

# Life Cycle Assessment of Novel Biomethane Systems

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# Life Cycle Assessment of Novel Biomethane Systems. Energy Performance and Climate Impact

## Abstract

Climate mitigation and supply of renewable energy are global challenges. The main cause of climate change is anthropogenic activities, including consumption of fossil energy sources and land use change. Biomethane, a biomass-derived renewable energy carrier, is interchangeable with fossil-based natural gas and can provide energy services (*e.g.* heat, electricity and vehicle fuel) and high-value products such as chemicals. However, the availability of feedstock suitable for anaerobic digestion, the limited grid infrastructure in certain regions and problems relating to storage and distribution are barriers to increased deployment of biomethane systems.

This thesis aims to provide decision support for the development and implementation of future biomethane systems, by describing the energy performance and climate impact of some promising novel technologies related to biomethane production, conversion of biomethane to high-value products and biomethane distribution in a life cycle perspective. Anaerobic digestion of maize and pyrolysis of willow for production of biomethane were assessed and compared, while gas-to-liquid (GTL) technologies were studied as potential routes for conversion of biomethane to liquid transportation fuels or platform chemicals. Gas hydrates were assessed as a means of biomethane distribution.

The results showed that transition from maize-based anaerobic digestion to willow-based pyrolysis for biomethane production improved energy performance (higher external energy ratio) and environmental performance (lower climate impact), mainly due to build-up of soil organic carbon and use of biochar as a soil amendment or as an energy source to replace fossil coal. Use of biomethane for production of dimethyl ether as a GTL fuel was competitive relative to the conventional compressed biomethane system regarding energy performance and climate impact. Formation and disassociation of gas hydrates was associated with high energy use, and thus technological development is required to overcome the high primary energy inputs and related high climate impact of gas hydrate distribution.

*Keywords:* Biomethane, pyrolysis, biochar, GTL products, gas hydrates, life cycle assessment, climate impact, energy performance

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# Livscykelanalys av nya biometansystem. Energiprestanda och klimatpåverkan

## Sammanfattning

Att bekämpa klimatförändringen och öka tillförseln av förnybar energi är globala utmaningar. Huvudorsaken till klimatförändringen är antropogena aktiviteter, bl.a. förbrukning av fossila energikällor och förändrad markanvändning. Biometan, en förnybar energibärare baserad på biomassa, kan substituera fossil-baserad naturgas och tillhandahålla energitjänster (*t.ex.* värme, el och fordonsbränsle) samt högkvalitativa produkter som kemikalier. Begränsad tillgång av råvaror som är lämpliga för anaerob omsättning till biometan (rötning), brist på gasnät i vissa regioner och problem med lagring och distribution är dock hinder för ökad användning av biometan.

Denna avhandling syftar till att ge beslutsstöd för utveckling och tillämpning av framtida biometansystem genom att beskriva energiprestanda och klimatpåverkan av flera lovande nya teknologier relaterade till biometanproduktion, omvandling av biometan till högkvalitativa produkter och biometandistribution i ett livscykelperspektiv. Rötning av majs och pyrolys av *Salix* för produktion av biometan utvärderades och jämfördes. Tekniker för omvandling av gas till flytande produkter analyserades som potentiella vägar för produktion av flytande transportbränslen eller plattformskemikalier. Distribution av biometan i form av gashydrater utvärderades.

Resultaten visade att övergången från majsbaserad biometanproduktion till *Salix*baserad pyrolys för biometanproduktion förbättrade energi- och miljöprestandan (lägre klimatpåverkan), främst på grund av ökad uppbyggnad av organiskt kol i marken och användning av biokol till jordförbättring eller som energikälla för ersättning av fossilt kol. Användning av biometan för framställning av dimetyleter som ett flytande bränsle hade bättre prestanda än konventionell komprimering av biometan avseende energiprestanda och klimatpåverkan. Bildandet och disassociationen av gashydrater var förknippad med hög energianvändning. Därför behövs teknisk utveckling för att reducera den höga primärenergiåtgången, vilken är kopplad till den höga klimatpåverkan.

*Nyckelord:* Biometan, pyrolys, biokol, GTL-produkter, gashydrater, livscykelanalys, klimatpåverkan, energiprestanda

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## Dedication

*Dedicated to my beloved parents, my lovely husband and little daughter*

With them the seed of Wisdom did I sow,  
And with mine own hand wrought to make it grow;  
And this was all the Harvest that I reap'd  
"I came like Water, and like Wind I go.

*Khayyam (Persian mathematician, 1048).*

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## List of publications

This thesis is based on the work contained in the following papers, referred to by Roman numerals in the text:

- I Moghaddam, E.A.\*, Ahlgren, S., Hulteberg, C. & Nordberg, Å. (2015). Energy balance and global warming potential of biogas-based fuels from a life cycle perspective. *Fuel Processing Technology* 132, pp. 74-82.
- II Moghaddam, E.A.\*, Ahlgren, S. & Nordberg, Å. (2016). Assessment of novel routes of biomethane utilization in a life cycle perspective. *Frontiers in Bioengineering and Biotechnology* 4: 89.
- III Moghaddam, E.A.\*, Ericsson, N., Hansson, P-A. & Nordberg, Å. (2019). Exploring the potential for biomethane production by willow pyrolysis using life cycle assessment methodology. *Energy, Sustainability and Society* 9: 6.
- IV Moghaddam E.A., Larsolle A., Tidåker P. & Nordberg, Å. Gas hydrates as a means of biogas and biomethane distribution. (manuscript).

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The contribution of Elham A. Moghaddam to the papers included in this thesis was as follows:

- I Planned the paper with the co-authors. Assisted in scenario development. Carried out the data collection and impact assessment together with the co-authors. Wrote the paper with assistance from the co-authors.
- II Planned and structured the paper with the co-authors. Carried out the data collection and impact assessment. Wrote the paper with inputs from the co-authors.
- III Planned and structured the scenario development and the paper with the co-authors. Carried out the data collection and impact assessment. Wrote the paper with inputs from the co-authors.
- IV Planned the paper with the co-authors. Assisted in the data collection and performed the impact assessment. Wrote the paper with inputs from the co-authors.

# Abbreviations

AD	Anaerobic digestion
CBG	Compressed biomethane
CH <sub>4</sub>	Methane
C	Carbon
CLCA	Consequential life cycle assessment
CO	Carbon monoxide
CO <sub>2</sub>	Carbon dioxide
N	Nitrogen
N <sub>2</sub> O	Nitrous oxide
P	Phosphorus
CHP	Combined heat and power
DM	Dry matter
DME	Dimethyl ether
ER	External energy ratio
EU-28	Member states of the European Union
FU	Functional unit
FTD	Fischer-Tropsch diesel
GHG	Greenhouse gas
GTL	Gas-to-liquid
GWh	Gigawatt hours
GWP	Global warming potential
ha	Hectare
ICBM	Introductory Carbon Balance Model
IPCC	International Panel on Climate Change
ISO	International Organization for Standardization
J	Joule
K	Potassium
km	Kilometre

LBG	Liquefied biomethane
LCA	Life cycle assessment
LHV	Lower heating value
MDEA	Methyldiethanolamine
N <sub>2</sub>	Nitrogen gas
Nm <sup>3</sup>	Normal cubic metres (at 0 °C and 1 bar)
PE	Primary energy
RED	Renewable Energy Directive
RES	Renewable energy source
s	Second
t	Tonne
SRC	Short-rotation coppice
SOC	Soil organic carbon
WC	Water content
WW	Wet weight
v	Volume
yr	Year



# 1 Introduction

Fossil energy has played a strongly positive role in global development, heralding the Industrial Revolution, followed by further technological, economic and social developments. However, annual carbon dioxide (CO<sub>2</sub>) emissions from fossil fuel combustion have increased from near to zero in 1880 to over 33 Gt CO<sub>2</sub> in 2015 (IEA, 2017). This has been accompanied by an average global temperature increase of 0.8 °C, two-thirds of which has occurred since 1975 (NOAA, 2017). An increase in the concentration of greenhouse gases (GHG) in the atmosphere is likely to be the dominant cause of this drastic global warming and climate change (Pachauri *et al.*, 2014). In 2015, around 82% of the world's total primary energy supply was based on fossil fuel sources, which contributed more than half of all total anthropogenic GHG emissions (IEA, 2017).

Overpopulation, overproduction and overconsumption have resulted in the world currently facing major energy supply challenges, such as limited fossil fuel reserves and lack of energy security. In 2016, transport (33.2%), households (25.7%) and industry (25.0%) were the largest fossil fuel consumers in the 28 European Union member states (EU-28) (Eurostat, 2018). The road transport sector is the largest and fastest growing final energy consumer, currently representing a 49.7% share of global oil consumption. Road transport produces 73% of the total GHG emissions from the transport sector (IEA, 2018).

In order to overcome the gap between energy supply and demand, while considering the destructive environmental impacts, renewable energy sources have attracted attention. The main renewable energy sources at present are biomass, hydropower, geothermal, wind and solar. They are all associated with flexible applications, emerging markets, a multitude of stakeholders, global affordability and availability, energy storage opportunities and high environmental and economic credibility (Marshall & Farahbakhsh, 2013; Borenstein, 2012; Del Río & Burguillo, 2008). Among the alternative renewable energy sources, biomass has taken the lead as it is available in different forms, *e.g.* agriculture and forestry materials including plant and animal waste, solid

municipal waste and waste from food processing (Ekpeni *et al.*, 2014). Biomass potentially extracts carbon dioxide from the atmosphere, following which it is further processed through bioenergy conversion, in what is termed the *decarbonisation* effect, which is an important aspect in the climate mitigation effect of bioenergy systems (Van Forest, 2011).

Biomethane is a versatile biomass-derived renewable energy carrier with the ability to produce energy services and high-value products. At present, biomethane is mainly produced by anaerobic digestion of wet biomass (*e.g.* manure, sewage sludge and organic waste). Energy crops such as maize and sugar beet and grasses are other feedstocks for biogas production (Weiland, 2010; Holm-Nielsen *et al.*, 2009). 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. The energy content of biogas can be increased through drying, cleaning and upgrading, resulting in biomethane with >97% CH<sub>4</sub>, which is suitable for injection to the gas grid and use as a transportation fuel. Unconverted biomass from the anaerobic digestion process remains as digestate, which is a nutrient-rich and sustainable soil amendment for biomass growth, replacing chemical fertilisers.

In Sweden, 2.07 TWh of biogas were produced in 2017, of which 65% was upgraded to biomethane for use in the transport sector (Energimyndigheten, 2018). Energy and carbon dioxide tax exemptions are the greatest driver of biogas use in the transport sector in Sweden. However, availability of gas infrastructure is an important factor in biomethane development. Biomethane can be distributed by the natural gas grid or a local biogas grid. In countries such as Sweden with limited gas grid infrastructure, biomethane is road-transported as compressed biomethane (CBG) or liquefied biomethane (LBG). Compressed biomethane, a gaseous fuel, has a low energy density compared with liquid fuels and is a suitable option for dispensing and distributing in the vicinity of a local biogas plant. Liquefying biomethane to LBG increases the energy density, which lowers the cost of distribution and makes it a suitable fuel for longer distances. However, this technology is energy-intensive (Sun *et al.*, 2015). Due to its gaseous nature, biomethane has issues related to fuel dispensing, storage and distribution. Filling stations for CBG and LBG are costly, limited and captive compared with standard liquid filling stations (Åhman, 2010). Regions with limited gas grid infrastructure, storage and distribution systems for biomethane are thus interested in technologies that can convert biomethane to liquid fuel with even higher energy density and more feasible transportability.

Converting biomethane to liquid biofuels would facilitate storage and distribution and enable a supply of biomethane to be offered to broader and larger markets. A novel route of biomethane conversion to vehicle fuel is gas-

to-liquid (GTL) technologies, which offer a means to exploit gaseous energy sources such as fuel, higher hydrocarbons and chemical products (Deshmukh *et al.*, 2010; Dry, 2002). The GTL technologies can potentially result in a product which can be blended with liquid fuels (Sajjad *et al.*, 2014). Existing GTL technologies include conversion of methane from natural gas to syngas, a mixture of carbon monoxide (CO) and hydrogen gas (H<sub>2</sub>) and subsequent synthesis to *e.g.* Fischer-Tropsch diesel (FTD), methanol and dimethyl ether (DME) through catalytic synthesis. Innovations within the GTL technologies, *e.g.* micro-channel technology, have led to improvements in the efficiency of production and in infrastructure. Through micro-channel technology, processes are accelerated by reducing heat and mass transfer distance, which also greatly reduces the size of the processing industry (LeViness *et al.*, 2011; Hu *et al.*, 2005). This technology has the potential to be applied for biomethane.

Apart from conversion to CBG, LBG and GTL fuels, biomethane can be stored and distributed through physical conversion of methane molecules. Gas hydration is a technology that can convert methane gas to clathrate hydrates. Clathrate hydrates of methane are compounds in which the methane molecules are physically trapped within the crystalline structure of frozen host water molecules. This structure is a stable source of methane gas existing under natural conditions of elevated pressure and low temperature, as found in marine sediments and permafrost regions that abound in conventional deposits of natural gas (Sloan & Koh, 2007). There are ongoing studies on using gas hydrate technologies in order to store and distribute biomethane (Budzianowski & Brodacka, 2017), which is an interesting option for use in biomethane systems.

An important barrier to increased biomethane production is the availability of biomass feedstock suitable for anaerobic digestion (Nanou, 2013; Van Foreest, 2011). However, options for producing biomethane through thermal processes (*i.e.* gasification and pyrolysis) are interesting because thermochemical gasification and pyrolysis technologies enable lignocellulosic biomass (*e.g.* short-rotation coppice willow) to be converted into a combustible gas that can be reformed and upgraded to methane. In pyrolysis, the feedstock is heated to between 400 °C and 800 °C under oxygen-limited conditions, resulting in three products: condensable gases (bio-oil), non-condensable gases and biochar (Laird, 2008). Pyrolysis products have a vast range of applications, such as energy services and chemical production (Kan *et al.*, 2016). Biochar is a potential by-product from the pyrolysis reactor that can be used as a soil amendment or heat source (Cao & Pawłowski, 2012).

To date, biomethane deployment has been hindered by issues mainly related to its gaseous nature and low energy content, lack of distribution infrastructure and product diversity. Thus, there is a need to evaluate and assess diversified

and prospective alternatives for biomethane production, conversion and distribution based on potential and regional conditions in a systematic perspective. Increased insights into potential designed systems and their environmental impacts could help decision-makers towards more efficient planning and use of resource and assist in reaching future environmental targets. Life cycle assessment (LCA) is a commonly used methodology in evaluating and assessing the potential environmental impacts of products, processes and services and is currently used in policy making for assessing the climate performance of bioenergy systems and biofuels. However, several methodological aspects related to LCA of bioenergy systems, such as definition of system boundaries, functional unit and impact allocation in a system with multiple products, should be considered carefully during application of the approach.



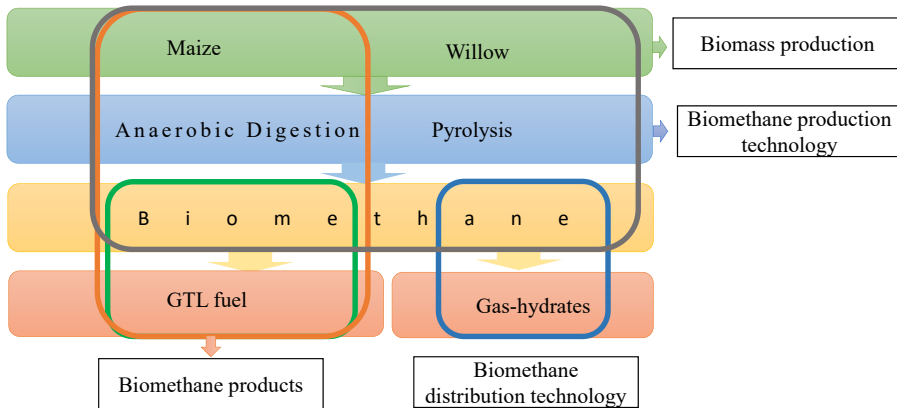
## 2 Aim, objectives and structure of the thesis

### 2.1 Aim and objectives

The aim of this thesis was to provide decision support for development and implementation of future biomethane systems. Specific objectives were to assess the energy performance and climate impact of novel technologies related to biomethane production, conversion of biomethane to high-value products and biomethane distribution in a life cycle perspective. The processes assessed were:

- Production of biomethane through pyrolysis of willow (Paper III).
- Conversion of biomethane by gas-to liquid technologies to Fischer-Tropsch diesel, methanol and dimethyl ether (Papers I & II).
- Distribution of biomethane via gas-hydrates (Paper IV)

In all papers, different scenarios covering prospective approaches were used and the novel technologies were compared against conventional and well-established technologies (Figure 1). The sensitivity of the results to different methodological choices and to changes in the background and foreground systems was also assessed.



- Paper I
- Paper II
- Paper III
- Paper IV

Figure 1. Structure of the work reported in Papers I-IV. Paper III covers the biomass production unit and conversion to biomethane through pyrolysis and AD. Paper I covers potential production of gas-to-liquid (GTL) transportation fuels from biomethane (e.g. Fischer-Tropsch diesel (FTD), methanol and dimethyl ether (DME)). Paper II expands the study boundaries to biomethane production from anaerobic digestion (AD) with maize as feedstock for production of GTL products (e.g. methanol and DME). Paper IV covers biomethane conversion to gas hydrates (e.g. biomethane hydrate, biogas hydrate) as a technology for biomethane distribution.

## 2.2 Structure of the thesis

In the remainder of this thesis, *Chapter 3* provides background to the novel and potential technologies assessed, which relates to *biomethane production*, *biomethane conversion to high-value products* and *biomethane distribution*. In *Chapter 4*, different scenarios and unit processes studied in Papers I-IV are described, as are study boundaries, the methodological approaches used in LCA and the functional units applied. *Chapter 5* presents the results on the energy performance and climate impact of the different system designs and scenarios studied in the thesis. *Chapter 6* provides a discussion on the most important findings in Papers I-IV, while conclusions and suggestions for future research are presented in *Chapter 7*.

