram of the high-pressure water scrubbing process.

related to small, medium and large biogas production plants. Real biogas plants (SI section 2) were considered with their biogas flowrates and compositions, as displayed in Table 3, section 4.1. In the simulations, it was assumed that the incoming gas has been pre-purified and is devoid of any impurities before entering the CC process (inlet gas contains only CO_2 and CH_4). It is important to note that all technologies were considered to be working under optimal conditions. This means that heat integration has been considered for the technologies using heat, namely CS. As CA is a widely used technology at large scales, this already benefits from heat integration (a heat exchanger that uses the output from the stripper to heat the output from the absorber).

In order to assess the influence of the operating parameters on the technology's performance, a sensitivity analysis was performed by using the medium scale biogas production site as benchmark. The sensitivity analysis was conducted by varying one parameter at a time, all other parameters were kept at their reference values (SI section 1 and 2). Two kind of parameters were considered: biogas related (flowrate and composition) and operating parameters specific to each technology.

3.3. Life cycle assessment

Assessing these CC technologies based on performance and energy consumption needs to be completed by an environmental analysis. Life Cycle Assessment (LCA) was performed according to the guidelines established by the International Organization for Standardization (ISO), series 14,040–14,046 (ISO, 2024).

3.3.1. Goal and scope

The system under study and its boundaries are displayed in Fig. 5. The overall system consists of the biogas production (including

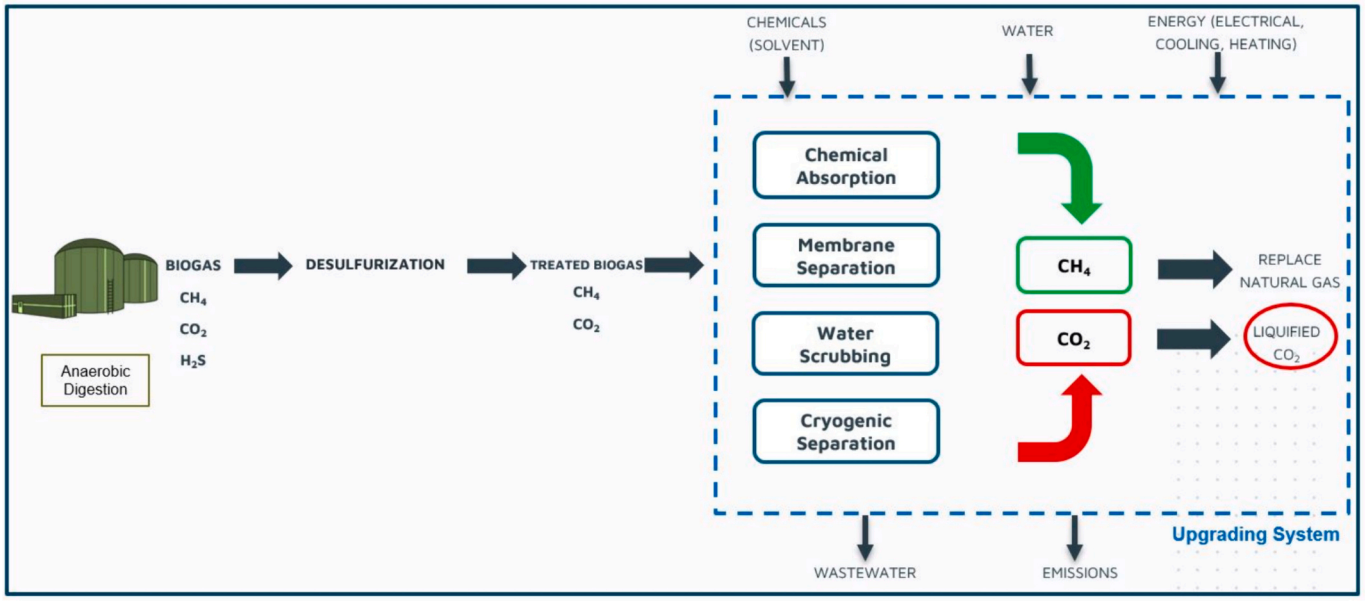

Fig. 5. System boundaries of the considered biogas-to-biomethane plant. The system is composed by the biogas production section and the upgrading – carbon capture section.

dehydration), the biogas pre-treatment (i.e. H_2S removal), and the upgrading process composed of one of the four CC processes, including the compression of biomethane and the compression/liquefaction of CO_2. This corresponds to a cradle-to-gate approach. Two co-products are obtained: high purity biomethane and high purity liquid CO_2 considered as a new valuable product. As the biomethane utilization is the same of that of the natural gas, a system expansion method was used to consider the co-production, i.e. the biomethane replaces the production of natural gas. Therefore, the selected functional unit corresponds to the production of 1 kg of liquified CO_2. This choice enables also the focus on the new product in the perspective of CO_2 valorization alternatives.

3.3.2. Life cycle inventory

The life-cycle process-system is composed of two parts: foreground system composed of the investigated processes, and the background, which commonly includes the processes linked to the foreground and providing utilities, transportation, raw materials, etc. The inventory follows the same categorization. The location of the whole process system is considered in France; hence the background data were selected according to this location. Anaerobic digestion, removal of H_2S are common processes to all scenarios, and their inventory were taken from Ecoinvent 3.9 database and from the literature (Cano et al., 2018). This part was considered as background in the life cycle system, along with all utilities and chemicals production. The four CC technologies represent the foreground part of the life-cycle system, whose inventory data was obtained from simulations. In this way, the adopted system modeling allows to specifically analyze and compare the four CC technologies. The four carbon capture technologies were simulated with the same amount of biogas produced at a medium scale AD plant, with an inlet flowrate of 364.25 Nm^3/h and containing 52 % CH_4 and 48 % CO_2. Small- and large-scale results were not included because only slight variations in the inventory (related to the production of 1 kg liquid CO_2) were detected when changing scales.

3.3.3. Life cycle impact assessment

The environmental impacts were calculated with ReCiPe 2016 v1.13 midpoint (H) method. This method was selected because it provides a harmonized implementation of cause-effect pathways for the calculation of impacts. Additionally, the proposed 18 impact categories in the ReCiPe method are relevant for the investigated system, providing a complete overview of the potential environmental issues (at midpoint).

LCA was implemented with Brightway v2 (open-source software). Brightway was coupled with ProSimPlus through an input/output interface (in Python) to automatize the whole calculation and facilitate the results generation and analysis. The process simulation results, in terms of material and energy flows, were sent to Brightway in which the LCA system was modeled and completed with background inventory from ecoinvent 3.9. The cut-off model was adopted for the background inventory.

A sensitivity analysis was performed on the LCA results by considering distinct heat sources and electricity mix for the CC processes.

