Technical University of Denmark



# Measurements of methane emissions from biogas production – Data collection and comparison of measurement methods

Energiforsk report 2015:158

Holmgren, Magnus Andreas ; Nørregaard Hansen, Martin; Reinelt, Torsten; Westerkamp, Tanja; Jørgensen, Lars; Scheutz, Charlotte; Delre, Antonio

Publication date: 2015

Document Version Publisher's PDF, also known as Version of record

Link back to DTU Orbit

Citation (APA):

Holmgren, M. A., Nørregaard Hansen, M., Reinelt, T., Westerkamp, T., Jørgensen, L., Scheutz, C., & Delre, A. (2015). Measurements of methane emissions from biogas production – Data collection and comparison of measurement methods: Energiforsk report 2015:158. Sweden: Energiforsk AB.

#### DTU Library Technical Information Center of Denmark

#### **General rights**

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.

- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

## MEASUREMENTS OF METHANE EMISSIONS FROM BIOGAS PRODUCTION

REPORT 2015:158





# Measurements of methane emissions from biogas production

Data collection and comparison of measurement methods

MAGNUS ANDREAS HOLMGREN, MARTIN NØRREGAARD HANSEN, TORSTEN REINELT, TANJA WESTERKAMP, LARS JØRGENSEN, CHARLOTTE SCHEUTZ, ANTONIO DELRE

ISBN 978-91-7673-158-1 | © 2015 ENERGIFORSK

Energiforsk AB | Telefon: 08-677 25 30 | E-post: kontakt@energiforsk.se | www.energiforsk.se

### **Authors' foreword**

This project was executed mainly by the authors

- Magnus Andreas Holmgren (SP)
- Martin Nørregaard Hansen (Agrotech)
- Torsten Reinelt and Tanja Westerkamp (DBFZ)
- Lars Jørgensen (DGC)
- Charlotte Scheutz and Antonio Delre (DTU)

and the measurement teams from AgroTech, DBFZ, DGC, DTU and SP. Invaluable contributions were also given by members of the project group

- Karine Adam, Ineris
- David Baxter, EC JRC & IEA Task 37
- Angelika Blom and Caroline Steinwig, Avfall Sverige
- Bernd Linke, ATB
- Sören Nilsson-Påledal, Tekniska Verken i Linköping
- Olga Oliveti Selmi and Zacaria Redad, GDF Suez/Engie
- Tobias Persson, Energiforsk
- Zacaria Redad, GDF Suez/Engie

Magnus Andreas Holmgren acted as project leader, and during his parental leave Johan Yngvesson, Gunilla Henriksson and Johanna Björkmalm at SP all made strong contributions to the project.

The work started in January 2014, with some main activities being performed in September 2014, namely a workshop on methane emissions arranged by IBBA and comparative measurements at a biogas plant in Linköping, Sweden.

This project was funded by the Swedish Energy Agency through Energiforsk – Swedish Energy Research Centre (former SGC). The project was also funded by Cosvegaz SA, GDF Suez/Engie, E.ON Gas Sverige, Stockholm Gas, Tekniska Verken i Linköping, Kraftringen and Avfall Sverige. In-kind contributions were given by DBFZ, DGC, DTU, Ineris and Tekniska Verken i Linköping.

We would like to especially thank Tobias Persson at Energiforsk for his strong dedication to the subject and for working so hard in making this project come true.



### Sammanfattning

Detta projekt samlar några av de mest erfarna organisationerna och människorna i Europa när det gäller mätning av metanutsläpp från produktionsanläggningar för biogas. Projektet har två huvudsyften. Dess första syfte är en litteraturstudie för att samla befintlig data och kunskap, främst i Europa, när det gäller metoder för att mäta metanutsläpp och deras resultat. Dess andra syfte är en jämförelse av några mer eller mindre etablerade mätmetoder under en jämförande mätning på en biogasanläggning. I vår kännedom har inte något liknande arbete utförts tidigare. Målet med projektet är att resultaten kan ligga till grund för ytterligare projekt, såsom en gemensam europeisk standardisering av mätmetoder och datautvärdering. Standardisering är viktigt för att säkerställa att anläggningars prestanda bedöms på samma sätt när de installeras i olika länder.

Biogas anses vara ett klimatneutralt bränsle eftersom kolet i biogasen binds från atmosfärisk koldioxid, CO<sub>2</sub>. Den CO<sub>2</sub> som frigörs vid förbränning av biogas anses därför vara biogen och inte fossil. Vidare så ersätter biogasen förbrukning av fossila bränslen vilket sänker de totala CO<sub>2</sub>-utsläppen. Biogas består huvudsakligen av metan (CH<sub>4</sub>), och eftersom metan i sig är en stark växthusgas så är det viktigt att samla kunskap om metanutsläppen som sker i form av förluster som kan uppstå i produktionskedjan för biogas, och det är också viktigt att minimera dessa utsläpp.

Metan är en växthusgas och bidrar till den globala uppvärmningen. Räknat per utsläppt ton så bidrar metan med 28-34 gånger mer påverkan jämfört med CO<sub>2</sub>. Utsläpp av ett ton metan motsvarar alltså 28-34 ton koldioxidekvivalenter. När man diskuterar metanutsläpp från biogasproduktion är det dock viktigt att komma ihåg att metanutsläpp också förekommer från naturgasanläggningar, vid alternativ behandling av gödsel och från deponier. Diskussioner har pågått i många delar av Europa och världen om det här problemet och flera initiativ för att bestämma storleken på metanförlusterna från biogasproduktion har gjorts. Mätningar har utförts med hjälp av många olika metoder och angreppssätt, och utsläppssiffror baserade på rena antaganden har också påträffats i litteraturen.

I biogas- och uppgraderingsanläggningar kan utsläpp av metan till luft förekomma i olika delar av systemet. Vanliga utsläppskällor på biogasanläggningar är ventilationer, buffert/lagertankar, rötkammare, biogödsellager och avvattningssystem. Vanliga utsläppskällor på uppgraderingsanläggningar är koldioxidrik restgas och ventilationsluft med spår av metan, samt anläggningens analysinstrument. Förutom det faktum att metan är en växthusgas så finns det en rad andra skäl till varför dessa utsläpp bör minimeras, inkluderande säkerhetsaspekter, anläggningsekonomi och lukt.

Den genomförda litteraturstudien sammanfattar ett antal studier av metanutsläpp på biogasanläggningar i ett antal olika länder. Från litteraturstudien kan man dra slutsatsen att utsläppsstudierna har genomförts med varierande metoder och angreppssätt. Den stora variationen i metoder gör att det är svårt att dra några generella slutsatser utifrån de tillgängliga resultaten. Vad som är en typisk



biogasanläggning varierar också mellan olika länder vilket gör jämförelsen ännu svårare. Rapporterade resultat av de totala metanförlusterna från biogasanläggningar sträcker sig typiskt mellan 1 - 3 % av den producerade mängden metan.

Det finns i allmänhet två fundamentalt olika angreppssätt för att mäta utsläpp av gaser från ett område eller en anläggning som har diffusa utsläppskällor, t.ex. en biogasanläggning. Dessa olika angreppssätt är fjärranalys och mätningar på plats.

Fjärranalys kan utföras med ett antal olika metoder som samtliga är inriktade på att bestämma hela anläggningens utsläpp genom att mäta metankoncentrationen i omgivningsluften vid anläggningen. I detta projekt prövades två olika tekniker för fjärranalys; dispersionsmodellering med bLs-modell ("backwards Lagrangian stochastic") och spårgasmetoden. Dispersionsmodellering baseras på mätningar av den genomsnittliga metankoncentrationen längs olika mätsträckor i vindriktningen nedströms anläggningen, samtidigt som olika väderförhållanden i omgivningen uppmäts. För detta ändamål kan ett s.k. Open-Path-system (t.ex. FTIR eller laser) användas tillsammans med meteorologiska mätningar. Spårgasmetoden kombinerar ett kontrollerat utsläpp av en spårgas från den studerade anläggningen. Eftersom denna metod använder en spårgas för att bestämma luftdispersionen så behövs inte någon matematisk modellering av denna. Fördelen med fjärranalys är att den totala mängden utsläpp från anläggningen bestäms, men metoderna kan inte användas för att lokalisera enskilda utsläppskällor eller kvantifiera deras andel av de totala utsläppen.

Mätningar på plats utförs genom att var och en av utsläppskällorna bestäms enskilt vid respektive källa. Med detta angreppssätt är det möjligt att skilja mellan stationära (t.ex. ventilation) och diffusa (t.ex. läckage) utsläppskällor. De enskilda utsläppens andel av de totala utsläppen kan också bestämmas. I detta projekt provades både direkta mätningar av metanhalt och gasflöde i utsläppspunkterna, samt provtagningssystem som provtar en stor gasvolym genom utspädning av utsläppet med luft. Speciella provtagningstekniker har också tillämpats vid exempelvis biogödsellager och biofilter, där delar av ytan täcks in med provtagningshuv. Totalt fyra olika team genomförde mätningar på plats i samband med den jämförande mätningen i Linköping.

Den jämförande mätningen inleddes också med att tre olika team genomförde läcksökning på anläggningen. Två av teamen hade tillgång till varsin IR-kamera för detektion av metanutsläpp som annars inte är synliga för blotta ögat. Det tredje teamet hade endast tillgång till ett traditionellt läcksökningsinstrument som måste föras intill ett läckage för att det ska kunna detekteras.

Slutsatsen av jämförelsemätningarna i Linköping är att de allmänna resultaten från olika metoder och arbetssätt är jämförbara. I Figur 1 ses resultat från de fyra team som mätte på plats på anläggningen. I Tabell 2 och Tabell 3 sammanfattas resultat från mätning på plats och med fjärranalys för två olika tidsperioder. Den studerade anläggningen är stor i storlek och de totala utsläppen är jämförelsevis låga. Den totala metanförlusten som rapporterats från mätningarna på plats i anläggningen varierar mellan 0,6-1,1 % av den producerade mängden metan. Det finns höga och okända osäkerheter i alla mätresultat och de beror på både analytiska osäkerheter och tidsvariation i utsläppskällor.

Några lärdomar gjordes från de jämförande mätningarna i Linköping. När schemat för mätningarna sattes inrättades det avsiktligt för att inte ha flera team i samma mätpunkt samtidigt som kunde påverka eller störa varandra. På grund av den stora tidsvariationen i utsläppskällorna har detta val dock gjort att en strikt jämförelse av resultaten inte kan göras. Variationer i driften under mätveckan inträffade som gjorde det ännu svårare att jämföra resultaten. För en framtida liknande mätkampanj rekommenderas det därför att så många mätningar som möjligt utförs parallellt, och mätningar bör kompletteras med internkalibreringar med gasflaskor med känd metanhalt.

Ett föreslaget nästa steg är att ta fram en gemensam handbok om mätningar av metanutsläpp från biogasanläggningar. Handboken ska hjälpa användaren att välja en mätmetod och ett angreppssätt anpassat till syftet med mätningen. Handboken bör lista för- och nackdelar för respektive metod och arbetssätt. Vidare bör den vägleda användaren i att analysera och förstå rapporterade värden från olika metoder och angreppssätt. Handboken skulle också vara en viktig referens vid ett framtida möjligt arbete med standardisering av metanutsläppsmätningar från biogasanläggningar.



Figur 1. Jämförelse av rapporterade värden för utsläppspunkter av låg magnitud. Utsläpp från rötrestlager och tryckvakter ingår inte i sammanställningen.

Tabell 1. Jämförelse av resultat från mätning på plats och med fjärranalys, tisdag 9 sept.

Utsläppskälla	Värde (kg CH <sub>4</sub> /h)
Summa utsläppskällor av låg magnitud	0.85
Aminskrubber, tryckvakt utblås, vänstra huset	9.2
Rötrestlager	4.4 / 6.5
Summa på plats mätningar	14.4 / 16.5
Fjärranalys spårgasmetoden (med std. avvikelse)	17.9 ± 3.1

Utsläppskälla	Value (kg CH4/h)
Summa utsläppskällor av låg magnitud	0.85
Aminskrubber, tryckvakt utblås, mellan husen	0.32
Rötrestlager	4.4 / 6.5
<u>Summa på plats mätningar</u>	5.6 / 7.6
Fjärranalys bLs modell (med std. avvikelse)	4.9 ± 1.5

### Summary

This project gathers some of the most experienced organizations and people in Europe regarding the measurement of methane emissions from biogas production plants. The project has two main objectives. The first objective is a literature study to gather existing data and knowledge, mainly in Europe, regarding methane emission measurement methods and results. The second objective is a comparison of some more or less established measurement methods during a joint comparative measurement campaign at a Swedish biogas plant, "Tekniska Verken i Linköping". To our knowledge similar work has not been performed previously.

Biogas is regarded as a climate-neutral fuel since the carbon in the biogas is fixed from atmospheric carbon dioxide, CO<sub>2</sub>. Biogas consists mainly of methane (CH<sub>4</sub>), and since methane in itself is a strong greenhouse gas, it is important to gather knowledge about the methane emissions in the form of losses that might occur in the biogas production chain, and subsequently it is important to minimize these emissions. When discussing methane emissions from biogas production, it is important to bear in mind that methane emissions do also occur in natural gas installations, in alternative manure treatment and in landfills.

From the literature study it can be concluded that a number of studies of the methane emissions from biogas plants have been performed in different countries, using different methods and approaches. The large variation in methods makes it hard to draw general conclusions from the existing data. A rather large variation between typical plants in different countries makes the comparison even harder. Reported results of the total methane losses from biogas plants typically range between 1 - 3 % of the produced methane.

There are in general two fundamentally different approaches to measure gas emissions from an area or facility that have diffuse emission sources, such as a biogas plant. They are remote sensing and on-site measurements. Comparative measurements were performed by six different teams, four of them applied on-site measurement methods and two performed remote sensing methods. The conclusion of the comparative measurements is that the general results from different methods and approaches are comparable. The studied plant is large in size and the overall emissions are comparably low. The total methane loss reported by the on-site measurement teams range between 0.6 - 1.1 % of the produced methane. There are high and unknown uncertainties in all measurement results and they are due to both analytical uncertainties and time variation in emission sources during the testing period of one week.

A suggested next step would be the production of a handbook on methane emission measurements from biogas installations. This handbook should aid the user in choosing a suitable measurement method and approach, depending on the purpose of the measurement task. It should list advantages and disadvantages of the respective methods and approaches. Further, it should guide the user in analyzing and understanding reported values using different methods and approaches. The handbook would serve as an important reference to future work on standardization of methane



emission measurements from biogas installations. Standardization is important in order to ensure that plant performance is deemed likewise when installed in different countries.

### **Table of contents**

1	Back	ground		14			
	1.1	Goal o	of the project	14			
2	Mea	sureme	nt methods	16			
	2.1	Remo	te sensing	17			
		2.1.1	The inverse dispersion modelling method	17			
		2.1.2	The tracer dispersion method	18			
	2.2	Leak d	detection	19			
	2.3	On-sit	te measurements	19			
		2.3.1	Methane concentration	19			
		2.3.2	Direct measurement of gas flow	20			
		2.3.3	Open area measurements	20			
		2.3.4	High volume sampler technique	22			
3	Biom	nethane	production and methane emission measurements in the				
	studi	ied coui	ntries	24			
	3.1	Biome	ethane production	24			
		3.1.1	Austria	24			
		3.1.2	Denmark	25			
		3.1.3	France	25			
		3.1.4	Germany	26			
		3.1.5	Sweden	26			
		3.1.6	Switzerland	27			
	3.2	Existir	ng measuring programs and performed measurements	27			
		3.2.1	Austria	27			
		3.2.2	Denmark	28			
		3.2.3	France	29			
		3.2.4	Germany	30			
		3.2.5	Sweden	35			
		3.2.6	Switzerland	36			
		3.2.7	Other countries	40			
4	Mea	leasurements at Tekniska verken in Linköping, Sweden					
	4.1	Plant description					
	4.2	Leak detection					
	4.3	On-sit	te emission measurements	49			
		4.3.1	Homogenization and hygienization tanks	49			
		4.3.2	Biofilter	52			
		4.3.3	Digestate storage	53			
		4.3.4	Activated carbon filter building	55			
		4.3.5	Chemical scrubber, CO <sub>2</sub> release	55			



		4.3.6	Chemical scrubber, compressor buildings	56
		4.3.7	Chemical scrubber, pressure relief vents	57
		4.3.8	Analysis instruments on site	57
		4.3.9	Comparison of on-site measurements	58
	4.4	Remo	te sensing emission measurements	60
		4.4.1	The backward Lagrangian stochastic method	60
		4.4.2	The tracer dispersion method	61
		4.4.3	Comparison of remote sensing results	61
	4.5	Comp	arison of on-site and remote sensing results	65
5	Concl	usion		67
	5.1	Contir	nued work	67
6	Litera	ture		68

#### Annex

#### A. AgroTech

Identification and quantification of methane emission from leaks at biogas plants

#### B. DBFZ - on-site method

Methane emissions from a biogas production site in Linköping

#### C. DBFZ - remote sensing method

Methane emissions from a biogas production site in Linköping

#### D. DGC

Methane emissions from biogas plant

#### E. DTU

Quantification of fugitive methane emissions from the biogas plant in Linköping

#### F. SP

Methane measurements at the Linköping biogas plant



### Abbreviations

Abbreviation	Explanation
ATEX	Appareils destinés à être utilisés en ATmosphères EXplosibles. 2 EU directives to protect employees from explosion risk in areas with an explosive atmosphere.
bLs	backward Lagrangian stochastic (model).
CHP	Combined Heat and Power (unit).
CRDS	Cavity Ring-Down Spectroscopy.
DBFZ	Deutsches Biomasseforschungszentrum. German Biomass Research Centre.
DGC	Dansk Gasteknisk Center. Danish Gas Technology Centre.
DTU	Danmarks Tekniske Universitet. Technical University of Denmark.
FID	Flame Ionization Detector.
FTIR	Fourier Transform InfraRed spectroscopy.
HBT	Hohenheimer Biogas yield Test.
IEA	International Energy Agency.
IR	InfraRed spectroscopy.
ISO	International Organization for Standardization.
LEL	Lower Explosive Limit. The lowest concentration of a gas in air capable of producing a flash of fire in presence of an ignition source. Methane gas has a LEL of 4.4 vol- %.
OTNOC	Other Than Normal Operating Condition.
ppm	parts per million
SP	Technical Research Institute of Sweden.
TDLAS	Tunable Diode Laser Absorption Spectroscopy.
VDI	Verein Deutscher Ingenieure. Association of German Engineers.



### Definitions

Phrase	Definition
Active area source	An area source of emission with gas flow being actively driven off, typically an open biofilter where a fan controls the gas flow through the filter.
Closed chamber	Sampling hood which encloses the sampling area and does not allow for air passing in or out of the chamber, also called static chamber.
Open chamber	Sampling hood which encloses the sampling area and allows for air entering the chamber with a known mass flow and allows for air leaving the chamber, also called dynamic chamber.
Passive area source	An area source of emission where gas flow is not being actively driven off.



### 1 Background

Biogas is regarded as a climate-neutral fuel since the carbon in the biogas is fixed from atmospheric carbon dioxide (CO<sub>2</sub>). The CO<sub>2</sub> released when combusting biogas is therefore regarded as being biogenic rather than fossil. Further, any consumption of fossil fuels replaced by biogas will lower the total CO<sub>2</sub> emissions. Biogas consists mainly of methane (CH<sub>4</sub>), and since methane in itself is a strong greenhouse gas, it is important to gather knowledge about the methane emissions in the form of losses that might occur in the biogas production chain, and subsequently it is important to minimize these emissions.

Methane is a greenhouse gas and contributes to global warming. Calculated per emitted ton, methane contributes 28-34 times more compared to CO<sub>2</sub>. Thus, a methane emission of one ton corresponds to 28-34 tons of carbon dioxide equivalents [1]. When discussing methane emissions from biogas production, it is important to bear in mind that methane emissions do also occur in natural gas installations, in alternative manure treatment and in landfills.

The Renewable Energy Directive (Directive 2009/28/EC) was adopted in 2009 by the EU. According to the Directive, by 2020, all member states shall have 10 % (on energy basis) renewable fuels in the transport sector. A number of sustainability criteria must be met in order for a biofuel to be accounted. To be accounted as sustainable the greenhouse gas emission savings from the use of the biofuel must be at least 35 % compared with the use of a fossil fuel. Over time the greenhouse gas constraint increase, the savings must be 50 % by the year 2017 and from the year 2018, biofuels produced in plants taken into operation after January 1st 2017, the savings must be at least 60 % [2]. Consequently the sustainability criteria are closely related to methane emissions and measurements of these at biogas plants.

Discussions have been ongoing in many parts of Europe and the world about this issue and several initiatives to determine the size of the losses have been made. Measurements have been performed using many different approaches and methods, and methane emission figures based on assumptions have also been found in literature.

In biogas and upgrading plants methane emissions to air can occur in different parts of the system. Common sources of emissions at biogas producing plants are ventilation, buffer/storage tank, digester, digestate storages and dewatering equipment. Common sources of emissions at biogas upgrading plants are the off-gas, ventilation and analysis instruments. Except for the fact that methane is considered a greenhouse gas there are other reasons to why these emissions should be minimized and these are: safety aspects, economy and odor [3].

#### 1.1 GOAL OF THE PROJECT

The project has two main objectives. The first objective is a literature study to gather existing data and knowledge, mainly in Europe, regarding methane emission measurement methods and results. The second objective is a comparison of some more



or less established measurement methods during a joint comparative measurement campaign at a Swedish biogas plant, Tekniska verken in Linköping. To our knowledge similar work has not been performed previously.

The goal of the project is that the results can serve as a basis for further projects, such as a common European standardization of measurement procedures and data evaluation. The latter is seen as particularly important in order to make available measurement results comparable. Standardization is important to ensure that plant performance is deemed likewise when installed in different countries.



### 2 Measurement methods

#### In general, there are two fundamentally different approaches to measure gas emissions from an area or facility that have diffuse emission sources, such as a biogas plant. They are remote sensing and on-site measurements.

Remote sensing measurement methods include a number of different methods all aiming for quantification of whole site emissions by measuring atmospheric methane concentrations often off-site but not always. In this project two remote sensing methods were tried out; the backward Lagrangian stochastic (bLs) model and the tracer dispersion method. The bLs model is based on measurements of average methane concentrations along different paths downwind the facility together with simultaneously measuring the meteorological conditions in the surroundings. For this purpose an Open-Path-System (e.g. FTIR or laser) can be used along with one or more meteorological stations. With this data the emission rate can be simulated with an inverse dispersion model or radial plume mapping. The tracer dispersion method combines a controlled tracer gas release from the treatment facility with time-resolved concentration measurements downwind of the facility [4]. As the method applies a tracer gas to determine the air dispersion, modelling is not needed. The advantage of both methods is that they determine the total amount of emission from the full scale facility - integrating the individual emissions from different on-site sources/leak areas. Thus the methods cannot be used to locate single on-site emission sources or quantify their share of the total emission.

On-site measurement methods aim to identify and quantify single sources of emission at the site. This approach enables the differentiation between stationary (e.g. gas upgrading) and diffuse (e.g. leaks) sources of emission.

Some advantages and disadvantages of these methods are listed in Tabell 3.

	Remote sensing	On-site
Advantages	Determination of total emission rate	Localization of single sources Quantification of single sources
	Longer time measurement series	Low detection limit for total emission rate
	No influence on plant operation/design	Weather independent
	No leakage search and encapsulating	
	Time effort independent of plant size	
	No modeling step involved (tracer disp.)	

Tabell 3. Advantages and disadvantages of remote sensing and on-site measurement



	Remote sensing	On-site		
Disadvantages	Depending on weather conditions (wind direction and wind speed)	Only short time window per single source Possibly not all sources		
	Disturbance by topography,	accessible		
	buildings, trees, etc (especially relevant for bLs)	Time consuming on large plants		
	Uncertainties of inverse dispersion modelling (only bLs)			
	Requires site access and prober tracer placement (only tracer dispersion)			
	Requires drivable roads in prober distance to the site (only tracer dispersion)			

#### 2.1 REMOTE SENSING

To date no international standard exists for the measurement of fugitive and diffuse methane emissions from anaerobic digestion facilities. However, there is a basic guideline on the investigation of diffuse sources (VDI 4285) where remote sensing methods are considered. In addition, a mandate from the European Commission to CEN has been issued to standardize measurement techniques for the emission monitoring of diffuse emissions from industrial sites, such as refineries [5]. Several remote sensing methods are descripted in detail by the EPA handbook [6].

#### 2.1.1 The inverse dispersion modelling method

Total methane emissions were quantified using an open path spectrometer sensitive to methane in the downwind plume, a meteorological station with 3D sonic anemometer and a backward Lagrangian stochastic model. This model is based on the Monin-Obukhov similarity theory [7]. The model approximates the emission source as a homogeneous area. If the measurement path is placed far enough downwind this is a viable simplification [8]. The spectrometer measures the path averaged methane concentration on path lengths of up to 500 m. Alternative to TDLAS devices the methane concentration can be determined by Open Path Fourier Transform Infrared Spectroscopy (FTIR) or mobile point measurement devices. The advantage of the open path devices is that a continuous measurement covering the same path is possible over a long time period. A 3D sonic anemometer is essential to register the turbulence parameters in the atmosphere. These are needed for the simulation of the emission rate. Apart from Windtrax other Lagrangian dispersion models are available, e.g. LASAT (Germany) or NAME (United Kingdom). This method has already been used to determine emissions from farms, landfills and biogas plants. The setup is illustrated in Figure 1.

For this project DBFZ applied the bLs method for whole site methane quantification, see Annex 3.





Figure 1. The setup of TDLAS for bLs modelling of the emission rate from area sources.

#### 2.1.2 The tracer dispersion method

Total methane emissions can be quantified using a mobile tracer dispersion method that combines a controlled release of tracer gas from the biogas facility with concentration measurements downwind of the facility, by using a mobile high-resolution analytical instrument ([9], [10], [11]). The tracer dispersion method is based on the principle that a tracer gas released at a source area, in this case a biogas facility, disperses into the atmosphere likewise the methane emitted from the same area. Since the ratio of their concentrations remains constant along their atmospheric dispersion, the methane emission rate can be calculated using the following expression when the tracer gas release rate is known:

$$E_{CH_4} = Q_{tr} * \frac{\int_{plume\ end}^{plume\ end} (C_{CH_4}) dx}{\int_{plume\ end}^{plume\ end} (C_{tr}) dx} \frac{MW_{CH_4}}{MW_{tr}}$$

where  $E_{CH_4}$  is the methane emission in mass per time,  $Q_{tr}$  is the tracer release in mass per time,  $C_{CH_4}$  and  $C_{tr}$  are the measured downwind concentrations in parts per billion (ppb) subtracted of their background concentrations and  $MW_{CH_4}$  and  $MW_{tr}$  are the molar weights of methane and tracer gas, respectively [9].



Figure 2. The principle of the dynamic plume method for quantifying GHG emissions from area sources.

The tracer gas should have a long atmospheric lifetime. Tracer gases often used include N<sub>2</sub>O, SF<sub>6</sub>, and acetylene. Downwind plume concentrations are measured driving along transects with an analytical equipment, which is a fast and has a high sensitivity capable of detecting methane and tracer gas concentrations down to ppb level every



second ([9], [11]). A GPS should be connected to the instrument for logging the measured concentrations to their geographical location. Correct simulation of the methane emitted from the source is very important to obtain precise emission rates. In order to obtain the best possible simulation of the source area, the tracer gas should be released from the part of the plant where the most elevated methane concentration are seen and/or expected. The principle is illustrated in Figure 2.

For this project DTU applied the tracer dispersion method for whole site methane quantification, see Annex 5.

#### 2.2 LEAK DETECTION

Leak detection, as the name suggests, is performed to detect (to locate) leakages. The methods used for leak detection can not necessarily be used to quantify the size of the emission, or to calculate the mass flow of e.g. methane.

The simplest form of leak detection can be performed without technical equipment, using only the human senses. Leaks can be detected by smell, sound, vision or touching a leaking object. With the additional help of leak detection spray, that helps to produce foam or bubbles at the leak, many leaks can be detected and hence hopefully avoided.

For performance of systematic leak detection it is however recommended to use a portable leak detection instrument, using a sensor that reacts to a rise in methane concentration. Common simple and cheap leak detection instruments have detection limits of 1-5 ppm, which will enable the user to find very small leaks. Some leak detection instruments will give a value of the methane concentration on a display. Even though the uncertainty of this value is comparably large it is allowed to use in the Swedish voluntary agreement system if the emission in a certain measuring point is less than 0.1 % of the total amount of methane produced in the plant and less than 10 % of the total emissions. Common sensors are semiconductor sensors or catalytic sensors [3].

In recent years methane lasers have been developed. These instruments measure the total methane concentration along the trajectory of a laser beam that is emitted from the instrument, giving the results in a path-integrated unit (ppm m). This technique makes it easier to perform leak detection over large areas.

The most advanced leak detection instrument available is the IR camera, which continuously shows an image where leaking methane can clearly be seen in the picture, which is otherwise not visible by the human eye. The IR camera is capable of producing both images and videos. With the aid of an IR camera, large areas can quickly be scanned for leaks. The IR camera is not capable of detecting very small leaks, an approximate limit of detection is 0.5 vol-% methane concentration, but the size of the leak (the mass flow) will also have an influence on the detection limit.

#### 2.3 ON-SITE MEASUREMENTS

#### 2.3.1 Methane concentration

The measurement of methane concentration in waste gases from stationary sources is standardized in EN ISO 25140:2010 and EN ISO 25139:2011. EN ISO 25140:2010 describes the continuous method of using a Flame Ionization Detector (FID), together



with a Non-Methane Hydrocarbon cutter which filters out other hydrocarbons than methane. This instrument usually has a wide range of concentration measuring ranges, from 1 ppm up to 100 000 ppm (10 vol-%). This technique has been applied by DGC and SP in this project, see Annexes 4 and 6. The manual sampling method where gas samples are taken in bags for later analysis with a gas chromatograph (GC) in a laboratory is described in EN ISO 25139:2011. This technique has been applied by DBFZ in this project, see Annex 2 (instead of gas bags evacuated glass vials with 22 ml volume were used).

