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(EvEmBi)

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# Evaluation and reduction of methane emissions from different European biogas plant concepts (EvEmBi)

Results of the Austrian Participation



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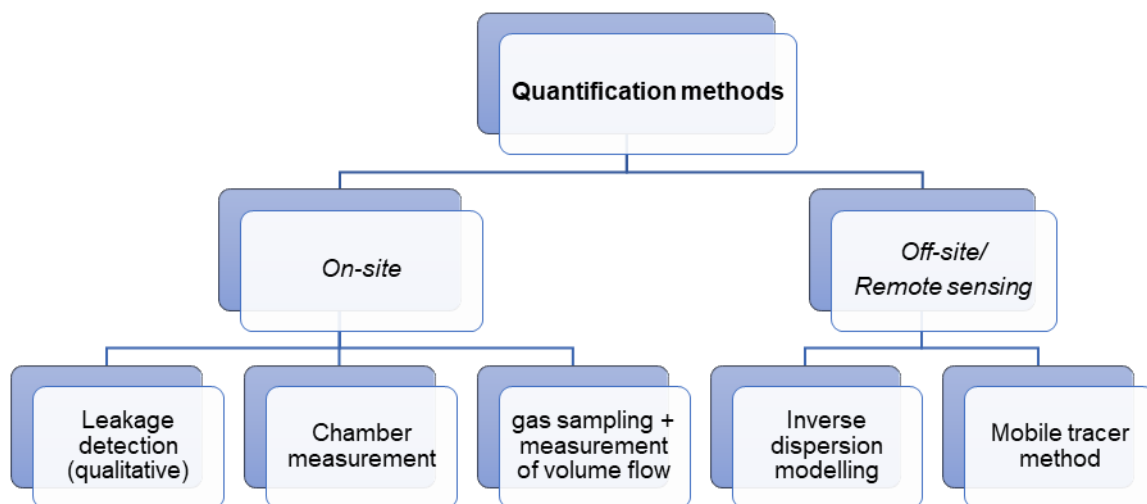
## 2 Introduction

Biogas or biomethane, resulting from the biological treatment of organic matter by anaerobic digestion, is a renewable energy source used for electricity production, heating and in transportation and can substitute natural gas. Therefore, biogas production is described as a sustainable strategy for reducing anthropogenic greenhouse gases (GHG). However, the positive environmental impact depends in particular on emissions that might occur within the biogas production and utilization chain. At biogas plants, GHG emissions can arise e.g. from open substrate and digestate storage as well as during upgrading of biogas to biomethane and combustion in combined heat and power (CHP) plants, respectively. Although the number of studies investigating the contribution of anaerobic digestion facilities to GHG emissions has increased during the last decade, there is still a lack of representative emission data.

As stated in the United Nations Framework Convention on Climate Change, the member countries are obliged to report their national GHG inventories according to the Intergovernmental Panel on Climate Change (IPCC) guidelines. This also requires high quality and comparable emission factor (EF) estimates from full-scale biogas plants treating diverse substrates. The current EFs used in IPCC are based on a limited amount of studies which result in a broad range of values and thus in a low accuracy of the EFs proposed. Currently, the Austrian National Inventory Report (Anderl et al., 2020) complains about the lack of national studies estimating methane (CH<sub>4</sub>) leakage losses from Austrian biogas plants. Anyhow, CH<sub>4</sub> emissions from anaerobic digesters were included in the report assuming a CH<sub>4</sub> loss of 2 % of the gas produced based on a study worked out in Germany before 2010 (FNR 2010).

In order to evaluate mitigation strategies as well as to improve the database for national emission inventories, accurate and comparable methods quantifying GHG emissions (especially methane emissions) from biogas plants are required. In recent years, several approaches have been established that determine individual emission sources as well as overall emissions from biogas plants using on-site and ground-based remote sensing (off-site) methods (see Figure 1). The on-site approach involves a leakage detection (e.g. using an optical gas imager [OGI] and/or a hand-held methane analyzer) and the quantification of methane emissions from different single sources (e.g. leaks, exhaust pipes, digestate storage) using several sub-methods. For channeled sources (e.g. off-gas from gas utilization units, encapsulated emission sources connected to a biofilter, encapsulated emission sources with forced ventilation) the methane emission flux is calculated based on measured CH<sub>4</sub> concentration and volume flow within the pipes. For area sources like open digestate storage tanks, dynamic or static chambers are usually used to determine emissions from multiple small area parts of the source, and the total emission is then calculated via extrapolation to the whole surface area. For leakages, dynamic chambers are usually applied to quantify the emission rate. Ground-based remote sensing approaches also include a number of different methods (e.g. inverse dispersion modelling [IDM], tracer gas dispersion method [TDM]) all aiming for quantification of overall or component plant emissions by sampling atmospheric methane concentrations at points upwind and downwind of the emissions source. With IDM, the integrated methane concentration on an open path is measured, e.g. by using an open-path tunable diode laser absorption spectrometer (OP-TDLAS). Together with data of an ultrasonic anemometer (USA) and of a temperature and pressure sensor, overall emissions from the biogas plant can be back-

calculated via an atmospheric dispersion model. TDM combines a controlled tracer gas release, e.g. acetylene, within the area of the plant with time-resolved measurements of downwind concentrations of both tracer and methane using a mobile analytical platform (e.g. vehicle carrying high resolution analytical instruments for gas detection). In contrast to direct on-site methods, remote sensing approaches are able to monitor time-independent and/or operational emissions over a longer period. However, they depend on transport processes in the atmosphere and are restricted by certain weather conditions and the topography or infrastructure of the surrounding area.



**Figure 1: Overview quantification methods to determine methane emissions from biogas plants.**

The main objective of the research project “EvEmBi” was to evaluate various biogas plant concepts (e.g. agricultural- and bio-waste- or waste water treatment biogas plants) used in practice with respect to their methane EFs. Using a previously developed measurement guideline for the harmonized implementation and evaluation of different measurement methods (project "European harmonization of methods to quantify methane emissions from biogas plants - MetHarmo", ERA-NET Bioenergy Program 2016-2018) (Clauß et al., 2019), comparable and representative methane EFs from a wide range of biogas plant technologies were determined for the European GHG inventory for the first time using both on-site and ground-based remote sensing methods. Between 2018 and 2021, 36 biogas plants in Austria, Germany, Sweden and Switzerland were investigated by eight measurement teams. Following the determination of EFs, emission reduction strategies were developed, implemented and evaluated based on a cost-benefit analysis for particular biogas plants. Findings were summarized in a general European position paper (EBA 2020a) as well as national (country specific) position papers on GHG emissions and abatement strategies (e.g. FvB 2020, KBVÖ 2020a). In addition, national voluntary systems for emission control of the biogas sector were developed (e.g. KBVÖ 2020b), based on elaborated minimum requirements (EBA 2020b).

The present report focuses on the Austrian project participation, which investigated 13 biogas plants using both on-site methods and IDM (sections 4.1 and 4.2). In section 3.4 a cost-benefit-analysis of identified mitigation measures is described, while section 5 presents the European and Austrian

voluntary system for emission control. The project results of the entire transnational consortium are summarized in section 6.

## 3 Material and Methods

### 3.1 Investigated biogas plants and measurement campaigns

Within two measurement series between 2018 and 2021, in total 13 biogas plants were investigated in Austria. An overview of the plant characteristics is given in Table 1.

**Table 1: Characteristics of the investigated Austrian biogas plants. Number of biogas plants are in brackets ().**

<b>Feed stock:</b>	Bio-waste <sup>1</sup> (6) Bio-waste <sup>1</sup> , manure (2) Energy crops, manure (5)
<b>Biogas utilization:</b>	CHP <sup>2</sup> (8) CHP <sup>2</sup> and BUU (2) BUU (3), with exhaust gas treatment (2)
<b>Digestate storage:</b>	Closed, gastight storage (9) Open/not-gastight storage (4)

BUU=biogas upgrading unit; CHP=combined heat and power; PSA=pressure swing adsorption;

<sup>1</sup> Bio-waste comprises household waste (bio-waste), food waste from supermarkets, restaurants and/or waste from food/beverage industry.

<sup>2</sup> Electrical capacity of CHP units: 250-830 kW<sub>el</sub>.

Within the first measurement series, individual plant components were investigated using on-site methods (see section 3.2). In addition, full-scale plant emissions were measured at five facilities, where the surrounding area met requirements for IDM measurements (see section 3.3). In total, measurements were performed over 1-3 days depending on the plant size and number of emission sources. Repeated measurements (second measurement series) were conducted after mitigation measures were implemented at individual plants.

Methane EF (methane losses in % of produced methane) were calculated by relating methane emission rates (kg h<sup>-1</sup>, STP) to the produced methane in the digester(s). For the latter, the average methane production of a reference year or the daily production on the measurement day was used, depending on the available data (information from plant operators). In case of biogas utilization units (methane slip), measurement results were related to the methane input to individual units, calculated based on operational data.

## 3.2 On-site methods

The on-site approach consists of two main steps to investigate methane losses from biogas plants: (1) leakage survey at biogas-bearing plant components and (2) quantification of emissions from individual sources. The used quantification methods as well as the applied measurement equipment are described in detail in Clauß et al, (2019), Liebetrau et al. (2017) and Reinelt et al. (2017).

### 3.2.1 Leakage detection and quantification

For leakage detection, an optical gas imaging infrared (IR) video camera (GF 320, Co. FLIR, Wilsonville, USA), a portable methane laser (LaserMethane mini gen2, Co. GROWCON, Abingdon, UK) as well as a portable flame ionization detector (FID, TVA1000B, Thermo Fisher Scientific Inc., Franklin, USA) were used to screen exposed biogas bearing plant components. A leakage was defined as (1) a methane release caused by technical or human failure and (2) an individual spot exceeding measured methane concentration of 0.1 vol.% CH<sub>4</sub>. In case of forced ventilation (air-inflated double membrane dome), a categorization was done after the determination of the emission mass flow (see section 3.2.3). When foil permeability exceeded 0.5 L CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> bar<sup>-1</sup>, methane release was classified as leak.

