



# Gas Hydrates as a Means for Biogas and Biomethane Distribution

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Biomethane is receiving great attention as a renewable energy gas with lower environmental impacts and diversified sources of production. However, availability of gas infrastructure is an important factor in biomethane development and use. Biomethane can be distributed by the natural gas or local biogas grid. Biomethane can also be road-transported as compressed biomethane (CBG) or liquefied biomethane (LBG). Biomethane could be distributed via gas hydration technology, where methane molecules are physically trapped within the crystalline structures of frozen host water molecules as gas hydrate compounds. Using life cycle assessment methodology, this study compared the energy performance and climate impact of two gas hydrate scenarios, biogas hydrate and biomethane hydrate, with that of a base case distributing biomethane as CBG. The technical system, from biogas upgrading, hydration, compression and road transport to filling station of biomethane as CBG, was included in the analysis. Results of this study show that distribution of biomethane as gas hydrates had a lower energy performance and higher climate impact than compressed biomethane distribution. The low energy performance was due to high electricity demand in hydrate formation and dissociation processes. The gas hydrate scenarios also had higher climate impacts as a result of high methane losses from hydrate formation and dissociation and emissions related to energy source use. Biogas upgrading to biomethane also significantly contributed to methane losses and climate impact of the scenarios studied.

**Keywords:** biogas hydrate, biomethane hydrate, compressed biomethane, life cycle assessment, primary energy inputs, climate impact

## OPEN ACCESS

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### Specialty section:

This article was submitted to  
Sustainable Energy Systems  
and Policies,  
a section of the journal  
Frontiers in Energy Research

**Received:** 02 June 2020

**Accepted:** 12 March 2021

**Published:** 07 April 2021

### Citation:

Moghaddam EA, Larsolle A,  
Tidåker P and Nordberg Å (2021) Gas  
Hydrates as a Means for Biogas  
and Biomethane Distribution.  
Front. Energy Res. 9:568879.  
doi: 10.3389/fenrg.2021.568879

## INTRODUCTION

Biomethane is a versatile biomass-derived renewable energy carrier with the ability to produce energy services and high-value products. The digestion of organic matter results in biogas containing approximately 45–65% by volume (v/v) of methane (CH<sub>4</sub>) and 25–30% v/v of carbon dioxide (CO<sub>2</sub>) with the remaining consisting of hydrogen sulfide (H<sub>2</sub>S), moisture and siloxanes. The energy content of biogas can be increased through drying, cleaning and upgrading, resulting in biomethane with > 97% CH<sub>4</sub> (Wellinger et al., 2013). In 2018, total biogas production in Sweden was around 2.1 TWh, of which 1.3 TWh was upgraded to biomethane. Approximately 42% of this biomethane was injected to gas grids in south-west Sweden and Stockholm. The remaining biomethane was stored and road-transported as compressed biomethane (CBG; 200 bar) in an “off-grid” solution due to lack of gas grid infrastructure. However, 44 GWh were converted to liquefied biomethane (LBG) at one plant (Swedish Energy Agency, 2018).

The greatest drivers of biogas use are a sustainable and renewable alternative that can contribute to a fossil-free transport sector and reduce organizations' environmental and climate impact. Energy and carbon dioxide tax exemptions, local alternative and circular economy, high consumer demands for sustainability, and possible competitive advantage are among the other driver of biogas use in the transport sector in Sweden (Geerolf, 2018).

The storage and transport system for CBG consists of steel or composite cylinders filled to a pressure of approximately 200 bar and road transport to central gas filling stations or industrial consumers (Hjort and Tamm, 2012; Budzianowski and Brodacka, 2017). Gas upgrading and compression are the most energy demanding stages, representing approximately 13% of the energy content of the biogas (Moghaddam et al., 2015). However, an issue related to CBG is the overall weight of the truck, including the load, in comparison with the amount of gas transported. Distribution as compressed biomethane (CBG) has a low energy density compared with liquid fuels and is a suitable option for dispensing and distribution in the vicinity of a local biogas plant. Another alternative for biomethane distribution is to liquefy biomethane at low temperature of  $-161^{\circ}\text{C}$  for road transportation as a liquid fuel. However, LBG is an energy intensive technology restricted to large-scale biomethane production units ( $>30$  GWh/year) and, due to the increase in temperature of the LBG tank during storage and transportation, LBG will evaporate, creating boil-off gas (Hjort and Tamm, 2012).

Selection of an appropriate transport and storage system for biomethane has a significant impact on the overall efficiency of biomethane system performance (Budzianowski and Brodacka, 2017). Apart from compression (i.e., CBG) and liquefaction (i.e., LBG), biomethane has the possibility to be stored and transported through physical conversion of  $\text{CH}_4$  molecules. In this method,  $\text{CH}_4$  is converted to clathrate hydrates, which are compounds in which  $\text{CH}_4$  molecules are physically trapped within the crystalline structures of frozen host water molecules. The required pressure for hydrate formation is around 30–100 bar and the temperature usually needs to be between  $-15$  and  $-32^{\circ}\text{C}$  (Budzianowski and Brodacka, 2017).

Gas hydrates is characterized by high storage capacity. Theoretically, a given volume of gas hydrates contains more than 150-fold the same volume of gas at standard temperature and atmospheric pressure, which shows that gas hydrates are promising for gas storage (Mori, 2003; Siazik et al., 2017). Compared to other biomethane storage and distribution methods (i.e., gas grid, gas liquification, gas compression) hydrate technology has the advantage of high safety and flexibility (Wu et al., 2019). In nature, gas hydrates are stable sources of gas existing under natural conditions of elevated pressure and low temperature found in marine sediments and permafrost regions, which abound in conventional deposits of natural gas (Sloan and Koh, 2007). Gas hydrates may be a suitable means of biomethane storage and transport in Sweden, due to low environmental temperatures and lack of grid infrastructure. Introducing methane hydrate in the logistics chain between the producer and consumer of biomethane would result in three

main stages; formation of hydrates, storage of hydrates and dissociation of hydrates.