Apart from these standardized methods there are also other techniques available to determine the methane concentration such as photo-acoustic, IR and FTIR analysers. The photo-acoustic technique has been applied by AgroTech in this project, see Annex 1.

#### 2.3.2 Direct measurement of gas flow

Determining the gas flow is a great challenge in these measurements since the situation varies a lot between different sampling points and between plants. In addition, the sampling points/sources of emission are rarely prepared for measurements. The main goal is to measure the flow but in many cases it is technically or practically difficult to perform. There are a number of measurement methods to use, for example pitot tube measurements with differential pressure readings, hot wire anemometer, vane anemometer etc.., and a standardized approach is described in EN ISO 16911-1:2013. The technique of direct measurement of the gas flow has been regularly applied by SP in this project, see Annex 6, but other teams also used the technique for some emission sources.

If measurements are difficult or impossible to perform there are other ways to determine the gas flow, e.g. the manufacturer's fan data for a forced ventilation release and as a last resort template values can be used.

#### 2.3.3 Open area measurements

Concentration and flow measurements as described above cannot be performed on open area sources, such as digestate storage tanks or biofilters. Different types of sampling hoods do exist, which can be used to encapsulate a small area of the emission source, enabling measurement and calculation of the total area source emission.

The simplest case of such a sampling hood is the sampling hood for biofilters, where the hood has a surface area of typically 1 m<sup>2</sup> at the bottom placed on the biofilter surface, and a sampling pipe area of typically ~0,0001 m<sup>2</sup> at the top, see Figure 3. In this way the very low gas flow through the biofilter surface is possible to measure. This type of sampling hood, for *active* area sources, is standardized by VDI [12]. This type of sampling hood was however not used in this project.





Figure 3. Sampling hood for biofilters.

For other area sources, such as digestate storage tanks, sampling hoods based on open chamber and closed chamber techniques can be used (see definitions). For a closed chamber a certain sample area is enclosed by the closed chamber and the gas concentration within the chamber is sampled periodically [13]. The measuring principle is based on the measurement of an increasing gas concentration inside the chamber volume. After putting the closed chamber on the emission surface a gradually increasing gas concentration can be determined. For this project DBFZ applied the closed chamber technique at some emission sources, see Annex 2.

For the closed chamber, samples are taken at certain time intervals (e. g. 5, 10, 15, 20, 25, 30 min). Then the emission rate can be calculated from the slope of the gas concentration, the chamber volume and the encapsulated surface area according to Equation 1.

$E_{Spec} = \frac{\partial c}{\partial t} \cdot \frac{V}{A} \cdot 0.06$		Equa	ition 1
	Espec	Surface specific emission mass flow	g m <sup>-2</sup> h <sup>-1</sup>
	$\frac{\partial c}{\partial t}$	Linear slope of gas concentration	mg m <sup>-3</sup> min <sup>-1</sup>
	V	Volume inside the chamber	m <sup>3</sup>
	А	Encapsulated surface area	m <sup>2</sup>

For an open chamber measurement a known flow of air is passed through the chamber allowing for calculation of the mass flow emission, see Figure 4. Both the open chamber and closed chamber sampling hoods, for *passive* area sources, are standardized by VDI [14]. For this project, SP applied the open chamber technique at the digestate storage tank, see Annex 6. The open chamber technique can also be applied when measuring the emission from leakages, as has been applied by DBFZ in this project, see Annex 2.





Figure 4. Schematic principle of an open chamber.

#### 2.3.4 High volume sampler technique

The basic technique of the high volume sampler is evacuating the leaking biogas from a hole or crevice together with a large amount of diluting air. The sample of diluted biogas is conveyed to a flow measuring device and FID analyzer and the total mass flow of leaking methane is calculated as the product of volume flow and methane concentration above background value in the diluting air.

To control the leaking biogas flow the actual leakage (the hole or crevice) is covered by a suction hood or wrapped in plastic sheet, allowing for dilution air and leaking biogas to be conveyed to the flow and concentration measurement.

For this project AgroTech and DGC applied the high volume sampler technique at most emission sources, see Annexes 1 and 4.



Figure 5. High volume sampler.

Figure 5 shows the principle of the sampling technique and a picture of a high volume sampler where the sampling hood is attached to a silo tank.



The sample system consists of:

- Sampling hood or
- Antistatic ventilation hose
- ATEX approved blower
- Control box including safety circuit in case of too high concentration of methane in the sample gas (alarm breaks in at 10 000 ppm methane equivalent to 25 % of LEL)
- Flow measurement (calibrated orifice)
- FID equipped with a Non-Methane Hydrocarbon cutter according to section 2.3.1.

The high volume sampler technique is described elsewhere and may not be exactly the same layout as described above, but the main principle is still the determination of coherent values of sample gas flow and methane concentration.

Also portable, intrinsically safe, battery-powered high flow samplers are commercially available. These instruments are mainly designed to determine the rate of gas leakage around various pipefittings, valve seals and compressor seals found in natural gas transmission, storage, and compressor facilities.



### 3 Biomethane production and methane emission measurements in the studied countries

This chapter details the biomethane (biogas) production in studied European countries and also gives an overview of the known methane emission measurements that have been performed.

#### 3.1 BIOMETHANE PRODUCTION

*This chapter is based primarily on the publication IEA Bioenergy Task 37 – Country Reports Summary 2014* [15]. *Unless stated otherwise, this is the source for all information given herein.* 

The biogas market has been established in a number of European countries with significant growth in the 2000 – 2020 period. The largest producers of biogas today are Germany, Italy, UK and France [16].

The largest European producers of biomethane (biogas upgraded to natural gas quality) are Germany, Sweden, Austria, Switzerland and the Netherlands [16]. Countries such as Denmark and UK are growing fast and are currently constructing several biogas upgrading plants due to the introduction of attractive financial incentives.

Biogas is produced from biogas in landfills and in anaerobic digesters. The feedstocks used for anaerobic digestion varies in different countries. The main production is based on energy crops in Germany while it is mainly based on various waste products in Sweden and UK [15].

Also the utilisation of the biogas varies between the countries. Electricity production is dominating in Germany and UK while automotive fuel is dominating in Sweden. The utilisation of biogas as automotive fuel is gaining more interest in several countries and is expected to grow significantly in countries such as Denmark, Germany and UK [15].

Below follows a description of the current situation and expected development in the countries that are further discussed in this report.

#### 3.1.1 Austria

Today the main production of biogas is derived from energy crops, sewage sludge and landfills. The annual biogas production corresponds to 1.5–2.5 TWh. Current trends are that high prices of biogas feedstock (e.g. maize) lead to severe difficulties to operate the plants economically. This has led to keen interest to investigate the possibility to use alternative substrates. There is a total of 368 biogas plants in Austria.

In Austria biogas is utilised mainly for electricity and heat production. Even though the aim is to upgrade more biogas to biomethane for use as automotive fuel, this change is taking place rather slowly. There are around 7,700 natural gas vehicles (NGVs) and about 180 compressed natural gas (CNG) filling stations. Three of the filling stations are situated at biogas upgrading plants. There are 11 biogas upgrading units in operation. All commercial technologies are represented (amine scrubber, water scrubber,



membrane and PSA). The upgrading plants are rather small, 600-800 Nm<sup>3</sup>/h, and have a combined capacity of around six million Nm<sup>3</sup> biomethane annually.

#### 3.1.2 Denmark

154 biogas plants are in operation in Denmark, with a yearly production of 1.2 TWh of biogas. Animal manure is the most important biogas feedstock, with a high prospective potential. According to the Danish Biogas Association, roughly 7% of the animal manure is today supplied to biogas plants in Denmark. The aim is to increase it to 50% by 2020. Along with manure, organic wastes from industry and sewage sludge also make a significant contribution to the biogas production today. In 2012 the Danish Energy Agency predicted a 4-fold increase (to 4.7 TWh) of the total biogas production by 2020. The Biogas Task Force concluded in 2013 that the increase will only be a doubling, to around 3 TWh by 2020. A limited number of biogas projects, representing an increase of about 400 GWh, have already reached a final decision.

Today biogas is mainly used for heat and power production in Denmark. The Danish government believes that biogas will be an important automotive fuel in the future, especially when replacing fossil fuels used by heavy duty vehicles. The first four Danish biogas upgrading plants are in operation and a number of biogas upgrading projects are at various stages of planning. There are seven biogas filling stations and more are about to be established. Currently, around 80 CNG cars are in operation in Denmark. In 2013 there was only one small biogas upgrading plant in operation in Denmark that was supplying biomethane to the natural gas network. In 2014, four new biogas upgrading plants were taken into operation. The use of biogas in Denmark as a transport fuel is only in its early days, and the main driving force is government policies in the form of certificates. At the end of 2014 there were 10 filling stations for compressed natural gas (CNG).

#### 3.1.3 France

The vision of the French Environment and Energy Management Agency is to produce 70TWh biogas annually by 2030 and that 600 biogas plants will be built every year. 50% of the biogas produced shall be injected into the grid, 30% shall be used to generate electricity and the remaining 20% shall be used to produce heat. In 2050, the aim is to produce 100TWh.

In France there are 256 biogas plants and 245 landfills. The number of farm AD plants is expected to double by the end of 2013. A recent study financed by ADEME on The estimation of feedstock for AD use shows that the potential resources for AD will give a probable potential of 56TWh by 2030. Based on its own calculation, an estimation of ADEME expects a theoretical production of 70TWh by 2030.

In France there is a strong development of on-farm and centralized biogas plants and for landfills to recover biogas for electricity generation (today only 90 out of 245 landfills utilize biogas). Around 120 on-farms AD plants were built by the end of 2013 and nearly 15 centralized units. In addition, 60 WWT and 80 agrofood industries AD plants are currently in operation. In 2010, a study showed a relatively low energy recovery from biogas, around 60% of raw energy, the main part coming from landfills.

There are only four biogas upgrading plants in operation, but more than 400 applications to inject biomethane into the natural gas grid, which indicate a significant increase of the number of upgrading plants in a nearby future. Today, all the



biomethane produced is injected into the natural grid or sold as fuel for vehicles. More than 13,500 vehicles, of which 3,500 are trucks, are in use in France with a daily consumption of 265,000 Nm<sup>3</sup>. 37 public filling stations and around 130 private filling stations are in operation.

#### 3.1.4 Germany

The share of renewable energies in energy generation is to be raised to 40-45 percent by 2025, to 55-60 percent by 2035, and to 80 percent by 2050. The reform of the Renewable Energy Sources Act (EEG) should play a key role in the success of the energy reforms. The introduction of specific growth targets for different technologies is a new development for the German renewables support scheme. The annual growth of biomass including biogas is limited to a maximum of 100 MW compared to 2,500 MW for onshore wind and solar power.

The 7960 biogas plants in the agriculture sector make the biggest contribution to biogas production today with electricity and heat supply of 25,120 GWh/year and 10,550 GWh/year, respectively. According to information from the Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Safety, in 2013 the main part of the biogas was used for electricity and heat production, while biogas utilization as an automotive fuel is rare. The share of energy consumption in Germany for electricity, heat and fuel amounted to 4.7%, 1% and 0.1%, respectively.

In 2014 a total number of 151 biogas upgrading plants were in operation with a feed-in capacity to the gas grid of 93,650 Nm<sup>3</sup>/h biomethane. Compared with data from the previous year, the number of plants increased by 25%. Amine scrubbing, water scrubbing and pressure swing adsorption (PSA) are the most commonly applied technologies. Due to the reduction in the feed-in tariffs, the number of new biogas upgrading plants will presumably be smaller than the 20-30 plants/year erected in previous years. Surveys have revealed that at least five of the projects under construction or in planning are not realized due to the 2014 EEG reform and/or a continuation of the projects are not yet finally clarified.

Based on data from Erdgas Mobil GmbH in 2013, about 170 filling stations with 100% biomethane have sold 300 TWh biomethane. This corresponds to 20% of natural gas consumption by the 95,000 registered gas vehicles in Germany.

#### 3.1.5 Sweden

In Sweden there is a governmental aim to produce 50 percent of the energy from renewables by 2020 (this has already been reached), but there are no specific targets for biogas production. Sweden also has a governmental vision to have a fossil free transportation sector by 2050. The results from a public inquiry on how to reach this vision are expected to be important for the future governmental support for biomethane production in Sweden.

In Sweden the production of biogas has been fairly constant at around 1.3–1.7 TWh for several years. The main reason is the difficulties in showing a reasonable profit for new investments and new biogas plants. Biogas produced in new plants has been balanced by the steady decline in landfill gas production.

In Sweden, around 50% of the biogas is used as CNG/biomethane. This part is increasing every year to meet the increasing demand from the increasing number of gas



vehicles. The main part of the remaining biogas is used for heat production. Nearly all upgraded biogas is used as automotive fuel, the annual biomethane production in Sweden is around 900 GWh. The biomethane is produced in 53 biogas upgrading plants with various technologies (~70% water scrubbers and, ~15% PSA, ~15% amine scrubbers). In one plant, with a capacity of 60 GWh, the biomethane is liquefied and sold as LBG (Liquefied Biogas). Of the methane used as an automotive fuel, the biomethane share was 58% on energy basis in 2013. It is used by 47,000 gas vehicles, of which 2,200 are buses and 750 are heavy duty vehicles. Around 210 filling stations dispense CNG/biomethane, five of these also have liquefied gas, LNG/LBG.

#### 3.1.6 Switzerland

The Federal Council has adopted the energy strategy 2050 in order to guarantee security of energy supply in the long term. The thrust of this strategy is to gradually phase out nuclear power and, on the other hand, to develop hydro power as well as the new renewable energies (sun, biomass, biogas, wind, wastes and geothermal heat) and to improve energy efficiency of buildings, appliances and transportation. Energy supply difficulties could be overcome by fossil-fuelled power generation and by energy imports. Concerning biogas, the energy strategy aims an annual electricity production of 1.6 TWh by 2050. In order to reach this goal, the focus is on coordinated energy research.

In Switzerland there are around 610 biogas plants and six landfills. The total gross biogas production was 1,129 GWh in 2013. The biogas is mainly used to produce electricity and heat in CHP plants, but the biomethane production is growing rapidly. There are 19 upgrading plants (mainly PSA units, amine scrubbers and organic physical scrubbers), two at agricultural sites, eight at wastewater treatment plants and seven at biowaste AD sites, with a total biomethane production of approximately 128 GWh. The target is to inject 300 GWh into the grid by 2016. Today more than 11,000 vehicles run on methane and 140 filling stations are in operation.

#### 3.2 EXISTING MEASURING PROGRAMS AND PERFORMED MEASUREMENTS

This chapter briefly describes existing measuring programs and other performed measurements in different countries which are known to the project group.

#### 3.2.1 Austria

In Austria a large project called "Klimoneff" was performed in the last years [17]. The focus lay on the quantification of fugitive methane emissions from agricultural biogas plants using a remote sensing method and on the analysis of digestate.

The remote sensing method in use was Open Path TDLAS in the downwind plume in combination with inverse dispersion modelling (refer to section 2.1). The emission quantifications were performed on five facilities. One of those was analysed in detail. For this biogas plant the results from seven days of data collecting gave a median value of 4 % loss of the produced methane when the open digestate storage tanks were filled. During periods when the storage tanks were empty, data from 6 measurement days led to quantification of 3 % methane loss. The four other plants were surveyed for two times five hours each. The estimated methane loss during these periods was given within the range from 1.6 up to 5.5 %. At least two of these plants had open digestate



storage tanks. For parts of these measurements operational problems of the facilities were reported.

#### 3.2.2 Denmark

Until 2012/13 there have been no studies in Denmark measuring and documenting methane leakages and emissions from biogas plants.

In April 2013 AgroTech and DGC initiated a project with the overall aim of reducing greenhouse gas emission from biogas plants in terms of methane losses. This is done by developing a method for identification and quantification of methane leakages. In the project, "Methane Emission from Danish Biogas Plants" funded by Energinet.dk under the ForskEL programme, nine biogas plants were screened for methane emissions and the identified leakages were subsequently quantified. One of the tasks in the Danish project is to test and evaluate the selected method against another method. Participating in the Linköping measurements in this current European project fulfil this demand. The project has developed an operational method for identification and measurement of methane emission from biogas plants. The developed method was utilized to identify and quantify methane emission at ten Danish biogas plants. In total 50 leakages were identified. The measured methane emission from individual leakages varied considerably from zero to 276,000 Nm<sup>3</sup> CH<sub>4</sub> per year. The total amount of methane emitted from the studied biogas plants summed up to 4.2 % of the total methane production at the studied plants.

In 2012/13 DTU Environment performed emission measurements at Avedøre wastewater treatment plant located in the Copenhagen area, Denmark [11]. In total, 9 measurement campaigns were conducted where the total emission of methane and N2O from the plant was quantified using the tracer dispersion method. The measurements performed on the plant showed that most of the methane emission occurred from the anaerobic sludge treatment, while the majority of the N<sub>2</sub>O emission derived from the biological nitrogen removal in the aeration tanks. The methane emission rate varied between 5 kg  $h^{-1}$  up to 92 kg  $h^{-1}$ , whereas the emission of N<sub>2</sub>O ranged from the detection limit (0.4 kg h-1) up to 10.5 kg h-1. The high emissions were observed during periods when the plant experienced operational problems. For example, the highest emission of methane was measured during a period with foaming problems in the anaerobic digesters, while the highest N<sub>2</sub>O emission was observed during sub-optimal operation of biological nitrogen removal in the secondary treatment of wastewater. The measured methane emissions accounted for between 2 and 33% of the total methane production at the plant. Similarly it was seen that between 0.15 (detection limit) and 4% of the total nitrogen into plant was released via N2O emissions.

From 2013 to 2015, DTU Environment has measured methane and N<sub>2</sub>O emissions using the tracer dispersion method from seven wastewater treatment plants, all with anaerobic digesters. The investigations are still ongoing and the results are not yet reported. Measured methane emissions varied between 3 and 30 kg h<sup>-1</sup> [18].

In 2013/14, DTU Environment measured methane emissions from three plants treating organic waste (household waste, garden waste and industrial waste) and producing biogas. Two of the plants have combined aerobic / anaerobic treatment, while one plant only performs anaerobic treatment. All three plants showed significant emissions of methane (20 to 40 kg h<sup>-1</sup>) [18].



#### 3.2.3 France

French regulation defines some requirements on methane monitoring: methane concentration in biogas and emissions from combustion plants (engine, turbines, boilers) should be monitored (CH<sub>4</sub>, CO<sub>2</sub>, NO<sub>x</sub>, SO<sub>x</sub>).



Studies have been conducted on anaerobic digestion in France for 2 years in order to identify:

- plant specific feedback on:
  - incidents / dysfunctions or OTNOC situations that could lead to methane emissions, and
  - measures that were taken to limit emissions, and to prevent future emissions (continuous improvement).
  - specific hotspot methane emissions, installations or activities where methane could be emitted directly to atmosphere.

The information was collected through a bibliographic review and situations applicable to French plants. In that way, a typology of anaerobic digestion plants was drawn up (for environmental and accidental risk analysis).

Several programs to quantify emissions (including methane) are in progress or have started during 2015. The objective is to identify and quantify biogas leakages at several stages [19].

Performed measurements:

#### - Stack emissions

Measurement campaigns were done in three farm installations on emissions from engines. Results showed that methane emission from engines are in the range of 1,7 - 3,2 % of methane production [20].

#### - Diffuse emissions

Some operators have voluntarily done methane emission detection by IR cameras to identify diffuse and fugitive sources. These measurements do not provide quantification but help operators to identify spot emissions and measures to be taken to limit emissions or identify equipment that needs to be monitored or repaired.

#### 3.2.4 Germany

Due to the "Renewable Energies Act" the number of biogas plants in Germany has grown to about 8,000 (agricultural biogas plants with CHP or biogas upgrading and bio-waste treatment plants) [21]. Consequently the avoidance of GHG emissions from biogas plants has been of paramount importance for a few years. Concerning the investigation of GHG emissions, please note that the biogas technology employed at agricultural biogas plants and bio-waste treatment plants differs significantly.

#### Agricultural biogas plants:

IR cameras and hand held methane lasers are used allowing remote sensing of leakages from plant components which are difficult to access. There are results showing the occurrence of leakages in gas-conducting plant components on biogas plants. One project investigated ten biogas plants and showed that biogas losses from leakages are relevant [22]. Eight plants had an overall number of 22 leakages and seven of them were evaluated as serious leakages (four occurrences of untight wires to adjust



agitators; bull eye (observation window), pipe penetration, leakage in double membrane roof).

Another project investigated a large number of biogas plants and evaluated the data concerning the frequency of occurrence of certain leakages [23]. Results from this project are shown in Figure 6.



Figure 6. Number and location of detected leakages (modified from [23]).

Typical leakages are the foil roofs (single and double membrane) and the wires to adjust the agitators, but also different leakages in gas-conducting plant components (e. g. CHPs, compressors, gas pipes) can occur.

In yet another project, ten agricultural biogas plants in Germany were investigated [24]. From these ten plants seven were based on wet fermentation, two on dry continuous fermentation (no. 1 and 6) and one on dry batch fermentation (no. 5). The measurement program included two measurement periods (summer and winter) in each plant, and every plant component was investigated (substrate storage, feeding unit, digestion, digestate storage and gas utilization).

The methane emissions were determined by an on-site method. First a plant survey was carried out to identify the emission spots and in the second step the emissions were quantified. The silage storage, the feeding units (e. g. screw conveyor, mixing tank) and the digestion (e. g. leakages, methane diffusion in the supporting air of double membrane roofs) were investigated by means of an open chamber system. The methane emissions from the open digestate storages were determined by a closed chamber. Not gastight covered storages and substrate storage tanks were investigated by means of the air injection method. The emissions from the gas utilization were directly measured in the exhaust pipe. The measured methane emission factors are shown in Figure 7.





Figure 7. Methane emissions in g CH4 kWh<sub>el</sub><sup>-1</sup> from ten agricultural biogas plants in Germany. The emission sources are classified as gas utilization (CHP/upgrading unit), open/not gastight covered digestate storages and miscellaneous [24].

The main emission sources were open or not gastight digestate storage tanks and the gas utilization. While plants 1, 2 and 6 had gastight covered digestate storages and consequently did not show any detectable methane emission from this source, the remaining plants emitted methane amounting to 0.40 - 20.14 g CH<sub>4</sub> kWhe<sup>-1</sup> (0.22 - 11.22 % CH<sub>4</sub> loss) from open/not gastight covered digestate storages. However, the methane emissions from the open/not gastight covered digestate storages were based on two single measurements only. So the results do not represent an average emission factor over a whole year.

Furthermore each plant showed a detectable methane slip in the exhaust of the gas utilization. The emission factors from the CHPs varied from 1.09 to 5.89 g CH<sub>4</sub> kWhel<sup>-1</sup> (0.61 – 3.28 % CH<sub>4</sub> loss) for the single CHP units. These methane emissions were caused by an incomplete combustion in the engine. The methane slip from the upgrading units (no. 8 and 9) amount to 9.58 and 2.19 g CH<sub>4</sub> kWhel<sup>-1</sup> (5.34 and 1.23 % CH<sub>4</sub> loss) which were caused by a defective or missing post combustion.

Miscellaneous emissions sources (e. g. leakages or the processing of fermentation residues) contributed marginally to the overall methane emissions from the investigated plants. An exception is a leakage from plant 3 which was not considered in Figure 7. This leakage (a not properly closed service opening) caused an emission factor of 5 % CH<sub>4</sub>-loss.

In the last project covered in this section, three agricultural biogas plants with an upgrading step to biomethane were investigated [25]. Each plant was based on wet fermentation and used a chemical scrubber for the upgrading process. Plants 1 and 2 used energy crops and plant 3 used residual materials (distiller's wash). While the digestate storages of plant 1 and 3 were gastight, the storage tanks from plant 2 were not gastight covered.

The methane emissions were determined by both an on-site method (comparable procedure to [24]) and a remote sensing method by means of an open-path TDLAS



system and the software tool Windtrax. The measurements were carried out simultaneously to ensure the comparability of the results, which are shown in Table 4.

	On-side method				Remote sensing method	
	MP-Result in % CH₄		Uncerta	ainty	Average	Uncertainty
			in % CH₄		in % CH₄	in % CH₄
BMA I – MP II	0.12		0.02		0.51	0.21
BMA I – MP III	0.13		0.02			
BMA II – MP I	0.12 <sup>1</sup>	0.67 <sup>2</sup>	0.03 <sup>1</sup>	0.30 <sup>2</sup>	2.0	0.8
BMA II – MP II	0.96 <sup>1</sup>	1.29 <sup>2</sup>	0.26 <sup>1</sup>	0.55 <sup>2</sup>		
BMA II – MP III	0.22 <sup>1</sup>	-	0.05 <sup>1</sup>	-		
BMA II – MP IV	0.72 <sup>1</sup>	1.33 <sup>2</sup>	0.22 <sup>1</sup>	0.66 <sup>2</sup>		
BMA II – MP V	0.09 <sup>1</sup>	-	0.02 <sup>1</sup>	-		
BMA III – MP I	0.14		0.04		0.22	0.12
BMA III – MP II	0.13		0.03			
BMA III – MP III	0.05		0.01			
BMA III – MP IV	0.05		0.01			
<sup>1</sup> Summation of only actually measured emission sources						

Table 4. Methane emissions in % CH4-loss from three biogas plants with an upgrading step to biomethane	in
Germany [25]	

<sup>2</sup> ... Extrapolation of measured emission factors on the overall system

BMA = biomethane plant; MP = Measurement Period

From the comparison of the two methods the following conclusions were drawn [25]:

- "Both methods were suitable for quantification of methane emissions of biogas plants."
- The on-site method is time consuming on large sites, but identifies individual sources. Only short periods can be measured at each emission spot. Recurrent measurements during the year may decouple the results from environmental conditions.
- "The remote sensing method provides meaningful values over the total emission rate of the plants and their temporal variability. The method is highly dependent on wind conditions and the topological and infrastructural conditions." Furthermore the measurement does not affect the plant operation.
- "At all three plants, the emission rate measured by the remote sensing method was higher than from the on-site method. A comparison of emission rates of two plants, one of them determined by the on-site method and the other by remote sensing, would not be justified due to the systematically lower estimate by the on-site method."
- "Malfunctions can have a significant impact on the total emission level of a biogas or biomethane plant. The continuous measurement of emissions from overpressure/underpressure valves is a promising way to represent the


relationship between the operating conditions of biogas plants and the resulting emissions."

### **Bio-waste treatment plants:**

In one project twelve German bio-waste treatment plants were investigated [26]. From these twelve plants four were based on wet fermentation, five on dry continuous fermentation and three on dry batch fermentation. The plants were chosen as a representative selection to the German plant inventory. The publication presents the results of the GHG emission measurements and the resulting carbon footprints from the plants.

It must be pointed out that bio-waste treatment plants are designed differently compared to agricultural biogas plants. Many plant components (e. g. substrate delivery and pretreatment, processing of fermentation residues, digestate storages, composting tunnel, batch digesters) can be encapsulated and exhausted. Then the exhaust is conducted to a biofilter and is treated there. Consequently each emission source which was conducted to the biofilter is part of this overall emission source. So the single emission sources (e.g. post composting process) cannot be compared between the single plants.

The plants are partly very different concerning their process technology and this has to be considered if the methane emissions should be interpreted in the right way. The results show that the emissions are dominated by the manner of plant operation. Figure 8 shows the component-specific emissions (as carbon dioxide equivalents) from each plant. Except for plant 6 (high ammonia emissions caused by non-operating acid scrubber), methane is the most important GHG.

Insufficient aeration or unaerated post-composting processes led to increased GHG emissions (especially methane, cp. Figure 8 plants no. 1, 2 and 12). In some plants the GHG emissions from the post-composting are included in "Emissions after biofilter" (cp. Figure 8, e.g. biogas plant 10), because the post-composting process was encapsulated and attached to the biofilter. Furthermore the results show that the open storage of fermentation residues can cause considerable GHG emissions and should be avoided.





Figure 8. GHG emissions in kg CO2-eq Mg<sup>-1</sup> bio-waste from twelve bio-waste treatment plants in Germany (modified from [26]).

Besides diffuse and stationary point sources and area sources, operational and timevariant methane emission sources have to be considered as well. In one study pressure relief vents from two agricultural biogas plants were investigated over long time periods (60 – 100 days) by means of a vane anemometer and temperature sensor permanently installed in the exhaust duct of the pressure relief vent. The methane losses measured were 0.06 CH<sub>4</sub>-loss (plant A, winter) and 3.88 % CH<sub>4</sub>-loss (plant B, summer). There were different influences which affect the methane emissions from pressure relief vents; the plant management during normal operation, OTNOC situations (mainly the outage of the primary gas utilization) and the atmospheric conditions. Additionally these influences are able to affect each other and create a compounded impact on the operational methane emissions from pressure relief vents [27].

## 3.2.5 Sweden

A pair of Swedish studies in 2003 and 2005 showed considerable methane emissions from biogas production plants and biogas upgrading plants. Therefore, in 2007, the Swedish Waste Management Association (Avfall Sverige) introduced a system called the "Voluntary Agreement" for biogas production plants and biogas upgrading plants with the aim to quantify and reduce methane emissions [3].

The purpose of the Voluntary Agreement is to control and minimize the emission of greenhouse gases. Carbon dioxide, methane and nitrous oxide are greenhouse gases that can occur in the biogas system. However, in the Voluntary Agreement system, only methane is included. Methane is the main component in biogas and by focusing on only methane, other factors can be included, such as economy and the safety aspect. The Voluntary Agreement consists of two main parts:

• Systematic leak detection including rectification of found leakages. This is mainly performed by the staff at each plant at least once a year.