For determining the extend of methane release from individual leakages, the dynamic chamber method was used based on a foil encapsulation. A constant flow of outside air was maintained through the head space of the chamber using a connected blower at the outlet (D060, Elektror airsystems GmbH, Tumeltsham, Austria) and the difference in concentration between inlet and outlet was measured. For each leakage, two different volume flows were applied to ensure that the emission source was not influenced by suction. Gas in the in- and outlet of the encapsulated point of measurement were sampled discontinuously via evacuated 20 ml glass vials and analyzed in the laboratory by a gas chromatograph (GC) equipped with a flame ionization detector (Agilent 7890A/7897A, Agilent Technologies, Santa Clara, USA) according to EN ISO 25139. The air flow was determined by two sensors (hot wire sensor: 06351032, Testo GmbH, Vienna, Austria; vane anemometer: 06359532, Testo GmbH, Vienna, Austria) according to ISO 16911-1:2013 and EN 15259:2007. Methane emissions were determined according to Equation (1):

$$E_{CH_4} = Q_{air} \cdot \rho_{CH_4} \cdot (c_{CH_4,out} - c_{CH_4,in}) \quad (1)$$

where  $E_{CH_4}$  is the CH<sub>4</sub> emission mass flow (mg h<sup>-1</sup>),  $Q_{air}$  is the volume air flow (m<sup>3</sup><sub>air</sub> h<sup>-1</sup> STP, dry),  $\rho_{CH_4}$  is the gas density of CH<sub>4</sub> (mg ml<sup>-1</sup>) and  $C_{CH_4,out}$  and  $C_{CH_4,in}$  (ppmv) are exhaust and background CH<sub>4</sub> concentrations, respectively. Due to the high effort related to this method (encapsulation and measurements), only major leakages were quantified.

### 3.2.2 Emission quantification of channeled sources

Methane emissions from channeled sources such as CHP units, BUU and exhaust air of substrate receiving halls after biofilters were quantified by measuring methane concentrations in the exhaust pipes according to EN ISO 25139 (discontinuous gas sampling and GC analysis see section 3.2.1). In case of biowaste treating facilities with exhaust air from substrate receiving halls, volume flow was directly

measured in the exhaust pipe before or after a biofilter, depending on an available measurement point (EN 15259:2007). Methane emissions were determined according to Equation (2):

$$E_{CH_4} = c_{CH_4} \cdot \rho_{CH_4} \cdot Q_{ex} \tag{2}$$

where  $E_{CH_4}$  is the  $CH_4$  emission mass flow ( $mg\ h^{-1}$ ),  $Q_{ex}$  is the volume exhaust flow ( $m^3\ h^{-1}$  STP, dry),  $\rho_{CH_4}$  is the gas density of  $CH_4$  ( $mg\ ml^{-1}$ ) and  $C_{CH_4}$  (ppmv) are exhaust  $CH_4$  concentrations.

In case of CHP units and BUU, volume flow could not be measured but was calculated by means of operational data. While data on the raw gas input and the product gas (biomethane) output was used for BUU units, the off-gas volume flow from CHP units was calculated based on the electrical power, the raw gas input and the (calculated) combustion-air ratio ( $\Lambda$ ) during measurements (see for more details Clauß et al., 2019).

### 3.2.3 Emission quantification of air-inflated double membrane domes

In case of methane diffusion of double membrane domes, methane emissions were quantified by measuring the methane concentration in the outgoing air from the air inflated outer layer (discontinuous sampling and GC analyses see section 3.2.1) as well as the air volume flow (emission quantification see equation 2). In order to meet requirements for volume flow measurements (EN 15259:2007), the inflation-air outlet was extended using an additional tube.

### 3.2.4 Emission quantification of area sources

The emission rate quantification from area sources such as open/non-gastight substrate and digestate storage tanks was carried out by either a floating static chamber ( $0.19\ m^2$ ,  $0.06\ m^3$ ) or the air-inflation method (Sneath et al., 2006), depending on the accessibility of the surface area. In case of the static chamber, the surface emissions ( $E_{CH_4}$  in  $mg\ CH_4\ m^2\ h^{-1}$ ) were determined by multiplying the linear increase (slope) of the headspace methane concentration inside the chamber ( $\partial C_{CH_4} / \partial t$  in  $mg\ CH_4\ m^3\ h^{-1}$ ; discontinuous gas sampling at defined time intervals [1 min]) and the volume to area ratio of the chamber ( $V_{ch}/A_{ch}$  in  $m^3\ m^{-2}$ ):

$$E_{CH_4} = \frac{\partial c_{CH_4}}{\partial t} \cdot \frac{V_{ch}}{A_{ch}} \tag{3}$$

Surface emissions were quantified by placing the chamber on 6 measurement points on the liquid digestate surface and results were extrapolated to the whole surface area.

The air-injection method was applied at non-gastight covered tanks, where the surface area was not accessible for chamber measurements. This method involved an aeration of the headspace of a tank by a blower (D060, Elektror airsystems GmbH, Tumeltsham, Austria) connected to the pressure side until a constant methane concentration was reached. The methane concentration in the headspace (start and end value) was measured via a sampling line by continuous analytical methods (FID). The volume flow was determined on the input side. The emission rate was calculated according to Equation (4):

$$E_{CH_4} = Q_{air} \cdot \rho_{CH_4} \cdot \frac{c_i}{1 - \frac{c_i}{c_0}} \tag{4}$$

where  $E_{\text{CH}_4}$  is the  $\text{CH}_4$  emission mass flow ( $\text{mg h}^{-1}$ ),  $Q_{\text{air}}$  is the volume air flow ( $\text{m}^3 \text{h}^{-1}$  STP, dry),  $\rho_{\text{CH}_4}$  is the gas density of  $\text{CH}_4$  ( $\text{mg ml}^{-1}$ ) and  $C_0$  and  $C_i$  (ppmv) are  $\text{CH}_4$  concentrations at the beginning and end of the measurement.

## 3.3 Remote sensing method – Inverse dispersion modelling

Inverse dispersion modelling (IDM) means to derive methane emission rates from measured concentrations at points upwind and downwind from the biogas plant combined with meteorological data using a dispersion model (for more detailed information see Clauß et al., 2019).

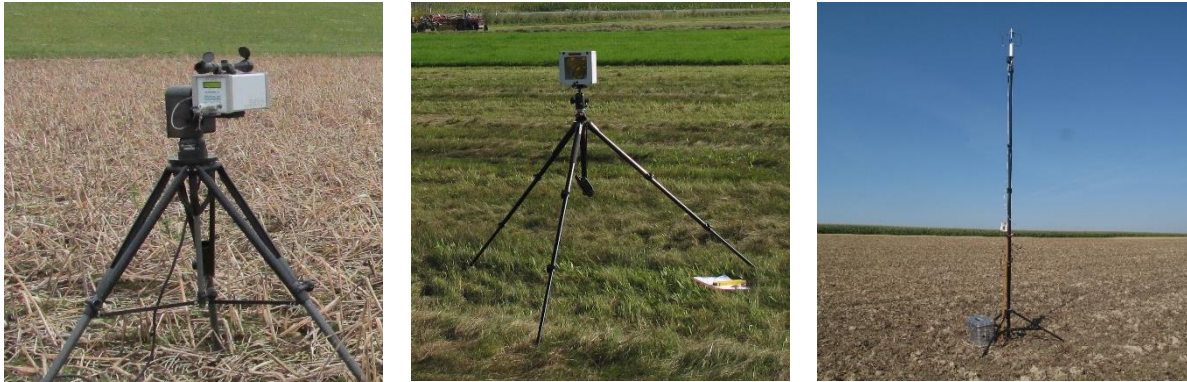
### 3.3.1 Measurement instrumentation

An OP-TDLAS system (GasFinder 2.0, GasViewMP Version 0.22, Boreal Laser Inc., Edmonton, Canada) and 1-2 distant reflectors were used to measure path-integrated  $\text{CH}_4$  concentrations downwind and upwind of the plants (see Figure 2). Using a digital scanning motor (Model PTU D300, Directed Perception Inc., CA, USA), the OP-TDLAS was automatically aligned to record path-integrated  $\text{CH}_4$  concentrations. The laser and reflectors were mounted on tripods at a height of 1.30-1.70 m and positioned 100-350 m apart. Depending on the dominant wind obstacles, the surroundings as well as the wind conditions at the respective plant, the laser paths were placed in distances of 50-100 m from the potential emission area meeting the placement criteria by Flesch et al. (2005, 2011) (at least 10 times the height of the highest obstacle). Background concentration upwind of biogas plants was measured prior to and after the measurement campaign and was assumed to be constant for the entire measurement period.

In order to ensure good quality data, the measured  $\text{CH}_4$  concentration was checked to have a return light level between 4,000 and 12,000 (unitless metric) and a coefficient of determination ( $R^2$ ) of the laser signal greater than 98 %.

Raw wind components ( $u$ ,  $v$ ,  $w$ ) and the sonic temperature were collected with a portable 3D ultrasonic anemometer (Model 81000, R.M. Young Company, Michigan, USA) (see Figure 2) at a frequency of 10 Hz and a height of 5 m above the soil surface. Raw averages were calculated (10-min intervals) using relations of the mean product of  $u$ ,  $v$ ,  $w$  and temperature ( $\langle u*u \rangle$ ,  $\langle u*v \rangle$ ,  $\langle u*w \rangle$ ,  $\langle v*v \rangle$ ,  $\langle v*w \rangle$ ,  $\langle w*w \rangle$ ,  $\langle u*T \rangle$ ,  $\langle v*T \rangle$ ,  $\langle w*T \rangle$ ,  $\langle T*T \rangle$ ) as an input for the dispersion model. Air temperature, moisture and barometric pressure were measured (every 30 seconds) using a datalogger (EASYlog 80 CL).  $\text{CH}_4$  concentrations and meteorological data were prepared in a time series of 10-min average.

A global positioning system was used to mark the 3D ultrasonic anemometer as well as the laser paths around the biogas plants.



