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Enhancing anaerobic digestion in urban wastewater management

DEPARTMENT OF CHEMICAL ENGINEERING | LUND UNIVERSITY
HAMSE KJERSTADIUS



Enhancing anaerobic digestion in urban wastewater management

Hamse Kjerstadius



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DOCTORAL DISSERTATION

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Abstract <p>The thesis investigates how anaerobic digestion could be utilized to improve wastewater management, specifically in regards to future expected regulation on sludge management in Sweden.</p> <p>Two possible paths of applying anaerobic digestion are investigated. First, the usage of thermophilic anaerobic digestion of sludge in order to achieve pathogen hygienization. Second, the usage of anaerobic digestion to treat wastewaters at decreased temperature. The evaluation of each path was made through practical lab scale experiments. Additionally, the benefits of each path was compared through desk top environmental impact studies and economic analysis.</p> <p>The results for the first path showed that thermophilic anaerobic digestion renders high pathogen hygienization even at relative short exposure times. However no additional beneficial impact on biogas production or the reduction of organic micropollutants was found. The results for the second path showed that the difficulty of operating the sensitive anaerobic digestion process at low temperatures can be partly overcome by simple engineering batch tests. Furthermore, the dissolved methane in the effluent wastewaters can be extracted using membrane contactors. Finally, the environmental impact assessment showed that increased resource recovery from wastewater, as well as decreased climate impact, can be achieved by applying anaerobic digestion on source separated domestic wastewater.</p> <p>The economic evaluation of the two paths showed that the implementation of source separation systems is expensive compared to implementing the needed thermophilic hygienization. However, source separation systems would greatly boost nutrient recovery from cities to agriculture which complies well with the goals of the Swedish Environmental Protection Agency.</p>		
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Enhancing anaerobic digestion in urban wastewater management

Hamse Kjerstadius



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*Av var och en efter förmåga,
åt var och en efter behov.*

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Lastly, my love goes out to my friend Duvan all the way over in California, thank you for your never ending support and all the adventures we have had during the past five years!

Abstract

The thesis investigates how anaerobic digestion could be utilized to improve wastewater management, specifically in regards to future expected regulation on sludge management in Sweden.

Two possible paths of applying anaerobic digestion are investigated. First, the usage of thermophilic anaerobic digestion of sludge in order to achieve pathogen hygienization. Second, the usage of anaerobic digestion to treat wastewaters at decreased temperature. The evaluation of each path was made through practical lab scale experiments. Additionally, the benefits of each path was compared through desk top environmental impact studies and economic cost assessment.

The results for the first path showed that thermophilic anaerobic digestion renders high pathogen hygienization even at relative short exposure times. However no additional beneficial impact on biogas production or the reduction of organic micropollutants was found. The results for the second path showed that the difficulty of operating the sensitive anaerobic digestion process at low temperatures can be partly overcome by simple engineering batch tests. Furthermore, the dissolved methane in the effluent wastewaters can be extracted using membrane contactors. Finally, the environmental impact assessment showed that increased resource recovery from wastewater, as well as decreased climate impact, can be achieved by applying anaerobic digestion on source separated domestic wastewater.

The economic assessment of the two paths showed that the implementation of source separation systems is expensive compared to implementing the needed thermophilic hygienization. However, source separation systems would greatly boost nutrient recovery from cities to agriculture which complies well with the goals of the Swedish Environmental Protection Agency.

Populärvetenskaplig sammanfattning

De näringsämnen som vi får i oss genom maten hamnar till slut i våra toaletter och förs till stadens avloppsreningsverk. Eftersom näringsämnen, som till exempel kväve och fosfor, är grundämnen finns det en möjlighet att återanvända dem som gödselprodukt på vår åkermark. Detta sker också i mindre utsträckning redan idag då 25 procent av allt avloppsslam som produceras återförs till jordbruksmark. Återförelse av slam är dock omdiskuterat eftersom avloppsslam, förutom de nyttiga näringsämnena, även innehåller tungmetaller, läkemedelsrester och andra kemikalier vars långtidseffekt på jordbruksmarken och människors hälsa är oklar. Naturvårdsverket har därför lagt fram ett förslag för skärpta regler som sätter gränsvärden för hur mycket tungmetaller och patogener (sjukdomsspridande mikroorganismer) som avloppsslam får innehålla om det skall användas som gödsel.

Den här avhandlingen undersöker hur avloppshanteringen kan ändras för att bättre svara mot Naturvårdsverkets föreslagna krav. Specifikt så undersöktes hur anaeroba (syrefria) processer kan appliceras för detta mål. Anaerob nedbrytning fungerar ungefär som en luftfri kompost, där avsaknaden av syre ser till att biogas bildas av de närvarande mikroorganismerna. Anaeroba mikroorganismer används redan idag vid avloppsreningsverk för att bryta ned organiskt material i avloppsslam och på så vis bilda biogas, ett klimatsmart biobränsle. Processerna är dock långt från optimerade.

Avhandlingen visar att man genom att öka drifttemperaturen på den anaeroba processen till 55-60 °C kan uppnå en fullgod minskning av patogener i avloppsslammet. Däremot verkar inte en ökad drifttemperatur ge någon ytterligare positiv effekt, såsom ökad biogasproduktion eller nedbrytning av läkemedelsrester. Det är också oklart om det räcker med en höjning av drifttemperaturen för att nå naturvårdsverkets mål. Avhandlingen visar därför hur ett alternativt sätt att nå naturvårdsverkets mål är att implementera källsorterande avloppssystem i våra städer. Med källsorterande avloppssystem separeras toalettavlopp från stadens övriga avlopp. Detta är bra eftersom toalettavlopp innehåller i stort sett allt organiskt material och näringsgämnena som vi släpper ut från våra hushållsavlopp. Genom att tillämpa anaeroba processer på källsorterat avlopp kan vi öka biogasproduktionen från avlopp samtidigt som vi ökar näringsgättervinnningen från städer. Kanske kommer vi i framtiden att få se hur källsorterande system införs i nya stadsdelar för att öka näringsåterförslaget tillräckligt mycket för att nå Naturvårdsverkets mål.

Källsorterande system håller redan nu på att införas i Helsingborg inom stadsrenoveringsprojektet H+ och pilotområden med källsorterande system är under uppbyggnad i städerna Hamburg, Amsterdam och Gent.

Avhandlingen tydliggör effekterna av två alternativa sätt att möta Naturvårdsverkets föreslagna krav. Genom att jämföra de två alternativen (ökad drifttemperatur eller införande av källsorterande avloppssystem) och tydligt presentera deras klimatpåverkan, möjligheter till näringsåtervinning samt deras ekonomiska kostnad så ges en god helhetsbild av vilka effekter som skulle följa om vi väljer endera vägen. På så vis bidrar avhandlingen med användbara resultat för planering av samhällets VA-system samt för stadsplanerare som vill bidra till ett ökat kretslopp mellan stad och land.

List of papers

This thesis comprises the following original papers, which are referred to in the text by their Roman numerals I-VI.

Paper I Kjerstadius, H., la Cour Jansen, J., de Vrieze, J., Haghghatafshar, S. & Davidsson, Å. (2013). Hygienization of sludge through anaerobic digestion at 35, 55 and 60 °C. *Water science & Technology*, 68(10), 2234-2239.

Paper II Davidsson, Å. Kjerstadius, H., Haghghatafshar, S., Fick, J., Olsson, M., Wachtmeister, H., Eriksson, E., la Cour Jansen, J. (2014). Effect of anaerobic digestion at 35, 55 and 60 °C on pharmaceuticals and organic contaminants. *Water science & Technology*, 69(6), 1282-1288.

Paper III Kjerstadius, H., de Vrieze, J., la Cour Jansen, J. & Davidsson, Å. (2016). Detection of acidification limit in anaerobic membrane bioreactors at ambient temperature. *Water Research*, 106, 429-438.

Paper IV Jahne-Kuchinke, S., Kjerstadius, H., Schumacher, M., Davidsson, Å., Modigell, M. (2016). Recovery of dissolved methane from anaerobic membrane bioreactor effluent using novel technology. *Proceedings of the 2nd IWA Conference on Holistic Sludge management*, 7-9 June 2016, Malmö, Sweden.

Paper V Kjerstadius, H., Haghghatafshar, S., Davidsson, Å. (2015). Potential for nutrient recovery and biogas production from blackwater, food waste and greywater in urban source control systems, *Environmental Technology*, 36(13), 1707-1720.

Paper VI Kjerstadius, H., Bernstad Saraiva, A., Spångberg, J. (submitted manuscript). Carbon footprint of urban source separation for nutrient recovery, *Environmental management* (submitted manuscript).

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My contribution to the papers

Paper I I designed the experimental plan together with Åsa Davidsson and Jes la Cour Jansen. I conducted the experimental work, with exception of the pathogens analysis which was performed by the National Veterinary Institute of Sweden. I interpreted all results and wrote the manuscript with input from my co-authors.

Paper II I designed the experimental plan together with Åsa Davidsson and Jes la Cour Jansen. I conducted the experimental work with exception of the analysis for micropollutants which was performed by the Department of Chemistry at Umeå University and the analysis for PAHs which was performed by the Department of Environmental Engineering at the Technical University of Denmark. I interpreted all results together with Åsa Davidsson and gave input to the manuscript.

Paper III I designed the experimental plan and design of the reactors together with input from Åsa Davidsson and Jes la Cour Jansen. I conducted the experimental work and analysis with help from the master thesis students acknowledged in the paper. The exception is the microbial analysis which was performed by the Center for Microbial Ecology and Technology at Ghent University. I interpreted all results and wrote the manuscript with input from my co-authors.

Paper IV I designed the experimental plan and set-up together with Sandra Jahne-Kuchinke. I conducted the main part of the experimental work and analysis together with help from Sandra Jahne-Kuchinke. I interpreted all results together with Sandra Jahne-Kuchinke and wrote parts of the manuscript.

Paper V I designed the study with input from Åsa Davidsson. I conducted all of the work. I interpreted the results with help from Åsa Davidsson. I wrote the manuscript with input from my co-authors.

Paper VI I designed the study with input from my co-authors. I conducted the main part of the work, with exception for the parts on food waste management and impact of replaced processes (both conducted by Anna Bernstad Saraiva) as well as the part on return to agriculture (conducted by Johanna Spångberg). I interpreted all results and wrote the manuscript with input from my co-authors.

Related publications

Kjerstadius, H., la Cour Jansen, J., Stålhandske, L., Eriksson, E., Olsson, M., Davidsson, Å., (2012). Rötning av avloppsslam vid 35, 55 och 60 °C (digestion of sewage sludge at 35, 55 and 60 °C), Report 2012-15, The Swedish Water and Wastewater Association. Stockholm

Kjerstadius, H., Bernstad Saraiva, A., Spångberg, J. (2016). Can source separation increase sustainability of sanitation management? Report Nr. 05 2016, VA-Teknik Södra, Lund.

Kärrman, E., Kjerstadius, H., Hagman, M., Davidsson, Å. (2017). Källsorterande system för spillvatten och matavfall – erfarenheter, genomförande, ekonomi och samhällsnytta (source separation systems for wastewater and food waste – experience, implementation, economy and societal benefit), Report, The Swedish Water and Wastewater Association, Stockholm.

Skambraks, A.-K., Kjerstadius, H., Meier, M., Davidsson, Å., Wuttke, M. & Giese, T. (2017) Source separation sewage systems as a trend in urban wastewater management: drivers for the implementation of pilot areas in Northern Europe. *Sustainable Cities and Society*, 28, 287-296.

List of abbreviations

AD	Anaerobic digestion
AnMBR	Anaerobic membrane bioreactor
AS	Activated sludge
BMP	Biomethane potential
BOD	Biochemical oxygen demand
COD	Chemical oxygen demand
CSTR	Continuous stirred tank reactor
FAC	Fatty acid capacity
FID	Flame ionization detector
FW	Food waste
HFMC	Hollow fiber membrane contactor
HRT	Hydraulic retention time
LCA	Life cycle assessment
LOQ	Limit of quantification
MET	Minimum exposure time
OLR	Organic loading rate
Pe	Person equivalent
SEK	Swedish krona
SEPA	Swedish environmental protection agency
SMA	Specific methanogenic activity
SRT	Solids retention time
TCD	Thermal conductivity detector
TS	Total solids
UASB-ST	Upflow anaerobic sludge blanket septic tank
VS	Volatile solids
VSS	Volatile suspended solids
WWTP	Wastewater treatment plant

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1. Introduction

Wastewater management has been a necessity since the emergence of densely populated cities. Water-based sewer systems were introduced to remove excrement from such urban areas to mitigate the spread of disease. The usage of sewer systems was very successful for disease control; however, it created environmental problems due to the discharge of the collected wastewaters at single point sources, causing a large impact on the receiving water bodies. To decrease the impact, wastewater treatment was introduced on a large scale for urban areas in Europe during the 20th century.

The core treatment of wastewater is focused on the removal of solids, organic material and nutrients from the water flow. Removal is performed via mechanical, chemical or microbiological processes, and the end products are discharged as atmospheric gases or as solids in sludge (Figure 1.1). Through these removal processes, wastewater treatment plants produce water that can be discharged into the receiving water body. The produced sludge fraction, containing solid material and nutrients, needs treatment and final disposal.

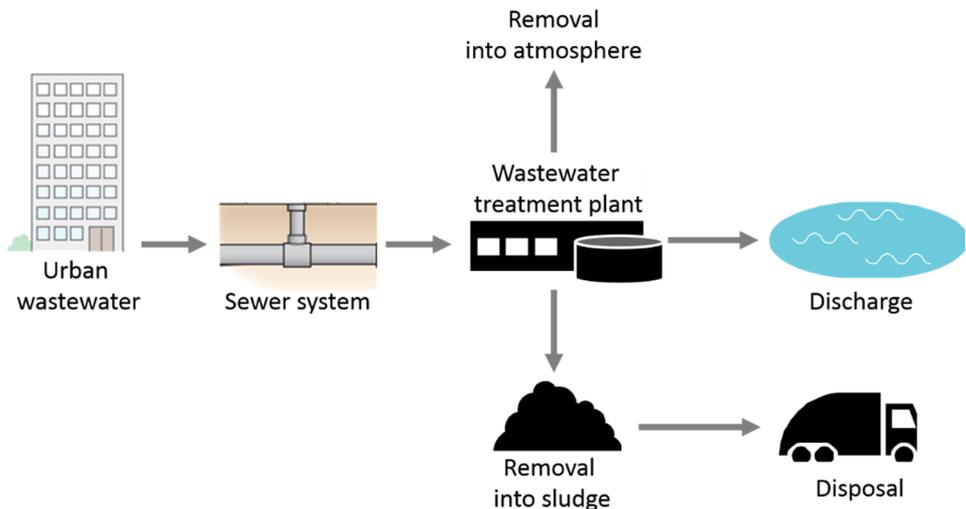


Figure 1.1.
Overall process of urban wastewater management.