• Emission measurements at discharge points to quantify the emissions and losses. This is performed by an external independent measurement consultant at least once every three years.

The system boundaries of the Voluntary Agreement specify the outer limits for the emission and only emissions within these boundaries are quantified. The following criteria are included [3]:

- Only the parts of the plant that the plant owner owns and can control are included.
- Only the parts that are associated with the production of biogas or cleaning/upgrading of the gas are included. Emissions that occur in association with the use of the gas or the digestate, and emissions that occur during transportation of substrate, digestate and gas, are not included in the system.

During the first three-year period of the Voluntary Agreement, 2007-2009, measurements and calculations were performed at 18 biogas producing plants and 20 upgrading plants. For the second period, 2010-2012, 21 biogas producing plants and 28 upgrading plants participated [28].

The average methane emission for the biogas producing plants during the first period was 1.6 % relative to the produced amount of raw gas. A moving average for the time period 2007-2012 was concluded to be 1.9 %. The emission levels seem to be quite constant, and it is more likely that the small increase in methane loss is due to the fact that additional sources of emissions were discovered at the plants during the second measuring period, 2010-2012. In addition, some of the plants that joined for the second period showed relatively high losses of methane [28].

The average methane emission for the biogas upgrading plants was 2.7 % relative to the produced amount of clean gas, for the first period, 2007-2009. The moving average for the time period 2007-2012 was 1.4 % which indicates that substantial improvements had been made at several of the participating plants. Also, newer plants that had joined the system for the second period have modern techniques with lower emission profiles (e.g. chemical scrubbers or post-treatment systems) [28].

The third time period 2013-2015 is ongoing, and the current discussion concerns the extension of the system to the many waste water treatment plants that have biogas production.

## 3.2.6 Switzerland

One project performed in Switzerland looked for leakages on gas-conducting plant components at twelve plants by means of an IR-camera (Esders GasCam) and a portable methane analyzer (Esders Goliath) [29]. The leakages were not encapsulated and quantified. The data validation was limited to qualitative analysis. In Table 5 the detected weak points are dedicated to a relative significance.

Table 5. Relative significance of the detected emission sources detected by the GasCam [29].

Emission source	Frequency	Reason	<b>Emission potential</b>
Connection of foil roof to tank wall	Very high	Constructional defect	Per leakage low → sum of leakages overall big
Pipe penetration	High	Constructional defect	Big



Emission source	Frequency	Reason	Emission potential
Stirrer adjustment	High	Missing maintenance	Low
Open digester overflow	High	Constructional defect	Medium
Bull eye	Medium	Constructional defect	Low
Stirrer	Medium	Missing maintenance	Medium
Membrane	Medium	Material fatigue	Very big
Pressure relief vent	Low	Constructional defect	Very big
Screw conveyor	Very low	Constructional defect	Big
Inspection opening	Very low	Constructional defect	Big

In the same project, methane emission measurements from open digestate storages were performed at three plants during four seasons. The measurements were performed with an open chamber system and complemented with batch tests in a laboratory (HBT – Hohenheimer biogas yield test) [29].

The measurements were not carried out directly in the storage. Instead the post digester, the open digestate storage and the solid digestate storage were sampled. The digestate samples were taken into the open chamber system ( $0.24 \text{ m}^3$  volume) and each measurement lasted 5 days. The flow velocity inside the chamber was adjusted to the wind velocity ( $0.1 - 2 \text{ m s}^{-1}$ ) over the open digestate storage. Simultaneously the samples were analyzed in the laboratory by the HBT. The results of the chamber measurements were extrapolated to calculate the methane loss. The results are presented in detail in Table 6, Table 7 and Table 8.

Post digester	Methane em	issions	Methane production	Methane emissions	Temperatur	re in °C
	Nm <sup>3</sup> Sample 60 d	Nm <sup>3</sup> d⁻¹ tank	Nm <sup>3</sup> d <sup>-1</sup>	% CH <sub>4</sub> -loss	Ambient	Digestate
			Biogas	plant 21		
Jun2011	0.212	24.7	925	2.7	12.5	21.0
Sep2011	0.10	1.1	925	0.1	12.8	17.1
Dec2011	0.031	3.6	925	0.4		
May2012	0.056	6.5	925	0.7	17.4	20.3
			Biogas	plant 13		
Jul2011	0.028	2.3	578	0.4	20.1	25.2
Oct2011	0.011	0.9	578	0.2	13.2	18.6
Jan2012	0.018	1.4	578	0.3	1.7	11.6
Mar2012	0.024	2.0	578	0.3	12.7	16.5
			Bioga	s plant 5		
Aug2011	0.058	3.2	784	0.4	20.4	21.6
Feb2012	0.000	0.0	784	0.0	0.0	0.9
Apr2012	0.026	1.5	784	0.2	12.1	12.5
Jun2012	0.075	4.2	784	0.5	15.8	19.3

Table 6. Methane emissions from digestate of the post digester [29].



Post digester	Methane	emissions	Methane production	Methane emissions	Temperature in °C
Average	0.046	4.3		0.5	

Table 7. Methane emissions from digestate of the liquid digestate storage [29].

Liquid storage	Methane emis	sions	Methane production	Methane emissions	Temperat	ure in °C
	Nm <sup>3</sup> Sample 60 d	Nm <sup>3</sup> d⁻¹ tank	$\mathrm{Nm}^{3}\mathrm{d}^{-1}$	% CH <sub>4</sub> -loss	Ambien t	Digestate
			Biogas p	ant 21		
Jun2011	0.124	13.7	925	1.5	14.5	18.6
Sep2011	0.080	8.8	925	1.0	13.7	20.1
Dec2011	0.011	1.2	925	0.1		
May2012	0.128	14.2	925	1.5	10.7	15.6
			Biogas p	ant 13		
Jul2011	0.033	2.5	578	0.4	18.6	22.8
Oct2011	0.014	1.1	578	0.2	10.1	18.4
Jan2012	0.047	3.6	578	0.6	2.1	9.1
Mar2012	0.031	2.3	578	0.4	11.5	17.7
			Biogas p	lant 5		
Aug2011	0.183	9.7	784	1.2	18.9	19.6
Feb2012	0.000	0.0	784	0.0	7.0	7.7
Apr2012	0.038	2.0	784	0.3	9.0	12.7
Jun2012	0.026	1.4	784	0.2	15.8	18.7
Average	0.060	5.1		0.6		

Table 8. Methane emissions from digestate of the solid digestate storage [29].

Solid storage	Methane emiss	sions	Methane production	Methane emissions	Temperat	ure in °C
	Nm <sup>3</sup> Sample 60 d	Nm <sup>3</sup> d <sup>-1</sup> tank	$Nm^3 d^{-1}$	% CH <sub>4</sub> -loss	Ambien t	Digestate
			Biogas pla	ant 21		
Jun2011	0.018	0.2	925	0.0	17.4	45.9
Sep2011	0.136	1.6	925	0.2	16.1	46.1
Dec2011	0.000	0.0	925	0.0		
May2012	0.000	0.0	925	0.0	14.7	35.1
			Biogas pla	ant 13		
Jul2011	0.026	0.2	578	0.0	20.0	51.0
Nov2011	0.000	0.0	578	0.0	2.7	25.8
Jan2012	0.000	0.0	578	0.0	2.2	22.3
Mar2012	0.000	0.0	578	0.0	15.2	39.5.
			Biogas p	lant 5		
Aug2011	0.200	1.1	784	0.1	23.4	55.0



Solid storage	Methane emissions		Methane production	Methane emissions	Temperat	ture in °C
Feb2012	0.000	0.0	784	0.0	3.2	5.9
Apr2012	0.000	0.0	784	0.0	10.4	25.7
Jun2012	0.021	0.1	784	0.0	21.3	42.0
Average	0.033	0.3		0.0		

The digestate from the post digester and the liquid digestate storage had comparable methane losses of about 0.5 - 0.6 % CH<sub>4</sub> and the methane loss from the solid digestate was lower than 0.1 % CH<sub>4</sub>. The residual methane potential from the post digester varied between 3.8 and 14.5 %, from the liquid digestate between 3.3 and 13.5 % and from the solid digestate between 0.4 and 3 %. The measured methane emissions from the open chamber system were 2 - 24 % for the post digester, 3 - 37 % for the liquid digestate and 0 - 20 % for the solid digestate. In conclusion the results showed that the separation did not abate methane emissions from not gastight digestate storages.

A specific project performed in Switzerland, but which is relevant to other countries as well, investigated the methane diffusion from single foil roofs [30]. The diffusion of the biogas storage foils was measured in a laboratory according to DIN 53380 (Testing of plastics - Determination of gas transmission rate - Part 2: Manometric method for testing of plastic films). The measurements were carried out with pure methane at a temperature of 41 °C, about 200 – 300 kPa absolute pressure and 0 % relative humidity. It must be pointed out that in reality biogas is water vapor saturated and consequently water condenses at the biogas storage foil. Furthermore biogas is a gas mixture of methane and carbon dioxide and the permeability coefficients of single gases in gas mixtures can possibly affect each other.

The single foil roofs from overall ten biogas plants and new foils from the manufacturers were investigated in a laboratory and their results are shown in Figure 9.



Figure 9. Methane diffusion in Ncm<sup>3</sup> m<sup>-2</sup> d<sup>-1</sup> bar<sup>-1</sup> from single foil roofs [30].



The methane diffusion from new and unused EPDM-membranes (thickness 1.50 mm) is at between 1 690 and 2 190 Ncm<sup>3</sup> m<sup>-2</sup> d<sup>-1</sup> bar<sup>-1</sup> in contrast to the manufacturer information of 400 or 785 Ncm<sup>3</sup> m<sup>-2</sup> d<sup>-1</sup> bar<sup>-1</sup>. A different temperature level between manufacturer information (23 °C) and the used temperature of 41 °C was also found. The methane diffusion from the investigated foil roofs was between 1 650 and 2 730 Ncm<sup>3</sup> m<sup>-2</sup> d<sup>-1</sup> bar<sup>-1</sup>. Their layer thicknesses were between 0.8 and 1.5 mm. The methane diffusion showed a dependence of the layer thickness. With increasing thickness the methane diffusion was reduced. In case of filled biogas storages, the EPDM-foil expanded by up to 40 %. This could increase the methane diffusion calculation by approximately 24 %. Other influences, such as the used substrates in the biogas plant, or the age of the foils did not have verifiable influence on the methane diffusion.

## 3.2.7 Other countries

One project performed in Canada investigated a Canadian biogas plant over a whole year [31]. An average of 3.1 % CH<sub>4</sub>-loss during normal operation was measured using a remote sensing method (TDLAS and the software Windtrax). The results of the whole year are presented in Table 9.

		Autumn	Winter	Spring	Summer	Average
Methane prod	uction in $m^3 h^{-1}$	183	183	70	224	164
Methane emissions in	Normal operation	2.9	2.7	5.2	1.7	3.1
% CH <sub>4</sub>	Flaring/ Venting	20	25		13	19
	Main- tenance	0.5		1.8		1.2

Table 9. Average seasonal fugitive emission rates as a percentage of seasonal methane production [31].



# 4 Measurements at Tekniska verken in Linköping, Sweden

## As a part of this project, comparative measurements of methane emissions were performed at the biogas plant in Linköping, Sweden, owned by Tekniska verken.

Measurements were performed during the working week September 8<sup>th</sup>-12<sup>th</sup> in 2014 by six measurement teams, see Table 10.

Team	Approach	Analyzer	Meas. days	Report
AgroTech, DK	On-site	Photo-acoustic	Tuesday	Annex 1
DBFZ, DE (1)	On-site	GC-FID	Tue-Fri	Annex 2
DBFZ, DE (2)	Remote	TDLAS	Tue-Thu	Annex 3
DGC, DK	On-site	FID-Cutter	Tue-Wed	Annex 4
DTU, DK	Remote	CRDS	Tue-Wed	Annex 5
SP, SE	On-site	FID-Cutter	Mon-Tue	Annex 6

Table 10. Measurement teams and details on equipment and time for measurement.

The complete measurement reports produced by the different teams are available as annexes to this report. These reports are generally detailed regarding the measurement methods and the application of methods at the Linköping plant. Hence, reporting of the results in this report does not go into much detail but rather tries to summarize and compare the results obtained by the different teams. For a basic understanding of the results presented later in this report it is however necessary to highlight some information concerning the measurement methods and analytical equipment applied by the different teams:

- **AgroTech (on-site).** AgroTech used an IR camera (FLIR GF 320) for the leak detection (together with DGC) and used a high volume sampler system during the emission measurements. Methane concentration measurements are made by a photo-acoustic analyser.
- **DBFZ (on-site).** DBFZ used an IR camera (FLIR GF 320), a portable methane laser (GROWCON LaserMethane mini Gen2) and a portable biogas monitor (Geotech BM 2000) for the leak detection. For the emission measurements both direct measurements and open/closed chamber methods were used. Methane concentration measurements follow EN ISO 25139:2009 (GC-FID in laboratory).
- **DBFZ (remote).** DBFZ used a TDLAS (GasFinder2.0, Boreal Laser Inc.) and a weather station with 3D sonic anemometer (Model 81000, RM Young) to measure path averaged concentrations and meteorological conditions in the



downwind plume of the site. Inverse dispersion modelling was performed using the software Windtrax [32].

- DGC (on-site). DGC used an IR camera (FLIR GF 320) for the leak detection (together with AgroTech) and used a high volume sampler system during the emission measurements. Methane concentration measurements follow EN ISO 25140:2010 (FID with Cutter).
- **DTU (remote).** DTU measured downwind plume concentrations of methane and the tracer gas acetylene using a mobile analytical platform holding a fast and high sensitive cavity ring down spectrometer (CRDS) capable of detecting methane and acetylene concentrations down to ppb level every second, a GPS and a weather station.
- SP (on-site). SP used a portable leak detection instrument (Sewerin EX-TEC PM4) for the leak detection and followed the guidelines in the Swedish Voluntary Agreement system (see section 3.2.5) during the emission measurements. In short, only well-defined and systematic emission sources are covered by the quantification method, excluding leaks and other diffuse sources. Direct measurements of the gas flow and methane content are made. Methane concentration measurements follow EN ISO 25140:2010 (FID with Cutter).

## 4.1 PLANT DESCRIPTION

The Åby biogas plant is one of Sweden's largest biogas plants, owned and operated by Tekniska Verken i Linköping. It is situated in Linköping 200 km south-west of Stockholm and has been in operation since 1996. The plant has an annual permit of treating 100,000 tons of organic waste, and the annual production of raw biogas was about 17,000,000 Nm<sup>3</sup> in 2013. Besides biogas the plant produces 80,000 tons of wet bio fertilizer, which is sold to farmers. Table 11 shows the distribution of the substrates treated at the plant.

% weight)

## Table 11. Substrate input in 2013.

The plant production layout comprises receiving and pre-treatment facilities, four digesters, gas storage/upgrading and biofilter. The layout is shown in Figure 10.





3 Receiving/homogenisation tank

**4** Hygienisation tank

6 Gas storage 7 Gas upgrading system 8 Flare

10 Control room and office 11 Office 12 Bio filter

Figure 10. Layout of Åby biogas plant, Tekniska Verken i Linköping.

#### 4.2 LEAK DETECTION

Leak detection was performed on Monday afternoon by all the on-site measurement teams. AgroTech and DGC performed it together as one team. The three different teams performed leak detection separately without influencing each other. Some already known emission points such as digestate storage, biofilter release and gas upgrading CO2 release were not part of the leak detection campaign, the focus was on trying to identify other sources of emission such as equipment leaks or emissions from storage tanks. Results were compared at a meeting during Monday evening and then the teams decided which emission points should be measured during the following days.

Leakage	DBFZ	AgroTech/DGC	SP
Upgrading unit, meas. device inside the right compressor unit	Detected	Detected	Not detected, does not look for leaks inside ventilated buildings
Upgrading unit, safety valve pipe on roof	Detected	Detected	Not detected
Carbon filter house	Detected	Detected	Detected
Hygienization tank, manhole and pipe	Not detected	Detected	Detected
Hygienization tank, one-hour holding tank	Not detected	Detected	Not detected
Homogenization tank	Detected	Detected	Not detected

#### Table 12. Results from leak detection campaign performed on Monday.



Leakage	DBFZ	AgroTech/DGC	SP
Digester no 3	Not detected	Not detected	A few minor gas leaks were detected on the roof

A brief outline of the results from the leak detection campaign is presented in Table 12. Pictures of the identified leaks are shown in Figure 11 and their placement on the plant is shown in Figure 12.

<u>Leakage # 1</u>

#### <u>Leakage # 4</u>

Carbon filter house

Upgrading unit – ventilation duct 1 (facing north – away from the biogas plant)





## <u>Leakage # 2</u>

Upgrading unit – ventilation duct 2 (facing east towards digester 4)

<u>Leakage # 5</u>

Carbon filter house – gas booster F22



#### Leakage # 3

Leakage # 6

Upgrading unit – safety valve pipe leading above the container

Carbon filter house – gas booster F21





Figure 11 a). Overview of detected leakages, arrows indicate precise location of the leakages (taken from DGC report).

#### Leakage # 7

<u>Leakage # 10</u>

Carbon filter house – gas filter

Homogenisation tank swan neck



Leakage # 8

Hygienisation tank man hole at the top of the tank <u>Leakage # 11</u>

Homogenisation tank hole in the concrete roof



Leakage # 9

Hygienisation tank overloading valve

<u>Leakage # 12</u>

Hygienisation tank (one-hour holding tank) swan neck at the top of the tank





Figure 11 b). Overview of detected leakages, arrows indicate precise location of the leakages (taken from DGC report).





MEASUREMENTS OF METHANE EMISSIONS FROM BIOGAS PRODUCTION

Figure 12. Overview of the plant and the detected leakages + other emissions sources (taken from DBFZ on-site report).



Results from the leak detection campaign depend on the detection equipment used, the experience of the personnel performing the leak detection and variations in plant conditions.

SP did not find the leaking safety valve pipe on the roof of the upgrading unit, since this area was not investigated by them. SP uses a portable leak detection device which needs to be situated in the emission point to allow for detection, whereas the other two teams use IR cameras that are able to detect leaks from a distance. Leak detection is not performed by the measurement teams in the Swedish Voluntary Agreement system (see section 3.2.5) but the plant owner needs to have routines and equipment to perform it themselves. On the other hand SP detected some minor leaks on top of digester no. 3 that the other teams did not find due to the relatively large detection limit of the IR cameras.

DBFZ did not detect the leaks at the hygienization tank and SP did not detect the leaks at the homogenization tank. Neither of these contributes to any great deal to the total emission measured later on (see section 4.3).

AgroTech/DGC detected a leakage with the IR camera at a swan neck exit on top of a one-hour holding tank which is part of the hygienization process. They were however in doubt whether the leakage detected with the IR camera was methane or just emitted heat from the hot tank. It can be hard to differ between methane and heat emission with the IR camera. The emission was investigated later during the week and was not found to be emitting methane.

The conclusion of the results from the leak detection campaign is that an IR camera is a very good tool to be able to find leaks at a biogas plant in an effective way, but since it is hard to differ between methane and heat with the cameras, a portable leak detection instrument is a necessary complement. It must be pointed out that a portable leak detector should preferably use a methane specific measuring principle. Instrumentation which is based on e.g. semiconductors often has cross sensitivity to hydrogen sulfide. Certain sources, such as the hygienization or the homogenization tank are then possibly overestimated concerning methane concentration measurements. For example, at the flange of the hygienization tank SP measured about 10 vol-% (thermal conductivity sensor) and DBFZ measured only 0.1 vol-% CH4 (IR analyzer).



## 4.3 ON-SITE EMISSION MEASUREMENTS

With all the reports from the teams finally available in January 2015, a comparison spread-sheet was made and circulated to the teams. The spreadsheet was discussed thoroughly during a telephone meeting with all the teams, and the revised version of the spread-sheet following this meeting is the basis of the results and how they are presented in this chapter. Individual results here are given in the unit g/h, whereas the emission rates are always given in the unit kg/h in other parts of this report. Table 13 indicates at what times the respective teams performed measurements at the individual sources.

Emission source	AgroTech	DBFZ	DGC	SP
Homogenization tank	Tues. 16:10- 17:00	Tues. 14:45-16:00	Wed. 10:30-12:05	N/A
Hygienization tank	Tues. 15:35- 16:05	Thur. 08:00-13:00	Wed. 15:30-18:15	N/A
Biofilter	N/A	Tues. 11:45-13:00	N/A	Mon. 14:00-16:00
Digestate storage	N/A	Tues. 16:00-18:45	N/A	Tues. 13:00-16:45
Activated carbon filter building	Tues. 17:40- 18:40	Wed. 08:00-13:00	Tues. 16:00-19:15	Tues. 15:00
Chemical scrubber, CO <sub>2</sub> release	N/A	Wed. 15:00-16:00	N/A	Tues. 8:55-10:20
Chemical scrubber, compr. buildings	Tues. 10:55- 12:50	Wed. 17:00-19:00	Tues. 11:30-12:50	Tues. 16:30
Chemical scrubber, prs. relief vents	Tues. 11:20- 12:15	Thur. 14:00-15:00	Tues. 14:20-15:00	N/A
Analysis instruments on site	N/A	N/A	N/A	Tues.

Table 13. Measurement of	lays and times for the re	spective emission sources.
--------------------------	---------------------------	----------------------------

### 4.3.1 Homogenization and hygienization tanks

SP did not measure these sources. The estimated methane mass flows were considered too low based on methane concentrations given by their portable leak detection instrument and not detectable gas flow. The preliminary measurements did not motivate performing proper measurements, also given that the nature of these sources would give values with a very high uncertainty and/or involve safety risks. Furthermore SP does not have the equipment or methods available that are necessary to perform measurements from the leaking flange (man-hole) on the hygienization tank.

AgroTech argues that they would normally not include the homogenization tank in a measurement campaign since it is not part of the digestion process and also since their measurement method is not suitable for releases with low or even non-existing pressure difference between the emission source and the ambient air (see discussion



below). They did however perform the measurements for this project to compare their results with the other teams.

The results from the measurements are presented in Table 14.

DBFZ DGC SP Emission source (g CH<sub>4</sub>/h) AgroTech Homogenization tank (sum) 104 0.46 6 N/A - hole 86 0.45 З N/A - pipe 18 0.01 3 N/A Hygienization tank (sum) 88 16 96 N/A - flange 7 10 6 N/A - pipe 81 6 90 N/A Total percentage methane loss 0.02 % 0.002 % 0.01 % N/A

Table 14. Emission measurements in homogenization and hygienization tanks

The results for the homogenization tank differ with a factor of 100 between the lowest and highest values. The homogenization tank is characterized by open access (holes and vent pipe) from the surrounding atmosphere to the confined tank volume meaning the tank "breathes" with the impact of atmospheric pressure, wind, stirring, supply of waste etc.. Therefore the pressure difference between the surroundings and the tank is close to zero. The larger the pressure difference between a volume of confined biogas and the atmosphere, the more precise the measurement result by the high volume sampler method (AgroTech and DGC) will be. Using the high volume sampler method will, at a source like this, always be a trade-off between affecting the source as little as possible with the risk of not detecting the entire emission or using a too high sample flow causing an overestimation of the emission by sucking out gas from the tank.

At the time of DGC measuring, 10 September 10:30-12:05, there were relatively strong winds and there was some heavy turbulence at the sample location. In order to minimize the impact on the measurement DGC built a "tent" to cover the hole and pipe. The air/sample flow was chosen as low as possible to continuously securing a positive flow across the sample plane of the tent (no back flow with loss of methane emission caused by wind turbulence). The velocity was checked by a vane anemometer covering the entire cross sectional area (grid measurements). The result was a sample flow of approx. 500 m<sup>3</sup>/h simulating a velocity of 0,4-0,5 m/s across the leakages.

Figure 13 shows how the tank breathes. DGC measured emissions ranging from zero (back ground level) to approx. 70 ppm. The fact that emissions occasionally fall to zero during the entire sample period indicates, that the sample flow applied is not affecting the emission excessively.

As for the swan neck (pipe) the emissions never fall to background level but vary between 3 and 30 ppm, see Figure 14. One possible reason is that the sample flow was too large creating a vacuum resulting in sucking out gas from the tank. But this could also be caused by another explanation. The pipe can act as a chimney which creates a natural draft conveying a small flow of gas from the tank.

The emission rate from a source where practically no pressure difference exists between atmosphere and the confined biogas volume is very prone to impacts by the high volume sampler method. It is believed that AgroTech overestimated the emission due to the difficulties described and DGC thinks that there is a small probability that



even their result could be overestimated and the measured emission may have been lower if a lower sample flow had been applied. It is further believed that DBFZ underestimated the emission since they used a closed chamber for their measurement (Figure 15), and this would also have had a great impact on the way that this emission source behaved.



Figure 13. DGC results and sampling set-up at homogenization tank – hole.





Figure 14. DGC results and sampling set-up at homogenization tank – pipe.



Figure 15. DBFZ sampling set-up at homogenization tank – hole.

For the hygienization tank the reported values are more in comparison. Strong variations in methane concentration were observed by the teams during their respective measurements.

To finalize the discussion on these emission sources, we would like to strongly point to the fact that they contribute very little to the total emission of the plant, and one team (SP) did not even perform measurements due to this fact.



## 4.3.2 Biofilter

AgroTech and DGC did not measure in this source since they do not have the equipment available that is necessary.

The results from the measurements are presented in Table 15.

Table 15. Emission measurements in biofilter release.

Biofilter	AgroTech	DBFZ	DGC	SP
Emission rate (g CH <sub>4</sub> /h)	N/A	152	N/A	204
(methane concentration mg/Nm <sup>3</sup> )	N/A	126	N/A	57
(gas flow Nm <sup>3</sup> /h)	N/A	1 209	N/A	3 564
Total percentage methane loss	N/A	0.02 %	N/A	0.02 %

DBFZ placed a plastic foil on the surface of the biofilter and took gas samples from it to analyze the methane concentration (Figure 16). Gas flow was measured in the pipe leading to the biofilter.

SP measured both methane concentration and gas flow in the pipe leading to the biofilter. This is done according to the Swedish Voluntary Agreement system since it is there defined that there are no available results that show that biofilters would have any effect on the methane concentration. Biofilters will (at best) have an effect on odorous substances.



Figure 16. DBFZ sampling set-up at biofilter.

Despite the different approaches the mass flows reported by DBFZ and SP are in good comparison, but there is however a large difference in the underlying measurements of methane concentration and gas flow (Table 15). DBFZ performed their measurement on Tuesday when the measured gas flow was ca 4 m/s, but when they quickly checked the gas flow again on Friday it was then about 10 m/s. It is therefore believed that the gas flow to the biofilter varies quite a lot over time, but this variation does not seem to have much impact on the emitted mass flow of methane.



#### 4.3.3 Digestate storage

AgroTech and DGC did not measure in this source since they do not have the equipment available that is necessary.

The results from the measurements are presented in Table 16.

Table 16. Emission measurements in digestate storage.

Digestate storage	AgroTech	DBFZ	DGC	SP
Emission rate (g CH <sub>4</sub> /h)	N/A	4 382	N/A	7 335
Total percentage methane loss	N/A	0.51 %	N/A	0.85 %

Looking at the reported results there seems to be a large difference in measurement results, but looking at the individual measurements it is revealed that this difference is not due to a difference in the measurement results but rather due to assumptions and the way the results are treated.

DBFZ performed measurements with a closed chamber at four points on the surface of the digestate storage. They chose one of the points where the surface layer was considered to be rigid, whereas the other three chosen points had a cracked surface layer. Further the assumption was made that 50 % of the total surface has a cracked surface layer and 50 % has a rigid surface layer, and the measurement results were corrected relative to this assumption.

SP on the other hand did not purposely choose their four sampling points due to the appearance of the surface layer, but rather chose the points on random. SP reported the mean value of the individual results from the four sampling points. SP used an open chamber for their measurement.

Looking at the individual measurement results in Figure 17, the DBFZ point on the rigid surface layer appears as an outlier with a notably lower value than the other measurements. All the other seven measurements are in agreement.

Figure 18 shows the digestate storage tank and the appearance of its surface layer. The open chamber used by SP can also be seen in this figure (if the reader looks closely), which also gives an indication of the task necessary to perform sampling in many sampling points. Four sampling points – all near the wall of the tank for practical reasons – could probably not be regarded as representative of the whole surface layer.





Figure 17. DBFZ and SP individual measurement results at digestate storage (4 sampling points each).



Figure 18. Digestate storage tank and the appearance of the surface layer.



#### 4.3.4 Activated carbon filter building

The results from the measurements are presented in Table 17.

Table 17. Emission measurements in activated carbon filter buil	ding
---	------

Emission source (g CH <sub>4</sub> /h)	AgroTech	DBFZ	DGC	SP
Activated carbon filter	97	138	133	80
<ul> <li>leakage 1st compressor (F22)</li> </ul>	75	68	111	N/A
<ul> <li>leakage 2nd compressor (F21)</li> </ul>	17	63	56	N/A
- leakage flange	5	4	9	N/A
- calculated sum of individual leaks	97	135	176	N/A
Total percentage methane loss	0.01 %	0.02 %	0.02 %	0.01 %

In the activated carbon filter building, three different leakages were detected. AgroTech applied their method on the individual leaks whereas SP measured the total emission from the building in the ventilation release. DBFZ and DGC used both approaches with very good agreement. DGC overestimated the individual leaks due to contaminated background air in the building. DBFZ used fresh air from outside the building for their individual measurements, giving an almost perfect agreement between individual leakages measurement and total emission measurement in the ventilation.