4. Results and discussion

4.1. Comparison of CC technologies at different scales

Simulations were carried out with the four modeled CC technologies to asses them at different scales, regarding their capacities to produce pure streams of biomethane and CO_2 with high recovery rates and low energy consumption. In order to attain the desired criteria of purity at different scales, some of the design parameters of the CC technologies were adjusted: the membrane surface for MS, the distillate steam flows rates in CS, the utility water flowrate (used by C-101 in Fig. 4) in HPWS of the large-scale site (necessary to cooldown the recycled water at ambient temperature). Table 2 presents the adapted parameter values, derived from a sensitivity analysis aimed at identifying the optimal values for the studied parameters. These selected values represent the best trade-offs between achieving optimal performance and minimizing energy consumption.

Table 2
Design parameters adaptation (in ProSimPlus) at each scale.

	Membrane Separation MS		
	Small Scale	Medium Scale	Large Scale
Surface membrane 1 (m^2)	77	445	1264
Surface membrane 2 (m^2)	75	437	1241
Surface membrane 3 (m^2)	117	677	1923
	Cryogenic Separation CS		
	Small Scale	Medium Scale	Large Scale
Distillate steam flow rate, ST-101 (kmol/h per 1 kmol/h of feed)	0.54	0.55	0.55
	High pressure water scrubbing HPWS		
	Small Scale	Medium Scale	Large Scale
Utility: Water flow rate (m^3/h)	8	8	14

The comparison was made in terms of purity and recovery of CH_4 and CO_2, as well as energy consumptions. Furthermore, the calculation of the theoretical energy production (TEP) by biomethane was added to set an upper limit to total energy consumption (TE). TEP was calculated by using the lower heating value of biomethane (35.22 MJ/Nm^3). As matter of fact, TE must not be larger than TEP, otherwise the process is not economically feasible. Table 3 presents the comparative overview of these four technologies operating under identical inputs at a given scale, and then for different scales, and with the same objective of the output streams: methane purity was imposed to be at least 96 mol% (compatible with the natural gas grid), and each CC technology aimed to achieve the highest possible CO_2 purity.

Regardless the scale, CA technology exhibits the highest energy consumption, mainly attributed to the energy-intensive solvent regeneration process. In other words, CA needs 3.4, 3.6 and 2.6 times more energy (in $kWh/kgCO_2$) than MS, CS and HPWS, respectively. The CA and MS perform the best for the purity and recovery of the gas streams, while HPWS achieves the lowest gas purities and recoveries. In addition, CA requires chemicals, HPWS and CA consume important amounts of water, which is not the case of MS and CS. Concerning the energy consumption, MS has the lowest total consumption, which is practically only electricity. MS and CS electricity consumptions are very close; however, CS requires in addition non-negligible cooling while MS operates at ambient temperature. Moreover, CS is not yet implemented at commercial scale for biogas separation. Additionally, biomethane produced by MS is directly available at the required pressure for grid injection. This is in contrast to HWPS technology, which demands high electrical energy consumption to compress gases at the required pressure for utilization. According to these results, MS technology emerges as the most suitable method, yielding high-quality biomethane and carbon dioxide products with minimal energy consumption.

Albeit not fair, the comparison between the three scales shows a slight diminution of the energy consumption, in kWh/Nm3 biogas, with the scale increase, varying between 6 % and 14 % for MS and CA respectively. A variation of 25 % was observed for HPWS for COE parameter. A slight increase in terms of TE $kWh/kg\ CO_2$ was observed with the scale increase. It is to be noted that the input conditions are not identical between the three scales, in Table 3 and, hence, the comparison between the scales using the results presented in Table 3 could be biased.

The simulation results were compared with available literature in Table 3. The available data are either simulation results or information from real sites, from original researches or from review articles. A good agreement is observed for the purity and recovery of the gas streams, when compared with the real or simulation data from the literature. Concerning the energy consumption, the simulation results obtained in

Table 3
Technical evaluation of the four CC technologies at different scales and comparison with literature data (1 - *10_methanobois_suivi_methanisation_rapport.pdf*, 2023; 2 - *5_methagri32_suivi_methanisation_rapport.pdf*, 2023; 3 - Dong et al., 2019; 4 - Starr et al., 2012; 5 - *2012_BioRegions_BiogasUpgradingTechnologyReview_ENGLISH.pdf*, 2024; 6 - Sgc, n.d.; 7 - *SGC166.pdf*, 2023; 8 - Weiland, 2010; 9 - Karimi et al., 2023; 10 - Esposito et al., 2019; 11 - Hosseinipour and Mehrpooya, 2019; 12 - Ahmed et al., 2021; 13 - Chen et al., 2015b; 14 - Hashemi et al., 2019; 15 - Yousef et al., 2017; 16 - Our companies – Rootselaar Group, 2024; 17 - Petersson and Wellinger, n.d.; 18 - Bekkering et al., 2010; 19 - Ardolino et al., 2021; 20 - Wantz et al., 2023; 21 - *1_gatinais_suivi_methanisation_rapport_vf (Inje).pdf*, n.d.; 22 - Patterson et al., 2011b).

	This study												Literature studies						
Input parameters	**Small** [1]				**Medium** [2]				**Large** [3]				CA – Sim	CA – Real	MS – Sim	MS – Real	CS – Sim	HPWS – Sim	HPWS – Real
Input biogas flowrate (Nm3/h)	62.14				364.25				911.22										
Input biogas composition	CH_4: 50.6 mol % CO_2: 49.4 mol %				CH_4: 52 mol % CO_2: 48 mol %				CH_4: 54.5 mol % CO_2: 45.5 mol %										
Input biogas temperature (°C)	35				35				37										
Input biogas pressure (Bar)	1.013				1.013				1.013										
Simulation results	**CA**	**MS**	**CS**	**HPWS**	**CA**	**MS**	**CS**	**HPWS**	**CA**	**MS**	**CS**	**HPWS**							
$P_{CH4\%}$	98	99	97	96	98	99	97	96	98	99	97	96	99 [4]^{r}	> 99 [5]^{L}	99 [6]^{r}	99 [5]^{M}	97 [7]^{r}	96 [8]^{r}	99 [5]^{L}
$P_{CO2\%}$	99.6	99.0	99.0	95.7	99.5	99.0	98.0	96.2	99.0	98.0	93.0	96.3	99.0 [9]^{r}	–	99.9 [10]^{L}	99.9 [10]^{L}	98.4 [11]^{r}	90.0 [12]^{r}	–
$R_{CH4\%}$	99.9	99.0	99.0	96.5	99.9	99.3	98.4	97.0	99.9	98.9	93.8	97.5	99.9 [13]^{L}	99.9 [5]^{L}	99.5 [13]^{L}	99.5 [5]^{M}	98.7 [14]^{r}	98.0 [13]^{L}	98.0 [5]^{L}
$R_{CO2\%}$	98.0	99.0	97.0	97.0	98.0	99.0	96.6	97.0	98.0	99.0	96.3	96.0	99.0 [19]^{r}	–	97.0 [19]^{r}	–	95.6 [15]^{L}	98.0 [19]^{r}	–
TEP (kW)	311	309	309	300	1877	1864	1847	1824	4937	4886	4635	4818	–	–	–	–	–	–	–
TE (kW)	158	88	83	95	686	280	261	322	1409	440	411	532	–	–	–	–	–	–	–
TE (kWh/kgCO2)	1.73	0.53	0.47	0.67	1.74	0.54	0.48	0.67	1.75	0.55	0.52	0.67	–	–	–	–	–	–	–
HEE (kWh/Nm^3 Biogas)	0.54	0.00	0.04	0.00	0.53	0.00	0.03	0.00	0.50	0.00	0.03	0.00	0.27 – 0.75 [19]^{r}	0.68 [16]^{S}	–	–	–	–	–
COE (kWh/Nm^3 Biogas)	0.780	0.028	0.185	0.138	0.760	0.028	0.180	0.120	0.730	0.028	0.180	0.110	–	–	–	–	–	–	–
ELE (kWh/Nm^3 Biogas)	0.029	0.196	0.171	0.290	0.029	0.195	0.171	0.280	0.030	0.195	0.170	0.270	0.030 – 0.150 [19]^{r}	< 0.150 [17]^{M}	0.190 ; 0.270 [22]^{r}	0.300 [21]^{S}	0.420 [22]^{r}	0.200 ; 0.330 [18]^{r} ; [19]^{r}	0.200 – 0.400 [20]^{S}
TE (kWh/Nm^3 Biogas)	1.349	0.224	0.396	0.428	1.319	0.223	0.381	0.400	1.260	0.223	0.380	0.380	–	–	–	0.270– 0.290 [10]^{L}	–	–	0.380 [20]^{S}

