

Swedish farm-scale biogas production- substrates and operating parameters

Gasen i botten

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Abstract

Biogas production from agricultural waste streams provides three value streams: production of fossil-free energy, reduced greenhouse gas (GHG) emissions and production of nutrient-rich digestate that can be used as fertiliser. However, farm-based biogas production is expanding rather slowly. One limitation is the low energy content and restricted degradation of manure. Previous research has identified strategies for improving gas production, but the other two value streams have received less attention. Therefore Swedish farm-scale biogas plants (FSBP) were evaluated in this thesis with the aim of identifying ways to improve overall efficiency in terms of high gas yields, high degree of degradation and high digestate nutrient concentration.

The results revealed large variations in the efficiency of Swedish FSBP, but with consistent correlations between gas production, degree of degradation and retention time. Co-digestion was found to be a commonly used strategy to improve biogas yield and digestate nutrient content. Detailed laboratory studies of different operating strategies showed that co-digesting manure with rapeseed oil, starch, albumin or cultivated energy crops had positive effects on volumetric gas yield and/or plant nutrient levels in digestate. Increased digester temperature improved biogas yield from cattle manure, whereas for poultry manure it instead resulted in decreased gas yield and instability, due to high ammonia levels. Residual methane potential in digestate was found to correlate positively with organic load and negatively with retention time, illustrating the importance of sufficient duration of degradation in reducing GHG emissions. Based on these findings, it can be concluded that measurement and evaluation of residual methane potential is a promising tool in understanding FSBP processes and assessing their efficiency.

Keywords: agricultural biogas production, fossil free energy, co-digestion, manure, residual methane potential, operating parameters.

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Gasen i botten. Lantbruksbaserad biogasproduktion i Sverige – substrat och driftsparametrar

Sammanfattning

Biogasproduktion från lantbrukets restprodukter tillför tre viktiga värden till samhället, produktion av fossilfri och lokal energi, minskning av växthusgaser och förbättrat växtnäringsvärde i den biogödsel som bildas i biogasprocessen. Tidigare forskning har framförallt fokuserat på att öka gasproduktionen för att effektivisera gasutbytet men den lantbruksbaserade biogasproduktionens hela värdekedja beaktas sällan. I denna avhandling har målet varit att identifiera hur totaleffektiviteten kan förbättras för dessa biogasanläggningar.

Resultaten visar att variationen i effektivitet och driftsparametrar är stor bland de undersökta biogasanläggningarna. Trots det finns korrelationer mellan gasproduktion, nedbrytningsgrad och uppehållstid vilket är intressanta samband då dessa nyckeltal relaterar till biogasproduktionens tre värdeområden. Samrötning används vanligen som strategi på befintliga biogasanläggningar för att förbättra både gasproduktion och koncentrationen av ammoniumkväve i biogödseln. I denna studie har rena fraktioner av fett, kolhydrater och protein, samt olika energigrödor, utvärderats i samrötning med gödsel. De olika samrötningssubstraten resulterade i olika grad av påverkan på gasproduktionen och växtnäringsvärde i biogödseln. För att öka biogasproduktionen kan temperaturen i reaktorn höjas till termofil temperatur. Detta kan dock vara problematiskt vid användning av proteinrika samrötningssubstrat som vid termofil rötning istället kan resultera i minskad gasproduktion och nedbrytningsgrad på grund av ammoniakhämmning i reaktorn. I denna studie visades också ett positivt samband mellan restmetanpotential i biogödseln och organisk belastning samt ett negativt samband mellan restmetanpotential och uppehållstid. Dessa samband är viktiga för att förstå biogasproduktionens roll i minskning av växthusgaser från jordbrukssektorn.

Nyckelord: lantbruksbaserad biogasproduktion, fossilfri energi, samrötning, gödsel, restmetanpotential, driftsparametrar

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Dedication

To my beloved family –Ella, Alva and Stefan

The most difficult thing is the decision to act, the rest in merely tenacity. The fears are paper tigers. You can do anything you decide to do. You can act to change and control your life; and the procedure, the process is its own reward
Amelia Earhart

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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 Ahlberg Eliasson, K., Nadeau, E., Levén, L., Schnürer, A*. (2017). Production efficiency of Swedish farm-scale biogas plants. *Biomass and Bioenergy*, 97, pp. 27-37.
- II Ahlberg Eliasson, K*, Liu, T., Nadeau, E., Schnürer, A. (2018). Forage types and origin of manure in co-digestion affect methane yield and microbial community structure. *Grass and Forage Science*, 73, pp. 1-18.
- III Ahlberg Eliasson, K*, Isaksson, S., Westerholm, M., Schnürer, A. (2018). Organic loading rate and temperature are determinative for methane yields and digestate residual production during anaerobic digestion of manure. (Submitted manuscript).
- IV Ahlberg Eliasson, K*, Singh, A., Isaksson, S., Schnürer, A. (2018). Co-substrate composition critical for efficiency during biogas production from cattle-manure. (Manuscript)

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The contribution of the author to the papers included in this thesis was as follows:

I Planned the study and analysed the data. Main writer of the manuscript.

II Planned the study, performed the laboratory work related to digester operation, and analysed the process data. Main writer of the manuscript.

III Planned the study, performed part of the laboratory work and analysed the process data. Main writer of the manuscript.

IV Planned the study, performed part of the laboratory work and operation and contributed to analysing the data. Main writer of the manuscript.

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Abbreviations

BMP	Bio methane potential
CHP	Combined heat and power
CSTR	Continuously stirred tank reactor
DD	Degree of degradation
EU	European Union
FAN	Free ammonia nitrogen
FSBP	Farm-scale biogas plant
GHG	Greenhouse gas
HRT	Hydraulic retention time
ILUC	Indirect land use change
OLR	Organic loading rate
RMP	Residual methane potential
SMP	Specific methane potential
TAN	Total ammonia nitrogen
TS	Total solids
VS	Volatile solids

1 Introduction

In the world-wide transition towards a sustainable society, reduction of greenhouse gas (GHG) emissions and sustainable food production are critical (Rockstrom *et al.*, 2017; FAO, 2014). However, GHG emissions from the agriculture sector are high, representing around 30% of total global emissions and 13% of national GHG emissions in Sweden (Clark & Tilman, 2017; Swedish Environmental Protection Agency, 2017). Agricultural sources of greenhouse gases are mainly dinitrogen oxide (N_2O) from denitrification in soil and enteric methane (CH_4) production from ruminants and fossil energy use for transportation, machinery and fertiliser production (Swedish Environmental Protection Agency, 2017; Strøm Prestvik *et al.*, 2013; Edström *et al.*, 2005). Nitrogen is essential for high-level food production from agricultural land and nitrogen recirculation is one of nine “planetary boundaries” for sustainable development (Rockström *et al.*, 2009). However, the Haber-Bosch process by which atmospheric nitrogen is synthesised to ammonia (NH_3) fertiliser is currently a significant contributor to the environmental impact of agriculture, as the process is very demanding of fossil energy (Erisman *et al.*, 2008).

Sweden has 16 environmental quality objectives, a number of which concern the agriculture sector (Olsson & Fallde, 2015; Swedish Environmental Protection Agency, 2014). Thus, the sector has great potential to contribute to reductions in greenhouse gas emissions and also to achieve other environmental improvements, such as nutrient recirculation. Establishment of biogas production systems can play an important role in the development of sustainable agricultural production. In this process, agricultural waste streams, including *e.g.* manure and crop residues, can be used for production of biogas while also creating long-term recirculation systems for nutrients (Bacenetti *et al.*, 2016; Kaporaju & Rintala, 2011; Holm-Nielsen *et al.*, 2009).

The importance of biogas production at farm-scale stems from the fact that it produces three value streams: Environmental benefits, fossil-free energy and nutrient-rich digestate (Figure 1), (Ahlberg-Eliasson, 2015). The environmental

values include the potential greenhouse gas reduction from conversion of manure to biogas and also from replacement of fossil energy (Bacchetti *et al.*, 2016; Holm-Nielsen *et al.*, 2009). The fossil-free biogas produced can be used as an energy source in several energy transitions, such as electricity, heat or vehicle fuel (Torrijos, 2016). Finally, the digestate has important value for nitrogen recirculation and management and for carbon sequestration (Björnsson *et al.*, 2016; Insam *et al.*, 2015; Holm-Nielsen *et al.*, 2009). Biodigested manure is also more homogeneous and has a lower content of total solids, and is thus easier to apply to arable land than untreated manure (Al Seadi *et al.*, 2013; Paavola & Rintala, 2008). These values in the farm-scale biogas system can contribute to social or economic systems, *e.g.* biogas production can result in job opportunities and development of rural areas (Olsson & Fallde, 2015).

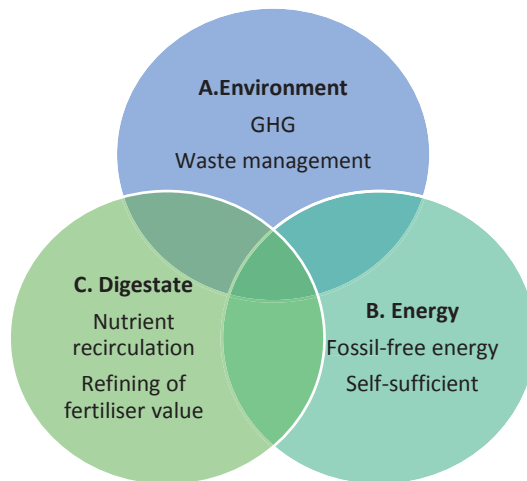


Figure 1. The three main value streams in agricultural biogas production: A) Environmental benefits, B) fossil-free energy and C) digestate fertiliser.

The outcome of the three value streams are strongly connected to the economic output of the system and form a value chain for agricultural biogas production (Figure 1). Different parameters, including technical configuration, operation of the biogas plant and chemical and physical parameters of the substrates, all affect these three essential value streams. Today, many manure-based biogas plants operating at farm-scale are in need of improvement in order to reach their full potential in terms of providing all these three value streams (Lebuhn *et al.*, 2014). This is mainly because manure generally has a high water content and low concentration of organic matter, and thus often gives low levels of biogas. To improve biogas production from manure, co-digestion with more energy-rich substrates can be an option, as previous research has revealed positive results on

gas production when manure is co-digested with other substrates (Tufaner & Avşar, 2016; Sondergaard *et al.*, 2015; Angelidaki & Ellegaard, 2003) Moreover, fundamental improvements in operating parameters such as temperature and organic load may be needed (Tufaner & Avşar, 2016; Mata-Alvarez *et al.*, 2014). However, there is a lack of information regarding the effects of co-digestion and of changes in operating parameters on the other value streams, such as nutrient levels in digestate and reduced risks of greenhouse gas emissions, all of which are important for farm-scale biogas production.

1.1 Aim and hypothesis

The aim of the work described in this thesis was to investigate the importance of substrates and operating parameters for agricultural manure-based biogas production and how these can be combined to increase the process efficiency. Efficiency was defined as improved energy production and digestate value, *i.e.* nutrient concentration and degree of degradation. Specific objectives were to:

- Evaluate Swedish farm-scale biogas plants in regard to substrates and gas production (**I**).
- Investigate how co-digestion of manure can be used to improve the efficiency in farm-scale biogas plants (**I, II, III and IV**).
- Evaluate effects of operating parameters, *e.g.* temperature and organic load, on the efficiency of farm-scale biogas plants (**II and III**).

1.2 Limitations

The main focus in the work was on different substrates and operating parameters used in farm-scale biogas production plants and how these affect the efficiency of the biogas process. Technical equipment, internal energy demand and different market solutions, including *e.g.* choices for energy utilisation, are only mentioned briefly in this thesis, even though these issues are also important for sustainable biogas production with low life cycle costs. Moreover, while many different types of substrates are currently used for biogas production in co-digestion with manure, in this thesis work the focus was mainly on cattle manure and a few types of forages and individual compounds (proteins, fats and carbohydrates).

