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Total methane emission rates and losses from 23 biogas plants

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ABSTRACT

Methane losses from biogas plants are problematic, since they contribute to global warming and thus reduce the environmental benefits of biogas production. Total losses of methane from 23 biogas plants were measured by applying a tracer gas dispersion method to assess the magnitude of these emissions. The investigated biogas plants varied in terms of size, substrates used and biogas utilisation. Methane emission rates varied between 2.3 and 33.5 kg CH_4 h^{-1}, and losses expressed in percentages of production varied between 0.4 and 14.9%. The average emission rate was 10.4 kg CH_4 h^{-1}, and the average loss was 4.6%. Methane losses from the larger biogas plants were generally lower compared to those from the smaller facilities. In general, methane losses were higher from wastewater treatment biogas plants (7.5% in average) in comparison to agricultural biogas plants (2.4% in average). In essence, methane loss may constitute the largest negative environmental impact on the carbon footprint of biogas production.
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1. Introduction

Biogas from anaerobic digestion, using various substrates such as manure, food waste, organic industrial waste and sludge from wastewater treatment, may result in several greenhouse gas (GHG) mitigation effects, including fossil fuel substitution, the possible balancing of energy sources in a supply system with a high proportion of wind and solar power and a reduction in methane (CH_4) emissions from manure management (Clemens et al., 2006; IPCC, 2011; Sommer et al., 2004). Fugitive CH_4 emissions from biogas plants, however, will reduce the environmental benefits of biogas production, mainly because of the relatively high global warming potential of CH_4, in that releasing just 1 kg of CH_4 into the atmosphere has the same effect with regards to global warming as the release of 28 kg of carbon dioxide (CO_2) integrated over a 100-year period (not including climate feedback) (Myhre et al., 2013). Data on the magnitude of these emissions are sparse, which in turn causes uncertainty with regards to the environmental assessment of biogas production concerning global warming (Meyer-Aurich et al., 2012; Møller et al., 2009). Recent studies suggest that the extent of CH_4 emissions expressed as a fraction of production lost to the atmosphere (also referred to as "CH_4 loss") may vary between facilities. Liebetrau et al. (2013), for instance, monitored CH_4 emissions from ten German biogas plants, using an on-site approach where individual leaks were identified and emission rates were subsequently measured. It was found that CH_4 emissions relative to the energy output of the biogas plants varied by approximately one order of magnitude between plants, and that open digestate storage tanks in many cases were the most significant emission source. Other sources of CH_4 emission from biogas plants may include unburnt CH_4 from gas engine exhausts, pressure relief valves, biogas upgrading units, ventilation from buildings, leaks in pipes, tanks, etc. (Kvist and Aryal, 2019; Angelidaki et al., 2018; Fredenslund et al., 2018; Liebetrau et al., 2013; Reinelt et al., 2017, 2016; Samuelsson et al., 2018).

An important step in understanding and subsequently reducing CH_4 emissions in the biogas sector is the reliable identification and quantification of single emission sources and the quantification of overall plant emissions. In general, two main approaches can be used for gas emission quantification: on-site and ground-based remote sensing approaches. The on-site approach measures emissions from various single sources at the plant, and it is the method most commonly used (Reinelt et al., 2016; Daniel-Gromke et al., 2015; Westerkamp et al., 2014; Liebetrau et al., 2013). Often a two-step procedure is followed where the first step includes a leakage search performed by using infrared cameras or handheld methane analysers. The second step includes quantification of each identified leakage or emission source often using the stationary or dynamic flux chamber technique. The ground-based remote sensing approach includes different methodologies and measures emissions from a good distance (for example one kilometre) away from the plant, thus providing plant-integrated emission numbers (Fredenslund et al., 2018; Delre et al., 2017; Groth et al., 2015; Yoshida et al., 2014; Hrad et al., 2014; Westerkamp et al., 2014; Flesch et al., 2011). Ground-based remote sensing techniques