P: purity, mol%; R: recovery rate, %; TE: Total energy consumption, kW or kWh/kg CO_2 produced or kWh/Nm3 Biogas; TEP: Total energy production, kW; HEE: Heating energy, kWh/Nm3 Biogas; COE: Cooling energy, kWh/Nm3 Biogas; ELE: Electrical energy, kWh/Nm3 Biogas.
Sim: literature, data obtained from simulation; Real: data from real sites;
^{S}: Small scale;
^{M}: Medium scale;
^{L}: Large scale;
^{r}: Review literature.

this work fall in the ranges found in the literature, except two cases. MS and CS energy (ELE) provided by (*1_gatinais_suivi_methanisation_rapport_vf (Inje).pdf*, n.d.; Patterson et al., 2011b) respectively are much higher all other ELE data; however, the data presented in these references are not peer-reviewed but originates from technical reports. In case of MS, another source could be the design of the membrane installation (membrane surface and module cascade), which needs to be optimized in order to minimize the energy consumption.

In conclusion, upon examining the results across simulations in different conditions, MS represents the best compromise among the investigated upgrading methods, with high purity and recovery rates and lowest energy consumption.

4.2. Sensitivity analysis

The parameters considered in the sensitivity analysis are presented in Table 4 along with their reference values and their variation intervals. Only the most influential parameter in each technology will be discussed here, while the other results are presented in SI. The results presented in figures below include the variation of purity and recovery of CO_2 and CH_4 streams and the most significant energy consumption item (heat or electricity) of each technology. The red dotted line indicates the nominal point.

4.2.1. Chemical absorption process

The overall performance of the process is notably influenced by key parameters such as heat losses in the stripper column, biogas flowrate, and biogas composition which stands out as the most significant. Fig. 6 illustrates the impact of varying the CO_2 composition on biomethane and CO_2 stream purity and recovery, and the reboiler duty expressed per raw biogas (kWh/Nm^3).

The CO_2 fraction in biogas can vary in a range of 30–50 mol% (Canu and Pagin, 2022). Thus, this study explores the impact of variation in the CO_2 fraction around the nominal point, 48 % (red line), on the overall biogas upgrading process. Results indicate that a higher CO_2 concentration in biogas leads to an increase in the reboiler duty per unit of treated raw biogas, as the biogas flowrate feeding the system remains

Table 4
Parameters investigated, their reference value for each technology and the interval of variation.

CA	MS	CS	HPWS
Stripper heat losses 0 kW; (0–30)	Pressure at 3rd stage 3.3 bar; (2.5–3.7)	Reflux ratio 2.8; (0.3–3.5) Vapor distillate ratio 0.55; (0.05–0.68)	Pressure of absorber column 9.7 bar; (6–10.5) GLCC pressure 4 bar; (2.8–6)
Biogas flowrate 364.25 Nm^3/h; (300–440) CO_2 Fraction 48 %; (30–55)			

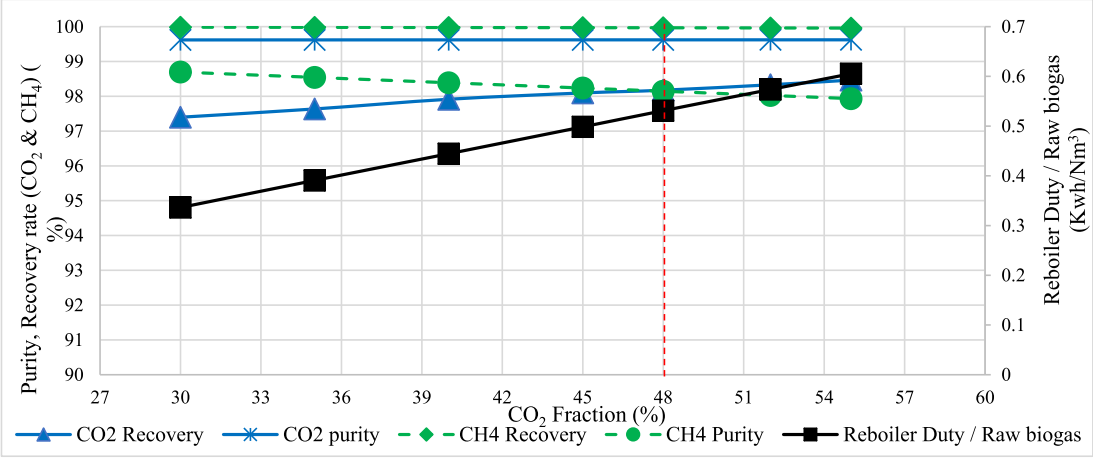

Fig. 6. Influence of the CO_2 fraction in the biogas on process performances.

constant. It is observed that the CO_2 purity and CH_4 recovery keep an almost constant value regardless the change on CO_2 content. Furthermore, CO_2 recovery increases linearly when there is a larger content of CO_2 while CH_4 purity decreases. Even if the CO_2 recovery increases, a larger CO_2 content implies a larger flowrate of CO_2 which, in this case, is translated into more CO_2 going to the CH_4 stream. However, its purity remains above the target value ($\geq$ 96 %, even when the biogas contains 55 % CO_2. For a 10 % variation in CO_2 fraction, the energy variation is of 10 %, while the variation in purities and recovery rates for both products are very small. Besides, the results of the sensitivity analysis for the other two parameters demonstrate lower influence on the process performance (see SI), and suggest that the optimization of a CA process should be conducted primarily with respect to the CO_2 fraction in the biogas.