It is believed that SP underestimated the gas flow in the ventilation since this was measured with a hotwire anemometer directly in the ventilation opening. DGC used the high volume sampler method also in the ventilation opening.

4.3.5 Chemical scrubber, CO<sub>2</sub> release

AgroTech and DGC did not measure in this source.

The results from the measurements are presented in Table 18.

Table 18. Emission measurements in chemical scrubber, CO <sub>2</sub> releas
--

CO <sub>2</sub> release	AgroTech	DBFZ	DGC	SP
Emission rate (g CH <sub>4</sub> /h)	N/A	N/A	N/A	85
(methane concentration mg/Nm <sup>3</sup> )	N/A	273	N/A	121
Total percentage methane loss	N/A	N/A	N/A	0.01 %

DBFZ performed measurements on Wednesday when the chemical scrubber was running on ca 50 % load and do not have the data available from the plant to be able to calculate the mass flow of methane. SP performed measurements on Tuesday when the chemical scrubber was running on full load.

The measurement point available for sampling is not suitable for flow measurement. To be able to calculate the emissions rate SP instead used the values of methane content and produced gas (Nm<sup>3</sup>/h) from the plant, which is standard practice. But since cooling air from the compressors is also released at this measurement point at the Linköping plant the calculation will not be correct in this case.

4.3.6 Chemical scrubber, compressor buildings

The results from the measurements are presented in Table 19.

Table 19. Emission measurements in	chemical scrubber,	compressor	buildings.
------------------------------------	--------------------	------------	------------

Emission source (g CH <sub>4</sub> /h)	AgroTech	DBFZ	DGC	SP
Right compressor building	84	198	285	363
(methane concentration mg/Nm <sup>3</sup> )	21	44	59	81
(gas flow Nm³/h)	5 100	4 476	4 944	4 475
Left compressor building	105	66	159	98
(methane concentration mg/Nm <sup>3</sup> )	13	12	25	22
(gas flow Nm³/h)	6 330	5 751	6 740	4 535
Total percentage methane loss	0.02 %	0.03 %	0.05 %	0.05 %

These sources illustrate the influence of both the methane concentration measurement and the gas flow measurement to the end result.

All teams but SP use background values of methane concentration to compensate the source measurements. In this particular case this would be the methane concentration of the air that is led in to the respective compressor buildings, and this concentration is then raised by any leakages in equipment inside the building, before the air is emitted. For these particular sources DBFZ and DGC used background values of 2 ppm, whereas AgroTech used values of 34 ppm and 12 ppm respectively. It is believed that AgroTech's measurement of background values was contaminated by the high emission of the leaking safety valve on the roof of the building. Particularly for the left compressor building, this correction for the background value has a relatively large influence on the reported result.

Taking the above in consideration when looking at the methane concentration measurement results in Table 19, it shows a good relation between teams for the left compressor building but with some larger variations for the right compressor building. It is believed that there was some variation in the concentration over time, and it might very well also be due to the leaking safety valve nearby.

SP use fan data for the gas flow values and since the fans are identical in the two buildings they report the same value for both buildings (slight difference due to temperatures). The fan data is in good agreement with the flow measurements performed by the other three teams for the right compressor building, but all measurements are consistently higher for the left compressor building for which we have no good explanation.



## 4.3.7 Chemical scrubber, pressure relief vents

SP was not able to perform measurements on the leaking pressure relief vent, since the compressor was not running on the Wednesday when measurements were going to be performed. DBFZ performed measurements on the Thursday and at this time the plant personnel had fixed the leaking safety valve. Instead DBFZ detected another leaking relief vent (not covered by the emission source list) and performed measurements on it.

The results from the measurements are presented in Table 20.

Emission source (g CH <sub>4</sub> /h)	AgroTech	DBFZ	DGC	SP
Prs. relief vent, left building	6 010	N/A	9 159	N/A
<u>Prs. relief vent, between buildings</u>	N/A	317	N/A	N/A
Total percentage methane loss	0.70 %	0.04 %	1.1 %	N/A

Table 20. Emission measurements in chemical scrubber, pressure relief vents.

The only comparable results are between AgroTech and DGC for the pressure relief vent on the left building. The difference in reported numbers is quite large. DGC are quite confident regarding the result obtained because two measurements were performed at different sample flows (duplicate determination):

- Measurement #1: Sampling hose was mounted directly on the vent pipe and sufficient sample flow controlled by checking for back flow of leaking biogas.
- Measurement #2: Sampling was mounted on the hose and the hood was equipped with plastic skirts to prevent wind turbulence causing leaking biogas to escape. This method resulted in less pressure loss in the sampling system and consequently a larger sample flow and lower CH<sub>4</sub> concentration.

Measurement #2 is the one reported by DGC. Table 21 indicates that the CH<sub>4</sub> emission measured by the two different sample flows is almost identical.

Measurement	Sample flow (Nm3/h)	CH4 (ppm)	Methane flow (g/h)
#1 (13:15 – 14:08)	507	24 650	9 000
#2 (14:22 – 15:03)	745	17 067	9 159

Table 21. DGC results for the pressure relief vent on the left building.

#### 4.3.8 Analysis instruments on site

SP made notes of the sample flows passing through the analysis instruments that are permanently installed at the site, since this is normal practice in the Swedish Voluntary Agreement System. No other team did this. The sample flow through the one analyzer installed in the activated carbon filter building is however included in the DBFZ and DGC measurements from this source.

The results from the measurements are presented in Table 22.

Table 22. Emission measurements in analysis instruments.

Analysis instruments	AgroTech	DBFZ	DGC	SP
Emission rate (g CH <sub>4</sub> /h)	N/A	N/A	N/A	41
Total percentage methane loss	N/A	N/A	N/A	<0.01 %



## 4.3.9 Comparison of on-site measurements

Figure 19 shows a comparison of the results reported by all the on-site teams for the emission sources of low magnitude, excluding the digestate storage and pressure relief vents. The exclusion of these sources makes for a more interesting comparison in this part, but they are of course very important to the end result.



Figure 19. Comparison of reported values for emission sources of low magnitude. Emissions from digestate storage and pressure relief vents are not included.

Figure 19 shows how the results compare presently with the equipment and methods applied by the different teams, as reported by them. The results show quite a large variation, with a coefficient of variation of 23 % for the total sum. The reported sums for these chosen emissions sources vary between 478 - 871 g CH<sub>4</sub>/h corresponding to 0.06 - 0.10 % total methane losses.

Results from the on-site measurements are summarized in Table 23.

Emission source (g CH <sub>4</sub> /h)	AgroTech	DBFZ	DGC	SP
Sum of sources of low magnitude	478	570	679	871
Prs. relief vent, left building	6 010 <sup>Tu</sup>	N/A	9 159 <sup>Tu</sup>	N/A
Prs. relief vent, between buildings	N/A	317 <sup>Th</sup>	N/A	N/A
Digestate storage	N/A	4 382	N/A	7 335
Sum on-site methods	6 488	5 269	9 838	8 206
Total percentage methane loss	0.75 %	0.61 %	1.14 %	0.95 %

Table 23. Results from on-site emission measurements.

<sup>Tu</sup> Results from measurements on Tuesday

Th Results from measurements on Thursday



There are large and unknown uncertainties in each reported result, but the large difference in the total percentage methane loss is mainly due to time variation of the emissions sources + the fact that the emission sources of high magnitude were not measured at the same time by all teams, or they were even not measured at all by some teams.

For the comparison of the emission sources of low magnitude there is clearly a difference in reported results. The difference is due to two reasons:

- 1. Difference in reported values for individual sources
  - a. Due to differences in emission over time
  - b. Due to differences in analysis methods
- 2. Missing values for individual sources, due to some measurements not being performed by some teams

The second reason makes it hard to compare results from the different teams applying somewhat different methods. It makes for an interesting comparison of the results if the missing values for each team are added to the result by using the mean value of the results from the other teams, for every single emission source. For example, DBFZ and SP measured 152 and 204 g CH<sub>4</sub>/h respectively in the biofilter release. The mean value of those measurements (178 g CH<sub>4</sub>/h) is added to the results for AgroTech and DGC. Using this method we end up with the results as illustrated in Figure 20.

The comparison in Figure 20 does not reflect the current situation, but illustrates what we could possibly end up with in the future if all teams worked on their methods and added the equipment necessary. The coefficient of variation is only 14 % for this case.



Figure 20. Comparison of estimated/hypothetic values for emission sources of low magnitude.



### 4.4 REMOTE SENSING EMISSION MEASUREMENTS

The following section reports the methane emission rates obtained by application of two remote sensing methods; the backward Lagrangian stochastic (bLs) method and the tracer dispersion method. Please note that the methane emissions rates are reported in kg methane per hour.

#### 4.4.1 The backward Lagrangian stochastic method

Measurements were carried out from Monday September 8<sup>th</sup> to Friday September 12<sup>th</sup>, 2014. On Monday, Wednesday and Friday measurement of the background methane concentration were performed. From Tuesday to Thursday successful downwind measurements and simulations with adequate wind conditions were realized.

The concentration readings of the TDLAS are sensitive to ambient temperature and were calibrated accordingly. The spectrometer used has a regression slope of  $-9.2 \times 10^{-3}$  per Kelvin. The paths downwind of the plant were placed at a height of about 1.5 m in distances of 80 to 200 m from the emission area depending on surroundings and wind conditions. The possibility to scan more than one path subsequently was used. Each measurement day, the spectrometer was scanning two or even three paths in intervals of 180 s. The input data for the simulation was filtered for invalid data and condensed to mean values in intervals of a quarter of an hour. The software calculates emission rates for the input intervals. For each data set, 50 000 air parcels were simulated backward in time starting from the measurement path. Depending on the concentration readings and the meteorological conditions the emission rate of the source area was calculated. The output of Windtrax was filtered using the following criteria. The friction velocity had to be larger than 0.15 m s<sup>-1</sup>. The absolute value of the Obukhov length had to be larger than 10 m and the fraction of the emission source area that had to be covered by air parcels touching the ground had to be higher than 90%.

On Tuesday only five valid 15-minute intervals were carried out (11:00 - 11:30 and 13:00 - 13:45) with emission rates ranging from 4 to 10 kg CH<sub>4</sub>h<sup>-1</sup> with rising tendency. The concentration measurement took place on two paths in the North of the plant and both paths were needed to perform valid simulations.

The measurements on Wednesday were performed using two paths south of the plant. Depending on the wind direction, valid emission rate calculations using only one of the paths were possible for a period in the morning (10:15 – 13:15) leading to an average emission rate of  $13.9\pm2.0$  kg CH<sub>4</sub> h<sup>-1</sup>. Using both paths for the simulation, the average emission over the same time period was 2.1 kg CH<sub>4</sub> h<sup>-1</sup> higher than with only one path. This was the first indication that an unknown source might be close to the second path leading to an overestimation of the known source area of the biogas plant. Using the results of the simulations with the concentration readings from both paths over a longer period (10:15 – 15:00) gave an average emission rate of  $18.9\pm4.2$  kg CH<sub>4</sub> h<sup>-1</sup>. Again, a rising emission rate was observed over the day.

On Thursday three measurement paths were set up South and South-West of the biogas plant. Two paths were located to the Southwest. It turned out that the reading was lower on the path that was located closer to the biogas plant than on the path further away. This lead to the conclusion that emission might come from water canals between these two paths. Taking only the closer of these two paths and the path to the South into account led to an average emission rate of  $4.9\pm1.5$  kg CH<sub>4</sub> h<sup>-1</sup> for the facility during the measuring period from 10:30 to 16:15.



## 4.4.2 The tracer dispersion method

Measurements were performed from Tuesday to Friday. During the first two days, area and plant methane screenings were performed. Successful quantifications were done in the afternoon on Tuesday and on the Wednesday.

Off-site and on-site methane screenings indicated methane releases from digesters, biogas upgrading units, the open digestate storage tank and the food waste pretreatment area. Off-site screenings indicated several methane sources in the area such as a local landfill, a wastewater treatment plant, and a storage area for biosolids. The latter was located about 300 m west of the biogas facility and occasionally resulted in overlapping downwind methane plumes. These plumes were disregarded.

The average methane emission rate measured during the afternoons of Tuesday (15:30-16:00) and Wednesday (17:00 – 19:20) was  $23.6\pm1.8$  kg CH<sub>4</sub> h<sup>-1</sup> (including all plume traverses). The corresponding average plant emission factor (EFs) was  $2.9\pm0.2\%$  based on the amount of upgraded biogas generated. The methane emission from the area with the digester tanks were estimated to account for approximately  $65\pm6\%$  of the total emission from the plant. The remaining part of the methane emission came from the pre-treatment area and the open digestate storage tank.

On Wednesday, the average methane emission was higher (24.5 $\pm$ 1.7 kg CH<sub>4</sub> h<sup>-1</sup>, which corresponds to 34.4 $\pm$ 2.4 Nm<sup>3</sup> CH<sub>4</sub> h<sup>-1</sup>) than the average emission measured on Tuesday (17.9 $\pm$ 7.8 kg CH<sub>4</sub> h<sup>-1</sup>, which corresponds to 25.1 $\pm$ 10.9 Nm<sup>3</sup> CH<sub>4</sub> h<sup>-1</sup>). This might be explained by the additional use of water scrubber during the second day, which is a technology known to release more CH<sub>4</sub> in the atmosphere than a chemical scrubber.

### 4.4.3 Comparison of remote sensing results

Table 24 provides an overview of the results obtained by the two remote sensing methods. Daily average methane emission rates are expressed in kg CH<sub>4</sub> h<sup>-1</sup>, while emission factors (EF) are provided in percentages. Emission factors were calculated as the ratio between the methane emission rate and the average methane production of upgraded gas for the specific day. All numbers are given in average value ± standard deviation. Methane emission rates were obtained for Tuesday, Wednesday and Thursday by the open-path method and on Tuesday and Wednesday by the tracer dispersion method. The daily average methane emission rates obtained by the backward Lagrangian stochastic method varied between 4.9±1.5 and 13.9±2.0 kg CH<sub>4</sub> h<sup>-1</sup> with EFs between 0.6±0.2 and 1.7±0.2% whereas emission rates obtained by the tracer dispersion method varied between 17.9±3.1and 24.5±3.4 kg CH4 h<sup>-1</sup> with EFs between 2.2±0.4 and 3.0±0.4%. In general, the emission rates obtained by the tracer dispersion method were higher than rates obtained by the backward Lagrangian stochastic method. However, one should be cautious when comparing the results as both methods showed daily variations in the emissions and as the measurements were not conducted in parallel (not within the same time interval) due to different requirements in terms of specific wind directions and wind speeds.

Both methods showed higher  $CH_4$  emissions on Wednesday 10<sup>th</sup>, which could be explained by the use of the water scrubber for the biogas upgrading process, a technology which is known to release more  $CH_4$  in the atmosphere than the chemical upgrading process.



Emission Rates				Emission factors (EF)						
	$(kg CH_4 h^{-1})$				(%)					
	DTU	-TDM DBFZ-BLSM DTU-TDM		DBFZ-BLSM		-TDM	DBFZ-BLSM			
Measuring day										
	Tue	Wed	Tue	Wed	Thu	Tue	Wed	Tue	Wed	Thu
	9 <sup>th</sup>	$10^{th}$	9 <sup>th</sup>	$10^{th}$	$11^{th}$	9 <sup>th</sup>	$10^{\text{th}}$	9 <sup>th</sup>	$10^{\text{th}}$	$11^{th}$
Average	17.9	24.5	7.0	13.9	4.9	2.2	3.0	0.9	1.7	0.6
SD	3.1	3.4	3.0	2.0	1.5	0.4	0.4	0.4	0.2	0.2
Count	3	18	5	9	17	3	18	5	9	17

Table 24. Results overview about remote sensing methods; tracer dispersion method (TDM) and the Backward Lagrangian Stochastic methodl (BLSM).

**SD**: Standard Deviation, **Count**: number of transects/simulations (dimensionless value)

**EF:** Emission Factor is calculated as the ratio between the emission rate and the average methane production for the specific day after upgrading processes.

BLSM daily averages were calculated from simulation results using the following measurement paths: Tuesday – path 1 and 2, Wednesday – path 2, Thursday – path 2 and 3. See measurement report in the annex for details.

The emissions measured on Wednesday by the two methods are the data best suited for comparison as both methods quantified emissions this day and because several emission rates were obtained. However, the open-path was applied in the morning (10:00-15:00) whereas the tracer dispersion method was applied in the afternoon (17:00-19:00). Figure 21 shows nineteen successful open-path simulations and eighteen plume traverses along the day with wind from North-Northeast in the morning and from East in the late afternoon. Table 25 reports the corresponding average emission rates with standard deviation. Notice that since DBFZ's data are based on both paths, reported values are different from those shown in Table 24. The average methane emission rate was 18.9 $\pm$ 4.2 kg CH<sub>4</sub> h<sup>-1</sup> using the bLs method whereas an emission rate of 24.5 $\pm$ 3.4 kg CH<sub>4</sub> h<sup>-1</sup> was obtained using the tracer dispersion method. Considering the standard deviation of means the emission rates obtained by the two different methods compares rather well. The average methane emission rate obtained by the tracer dispersion method is a bit higher in comparison to the average rate obtained by the bLs method. However, the emission rates measured during the day show an increasing trend. Especially the emission rates measured during the morning shows a general increase from 10:00 to 15:00. Thus it cannot be ruled out that the small difference in average emission measured in the morning and in the afternoon is actually due to an increase in the emission during the day. The biogas generation (before upgrading) decreased during the day from about 1852 Nm<sup>3</sup> h<sup>-1</sup> at 10:00 to 1792 Nm<sup>3</sup> h<sup>-1</sup> at 19:30. In the same period the emission rate changed from about 19 to 42 Nm<sup>3</sup> h<sup>-1</sup>. We do however not know if the decrease in gas production was associated with venting from the reactors. In the same time the upgrading process slightly shifted from the chemical scrubber to the water scrubbers. While the amount of upgraded methane was on the same level all the time, the part upgrade by water scrubbers shifted from 42% to 55%. Assuming 2% methane loss for the water scrubbers and 0.01 % for the chemical scrubber [28], according to the on-site results this gives a rise of methane loss of 3 Nm<sup>3</sup> h<sup>-1</sup> during the period from 11:00 to 19:00. The shift from amine scrubber to water scrubber during Wednesday is illustrated in Figure 22.



Both methods are associated with uncertainties. The tracer dispersion method is sensitive to the simulation of the source - accurate placement of the tracer release bottle. In this study correct simulation of the source is not considered to be a problem as the area of the source is relatively small especially in comparison to the measuring distance. Also the good match of the tracer and the methane plume indicated a good simulation of the methane emission from the plant. If methane is emitted from elevated heights such as the top of the anaerobic digesters this could lead to an underestimation of the measured emission. However, an evaluation using a Gaussian plume model shows that this effect will be minor (<1% of the emission rate, assuming a Pasquill stability class B – slight insolation) due to the long distance between the plant and the plume transect. The tracer dispersion method is also sensitive to interference of other local methane areas especially if these are located between the methane source and the transect used for plume traversing. Methane emissions from an area with water canals west-southwest to the facility were observed when working with the TDLAS. The amount of methane emitted from a part of this area was simulated to about 1 kg h<sup>-1</sup> by adding an additional source area located between the two paths in the South-Southwest of the plant to the inverse dispersion model. Such a small emission would have a negligible effect on the emission rate measured using the tracer dispersion method. However, if other areas also emit methane and potentially at higher rates or located closer to the transect this would lead to an overestimation of the measured methane emission. No indications of a significant additional methane source close to the measuring transect was seen as this would result in additional sharp peaks in the methane plume profile.

The bLs modelling is sensitive to the determination of the atmospheric stability. Especially if the emission sources are not at ground level knowledge about the atmospheric mixing is important to gain reliable results. Results from periods when the atmospheric mixing was poor were filtered out. However, the filtering parameter was at the close to the lower limit on Thursday which indicates that there might have been a slight underestimation of the emission rate on that day. In addition, it is necessary to position the measurement paths far enough downwind from the source. A rule of thumb is that 5 to 10 times of the source heights are sufficient to neglect the height of the source in the dispersion model. With a distance of 80 to 200 meters this was ensured. Disturbing structures such as trees may influence the dispersion, too. The 3D sonic was always placed as good as possible to ensure the correct depiction of the turbulences.

The resolution of the TDLAS device is 1 ppm·m, which corresponds e.g. to 5 ppb average on a 200 m path. This adds a minor uncertainty to the results. Furthermore, it is essential to resolve the emission plume from the natural methane background and to determine this background as careful as possible. Changes of the background concentration during the downwind measurements can influence the result of the inverse dispersion model. During this campaign this could have been a large uncertainty for the bLs method. The background was measured before and after the downwind measurements, and when one of the paths was not touched by the emission plume. But due to several other emission sources in the close environment, such as the landfill and the waste water treatment plant, the background concentration might have varied during the campaign.

However, if all measurements are carried out carefully and averaged, the uncertainty of the model is estimated to be approximately 10 % [33]. The bigger challenge is to find





out if deviations in the results correlate with changes in the operational conditions at the plant.

Figure 21. Methane emission rates measured on Wednesday 10<sup>th</sup>. DBFZ data referred to simulations performed with concentration data of both measurement paths.



Figure 22. Upgraded biogas production during Wednesday 10<sup>th</sup>.

Table 25. Average emission rates measured on Wednesday, using the tracer dispersion method (TDM) and the Backward Stochastic Lagrangian Model (BSLM). BSLM data are based on values from both paths.

	Emission rates on Wednesday 10 <sup>th</sup>				
	$(\text{kg CH}_4 \text{ h}^{-1})$				
	DTU-TDM	DBFZ-BSLM			
Measuring time interval	17:00-19:30	10:00-15:00			
Average	24.5	18.9			
SD	3.4	4.2			
Counts	18	19			



## 4.5 COMPARISON OF ON-SITE AND REMOTE SENSING RESULTS

The results of the two remote sensing methods could only be compared on the Wednesday (see above), and during this day the water scrubber unit was running, which makes comparison with the on-site methods impossible for this day. In addition, the remote sensing results have shown that there are strong diurnal variations in the emission rates in addition to the operational changes from day to day.

Two individual comparisons can however be made between on-site and remote sensing methods. The DTU remote sensing (tracer dispersion method) reported values for Tuesday evening and the DBFZ remote sensing (the backward Lagrangian stochastic method) reported values for Thursday (its most reliable values) are taken for comparison. These results can be compared with the on-site methods since the water scrubber was not in operation during these periods. An important difference between the days is however that the pressure relief vent on the left compressor building was emitting methane on Tuesday, and instead the pressure relief vent between the compressor buildings was emitting methane on the Thursday.

For the comparison we use the following values from the on-site methods:

- Homogenization tank, 6 g CH<sub>4</sub>/h (DGC)
- Hygienization tank, 67 g CH<sub>4</sub>/h (mean value AgroTech, DBFZ, DGC)
- Biofilter, 178 g CH<sub>4</sub>/h (mean value DBFZ, SP)
- Activated carbon filter building, 136 g CH<sub>4</sub>/h (mean value DBFZ, DGC)
- Chemical scrubber, CO2 release, 85 g CH4/h (SP)
- Chemical scrubber, compressor buildings, 340 g CH<sub>4</sub>/h (mean value all teams)
- Analysis instruments, 41 g CH<sub>4</sub>/h (SP)
- Total 0.85 kg CH<sub>4</sub>/h

For the pressure relief vents we use the DGC reported value of 9.2 kg CH<sub>4</sub>/h for the left building and the DBFZ reported value of 0.32 kg CH<sub>4</sub>/h for the vent between the buildings.

The reasoning behind choosing these values can be found in the discussions in sections 4.3.1 to 4.3.8.

For the digestate storage tank it is hard to choose one value, comparisons are therefore made with both the DBFZ reported value 4.4 kg CH<sub>4</sub>/h and the mean value of all individual measurement points by DBFZ and SP which is 6.5 kg CH<sub>4</sub>/h.

Results from the comparisons are presented in Table 26 and Table 27.

Table 26. Comparison of on-site and remote sensing results, Tuesday.

Emission source	Value (kg CH₄/h)
Sum of sources of low magnitude	0.85
Chemical scrubber, pressure relief vent, left building	9.2
Digestate storage	4.4 / 6.5
Sum on-site methods	14.4 / 16.5
DTU remote sensing method (with std. deviation)	17.9 ± 3.1



Table 27. Comparison of on-site and remote sensing results, Thursday.

Emission source	Value (kg CH₄/h)
Sum of sources of low magnitude	0.85
Chemical scrubber, pressure relief vent, between buildings	0.32
Digestate storage	4.4 / 6.5
Sum on-site methods	5.6 / 7.6
DBFZ remote sensing method (with std. deviation)	4.9 ± 1.5

The comparisons in Table 26 and Table 27 indicate a fair agreement between on-site and remote sensing methods, given that there are high and unknown uncertainties in all measurements. The remote sensing methods are affected by other emission sources that are not included in the on-site measurements (landfill, waste water treatment plant etc..). On Thursday, the weather was mostly sunny with light wind only. Although the DBFZ results during periods with too pure wind conditions were filtered out, a slight underestimation of the emission rates on that day cannot be ruled out completely. For the on-site methods there is a large uncertainty in the emission from the digestate storage tank.



## 5 Conclusion

From the literature study it can be concluded that a number of studies of the methane emissions from biogas plants have been performed in different countries, using different methods and approaches. The large variation in methods makes it hard to draw general conclusions from the existing data. A rather large variation between typical plants in different countries makes the comparison even harder.

The conclusion of the comparative measurements in Linköping is that the general results from different methods and approaches are comparable. The studied plant is large in size and the overall emissions are comparably low. There are high and unknown uncertainties in all measurement results and they are due to both analytical uncertainties and time variation in emission sources.

## 5.1 CONTINUED WORK

A few lessons were learned from the comparative measurements in Linköping. When the time schedule for the measurements was set it was purposely set up to not have the measuring teams influencing or disturbing each other. Due to the large time variation in emission sources, this however made strict comparison of the results impossible. Variations in plant operation during the week occurred which made it even more hard to compare results. For a future measurement campaign it is strongly recommended to have as many measurements as possible performed in parallel, and measurements should be complemented with intercalibrations with gas bottles with known methane content.

A suggested next step would be the production of a handbook on methane emission measurements from biogas installations. This handbook should aid the user in choosing a suitable measurement method and approach depending on the purpose of the measurement task. It should list advantages and disadvantages of the respective methods and approaches. Further, it should guide the user in analyzing and understanding reported values using different methods and approaches. The handbook would serve as an important reference to future work on standardization of methane emission measurements from biogas installations.



## 6 Literature

- IPCC, "Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental," Cambridge University Press, Cambridge, UK, 2013.
- [2] 10 07 2014. [Online]. Available: http://www.f3centre.se/renewable-fuels/fact-sheets/eu-sustainability-criteria-biofuels.
- [3] A. Peterson och M. A. Holmgren, "The Swedish Voluntary system for control of methane emissions," IEA Bioenergy Task 37, 2012.
- [4] C. Scheutz, J. Samuelsson, A. Fredenslund och P. Kjeldsen, "Quantification of multiple methane emission sources at landfills using a double tracer technique," *Waste Management*, vol. 31 (5), pp. 1009-1017, 2011.
- [5] EC DG Environment, "STANDARDISATION MANDATE TO CEN, CENELEC AND ETSI UNDER DIRECTIVE 2010/75/EU FOR A EUROPEAN STANDARD METHOD TO DETERMINE FUGITIVE AND DIFFUSE EMISSIONS OF VOLATILE ORGANIC COMPOUNDS (VOC) FROM CERTAIN INDUSTRIAL SOURCES TO THE ATMOSPHERE," European Commission, Brussels, 2012.
- [6] D. K. Mikel och R. Merrill, "EPA Handbook: Optical Remote Sensing for Measurement and Monitoring of Emissions Flux," U.S. Environmental Protection Agency, Research Triangle Park, NC, 2011.
- [7] T. Flesch, J. Wilson, L. Harper, B. Crenna och R. Sharpe, "Deducing Ground-to-Air Emissions from Observed Trace Gas Concentrations: A Field Trial," J. Appl. Meteorol., vol. 43, p. 487, 2004.
- [8] Z. Gao, R. Desjardins och T. Flesch, "Assessment of the uncertainty of using an inverse-dispersion technique to measure methane emissions from animals in a barn and in a small pen," *Atmos. Environ.*, vol. 44, p. 3128, 2010.
- [9] J. Mønster, J. Samuelsson, P. Kjeldsen, C. Rella och C. Scheutz, "Quantifying methane emission from fugitive sources by combining tracer release and downwind measurements – a sensitivity analysis based on multiple field surveys," *Waste Management*, vol. 34 (8), pp. 1416-1428, 2014.
- [10]J. Mønster, J. Samuelsson, P. Kjeldsen och C. Scheutz, "Quantification of methane emissions from 15 Danish landfills using the mobile tracer dispersion method," *Waste Management*, vol. 35, pp. 177-186, 2015.
- [11]H. Yoshida, J. Mønster och C. Scheutz, "A plant integrated measurement of methane and nitrous gas from a municipal wastewater treatment plant," *Water Research*, vol. 61, pp. 108-118, 2014.
- [12]"VDI 3477 Biological waste gas purification Biofilters," Verein Deutscher Ingenieure e.V., Düsseldorf, 2004.
- [13]P. Rochette och S. McGinn, "Methods for Measuring Soil-Surface Gas Fluxes," i Soil-Water-Solute Process Characterization: An Integrated Approach, Boca Raton, USA,



CRC Press, 2005.

