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Scenarios for upgrading and distribution of compressed and liquefied biogas — Energy, environmental, and economic analysis

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ABSTRACT

In the transition towards fossil-free transports, there is an increasing interest in upgraded biogas, or biomethane, as a vehicle fuel. Liquefied biogas has more than twice as high energy density as compressed biogas, which opens up the opportunity for use in heavy transports and shipping and for more efficient distribution. There are several ways to produce and distribute compressed and liquefied biogas, but very few studies comparing them and providing an overview. This paper investigates the energy balance, environmental impact and economic aspects of different technologies for upgrading, liquefaction and distribution of biogas for use as a vehicle fuel. Furthermore, liquefaction is studied as a method for efficient long-distance distribution.

The results show that the differences between existing technologies for upgrading and liquefaction are small in a well-to-tank perspective, especially if the gas is transported over a long distance before use. Regarding distribution, liquefaction can pay back economically after 25–250 km compared to steel container trailers with compressed gas, and reduce the climate change impact after 10–30 km. Distribution in gas grid is better in all aspects, given that it is available and no addition of propane is required. Liquefaction can potentially expand the geographical boundaries of the market for biogas as a vehicle fuel, and cost reductions resulting from technology maturity allow cost-effective liquefaction even at small production capacities.

1. Introduction

Biogas production combines energy production and waste management in a way that can serve many different purposes and give benefits in several areas of society (Guenther-Lübbers et al., 2016; Hagman et al., 2018; Martin and Parsapour, 2012). It has been claimed that biogas can help meet as many as seven (Dada and Mbohwa, 2018), nine (World Biogas Association, 2017) or even all 17 (Hagman and Eklund, 2016) of the UN sustainable development goals (United Nations, 2015). In the European Union (EU), the production of biogas has increased steadily over the last few decades (EurObserv'ER, 2017). Although most of the biogas produced is used to generate heat and electricity (Persson and Baxter, 2014), there has also been an increase in biogas upgrading to natural gas quality to inject it in natural gas grids or use it as vehicle fuel (Hoyer et al., 2016; Pettersson and Wellinger, 2009).

The use of biogas in the transport sector is very limited, except in Sweden and in Switzerland (Persson and Baxter, 2014), but a growing interest has led to studies on increasing the use of biogas in vehicles in other countries. In Denmark, Cong et al. (2017) suggested an increased use of biogas in heavy transports to reduce CO_2 emissions, while Patterson et al. (2011) found that biogas as a vehicle fuel would have environmental benefits and could be financially competitive with other biofuels on the UK market. In the EU Renewable Energy Directive, the goal for year 2020 is that 10% of the energy used in transport should come from renewable sources (European Commission, 2009). Sweden has set the goal to reduce the greenhouse gas emissions from the transport sector by at least 70% from 2010 to 2030, and that the whole transport sector will be completely free from fossil greenhouse gas (GHG) emissions by 2045 (Swedish Energy Agency, 2017).

In order to use biogas as vehicle fuel, it has to be cleaned from CO_2 and other impurities to increase the methane content and thereby the heating value. A methane content of at least $95\%_{vol}$ is

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Nomenclature

Abbreviations

AF	annuity factor
AS	amine scrubbing
CBG	compressed biogas (biomethane)
EU	European Union
LBG	liquefied biogas (biomethane)
LHV	lower heating value
MR	mixed refrigerant
MS	membrane separation
OS	organic scrubbing
PEF	primary energy factor
PSA	pressure swing adsorption
WS	water scrubbing

Symbols

CH_4	methane
CO_2	carbon dioxide
H_2S	hydrogen sulfide
i	interest rate
N	depreciation period
p	pressure
W	compressor work
η	compressor efficiency

required, and the last Swedish standard requires a methane content of 97 ± 1%$_{vol}$ (Swedish Standards Institute, 2015). Technologies for upgrading of biogas to vehicle fuel quality have been described in several studies. The technical report by Bauer et al. (2013) is one of the most comprehensive works in the field, covering both the technical and the economic aspects of the conventional upgrading technologies. It is frequently cited in newer scientific articles; for example, Singhal et al. (2017) took most of their data on biogas upgrading technologies from this report. The reports by Bailón Allegue and Hinge (2012) and Pettersson and Wellinger (2009), as well as the article by Ryckebosch et al. (2011), also provide useful information on biogas upgrading technologies, although the references they use may not be entirely up to date with the development since they were published. In a more recent review, Miltner et al. (2017) refer back to many of these articles as well as other sources, but do not go into the energy demand of existing upgrading technologies.

By cooling natural gas or upgraded biogas below the boiling point of methane (−161.5 °C), the gas can be condensed to liquid form. The energy density of liquefied CH_4 is about 600 times higher than that of gaseous CH_4 at atmospheric pressure and 2.5 times higher than CH_4 at 250 bar (Benjaminsson and Nilsson, 2009). Due to the relatively high energy density, lower sulfur content and life cycle GHG emissions compared to diesel or heavy fuel oil (DMA, 2012; Edwards et al., 2014), liquid natural gas (LNG) or liquid biogas (LBG) have emerged as an alternative fuel for heavy road transports as well as sea transports. Several truck manufacturers have started producing engines that can run on methane (Daimler, 2018; Isuzu, 2018; Iveco, 2015; Scania, 2017; Volvo Lastvagnar, 2017). Brynolf et al. (2014) investigated the environmental aspects of using LNG, LBG or methanol as marine fuels, finding clear advantages for all of the alternative fuels compared to conventional fuel oil. Their results also showed that only fuels from renewable sources, LBG or bio-methane, could lead to significant reductions of climate change impact.

The higher energy density of liquefied gas compared to compressed gas can also be an advantage for long-range distribution in areas without an existing natural gas grid, which is the case in most areas in Sweden (Benjaminsson and Nilsson, 2009). Ahmadi Moghaddam et al. (2015) compared the energy efficiency and global warming potential of five different biogas-based fuels for city buses: compressed biogas (CBG), LBG, dimethyl ether, Fischer-Tropsch diesel and methanol. Taking a life cycle perspective, their study pointed at advantages of liquefying the upgraded gas or converting it to dimethyl ether or methanol for distribution over longer distances. Pettersson et al. (2006) calculated that it would be more cost-effective to condense the gas and distribute it on a 21 ton LBG trailer than to transport it in compressed form in steel containers for distances above 90 km, or even shorter if there is also a market for the liquid CO_2 produced. According to Benjaminsson and Nilsson (2009), distribution of LBG in a 25 ton trailer would only be economic compared to CBG in steel containers for distances above 200 km and annual amounts above 100 GWh, while for smaller amounts the specific costs for liquefaction would be too high. Börjesson et al. (2016) published a technical report on methane as a vehicle fuel, where they compared costs, GHG emissions and energy efficiency of CBG and LBG scenarios against compressed natural gas (CNG), LNG and diesel. A transport distance of 200 km was assumed for biogas upgraded through water scrubbing or amine scrubbing and 600 km for biogas produced through thermal gasification. Their results showed that bio-methane fuels could be cost-competitive against fossil fuels in a Swedish context, while also leading to significant reductions of GHG emissions.

The many existing technologies for upgrading, liquefaction and distribution of biogas present a large number of pathways from producer to user. Up to this point, very few studies have attempted to describe and compare these pathways from a technical and economic point of view; in fact, previous studies have either described the technologies without placing them in a context, or included only one or two upgrading and liquefaction technologies and a limited set of distribution options. The aim of this paper is to analyze the energy balance, environmental impact and economic viability of a wider range of scenarios for upgrading, liquefaction and distribution of biogas for use as vehicle fuel. In addition, liquefaction as a means for efficient long-distance distribution of biogas is assessed by investigating the break-even distances for primary energy, climate change impact and costs compared to distribution of compressed biogas. Through this approach, the study will contribute to the research on technical and economic conditions for different ways to produce and distribute upgraded biogas. The analysis is based on Swedish conditions for distribution, implying a limited availability of gas grids, although grid distribution is also included in the analysis for comparison.