Sludge management was introduced as an auxiliary process to stabilize the produced sludge, i.e., reduce its volume and activity, before final disposal. Microbiologically mediated anaerobic digestion has long been a favored method for sludge stabilization. During digestion, which is normally performed at 35-37 °C to achieve sufficiently high reaction rates, organic material is degraded in several conversion steps. As the end product, anaerobic digestion produces biogas, an energy rich biofuel. In Sweden, biogas is utilized to reduce climate impact by replacing fossil energy as a vehicle fuel. The remaining stabilized sludge requires disposal and has to some extent been used as fertilizer in agriculture. This practice may appear suitable since sludge contains some of the nutrients found in human feces and urine. However, because urban wastewater is a mixture of many input sources, the sludge also contains metals and organic micropollutants that are potentially unsuitable for return to agriculture.

The Swedish Environmental Protection Agency (SEPA) has been working on a more stringent regulation for the management of sludge intended for return to agriculture during the past 15 years (SEPA, 2013, 2010, 2002). Their latest regulation proposal (SEPA, 2013) contains demands for sludge hygienization (pathogen destruction), limits for metal content and target goals for the return of nutrients from wastewater to farmland. Since similar limits and goals have been mentioned in all three published regulation proposals (SEPA, 2013, 2010, 2002), it is likely that such demands will be part of a future legislation on sludge management. Due to anaerobic digestion being the most common method for sludge treatment in Sweden, this legislation will impact the application of anaerobic digestion in wastewater management. It is thus of high interest to investigate how anaerobic digestion can be enhanced to comply with expected future demands. Such investigations of anaerobic digestion systems should cover both the required adjustments to existing systems and new applications of anaerobic digestion that may comply with the expected sludge regulation as well as providing further benefits.

A specific path of interest is the effects of increased operational temperature up to the thermophilic range for existing anaerobic digestion systems utilized as auxiliary processes at wastewater treatment plants. Since demand for sludge hygienization seems likely, an increased operational temperature would be required to achieve satisfactory pathogen destruction (Gray and Hake, 2004; Sahlström, 2003). An increase in operational temperature could also have a potential beneficial effect on biogas production (De Vrieze et al., 2016; Davidsson, 2007) or the degradation of organic micropollutants (Cirja et al., 2008; Grandclément et al., 2017) due to the increased microbial conversion rates at elevated temperatures. Although the potential impact of organic micropollutants present in sludge returned to agriculture is unknown (SEPA, 2013), any reduction would be beneficial since it might mitigate the potential risks (SEPA, 2008).

However, changes to the present application of anaerobic digestion may not be sufficient to meet both the expected demands for low metal content in sludge and the suggested goals for nutrient recovery. If implemented, target goals for nutrient recovery could create a need for a shift of focus from removal to recovery in Swedish wastewater management.

An alternative path of interest is to investigate how anaerobic digestion could be utilized as a core process to facilitate nutrient recovery. Applying anaerobic digestion as a core process by directly treating wastewater rather than sludge has long been suggested as a possible path to achieve more energy efficient wastewater treatment (Schink, 1988; Zeeman et al., 2008; Zeeman and Lettinga, 1999). Applying anaerobic digestion as a core process has the benefits of recovering energy as biogas and mineralizing nutrients, which facilitates their extraction. In contrast, energy is spent to remove nutrients in the current core process of wastewater treatment, the activated sludge process (Gikas, 2016). However, applying anaerobic digestion as a core process faces challenges. Due to the low concentration of organic material in wastewater, compared to sludge, it is less energetically favorable to heat the digestion process to the normally used mesophilic optimum of 35-37 °C. Thus, anaerobic digestion of wastewater should be performed at lower temperatures, which creates operational challenges due to the decreased reaction rates of the microbiological conversions involved (Petropoulos et al., 2016). This issue could be partially overcome by using source separation of wastewaters, for which anaerobic digestion would be used to treat only the wastewaters with higher concentrations of organic material (Otterpohl et al., 1999; Zeeman et al., 2008).

Finally, the potential gain of either path needs to be weighed against the potential environmental impact and associated economic cost of their implementation. Comparison of these two paths can assess whether either of these changes to our wastewater systems is more likely to enable efficient management and answer to our needs.

1.2 Aim

The overall aim of this thesis was to evaluate how anaerobic digestion can be enhanced as an auxiliary or core process in Swedish wastewater management. The evaluation was performed especially in regards to expected future legislation.

Particular attention was given to i) thermophilic anaerobic digestion, ii) stability of low-temperature anaerobic digestion and iii) source separation systems with anaerobic digestion as core process.

The following hypotheses were made:

Hypothesis #1: Thermophilic anaerobic digestion achieves sufficient hygienization of sludge to comply with the proposed SEPA regulation while having beneficial effects on biogas production and the removal of organic micropollutants.

Hypothesis #2: The operational challenges of utilizing anaerobic digestion as a core process at low temperatures can be overcome by engineering solutions.

Hypothesis #3: Source separation systems with anaerobic digestion as core process will increase the sustainability of wastewater management in regards to climate impact and nutrient recovery.

1.3 Outline of thesis

To achieve the aim of the thesis, two possible future paths were investigated (Figure 1.2).

In the first path, anaerobic digestion remains an auxiliary process in wastewater management and is utilized to treat sludge. The work within the path (**Paper I and II**) was centered on practical anaerobic digestion experiments at thermophilic temperature.

Paper I investigates the effect of 55 and 60 °C thermophilic anaerobic digestion on process performance and the hygienization of sludge. Comparison is made to the proposed SEPA regulation and to results for 37 °C mesophilic conditions.

Paper II evaluates the reduction of organic micropollutants in sludge through thermophilic anaerobic digestion at 55 and 60 °C to determine whether substantial reduction is achieved.

In the second path, anaerobic digestion is used as a core treatment process for wastewater, treating wastewater with little or no prior treatment. This implies changes in wastewater management, especially in regards to implementing source separation systems for domestic wastewater in urban areas. The research within the path (**Paper III and IV**) was centered on practical anaerobic digestion experiments at low (24 °C) temperature, which was deemed representative of the challenge of treating more dilute wastewaters with little or no heating.

Paper III proposes and evaluates a method for identifying a safe loading rate to avoid failure of the microbial anaerobic digestion process due to over-loading.

Paper IV evaluates the use of membrane extraction of dissolved methane from reactor effluent to achieve reduced climate impact from low-temperature anaerobic digestion.

Paper V and VI, representing a broader perspective, compare the path using anaerobic digestion as an auxiliary process to the path applying anaerobic digestion as a core process.

Paper V is a desk top comparison of the potential for nutrient recovery and biogas production for a conventional system and a source separation system, representing the two paths in Figure 1.2.

Paper VI is a life cycle assessment of the climate impact of two of the systems from **Paper V**, aimed at identifying the most important issues in regards to climate impact of wastewater management for a conventional and a source separation system.

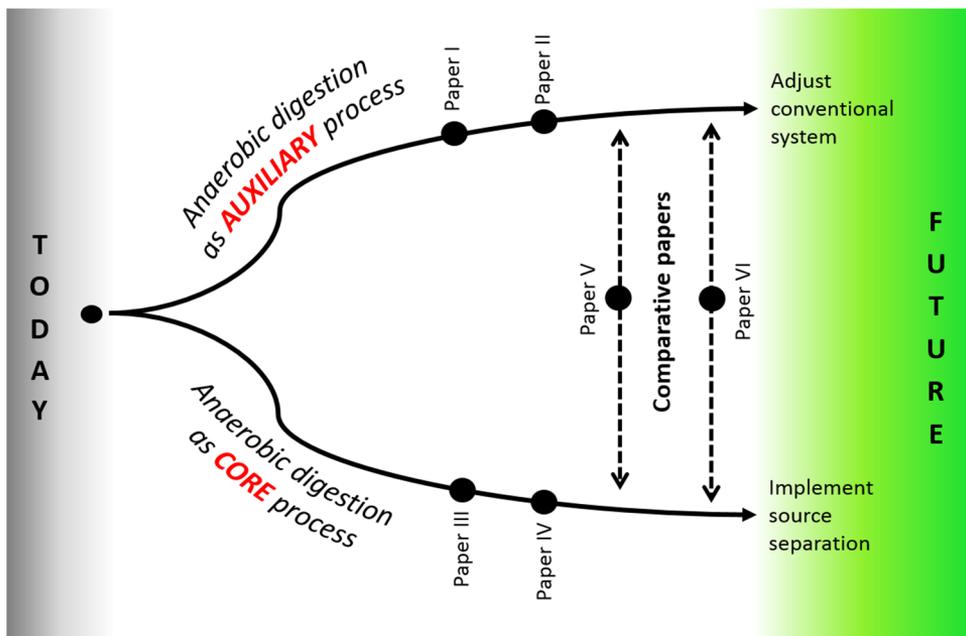


Figure 1.2.
Graphical outline of the thesis structure.

1.4 Delimitations

The following delimitations to the thesis work should be stated.

First, due to the proposed SEPA (2013) regulation being published during this thesis work, the research in **Papers I and II** is based on the proposed SEPA (2010) regulation. The practical implication is that no limits for organic micropollutants in sludge were suggested in the SEPA (2010) proposal. This explains why **Paper II** focuses on a broad spectrum of micropollutants rather than those presented in the proposed SEPA (2013) regulation.

Second, this thesis focuses on urban wastewater management in, and technological solutions relevant for, Southern Sweden and equivalent areas only. Thus, the systems selected to represent a conventional and a source separation system in **Paper V and VI** may not be suitable elsewhere.

Third, the assumed discharge concentration limits in **Paper VI** (10 mg N L^{-1} and 0.5 mg P L^{-1}) are selected to represent a reasonable tradeoff between current standard limits in Southern Sweden and discharge limits in other northern European countries. Thus, the results in **Paper VI** should be seen as comparative rather than as an absolute truth for which specific discharge demands need to be known.

Lastly, the cost evaluation presented in the discussion aims to highlight only the magnitude of costs involved, not the actual cost, which will depend on local prerequisites. Since the work in Kärman et al. (2017) clearly indicates large variation in the reported assessment of economic costs for wastewater systems, the results from this report should only be seen as an approximation and used to highlight differences between the investigated systems and other economic costs.

2. Anaerobic digestion in wastewater management

2.1 Urban wastewater management

2.1.1 The current goals of urban wastewater management

Conceptually, conventional urban wastewater management has two goals. First, to mitigate the spread of disease by removing excrement from urban areas. This goal was achieved in Sweden by implementing sewer systems on a large scale in the first half of the 1900s (SEPA, 2014). The second goal is to protect recipient waters from deterioration and eutrophication. This was achieved through the implementation of removal steps for organic material and nutrients from wastewater treatment plants (WWTPs) on a broad scale during the second half of the 1900s (SEPA, 2014). Due to these implementations, conventional urban wastewater management has three characteristics i) a sewer net that mixes all urban wastewaters; ii) a centralized WWTP; and iii) treatment at the WWTP is focused on removal.

2.1.2 The conventional wastewater treatment plant

A conceptual layout of a conventional wastewater treatment plant, together with a mass balance of selected constituents of wastewater, is presented in Figure 2.1. Wastewater treatment at a conventional WWTP includes mechanical treatment (screens, grit chamber and primary settling) and biological treatment with activated sludge (AS) and secondary settling. AS is defined as nitrification/denitrification microbiological processes occurring with suspended sludge in aerated and anoxic tanks, as described elsewhere (Ekama and Wentzel, 2008). The sludge produced during primary and secondary settling is treated by anaerobic digestion (AD). Of the two microbiological processes, AS is the core process used to treat the main water flow, while AD is an auxiliary process implemented to minimize and stabilize the produced sludge. In a conventional wastewater treatment plant, nitrogen is mainly removed by nitrification/denitrification in the AS (Ekama and Wentzel, 2008; SEPA, 2014), while a smaller fraction ends up in the produced sludge or is

discharged in the effluent wastewater. Phosphorus is mainly removed by chemical precipitation, which may be performed during mechanical (pre-precipitation) or biological (simultaneous precipitation) treatment using coagulating agents, commonly ferric or aluminum salts (Lindquist, 2003). Additional phosphorus removal may also be achieved in the effluent water from biological treatment (post-precipitation). The precipitated phosphorus ends up in the dewatered sludge. Organic material, represented by chemical oxygen demand (COD), is partly removed by microbial degradation in the AS and by anaerobic digestion, while the remains end up in the dewatered sludge or are discharged in the effluent wastewater.