4.2.2. Membrane separation process

The pressure of the third membrane stage has a significant influence on both the process performance and the energy consumption, as well as other parameters (Scholz et al., 2015). Fig. 7 illustrates the influence of changing this pressure while keeping the same inlet pressure at the first stage (16 bar). The influence of the biogas flowrate and composition is illustrated in SI.

A decrease in the inlet pressure of the 3rd membrane stage implies a higher transmembrane pressure which increases the flowrate going into this stage. As there is a lower input pressure in stage three, a lower flowrate of permeate will be produced. Therefore, the CO_2 recovery will decrease. For instance, the flowrate leaving the first stage decreases from 28.22 to 8.40 kmol/h in the permeate side, leading to a decrease from 7.67 to 7.21 kmol/h at the permeate side of the third stage.

CH_4 recovery is determined by the retentate stream, and as the pressure at the retentate side remains constant, the performance of the second membrane stage also remains constant. Reducing the pressure at the inlet of the third membrane stage will lower the pressure of the recycled stream entering the compressor leading to an increase of electrical consumption. Therefore, this is a parameter that requires careful control to avoid process variations, as any deviation could lead to increased energy consumption and performance losses. Noticeably, the energy consumption decreases by 50 % when this pressure increases from 2.5 to 2.65 bar, (5.5 % of pressure variation) and the CH_4 purity increases by 5 %. Additionally, a 10 % decrease from the nominal point ($\sim$ 3.3 to 3 bars) does not lead to any change in performance. However, when there is a 10 % increase, i.e. 3.6 bar, the recoveries and purities of CH_4 and CO_2 streams slightly decrease but remain within the desired target. The influence of biogas flowrate and composition is quite linear for all performance criteria (SI): a 10 % increase in flowrate or CO_2 fraction results in 10 % or 3.5 % increase in electricity consumption, respectively.

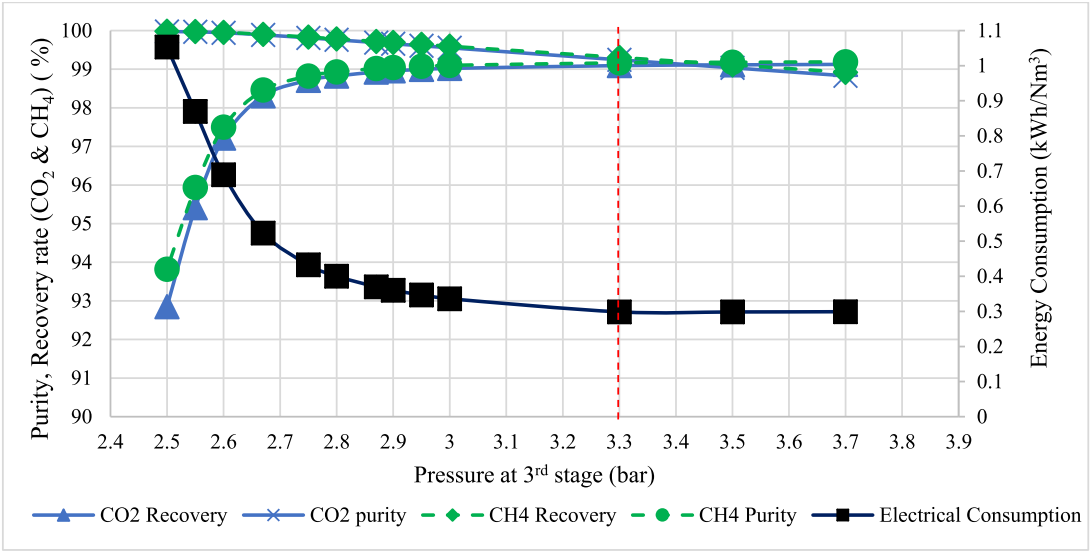

Fig. 7. Influence of pressure at permeate side on the process performances.

4.2.3. Cryogenic separation process

The reflux ratio, biogas flow rate, and ratio of distillate vapor are critical parameters affecting process performance, specifically influencing the purities of CH_4 and CO_2. Fig. 8 presents the results of varying the reflux ratio, while the other results are provided in SI.

The range of variation from 0.3 to 3.5 was chosen based on the numerical convergence of the simulations of the distillation columns. Increasing the reflux ratio adversely impacts the column's energy consumption, resulting in an increase of the reboiler duty in each column (total reboiler duty). Moreover, increasing the reflux ratio from 0.3 to 2.8 results in a significant enhancement in CH_4 purity. The trends of CO_2 recovery and of CH_4 purity are similar, and align with the product specifications (purity ≥96 % mol). The same happens for CO_2 purity (equivalent trend to CH_4 recovery). However, continuing increasing the reflux ratio to 3.5 exhibits only a slight positive impact, therefore it is not necessary to increase it beyond 3, while the required energy for the reboiler increases continuously. An increase in the total reboiler duty of 7.5 % was obtained when increasing by 26 % the reflux ratio (between 2.3 and 3.1), while there is no significant impact on purities and recovery rates. The vapor distillate ratio is a design parameter, very sensitive for all performance criteria. It should be obtained by process optimization (SI). Concerning the biogas flow rate and composition, there is practically no influence on recovery/purity, while a flowrate and CO_2 fraction variation of 10 % induces a variation in energy demand of 10 % and 20 % respectively.

4.2.4. High pressure water scrubbing process

The performance of this CC process is significantly influenced by the pressure inside the absorber column, in addition to the pressure of the gas-liquid cylindrical cyclone (GLCC) and the biogas flow rate. Fig. 9 displays the variation of the pressure of the absorber column, which primarily affects the purity of the biomethane stream. The impact of varying the other parameters is illustrated in SI.

When the absorber pressure was lower than 8 bars, lower CH_4 purity (<96 mol%) was observed. CO_2 recovery decreases because the CO_2 losses will increase. CO_2 purity and CH_4 recovery slightly decrease. In the other hand, decreasing the pressure will decrease the methane solubility in the liquid phase and leads to an increase in CH_4 recovery in the absorber. Regarding energy consumption of the process, there is an increase in electrical energy, as more energy is required to compress biogas and water at high pressure. Therefore, setting absorber pressure requires careful consideration to achieve the desired methane purity without excessive energy consumption and minimizing methane leaks. It is important to highlight that there is a slight increase in the electrical energy consumption when increasing the absorber column pressure. For instance, a 7.6 % increase in the absorber pressure results in a 4.13 % increase of electrical energy consumption. The biogas flowrate has practically no influence on the purities and recoveries (SI), but a 10 % increase leads to an energy consumption increase of 10 %, while a 10 % increase of CO_2 fraction in biogas leads to 7.5 % more energy demand.