The paper is organized into five sections. The Introduction is followed by a Methodology section, describing the modelling, the studied scenarios for production and distribution and the data collection. In the third section, results from the calculations are presented in the form of energy balance, environmental impact and cost analysis. Moreover, there is an analysis in this section on when liquefaction is advantageous compared to compression. The results and methods are then elaborated in the Discussion section, and the main findings and contributions are summarized in the Conclusion section.

2. Methodology

In this paper, technologies for upgrading, liquefaction and distribution of biogas are described and analyzed in form of scenarios.

Each scenario represents a possible pathway for the biogas from producer to user, including all the intermediary processes to get there, as illustrated in Fig. 1. Biogas production through anaerobic digestion was assumed to be a common starting point for all scenarios, and was therefore not included in the analysis. The scenarios were categorized according to the end product, which could be either compressed biogas (CBG) or liquid biogas (LBG) of vehicle fuel quality. The analysis consisted in two parts:

1. Comparison between different technologies to achieve the same product (either CBG or LBG)
2. Break-even analysis to find the distribution distance at which it is more efficient to transport biogas in liquid than in compressed form.

Both of these parts included three different perspectives: energy use, environmental impact and life cycle costs. The data used in modelling and calculations were collected through a literature review, with industrial contacts assisting in filling in some gaps. The data that ended up being used was selected based on its timeliness and proximity to an original source of information, such as an industry. Furthermore, previously published data was used as far as possible, in order to have an existing report or article for the reader to refer back to. Hence, priority was given to relatively new reports containing first-hand information.

2.1. Modelling

The energy balances were calculated both in terms of final energy use, divided among different energy carriers (electricity, heat and diesel), and primary energy use. The primary energy factors (PEF) used are applicable to Sweden, with electricity reflecting the Nordic electricity generation mix, and steam was considered to be generated by a wood-chip boiler. The energy balances included energy use for upgrading, liquefaction, compression, distribution and fueling.

Life cycle assessments (LCA) of the different scenarios were conducted in accordance with ISO 14040 and 14044, following the guidelines for an attributional LCA (ISO, 2006a, 2006b). Modelling and calculations were done with SimaPro 8 (Goedkoop et al., 2016), using the Ecoinvent 3 database (Weidema et al., 2013) and the ReCiPe heuristic midpoint method for impact assessment (Goedkoop et al., 2013; PRé, 2014), which is commonly used in scientific LCA studies. To make the results more manageable and presentable alongside the energy and economic analyses, the figures only include climate change impact. Results for terrestrial acidification, freshwater eutrophication, photochemical oxidation and ozone depletion are given in Table A1 in the Appendix.

Uncertainty analyses of the environmental impact were performed using Monte Carlo analysis and a lognormal distribution, where the value of a parameter lies between the mean value divided by the variance (σ^2) and the mean value multiplied by σ^2 (Weidema et al., 2013). In most cases, data for energy, water and chemical demands were considered fairly reliable and representative and σ^2 was set to 1.1. For cryogenic technology, data was deemed more uncertain and σ^2 was set to 1.2. For methane slip and CBG distribution by truck, σ^2 was set to 1.2 and 1.5, respectively, to account for the methane slip range presented in literature and for the range of number of containers carried.

Life cycle costs (LCC) were calculated considering investment, operation and maintenance costs. To account for capital costs and equipment lifetime, the specific costs for the different scenarios were calculated on a yearly basis using the annuity factor (Equation (1)), with a depreciation period (N) of 15 years and a 6% interest rate

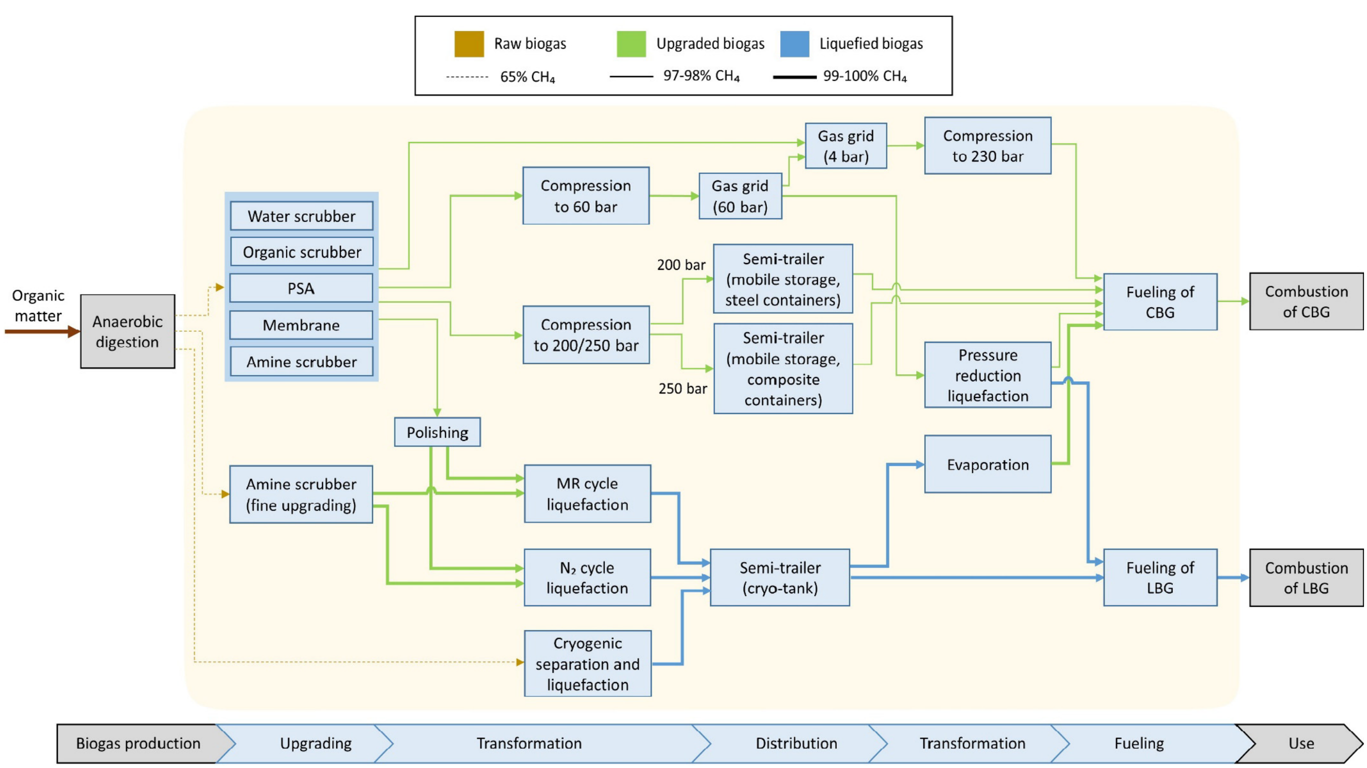

Fig. 1. Schematic view of the modelled scenarios for upgrading, liquefaction and distribution of biogas. Going from the left to the right, each of the indicated pathways represent a possible scenario from biogas production to end user. The color and weight of the lines indicate the state and methane content of the gas.

(i), in line with similar economic studies (Börjesson et al., 2016; Larsson et al., 2015). In addition to energy costs, operation and maintenance costs were considered to be 2.5% of the investment costs, which is the norm for industrial technology investment, in which values usually vary between 2% and 2.5% (Larsson et al., 2015; Pettersson et al., 2006).

$$AF = \frac{i}{1-(1+i)^{-N}} \tag{1}$$

The specific transport cost, which was set to 1.80 €/km, includes the costs for diesel, driver, truck and other related costs. Time for loading and unloading of trucks was set to 1 h in the CBG scenarios, and 4 h for LBG, with a cost of 64.50 €/hour. These costs were taken from Pettersson et al. (2006) and updated for inflation. Distribution costs for road transport included storage capacity for CBG in containers equivalent to three times the daily production of upgraded biogas (Börjesson et al., 2016). Costs for fueling included only the electricity cost required for fueling, and excluded investment and other operation costs for fueling stations, as these costs depend on the capacity of individual stations. The costs for distribution by high- and low-pressure grids exclude investment, operation and maintenance costs of these grids, i.e. the costs consist only of energy costs for the operation of the gas grids.