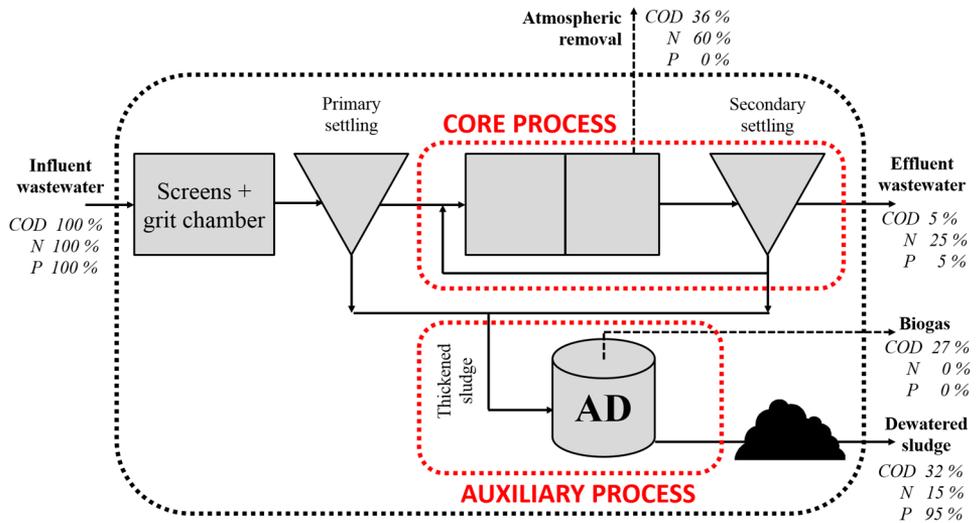


Figure 2.1. Mass balance over a conceptual layout of a conventional wastewater treatment plant with activated sludge (AS) as the core process and anaerobic digestion (AD) as an auxiliary process to stabilize the produced sludge. The black dotted line indicates the mass balance boundary. The mass balance is based on Siegrist et al. (2008) for COD and nitrogen (N) and **Paper V** for phosphorus (P).

2.2 Sludge management and anaerobic digestion

2.2.1 Sludge production and treatment

Sludge is a byproduct of urban wastewater management and is generated mainly from organic material during primary settling and from excess biomass during secondary settling. To decrease the amount of organic material in sludge and its microbiological activity, sludge is stabilized via anaerobic digestion.

2.2.2 The anaerobic digestion process

Anaerobic digestion consists of several microbiologically mediated processes. The most relevant processes for this thesis are hydrolysis, acidogenesis, acetogenesis and methanogenesis, as schematically indicated in Figure 2.2. Via these metabolic steps, larger organic molecules are degraded to intermediates and finally to the end products methane and carbon dioxide. Due to the anaerobic conditions, the Gibbs free energy of the microbial conversions is generally low compared to aerobic conditions, and individual reactions may occur close to energetic equilibrium (McCarty and Mosey, 1991). The exemption is acidogenesis, which generally has the highest Gibbs free energy and conversion rates of the processes presented in Figure 2.2 (Van Lier et al., 2008). As a consequence of the relatively rapid acidogenesis, anaerobic digestion processes are sensitive to over-loading, which causes accumulation of volatile fatty acids. At sufficient concentrations, the accumulation inhibits methanogenesis and subsequently causes process deterioration. To increase the low growth rates of anaerobic microorganisms, anaerobic digestion processes are commonly performed at mesophilic (35-37 °C) temperature in Swedish WWTPs. Compared to the aerated microbiological processes in the activated sludge process, anaerobic digestion produces relatively little excess sludge, and the main fraction of energy is recovered as methane rather than biomass and heat, as in activated sludge (Schink, 1988).

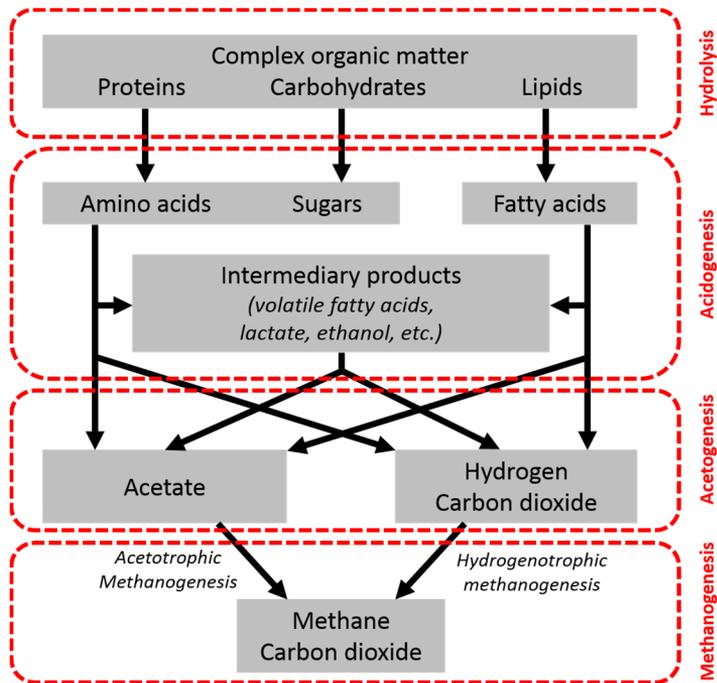


Figure 2.2. Selected anaerobic digestion processes that are relevant to this thesis. Image based on Khanal (2008) with permission by Wiley-Blackwell.

2.2.3 Sludge disposal

The stabilized sludge is dewatered and transported from the WWTP for disposal. In Sweden, sludge is primarily (Statistics Sweden, 2016) used for soil production (29%), returned to agriculture (25%) or used to cover landfills (24%). The return of sludge to agriculture is beneficial in regards to nutrients (mainly nitrogen and phosphorus) present in the sludge. However, sludge also contains unwanted metals and organic micropollutants, and sludge use in agriculture is heavily debated in Sweden, causing a relative low return (25%) compared to the European Union average (>35%) (Bengtsson and Tillman, 2004; Börjesson et al., 2014; Linderholm et al., 2012; Wiechmann et al., 2013). To increase sludge quality and achieve a higher rate of sludge return to agriculture, the Swedish Water and Wastewater Association (SWWA) initiated a voluntary certification system for sludge, termed Revaq, in 2008 (SEPA, 2013), together with the Federation of Swedish Farmers. Approximately 50% of the Swedish population is connected to WWTPs with Revaq certification, and these plants supply most of the sludge that is currently being returned to agriculture (Mattsson and Finnson, 2016).

2.3 Expected regulation and goals

The Swedish Environmental Protection Agency (SEPA) is presently working on a more stringent regulation for sludge return to agriculture (SEPA, 2013). The main purpose of the regulation is to ensure that phosphorus from waste and wastewater fractions can be returned to agriculture or productive land without risk to human health or the environment. SEPA also states that the regulation is part of the work to fulfill Swedish national environmental objectives, particularly the goals of “A non-toxic environment” and “Zero eutrophication” (SEPA, 2013). To contribute to the objective of “A non-toxic environment”, demands are suggested for sludge hygienization as well as concentration limits for metals and selected micropollutants. In relation to the environmental objective of “Zero eutrophication”, the proposed regulation contain target goals for the recovery of nutrients from wastewater, which would decrease mineral fertilizer imports.

2.3.1 SEPA proposed regulation for sludge reuse in agriculture

For sludge intended for application in agriculture, the latest version of the proposed SEPA regulation includes i) stricter demands for metal content, ii) demands for pathogen hygienization, and iii) target goals for nutrient recovery from wastewater, as presented in Table 2.1.

Table 2.1.

Suggested concentration limits and target goals in the proposed SEPA (2013) regulation. TS=Total solids.

Target goals for nutrient recovery from wastewater	Concentration limits of pathogens	Concentration limits of metals from the year 2030 [mg kg TS ⁻¹]
40% of phosphorus	<i>E. coli</i>	Pb [25]
	[<3 log ₁₀ g TS ⁻¹]	Cd [0.8]
		Cu [475]
10% of nitrogen	<i>Enterococcus</i>	Cr [35]
	[<3 log ₁₀ g TS ⁻¹]	Hg [0.6]
		Ni [30]
	Salmonella	Ag [3]
	[Absent in samples of 25 g]	Zn [800]

2.3.2 How the SEPA proposal affects sludge management

The SEPA regulation proposal includes suggested methods to reach the hygienization limits for pathogens. These methods include thermophilic anaerobic digestion with minimum exposure times and pasteurization (**Paper I**). Since a majority of Swedish WWTPs are operated at mesophilic temperature and do not currently include pasteurization, management of sludge intended for reuse in agriculture will have to change if the regulation proposal is accepted by the Swedish parliament. However, the proposal does not include suggested methods to reach the concentration limits for metals or the target goals for nutrient recovery. As will be made evident by this thesis, it is unclear whether changes to the conventional system alone will be sufficient to meet all the SEPA demands presented in Table 2.1. Thus, implementing anaerobic digestion as core process on source separated wastewater, as investigated in hypothesis #3, may be a suitable method to achieve the suggested target goals for nutrient recovery.

2.4 Anaerobic digestion as core process

In conventional wastewater treatment, suspended activated sludge (AS) systems are the core process, achieving removal of organic material and nitrogen via nitrification and denitrification. In addition to the obvious drawback of achieving nutrient removal rather than recovery, activated sludge systems are energy intensive due to aeration requirements, produce large amounts of sludge and emit nitrous oxide (Gikas, 2016; Gustavsson and Tumlin, 2013; Schink, 1988). In contrast, the introduction of anaerobic digestion as the core process in wastewater (Figure 2.3 right) to replace the AS processes would potentially enhance the energy balance and reduce the climate impact of wastewater management (Remy, 2010; Schink, 1988). The main challenge of applying anaerobic digestion as a core process is the low concentration of organic material in wastewater compared to thickened sludge digested in systems with anaerobic digestion as an auxiliary process (Figure 2.3 left). The low concentration in wastewater, as compared to thickened sludge, makes heating of the anaerobic process to standard mesophilic digestion (35-37 °C) less energetically favorable. Thus, low-temperature anaerobic digestion with little or no heating would be required. A further challenge is reaching the discharge limits for nitrogen and phosphorus. Due to the exclusion of stormwater from source separated wastewater, dilution is low, and reaching discharge limits (assumed to be 10 mg N L⁻¹ and 0.5 mg P L⁻¹ in Kjerstadius et al., 2016) would require a polishing step after nutrient recovery, as indicated in Figure 2.3, to remove the excess nutrients. Polishing could consist of activated sludge, chemical precipitation or novel nutrient recovery technologies (Romero-Güiza et al., 2015). The appropriate method for

polishing would be determined by local discharge demands and the selected nutrient recovery technologies.

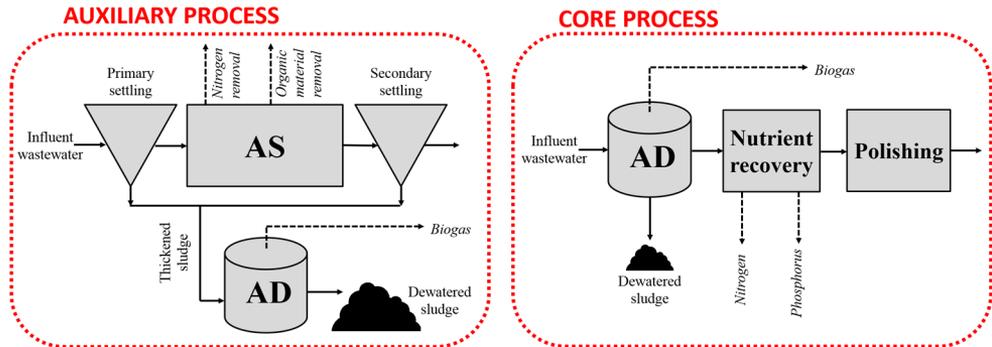


Figure 2.3.

Conceptual comparison of systems with anaerobic digestion (AD) as an auxiliary process (left) and anaerobic digestion as core process (right). AS = Activated sludge.

2.4.1 Source separation of wastewaters

One method to facilitate the problems of applying anaerobic digestion directly to wastewater is source separation. This increasingly emphasized concept (McConville et al., 2017; Larsen et al., 2013; Otterpohl et al., 1997) includes separation of domestic wastewaters from each other and from other urban wastewaters. Separate collection of toilet wastewater (blackwater), containing most of the domestic organic and nutrient load, facilitates more energy efficient treatment since the blackwater flow is small (Otterpohl et al., 1997). The concentration of blackwater collected with vacuum sewers is approximately 10-12 g COD L⁻¹ (Wiersma and Elzinga, 2014; Zeeman et al., 2008). The high concentration, compared to the concentration of approximately 200 mg COD L⁻¹ in mixed wastewater (Henze et al., 1997), makes it potentially suitable to treat the fraction by anaerobic digestion as the core process, preferably together with food waste, to further increase the organic load. Separation of blackwater from the other domestic wastewater from bathrooms, washing machines and kitchen sinks (collectively known as greywater) also facilitates recovery of water and heat from the greywater flow (Hellborg Lapajne, 2016; Larsen, 2015). Presently, several pilot areas with source separation systems are under installation in Europe (Skambraks et al., 2017) and a re-newed interest is also seen in Sweden (McConville et al., 2017).

3. Methods

The chapter briefly describes the most relevant methods used in **Papers I to VI** and the related publications. The selected methods are i) batch anaerobic digestion experiments, ii) continuous anaerobic digestion experiments, or iii) comparative desk top studies.

3.1 Batch tests

Batch tests were performed by applying sludge from continuous anaerobic reactors in a closed bottle together with a substrate and subsequently measuring the resulting methane production during incubation at a selected temperature. Batch tests were used to determine the potential for biogas production (**Paper V**), the hydrolysis rate constant (**Paper III**) and the specific methanogenic activity (**Paper III**).