2 Biogas production in the European Union and Sweden

2.1 Biogas production in the European Union

In the European Union (EU), the overall goal set by the Renewable Energy Directive (RED) is to reach 20% of renewable energy by 2020, including a minimum of 10% renewable fuels in the transportation sector (European Parliament, 2009). Production of biogas in the EU-28 has increased considerably in recent decades, from approximately 47 TWh in 2005 to 193 TWh in 2015 (Eurostat, 2016). Among the EU member states, Germany has the highest number of biogas plants, followed by Italy, the UK and France (Table 1) (EBA, 2016). The feedstock used for biogas production varies between countries. Agricultural substrates, *i.e.* manure, crop residues and cultivated energy crops, are mainly used for biogas production in Germany, Italy and Czech Republic, where approximately 75% of all biogas plants operate with these materials as the major substrate (EBA, 2016; Torrijos, 2016). In total, approximately 13 TWh are produced from anaerobic digestion of manure in the EU. Energy crops contribute approximately 50% of the total energy in biogas in the EU (equivalent to 88 TWh) (Kampman *et al.*, 2017). However, indirect land use change (ILUC) criteria limit the areal production of energy crops, thus also restricting their use for biogas production (European Parliament, 2009). Cultivation of energy crops is considered a problem in terms of land use and competition with food production. Problems such as soil erosion and loss of biodiversity can also result from production of energy crops (Herrmann *et al.*, 2017). However, assessments in Sweden have shown that at least 10% of arable land is currently not cultivated, suggesting that energy crops could be cultivated and used for energy production purposes without competition with food production (Prade *et al.*, 2017; EBA, 2016; Swedish Government, 2007). While overall biogas production in the EU

has increased during recent years, the untapped potential is still high. According to estimates by the European Biomass Association, agricultural substrates such as manure, crops and straw currently available in Europe have the potential for annual production of 58.9 billion m³ of biomethane, which is equivalent to 364 TWh (AEBIOM, 2009).

Table 1. *The top 12 European countries in terms of biogas production and the number of biogas plants in each. Adapted from (EBA, 2016).*

Country	Number of biogas plants
Germany	10786
Italy	1491
United Kingdom	831
France	763
Switzerland	633
Czech Republic	554
Austria	439
Sweden	279
Poland	277
Netherlands	268
Belgium	204
Denmark	152

2.2 Biogas production in Sweden

From a historical view, biogas production in Sweden developed from the requirement for sewage sludge treatment, followed by the requirement for fossil-free fuels in the transportation sector (Olsson & Fallde, 2015). From 2005 to 2016, Swedish biogas production increased from 1285 GWh to 2018 GWh (Figure 2). The total number of biogas plants in 2016 was 279, with the largest amounts of gas coming from co-digestion plants (industrial), mainly digesting food waste, manure and other organic materials, and from digestion of sewage sludge (Swedish Energy Agency, 2017). Energy production at farm-scale biogas plants is showing a steady increase and reached 49 GWh in 2016. The main substrate used in farm-scale biogas plants in Sweden is manure, but crop residues, energy crops and other organic waste streams are also used (I), (Swedish Energy Agency, 2017). The total number of biogas plants on farms has increased over recent years and the current number is 41 (Swedish Energy Agency, 2017). The driving forces for agricultural biogas production are manure management and the different subsidies available for investment within the

Rural Development Programme through the EU. In Sweden, a relatively high proportion of total biogas production is used for upgrading to vehicle fuel, in total approximately 1300 GWh or 64% (Swedish Energy Agency, 2017). The methane content in upgraded biogas used for vehicle fuel must be $\geq 97\%$ according to standardisation legislation (Awe *et al.*, 2017). However, upgraded biogas is mainly produced in large co-digestion plants or at wastewater treatment plants, while most of the farm-scale biogas plants in Sweden instead have a combined heat and power (CHP) unit installed (I), (Swedish Energy Agency, 2017).

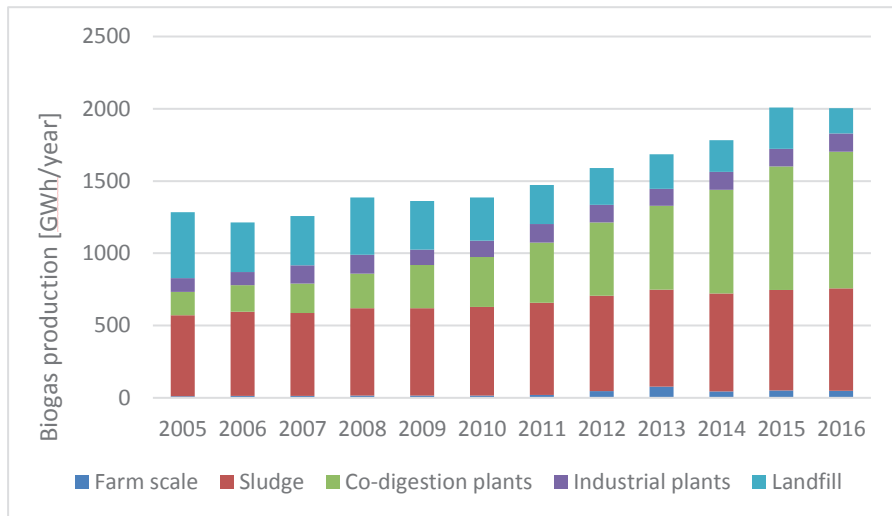


Figure 2. Swedish biogas production 2005-2016 by category of production plants (gasification with total production 14 GWh in 2016 excluded).

The digestate produced in biogas plants in Sweden is governed by regulations. Digestate from sludge digestion is covered by a domestic certification system called REVAQ and this type of digestate is only permitted to be used for crops not intended for feeding animals or humans, *e.g.* energy crops and forest (SWWA, 2018). Digestate from co-digestion plants is covered by certification regulation SPCR 120, which regulates the quality of the substrate/substrates used for digestion and use of the digestate based on concentrations of nutrients, heavy metals and levels of pathogens. About 20 commercial biogas plants have SPCR 120 certification in Sweden. The certified digestate is permitted for use on agricultural land and 96% of the digestate produced in co-digestion is now used as a fertiliser in crop cultivation (Swedish Waste Management Association, 2018; Swedish Board of Agriculture, 2017; Swedish Energy Agency, 2017). For biogas plants digesting manure, the same regulation that covers spreading and

storage of manure applies and in this case 100% of the digestate produced is used as fertiliser on agricultural land (Swedish Energy Agency, 2017). Biogas plants in Sweden are also governed by the EU regulations on animal by-products (EG 1069/2009), which restrict the use of substrates such as slaughterhouse waste, household waste and manure. All biogas plants need a permit from the Swedish Board of Agriculture stating which substrate/substrates that specific plant is allowed to use (European Parliament, 2009).

The potential for production of fossil-free energy from available agricultural substrates, manure, energy crops and crop residues far exceeds actual production today (Martin, 2015; Swedish Energy Agency, 2010; Linné *et al.*, 2008). According to estimates by the Swedish Energy Agency, the total biogas potential from manure is 2.3 TWh and that from crop residues is 3.1 TWh. When cultivated energy crops are included in the calculations, the total biogas potential from agricultural substrates, *i.e.* manure, crop residues and energy crops, is approximately 14 TWh (Swedish Energy Agency, 2010). Co-digestion with more energy-dense substrates can further increase biogas production at farm-scale biogas plants (**I**, **II** and **III**). Swedish researchers have also identified possibilities for increased gas production by more efficient digestion, for example by increasing the retention time, at existing biogas plants in Sweden digesting industrial and household waste (Linné & Persson, 2017). According to those authors, 5-12% more biogas can be obtained from these existing plants. However, no corresponding research has so far been done to farm-scale biogas plants in Sweden.

2.3 Subsidies for biogas production, impact on production structure

The increase in biogas production in Europe is primarily a result of domestic subsidy systems, mainly consisting of feed-in tariffs for electricity production to the grid. These subsidy systems have historically resulted in an increase in agricultural biogas plants, especially in Germany, where the total number of biogas plants has increased from approximately 400 in 1997 to over 10 000 today (EBA, 2016; Torrijos, 2016; Lebuhn *et al.*, 2014). However, according to the European Commission, the long-term goal is to move away from feed-in tariffs and instead use other subsidy systems, like a feed-in premium or quota system. These modifications in the EU have changed the subsidy system in member states in different ways (EBA, 2014). However, the financial support systems still vary between EU member states. A recent study by Rogstrand (2017) summarises the regulations and financial systems for 15 countries in the European Union and divides the support system into 14 categories. It was

concluded in that study that France, Denmark and the UK have the most well-developed financial structure for further expansion of biomethane (Rogstrand, 2017). The subsidy system of one country can also have consequences for other countries, *e.g.* a recent change to the Danish subsidy system has resulted in import of Danish biogas to Sweden, which has caused problems for domestic biogas plants supplying upgraded biogas to the grid (Andersson, 2017; Haaker, 2017; Johnsson, 2017).

2.3.1 Subsidy structure in Sweden

In Sweden, biogas plants that digest manure are eligible for a subsidy. The main aim is to reduce greenhouse gas emissions from manure and increase the level of manure digested to biogas. Therefore, only biogas plants that use manure get the subsidy, which is equal to 4 c€ per kWh produced (Swedish Board of Agriculture, 2015). The main difference between the Swedish subsidy structure compared with that in *e.g.* Germany and Denmark is that only manure is eligible for the subsidy in the Swedish system. Nevertheless, the subsidy has greatly improved the economic situation for farm-scale biogas plants in Sweden. Unfortunately, there are some difficulties with interpretation of the regulations, mostly relating to reported data from the plants (Ahlberg Eliasson & Birgersson, 2017). However, the Swedish subsidy has led to an increase in the use of manure for biogas production and to more biogas being produced from each ton of manure (Niemi Hjulfors & Edström, 2017).

In addition to the subsidy for manure digestion, biogas plants on farms are also offered investment support through the Rural Development Programme, covering on average 40% of the investment cost of the biogas plant (Swedish Board of Agriculture, 2018). If biogas is used for electricity production, irrespective of substrate, the electricity is included in the system of green certificates. For 2017 the average price of electricity green certificates was 6.3 €/MWh and the price has decreased since 2012 (Figure 3), (Svensk Kraftmätning, 2018; Norges vassdrags-og energidirektorat & Energimyndigheten, 2016). This decrease in energy prices for electricity on the Nordic energy market has had a negative impact on farm-scale biogas production.

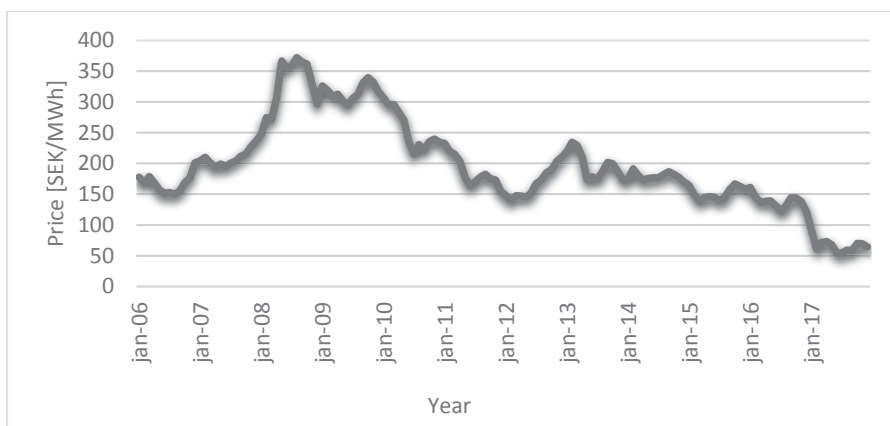


Figure 3. Change in the price of an electricity certificate in Sweden, 2009-2017. Adapted from Svensk Kraftmätning (2018).

2.4 Technology, gas production and digestate at Swedish farm-scale biogas plants

The majority of Swedish farm-scale biogas production plants use semi-continuous digestion with one main digester (Figure 4). The temperature range is in most cases mesophilic (35-40 °C), with only three Swedish farm-scale plants running at thermophilic temperature (50-55 °C) in 2015 (I). The substrate slurry is pumped into the digester from a mixing tank. Substrate with a high dry matter content is either mixed together with the slurry or fed into the digester separately (I). A majority of existing farm-scale biogas plants use the biogas they produce in a combined heat and power unit for energy conversion. The electricity is used on the farm and/or sold to the grid and the heat is often used for internal heating or in local heating systems. At a few biogas plants, the biogas produced is directly combusted in a boiler for heat production and used in local industries (I). About six biogas plants at farm-scale upgrade biomethane to vehicle fuel (I) (Swedish Energy Agency, 2017).