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encounter inverse dispersion techniques using for instance open-path lasers and tracer gas dispersion methods. Recent measurement comparison studies have found that methods measuring the plant's total CH_4 emission often result in a higher emission rate in comparison to on-site measurements, where the total emission is obtained by summing up those measured from single sources (Fredenslund et al., 2018; Reinelt et al., 2017). The reason for the discrepancy between on-site and ground-based remote sensing approaches is most likely that single sources are overlooked and/or not identified, or they are technically not quantifiable when using a particular measurement technique (e.g. open tanks). For GHG emission reporting or environmental assessment, the plant's total emissions are important; however, if the purpose of measuring CH_4 emissions at a biogas plant is to identify mitigation options, and thereby provide options to improve the environmental benefits of biogas production, on-site methods are needed.

The objective of this study was to quantify CH_4 emission rates and losses from full-scale biogas plants. The study focused primarily on large, centralised, manure-based biogas plants, which produce the bulk of biogas in Denmark. Production capacity at this type of facility was in the expansion phase nationally at the time of this study. In addition, CH_4 emission rates and losses were measured at biogas plants located at wastewater treatment plants (WWTPs). Landfill gas extraction and utilisation sites were not included. The paper compiles the results taken from several biogas plants, in order to provide an estimate of CH_4 losses from biogas production and to assess the importance of minimising this issue. CH_4 emissions were measured using the tracer gas dispersion method, which measures plant-integrated emission rates. The environmental importance of fugitive CH_4 emissions from biogas plants was evaluated by performing CO_2 footprint calculations for a generic, manure-based agricultural biogas plant.

2. Methodology

2.1. Site descriptions

The biogas plants included in this study all utilise continuously stirred anaerobic digesters to produce biogas, and they all are commercially operated facilities. They varied in terms of feedstocks, size, rate of gas production, type of gas utilisation and other factors. Table 1 provides an overview of the main characteristics of the plants.

Thirteen of the biogas plants (plants 1–13) are categorised herein as "agricultural", which means that the feedstocks consist mainly of manure, energy crops and agricultural waste, though they can also receive other feedstocks such as slaughterhouse waste or food waste. Out of the 13 agricultural plants, nine receive manure as the main feedstock (>75% of dry matter input is manure), whereby organic waste (organic industrial waste and/or food waste) is used as a supplement to increase gas production. Two biogas plants (plants 8 and 12) rely on energy crops (grass, maize silage and forage rye), one (plant 5) receives mainly organic waste (~80% of dry matter input) but also receives manure and one plant (plant 13) mainly uses food waste. Plant 13 (and possibly also plant 5) could depending on definition be termed a waste treatment biogas plant as it mainly treated slaughterhouse waste, food industry waste and household food waste. However, as this plant was the only one of this type and also as the generated digestate is spread on farmland, the plant was included in the agricultural biogas plant category.

Five of the agricultural biogas plants (plants 1, 2, 5, 6 and 8) were recently constructed (constructed in 2013 or later), whereas the remaining agricultural plants generally were constructed in the 1980s or 1990s. For a number of reasons, the 2000s saw very low levels of investment in Danish biogas production (Raven and Gregersen, 2007), whereas increases and diversification in subsidies in recent years have led to a "second wave," with most new production capacity emanating from large facilities that upgrade and inject the biogas into the Danish natural gas distribution grid. At the smaller and older plants, it is more common that the biogas is utilised on-site in a combined heat and power (CHP) unit.

Biogas plants 14–23 are categorised herein as "wastewater treatment biogas plants". These plants utilise sludge from wastewater treatment to produce biogas, and they are all located on the grounds of the WWTP from where the sludge originates. They can thus be considered part of a larger plant, the primary function of which is to remove pollutants from wastewater before discharge to a recipient, with energy production as secondary function. The biogas plants categorised as "agricultural biogas plants" all rely on gas production for revenue. Although the wastewater treatment biogas plants receive revenue from their gas production, their primary function is to stabilise and reduce the volume of sludge, and thereby the costs of further sludge treatment. The WWTPs may thus arguably have less incentive to minimize loss of methane compared to the agricultural biogas plants.