3.1.1 Biomethane potential tests

Batch biomethane potential (BMP) tests were performed to assess the overall degradation of complex substrates during anaerobic digestion. The test, described by Hansen et al. (2004), was performed by applying anaerobic sludge from a continuous process together with a selected ratio of substrate in a closed 2 L bottle. Methane production was measured by manual injection into a gas chromatograph (**Paper III and V**). The test can be used to determine the potential for biogas production, and BMP tests were performed within **Paper V** to determine the potential for biogas production from blackwater and food waste. The determined potentials were used in **Papers V and VI** to calculate the possible energy recovery in the form of biogas from applying anaerobic digestion as core process, thus contributing to answering hypothesis #3 about the sustainability and energy recovery potential of source separation systems. BMP tests were also used in **Paper III** to assess the decreased rate of hydrolysis (Figure 2.2) in low-temperature processes by determining the hydrolysis rate constant. For this application, as previously reported elsewhere (Angelidaki et al., 2009; Haghightafshar et al., 2015; Koch and Drewes, 2014), it was assumed that hydrolysis was the rate-limiting

step of the anaerobic digestion process. To calculate the hydrolysis rate constant, a first-order kinetics model (Eq. 1) was fitted to the measured data (Figure 3.1) using a non-linear solver with minimization of the standard error as the goal function (Brown, 2001). In Eq. 1, B is the cumulative methane production at a given time t , B_{∞} is the ultimate methane production and k_{hyd} is the hydrolysis rate constant. The results from **Paper III** were used to answer hypothesis #2 about overcoming the operational challenges of low-temperature anaerobic digestion.

$$B = B_{\infty} * (1 - e^{-k_{hyd}*t}) \quad (\text{Eq. 1})$$

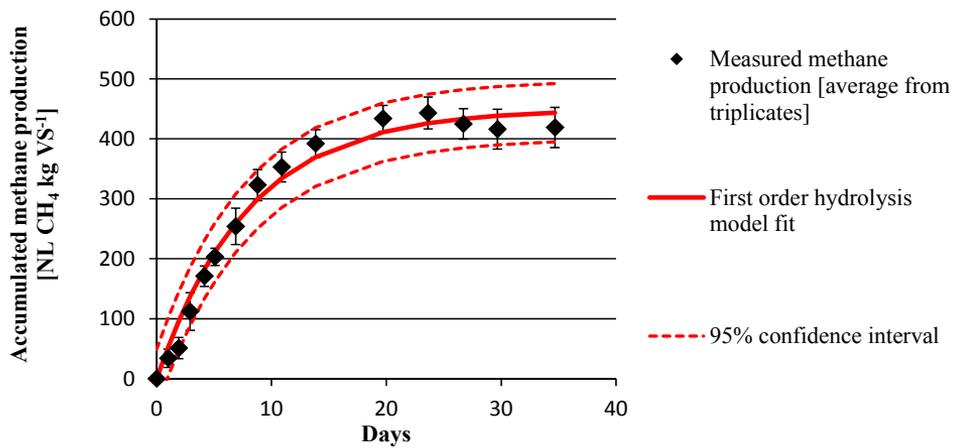


Figure 3.1.

Batch 2 L bottles used in the BMP tests (top) and the fitted first-order kinetics model used to determine the hydrolysis rate constant from the measured experimental data (bottom). VS = volatile solids.

3.1.2 Specific methanogenic activity tests

Specific methanogenic activity (SMA) tests were used to determine the activity of the two methanogenic groups, acetotrophic methanogens and hydrogenotrophic methanogens. Such tests are useful since methanogens are usually assumed to be the most sensitive of the anaerobic microbiological consortia to process overloading (Astals et al., 2015). Because they were used for pure substrates (acetic acid or H₂/CO₂ gas mixture), the tests were performed in relatively small (120 mL) batch bottles (Figure 3.2), compared to the 2 L BMP tests used for complex substrates. The gas production rate was measured using a pressure transducer and gas chromatograph (Collins et al., 2003; **Paper III**). Assuming monod-type kinetics the maximum activity of the methanogenic group was assessed using a non-linear fit of Equation 2 to the measured data (Figure 3.2), using methodology from Brown (2001). In Eq. 2, SMA is the specific methanogenic activity for bottles with initial substrate concentration S, SMA_{max} is the maximum specific methanogenic activity and K_s is the half saturation constant.

$$SMA = SMA_{max} \frac{S}{K_s + S} \quad (\text{Eq. 2})$$

Together with the BMP test, the SMA test was used to identify suitable organic loading rates for stable continuous reactor operation using the acidification limit test developed in **Paper III**. This application contributed to answering hypothesis #2 about overcoming operational challenges with low-temperature anaerobic digestion.

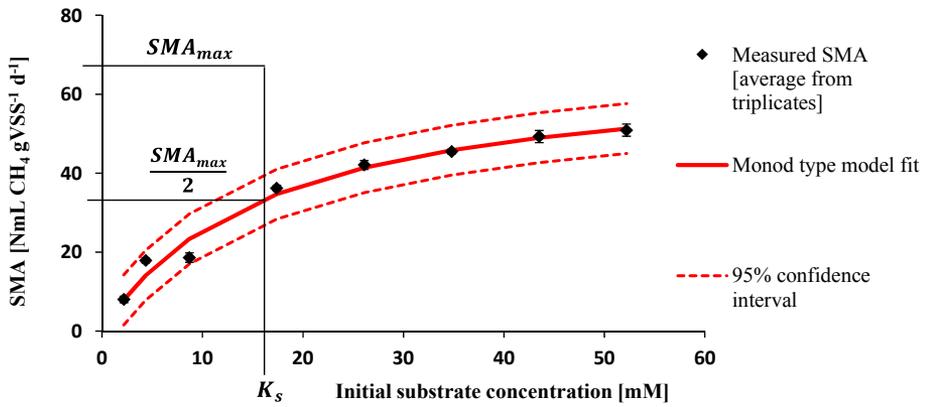
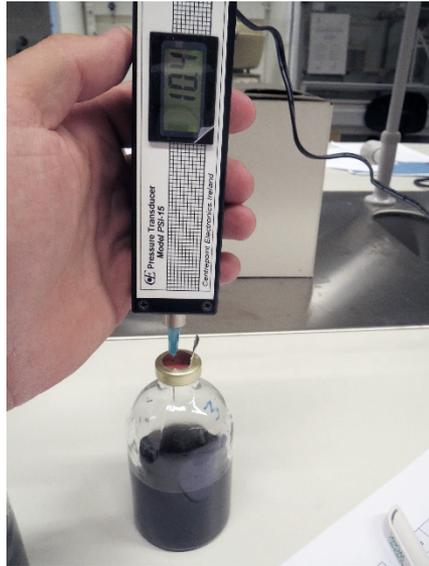


Figure 3.2.

Batch 120 mL bottle and pressure transducer used in the SMA tests (top) and the fitted monod-type kinetics model to experimental data (bottom). Figure with fitted monod-type kinetics based on Paper III and re-used with permission by Elsevier. VSS = volatile suspended solids.

3.2 Continuous tests

Continuous digestion experiments mimic full-scale operation conditions and were performed to evaluate the hygienization effects on pathogens (**Paper I**), the removal of micropollutants (**Paper II**), the process stability of anaerobic digestion at low temperature (**Paper III**) and the release of methane from reactor effluent (**Paper IV**).

3.2.1 Continuous stirred tank reactor

The completely mixed nature of the ideal continuous stirred tank reactor (CSTR) implies that the solids retention time (SRT) equals the hydraulic retention time (HRT). This assumption has implications for pathogen and organic compound removal, which, if not degraded during the process, remain until they are washed out. Due to risk of short-cutting between influent and effluent, CSTRs can be operated semi-continuously, allowing a minimum exposure time (MET) between feeding and removal to ensure sufficient pathogen removal (**Paper I**). CSTRs were utilized in **Paper I and II** to simultaneously determine the hygienization of pathogens and the reduction of organic micropollutants. The papers constitute the main contribution to answering hypothesis #1 about the hygienization of sludge and the removal of micropollutants. A graphical presentation of the CSTR setup utilized in the thesis (**Paper I and Paper II**) is given in Figure 3.3.

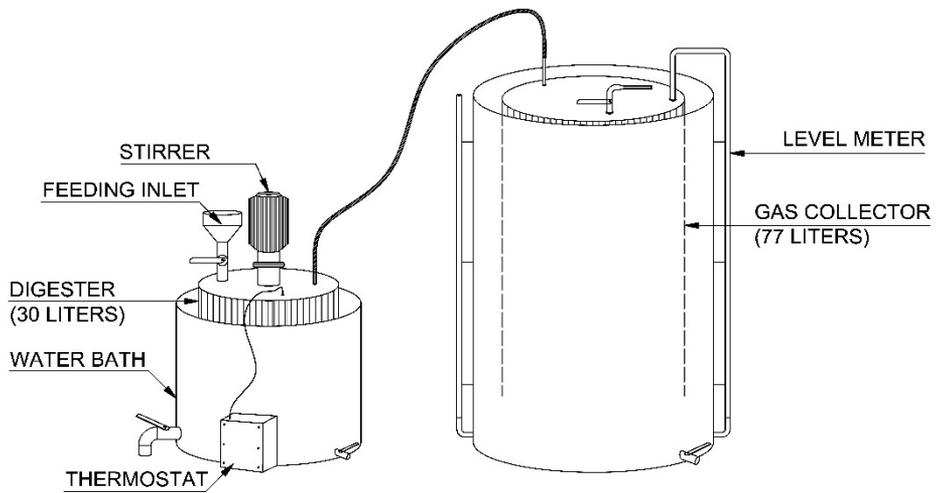


Figure 3.3.

Photo and layout of one of the CSTRs and gas bells used in Papers I and II. The gas bell was used to measure the amount of biogas produced by means of water displacement. Layout with courtesy of Salar Haghighatafshar.

3.2.2 Anaerobic membrane bioreactor

The anaerobic membrane bioreactor (AnMBR) utilizes a membrane to separate the SRT from the HRT (Hai et al., 2014). Thus, AnMBRs can be used to perform anaerobic digestion as core process since these reactors can treat wastewater directly without thickening of the wastewater in to sludge. However, since wastewater has much lower concentrations of organic material, heating the larger flow of wastewater to the mesophilic optimum might not be energetically favorable. Thus, low-temperature anaerobic digestion, utilizing little or no heating, is required for a better energy balance. Due to the lower growth rates of microorganisms at decreased temperatures, there is need for a method to determine what organic loading rate (OLR) can be applied without risking over-loading of the digestion process. Such a method was developed and tested in **Paper III**. Furthermore, low temperature increases the solubility of methane in water, which together with the short HRT of AnMBRs, increases the amount of methane leaving the anaerobic process via the reactor effluent and subsequently entering the atmosphere as greenhouse gas. A method to extract the dissolved methane using membrane contactors was evaluated in **Paper IV**. Together, **Papers III and IV** constitute the main work addressing hypothesis #2 about overcoming operational challenges of low-temperature anaerobic digestion. The CSTR combined with an external microfiltration membrane is shown in Figure 3.4.

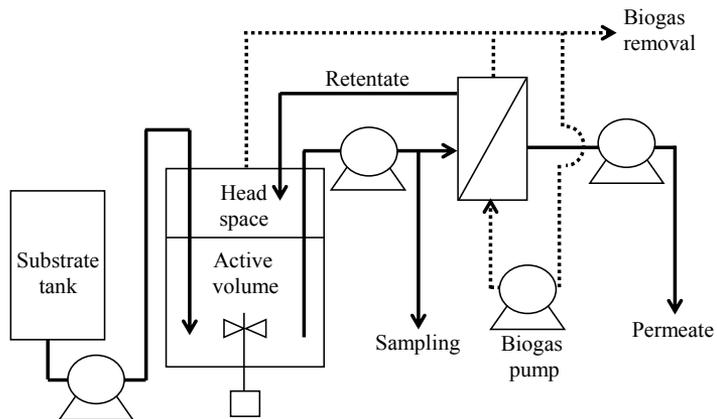


Figure 3.4.

Photo and layout of the AnMBR with external filtration tank utilized in Papers III and IV. Layout image is taken from Paper III and is reprinted with permission by Elsevier.

3.3 Comparative desk top studies

Comparative desktop studies were performed to compare systems representative of the two paths in Figure 1.2. These studies included mass balances (**Paper V**), life cycle assessments (Kjerstadius et al., 2016 and **Paper VI**) and economic cost evaluation (Kärroman et al., 2017).

3.3.1 Life cycle assessment

Life cycle assessments (LCAs) were performed to compare the environmental impact of wastewater systems by normalization against a selected functional unit. All relevant material and energy requirements for the production and operation were accounted for using a life cycle inventory, compiling the total energy use and mass balances for the materials. Secondary effects, such as replacement of fuel, energy or material, were taken in to account via expansion of the system boundaries. LCAs and the associated mass balances were performed in **Papers V and VI** to compare conventional wastewater management (with anaerobic digestion as an auxiliary process) to source separation systems (with anaerobic digestion as core process). The results were presented for the impact category climate impact and potential for recovery of phosphorus and nitrogen. **Papers V and VI** constitute the main work addressing hypothesis #3 about the sustainability of systems with anaerobic digestion as core process.

3.3.2 Life cycle cost assessment

When evaluating the environmental and energetic sustainability of wastewater systems, the associated economic cost is relevant for real life implementations. The economic cost of wastewater systems, due to having a long life span of >50 years, should take in to account both installation cost and operational cost. This type of evaluation was performed in Kärroman et al. (2017) using a Life Cycle Cost (LCC) evaluation of the systems investigated in **Papers V and VI**. The evaluation used the net present value method, as described in Kärroman et al. (2017), which puts a rate on the installation cost and the cost of capital, which increases the cost of installations with long economic life spans. Since the investigation covered systems that supply a service rather than an economic gain, the cost of capital was set to 4%, as recommended by SEPA (2003). Although they do not address any of the hypotheses of this thesis, the results from the LCC contribute to the grander discussion.

3.4 Additional analysis methods

The following methods are general methods used in several papers. Specific methods relevant to only one paper, like analysis of pathogens, are presented in their respective papers.

For sludge and water samples, total solids (TS), volatile solids (VS) and volatile suspended solids (VSS) were measured according to APHA (2005). Chemical oxygen demand (COD) and ammonium nitrogen were measured spectrophotometrically (DR 2800 spectrophotometer using Dr. Lange cuvettes 314, 514 and 303). Total alkalinity was measured according to APHA (2005), while partial (bicarbonate) and intermediate (VFA) alkalinity was measured according to Ripley et al. (1986). Volatile fatty acids (acetic acid and propionic acid) were measured using a gas chromatograph (Agilent 6850A) equipped with a flame ionization detector (FID) and a 30 m (length) by 0.53 mm (diameter) by 1.0 mm (film) HP-FFAP column. Temperature and pH were measured using portable equipment (WTW pH 3110).

For gas samples, the biogas flow of the AnMBRs was measured using a thermal mass flow meter (Vögtlin GSM-B), while biogas production in the CSTR reactors was measured daily using water displacement. Methane content was measured using a pressure lock syringe and a gas chromatograph (Varian 3800) equipped with a thermal conductivity detector (TCD) and a 2.0 m (length) by 3.2 mm (diameter) by 2.0 mm (film) HayeSep mesh column. Alternatively, for the CSTR equipment, biogas composition (CH_4 , CO_2 , and H_2S as volumetric percentages) was determined using a portable gas analyzer (Sewerin SR2-DO).