**Figure 2: Measurement instrumentation for the inverse dispersion modelling method: OP-TDLAS (left), reflector (middle) and 3D ultrasonic anemometer (right).**

### 3.3.2 Inverse dispersion modelling - quantification of methane emission rates

The backward-time Lagrangian stochastic model (bLs) was applied to infer the CH<sub>4</sub> emission rates of whole plant emissions from the biogas plants based on the measured (upwind and downwind) CH<sub>4</sub> concentrations and meteorological data. The bLs model is implemented by the software product WindTrax (Thunderbeach Scientific, version 2.0.8.9), which assumes idealized wind conditions appropriate for simple undisturbed terrain (using Monin–Obukhov similarity theory) (Flesch et al., 1995).

The emission rate  $E_{CH_4}$  was calculated from the measured gas concentration  $C_{CH_4}$  (background compensated  $C_{BG}$ ) and the dispersion model prediction of the ratio of concentration at the gas sensor to the emission rate  $(C/Q)_{sim}$  according to the following equation (5) (Flesch et al., 2005b):

$$E_{CH_4} = \frac{(C_{CH_4} - C_{BG})}{(C/Q)_{sim}} \quad (5)$$

WindTrax simulates turbulence dispersion by modelling the random movement of thousands of particles backward in time as they travel upwind from the concentration sensor (measurement path) being displaced by horizontal and vertical aerodynamic forces. The particles are released from each point spaced evenly along the path length of the sensor. The simulated ratio of concentration at the sensor to the emission rate  $(C/Q)_{sim}$  is calculated from the number of points along the measurement path ( $P$ ), the total number of gas particles released at the measurement site ( $N$ ) and the modelled vertical velocity at “touchdown”, summed across all instances where a particle impacts (touch-downs) the ground within the emission source area ( $w_0$ , m s<sup>-1</sup>) (Flesch et al., 2004, 2005b).

$$(C/Q)_{sim} = \frac{1}{P} \sum_{i=1}^P \left( \frac{1}{N} \sum \left| \frac{2}{w_0} \right| \right) \quad (6)$$

Each of the 10-min-averaged measurements are used as input data to back-calculate the CH<sub>4</sub> emission using 50,000 particle projections and 30 points along the measurement path. Since the edge of the emission plume can give a poor estimate of the overall emission, periods where the laser path touchdown field covered less than 60% of the biogas plant area were removed.

Inputs to the dispersion model were the geometry of the source(s), the location and height of the laser path and the 3D ultrasonic anemometer as well as the anemometer data (expressed as relations of the mean products of  $u$ ,  $v$ ,  $w$  and temperature). The whole biogas plant area was defined as a spatially homogenous single area source.

The inverse dispersion technique to estimate the source strength depends on a good description of atmospheric transport, which is known to be difficult in extreme stability conditions and/or low wind speeds. The criteria of Flesch et al. (2004; 2007) were used to exclude observation periods that might provide inaccurate emission calculations. Only the measurement periods that met the requirements were used to determine emission rates:  $|L| \geq 10$  (strongly stable/unstable atmosphere) and  $u^* > 0.15 \text{ m s}^{-1}$  (low wind conditions).

## 3.4 Cost-benefit analysis

Implemented emission mitigation measures were evaluated by cost-benefit analysis (CBA). For each measure, net present costs (discounted investment and operational costs) were compared to the net present benefit. For the latter, it was distinguished between benefit A and B. Benefit A represents the benefit for the plant operator when avoiding methane losses (business economic repair investment). Benefit B (environmental economic repair investment) additionally includes benefits through internalizing the external costs of methane emissions by emission trading. Subtracting the (discounted) costs of the measure from benefit A and B, respectively, results in the net present values (NPV) A and B.

The calculations were based on the following assumptions:

- Economic lifetime: 10 years
- Interest rate: 2 %
- System boundary: gate-to-gate
- $\text{CO}_2$  equivalents ( $\text{CO}_{2\text{eq}}$ ) of  $\text{CH}_4$ : 28
- Emission trading course of  $40 \text{ € t}^{-1} \text{ CO}_{2\text{eq}}$  (Ember, 2021)

In case both calculated NPV are positive (NPV A, B > 0), the mitigation measure is evaluated as cost-effective (+++). In contrast, if only NPV B is > 0, cost-effectiveness can only be achieved by internalizing the external costs of methane losses (++) . Eventually, a mitigation measure is evaluated with (+), when methane emissions can be reduced but both NPV are negative (NPV A, B < 0).

## 4 Results and Discussion

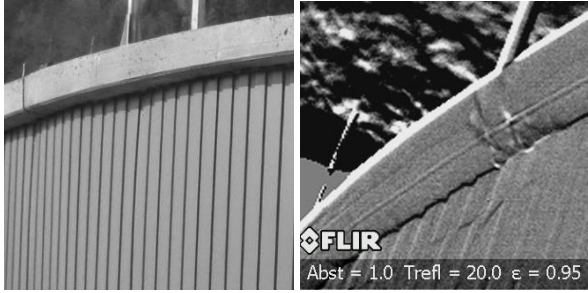
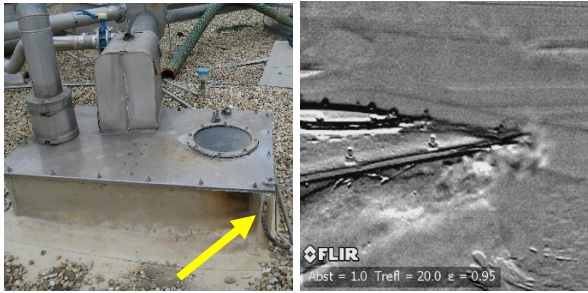
### 4.1 On-site measurements

#### 4.1.1 Leakage detection

Leak detection was performed at 12 biogas plants. In total, 22 concrete roof digesters, 10 single membrane domes and 9 air-inflated double membrane domes were investigated. Individual leakages are listed in Table 2-Table 4. Figure 3 shows the number of leaks according to digester technology and location. As described in section 3.2.1, not all of the detected leakages were quantified. Methane losses are shown in section 4.1.2.



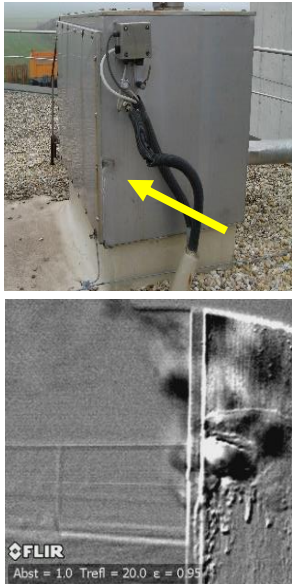

At concrete roof digesters, leakages were found at 27 % of the investigated tanks. Half of the leaks were caused by cracks in the concrete wall (of two digesters at one biogas plant), the other half by leaking service boxes and an overflow. Methane losses due to cracks in the concrete were detected between digester wall and roof. Five major leaks (> 0.1 vol% CH<sub>4</sub>) and several smaller ones were found (see Table 2). In addition, four leaks were detected at service boxes on top of concrete roof digesters serving as agitator, pipe leadthrough (substrate/biogas) and/or pressure relief valve. Methane was released due to leaking gaskets and damages in the material, respectively (see Table 2).

Table 2: Overview of detected leakages at concrete roof digesters.

Component	Leak number	Description	Maximum CH <sub>4</sub> concentration [vol%]	Picture/ picture OGI camera
Concrete wall/roof (n=5)	L1-L5	Leaks between wall and roof of two digesters due to cracks in the concrete wall	0.15 %	
Pipe leadthrough (n=2)	L6	Leaking gasket	40 %	

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	L7	Leaking gasket	2.8 %	
<b>Pressure relief valve (n=1)</b>	L8	Leaking gasket	0.5 %	
<b>Agitator (n=1)</b>	L9	Damaged surface	10 %	
<b>Overflow (n=1)</b>	L10	Ventilation settings	0.2 %	

n=number of detected leaks

At the 10 investigated single membrane domes, leakages were detected at only one tank at the wires to adjust the submerged agitators (see Table 3).


**Table 3: Overview of detected leakages at single membrane domes.**

Component	Leak number	Description	Maximum CH <sub>4</sub> concentration [vol%]	Picture
Wire to adjust agitator (n=2)	L18-L19	Frost damage	L18: 0.2 % L19: 0.4 %	

n=number of detected leaks





Leaks were found at more than half of the air-inflated double membrane domes (56 %). At two digesters, increased methane concentrations were measured beneath the membrane fixation (L11-L12, see Table 4). It was assumed that cracks in the concrete caused the loss of biogas. However, the location of the leaks could not be identified due to the isolation of the digester wall. Other leaks were detected at the flange connection of one pressure relief valve, at a wire for the adjustment of a submerged agitator (due to insufficient lubrication) and at a bull's eye (due to a leaking gasket). At two domes, the inner membrane showed increased foil permeability ( $>0.5 \text{ L CH}_4 \text{ m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$ ), determined by quantification measurements (see section 4.1.2). At one of these membranes, the increased methane release ( $110.6 \text{ L CH}_4 \text{ m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$ ) could be attributed to a fist-sized hole that was detected after the foil had been replaced.