2.2. Studied scenarios

The analysis covered the way from raw biogas from anaerobic digestion, with a CH_4 content of 65%, until vehicle fueling (Fig. 1). The upgrading technologies considered included water-, organic- or amine scrubbing, pressure swing absorption or membrane separation, thus covering the most common technologies used in Europe (Hoyer et al., 2016). Distribution of upgraded gas was considered by low-pressure (4 bar) or high-pressure (60 bar) gas grid or by truck in steel or composite containers with one, two or three trailers per truck. The compression pressure for transportation by truck was considered to be 200 bar for steel containers and 250 bar to composite containers. Liquefaction was considered through mixed refrigerant (MR) cycle, nitrogen (N_2) cycle, pressure reduction from high-pressure grid or cryogenic liquefaction of raw biogas, and LBG was assumed to be distributed by truck in an 18–30 ton cryogenic tank.

As there is a large number of possible scenarios, results for only a few of them are included in the figures in section 3, and a more comprehensive summary is given in Table A1 in the Appendix. The CBG scenarios shown in section 3 include upgrading through water scrubbing (WS), organic scrubbing (OS), pressure swing adsorption (PSA), membrane separation (MS) and amine scrubbing (AS). For AS, either district heat (DH) or steam is used as heat source. Distribution scenarios for CBG include truck transport with two steel containers for all upgrading technologies over a distance of 100 km. For WS, which is the most common upgrading technology (Hoyer et al., 2016), distribution scenarios also include composite containers, high- and low-pressure gas grid. For the LBG scenarios, upgrading was considered by WS or AS with DH, followed by liquefaction with MR cycle, N_2 cycle, or pressure reduction, or direct upgrading and liquefaction with cryogenic technology. The LBG is distributed over the same distance (100 km) in a 25-ton cryogenic tank, or, in case of pressure reduction liquefaction, in gaseous form via a high-pressure grid to the place where the LBG is used. Energy use and climate change impact are presented as a total and per process of each scenario, while the costs, which are more size-dependent, are shown as a function of yearly production capacity.

An extra CO_2 polishing step before MR- or N_2 cycle liquefaction was considered for gas upgraded through WS, OS, PSA or MS, whereas AS is able to comply with the CO_2 requirements for liquefaction at the cost of a higher energy use (Bauer et al., 2013; Karlsson, 2018).

Removal of hydrogen sulfide (H_2S) and other trace gases was not included in the analysis, as early calculations indicated that the importance of these processes would be very small in relation to the energy for upgrading. Also, they were considered to be equal for all scenarios. Although water scrubbers are less sensitive to impurities than other technologies and are able to separate H_2S from methane, the H_2S has to be removed at some point to avoid emitting the toxic gas to the atmosphere (Abatzoglou and Boivin, 2009; Bauer et al., 2013).

In the break-even analysis (section 3.4), upgrading was considered through WS and liquefaction through MR cycle, as MR cycle was found to be the most competitive liquefaction technology and WS one of the best upgrading technologies. WS is also representative as it is the most common technology for biogas upgrading worldwide (Fagerström and Murphy, 2018). Distribution was considered to take place either in the form of CBG in steel or composite containers, in the form of LBG in cryogenic tanks, or in a high- or low-pressure gas grid. The distribution by truck considered one or three CBG trailers per truck, or a cryogenic tank with a capacity of 18 or 30 ton of LBG. Low-pressure gas grid was considered an option for local distribution only, for distances up to 100 km. The economic analysis was based on a production capacity of 30 or 120 GWh/year.

2.3. Data collection

The data used in modelling and calculations (Table 1) were collected through a literature review, with industrial contacts assisting in filling in some gaps. Data for upgrading technologies were mainly taken from Bauer et al. (2013), which in turn is largely based on contacts within the biogas industry and has been shown to have good agreement with actual figures from companies (Hoyer et al., 2016). The data shown regarding energy use, material use and emissions represent the average values used in the calculations. The investment costs are very much size dependent; the lower figures in the respective range are valid for large scale plants and the higher figures for small scale plants.

Energy use for compression was calculated according to the ideal gas law, using equation (2):

$$W = p_1 v_1 ln\frac{p_2}{p_1}\eta \tag{2}$$

where W is the compressor work (J/mol), p_1 is the initial pressure, p_2 is the pressure after compression, v_1 is the initial specific volume of the gas and η is the compressor efficiency, which was set to 50%.

Methane contents in off-gas were considered to be treated through catalytic oxidation in scenarios involving water/organic physical scrubber, MS and PSA, while the low methane slip of the AS makes such treatment redundant (Bauer et al., 2013). The use of catalytic oxidation was assumed to increase the electricity demand by 0.05 $kWh/Nm^3_{raw\ biogas}$ (Bauer et al., 2013), being able to convert 95% of the methane in the off-gas to CO_2 and water (Herbst et al., 2010). Foam formation in WS was assumed to be

Table 1
Data used in modelling of energy balance, environmental impact and life cycle costs of pathways for compressed and liquefied biogas. Data from: [1](Bauer et al., 2013); [2](Karlsson, 2018); [3](Tybirk et al., 2018); [4](Olgemar and Partoft, 2017); [5](Pettersson et al., 2006); [6]Estimated from (Berg and Clodic, 2017); [7](Benjaminsson and Nilsson, 2009); [8](Heisch, 2012).

Process	Electricity	Heat	Water	Chemicals	Purity	Methane slip	Investment cost
	kWh/Nm³	kWh/Nm³	m³/Nm³	kg/Nm³	%	%	€/(Nm³/h)
Water scrubber	0.36[1]		0.00034[1]	5E-05[1]	98%[1]	1%[1]	1490–5 820[1]
Organic scrubber	0.33[1]			5E-05[1]	98%[1]	1%[1]	1420–4 720[1]
Amine scrubber							1630–3 300[1]
Steam, vehicle fuel quality	0.17[2]	0.17[2]	4.6E-05[1]	5E-05[1]	99.8%[1,2]	0.06%[1,2]	
District heat, vehicle fuel quality	0.21[2]	0.17[2]	4.6E-05[1]	5E-05[1]	99.8%[1,2]	0.06%[1,2]	
Steam, liquefaction quality	0.28[2]	0.17[2]	4.6E-05[1]	5E-05[1]	99.995%[1,2]	0.06%[1,2]	
District heat, liquefaction quality	0.34[2]	0.17[2]	4.6E-05[1]	5E-05[1]	99.995%[1,2]	0.06%[1,2]	
Pressure swing adsorption	0.38[1]				98%[1]	1.8%[1]	1490–2 930[1]
Membrane separation	0.35[1]				98%[1]	0.5%[1]	1970–6 460[1]
Catalytic oxidation of off-gas	0.08[1]						
Polishing for liquefaction	0.14[2]				99.995%[2]		1400–2600[2]
Mixed-refrigerant cycle	0.57[2]				99.995%[2]	0%[3]	4650–21000[4]
N_2 cycle	0.72[2]				99.995%[2]	0%[3]	8025[1]
Cryogenic liquefaction	1.08[3,5]				99.9%[4]	0%[3]	6400–26700[6]
Pressure reduction liquefaction	0.01[5]						
Fueling, CBG							
From grid	0.3[7]						
From container	0.07[7]						
From LBG	0.038[8]						
Fueling, LBG	0.0032[8]						

avoided by addition of rapeseed oil (Kougias et al., 2015). The heat demand used in calculations for AS was the net heat demand, assuming that 80% of the heat used in the stripper can be reused at a lower temperature in other parts of the biogas process (Bauer et al., 2013).