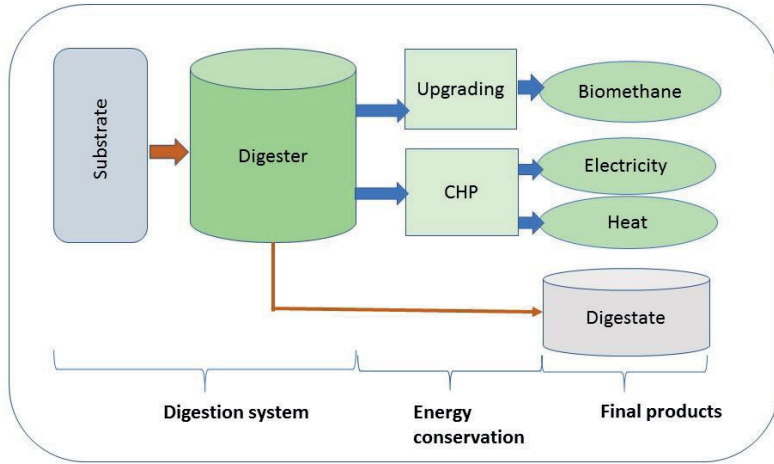


Figure 4. Main components of the type of biogas plant commonly used at farm-scale in Sweden

In Sweden, most of the technology used in farm-scale biogas plants is developed by hand. Each biogas plant is often in need of individual solutions for technology and design. The farm-scale biogas plant in general has a lower level of automation and has limited possibilities for online monitoring and control compared with larger industrial biogas (Ahlberg-Eliasson, 2015). The average methane yield, measured as $\text{Nm}^3 \text{CH}_4/\text{ton}$ wet weight (ww) substrate is $15 \text{ Nm}^3 \text{CH}_4/\text{ton}$, but it varies from $7 \text{ Nm}^3 \text{CH}_4/\text{ton}$ in Swedish farm-scale plants operating with mono-digestion to $45 \text{ Nm}^3 \text{CH}_4/\text{ton}$ in biogas plants using manure combined with co-substrates such as slaughterhouse waste and industrial food waste (I). This production level is lower than in German farm-scale plants, which have an average production level of $86 \text{ Nm}^3 \text{CH}_4/\text{ton}$ ww substrate (range $28\text{-}141 \text{ Nm}^3 \text{CH}_4/\text{ton}$ ww), using mainly maize silage as substrate (FNR, 2010). The overall production level in Swedish farm-based plants is also lower than in Danish farm-scale plants digesting manure, often in co-digestion with different kinds of energy crops, which achieve an average production level of $27 \text{ Nm}^3 \text{CH}_4/\text{ton}$ ww substrate (Møller & Nielsen, 2015). The difference in production level between different countries is likely caused by differences in feedstock composition, which several studies has been shown to be of importance for achieving high biogas yields (I, II, III and IV) (Swedish Energy Agency, 2010; Holm-Nielsen *et al.*, 2009; Angelidaki & Ellegaard, 2003).

In Sweden, the digestate from farm-scale biogas plants is mainly used on the farm where the biogas plant is located. The average concentration of ammonium in the digestate of 27 biogas plants evaluated in 2015 was 2.6 kg/ton (I). This value corresponds to an increase of 22% compared with the concentration in the substrate/substrates used in Swedish farm-scale plants. The levels of ammonium in cattle and pig manure reported in this Swedish evaluation are in line with previous findings for European biogas plants operating with manure and/or manure in co-digestion (Alburquerque *et al.*, 2012; Poetsch *et al.*, 2004). Storage of digested manure is governed by the same regulations as storage of undigested manure, *i.e.* manure and covered storage are only mandatory at some production units (depending on numbers of animals), as stipulated by environmental restrictions. Although most Swedish farm-scale plants have covered storage for their digestate (Figure 5), some do not (I). As regards spreading digestate on arable land, there is a restriction of maximum 22 kg phosphorus and 170 kg total nitrogen yearly per hectare (Swedish Board of Agriculture, 2017). None of the digestates from Swedish farm-scale biogas plants that were evaluated in Paper I risked exceeding the maximum level of nitrogen allowed. More often, the average phosphorus concentration in the digestate samples, which was 0.7 kg/ton, limited the volume of digestate that could be applied to farm land (I).



Figure 5. Aerial view of a Swedish farm-scale biogas plant (FSBP) with, main digester, second digester, covered storage and final uncovered storage. Photo: Mattias Bergström.

3 Anaerobic digestion process

The efficiency of a biogas production system, as described in this thesis, is defined as high gas production, high degradation of substrate/substrates used and high nutrient concentration in the digestate. The efficiency of the biogas process depends on different factors, such as substrate and operating parameters, as well as the prevailing microbiological system

3.1 Operating parameters

Different operating parameters, together with the substrate, affect the development of the microbial community, the biogas yield and the digestate quality (**I**, **II**, **III** and **IV**) (Schnürer *et al.*, 2016; Labatut *et al.*, 2014; Nasir *et al.*, 2014; Al Seadi *et al.*, 2013). One important parameter for management is the operating temperature. Most farm-scale biogas production units in Sweden use the mesophilic temperature range (37-40 °C), although a few plants operate at thermophilic temperature (50-55 °C). Some of the latter plants started at this temperature from the outset and some changed to it from mesophilic temperature (**I**). Digestion of manure at thermophilic compared with mesophilic temperature can result in faster gas production rates, higher yields and higher degree of degradation (Moset *et al.*, 2015; Labatut *et al.*, 2014). These effects can also allow for shorter retention times (Ward *et al.*, 2008). With higher digestion temperature, the rheological behaviour of the slurry changes and, as a consequence, stirring can be less energy demanding (Brambilla *et al.*, 2013). High temperatures can also reduce the level of pathogens (Watcharasukarn *et al.*, 2009). However, the overall outcome of higher operating temperature during manure digestion is dependent on the substrate/s used (**III**) (Labatut *et al.*, 2014). As a result of increasing temperature, the equilibrium between ammonium and ammonia is pushed to higher ammonia concentration (Westerholm *et al.*, 2016; Rajagopal *et al.*, 2013). Ammonia is toxic to the microbial process and at high

concentrations can thus cause inhibition of methanogenesis and process instability (Yenigun & Demirel, 2013). This has been observed *e.g.* during thermophilic digestion of poultry and swine manure, alone or in co-digestion (III) (Hansen 1998, Rajogopal, Rodriguez Verde 2018, Yenguiin a Dermiel 2013).

Another important management parameter is the hydraulic retention time (HRT), which needs to be sufficiently long to allow degradation of the substrate used. Short HRT increases the risk of washing out important microorganisms, resulting in ineffective degradation, while long HRT results in inefficient use of digester volume (Ruile *et al.*, 2015). Substrates containing high concentrations of lignocellulose, *e.g.* some energy crops and manure, need relatively long retention times (>30 days) to be degraded compared with more easily degradable materials such as starch- and sugar-rich substrates which can be degraded using shorter retention times (IV) (Linke *et al.*, 2013). Typical retention times in Swedish farm-scale biogas plants range between 31 and 57 days (I). These retention times are similar to those reported in other European studies, although there is wide variation depending on the substrate used. For example, biogas plants in Germany using energy crops in the substrate mix have longer retention times, on average 100 days (Møller & Nielsen, 2015; FNR, 2010; Hopfner-Sixt *et al.*, 2006).

The organic loading rate (OLR), representing the amount of organic matter fed into the digester every day, is another important operating parameter. Organic loading rates of between 2 and 6 kg volatile solids (VS)/m³ active digester volume are commonly found in agricultural biogas plants (I) (Schnürer, 2016; Angelidaki & Ellegaard, 2003). Moreover, the OLR and HRT are often interlinked and an increase in load, *e.g.* in the case of manure, often results in shortening of the retention time, which can impact negatively on the efficiency (III). Moreover, higher organic loads can also cause problems with accumulation of volatile fatty acids (VFA) and process failure (III) (Schnürer, 2016).

3.2 The microbiology of biogas production

Anaerobic digestion involves different groups of microorganisms that degrade the substrate in a series of steps (Figure 6), (Angelidaki *et al.*, 2011). The main groups of microorganisms involved in degradation are anaerobic or facultative anaerobic bacteria and archaea, including anaerobic methane producers. There is a difference between the functionality of these types of microorganisms in the different steps of anaerobic digestion (Angelidaki *et al.*, 2011). In general, a high diversity of microorganisms is believed to give stability and robustness to the

degradation steps in the biogas process (Schnürer, 2016; Angelidaki *et al.*, 2011). The structure of the microbial community depends on factors such as substrate composition, temperature, retention time, stirring and pH value, with ammonia level and temperature being the strongest drivers (Müller *et al.*, 2016; De Vrieze *et al.*, 2015; Sundberg *et al.*, 2013). The four steps of anaerobic degradation of substrates are: hydrolysis, fermentation, anaerobic oxidation and methane formation (Figure 6). During hydrolysis, organic substrates, *i.e.* proteins, carbohydrates and fats, are degraded by hydrolytic bacteria to amino acids, simple sugar molecules and fatty acids, respectively. The rate of degradation in hydrolysis depends on the composition of the substrate. In agricultural substrates such as manure and energy crops, the concentration of lignocellulose is often high, restricting microbial degradation and slowing the hydrolysis step (Liu *et al.*, 2015; Frigon & Guiot, 2010; Angelidaki & Ellegaard, 2003). This explains the importance of pre-treatment for these type of substrates, in order to break up the complex structure of lignocellulose and improve the access for microbial degradation. In this regard, the retention time is also important in providing enough time for hydrolysis (Azman *et al.*, 2015; Tomei *et al.*, 2009; Vavilin *et al.*, 2008).

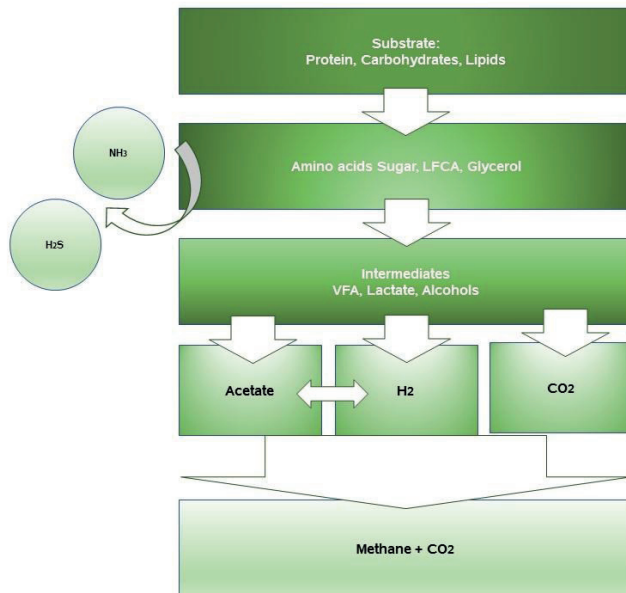


Figure 6. Schematic diagram of anaerobic degradation of organic compounds

In the fermentation step, products from the hydrolysis step, *i.e.* amino acids, sugars and fatty acids, are degraded to *e.g.* short-chain fatty acids, alcohols,

ammonia and hydrogen sulphate (H₂S), (Figure 6). In this step and in the next degradation step, hydrogen is formed. During the third step, anaerobic oxidation, products from the fermentation step are further utilised by acetogenic bacteria, producing hydrogen, carbon dioxide and acetate as the main end-products. The acetogenic bacteria use protons as final electron acceptors, resulting in production of hydrogen gas. If the partial pressure of hydrogen is high, this oxidation step will not occur for thermodynamic reasons (Schink, 1997). Hydrogen-consuming microorganisms such as methane-forming archaea that are active in the final step are consequently critical for the conversion of organic acids. An appropriate balance between the hydrogen producers and the hydrogen consumers is essential in order to achieve stable anaerobic digestion (Lebuhn *et al.*, 2014).

The final methane-forming step can be divided into two main pathways; degradation of acetate to methane and carbon dioxide, which is performed by acetoclastic methanogens, and formation of methane from hydrogen and carbon dioxide, which is performed by the hydrogenotrophic methanogens (Angelidaki *et al.*, 2011). In addition, an alternative pathway for methane formation from acetate, via so-called syntrophic acetate oxidation (SAO), can proceed depending on prevailing conditions (Westerholm *et al.*, 2016). In the SAO pathway, bacteria oxidise acetate to form hydrogen and carbon dioxide, which are thereafter used by hydrogenotrophic archaea for methane formation. (Westerholm *et al.*, 2016). Technical parameters such as temperature, retention time and ammonia concentration have been shown to influence the degree to which the pathway SAO is active (Westerholm *et al.*, 2016).