*Formation of hydrates:* Gas hydrates are produced by bringing liquid water into contact with gas at the appropriate temperature and pressure. The formation of gas hydrates can take place in a continuous stirred tank reactor, where the gas is injected into liquid water. The necessary cooling is provided by an ice/water slurry, which is injected into the reactor. During the formation process, gas mixes of  $\text{CH}_4$  and  $\text{CO}_2$  molecules are captured in polyhedral structures consisting of hydrogen bonded water molecules. The molecular structure is theoretically  $8\text{G}46\text{H}_2\text{O}$ , and the gas to water mole ratio is 0.1739, which represents a hydration number of 1:5.75 (Kang et al., 2009). According to Uchida et al. (2005), guest molecules of  $\text{CH}_4$  and  $\text{CO}_2$  are incorporated into hydrate structures in proportion to their concentration in the feed gas.

*Storage and transport of gas hydrates:* After formation, hydrates are stored in containers at near adiabatic conditions. Gas hydrates remain stable during storage and transport at atmospheric pressure and freezing temperatures ( $-5$  to  $-15^{\circ}\text{C}$ ), representing similar physical conditions to those of simple hydrates (Gudmundsson et al., 1999, 1994). During storage of mixed gas hydrates, the cage structures enclathrating  $\text{CH}_4$  molecules are more stable than those enclathrating  $\text{CO}_2$  molecules. This allows loss of  $\text{CO}_2$  and increased concentration of  $\text{CH}_4$  in the structured ice. However, the decomposition rate of frozen gas at atmospheric temperature is negligible. Some studies report no gas hydrate decomposition within years of storage (Ershov and Yakushev, 1992; Sun et al., 2019). Gas-hydrate compounds are distributed to a transportation unit with a suitable storage container to transport/store the hydrate at adiabatic conditions at atmospheric pressure or at a slight gauge pressure at a temperature below  $0^{\circ}\text{C}$ ., preferably at  $-10^{\circ}\text{C}$ . to  $-15^{\circ}\text{C}$  (Gudmundsson, 1996).

*Dissociation of hydrates:* Dissociation of hydrates can be performed in three different ways; by increasing the temperature, by decreasing the pressure or by adding an inhibitor to the hydrate. During thermal dissociation,  $\text{CO}_2$  is more easily released than  $\text{CH}_4$ . However, hydrates with a higher  $\text{CO}_2$  concentration tend to decompose more than those with less  $\text{CO}_2$  (Pang et al., 2009; Kwon et al., 2011; Wang et al., 2017).

When proposing novel systems for distribution and transport of biomethane, there is a need to analyze the energetic and environmental performance in a systems perspective, including a comparison with conventional techniques. The approach adopted in this study was to evaluate the energy performance and climate impact associated with implementation of gas hydrates from biogas and biomethane produced in a small-scale anaerobic digestion unit using life cycle assessment (LCA). The LCA methodology is applicable for compiling and evaluating potential environmental impacts of a product system throughout its life cycle (ISO, 2006a,b) and has been extensively used for comparisons of different bioenergy systems (e.g., Cherubini and Strømman, 2011). Through a life cycle perspective, environmental hotspots can be highlighted, for identification of potential improvements. This is important when evaluating future-oriented

scenarios, including technology not yet implemented in a specific setting.

## Methods

Life cycle assessment methodology was used to assess the energy performance and climate impact of three different biogas distribution system scenarios related to a small-scale biogas production unit with CBG as the final product. The scenarios were a conventional system based on road transport of CBG and two future-oriented scenarios based on gas hydration (i.e., biogas hydrates and biomethane hydrates). Climate impact and energy inputs were calculated throughout the life cycle, from raw biogas upgrading to gas compression, hydrate formation and dissociation in the gas hydrate scenarios, storage and road transport to filling station. Distribution of biomethane as CBG was assumed as the base scenario, to which gas hydrate distribution technologies were compared. The LCA method used is described in ISO standards 14040 and 14044 (ISO, 2006a,b) and is similar to the methodology described in the sustainability criteria for biofuels in the European Union Directive (2009). Further, an attributional modeling approach was used, in which the inputs and outputs were attributed to the functional unit of a product system by linking and/or partitioning the unit processes of the system (Curran, 2015). This can be compared with consequential LCA modeling, which examines the environmental consequences of marginal changes in a life cycle, often with a market-oriented approach (Zamagni et al., 2012).

Energy use was studied as primary energy (PE) input. Factors used for conversion of data on electricity and diesel to PE has been assessed from Ecoinvent (2017) and are presented in **Table 1**. The PE factor is defined as the ratio between PE input and delivered useful energy. Included in PE are extraction of fuel, transportation and conversion, transmission and distribution losses. The electricity required for the processes was assumed to be Swedish electricity mix, mainly consisting of electricity produced from hydropower (43%) and nuclear power (41%) (Byman, 2016). Energy performance was assessed by both the total PE inputs and the ratio between delivered biomethane and primary energy inputs. It was assumed that the surplus low-grade heat generated left the system and it was not accounted for or valorized in calculations.

The climate impact, expressed as global warming potential (GWP), considered the upstream and downstream emissions

of greenhouse gases (GHGs), i.e., CO<sub>2</sub>, CH<sub>4</sub> and nitrous oxide (N<sub>2</sub>O), during the life cycle of biomethane distribution systems in the three different scenarios. Thus, emissions occurring inside the system boundaries, such as emissions related to input resources (i.e., input electricity and diesel), transportation and different technical units (i.e., gas compression, biogas upgrading, gas hydration and dissociation) were included in the calculations. The emissions were calculated as CO<sub>2</sub>-equivalents (CO<sub>2</sub>-eq.), using characterization factors for a 100-year perspective based on IPCC (2006). The characterization factor used when calculating GWP100 was 28 and 30 for biogenic and fossil CH<sub>4</sub>, respectively, and 265 for N<sub>2</sub>O (Stocker et al., 2013). Biogenic CO<sub>2</sub> was not included in the GHG accounting.

## Functional Unit and System Boundaries

The functional unit (FU) was defined as 1 Nm<sup>3</sup> biogas (60% CH<sub>4</sub>, 40% CO<sub>2</sub> at 1 bar and 0°C) leaving the anaerobic digester and entering the system boundary for each scenario. The anaerobic digestion process (handling and use of substrate, plant operation and use of digestate) was assumed to be identical for all three scenarios assessed and was therefore not included in the analysis. The system boundary of the study encompassed the following processes:

- Biogas cleaning and upgrading,
- Compression to CBG,
- Physical conversion to biogas hydrate and biomethane hydrate,
- Storage and distribution to a gas filling station.

Embodied impacts of capital equipment and infrastructure production of capital goods, such as machinery and buildings, were not included in the calculations, due to their minor influence on the results (Grant and Bengtsson, 2017).