## 3 Background

Renewable energy sources have had the highest growth rate of all commercial energy sources, reaching 18% of the total global energy demand in 2017, while the share of fossil fuels has remained stable at around 80% since 2000 (WBA, 2017). National and international environmental policies have an important role in directing the future of technological developments and consumption and production patterns of the global population. The Renewable Energy Directive of the European Commission establishes an overall policy for the production and promotion of renewable energy sources in the EU. Through the Renewable Energy Directive, the EU is committed to meeting at least 20% of its total energy needs with renewables by 2020, to be achieved through individual national targets. All EU countries must also ensure that at least 10% of their transport fuel comes from renewable sources by 2020 (EC, 2009). The Renewable Energy Directive specifies national renewable energy targets for each country, taking into account its starting point and overall potential for renewables. Sweden's national target has been set to 49% of renewable energy in gross final energy by 2020.

### 3.1 Bioenergy and biomethane

The World Energy Council defines bioenergy as energy from organic matter, *i.e.* all materials of biological origin that are not embedded in geological formations (fossilised). It thus includes conventional biomass (*e.g.* forestry and agricultural biomass and residues), modern biomass and biofuels (World Energy Council, 2016; Demirbas, 2009; Goldemberg & Coelho, 2004). From 2000 to 2014, biogas and liquid biofuels showed the highest increase in contribution to total primary energy supply of biomass (11.2% and 15.6%, respectively) (WBA, 2017).

In 2017, global biogas production reached 58.7 billion Nm<sup>3</sup>, which is equal to 352 TWh (WBA, 2017), with half of the production occurring in Europe (170 TWh). Biogas potential in Sweden is estimated to be around 14-17 TWh per year, mostly sourced in agricultural biomass (Linné *et al.*, 2008). Sweden has ambitious targets to have a fossil fuel-independent transport sector by 2030 and to become a fossil fuel-free country by 2045. It is worth noting that the Swedish Government has also decided to separate out and biologically treat at least 50% of the food waste from households, shops and restaurants in order to re-utilise the nutrients (Miljömålen, 2018).

### 3.1.1 Current biomethane conversion technologies

Anaerobic digestion is a series of biological processes in which microorganisms convert biodegradable organic matter in the absence of oxygen. Typical process conditions for anaerobic digestion are a temperature of around 37 °C (mesophilic) or 55 °C (thermophilic) and a pH of approximately 7 (Nanou, 2013). Historically, anaerobic digestion was mainly used as a waste treatment method. Today, it has become a multi-purpose integrated technology in waste treatment, nutrient cycling and production of biogas as a renewable energy source (Mohan *et al.*, 2016; Surendra *et al.*, 2014). Extensive studies have been carried out on anaerobic digestion of energy crops, agricultural wastes, food waste and household waste, in order to achieve higher methane yields.

Digestate, the residue of anaerobic digestion, is used as a nutrient source, which reduces the demand for chemical fertilisers and consequently results in lower GHG emissions (Adams *et al.*, 2015). The digestate, an organic product, also improves soil biochemical activity and the structural stability of agricultural soil, which plays an important role in a variety of processes such as soil aeration, water infiltration, root penetration and soil erosion (Beni *et al.*, 2012; Weiland, 2010; Holm-Nielsen *et al.*, 2009; Møller *et al.*, 2009). Digestate can be dewatered in a solid-liquid separation process in order to increase its nutrient value and availability as a biofertiliser and lower the logistics costs (Czekala, 2017; Whiting & Azapagic, 2014).

In order to use biogas as a vehicle fuel or for injection to the gas grid, it must be purified and upgraded. Through purification, gas contaminants (*e.g.* water vapour (H<sub>2</sub>O), hydrogen sulphide (H<sub>2</sub>S) and particles) are removed. Biogas is upgraded by increasing the methane content through removal of carbon dioxide, resulting in an increased energy content. Different upgrading technologies are used for the removal of carbon dioxide from the biogas stream. These are classified based on absorption, adsorption and membrane separation (Bauer *et al.*, 2013). Most common upgrading technologies are based on absorption,

performed in either a water scrubber or a chemical scrubber. The water scrubber, which is currently most widely used (Thrän *et al.*, 2014), is based on the principle that carbon dioxide has higher solubility in water than methane. Carbon dioxide is separated from water in a desorption column for re-use of the water in the absorption column. The chemical scrubber applies a chemical solution that absorbs and chemically binds to the carbon dioxide molecules. In the next stage, the chemical solution is regenerated by heating the chemical solution, releasing the carbon dioxide as a gas.

### 3.1.2 Current biomethane use and distribution technologies

In 2017, 65% of the biogas produced in Sweden was upgraded to biomethane for transportation fuel, 19% was used for heat and 3% was used for electricity (Energimyndigheten, 2018). The share of biofuels in the transportation sector increased by 19% between 2000 and 2016. The main biofuel in the transport sector was biodiesel, followed by bioethanol and biogas, representing 86%, 7% and 7% of the total, respectively (Energimyndigheten, 2018). Today, more than 70% of the methane used as vehicle fuel is biomethane and less than 30% is natural gas. The main barriers to biogas implementation in Sweden are the limited gas infrastructure and storage and distribution systems. In regions with limited gas grids, biomethane is transported mainly as compressed biomethane (CBG).

Compressed biomethane is stored in steel or composite cylinders under a pressure of 200 bar at ambient temperature. The cylinders have a capacity of 2000 Nm<sup>3</sup> biomethane and are transported in trucks with swap bodies that can accommodate a total of 4500 Nm<sup>3</sup> gas (Hjort & Tamm, 2012). Biomethane is also road-transported as LBG, where biomethane is cooled to -161 °C and liquefied by the closed Brayton cycle technology, using nitrogen gas (N<sub>2</sub>) as the refrigerant fluid. Liquefied biomethane is stored in a vacuum-insulated vessel and delivered by semi-trailer with a tank capacity of 33 000 Nm<sup>3</sup>. Biomethane fuel is supplied as CBG at high pressure (> 250 bar), low pressure and as LBG (Åhman, 2010).

## 3.2 Novel technologies of biomethane

### 3.2.1 Biomethane production technology: Pyrolysis

Pyrolysis of organic material has the flexibility to generate varying proportions of different gaseous, liquid and solid products, based on varying operating

parameters such as temperature or heating rate (Kan *et al.*, 2016). Slow pyrolysis occurs under lower process temperatures (300-550 °C) and longer hot-vapour residence times (minutes or hours) favour the production of solids (*e.g.* charcoal), while fast pyrolysis at higher temperatures (300-1000 °C) and shorter residence times (< 2 s) increases the yield of liquids (Jouhara *et al.*, 2018). Essential features of the pyrolysis feedstock are particle size typically less than 5 mm and moisture content less than 10%.

Based on the feedstock preparation process, many types of raw material can be used, including lignocellulosic material and industrial and domestic residues. The pyrolysis reactor represents only 10-15% of the total system energy use and costs, while the main costs and energy use are related to logistical operations such as biomass delivery, storage and handling, drying, grinding, product collection, storage and upgrading (Palz *et al.*, 2015). Conversion of pyrolysis products to syngas has been examined in a number of studies (Kan *et al.*, 2016; Yue *et al.*, 2016). Further synthesis of biomethane via gas reforming and methanisation of syngas has been studied by Görling *et al.* (2013) and Larsson *et al.* (2013).

Pyrolysis is considered a key component of the European Commission's R&D programme, with the main aim of producing an environmentally friendly and cost-efficient liquid fuel (Faaij, 2006). Pyrolysis seems more promising in that regard than other routes such as gasification, due to its higher energy performance, shorter reaction time, lower infrastructure requirement and suitability for a large variety of feedstocks, from industrial waste to lignocellulosic material.

### 3.2.2 Biomethane conversion to high-value products: GTL technologies

Syngas production through steam reforming of biomethane to carbon monoxide and hydrogen offers the possibility to produce several top-quality synthesis fuels, such as Fischer-Tropsch synthetic diesel, methanol, DME *etc.*

The GTL technologies are based on a group of chemical reactions for the production of synthetic fuels from syngas. The Fischer-Tropsch process is one of several catalytic processes carried out on syngas for production of mainly FTD, olefins, waxes, LPG, kerosene, naphtha *etc.* Methanol synthesis is one of the GTL group reactions for the production of methanol and further processing to DME, ethylene, propylene, oxygenates, gasoline and other advanced fuels. Recent developments in micro-channel technology for GTL production have led to great improvements in production efficiency and infrastructure establishment (Jin *et al.*, 2016; LeViness *et al.*, 2011; Cao *et al.*, 2005). The main characteristic of micro-channel technology is parallel arrays of micro-channels, with typical

diameter in the 0.1-5.0 mm range. Through reduction of heat and mass transfer distance, reaction processes can be accelerated and system volumes can be reduced 10-fold or more compared with the conventional hardware (LeViness *et al.*, 2011; Tonkovich *et al.*, 2008; Hu *et al.*, 2005).

Small-scale GTL technology can provide future possibilities for converting biogas from anaerobic digestion to liquid fuels, which would facilitate product distribution and increase flexibility of use. Furthermore, the hydrogen in the syngas could be used for generating ammonia-nitrogen (NH<sub>3</sub>-N) through the Haber-Bosch process. Ammonia is an important platform chemical and a precursor to nitrogen fertilisers. Syngas manufacture from biomethane through steam reforming produces H<sub>2</sub>, which together with nitrogen from the air, act as the starting elements for the production of ammonia from renewable energy sources (Ahlgren, 2008).

### 3.2.3 Biomethane distribution technology: Biogas/biomethane hydrates

Selection of an appropriate distribution technology for biomethane has a significant impact on the overall efficiency of the biomethane system (Budzianowski & Brodacka, 2017). Apart from compressing biomethane (*i.e.* CBG) and liquefying it (LBG), biomethane can be stored and transported through physical conversion to methane hydrate molecules.

Formation of methane hydrate requires a pressure of around 3-10 MPa and temperature usually needs to be between -15 and -32 °C (Budzianowski & Brodacka, 2017). Gas hydrates are stable sources of gas existing under natural conditions of elevated pressure and low temperature in marine sediments and permafrost regions (Sloan & Koh, 2007). This justifies the use of gas hydrates for biomethane distribution under Swedish conditions of cold climate and lack of grid infrastructure. A given volume of gas hydrates at high pressure and low temperature contains more than 150 times the volume of the same gas at standard temperature and atmospheric pressure, indicating that gas hydrates are a promising material for gas storage (Veluswamy *et al.*, 2018; Siazik *et al.*, 2017; Mori, 2003). Introducing methane hydrate into the logistics chain between the producer and consumer of biomethane would result in three main process stages: formation of hydrates, storage of hydrates and disassociation 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.

### 3.3 Life cycle assessment studies

#### 3.3.1 Life cycle assessment (LCA)

Life cycle assessment is a standardised analytical methodology that investigates and assesses the environmental impacts of a product or process throughout its entire life cycle. The assessment is based on reliability and transparency in defining the goal and scopes of the LCA, data gathering (inventory analysis), assessment of potential impacts (impact assessment) and interpretation, which are the four main steps of LCA (Figure 2). The standards for the different steps of an LCA are provided by the International Organization for Standardization (ISO), in ISO 14040 (2006) and 14044 (2006). The general approach in an LCA is to consider the broader environmental implications of a product or process by aiming to account for resources used and emissions from the whole life cycle, in order to have a more complete assessment of the overall impacts or to compare the different environmental impacts of different systems, products and services.

The four main steps of LCA (goal and scope definition, inventory analysis, impact assessment, interpretation) are applied in an iterative process, meaning that all four are dependent and influenced by each step. The first step of the LCA is to specify a goal and scope. During the inventory step, a life cycle model is constructed and calculations are made for the emissions produced and resources consumed. The data collected in the inventory phase are related to various environmental impacts in the impact assessment phase. Throughout the process, interpretations are made and finally potential impacts can be identified (Baumann & Tillman, 2004).

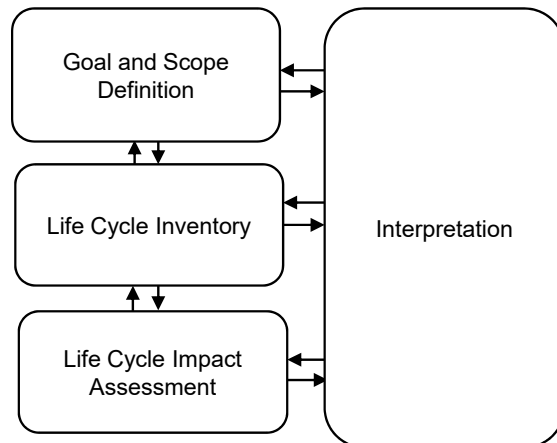


Figure 2. The four steps in life cycle assessment (LCA), based on standards ISO 14040 and ISO 14044.



In order to compare the different environmental impacts in an LCA study, a functional unit (FU) to which all resources and emissions are related is established. Products and processes in the system studied have to fulfil the chosen functional unit in order to be considered equivalent.

Many processes contribute to more than one product, as multi-function processes or intertwined product systems. Moreover, waste or co-products of a product system may act as raw material to another system. The environmental impacts of the system must then be allocated fairly to all these multiple products. The allocation problem is a challenging fundamental problem in LCA methodology, as it plays an important role in the final results and assessment. Based on the ISO standard, allocation should be avoided by dividing the process into sub-processes or by including additional functional units. Several allocation techniques and procedures have been developed, such as partitioning the input and/or output flows of a process in relation to the economic or physical properties of the product system under study.

### 3.3.2 Life cycle perspective

A life cycle is the life scenario of a product, process or service including phases of *raw material extraction, manufacturing, use* and *end-of-life* (ISO, 2006), also called ‘cradle-to-grave’ study. Gathering and preparing large amounts of data for quantification of environmental impacts for the different phases of a life cycle study is time- and resource-consuming. A solution is to simplify the process. A simplified life cycle assessment (LCA) is an efficient tool in evaluating and assessing the environmental impacts and its attributes to a product, process or service in the initial product development stages (Graedel & Saxton, 2002). 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 service or product system that could be neglected during the analysis, without significantly affecting the overall results, leads to a reduced amount of data (Pelton & Smith, 2015). For instance in the case of comparison of different biofuels from cellulosic feedstock, since the biofuel production process has a major influence on the overall results of the system, it is possible to exclude other phases in a simplified comparable assessment, although the reliability of the results is more limited (Pigosso & Sousa, 2011).

A complete life cycle study helps identify the hotspots and areas of improvement for better environmental performance. Hotspot identification aids systems and decision-makers in avoiding and lowering environmental impacts. Use of a life cycle perspective can avoid sub-optimising the environmental

performance of a system and increase understanding of the trade-offs between different study units (Tasala Gradin, 2016; Finnveden *et al.*, 2009; Hur *et al.*, 2005; Graedel & Saxton, 2002).

### 3.3.3 Life cycle assessment of bioenergy systems

Life cycle assessment is a commonly used tool to assess the environmental impacts of biofuels and bioenergy systems. However, the use of different input data, functional units, allocation methods and reference systems, along with indirect effects such as land use change and nitrogen-based soil emissions, may complicate the overall picture (Cherubini & Strømman, 2011).

Many bioenergy systems produce more than one product in the same production plant and the additional product/s can be categorised either as co-product or by-product. Co-production situations in the bioenergy sector can be handled through system expansion or allocation. The conventional solution to co-product handling in a LCA study is allocation, in which the environmental impact of the production process is distributed over the multiple products, based on a chosen allocation key (*e.g.* the mass, energy content or economic value of the co-products). The core problem in co-product allocation is the difficulty in finding an objective allocation key and justifying the choice of this allocation key. Therefore, several allocation methods can be considered in an LCA study to examine the sensitivity of results to this methodological choice (Tufvesson *et al.*, 2013; Rehl *et al.*, 2012).

The alternative to allocation is system expansion, which considers a situation of expanding the product system to include the additional functions related to the co-products (ISO 14044). In other words, the production system is credited for displacing production of the co-products in alternative systems and the impacts are subtracted from the bioenergy system. The basic concepts in system expansion are most easily understood when considering by-product handling in a system. According to ISO 14041, system expansion provides more realistic modelling of the impact of a product-related system (Weidema & Norris, 2002).

It should be emphasised that LCA aspects such as functional unit, system boundaries and reference system can change the bioenergy results. The choice of functional unit is based on the study goal. For instance, the EU Renewable Energy Directive uses the energy content of biofuels (MJ) as its functional unit, while many studies include the distance travelled by vehicles (km) or the agricultural land used in biofuel feedstock production. The use of such a wide array of functional units makes comparisons of LCA studies challenging. In addition, a comparative LCA could best support decision-making by being

spatially explicit, in order to address regional characteristics and differences (Langfitt, 2017; Choudhary *et al.*, 2014).

### 3.4 LCA case studies of biomethane

Many LCA studies have been carried out on biogas energy systems, mostly based on anaerobic digestion of different feedstocks for heat and electricity production (*e.g.* Zhang *et al.*, 2016; Sawatdeenarunat *et al.*, 2015; Börjesson & Mattiasson, 2008). Many studies have also been conducted on sustainability measures and environmental assessments (Bacenetti *et al.*, 2014; Gissén *et al.*, 2014). To date, LCA has been the most commonly used approach in studying the environmental impacts of agricultural biogas plants, as it enables illustration of these impacts and highlights potential mitigation strategies (Bacenetti *et al.*, 2016; Hijazi *et al.*, 2016; Styles *et al.*, 2015; Evangelisti *et al.*, 2014; Tonini *et al.*, 2012; Patterson *et al.*, 2011). However, a major challenge to the credibility of different LCA studies is the discrepancy in outcomes due to different methodological choices and limitations (Bacenetti *et al.*, 2016; Styles *et al.*, 2015). For example, considerations relating to land use change and multi-functionality handling have large impacts on the results of different studies (Notarnicola *et al.*, 2017).