4. Results

To clarify the contribution to each of the three hypotheses, the results are divided into three sections. This corresponds to one section each for the two possible paths in Figure 1.2, the utilization of anaerobic digestion as an auxiliary (**Paper I and II**) or core process (**Paper III and IV**), respectively, and one section on the results of the comparative studies (**Paper V and VI**).

4.1 Adjusting anaerobic digestion as an auxiliary process

The continued use of anaerobic digestion for treatment of sludge at mesophilic or thermophilic temperatures is investigated in **Paper I and II**. Especially, this covers hygienization of pathogens (**Paper I**) as well as the effect on process performance (**Paper I**) and reduction of organic micropollutants (**Paper II**).

4.1.1 Sludge hygienization at thermophilic temperature

As presented in **Paper I**, the SEPA proposal for sludge hygienization includes two suggested standard processes: i) pasteurization (70 °C for 1 h) and ii) thermophilic anaerobic digestion with a minimum exposure time (MET) for anaerobic digestion at 52 °C, 55 °C or 60 °C (SEPA, 2013). The increased temperature causes pathogen destruction or inactivation (Sahlström, 2003), while the MET decreases the risk of short-cutting through the CSTR. The resulting hygienization (**Paper I**) for the tested METs of 2 h, 2.5 h, 6 h and 24 h is presented in Table 4.1. Since thermophilic temperature enables the possibility of shorter HRTs due to the increased microbial activity, both 15 d and 7 d HRT were tested for thermophilic temperatures. The results show that thermophilic anaerobic digestion at either 55 °C or 60 °C, even with a short MET of 2 h, is sufficient to achieve the suggested concentration limits of cultivable pathogens. Thus, satisfactory results for hygienization, compared to pasteurization, can be achieved by increasing the operation temperature and applying MET control at existing wastewater treatment plants in Sweden. In contrast, mesophilic anaerobic digestion requires more than 24

h MET, which would be practically unreasonable to implement at a conventional WWTP due to the continuous production of sludge.

Table 4.1.

Minimum exposure time (MET) required to meet the suggested pathogen concentration limits of SEPA (2013) for anaerobic digestion (35, 55 or 60 °C) and pasteurization (70 °C). Tested METs were 2 h, 2.5 h, 6 h and 24 h.

Temperature	35 °C	55 °C	55 °C	60 °C	60 °C	70 °C
HRT	15 d	15 d	7 d	15 d	7 d	-
<i>E. coli</i> <3 log ₁₀ g TS ⁻¹	> 24 h	2 h	2 h	2 h	2 h	60 min
<i>Enterococcus</i> <3 log ₁₀ g TS ⁻¹	> 24 h	2 h	2 h	2 h	2 h	60 min
<i>Salmonella</i> Absent in samples of 25 g	> 24 h	2 h	2 h	2 h	2 h	60 min

4.1.2 Process performance of thermophilic anaerobic digestion

Although literature data from anaerobic digestion at temperatures as high as 82 °C exist (Nozhevnikova et al., 1999), no literature on the digestion of sludge from WWTPs at ≥ 60 °C was found prior to the work in **Paper I**. Similar work on the digestion of manure and agricultural residues in CSTRs was found, which showed contradictory results for the effect on process stability and biogas production at 60 °C compared to 55 °C or 35 °C (Kjerstadius et al., 2012). The operational results from **Paper I** (Table 4.2) indicate a stable process for 60 °C operation compared to the stated inhibitory conditions (Chen et al., 2008). Furthermore, an increased operation temperature resulted in an increased hydrolysis rate constant (Haghighatafshar et al., 2015). However, the increased hydrolysis rate did not result in increased biogas production at the thermophilic temperature. In contrast, operation at 60 °C decreased the methane yields by approximately 10% due to lower average gas production and lower methane content. A decreased methane content at higher operating temperatures was also observed by (Kim and Lee, 2016) and is likely an effect of decreased CO₂ solubility. The decreased methane production may be due to incomplete microbial adaptation following the increase of operational temperature from 55 °C to 60 °C, which was performed 60 days (corresponding to >3 HRT) before the measurement of the results in Table 4.2 was initiated. The result promoting this theory was the severely decreased population of *Methanosarcinaceae* at 60 °C compared to the reactors operated 55 °C (**Paper I**), which indicates decreased methanogenic capacity.

Table 4.2.

Selected process data from 36 days of steady state operation (following a minimum of ≥ 3 HRT). The results are presented with the number of data points (n) and the standard deviation (\pm) or min/max values (in brackets). Denotations made for no data (n.d.).

		35 °C	55 °C	55 °C	60 °C	60 °C
Temperature		35 °C	55 °C	55 °C	60 °C	60 °C
HRT		15 d	15 d	7 d	15 d	7 d
Parameter	Unit					
k_{hyd}	d ⁻¹	0.32 ¹	0.44 ¹	n.d.	0.50 ¹	n.d.
OLR	kg VS m ⁻³ d ⁻¹	1.9±0.2	1.9±0.2	4.1±0.5	1.9±0.2	4.1±0.5
pH	pH	7.3 (7.2-7.3)	7.4 (7.3-7.5)	7.4 (7.3-7.5)	7.4 (7.4-7.5)	7.4 (7.3-7.4)
Acetate	mg L ⁻¹	13±5	71±37	32±12	166±69	281±207
Propionate	mg L ⁻¹	3±11	16±21	2±3	43±59	113± 170
Gas production	L day ⁻¹	19±2	19±2	38±5	17±2	37±5
Methane content	Vol-%	66±1	65±1	67±1	63±1	62±1
Hydrogen sulfide	ppm	0±1	20±18	2±1	6±4	127±54
Methane production	NL CH ₄ kg VS ⁻¹	312±20	311±14	301±17	278±17	275±23

1) Value obtained by fitting the data to a first-order hydrolysis model (Haghighatafshar et al., 2015).

4.1.3 Reduction of organic micropollutants

Some organic micropollutants in urban wastewater, such as pharmaceutical residues, are separated into the sludge phase (Petrie et al., 2015) at wastewater treatment plants. Using the same experimental set up as in **Paper I**, the degradation of 99 organic micropollutants and 15 PAHs (polycyclic aromatic hydrocarbons) during anaerobic digestion was investigated in **Paper II**. Results are presented in Table 4.3 for compounds selected on basis of i) being mentioned in the EC (2012) suggested amendment of the Water Framework Directive (Diclofenac and Ethinylestradiol); ii) that occur at the highest concentration (Ciprofloxacin, Dipyridamol, Sertraline, Irbesartan and Ketoconazole) or iii) show trends of

reduction (Irbesartan and Trimethoprim). The lower concentration of TS in the digested samples depends on the degradation of organic material during anaerobic digestion. Although the concentrations in the mesophilic sludge were generally similar to those in mesophilic sludge in other Swedish studies (Fick et al., 2010; Wahlberg et al., 2010; **Paper II**), the high variation in the overall results demonstrated the difficulty in measuring organic compounds in the ng to μg scale in a sludge matrix. Although the high standard deviations in Table 4.3 could be explained by the small numbers of samples ($n=2$ or 3), the analysis method also had low recovery for spiked samples (**Paper II**). In conclusion, the results showed substantial ($>50\%$) reduction of 2 out of 99 substances, indicating that anaerobic digestion at either mesophilic or thermophilic temperature does not result in broad spectrum reduction of organic micropollutants.

Table 4.3.

Selected results ($\mu\text{g kg TS}^{-1}$) for the concentration of organic micropollutants in raw sludge and digested (24 h MET) or pasteurized ($70\text{ }^{\circ}\text{C}$) sludge. Number of data points (n) = 2 for all samples except for pasteurization ($70\text{ }^{\circ}\text{C}$, 60 min) where $n=3$. The table includes the standard deviation (\pm) and limit of quantification (LOQ) of the analysis method.

		Raw sludge	35 $^{\circ}\text{C}$ 15 d HRT	55 $^{\circ}\text{C}$ 15 d HRT	60 $^{\circ}\text{C}$ 15 d HRT	70 $^{\circ}\text{C}$ 60 min
Total solids		4.9 %	2.0 %	2.1 %	2.3 %	4.9 %
Compound	LOQ					
Diclofenac	10	<10	<10	<10	<10	<10
Ethinylestradiol	10	<10	<10	<10	<10	<10
Ciprofloxacin	10	2 100 ± 800	4 100 ± 600	1 500 ± 30	1 400 ± 500	2 400 ± 570
Dipyridamol	50	190 ± 130	490 ± 30	470 ± 270	320 ± 10	190 ± 30
Sertraline	10	280 ± 10	670 ± 40	810 ± 250	180 ± 110	340 ± 110
Ketoconazole	50	200 ± 60	160 ± 50	220 ± 90	90 ± 0	110 ± 0
Irbesartan	0.5	1 900 $\pm 2 600$	540 ± 380	20 ± 20	60 ± 10	240 ± 100
Trimethoprim	0.1	19 ± 21	<0.1	1 ± 0	5 ± 0	30 ± 20

4.2 Operational issues of applying anaerobic digestion as core process

As explained in section 2.4, low-temperature anaerobic digestion is required for an energetically favorable application of anaerobic digestion as core process to treat wastewater. Selected operational challenges of low-temperature anaerobic digestion at the ambient temperature of synthetic dairy wastewater were evaluated in **Papers III and IV**.

4.2.1 Acidification limit

A decreased operation temperature results in lower reaction rates for all microbiological steps in the anaerobic digestion food chain, as expected based on the Arrhenius equation (Pavlostathis and Giraldo-Gomez, 1991). However, not all process steps are affected equally. Due to the different reaction rates of its degradation steps, anaerobic digestion is susceptible to possible process failures due to accumulation of intermediary products and subsequent inhibition (Van Lier et al., 2008). This is especially true when performing anaerobic digestion at low temperatures (Bowen et al., 2014). The application of anaerobic digestion as core process in wastewater treatment thus requires the means to assess a safe operation space in regards to OLR. The test for acidification limit was developed and evaluated in **Paper III** for this purpose. The assumption that either the hydrolysis or the methanogenesis is the rate-limiting step of the anaerobic digestion is essential for this method.

The acidification limit relates the load of organic material to the rate of hydrolysis and the rate of methanogenesis in the anaerobic digestion food chain. Thus, it creates a ratio, the Fatty Acid Capacity (FAC), between the existing activity (V_{active}) and the activity that is required (V_{needed}) to avoid over-loading of the anaerobic process (Eq. 3). If the $FAC \leq 1$, the process is overloaded and eventually fails due to accumulation of intermediary products.

$$Fatty\ acid\ capacity\ (FAC) = \frac{V_{active}}{V_{needed}} \quad (Eq. 3)$$

This method was evaluated (**Paper III**) for two anaerobic membrane bioreactors treating synthetic dairy wastewater at 24 °C. Batch BMP tests for hydrolysis rate constant (k_{hyd}) and specific methanogenic activity (SMA) test for acetotrophic (SMA_{ac}) and hydrogenotrophic (SMA_{hy}) populations were performed throughout the startup of the reactors to calculate the FAC at an increased organic loading rate

(OLR) (Figure 4.1). The results showed a clear adaptation of the inoculum in the first reactor (AnMBR #1), with increasing numbers for k_{hyd} and SMA, keeping $\text{FAC} > 1$ for all applied OLRs. In contrast, the other reactor (AnMBR#2) showed decreasing methanogenic activity and reactor failure at increased OLR, coinciding with $\text{FAC} < 1$, the theoretical limit for process failure. Therefore, it is plausible that the acidification limit method is suitable to identify safe applications of OLR for low-temperature anaerobic digestion; however, further evaluation is needed for confirmation.

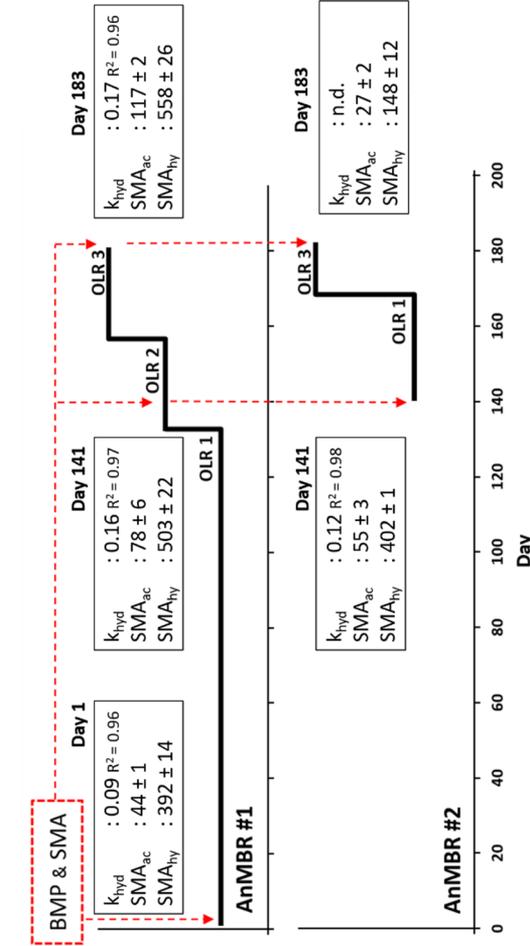
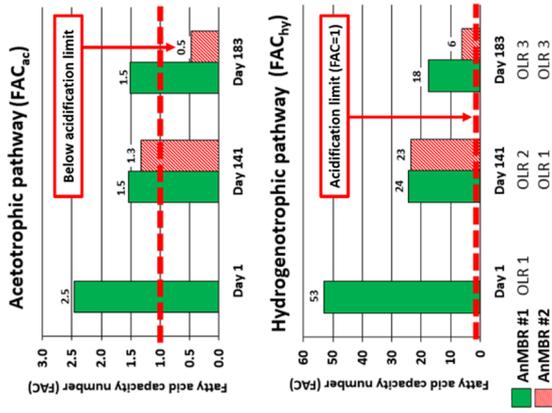


Figure 4.1.