## Sensitivity Analysis

Assessment of future-oriented scenarios, such as the hydrate scenarios, is associated with high un-certainty since little data are available from implementation in real settings. Since assumptions made in a scenario study might be critical for the outcome and the results, a sensitivity analysis was performed. In this, the influence of transport distance, choice of electricity mix, CH<sub>4</sub> emission levels from the upgrading unit, hydration efficiency and CH<sub>4</sub> emissions during gas handling in hydration and dissociation processes on the total PE input and GHG emissions was evaluated.

## System Description

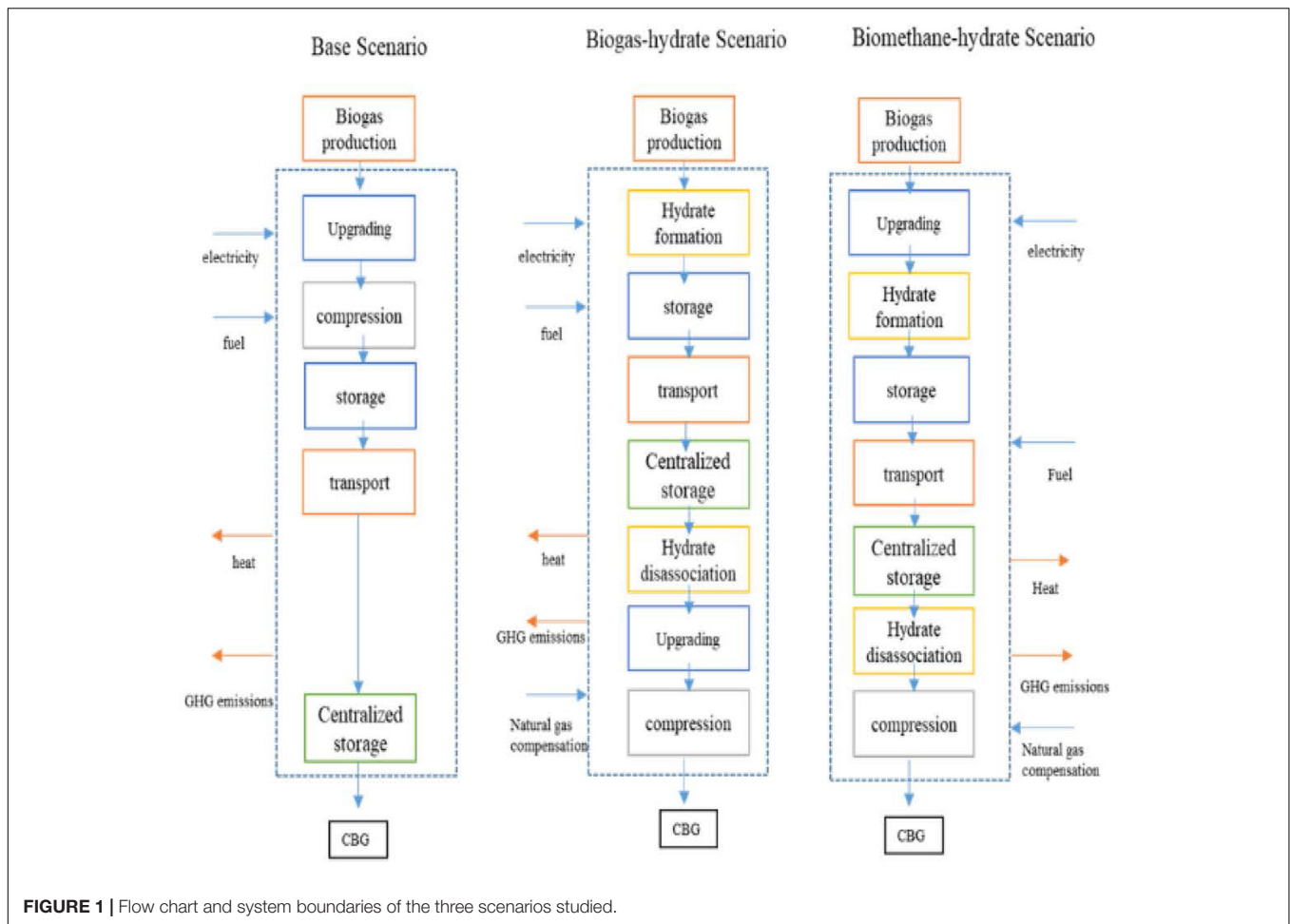
### System Description Overview and General Assumptions

The assessment was based on small-scale biogas production of 40 Nm<sup>3</sup>/h of raw biogas (1 bar and 0°C) with a composition of 60% CH<sub>4</sub>, and 40% CO<sub>2</sub>. The annual gross energy equivalent of the biogas produced was 2096 MWh, based on 9.97 kWh/Nm<sup>3</sup> CH<sub>4</sub>. The base scenario reflected conventional management of biogas, i.e., raw biogas was assumed to be cleaned, upgraded and compressed in cylindrical tanks to CBG at the biogas plant

**TABLE 1** | Primary energy (PE) and climate impact factors for different energy carriers.<sup>1</sup>

Energy carrier	Specifications	PE factor (MJ/MJ)	Climate impact (g CO <sub>2</sub> -eq./MJ)
Electricity	Swedish electricity mix	1.6	16
	Hydropower	1.0	1.1
	Natural gas	2.0	100
Fuel	Diesel, low-sulphur	1.3	77
	Natural gas	Fossil	1.1

<sup>1</sup>Ecoinvent ver.3-4 (2017).



and transported to a centralized facility for storage and further use (Figure 1). In the biogas hydrate scenario, raw biogas was assumed to be converted to hydrate at the biogas plant and transported in containers to a centralized facility for storage and further gas handling, thereafter dissociation of hydrate, upgrading and compression to CBG. In the biomethane hydrate scenario, raw biogas was assumed to be upgraded to biomethane and converted to hydrate at the biogas plant for transportation in containers to a centralized facility for storage and thereafter dissociation of hydrate and compression to CBG. The output from all three scenarios was CBG at 200 bar. In all scenarios, storage time at the biogas plant was determined by the storage container capacity of one transport of containers. The assumed storage time at the final destination was set to 14 days. The transport distance from the biogas plant to the central facility was set to 100 km. Model implementation and scenario calculations were performed using a previously developed calculation and modeling platform (Norberg et al., 2013). The surplus low value heat leaving the system was not valorized or accounted for. In all scenarios, the upgrading unit had a  $\text{CH}_4$  loss of 1% of the biomethane produced (Lantz et al., 2009). Handling operations during gas hydration and dissociation were assumed to result in a  $\text{CH}_4$  loss of 2% of the biomethane produced. The lower biomethane output from the hydrate scenarios due to gas losses