3.2.1 The microbial community in manure based processes

The most dominant microorganisms in anaerobic digestion, which are abundant in many processes irrespective of operating conditions and substrates, are members of the phyla Firmicutes and Bacteroidetes. Members of these phyla have a wide metabolic capacity, which most likely explains their high abundance in many biogas plants (**II** and **III**) (Schnürer, 2016; Azman *et al.*, 2015; De Vrieze *et al.*, 2015). Actinobacteria and Proteobacteria are also commonly found phyla during manure digestion (**II** and **IV**) (Liu *et al.*, 2017; Chen *et al.*, 2016; Schnürer, 2016; Sun *et al.*, 2015). For substrates such as manure and energy crops, degradation of lignocellulose is of particular importance. The phyla Firmicutes and Actinobacteria are known to contain many members with cellulose-degrading ability (Bozan *et al.*, 2017; Azman *et al.*, 2015; Saini *et al.*, 2015). The phylum Bacteroidetes also contains cellulose degraders (Sun *et al.*, 2016; Azman *et al.*, 2015; Sun *et al.*, 2013). The abundance of these phyla in

manure digestion varies depending on the operating parameters employed, with higher levels of Firmicutes relative to Bacteroidetes often associated with thermophilic temperature and co-digestion with easily accessible carbohydrates such as starch (II and IV) (Sun *et al.*, 2015). Interestingly, several studies have indicated that higher abundance of Firmicutes compared with Bacteroidetes is correlated with high methane yield (II) (Chen *et al.*, 2016). In swine and poultry manure, the level of ammonia is often high and in several studies, in this thesis and elsewhere, this parameter has also been shown to give high abundance of Firmicutes (IV) (Müller *et al.*, 2016; De Vrieze *et al.*, 2015). However, when albumin was co-digested with cattle manure in Paper IV, giving an increase in the ammonium level from 1.9 to 3.8 g/L, no increase in Firmicutes could be observed. However, in line with previous studies an increase of the abundance of syntrophic acetate oxidising bacteria was seen in Paper IV, likely a response to the increasing ammonia level. Co-digestion of manure with other substrates, like fat and starch, have also shown substrate specific responses in microbial community, with enrichment of lipid degrading *Syntrophomonas* or cellulose degrading *Ruminoclostridium* (IV).

In the anaerobic digestion process, methane-forming archaeal microorganisms belonging to the phylum Euryarchaeota are typically members of the orders Methanobacteriales, Methanosarcinales and Methanomicrobiales (Angelidaki *et al.*, 2011). These orders all contain hydrogenotrophic methanogens. Aceticlastic methanogens belong to the order Methanosarcinales, families Methaosarcinaceae and Methanosaetaceae. The abundance of different methanogens is connected to the concentration of acetate, but technical parameters such as stirring, feeding pattern, load and ammonia level can also influence their abundance (Schnürer, 2016). The genera *Methanosarcina* and *Methanobacterium*, both belonging to the order Methanobacteriales, are common in manure-based biogas digestion (II, III and IV) (Sun *et al.*, 2015; Demirel *et al.*, 2008). *Methanosarcina* appears to be a robust microorganism for methane formation, as it can use several different substrates and can also handle stress factors such as high ammonium concentrations, temperature fluctuations and high organic loads (III and II) (De Vrieze *et al.*, 2012). In general, the genus *Methanosarcina* or hydrogenotrophic methanogens belonging to the genus *Methanothermobacter* (order Methanobacteriales) or the genus *Methanoculleus* (order Methanomicrobiales) are often seen to increase in abundance with increasing process temperature (De Vrieze *et al.*, 2015; Sun *et al.*, 2015; Sundberg *et al.*, 2013). *Methanoculleus* is also often present in high abundance at high ammonia levels (Westerholm *et al.*, 2016). However, following a change from mesophilic to thermophilic temperature during mono-digestion of cattle

manure and co-digestion of cattle and poultry manure in Paper **III**, no such change in abundance could be detected.

3.2.2 Disturbance of the biogas process

The microorganisms in the anaerobic digestion process, especially methanogens, are sensitive and are easily disturbed by various parameters (Chen *et al.*, 2014). Parameters such as pH and compounds or derivatives from the degradation process, such as fatty acids, ammonia and hydrogen sulphide, can disturb or even be toxic for the microorganisms in the process, both bacteria and methanogens (Chen *et al.*, 2014). If the methanogens do not fulfil their metabolism, anaerobic oxidation is inhibited and, as a consequence, volatile fatty acids accumulate. This gives a less efficient process and can also result in complete process failure. Different volatile fatty acids can accumulate, with propionate suggested as an indicator of more severe process disturbance (Schnürer, 2016; Marchaim & Krause, 1993). In addition to toxic effects, instability can also be caused by temperature fluctuations, mixing strategies, high organic loading rate and too short retention time (Schnürer *et al.*, 2016; Lindmark *et al.*, 2014) (see also section 3.1).

A common cause of disturbance of biogas processes is ammonium release during mineralisation of organic nitrogen (**III** and **IV**). Ammonium is an important component for microbial growth and for the fertiliser value of the digestate (Zarebska *et al.*, 2015; Rajagopal *et al.*, 2013; Angelidaki & Ahring, 1993). Ammonium (NH_4^+) is in equilibrium with ammonia (NH_3) (Eq. 1), with the latter suggested to be the inhibitory component.



Under high temperature and pH values, the equilibrium shifts towards the toxic ammonia gas. High levels of free ammonia nitrogen can cause inhibition of the biogas process, with acetoclastic methanogens being known to be particularly sensitive (Yenigun & Demirel, 2013). However, bacteria active in the hydrolysis step have also been suggested to be sensitive to high ammonia levels (Liu *et al.*, 2017; Sun *et al.*, 2016). Inhibition of the biogas process is often seen at ammonia concentrations around 0.15-0.5 g/L. However, both higher and lower values have been reported to cause inhibition, typically depending on temperature and pH (Westerholm *et al.*, 2016; Rajagopal *et al.*, 2013). In the work presented in this thesis (**I**, **III** and **IV**), ammonia inhibition and related volatile fatty acid accumulation was found to be a common cause of disturbance in Swedish farm-

scale biogas production plants, an effect often related to the use of swine/poultry manure (**I**, **III** and **IV**). Substrates such as slaughterhouse waste and manure, especially pig and poultry manure, contain high concentrations of protein, and therefore pose a risk of high free ammonia nitrogen concentrations and process inhibition (**III**) (Schnürer, 2016; Hansen *et al.*, 1998). It is worth noting is that, over time, process microorganisms can adapt to high concentrations of free ammonia nitrogen, often by a shift to the syntrophic acetate oxidation pathway (**IV**) (Wang *et al.*, 2016; Westerholm *et al.*, 2016; Rajagopal *et al.*, 2013; Yenigun & Demirel, 2013).

In the biogas process, hydrogen sulphide (H₂S) is also formed from degradation of proteins, *i.e.* sulphur-containing amino acids (Rasi *et al.*, 2011). Hydrogen sulphide can be toxic for process microbes and can inhibit the methanogens (Meyerjens *et al.*, 1995). It can also precipitate metals that are necessary as co-factors for enzyme activity (see section 3.3). Reduced enzymatic activity slows down degradation and can result in negative effects on the methanogens, resulting in decreased degradation rate (Choong *et al.*, 2016; Moestedt *et al.*, 2013; Chen *et al.*, 2008; Hansen *et al.*, 1999). A majority of the farm-scale biogas plants currently operating in Sweden have problems with H₂S in the raw biogas (**I** and **III**) (Ahlberg-Eliasson, 2015). From a technical point, H₂S causes corrosion problems on equipment, *e.g.* pipes and the CHP unit, at the biogas plant, and it is recommended that the level be kept under 1000 ppm (Rasi *et al.*, 2011). As a consequence, lowering the H₂S concentration is important in farm-scale biogas plants, and therefore iron (Fe) is often used as a process additive to precipitate sulphide as iron sulphide (**III**) (Ryckebosch *et al.*, 2011; Meyerjens *et al.*, 1995). An alternative approach is aeration at the top of the digester to oxidise sulphide in the gas phase, an approach which is used by some farm-scale biogas plants in Sweden.

3.3 Substrates for farm-scale biogas production

Any candidate substrate for biogas production must have a suitable combination of degradable carbohydrates, fats and proteins, as well as trace metals and buffering components (Schnürer *et al.*, 2016). Other factors to consider are the energy content and achieving an optimal composition for microbial activity. According to Buschwell's formula degradation of proteins, fats and carbohydrates results in different yields of biogas and different concentrations of methane in the biogas. Carbohydrates yield approximately 430 m³ CH₄/ton VS and the methane concentration in the raw biogas is typically around 50%. Proteins yield approximately 500 m³ CH₄/ton VS, with approximately 60%

methane concentration, while the methane potential for fats is 1000 CH₄/ton VS, with 70% methane in the biogas (Angelidaki *et al.*, 2011).

For the biogas process, the ratio between carbon and nitrogen (C/N ratio) is important and a value between 15 and 30 is reported to be optimal for microbial growth (Mata-Alvarez *et al.*, 2014; Esposito *et al.*, 2012). If the C/N ratio is too high or too low for a single substrate, co-digestion can be used to adjust to an optimum C/N ratio (II) (Mata-Alvarez *et al.*, 2014; Risberg *et al.*, 2013; Nasir *et al.*, 2012). Trace metals are also essential for the anaerobic process and for microbial activity. Iron, cobalt (Co), molybdenum (Mo), selenium (Se) and nickel (Ni) have been suggested to be of particular importance (Choong *et al.*, 2016; Demirel & Scherer, 2011). According to a review by Choong *et al.* (2016), the micronutrients required by the methanogens follow the order of importance: Fe>>Zn>Ni>Cu≈Co≈Mo>Mn. When present in high concentrations, some metals, *e.g.* copper and zinc, can cause inhibition of the methanogens (Lebuhn *et al.*, 2014).

3.3.1 Manure

Manure is a waste material from animal production and therefore often a free substrate available for biogas production. Manure has high buffering capacity and a good proportion of trace metals important for the microorganisms in the digester (Tufaner & Avşar, 2016; Mata-Alvarez *et al.*, 2014). The proportion of nutrients and the final biogas potential in the manure are related to the feed composition and the digestion efficiency of the animal (II) (Møller *et al.*, 2012; Amon *et al.*, 2007; Van Soest, 1994). A large proportion of manure consists of the fibre fraction, comprising undegradable cellulose, hemicellulose and lignin not digested by the animal (Demirel & Chen, 2004). Thus manure generally has quite low biogas potential of between 100 and 300 CH₄ m³/ton VS, but with large variations between different types of manure. Pig and poultry manure often gives slightly higher values than cattle manure, due to a higher content of proteins in the former (III) (Schnürer & Jarvis, 2017; Møller *et al.*, 2004; Kaparaju *et al.*, 2002).

In Sweden, the most common types of manure used for biogas production are cattle and pig slurry. The content of total solids (TS) varies from 3 to 12 % and the manures can therefore be pumped into the system (I) (Björling *et al.*, 2017; Risberg *et al.*, 2017). As described above, types of manure with a relatively high protein content, such as pig and poultry manure, can result in process disturbance due to high ammonia levels (III) (Schnürer *et al.*, 2016; Rajagopal *et al.*, 2013). This is unfortunate, as high ammonium concentration is an important determining component for the value of the digestate as a fertiliser (III) (Monlau

et al., 2015; Möller & Müller, 2012) (see also section 3.4). Moreover, high ammonia concentrations represent a possible option for *in situ* hygienisation, *e.g.* it has been reported that decreased levels of coliform bacteria in digestate from pig and poultry manure are negatively correlated ($R^2=0.84$) with the ammonia concentration (Luo *et al.*, 2017; Ottoson *et al.*, 2008). Common strategies to deal with the problem of high levels of ammonia are dilution and approaches shifting the equilibrium to ammonium, for example by decreasing the temperature and/or adjusting the pH (III) (Nasir *et al.*, 2012).

Another problem with manure is the high water content, which results in low organic loads and long retention times, typically resulting in a less efficient process, *i.e.* low gas production (III) (Tufaner & Avşar, 2016). Moreover, despite the relatively high water content in liquid manure, the rheological behaviour of the manure can still be a problem, particularly if mixing and pumping are inefficient. This is often the case in Swedish farm-scale biogas plants and causes high internal energy demand (Ahlberg-Eliasson, 2015). The viscosity of manure has been shown to be positively correlated with the total solids content and negatively correlated with increased temperature (El-Mashad *et al.*, 2005; Landry *et al.*, 2004).