Left: Results from the BMP test for the hydrolysis rate constant [d^{-1}] and SMA [$mg\ COD-CH_4\ gVSS^{-1}d^{-1}$] for the acetotrophic and hydrogenotrophic methanogenic pathways.

Right: Results for the fatty acid capacity for the two methanogenic pathways during the operation of two AnMBRs with different strategies to increase the OLR. The figures are based on Paper III and are published with permission from Elsevier. R^2 = coefficient of determination. n.d. = no data

4.2.2 Extraction of dissolved methane

Low-temperature increases the solubility of methane in the reactor liquid, which can enter the atmosphere as a greenhouse gas upon discharge of the reactor effluent. Due to the particle-free nature of the effluent of an AnMBR, dissolved methane can be extracted using a hollow fiber membrane contactor (HFMC) with a sweep gas to transport the extracted methane (Figure 4.2). This method was tested in **Paper IV** with nitrogen or air as the sweep gas. To analyze dissolved methane, the salting out method, as presented by Daelman et al. (2012), was adapted. The results (Figure 4.3) showed a great extraction of methane (>90%), even at low sweep gas flow rates. Similar results were recently published (Cookney et al., 2016, 2012; Henares et al., 2016), which supports the potential to use membranes to extract dissolved methane from anaerobic digesters. The results also indicated a super-saturation of methane in the reactor permeate, a finding also supported elsewhere (Crone et al., 2016). Thus, Henry's law could not be used to accurately predict the amount of dissolved methane in the reactor, most likely due to the prerequisite of equilibrium not being achieved in the anaerobic digester as a result of the continuous production of biogas (**Paper IV**).

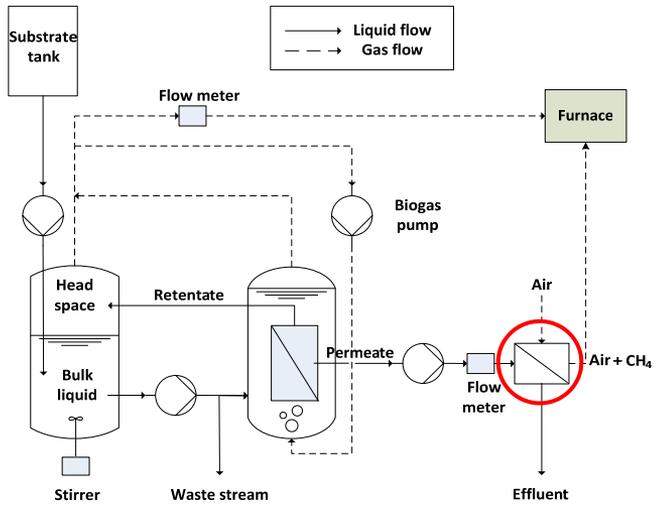


Figure 4.2.

Layout of the experimental plan for the extraction of dissolved methane from an AnMBR. The hollow fiber membrane contactor (HFMC) is indicated by the red circle.

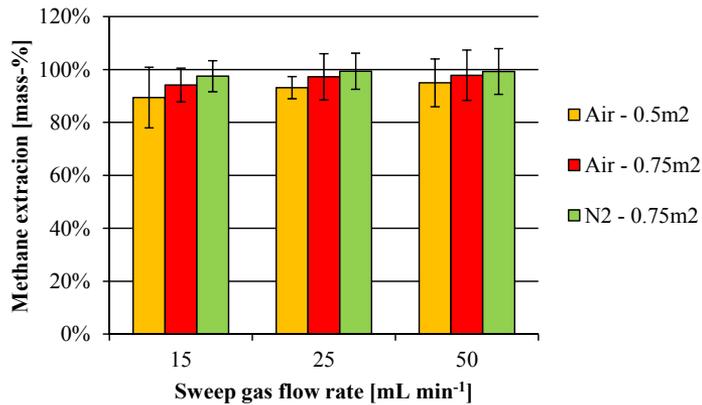


Figure 4.3.

Extraction ratio of methane depending on the flow rate of the sweep gas (air or nitrogen) and the total membrane area of the HFMC extraction modules (0.5 m² or 0.75 m²). Standard deviation (bars) calculated from subtracted average values (n=9 or 10) on measurements before and after HFMC.

4.3 Source separation with anaerobic digestion as core process

Source separation, within this thesis, includes the separate collection, transport and treatment of blackwater, greywater and food waste. Due to the relatively high concentration of organic material in blackwater and food waste, these streams can be treated using anaerobic digestion as core process. An example is presented in Figure 4.5 (based on **Paper V**). Core treatment was assumed performed with an upflow anaerobic sludge blanket septic tank (UASB-ST), as described by Kujawa-Roeleveld et al. (2006), which separates the SRT from the HRT, thus allowing low-temperature anaerobic digestion with associated lower growth rates. Nutrient recovery was assumed performed on the digester effluent through struvite precipitation and ammonia stripping, as described in **Paper V**, **Paper VI** and Kjerstadius et al. (2016). Separate treatment of greywater in an activated sludge process was calculated based on data from Wiersma and Lettinga (2014). Importantly, the system described in Figure 4.4 would require a polishing step to reach the discharge limits for nitrogen and phosphorus. This was represented by post-precipitation in Figure 4.4, calculated using empirical values from Lindquist (2003), and by assuming increased denitrification with methanol as the carbon source according to USEPA (2013).

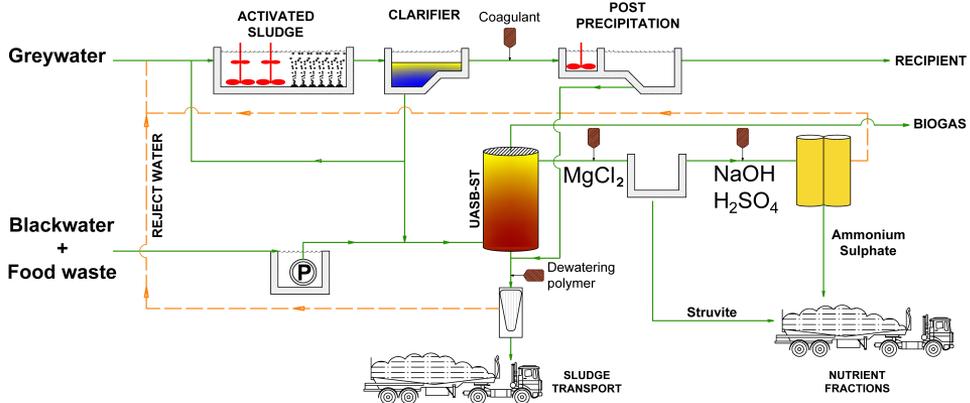


Figure 4.4.

A process scheme of the source separation system with anaerobic digestion as the core process and nutrient recovery steps. The anaerobic digester is termed UASB-ST (upflow anaerobic sludge blanket septic tank). Figure based on **Paper V** by permission from ACS Journal Publishing Agreement.

4.3.2 Potential of source separation

The mass balances performed in **Paper V** show that source separation systems can enhance the recovery efficiency of wastewater treatment. A comparison of the potential against a conventional Swedish system (**Paper V** updated in Kjerstadius et al., 2016 and **Paper VI**) shows increased potential for the recovery of energy and nutrients. Increased energy recovery is achieved by applying anaerobic digestion as the core process, thus transforming more organic carbon into methane. Increased nutrient recovery is potentially achieved by struvite precipitation and ammonium stripping from the digester effluent. Furthermore, the application of a heat pump to the source separated greywater increases the heat recovery efficiency compared to heat recovery from mixed wastewater (Hellborg Lapajne, 2016; Kjerstadius et al., 2016). The realistic potential of heat recovery from greywater has been approximated to between 450 kWh capita⁻¹ year⁻¹ (**Paper VI**; STOWA, 2014) and 800 kWh capita⁻¹ year⁻¹ (Larsen, 2015). For the city of Helsingborg, which is constructing the H+ pilot area with source separation (Skambraks et al., 2017), the current heat recovery at the WWTP is highly variable (Table 4.4) since the low coefficient of performance (COP) of the heat pump, due to the addition of stormwater to the sewer system, makes heat recovery highly dependent on electricity cost (Hellborg Lapajne, 2016).

Table 4.4.

Demand for electricity (kWh_{el}) and heat (kWh_{heat}) and potential for recovery of energy and nutrients for the collection and treatment of food waste (FW), blackwater and greywater. WWTP = wastewater treatment plant. n.d. = no data.

	Conventional system Paper VI	Conventional system in Helsingborg NSVA (2014)	Source separation system Paper VI	Source separation Waterschoon STOWA (2014)
Wastewater treatment				
Demand – sewer pumps [kWh _{el} capita ⁻¹ year ⁻¹]	5	n.d.	8	40 ²
Demand – WWTP _{treatment} [kWh _{el} capita ⁻¹ year ⁻¹]	42	57 ¹	21	52 ²
Demand – WWTP _{treatment} [kWh _{heat} capita ⁻¹ year ⁻¹]	49	n.d.	140	50 ²
Demand – heat pump [kWh _{el} capita ⁻¹ year ⁻¹]	114	80-300 ³	90	264 ²
Recovery – heat pump [kWh _{heat} capita ⁻¹ year ⁻¹]	445	230-850 ^{3,5}	420	477 ²
Energy recovery – biogas production	2.6 (FW) 5.4 (WWTP)	2.6 ⁴ (FW) 5.4 ³ (WWTP)	12.8	12.2
Sludge				
Total solids [kg TS capita ⁻¹ year ⁻¹]	16.8	20.6	21.0	9.2
Phosphorus [kg P capita ⁻¹ year ⁻¹]	0.68	0.59	0.22	n.d.
Nitrogen [kg N capita ⁻¹ year ⁻¹]	1.39	1.20	0.39	n.d.
Biofertilizer				
	Food waste digestate	Food waste digestate	Struvite + ammonium sulfate	Struvite
Total solids [kg TS capita ⁻¹ year ⁻¹]	2.7	2.7 ⁴	-	-
Phosphorus [kg P capita ⁻¹ year ⁻¹]	0.02	0.02 ⁴	0.51	0.32
Nitrogen [kg N capita ⁻¹ year ⁻¹]	0.20	0.20 ⁴	0.23 / 3.63	0.14

1) Calculated assuming 120 000 capita connected to Öresundsverket WWTP. 2) Calculated as primary energy. 3) Includes industrial wastewater and stormwater, calculated assuming 120 000 capita connected to Öresundsverket WWTP. 4) Paper V. 5) Results from Hellborg Lapajne (2016).

4.3.3 Carbon foot print of source separation with anaerobic digestion as core process

The potential benefit of source separation systems, highlighted in Table 4.4, should not have an adverse impact on other environmental issues. The long-term impact of source separation systems on climate impact was therefore assessed in **Paper VI** using an attributional life cycle assessment (LCA) with the aim being to identify the most important aspects to minimize the impact. The paper was especially interesting since the study covered the Swedish electricity and heat mix, which has a very low impact per kWh compared to previous studies (Kjerstadius et al., 2016). The results (Table 4.5) from **Paper VI** showed that source separation has potential to decrease climate impact under the Swedish conditions. The impact, measured as carbon footprint, was $-37 \text{ kg CO}_2\text{-eq. capita}^{-1} \text{ year}^{-1}$, which was slightly lower than that of the conventional system ($-13 \text{ kg CO}_2\text{-eq. capita}^{-1} \text{ year}^{-1}$). Important aspects to decrease carbon footprint were identified: i) increased biogas production, ii) increased heat recovery via heat pump, iii) decreased emissions of N_2O from activated sludge processes, iv) increased nutrient recovery and decreased emissions from sludge management. A qualitative sensitivity analysis (**Paper VI** and Kjerstadius et al., 2016) showed that changes to parameters ii-iv resulted in a final impact between -37 to $10 \text{ kg CO}_2\text{-eq. capita}^{-1} \text{ year}^{-1}$ for the source separation system and -12 to $48 \text{ kg CO}_2\text{-eq. capita}^{-1} \text{ year}^{-1}$ for the conventional system. Combined with the potential for increased nutrient recovery (Table 4.5), source separation systems thus have potential to increase sustainability compared to conventional systems.

Table 4.5.

Results for carbon footprint and nutrient recovery in comparison with international studies. The results from Paper VI include the range given by the sensitivity analyses.

	Hillenbrand (2009)	Remy (2010)	Meininger (2010)	Witteveenbos (2014)	Thibodeau (2014)	Paper VI
Carbon footprint [kg CO ₂ -eq. capita ⁻¹ year ⁻¹]						
Conventional system	244	140	-	32-40	52.8	-12 to 48
Source separation system	315	85	-	-22	65.3	-37 to -10
Return of nitrogen to agriculture [kg N capita ⁻¹ year ⁻¹]						
Conventional system	-	0.40 ¹	0.11 ⁴	-	0.39 ^{2,5}	0.54-0.79
Source separation system	+4.29 ³	3.24 ²	3.09	-	2.12 ^{2,5}	3.82-3.89
Return of phosphorus to agriculture [kg P capita ⁻¹ year ⁻¹]						
Conventional system	-	0.49	0.03 ⁴	-	0.54 ^{2,5}	0.19-0.31
Source separation system	+0.54 ³	0.72	0.44	-	0.60 ^{2,5}	0.57-0.61

1) Assumed 100% sludge to agriculture. 2) Return of the entire treated wet fraction. 3) Results given only as excess return with source separation system compared to the conventional system. 4) No nutrients are returned from the WWTP, only from food waste management. 5) Value is for plant available nutrients after emissions and run-off.

5. Discussion

The discussion section is divided into five sections. These sections cover the evaluation of the hypotheses of the thesis, a discussion about meeting the SEPA demands, a comparative economic analysis used to place the results in a grander context and finally related aspects and outlook.