4.3. LCA results

4.3.1. Life cycle inventory results

LCA was applied to the medium scale plant with the four investigated technologies for the production of 1 kg of high purity liquid CO_2. The produced biomethane was considered for injection into the natural gas network and thus allocated to the production of natural gas.

The foreground inventory is composed of different energy consumptions split according to the type of process in: cooling, heating, compression, pumps and other electricity consumptions. Even if the recirculation of water and amines is implemented in the process, a certain amount must be completed to compensate for losses or amine degradation.

In addition, CO_2 and CH_4 can be emitted by leakage. It was considered, as in (Lombardi and Francini, 2020), that a small amount of the outlet streams is emitted, corresponding to 0.1 %, 1 % and 2 % of the recovered CO_2 stream for CA, MS and HPWS, respectively, and with their respective compositions (traces of biomethane, water). The emissions for CS were assumed as 0.1 % of the recovered CO_2 stream. Concerning the biomethane stream, no leakage was included in calculation because the subtraction of the avoided fossil methane production having the same leakages practically leads to their compensation.

Table 5 presents the inventory results related to the production of 1 kg liquid CO_2 (functional unit). If the heat source necessary for the CC processes is a portion of the on-site produced biomethane, in this case the net CH_4 production decreases and the CO_2 emissions increase due to the combustion in the heating system. This is considered as a likely option and the inventory was adapted to this scenario. Biomethane was chosen and not raw biogas in order to not distort the technical performances of the simulated CC technologies by using a part of the raw biogas to produce heat, which would have led to the modification of the treatment capacities and the impossibility to validate the simulations versus the site/literature data). Hence, different heat sources were considered for a sensitivity analysis: 1) natural gas from the grid (scenarios CA, CS), 2) biomethane produced on site (grid-compatible) (scenarios CA_2, CS_2).

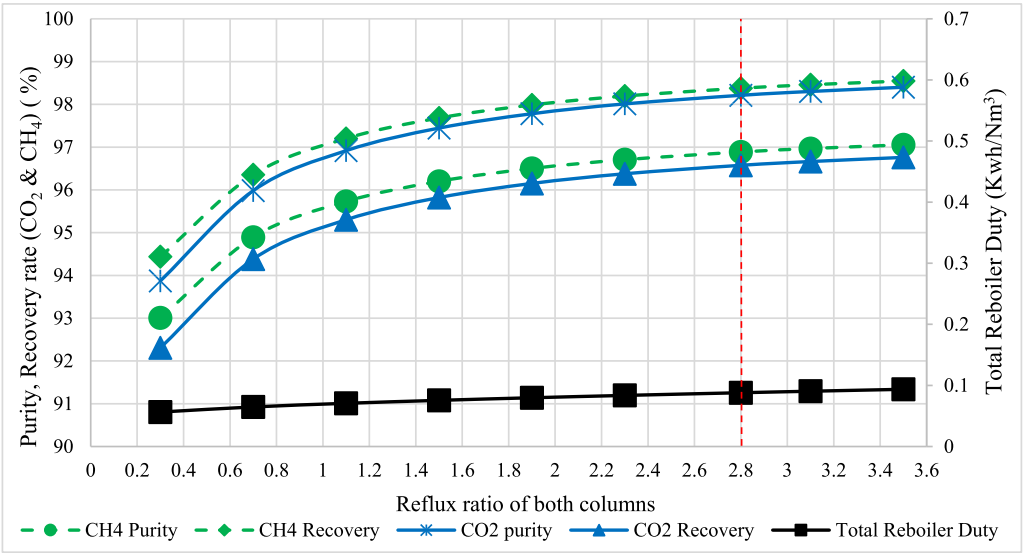

Fig. 8. Influence of reflux ratio in both distillation columns on the process performances.

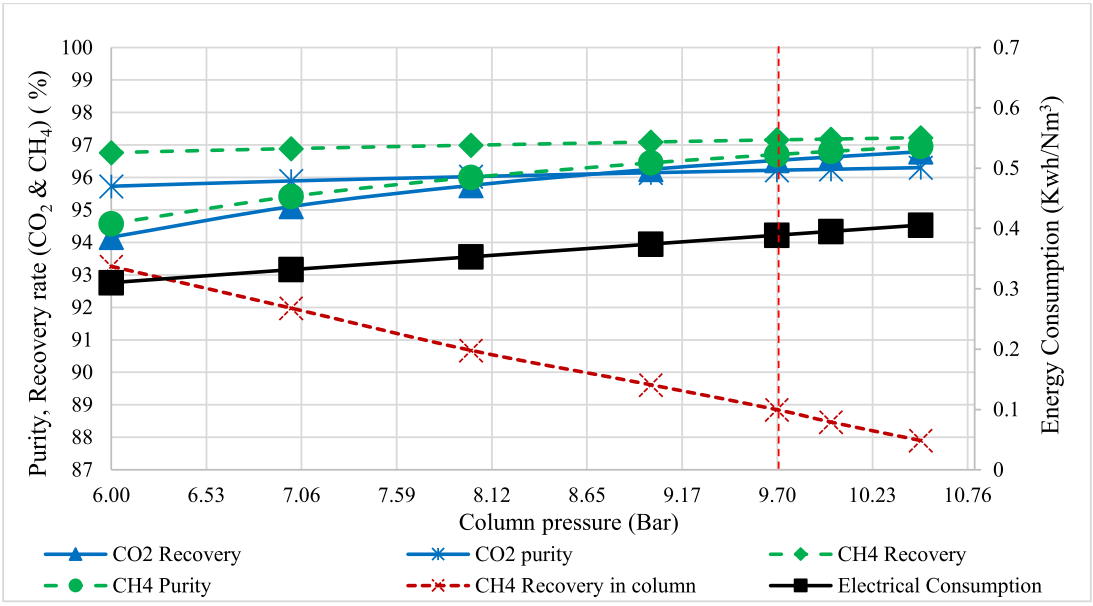

Fig. 9. Influence of absorption pressure on the process performances.

Table 5
Life cycle inventory for the four CC technologies, related to the production of 1 kg CO_2 liquid.