LCA studies of the novel technologies proposed within the context of this thesis have not to the authors knowledge been previously performed.

## 4 Methodological approach

This chapter starts with an overall description of the different scenarios, followed by the scope and methodological choices related to the LCAs performed on the different scenarios in Papers I-IV. The methodology used to calculate the energy performance and climate impact assessment is presented in detail. This is followed by a detailed description of the unit processes.

### 4.1 Description of scenarios studied in this thesis

The scenarios studied in Papers I-IV are shown in Figure 3. Paper III assessed alternative biomethane production by transition of maize cropping for anaerobic digestion with willow cropping for pyrolysis. Biomethane production at the maize anaerobic digestion plant and at the willow pyrolysis plant corresponded to 149 and 113 GWh yr<sup>-1</sup>, respectively. The climate impact of biochar from pyrolysis used as a carbon sequestration source (by adding biochar to soil) or for generation of energy services (biochar to energy) was also investigated.

Paper I studied production of three GTL fuels (FTD, methanol and DME) and two conventional biofuels (CBG and LBG) as reference scenarios based on biogas production in a relatively large biogas plant (60 GWh yr<sup>-1</sup>). The production chain from upgrading of raw biogas to conversion in engines was also studied.

Paper II investigated production of methanol, DME and ammonia as biofuels or platform chemical from anaerobic digestion of maize, with combined heat and power (CHP) production as the reference scenario. The assessment was based on a biogas production plant producing 100 GWh yr<sup>-1</sup>, which corresponds to Sweden's largest anaerobic digestion facility. All stages from biomass production until production of end products were included.

Paper IV studied technologies for biomethane distribution from a small (local) biogas plant with a production capacity of 2 GWh yr<sup>-1</sup>. Transport of gas

hydrates was assessed in comparison with biomethane transport as compressed biomethane (CBG).

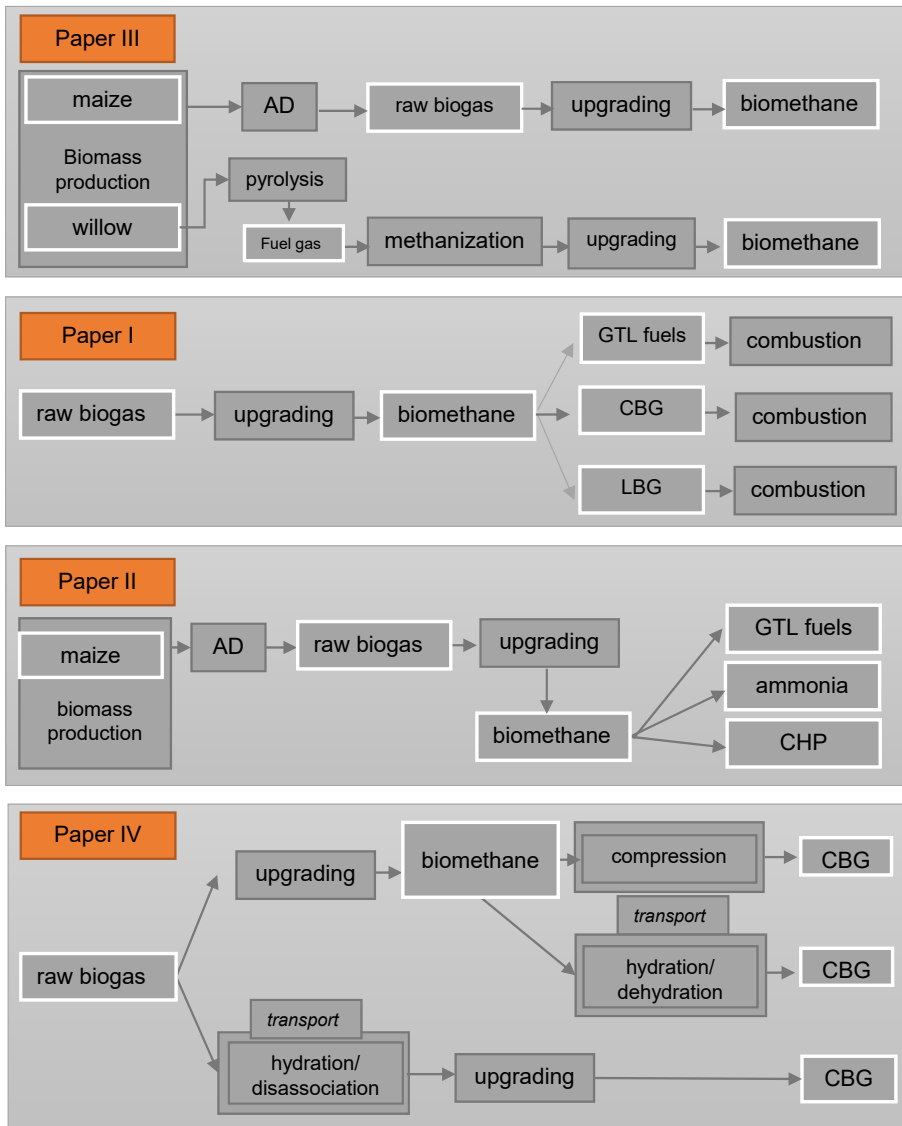


Figure 3. Scenarios studied in Papers I-IV, which included potential biomethane production technologies (pyrolysis), biomethane products (gas-to-liquid (GTL) fuels) and biomethane storage and distribution methods (*i.e.* biomethane hydrate and biogas hydrate). CBG = compressed biomethane, LBG = liquefied biomethane, AD = anaerobic digestion, CHP = combined heat and power plant.

## 4.2 Scope and methodological choices related to LCA

Life cycle assessment was used to assess the energy performance and climate impact of novel and potential technologies in systems for *biomethane production* (Papers III), *biomethane conversion to high-value products* (Paper I and II) and *biomethane distribution* (Paper IV). Energy performance and climate impact related to the different unit processes included in the studies, along with by-product handling through allocation and system expansion and the functional units used both in the papers and in the thesis are described.

### 4.2.1 Energy performance

Energy performance of the bioenergy systems examined in this thesis was assessed as the external energy ratio (*ER*) according to Murphy *et al.* (2011), which is defined as the ratio between the delivered energy service (*E<sub>out</sub>*) and the total external energy input (*E<sub>in</sub>*) used to generate the energy service excluding the energy in the feedstock biomass (Equation 1). The external energy inputs include the total upstream primary energy input and the direct energy used in the operations of the system.

$$ER = \frac{E_{out}}{E_{in}} \quad (1)$$

The fraction of the biomass feedstock used within the bioenergy system (*e.g.* for drying, heat generation), losses (*e.g.* methane and heat) and by-products (external heat and steam, biochar) were excluded from *E<sub>out</sub>*. Inputs related to the production of capital goods such as machinery and buildings were not included in the calculations, as it was assumed that this would have only slight effects on the overall results (Bauer *et al.*, 2007).

### 4.2.2 Climate impact

As a climate impact indicator, global warming potential (GWP) was assessed in different systems (Papers I-IV) using the IPCC methodology (Myhre *et al.*, 2013). The assessment was limited to the impact of three major greenhouse gases (GHGs) contributing to global warming: carbon dioxide (CO<sub>2</sub>), nitrous oxide (N<sub>2</sub>O) and methane (CH<sub>4</sub>). The GHG emissions recorded were:

- Upstream emissions related to inputs from energy services (*e.g.* electricity, heat, fuel) and products (*e.g.* chemicals and mineral fertiliser production)

- Downstream emissions related to fuel combustion
- Emissions as losses through technical conversion and storage
- Digestate storage management and application to soil
- Soil nitrous oxide (N<sub>2</sub>O) emissions due to crop fertilisation and nutrient application
- Emissions from soil organic carbon (SOC) stock changes

#### *Biogenic soil emissions of N<sub>2</sub>O*

Application of mineral fertilisers and digestate is a significant source of N<sub>2</sub>O emissions. Direct pathways include microbial nitrification and denitrification of fertiliser and manure nitrogen in agricultural soils or animal waste management systems. Indirect pathways involve nitrogen losses from agricultural soil and animal waste management systems via volatilisation, leaching, runoff or harvest of crop biomass. In Papers II and III, N<sub>2</sub>O emissions, as the result of mineral fertiliser and digestate application, were calculated based on IPCC (2006) guidelines.

#### *Biogenic carbon fluxes*

In Paper III, the carbon dioxide fluxes between the atmosphere and the biosphere were modelled and included in the assessment of the climate impact. The biosphere was divided into three different pools: soil organic carbon (SOC), digestate and biochar (Figure 4). The carbon stocks of each pool were studied through the use of different models.

The introductory carbon balance model (ICBM) (Andr n *et al.*, 2004) was used to calculate the annual carbon dioxide flux related to soil organic carbon and inputs from digestate application.

Biochar produced from the pyrolysis process was applied to soil with the potential to create a carbon sink. Mineralisation of carbon from the biochar sink to carbon dioxide was calculated annually for the willow pyrolysis scenario over the course of the study described in Paper III. Biochar carbon losses after a given period of time were calculated using an equation presented by Zimmerman (2010).

Dead biomass was not considered, since live biomass was either used for biomethane production in the energy conversion facilities (*i.e.* anaerobic digestion, pyrolysis) or used as input to the soil organic carbon or biochar pools.

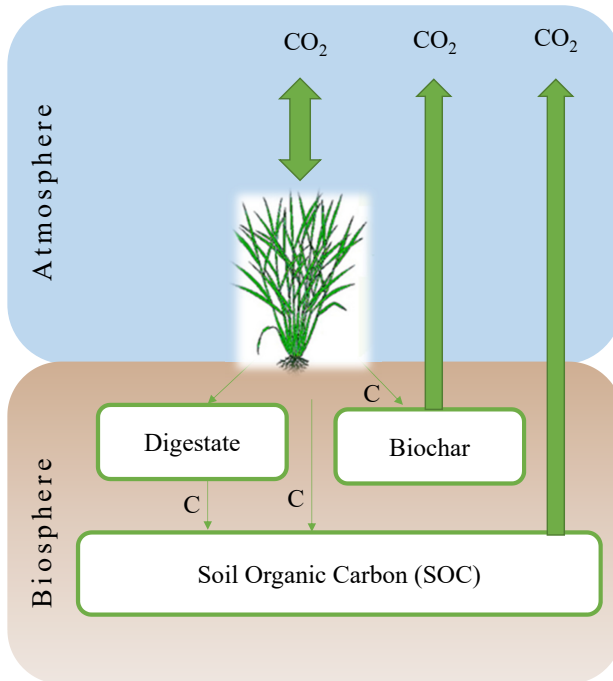


Figure 4. Carbon (C) flows between different pools within the biosphere and carbon dioxide ( $\text{CO}_2$ ) fluxes between the biosphere and atmosphere, which were modelled in Paper III.

#### 4.2.3 Allocation and system expansion

Multi-functionality issues, which arise in bioenergy systems generating more than one product, were handled by allocating the climate impact and primary energy inputs to all products (Papers I, III and IV), and system expansion (Papers II and III).

In Papers I and IV, the bioenergy system resulted in multiple products, *i.e.* biofuels or chemicals, heat and steam. Physical allocation was based on the lower heating value (LHV) of the products. In Paper I, the sensitivity of the results to choice of allocation methodology was examined based on equivalent electricity produced or power generation efficiency, which has previously been used to represent the overall exergy, since LHV does not take the exergy of the various energy sources into account (Tunå *et al.*, 2012; Andersson *et al.*, 2006).

In Paper II, the climate impact mitigation prospect from implementation of the potential products from different scenarios studied was quantified by considering complete substitution by fossil alternatives.



In Paper III, physical allocation and system expansion were both tested, to investigate the effect of different approaches on the outcome of the study. Allocation was performed in the willow scenario based on the LHV of the biomethane. The climate impact mitigation prospect from implementation of biomethane, biochar and heat produced from the scenarios studied was also quantified, by considering complete substitution of fossil alternatives. The system expansion was performed by assuming that the exported heat replaced heat from a natural gas combined heat and power (CHP) plant and that the biochar replaced hard coal in industrial processes.

#### 4.2.4 Functional units

Different functional units (FU) were used in the papers. In Papers I and IV, an input-based FU of 1 Nm<sup>3</sup> of raw biogas was used to assess alternative transportation fuels (Paper I) and 1 Nm<sup>3</sup> of biomethane was used to assess alternative distribution methods for biomethane (Paper IV). In Papers II and III, one hectare of land under cultivation (*e.g.* maize and willow) was chosen as the FU, in order to study the biomass production process and land use efficiency. In Paper III, a second FU based on energy (GJ) of delivered product was used in order to compare the biomethane output from the studied scenarios with its fossil-based counterparts. In Chapter 5 of this thesis, the overall results are presented and discussed based on a FU of 1 Nm<sup>3</sup> of biomethane.

### 4.3 Description of unit processes

In this section, different unit processes are described. Biomass cropping system including crop cultivation and harvest operations, along with nutrient (*e.g.* fertiliser, digestate) and biochar application to soil are described in sub-section 4.3.1 *Biomass cropping systems*. Pre-treatment of biomass, conversion facilities of AD and pyrolysis along with biomethane upgrading technologies are described in sub-section 4.3.2 *Biomass conversion to biomethane*. Conversion technologies of biomethane for syngas production and synthesis of GTL products and ammonia are described in sub-section 4.3.3 *Biomethane conversion to high-value products*. The hydration of biogas and biomethane as an alternative distribution technology is described in sub-section 4.3.4 *Hydrates of Biogas/biomethane*. Finally, transport between different unit processes is described in sub-section 4.3.5 *Transport*.

#### 4.3.1 Biomass cropping systems

*Maize cultivation:* Energy inputs and related GHG emissions from all operations in maize cultivation and management were studied, including seed preparation, soil preparation, weed control, sowing, herbicide production and application, fertiliser production and application, harvesting, field transport, transport of biomass to the energy conversion facility and return transport of liquid and solid digestate and application to the field.

Growing maize (*Zea mays*) as a feedstock for biomethane production through anaerobic digestion was studied in Papers II and III. Tillage operations for the maize crop, harvesting and transport were modelled. The total amounts of nitrogen (N), phosphorus (P) and potassium (K) applied to the annual maize plantation were 149, 26 and 119 kg per hectare, respectively, which included the nutrients in the mineral fertiliser and digestate applied (Papers II and III). Herbicides and pesticides were applied during the growing stages when required. Maize was harvested during autumn by a whole-crop chopper (30% dry matter (DM)) and transported to the energy conversion facility by truck, where it was ensiled. Losses from the storage and during transport were assumed to be 15% of DM.

The operations studied during maize cultivation were identical in Papers II and III, with the exceptions of yield and the level of digestate and nutrients applied.

*Willow plantation:* The energy inputs and related GHG emissions from all operations in willow cultivation and management were studied, including soil preparation, physical and chemical weed control, seedling production and plantation, application of herbicides and fertiliser production and application, full-stem harvest, transport to a corner of the field, temporary storage (< 7 days), transport to the energy conversion facility and return of the biochar by-product to soil. It was assumed that the biochar was applied annually to the soil under another plantation.

Growing willow (*Salix* spp.) on agricultural land as a feedstock for biomethane production through pyrolysis was studied in Paper III. A complete willow plantation managed in two rotations of 22 years was considered, with a three-year coppicing cycle and a fallow year in between, resulting in a total study period of 45 years.

In each rotation, one year before willow establishment the soil was assumed to be prepared mechanically by weed harrowing/ploughing and chemically by application of pesticides. Herbicides were applied prior to ploughing and during the establishment year. Willow seedlings were planted in late spring. The plants

were cut after the first year of growth to promote production of multiple stems. Subsequent harvesting was performed during winter, when the soil generally had higher carrying capacity, using a whole-stem harvester. After each harvest, the willow stools were left to re-grow (re-sprout) in the following spring, and harvested again after three years. The stems were assumed to be cut and stored in bundles near the field for natural drying. During storage, the moisture content was assumed to be reduced to 20%, resulting in an increased net calorific value. Dry matter losses during the first six months of storage were estimated to be  $0.3 \text{ t ha}^{-1} \text{ yr}^{-1}$ . In the final year in each rotation, the remaining roots and stools were removed to prepare the soil for the new rotation. Fertilisation was performed using general mineral fertilisers. Phosphorus and potassium were applied in the second year of every cycle, and nitrogen was applied annually starting from the second cycle. The amount of nitrogen applied was based on willow yield, so that equal amounts were added with fertiliser and removed by harvest.

Willow yield was set to  $20 \text{ t DM ha}^{-1} \text{ yr}^{-1}$  for the first harvest and  $30 \text{ t DM ha}^{-1} \text{ yr}^{-1}$  for the subsequent years. The average annual yield at full production was considered to be  $9 \text{ t DM ha}^{-1} \text{ yr}^{-1}$  with an average moisture concentration of 48%. The first coppicing cycle of each rotation yielded two-thirds of the total yield of the rotation.

#### 4.3.2 Biomass conversion to biomethane

*Anaerobic digestion* In Papers II and III, anaerobic digestion of maize was studied for biomethane production. Maize was assumed to be ensiled before maceration and homogenisation of the biomass, in order to make the biomass more accessible to microorganisms and improve the biogas yield. Thereafter biomass was pumped into a biogas reactor operated as a wet fermentation process under mesophilic conditions. The main product from the anaerobic digestion process was raw biogas (~60%  $\text{CH}_4$ ), which was upgraded to biomethane (97%  $\text{CH}_4$ ) in an upgrading unit. In Papers I and IV, biogas production through anaerobic digestion was a common unit in all the routes studied and therefore it was excluded from the analysis.