5.1 Addressing the hypotheses

5.1.1 Benefit of thermophilic anaerobic digestion (Hypothesis #1)

The results in **Paper I** indicate the possibility to operate digesters at thermophilic temperature (55 °C and 60 °C) to achieve hygienization of sludge from pathogens, even at 2 h MET. However, the expected increased biogas production due to the increase in the hydrolysis rate constant was not observed, making beneficial economic effects from excess biogas production doubtful. Similar results were seen by Kim and Lee (2016), who did not find a significant change in methane yield between 50 and 60 °C when using whey permeate as substrate; however, a slight increase was found compared to mesophilic operation. An increased COD conversion from substrate into methane during thermophilic anaerobic digestion has been reported (De Vrieze et al., 2016; Kim and Lee, 2016) but was not found in the present work (conclusion based on VS-degradation). Conflicting results showing either no change (Gavala et al., 2003) or an increase in biogas production (Davidsson, 2007; Kim et al., 2002) at thermophilic temperature have also been published. Likely, the effect on the overall energy efficiency from a shift from mesophilic to thermophilic temperature may be negative or positive, depending on the substrate, mixing and reactor insulation, as concluded by (De Vrieze et al., 2016). Lastly, the potential increase in biogas production following pasteurization was not assessed in **Paper I** and was not found in similar trials (Grim et al., 2015).

Although the potential risk constituted by the return of organic micropollutants to agriculture via sludge is not known (SEPA; 2013), the issue is receiving increased attention (SEPA, 2008). **Paper II** demonstrated the difficulty of measuring a reduction in organic micropollutants in the ng to µg range in the sludge matrix. A

later study by Malmborg and Magnér (2015) showed better recovery rates and reported some reduction (in general 30%) of 12 compounds by anaerobic digestion, irrespective of mesophilic or thermophilic temperature. Similarly to the present study, Trimethoprim was reduced below the LOQ, and anaerobic digestion generally had a stronger effect on reduction than pasteurization. However, the results in **Paper II**, in agreement with the study of Malmborg and Magnér (2015) as well as the well documented presence of organic micropollutants in sludge from WWTPs (Fick et al., 2010; Wahlberg et al., 2010), indicate that anaerobic digestion does not provide a substantial reduction in organic micropollutants, regardless of the operational temperature. The achieved reduction was also much lower than obtained for aerated processes (Falås et al., 2016). Finally, the environmental impact of organic micropollutants from sludge application in agriculture, as well as their vectors for spreading and degradation, are still relatively unknown (Verlicchi and Zambello, 2015).

In summary, hypothesis #1 can be partly confirmed. Thermophilic anaerobic digestion achieves sufficient hygienization of sludge to comply with proposed SEPA regulation, but the treatment does not have a general effect on the removal of organic micropollutants. A beneficial effect on biogas production from thermophilic anaerobic digestion could not be confirmed.

5.1.2 Applying anaerobic digestion as core process (Hypothesis #2)

The idea to replace AS with anaerobic digestion as the core process in wastewater treatment is not new. The potential benefits (less sludge production, increased biogas production, decreased electricity demand) have long been known (Schink, 1988; Zeeman et al., 2008). However, applying anaerobic digestion to wastewater (municipal wastewater with an organic content of approximately 200 mg COD L⁻¹ and source separated blackwater ranging approximately 10 000-12 000 mg COD L⁻¹) makes heating the process to mesophilic temperature questionable from an energy perspective. Low-temperature digestion at ambient wastewater temperature is thus ideal, and the recent increase in work on AnMBR has made such applications reasonable (Hai et al., 2014). However, the work for operators to achieve stable anaerobic processes at lower temperatures is more challenging due to the slow growth rates of anaerobic microorganisms. The results in **Paper III** showed that a stable OLR can be successfully identified. However, although the developed method aims to be relatively easy to implement, it remains to be seen whether it is relevant for professionals in the wastewater field. On the other hand, the successful results of the extraction of dissolved methane in **Paper IV** indicate that the emissions of greenhouse gases can be greatly decreased by using existing membrane technology. Realistically, the results from **Paper III and IV** are part of the solution

to achieve a more holistically beneficial wastewater management by demonstrating methods to achieve stable low-temperature anaerobic digestion.

In summary, hypothesis #2 can be confirmed. Stable low-temperature anaerobic digestion can be achieved; thus, the operational challenges of using anaerobic digestion as the core process can be overcome.

5.1.3 Increased sustainability with source separation systems (Hypothesis #3)

In regards to the climate impact of wastewater management, Sweden is potentially different due to the low impact of electricity and heat generation compared to more fossil-fuel-dependent countries. This can be seen in Table 4.5, where the impacts of both conventional and source separation systems are low compared to international studies. However, some of the differences could be attributed to differences in the studied systems (**Paper VI** and Kjerstadius et al., 2016). The study by Thibodeau (2014) showed a greater impact on climate change with source separation, mainly due to the system including the return of the entire blackwater fraction by truck to farmland. Clearly, the system configuration and local conditions are important factors in determining the carbon footprint. The choice of the system boundary and in-data can have a major impact on the final results, and LCAs should be designed to answer specific aims while minimizing the ambiguity due to the choice of the system boundary and in-data. Thus, no general conclusion in regards to which system has a more beneficial impact on climate change should be made. However, source separation systems were shown to decrease the climate impact in a Swedish context (**Paper VI**). The decreased impact, ranging from -25 to -58 kg CO₂-eq. capita⁻¹ year⁻¹, was mainly due to increased biogas production, minimized greenhouse gas emissions and better nutrient fraction management. Additionally, in all international studies, as well as in **Paper VI**, source separation systems were seen to increase the nutrient recovery of phosphorus and nitrogen (Table 4.5). The increase compared to the conventional system ranged from 0.30 to 0.38 kg P capita⁻¹ year⁻¹ and from 3.10 to 3.28 kg N capita⁻¹ year⁻¹. The potential for nutrient recovery may have increased importance in regards to sustainability in the future due to the increased need for urban nutrient recovery (Matassa et al., 2015) and the need for decreased interference with global nutrient cycles (Steffen et al., 2015).

In summary, hypothesis #3 can be partly confirmed. Source separation systems with anaerobic digestion as the core process can increase the sustainability of wastewater management.

5.2 Meeting the SEPA demands and target goals

This thesis investigated paths to meet the demands on the quality of sludge intended for return to agriculture and the target goals for nutrient recovery in regards to the proposed SEPA regulation (SEPA, 2013). Specifically, the demands considered were hygienization and metal concentrations, and the target goals for recovery were 40% of the phosphorus and 10% of the nitrogen present in the influent wastewater to WWTPs. The results in this thesis provide clear indications of how the hygienization of sludge can be achieved, as well as for the potential for nutrient recovery. However, a broader discussion of the effect of more stringent metal demands and a quantitative evaluation of the target goals for nutrient recovery is required to put the results into perspective. Although not part of the hypotheses, such an investigation addresses the aim of the thesis.

5.2.1 Meeting the demands for metal concentrations

The SEPA proposal includes more stringent demands for metal content in sludge intended for return to agriculture (Table 2.1). Minimizing the metal content in sludge is one of the long-term goals of the Swedish voluntary certification system Revaq, and the WWTPs connected to the certification system produce almost half of the sludge from Swedish wastewater treatment (SEPA, 2014). Sludge from Revaq-certified treatment plants has, on average, a lower metal content than sludge from other Swedish WWTPs (Table 5.1). Although the average values for Revaq-WWTPs are lower than the SEPA regulation proposal, it is clear from the results of Mattsson and Finnson (2016), reprinted in Table 5.1, that not all Revaq-certified WWTPs meet the suggested SEPA demands. The problem is accentuated by the fact that many of the pollutants in Swedish WWTPs originate from diffuse, rather than point, sources (Olofsson et al., 2013). Especially problematic are the levels of cadmium, mercury, chromium and copper. This conclusion is similar to that of the Swedish Water and Wastewater Association, who in their reply to the SEPA proposal (SWWA, 2014), reported that 10-35% of Swedish WWTPs will likely have trouble reaching the suggested limit for lead, cadmium, mercury or copper.

In contrast, the metal content in source separated blackwater is low enough to meet the suggested SEPA limits, both for raw blackwater and treated sludge (**Paper V**; **Paper VI**; Kjerstadius et al., 2016). This is also true for the associated nutrient recovery fractions of ammonium sulfate and struvite (Kjerstadius et al. 2016). An exception to this is sludge from a pilot area with anaerobic digestion as the core process (STOWA, 2014), but the high metal content (Table 5.1) can likely be explained by the high TS-destruction and very long sludge age in the digester due to operating below maximum capacity. Considering that the metal content in

blackwater almost exclusively originates from dietary sources (Tervahauta et al., 2014), the metal concentrations are understandably low compared to sludge from conventional WWTPs. Thus, source separation appears to be an advantageous method to achieve lower return of metals to agriculture while still meeting the suggested SEPA limits.

Table 5.1.

Suggested legal limits for metals in sludge to be returned to agriculture (mg kg TS^{-1}) together with the levels in sludge from a conventional system and source separation systems. Red fields indicate concentrations above the proposed regulations.

	Pb	Cd	Cu	Cr	Hg	Ni	Ag	Zn	Ref
Present regulation	100	2	600	100	2.5	50	-	800	1
Proposed regulation	25	0.8	475	35	0.6	30	3	700	2
WWTP sludge in Sweden									
All WWTPs National average	19	0.9	348	24	0.5	16	n.d.	568	1
Revaq-WWTPs Average	16	0.67	312	30	0.48	15	1.6	493	3
Revaq-WWTPs 75th percentile	18	0.77	375	35	0.62	19	2.0	590	3
Revaq-WWTPs 90th percentile	24	0.92	455	41	0.75	21	2.5	627	3
Source separation systems									
Sludge from source separation	22	0.68	122	19	0.17	5.8	n.d.	313	4
Struvite from source separation	0.36	0.21	0.36	0.10	0.15	1.43	n.d.	1.05	4
Sludge from digester in Sneek	43	0.8	268	16	0.6	16	n.d.	975	5

1) SBS (2016). 2) SEPA (2013). 3) Mattsson and Finnson (2016). 4) Paper VI. 5) STOWA (2014).

5.2.2 Meeting target goals for nutrient recovery

Presently, 25% of the sludge in Sweden is returned to agriculture, and almost all of the sludge being returned to farmland originates from wastewater treatment plants connected to Revaq (Mattsson and Finnson, 2016). The SEPA suggested target goals for nutrient recovery from wastewater to farmland (40% of phosphorus and 10% of nitrogen) are far from being achieved by the current sludge return. As evident from Table 5.2, there is an annual national deficit of more than 1.7 Mkg of nitrogen and 0.9 Mkg of phosphorus to reach the target goals. The deficits correspond to an increase in sludge return from 25% to approximately 43-45% (assumed potential to be $0.66 \text{ kg P capita}^{-1} \text{ year}^{-1}$ and $1.12 \text{ g N capita}^{-1} \text{ year}^{-1}$, as calculated from SBS, 2016). Similar results were observed in **Paper VI** (calculated in Kärman et al., 2017), which concluded that approximately 40%, or more, of the sludge from conventional WWTPs needs to be returned to comply with the SEPA (2013) target goals.

However, as demonstrated by the discussion of metal demands in sludge in section 5.2.1, it is unclear whether the sludge return rates will increase due to fewer treatment plants meeting the more stringent demands for metal content. Thus, one option is to complement the existing conventional system in a few urban areas with source separation systems to boost the nutrient recovery and meet the target goals for nutrient recovery and the demands for metal content. As shown in Table 5.2, this would require source separation systems for between 0.4 and 1.3 million capita (to reach the goals for nitrogen and phosphorus) out of the approximately 8 million person equivalent (pe) load to larger (>2000 pe) WWTPs on the national level, assuming that each pe corresponds to 1 capita load of nutrients, according to **Paper VI**. Such an assumption can be justified since reported data from collected statistics of the SWWA show that only 7% of the pe load to these WWTPs (>2000 pe) are due to industries (SWWA, 2016).

Table 5.2.

Required amount of capita to reach the SEPA target goals for nutrient recovery.

	Nitrogen [tonnes year ⁻¹]	Phosphorus [tonnes year ⁻¹]	Comment
Total load to Swedish WWTPs	40 194	5 641	~8 M pe BOD load to Swedish WWTPs (SBS, 2016) assuming each pe load to 0.701 kg P capita ⁻¹ year ⁻¹ and 4.993 kg N capita ⁻¹ year ⁻¹ (Paper VI).
Target goal for recovery	4 019	2 256	40% of phosphorus and 10% of nitrogen (SEPA, 2013).
Current recovery (25% sludge return to agriculture)	2 263	1 321 ¹	Calculated from total sludge production (tons of TS) and average content of phosphorus and nitrogen (mg nutrient kg TS ⁻¹) from SBS (2016). 25% sludge return is Swedish national average (SBS, 2016)
Deficit to reach target goal	1 756	935	Gap between goal and current actual recovery.
	To reach goals for nitrogen [capita]	To reach goals for phosphorus [capita]	
Needed capita with source separation	426*10 ³	1 268*10 ³	Calculated from Paper VI using the full potential return of 0.738 kg P capita ⁻¹ year ⁻¹ and 4.119 kg N capita ⁻¹ year ⁻¹ (Paper VI).

1) Agrees with SEPA (2013) who states 1 340 tonnes N year⁻¹.

5.3 Economic cost of meeting SEPA goals

Albeit not a part of the hypotheses, a comparison of the economic cost of the different paths in Figure 1.2 in relation to the SEPA goals for nutrient recovery was performed to help answer the aim of the thesis. It should be clearly noted that economic cost evaluations of similar systems have been shown to differ greatly between studies (Kärrman et al., 2017). Thus, the present comparison should be interpreted as an example calculation to indicate the magnitude of costs in order to view the potential of the investigated paths from a more realistic perspective.

5.3.1 Economic cost of implementing source separation

As calculated in Table 5.2, the required net size of areas with source separation needed to boost nutrient recovery from the current values to the SEPA target goals ranges between 0.4 and 1.3 million capita out of the approximately 8 million capita in Sweden connected to larger WWTPs. The corresponding annual economic cost of implementing source separation for the required number of people in urban areas was calculated (Table 5.3) to range between 2.3 and 7.0 billion Swedish krona (SEK). This corresponds to an increased cost of between 544 and 1 619 MSEK compared to re-implementing and operating a typical conventional system as we have today. However, the factor of scale is not included here, since the cost of source separation systems was calculated for areas with 12 000 capita, while the cost of conventional systems was calculated for areas with 100 000 capita (Kärrman et al., 2017). Of the increased cost presented in Table 5.3, roughly half (280 to 833 MSEK) would fall to the water utilities, constituting an increased cost of 35 to 100 SEK capita⁻¹ year⁻¹ if divided evenly across all water collectives (assuming 8 million capita connected to the water collectives, calculations not shown).