Inventory	CA	CA_2	MS	CS	CS_2	HPWS	Ecoinvent 3.9 data-set
Production							
Biomethane net, Nm^3	0.572	0.439	0.562	0.577	0.571	0.578	Avoided natural gas production Market for natural gas, low pressure [FR]
Consumptions							
Biogas, Nm^3	1.080	1.080	1.079	1.093	1.093	1.105	Anaerobic digestion of manure [RoW]
Electricity for cooling, kWh	1.582	1.574	0.213	0.411	0.411	0.244	Market for electricity, low voltage [FR]
Electricity for compression, kWh	0.122	0.113	0.111	0.000	0	0.111	Market for electricity, low voltage [FR]
Electricity, other consumptions, kWh	0.032	0.032	0.211	0.187	0.187	0.312	Market for electricity, low voltage [FR]
Heat from natural gas, MJ	4.129	–	–	0.530	–	–	Market for heat, district or industrial, natural gas [Europe without Switzerland]
Heat from biomethane, MJ Combustion on site	–	4.129	–	–	0.530	–	
Water, kg	0.037	0.037	–	–		3.5×10^{-3}	Market for tap water [RER]
MEA, kg	1.99×10^{-4}	1.99×10^{-4}	–	–	–	–	Market for monoethanolamine [GLO]
Emissions							
Leakage, CO_2 non-fossil, kg	9.99×10^{-4}	9.99×10^{-4}	1.01×10^{-2}	9.94×10^{-4}	9.94×10^{-4}	2.01×10^{-2}	Carbon dioxide, non-fossil, air
Leakage, CH_4 non-fossil, kg	1.26×10^{-7}	1.26×10^{-7}	2.8×10^{-5}	6.60×10^{-6}	6.60×10^{-6}	2.34×10^{-4}	Methane, non-fossil, air
Combustion on site, CO_2 non-fossil, kg	–	0.261	–	–	0.012	–	Carbon dioxide, non-fossil, air

4.3.2. Environmental impact results

Given the specificity of the inventory essentially composed by energy consumptions, the impact results are expected to be strongly determined by the type of energy used. The ReCiPe midpoint impact results are presented in Fig. 10 by the means of normalization of each impact category with respect to its maximum positive value. The impact value is composed of the contributions of biogas production/ purification and of the CC technology. Negative values correspond to the avoided natural gas production.

In Fig. 10, CA, CA_2, CS and CS_2 correspond to the scenarios mentioned in Table 5, i.e. the heat source is either a portion of the biomethane produced on site or the natural gas from the gas grid. The impact net values (after subtraction of avoided impact) are presented in Table 6. The colors represent the ranking of the six scenarios by impact category.

CA and CA_2 demonstrate the highest values for all impact categories. This is due to the important energy consumption, especially heat.

Conversely, MS displays the lowest impacts overall and confirms the best performances from the environmental point of view. CS and CS_2 perform similarly and better than HPWS. The negative impact categories SOP and FFP are due to the avoided infrastructure for natural gas extraction and the natural gas itself as natural resource. It should be mentioned that the final utilization, i.e. the gas combustion was not included in the system's life cycle boundaries. If the final combustion is

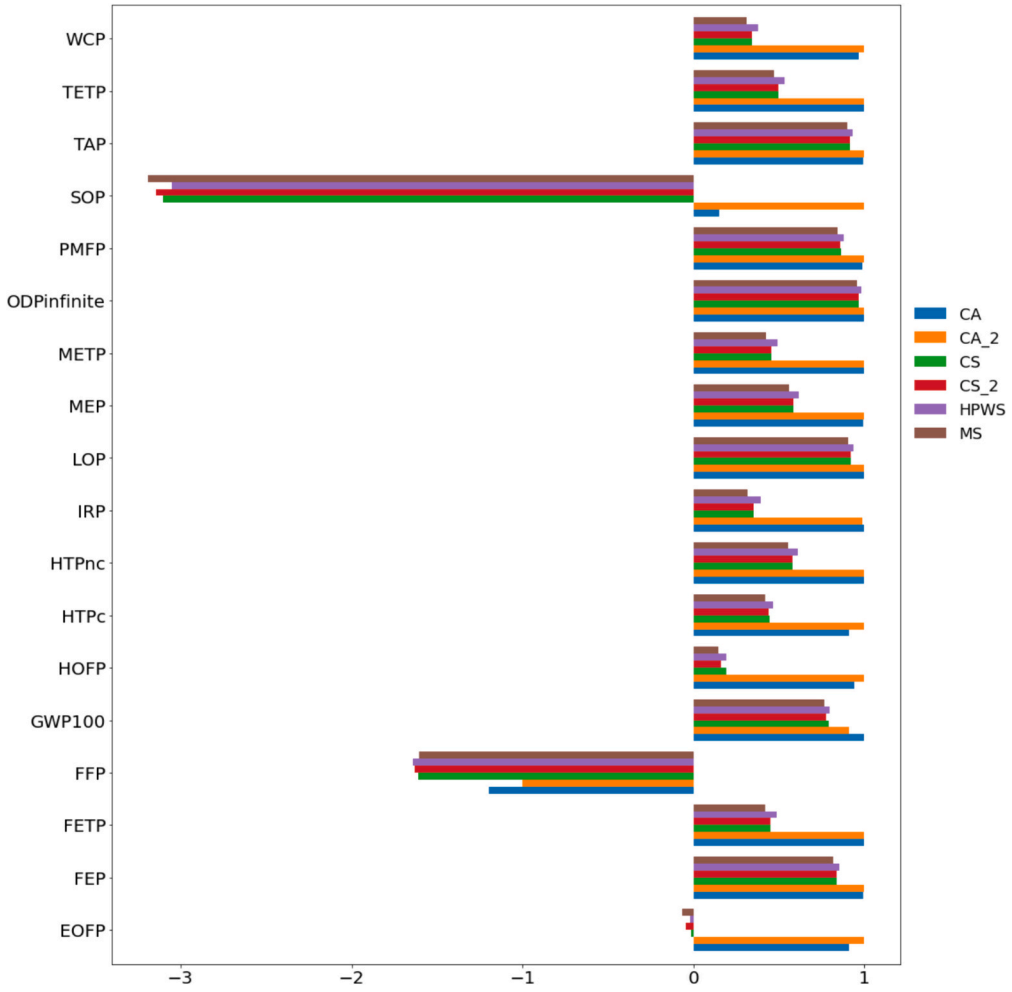

Fig. 10. Comparison of the LCA results (ReCiPe midpoint impacts) for the four CC technologies. Each impact category was normalized with respect to the highest value observed.

included, then GWP100 category would also have a negative value due to the avoided fossil CO_2 emission. To better understand the impact results, the contribution of the different processes is presented in Fig. 11 for GWP100 impact and in SI section 4 for all other impact categories.

Biogas production is the activity with the largest contribution to global warming. This contribution is almost the same for all the scenarios since the same inputs were used in the simulations of the four CC technologies. Insignificant variations are due to functional unit used, i.e. 1 kg of CO_2 liquid. Scenario CA_2 assumes that a part of the produced biomethane is burned in a boiler. Hence, the net biomethane production is lesser, but this avoids fossil CO_2 emissions from heat fossil sources. Moreover, CA process demands higher energy consumption compared to the other three technologies. To mitigate the environmental impact of emissions from energy production sources, it is crucial for this technology to be powered by green energy. As mentioned above, MS, CS, CS_2 and HPWS behave very closely in global warming category, with MS outperforming the other technologies.