- [14]"VDI 3880 Olfactometry Static sampling," Verein Deutscher Ingenieure e.V., Düsseldorf, 2011.
- [15]T. Persson och D. Baxter, "IEA Bioenergy Task 37 Country Reports Summary 2014," IEA Bioenergy Task 37, 2015.
- [16]A. Wellinger, "EBA Biogas Report 2013," European Biogas Association, 2013.
- [17]M. Huber-Humer, M. Hrad, M. Piringer, L. Kamard, G. Bochmann, M. Tauber och R. Kirchmayr, "Klimagasmonitoring zur Optimierung der Energiebilanz und Verfahrenseffizienz bei Biogasanlagen," 2014.
- [18]C. Scheutz, Interviewee, [Intervju]. 2015.
- [19]K. Adam, Interviewee, Ineris. [Intervju]. 2015.
- [20]I. Zdanevitch, G. Masselot, S. Collet och O. Bour, "Étude de la composition du biogaz de méthanisation agricole et des émissions en sortie de moteur de valorisation," Ineris, Verneuil-en-Halatte, 2009.
- [21]M. Scheftelowitz, J. Daniel-Gromke, V. Denvsenko, K. Hillebrand, A. Krautz, V. Lenz, J. Liebetrau, K. Naumann, A. Ortwein, N. Rensberg, W. Stinner, M. Trommler, T. Barchmann, J. Witt, M. Zeymer, K. Schaubach, D. Büchner, D. Thrän, W. Peters, S. Schicketanz, C. Schultze, P. Deumelandt, F. Reinicke, H. Gröber, M. Beil och W. Beyrich, "Stromerzeugung aus Biomasse," Deutsches Biomasseforschungszentrum, Leipzig, 2014.
- [22]W. Schreier, "Untersuchung von Gasleckagen bei Biogasanlagen (Investigation of gas leakages from biogas plants)," IR\_BGA Sachsen B66711, 2011.
- [23]J. Clemens, "Erfahrungen bei der Untersuchung von Biogasanlagen auf Gasdichtheit (Diffuse emissions from biogas plants – practical experience)," Gwf Gas | Erdgas, vol. 3, pp. 128-130, 2014.
- [24]J. Liebetrau, T. Reinelt, J. Clemens, C. Hafermann, J. Friehe och P. Weiland, "Analysis of greenhouse gas emissions from 10 biogas plants within the agricultural sector," *Water Sci. Technol.*, vol. 67, pp. 1370-1379, 2013.
- [25]T. Westerkamp, T. Reinelt, K. Oehmichen, J. Ponitka och K. Naumann, "KlimaCH4 – Klimaeffekte von Biomethan (Climate effects of the biomethane economy)," DBFZ report no. 20, Leipzig, 2014.
- [26]J. Daniel-Gromke, J. Liebetrau, V. Denysenko och C. Krebs, "Digestion of bio-waste - GHG emissions and mitigation potential," *Energy, Sustainability and Society*, vol. 5 (3), pp. 1-12, 2015.
- [27]T. Reinelt, J. Liebetrau och M. Nelles, "Operational methane emissions from pressure relief vents on two agricultural biogas plants," i *Proceedings to the International Conference on Solid Waste*, Hong Kong, 2015.
- [28]M. A. Holmgren, "Sammanställning av mätningar inom frivilligt åtagande 2007-2012 (Compilation of measurement results within the voluntary agreement 2007-2012)," Avfall Sverige U2012:15, Malmö, 2012.


- [29]M. Sax, M. Schick, A. Soltermann-Pasca och L. Van Caenegem, "Methanverluste bei landwirtschaftlichen Biogasanlagen (Methane losses from agricultural biogas plants)," Bundesamt für Energie BFE, Bern, 2013.
- [30]E. Büeler, "CH4-Emissionen bei EPDM-Gasspeichern und deren wirtschaftlichen und ökologischen Folgen (CH4 emissions from EPDM biogas storages and their economic and ecologic consequences)," Bundesamt für Energie BFE, Bern, 2011.
- [31]T. Flesch, R. Desjardins och D. Worth, "Fugitive methane emissions from an agricultural biodigester," *Biomass and Bioenergy*, vol. 35 (9), pp. 3927-3935, 2011.
- [32]"Thunder Beach Scientific," 2011. [Online]. Available: www.thunderbeachscientific.com.
- [33]L. Harper, T. Flesch, K. Weaver och J. Wilson, "The Effect of Biofuel Production on Swine Farm Methane and Ammonia Emissions," J. Environ. Qual., vol. 39, pp. 1984-1992, 2010.



A. AgroTech Identification and quantification of methane emission from leaks at biogas plants





Identification and quantification of methane emission from leaks at biogas plants

Linköping



Report

AGROTECH October 2014



AgroTech A/S Institut for Jordbrugs- og FødevareInnovation Institute for Agri Technology and Food Innovation Agro Food Park 15 . DK-8200 Aarhus N Tel. +45 8743 8400 . Fax +45 8743 8410 www.agrotech.dk . info@agrotech.dk

## Identification and quantification of methane emission from leaks at biogas plants

Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.



AgroTech A/S Institut for Jordbrugs- og FødevareInnovation Institute for Agri Technology and Food Innovation Agro Food Park 15 DK-8200 Aarhus N Tel +45 8743 8400 Fax +45 8743 8410 www.agrotech.dk info@agrotech.dk

### **1. PREFACE**

This report and the measurements described herein have been conducted in the project "Methane Emission from Danish Biogas Plants" supported by the Danish ForskEL program administered by Energinet.dk.

The goals of the project are to:

- Develop a method for measurements of methane emission from biogas plants
- Measurements of methane emission from approximately 10 Danish biogas plants.

The partners in the projects are Danish Gas Technology Centre and Agrotech. For more information about the project please contact Kasper Stefanek, Agrotech: 30 91 12 55 or kps@agrotech.dk

### 2. MATERIAL

#### Leakage detection

For identification of methane leakages on the biogas plants a FLIR GF 320 optical gas imaging camera was used (figure 1). The camera is adjusted to the wavelength range between 3.2 and 3.4  $\mu$ m and due to the absorption of infrared radiation in this range by many hydrocarbons, some gasses including methane becomes visually with the camera.



Figure 1. Optical gas imaging camera (FLIR GF 320).

### Overall description of measurement system for quantification of methane emission

The emission of methane from leakages identified at biogas plants was measured by a dynamic chamber measuring system. This system consisted of the following components.

- Air sucking ventilator
- Air flow regulating system
- Air flow measurement system
- Flexible air hose
- Suction unit
- Gas concentration measuring system

The different components are described in detail in the following sections



#### Air sucking ventilator

The air sucking ventilator was an ATEX ventilator (M SX 200-3) having a max air capacity of 1.000 m<sup>3</sup> air h<sup>-1</sup>. The ventilator was attached to the end of an air tube system (id = 160 mm) attached an air flow regulator and an air flow measuring system.

#### Air flow regulating system

The air flow regulator was an air iris system allowing regulation of the air flow between 10 and 100 per cent of max airflow capacity (Lindab, DAU flow damper).

#### Air flow measuring system

The volume of air drawn by the air sampling system was quantified by measuring the pressure difference in high-pressure in upstream air and low-pressure in downstream air before and after an inserted orifice plate system at the air flow tube (Lindab, FM/FMDRU 315-250). The volumetric flow rate (Q) for this system is given by:

$$Q = C A_2 \sqrt{\frac{2(p_1 - p_2)}{\rho \left[1 - (A_2 / A_1)^2\right]}}$$

where

 $Q = Volumetric flow rate, | s^{-1}$ 

A1 = Area of pipe upstream from restriction

A2 = Flow area of pipe at restriction

- p1 = pressure upstream from restriction, Pa
- p2 = pressure at restriction, Pa
- $\rho$  = density of fluid
- C = correction factor for energy losses

For the specific type of orifice plate system, the above equation can be reduced to

$$Q = 73.3 \times \sqrt{p1 - p2}$$

#### Flexible air hose

The air sampled above the point of leakage was drawn by a flexible ATEX air hose (GeoFlex P2 PU id=160 mm) to the air sampler unit. During sampling the hose was connected to the air sampler unit and the air sucking unit.

#### Air sucking unit

The flexible air hose can be fitted with a variety of different designs and types of sucking units depending on the type, shape and area of identified leaks (figure 2). Often the sucking unit has to be built and designed at the site to fit the specific type of leak, and often the sucking unit has to be covered by air tight curtain system to encapsulate and control the airflow above the leaking area.



**Figure 2.** Different examples on design and designs of air suckling devices depending on type and area of methane leak. The air suckling devices has to be fitted to the shape and type of the leakage. The air suckling device shown left is involved with sampling of methane emission from single point leakage. The air suckling device shown to the right is designed for measurement of methane emission from a mixing pump cover situated above a digestion tank.

#### Gas concentration measuring system

The concentration of methane in background air and in air sucked from the point of leakage were continuously sampled and analysed by a photo-acoustic multigas analyser system (INNOVA, 1412, LumaSense Technology) coupled to a multipoint sampler (INNOVA 1312, LumaScense Technology). The measuring system allowed a semi-continuous measurement of methane concentration in inflowing and outflowing air to and from the air flow sampling system. This allowed that the emission of methane ( $E_{CH4}$ ) from the point of leakage could be calculated by:

$$E_{CH_4} = \frac{1}{t} \sum_{j=1}^{j=N} (V_j) (C_{ij} - C_{bj})$$

Where

 $E_{CH4}$  = Emission of methane from a leakage, mg CH<sub>4</sub> h<sup>-1</sup>

V = Air flow during the methane emission measuring system, m<sup>3</sup> air h<sup>-1</sup>

 $C_i$  = Methane concentration in air drawn out of the sucking unit, mg CH<sub>4</sub> m<sup>-3</sup> air

 $C_b$  = Methane concentration in background air, mg CH<sub>4</sub> m<sup>-3</sup> air

- J = Sample number
- t = Length of sampling period, h

N = Total number of samplings during the sampling period.

### 3. METHODS

#### Leakage detection

Methane leakages were identified with an optical gas imaging camera (FLIR GF 320). The biogas plant was scanned with the camera at a day with very low wind speed (08.09.2014).

#### **Measurement of methane emission**

The methane emission from the identified leakages was measured with the measurements system developed by Agrotech (described in "2. Materials" in this report).

Before sampling, an appropriate air sampling unit was chosen or designed to fit the specific type and area of leak.

The sampling system and the sampling unit were sampled by the flexible air tube, where after the air sucking ventilator and the gas concentration measuring unit were stared. The methane concentration was then measured in upwind (background) air. After that the sampling unit was placed above the leaking area. Care was taken that the sampling unit covered the area of leakage, and that methane was not sucked out of the leaking area by creating under-pressure in the sampling unit. This was ensured by use of the methane camera, and by regulation of the air flow rate of the sampling system.

The methane emission was measured in a period of time depending on the type of leakage and the observed variation in methane emission rate. When observing low emission rate and low variation in emission rate the measuring time was approximately set to 20 minutes per leakage. When observing higher emission rate and high variation in emission rate, the measuring time per leakage was set to between 30 and 90 minutes per leakage.

During sampling the air flow during the sampling system was quantified by measuring the pressure difference before and after the inserted orifice plate system.

Following air sampling above the leakage, the methane concentration in background air was again measured by sampling upwind air by the sampling system. When difference was observed between methane concentration measured before and after the sampling period, the background concentration was calculated as the average of the two.

By scanning of the biogas plant 12 leakages were identified (table 1 and pages 10-21).

The total methane emission from the identified leakages is 84,096 m<sup>3</sup> methane/year.

Table 1.	Identified	methane	leakages,	and	measured	methane	emission	from	each	indi-
vidual le	akage and	in total.								

Leakage	Description	Methane emission [m <sup>3</sup> year <sup>-1</sup> ]	Methane emis- sion [kg h <sup>-1</sup> ]
1	Upgrading unit – ventilation duct 1 (facing north)	1,089	0.084
2	Upgrading unit – ventilation duct 2 (facing east - towards digester 4)	1,363	0.105
3	Upgrading unit - Safety valve pipe leading above the container	77,882	6.010
4	Carbon filter container – Ventilation duct leading south of the container	No measurements performed <sup>a</sup>	No measurements performed <sup>a</sup>
5	Carbon filter container – booster pump 1 (left pump, viewed from the door	974	0.075
6	Carbon filter container – booster pump 2 (right pump, viewed from the door)	226	0.017
7	Carbon filter container –leak in gas filter	60	0.005
8	Hygienisation tank – leak in man hole cover at top	93	0.007
9	Hygienisation tank – Overloading valve	1,052	0.081
10	Homogenisation tank (swan neck pipe)	234 <sup>b</sup>	0.018 <sup>b</sup>
11	Homogenisation tank – hole in the concrete roof	1,117 <sup>b</sup>	0.086 <sup>b</sup>
12	Hygienisation tank (1 hour holding tank) – swan neck situated at the top of the tank situated most west	No measurements performed <sup>c</sup>	No measurements performed <sup>c</sup>
	Total	84,096	6.488

<sup>a</sup> The quantification was instead performed at the individual leaks identified inside the carbon filter container (leakage 5, 6 and 7)

<sup>b</sup> Very high variation observed during measurements. The measured values are considered uncertain and dubious

<sup>c</sup> A possible leakage was observed. However as the identification was considered to be caused by heat production and not methane emission, the methane emission was not quantified for this point.



Customer: Svensk Biogas Linköping AB Åby Västergård 58115 Linköping

### No video available

We were not able to detect methane from the ventilation duct with the optical gas detection camera, but detected the sources of the leaks inside the building. It was decided in the project not to measure on the sources, but on the ventilation duct. **Measurements performed by:** Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detection of leakages:** 08.09.2014





Customer: Svensk Biogas Linköping AB Åby Västergård 58115 Linköping

### No video available

We were not able to detect methane from the ventilation duct with the optical gas detection camera, but detected the sources of the leaks inside the building. It was decided in the project not to measure on the sources, but on the ventilation duct. Measurements performed by: Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detection of leakages:** 08.09.2014





Customer: Svensk Biogas Linköping AB Åby Västergård 58115 Linköping



Measurements performed by: Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detection of leakages:** 08.09.2014





Customer: Svensk Biogas Linköping AB Åby Västergård 58115 Linköping



Measurements performed by: Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detection of leakages:** 08.09.2014





**Customer:** Svensk Biogas Linköping AB Åby Västergård 58115 Linköping



gaard Hansen, Agrotech. **Detection of leakages:** 

08.09.2014





Customer: Svensk Biogas Linköping AB Åby Västergård 58115 Linköping

### No video available

Measurements performed by: Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detection of leakages:** 08.09.2014





**Customer:** Svensk Biogas Linköping AB Åby Västergård 58115 Linköping



Measurements performed by: Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detection of leakages:** 08.09.2014





Customer: Svensk Biogas Linköping AB Åby Västergård 58115 Linköping



Measurements performed by: Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detection of leakages:** 08.09.2014





Customer: Svensk Biogas Linköping AB Åby Västergård 58115 Linköping



Measurements performed by: Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detection of leakages:** 08.09.2014





**Customer:** Svensk Biogas Linköping AB Åby Västergård 58115 Linköping



Measurements performed by: Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detektion af lækager:** 08.09.2014

Emissionsmåling: 09.09.2014





Customer: Svensk Biogas Linköping AB Åby Västergård 58115 Linköping



Measurements performed by: Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detektion af lækager:** 08.09.2014

Emissionsmåling: 09.09.2014





Customer: Svensk Biogas Linköping AB Åby Västergård 58115 Linköping



Measurements performed by: Kasper Stefanek, Søren Gustav Rasmussen and Martin Nørregaard Hansen, Agrotech.

**Detektion af lækager:** 08.09.2014

Emissionsmåling: 09.09.2014



### B. DBFZ - on-site method Methane emissions from a biogas production site in Linköping





# Methane Emissions from a Biogas Production Site in Linköping

Report on-site method (Campaign from 08/09/2014 until 12/09/2014)

**Torsten Reinelt** 

DBFZ Deutsches Biomasseforschungszentrum gemeinnützige GmbH

Torgauer Straße 116 04347 Leipzig

Tel.: +49 (0)341 2434-112 Fax: +49 (0)341 2434-133

www.dbfz.de info@dbfz.de



Funding body / Principal	Swedish Gastechnology Center for Swedish Energy Agency	
Contact:	DBFZ Deutsches Biomasseforschungszentrum gemeinnützige GmbH Torgauer Straße 116 04347 Leipzig Tel.: +49 (0)341 2434-112 Fax: +49 (0)341 2434-133 E-Mail: info@dbfz.de Internet: www.dbfz.de DiplIng. (FH) Torsten Reinelt Tel.: +49 (0)341 2434-374 E-Mail: torsten.reinelt@dbfz.de	
Date:	21.01.2015	
Project No. DBFZ:	3270002	
Project No. Funding body:	GERG 1.73	
No. of pages	30	



### **Table of Contents**

Abbrevi	ations		IV		
Chemic	al symbols	s and formula	IV		
1	Introduction1				
2	Plant des	cription	1		
3	Method a	nd material	2		
3.1	Identifica	tion of leakages	. 2		
3.2	Quantific	ation measurements	. 6		
	3.2.1	Basic measurements	. 6		
	3.2.2	Ventilation pits from buildings	. 9		
	3.2.3	Digestate storage and homogenization tank	. 9		
	3.2.4	Leakages	10		
	3.2.5	Biofilter	12		
	3.2.6	Exhaust from the chemical scrubber	12		
4	Results		12		
4.1	Plant surv	vey	L3		
4.2	Emission	rates and emission factors	14		
	4.2.1	Ventilation pits from buildings	14		
	4.2.2	Digestate storage	15		
	4.2.3	Leakages	20		
	4.2.4	Biofilter	23		
	4.2.5	Chemical scrubber	24		
	4.2.6	Overall methane emission	24		
5	Literature				



### Abbreviations

Abbreviation	Explanation
DBFZ	Deutsches Biomasseforschungszentrum
FM	Fresh matter
TDLAS	Tunable Diode Laser Absorption Spectroscopy
TIC	Total inorganic carbonate buffer
TS	Total solids
VOA	Volatile organic acids
VS	Volatile solids
GC	Gas chromatograph
FID	Flame ionization detector
ECD	Electron capture detector
IR	Infrared

### Chemical symbols and formula

Symbol	Explanation
$\frac{\partial c}{\partial t}$	Linear slope of gas concentration
Ý	Volume flow
A	Surface area
CH <sub>4</sub>	Methane
Cin	Background concentration
Cout	Exhaust concentration
E	Emission mass flow
m <sup>3</sup> STP	m <sup>3</sup> at 0°C and 101.325 kPa
N <sub>2</sub> O	Nitrous oxide
NH4-N	Ammonia nitrogen



V	Volume
ρ	Density of the target gas
EF	Emission factor



### 1 Introduction

From 8<sup>th</sup> until 12<sup>th</sup> September 2014 methane emission measurements were performed by using different methods at a bio-waste treatment plant located in Linköping, Sweden. This preliminary report presents the available results of the onsite method up to now. The measurements were performed by Torsten Reinelt and Martin Apelt from DBFZ.

### 2 Plant description

The investigated biogas plant is a bio-waste treatment plant upgrading the produced biogas to biomethane. The provided biomethane is used as transportation fuel. The plant features are listed in the following.

#### **Biogas plant features**

General description:

Plant operator:	Swedish Biogas in Linköping AB		
Location:	Linköping, Sweden		
Construction year:	1996		
Substrate input in 2013:	Slaughterhouse waste cat. 3	20	% by weight
	Food industry	35	% by weight
	Alcohol	0.5	% by weight
	Thin stillage	1.5	% by weight
	Fat	2	% by weight
	Glycerol	1	% by weight
	Food waste	40	% by weight
Annual biogas production in 2013:	17 169 600 m³ STP raw biogas		
Annual average methane content in 2013:	64 % by volume		
Plant technology:			
Fermentation system:	Wet fermentation		
Fermentation stages:	Three parallel 1 <sup>st</sup> stage digesters digester	s and	one 2 <sup>nd</sup> stage
Digestate storage:	Open storage		
Biogas storage:	External tank		
Biogas upgrading technology:	Basically chemical scrubbing and demand	l wate	r scrubbing on
Miscellaneous:	Biofilter which treats the ducted pretreatment hall	exhau	st air from the
	Hygienization tanks (70 °C for one	e hour	)



#### Operating conditions during the measurement period

The plant has three known methane emissions sources, including the off-gas from the biogas upgrading as well as the diffuse emissions from the open digestate storage and the biofilter.

During the measurement campaign the methane content of raw biogas amounted to 62 % on Tuesday and Wednesday, and 60 % on Thursday. The methane content of the upgraded biogas was 96.4 % on Tuesday, 97.1 % on Wednesday and 96.6 % on Thursday.

The production rates are listed in Table 1 according to the values from the flowmeters of the biogas plant. The values on Tuesday the 9<sup>th</sup> led to a loss of methane in the upgrading process of 54 m<sup>3</sup> h<sup>-1</sup> STP during the upgrading process. This high value representing 4.7 % of the produced methane might have slipped through an open safety valve at the upgrading unit. Alternatively it might be due to the uncertainty of the flow meters. Nevertheless on the following days the deviation between the methane rate of the raw biogas and the methane rate of the upgraded biogas was less than 10 m<sup>3</sup> h<sup>-1</sup> STP.

On Wednesday the chemical scrubber was operated in turndown (50 % capacity). In addition, the water scrubber was in operation.

	Unit	Tue 09/09/2014	Wed 10/09/2014	Thu 11/09/2014
Type of upgrading		Chemical scrubber	Water scrubber, Chemical scrubber	Chemical scrubber
Raw biogas production		1 922	1 830	1 990
Upgraded biogas production	m <sup>3</sup> h <sup>-1</sup> STP	1 180	1 160	1 230
Methane production		1 138	1 126	1 188
Methane loss during upgrading		54	9	6

Table 1: Production rates during measurement period

### 3 Method and material

The on-site method is based on two working steps. First a survey on the biogas plant was carried out looking for spots with increased methane concentrations indicating leakages from biogas-bearing plant components. The next step was the quantification of the emission mass flow from the discovered leakages and from already known emissions sources. Depending on the source a measurement method was applied and gas samples were taken and analyzed in the lab.

### 3.1 Identification of leakages

The localization of leakages was investigated by three measurement techniques, an infrared (IR) camera, a portable methane laser and a portable biogas monitor. The single systems are listed and shortly described in the following.



The IR camera uses the specific characteristic from volatile organic compounds to absorb much infrared radiation at certain wave lengths. Methane has different absorption maximums in the infrared spectrum. The camera uses the wavelength range from 3.2 to 3.4 µm. The camera is shown in Figure 1.



Figure 1: Infrared camera during use on the exploratory biogas plant of the DBFZ

The incoming radiation is filtered to the wave length range specified above. If the radiation subsequently encounters the detector, a photon flux is induced. A gas cloud between background and lens changes the energy flux. The cloud has to have a different temperature compared to the background, but it is not crucial if the temperature of the cloud is higher or lower than the background temperature. The detector consists of a cooled Focal-Plane-Array that is a layout of light sensitive detector elements registering the photon flux based on the photoelectrical effect. By means of a special image overlay technique (HSM mode) the gas from a leakage is visualized as a visible gas cloud on the camera display. The visualized gas cloud can be documented as movie or picture.

Features of the IR camera:

Device:	Imaging IR camera
Producer:	FLIR
Туре:	GF 320
Measurement principle:	Passive infrared
Measurable gases:	Methane, Ethane, Propane, Butane, Ethylene and others
Temperature range:	-40 – 350 °C
Temperature uncertainty:	$\pm 1$ °C for temperature range 0 – 100 °C and $\pm 2$ % of the reading for the range > 100 °C
Leakage range:	Depending on temperature difference/distance to source; detection limit for methane: $6 I h^{-1}$ (3 m distance and no wind)
Last calibration:	Manufacturer's calibration
Explosion protection:	Not protected



Like the IR camera the portable methane laser is a remote sensing measurement technique, too. In contrast to the camera the laser is based on an active IR measurement principle. From the device an IR laser beam with a certain wave length (1 653 nm) is emitted and reflected back from a surface to the detector in the device. The intensity of the reflected laser light decreases exponentially with raised distance from laser source to reflection surface. Due to the installed laser diode the device is sensitive for methane. From the measured absorption the device calculates path integrated methane concentration stated in ppm m. The measured value has to be divided by the distance to the reflection surface to get path averaged concentration in ppm. This principle is schematically shown in Figure 2.



Figure 2: Use of a portable methane laser to detect leakages

Features of the methane laser:

Device:	Portable methane laser
Producer:	GROWCON
Туре:	LaserMethane® mini Gen2
Measurement principle:	TDLAS (Tunable Diode Laser Absorption Spectroscopy)
Measurement range:	1 – 50.000 ppm m (depending on distance/reflection surface)
Measurement uncertainty:	± 10 % (1000 ppm m and 2 m distance)
Last calibration:	Self-calibrating by integrated gas measuring cell
explosive protection:	II 2G Ex ib IIA T1

The determination of the methane concentration at the direct leakage spot was done by a portable biogas monitor shown in Figure 3.

#### Method and material





Figure 3: Available biogas monitors [DBFZ, Torsten Reinelt]

Features of the biogas monitor:

Device:	Portable biogas monitor
Producer:	Geotech GmbH
Туре:	BM 2000 (cp. left device in Figure 3)
Volume flow of integrated pump:	0.3 l min <sup>-1</sup>
Integrated air pressure sensor:	900 1 100 mbar (± 5 mbar)
Last calibration	2014
Explosion protection:	Ex II 2G EEx ibd IIA T1 Gb

The measurement uncertainties of the BM 2000 are shown in Table 2.

Table 2: Measurement range and uncertainties of the BM 2000

		CH4	CO <sub>2</sub>	02
		in Vol. %	in Vol. %	in Vol. %
Measurement range		0 - 100 (IR)	0 - 100 (IR)	0 – 25 (Electrochemical)
Uncertainty	0 – 5 Vol. %	± 0.5	± 0.5	± 1.0
	5 - 15 Vol. %	± 1.0	± 1.0	± 1.0
	> 15 Vol. %	± 3.0	± 3.0	± 1.0



### 3.2 Quantification measurements

### 3.2.1 Basic measurements

#### Measurement of the gas concentration:

The directly acting greenhouse gases methane and nitrous oxide were determined. The used methods are listed in Table 3.

Table 3: Analytical measurement methods

Gas	Sampling	Measurement method	Measurement device	Standard
Methane (CH <sub>4</sub> )	Evacuated vials - (less than 10 mbar absolute pressure)	Gas chromatograph with an auto sampler and flame ionization detector (FID) for CH <sub>4</sub> and electron capture detector (ECD) for N <sub>2</sub> O	Agilent 7890A GC System	VDI 2466 sheet 1 (Verein Deutscher Ingenieure, 2008)
Nitrous oxide (N2O)				VDI 2469 sheet 1 (Verein Deutscher Ingenieure, 2005)

#### Measurement of the volume flow:

The volume flow was determined by measuring the flow velocity by means of vane anemometers. The determined flow velocities were referred to the inner cross-sectional area of the pipes. Furthermore the temperature and the relative air humidity and the pressure were determined. The used measurement devices are listed in Table 4. The volume flow under normal conditions was determined by Equation 1.

$\dot{V}_{STP} = \frac{\left(p_{Air} - \left(\left(\frac{rH}{100}\right) \cdot \frac{rH}{100}\right)\right)}{101}$	Equation 1	
V <sub>STP</sub>	Volume flow at 0°C and 101.325 kPa	m <sup>3</sup> h <sup>-1</sup> STP
Ý	Volume flow	m <sup>3</sup> h <sup>-1</sup>
p <sub>Air</sub>	Air pressure	kPa
θ	Temperature	°C
rH	Relative air humidity	%



Table 4: Used measurement equipment

Device	Measurement range		Accuracy
PCE 007	0 - 45	m s <sup>-1</sup>	$\pm 3$ % from upper range value and 0.1 m s <sup>-1</sup>
	0 - 45	°C	±1 °C
Ahlborn FVAD 15 S120	0.6 - 20	m s <sup>-1</sup>	$\pm 1$ % from upper range value and
			±1.5 % from measurement value
Testo 416	0.6 – 40	m s <sup>-1</sup>	0.2 m s $^{\rm 1}$ and ±1.5 % from measurement
			value
Ahlborn pressure sensor FDA 612 SA	700 - 1 050	mbar	±0.5 % from upper range value
Ahlborn temperature sensor FPA 32 P	- 40 – 500	°C	± 0.3 K (at 0°C)
Extech SD 700	0 - 50	°C	± 0.8 °C
	700 - 1 100	hPa	± 2 hPa ( 10.0 - 1 000.0 hPa)
			± 3 hPa (1000.1 - 1 100.0 hPa)
	10 - 90	% RH	± 4 % RH (10 – 70 % RH)
			± 4 % from measurement value and
			± 1 % RH (70 – 90 % RH)

The volume flow of the used ATEX classified blowers was measured by the anemometers PCE 007 (PCE Deutschland GmbH, Meschede, Germany) and S120 (Ahlborn Mess- und Regelungstechnik GmbH, Holzkirchen, Germany). The PCE anemometer was calibrated by the Testo 416 anemometer in the laboratory and the measured volume flows were corrected with a linear regression. The calibration of the PCE anemometer is shown in Figure 4.

The measurement of the flow velocity from the ventilation pits was carried out with the PCE anemometer and from the inlet air to the biofilter with the Testo anemometer.




Figure 4: Linear calibration of PCE anemometer for measurement of the volume flow of the ATEX blower

#### Calculation of the emission mass flow and the emission factor:

By measuring the volume flow and the gas concentration of the target gas, the emission mass flow can be calculated according to Equation 2.

$$E = \frac{c \cdot \dot{V}_{STP}}{1\,000}$$

$$E \quad Emission mass flow \qquad g h^{-1}$$

$$c \quad Mass concentration \qquad mg m^{-3}$$

$$\dot{V}_{STP} \quad Volume flow under normal conditions \qquad m^3 h^{-1} STP$$

The emission factor that relates the emission mass flow from a source to the methane production of the biogas plant is determined according to Equation 3.