As a final point, the costs of wastewater infrastructure, which are dominated by sewer system costs, constitute the majority (>70%) of the annual cost (Kärrman et al., 2017). Thus, the only reasonable opportunity for implementation of source separation systems, from an economic perspective, is in new urban areas or in existing urban areas that need to replace their infrastructure system for wastewater.

Table 5.3.

Required amount of capita to reach SEPA target goals for nutrient recovery (40% phosphorus and 10% nitrogen from wastewater to farmland) and the associated annual economic cost.

	To reach goals for nitrogen [capita]	To reach goals for phosphorus [capita]	
Required capita with source separation	426*10 ³	1 268*10 ³	Calculated from Paper VI using the full potential return of to 0.738 kg P capita ⁻¹ year ⁻¹ and 4.119 kg N capita ⁻¹ year ⁻¹ .
	To reach goals for nitrogen [MSEK year ⁻¹]	To reach goals for phosphorus [MSEK year ⁻¹]	
Cost of source separation system	2 347	6 981	Calculated using the net present value method (4% depreciation cost). Total cost was 5 506 SEK capita ⁻¹ year ⁻¹ (Kärman et al., 2017).
Excess cost compared to the conventional system	544	1 619	Total cost for conventional system was 4 229 SEK capita ⁻¹ year ⁻¹ (Kärman et al., 2017).
Excess cost for water utility	280	833	Total costs for water utilities were 2 527 SEK capita ⁻¹ year ⁻¹ (source separation) and 1 870 SEK capita ⁻¹ year ⁻¹ (conventional) (Kärman et al., 2017).

5.3.2 Economic cost of sludge hygienization

The proposed SEPA (2013) regulation include proposed methods (thermophilic anaerobic digestion and sludge pasteurization) to achieve satisfactory hygienization of the sludge. The cost of these hygienization methods (including the required infrastructure upgrade) was assessed in SEPA (2013), assuming that smaller WWTPs transport their sludge to larger WWTPs where the hygienization is performed. The economic cost of treating all sludge in Sweden in this manner was assessed to be 1.2 billion SEK or an annual cost of 320 MSEK (200 MSEK for infrastructure and 130 MSEK for operations) (SEPA, 2013). Using these numbers the cost of hygienization for 50% of the sludge, roughly corresponding to the amount required to reach the target goals for nutrient recovery as stated in section 5.2.2, is approximately 160 MSEK year⁻¹ (50% of the total cost). This is a rough approximation that assumes that demands for metal concentrations can be met via upstream work to decrease metal input into the sewage system. Information spreading to upstream users to reduce the metal concentrations in sludge is part of the Revaq certification system and has likely contributed to reduced metal levels (Mattsson and Finnson, 2016). Such informative work has been calculated to cost 180 MSEK (for all WWTPs in Sweden) or 100 MSEK (for the larger WWTPs treating 80% of the annual pe load) (SEPA, 2013).

5.3.3 Comparison of economic costs

A comparison of the total approximated cost for the two paths investigated in the thesis (Figure 1.2) is presented in Table 5.4, including changes to the conventional system (sludge hygienization) or the introduction of a source separation system to boost nutrient recovery (assuming 25% sludge return still occurs). It is clear from Table 5.4 that the introduction of source separation systems is relatively expensive compared to sludge hygienization.

However, if the sludge return to agriculture does not increase to the required levels calculated in section 5.2.2, sludge incineration might be needed to dispose of the sludge. The cost of sludge incineration has been stated as an investment cost of 5 000 MSEK (SWWA, 2014) and an annual cost of 150-250 SEK household⁻¹ year⁻¹ (SWWA, 2013). For a final comparison, all the presented costs are compared (Table 5.5) to the total cost of water services (3 000-5 000 SEK household⁻¹ year⁻¹, SWWA (2013)). This comparison shows that introducing source separation is much more expensive than sludge hygienization. However, the introduction of anaerobic digestion as core process to meet SEPA demands shows potential compared to the cost of sludge incineration. Furthermore, the cost of all alternatives is relatively small compared to the total present cost for water services

Table 5.4.

Approximation of the national annual cost for water utilities to reach SEPA goals with changes to the conventional system or by introducing source separation systems. The assumptions are stated in the text.

Conventional	MSEK year ⁻¹	Source separation	MSEK year ⁻¹
Sludge hygienization (50% of sludge)	160	Construction and operation for greenfield area (increased cost compared to conventional system)	280-830
Upstream information (80% of pe load)	100	Needed hygienization at existing plants (25% of sludge)	80
<i>NET COST</i>	260		360-910

Table 5.5.

Comparison of costs for different future applications of sludge management in Sweden compared to the total annual cost for water services.

	Needed sludge hygienization	Needed source separation	Sludge incineration	Total cost for water services
Cost per household [SEK hh ⁻¹ year ⁻¹]	70	90-240	150-250	3 000 to 5 000

5.4 Related aspects

5.4.1 Using sludge and wastewater fractions in agriculture

Sludge reuse in agriculture is a heavily debated topic in Sweden (Bengtsson and Tillman, 2004; Börjesson et al., 2014; Linderholm et al., 2012), and the degree of return to agriculture, 25% national average (SEPA, 2013), is much lower than in many other European countries and the EU average of 37% (Wiechmann, 2013). Although the positive sides of nutrient recovery are generally shared, the debated issues are the presence of unwanted substances, such as heavy metals, pathogens and organic micropollutants (Bengtsson and Tillman, 2004). Current assessments in these regards (SEPA, 2013) report little evidence of harmful effects on agriculture from the current spreading of sludge, both in regards to heavy metals and organic micropollutants. This claim is further supported by a long-term study of metal uptake in soil and plants from sludge spreading for 14-53 years in Southern Sweden (Börjesson et al., 2014). However, as the Swedish sludge debate stands between a precautionary frame and a proof-first frame, scientific studies on the impact will not end the debate. Thus, it is important to realize that more factors than sludge quality, in regards to SEPA demands for metals and pathogens, determine the degree of sludge return to agriculture in Sweden. Likewise, the nutrient products recovered from source separated wastewater also have to be integrated into a local market to successfully complete the potential sustainable loop of such systems (Otterpohl et al., 1997).

5.4.2 Organic micropollutants in the nutrient fractions from source separation

Much of the pharmaceuticals we consume in our households end up in the blackwater load (Levén et al., 2016). Microbiological treatment of source separated wastewater has been shown to remove a selected range of pharmaceuticals from the water phase, mainly via sorption to sludge (Butkovskyi et al., 2015). Butkovskyi et al. (2015) concluded that post-treatment was needed for both the water and sludge (if being used as fertilizer) phases to decrease the concentration of pharmaceutical residues. Thus, anaerobic digestion (in agreement with the results in **Paper II**), does not greatly reduce pharmaceutical residues, even when applied to concentrated blackwater. That anaerobic digestion is insufficient to achieve a major reduction of organic micropollutants agrees with Falås et al. (2016).

In regards to sludge application to soil, blackwater has been shown to render similar loads compared to the return of sludge for conventional systems (Leven et al., 2016).

Thus, increased treatment or extraction of nutrients from source separated fractions (as described in **Paper V and VI**) would be necessary to achieve a reduction in the return of organic micropollutants to agriculture with source separation systems. In regards to increased treatment, source separation has the advantage that blackwater, being a minor fraction of the total domestic wastewater flow, has concentrations of organic micropollutants up to 100 times greater than in combined municipal wastewater (Leven et al., 2016; Butkovskiy et al., 2015). Thus, the application of an energy intensive oxidation treatment (like ozone) may be justified for the smaller blackwater flow, while such treatment would be more costly for the larger combined wastewater flow.

5.5 Outlook

Time perspectives in wastewater management are long due to the long life time of sewers. Thus, the current wastewater management is an adaptation to the sewer systems built approximately 100 years ago that are still in place, albeit with separate stormwater piping often being installed when renovating the sewer net. Adaptation of this system has included measures to protect receiving water bodies from nutrients by removing them from wastewater (SEPA, 2014). The central role of biological systems for nitrogen removal in conventional wastewater treatment is a direct cause of this focus on removal. Activated sludge processes are energy demanding (Gikas, 2016; Larsen, 2011) and produce N₂O emissions, which constitute a large part of the WWTP carbon footprint (Gustavsson and Tumlin, 2013). Furthermore, the removal of nitrogen from wastewater treatment plants greatly decreases the potential for recovery (**Paper VI**), a fact that directly opposes the increased need for nitrogen recovery (Jönsson, 2011; Matassa et al., 2015). Due to the large impact on the global nitrogen cycle from anthropogenic fertilizer usage (Steffen et al., 2015), increased recycling of nitrogen will likely be needed in the future. Although the greatest losses of nitrogen occur from diffuse sources in agriculture (Matassa et al. 2015), urban areas constitute a concentrated fraction of nitrogen, which can be collected with source separation systems (McConville et al., 2017). The results in this thesis suggest that a shift from conventional wastewater treatment with nitrogen removal to source separation systems with nutrient recovery would greatly aid such a collection. Furthermore, source separation systems have been shown to be beneficial in several other aspects, such as water reuse and decreased energy usage (Larsen, 2011; Skambraks et al., 2017). Large parts of the sewer system in Sweden will need to be replaced in the coming decades, and society thus faces rapid increases in economic cost for wastewater management (Malm and Svensson, 2011). The annual cost, calculated from Malm et al. (2013), corresponds to approximately 800 SEK household⁻¹ year⁻¹, which is a large cost compared to the

total cost of water services given in Table 5.5. Since the cost for sewer systems constitutes the majority of the cost for wastewater management (Kärman et al., 2017), it is difficult to justify replacement of combined sewer system before their technical life span has ended. Facing the upcoming situation of great needs for renovation or replacement of the sewer network, it is worth discussing why we should give wastewater management systems focused on removal another chance. The results in this thesis suggest that a system with source separation and anaerobic digestion as the core process is a suitable replacement, in order to change the focus of wastewater management from removal to recovery.

6. Conclusions

This thesis investigated how anaerobic digestion can be enhanced as an auxiliary or core process in Swedish wastewater management. The work focused on two possible paths to meet the target goals and demands in the proposed SEPA (2013) regulation. One path included adjustments (thermophilic anaerobic digestion for sludge hygienization) to the existing system, with anaerobic digestion as an auxiliary process. The other path included implementation of source separation systems with low-temperature anaerobic digestion as the core process. Based on the work presented in this thesis, it is concluded that:

- Satisfactory hygienization of pathogens can be achieved using thermophilic anaerobic digestion, albeit without an increased effect on the removal of organic micropollutants. The economic cost of introducing hygienization to Swedish WWTPs is relatively low. However, it is unclear whether the SEPA goals for nutrient recovery can be met by changes to the conventional system due to the suggested stringent demands on metal content in sludge as well as the low degree of sludge return to agriculture.
- The challenges of stable operation of anaerobic digestion at low temperatures can be met by applying the presented acidification limit test based on simple batch experiments. The associated challenge of extracting an increased amount of dissolved methane from the reactor effluent at low temperature can be met by using membrane contactors. These potentials facilitate the introduction of low-temperature anaerobic digestion as a core process in wastewater management.
- Source separation systems with anaerobic digestion as core process has great potential to increase nutrient recovery while constituting an opportunity to decrease the carbon footprint of wastewater management. The economic cost of introducing source separation systems is high compared to introducing sludge hygienization for the conventional system, but the cost is similar to the cost of introducing sludge incineration.

7. Future research

- The associated effects of sludge incineration need to be determined. Sludge incineration is seen as a viable option to recover phosphorus in Sweden (SEPA, 2013). However, other nutrients, mainly nitrogen, are lost during incineration, which increases the risk of sub-optimization since incineration demands large infrastructure to be economically viable. The Swedish Water and Wastewater Association clearly state that a shift to sludge incineration is a practically irreversible step for Swedish wastewater management, which will cause increased emissions of greenhouse gases, and imply that no return of nutrients, except phosphorus, will likely occur (SWWA, 2014).
- Better nutrient recovery steps for nitrogen in source separation systems are needed. The life cycle analysis in **Paper VI** showed that the climate impact and energy demand of ammonium stripping is relatively large due to the excessive use of chemicals needed due to the dilute wastewater.
- Possible improvements in nutrient recovery in the conventional system should be investigated. The calculations by De Vrieze et al. (2016) showed that ammonium stripping of the reject water from anaerobic digesters is economically viable for concentrations of 1 000-1 500 mg N L⁻¹. Such work demonstrates the opportunity for partial recovery of nutrients in the highly concentrated reject water stream in conventional systems. Furthermore, some increases in biogas production with the conventional system can likely be achieved with pretreatment methods and changes to the operational procedures (Carlsson et al., 2016).
- The effects of thermophilic and low-temperature anaerobic digestion on sludge dewatering should be investigated since even small effects will likely affect sludge transportation costs. The cost of sludge transportation may be substantial, and it has previously been shown that thermophilic anaerobic digestion affects sludge dewatering.

8. References

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This is a thesis on the management of wastewater in our cities. Especially in regards to the microbiological workers of anaerobic digestion, and the aid they can supply to our society when implemented in technological systems.

The thesis investigates two possible future paths to utilize anaerobic digestion in wastewater management. Both paths relates to the current regulation proposal on sludge management by the Swedish Environmental Protection Agency. In the first path we enhance our current system by increasing the operational temperature of anaerobic digesters at our wastewater treatment plants to thermophilic temperature. In the second path we enhance our current system by implementing source separation wastewater systems in parts of our cities in order to increase resource recovery from wastewater.

The results presented in the thesis has been obtained by both practical and theoretical experiments and, most importantly, with a lot of love for the wonders of science.