The size of the plants varied in size in terms of treated feedstocks, from 30,000 to 600,000 tonnes (wet weight) per year for agricultural biogas plants, while the WWTPs treated between 60,000 and 805,000 PE, which corresponded to a load to the on-site biogas plants of between 3,000 and 112,000 tonnes (wet weight) per year.

The biogas plants differ with regards to gas utilisation (Table 1). At 12 plants, all or some of the produced biogas is utilised on site in a CHP unit. At plants 3, 4 and 7 some of the gas (~20–30%) is used on site in a CHP unit providing process heat for the biogas reactors. The generated electricity is sold to the grid. The remaining part of the gas is routed off site to a nearby power plant (where it is used in a CHP unit). At eight plants, all or some of the biogas is upgraded to biomethane, using technologies such as water scrubbers or chemical scrubbers. At these facilities, the gas is either compressed and transported off site or is injected into a natural gas distribution network. At four plants (5, 11, 14 and 21), all gas utilisation occurs off site. An example of this type is plant 5, where the biogas is led to a nearby power plant (and used in a CHP unit) to generate electricity to the grid and heat to a district heating network.

Open digestate storage units may be significant emitters of CH_4 from biogas plants (Samuelsson et al., 2018; Reinelt et al., 2017; Baldé et al., 2016; Liebetrau et al., 2013). Table 1 lists those facilities, which store digestate in open tanks on site. All biogas plants were equipped with gas storage units with capacities typically corresponding to ~1 to 2 days gas production.

In all, the 23 biogas plants included in this study represent a variety of continuously stirred reactor biogas plant types with regards to amounts of feedstock utilised, feedstock types, gas production rates and gas utilisation.

2.2. Tracer gas dispersion method

CH_4 emission rates from each biogas plant were quantified using a tracer gas dispersion method, whereby a gaseous tracer (here acetylene gas – C_2H_2) is continuously released at the biogas plant, and concentrations of CH_4 and C_2H_2 are then measured while traversing the CH_4/C_2H_2 plume at distances up to ~2 km away, using a vehicle-mounted, high-precision gas analyser. The method has been applied to quantify fugitive emissions from various facilities such as landfills, composting facilities, WWTPs and biogas plants (Andersen et al., 2010; Fredenslund et al., 2018; Mønster et al., 2014; Scheutz et al., 2011; Yoshida et al., 2014). An advantage of this method compared to on-site methods, where emission sources are quantified individually, is the measurement

Table 1
Overview of the main characteristics of the investigated biogas plants.