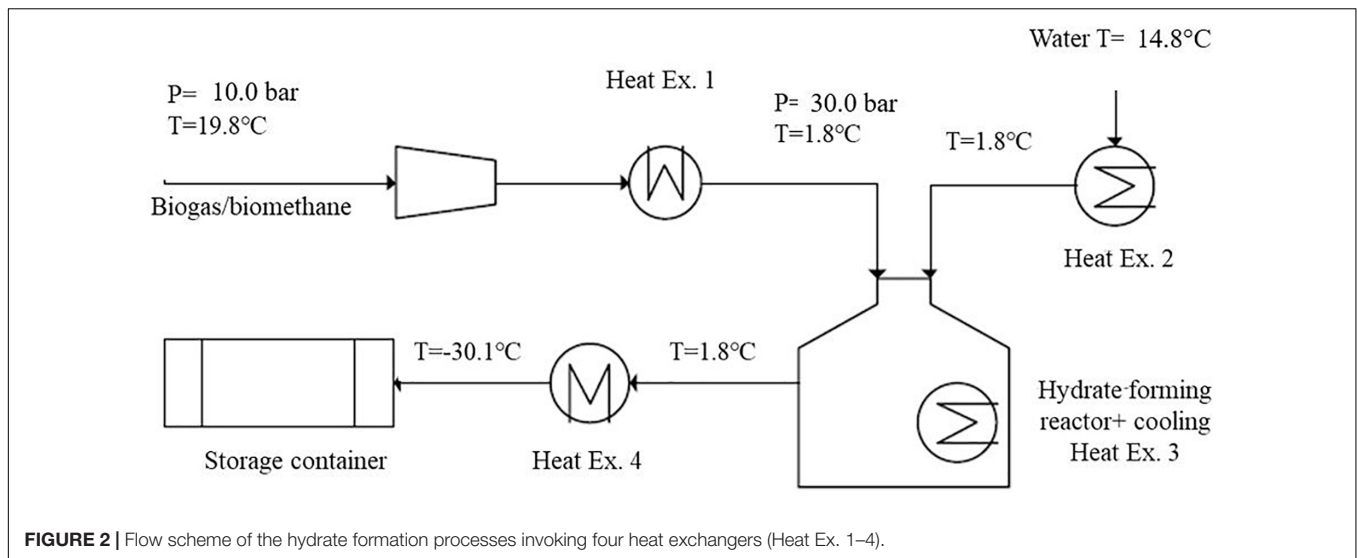
was compensated for by natural gas, in order to reach an equal output to that in the base scenario.

### Base Scenario

The small-scale upgrading was based on a decentralized water scrubbing technique with a lower pressure requirement and lower energy demand than conventional water absorption technologies (Bauer et al., 2013). The required electricity was  $0.20 \text{ kWh/Nm}^3_{\text{rawbiogas}}$  and compression of bio-methane to 200 bar based on four compression stages required  $0.23 \text{ kWh/Nm}^3_{\text{rawbiogas}}$  according to Bauer et al. (2013). The compressed biomethane was assumed to be transported in pressurized steel vessels on a hook-lift truck with a capacity of  $4,000 \text{ Nm}^3$  biomethane per truck (Norberg et al., 2013) and diesel consumption of  $0.48 \text{ L/km}$  (Benjaminsson and Nilsson, 2009). The storage time for CBG at the biogas plant was 6.9 days, based on the storage container capacity for one transport.

### Biogas Hydrate Scenario

The hydrate formation process was divided into compression work, gas cooling, water cooling, hydrate formation, and pumping and mixing (Figure 2). The equilibrium pressure and temperature profile were obtained from Arca et al. (2011). The biogas was assumed to be compressed in two stages



with equivalent compression ratio and compressor efficiency of 0.8 (isentropic work). The gas cooling power demand was calculated from 20 to 2°C for a heat exchanger (Heat Ex. 1) with a thermal exchange efficiency of 0.75. The power for cooling the water entering together with the gas to the hydrate formation reactor was correspondingly calculated from an inlet temperature of 15 to 2°C and a heat exchanger efficiency of 0.75 (Heat Ex. 2). The cooling power required for the hydrate formation was calculated based on the hydrate dissociation enthalpy for CH<sub>4</sub> and CO<sub>2</sub> (54.5 and 61 kJ/mol, respectively) according to Kwon et al. (2011), and with a cooling efficiency of 0.75 (Heat Ex. 3). A gas-to-water stoichiometric ratio of 1:5.75 for hydrate formation was assumed, with a water conversion efficiency of 0.8 (Arca et al., 2011), thus resulting in a ratio of 1:7.1875 for the calculations. The power for cooling the hydrate to a storage temperature of -30°C was calculated for a heat exchanger (Heat Ex. 4) with a thermal efficiency of 0.75. A compression chiller with a coefficient of performance (COP) of 3.0 was assumed for the gas and water cooling. The power demand for pumping and mixing inside the hydrate formation reactor was approximated to 5% of the entire process demand. After formation, the hydrate was assumed to be transferred to, and stored in, gastight freezing containers consisting of three jacketed vessels with a total inner volume of 11.1 m<sup>3</sup>. The density of hydrate was set to 0.9 t/m<sup>3</sup> (Wang et al., 2017), leading to a weight capacity of 10 tons of hydrate per container. Ethanol was selected to be the refrigerant surrounding the vessels and each jacketed vessel was assumed to be isolated with 10 cm of styrofoam. The freezing containers were assumed to be transported by a truck equipped with a hook lift and with a total capacity of three containers, resulting in a storage time of 4 days at the biogas plant.