A specific type of manure is deep litter manure, defined as having a total solids content of above 25%, which consists of animal excreta and straw used for bedding material (Björling *et al.*, 2017). Due to the high concentration of total solids, deep litter can be used for dry digestion, but it can also be used as a co-substrate with liquid manure in continuously stirred tank reactor (CSTR) processes (I and III) (Kothari *et al.*, 2014). Deep litter is suitable for anaerobic digestion as it contains high amounts of organic nitrogen, mineralised to ammonium, in combination with high carbon levels. Straw can also provide important attachment points for cellulolytic microorganisms in the digester. The importance of attachment for fibrolytic bacteria degrading fibres has been well documented (Vavilin *et al.*, 2008; Sung *et al.*, 2007; McAllister *et al.*, 1994). However, the deep litter fraction is heterogeneous, resulting in a wide variation in biomethane potential values (Angelidaki & Ellegaard, 2003). Moreover, it needs some form of pre-treatment, for example milling to reduce particle size (Figure 7), typically resulting in higher internal energy demand and investment costs (Schnürer & Jarvis, 2017; Ahlberg Eliasson, 2012).



Figure 7. Equipment for pre-treatment and feeding of deep litter manure. Photo Karin Ahlberg Eliasson and Lars-Erik Jansson.

3.3.2 Energy crops

Common energy crops used in farm-scale biogas production in Sweden are maize, perennial forages and whole crop cereals (I) (Gissén *et al.*, 2014). Crops are feasible for use in biogas production due to their high energy concentration and biomethane potential, but with variations depending on crop type. Reported values for maize and whole-crop cereals range between approximately 260 and 360 m³ CH₄/ton VS. For perennial forages, *e.g.* ley silage, typical values for biomethane potential are around 300 m³ CH₄/ ton VS (II) (Herrmann *et al.*, 2016; Sondergaard *et al.*, 2015; Gissén *et al.*, 2014; Lehtomaki, 2006). Maturity stage affects the energy value of the crop and BMP values decreasing with increasing crop maturity. This effect is caused by an increased proportion of undigestible neutral detergent fibre (NDF) in total solids, resulting in decreasing organic matter digestibility (II) (Surendra & Khanal, 2015; Nizami *et al.*, 2009; Van Soest, 1994). For example, early-harvested ley has been shown to result in higher BMP values than late-cut ley (Gissén *et al.*, 2014). Therefore, use of energy crops for biogas production is a balance between harvest time and yield (Frigon & Guiot, 2010; Lehtomaki & Bjornsson, 2006). Early-cut crops in general contain more easily degradable compounds. However, to maximise the VS yield per hectare, crops should be harvested late (Amon *et al.*, 2007). Forage crops, such as heavily fertilised grass leys and legume-rich leys rich in protein, can add nitrogen to the biogas substrate mix, which is especially valuable for the final nitrogen concentration in the digestate, although this also can cause problems due too high FAN concentrations (II) (Benke *et al.*, 2017; Muller-Stover *et al.*, 2016). For energy crops, the concentration of essential trace metals is often low and thus biogas processes operating with high inputs of energy crops typically need to be complemented with trace minerals or, alternatively, they need to be co-digested with more nutrient-rich materials (Demirel & Scherer, 2011). However, the trace energy concentration in energy crops differs according to type of crop, soil and fertilisation during cultivation (Demirel &

Scherer, 2011; Albrecht & Beauchemin, 2003). For example, trace metal concentrations in ley silage have been shown to be higher compared to the concentration in cereals (II) (Gissén *et al.*, 2014).

In general, energy crops need pre-treatment, including ensiling of the crop, if the biomass is to be used in a biogas process and choice of pre-treatment method will affect final biogas production (Surendra & Khanal, 2015; Kreuger *et al.*, 2011; Lehtomaki, 2006; Mshandete *et al.*, 2006). A factor to take into consideration in this regard is that pre-treatment is typically associated with additional costs (Franco *et al.*, 2016; Møller & Nielsen, 2015; Ahlberg Eliasson, 2012; Banemann, 2010).

Another factor to consider when using energy crops in a biogas system is the relatively high environmental impact and production costs compared with waste substrates such as manure. As mentioned earlier in this thesis (section 2.1), the regulatory system in the EU and the ILUC criteria limit the possibilities for cultivation of energy crops on arable land. The effect of nitrate leaching from the widespread cultivation of energy maize in Germany is under discussion and recent results show that maize risks having lower eco-efficiency than other crops (Herrmann *et al.*, 2017; Svoboda *et al.*, 2013). Perennial ley crops and whole-crop cereals have instead been shown to have a low environmental impact compared with maize silage (Börjesson *et al.*, 2015). Leys and whole-crop cereals have also been shown to increase gas yields and improve digestate quality in co-digestion with manure (II) (Börjesson *et al.*, 2015; Gissén *et al.*, 2014; Luscher *et al.*, 2014)

3.3.1 Other substrates

In Sweden, substrates such as slaughterhouse waste and waste fat from restaurants are used for co-digestion with manure in some farm-scale biogas plants (I). Slaughterhouse waste contains different ratios of lipids and proteins and is very rich in energy. The high protein content results in high ammonium levels, which is important for the digestate value. However, the composition of different wastes in this category differs greatly, and a recent study reported BMP values ranging from 602 to 855 m³ CH₄/ton VS and volatile solids concentrations ranging from 12 to 35 % of wet weight (Moukakis *et al.*, 2018). During degradation of fats, long-chain fatty acids (LFCA) are produced and these can cause inhibition and foaming problems in the digester (Schnürer & Jarvis, 2017; Moeller & Zehndorf, 2016; Kougias *et al.*, 2013). However, with a good balance between the different substrates and a suitable feeding strategy, the process can still be managed successfully (IV) (Hamawand, 2015; Pitk *et al.*, 2014; Palatsi *et al.*, 2011).

Some of the farm-scale biogas production plants in Sweden also use other types of energy-rich substrates containing easily degradable carbohydrates, such as industrial waste from commercial bakeries and confectionary manufacturers (I). In addition, different waste products from the dairy industry are used, for example energy-rich residues such as milk or cream, or whey, which is typically rich in protein. These often result in high gas production, but also pose a risk of ammonia inhibition. Even though these substrates are used in limited amounts in Swedish farm-scale biogas production plants, they are still important substrates for co-digestion with manure, in order to increase the final gas production and increase the nutrient content in digestate (I, II and IV).

3.4 The digestate

The final concentration of nutrients in the digestate is dependent on the quality of substrates and the performance of the biogas process. Operating parameters such as substrate composition, temperature and retention time are of particular importance for the overall fertiliser value of the digestate (I, II and III) (Bacenetti *et al.*, 2016; Nkoa, 2014; Al Seadi *et al.*, 2013; Alburquerque *et al.*, 2012). The reduction in total solids during the anaerobic digestion process improves the spreadability of the manure. This reduction in total solids has been shown to be >28% in case of manure digestion (I) (Al Seadi *et al.*, 2013). For farm-scale biogas plants, one main value of the anaerobic process is the mineralisation of nitrogen to ammonium, the nitrogen source available directly to crops (Cabello *et al.*, 2009). When manure is digested as a single substrate or in co-digestion with other substrates in farm-scale plants, the ammonium concentration is generally higher in the digestate than in the substrate (I) (Schnürer & Jarvis, 2017; Insam *et al.*, 2015). Moreover, digestate from biogas plants operating with swine manure often have higher concentrations of ammonium than plants operating with cattle manure (I and III) (Herrmann *et al.*, 2017; Alburquerque *et al.*, 2012). Poultry manure also results in high final ammonium concentrations (Nie *et al.*, 2015). For the farm-scale biogas plants in Sweden studied in Paper I, the average increase in ammonium was 0.5 kg/ton in all plants evaluated (Figure 8). The final concentration in the digestate from biogas plants operating with pig manure in mono- or co-digestion was on average 2.5 kg/ton, while in plants with cattle manure in mono- or co-digestion the concentration in digestate averaged 1.8 kg/ton (I). The levels in the Swedish farm-scale biogas plants studied in this thesis are in line with values reported in previous studies of digestate originating from manure, although there are wide variations typically depending on origin of the manure and operating parameters in the digester (II) (Sambusiti *et al.*, 2015; Al Seadi *et al.*, 2013; Alburquerque

et al., 2012; Triolo *et al.*, 2011). The concentrations of trace metals in the digestate depend on concentrations in the substrate, but also on the operation or the biogas process, *e.g.* if process additives are used. In addition, losses of cadmium, zinc and magnesium in the form of struvite and metal sulphides can occur (II) (Zirkler *et al.*, 2014).

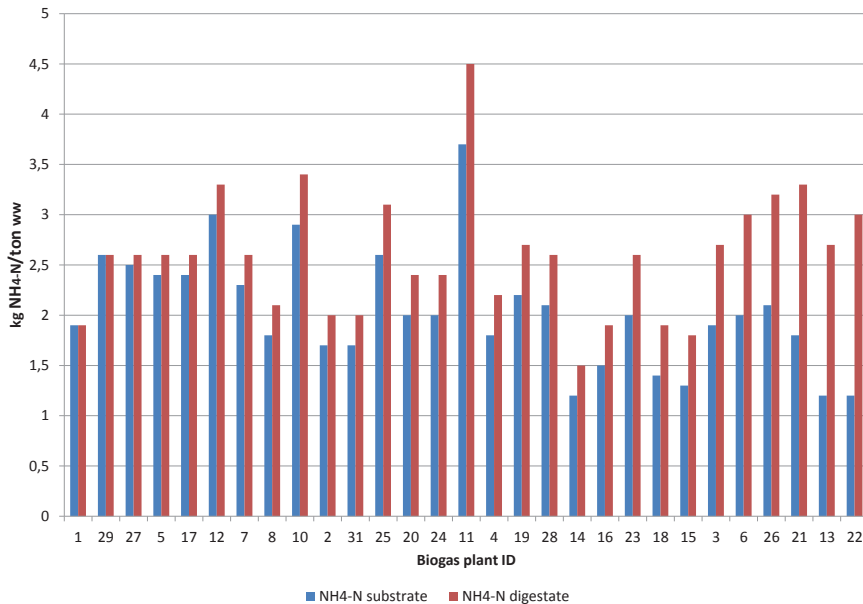


Figure 8. Ammonium-nitrogen concentration (kg/ton wet weight) in substrate and in digestate from farm-scale biogas plants in Sweden (I).

The hygienisation effect, *i.e.* reduction of pathogens, and the reduction of odour from the digested manure after the anaerobic digestion process are also important outcomes of the biogas process (Arikan *et al.*, 2018; Lebuhn *et al.*, 2014; Holm-Nielsen *et al.*, 2009). The amounts of pathogens and weed seeds in the digestate are related to the concentrations in substrate and to operating parameters such as retention time and temperature (Froschle *et al.*, 2015; Luo & Angelidaki, 2014; Al Seadi *et al.*, 2013). The techniques used for storage and spreading of the digestate will also affect the final value of the biogas process. During storage, factors such as crusting and temperature affect the risk of greenhouse gas emissions and ammonia losses (Elsgaard *et al.*, 2016; Insam *et al.*, 2015; Rhode *et al.*, 2012; Wood *et al.*, 2012). Finally, the type of crop, crop rotation and soil also affect how efficiently the crop uses the nutrients in the digestate (Insam *et al.*, 2015; Albuquerque *et al.*, 2012; Stinner *et al.*, 2008).