Agricultural biogas plants	Type of feedstock and annual total amount treated at the plant (in tonnes wet weight per year)	On site gas utilisation (CHP[1]/biogas upgrade)	Digestate storage (open/closed)
1*	Manure, maize silage, organic waste (600,000)	Biogas upgrade: chemical scrubber, gas grid injection	Closed
2*	Manure, slaughterhouse waste (240,000)	Biogas upgrade: water scrubber, gas grid injection	Closed
3	Manure, organic waste (300,000)	CHP (partly off site)	Closed
4	Manure, slaughterhouse waste, other organic waste (235,000)	CHP (partly off site)	Closed
5*	Industrial waste, manure (200,000)	None – routed for off-site use in a CHP	Closed
6*	Manure, maize silage (118,000)	Biogas upgrade: chemical scrubber, gas grid injection	Closed
7	Manure, slaughterhouse waste, other organic wastes (225,000)	CHP (partly off site)	Closed
8*	Maize silage, forage rye	CHP and biogas upgrade: chemical scrubber, gas grid injection	Closed
9	Manure, organic waste, maize silage (170,000)	CHP	Closed
10	Manure, organic waste (37,000)	CHP	Closed
11	Manure, maize and grass silage, glycerol (30,000)	None – routed for off-site use in a CHP	Closed
12	Grass and maize silage, manure	CHP	Open
13	Organic waste (slaughterhouse waste, industrial food waste and household food waste) (104,000)	Biogas upgrade: chemical and water scrubber, gas to vehicle fuel	Open
Wastewater treatment biogas plants	Feedstock (amount given in person equivalent)	On site gas utilisation (CHP[1]/biogas upgrade)	Digestate storage (open/closed)
14	Sludge from wastewater treatment (750,000 PE[2])	None – routed for off-site biogas upgrading	Closed
15	Sludge from wastewater treatment (265,000 PE)	CHP	Open
16	Sludge from wastewater treatment (150,000 PE)	CHP	Closed
17	Sludge from wastewater treatment (420,000 PE)	Biogas upgrade: chemical scrubber, gas grid injection	Closed
18	Sludge from wastewater treatment (95,000 PE)	CHP	Closed
19	Sludge from wastewater treatment (60,000 PE)	CHP	Open
20	Sludge from wastewater treatment (125,000 PE)	CHP	Closed
21	Sludge from wastewater treatment (805,000 PE), industrial food waste and sewage sludge from small WWTPs	None – routed for off-site biogas upgrading, gas to vehicle fuel	Open
22	Sludge from wastewater treatment (95,000 PE), food waste	Biogas upgrade: chemical scrubber, gas to vehicle fuel	Open
23	Sludge from wastewater treatment (120,000 PE)	Biogas upgrade: chemical scrubber, gas to vehicle fuel	Open

[1] CHP: Combined heat and power.
[2] PE: Person equivalent.
* Constructed in 2013 or later.

of the biogas plant's total CH_4 emission, with little risk of underestimating them due to undetected emission sources (Fredenslund et al., 2018).

The method and instrumentation are described in detail in Mønster et al. (2014) and Yoshida et al. (2014). The overall error of the method has been the subject of a recent validation study, and it was found very likely to be less than 20% (Fredenslund et al., 2019). The potential error of the tracer gas dispersion measurement technique was determined to 15% by establishment of an error budget including the analytical error, error in the tracer gas release rate, data processing, and error in tracer gas placement and source simulation. The error of a measurement is the combined error of the method and the variability of the quantification, which was found to be about 20% in a controlled release test and comparable to the error obtained by comparison of the measured emission rate and the known controlled release rate (Fredenslund et al., 2019).

Emission rates are calculated using Eq. (1):

$$E_{target} = Q_{tracer} \times \frac{\int_{plume\ start}^{plume\ end} (C_{target} - C_{target,backgruound})dx}{\int_{plume\ start}^{plume\ end} (C_{tracer} - C_{tracer,backgruound})dx} \times \frac{MW_{target}}{MW_{tracer}} \tag{1}$$

where E_{target} is the emission rate of CH_4 in kg h^{-1}; Q_{tracer} is the release rate of the acetylene tracer gas in kg h^{-1}; C_{target} and C_{tracer} are the measured downwind concentrations in parts per billion (ppb); $C_{target,\ background}$ and $C_{tracer,\ background}$ are the measured background concentrations in parts per billion (ppb) and MW_{target} and MW_{tracer} are the molar weights of the two gases.

The measurements were taken by driving through the downwind plumes several times (typically 10 to 20 traverses per measurement campaign). Each plume traverse resulted in one CH_4 emission measurement, calculated using Eq. (1). The CH_4 emission rate (in kg h^{-1}) was calculated as the average value of the individual plume traverses, and any uncertainty was estimated as the standard error of the mean of the measurements (Fredenslund et al., 2019).

CH_4 loss (%) was determined as the ratio of the measured CH_4 emission to the CH_4 production of the biogas plants, logged the day the measurement was performed.

2.3. Measurement campaigns

The measurements were performed July 2013 through June 2018. At six biogas plants, this happened on a single day, whereas for the remaining 17 plants, measurements were repeated up to a maximum of six days (Table 2).