Distribution by truck was considered either as CBG in steel or composite containers with 1–3 trailers per truck, or as LBG in a tank carrying 18–30 ton (Table 2). With CBG trailers it was assumed that 85% of the gas volume can be evacuated from the containers (Karlsson, 2018), while the cryogenic tank was assumed to be emptied to 100% at the fueling station. For gas grid distribution, the upgraded gas was assumed to be either injected directly into a local low-pressure grid, or compressed and injected into a regional high-pressure grid with an addition of 0.24 kg propane per 1 kg biogas, which corresponds to a volumetric ratio of 92% biogas and 8% propane (Benjaminsson and Nilsson, 2009). For scenarios including pressure reduction liquefaction, it was assumed that 10% of the gas is liquefied and the remaining 90% is fueled as CBG (He and Ju, 2013; Tan et al., 2016).

The electricity price (Table 3) was the average price in 2017 in Sweden for non-household consumers, without taxes, and the heat price was assumed at 0.05 €/kWh for both district heat and steam. All costs were converted to Euros using the average exchange rate in 2017 of 0.104 SEK/€ (European Central Bank, 2017) and corrected for inflation. The cost for diesel used for distribution was not calculated separately, but was included in the total transport costs. The primary energy factors used in energy calculations were representative for Nordic electricity mix, Swedish district heating, steam from a wood chip boiler and fossil diesel.

3. Results and analysis

3.1. Energy balance

Fig. 2 shows the final and primary energy use for different biogas upgrading scenarios, divided into CBG and LBG as final product. In the CBG scenarios, the final energy use of the different upgrading technologies does not vary considerably, but amine scrubbing (AS) shows a lower primary energy use than the other technologies, due to the lower primary energy factor (PEF) of heat compared to electricity. AS with district heating as heat source has the lowest primary energy use, followed by AS with steam. Comparing these two cases, the much lower PEF of district heating is somewhat compensated by a higher electricity demand for upgrading.

Regarding distribution of CBG, distribution in steel containers has the highest final energy use, mainly because the amount of biogas transported is much lower in comparison to composite containers, which leads to higher fuel consumption per MJ of distributed biogas. Distribution in low-pressure grid results in the lowest energy use, both for final and primary energy, but is restricted to shorter distances, typically below 100 km. Distribution in high-pressure grid has a higher energy use due to the compression to 60 bar, followed by a pressure reduction necessary to allow injection to the low-pressure grid, and a recompression for fueling at 200 bar.

In the LBG cases, upgrading demands more electricity because of the polishing required for liquefaction, while the heat demand remains unchanged. AS results in lower energy use than water scrubbing (WS). Liquefaction with mixed-refrigerant (MR) has lower energy use than nitrogen (N_2) liquefaction, because of the better fit between the cooling curve of the refrigerant and methane. Cryogenic upgrading and liquefaction results in intermediary energy use when compared to conventional upgrading and liquefaction options. In these LBG cases, it is worth noting that the energy required for distribution and fueling is much lower than in the CBG cases, which indicates that for longer distances, the distribution of LBG becomes more energy efficient. Liquefaction through pressure reduction from a high-pressure grid is the most energy efficient option, although only a small fraction (about 10%) of the gas can be liquefied that way.

Table 2
Loading capacity, net distribution capacity and fuel use for distribution by truck. Data from: [1](Börjesson et al., 2016); [2](Pettersson et al., 2006); [3](Benjaminsson and Nilsson, 2009); [4](Karlsson, 2018).

	Investment cost	Loading capacity	Trailers/truck	Net distribution capacity				Fuel use
	€/trailer	Nm^3/trailer		%	Nm^3/truck	ton/truck	MJ/truck	kWh/km
CBG, steel	95230[1]	2000[3]	1–3[3]	85%[4]	1700–5100	1.2–3.6	59180–177540	4.61[1]
CBG, comp.	222200[1]	5250[4]	1–3[4]	85%[4]	4463–13388	3.2–9.5	155350–466050	4.61[1]
LBG, tank	320000[2]	25000–42000[1,3]	1[3]	100%[1]	25000–42000	18–30	903830–1506 380	3.92[1]

Table 3
Primary energy factors and energy prices for the energy carriers included in the studied scenarios. Data from: [1](Swedish Energy Agency, 2006); [2](Gode et al., 2011); [3](Uppenberg et al., 2001); [4](Eurostat, 2017).

Energy carrier	Primary energy factor	Energy price
	kWh_{PE}/kWh_{FE}	€/kWh
Electricity	1.6[1]	0.065[4]
District heat	0.79[2]	0.05
Steam	1.31[2]	0.05
Diesel	1.06[3]	

3.2. Environmental impact assessment

Fig. 3 shows the climate change impact of different scenarios. The error bars in the figure indicate the 95% confidence interval for each scenario. Comparing the different upgrading technologies, the climate change impact is quite similar for WS, OS and MS. PSA has the highest impact due to the higher methane slip, even though most of the methane is assumed to be oxidized to CO_2. AS with district heating has the lowest impact, while the climate change impact of AS with steam is in the same range as that of WS, OS and MS. The use of water and chemicals in WS, OS and AS is not high enough to make an impact compared to the energy use. The total uncertainties of the CBG scenarios with truck distribution are 16%–40%, most of which is attributed to the transport phase where the uncertainty reflects the use of one or three trailers per truck instead of two. The higher capacity of composite containers results in a lower impact per distributed MJ of CBG. Low-pressure grid also has a lower impact, while distribution via high-pressure grid has a much higher impact due to the use of propane additive.

For LBG, the addition of propane for grid distribution is also the reason behind the larger environmental impact of the scenarios including pressure reduction liquefaction. For other scenarios, however, distribution is a much smaller part of the total impact than in the CBG scenarios, and the total climate change impact is lower for those LBG scenarios than for the CBG scenarios at a distribution distance of 100 km. MR liquefaction has a slightly lower climate change impact compared to N_2. Scenarios including AS have a lower impact than scenarios including WS, as the AS requires less extra electricity for producing biogas with very low CO_2 content. Cryogenic technology has a higher impact than MR, but lower than N_2, with upgrading through WS. The calculated uncertainties are within 6%–14% for all scenarios.

3.3. Economic analysis

Fig. 4 shows the life cycle costs for different scenarios as a function of yearly production capacity. All scenarios are subject to a higher specific cost for small production capacities, and decreasing specific costs as the production capacity increases. This trend is particularly clear for the liquefaction technologies, where the specific cost for a scenario including MR liquefaction is 2.7 times higher for 10 GWh/year than for 120 GWh/year. Thus, liquefaction becomes more interesting from an economic point of view for large-scale facilities and longer distances. At a distribution distance of 100 km, liquefaction becomes cost competitive against distribution with steel containers for a yearly production of 40–100 GWh, depending on the liquefaction technology and truck capacity, and liquefaction is only cost competitive against distribution in composite containers with AS upgrading for production capacities over 115 GWh/year.

3.4. Break-even for distribution distance

This section presents eight different distribution scenarios comparing primary energy use, climate change impact and life cycle cost as a function of the distance between the biogas production facility and the filling station. All scenarios include water scrubber upgrading, and the LBG scenarios include MR liquefaction, implementing the primary energy factors and transport costs presented in section 2.

3.4.1. Break-even analysis for primary energy use

The break-even transport distance when comparing distribution of CBG and LBG by truck varies substantially, depending on the number of trailers per truck (1–3), the use of steel or composite containers and the capacity of the LBG tank (Fig. 5). The primary energy factor for electricity is much higher than the primary energy factor for diesel, making the upgrading and liquefaction more important than the distribution from a perspective of primary energy use. Distribution in low- and high pressure grids have the lowest primary energy use for distances below 100 km and above 290 km, respectively. The break-even distance for CBG distribution in steel containers is in the range of 130–450 km, and for composite containers at 350–1750 km.

3.4.2. Break-even analysis for climate change impact

Distribution in liquid form reduces the climate change impact (Fig. 6) compared to distribution of compressed gas in steel containers already at 10–20 km transport distance, depending on the number of trailers the CBG truck is carrying. For N_2 cycle and cryogenic liquefaction (not shown in the figure), the break-even distance is 20–30 km and 15–20 km, respectively. If the compressed gas is transported in composite containers, the climate change impact break-even distances are 40–70 km with MR, 65–120 km with N_2 cycle and 40–80 km with cryogenic liquefaction.