The hydrate was assumed to be cooled to -30°C at the plant and no further cooling power in transport and storage at the centralized facility was needed. The maximum temperature of the hydrate before dissociation was set to -10°C. Heat required for dissociation of the hydrates was the heat for melting the hydrate from -10 to 0°C and the

dissociation enthalpy. This heat was supplied by a compression heat pump with the COP set to 3, including recovery of heat from subsequent compression of biomethane to 200 bar (Figure 3). Upgrading at the central facility was by conventional water scrubber with a capacity of 320 Nm<sup>3</sup> rawbiogas/h. The electricity required for upgrading was 0.30 kWh/Nm<sup>3</sup> rawbiogas and subsequent compression of biomethane to 200 bar required 0.16 kWh/Nm<sup>3</sup> rawbiogas according to Bauer et al. (2013).

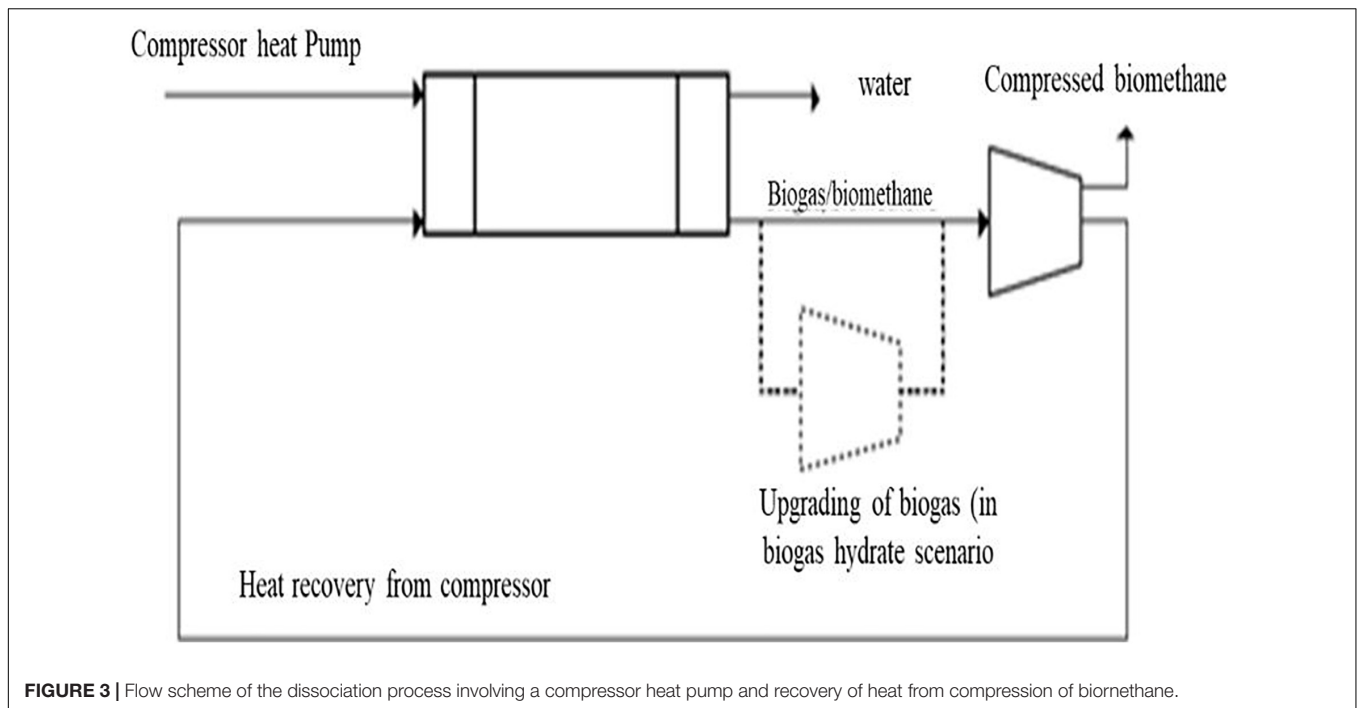
### Biomethane Hydrate Scenario

Biogas was upgraded to biomethane in a decentralized upgrading unit similar to that in the base scenario. The equilibrium pressure and temperature profile for methane hydrate were taken from Arca et al. (2011). The working pressure was set to 40 bar and the temperature to 2°C. Electricity demand for hydrate formation, including cooling of the hydrate to -30°C, was calculated with the same gas and water temperatures and efficiencies as for the biogas hydrate scenario. Calculation of weighted averages of specific heat capacity and dissociation enthalpy was based on a composition of 97% CH<sub>4</sub> and 3% CO<sub>2</sub>. Storage and transport of biomethane hydrate were as assumed in the biogas hydrate scenario. The storage time at the biogas plant was 8 days, based on the storage container capacity for one transport. The heat requirement for hydrate dissociation at the centralized facility was based on melting the hydrate from -10°C and the dissociation enthalpy. Recovery of heat from subsequent compression of biomethane to 200 bar was performed by a similar heat pump as used in the biogas hydrate scenario.

## RESULTS

### Energy Input

The PE input per FU for different units in each scenario is presented in Table 2. Total PE input in the base, biogas hydrate and biomethane hydrate scenario was 3.5, 11.5 and 8.2 MJ/Nm<sup>3</sup> rawbiogas, respectively. The high energy inputs in



**FIGURE 3 |** Flow scheme of the dissociation process involving a compressor heat pump and recovery of heat from compression of biomethane.

the gas hydrate scenarios derived from high levels of electricity demand in the hydrate formation and dissociation units. The PE input to hydrate formation and dissociation in the biogas hydrate scenario was  $6.4 \text{ MJ}/\text{Nm}^3_{\text{rawbiogas}}$  (55.6% of the total PE input), while the corresponding PE input in the biomethane hydrate scenario was  $4.1 \text{ MJ}/\text{Nm}^3_{\text{raw biogas}}$  (50.0% of the total PE input). In the biogas hydrate scenario, the centralized upgrading of biogas had a higher PE input ( $2.5 \text{ MJ}/\text{Nm}^3_{\text{rawbiogas}}$ ) than the decentralized upgrading unit used in the biomethane hydrate and base scenarios ( $1.7 \text{ MJ}/\text{Nm}^3_{\text{rawbiogas}}$ ). The small-scale decentralized upgrading unit had a lower electricity demand for increasing the pressure in the water scrubber, compared with the large-scale centralized water scrubber. The compression work for producing CBG in the biogas hydrate scenario was slightly lower than in the other scenarios, since the compression started at a higher pressure due to the pressure build-up in the upgrading

process. The main PE input in the base scenario was upgrading of biogas and compression to 200 bar, corresponding to 80% of the total PE input.