All measurements were performed using the same analytical equipment and the method described in Section 2.2. The measurements were performed during normal operation of the biogas plants. No malfunctions were reported by the plants for the periods of measurement. As implementation of the method required certain adjustments in each case, some variability with regards to tracer gas release rates and number of release points exists.

Table 2
Overview of measurements performed.

Plant number	Days of measurement campaigns	Number of plume traverses	Tracer gas release rate (kg C_2H_2 h^{-1})
Agricultural biogas plants			
1	1	20	2.24
2	2	66	1.07
3	1	14	2.29
4	3	54	1.90
5	2	32	1.44
6	2	42	0.97
7	3	54	0.83
8	5	166	1.32
9	2	39	1.24
10	1	17	0.91
11	2	29	1.50
12	4	138	1.51
13	2	21	0.44
Wastewater treatment biogas plants			
14	6	82	0.57
15	1	21	0.92
16	4	63	0.51
17	2	37	1.68
18	2	40	0.93
19	4	89	0.48
20	1	16	0.90
21	1	16	–
22	3	81	0.91
23	3	82	0.78

The average tracer gas release rate varied between 0.11 and 2.29 kg C_2H_2 h^{-1}, and the number of tracer gas release points varied between one and three. The measurement distance varied from a few hundred metres up to more than 1 km, according to the availability of drivable roads downwind and the detectability of elevated concentrations of CH_4 and C_2H_2 in the plume – low emission rates and high wind speeds increase dilution, and so it may be necessary to traverse the plume closer to the source of emission.

2.4. Impact of methane emissions on the overall CO_2 footprint of biogas plants

The impact of CH_4 loss on the overall CO_2 footprint of biogas plants was evaluated by using a calculation model provided by the Danish Ministry of Environment for environmental impact assessments of biogas projects (Danish Nature Agency, 2014). The model considers the following factors in determining the overall CO_2 footprint of biogas plants:

- Substitution of fossil fuels
- Substitution of chemical fertiliser
- Transportation of feedstock and digestate
- Change in manure management compared to conventional storage and use of manure in agriculture (fewer GHG emissions from manure storage at farms when manure is digested before storage)
- Energy use of the biogas plant
- Direct GHG emissions from biogas production and utilisation

Emissions and savings were determined by considering five levels of direct CH_4 loss from an agricultural biogas plant: 1%, 2%, 5%, 10% and 20%. Losses of produced biogas contribute directly (CH_4 emitted into the atmosphere) and indirectly (less substitution of fossil fuel as a result of lost biogas production), so both losses were included in the model.

In this assessment, we considered a generic agricultural biogas plant receiving 50,000 tonnes yr^{-1} of cattle manure, 60,000 tonnes yr^{-1} pig manure and 5000 tonnes yr^{-1} organic waste, which in combination produced 2.2 million m^3 CH_4 yr^{-1}. This calculation example is similar to one described by The Danish Nature Agency (2014). Two biogas utilisation options were considered: CHP and biogas upgrade and injection into the natural gas grid.

Table 3 provides an overview of emission factors as well as energy use and CHP energy conversion efficiencies. Two emission factors regarding the use and production of electricity were considered in terms of CHP gas utilisation, namely average and marginal. The average emission factor corresponds to the average emissions associated with the provision of electricity in Denmark, whereas the marginal factor is derived from an estimate of which electricity sources are reduced when production from (for example) biogas plants is increased – also in Denmark. The provision of electricity, on average, consisted of 17% coal, 6% natural gas, 55% wind, hydro and solar, 18% waste incineration, biomass and biogas, 1% oil and 3% nuclear – as reported by the Danish national authority on electricity production (Energinet.dk, 2018), whereas the provision of marginal electricity consisted of 80% coal, 15% natural gas and 5% renewables from a recent study on CO_2 emissions caused by

Table 3
Overview of parameters used in carbon footprint calculations.