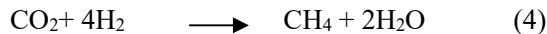
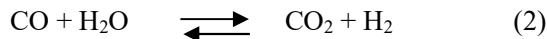
*Upgrading technologies* In Papers I-IV, biogas was upgraded to biomethane (97%  $\text{CH}_4$ ). The two upgrading technologies included in Papers I-IV were the water scrubber and chemical/amine scrubber. Water scrubbing is the most common upgrading technology applied and is based on  $\text{CO}_2$  separation from raw biogas through dissolving into water under high pressure, normally 6-10 bar (Bauer *et al.*, 2013). A chemical scrubber is based on an activated methyldiethanolamine (MDEA) system, with an electricity requirement of 0.12-

0.14 kWh/Nm<sup>3</sup><sub>biomethane</sub> and a heat requirement of 0.55 kWh/Nm<sup>3</sup><sub>biomethane</sub>. The choice of the upgrading technology in Papers I-IV was based on the availability of heat energy sources.

*Digestate* All nitrogen, phosphorus and potassium in the feedstock was assumed to end up in the digestate, which was applied as an organic fertiliser to maize crops, resulting in a reduced need for mineral fertilisers in crop production. The digestate was phase-separated into solid and liquid fractions for efficient management and application to field.

*Pyrolysis and methanisation* Before the pyrolysis reactor unit, the willow feedstock was assumed to be further reduced in size to 3 mm by comminution and dried to a moisture content of 7% in order to increase the heating rate and reduce the reaction time. Biomass was pyrolysed in a bubbling fluidised bed reactor with external heating and vapour recirculation to generate fuel gas and biochar. The heat of pyrolysis was supplied by burning 12% of the total biomass input.

After the pyrolysis gas was cleared of solid particles and sulphur by adsorption filters, it was assumed to be converted to biomethane in a fuel synthesis step. The fuel synthesis step included pre-reforming of long hydrocarbon chains into mainly CO<sub>2</sub>, CO, CH<sub>4</sub> and H<sub>2</sub> in the presence of a nickel catalyst (at 500 °C, atmospheric pressure) in an adiabatic fixed bed reactor. This was assumed to be followed by a combined water-gas shift (Eqs. 2 and 3) and a methanisation reaction (Eq. 4), where CO and H<sub>2</sub> were converted to CH<sub>4</sub> in the presence of a nickel catalyst (at 300 °C and 10 bar) in an isothermal reactor. After the fuel synthesis, the gas contained mostly CH<sub>4</sub>, CO<sub>2</sub>, H<sub>2</sub>O and traces of CO and H<sub>2</sub>. A water scrubber was assumed to be used to upgrade the gas to vehicle fuel quality (97% CH<sub>4</sub>). The pyrolysis plant was designed for maximum fuel gas production, which means that all condensable and non-condensable gases from the pyrolysis reactor were converted to fuel gas.



*Biochar* Biochar, the by-product of the pyrolysis reaction, was assumed to be quenched by adding water in order to avoid spontaneous ignition. The biochar was then transported to the field and applied to soil using a lime spreader, to act

as a carbon sequestration agent and potential soil structure improver. In another case, biochar was transported to an industrial unit where it replaced coal as the energy source.

#### 4.3.3 Biomethane conversion to high-value products

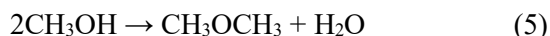
In Papers I and II, biomethane ( $\text{CH}_4$ ) was converted to syngas ( $\text{CO} + \text{H}_2$ ) by steam reforming. The steam reformer was assumed to be externally heated by burning part of the feed (biomethane) in a combustion reactor. The syngas produced by steam reforming contains  $\text{H}_2$ ,  $\text{CO}$ ,  $\text{CO}_2$ ,  $\text{N}_2$  and  $\text{H}_2\text{O}$  (water vapour). A water-gas-shift reactor and  $\text{CO}_2$  removal unit was included in order to clean and extensively condition the biomethane to meet the specifications of catalytic synthesis processes for further product (*e.g.* chemicals and fuels) synthesis. The transformation of syngas to GTL products was conducted in micro-channel based reactors.

*Fischer-Tropsch diesel (FTD) production (Paper I)* Biomethane was converted to FTD in three main steps: *Fischer-Tropsch synthesis, hydrocracking and reforming*. In the Fischer-Tropsch synthesis unit, syngas was converted into liquid hydrocarbons suitable for the manufacture of fuels and chemicals. Fischer-Tropsch diesel was then produced from syngas in a pressure- and temperature-controlled reactor. In addition to the diesel fuel fraction, the process yields wax, kerosene, fuel gas and naphtha. Wax was assumed to be cracked down in order to increase the fuel yield. Flue gas was combusted in order to provide the process with heat and energy. The Fischer-Tropsch reactor was modelled in Paper I as an isothermal plug with varying length, operating at 250 °C, and catalysed by iron (Fe) and cobalt (Co). During a refinery stage, low-pressure steam and heat were produced. The hydrocarbons, which are heavier than diesel fuels, are good feedstock for a cracker to produce more vehicle fuel. Part of the tail-gas produced in the hydrocracking was burnt to produce heat and power for the plant. The remaining tail-gas was assumed to be recirculated to the reformer.

*Methanol production (Papers I and II)* Methanol production was simulated using a steam-raising type of reactor based on micro-channel technology. Synthesis of methanol is highly exothermic, occurring over a catalyst bed at moderate temperatures. The methanol reactor was cooled by boiling feed water. The product stream was condensed and methanol and water were removed in a reforming unit supplied by energy from the methanol synthesis. The gas was recycled to the inlet of the reactor except for a small part that was removed as

tail-gas. Methanol and water were distilled to yield 99.9% pure methanol output (Lundgren *et al.*, 2013).

*Dimethyl ether (DME) production (Papers I and II)* The methanol synthesis described above was considered an input to DME synthesis. Methanol was dehydrated in the presence of a catalyst, resulting in the production of DME (Eq. 5).



The product was cooled, and methanol, DME and water were separated in a two-step distillation process. Methanol was recycled back to the reactor inlet. The purge stream was burnt to produce heat and power. The simulations were based on pressurised storage of DME, requiring no energy for storage.

*Ammonia synthesis (Paper II)* Synthesis of ammonia ( $\text{NH}_3$ ) was conducted over an iron catalyst at pressures around 100-250 bar and temperatures of 350-550 °C . The process is exothermic, resulting in high-pressure steam. Since the conversion efficiency of ammonia is low (20-30%) the unreacted gas is circulated while ammonia is separated by condensation. Syngas was assumed to be compressed and directed to an ammonia synthesis unit, where the hydrogen reacts in the presence of an iron catalyst with nitrogen, derived from process air (100-250 bar and 350-550 °C) to form anhydrous liquid ammonia in what is known as the Haber-Bosch process (Eq. 6).



It was assumed that the heat released was used in the district heating grid. The final product was assumed to be refrigerated and stored at low pressure (Ahlgren *et al.*, 2008).

#### 4.3.4 Hydrates of biogas/biomethane

Modelling biogas/biomethane hydrates was performed in Paper IV and compared with compressed biomethane (CBG) as the conventional route of biomethane transport. The hydrate formation process was divided into compression work, gas cooling, water cooling, hydrate formation, and pumping and mixing. The formation of hydrate was performed as gas bubbled into a continuous water phase. The biogas and biomethane was supplied with a pressure of 3 MPa and 4 MPa, respectively. The power for cooling the water

entering together the hydrate formation reactor with the gas was correspondingly calculated from an inlet temperature of 15 °C to 2 °C and a heat exchanger. A compression chiller was assumed for the gas and water cooling.

The power for cooling the hydrate to a storage temperature of -30 °C was calculated for a heat exchanger. 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 transferred to and stored in gastight freezing containers, assumed to consist 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.*, 2009), leading to a weight capacity of 10 tons of hydrate per container.

The heat needed for hydrate dissociation at the centralised facility was based on melting the hydrate from -10 °C and the dissociation enthalpy. The compression heat pump used in the biogas hydrate scenario was the same as in the CBG scenario, including recovery of heat from the subsequent compression of biogas to 200 bar.

The upgrading of biogas in the biomethane hydrate and biogas hydrate scenario was assumed to use the same technique as in the CBG scenario. In the CBG and biomethane hydrate scenarios, biogas was upgraded in a small-scale decentralised water scrubber with a lower pressure requirement and lower energy demand than the conventional water scrubber in the biogas hydrate scenario.

#### 4.3.5 Transport

Transport was constrained to the biomass supply chain logistics to the energy conversion facilities and the distribution of biomethane products to the final use point. In this thesis, all transport was assumed to be carried out with diesel-powered trucks.

*Transport of biomass, digestate and biochar (Papers II and III)* Transport included delivery of biomass to the energy conversion facility and delivery of digestate (Papers II and III) and biochar (Paper III) from the conversion plant back to the field. In both cases, delivery of inputs (*i.e.* fertilisers and chemicals) to the crop cultivation site was included in the biomass supply chain. Transportation work and distance were calculated using the model described in Overend (1982). The assumptions related to the road tortuosity factor and available land were similar in both Papers II and III. Digestate and biochar were assumed to have the same transport distance.

*Transport of biomethane products (Papers I and IV)* In Papers I and IV, transport included the delivery of biomethane products (*i.e.* GTL products, CBG, LBG, biomethane/biogas hydrates) to a final use point. Different forms of transport were assumed based on the physical and chemical characteristics of the products. In both papers, the distance travelled was calculated based on Hjort and Tamm (2012). The assumptions were based on the existing benchmarks for biomethane transport as CBG and LBG. Calculations of fuel consumption for the distribution of the fuel were based on data from NTM (2006) and Berggren (1999).



## 5 Results

The results from the different studies are presented in three main sub-chapters: biomethane production technology (Paper III), biomethane conversion to high-value products (Papers I and II) and gas hydrates for biomethane distribution (Paper IV). Results in all cases are presented for a functional unit of 1 Nm<sup>3</sup> of biomethane.

### 5.1 Biomethane production technology (Paper III)

Pyrolysis as a potential technology for biomass conversion to biomethane was studied in comparison with anaerobic digestion, a well-established technology, in Paper III. It was assumed that a maize plantation for supplying biomass to anaerobic digestion for biomethane production, used as the reference scenario, was replaced by willow plantation for biomethane production via pyrolysis. The pyrolysis reactor produced biochar as a by-product. This section presents energy performance and climate impact results for the scenarios described in Paper III.

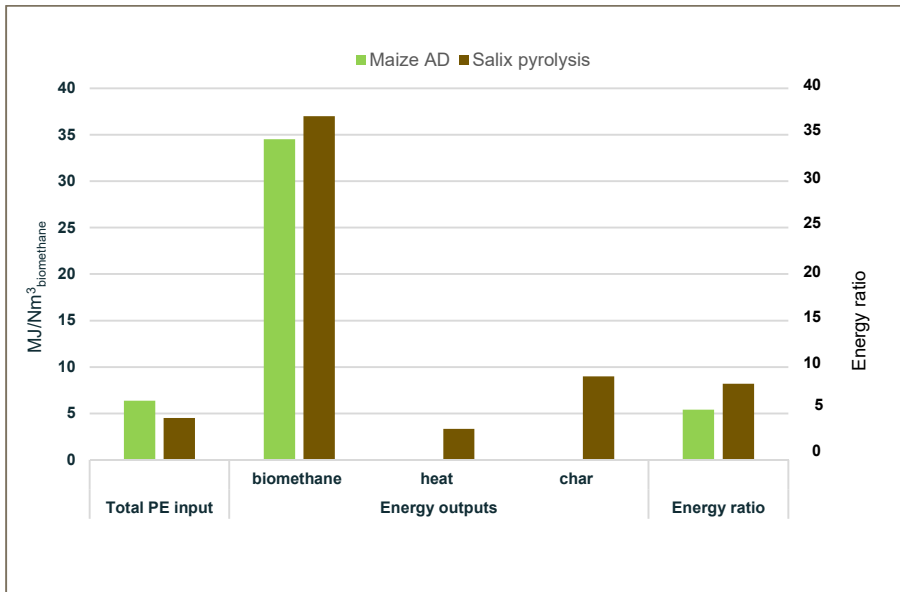


Figure 5. Primary energy (PE) inputs, outputs (*biomethane, heat, biochar*) and external energy ratio of maize anaerobic digestion (AD) and willow pyrolysis (Paper III).

The energy performance (Figure 5) and climate impact (Figure 6) showed that biomethane production based on willow pyrolysis was competitive relative to maize anaerobic digestion. The pyrolysis process had a higher external energy ratio (ER = 8) than anaerobic digestion (ER = 5) for biomethane production. The main reason for the higher external energy ratio of willow pyrolysis was the lower primary energy inputs (Figure 5) and efficient heat recovery in the pyrolysis plant. Primary energy inputs to the agricultural operations for supply of willow and maize as feedstocks were approximately similar. However, the energy inputs to field preparation operations and chemicals (*i.e.* herbicides and pesticides) were higher for maize, while the fertiliser inputs were lower due to the by-product digestate application to crops, although the digestate by-product handling operations were energy-intensive.

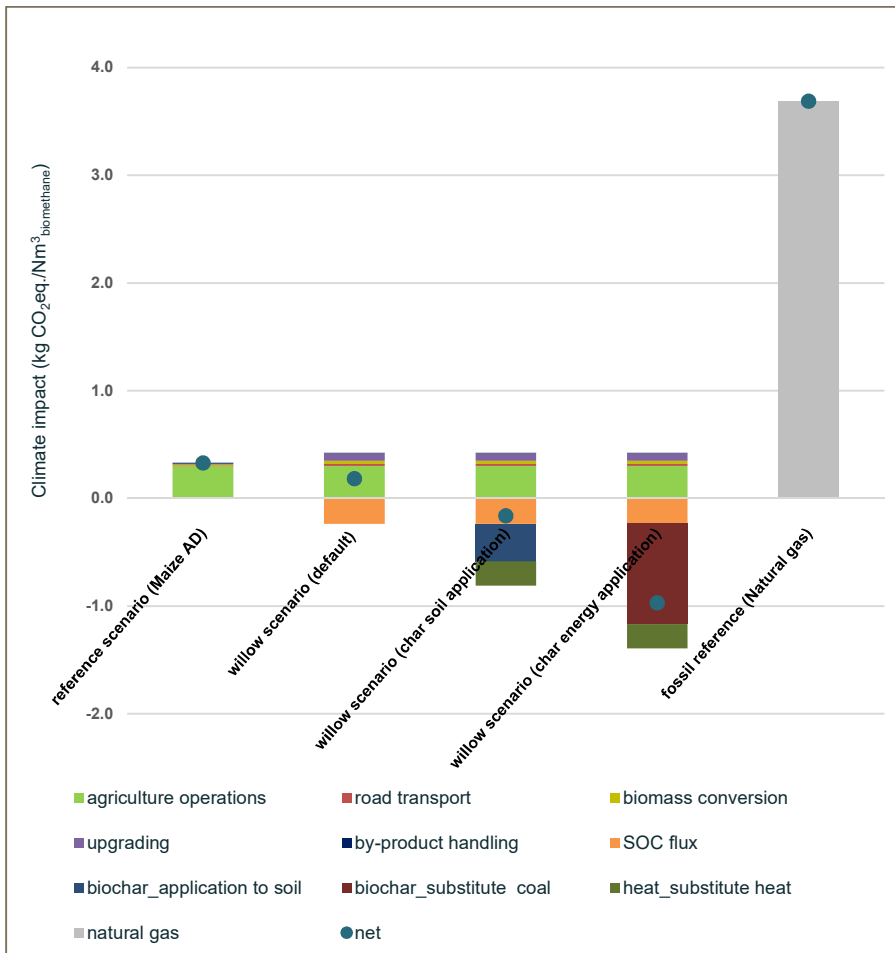


Figure 6. Climate impact ( $kg\ CO_2eq./Nm^3_{biomethane}$ ) of biomethane production via anaerobic digestion (AD) of maize (reference scenario) and pyrolysis of willow. Different cases for the willow scenario were assumed based on biochar handling; no biochar application, biochar application as soil amendment and biochar application as energy source. Natural gas is presented as a fossil fuel reference (Paper III).

The climate impact of the default willow pyrolysis scenario ( $0.2\ kg\ CO_2eq./Nm^3_{biomethane}$ ) was lower than that of the maize anaerobic digestion scenario ( $0.3\ kg\ CO_2eq./Nm^3_{biomethane}$ ), due to lower methane losses during biomass conversion to biomethane and the upgrading step (Figure 6). The change in land use, as willow plantation replaced previous annual maize cropping associated with different management practices, resulted in an increase in the soil organic carbon (SOC) pool. No SOC stock changes were considered from the digestate contribution to soil in the maize anaerobic digestion scenario, since the SOC was assumed to have reached steady state before the start of the

study period. Replacing willow plantation resulted in an increase in the SOC pool from the initial level of 130 t C/ha to 155 t C/ha. Use of biochar as a soil amendment and a source of carbon sequestration with low decay rates resulted in a climate mitigation effect from the willow pyrolysis scenario of  $-0.2 \text{ kg CO}_2\text{eq./Nm}^3_{\text{biomethane}}$ . However, the best climate mitigation effect ( $-1.0 \text{ kg CO}_2\text{eq./Nm}^3_{\text{biomethane}}$ ) occurred when the biochar from the pyrolysis plant was used as a source for heat, substituting mainly coal, in industries. The climate impact for the equivalent amount of natural gas compared with the biomethane produced in the maize anaerobic digestion and willow scenarios was  $4 \text{ kg CO}_2\text{eq./Nm}^3_{\text{biomethane}}$ . (Figure 6).