**Table 4: Overview of detected leakages at double membrane domes.**

Component	Leak number	Description	Maximum CH <sub>4</sub> concentration [vol%]	Picture
Concrete wall (n=2)	L11-L12	Assumed cause: damage in inner foil and/or crack in concrete wall	L11: 0.9 % L12: 1.5 %	

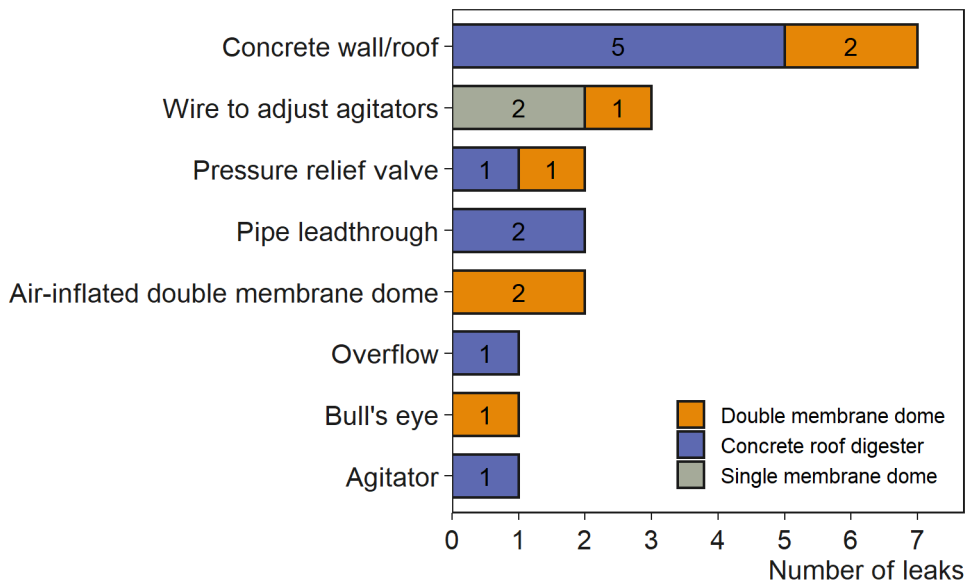
# Energieforschungsprogramm - 11. Ausschreibung

Klima- und Energiefonds des Bundes – Abwicklung durch die Österreichische Forschungsförderungsgesellschaft FFG

<b>Air-inflated double membrane dome (n=2)</b>	L13	Leaking membrane; fatigue	inner material	0.1 %	
	L14	Increased permeability	foil	0.003 % <sup>1</sup>	
<b>Wire to adjust agitator (n=1)</b>	L15	Insufficient lubrication		3 %	
<b>Bull's eye (n=1)</b>	L16	Leaking gasket; construction deficit		0.15 %	
<b>Pressure relief valve (n=1)</b>	L17	Leaking connection	flange	2.4 %	

n=number of detected leaks

<sup>1</sup> leak classification after quantification measurements



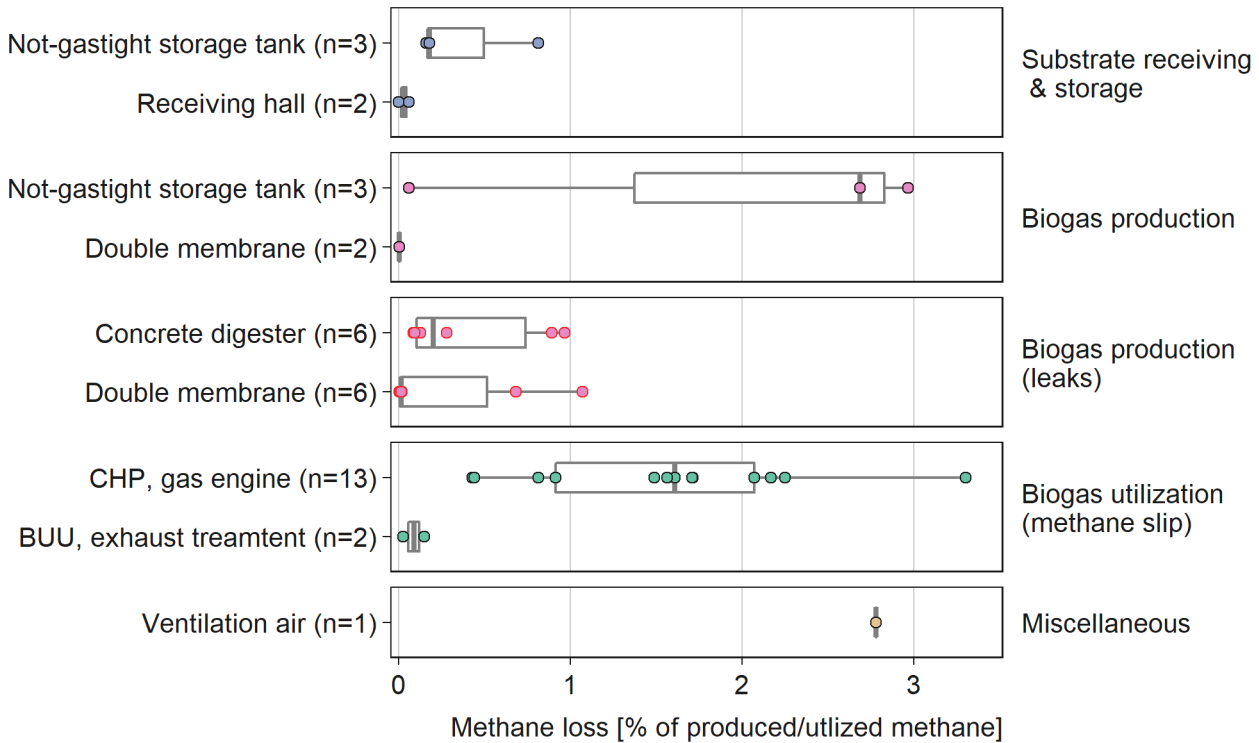
**Figure 3: Number of leakages according to technology and location.**

The detected leakage hotspots match with those from previous studies. Most of the leakages were caused by construction defects (i.e. badly manufactured pipe leadthroughs), poor maintenance (i.e. insufficient lubrication, leaking gaskets and flanges) or material fatigue (Schreier, 2011, Sax et al., 2013, Liebetrau et al., 2013, Westerkamp et al., 2014, Clemens et al., 2014, Reinelt et al., 2017).

## 4.1.2 Methane emissions of component sources

Figure 4 shows an overview of component specific EF within different process stages (substrate receiving and storage, biogas production, biogas utilization). Major emissions were caused by the off-gas of CHP units (0.4-3.3 % of utilized methane) and open/not gastight digestate storage tanks (2.7 and 3.0 % of produced methane). Additional methane loss arose from several substrate storage tanks (0.2-0.8 %) as well as leakages (0.0-1.1 %). At one plant, high methane emissions (2.8 %) were determined in the exhaust air of bio-filters, treating ventilation air from a substrate pretreatment hall as well as from a buffer and sedimentation tank. However, as measurements covered only short time periods, results of substrate receiving halls, storage tanks and digestate storage tanks give limited information on average emissions.

In the following sections (4.1.2.2 and 4.1.2.3), individual process stages and emission sources are addressed in detail.



**Figure 4: Overview of component specific emission factors (EF) within different process stages. For the methane slip of biogas utilization units, methane emissions are related to the amount of utilized methane, otherwise to the methane production in the digester(s). BUU=biogas upgrading unit. CHP= combined heat and power plant.**

### 4.1.2.1 Substrate receiving and storage

Within the substrate receiving and storage stage, investigated emission sources include not-gastight substrate storage tanks as well as the ventilation air of substrate receiving and pretreatment halls. Especially at bio-waste treating facilities, the ventilation air of receiving halls is treated by bio-filters for odour reduction. Methane emission rates and factors are shown in Table 5.

**Table 5: Methane emission rates [g h<sup>-1</sup>] and methane emission factors (EF) [% of produced methane] from substrate receiving and storage. Standard deviations are given in brackets ().**

Plant ID	Emission source	Substrate	CH <sub>4</sub> emission rate [g h <sup>-1</sup> ]	CH <sub>4</sub> EF [% CH <sub>4</sub> produced]
1	Not-gastight substrate storage	manure. bio-waste	630 (197)	0.8 (0.3)
6	Not-gastight substrate storage	process water (digestate)	138 (9)	0.2 (0)
2	Not-gastight substrate storage	bio-waste	232 (37)	0.2 (0)
6	Ventilation (substrate receiving hall) <sup>1</sup> air	bio-waste	44 (9)	0.1 (0)
3	Ventilation (substrate receiving hall) <sup>1</sup> air	bio-waste	1 (0)	0.0 (0)

<sup>1</sup> receiving and processing hall

<sup>2</sup> Measurements were performed in the inlet to two out of four bio-filters.

Highest emissions (0.8 % of produced methane) were released from a substrate tank in which manure and bio-waste were stored together. However, at two other biogas plants, storage tanks containing bio-waste and liquid digestate (process water) emitted less methane (0.2 %). At three plants not-gastight substrate storage tanks were not included in the quantification measurements as methane concentrations were low during leak detection ( $<0.1 \text{ vol\% CH}_4$ ). Methane emissions from ventilation were low ( $\leq 0.1 \text{ %}$  of produced methane). It should be noted that results give limited information on average emissions, as emissions can vary depending on the mode of operation (delivery, pretreatment). In addition, part of the emission plume can escape through open doors.

Previous studies revealed similar results. For receiving halls, Reinelt et al. (2017) observed almost negligible emissions of  $< 0.1 \text{ %}$  (of produced methane) at a bio-waste treating facility, Fredenslund et al. (2018) determined 0.2 % at a manure-based biogas plant. For substrate storage and mixing tanks, methane losses ranged from  $<0.1$  to 3.0 % (Sax et al., 2013; Liebetrau et al., 2013; Fredenslund et al., 2018). Various factors as agitation, the filling level and the type of substrate have shown to affect emissions from substrate tanks (Hafermann et al., 2009, Liebetrau et al., 2013, Sax et al., 2013, Schories et al., 2018). During agitation, Sax et al. (2013) observed an increase in emissions from 0.4 to 3.0 %. By far the highest emissions were recorded when substrate was hydrolyzed before mixing with digestate (pH-levels nearly neutral from 6.3 to 7.1), ranging from 2.4 up to 10.8 % (Schories et al., 2018).

## 4.1.2.2 Biogas production

### 4.1.2.2.1 Methane emissions from foil roofs

In this study, foil roof emissions by membrane diffusion were measured at the inflation air outlet of air-inflated double membrane domes. Except for one digester, methane emissions were very low ( $\leq 0.01 \text{ %}$  of the produced methane), see Table 6. However, in addition to a leaking foil causing a methane loss of 0.68 %, also a methane release of 0.01 % was classified as leak as foil permeability exceeded the set threshold value of  $0.5 \text{ L CH}_4 \text{ m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$  (see section 3.2.1).