Diesel consumption for biomethane hydrate was  $0.4 \text{ MJ}/\text{Nm}^3_{\text{rawbiogas}}$ , while it was higher for the base and biogas scenarios, corresponding to 0.7 and  $1 \text{ MJ}/\text{Nm}^3_{\text{rawbiogas}}$ , respectively. The higher fuel consumption in the base and biogas hydrate scenarios was due to higher total mass transported as a result of heavy steel tanks used in the base scenario and higher density of biogas hydrates in comparison to biomethane hydrates.

The difference in biomethane ( $\text{CH}_4$ ) output from the base and the hydrate scenarios was considered by compensation of natural gas in the two hydrate scenarios. The energy required for natural gas compensation in the hydrate scenarios was  $0.8 \text{ MJ}/\text{Nm}^3_{\text{rawbiogas}}$ , corresponding to 2% total loss of biomethane during the gas hydrate formation and dissociation processes.

**TABLE 2 |** Primary energy (PE) inputs ( $\text{MJ}/\text{Nm}^3_{\text{rawbiogas}}$ ) for the three scenarios studied.

	Base	Biogas hydrate	Biomethane hydrate
Decentralised upgrading/centralised upgrading	1.7	2.5	1.7
Decentralised compression/centralised compression	1.1	0.8	1.2
Hydrate formation		5.5	3.3
Hydrate disassociation		3.0	1.9
Transport	0.7	1.0	0.4
Natural gas compensation	–	0.8	0.8
Total	3.5	13.6	9.3

## Electricity Requirement for Formation and Dissociation of Hydrate

The higher PE energy inputs to the hydrate scenarios were mainly due to the electricity demand for the hydrate formation and dissociation processes. The electricity demand ( $\text{MJ}_{\text{el}}/\text{Nm}^3_{\text{rawbiogas}}$ ) for formation of hydrate, cooling for storage and dissociation of hydrates is presented in **Table 3**. Cooling for hydrate formation and heating for dissociation were the most energy-demanding operations, making up approximately 30% and 45% of the total electricity demand, respectively, in both hydrate scenarios (**Table 3**). No electricity was required during storage in either of the scenarios.

## Energy Performance

The PE inputs, energy outputs and energy ratio (Outputs/Inputs) of the three scenarios are presented in **Table 4**. The output of all scenarios studied consisted of biomethane (97% CH<sub>4</sub>), which in the hydrate scenarios was slightly lower due to losses in gas handling during hydrate formation and dissociation. The base scenario had the highest energy ratio, due to the relatively low PE input, whereas the biomethane hydrate scenario and biogas hydrate scenario had much lower energy ratio as a result of the high PE input.

## Climate Impact: Global Warming Potential (GWP)

The contribution to climate impact of the biogas and biomethane hydrate scenarios was approximately 4-fold and 3-fold higher, respectively, than in the base scenario (**Table 5**). The upgrading unit was the highest contributor in the base and biogas hydrate scenarios. Total global warming potential in the base, biogas hydrate and biomethane hydrate scenarios was 141, 567 and 416 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>rawbiogas</sub>, respectively. Loss of CH<sub>4</sub> from the upgrading unit corresponded to 120 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>rawbiogas</sub> for all scenarios. The difference in total climate impact from the upgrading unit in the three scenarios was related to the energy (i.e., electricity) demand. The centralized upgrading unit in the biogas hydrate scenario had a higher electricity demand than the decentralized upgrading unit in the biomethane hydrate and base scenarios. The hydrate formation and dissociation units

**TABLE 3** | Electricity demand for the formation, storage and dissociation of hydrates.

	Biogas hydrate (60% CH <sub>4</sub> , 40% CO <sub>2</sub> )	Biomethane hydrate (97% CH <sub>4</sub> , 3% CO <sub>2</sub> )
	MJ <sub>el</sub> /Nm <sup>3</sup> <sub>rawbiogas</sub>	
Compression of gas	0.5	0.4
Cooling of gas, water and formation of hydrate	1.2	0.8
Pumping and mixing	0.2	0.2
Cooling of hydrate to storage temperature	0.2	0.1
Heating for dissociation	1.9	1.2
Total electricity demand	4.1	2.7

**TABLE 4** | Primary energy (PE) inputs, outputs and losses, and energy ratio (O/I) for the three scenarios studied.

	Input	Output	Losses	Energy ratio (O/I)
	(MJ/Nm <sup>3</sup> <sub>rawbiogas</sub> *)			
	Total	Biomethane	Methane	
Base (CBG)	3.5	35.6	0.4	10.0
Biogas hydrate	11.5	35.3	0.7	3.0
Biomethane hydrate	9.3	35.3	0.7	3.7

\*Based on lower heating value, LHV.

**TABLE 5** | Global warming potential (g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>rawbiogas</sub>) in the three scenarios studied.

	Base (CBG)	Biogas hydrate	Biomethane hydrate
Upgrading/centralised upgrading	131	258	136
Compression/centralised compression	3	7	7
Hydrate formation		133	119
Hydrate disassociation		150	143
Transport	9	13	5
Natural gas compensation		6	6
Total	143	567	416

produced high levels of GHG emissions, due to CH<sub>4</sub> losses and high levels of energy input in comparison with the base scenario. Methane losses during gas handling in the hydrate formation process were 79 and 84 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the biogas and biomethane hydrate scenario, respectively. The GHG emissions related to electricity input in hydrate formation corresponded to 54 and 35 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the biogas and biomethane hydrate scenarios, respectively.

Methane losses during gas handling in the hydrate dissociation process were 102 and 113 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the biogas and biomethane hydrate scenarios, respectively. The GHG emissions related to energy input in hydrate dissociation corresponded to 48 and 30g CO<sub>2</sub> eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the biogas and biomethane hydrate scenario, respectively. The climate impact from compression of biomethane to CBG only related to electricity use. The climate impact associated with transport of biogas hydrate was higher than in the biomethane hydrate and base scenarios (**Table 5**).

The climate impact of natural gas compensation in the hydrate scenarios was 6 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>rawbiogas</sub>, corresponding to 2% total loss of biomethane in the gas hydrate formation and dissociation units.

## Sensitivity Analysis

Sensitivity analysis was performed to examine the impact of data uncertainties on the energy performance and climate impact. The parameters tested were transport distance, choice of electricity mix, CH<sub>4</sub> losses from the upgrading unit, CH<sub>4</sub> losses during gas handling in the hydrate scenarios and hydration efficiency (**Tables 6, 7**).