## 5.2 Biomethane conversion to high-value products (Papers I and II)

Three GTL fuels (FTD, methanol and DME) were studied as potential high-value products from biomethane conversion in Papers I and II. Potential products were compared with conventional uses of biomethane as biofuels (*i.e.* CBG and LBG) in Paper I, and with ammonia and heat and power production (*i.e.* CHP) in Paper II. Biogas conversion to products included the process of biogas ( $\sim 60\% \text{ CH}_4$ ) upgrading to biomethane ( $97\% \text{ CH}_4$ ) and the fuel synthesis process, presented here per FU of  $1 \text{ Nm}^3$  biomethane. The process ended with biogas being transformed to the GTL products. There was substantial co-production of heat and steam in the various scenarios, while the primary energy input and GHG emissions were allocated values relative to the LHV of the main products (*i.e.* GTL products).

The primary energy inputs, outputs and the energy ratio of the biogas conversion process to GTL products are presented in Figure 7. The GTL fuels methanol and DME had relatively high external energy ratio (5.6 and 5.5, respectively) compared with FTD (2.4). In addition, Fischer-Tropsch synthesis results in high levels of steam and heat production, as by-products, in the refinery stage, which reduces the fuel output.

Product yield, defined as the GTL product output (MJ) compared with the input biogas (MJ), varied between the different systems. Both DME and methanol had a relatively high product yield, corresponding to 84% and 71% of the input biogas, respectively, while FTD had the lowest product yield, corresponding to 40% of the input biogas (Figure 7).

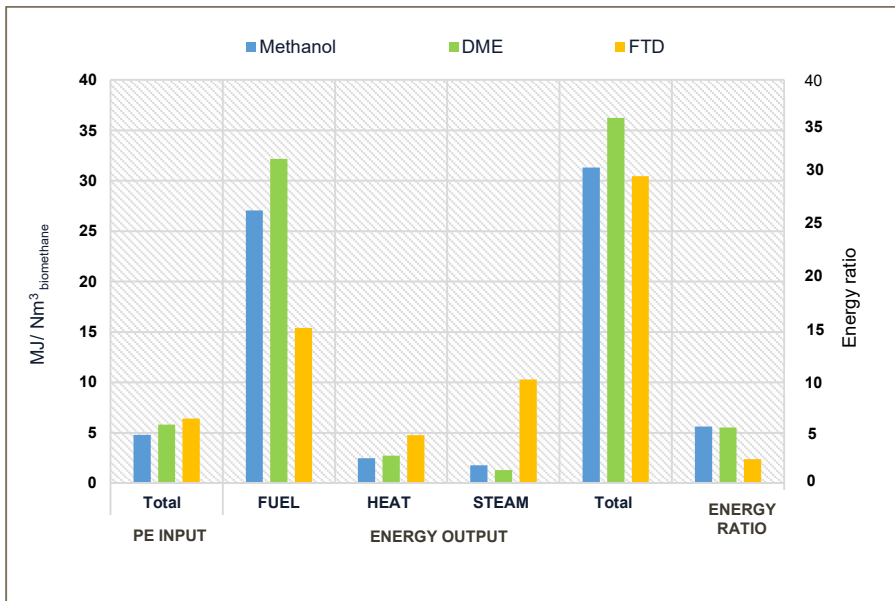


Figure 7. Total primary energy (PE) inputs, energy outputs (*gas-to-liquid (GTL) product, heat, steam and total output*) and external energy ratio of the biogas conversion to GTL products (DME = dimethyl ether, FTD = Fischer-Tropsch diesel) (Papers I and II).

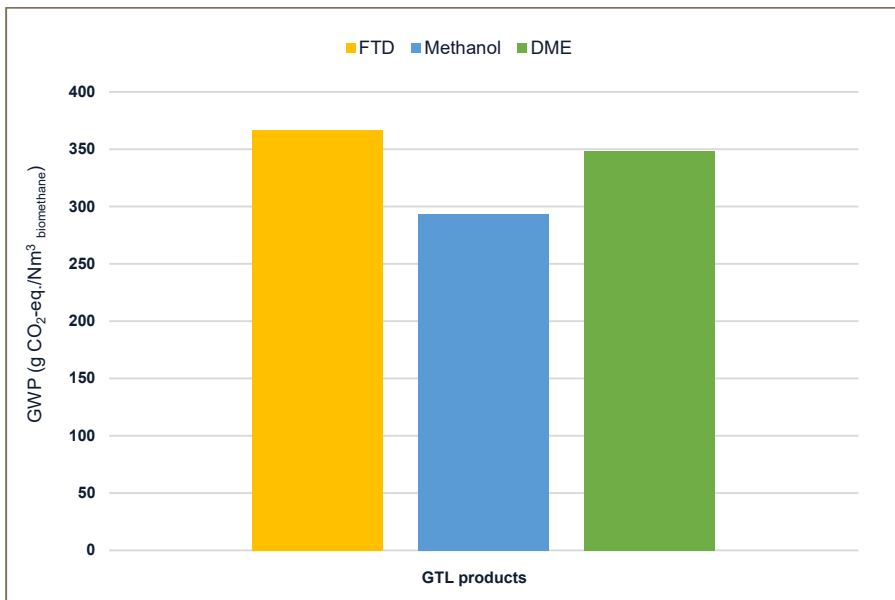


Figure 8. Climate impact (global warming potential (GWP),  $g\ CO_2\text{-eq./Nm}^3\ \text{biomethane}$ ) from biogas conversion to gas-to-liquid (GTL) products (FTD = Fischer-Tropsch diesel, DME = dimethyl ether) (Papers I and II).

Climate impact assessment results for the GTL products studied in Papers I and II are presented per FU of  $1 \text{ Nm}^3_{\text{biomethane}}$  in Figure 8. In the calculations, GHG emissions were allocated to the LHV of the GTL products. As can be seen, FTD had the highest climate impact among the GTL fuels studied, which was due to due to high energy inputs and gas losses during fuel synthesis.

### 5.2.1 Biogas conversion to GTL fuels (Paper I)

Paper I assessed the primary energy inputs and climate impact of different unit processes of biogas conversion to GTL products as biofuels (*i.e.* FTD, methanol and DME) and conventional biomethane-based biofuels (*i.e.* CBG and LBG). The unit processes included upgrading of biogas to biomethane, the fuel synthesis phase (*i.e.* compression, liquefaction, syngas and fuel synthesis), transport to fuelling station and the engine fuelling phase.

Gas-to-liquid fuels had lower total primary energy inputs than CBG and LBG (Figure 9). The fuel synthesis step for GTL fuels demanded high electricity inputs, especially for FTD due to catalytic reactions in reducing long hydrocarbons in the fuel synthesis and hydrocracking step.

Heat generation during the GTL fuel synthesis was recirculated and used for the biogas upgrading by an amine scrubber. The amine scrubber demanded lower electricity inputs ( $0.4\text{-}1.0 \text{ MJ/Nm}^3_{\text{biomethane}}$ ) in the upgrading stage of GTL fuels compared with the water scrubbers in the CBG and LBG scenarios. Water scrubbers operate under high pressure levels, which demands high electricity input ( $0.9\text{-}1.0 \text{ MJ/Nm}^3_{\text{biomethane}}$ ) in comparison with amine scrubbers.

The highest energy inputs among the biofuels studied were related to LBG production. In the LBG scenario, gas upgrading was performed by water scrubber along with an extra  $\text{CO}_2$  polishing step and liquefaction, which led to the highest energy inputs among the different systems studied. In the CBG scenario, upgrading and compression represented the largest share of primary energy inputs. Transport and vehicle fuelling made a small contribution to the overall primary energy inputs in the different scenarios studied in Paper I. Energy input to CBG transport was highest among all biofuels studied, as a result of low gas density and a high proportion of steel in comparison with the amount of gas transported. Filling stations for CBG and LBG were energy-intensive, since they were assumed to dispense biomethane under high pressure ( $>250 \text{ bar}$ ) (Figure 9).

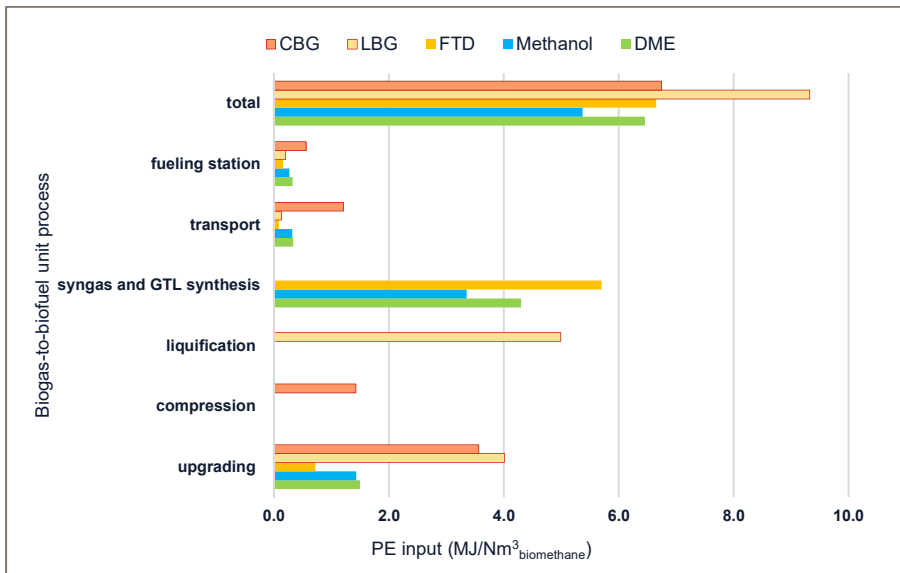


Figure 9. Primary energy (PE) inputs ( $MJ/Nm^3$  biomethane) to the biogas-to-biofuel unit processes (CBG = compressed biomethane, LBG = liquefied biomethane, FTD = Fischer-Tropsch diesel, DME = dimethyl ether, GTL = gas-to-liquid) (Paper I).

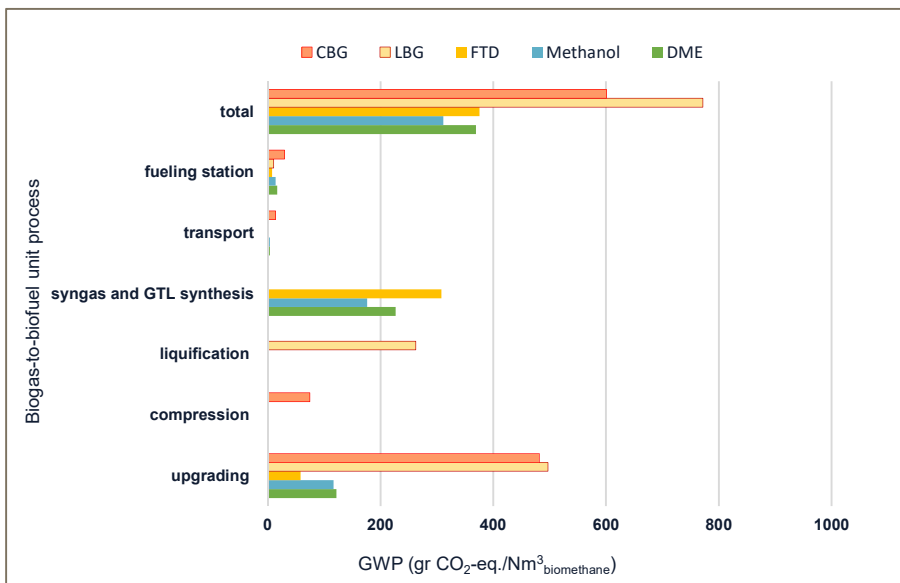


Figure 10. Climate impact ( $g CO_2\text{-eq./Nm}^3$  biomethane) from the biogas to biofuel unit processes (CBG = compressed biomethane, LBG = liquefied biomethane, FTD = Fischer-Tropsch diesel, DME = dimethyl ether, GTL = gas-to-liquid) (Paper I).

Gas-to-liquid fuels had a relatively lower total climate impact in comparison with the other biofuels studied in Paper I (Figure 10). This was mainly due to using the amine scrubber as the upgrading technology. Methane loss levels in the amine scrubber (0.1%) corresponded to 0.2 g CH<sub>4</sub>/kWh<sub>biomethane</sub> in the GTL scenarios, while methane losses in the water scrubber (1%) corresponded to 1.2 g CH<sub>4</sub>/kWh<sub>biomethane</sub> in the CBG and LBG scenarios.

However, the energy and emissions related to the syngas and fuel synthesis step in the GTL scenarios, especially for FTD, were relatively high. There were no significant direct emissions related to the GTL fuel synthesis step except for N<sub>2</sub>O emissions from FTD production (7% of total emissions in the FTD fuel synthesis step).

Apart from the energy and climate performance, fuel yield (output), thermal energy density and distance travelled per unit fuel are of interest in assessing the performance of the fuels studied. Based on their LHV, CBG and LBG had a thermal energy density of approximately 46 MJ/kg, while FTD, methanol and DME had an energy density of 42, 20 and 28 MJ/kg, respectively. However, CBG and LBG had the highest fuel yield (34 MJ/Nm<sup>3</sup><sub>biomethane</sub>) and FTD had the lowest (15 MJ/Nm<sup>3</sup><sub>biomethane</sub>).

Comparison of the distance travelled by the biofuels from the different systems showed that DME-fuelled engines travelled the longest distance per functional unit of 1 Nm<sup>3</sup> of biomethane, whereas engines fuelled by methanol and FTD travelled the shortest distance, due to the low fuel output from the FTD system (Figure 11). The energy and climate performance of using the studied biofuels to drive a certain distance (1 km) (Paper I) are presented in Figures 12 and 13. Comparison of primary energy input per km distance travelled by different fuels indicated that DME performed best, due to relatively high fuel output and high thermal energy density, and that CBG- and LBG-fuelled engines travelled a similar distance. Methanol had the highest primary energy input per km distance travelled. FTD and LBG also demand high energy inputs per distance travelled, mainly due to their energy-intensive production processes (Figure 12).



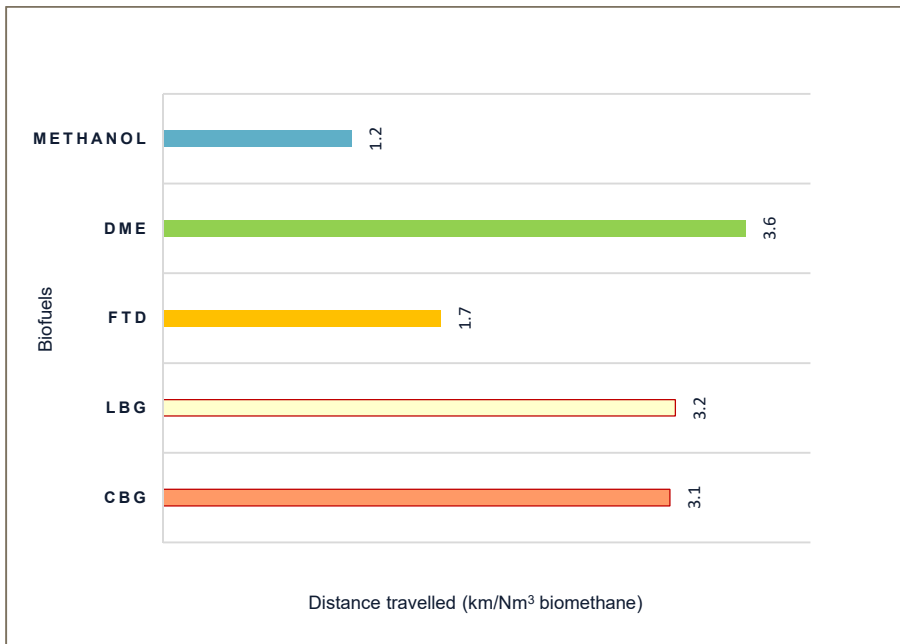


Figure 11. Distance travelled ( $km/Nm^3_{biomethane}$ ) by studied biofuels produced from biogas (CBG = compressed biomethane, LBG = liquefied biomethane, FTD = Fischer-Tropsch diesel, DME = dimethyl ether) (Paper I).

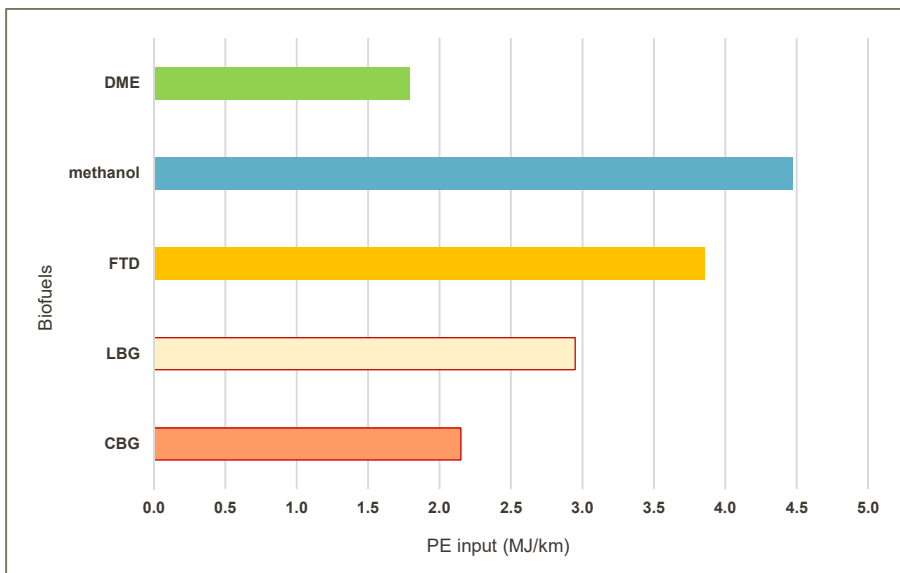


Figure 12. Primary energy (PE) inputs (MJ) per unit distance (km) travelled by different biofuels studied in Paper I (CBG = compressed biomethane, LBG = liquefied biomethane, FTD = Fischer-Tropsch diesel, DME = dimethyl ether).