The contribution of the chemicals (amines) and water is negligible in comparison with energy contribution. However, water consumption is better accounted for in WCP category (SI section 4) where the main contributor is the electricity production. Water consumption is correlated to the electricity mix (France) because of the high proportion of nuclear power (water for cooling). IRP is another category highly affected by this electricity mix consumption. Other impacts with similar trends are FETP, METP, TETP, MEP, HTPnc.

Concerning the gas leakages, biomethane release contributes to global warming in an extent dependent on the onsite installation. The leakage is a blur parameter, which requires specific measurement on sites to be known. An estimation was made in this work in the range of 1 %, leading to a contribution to global warming of 0.008 kg CO_2 eq for HPWS (0.7 % of GWP100 impact, the highest), 0.001 for MS (0.09 %), 0.0002 for CS (0.02 %), and still lower for CA. An increase of the leakage fraction will not change the conclusions; e.g. for 5 % losses, a contribution of maximum 3.5 % could be observed.

From the impact results, MS has always the lowest net impact value, however, the difference with CS and HPWS is not important. In case of global warming, the difference between CS, CS_2, HPWS and MS is maximum 3.2 %, which cannot clearly discriminate these three CC technologies. In contrast, the difference between CA variants and MS amounts 20–30 %.

In their study, Florio et al. (Florio et al., 2019) reached similar conclusion. Comparing the environmental benefits of biomethane produced by five different biogas upgrading technologies (CA, MS, PSA, CS, and HPWS), the authors showed that MS had the lowest environmental impacts across all the examined impact categories.

From the sensitivity analysis, variations in energy consumption could be induced in case of variation of the input biogas stream, i.e. composition and flowrate (section 3.2). Supposing the process is implemented with the optimal design and operating parameters, a variation of 10 % in the biogas input flowrate leads to 10 % variation in electricity consumption (kWh/Nm^3 biogas treated) of MS, CS and HPWS and no influence on CA energy consumption. A 10 % increase in CO_2

Table 6
ReCiPe midpoint impact results using the French electricity mix.

Impacts			CA	CA_2	MS	CS	CS_2	HPWS
Photochemical oxidant formation potential ecosystems	EOFP	kg NOx-Eq	4.60×10^{-4}	5.05×10^{-4}	-3.21×10^{-5}	-6.33×10^{-6}	-2.37×10^{-5}	-9.49×10^{-6}
Freshwater eutrophication potential	FEP	kg P-Eq	2.75×10^{-4}	2.77×10^{-4}	2.27×10^{-4}	2.32×10^{-4}	2.32×10^{-4}	2.37×10^{-4}
Freshwater ecotoxicity potential	FETP	kg 1,4-DCB-Eq	7.62×10^{-2}	7.61×10^{-2}	3.20×10^{-2}	3.45×10^{-2}	3.44×10^{-2}	3.71×10^{-2}
Fossil fuel potential	FFP	kg oil-Eq	-3.06×10^{-1}	-2.55×10^{-1}	-4.10×10^{-1}	-4.11×10^{-1}	-4.16×10^{-1}	-4.19×10^{-1}
Global warming potential	GWP100	kg CO_2-Eq	1.42×10^{0}	1.29×10^{0}	1.08×10^{0}	1.13×10^{0}	1.10×10^{0}	1.13×10^{0}
Photochemical oxidant formation potential humans	HOFP	kg NOx-Eq	5.40×10^{-4}	5.73×10^{-4}	8.40×10^{-5}	1.10×10^{-4}	9.39×10^{-5}	1.09×10^{-4}
Human toxicity potential: carcinogenic	HTPc	kg 1,4-DCB-Eq	2.49×10^{-2}	2.74×10^{-2}	1.15×10^{-2}	1.22×10^{-2}	1.20×10^{-2}	1.27×10^{-2}
Human toxicity potential: non-carcinogenic	HTPnc	kg 1,4-DCB-Eq	1.06×10^{0}	1.07×10^{0}	5.90×10^{-1}	6.20×10^{-1}	6.19×10^{-1}	6.51×10^{-1}
Ionizing radiation potential	IRP	kg CO-60-Eq	9.89×10^{-1}	9.79×10^{-1}	3.13×10^{-1}	3.49×10^{-1}	3.49×10^{-1}	3.88×10^{-1}
Agricultural land occupation	LOP	m^2*a crop-Eq	5.48×10^{-2}	5.50×10^{-2}	4.99×10^{-2}	5.07×10^{-2}	5.07×10^{-2}	5.16×10^{-2}
Marine eutrophication potential	MEP	kg N-Eq	3.67×10^{-5}	3.70×10^{-5}	2.07×10^{-5}	2.17×10^{-5}	2.17×10^{-5}	2.27×10^{-5}
Marine ecotoxicity potential	METP	kg 1,4-DCB-Eq	9.61×10^{-2}	9.60×10^{-2}	4.08×10^{-2}	4.39×10^{-2}	4.39×10^{-2}	4.72×10^{-2}
Ozone depletion potential	ODPinfinite	kg CFC-11-Eq	3.19×10^{-6}	3.19×10^{-6}	3.05×10^{-6}	3.09×10^{-6}	3.09×10^{-6}	3.13×10^{-6}
Particulate matter formation potential	PMFP	kg PM2.5-Eq	1.56×10^{-3}	1.58×10^{-3}	1.33×10^{-3}	1.36×10^{-3}	1.36×10^{-3}	1.39×10^{-3}
Surplus ore potential	SOP	kg SO2-Eq	2.05×10^{-4}	1.37×10^{-3}	-4.38×10^{-3}	-4.25×10^{-3}	-4.31×10^{-3}	-4.19×10^{-3}
Terrestrial acidification potential	TAP	kg SO2-Eq	6.50×10^{-3}	6.53×10^{-3}	5.89×10^{-3}	5.99×10^{-3}	5.98×10^{-3}	6.08×10^{-3}
Terrestrial ecotoxicity potential	TETP	kg 1,4-DCB-Eq	4.91×10^{0}	4.93×10^{0}	2.31×10^{0}	2.46×10^{0}	2.46×10^{0}	2.62×10^{0}
Water consumption potential	WCP	m^3	6.27×10^{-3}	6.48×10^{-3}	2.01×10^{-3}	2.23×10^{-3}	2.23×10^{-3}	2.46×10^{-3}