<b>FF -</b>	E		Equa	tion 3
CF -	$\rho_{CH4} \cdot \dot{V}_{methane} \cdot 1000$			
	EF	Methane emission factor	% CH <sub>4</sub> loss	
	E	Emission mass flow	g h⁻¹	
	$ ho_{CH4}$	Density of methane	kg m⁻ <sup>3</sup>	



$\dot{V}_{methane}$	Methane production of the plant	m <sup>3</sup> h <sup>-1</sup> STP
Ϋ́ <sub>STP</sub>	Volume flow under normal conditions	m <sup>3</sup> h <sup>-1</sup> STP

# 3.2.2 Ventilation pits from buildings

The ventilation pits of buildings were investigated directly at the exhaust duct. In case of the compressor buildings running ventilators were installed, venting the whole building. There the flow velocity was measured at twelve and the emitting gas concentration at four points of the exhaust duct, which are shown in Figure 5. The background concentration sampling was done in front of the air supply duct of the building.



Figure 5: Measurement points of flow velocity (left) and points of concentration measurements (right)

In case of the building which was connected to the activated carbon filter the ventilator was out of operation. Consequently the exhaust duct was encapsulated and ventilated with the ATEX classified blowers. In this case the measurement of the gas concentration and the volume flow were carried out like the determination of leakages (cp. section 3.2.4).

# **3.2.3** Digestate storage and homogenization tank

To determine the methane emissions from the open digestate storage and two emission sources from the homogenization tank, the closed chamber method (Rochette and Mc Ginn, 2005) was used.

The measuring principle is based on the measurement of an increasing gas concentration inside the chamber volume. After putting the closed chamber on the emission surface a gradually increasing gas concentration was determined. The chamber is shown in Figure 6.





Figure 6: Use of the closed chamber (left: on a hole of the homogenization tank; right: on the surface of the open digestate storage)

Samples were taken after certain time intervals (0, 3, 6, 9, 12, 15, 20, 25, 30 min). The emission rate was calculated from the slope of the gas concentration, the chamber volume and the encapsulated surface area according to Equation 4.

$E_{Spec} = \frac{\partial c}{\partial t} \cdot \frac{V}{A} \cdot 0.06$				Equation 4
	Espec	Surface specific emission mass flow	g m-2 h-1	
	∂c ∂t	Linear slope of gas concentration	mg m <sup>-3</sup> min <sup>-1</sup>	
	V	Volume inside the chamber	m <sup>3</sup>	
	А	Encapsulated surface area	m²	

In case of a pipe bend on the homogenization tank the volume of this pipe bend had to be subtracted from the volume of the chamber.

#### 3.2.4 Leakages

Identified leakages were quantified by means of an open chamber system. The encapsulation of gasbearing plant components was done by a flexible enclosure made of foil. The open chamber had an input and output pipe and two connected blowers which produced an air flow through the chamber. The methane from the leakage and the fresh air were mixed inside the chamber. Gas samples were taken in the in- and output stream of the chamber. Finally the emission mass flow of the leakage was calculated from the concentration difference and the flow rate of the blower according to Equation 5.

$\mathbf{F} = \frac{\dot{\mathbf{V}} \cdot \boldsymbol{\rho} \cdot (\mathbf{c}_{out} - \mathbf{c}_{in})}{\mathbf{V} \cdot \boldsymbol{\rho} \cdot (\mathbf{c}_{out} - \mathbf{c}_{in})}$				Equation 5
1 000				
	E	Emission mass flow	g h-1	
	Ý	Volume flow	m <sup>-3</sup> h <sup>-1</sup> STP, dry	
	ρ	Density of the target gas	kg m <sup>-3</sup>	
	Cout	Exhaust concentration	ppm (mg kg-1)	
	Cin	Background concentration	ppm (mg kg-1)	



The measurement method is schematically shown in Figure 7 and field-tested at a leakage on a compressor in Figure 8.



Figure 7: Schematic of the measurement setup to quantify diffuse leakages



Figure 8: Measurement setup at a leakage on the 1st compressor besides the active carbon filter



# 3.2.5 Biofilter

The gas sampling at open biofilters occurred in the exhaust after the polluted gas stream passed through the material of the biofilter. To grasp the exhaust stream, a foil was laid on the surface and fixed at the sides. The foil was arched by the air flow through the biofilter. A heated sample line was placed underneath the foil. The sample method is described by Cuhls and Liebetrau (2013) in a German biogas method collection. The volume flow was measured in the raw gas pipe to the biofilter.



Figure 9: Measurement setup at an open biofilter

# **3.2.6** Exhaust from the chemical scrubber

The measurement of the methane gas concentration was directly sampled from the exhaust pipe according to VDI 2466 Sheet 1 (VDI, 2008). The sample point did not allow volume flow measurements by a vane anemometer.

#### 4 Results

#### 4.1 Plant survey







Figure 10 shows the discovered and already known emission sources. The sources are marked in terms of color. Red marked leakages showed more than 10 vol. % methane at the direct emission spot (determined by biogas monitor). Green marked leakages showed less than 1 vol. % methane at the direct emission spot. The emission source with number 6 is grey marked, because a methane concentration wasn't measured, but the methane emission from the source was quantified by an open chamber. Yellow marked source have been already known.

# 4.2 Emission rates and emission factors

# 4.2.1 Ventilation pits from buildings

Inside the right compressor unit a leakage on a measurement device was located which is shown in Figure 11. The methane concentration was up to 10 vol. % at the direct leakage spot. Due to the complicated casing around the device, it was decided that the leakage wouldn't be encapsulated. The methane emission was measured at the ventilation pit of the building. In the left compressor building no leakages were identified. But a limiting factor was that the left building was only temporary amenable for leakage detection.



Figure 11: Leakage at measurement device flanged to biogas pipe

Consequently the methane emissions from the right compressor building were higher than from the left one. The measured values are listed in Table 5. However, methane emissions occurred evidently in both buildings.



		Unit	Left compressor unit	Right compressor unit	
<b>.</b> .	Height	m	0.58		
Dimensions exhaust duct	Length	m	0.785		
	Surface area	m²	0.4	.6	
Flow velocity		m s <sup>-1</sup>	3.9 ± 10 %	3.0 ± 15 %	
Volume flow		m <sup>3</sup> h <sup>-1</sup> stpd	5 751	4 476	
Mass concentration (without background)		mg m <sup>-3</sup>	12 ± 15 %	44 ± 8 %	
E		g CH4 h <sup>-1</sup>	66	198	
EF		% CH4 loss	0.008 0.024		

Table 5: Methane emissions from the ventilation of the compressor units (right compressor unit includes the leakage shown inFigure 11)

#### 4.2.2 Digestate storage

#### Determination by Closed Chamber method:

As described in chapter 3.2.3 the closed chamber method determines the slope of the gas concentration inside the chamber volume. This is exemplified in Figure 12. The determined slope is emphasized in bold font.







In some cases the measured slope was not linear, but exponential which is shown in Figure 13. If the coefficient of determination was below 0.95, the slope was corrected by sorting the outlier (cp. Figure 14).



Figure 13: Concentration gradient determined by a closed chamber (sample point 1)



Figure 14: Corrected concentration gradient (sample point 1)



		Surface layer		Cracked surface layer		Overall storage (50 % cracked and 50 % thick surface layer)	
		CH4	N <sub>2</sub> O	CH4	N20	CH4	N <sub>2</sub> O
n		1	1	3	3		
∂c ∂t	mg m <sup>-3</sup> min <sup>-1</sup>	52.70	1.15	220.46	2.30		
E <sub>spec</sub>	g m <sup>-2</sup> h <sup>-1</sup>	1.92	0.04	6.23	0.07	4.08	0.05
$\sigma_{\text{Espec}}$	g m <sup>-2</sup> h <sup>-1</sup>			0.75	0.05		
Е	g h-1					4 382	58
EF	% CH4-loss g N <sub>2</sub> O m <sup>-3</sup> <sub>CH4</sub>					0.53	5

#### Table 6: Measurement results from the open digestate storage

If the emitted mass flows of methane and nitrous oxide are converted into  $CO_2$ -eq by using the GWP's from Myhre et al. (2013), it results that 11 % of the overall  $CO_2$ -eq emissions of the digestate storage arise from nitrous oxide and 89 % from methane. In a digestate sample from the post digester was measured a high ammonium nitrogen content about 3 g l<sup>-1</sup> and a high pH about 8.18 (cp. Table 7). These digestate conditions and the surface layer promote the production and emission of nitrous oxide, because the denitrification is promoted by more oxygen input and the high nitrogen content.

#### Residual gas potential from the digestate:

For the modelling of the diffuse methane emissions from the open digestate storage the digestate temperature and the residual biogas potential from the digestate of the 2<sup>nd</sup> stage digester was determined experimentally. The results are listed in the following. Figure 15 shows altogether three temperatures (air temperature and digestate temperature in two depths). It is known that the temperature of the digestate has great influence on the residual methane production (O<sup>^</sup>ROURKE, 1968). The digestate characteristics from the post digester shown in Table 7 suggest that besides methane the emission of nitrous oxide is possible.







Figure 15: Digestate and air temperature during the investigation period (declared values are average ± standard deviation)

Parameter	Value	
Sample number	BK 14-1155	
TS	3.66	%ғм
VS	69.24	%тs
рН	8.18	
VOA	1.01	g  -1
VOA/TAC	0.14	gvoa gcaco3 <sup>-1</sup>
NH4-N	3.02	g  -1

Table 7: Digestate characteristics







Figure 16: Specific biogas production at 37 °C after 60 days (error bars indicate standard deviation, n = 3)

The residual biogas potential amounted to 262 m<sup>3</sup><sub>STP</sub> Mg<sub>VS</sub><sup>-1</sup>. The used method is described in VDI 4630 (VDI, 2006). By using Equation 6 to Equation 8, the overall methane emission potential ( $EF_{digestate at 37 \ ^\circ C}$ ) of the digestate was calculated. The results are shown in Table 8.

Mass loss fac	tor = $1 - \frac{\dot{V}_{raw \ biogas} \cdot \rho_{raw \ biogas}}{1000 \cdot \dot{m}_{substrate}}$	Equation 6
V <sub>raw biogas</sub> ρ <sub>raw biogas</sub> ṁ <sub>substrate</sub>	Raw biogas production in 2013 Density of raw biogas (at 64 % methane content $\rightarrow$ data of 2013) Substrate input	m <sup>-3</sup> d <sup>-1</sup> STP 1.17 kg m <sup>-3</sup> Mg d <sup>-1</sup>
ṁ <sub>digestate</sub> = № ṁ <sub>digestate</sub> ṁ <sub>substrate</sub>	Mass loss factor · ṁ <sub>substrate</sub> Digestate input to the storage Substrate input	Equation 7 Mg d <sup>.1</sup> Mg d <sup>.1</sup>
EF <sub>digestate</sub> at 3	$_{7^{\circ}C} = \frac{Y_{CH4, digestate} \cdot \dot{m}_{digestate}}{\dot{V}_{methane}} \cdot 100$	Equation 8
<sup>I</sup> CH4, digestate	Methane potential from digestate	m³ CH₄ Mg⁻¹ <sub>FM</sub>



$\dot{V}_{methane}$	Methane production from the biogas plant	m³ d⁻¹ STP
m <sub>digestate</sub>	Digestate input to the storage	Mg d <sup>-1</sup>

Table 8: Estimation of the residual methane potential (37 °C) in comparison to the methane production of the biogas plant

Input 2013	Raw bio meti producti	gas and hane on 2013	VS (diges- tate)	Methane potential from digestate (digested at 37 °C)		Mass loss factor	Digestate mass	EF <sub>digestate</sub> at 37°C
Mg d <sup>-1</sup>	m <sup>3</sup>	d-1	%	m <sup>3</sup> CH <sub>4</sub> Mg <sup>-1</sup> vs	m <sup>3</sup> CH <sub>4</sub> Mg <sup>-1</sup> <sub>FM</sub>		Mg d <sup>-1</sup>	% CH4
257.5	47 040	30 106	2.5	157.2	4.0	0.79	202.5	2.7

Compared with the results from the chamber measurements, the digestate storage emitted a methane amount about 20 % from the residual methane potential determined at 37 °C.

#### 4.2.3 Leakages

#### 4.2.3.1 Activated carbon filter

The building next to the activated carbon filter showed a total of three leakages. Every single leakage and the whole building were investigated separately. The results are listed in Table 9.

Table 9: Summary of methane emissions from the activated carbon filter

Component	Determination	Mass concentration without background	E	EF
		mg CH₄ m⁻³	g CH4 h <sup>-1</sup>	% CH <sub>4</sub> loss
Overall Container	measured	722 ± 5 %	138	0.017
Overall Container	calculated <sup>1</sup>		135	0.016
Leakage 1 (1 <sup>st</sup> compressor)	measured	600 ± 5 %	68	0.008
Leakage 2 (2 <sup>nd</sup> compressor)	measured	660 ± 3 %	63	0.008
Leakage 3 (flange)	measured	54 ± 8 %	4	< 0.001
Hose	calculated <sup>2</sup>		3	< 0.001

<sup>1</sup>... Sum of leakage 1, 2 and 3

<sup>2</sup>... Difference of overall container (measured) and overall container (calculated)

The results show a good match between the measurement and summation of single leakages and the measurement of the exhaust air from the whole building.



#### 4.2.3.2 Hygienization tank

The hygienization tank showed two leakage spots. The first one was a not gastight closed flange and the second one an open pipe connected between homogenization and hygienization tank. The results are shown in Table 10.

Table 10: Summary of methane emissions from the hygienization tank

Component of the hygienization tank	Mass concentration without background	E	EF	
	mg CH₄ m⁻³	g CH4 h <sup>-1</sup>	% CH <sub>4</sub> loss	
Flange	101 ± 5 %	10	0.001	
Pipe	58 ± 19 %	6	0.001	

During the investigation of the pipe a stirrer was active led to a short rise of the measured methane concentration. This shows the effect of stirring periods.



Figure 17: Concentration run during encapsulation of the pipe of the hygienization tank

#### 4.2.3.3 Pressure relief vents from the compressor units

Two activated pressure relief vents connected to the compressor units were detected (see Figure 18 and Figure 19). The vents weren't activated simultaneously. The DBFZ investigated the second one by



an open chamber and the first one was calculated to compare the result with the measurements from the other institutes. The results from these pressure relief vents are shown in Table 11.



Figure 18: First pressure relief vent (08 and 09/09/2014, 80 vol.  $\%~CH_4)$ 



Figure 19: Second pressure relief vent (11/09/2014, 20 vol. % CH<sub>4</sub>)

Table 11: Methane emissions from the pressure relief vent of the compressor unit

Pressure relief vent		Mass concentration without background	E	EF
		mg CH₄ m⁻³	g CH4 h <sup>-1</sup>	% CH4 loss
Small one (20 % CH <sub>4</sub> )	measured	4 972 ± 5 %	317	0.038
High one (80 % CH <sub>4</sub> )	calculated	19 888*	1 269	0.154

\* ... Mass concentration was calculated by multiplying the ratio of 80 vol. %/20 vol. % with the mass concentration of the measured one assuming that the volume flows are equal



#### 4.2.3.4 Homogenisation tank

The both leakages from the homogenisation tank were measured by a closed chamber like the digestate storage. The results are shown in Table 12. The emitted methane volumes were very low compared to the other leakages.

Table 12: Measurement results from the open digestate storage

		Hole		Pipe		Overall tank	
		CH4	N <sub>2</sub> O	CH4	N <sub>2</sub> O	CH4	N <sub>2</sub> O
∂c ∂t	mg m <sup>-3</sup> min <sup>-1</sup>	43.53	6.15	1.71	0.28		
E <sub>spec</sub>	g m <sup>-2</sup> h <sup>-1</sup>	1.59	0.22	0.05	0.01		
Е	g h-1	0.45	< 0.01	0.01	0.06	0.46	0.07
EF	% CH4-loss g N <sub>2</sub> O m <sup>-3</sup> cн4					0.6 • 10-4	0.6 · 10-4

# 4.2.4 Biofilter

The biofilter showed emissions of methane and nitrous oxide. The concentration runs from both gases are shown in Figure 20 and the results in Table 13.



Figure 20: Mass concentration of methane and nitrous oxide from the biofilter



Gas	Mass concentration	Volume flow (raw gas)	E	EF
	mg m <sup>-3</sup>	m <sup>-3</sup> stpd h <sup>-1</sup>	g h <sup>-1</sup>	% CH <sub>4</sub> loss/g N <sub>2</sub> O m <sup>-3</sup> <sub>CH4</sub>
CH4	126 ± 32 %	1 209	152	0.018
N <sub>2</sub> O	8 ± 19 %	1200	10	0.01

#### Table 13: Methane and nitrous oxide emissions from the biofilter

# 4.2.5 Chemical scrubber

Because the sample point of the off-gas pipe did not allow a volume flow measurement, the flow should be determined from operational data of the plant. According to the operator at that time of the emission measurement the chemical scrubber was operated in turndown (50 % capacity only). Furthermore it was determined that the concentration of carbon dioxide in the off-gas was less than 40 vol. %. A leakage in the sample line was excluded, because the concentration was checked directly at the sample point with the biogas monitor. Because of the unclear composition from the off-gas it is resigned to calculate an off-gas volume flow from operational data. The measured mass concentration of methane (half hour average) is 273 mg m<sup>-3</sup> ± 58 %.

# 4.2.6 Overall methane emission

The addition from each investigated emission source leads to an overall methane emission about 0.64 CH<sub>4</sub>-loss from the whole plant. The results are summarized in Table 14.

Component of the biogas plant	E	EF
	g CH <sub>4</sub> h <sup>-1</sup>	% CH4 loss
Activated carbon filter	138	0.017
Biofilter	152	0.018
Compressor units (including ventilation pits and pressure relief vent)	264	0.032
	317	0.038
Digestate storage	4 382	0.53
Homogenization tank	0.5	0.6 · 10-4
Hygienization tank	16	0.002
Off-gas chemical scrubber		
Overall methane emission	5 269.5	0.64

Table 14: Overall methane emissions from the whole biogas plant



#### 5 Literature

- Cuhls, C., Liebetrau, J., 2013. Emissionsmessungen an Anlagen zur biologischen Abfallbehandlung (Emission measurements at bio-waste treatment plants). In: Mesmethodensammlung Biogas – Methoden zur Bestimmung von analytischen und prozessbeschreibenden Parametern im Biogasbereich. Schriftenreihe des BMU-Förderprogramm "Energetische Biomassenutzung". [Jan Liebetrau, Diana Pfeiffer, Daniel Thrän (eds.)], Leipzig, pp. 82 – 89. Link: https://www.energetische-biomassenutzung.de/fileadmin/user\_upload/Downloads/Ver%C3% B6ffentlichungen/07\_Messmethodensamm\_Biogas\_web.pdf
- Myhre, G., D. Shindell, F.-M. Bréon, W. Collins, J. Fuglestvedt, J. Huang, D. Koch, J.-F. Lamarque, D. Lee, B. Mendoza, T. Nakajima, A. Robock, G. Stephens, T. Takemura and H. Zhang, 2013: Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
- O'ROURKE, J. T.: *Kinetics of Anaerobic Treatment of reduced Temperatures*. Stanford, Stanford University, Dissertation, 1968
- Rochette, P., Mc Ginn, S. M., 2005. Methods for Measuring Soil-Surface Gas Fluxes. In: Soil-Water-Solute Process Characterization: An Integrated Approach. [Álvarez-Benedí, J. and Munoz-Carpena, R. (eds.)] CRC Press, Boca Raton, USA.
- Verein Deutscher Ingenieure (VDI), 2005. VDI 2469 Messen gasförmiger Emissionen Messen von Distickstoffmonoxid – Manuelles gaschromatographisches Verfahren (Gaseous emission measurement – Measurement of nitrous oxide – Manual gas chromatography method). In VDI/DIN-Handbuch Reinhaltung der Luft, Vol. 5.
- Verein Deutscher Ingenieure (VDI), 2006. VDI 4630 Vergärung organischer Stoffe, Substratcharakterisierung, Probenahme, Stoffdatenerhebung, Gärversuche (Fermentation of organic materials, Characterisation of the substrate, sampling, collection of material data, fermentation tests). In: VDI-Handbuch Energietechnik, Landwirtschaft/Landtechnik.
- Verein Deutscher Ingenieure (VDI), 2008. VDI 2466 Messen gasförmiger Emissionen Messen von Methan – Manuelles gaschromatographisches Verfahren (Gaseous emission measurement – Measurement of methane – Manual gas chromatography method). In VDI/DIN-Handbuch Reinhaltung der Luft, Vol. 5.

# C. DBFZ - remote sensing method Methane emissions from a biogas production site in Linköping





# Methane Emissions from a Biogas Production Site in Linköping

# TDLAS Measurement Report (Campaign from 08/09/2014 until 12/09/2014)

**Tanja Westerkamp** 

DBFZ Deutsches Biomasseforschungszentrum gemeinnützige GmbH

Torgauer Straße 116 04347 Leipzig

Tel.: +49 (0)341 2434-112 Fax: +49 (0)341 2434-133

www.dbfz.de info@dbfz.de



Funding body / Principal	Swedish Gastechnology Center for Swedish Energy Agency
Contact:	DBFZ Deutsches Biomasseforschungszentrum gemeinnützige GmbH Torgauer Straße 116 04347 Leipzig Tel.: +49 (0)341 2434-112 Fax: +49 (0)341 2434-133 E-Mail: info@dbfz.de Internet: www.dbfz.de Dr. Tanja Westerkamp Tel.: +49 (0)341 2434-322 E-Mail: tanja.westerkamp@dbfz.de
Date:	05.11.2014
Project No. DBFZ:	3270002
Project No. Funding body:	GERG 1.73
No. of pages	13



# **Table of Contents**

Abbrevi	ations	.IV
1	Introduction	5
2	Site description	5
3	Method and material	7
4	Results	8
4.1	Measuring conditions and setup	. 8
12		40
4.2	Emission rates	12



# **Abbreviations**

Abbreviation	Explanation
CH <sub>4</sub>	Methane
DBFZ	Deutsches Biomasseforschungszentrum
TDLAS	Tunable Diode Laser Absorption Spectrometry
m <sup>3</sup> STP	m <sup>3</sup> at 0 °C and 101,3 kPa



#### 1 Introduction

From 8<sup>th</sup> until 12<sup>th</sup> September 2014 measurements were performed at the Linköping Biogas Plant to compare different methods to estimate methane emissions from biogas plants. This report presents the results of the indirect measurement with Tunable Diode Laser Absorption Spectrometry (TDLAS) and inverse dispersion modelling using the software Windtrax. The measurements were performed by Tanja Westerkamp and Carsten Tilch from DBFZ. The resulting emission rates are summarized in Table 4-3.

# 2 Site description

Important information about the biomethane production site in general, during the measurement period and about the surroundings is given here to be able to rank the results of the emission measurements.

#### **Plant parameters**

Table 2-1: Biogas plant parameters

Plant operator	Swedish Biogas in Linköping AB
Location	Linköping
Construction year	1996
Substrates	Food waste, slaughterhouse waste
Annual production 2013	16 206 000 m <sup>3</sup> STP raw biogas (flowmeter value)
Biogas treatment	Chemical scrubber, water scrubber

#### Operating conditions during the measurement period

The three relevant days of the measurement period for the calculation of emission factors are from Tuesday, 09/09/2014 until Thursday, 11/09/2014. These were the days where TDLAS data for inverse dispersion modeling was successfully generated. The raw biogas had a methane content of 62% on Tuesday and Wednesday, and 60% on Thursday. The methane content of the upgraded biogas was 96.4% on Tuesday, 97.1% on Wednesday and 96.6% on Thursday.

The production rates for the three days for raw and upgraded biogas are listed in Table 2-2 according to the values from the flowmeters of the biogas plant. Using the given methane content of the gases, the methane production rate is calculated for raw biogas and upgraded biogas, respectively. The values for Tuesday the 9<sup>th</sup> lead to a methane loss in the upgrading process of 54 m<sup>3</sup> h<sup>-1</sup> STP. This high value representing 4.7% of the produced methane might have slipped through a permanent open safety valve at the upgrading unit. Alternatively, it might be due to the uncertainty of the flowmeters. But the two other days the deviation between the methane rate of the raw biogas and the methane rate of the upgraded biogas is less than 10 m<sup>3</sup> h<sup>-1</sup> STP. It is of interest if the measurements can shed light on this issue.

On Wednesday the chemical scrubber was operated in turndown (50% capacity). In addition, the water scrubber was in use. Tuesday and Thursday the chemical scrubber was operated under full load.



Table 2-2: Production rates during measurement period according to the flowmeters at the plant (Lines with \* are calculated using the methane contents of the gases.)

	Unit	Tue 09/09/2014	Wed 10/09/2014	Thu 11/09/2014
Raw biogas production		1 922	1 830	1 990
Methane in raw biogas*		1 192	1 135	1 194
Upgraded biogas production		1 180	1 160	1 230
Methane in upgraded biogas*	m <sup>3</sup> h <sup>-1</sup> SIP	1 138	1 126	1 188
Calculated methane loss during upgrading according to flowmeters*		54	9	6

#### Surrounding area

In the north east a landfill is situated, while wastewater treatment is performed in the south-west of the plant. In the west of the plant were some ditches and ponds. All these sites might influence the results of the remote sensing measurements. North of the biogas plant, there were several short rotation coppices with heights of 4 to 5 meters making measurements during south wind periods a challenge. All these "obstacles" in the surrounding area are shown on the map in Figure 2-1.



Figure 2-1: Map of the surrounding area (based on www.openstreetmap.org)



#### 3 Method and material

The method whose results are presented here is optical remote sensing with a tunable diode laser absorption spectrometer (TDLAS) followed by an inverse dispersion simulation to determine the whole plant's methane emission rate. Remote sensing was carried out with a GasFinder 2.0 (Boreal Laser Inc.) and a weather station with 3D sonic anemometer (RM Young), shown in Figure 3-3-1. The spectrometer measures the path integrated concentration of methane between the device and a reflector on a path of a few hundred meters. The concentration readings are sensitive to ambient temperature and were calibrated accordingly. The spectrometer in use has a regression slope of -9.2x10<sup>-3</sup> per Kelvin. The paths downwind of the plant were placed at a height of about 1.5 m in distances of 80 to 200 m from the emission area depending on surroundings and wind conditions. Lengths were measured with a laser rangefinder with an accuracy of 1 m. Concentration and weather condition data were sampled with a frequency of 1 Hz. To determine the emission rate of the whole site the software WindTrax (Version 2.0.8.8) was used. The underlying backward Lagrangian stochastic (bLs) model is based on the Monin-Obukhov similarity theory. The input data for the simulation is filtered for invalid data and condensed to mean values in intervals of a quarter of an hour. The software calculates emission rates for the input intervals. For each data set 50 000 air parcels were simulated backward in time starting from the measurement path. Depending on the concentration readings and the meteorological conditions the emission rate of the source area is calculated. The model assumes that the source area is homogeneous and close to the ground. This simplification is possible when the measurement paths are placed behind the disturbances of the wind field in the downwind plume of the source. The output of Windtrax was filtered using the following criteria. The friction velocity had to be larger than 0.15 m s<sup>-1</sup>. The absolute value of the Obukhov length had to be larger than 10 m and the fraction of the emission source area that had to be covered by air parcels touching the ground had to be higher than 90%.



Figure 3-3-1: TDLAS back view (left) and in the downwind plume of the biogas plant together with the weather station (right). (Photos: DBFZ/Carsten Tilch)



#### 4.1 Measuring conditions and setup

Table 4-1 gives an overview on weather conditions and the setup of the measurement paths including some remarks on the daily observations. Measurements were performed on two or three paths each day. For this purpose the spectrometer was mounted on a pan-and-tilt unit scanning two or three reflectors subsequently in 2 to 3 min intervals.

Date	Wind conditions	Weather	Measurements	Comments
08/09/2014	SE changing to SW, 1-2 m s <sup>-1</sup>	rainy and misty/hazy, 15°C	background in the south on two paths	Path close to the solid residuals of the wastewater treatment plant had slightly higher concentrations
09/09/2014	S-SE, 2 m s <sup>-1</sup>	Cloudy/rainy, 13°C	two paths in the north with 320 m and 106 m	Measurement north of highway, slightly above background, few peaks in the morning, enhanced values from 11am on
10/09/2014	N-NE, 4-5 m s <sup>-1</sup>	several times from rainy to sunny, 12- 16°C	two paths in the south with 217 m and 134 m, background in the afternoon in the northeast	Signals well above background, ammonia emissions not detectable
11/09/2014	N-NE, 1-2 m s <sup>-1</sup>	sunny, 16°C	three paths in the south-southwest	Additional methane emissions from ponds and ditches in the southwest
12/09/2014	low wind speed, various directions	sunny, 16 to 20°C	one path in the south	No reliable simulation was possible.

Table 4-1: Measurement conditions and setup

Inverse dispersion modeling was carried out for three days of the measurement period. The results are shown in Table 4-3. The path setup for these three days and some remarks on the emission simulations are given in Table 4-2.



Table 4-2: Path setup and description. Measurement paths are marked by red dotted lines and the sonic anemometer by a red star. The source area is marked in green. (Based on www.openstreetmap.org)



#### 09/09/2014

The measurement took place on two paths in the north of the short rotation coppice at a large distance to the emission source. In addition wind was low from volatile directions. This is not a perfect situation for the inverse dispersion modelling. Looking at the result this should be considered.

#### 10/09/2014

Two paths were set up. Simulations were performed using data of both paths and using only data of path 2. Results were higher when using both paths. Regarding the results of 11/09/2014 only the simulation results of the latter simulations were used to calculate the emission factor for the day.