4 Improved efficiency in farm-scale biogas production

Efficiency in a biogas system is defined in this thesis as high utilisation of available substrate in order to increase biogas production and, as a result, improve the degradation of available organic matter and nutrients in digestate and reduce the greenhouse gas emissions (**II**, **III** and **IV**). The level of efficiency depends on the chemical and physical characteristics of the substrate/substrates used, the operation of the biogas plant and also the technology used in the biogas system. The chemical, physical and technical parameters affect the final output values of the entire biogas system (Figure 9). Chemical parameters include *e.g.* concentration of macro- and micronutrients in the substrate and whether the nutrients are accessible for conversion to biogas by the microorganisms. Chemical parameters also affect the digestate quality, *i.e.* its ammonium concentration. Physical parameters include *e.g.* particle size, viscosity and temperature. Some of these parameters can be influenced by operating conditions, for example by pre-treatment of substrates in order to reduce the particle size. Technical parameters are defined as conditions that can be manipulated by technical decisions made by the operator of the plant. These technical parameters can affect the physical parameters and vice versa. For example, particle size is determined by methods for pre-treatment and the viscosity by the stirring conditions, while the digestibility of the substrate can also be affected by operating conditions such as retention time and temperature (**II** and **III**) (Baudez *et al.*, 2013; Kaparaju *et al.*, 2008; Mshandete *et al.*, 2006). The technical parameters can be actively chosen to improve the degradation process and the final output of gas and degradation (**II** and **III**) (Moset *et al.*, 2015; Moestedt *et al.*, 2013; Risberg *et al.*, 2013). The chemical and also the physical parameters are related to the type of substrate and therefore it is not always possible to actively change these parameters

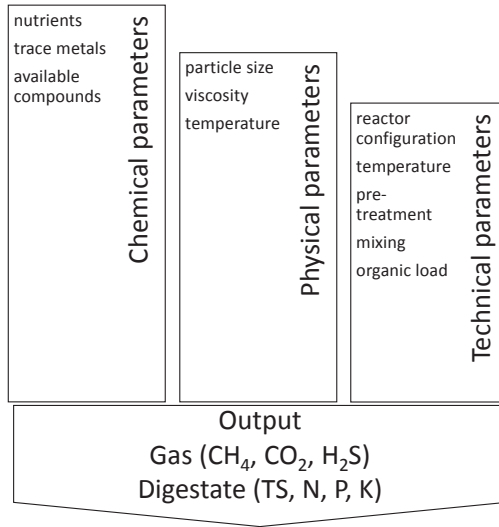


Figure 9 Schematic description for connection between substrate characteristics and the technology used at the biogas plant (chemical-, physical- and technological parameters)

4.1 Evaluation of efficiency

Efficiency in a biogas system as defined in this thesis is mainly evaluated using four points for assessment. These are: degree of degradation, gas production, residual methane potential and mineralisation of nutrients. These factors can be improved *e.g.* by co-digestion and/or altering technical parameters such as temperature, retention time and organic load (**I**, **II** and **III**) (Moset *et al.*, 2015; Mata-Alvarez *et al.*, 2014).

4.1.1 Degradation of organic matter, digestibility of compounds.

Reduction of organic matter, degree of degradation (DD) or reduction in volatile solids (VS) between substrate and digestate describes the utilisation of organic matter for biogas production and is calculated according to Eq. 2 (**I**).

$$DD = 1 - \frac{VS \text{ digestate}}{VS \text{ substrate}} \quad (2.)$$

The variation in content of degradable compounds in manure results in wide differences in degree of degradation. For example, cattle manure typically shows

a lower degree of degradation (25-40%) than swine manure (45-60%) (I) (Møller *et al.*, 2004). Degree of degradation often shows a correlation with operating parameters such as organic loading rate and hydraulic retention time (HRT). For the Swedish biogas plants evaluated in Paper I, a correlation ($k=0.71$) was found between DD and HRT (Figure 10). A positive correlation was also found in Paper I between DD and reduction in carbon and nitrogen ($k=0.78$ and $k=0.70$, respectively) and between DD and specific methane potential (SMP) ($k=0.54$).

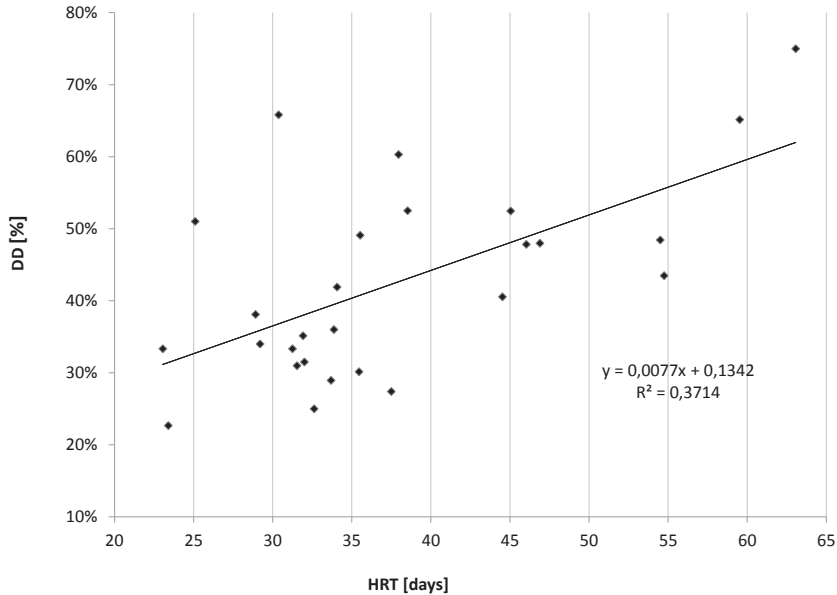


Figure 10. Relation between degree of degradation (DD) and hydraulic retention time (HRT) in 27 Swedish biogas plants.

Compared with degree of degradation, determination of digestibility (Eq. 3) is a more accurate method to evaluate degradable compounds, as digestibility takes the actual mass balance into account. The digestibility value is often used in feed-related research and is used in practice when formulating feeds for animals and to determine the mass balance in anaerobic biogas processes (II) (Rustas *et al.*, 2011; Triolo *et al.*, 2011; Mertens, 2007).

$$\text{Digestibility} = 100 \times \frac{m_{in} - m_{out}}{m_{in}} \quad (3.)$$

4.1.2 Residual methane potential

Residual methane potential (RMP) is defined as the amount of methane that it is possible to produce from the digestate. The RMP is a measure of the amount of methane that could have been obtained from a certain substrate if conditions had been optimal. It is also a measure of the potential risk of methane losses during storage of the digestate (Elsgaard *et al.*, 2016; Moset *et al.*, 2015; Rico *et al.*, 2011). Digestate composition, *i.e.* origin of substrate, organic loading rate and retention time, seem to have a high impact on the final RMP (III, IV) (Moset *et al.*, 2015; Seppala *et al.*, 2013). For the Swedish farm-scale plants studied in this thesis, only a few measurements of RMP had been performed. In Ahlberg Eliasson *et al.* (2017), the RMP at 11 farm-scale biogas plants was evaluated after 30 days of incubation and was found to be between 48 and 145 mL CH₄/g VS, corresponding to 25% of total gas production at these plants. In Denmark and Germany, the corresponding figures are 15% and 7%, respectively, of total gas production left in the digestate (Møller & Nielsen, 2015; FNR, 2010). Different ways to evaluate RMP are described in the literature. These differ in terms of *e.g.* the temperature applied for incubation and the type of inoculum used, which can impact on the final results (Moset *et al.*, 2015; Thygesen *et al.*, 2014). In this thesis, RMP was analysed using the same equipment as used for biochemical methane potential (BMP) evaluation. The temperature was set to the same level as the digester temperature, *i.e.* mesophilic or thermophilic temperature. In this way, the set-up can be said to assess the results of an increased retention time for the substrate (III and IV) (Ahlberg Eliasson *et al.*, 2017). The RMP of digestate originating from manure in co-digestion has been shown to be negatively correlated with hydraulic retention time (III) (Seppala *et al.*, 2013). Interestingly, in Paper III RMP was also shown to correlate positively with gas yield ($k=0.74$). This suggests a possible risk of increasing RMP value if more substrate is added in order to increase the gas production, resulting in an overall decrease in retention time and lower utilisation rates (III and IV). A previous study evaluating RMP in a process using co-digestion of crops and manure also observed an increase in RMP when the ratio of manure increased (Lindorfer *et al.*, 2008). Manure is a substrate with a high concentration of undegradable fibre, which may explain the observed overall decrease in degradation and higher RMP when the proportion of manure in the substrate mix increases. This suggests that improved degradation can be achieved by increasing the hydraulic retention time (II and III) (Moset *et al.*, 2015).

The production of residual methane can be used to calculate the efficiency of a process using Eq. 4 (Rico *et al.*, 2015). This method can be a promising way to assess efficiency in a biogas plant, since the calculation considers methane

production (MP), retention time (HRT) and residual methane yield (RMY) (**III** and **IV**) (Rico *et al.*, 2015).

$$\text{Efficiency \%} = 100 \times (MP \times HRT) \div (MP \times HRT + RMY) \quad (4.)$$

According to Eq. 4, the efficiency of a biogas production process is correlated negatively with its RMP ($k = -0.59$). In Paper **III**, the efficiency also showed a negative correlation with OLR ($k = -0.79$).

4.2 Co digestion and changes in operating parameters for improved efficiency

In co-digestion, substrates are used together to complement the nutrient composition, with the aim of maximising microorganism growth in the anaerobic digestion process (Mata-Alvarez *et al.*, 2014; Esposito *et al.*, 2012). The advantages of co-digestion in comparison with mono-digestion include *e.g.* increased volumetric biogas production, improved degradation and process stability, increased value of the digestate and overall improved efficiency (**I**, **II**, **III** and **IV**) (Tufaner & Avşar, 2016; Mata-Alvarez *et al.*, 2014; Esposito *et al.*, 2012).

If co-digestion improves the degradation of the substrates used, it can result in a so-called priming effect. Priming is suggested to be caused by easily degradable organic matter, such as pure fat or carbohydrates, resulting in increased production of enzymes that improve degradation of complex organic materials (Insam & Markt, 2016; Aichinger *et al.*, 2015). The priming effect, which is well described for soil, has been proposed to occur also in biogas processes, for example during co-digestion of sludge and protein rich whey (Insam & Markt, 2016; Aichinger *et al.*, 2015).

4.2.1 Operating parameters and impact on the degradation of manure

At farm-scale biogas plants in Sweden, the substrates most commonly used for co-digestion with manure are energy crops and crop residues, slaughterhouse waste, waste fat and different kinds of industrial waste products (**I**). The pros and cons of these substrates are explained in section 3.3.

When additional substrates are used for co-digestion with manure, the organic load (OLR) can increase. Depending on the character of the co-substrate, *i.e.* total solids content, the HRT often decreases as a consequence of the extra volume of substrate added to the digester on a daily basis. Several researchers

have described this negative relationship between OLR and HRT (**I**) (Tufaner & Avşar, 2016; Nizami *et al.*, 2009; Lindorfer *et al.*, 2008). The connection between HRT and OLR is also important for the efficiency value, as described in section 4.1.2, (**III**) (Rico *et al.*, 2015). In manure-based biogas systems, HRT has been shown to be positively correlated with specific methane potential (SMP), degree of degradation (DD) and gas yield (GP) (**I**). Likewise, OLR has been reported to show positive correlations with volumetric and daily methane production (**I** and **II**), (Miranda *et al.*, 2016; Labatut *et al.*, 2014; Angelidaki *et al.*, 2011). If this increase in OLR does not affect HRT to any great extent, even the DD can be improved by co-digestion (**II** and **IV**). As an example, increasing the OLR by addition of soy-wheat meal (a protein-rich substrate) in Paper **II** in this thesis resulted in increased gas production and a tendency for increased DD (Figure 11).

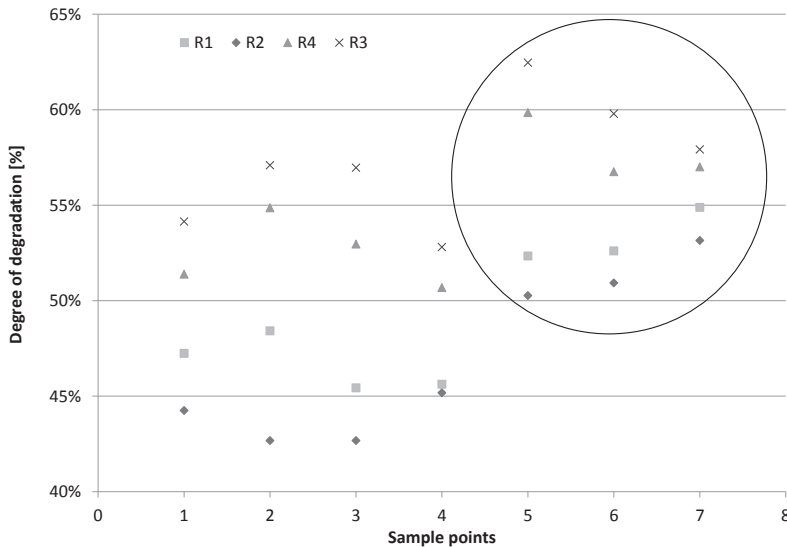


Figure 11. Degree of degradation (DD) in four laboratory-scale biogas reactors (R1-R4) operating with two different kinds of manure and energy crops, before and after (black circle) an increase in organic loading rate (OLR) (**II**).