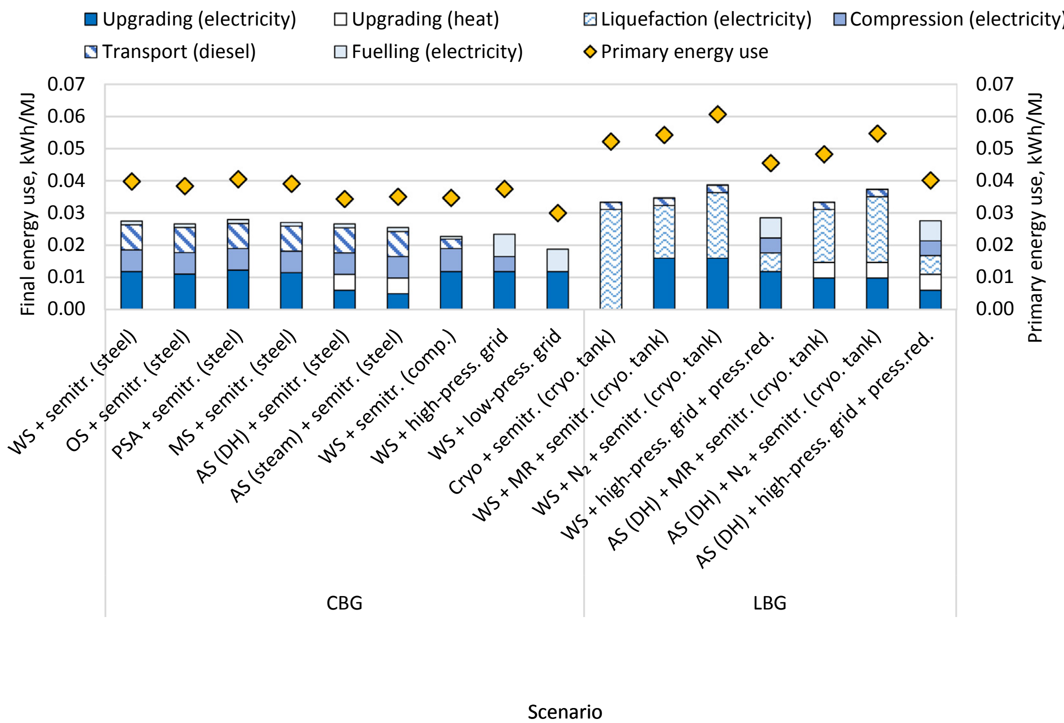

Fig. 2. Final energy use (bars) and primary energy use (diamonds) of scenarios for production and distribution of compressed (CBG) and liquefied biogas (LBG).

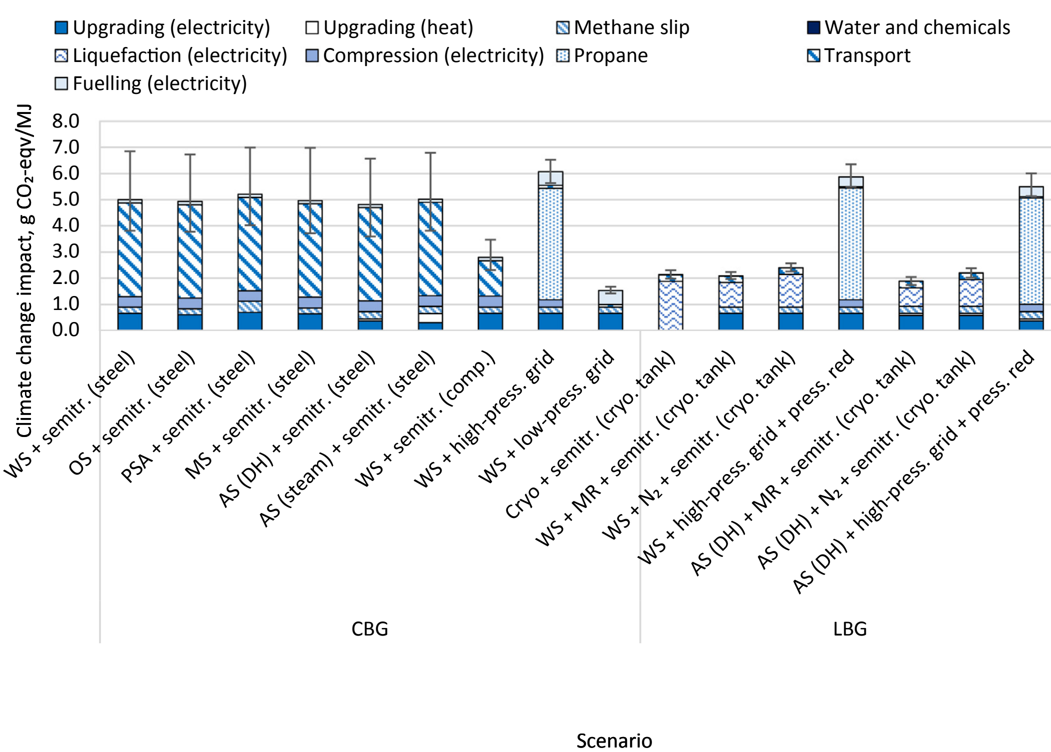

Fig. 3. Climate change impact of scenarios for production and distribution of compressed and liquefied biogas. "Transport" includes distribution over a distance of 100 km. The error bars indicate the 95% confidence interval for environmental impact.

Distribution in a low-pressure gas grid is the alternative with the lowest climate change impact. The high-pressure gas grid has a higher impact, mainly due to the addition of propane, but reaches break-even with CBG distribution in steel containers at 105–140 km and with distribution in composite containers at 290–390 km.

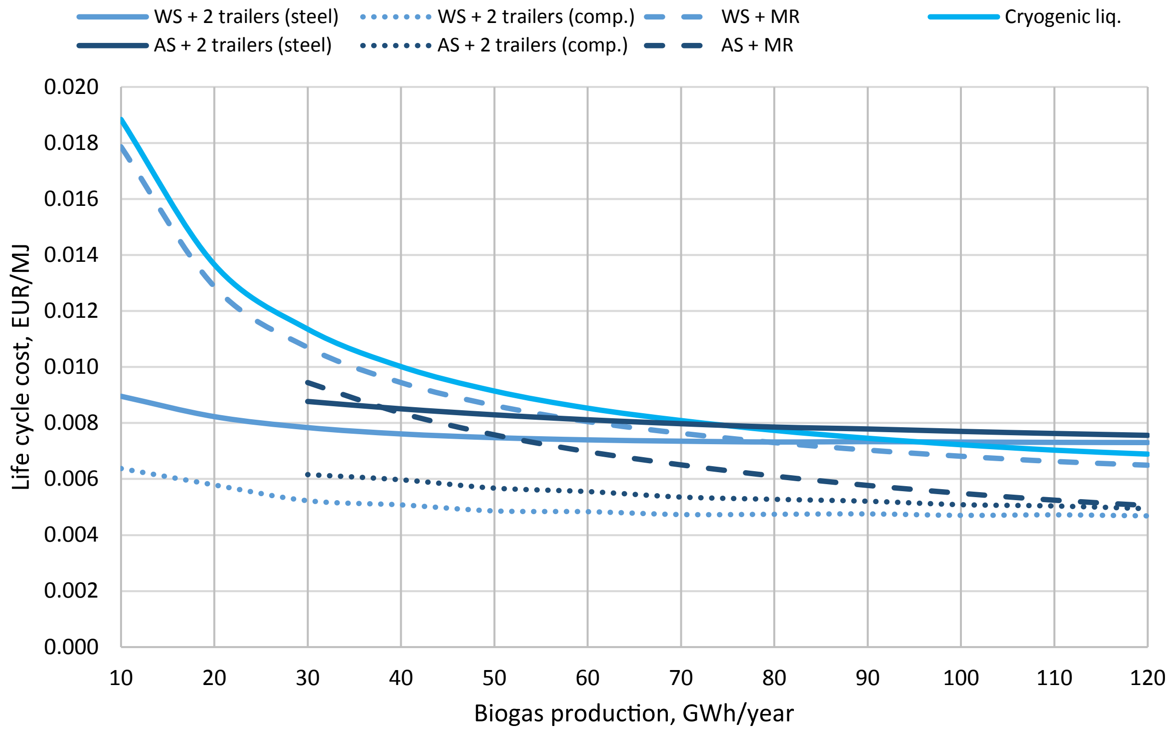

Fig. 4. Life cycle cost of scenarios for upgrading, liquefaction and distribution of biogas as a function of annual production capacity, for a distribution distance of 100 km.