#### 11/09/2014

Three paths were set up. Concentration readings on path 1 were higher than on path 2 despite the larger distance to the source area. Simulations were carried out using data of path 3 in combination with data of path 1 or path 2, and using all paths' data. It is probable that ponds and ditches between path 1 and 2 caused the higher values on path 1 (see also Subsection 4.2). Only simulation results using data of path 2 and 3 were taken for the calculation of the emission factor of the biogas plant.



# 4.2 Discussion of uncertainties

Several methane emission sources in the surrounding area of the biogas plant influence the methane background concentration. Changing wind direction could show that the concentration readings on the measurement paths could be influenced by the residual storage site of the wastewater treatment plant. Unless the background concentrations for the simulations were carefully chosen there is an uncertainty because background and downwind concentrations could not be measured simultaneously. But the largest uncertainty during the measurements was caused by ditches and ponds in the south-west of the plant. Figure 4-1 shows upcoming gas bubbles of one of these ditches. As described in Table 4-2 there were higher methane concentration readings on the path downwind of this area than closer to the biogas plant. The reason could be that methane is emitted by the ditches.



Figure 4-1: Ditch in the south-west of the biogas plant close to the southern gate. The black spots indicate places where gas bubbled up (Photo: DBFZ/Carsten Tilch).

Another explanation for higher readings on the path further away from the biogas plant might be that emissions from high or hot emission spots situated at the biogas plant could descend further away from the plant. But the simulations using the data from 10/09/2014 indicate that this is probably not the case. Using both paths 19 valid simulation intervals could be calculated. Using only path 2, only nine intervals gave valid results due to unfavorable wind directions during the other intervals leading to a covering of the source area lower than 90%. Figure 4-2 shows the results of both simulations in comparison. It strongly suggests that the simulations with the data of both paths overestimate the emission rate of the biogas plant by about 2 kg h<sup>-1</sup> in average compared to the simulations using only path 2. For this day's average emission rate only those intervals were taken where the use of path 2 only was sufficient to calculate the emission rate with a covering of over 90% of the source area (cp. 2<sup>nd</sup> row of Table 4-2).



Figure 4-2: Results of the inverse dispersion modelling in intervals of 15min using one or two measurement paths. Error bars indicate the standard deviation within each model run.

The idea to analyze the influence of the bubbling ditches rose on the 11/09/2014 when methane concentrations were measured on three different paths. The wind was unstable from varying directions. It turned out that the measured concentrations on path 1 were higher than on path 2 which was closer to the biogas plant (cp. 3<sup>rd</sup> row of Table 4-2). Simulations were performed using different combinations of path concentration data. Apart from the simulation where the data of path 1 was used all calculations came to results of the same magnitude. Path 2 alone was sufficient for five simulation intervals in the morning when the wind came from northeast. Path 3 alone was sufficient for eight simulation intervals in the afternoon when the wind came from north-northwest. That the results of path 2 only and path 3 only are comparable gives a good indication that the simulation results are reliable.



Figure 4-3: 15min interval emission rates calculated with several combinations of path concentration data

In addition, a simulation was performed where the data of all paths was used and a second source area was added at the location of the bubbling ditches. For each path the program simulates 50 000 air parcels loaded with methane on their way back in time according to the present meteorological conditions. The air parcels measured on path 1 were loaded partly when touching the "biogas plant



emission area" and partly when touching the "ditch emission area". This led to similar results for the emission rate of the biogas plant as the simulations using paths 2 and/or 3.

The methane emission rate of the ditches was calculated to be about 1 kg h<sup>-1</sup> with 50% uncertainty.

The strong overestimation of the emissions of the biogas plant, when path 1 is used and a second emission area is absent, can be explained by the large distance to the plant compared to the additional "ditch source" which is located close to path 1. The overestimation was stronger the more the wind was coming from the east because the weight of the calculation shifts to path 1 in that case while path 3 is not seeing particles from the whole biogas plant area. Windtrax calculates a least squares solution when more measured paths than emission sources are present in the simulation. The rising question is whether the bubbling water comes from the biogas plant or from the wastewater treatment site or has different origin.

# 4.3 Emission rates

The calculated fugitive methane emission rates of the three days for which simulations were possible are given in Table 4-3. Normal conditions were assumed to be at a temperature of 0°C and a pressure of 101.3 kPa. The uncertainties are given as standard deviation of the results of the single 15 min intervals. For the 09/09/2014 the whole range of values is taken as uncertainty as there were only five valid simulation intervals. In addition, the measurement took place behind the short rotation coppice.

On the 10/09/2014 the chemical scrubber was operating at less than 50% load and the water scrubber was additionally operated. For that day the calculated emission rate is higher than on the two other days. This is in good agreement with the Swedish measurement program that indicates that water scrubbers have a larger methane slip than chemical scrubbers. Emission rates on the other two days were in the same range with overlapping uncertainties. The methane loss of 54 m<sup>3</sup> h<sup>-1</sup> STP according to the flowmeters on Tuesday the 9<sup>th</sup> is not reproduced by the remote method with TDLAS and inverse dispersion modelling.

For Tuesday and Thursday with only chemical scrubbing in operation the estimated emission factor is between 0.6 and 0.9 %. For Wednesday when half of the raw biogas was upgraded by water scrubbing the estimated emission factor is about 1.7 %.

Date	Produced methane (m <sup>3</sup> h <sup>-1</sup> STP)	No of inter- vals	Simulated emission rate (kg h <sup>-1</sup> )	Uncertainty (kg h⁻¹)	Simulated emission rate (m <sup>3</sup> h <sup>-1</sup> STP)	Uncertainty (m <sup>3</sup> h <sup>-1</sup> STP)	Simulated emission factor (% CH <sub>4</sub> )	Uncertainty (% CH <sub>4</sub> )
09/09	1 138	5	7.0	3.0	9.8	4.2	0.9	0.4
10/09	1 126	9	13.9	2.0	19.4	2.8	1.7	0.2
11/09	1 188	17	4.9	1.5	6.8	2.1	0.6	0.2

Table 4-3: Daily averages of the emission rates and emission factors (m<sup>3</sup> STP: m<sup>3</sup> at 0 °C and 101.3 kPa) determined by TDLAS and inverse dispersion technique



As the upgrading process was different on the 10/09/2014 when the water scrubber was in use it is not useful to create an overall average for the whole period.

# D. DGC Methane emissions from biogas plant





# Svensk Biogas i Linköping AB Methane emissions from biogas plant

Measurement report February 2015

# Svensk Biogas i Linköping AB

Methane emissions from biogas plant

Lars Jørgensen

Danish Gas Technology Centre Hørsholm 2015

Title	:	Svensk Biogas i Linköping AB
Report Category	:	Measurement Report
Author	:	Lars Jørgensen
Date of issue	:	16.02.2015
Copyright	:	Danish Gas Technology Centre
File Number	:	738-87; h:\738\87 metanemissioner for biogasanlæg\sgc projekt\målinger linköping\rapporter\dgc\measurement_report_final_rev1_16022015.docx
Project Name	:	SGC methane emissions
#### Table of Contents

P	a	q	e
	·,	3	-

1

1	Summary and results	3
2	Introduction and background	4
3	Plant description	5
4	Measuring method applied by DGC	7
5	Equipment and instruments	9
5.1	Optical IR camera	9
5.2	Volume flow	9
5.3	Methane concentration	9
6	Uncertainty	10
7	Performed measurements	11
7.1	Approach and purpose of the comparative measuring campaign	11
7.2	Detected methane mission sources	11
7.3	Measurement periods	
7.4	Operating conditions	13
8	Result of the methane emission quantification measurements	14
8.1	Overview	14
8.2	Upgrading unit – ventilation duct 1	
8.3	Upgrading unit – ventilation duct 2	
8.4	Upgrading unit – safety valve pipe leading above the container	16
8.5	Carbon filter house – Ventilation exit	16
8.6	Carbon filter house – gas booster F22	17
8.7	Carbon filter house – gas booster F21	17
8.8	Carbon filter house – gas filter	
8.9	Hygienisation tank – manhole at the top of the tank	
8.1	0 Hygienisation tank – overloading valve	
8.1	1 Homogenisation tank – swan neck	
8.1	2 Homogenisation tank – Hole in the concrete roof	
8.1	3 Hygienisation tank (one-hour holding tank) – swan neck at the top of the tank	21
9	Appendices	

Appendix 1 – Linköping Biogas Plant layout	23
Appendix 2 – Data from Linköping biogas plant 8-9 September	24
Appendix 3 – Data from Linköping biogas plant 10-11 September	25
Appendix 4 – Registration of ambient air conditions	26

#### 1 Summary and results

Danish Gas Technology Centre (DGC) performed measurements of methane emission from leaks and point sources at the biogas plant "Svensk Biogas i Linköping" from 8 September to 10 September 2014.

The measurements were carried out as part of the project *Climate impact* from biogas production, data collection and comparative study of measurement and calculation methods in Europe, see chapter 2. Steen D. Andersen and Lars Jørgensen, DGC, performed the measurements.

The main results are presented in Table 1.

Leakage ID	Sample location	Sample gas flow	Methane in sample gas	Background conc.	Methane emission <sup>2)</sup>	Methane emission <sup>2)</sup>
#		Nm <sup>3</sup> /h dry	ppm, dry	ppm, dry	Nm³/h dry	kg/h
1	Upgrading unit – ventilation duct 1 <sup>3)</sup>	6,740 <sup>1)</sup>	35	2.0	0.220	0.159
2	Upgrading unit – ventilation duct 2 <sup>4)</sup>	4,944 <sup>1)</sup>	82	2.0	0.396	0.285
3	Upgrading unit – safety valve pipe at roof	745	17,067	2.0	12.721	9.159
4	Carbon filter house – ventilation exit	604	315	9.0	0.185	0.133
5	Carbon filter house – gas booster F22	457	346	9.0	0.154	0.111
6	Carbon filter house – gas booster F21	456	181	9.0	0.078	0.0564
7	Carbon filter house – gas filter	452	37	9.0	0.013	0.0090
8	Hygienisation tank – manhole at tank top	522	19	2.5	0.0086	0.0062
9	Hygienisation tank – overloading valve	376	336	2.5	0.126	0.0904
10	Homogenisation tank – swan neck	458	11	2.5	0.0038	0.0027
11	Homogenisation tank – hole in the concrete roof	449	12	2.5	0.0045	0.0032
12	One-hour holding tank – swan neck at tank top	-	Not detected	-	Not detected	Not de- tected
Sum of le	akage 1, 2, 3, 4, 8, 9, 10, 11	and 12			13.7	9.8

 Table 1
 Measurement results – overview

Grid measurement across the ventilation exit openings

<sup>2)</sup> Corrected for background concentration

<sup>3)</sup> Left compressor house

<sup>4)</sup> Right compressor house

#### 2 Introduction and background

Danish Gas Technology Centre (DGC) is participating in the Danish project *Methane Emission from Danish Biogas Plants* together with AgroTech A/S. One of the major tasks of this project is to reduce greenhouse gas emission from biogas plants in terms of methane losses. This will be done by developing a method for identification and quantification of leakages from biogas plants. The developed method must be evaluated and compared to other methods.

This is done by participating in the joint project *Climate impact from biogas production, data collection and comparative study of measurement and cal- culation methods in Europe* carried out by Sveriges Tekniska Forskningsinstitut (SP). The project is financed by the Swedish Energy Agency through Swedish Gas Technology Centre (SGC). DGC is one of a number of participating parties.

The other participants in the comparative measurements are:

- SP Technical Research Institute of Sweden
- DBFZ Deutsches Biomasseforschungszentrum gemeinnützige GmbH
- Technical University of Denmark, Department of Environmental Engineering
- AgroTech A/S

The measurements presented in this report are DGC's contribution to the European project.

#### 3 Plant description

"Svensk Biogas i Linköping" is Sweden's largest biogas plant. It is situated in Linköping 200 km south west of Stockholm and has been in operation since 1996. The plant has an annually permit of treating 100,000 tonnes of waste, and the annual production of raw biogas was 15 m Nm<sup>3</sup> in 2012. Besides biogas the plant produces 80,000 tonnes of wet bio fertilizer, which is sold to farmers.

- Waste from slaughterhouse cattle/chicken/swine (e.g. production waste, blood)
- Food waste from supermarkets
- Organic waste from households
- Ethanol/residuals from ethanol production
- Main substrate feed is slaughterhouse waste ( $2012 \approx 80,000$  tonnes)
- Other types of substrates
  - $\circ$  Sewage sludge
  - Silage from grass
  - o Grains

Table 2 indicates the distribution of the substrates.

Table 2Distribution of substrates

Туре	Proportion (% weight)
Slaughterhouse cat. 3	27
Food Industry	37
Alcohol	1
Thin stillage	3
Fat	2
Glycerol	2
Food waste	28

The plant production layout comprises receiving and pre-treatment facilities, four digesters, gas storage/upgrading and bio filter. The layout is shown in Figure 1.



2 Receiving hall 3 Receiving/homogenisation tank 4 Hygienisation tank

5 Digesters, 4 units 6 Gas storage 7 Gas upgrading system 8 Flare

9 Manure well 10 Control room and office 11 Office 12 Bio filter

Figure 1 Layout Swedish Biogas in Linköping AB

#### 4 Measuring method applied by DGC

Methane emissions from known point sources and leakages at biogas plants are detected by an IR camera and subsequently quantified by measuring coherent values of a sample volume flow and methane concentration giving the possibility to calculate methane volume flow and mass flow from the point source.

A high volume sampler evacuates leaking biogas from crevices and holes and the sample flow (leaking biogas diluted in air) is measured by an orifice placed in the exit duct of the blower. The high volume sample system consists of a customized suction hood or custom built enclosure (bagging), ventilation hose and a high volume blower. See Figure 2.



Figure 2 High volume sampler

The methane concentration in the sample flow is measured by an FID hydrocarbon analyser. To ensure that only methane is detected, a non-methane hydrocarbon cutter is placed upstream the FID analyser.

If the source is a ventilation opening in a building, the flow is determined by a vane anemometer in grid pattern.

#### **5** Equipment and instruments

#### 5.1 Optical IR camera

- FLIR GF320

The IR camera was operated by AgroTech.

#### 5.2 Volume flow

Ventilation openings

- TSI Air Velocity Meter, Model 9555 Series
- Temperature measurement (Thermocouple type K)

#### Leakages and holes

Custom built high volume sampler including:

- Customized sample hoods for evacuating methane from leakages
- ATEX air blower for evacuating gas leaks
- Calibrated orifice (Lindab FMU 200-160)
- Temperature measurement (Thermocouple type K)

The physical conditions (relative humidity, temperature and atmospheric pressure) of the surrounding air were measured by Testo 511, Testo 174H and Elpro Ecolog TH1.

#### 5.3 Methane concentration

In order to measure only methane a heated non-methane hydrocarbon cutter were used before analysing the sample gas with an FID instrument.

#### Methane cutter

- Model 320 heated non-methane hydrocarbon cutter from The Signal Instrument Company Limited.

#### FID analyser

- Thermo FID from M&A Analysentechnik GmbH
- Range: 0 1000000 ppm (selectable range)
- Reproducibility:  $\pm 1$  % of range
- Linearity:  $\pm 1$  % of range

The analyser is calibrated with certified and traceable reference gasses before and after the measurements.

### 6 Uncertainty

\_

The main constituents of the combined uncertainty of the methane emission measurements are related to:

- Determination of volume flow or sample flow
- Determination of methane concentration
  - Sample conditions
    - Weather
    - o Physical characteristics of the sample location
    - Steadiness of the methane emission

DGC estimates an overall uncertainty of 15-25 % of measured values depending on above mentioned conditions.

#### 7 Performed measurements

## 7.1 Approach and purpose of the comparative measuring campaign

Prior to the measurement campaign the participating companies and laboratories discussed and agreed on a procedure to carry out the measurements.

- 1. Each and every participant performs a survey of the biogas plant in order to detect methane leakages and emissions.
- 2. The found leakages are presented at a meeting, and a gross list of leakages is produced.
- 3. Every participant performs quantification of all methane leakages on the gross list.
- 4. If the method permits, each participant presents the following results:
  - a. Quantification of each leakage.
  - b. Sum of leaks found by the company itself.
  - c. Sum of all leakages.

#### 7.2 Detected methane mission sources

12 sources of methane emission were detected by the participants. Figure 3 shows an overview based on a list prepared by AgroTech and DGC and subsequently submitted to all the teams.

DGC and AgroTech performed a joint effort in detecting leakages and did find all of the 12 leakages shown in Figure 3. For some of the leakages, however, it was hard to determine whether the observation was actually methane or flicker on the camera screen caused by hot and damp air.

#### DGC-report

#### Leakage # 1

Upgrading unit – ventilation duct 1 (facing north – away from the biogas plant) Leakage # 5

Leakage # 6

Leakage # 7

Carbon filter house – gas booster F22

Holes at flange of electric motor

Carbon filter house - gas booster F21

eles at flange of electric moto



<u>Leakage #2</u> Upgrading unit – ventilation duct 2 (facing east towards digester 4)



<u>Leakage #3</u> Upgrading unit – safety valve pipe leading above the container



<u>Leakage # 4</u> Carbon filter house – ventilation duct leading from the container





Carbon filter house - gas filter

<u>Leakage # 8</u> Hygienisation tank – man hole at the top of the tank



<u>Leakage # 9</u> Hygienisation tank – overloading valve



<u>Leakage # 10</u> Homogenisation tank – swan neck



<u>Leakage # 11</u> Homogenisation tank – hole in the concrete roof



<u>Leakage # 12</u> Hygienisation tank (one-hour holding tank) – swan neck at the top of the tank



Figure 3 Overview of detected leakages (arrows indicate precise location of the leakages)

#### 7.3 Measurement periods

DGC performed measurements in the period 8 September to 10 September 2014. Table 3 shows the measuring periods in detail.

Date	Time	Leakage #	Activity or sample location
8 Sep.	14:00-18:00	-	Scanning for leaks
9 Sep.	12:10-12:51	1	Upgrading unit – ventilation duct 2
	11:30-12:02	2	Upgrading unit – ventilation duct 1
	14:22-15:03	3	Upgrading unit – safety valve pipe at roof
	18:50-19:15	4	Carbon filter house – ventilation exit
	15:55-16:35	5	Carbon filter house – gas booster F22
	16:45-17:15	6	Carbon filter house – gas booster F21
	17:38-18:08	7	Carbon filter house – gas filter
10 Sep.	15:25-16:20	8	Hygienisation tank – man hole at tank top
	17:15-18:15	9	Hygienisation tank – overloading valve
	10:30-11:10	10	Homogenisation tank – swan neck
	11:25-12:05	11	Homogenisation tank – hole in the concrete roof
	Approx. 14:00	12	One-hour holding tank – swan neck at tank top

Table 3Measurement periods

#### 7.4 Operating conditions

Production data from Linköping biogas plant was provided by process engineer Sören Nilsson Påledal, Tekniska Verken FoU Biogas.

The production data is attached in Appendix 2 and 3.

#### Result of the methane emission quantification 8 measurements

#### 8.1 Overview

An overview of the results is shown in Table 4.

The individual measurements are described in Chapter 8.2 - 8.10.

Leakage ID	Sample location	Sample gas flow	Methane in sample gas	Background conc.	Methane emission <sup>2)</sup>	Methane emission <sup>2)</sup>
#		Nm <sup>3</sup> /h dry	ppm, dry	ppm, dry	Nm <sup>3</sup> /h dry	kg/h
1	Upgrading unit – ventilation duct 1 <sup>3)</sup>	6,740 <sup>1)</sup>	35	2.0	0.220	0.159
2	Upgrading unit – ventilation duct 2 <sup>4)</sup>	4,944 <sup>1)</sup>	82	2.0	0.396	0.285
3	Upgrading unit – safety valve pipe at roof	745	17,067	2.0	12.721	9.159
4	Carbon filter house – ventilation exit	604	315	9.0	0.185	0.133
5	Carbon filter house – gas booster F22	457	346	9.0	0.154	0.111
6	Carbon filter house – gas booster F21	456	181	9.0	0.078	0.0564
7	Carbon filter house – gas filter	452	37	9.0	0.013	0.0090
8	Hygienisation tank – manhole at tank top	522	19	2.5	0.0086	0.0062
9	Hygienisation tank – overloading valve	376	336	2.5	0.126	0.0904
10	Homogenisation tank – swan neck	458	11	2.5	0.0038	0.0027
11	Homogenisation tank – hole in the concrete roof	449	12	2.5	0.0045	0.0032
12	One-hour holding tank – swan neck at tank top	-	Not detected	-	Not detected	Not de- tected
Sum of le	akage 1, 2, 3, 4, 8, 9, 10, 11	and 12			13.7	9.8
<sup>1</sup> Grid measurement across the ventilation exit openings						

Table 4Measurement results – overview

Grid measurement across the ventilation exit openings

2) Corrected for background concentration

3) Left compressor house

4) Right compressor house

#### 8.2 Upgrading unit – ventilation duct 1



Flow measurement:

- 4 x 4 grid, i.e. 16 points, vane anemometer

Concentration:

- Uniform distribution over grid section, one point measurement

Methane emission: <u>0.159 kg/h</u>

#### 8.3 Upgrading unit – ventilation duct 2



Flow measurement:

- 4 x 4 grid, i.e. 16 points, vane anemometer

Concentration:

- Uniform distribution over grid section, one point measurement

Methane emission: <u>0.285 kg/h</u>



## 8.4 Upgrading unit – safety valve pipe leading above the container

Methane emission from the vent pipe was evacuated together with dilution/excess air by placing the ventilation suction hose directly over the vent pipe exit. This was later changed to a suction hood supported by plastic wrapping allowing for more dilution air and less vacuum at the vent pipe exit. However, the subsequent calculation of the methane mass flow showed equivalent results of the two sample techniques.

Methane emission: <u>9.159 kg/h</u>

#### 8.5 Carbon filter house – Ventilation exit



A suction hood was placed over the vent exit. Dilution air was secured by the ventilation intake grid of the building placed in the opposite wall. The methane concentration was steady after approx. 15 minutes.

Methane emission: <u>0.133 kg/h</u>



#### 8.6 Carbon filter house – gas booster F22

A plastic tub/container supported by additional plastic wrapping was used to encapsulate the gas booster. A large opening for sufficient dilution air was at the bottom of the line-up.

Methane emission: <u>0.111 kg/h</u>



#### 8.7 Carbon filter house – gas booster F21

A plastic tub/container supported by additional plastic wrapping was used to encapsulate the gas booster. A large opening for sufficient dilution air was at the bottom of the line-up.

Methane emission: 0.0564 kg/h

#### 8.8 Carbon filter house – gas filter



The gas filter was encapsulated in the same way as for the gas boosters (no picture of the actual encapsulation line-up).

Methane emission: <u>0.0090 kg/h</u>

#### 8.9 Hygienisation tank – manhole at the top of the tank



The manhole flange was covered by a suction hood and additional plastic wrapping (no picture of encapsulation).

Methane emission: <u>0.0062 kg/h</u>



#### 8.10 Hygienisation tank – overloading valve

The top of the moisture collection overflow container was covered by a suction hood and plastic wrapping. To ensure sufficient dilution air a hole was cut in the suction hood.

The emitted gas was very damp and did subjectively vary a lot in flow. Occasionally, small amounts of gas escaped the sampling hood, but this is not considered to have significant impact on the outcome of the measurement.

DGC observed that other components than methane must have been present in the gas. Looking at the trend curve, a steady emission of approx. 200 ppm was observed in the period 18:05-18:20. The peak of approx. 400 ppm 18:20-18:25 is reflecting the FID analyzer with by-passed non-methane cutter.

Methane emission: <u>0.0904 kg/h</u>



#### 8.11 Homogenisation tank – swan neck

In order to minimize the potential influence of sucking biogas from the tank/swan neck a large encapsulation was built to cover the entire swan neck. The sample flow was adjusted to an average flow of approx. 0.5 m/s across the neck at the concrete roof (measured perpendicular to the flow direction by a vane anemometer).

Methane emission: <u>0.0027 kg/h</u>



#### 8.12 Homogenisation tank – Hole in the concrete roof

In order to minimize the potential influence of sucking biogas from the tank a large encapsulation was built to cover the hole. The sample flow was adjusted to an average flow of approx. 0.5 m/s across the hole in the concrete roof (measured perpendicular to the flow direction by a vane anemometer).

*Methane emission:* <u>0.0032 kg/h</u>

# 8.13 Hygienisation tank (one-hour holding tank) – swan neck at the top of the tank

No emission detected.

## 9 Appendices

Appendix 1 – Linköping Biogas Plant layout

- Appendix 2 Data from Linköping biogas plant 8-9 September
- Appendix 3 Data from Linköping biogas plant 10-11 September
- Appendix 4 Registration of ambient air conditions



Appendix 1 – Linköping Biogas Plant layout

## Appendix 2 – Data from Linköping biogas plant 8-9 September

Copy of e-mail from process engineer Sören Nilsson Påledal, Tekniska Verken FoU Biogas.

The diameter of the manure well is approx. 37 m. Production at the chemical scrubber between 9-17 Tuesday 9 september Raw biogas 1922 Nm3/h according to flowmeters Methane content Raw biogas 62 vol% Upgraded biogas 1180 Nm3/h Methane content Upgraded biogas 96,4 vol% Produktion 8 september 1200 – 9 september 1200 Raw biogas 1615 Nm3/h according to flowmeters Methane content Raw biogas 62 vol% Production for 2013 for the chemical scrubber (except waterscrubbers) Raw biogas 1850 Nm3/h according to flowmeters (16 206 000 Nm3 for whole year) Methane content Raw biogas 64 vol% Upgraded biogas 1215 Nm3/h (10 643 400 Nm3 for whole year) Methane content Upgraded biogas 96,6 vol% The water scrubbers were only used from Wednesday 10/9 09:45 to Thursday 11/9 09:00 during the week. During this time the chemical scrubber was used for less than 50 % (one compressor and dryer not in use) and there were no leakages from the security valve pipe.

## Appendix 3 – Data from Linköping biogas plant 10-11 September

Copy of e-mail from process engineer Sören Nilsson Påledal, Tekniska Verken FoU Biogas.

Production at the chemical scrubber between 9-17 Wednesday 10 september Raw biogas 1830 Nm3/h according to flowmeters Methane content Raw biogas 62 vol% Upgraded biogas 1160 Nm3/h Methane content Upgraded biogas 97,1 vol%

Production at the chemical scrubber between 9-17 Thursday 11 september Raw biogas 1990 Nm3/h according to flowmeters Methane content Raw biogas 60 vol% Upgraded biogas 1230 Nm3/h Methane content Upgraded biogas 96,6 vol%

Date	Time	Air humidity	Atmospheric pressure	Air temperature
yyyy-mm-dd	hh:mm	RH %	hPa	°C
2014-09-09	11:30	76.0	1001.8	18.8
2014-09-09	12:25	65.1	1001.9	17.5
2014-09-09	12:50	62.2	1002.0	17.0
2014-09-09	13:21	74.7	1001.9	16.8
2014-09-09	15:00	97.4	1002.9	13.3
2014-09-09	17:09	94.6	1001.7	15.8
2014-09-09	17:44	85.2	1001.9	16.4
2014-09-09	18:01	81.1	1001.9	16.3
2014-09-09	19:21	93.3	1002.4	13.1
2014-09-10	10:42	79.5	1008.8	16.3
2014-09-10	10:59	82.0	1009.0	15.6
2014-09-10	11:39	76.1	1009.4	17.3
2014-09-10	11:53	70.3	1009.5	18.5
2014-09-10	12:07	73.4	1009.6	17.6
2014-09-10	15:28	73.3	1011.4	18.6
2014-09-10	15:48	76.1	1011.4	17.8
2014-09-10	16:00	77.6	1011.7	17.2
2014-09-10	16:12	77.7	1011.8	17.4
2014-09-10	17:08	69.6	1012.0	17.6
2014-09-10	17:30	71.4	1012.2	17.4
2014-09-10	17:39	71.1	1012.2	17.6
2014-09-10	17:58	68.0	1012.4	17.7
2014-09-10	18:12	69.5	1012.6	17.4
2014-09-10	18:28	69.3	1012.8	16.8

## Appendix 4 – Registration of ambient air conditions

E. DTU Quantification of fugitive methane emissions from the biogas plant in Linköping





# Quantification of fugitive methane emissions from the biogas plant in Linköping (SE)



Atmospheric concentrations of methane and tracer gas above background level at approximately 900 meters downwind from the biogas plant measured on September 10<sup>th</sup>, 2014. The triangle marks the placement of the tracer gas release on the facility.

Antonio Delre, Jacob Mønster & Charlotte Scheutz

DTU Environment Department of Environmental Engineering

October 2014

## Contents

1.	Introduction and purpose	1
2.	Description of the measurement method	1
3.	Description of the measurement campaign.	2
4.	Results and discussion	4
4.1	Biogas plant layout and gas production	4
4.2	Methane screening of the area surrounding the biogas plant	6
4.3	Initial on-site methane screening of the biogas plant	7
4.4	Whole plant fugitive emissions	8
5.	Conclusion	4
6.	References	4



## 1. Introduction and purpose

Greenhouse gas (GHG) emissions from facilities treating organic waste are often difficult to quantify due to the diffusive nature of the emissions combined with large temporal variation and the challenging physical structure of the facility. Only over the last few years, the scientific community has developed methodologies and strategies of GHG quantifications from biogas facilities. However, there is no single measurement method that has been recognized as a standard method yet. The Technical University of Denmark has recently implemented a novel analytical setup enabling mobile measurements of small (ppb level) changes in atmospheric methane concentrations. This enables detection and quantification of methane emission sources by performing measurements downwind from the source in combination with release and measurement of a tracer gas. The analytical setup and the dynamic tracer dispersion method have been tested at a number of landfills and wastewater treatment plants since November 2011 (Mønster et al., 2014b; Yoshida et al., 2014), building up a sound knowledge on quantification of fugitive methane emissions from full-scale facilities.