Parameter	Value	Reference
Emission factors		
Provision of electricity (average)[a]	0.053 kg CO_2-eq. MJ^{-1}	Energinet.dk (2018)
Provision of electricity (marginal)[b]	0.24 kg CO_2-eq. MJ^{-1}	Ea Energianalyse (2016)
Provision and consumption of natural gas	0.057 kg CO_2-eq. MJ^{-1}	Danish Energy Agency (2018)
Provision of heat (district heating, Danish average value)[c]	0.056 kg CO_2-eq. MJ^{-1}	Danish Nature Agency (2014)
Production of N fertiliser	7.0 kg CO_2-eq. kg N^{-1}	Danish Nature Agency (2014) and Wood and Cowie (2004)
Production of P fertiliser	0.5 kg CO_2-eq. kg P^{-1}	Danish Nature Agency (2014) and Wood and Cowie (2004)
Transportation of digestate, manure, etc.	0.09 kg CO_2-eq. $tonne^{-1}km^{-1}$	(Danish Nature Agency, 2014)
Emission of CH_4	28 kg CO_2-eq. kg CH_4^{-1}	Myhre et al. (2013)
Manure management, cattle	−15 kg CO_2-eq. tonne $manure^{-1}$	Danish Nature Agency (2014)
Manure management, pigs	−23 kg CO_2-eq. tonne $manure^{-1}$	Danish Nature Agency (2014)
Other factors		
Process heat	8.4% of energy output	Danish Energy Agency (2017a)
Electricity use, biogas plant	3.7% of energy output	Danish Energy Agency (2017a)
Electricity use, biogas upgrade and compression	5.3% of energy output	Danish Energy Agency (2017a)
Electrical efficiency, CHP unit	44%	Danish Energy Agency and Energinet.dk (2014)
Total efficiency, CHP unit	92%	Danish Energy Agency and Energinet.dk (2014)

[a] Provision of electricity, average: 17% coal, 6% natural gas, 55% wind, hydro and solar, 18% waste incineration, biomass and biogas, 1% oil and 3% nuclear.
[b] Provision of electricity, marginal: 80% coal, 15% natural gas and 5% renewables.
[c] Average value of Danish district heating networks utilising various energy sources (waste incineration, solar, surplus heat from coal and biomass electricity production and more).

increasing electricity demand in Denmark (Ea Energianalyse, 2016). The differences in energy mix in the provision of average electricity, and provision of marginal electricity cause a relatively large difference in emission factors at 0.053 kg CO_2-eq MJ^{-1} (average) and 0.24 kg CO_2-eq MJ^{-1} (marginal). In the scenario considering the biogas upgrade, for simplicity we only considered the average emission factor for electricity use. In all scenarios, the consumption of heat by the biogas plant was presumed to be in the form of natural gas. In both CHP scenarios, the same emission factor for heat substitution was used, namely an average value of district heating networks in Denmark.

Both feedstock and digestate were assumed to be transported 5 km to and from the biogas plant. The anaerobic digestion of organic waste and the land application of digestates, and thereby recycling of the contained nutrients, was assumed to result in the reduced use of chemical fertiliser at 10 tonnes N yr^{-1} and 5 tonnes P yr^{-1}. The nutrient content of the manure was not considered to contribute to the reduced use of chemical fertiliser, since these nutrients would be applied to agricultural land anyway as raw manure without digestion.

3. Results and discussion

3.1. Measured CH_4 emission rates

Table 4 lists the measured CH_4 emission rates and losses for the 23 biogas plants in this study. The table also lists the biogas production rate of each plant, which was reported by individual plant operator in each case in the form of average daily production at the time of measurement. In those cases where CH_4 emission rates were measured over several campaigns, the listed CH_4 emission rates, gas production rates and CH_4 losses are average values.