Figure 13 shows the climate impact related to distance travelled (g CO<sub>2</sub>-eq/km) of different fuels studied in Paper I, including GHG emissions related to the production process. The GHG emissions from the fuel combustion phase were considered climate-neutral, as they are of biogenic origin. As can be seen from Figure 13, DME had the best performance in terms of climate impact per km distance travelled, including the GHG emissions through biofuel production and emissions related to engine combustion. Methanol had the highest GHG emissions per km distance travelled, due to its low energy density. Fischer-Tropsch diesel is a high energy density fuel but, due to the low fuel outputs from the production stage, the ultimate climate impact was higher. Overall, taking into account the energy performance and climate impacts of input biomethane conversion to biofuels and biofuel performance, DME showed the best results among the different fuel conversion scenarios studied.

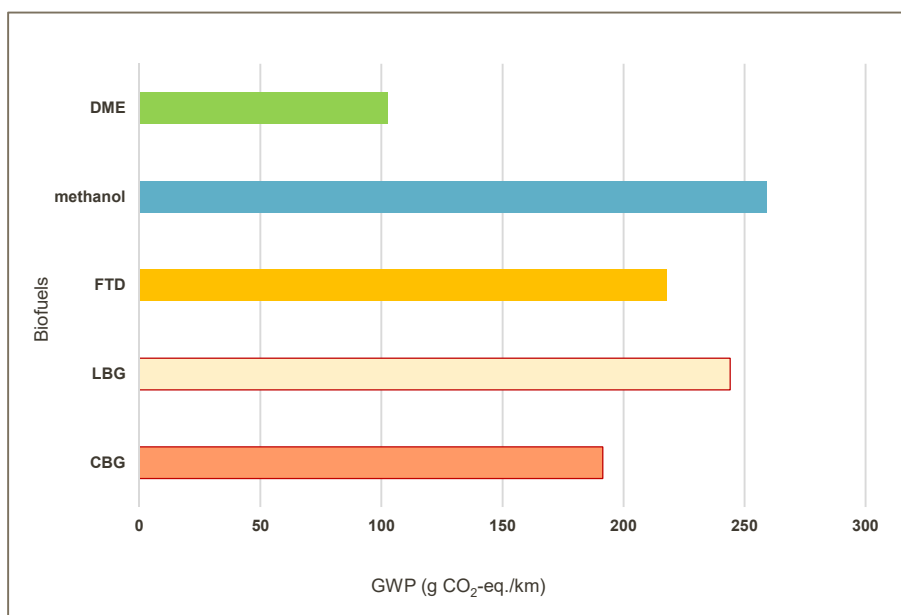
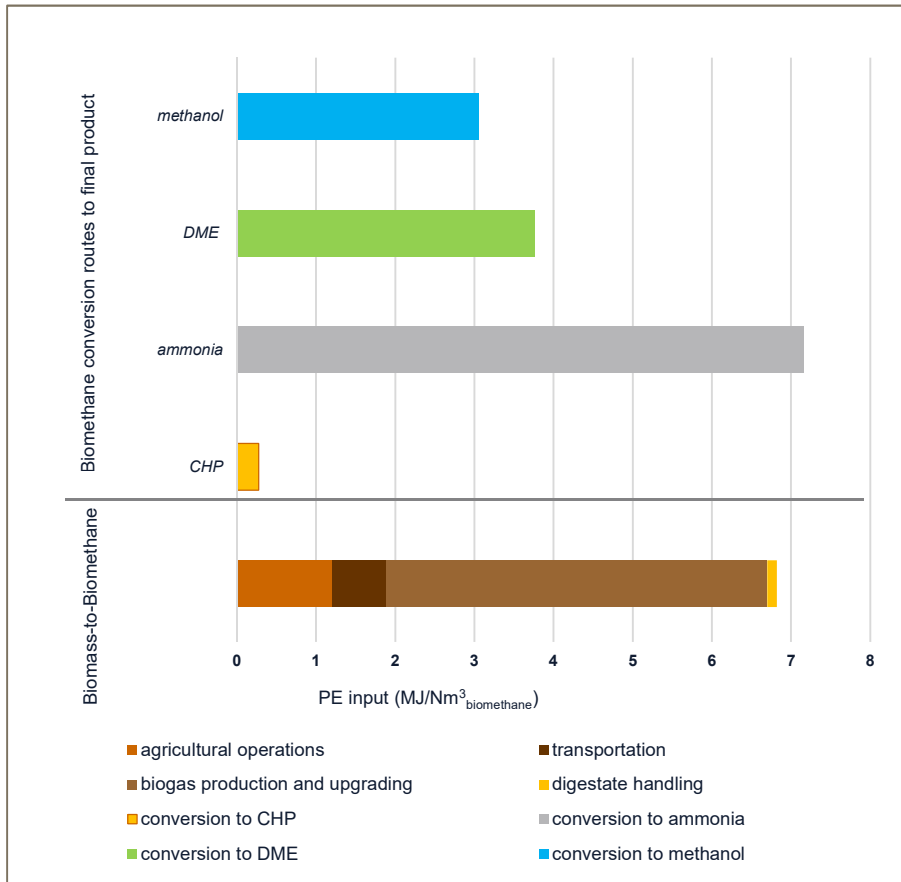


Figure 13. Climate impact (global warming potential (GWP), g CO<sub>2</sub>-eq.) in distance (km) travelled by biofuels studied in Paper I (CBG = compressed biomethane, LBG = liquefied biomethane, FTD = Fischer-Tropsch diesel, DME = dimethyl ether).

### 5.2.2 Biomass conversion to GTL fuels and ammonia (Paper II)

In Paper II, conversion of biogas to DME, methanol and ammonia was compared with electricity production in a CHP unit as the reference scenario. It was assumed that biomethane for all conversion routes was produced from anaerobic

digestion of maize and then upgraded. The output biomethane was converted to products in different process routes (see *Chapter 4, Figure 3*). Digestate from the anaerobic digester was applied to the maize plantation to partly replace chemical fertilisers.



*Figure 14.* Primary energy (PE) inputs ( $MJ/Nm^3$  biomethane) to the different unit processes of biomass-to-biomethane conversion (*i.e.* agricultural operations, transportation, biogas production and upgrading and digestate handling) and the different routes for biomethane conversion to products (*i.e.* diemethyl ether (DME), methanol and ammonia and electricity production in a combined heat and power (CHP) unit) (Paper II).

Results from Paper II showed that the biomass-to-biomethane supply chain, *i.e.* biomass production, transport of biomass and digestate, anaerobic digestion, upgrading to biomethane and digestate handling operations, made a large contribution to both the total energy inputs (Figure 14) and climate impact

(Figure 15) of all routes studied. Combined heat and power was the best route for biomethane conversion, supporting findings in previous studies (e.g. Goehner *et al.*, 2013). The CHP production process had the lowest energy requirement, mainly related to operating equipment, with no considerable climate impact. The DME and methanol conversion routes had approximately similar energy inputs and climate impacts, mainly through energy and emissions related to electricity demand by the syngas reformer.

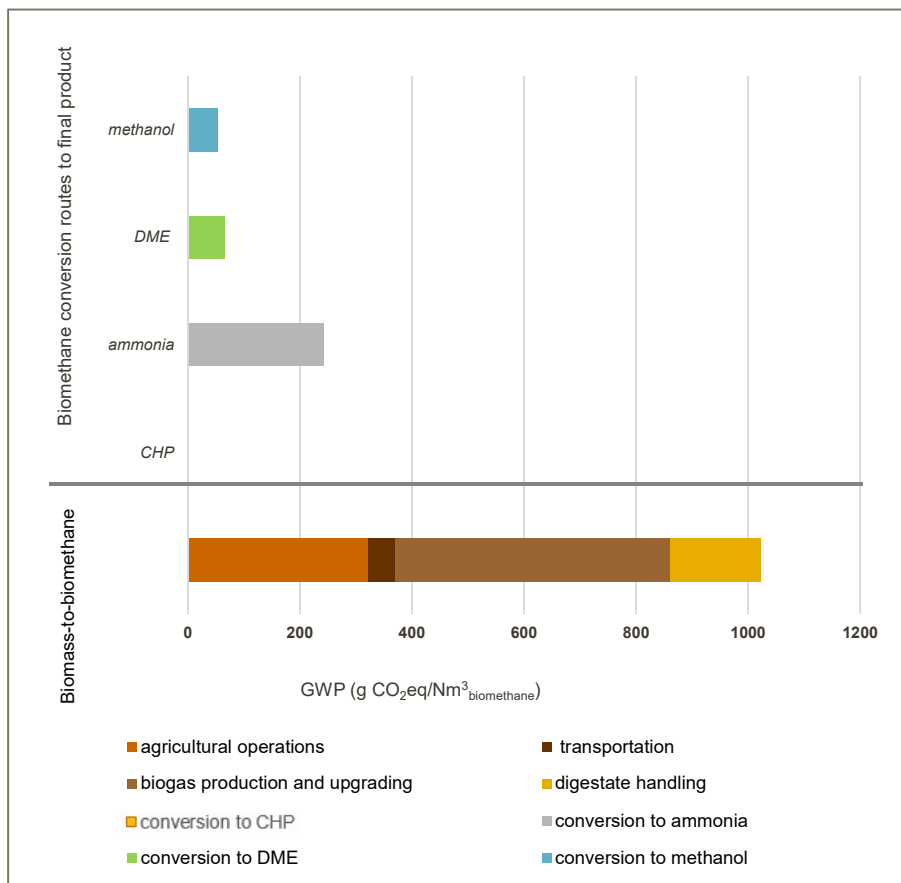


Figure 15. Climate impact ( $g\ CO_2\text{-eq./Nm}^3\text{ biomethane}$ ) of the different unit processes of biomass-to-biomethane conversion (i.e. agricultural operations, transportation, biogas production and upgrading and digestate handling) and the different routes for biomethane conversion to products (i.e. dimethyl ether (DME), methanol, ammonia and electricity production in a combined heat and power (CHP) unit) (Paper II).

Paper II did not include the use phase of the different products studied due to their different applications as biofuels, biochemicals and electricity. In order to elaborate the climate impact of the studied products, the net emissions of equivalent fossil fuel substitutes were studied and are presented in Figure 16. The climate impact mitigation prospects for the implementation of bio-based products considering complete substitution of fossil substitutes were quantified. The fossil alternatives for the main products and the by-products (heat and steam) were based on natural gas conversion. The net GHG emissions for the different products were calculated based on the differences in emissions between the biomethane-based production routes and their fossil-based alternatives, on a functional unit (FU) basis. Electricity production in a biomethane-based CHP unit had the highest climate benefits in comparison with fossil substitutes (*i.e.* natural gas-based CHP). While ammonia showed a higher climate benefit than DME and methanol, it should be noted that the total net emissions in the case of the biofuels did not consider the emissions related to product use (*i.e.* engine composition), which is a major contributor to the climate impact of fuels. In other words, emissions of CO<sub>2</sub> in the fossil fuel alternatives occur in different stages. For fossil-based ammonia and CHP, all CO<sub>2</sub> emissions occur in the production phase. For fossil DME and methanol (if used as engine fuels), most CO<sub>2</sub> emissions occur in the use phase. To make a fair comparison, we therefore added the CO<sub>2</sub> emissions for combustion of fossil-based DME and methanol. Based on the results in Paper II, it was concluded that production of ammonia from non-fossil sources is not competitive relative to the alternative of biomethane-based CHP, DME and methanol and their fossil-based substitutes (Figure 16).

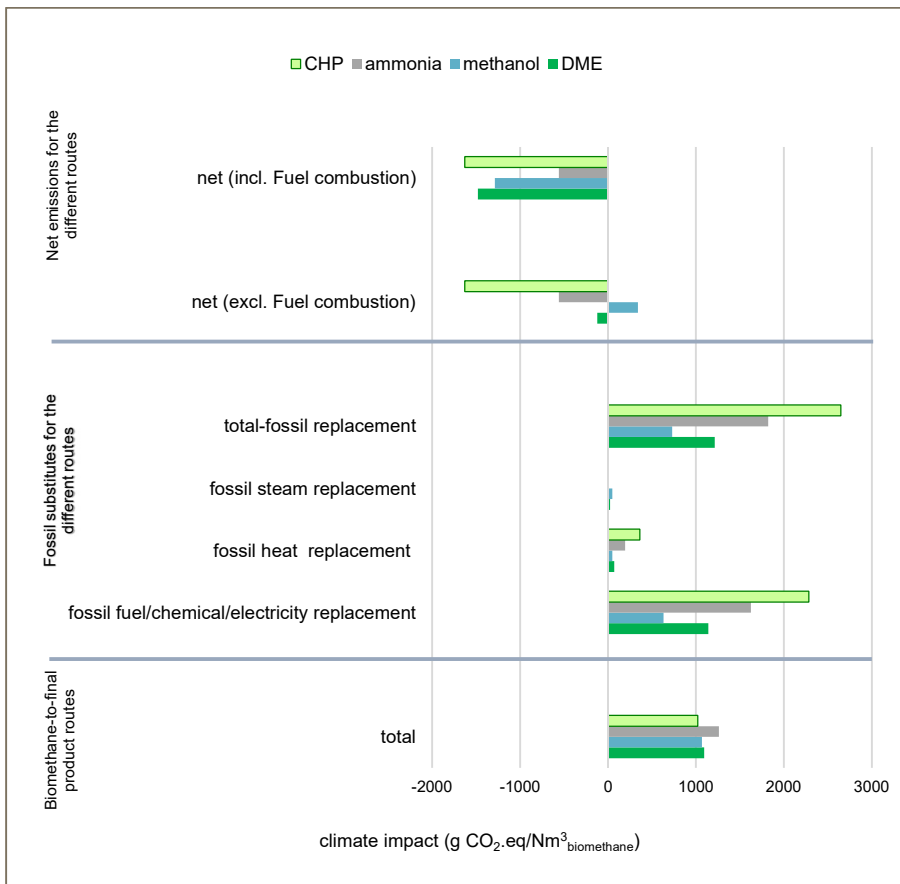


Figure 16. Climate impact (g CO<sub>2</sub>-eq./Nm<sup>3</sup> biomethane) from the different products studied and the fossil substitutes and net greenhouse gas (GHG) emissions for biomass to product conversion. CHP = combined heat and power, DME = dimethyl ether.

### 5.3 Gas hydrates for biogas/biomethane distribution (Paper IV)

Future-oriented scenarios of biomethane distribution in the form of gas hydrates (*i.e.* biogas hydrates and biomethane hydrates) were studied and compared with biomethane distribution in pressurised vessels (*i.e.* CBG). Two scenarios of biogas hydrate and biomethane hydrate were modelled and the energy performance and climate impact were calculated throughout their life cycle, from raw biogas upgrading, hydrate formation and disassociation, and transport to filling station to be used as CBG.

The energy performance of the three scenarios is compared in Figure 17. The output of all scenarios studied consisted of biomethane (97% CH<sub>4</sub>), which in the

hydrate scenarios was slightly lower due to methane losses during gas hydrate formation and dissociation process. The CBG in the base scenario had the highest external energy ratio (16.6), due to the relatively low primary energy input ( $5.8 \text{ MJ}/\text{Nm}^3_{\text{biomethane}}$ ), whereas the biomethane hydrate scenario and biogas hydrate scenario had a much lower external energy ratio (6.6 and 5, respectively) due to their high demand for electricity for pressure increase and temperature reduction. Total primary energy inputs in the biomethane hydrate and biogas hydrate scenario were 15.0 and 22  $\text{MJ}/\text{Nm}^3_{\text{biomethane}}$ , respectively (Figure 17).

In the CBG and biomethane hydrate scenario, biogas was initially upgraded in a decentralised upgrading unit and was further compressed in the CBG scenario and hydrated in the biomethane hydrate scenario. In the biogas hydrate scenario, biogas was preliminarily hydrated and distributed and, after disassociation, was upgraded in a centralised upgrading unit. This centralised biogas upgrading unit had higher primary energy inputs ( $4.0 \text{ MJ}/\text{Nm}^3_{\text{biomethane}}$ ) than the decentralised upgrading unit assumed in the biomethane hydrate and the CBG scenarios ( $2.0 \text{ MJ}/\text{Nm}^3_{\text{biomethane}}$ ). A small-scale decentralised upgrading unit has a lower electricity demand for increasing the pressure in the water scrubber than a large centralised water scrubber. The compression work for 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.

In the hydrate scenarios, thermo-physical conversion of biogas/biomethane to hydrates required high pressure and relatively low temperatures. The primary energy input to hydrate formation and dissociation in the biogas hydrate scenario was  $14 \text{ MJ}/\text{Nm}^3_{\text{biomethane}}$  (62.5% of total PE input), while the corresponding primary energy input for the biomethane hydrate scenario was  $8.6 \text{ MJ}/\text{Nm}^3_{\text{biomethane}}$  (56% of total PE input). The cooling process, which included a compression chiller for the formation of hydrates, and the heating for dissociation of hydrates with a heat pump, was the most energy-demanding operation in the hydrate scenarios, making up around 30% of the total primary energy input. The electricity demand for biomethane compression in the CBG scenario was  $2.8 \text{ MJ}/\text{Nm}^3_{\text{biomethane}}$ .

In the gas hydrate scenarios, approximately 3% methane loss was assumed from the upgrading unit and the hydrate formation and disassociation process, which required natural gas compensation. The energy input for the natural gas compensation in the hydrate scenarios was  $1.3 \text{ MJ}/\text{Nm}^3_{\text{biomethane}}$ . Thus natural gas compensation decreased the energy performance and climate impact for the hydrate scenarios.

Diesel consumption for CBG and biomethane hydrate transport was approximately similar, corresponding to 1.0 and 0.6 MJ/Nm<sup>3</sup><sub>biomethane</sub>, respectively. The higher primary energy use for transport of biogas hydrate (1.6 MJ/Nm<sup>3</sup><sub>biomethane</sub>) was due to 75% increased hydrate mass transport when CO<sub>2</sub> was included in the gas hydrate structure.

The contribution to climate impact was much higher for the biogas and biomethane hydrate scenarios compared with the CBG scenario (Figure 18). The upgrading unit was the single largest contributor to climate impact for all scenarios. Loss of CH<sub>4</sub> from the upgrading unit corresponded to 200 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>biomethane</sub> for all scenarios. The difference in total climate impact from the upgrading unit in the three scenarios was related to the electricity demand.