The sensitivity to transport distance was analyzed in order to determine the influence of larger transport distances for the three scenarios. Increased transport distance to 1,000 km was compared with 100 km in the base scenario. The results showed a large relative increase in PE input and climate impact per FU for the base scenario compared with the hydrate scenarios. Increasing the transport distance to 1,000 km increased the PE inputs to 11.0, 17.0 and 11.7 MJ/Nm<sup>3</sup><sub>rawbiogas</sub> in the base, biogas hydrate and biomethane hydrate scenarios, respectively. The climate impact with 1,000 km transport distance was 200.0, 612.0 and 429.0 g CO<sub>2</sub> eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the base, biomethane hydrate and biogas hydrate scenarios, respectively. The amount

**TABLE 6** | Change (%) in primary energy (PE) input per functional unit when selected input parameters were changed in the three scenarios studied.

Scenarios	Electricity mix		1,000 km transport	100% Hydration efficiency
	Natural gas	Hydropower		
Base (CBG)	+104	-44	+215	-
Biogas hydrate	+124	-40	+75	-6
Biomethane hydrate	+126	-50	+43	-6

of biomethane transported in the different scenarios was within the same range, while the total mass transported per trip was higher in the base scenario than in the hydrate scenarios (**Supplementary Table 1**).

In the base scenario, the Swedish electricity mix was used for the LCA calculations. In the sensitivity analysis, the results were compared with those for natural gas-based electricity, as a representative of fossil-based electricity, and hydro-based electricity, as a complete fossil-free electricity. The PE factors and climate impact used in the sensitivity analysis for the different electricity mixes are presented in **Table 1**. Choice of fossil-free electricity considerably decreased the PE input and climate impact in all scenarios. With the fossil-free option, the PE inputs decreased to 1.6, 6.5 and 4.0 MJ/Nm<sup>3</sup><sub>rawbiogas</sub> and the climate impact decreased to 126.0, 482.0 and 366.0 g CO<sub>2</sub> eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the base, biogas hydrate and biomethane hydrate scenarios, respectively. Choice of fossil-based electricity significantly increased the PE input and climate impact of the scenarios, especially the gas hydrate scenarios due to the high electricity input in the hydrate formation and dissociation processes.

With the fossil-based electricity mix, the PE inputs increased to 6.0, 26.0 and 18.6 MJ/Nm<sup>3</sup><sub>rawbiogas</sub> and the climate impact increased to 252.0, 1242.0 and 765.0 g CO<sub>2</sub> eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the base, biogas hydrate and biomethane hydrate scenarios, respectively.

Methane losses from the upgrading stage played an important role in the climate performance of all scenarios. Reducing the CH<sub>4</sub> losses to 0.5% (1% in the original scenario) resulted in a total climate impact of 80.0, 454.0 and 341.0 g CO<sub>2</sub> eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the base, biogas hydrate and biomethane hydrate scenarios, respectively. Increasing the CH<sub>4</sub> losses to 2%

resulted in a climate impact of 260.0, 663.0 and 515.0 g CO<sub>2</sub> eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the base, biogas hydrate and biomethane hydrate scenarios, respectively.

Gas handling in the hydrate formation and dissociation units led to gas losses, including losses of CH<sub>4</sub>, which contributed to the climate impact and reduced the performance of the hydrate scenarios. Reducing the CH<sub>4</sub> losses during gas handling from 2 to 0.5% in the hydrate scenarios resulted in a climate impact of 311.0 and 270.0 g CO<sub>2</sub> eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the biogas hydrate and biomethane hydrate scenarios, respectively. Doubling the CH<sub>4</sub> losses to 4% during gas handling in the hydrate scenarios increased the climate impact to 907.0 and 520.0 g CO<sub>2</sub> eq./Nm<sup>3</sup><sub>rawbiogas</sub> in the biogas hydrate and biomethane hydrate scenarios, respectively.

Based on the assumption of hydrate formation from liquid water, the efficiency of gas contact with water plays a key role in the formation of gas hydrates. Changing the hydration efficiency from 80% to 100% based on a hydration number of 1:5.75 were tested in the sensitivity analysis. In a modeling study by Anderson (2004), the hydration number was shown to vary with temperature and pressure. Within the temperature range 0–45°C, the hydration number was calculated to be highest (6.12) at 12°C and lowest (5.71) at 20°C. With the ideal hydration number for a structure I-hydrate (1:5.75) and an efficiency of 80% used in this study, the hydrate would contain 137.0 m<sup>3</sup>CH<sub>4</sub>/m<sup>3</sup> hydrate (Norberg et al., 2013). Assuming that the ideal hydration number could be obtained with 100% efficiency, i.e., containing 164 m<sup>3</sup>CH<sub>4</sub>/m<sup>3</sup> (Pellenbarg and Max, 2000), the total PE input would decrease to 10.8 and 7.7 MJ/Nm<sup>3</sup><sub>rawbiogas</sub> and the climate impact would decrease to 544.0 and 403.0 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>rawbiogas</sub> for the biogas hydrate and biomethane hydrate scenarios, respectively.

## DISCUSSION

The gas hydrate scenarios examined in this study had a lower energy output/input ratio and a higher climate impact than in the base scenario, where biomethane was assumed to be distributed as CBG. The main processes in the hydrate scenarios associated with high energy use and climate impact were hydrate formation and dissociation. Comparing the hydrate scenarios, the biomethane hydrate scenario showed better energy performance than the biogas hydrate scenario, while the climate impact was approximately similar for both gas hydrate scenarios. This indicates that biogas should preferably be upgraded before

**TABLE 7** | Change (%) in global warming potential per functional unit when selected input parameters were changed in the three scenarios studied.

Scenarios	Methane loss				Electricity mix		1,000 km transport	100% Hydration efficiency
	Upgrading		Hydration formation and disassociation		Natural gas	Hydro-power		
	0.5%	2.0%	0.5%	4.0%				
Base (CBG)	-43	+85	-	-	+79	-12	+42	-
Biogas hydrate	-20	+17	-45	+60	+90	-15	+8	-4
Biomethane hydrate	-18	+24	-35	+25	+103	-12	+3	-3



hydration. However, the futuristic approach of the hydrate scenarios means that considerable uncertainties are embedded in the results, since assumptions had to be made due to lack of experiences and data. The lack of a detailed technical description and performance data on the hydrate formation and dissociation processes is clearly a limitation. In the absence of such data, the energy calculations were based on thermodynamic considerations and assumptions on conversion efficiencies. Furthermore, the CH<sub>4</sub> losses related to hydrate formation and dissociation had to be assumed and were taken to be in the same range as for biogas upgrading.