**Table 6: Methane emission rates [ $\text{g h}^{-1}$ ], methane emission factors (EF) [% of produced methane] and foil permeability [ $\text{L CH}_4 \text{ m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$ ] of air-inflated double membrane domes. Standard deviations are given in brackets ().**

Plant ID	CH <sub>4</sub> emission rate [ $\text{g h}^{-1}$ ]	CH <sub>4</sub> EF [% CH <sub>4</sub> produced]	Foil permeability [ $\text{L CH}_4 \text{ m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$ ]
11-2	609.4 (57.5)	0.68 (0)	110.6
4-2	1.5 (0.4)	0.01 (0)	0.8
11-1	2.1 (0.7)	0.00 (0)	0.3
4-1	14.8 (1.6)	0.00 (0)	0.2

In Clemens et al. (2014), 63 % of the investigated 202 double membrane domes exceeded a foil permeability of  $1 \text{ L CH}_4 \text{ m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$ , 29 % released more than  $3 \text{ L CH}_4 \text{ m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$ . In contrast, only three foils of 35 single membrane domes showed a permeability of  $> 1 \text{ L CH}_4 \text{ m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$  and one emitted  $> 3 \text{ L CH}_4 \text{ m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$ . Liebetrau et al. (2013) investigated nine double membrane domes and

detected occasionally high methane concentrations in the inflation air. Methane losses ranged up to 0.02% (median <0.01%).

#### 4.1.2.2.2 Methane emissions from open and not-gastight digestate storage

Measurements were performed at three bio-waste treating plants in winter/spring season (Feb. 2019, Mar. 2019/21). While two of the digestate storage tanks emitted up to 3.0 % of the produced methane, emissions were relatively low at plant no. 2 (0.1 % methane loss), see Table 7. However, the low emissions of this tank can be explained by the low air temperature and storage volume as well as by an upstream solid-liquid separation. In addition, at plant no. 2 most of the digestate is stored in another upstream storage tank. Due to problems during measurements, results of the main digestate storage tank are not shown.

**Table 7: Methane emission rates [g h<sup>-1</sup>] and methane emission factors (EF) [% of produced methane] from open and not-gastight digestate storage. Standard deviations are given in brackets ().**

Plant ID	Feedstock	Air temperature [°C]	Digestate temperature <sup>1</sup> [°C]	Maximum storage volume [m <sup>3</sup> ]	Filling level [%]	Surface area <sup>2</sup> [m <sup>2</sup> ]	CH <sub>4</sub> emission rate [g h <sup>-1</sup> ]	CH <sub>4</sub> EF [% CH <sub>4</sub> produced]
1	bio-waste, manure	0-10	n.m.	1780	55	-	1988 -	3.0 -
6	bio-waste	12	14	3600	97	616	2077 (103)	2.7 (0.1)
2-2	bio-waste	5	n.m.	-	10	-	87 (6)	0.1 (0)

n.m.=not measured

<sup>1</sup> Temperature was measured 1.7 m below digestate surface.

<sup>2</sup> The investigated digestate storage tanks had a liquid surface layer (without a crust).

So far, the most extensive studies on digestate storage tanks were performed by Baldé et al. (2016) and Maldaner et al. (2018). Both investigated a farm-scale digestate storage tank in Canada over one year. By performing IDM measurements Baldé et al. (2016) determined annual cumulated methane emissions of 12 % of produced methane. In contrast, Maldaner et al. (2018) quantified four times lower average annual emissions, scaled by volatile solid (VS) content, using a micrometeorological mass balance method. Baldé et al. (2016) explained the high emissions with the long retention time during summer months (high temperature and storage volume) but also with the high volume of the permanent sludge layer (high VS content) and the lack of solid-liquid separation. In contrast, the AD plant investigated by Maldaner et al. (2018) had a solid-liquid separation in place and the sludge layer was removed before starting the measurements. In addition, the hydraulic retention time (HRT) was longer at this plant and a greater fraction of off-farm materials was digested together with manure. It was, therefore, concluded that substrate characteristics, process parameters (HRT, solid-liquid separation) and storage conditions (residual sludge, temperature, retention time) have a major impact on methane formation during digestate storage. The effect of solid-liquid separation was also confirmed by other studies (Amon et al., 2006, Gioelli et al., 2011, VanderZaag et al., 2018). A wide range of emissions was measured by Liebetrau et al. (2013) investigating 7 agricultural biogas plants (based on energy crops and co-digestion of energy crops and manure). Methane losses ranged between 0.22 to 11.22 % quantified by on-site

methods over short time periods. Relatively low emissions were determined by intermittent on-site measurements at an energy crop-based AD plant in Germany, covering different seasons (Westerkamp et al., 2014). The sum of methane emissions from multiple tanks ranged between <0.1 and 0.23 %.

Vergote et al. (2020) conducted continuous on-site measurements (refinement of closed chamber method) over three months at a farm-scale AD plant in Belgium (mono-digestion of cattle manure). Methane losses were quite high with daily average emissions between 3.9 to 8.2 %. However, the measurement period reflected worst case conditions with high digestate temperature and high storage volume. Reinelt et al. (2017) investigated a bio-waste treating facility comparing different measurement methods. Methane emissions from digestate storage ranged between 0.53 and 0.89 %, depending on the used method.

One major factor influencing methane emissions from individual biogas plants has shown to be the digestate temperature which can be considerably higher than the actual air temperature. The positive correlation of temperature and methane emissions was confirmed by lab-scale and pilot-scale studies as well as by actual measurements (Clemens et al., 2006, Rodhe et al., 2015, Westerkamp et al., 2014, Hrad et al., 2014, Baldé et al., 2016, Maldaner et al., 2018, Vergote et al., 2020). In addition, short-term increases in methane emissions were observed during agitation of the digestate and due to other disturbances, such as increased wind speeds and precipitation (Hrad et al., 2015, Groth et al., 2015, Baldé et al., 2016).

#### 4.1.2.2.3 Methane emissions from leakages

Methane losses by leakages ranged between <0.01 and 1.1 % of produced methane (see Table 8). Highest emissions were measured beneath the membrane fixation of an air-inflated double membrane dome, assumed to be caused by a crack in the concrete wall. However, the emissions of approx. 3 kg CH<sub>4</sub> h<sup>-1</sup> or 1.1±0.1 % are potentially overestimated. Due to the high methane concentration during measurements, the procedure for the dynamic chamber method had to be adapted. Instead of two different volume flows (to control for influencing the source by suction), only one high volume flow was applied due to safety reasons. However, the air flow velocity through the chamber (5.3-5.4 m s<sup>-1</sup>) was comparable to the wind speed during measurements (peaks up to 4.1 and 8.3 m s<sup>-1</sup>). However, most of the other leaks at double membranes showed low methane emissions (<0.02 %). One exception was a leaking inner membrane (0.7 % methane loss).

At concrete digesters, highest emissions were caused by leakages at two service boxes (pipe leadthrough, agitator) on the top of concrete roofs (L6, L9) – releasing 1.0 and 0.9 % of the produced methane. At two other service boxes, in contrast, leakages (L7, L8) released considerably less methane (0.1 % of produced methane). As described in section 4.1.1, several leaks were found at two concrete digesters between wall and roof. Emissions were quantified at two of the five major leakage spots (0.1 and 0.3 % methane loss).

The two detected leakages at single membrane domes were repaired previous to the quantification measurement and methane losses, therefore, not determined.

**Table 8: Methane emission rates [g h<sup>-1</sup>] and methane emission factors (EF) [% of produced methane] of different leakages. Standard deviations are given in brackets ().**

Digester technology	Plant ID	Leak number	Leak location	CH <sub>4</sub> emission rate [g h <sup>-1</sup> ]	CH <sub>4</sub> EF [% CH <sub>4</sub> produced]
Double membrane dome	5	L11	Concrete wall/roof	2983 (207)	1.07 (0.07)
	11	L13	Air-inflated double membrane dome	609 (58)	0.68 (0)
	4	L15	Wire to adjust agitator	16 (7)	0.02 (0.01)
	4	L14	Air-inflated double membrane dome	1 (0)	0.01 (0)
	11	L16	Bull' Eye	5 (3)	0.01 (0)
	5	L12	Concrete wall/roof	27 (5)	0.00 (0)
Concrete roof digester	2	L6	Service box (pipe leadthrough)	1412 (116)	0.97 (0.08)
	2	L9	Service box (agitator)	1299 (114)	0.89 (0.08)
	1	L1	Concrete wall/roof	216 (65)	0.28 (0.08)
	1	L2	Concrete wall/roof	98 (32)	0.13 (0.04)
	3	L7	Service box (pipe leadthrough)	69 (24)	0.09 (0.03)
	3	L8	Service box (pressure relief valve)	63 (21)	0.09 (0.03)

In previous studies, the majority of leakages showed quite low emissions compared to the overall methane production – with a few exceptions such as 5 % methane loss caused by a not properly closed manhole (Liebetrau et al., 2013). In Westerkamp et al. (2014), one of several leakages on a concrete roof has been investigated - emitting 0.05 % of the produced methane or 730 g h<sup>-1</sup>.

### 4.1.2.3 Biogas utilization

#### 4.1.2.3.1 Methane slip of CHP units

The 13 investigated CHP units showed a high variability in methane emissions, ranging between 0.4 and 3.3 % of utilized methane (see Table 9). At one unit (plant no. 6), the high methane slip of 3.2 % could be attributed to malfunction. In this case, maintenance reduced the methane emissions by 35 %. However, at another CHP unit (plant no. 4) maintenance had no effect on methane emissions. The methane slip was > 3 %. Relatively low methane emissions (0.9 %) were quantified at plant no. 13, after a revision of the CHP unit.