The hydrate formation and disassociation units corresponded to high levels of GHG emissions related to electricity input in hydrate formation. These emissions corresponded to 60 and 38 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>biomethane</sub> in the biogas and biomethane hydrate scenarios, respectively. Methane losses during gas handling in hydrate formation were around 240 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>biomethane</sub> in both the biogas and biomethane hydrate scenarios. The GHG emissions related to energy input in the hydrate disassociation step corresponded to 101 and 72 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>biomethane</sub> in the biogas and biomethane hydrate scenarios, respectively. Methane losses during gas handling in hydrate disassociation were 148 and 166 g CO<sub>2</sub>-eq./Nm<sup>3</sup><sub>biomethane</sub> in the biogas and biomethane hydrate scenarios, respectively. There were no significant methane losses during decentralised compression in the CBG scenario, as the biogas upgrading and compression units were integrated. The global warming potential (GWP) associated with transport of biogas hydrate was twice that in the biomethane hydrate and CBG scenarios.



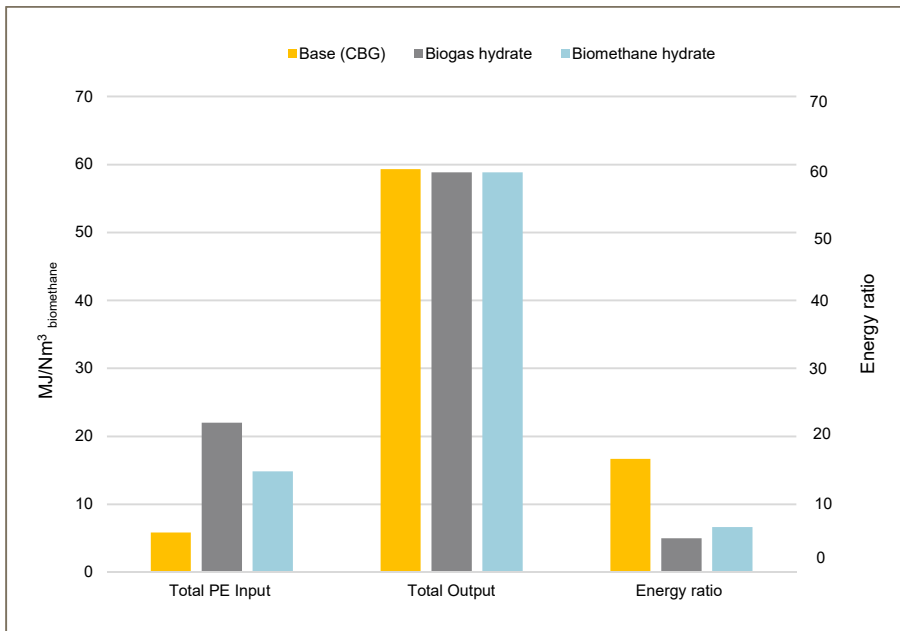


Figure 17. Primary energy (PE) input, energy output and external energy ratio of biogas conversion to high-value products (CBG = compressed biomethane) (Paper VI).

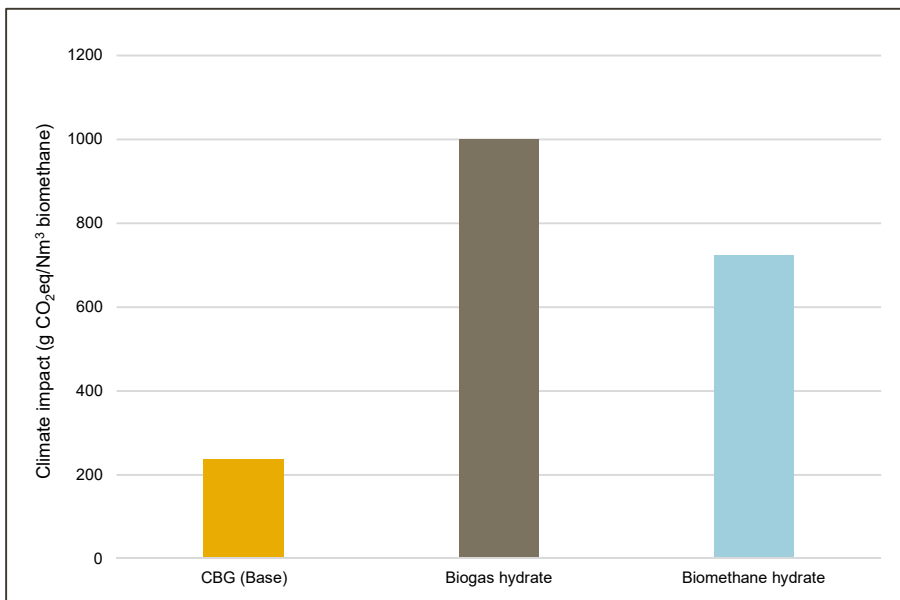


Figure 18. Climate impact (g CO<sub>2</sub>-eq./Nm<sup>3</sup>biomethane) of the biogas/biomethane distribution scenarios (CBG = compressed biomethane).

## 6 Discussion

Biomethane can be produced by a multitude of crops, cropping systems and conversion technologies and has a wide range of applications (*i.e.* heat, electricity, chemicals). The large number of possible combinations of various biomass streams, conversion options, scale ranges, logistics and applications makes it difficult to identify the most optimal system from an energy and climate point of view. In this thesis, different system designs of some novel and futuristic technologies applicable to biomethane were presented and analysed. The systems designed were studied within a life cycle assessment (LCA) framework with a futuristic aspect where the input data were the result of modelling and several combined literature studies. Life cycle assessment of biomethane-based bioenergy systems encompassed feedstock production and logistics, biomass conversion to biogas, biogas upgrading to biomethane, biomethane conversion to other products, by-product handling, biomethane use and distribution. The futuristic nature of the technologies studied meant that detailed technology design and performance data could not be completely verified. However, the data were sufficient for assessing the energy and climate performance of the novel technologies, as the core of the systems studied. In this chapter, a general discussion is provided on the results of the different studies presented in Papers I-IV.

### 6.1 Cropping systems for biomethane production

Cropping systems for biomethane production have different environmental impacts based on the crop and its production system, which differ in respect to crop life cycle length, yield, nutrient and chemical demand, nitrogen losses, soil carbon fluxes, and machinery and other management operations. These factors influence the magnitude of the components contributing to energy performance and climate impact of the overall bioenergy system.

The choice of perennial crops such as willow over annual crops such as maize for bioenergy production is mostly promoted in relation to the combined global food and energy security challenges (Bommarco *et al.*, 2018; Valentine *et al.*, 2012; Karp & Richter, 2011). In addition, it has been shown that perennial crops for bioenergy production have lower environmental impacts than annual crops due to their life cycle length and extensive root system, which allow more efficient nutrient and water use (Rowe *et al.*, 2009; Tilman *et al.*, 2009; Uellendahl *et al.*, 2008; Tilman *et al.*, 2006). Perennial crops establish a viable and extensive root system throughout the cropping year, which leads to lower amounts of drainage water leaving the soil and potentially avoiding nitrogen leaching from the root zone (Dimitriou *et al.*, 2017; Pugesgaard *et al.*, 2014). Perennial energy crops have been suggested as a measure for achieving the targets set by the EU Water Framework Directive, through their permanent, deep root systems and long growing season (Kaspersen, 2015; Bennion & Battarbee, 2007). However, there are concerns related to regional water source issues in cultivation of perennial crops for bioenergy use and these should be considered in sustainability assessments of bioenergy systems (Georgescu *et al.*, 2011).

The data presented in this thesis show the energy and climate benefits of replacing maize-based anaerobic digestion with willow-based pyrolysis for biomethane production (Paper III). These data support findings in other studies that short-rotation willow (*Salix* spp.) is a suitable perennial crop for bioenergy production (*i.e.* heat, electricity and biofuels), with low environmental impacts (*i.e.* global warming, nitrate leaching and eutrophication) and high net energy in comparison with annual crops (Pugesgaard *et al.*, 2014; Karp & Rochter, 2011; Börjesson & Tufvesson, 2011). Furthermore, willow plantation has a low demand for machine operations and diesel inputs in comparison with annual crops (Pugesgaard *et al.*, 2014).

The results obtained in this thesis also show that land use change from annual crops to perennial crops such as willow can lead to higher soil organic carbon (SOC) levels, which is in line with findings by Don *et al.* (2012), Lemus & Lal (2005) and Zan *et al.* (2001). However, it should be taken into consideration that the potential for increasing SOC levels is largely influenced by soil characteristics such as initial SOC stocks and on-site conditions (Don *et al.*, 2012; Zan *et al.*, 2001). Perennial lignocellulosic biomass species, such as willow, develop a coarse, deep and extensive root system, which leads to higher SOC levels, resulting in a lower climate impact or in some cases a climate benefit (Dimitriou & Mola-Yudego, 2017; Hammar *et al.*, 2014; Baum *et al.*, 2013; Toenshoff *et al.*, 2013). Soil organic carbon sequestration is affected by land and crop management decisions, which impact the quantity and quality of crop residues added to the soil and the rate of decomposition (Jarecki & Lal, 2003;

Paustian *et al.*, 2000). Consideration of soil carbon fluxes in assessment of bioenergy systems is important due to their role in climate concerns, crop yields and soil quality, but such fluxes have not been fully covered in previous studies.

Crop yield is also an important factor in the net energy performance and climate impact of bioenergy production (Whitaker *et al.*, 2010). Plant yield and carbon allocation within the plant determine the amount of carbon input to soil (Ericsson, 2015).

Loss of nitrogen from agricultural soil (*i.e.* as  $N_2O$ ,  $N_2$  and  $NO_3^-$ ) through different microbiological and geophysical processes (*e.g.* nitrification, denitrification, runoff, erosion, leaching and volatilisation) results in various environmental impacts, such as global warming, water pollution, loss of biodiversity and human health issues (Sutton *et al.*, 2011). In this thesis, although the climate performance of all biomethane scenarios was found to be better than that of their fossil alternatives, the eutrophication potential of water sources was higher as a result of nitrogen leaching from soil in cropping systems (Paper II). These results support findings in other studies that replacing the fossil energy system with biomethane systems based on energy crops (*i.e.* maize, miscanthus) leads to an increase in freshwater eutrophication (*e.g.* Wagner *et al.*, 2018; Kiesel *et al.*, 2016).

Addition of nitrogen sources (*i.e.* fertilisers and digestate) to cropping systems increases emissions of nitrous oxide ( $N_2O$ ), which is a potent GHG (Solomon *et al.*, 2007). In the case of application of chemical fertilisers, the climate impact from the fertiliser manufacturing process should also be accounted for. In this thesis, it was found that nitrous oxide emissions were the dominant source of GHG emissions in the bioenergy cropping systems of maize and willow (Papers II and III), confirming findings in previous research (Crutzen *et al.*, 2016; Don *et al.*, 2012). However, the nitrous oxide emissions from willow plantation were 67% lower than from maize, due to lower nutrient inputs (*e.g.* nitrogen fertiliser) to willow.

In practice, an optimal bioenergy system also relies on available bioenergy plants and a short distance between the bioenergy cropping unit and the bioenergy conversion plant. The logistics and transport requirements of biomass vary based on the energy density and water content. The relatively low energy density and high water content of biomass make long-distance transport costly and inefficient, and the technology therefore requires an optimised supply chain design.

## 6.2 Technologies for biomass conversion to biomethane

Apart from the efficiency of the cropping system, the choice of biomass conversion technology and its conversion efficiency are important for the total performance of the bioenergy system (Börjesson & Tufvesson, 2011). In this thesis, the reforming and upgrading of pyrolysis gas was shown to be a promising potential route for biomethane production compared with conventional anaerobic digestion. Studies conducted by Larsson *et al.* (2013) showed that biomethane production via pyrolysis even had higher yields of biomethane (based on the biomass) than gasification as an alternative thermochemical technology for biomethane production from lignocellulosic biomass (Paper III). Moreover, uncertainty about the composition of the input feedstock is less limiting for the biomethane output from pyrolysis, since all components of the pyrolysis gas are reformed and converted to biomethane. Under the system assumed in this thesis, all pyrolysis vapours (condensable and non-condensable) were reformed for biomethane production, leading to higher fuel output. Based on findings in other pilot studies (Görling *et al.*, 2013; Larsson *et al.*, 2013), there are no pollutants such as sulphur which could deactivate the catalyst. Experimental studies on pyrolysis of different raw materials would be of interest, regarding different feedstock composition, energy properties and products.

The performance of thermochemical conversion pathways relies on the choice of appropriate biomass feedstock (Tanger *et al.*, 2013). Pyrolysis as a thermochemical conversion technology is most suitable for conversion of woody biomass, with high heating value and low water content, into different forms of energy carriers (*i.e.* liquid, gas and solid) (Kan *et al.*, 2016). The pyrolysis system examined in this thesis was designed based on possible process integration and polygeneration of biomethane, biochar and heat, where the biochar was used as a soil amendment or as an energy carrier to replace fossil coal to exploit the difference in climate mitigation potential. The results show that biochar can create a substantial carbon sink in soil, leading to a potential climate mitigation effect, as also reported in studies by Gaunt & Lehmann (2008) and Laird (2008). The use of biochar as an energy carrier to replace fossil coal led to even higher mitigation effects than its use as a soil amendment. The biochar output from the pyrolysis plant corresponded to approximately 19% of the total useful energy output, or 20 GWh, with a net calorific value of 20.0 MJ/kg based on 4% water content.

On the other hand, heat integration within the pyrolysis system decreased the use of external energy inputs and increased the energy ratio of the system. A pyrolysis reactor can install an amine scrubber as the upgrading technology, due to residual heat produced in the plant. The climate impact of the pyrolysis reactor

and fuel synthesis reactor mainly derived from emissions from the energy carriers and there were no marked gas losses from these two unit processes. In the pyrolysis and anaerobic digestion routes, methane losses during upgrading of gas made large contributions to the climate impact from both scenarios, which draws attention to the choice of technology, for instance use of an amine scrubber instead of a water scrubber.

Based on the overall energy performance and climate mitigation effect, a combined biomethane and biochar production system via pyrolysis of willow is a potential option if an available market exists for the two products. In Sweden, there is currently market interest in biomethane as a transportation fuel, specifically for public transport buses supported by national environmental targets. Biochar, the other main by-product from pyrolysis of willow, can potentially find regional or local markets for use as a heat source or other applications such as displacement of coke in the steel industry.

Both biomass conversion plants studied had similar rates of energy content in the input (approximately 17 MW) and output (approximately 12 MW) flows during operation. However, this is without considering the rate of energy content in the biochar and the exported heat output flows. The rate of energy content lost in the form of gas (*i.e.* methane) was higher in the anaerobic digestion plant during the digestion and upgrading steps.

The pyrolysis system was found to have a net heat surplus (7 MW), mainly from the compression work (2.9 MW), cooling (1.2 MW) and chemical reaction heat from methanisation (0.9 MW). Excess low-temperature heat from the upgrading unit was assumed to be used for drying of feedstock. Excess high-temperature heat (>300 °C) was assumed to be exported from the system for other use. Another proposed application of this excess heat is to replace the additional biomass input for the heat demand of the pyrolysis reactor or drying of the biochar.

### 6.3 Technologies for biomethane conversion to high-value products

Liquefying biomethane through gas-to-liquid (GTL) technologies in order to exploit its potential for use as a biofuel or chemical was studied in another part of the research in this thesis. Dimethyl ether (DME) had the best performance regarding primary energy inputs and climate impact per input biomethane (Nm<sup>3</sup>) and per distance (km) driven (Paper I). It also had the highest output fuel of all the GTL fuels studied. Assessment of fuel combustion in engines showed that, although methanol had overall high energy performance and low global warming potential, owing to its lower heating value (LHV) larger volumes of

fuel must be consumed for similar distances, resulting in larger and heavier fuel tanks. The results in this thesis show that converting biogas to compressed biomethane (CBG) leads to a high specific fuel productivity if the CBG is utilised on a local market (Paper I).

In terms of land use efficiency, combined heat and power (CHP) had the highest energy and climate performance, which was mainly related to the biomass-to-biomethane unit, with a low contribution from the CHP unit itself (Patterson *et al.*, 2011). However, the production of DME and methanol for use as transportation fuel or chemical was shown to be beneficial (Paper II). Production of ammonia from biomethane was not competitive with the alternative biomethane-based CHP, DME and methanol and their fossil-based substitutes.

Liquefying biomethane through GTL technologies would result in other energy carriers such as steam and heat of sufficient quality to be exported to use in other industries. Fischer-Tropsch diesel (FTD) showed a relatively low fuel yield and a high steam yield. Thus, locating an FTD plant close to an industry or CHP plant would facilitate more efficient utilisation of the steam produced.

On the other hand, all GTL fuels had better performance than liquefied biomethane (LBG) produced through cryonic technologies. Cryonic technologies require high electricity input and the technology is economically applicable for large biomethane production plants (50 GWh annual output). Filling stations receive methane as CBG or LBG and dispense it to vehicles at high pressure (>250 bar), low pressure or as LBG (Åhman, 2010). Biomethane cannot be stored as LBG for more than 30 days and it is therefore considered a suitable technology for distributing biomethane (Budzianowski & Brodacka, 2017). On the other hand, the mismatch of biogas production and seasonal peak fuel requirements could be solved, as liquid fuels can be stored (Budzianowski & Brodacka, 2017).

## 6.4 Technologies for biogas/biomethane distribution as gas-hydrates

Gas hydrates were studied in Paper IV as a means to distribute biomethane in comparison with the conventional method of road transport (*i.e.* as CBG). Gas hydration was assumed as a conversion process where biomethane gas molecules are trapped in crystalline structures of water. Biomethane hydrates could also be an efficient means of biomethane storage, but this characteristic was not assessed in the thesis due to limited availability of data.