Increasing the OLR by using only manure has been shown to increase gas production, even with shortened retention times, at both thermophilic and mesophilic temperature (Moset *et al.*, 2015). However, high OLR can also affect the degree of degradation, resulting in higher residual methane potential (**III**). This illustrates the trade-off effect between higher gas production due to

increased OLR and shortened HRT, on one hand, and decreased degree of degradation and high RMP of the digestate, on the other hand (**I** and **III**) (Moset *et al.*, 2015; Labatut *et al.*, 2014; Lindorfer *et al.*, 2008).

4.2.2 Example of how choice of co-substrate impact on gas production and nutrient concentration in digestate

Co-digestion can also be a way to improve the nutrient concentration in the digestate, such as giving a high ammonium level. This was for example shown for two biogas plants using different operational strategies (Table 2) (**I**). These two plants run with the main aim of producing biogas, but use different ways to economically optimise their biogas production (Table 2). Plant A is an organic dairy farm in need of organic fertiliser and thus optimises the biogas process to achieve high nitrogen concentrations in the digestate. Plant A uses poultry manure in co-digestion with cattle manure to obtain a relatively high final ammonium concentration, 3.0 kg/ton. The SMP in plant A is approx. 250 m³ CH₄/ton VS (Table 2). Plant B is a small producer of cattle (beef) and the biogas from this facility is sold as heat to a greenhouse production unit nearby. Consequently, the main economic output for this plant is to produce as much energy as possible, so a sugar-rich residue from a confectionary factory is used as co-substrate. This material is quickly degraded and can be added in comparatively high amounts, with 50/50 proportions of confectionary and manure on a volatile solids basis fed into the digester (**I**). The final SMP for this plant is high, 343 m³ CH₄/ton VS. However, the final ammonium level is only 1.9 kg/ton (Table 2). This example shows how the characteristics of the substrates used for co-digestion affect the values of the different outputs of the biogas process (Figure 9).

Table 2. Comparison of two operating strategies¹. Effects on the specific methane potential (SMP) and final total nitrogen (tot-N) and ammonium nitrogen (NH₄⁺-N) concentration in digestate².

Substrate / digestate	SMP	N-tot	NH ₄ ⁺ -N
Digestate from cattle manure	178	3.9	2.0
Digestate plant A	250	4.7	3.0
Digestate plant B	343	3.8	1.9

- 1) Substrate in plant A: cattle and poultry manure, substrate in plant B: cattle manure and residues from confectionary
- 2) Results from Paper I

To examine in depth this effect of different substrates, an increase in OLR of 0.5 g VS/L as protein, fat or carbohydrates, or a mixture of all these, in co-digestion with manure was evaluated (IV). The results showed that addition of protein increased the ammonium-nitrogen concentration in the digestate, but at the same time the digester was obstructed by high volatile fatty acid levels, causing disturbance and less efficient degradation in the process, *i.e.* a decrease in specific methane potential (SMP). The addition of pure fat resulted in an increase in both gas yield and SMP. The addition of starch (carbohydrate) also increased the gas yield compared with digestion with only manure. However, addition of fat and starch did not affect the ammonium content in the digestate (IV). Interestingly, the RMP did not differ to a great extent between the co-digestion substrate mixes, despite variations in degree of degradation (IV).

To conclude, co-digestion can be used for several purposes, *e.g.* to achieve increased gas production and improve the fertiliser value of the digestate. Important aspects to consider when selecting a co-substrate for farm-scale biogas production are:

- Aspects of hygienisation, according to prevailing rules and regulation systems (European Parliament, 2009).
- How the co-substrate will affect organic load and retention time, consequently influencing the environment and degradation in the digester, for example pH, concentration of fatty acids, gas quality and/or ammonium concentration (III and IV) (Tufaner & Avşar, 2016; Mata-Alvarez *et al.*, 2014; Pitk *et al.*, 2014).
- How the co-substrate will affect the overall efficiency and residual methane potential (III, and IV) (Rico *et al.*, 2011).
- Costs associated with the co-substrate, including costs of storage and transportation, and how the final economic outcome of the biogas plant can be affected by the increased cost of statutory systems when co-substrate is added and/by the higher revenue if the digestate value increases (Boldrin *et al.*, 2016; Pöschl *et al.*, 2010; Paavola & Rintala, 2008).

5 Future perspectives and implications for farm-scale biogas production

5.1 SWOT analysis for FSBP in Sweden

To summarise the potential and implications of the finding reported in this thesis for Swedish farm-scale biogas production, a SWOT analysis was performed (Table 3). A SWOT analysis can be used as a way to evaluate and describe the direction of new technology strategies, for example biogas development (Iglinski *et al.*, 2016). The SWOT factors are divided into four fields: Strength and Weaknesses, here relating to the internal development of the biogas plant, and Opportunities and Threats, here reflecting the external implications *e.g.* for society and the environment. In the SWOT performed in this thesis, no prioritisation was made between these factors. A few previous studies have performed a SWOT analysis with the aim of contributing to the farm-scale biogas sector (Ahlberg-Eliasson, 2015; Brudermann *et al.*, 2015; Martin, 2015). The results from these studies were included in the present analysis (Table 3). Moreover, others have conducted similar analyses on other sectors involved in future energy transition (Iglinski *et al.*, 2016; Wilson *et al.*, 2015), resulting in interesting conclusions that can apply to the biogas sector.

Table 3. *SWOT analysis of farm-scale biogas production in Sweden¹.*

<p>Strengths</p> <p>Self-sufficient and reduced energy costs ^{a, d, e}</p> <p>Utilisation of resources ^{a, b}</p> <p>Nutrient circulation ^{a, b, I-III}</p> <p>Known technology ^{d, e, I}</p> <p>Subsidies/support ^{d, III}</p> <p>New markets and products^d</p>	<p>Weaknesses</p> <p>Difficult economically/low profitability ^{a, d, I}</p> <p>Internal energy efficiency ^{a, b, e, d}</p> <p>Substrate costs and availability ^{b, e, c, IV}</p> <p>Investment costs ^{c, d}</p> <p>Technology ^{a, b, d}</p> <p>Low efficiency/utilisation ^{a, b, d, III}</p>
<p>Opportunities</p> <p>Climate change/environmental transition ^a</p> <p>Energy security ^b</p> <p>Circular economy ^{a, b}</p> <p>Development rural areas and regional development ^{a, b}</p> <p>New substrates ^c</p> <p>Available substrates</p> <p>Digestate regulations ^c</p>	<p>Threats</p> <p>Political decisions and market uncertainties ^{a, c, e}</p> <p>Subsidies ^{c, d, e, III}</p> <p>Energy prices ^d</p> <p>Food versus fuel ^{a, c}</p> <p>Other energy sources ^{c, b}</p> <p>Substrates price/supply ^c</p> <p>Digestate regulations ^c</p>

1) References: ^aBrudermann et al. (2015), ^bIglinski et al. (2016), ^cMartin (2015), ^dAhlberg-Eliasson (2015), ^eSchaper and Theuvsen (2007), ^{I, II, III, IV}Papers **I-IV** in this thesis

The strengths of farm-scale biogas plants, as mentioned earlier in this thesis, are the importance of nutrient circulation, self-sufficiency of energy and utilisation of available resources (**I** and **II**) (Nkoa, 2014; Pucker *et al.*, 2013; Börjesson & Berglund, 2007). Different outputs from farm-scale biogas plants can also be developed into new products, with potential to reach new markets. For example, emerging small-scale techniques for upgrading raw biogas to vehicle fuel are promising to use at farm-scale (Bauer *et al.*, 2013). Another example is valorisation of digestate, for example to biochar by pyrolysis, different chemicals or better developed fertiliser products (Buisman, 2017; Monlau *et al.*, 2015). Studies seeking better use of nutrients in the digestate, *e.g.* from recovery of struvite precipitate and different kinds of ammonia stripping, show promising results (Amini *et al.*, 2017; Kjerstadius *et al.*, 2017; De Vrieze *et al.*, 2016). Both the biogas plants evaluated in Paper **III** have recently invested in equipment for separation of digestate into a dry and a liquid fraction, resulting in a nitrogen-rich liquid fraction suitable for use as a fertiliser and a hygienised fibre fraction. This dried fraction shows promise for use as bedding in animal houses (Sheets *et al.*, 2015). Other promising technologies that can be developed into new economic values are power to gas and solutions for “energy on demand”, which can be one way to meet the market demand for electricity (Willegghems & Buysse, 2016; Lebuhn *et al.*, 2014). Opportunities that deserve highlighting include for example the value chain of local energy production and the

development of rural areas, including *e.g.* jobs and employment in the biogas sector (Iglinski *et al.*, 2016). For example, Germany and the UK, the countries with the highest biogas production in the EU, also have the highest number of employees in the biogas sector, approximately 34 000 and 4300, respectively. Another opportunity for farm-scale biogas production plants is the large amount of agricultural waste products still available for biogas production, thus representing potential for increased energy production (Sondergaard *et al.*, 2015).

The main weakness for the individual biogas plant at farm-scale is the difficult economic situation, mainly caused by problems with technology and low energy prices, especially for electricity (Ahlberg-Eliasson, 2015). Some of the existing farm-scale biogas plants in Sweden are based on co-digestion of energy-rich substrates together with manure (I). This co-digestion contributes to higher gas production, but also results in a dependence on external substrates. These substrates often result in higher internal costs of energy and transportation and require higher investment in *e.g.* pre-treatment methods and/or storage. Energy-rich substrates for co-digestion, such as energy crops, are important to increase the gas yield in existing plants. (I and IV). However, demand for these substrates can also result in a risk of competition between biogas producers and other sectors. This includes the demand from other energy suppliers when a substrate can be used for other energy transition systems (Martin, 2015). This aspect can be seen as an opportunity for agriculture to pursue diversification of production from biomass originating from *e.g.* energy crops. It can also result in a higher demand from other parts of the society for energy transition (EBA, 2013). The greenhouse gas impact of any biogas system depends on the technical system used, but also on the operating parameters.

As regards threats, government policies and rules, including subsidies regulations, are among the main factors that can create market uncertainties for farm-scale biogas plants (Brudermann *et al.*, 2015; Martin, 2015; Schaper & Theuvsen, 2007). For example, the Swedish subsidy for manure digestion is under discussion owing to uncertainties in the system used for calculation (III) (Ahlberg Eliasson & Birgersson, 2017). Braudmann *et al.* (2015) concluded that external factors, opportunities and threats have greater impacts on the farm-scale biogas sector than internal factors, strengths and weaknesses. This was also found in practical evaluation of Swedish farm-scale biogas plants during the period 2010-2015 (I) (Bergström-Nilsson *et al.*, 2016; Ahlberg-Eliasson, 2015).

5.2 Need of further research and knowledge

Overall knowledge about the three value streams in biogas production from agricultural substrates, *i.e.* the environmental, energy and digestate values, generally needs to be improved. Better information about these three value streams, as provided in this thesis, provides a more complete understanding of the biogas sector and can possibly also act as a good background for more detailed research. Many of the factors in the SWOT analysis summarised in Table 3 are related to the overall need for increased knowledge about the farm-scale biogas sector. One method to work for gradual improvements in the biogas sector is benchmarking, which involves knowledge transformation and transparency of information and data collected. In bench marking for biogas production, bottle necks can be identified and gradual improvements can be achieved over time (Lebuhn *et al.*, 2014; Yngvesson *et al.*, 2013) Different indicators can be useful for benchmarking at farm-scale biogas plants, for example biogas production per ton added substrate, gas quality, retention time, organic load and residual gas production (**I**, **II**, **III** and **IV**) (Liebetrau & Jacobi, 2016; Lebuhn *et al.*, 2014). Benchmarking can also identify possibilities for improvements and position the individual biogas plant within a larger system in need of several local systems (Olsson & Fallde, 2015).