**Table 9: Methane slip of combined heat and power plants: methane emission rates [g h<sup>-1</sup>] and methane emission factors (EF) [% of utilized methane]. Standard deviations are given in brackets ().**

Plant ID	Capacity [kW <sub>el</sub> ]	Load [%]	Lambda	CH <sub>4</sub> emission rate [g h <sup>-1</sup> ]	CH <sub>4</sub> EF [g kWh <sup>-1</sup> ]	CH <sub>4</sub> EF [% CH <sub>4</sub> utilized]
4	450	100	1.6	2666 (480)	5.9 (1.1)	3.3 (0.6)
4	450	75	1.4	1939 (679)	5.7 (2)	3.2 (1.1)
6 <sup>1</sup>	527	68	1.4	2108 (13)	5.9 (0)	3.2 (0)
2-1	500	110	1.5	2192 (367)	4.0 (0.7)	2.2 (0.4)
11	509	100	1.4	1932 (401)	3.8 (0.8)	2.2 (0.5)
6	527	61	1.5	1220 (408)	3.8 (1.3)	2.1 (0.7)
7	830	87	1.4	1977 (435)	2.7 (0.6)	1.7 (0.4)
12	500	100	1.3	1531 (722)	3.1 (1.4)	1.7 (0.8)
8	250	100	1.4	740 (98)	3.0 (0.4)	1.6 (0.2)
1	350	97	1.4	1017 (148)	3.0 (0.4)	1.6 (0.2)
2-2	330	51	1.4	472 (38)	2.8 (0.2)	1.5 (0.1)
13	330	100	1.5	554 (105)	1.7 (0.3)	0.9 (0.2)
3-1	124	100	1.3	191 (119)	1.5 (1)	0.8 (0.5)
3-2	180	100	1.3	150 (26)	0.8 (0.1)	0.4 (0.1)
3-3	124	100	1.2	100 (5)	0.8 (0)	0.4 (0)

<sup>1</sup> Malfunction.

The methane slip, resulting from incomplete combustion, has shown to be affected by various factors including the design of the combustion process, engine settings (ignition time, valve overlap, combustion-air ratio), load and age-related wear of components (Aschmann et al., 2008, Aschmann et al., 2010, Aschmann and Effenberger, 2012, Aschmann et al., 2019). Previous studies revealed a wide range of methane emissions from CHP gas engines. Methane slips of 0.4-6.0 % (n=13) were measured by Kretschmann et al. (2012), 0.6-3.3 % (n=9) by Liebetrau et al. (2013) and 0.4-3.2 % (n=6) by Aschmann et al. (2014).

#### 4.1.2.3.2 Methane slip of biogas upgrading units

Measurements were performed at two biogas upgrading units (BUU) based on pressure swing adsorption (PSA) technology with exhaust gas treatment by a micro-gas turbine. With exhaust gas treatment, the methane slip of the BUU could be reduced to 0.03 and 0.15 % of the utilized methane, respectively (see Table 10).

**Table 10: Methane emission rates [g h<sup>-1</sup>] and methane emission factors (EF) [% of utilized methane] from biogas upgrading after exhaust gas treatment. Standard deviations are given in brackets ().**

Plant ID	Technology biogas upgrading	Technology exhaust treatment	CH <sub>4</sub> emission rate [g h <sup>-1</sup> ]	CH <sub>4</sub> EF [% CH <sub>4</sub> utilized] <sup>1</sup>
9	Pressure swing adsorption	Micro-gas turbine	114 (50)	0.03 (0.02)
10	Pressure swing adsorption	Micro-gas turbine	16 (10)	0.15 (0.07)

<sup>1</sup> related to methane input to biogas upgrading unit

Liebetrau et al. (2013) measured 5.3 % methane slip at a BUU unit (PSA), when the exhaust gas treatment was out of order. Lozanovski and Brandstetter (2015) investigated two PSA units. At measuring points upstream of the exhaust gas treatment, methane slips were 1.0 and 1.6 %. The effect of exhaust gas treatment was examined by Kvist and Aryal (2019). They found that a regenerative thermal oxidizer (RTO) could reduce the methane slip of a BUU (membrane separation) by 99.5 %.

## 4.2 Remote sensing measurements – Inverse dispersion modelling

Remote sensing measurements (IDM) were performed at five Austrian biogas plants. An overview of the individual measurement days is given Table 11, including information on meteorological conditions as well as measurement results.

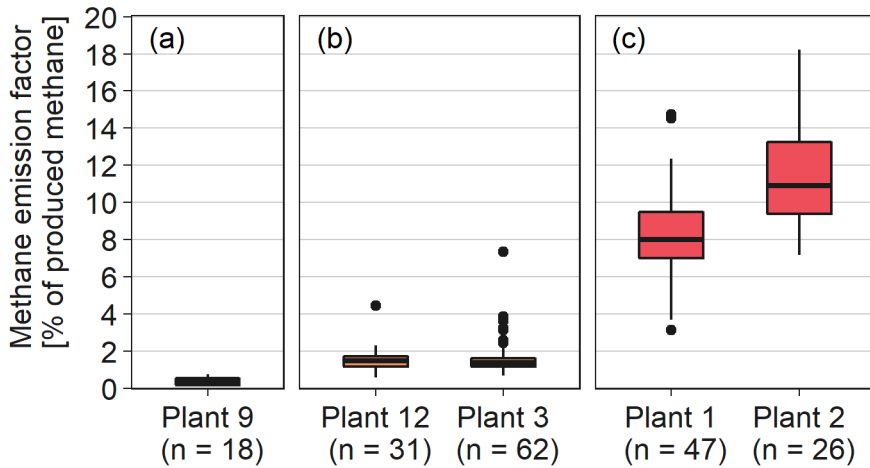
**Table 11: Overview of remote sensing measurements: meteorological conditions, methane emission rates [kg h<sup>-1</sup>] and methane emission factors (EF) [% of produced methane] from full-scale biogas plants. Standard deviations are given in brackets ().**

Plant ID	Date	Ø Wind direction	Ø Wind speed [m s <sup>-1</sup> ]	Ø Air temp. [°C]	n	CH <sub>4</sub> emission rate [kg h <sup>-1</sup> ]	CH <sub>4</sub> EF [% CH <sub>4</sub> produced]
1	03.04.2019	SW-SOO	5.7	18	34	6.3 (2.3)	8.1 (2.9)
	18.03.2021	N-NW	3.6	2	13	5.9 (1.4)	8.8 (2.1)
2	26.03.2019	N-NW	5.0	10	26	16.6 (4.2)	11.3 (2.8)
3	08.03.2019	SW-W	3.1	11	22	1.7 (1.1)	2.3 (1.5)
	25.02.2020	WSW-W	2.5	13	25	1.0 (0.3)	1.4 (0.5)
	12.10.2020	WSW-WNW	2.2	8	15	0.9 (0.2)	1.3 (0.3)
9	12.03.2020	SW-NW	5.0	18	18	0.3 (0.2)	0.4 (0.2)
12	27.02.2020	W-WSW	6.5	5	10	1.5 (0.9)	1.7 (1)
	15.09.2020	O-OSO	4.3	25	12	1.2 (0.3)	1.3 (0.4)
	20.10.2020	O-OSO	2.2	12	9	1.4 (0.4)	1.6 (0.4)

n=number of 10-minute intervals.

Full-scale plant emissions differed considerably between the biogas plants, ranging between 0.4 and 11.3 % of the produced methane in the digester(s). In Figure 5, the biogas plants are categorized according to the following characteristics: digestate storage (open/closed), biogas utilization (with/without

exhaust gas treatment), leakages (no leaks detected/leaks detected). Lowest emissions were measured at plant no. 9, having implemented the best available technology with regard to methane emissions – a closed digestate storage tank and exhaust gas treatment for the off-gas of the biogas upgrading unit. In addition, no leaks were detected during remote sensing measurements. In contrast, highest methane emissions were determined at two plants with open/not-gastight digestate storage and leaks contributing to overall emissions.



**Figure 5: Methane emission factors (EF) [% of produced methane] from full-scale biogas plants by plant concept: (a) closed digestate storage, biogas utilization with exhaust gas treatment, no leaks, (b) closed digestate storage, biogas utilization without exhaust gas treatment, no leaks (c) open/not-gastight digestate storage, biogas utilization without exhaust gas treatment, leaks detected. n=number of measurements (10-minute intervals).**

Occasionally, emission peaks were captured during single measurement days, classified as OTNOC (other than normal operating conditions) events. These periods with increased methane emissions are listed in Table 12 and are not included above (Table 11, Figure 5). Average full-scale plant emissions were subtracted from individual 10-min intervals during OTNOC events - Table 12 thus shows the additional emissions caused by specific events. Two OTNOC events could be attributed to the activation of pressure relief valves (PRV). At event 3, gas pipes were blocked by foam. As a result, biogas (18.8 % of the production) was released by the safety valves. During event 4, PRVs were made frost-proof by replacing the water seal with a frost protection agent. This procedure, done once a year, caused methane losses of 3.0 %. For the other OTNOC periods (event 1, 2 and 5), the underlying cause could not be identified. Event 1 and 2, occurring in the course of the same measurement day, showed the highest emissions. Within these time periods, additional 19 and 20 % of the produced methane were released.

**Table 12: Methane emission rates [kg h<sup>-1</sup>] and methane emission factors (EF) [% of produced methane] caused by other than normal operating conditions (OTNOC). Standard deviations are given in brackets ().**

Emission source	Event no.	n	CH <sub>4</sub> emission rate [kg h <sup>-1</sup> ]	CH <sub>4</sub> EF [% CH <sub>4</sub> produced]
Safety valves	Event 3	6	13.8 (8)	18.8 (10.9)
	Event 4	29	2.7 (1)	3.0 (1.1)
Unknown	Event 2	5	29.5 (11)	20.2 (7.5)
	Event 1	6	27.7 (13.9)	19.0 (9.5)
	Event 5	24	1.6 (0.3)	1.7 (0.4)

n=number of measurements (10-minute intervals).

In previous studies, a wide range of methane emissions from full-scale biogas plants was determined by remote sensing methods using IDM or TDM. At agricultural and bio-waste treatment plants, methane losses ranged between 0.4 and 8.6 % of the total methane production (Westerkamp et al., 2014, Hrad et al., 2015, Groth et al., 2015, Reinelt et al., 2017, Fredenslund et al., 2018, Scheutz and Fredenslund, 2019, Bakkaloglu et al., 2021), at waste water treatment plants between 2.2 and 14.9 % (Delre et al., 2017, Samuelsson et al., 2018, Fredenslund et al., 2018, Scheutz and Fredenslund, 2019).