3.4.3. Break-even analysis for life cycle costs

The life cycle costs for a production capacity of 120 GWh/year (Fig. 7) present break-even points that are intermediary to the two cases previously analyzed. Comparing the distribution scenarios for CBG and LBG by trucks, the higher investment costs for equipment and operational costs due to higher energy use of biogas liquefaction is compensated by more efficient transportation. The LBG scenarios with MR liquefaction are cost-effective at distances above 25–120 km for steel containers and at 110–460 km for composite containers. Low and high-pressure gas grids are potentially a cost-effective way of distributing biogas, but the results presented for these scenarios do not include investment and operational costs other than energy use. Looking at the other liquefaction alternatives, if N_2 liquefaction is considered, the break-even distances are at 40–160 km and 160–530 km, for steel containers and composite containers, respectively. Cryogenic liquefaction becomes economically beneficial compared to CBG distribution above 30–120 km for steel containers and 130–510 km for composite containers. For scenarios with a production capacity of 30 GWh/year (Fig. 8), MR liquefaction is cost-effective at distances above 80–250 km for steel containers and at 290–1100 km for composite containers.

4. Discussion

It is evident from the results of the environmental analysis that keeping the methane losses down is of paramount importance to minimize the climate change impact. Even though the methane slip from the upgrading is assumed to be reduced by 95% through catalytic oxidation, the climate change impact of the methane slip is around 35%–60% of the impact of the electricity for upgrading. For distribution systems, the methane losses are usually described as very low or negligible (Benjaminsson and Nilsson, 2009), and in this study such losses were not included. As noted also by Paolini et al. (2018), a high methane slip can counteract the environmental benefits of biogas production.

With transportation in steel containers, the number of trailers per truck can in reality be limited to two, due to regulations on maximum carriage weight (Benjaminsson and Nilsson, 2009; European Commission, 2015; Transportstyrelsen, 2015). There could also be other factors limiting the number of trailers or the size of the cryogenic tank used for biogas distribution, such as investment cost for the trailers or the tank, production capacity, demand and storage capacity at the fueling station and the number of stations served by each transport, but this is something that might require further investigation. In case the carriage weight is the limiting factor, it would be possible to combine trailers with steel and composite containers on the same truck (Benjaminsson and Nilsson, 2009). The costs, energy use and environmental impact would then end up somewhere in between those of steel containers and composite containers.

As shown in this study, liquefaction can have great economic and environmental advantages for long-range distribution, with benefits already at relatively short distances. In the absence of a national gas grid, as in Sweden, liquefaction can really extend the viable distribution range for upgraded biogas. Distribution by grid or pipeline would otherwise be preferable, given that the use of propane additive can be avoided. It has been shown that a certain share of the natural gas in the grid could be exchanged with biogas without any negative effects for the customers (Kristensson et al., 2007), and Börjesson et al. (2016) chose to

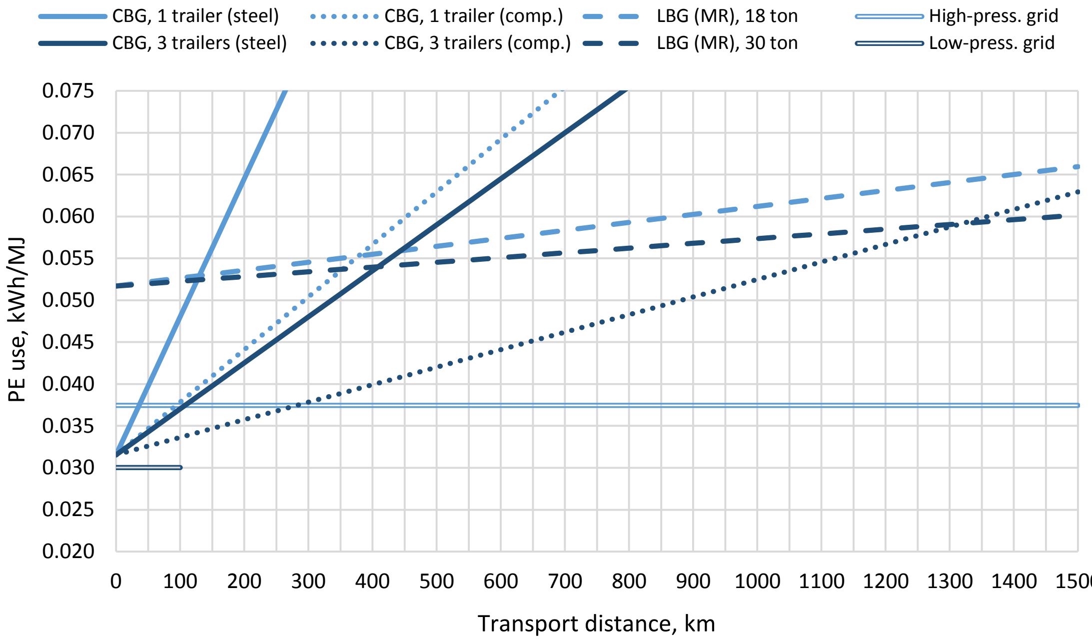

Fig. 5. Primary energy use for CBG pathways as a function of transport distance from production site to customer area.

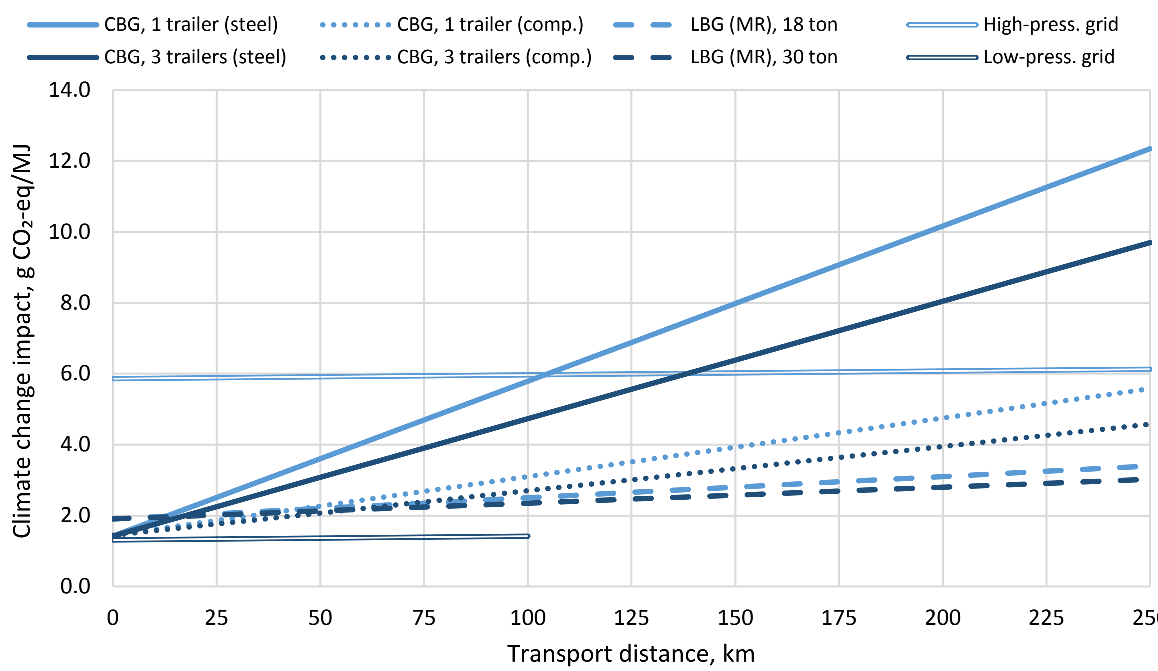

Fig. 6. Climate change impact for CBG pathways as a function of transport distance from production site to customer area.

not include propane additive in their study. Ideally, from an environmental point of view, biogas would be the standard for gas grids rather than natural gas. Benjaminsson and Nilsson (2009) proposed to reduce the heating value of natural gas injected to the grid by adding oxygen. Investment costs for gas grids, which were not included in this study, could be a barrier against the expansion of this infrastructure. However, considering the benefits in form of reduced energy use, environmental impact and costs during operation, it should be an alternative worthy of further investigation. Road transports were in this study assumed to be performed with conventional diesel trucks. With new engine technology, the trucks used for distribution of CBG and LBG could run on methane instead of diesel, thus decreasing the environmental impact of the transport phase. Changing diesel for biomethane would also affect the costs for distribution and thereby the point of breakeven for CBG and LBG.