The results from the sensitivity analysis indicated that changing to hydropower electricity, increasing the transport distance, reducing CH<sub>4</sub> losses from both upgrading and hydrate handling and increasing the hydration efficiency to an ideal hydration number would not make the hydrate scenarios competitive in comparison with conventional CBG storage and distribution. However, when the transport distance was increased to 1,000 km, the PE inputs and climate impact in the hydrate scenarios were less affected than those in the base scenario, since the conventional CBG transport included heavier total mass transport due to the steel tanks used for compressed biomethane.

Methane losses made an important contribution to the climate impact (45% and 35% for the biogas hydrate and biomethane hydrate scenarios, respectively). Choice of electricity mix is a notoriously influential factor for the outcome of LCA studies. Changing from the Swedish electricity mix to hydropower in the present case reduced the PE inputs of all scenarios by ~50%, while using natural gas-based electricity more than doubled the PE input.

The results obtained in this study indicate that using hydrate as a means of transporting gas from a production site for use at another site is not favorable in terms of energy use. In future studies, it would be interesting to study integration of hydrates for storage at centralized biogas plants.

In the present study, cooling for hydrate formation and heating for hydrate dissociation was assumed to be performed by a compression chiller and heat pump, respectively. This resulted in high electricity consumption, which understandably resulted in an increase in both the PE input and the climate impact. Therefore, integration with e.g., return water in a district heating system or using surplus heat from a combined heat and power plant for dissociation heat could help to improve the energy balance. Moreover, expansion of the system to include the anaerobic digestion process, where low-grade heat from cooling during hydrate formation could be used, would likely improve the overall efficiency of an integrated system. In addition to system integration, it is also important to consider future developments in hydrate technologies. In CH<sub>4</sub>-CO<sub>2</sub> hydrate mixtures, the CO<sub>2</sub> is more easily released than the CH<sub>4</sub> (Kwon et al., 2011), a difference that could be exploited in an upgrading process (Arca et al., 2011; Silva, 2016). This would make it possible to integrate dissociation and upgrading in one process (Sun et al., 2015; Castellani et al., 2018). Furthermore, the use of detergents such as sodium dodecyl sulfate has been shown to enhance the hydrate formation rate at lower pressures with reduced stirring, resulting in lower energy

input (Zhong and Rogers, 2000). Thus, system integration and technical developments could reduce the limitations of hydrate use on an industrial scale.

Within the present study boundaries, material consumption and capital investments were not considered, but they are high in all scenarios. For instance, in the base scenario, high-pressure capsules for compressed biomethane (CBG) storage and transport are made from steel, steel alloys or aluminum, along with other metallic elements. Gas hydrates are confined in a water medium, which does not require very high pressure or low temperatures.

European Department of Transportation regulations classify CBG as a flammable and hazardous material. Major requirements for road transportation include approved tanks (e.g., seamless steel cylinders), specific tank pressure and vapor content (i.e., less than 10 ppm H<sub>2</sub>O) and a minimum methane content of 97%. Additional compression at the biomethane filling point is also required, which leads to gas loss and GHG emissions. Transport of biomethane as CBG is not a solution for very long distances due to technical requirement and investment cost. As previously mentioned LBG is an energy intensive technology restricted to large-scale biomethane production units.

Gas hydrate technology could be considered an alternative to CBG and LBG systems, as it could be distributed in medium quantities over an average distance and would be very efficient for large-scale biomethane storage units due to high withdrawal flow rate (Mimachi et al., 2015; Budzianowski and Brodacka, 2017). During storage, CH<sub>4</sub> emissions occur when the temperature is over 4°C and the storage pressure is lower than atmospheric pressure, which is of interest under the prevailing climate conditions in Sweden. Thus, there is promising scope for technical developments that could reduce the limitations of future hydrate use in industrial scale (Moghaddam, 2019).

A life cycle perspective was applied in this study, covering the energy inputs and GHG emissions from using biogas and biomethane hydrates for storage and transport in comparison with CBG. Production of raw material, anaerobic digestion and the filling unit were identical in all scenarios and were omitted from the analysis. Performing a complete LCA study could delay use of results in decision making and product development due to complexity, slowness and lack of detailed data in the initial stages. Identification of different parts of a services or product system that could be neglected during the analysis, without significantly affecting the overall results, leads to a reduced amount of data (Pelton and Smith, 2015). The results obtained could be combined with an extended economic input-output (EEIO) study to identify key hotspot inputs and stages within the biomethane life cycle and compare the environmental benefits against a baseline scenario. Implementing hotspot system analysis (HSA) would add a further dimension, particularly as regards sustainability purchasing criteria.

## CONCLUSION

The current study aimed to investigate gas hydrates as an alternative technology for biogas road distribution

which is one of the current barriers to the wider implementation of biogas as a source of renewable energy as the temporary supply problems have affected the trust for the vehicle biogas market in Sweden.

The study concludes that the hydration conversion process and dehydration require a high electricity input and contribute to high levels of GHG emissions. Even with suggested improvements to the biomethane hydrate production process, such as minimizing CH<sub>4</sub> losses from hydrate formation and dissociation and from gas upgrading to biomethane, the total energy requirement was 227.0 g CO<sub>2</sub> eq./Nm<sup>3</sup><sub>rawbiogas</sub>, which was almost twice the climate impact in the base (CBG) scenario.

To reduce the limitations for future use of hydrate as a mean for biogas and biomethane distribution, it is important to consider aspects on system integration and technical development. A combination of technological development (e.g., hydration efficiency and gas upgrading) and use of a completely clean source of electricity (e.g., 100% hydropower electricity mix) would significantly increase the potential for biomethane storage and distribution via gas hydrates.

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## DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**, further inquiries can be directed to the corresponding author/s.

## AUTHOR CONTRIBUTIONS

ÅN supervised the work. EM collected the data, carried out the LCA study, and wrote the manuscript. AL and ÅN conceptualized the work, PT, AL, and ÅN contributed to data inventory and reviewing and editing the article. All authors contributed to the article and approved the submitted version.

## SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fenrg.2021.568879/full#supplementary-material>

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**Conflict of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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