OTNOC events leading to flaring and the activation of safety valves, respectively, were also covered by previous remote sensing measurements using IDM (Flesch et al., 2011, Groth et al., 2015). Reinelt et al. (2016) as well as Reinelt and Liebetrau (2019) investigated trigger events and emissions from PRV by on-site monitoring.

## 4.3 Implementation of mitigation measures and cost-benefit analysis

After the first measurement series, three mitigation measures were implemented by Austrian plant operators to reduce methane emissions (in addition to fixing leakages). The effectiveness of these measures was evaluated in the second measurement series. At two biogas plants, maintenance of the CHP unit was arranged after a methane slip of > 3 % (of the utilized methane) was quantified. While at one plant maintenance of the CHP unit successfully reduced emissions by 35 %, the methane slip of the second unit stayed on a high level. As a third mitigation measure, the leaking inner membrane of an air-inflated double membrane dome was replaced. With that, methane emissions of the digester could be reduced from 0.7 % (of produced methane) to < 0.01 %.

The two successful mitigation measures were evaluated by cost-benefit analysis. Results are shown in Table 13. As the maintenance of the CHP unit was included in a maintenance contract, no additional costs arose for the plant operator for this measure. Thus, both, net present value A and B were positive. For replacing the inner foil of the double membrane dome, great part of the costs is caused by biogas losses when emptying the digester (reduced HRT of the substrate). Therefore, cost-effectiveness depends on whether the mitigation measure can be implemented together with other maintenance work.

The net present value is only positive in case the replacement is scheduled around a next revision or external costs of methane losses are internalized.

**Table 13: Cost-benefit analysis of mitigation measures.**

Mitigation measure	CH <sub>4</sub> EF [% of produced/utilized CH <sub>4</sub> ] <sup>1</sup>		Net present costs	Evaluation: NPV A, B < 0 (+) NPV A < 0 < NPV B (++) NPV A, B > 0 (+++)
	1 <sup>st</sup> measurement series	2 <sup>nd</sup> measurement series		
Maintenance of CHP unit after malfunction	3.2	2.1	0 € (costs included in maintenance contract)	+++
Exchange of leaking inner membrane of air-inflated double membrane dome	0.7	<0.01 <sup>2</sup>	15.000 € (material construction costs) plus costs of lost biogas production <sup>3</sup>	++/+++ <sup>4</sup>

CHP=combined heat and power; EF=emission factor [% produced/utilized methane]; NPV = net present value;

<sup>1</sup> The EF of the CHP unit is given in % of utilized methane, the EF of foil roof emissions in % of produced methane.

<sup>2</sup> Measurements were performed at another double membrane dome of the same biogas plant, at which the inner foil was recently replaced.

<sup>3</sup> Over three months, one third of the biogas production is lost by emptying the digester (information by plant operator).

<sup>4</sup> If costs of lost biogas production are considered: (++); if costs of lost biogas production are not considered (measure is done at next scheduled revision): (+++).

## 5 European and national voluntary systems

### European and national position paper

Based on the findings of the first measurement series as well as a literature study on possible leaks within a biogas plant, a first position paper was prepared by the European Biogas Association (EBA). Each national biogas association partner then discussed this draft paper within their national community. In Austria, this was done by the Austrian Compost and Biogas Association (KBVÖ) within several meetings of the state's delegates of the biogas department from KBVÖ. The European position paper was then adapted and published under the title "Methane emission mitigation strategies - Information sheet for biogas industry" (EBA. 2020a). The paper covers topics such as the current situation and perspectives of regulations in Europe, emission sources and recommendations for minimizing emissions from biogas plants as well as activities of the biogas industry to reduce methane emissions.

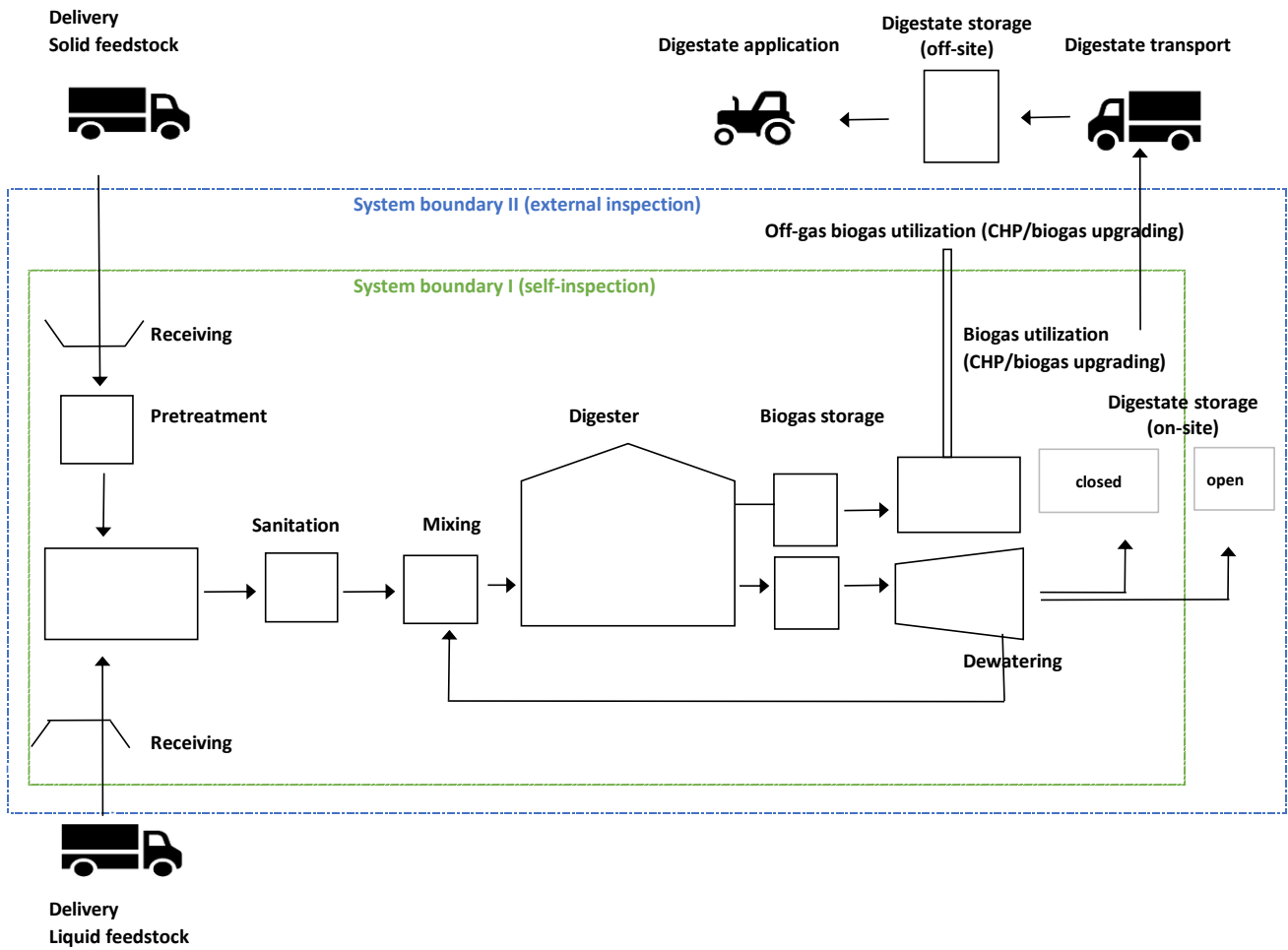
Based on the European paper, an Austrian position paper was elaborated. In addition to the European paper, it includes national legal requirements for biogas plant developer and operator. The paper "Erhebung und Verminderung von Methanemissionen in Biogasanlagen. Teil I: Einführung in die

Thematik“ (KBVÖ. 2020a) was published within the newsletter and website and has been introduced at several meetings.

## **European and national voluntary systems**

Based on the established voluntary systems for emission mitigation in the biogas sectors in Sweden and Denmark, minimum requirements for a European system were elaborated in the same way as for the position paper. Within several meetings KBVÖ tried to convince European partners to an agreement of a European wide system with identical requirements and data management, especially concerning a European-wide data base with collected measurement results. Due to the expected amount of data, the latter would allow advanced analyses on best practice scenarios improving emission mitigation of this technique in the future. Sadly, no agreement could be achieved on such a common approach. The document “Minimum requirements for European voluntary systems for self and external inspection of possible methane emissions on biogas and biomethane plants” was published by EBA (2020b).

KBVÖ started to discuss the elaboration of a national voluntary system with plant operators and main construction companies, keeping in mind the reasons and goals behind such a voluntary system without the need of high requirements and expenses. It became clear that the definition of boundaries, as shown in Figure 6, is of high importance in order to allow comparisons to other plants.



**Figure 6: System boundaries for self- and external inspection within the Austrian voluntary system for emission control.**

As plant operators have no direct influence on companies who deliver feedstock and transport digestate, the transport sector outside of the plant has been excluded from the voluntary system. A second boundary was defined to distinguish between self-detection and external detection of possible leakages and main emission sources. While self-inspection is done by the plant operator, external inspection of exhaust gas of CHP and biomethane upgrading systems as well as open digestate storage tanks should be done by third parties (e.g. accredited laboratories). Both forms of inspections also differ in terms of intervals and measurement requirements. While a leakage survey should be performed on a monthly/yearly basis by the plant operator, a leakage survey as well as a quantification of emission sources should be done by third parties every 2.5 and 5 years (or after bigger maintenance), respectively. In addition, the voluntary system defines requirements for the documentation of the emission measurements.

**Table 14: Overview of monitoring program within the Austrian voluntary system for emission control.**

<b>Monitoring</b>	<b>Measurements</b>	<b>Interval</b>
Self-inspection	Leakage detection	Monthly/yearly
External inspection I		Every 2.5 years
External inspection II	Quantification of methane emissions	Every 5 years

The Austrian Voluntary System aims to sustainably reduce unintended emissions through a systematic approach by regular monitoring and quantification of methane emissions. It has been announced via website and newsletter (KBVÖ. 2020b) but much more important it was also introduced to plant operators at 3 workshops in August 2020.