Some of the farm-scale biogas plants evaluated in this thesis have limited possibilities regarding techniques and equipment used for measurements, and therefore new methods are required for accurate evaluation of these plants (Liebetrau & Jacobi, 2016; Ahlberg-Eliasson, 2015). In terms of gas quality, it is important to be able to measure the concentrations of methane, carbon dioxide, oxygen and hydrogen sulphide on a daily basis, in order to understand and assess gas production. During *e.g.* ammonia inhibition, the concentration of carbon dioxide increases in the raw biogas when the microbes are inhibited. This shift to a higher concentration of carbon dioxide occurs relatively quickly and can therefore be an important parameter to measure on a daily basis (**III**) (Schnürer, 2016; Schnürer *et al.*, 2016). Other important parameters for process evaluation are for example the concentration of volatile fatty acids, buffering capacity, pH and the concentrations of nutrients in the digestate and substrate. For the digestate and substrate, simplified methods to evaluate characteristics online are needed. Near-infrared reflectance (NIR) is under evaluation for estimation of nitrogen- and ammonium concentrations, and other parameters such as biomethane potential, with promising results (Finzi *et al.*, 2015; Triolo *et al.*, 2014; Stockl *et al.*, 2013). However, more development is needed before NIR methods can be routinely used for applications at biogas plants (Finzi *et al.*, 2015; Delin *et al.*, 2012).

Under the Swedish subsidy system for manure, there are requirements to assess real and potential methane losses at the biogas plant (Swedish Board of Agriculture, 2015). This is also requirements due to regulations at EU and national level and effective methods have been developed for biogas plants with high levels of equipment for measurement (Lantz, 2017). For small-scale biogas production units, appropriate technology and methods are in need of development, *e.g.* simpler and less expensive equipment and methods without risking the overall quality of the results.

To exploit the advantages with co-digestion, especially those explained in this thesis, more knowledge is needed about the effect of extra substrate, particularly the economic outcome since co-substrate often results in higher costs for transportation, pre-treatment and storage, but on the other hand can result in higher efficiency. In full-scale biogas plants there is a trade-off between investment cost in digester volume, with the aim of enabling long retention times, and the amount of gas that can be obtained from the extra volume added. Different kinds of two-step digestion system are one way to increase the retention time and improve degradation (Ruile *et al.*, 2015; Weiland, 2010).

To summarise, process evaluation and benchmarking can increase the value chain for biogas production. In the case of the digestate, where there is a need to find a market for a refined fertiliser, knowledge from both the supply and demand side needs to be taken into consideration (Figure 12). This includes *e.g.* indicators of the biogas process and consumer requirements. If this information is used appropriately, new products with a price level that suits the market can be created.

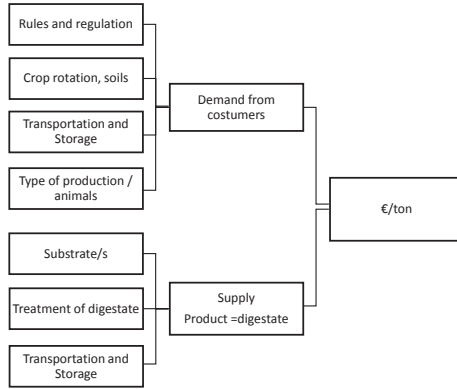


Figure 12. Examples of parameters from the supply and demand side in the case of digestate.

6 Conclusions

Agricultural farm-scale biogas production is a promising component of a local and national fossil-free energy supply. Biogas production fills a gap in the sustainable energy structure, as it can be stored and used in several different pathways, *e.g.* for fuel, heat conversion, electricity production and production of different refined products. In farm-scale biogas plants in Sweden, manure is the most common substrate. To increase the overall efficiency of the digestion process, co-substrates are often used. The components in substrate (proteins, fats and carbohydrates) can increase the three value streams denoting efficiency (digestate value, energy production and reduction of greenhouse gases) to differing extents when the substrate is co-digested with manure. One commonly used substrate in Swedish farm-scale plants is waste fat. Fat typically increases gas production, but does not improve the digestate value, *e.g.* ammonium concentration. The proportion of fat used in existing biogas plants is low, which probably explains why no major process disturbances, like foam and overfeeding, are seen in Swedish farm-scale plants. Substrates rich in starch, such as whole-crop silage, often result in increased gas production and can potentially also result in improved degradation. Substrates rich in proteins can cause process disturbance, due to a strong increase in volatile fatty acid concentrations as a result of ammonia inhibition. However, protein-rich substrates usually used in farm-scale biogas plants, such as poultry manure and slaughterhouse waste, contribute to high ammonium concentration in the digestate, increasing its value. Therefore there is a trade-off effect between increased biogas production and the risk of process failure when these substrates are used in co-digestion with manure.

Degradation of agricultural substrates is also influenced by operating conditions, such as retention time, organic load and temperature. Thermophilic temperature is promising to use if the substrate mix does not have too high proportion of proteins, as that raises the risk of ammonia inhibition in the process. The organic load in the manure-based system can be increased with

energy-rich co-substrates if the retention time is sufficient. The process operating parameters also have an impact on the untapped residual methane potential (RMP) of the digestate. There is a correlation between RMP and operating parameters such as retention time and load, which suggests that RMP can be a promising tool for assessing the efficiency of manure-based biogas production. In addition, RMP can be used to calculate the overall efficiency.

To further develop the biogas sector at farm-scale in Sweden, the positive and negative findings of the SWOT analysis in this thesis need to be taken into consideration when new investments and projects are planned. Specifically, overall knowledge about the sector needs to be improved, both by the sector itself and by authorities responsible for the subsidy and regulatory systems. To fully describe the multiple values of farm-scale biogas systems, all values of biogas production from agricultural waste streams, such as energy production, decreased greenhouse gas emissions and digestate value as fertiliser, are important. This way of assessing farm-scale biogas production systems can be used for the individual biogas plant, but also in a broader context when evaluating farm-scale biogas production for subsidy systems and formulating policies and regulations.

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Popular science summary

In Sweden, there are approximately 40 farm-scale biogas plants, using mainly manure as their substrate. Since biogas is a fossil-free energy source, this reduces greenhouse gas emissions from livestock-based agriculture. The digestate produced from the biogas process contains a higher proportion of ammonium-nitrogen than the unprocessed manure and this increase in fertiliser value is also positive for agriculture. Despite these benefits, Swedish biogas plants suffer from low profitability due to low energy prices and high maintenance and operating costs. A subsidy for biogas produced from manure was introduced in Sweden in 2015 and has improved the economic situation somewhat. Despite this, profitability is still low and therefore the overall efficiency, defined as increased gas production and higher plant-available ammonium-nitrogen in digestate, of existing biogas plants needs to be improved.

Anaerobic degradation of organic material into biogas (methane and carbon dioxide) involves complicated interactions between many different microorganisms that need the right composition of nutrients and right environment to produce the end product, biogas. Farmers operating biogas plants that digest manure can increase their production of biogas, and thus improve their efficiency, by mixing the manure with more energy-rich co-substrates. Gas production can also be increased by providing the right conditions for degradation of the specific substrates by adjusting the operating parameters in the digester. Using data from Swedish farm-scale biogas plants and from laboratory experiments on different substrates and operating parameters, this thesis examined how overall efficiency can be increased in farm-scale biogas production.

The results showed that co-digestion of manure and energy-rich substrates can increase overall efficiency in farm-scale biogas plants. Common substrates used for co-digestion include energy crops, slaughterhouse waste and waste fat. Energy crop substrates result in higher gas production, but may require a longer retention time for degradation to biogas than other substrates. Maize silage is currently commonly used for biogas production, but producing the maize crop

results in a relatively high environmental impact. In contrast, ley silage and whole-crop barley silage substrates have a low environmental impact and also contribute to increased gas production. Fat-rich substrates such as waste fat and protein-rich substrates such as slaughterhouse waste also increase gas production, but under certain conditions, e.g. high digester temperature, may cause disturbances in the digester.

The increased gas production observed during co-digestion of manure with fat- or starch-rich substrates is usually due to fast degradation of the easily degradable compounds in the co-substrate. The co-substrate also affects the nutrient concentration in the digestate. Substrates with a high nitrogen concentration, e.g. slaughterhouse waste, clover-rich ley biomass and chicken manure, contribute with more ammonium-nitrogen to the digestate. The final efficiency of the digestion process also depends on operating parameters such as temperature, how much material is fed into the biogas plant and how long the material remains in the digester. An important parameter investigated in this thesis is how much residual methane potential remains in the digestate after the biogas process. Future efforts to improve the overall efficiency of farm-scale biogas production must seek to increase the amount of fossil-free energy (biogas) produced, achieve better management of the digestate and reduce greenhouse gas emissions from agriculture.

Populärvetenskaplig sammanfattning

I Sverige finns idag fyrtiotalet biogasanläggningar som är lantbruksbaserade. Dessa producerar i huvudsak biogas av gödsel. Biogas är en fossilfri energikälla som därmed bidrar till att minska utsläppen av växthusgaser från lantbruket. Den biogödsel som bildas i biogasprocessen innehåller högre andel växttillgängligt kväve, jämfört med örötad gödsel, vilket är positivt för lantbruket. Trots alla fördelar har de svenska biogasanläggningarna under de senaste åren haft en tuff ekonomisk situation på grund av låga energipriser samt höga teknik- och driftskostnader. I Sverige finns sedan 2015 en subvention till biogas producerad av gödsel, vilket förbättrat den ekonomiska situationen något. Trots det finns det fortsatt lönsamhetsproblem inom sektorn och effektiviteten i befintliga biogasanläggningar behöver förbättras. Totaleffektiviteten i en biogasanläggning kan definieras just som ökad gasproduktion och högre växtnäringsinnehåll i biogödseln.

Den biologiska nedbrytningen av organiskt material till biogas (metan och koldioxid) sker i samspel mellan många olika mikroorganismer. De är i behov av rätt sammansättning näringsämnen och rätt miljö för att kunna producera biogas. Biogasanläggningar som rötar gödsel kan höja sin produktion av biogas och därmed nå en bättre ekonomi genom att blanda gödsel med andra energirika substrat. Ökad gasproduktion kan också nås genom att säkerställa rätt förutsättningar för nedbrytning av de material som tillförs genom att anpassa driftsparametrarna. Denna undersökning bygger på en utvärdering av svenska biogasanläggningar på lantbruk samt av laboratorieförsök där olika substrat och driftsparametrar studerats med målsättningen att visa på hur totaleffektiviteten kan höjas vid gödselrötning i befintliga biogasanläggningar.

Resultaten visar att samrötning mellan gödsel och energirika substrat kan höja totaleffektiviteten, jämfört om man bara rötar nöt- eller svingödsel. Vanliga material som används i samrötning är till exempel energigrödor, slakteriavfall och fettavskiljarslam. Energigrödor bidrar till högre gasproduktion men kan samtidigt kräva en längre tid för nedbrytning till biogas än om man bara rötar

gödsel. Traditionellt har majsensilage använts till biogasproduktion, majs har dock relativt hög miljöbelastning. Vallgrödor och helsädesensilage av till exempel korn eller rågvete, har också visat sig fungera bra i biogasanläggningar och har förhållandevis låg miljöbelastning. Andra energirika material som innehåller mycket protein till exempel slakteriavfall höjer gasproduktionen. Men kan under vissa förutsättningar orsaka driftstörningar i metanbildningen. Fettrika substrat till exempel fettavskiljarslam tillför mycket energi i samrötning och kan ge upphov till hög gasbildning. Den ökade gasbildningen vid samrötning orsakas vanligen av den snabba nedbrytningen av energin i samrötningssubstratet, det har i denna studie kunnat visas vid samrötning av nötgödsel med fett- och stärkelsesrika substrat. Samrötningssubstratet påverkar också näringsinnehållet i biogödsel. Kväverika substrat t.ex. slakteriavfall, energigrödor med hög klöverandel och kycklinggödsel, bidrar till mer ammoniumkväve i biogödseln. Hur väl samrötningen lyckas beror även på driftsparametrar såsom temperatur, hur mycket material som matas in i biogasanläggningen och hur länge materialet är kvar i röt-kammaren. En viktig parameter som har undersökts i denna studie är hur mycket biogaspotential det finns kvar i biogödseln efter rötning, så kallad restmetanpotential. För att uppnå en hög totaleffektivitet är det viktigt att så mycket som möjligt av potentialen i materialet tillvaratas. Det görs genom rätt förutsättningar i valet av driftsparametrar samt rätt sammansättning av substrat. För att långsiktigt kunna producera biogas på lantbruk i Sverige behöver hela värdekedjan lyftas. Värdekedjan består av; produktion av fossilfri energi, ett ökat växtnäringsvärde i biogödseln samt minskade utsläpp av växthusgaser från animalieproduktionen.

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