In cryogenic separation and liquefaction, recovery of liquid CO_2 would be a possibility to reduce the environmental impact and improve the economic balance. With some additional energy for

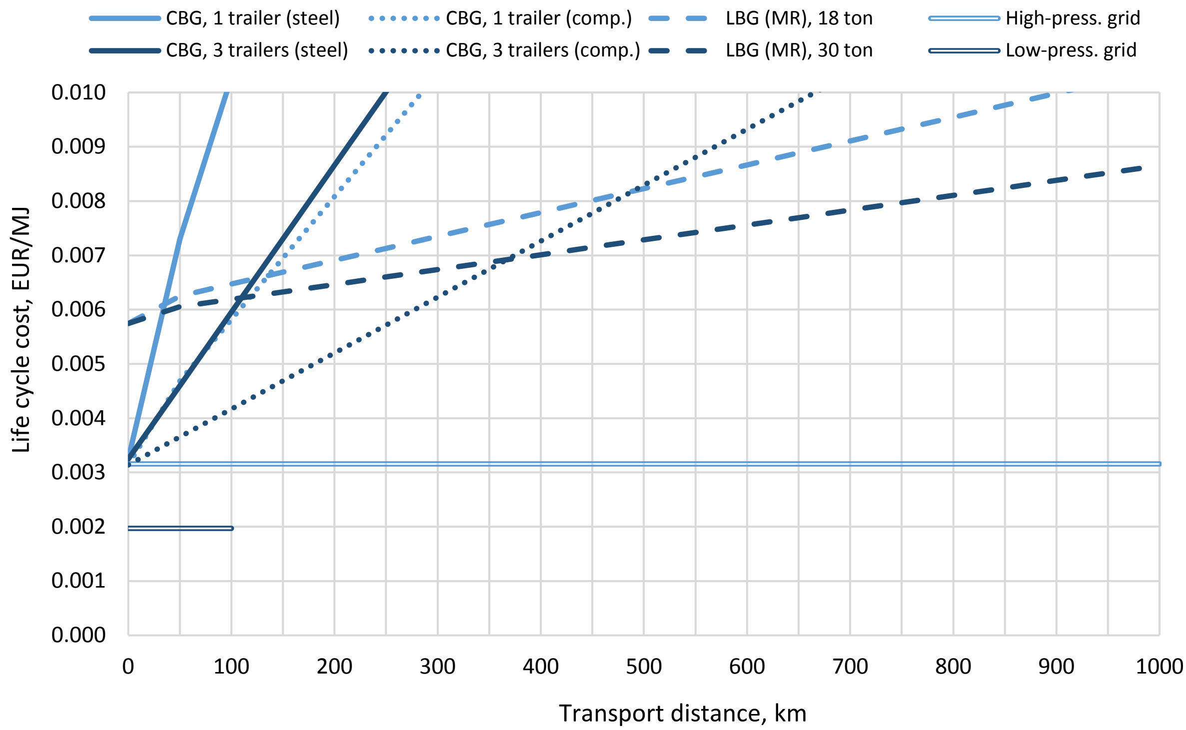

Fig. 7. Life cycle cost for CBG pathways as a function of transport distance from production site (120 GWh/a) to customer area.

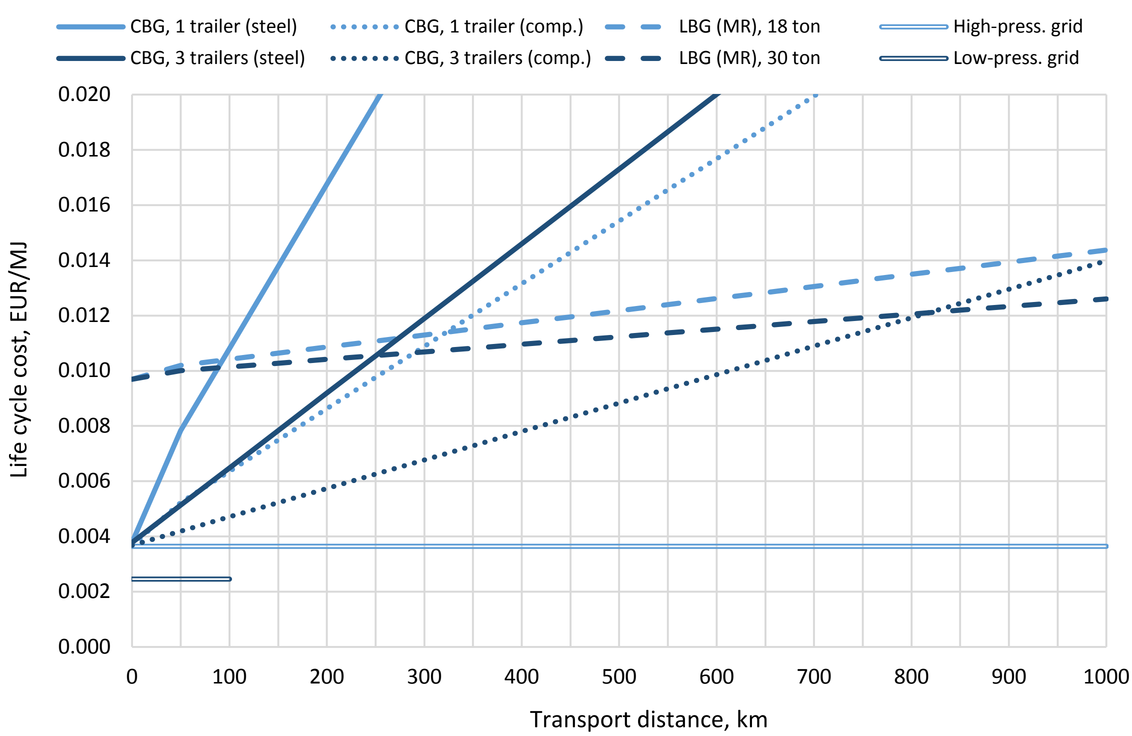

Fig. 8. Life cycle cost for CBG pathways as a function of transport distance from production site (30 GWh/a) to customer area.

cooling, liquid CO_2 could also be produced in upgrading plants utilizing amine scrubbing, membrane separation or PSA. Pure CO_2 can be used to enhance the yield in green houses, and is widely used in carbonated drinks. In its solid form, also known as dry ice, CO_2 can replace diesel-driven mechanical refrigerators as a refrigerant in cold transports (Hoyer et al., 2016; Pettersson et al., 2007). The possibilities with CO_2 as a useful by-product from biogas processes could therefore be a relevant subject of future studies.

Since LBG needs to be kept at a very low temperature to remain in its liquid form, storage time can be an issue. If LBG is left unused in a cryogenic tank, it will eventually be heated up by the higher surrounding temperature and start to evaporate. In order to avoid overpressure without emitting any boil-off methane, it can either be re-liquefied (Tybirk et al., 2018) or used as a fuel in its gaseous phase. The so-called LCNG fueling stations can deal with this by supplying both LBG and CBG from the storage tanks (Pettersson et al., 2006). LCNG fueling stations are also ideal if combined with pressure reduction liquefaction from a high-pressure grid, as part of the energy of the transported gas is used to liquefy a fraction of the biogas, supplying both LBG and CBG at the fueling station. However, this would require a certain balance of LBG and CBG demands at the site.