## **6 Determination of representative EFs for biogas plant concepts in Europe**

For the determination of component specific as well as plant concept specific methane EFs, a large number of measurements has been carried out by eight measurement teams. A total of 36 biogas plants in four countries were investigated using both the on-site approach (four teams) for the determination of methane emissions from individual biogas plant components and remote sensing methods (three IDM teams, one TDM team) for the entire biogas plant. An overview of plant characteristics is shown in Table 15.

**Table 15: Overview of investigated European biogas plants.**

Plant ID	CH <sub>4</sub> production [kg h <sup>-1</sup> ]	Substrate	Biogas utilization	Biogas upgrading technology	Digestate storage (open/closed)
I-1	77	energy crops, manure	BUU	PSA, EGT	closed
I-2	64	energy crops, manure	BUU	PSA, EGT	closed
I-3	501	energy crops, manure	BUU	Chemical scrubber	closed
I-4	529	energy crops	BUU (67%), CHP (33%)	Chemical scrubber	closed
I-5	46	energy crops, manure	CHP		closed
I-6	90	energy crops, manure	CHP		closed
I-7	90	energy crops, manure	CHP		closed
I-8	13	manure	CHP		open
I-9	36	energy crops	CHP		closed
I-10	85	energy crops, manure	CHP		closed
I-11	153	energy crops, manure	CHP		closed
I-12	134	energy crops	CHP		closed
I-13	203	energy crops, manure	CHP		closed
I-14	102-135	energy crops, manure	CHP		closed <sup>1</sup>
I-15	98	energy crops, manure	CHP		closed
I-16	81	energy crops, manure	CHP		closed
I-17	18	manure, bio-waste	CHP		open
I-18	62	manure, bio-waste	CHP		closed
I-19	37	manure, bio-waste	CHP		open
I-20	130	energy crops, manure	CHP (12 %), off-site (88 %)		closed
I-21	248	energy crops, manure	CHP (83 %), off-site (17 %)		open
I-22	279	bio-waste	BUU	Membrane	open
I-23	167	bio-waste	BUU	Chemical scrubber	open
I-24	132	bio-waste	BUU	Chemical Scrubber	open
I-25	321	bio-waste	BUU	Water scrubber, EGT <sup>2</sup>	closed <sup>1</sup>
I-26	465	bio-waste	BUU	Chemical scrubber	open
I-27	542	bio-waste	BUU	PSA, EGT	open
I-28	73	bio-waste	CHP		closed
I-29	100	bio-waste	CHP		closed
I-30	77	bio-waste	CHP		closed
I-31	100	bio-waste	CHP		open
I-32	73	bio-waste	CHP		closed
I-33	67-78	bio-waste	CHP (85%), BUU (15%)	PSA	open
I-34	146	bio-waste	CHP (90%), BUU (10%)	PSA	open
I-35	133	wastewater	BUU	Chemical scrubber	open
I-36	80	wastewater	off-site		open

BUU: biogas upgrading unit; CHP: combined heat and power; EGT: exhaust gas treatment; n.s.: not specified; PSA: pressure swing adsorption

<sup>1</sup> gastight covered after first measurement campaign

<sup>2</sup> exhaust gas treatment installed after first measurement campaign

The average methane EF from all member countries determined with the remote sensing approach ranged from 0.4 to 11.3 %, relative to the amount of methane produced. At biogas plants, where no leakages were detected, EF were 1.3 % (median) for plants with closed digestate storage (n=12) and 2.1 % when digestate was not gas-tight stored (n=4). A wide range of methane emissions from agricultural and bio-waste treating plants was also quantified in previous studies, between 0.4 and 8.6 % of the total methane production (Westerkamp et al., 2014, Hrad et al., 2015, Groth et al., 2015, Reinelt et al., 2017, Fredenslund et al., 2018, Scheutz and Fredenslund, 2019, Bakkaloglu et al., 2021).

An overview of component specific EF combined from all member countries is given in Table 16. At most of the investigated biogas plants, major methane emissions arose from biogas production (open digestate storage tanks) and from biogas utilization (methane slip). Within the substrate receiving and storage stage, high emissions (up to 2.4 % of produced methane) were occasionally quantified in the ventilation air from several sources (receiving halls and storage tanks). Elevated emissions of up to 3.0 % were also determined by Fredenslund et al. (2018). However, not-gastight storage tanks emitted 0.2 % of the produced methane (median, n=6) and receiving halls 0.01 % (n=3). Results are comparable with previous studies (see section 4.1.2.1). Within the biogas production stage, emissions occurred primarily due to open digestate storage. Specific to a certain filling level, temperature and substrate, methane losses ranged from 0.1 % to 5.6 %, compared to <0.1-12 % determined in recent studies (see section 4.1.2.2.2). Methane emissions from biogas utilization showed a large variation. The methane slip (median) of different technologies ranged from <0.1 to 3.0 %. Consistently low emissions were determined when exhaust gas treatment was installed or chemical scrubbing was used to upgrade biogas to biomethane. Highest emissions were caused by three CHP units (pilot injection) and two BUU water scrubbing units with median values around 3 %. However, for both technologies lower emissions can be achieved according to previous studies. For the CHP units, median values of 1.4 % (n=2) and 2.6 % (n=4) were measured by Liebetrau et al. (2013) and Aschmann et al. (2014), respectively. For water scrubbing units, 1.3 % (n=1) was determined by Lozanovski and Brandstetter (2015), 1.5 % (n=1) by Liebetrau et al. (2013), 3.1 % by Fredenslund et al. (2018) and 1.1-2.0 % (n=4) were measured by Kvist and Aryal (2019). Similar to previous studies (see section 4.1.2.3.1), the 21 investigated CHP gas engines showed a wide range of methane emissions, starting with 0.4 % methane slip up to 3.3 %. The median value (1.6 %, n=21) was lower compared to the pilot injection technology (3.0 %, n=3). Individual leakages caused methane losses of 0.04 % of produced methane (median, n=43).

**Table 16: Component specific emission factors (EF) from all member countries according to process stage.**

Process stage	Component	CH <sub>4</sub> EF [% CH <sub>4</sub> produced/utilized] <sup>1</sup>			
		n	median	min	max
Substrate receiving & storage	Receiving hall & storage tank	3	1.05	0.83	2.44
	Not-gastight storage tank	6	0.17	0.00	0.81
	Receiving hall	3	0.01	0.00	0.06
	Not-gastight mixing tank	1	0.00	0.00	0.00
Biogas production	Not-gastight storage tank	8	2.83	0.06	5.62
	Double membrane	25	0.00	0.00	0.01
Biogas production (leaks)	Concrete digester	8	0.20	0.09	0.97
	Double membrane	23	0.01	0.00	1.07
Biogas utilization (methane slip)	CHP, pilot injection	3	3.00	2.74	3.74
	BUU, water scrubbing	2	2.94	1.98	3.89
	CHP, gas engine	21	1.61	0.43	3.30
	BUU, exhaust treatment	3	0.10	0.03	0.15
	BUU, chemical scrubbing	4	0.06	0.04	0.11
Biogas utilization (ventilation air)	Biogas analysis instrument	1	0.05	0.05	0.05
	CHP, processing room	10	0.04	0.00	0.22
	BUU, chemical scrubbing	1	0.00	0.00	0.00
Biogas utilization (leaks)	Biogas analysis instrument	1	0.30	0.30	0.30
	BUU, water scrubbing	3	0.28	0.20	0.38
	BUU, chemical scrubbing	3	0.25	0.18	2.14
	CHP, processing room	5	0.05	0.01	0.21
Miscellaneous	Ventilation air	2	2.21	1.64	2.78

BUU=biogas upgrading unit. CHP= combined heat and power plant.

<sup>1</sup> EF of biogas utilization units are given in % of utilized methane, others in % of produced methane.

All measured data as well as data from previous studies were analyzed for further use in an emission quantification model, adapted from an approach developed for the natural gas sector (Balcombe et al., 2018). For selected technologies, probability density functions were calculated and used for Monte-Carlo simulations. The aim of this model is to estimate methane emissions for different biogas plant concepts, thereby assessing the variation in emissions and the causes of these variations as well as the impact of emissions-reducing equipment. The greater understanding of methane emissions profiles is important to analyze the impact of different technological options across the whole biogas-biomethane value chain and identifies a series of options as best technological option scenarios. However, further research is needed to increase the amount of emission data. The model has to be specified for open or not gastight covered digestate storages and more precise data from the biogas plant inventory is necessary. The status quo of the model is described in Clauß et al. (2019b).

## 7 Conclusion and Recommendations

In summary, it can be said that the successful provision of cross-national comparable emission factors based on harmonized guideline for the European Biogas Plant Inventory has attracted EU-wide interest.

In addition to the emission measurements, however, the knowledge transfer of methane mitigation measures also played a major role in the EvEmBi project. The publication of the national and European position papers triggered a discussion with the European commission about a European (voluntary) monitoring system especially for the biogas sector, which can be considered a great success. The consistently positive feedback from the operator training sessions shows that there is both interest on behalf of plant operators and a need for action in terms of knowledge exchange. The continuation of the emissions workshops is considered very positive by the national associations and holds great potential for the avoidance of unintentional methane emissions.

After the first measurement campaign, mitigation measures were implemented at selected plants. Thus, in the second campaign, some successful reduction measures could be proven and included in the cost-benefit analysis. Mitigation measures included an additional maintenance of a CHP unit and the foil replacement of an air-inflated double membrane dome. Based on on-site and remote sensing methods it could be shown that certain components have an increased risk of methane emissions, so regular leak detection is important to keep the plant operating cost-effectively and safely.

Building on the results of the EvEmBi project, the data base on methane emissions from diverse technologies should be further expanded (also by performing emission measurements in the scope of the voluntary agreements) in order to enable representative, technology-based emission factors and benchmarks.

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# Energieforschungsprogramm - 11. Ausschreibung

Klima- und Energiefonds des Bundes – Abwicklung durch die Österreichische Forschungsförderungsgesellschaft FFG

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