The results presented in this thesis show that the hydration conversion process and dehydration require a high electricity input and contribute to high

levels of GHG emissions (Paper IV). 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.

The future-oriented approach for the use of hydrates in systems for utilising biogas as a vehicle fuel has several inherent uncertainties, as mentioned previously. The results in this thesis indicate that the energy use is not favourable when considering using hydrate as a means for transporting gas from a production site to be used at another site. In future work, it would be interesting to study integration of hydrate for storage at a centralised biogas plant and integration with *e.g.* return water in a district heating system or surplus heat from a CHP plant for dissociation heat. Furthermore, expansion of the system to include the anaerobic digestion process, where low-grade heat from the cooling during hydrate formation could be utilised, would likely improve the overall efficiency of an integrated system. In addition to system integration assessment, future development of hydrate technologies is important to consider. 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). It has been suggested that this difference could be exploited in an upgrading process (Arca *et al.*, 2011), which would make it possible to integrate dissociation and upgrading in one process. Furthermore, the use of detergents such as sodium dodecyl sulphate has been shown to enhance the hydrate formation rate at lower pressures with reduced stirring, resulting in lower energy input (Zhong & Rogers, 2000).

However, Budzianowski and Brodacka (2017) claim that 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. 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.

## 6.5 LCA of futuristic bioenergy systems

Life cycle assessment has been proven to be a useful tool for impact assessment of futuristic and novel technologies, thus providing guidance for technology development (Arvidsson *et al.*, 2018; Gavankar *et al.*, 2015; Sandén *et al.*, 2005). Futuristic technologies are immature, with small-scale production which may



not have reached the market or may have reached only developed minor niche markets (Arvidsson *et al.*, 2018). Available data and knowledge related to futuristic technologies are limited and sparse, and assessments are mostly based on research publications, patents, prototypes, laboratory experiments and modelling (Khanna & Bakhshi, 2009). Performing LCA of futuristic technology which could adequately represent the environmental impacts of the technology when it has achieved maturity or large-scale production is challenging, due to limited availability of data and methodological settings (Gavankar *et al.*, 2015). One of the most important challenges in performing prospective LCA is setting technology performance parameters, which are functions of time and scale. In the work reported in this thesis, different possible ranges for the parameters in the foreground and background systems were assumed based on modelling and process descriptions in scientific articles. For instance, in the foreground system, scenario ranges were designed to illustrate the potential environmental impacts (*e.g.* different ranges of methane loss from gas upgrading units in Papers II-IV, hydration efficiency in Paper IV). Background system modelling results were also studied by scenario ranges, *e.g.* for electricity mixes (*e.g.* 100% green electricity mix range to 100% fossil electricity mix) representing different primary energy inputs and GHG emission intensities when studying electricity input (energy carriers) in different bioenergy plants.

## 6.6 Uncertainties

Loss of methane from biomethane plants is a fundamental issue which, apart from the significant climate impacts, reduces the yield of the bioenergy plant. These methane losses arise from the anaerobic digestion process and in the upgrading of biogas to biomethane. However, there are no accurate data on methane losses and many operators and research material refer to default values presented by Bauer *et al.* (2013).

On the other hand, biogenic carbon fluxes have been mostly ignored in climate assessment of bioenergy systems based on crops, although they highly influence the overall performance of bioenergy systems, especially those involving land use change. However, assessment of soil carbon stock changes is associated with large uncertainties such as microbiological factors (*i.e.* humification factor) and ecosystem factors (*e.g.* soil properties and climate).

The relatively large share of GHG emissions from cropping systems is related to soil N<sub>2</sub>O emissions as a result of nutrient application (*i.e.* mineral fertilisers, digestate and crop residues). The methodology used for calculating N<sub>2</sub>O emissions from soil is based on IPCC (2006), the results of which are not intended for use in a life cycle inventory. The main uncertainty relating to

calculation of N<sub>2</sub>O emissions is the emission factors used. In Paper II, different ranges of factors were tested in a sensitivity analysis which showed relatively high influence of different emission factor values.

Uncertainties such as dynamics related to GWP emission factors and the time dependency of emissions are important factors to consider when assessing the climate performance of a bioenergy system, as they may yield different results.

## 6.7 Future research

Biomethane development is mainly limited by availability of feedstocks, access to markets and higher cost in comparison with natural gas. Integrated bioenergy systems based on biomethane could be considered as a potential biomethane market developer. Small-scale conversion plants are generally less efficient than large-scale facilities in conversion of biomass to biofuel or biochemicals. This is the cause of many limitations in bioenergy commercialisation, as many farms and small scale-biogas units have less efficient conversion rates (Dornburg & Faaij, 2001).

Based on research conducted in this thesis, small GTL plants can be established in the vicinity of farm-scale and remote biogas plants. On the other hand, there is technological and market interest in offering public electric vehicles in the near future, which could hinder the development of biomethane-based transport. However, it should be borne in mind that heavy-duty and agricultural machinery uses approximately 40% of the commercial diesel fuel. Providing GTL plants for agricultural production units would provide a clean fuel for agricultural machinery and local fuel transport. By-products of heat and steam could be integrated for warming and electricity production or for storing biomethane as hydrates over longer periods. It would be of future research interest to study integrated farm-scale biomethane production based on the different technologies studied in this thesis.

A more holistic view on integrating different unit processes in biomethane-based bioenergy plants, *e.g.* integrating the most efficient biomass and cropping system with a suitable conversion technology and considering the final use of biomethane, would be beneficial in future research. The novel technologies studied within the context of this thesis provide valuable by-products (*e.g.* heat, steam, biochar), which in case of system integration could potentially provide an optimal system in terms of energy and climate performance, while also providing sustainable transportation fuel. It would be highly interesting to study the environmental impact and economic effects of different system integrations. Previous research shows that LCAs of novel technologies based on immature data should be interpreted in conjunction with their technology and

manufacturing readiness levels, reinforcing the need for standardising and communicating information on these readiness levels and scales of production in life cycle inventory practices (Gavankar *et al.*, 2015).

The rapid development of biofuels as a potentially sustainable and cleaner replacement for conventional fuels represents a unique challenge for the chemical industry that requires simultaneous consideration of economic, social and ecological aspects. Thus reduced environmental impacts is one of the main system design objectives. Biomethane is the renewable energy carrier which can potentially best replace fossil sources (*e.g.* natural gas) in chemical production. Gas-to-liquid and pyrolysis plants can be process-designed for production of both fuel and chemicals.

## 7 Conclusions

Conclusions regarding technologies for biomethane production:

- Transitioning from maize-based anaerobic digestion to a willow-based pyrolysis system for biomethane production led to increased energy performance and negative global warming potential.
- Biomethane production via willow pyrolysis had a lower climate impact than anaerobic digestion of maize, mainly due to build-up of soil organic carbon stocks under willow cultivation and the use of biochar.
- Biochar use as an energy source in replacing fossil coal had higher climate mitigation effects than use as a soil amendment or as a carbon sink.

Conclusions regarding novel technologies for biomethane use:

- Assessment of the energy performance and climate impact of GTL products (*i.e.* FTD, methanol, DME) in comparison with current in-use biomethane products (*i.e.* CBG, LBG, CHP) showed that DME and methanol could be feasible alternatives in terms of low primary energy inputs and low climate impact for the enhancement of biomethane utilisation.
- CBG had the highest specific fuel productivity among the biofuels studied and a relatively low primary energy input. However, on increasing the fuel distribution distance from 100 km to 1000 km, DME showed the highest specific fuel performance with low primary energy inputs and lower climate impact.
- Biomass cropping operations were the second largest contributor to primary energy inputs and the climate impact of biomass-to-biomethane production processes.

Conclusion regarding technologies for biomethane distribution:

- Gas hydrate formation and disassociation processes demanded high electricity inputs. The climate impact of the gas hydrate scenarios was higher than that of CBG due to high gas (*i.e.* CH<sub>4</sub>) losses in the formation and disassociation processes and higher energy-related emissions.

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## Popular science summary

### *Background*

Fossil energy has played a strongly positive role in global economic development, followed by further technological, economic and social advances. Increasing concentrations of greenhouse gases (GHGs), including carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O), in the atmosphere, as a result of fossil source consumption, are likely to be the dominant cause of drastic global warming and climate change. In 2015, around 82% of the world's total primary energy supply was based on fossil fuel sources (*i.e.* oil, natural gas and coal), which contributed more than half of all anthropogenic GHG emissions (IEA, 2017). As fossil fuels are finite, there is a need to develop sustainable, available and abundant renewable energy sources for the global energy and material demand.

National and international environmental policies and agreements promote a change from fossil energy to renewable energy and have an important role in directing the future of technological developments. Technical development in the field of renewables is essential in lowering the current high fossil fuel consumption, while still providing innovation and sustainable global growth.

An attractive renewable energy source is biomethane or *renewable natural gas* derived from biogas. Biogas consists of CH<sub>4</sub> and CO<sub>2</sub>, but the CO<sub>2</sub> can be removed to increase the energy content, achieving a gas with almost only biomethane. Biomethane is chemically similar to natural gas and has the potential to produce a broad range of chemicals and energy services (*i.e.* electricity, heat and fuel). Biogas is currently mainly produced through anaerobic digestion of organic material (*i.e.* manure, organic waste, sewage, dead animal and plant matter) by microorganisms. Biogas has significant environmental advantages and can contribute substantially to mitigating GHG emissions through different strategies such as replacing fossil fuel consumption, waste management and use of the digestate as a replacement for mineral

fertilisers. In Sweden, more than 65% of the biogas produced is upgraded to biomethane and mostly used in the transport sector.

Compressed biomethane (CBG) is a suitable option for distribution in the vicinity of a local biogas plant. However, due to the limited gas infrastructure in Sweden, biomethane is also transported as CBG over longer distances. This is not efficient because of the rather low energy density of CBG compared with liquid fuels. Another option is to produce liquefied biomethane (LBG), which has a higher energy density and is more suitable for longer distribution distances. However, this involves cooling the gas to -161 °C, a process restricted to large-scale biomethane production units. Other important barriers to increased deployment of biomethane are low availability of biomass feedstock and limited conversion routes for biomethane production.

### *The research in this thesis*

This thesis investigated futuristic and potential technologies in the field of biomethane production, use and distribution, and compared them with conventional and in-use technologies of their kind. The technologies examined were:

- Gas-to-liquid (GTL) technologies, where biomethane is converted from gaseous to liquid biofuels (e.g. diesel, ethers and alcohols). This enables conversion of biogas into other energy carriers with higher energy density.
- Pyrolysis, which involves thermal conversion of biomass to biomethane to fuel gas and by-products such as biochar in the absence of oxygen and at relatively low temperatures (400-800 °C). This process enables use of a wide range of feedstock, including woody biomass. Biochar can be used in agriculture as a soil amendment or in industries as an energy source.
- Biomethane hydrate technology, which can be used for capturing methane in ice structures of water called methane hydrates. These structures are naturally present in deep sea floor sediments. Gas hydrates were studied in this thesis as a technology to distribute biomethane as solid mass.

In order to assess the performance and compare the new and novel technologies with existing technologies, life cycle assessments (LCAs) were conducted. LCA is an internationally agreed tool for supporting decision-making related to environmental planning. In this thesis, it was applied to study the different technologies and assess their energy and climate performance through their life cycle.



### *Main findings of the thesis*

The results showed that biomethane conversion to dimethyl ether (DME) by GTL technology had better energy and climate performance than using biomethane as compressed or liquefied biomethane (CBG, LBG).

Pyrolysis of willow had better energy and climate performance than anaerobic digestion of maize. Using biochar from pyrolysis as a soil amendment or an energy source to replace coal in industries resulted in considerable climate cooling effects.

The electricity demand of biomethane hydrate formation and disassociation was found to be high. Gas handling during biomethane hydrate formation and disassociation resulted in gas losses and high climate impacts. This indicates a need for further technological developments and future studies in order to improve the energy performance and reduce the climate impact of biomethane hydrate distribution systems.

Finally, the work in this thesis showed that the biomethane produced and used through the novel technologies studied had better energy performance and lower climate impacts than the fossil counterparts. However, life cycle cost assessments (LCCAs) are needed to provide better decision support for future technology development and implementation.

# Populärvetenskaplig sammanfattning

## *Bakgrund*

Fossil energi har spelat en mycket viktig roll för den globala ekonomiska utvecklingen, vilket lett till ytterligare tekniska och sociala framsteg. Ökande koncentrationer av växthusgaser, t.ex. koldioxid (CO<sub>2</sub>), metan (CH<sub>4</sub>) och kväveoxid (N<sub>2</sub>O), i atmosfären är en följd av konsumtionen av fossila energikällor, vilket är den dominerande orsaken till den omfattande globala uppvärmningen och klimatförändring. År 2015 var cirka 82 % av världens totala primära energiförsörjning baserad på fossila bränslen (dvs. olja, naturgas och kol), vilka bidrog med mer än hälften av alla antropogena växthusgasutsläpp (IEA, 2017). Eftersom fossila bränslen är en ändlig resurs är det nödvändigt att utveckla hållbara, tillgängliga och rikligt förekommande förnyelsebara energikällor för att tillgodose den globala efterfrågan på energi och material.

Nationella och internationella miljöavtal främjar en förändring från fossil energi till förnybar energi och har en viktig roll för att styra framtida teknisk utveckling. Teknisk utveckling inom förnybara energikällor är avgörande för att sänka den nuvarande förbrukningen av fossila bränslen och samtidigt bidra till hållbar global tillväxt.

En attraktiv förnybar energikälla är biogas, som kan substituera fossil naturgas. Biogas består framför allt av CH<sub>4</sub> och CO<sub>2</sub>, men CO<sub>2</sub> kan avlägsnas för att öka energiinnehållet och ge en gas med nästan bara biometan. Biometan är kemiskt lik naturgas och har potential som råvara för produktion av ett brett spektrum av kemikalier och energitjänster (dvs. el, värme och bränsle). Biogas produceras för närvarande huvudsakligen genom anaerob (syrefri) omsättning av organiskt material (dvs. gödsel, organiskt avfall, avloppsvatten, slakteriavfall och växtmaterial) med hjälp av mikroorganismer, s.k. rötning. Biogas har betydande miljömässiga fördelar och kan väsentligt bidra till att reducera växthusgasutsläppen genom att reducera fossil bränsleförbrukning, förbättra avfallshanteringen och ersätta mineralgödselmedel genom användning av

rötresten. I Sverige uppgraderas mer än 65 % av den producerade biogasen till biometan, som används mest inom transportsektorn.

Komprimerad biometan (CBG) är ett lämpligt alternativ för distribution i närheten av lokala biogasanläggningar. På grund av den begränsade gasinfrastrukturen i Sverige transporteras biometan också som CBG över längre sträckor. Detta är inte effektivt på grund av den relativt låga energitätheten för CBG jämfört med flytande bränslen. Ett annat alternativ är att producera flytande biometan (LBG), som har högre energitäthet och är mer lämplig för längre distributionsavstånd. Detta innebär dock att gasen kyls till  $-161^{\circ}\text{C}$ , en process som är begränsad till storskaliga biometanproduktionsenheter. Andra viktiga hinder för ökad användning av biometan är begränsad tillgänglighet av lämpliga biobaserade råmaterial för mikrobiologisk omsättning till biometan.

### *Forskningen i denna avhandling*

Denna avhandling undersökte nya potentiella teknologier för produktion, användning och distribution av biometan, och jämförde dem med konventionella tekniker och system. De undersökta teknikerna var:

- Teknik för "gas-till-vätska" (GTL), där biometan omvandlas från ett gasformigt biobränsle till flytande form (*t ex.* diesel, etrar och alkoholer). Detta möjliggör omvandling av biogas till energibärare med mycket högre energitäthet.
- Pyrolys, som innebär termisk omvandling av biomassa till biometan och biprodukter såsom biokol i frånvaro av syre och vid relativt låga temperaturer ( $400\text{-}800^{\circ}\text{C}$ ), möjliggör användning av ett brett utbud av råvaror, inklusive träbaserad biomassa. Biokol kan användas inom jordbruket som jordförbättring eller i industrier som energikälla.
- Teknik för bildning av gashydrater, vilket innebär att biogas eller biometan kan fångas i is-strukturer. Dessa strukturer är naturligt förekommande i djupa havssediment. Gashydrater studerades i denna avhandling som en möjlighet för att distribuera biometan i fast fas.

För att bedöma prestanda och jämföra de nya teknikerna med befintlig teknik, genomfördes livscykelanalyser (LCA). LCA är en internationellt överenskommen metodik för att stödja beslutsfattande i samband med miljöplanering. I denna avhandling tillämpades den för att studera olika teknologier och utvärdera deras energi- och klimatprestanda i ett livscykelperspektiv.

### *Huvudresultat i avhandlingen*

Resultaten visade att konvertering av biometan till dimetyleter (DME) med GTL-teknik hade bättre energi- och klimatprestanda jämfört med användning av komprimerad eller flytande biometan (CBG eller LBG).

Pyrolys av Salix hade bättre energi- och klimatprestanda i systemperspektiv än rötning av majs. Att använda biokol från pyrolys som jordförbättring eller som energikälla för att ersätta kol i industrin resulterade i betydande kylning av klimatet.

Elbehovet för bildning och disassociation av biometanhydrat var hög. Gashanteringen vid bildning och disassociation av biometanhydrat resulterade i gasförluster med hög klimatpåverkan. Detta indikerar behovet av ytterligare teknisk utveckling och framtida studier för att förbättra energiprestanda och minska klimatpåverkan vid användning av biometanhydrat för distribution.

Slutligen visade arbetet i denna avhandling att biometan som produceras och används med de nya tekniker som ingick i studien hade bättre energiprestanda och lägre klimatpåverkan än sina fossila motsvarigheter. För att ge bättre beslutsstöd inför framtida teknikutveckling och implementering behövs dock livscykelkostnadsbedömningar (LCCA).

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