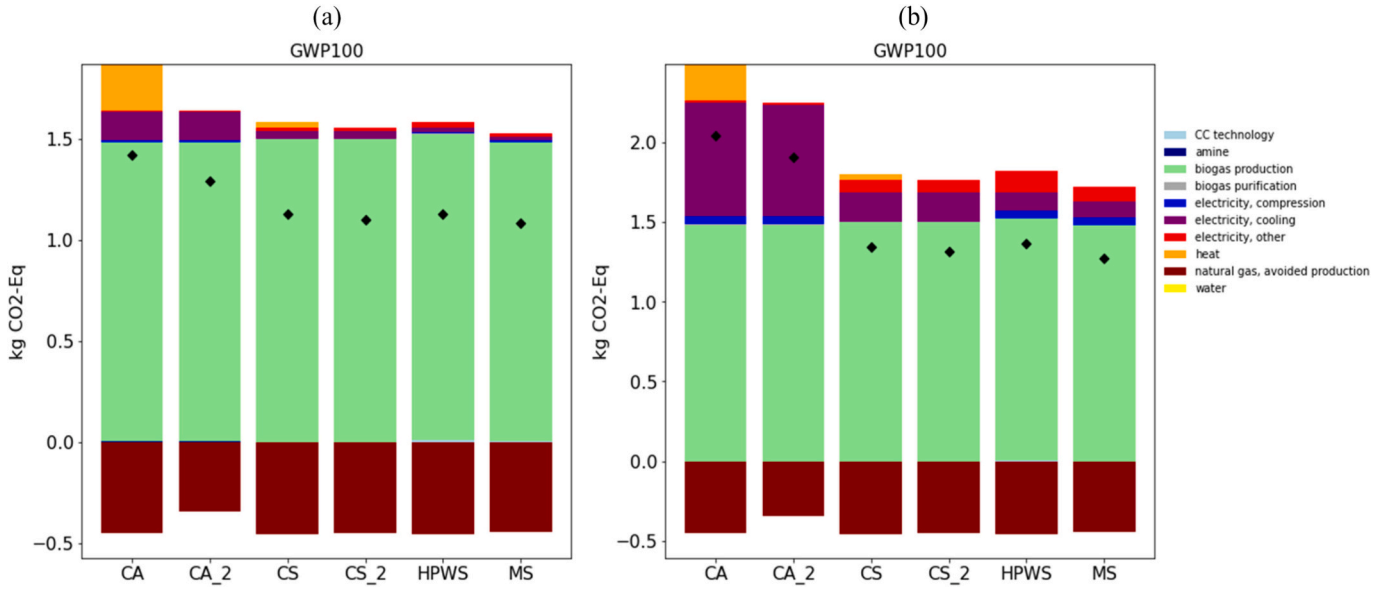

Fig. 11. Contribution analysis for the global warming (GWP100) impact. The black dots indicate the net value of the impact. Left: electricity mix France; Right: electricity mix Germany.

fraction in biogas stream determines an increase of 10 % of heat consumption in CA, 3.5 % increase in electricity consumption in MS and no influence on CS and HPWS.

Variations in biogas composition and flowrate in a biogas plant are common, depending on seasonal variation in the feed material (e.g. 3.5 % in CO_2 fraction, 27 % in flowrate, with respect to the nominal point, in case of medium scale example (*5_methagri32_suivi_methanisation_rapport.pdf*, 2023). The induced energy variations will affect the impact results especially when energy is the main contributor. Energy variation could be higher than the differences observed for the GWP100 results of MS, CS and HPWS, and in consequence leads to the conclusion that MS, CS and HPWS behave similarly with respect to this impact category.

A sensitivity study was performed with respect to the electricity mix, by replacing the French mix (0.08699 kg CO_2eq/kWh) with a more fossil based mix (electricity mix Germany, 0.39108 kg CO_2eq/kWh; market of electricity, low voltage [DE]). The ranking of the CC technologies remains the same (see LCA results in SI Table S6). However, more the electricity is consumed, more the impacts increase, and the global environmental performance of technologies decreases: for example, GWP100 impact of MS increases by 17 % when switching from the French mix to German mix. A higher CO_2 emitting electricity mix will better discriminate between MS, CS and HPWS global warming impact:

MS impact is 7 % lower than CS and 60 % lower than CA.

5. Conclusions

The objective of this study was to compare four carbon capture technologies for biogas separation with the aim to achieve high biomethane purity (at least 96 mol%) suitable for direct injection into the natural gas pipeline network and the highest possible carbon dioxide purity in liquid phase, for storage and future use in various applications. The technical performance in terms of purity and recovery of biomethane and CO_2 product, and energy consumption, were evaluated, as well as the environmental impacts, calculated with LCA method.

Simulations conducted for different input biogas compositions and flowrates provided coherent results compared with the available literature data or onsite collected data. The comparison between the three scales showed similar results in terms of purity and recovery for CH_4 and CO_2 with a slight diminution of the energy consumption as the scale increases. All investigated CC technologies can supply high purity products but at the expense of different energy consumptions: electricity from 0.2 kWh/Nm^3 biogas treated for MS to 0.8 kWh/Nm^3 for CA, heat 0.5 kWh/Nm^3 for CA and no heat for MS and HPWS. The results indicated that CA yields very high performance in terms of product purity (higher than 98 % for both products) but also the highest impact values for all impact categories, due to the high energy consumption. MS technology resulted in significantly lower energy consumption and thus lower environmental impacts. The two other CC technologies, CS and HPWS, demonstrated good technical performances, with slightly lower recoveries and purities with respect to CA, and higher energy demand in comparison to MS. The environmental impacts of MS, CS and HPWS are close and their discrimination depends on the type of energy used, i.e. a fossil-based energy induces higher differences in the impact values than a decarbonated/renewable one. However, if a non-fossil mix is considered, carbon capture technologies such as MS, CS and HPWS will be equivalent in terms of GWP, which will conduct to include additional criteria as other environmental impacts, equipment and energy costs, etc.

As the performance of the CC technology depends upon the input biogas stream and on the operating strategy, for example the energy production onsite, the rigorous choice of the LCA boundaries is crucial for a relevant comparison. The CC technologies cannot be evaluated per se, isolated from the upstream (or downstream) processes, but only integrated in the whole supply chain, and after the optimization of the design and operation parameters. The range of uncertainty of the LCA results depends on the range of input biogas fluctuations in terms of flowrate and composition, which is inherently due to seasonal variations, and thus is site dependent.

Future enhancements in these technologies through the incorporation of more suitable and innovative materials, such as mixed-matrix membranes or alternative solvents, have the potential to significantly reduce the energy demand in the operation and the performance of these processes. Another challenging future work will involve exploring the utilization of CO_2 in various applications.

CRediT authorship contribution statement

Mohamad Kanso: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Investigation, Formal analysis, Data curation. **Ligia Tiruta-Barna:** Writing – review & editing, Validation, Supervision, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Carlos Eduardo Robles-Rodriguez:** Writing – review & editing, Validation, Supervision, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

This work was supported by the French National Research Agency (ANR) under the "Investissements d'Avenir" program, reference ANR-18-EURE-0021.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.spc.2024.07.010.

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