Economic break-even results found in this study for distribution of CBG in steel containers or LBG in tanks, at distances of 25–160 km for 120 GWh/year, are similar or a bit lower compared to previous studies. Pettersson et al. (2006) found the economic break-even for liquefaction to be around 90 km, while Benjaminsson and Nilsson (2009) suggested that liquefaction would only economically advantageous for distances greater than 150–200 km and production capacities above 100 GWh/year. These differences are partially justified by different assumptions on truck transportation capacities and energy and fuel costs, but also because investment costs and operational costs related to energy use are now lower as liquefaction technologies mature and establish themselves on the market. One important finding in this study is that liquefaction can be cost-effective even at smaller production sizes, being advantageous at capacities as low as 30 GWh/year for distribution distances over 80 km. Thus, liquefaction could in many cases be seen as a realistic way to enable distribution of biogas to CBG customers further away from the production site.

Similar to investment costs, the specific energy use for a process is typically lower for larger facilities. In the case of biogas upgrading and liquefaction, this type of scale dependence is not clearly described in the literature. Nevertheless, it is likely that such a relationship exists, which would imply lower specific energy use, environmental impact and operational costs for larger upgrading and liquefaction plants than for smaller ones.

The results presented in this paper in terms of primary energy use, environmental impact and costs of biogas upgrading and liquefaction technologies are valid for the specific set of boundary conditions and assumptions. Variations depending on the electricity system, the methane content of the raw biogas and the transport costs would be an interesting topic for future studies. It would also be relevant to compare scenarios for biogas production with fossil natural gas or other competing fuels.

5. Conclusions

In this paper, the energy balance, environmental impact and economic viability of different scenarios for upgrading, liquefaction and distribution of biogas for use as vehicle fuel were investigated, and liquefaction was assessed as a means for efficient long-distance distribution of biogas. The main findings of this research were:

- The differences between different technologies for upgrading and liquefaction are quite small in a life cycle, or well-to-tank, perspective, especially if the gas is transported over a long distance before it is used.
- Amine scrubbing differs from other upgrading technologies through its lower electricity demand and higher methane purity and could, depending on the heat source, have a lower primary energy use and environmental impact than other technologies.
- When it comes to liquefaction, mixed-refrigerant appears to be the most efficient technology currently on the market.
- Liquefaction can be a good option for distributing biogas over longer distances.

Although liquefaction of biogas more than doubles the energy use compared to just upgrading it, it can still be worthwhile if the distance to the customer is long enough. Compared to CBG in steel containers, distribution of LBG has a lower climate change impact at distances above 10–30 km and can be a cost-effective option above 25–250 km, depending on the size of the carriage, the method for liquefaction and the production capacity. Investment cost reduction and energy use improvements allow cost-effective liquefaction at smaller production capacities than was previously possible. Thus, liquefied biogas may not only open up for use of biogas in heavy transports, but also expand the geographical area for the CBG market.

The emerging technology for cryogenic separation and liquefaction has the potential to compete, but is less common and proven. Pressure reduction liquefaction requires less energy than other liquefaction technologies, but the fraction of LBG obtained is very low as most of the gas remains in its gaseous form.

Building upon the results of this paper, future studies could include assessing the impact of using CBG or LBG in the distribution trucks instead of diesel, investigating the feasibility of utilizing CO_2 from biogas upgrading, and comparing CBG and LBG pathways to fossil fuels such as natural gas.

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.

CRediT authorship contribution statement

Marcus Gustafsson: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Writing - review & editing, Visualization. **Igor Cruz:** Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Writing - review & editing, Visualization. **Niclas Svensson:** Conceptualization, Writing - review & editing, Visualization, Supervision. **Magnus Karlsson:** Conceptualization, Writing - review & editing, Visualization, Supervision.

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Appendix

Table A1
Environmental impact potential, primary energy use and life cycle cost for studied scenarios of CBG and LBG production and distribution. The values presented refer to cases with an annual production of 120 GWh, a distance from production site to filling station of 100 km and, in case of distribution by truck, two CBG trailers or one 25 ton cryogenic tank per truck.

End product	Upgrading	Liquefaction	Distribution	Climate change	Acidification	Freshwater eutrophication	Photochemical oxidation	Ozone layer depletion	Primary energy use	Life cycle cost
				kg CO_2-eq/MJ	kg SO_2-eq/MJ	kg P-eq/MJ	kg NMVOC/MJ	kg CFC-11-eq/MJ	kWh/MJ	€ cent/MJ
CBG	WS		Low-pressure grid	1.53E-03	5.36E-06	5.31E-07	3.92E-06	1.29E-09	0.032	0.19
	WS		High-pressure grid	6.08E-03	2.96E-05	9.45E-07	2.98E-05	2.70E-09	0.037	0.31
	WS		Semitr. (steel)	5.00E-03	1.67E-05	8.17E-07	2.19E-05	1.25E-09	0.041	0.71
	WS		Semitr. (comp.)	2.80E-03	9.41E-06	6.21E-07	1.06E-05	1.11E-09	0.035	0.45
	Cryo	Cryo	Semitr. (cryo. tank)	2.44E-03	9.52E-06	8.43E-07	8.21E-06	1.67E-09	0.055	0.63
	WS	MR	Semitr. (cryo. tank)	2.40E-03	8.41E-06	7.30E-07	7.51E-06	1.43E-09	0.054	0.56
	WS	N_2	Semitr. (cryo. tank)	2.71E-03	9.68E-06	8.58E-07	8.43E-06	1.70E-09	0.061	0.70
	OS		Semitr. (steel)	4.94E-03	1.64E-05	7.93E-07	2.17E-05	1.21E-09	0.039	0.66
	PSA		Semitr. (steel)	5.22E-03	1.68E-05	8.32E-07	2.21E-05	1.29E-09	0.046	0.71
	MS		Semitr. (steel)	4.97E-03	1.66E-05	8.06E-07	2.18E-05	1.23E-09	0.040	0.73
	AS (DH)		Semitr. (steel)	4.65E-03	1.56E-05	7.17E-07	2.06E-05	1.00E-09	0.040	0.74
	AS (steam)		Semitr. (steel)	4.85E-03	1.75E-05	8.28E-07	2.42E-05	9.62E-10	0.037	0.69
LBG	Cryo	Cryo	Semitr. (cryo. tank)	2.38E-03	9.26E-06	8.17E-07	8.03E-06	1.62E-09	0.053	0.62
	WS	MR	Semitr. (cryo. tank)	2.34E-03	8.16E-06	7.04E-07	7.33E-06	1.38E-09	0.053	0.59
	WS	N_2	Semitr. (cryo. tank)	2.65E-03	9.43E-06	8.33E-07	8.25E-06	1.65E-09	0.059	0.69
	WS	Press. red.	High pressure grid	5.55E-03	2.75E-05	7.28E-07	2.83E-05	2.25E-09	0.036	0.21
	OS	MR	Semitr. (cryo. tank)	2.28E-03	7.91E-06	6.80E-07	7.17E-06	1.34E-09	0.051	0.58
	PSA	MR	Semitr. (cryo. tank)	2.77E-03	9.14E-06	8.05E-07	8.13E-06	1.59E-09	0.053	0.59
	MS	MR	Semitr. (cryo. tank)	2.31E-03	8.04E-06	6.93E-07	7.25E-06	1.36E-09	0.052	0.24
	AS (DH)	MR	Semitr. (cryo. tank)	1.22E-03	3.79E-06	2.34E-07	4.22E-06	3.53E-10	0.055	0.62
	AS (steam)	MR	Semitr. (cryo. tank)	1.42E-03	5.75E-06	3.44E-07	7.76E-06	3.12E-10	0.051	0.58

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