The objective of this study was to quantify the methane emission from a Swedish biogas plant using the tracer dispersion method. The study was part of a large comparison study where other groups performed parallel methane detections and emission quantification using a range of different technologies including on-site measurements and remote sensing approach coupled to backward Lagrangian Stochastic inverse modelling.

## 2. Description of the measurement method

Total methane emissions were quantified using a mobile tracer dispersion method that combines a controlled release of tracer gas from the biogas facility with concentration measurements downwind of the facility, by using a mobile high-resolution analytical instrument (Mønster et al., 2014a; Yoshida et al., 2014).



Figure 1. The principle of the dynamic plume method for quantifying GHG emissions from area sources

The tracer dispersion method, shown in Figure 1, is based on the principle that a tracer gas released at a source area, in this case a biogas facility, disperses into the atmosphere likewise the methane emitted from the same area. Since the ratio of their concentrations remains constant along their atmospheric dispersion, the methane emission rate can be calculated using the following expression when the tracer gas release rate is known:

 $E_{CH_4} = Q_{tr} * \frac{\int_{plume\ end}^{plume\ end} (C_{CH_4}) dx}{\int_{plume\ end}^{plume\ end} (C_{tr}) dx} \frac{MW_{CH_4}}{MW_{tr}}$ 

where  $E_{CH_4}$  is the methane emission in mass per time,  $Q_{tr}$  is the tracer release in mass per time,  $C_{CH_4}$  and  $C_{tr}$  are the measured downwind concentrations in parts per billion (ppb) subtracted of their background concentrations and  $MW_{CH_4}$  and  $MW_{tr}$  are the molar weights of methane and tracer gas, respectively (Mønster et al., 2014a). In this study, acetylene (C<sub>2</sub>H<sub>2</sub>) was used as tracer due to its long atmospheric lifetime. Downwind plume concentrations were measured driving along transects with a cavity ring down spectrometer (CRDS) from Picarro (model G2203), which is a fast and high sensitive gas analyzer capable to detect methane and acetylene concentrations down to ppb level every second (Mønster et al, 2014a; Yoshida et al., 2014). A GPS was connected to the instrument for logging the measured concentrations to their geographical location. In order to obtain the best possible simulation of the source area, the tracer gas was released from the part of the plant where the most elevated methane concentration was seen and/or expected.

## 3. Description of the measurement campaign.

Measurements were performed from September 9<sup>th</sup> to September 12<sup>th</sup>, 2014. During the first two days, area and plant methane screenings were performed, followed by tracer release and methane emission quantification. The absence of favourable wind direction and speed during 11<sup>th</sup> and 12<sup>th</sup> allowed only a further confirmation of the plant screening. Different tracer gas release rates were tried out to have sufficient tracer gas for quantification in the downwind plume, but also to have tracer gas release for enough time to perform several plume traverses. Successful quantifications were done in the afternoon on Tuesday 9<sup>th</sup> and Wednesday 10<sup>th</sup> with a total tracer gas release of 0.44 kg h<sup>-1</sup>. The tracer gas was released in one point from one gas bottle placed next to the gasholder in the digesters area. Figure 2 shows the securing of the acetylene cylinder, while Figure 3 shows its location.



Figure 2. Acetylene cylinder secured at the digesters area



Figure 3. Acetylene cylinder placement close to digester tanks marked with red circle

The measurements were performed during a period with stable weather conditions. On Tuesday 9<sup>th</sup> the sky was cloudy with light rain, the atmospheric pressure was around 1003 mbar and the air temperature was about 13 °C. On Wednesday 10<sup>th</sup> the sky was partly cloudy, the atmospheric pressure was around 1014 mbar and the air temperature was about 17 °C. On both days the wind blew from east with an average velocity of 1 m/s. While on Tuesday 9<sup>th</sup> twenty-one plume traverses were completed, on Wednesday 10<sup>th</sup> twenty-four plume traverses were carried out. The following two days the calm (no wind), sunny and warm weather resulted in a fast plume rising, which made methane measurements of downwind plumes at ground level far from the plant impossible.

## 4. Results and discussion

#### 4.1 Biogas plant layout and gas production

The biogas plant in Linköping processes source separated household waste ( $\approx$ 50%), industrial food waste ( $\approx$ 25%), like dairy wastewater, and slaughterhouse waste ( $\approx$ 25%). After pretreatment such as screening, grinding, watering and mixing, the substrate undergoes pasteurization followed by anaerobic digestion. The biogas plant has two digester tanks and one tank for biogas storage. 95% of the biogas is upgraded by a chemical scrubber, which sometimes is supported by a water scrubber that processes only about 5% of the upgraded biogas over one year. The digestate is stored in an open tank for 15-20 days before being delivered to farmers for land application. Figure 4 and Figure 5 give a visual overview of some of the process units. Notice that even though Figure 5 dates back to September 2011, it gives a very good understanding about the main units' locations in the plant.



Figure 4. Linköping biogas plant. From left to right: a) Food waste receiving area and pretreatment unit and thermal process tank, b) Main process units including digester tanks and biogas upgrading systems, and c. Digestate storage in open tank.



Figure 5 - Linköping biogas plant layout; a) Food waste pretreatment unit and thermal process tank, b) Substrate inlet and pasteurization tank, c) Digesters, gasholder and flare, d) Digestate storage in an open tank, e) Chemical scrubber and f) Water scrubber

Table 1 and Table 2 report biogas production during the days when the methane emissions quantifications were performed. Table 1 and Table 2 show the biogas and methane production before and after upgrading, respectively.

Lapse of time	Raw Biogas (Nm <sup>3</sup> h <sup>-1</sup> )	CH₄ content (%)	CH₄ production before upgrading (Nm <sup>3</sup> h <sup>-1</sup> )
Tuesday 9 <sup>th</sup> from 15:30 to 16:00	1910	60.0	1146
Wednesday 10 <sup>th</sup> from 17:00 to 19:30	1800	63.0	1134

Table 1. Methane production before t	biogas upgrading process
--------------------------------------	--------------------------

Table 2. Methane production after biogas upgrading process

Lapse of time	Upgraded Biogas (Nm <sup>3</sup> h <sup>-1</sup> )	CH₄ content (%)	CH <sub>4</sub> production after upgrading (Nm <sup>3</sup> h <sup>-1</sup> )
Tuesday 9 <sup>th</sup> from 15:30 to 16:00	1180	96.2	1142
Wednesday 10 <sup>th</sup> from 17:00 to 19:30	1184	96.0	1133

The volume of biogas is expressed according to standard temperature and pressure (STP) i.e. 0 °C and 1 atmosphere (DIN 1343). During measurements conducted on Wednesday 10<sup>th</sup>, biogas upgrading occurred with water scrubber support.



#### 4.2 Methane screening of the area surrounding the biogas plant

Figure 6 shows different potential sources known to emit methane into the atmosphere in the surroundings of the biogas plant. North of the biogas plant is a sorting facility (receiving non-organic waste), while Northeast of the biogas plant is a landfill close to an incineration plant. The landfill is expected to release significant amounts of methane, whereas no methane emissions are expected from the incineration plant. South of the biogas plant is a wastewater treatment plant (WWTP), which stores its biosolids in heaps located relatively close to the biogas plant. The storage of biosolids could potentially emit methane.



Figure 6. Location of treatment facilities in the surroundings of the biogas plant

The methane screening of the area is showed in Figure 7 where methane concentrations above background level are marked in red. Measurements were carried out with wind blowing from the East. Area screening upwind and downwind the biogas plant allowed distinguishing atmospheric methane plumes from different sources. Figure 7 shows methane concentrations measured at two different distances downwind the landfill. The lower concentrations measured at a further distance from the landfill underlines the atmospheric gas dispersion. Furthermore, methane plumes from the biogas plant, the biosolids storage and WWTP were observed. Notice that upwind of the three sources, the methane concentration was close to background. Emissions from the biogas plant are depicted in yellow. The distinction between emission from the WWTP biosolids storage and biogas plant was carried out making sure that the tracer release simulated the biogas plant well and that good mixing between methane and acetylene ( $C_2H_2$ ) was obtained. Screening inside the sorting facility did not highlight any relevant methane releases.



Figure 7. Methane screening of biogas plant surroundings with wind blowing from east.

#### 4.3 Initial on-site methane screening of the biogas plant

A plant methane screening is usually used for identification of hotspots emission areas for optimal placement of the tracer gas cylinder to obtain the best methane source simulation. Methane might be released from the top of the digester tanks, which will be difficult to see based on on-site measurements due to the elevated release height in comparison to the measuring height (2m). Therefore in addition, information about the biogas plant and the methane plumes were used as support to identify the tracer gas placement.

Figure 8 shows methane concentrations measured during the plant screening at two different screening campaigns during calm and warm weather conditions with vertical plume rise from the area. Therefore, the detected methane concentrations are related to releases from the closest process units. The on-site screening indicated methane emissions from the open digestate storage tank, from the food waste pre-treatment area, from the biogas upgrading units and from the digester tanks.


Figure 8. Biogas plant screening during calm and warm weather with plume vertical mixing. The maximum methane concentration above background was 9.0 ppm and 12.8 ppm during the two screenings, respectively.

#### 4.4 Whole plant fugitive emissions

The placement of the tracer gas release was chosen in order to match biogas plant emissions and to distinguish methane released from the close WWTP biosolids storage. One tracer cylinder was chained up at the digesters area as reported in section 3. Figure 9 shows an example of downwind plumes, which was detected at 17:34 on Wednesday 10<sup>th</sup> along a road 800-900 m away from the plant, distant enough to consider the plant as point source. The tracer location is marked with a yellow triangle, while methane and acetylene plumes are showed in red and yellow, respectively. Peak concentrations above background level were 0.2 ppm of CH<sub>4</sub> and 3.1 ppb of C<sub>2</sub>H<sub>2</sub>. The tracer and the methane plume from the biogas plant follow each other nicely indicating a good simulation of the methane emission from the biogas plant. It should be noted that the way plumes follow each other underlines their good mixing. The figure also shows the adjacent methane plume coming from WWTP biosolids storage in southern direction. With wind from the East, it is possible to distinguish the two plumes from the biogas plant and the biosolids storage area from each other. However, if the wind shifts more to the North the plumes will blend together.



Figure 9. Atmospheric concentrations of methane and tracer gas above background level at approximately 900 meters downwind from the biogas plant measured on September 10th, 2014 at 17:34. The yellow triangle marks the placement of the  $C_2H_2$  tracer gas release on the facility.

Further confirmation of correct tracer placement and emission source simulation is showed in Figure 10b by calculation of the coefficient of determination (R^2). The R^2 coefficient is obtained by plotting the plume concentrations of methane and acetylene to each other, and is an indicator of the degree of plume mixing. In this case, the R^2 was very high (0.94) indicating a good simulation of the methane plume.

For plume integration, the methane plumes form the biogas plant and the biosolids storage area have to be separated from each other. For most of the plume traverses, this was possible as there was very little overlap of the plumes. The methane plume separation was done using the tracer gas plume and tracking back the methane and tracer gas plume to the biogas plant using the wind direction. An example of methane plume separation is shown in Figure 10a. It is evident that the methane and the tracer gas plume from the biogas plant follows each other very nicely. However, it is also clear that there is another methane plume to the left, which comes from another source (the biosolids storage area) as there is no sign of the tracer. As the overlap of the plumes in this case was very little it is valid to separate the two plumes as shown in Figure 10a and b.



Figure 10. Data elaboration and confirmation of tracer placement; a) Concentrations over time b) Coefficient Of Determination (R^2) calculation.

Out of the more than forty plume transects performed on September 9<sup>th</sup> and 10<sup>th</sup>, 21 transects were useful for calculating the whole biogas plant methane emission. Plume transects where methane plumes were overlapping when the wind shifted more to North were thus not included. Single emission rates for each individual plume transects are listed in Table 3 and expressed both in kg CH<sub>4</sub> h<sup>-1</sup> and Nm<sup>3</sup> CH<sub>4</sub> h<sup>-1</sup> (STP: 1 atm, 0°C). Furthermore, emission factors (EF) related to raw biogas production and to upgraded biogas are reported. The EF is calculated as the ratio between the methane emission and the methane production for the specific measuring hours – both expressed in Nm<sup>3</sup> CH<sub>4</sub> h<sup>-1</sup>.

The average methane emission rate from the biogas plant was found to be  $23.6\pm1.8 \text{ kg CH}_4 \text{ h}^{-1}$ , which corresponds to  $33.0\pm2.6 \text{ Nm}^3 \text{ CH}_4 \text{ h}^{-1}$ . The confidence interval was calculated considering a t-distribution at significance level ( $\alpha$ ) of 5%, giving a confidence interval of 95% ( $1 - \alpha$ ). This means that there is 95% probability that the right value falls within the confidence interval<sup>1</sup>. The smaller the confidence interval (*Conf.*) is, the higher is the accuracy of the methane quantification. In this case, the good accuracy was reached due to the high number of transverses included in the calculation (Mønster et al., 2014a). The plant emission factors, both referred to raw produced biogas (see Table 1) and upgraded biogas (see Table 2), were 2.9\pm0.2%, which is in the range of what was seen at other Swedish plants (Holmgren, 2014).

<sup>&</sup>lt;sup>1</sup> The confidence interval was calculated as following:  $\bar{y} \pm SEM * t_{\nu,\alpha/2}$ 

where  $\bar{y}$  is the average value; *SEM* is the standard error of means (*SEM* = *SD*/ $\sqrt{Count}$ ); *SD* is the standard deviation of the sample; *Count* is the number of transverses.  $t_{\nu,\alpha/2}$  is the probability point of *t* distribution with  $\nu$  degrees of freedom and significance level  $\alpha$  as known as tail area probability. Therefore, such as confidence interval provides the range of values within the right value falls with  $(1 - \alpha)$  probability.

Dav	Peak time	Emission	Emission	EF Raw biogas	EF Upgraded biogas	
	(hh:mm)	(kg CH₄ h <sup>-1</sup> )	(Nm <sup>3</sup> CH₄ h <sup>-1</sup> )	(%)	(%)	
Tuesday 9 <sup>th</sup>	15:36	20.8	29.1	2.5	2.6	
	15:46	18.4	25.8	2.3	2.3	
	15:59	14.6	20.4	1.8	1.8	
	17:11	22.6	31.7	2.8	2.8	
	17:22	22.7	31.8	2.8	2.8	
	17:26	24.8	34.7	3.1	3.1	
	17:34	16.6	23.2	2.0	2.0	
	17:40	19.6	27.5	2.4	2.4	
	17:49	23.2	32.5	2.9	2.9	
	18:01	25.9	36.3	3.2	3.2	
	18:06	26.9	37.6	3.3	3.3	
Wednesday 10 <sup>th</sup>	18:33	28.4	39.7	3.5	3.5	
	18:39	20.2	28.3	2.5	2.5	
	18:44	23.7	33.2	2.9	2.9	
	18:50	26.0	36.4	3.2	3.2	
	18:55	26.0	36.4	3.2	3.2	
	19:03	24.7	34.6	3.0	3.1	
	19:06	29.8	41.7	3.7	3.7	
	19:09	25.9	36.3	3.2	3.2	
	19:16	25.3	35.4	3.1	3.1	
	19:22	29.4	41.2	3.6	3.6	
Average		23.6	33.0	2.9	2.9	
Conf.*		1.8	2.6	0.2	0.2	
SD		4.1	5.7	0.5	0.5	
Count		21	21	21	21	
SEM		0.9	1.2	0.1	0.1	

Table 3. Methane emission rates and Emission Factors (EF) measured at the biogas plant.

Conf.: Confidence Interval calculated with t distribution at significance level  $\alpha$ =5%

SD: standard deviation, Count: number of transects (dimensionless value), SEM: standard error of means.

Using information about the plant layout and the wind direction during measurements, quantification of emissions coming from digesters and gasholder area could be estimated. Wind blowing from East, as showed in. Figure 11, generates a downwind plume whose northern part (marked in green) describes emissions coming from digesters and gasholder area, whereas the southern part (marked in light blue) shows emissions coming from other process units.



Therefore, the calculation for emissions generated from digesters and gasholder area was carried out integrating only the green part of the acetylene and methane plumes.



Figure 11. Estimation of methane emissions from digesters and gasholder area. Only the green part of the plume was used for this calculation. Downwind plume measured on September 10th, 2014 at 18:49.

Table 4 lists the methane emission estimated from the area with the digesters and the gasholder for each successful transect. The values are compared with the whole plant methane emissions in order to estimate the percentage of methane emissions coming from this area. The average emission from the area with the digester tanks and the gasholder was  $14.9\pm0.9$  kg CH<sub>4</sub> h<sup>-1</sup>, which corresponds to  $65\pm6\%$  of the total emission of the biogas plant. The standard deviation of 14% (see Table 4) underlines the high uncertainty of this value, which therefore can be used only for a rough estimation.

Day of September 2014	Peak time (hh:mm)	Whole plant Emission (kg CH₄ h⁻¹)	Emission estimation from Digesters Area (kg CH4 h <sup>-1</sup> )	Emission share from Digesters Area (%)	
	15:36	20.8	18.1	87	
Tuesday 9 <sup>th</sup>	15:46	18.4	15.4	83	
	15:59	14.6	11.7	80	
	17:11	22.6	16.2	72	
	17:22	22.7	16.7	74	
	17:26	24.8	17.8	72	
	17:34	16.6	14.4	87	
	17:40	19.6	15.5	79	
	17:49	23.2	14.7	63	
	18:01	25.9	16.2	62	
	18:06	26.9	13.3	49	
Mada and a start 10 <sup>th</sup>	18:33	28.4	13.5	48	
weanesaay 10	18:39	20.2	10.5	52	
	18:44	23.7	17.0	72	
	18:50	26.0	17.8	68	
	18:55	26.0	12.4	48	
	19:03	24.7	14.5	59	
	19:06	29.8	13.5	45	
	19:09	25.9	15.3	59	
	19:16	25.3	13.7	54	
	19:22	29.4	14.3	49	
Average		23.6	14.9	65	
Conf.		1.8	0.9	6	
SD		4.1	2.0	14	
Count		21	21	21	
SEM		0.88	0.45	3	

Table 4. Estimation of CH4 fugitive emissions from digesters area

Conf.: Confidence Interval calculated with t distribution at significance level  $\alpha$ =5%

SD: standard deviation, Count: number of transects (dimensionless value), SEM: standard error of means.

More information can be drawn comparing results from different days. Table 5 reports these calculations from data listed in Table 3. The average emission rate measured on Tuesday 9<sup>th</sup> was 17.9±7.8 kg CH<sub>4</sub> h<sup>-1</sup>, which corresponds to 25.1±10.9 Nm<sup>3</sup> CH<sub>4</sub> h<sup>-1</sup>, and on Wednesday 10<sup>th</sup> 24.5±1.7 kg CH<sub>4</sub> h<sup>-1</sup>, which corresponds to 34.4±2.4 Nm<sup>3</sup> CH<sub>4</sub> h<sup>-1</sup>.

EFs calculated for data collected on Tuesday 9<sup>th</sup> are 0.8% smaller than those calculated for data collected on Wednesday 10<sup>th</sup>. The different number of transects between the two quantification days, 3 on 9<sup>th</sup> vs 18 on 10<sup>th</sup>, involves different confidence interval underlining a more accurate value on Wednesday (0.2%) than Tuesday (1.0%). Nevertheless, the higher fugitive methane emissions on Wednesday 10<sup>th</sup> compared to Tuesday 9<sup>th</sup> could be explained by the additional use of water scrubber in the second day (see introduction to section 4), which is a technology known to release more CH<sub>4</sub> to the atmosphere than chemical scrubber (Petersson, 2012). Notice that also in this case the two EFs have the same value.



	Whole plant emissions		Whole plant emissions		Emission Factor (%)		Emission Factor (%)	
	(kg CH₄ h <sup>-1</sup> )		$(Nm^3 h^{-1})$		Raw biogas		Upgraded biogas	
Day of	Tue	Wed	Tue	Wed	Tue	Wed	Tue	Wed
sep-14	$\boldsymbol{9}^{th}$	10 <sup>th</sup>	$9^{th}$	10 <sup>th</sup>	$9^{th}$	10 <sup>th</sup>	$9^{th}$	10 <sup>th</sup>
Average	17.9	24.5	25.1	34.4	2.2	3.0	2.2	3.0
Conf.	7.8	1.7	10.9	2.4	1.0	0.2	1.0	0.2
SD	3.1	3.4	4.4	4.8	0.4	0.4	0.4	0.4
Count	3	18	3	18	3	18	3	18
SEM	1.8	0.8	2.5	1.1	0.2	0.1	0.2	0.1

Table 5. Whole plant fugitive emissions in two different measurements days

Conf.: Confidence Interval calculated with t distribution at significance level  $\alpha$ =5%

SD: standard deviation, Count: number of transects (dimensionless value), SEM: standard error of means.

# 5. Conclusion

Off-site and on-site methane screenings indicated methane releases from digesters, biogas upgrading units, digestate storage tank and pre-treatment area. Methane emission from Linköping biogas plant, during the afternoons of September 9<sup>th</sup> and September 10<sup>th</sup>, was successfully quantified using the tracer dispersion method. The emission rate measured was 23.6±1.8 kg CH<sub>4</sub> h<sup>-1</sup> (corresponding to 33.0±2.6 Nm<sup>3</sup> CH<sub>4</sub> h<sup>-1</sup>). Plant emission factors (EFs) referred to raw produced biogas and upgraded biogas were 2.9±0.2%. The methane emission from the area with the digester tanks were estimated to account for approximately 65±6% of the total emission from the plant. The remaining part of the methane emission came from the pre-treatment area and the digestate storage in an open tank.

On Wednesday 10<sup>th</sup> the methane emission was higher (24.5±1.7 kg CH<sub>4</sub> h<sup>-1</sup>, which corresponds to 34.4±2.4 Nm<sup>3</sup> CH<sub>4</sub> h<sup>-1</sup>) than the emission measured on Tuesday 9<sup>th</sup> (17.9±7.8 kg CH<sub>4</sub> h<sup>-1</sup>, which corresponds to 25.1±10.9 Nm<sup>3</sup> CH<sub>4</sub> h<sup>-1</sup>). This might be explained by the additional use of water scrubber during the second day, which is a technology known to release more CH<sub>4</sub> in the atmosphere than chemical scrubber.

Stable weather conditions characterized each methane quantification day during measurements. In both days, wind blew from East with an average velocity of 1 m/s. The following two days (September 11<sup>th</sup> and 12<sup>th</sup>) the calm, sunny and warm weather resulted in a fast plume rising, which made impossible methane measurements of downwind plumes at ground level far from the plant.

# 6. References

- Holmgren, M.A., 2014. Methane Emission from Swedish Biogas Plants, in: 2nd IBBA Workshop on Methane Emissions in Kiel, Germany, September 4th 2014. SCG - Swedish Gas and Technology Center.
- Mønster, J.G., Samuelsson, J., Kjeldsen, P., Rella, C.W., Scheutz, C., 2014a. Quantifying methane emission from fugitive sources by combining tracer release and downwind

measurements - a sensitivity analysis based on multiple field surveys. Waste Manag. 34, 1416–28.

Mønster, J., Samuelsson, J., Kjeldsen, P., Scheutz, C. 2014b. Quantification of methane emission from 15 Danish landfills using mobile tracer dispersion method. Waste Management. *In press.* 

Petersson, A., 2012. The Swedish Voluntary system for control of methane emissions.

Yoshida, H., Mønster, J., Scheutz, C., 2014. Plant-integrated measurement of greenhouse gas emissions from a municipal wastewater treatment plant. Water Res. 1, 108–118.

F. SP Methane measurements at the Linköping biogas plant







Contact person Magnus Andreas Holmgren Energy Technology +46 10 516 56 18 magnus.andreas.holmgren@sp.se Date Reference 2015-01-27 ETf6436

Page 1 (3)

SGC

# Methane measurements at the Linköping biogas plant

## Introduction

Measurements were performed by 4 different teams on site and 2 different teams with remote sensing. This report gives the results from the measurements performed on site by SP Technical Research Institute of Sweden. Measurements were mainly performed by Magnus Andreas Holmgren, partly with assistance from Johan Yngvesson.

## Equipment and execution

Leak detection and measurements on the biofilter were performed on the 8<sup>th</sup> of September. All other measurements were performed on the 9<sup>th</sup> of September.

Methane concentration was analysed on site with a FID analyser equipped with a Non Methane Hydrocarbon Cutter (JUM). The FID was calibrated with 900 ppm or 8000 ppm  $CH_4$  in synthetic air.

### Methods and results

The methods used follow the Swedish handbook on methane measurements (SGC report no 227) and the limitations and recommendations given by the Swedish Voluntary Agreement system (Avfall Sverige report no 2007:02 rev. 2009).

#### Homogenisation tank

Minor methane emissions were identified with the portable leak detector, but the levels were so low that given by recommendations in AS report 2007:02 rev. 2009 it was decided to disregard measurements.

#### Hygenization tank

A minor methane emission was identified with the portable leak detector at the pipe coming from the homogenisation tank, but the levels were so low that given by recommendations in AS report 2007:02 rev. 2009 it was decided to disregard measurements.

A minor leak was detected with the portable leak detector at a manhole. Leaks are not quantified in the Swedish system and hence SP has no equipment to measure the mass flow from this leak.

#### SP Technical Research Institute of Sweden

Postal address SP Box 857 SE-501 15 BORÅS Sweden Office location Västeråsen Brinellgatan 4 SE-504 62 BORÅS

Phone / Fax / E-mail +46 10 516 50 00 +46 33 13 55 02 info@sp.se This document may not be reproduced other than in full, except with the prior written approval of SP.





### <u>Biofilter</u>

In the Swedish system biofilters are regarded as having no effect on the methane concentration in the treated gas. Hence measurements were performed in the pipe directly before the biofilter. The methane concentration was analysed to be 80 ppm with the FID (with NMHC). The gas flow was measured with a pitot tube and a differential pressure sensor as 3 564 Nm3/h (with correction for pressure and temperature). The resulting emission mass flow is 204 g  $CH_4/h$ .

#### Digestate storage

The emission was quantified with an open chamber technique using a VDI-hood manufactured by OdourNet in Germany (according to specifications given in VDI 3880). The FID was used to analyse the methane concentration of the offgas from the VDI-hood. Measurements were performed on 4 different points on the large surface area of the storage tank. The resulting mean emission mass flow rate is 7 335 g  $CH_4/h$ .

#### Activated carbon filter (building)

Measurements were performed in the 2 air ventilation openings in the building. The methane concentration was analysed to be 1 400 ppm with the FID (without NMHC) using a pump to take a gas sample in a sample bag. Flow was measured with a hot-wire anemometer in the ventilation openings giving the result 81 Nm<sup>3</sup>/h. The resulting emission mass flow is 80 g  $CH_4/h$ .

#### Chemical scrubber

The methane concentration was analysed with the FID, with a mean value of 169 ppm. It is not possible to measure the flow at the sampling point, instead the flow is calculated from the product gas flow and methane concentration (given by the plant). This gives an emission mass flow of 85 g  $CH_4/h$ .

#### Right compressor building

The methane concentration was analysed with the FID, with a mean value of 113 ppm. Gas flow was given by fan data found to be 1,375 m<sup>3</sup>/s. With correction of the gas flow with temperature, this gives an emission mass flow of 363 g  $CH_4/h$ .

#### Left compressor building

The methane concentration was analysed with the FID, with a mean value of 30 ppm. Gas flow was given by fan data found to be 1,375 m<sup>3</sup>/s. With correction of the gas flow with temperature, this gives an emission mass flow of 98 g CH<sub>4</sub>/h.

#### Pressure relief vents

These leaks were not identified by SP. When being informed by the other teams about one major leak it was decided to perform measurements, but unfortunately when we tried to perform measurements on the Wednesday, the machine was not in operation and hence there was no leak at this time.

#### Analysis instruments on site

Four gasflows through different analysis instruments were identified. The flows were read on the rotameters and the methane concentrations were given by the analysis instruments on site, resulting in an emission flow rate of 41 g  $CH_4/h$ .





Overall emission

The overall emission from the points measured is 8 206 g CH<sub>4</sub>/h.

# SP Technical Research Institute of Sweden Energy Technology - Combustion and Aerosol Technology

Performed by

Magnus Andreas Holmgren

# MEASUREMENTS OF METHANE EMISSIONS FROM BIOGAS PRODUCTION

Metanutsläpp från biogasproduktion, hur stora är de? Och hur kan vi hitta och mäta dem? Biogas är ett klimatneutralt bränsle eftersom det produceras från biomassa som bundit atmosfärisk koldioxid. Men eftersom dess huvudsakliga beståndsdel, som är metan, är en kraftfull växthusgas är det viktigt att så lite som möjligt av biogasen kommer ut i atmosfären.

Här redovisas resultaten av ett projekt, som med stöd av den europeiska gasbranschens forskningsorganisation GERG, har låtit några av Europas mest erfarna organisationer och experter jämföra olika metoder och angreppssätt för mätning av metanutsläpp.

Mätningarna på biogasanläggningen i Linköping visade till exempel att resultaten av olika metoder är jämförbara även om osäkerheten i alla typer av mätningar är stor. Ungefär en procent av all producerad metan visade sig gå förlorad till atmosfären. Nästa steg är att ta fram en handbok som kan hjälpa användaren att jämföra och välja mellan olika mätmetoder.

#### Another step forward in Swedish energy research

Energiforsk – Swedish Energy Research Centre is a research and knowledge based organization that brings together large parts of Swedish research and development on energy. The goal is to increase the efficiency and implementation of scientific results to meet future challenges in the energy sector. We work in a number of research areas such as hydropower, energy gases and liquid automotive fuels, fuel based combined heat and power generation, and energy management in the forest industry. Our mission also includes the generation of knowledge about resource-efficient sourcing of energy in an overall perspective, via its transformation and transmission to its end-use. Read more: www.energiforsk.se