Overall, the average CH_4 emission rates varied between 2.3 and 33.5 kg CH_4 h^{-1}. CH_4 losses (CH_4 emission relative to CH_4 production) varied between 0.4 and 14.9%, with the average being 4.6%. These results are comparable to Liebetrau et al. (2013), who found CH_4 losses from single, dominant sources (CHP units and open digestate storage) equating to between 0.22 and 11.2% of the utilised gas at 10 biogas plants. They are also comparable to the results of a study of a Canadian biodigester, where losses under normal operating conditions corresponded to 3.1% of CH_4 production (Flesch et al., 2011).

In general, CH_4 losses were higher from wastewater treatment biogas plants (average 7.5%) than from agricultural plants (2.4%) (Table 4). At seven of the 23 biogas plants, the average measured CH_4 loss was higher than the overall average (4.6%) (Table 4). Of these seven plants, six were WWTPs. The agricultural biogas plant that emitted more than 4.6% (plant 11, Table 4) actually had the lowest level of biogas production (Table 4). Of the agricultural plants, the highest CH_4 loss was 8.4% (biogas plant 11). The reported loss was based on two measurement campaigns, which both showed high CH_4 emissions. There was no on-site gas utilisation and no open mixing tanks, digestate storage tanks or similar. A specific reason as to why the biogas plant had a higher loss than

Table 4
Overview of measured average CH_4 emission rates and losses.

Plant number	On-site sources included in the measured emission (CHP or biogas upgrade unit)	Average biogas production kg CH_4 h^{-1}	Average CH_4 emission rate kg CH_4 h^{-1}	Average CH_4 loss %	Estimated revenue loss[e] k€ y^{-1}	Off-site sources not included in the measured emission (CHP or biogas upgrade unit
1	Biogas upgrade unit	1469	6.5 ± 0.6	0.4 ± 0.04	27.4 ± 2.3	–
2[a]	Biogas upgrade unit	1083	19.1 ± 2.5	1.8 ± 0.23	80.9 ± 10.6	–
3	CHP[f]	888	23.2 ± 1.7	2.6 ± 0.19	98.5 ± 7.2	CHP
4	CHP[f]	858	6.4 ± 0.5	0.7 ± 0.06	27.0 ± 2.0	CHP
5	–	498	3.0 ± 0.3	0.6 ± 0.06	12.9 ± 1.4	CHP
6[a]	Biogas upgrade unit	411	10.7 ± 0.5	2.6 ± 0.12	45.2 ± 2.1	–
7[a]	CHP[f]	404	6.4 ± 0.2	1.6 ± 0.06	27.1 ± 1.0	CHP
8	CHP and biogas upgrade unit	400	2.3 ± 0.4	0.6 ± 0.10	9.9 ± 1.6	–
9	CHP	333	14.9 ± 0.9	4.5 ± 0.26	63.1 ± 3.6	–
10	CHP	234	6.1 ± 0.8	2.6 ± 0.35	26.1 ± 3.5	–
11	–	74	6.4 ± 0.4	8.6 ± 0.50	27.2 ± 1.6	CHP
12	CHP	127	2.6 ± 0.4	2.1 ± 0.35	11.0 ± 1.9	–
13[b]	Biogas upgrade unit	815	21.2 ± 3.3	2.6 ± 0.40	90.0 ± 13.8	–
Plant average CH_4 loss, **agricultural**: 2.4%						
Production weighted average CH_4 loss, **agricultural**: 1.7%						
14[c]	–	440	9.8 ± 0.7	2.2 ± 0.15	41.7 ± 2.8	Biogas upgrade unit
15[a]	CHP	162	13.5 ± 0.5	8.3 ± 0.33	57.3 ± 2.3	–
16[c]	CHP	100	2.6 ± 0.4	2.6 ± 0.39	11.1 ± 1.6	–
17	Biogas upgrade unit	96	12.3 ± 1.2	12.8 ± 1.29	52.0 ± 5.2	–
18	CHP	88	8.1 ± 0.5	9.1 ± 0.60	34.2 ± 2.3	–
19[c]	CHP	85	2.6 ± 0.1	3.0 ± 0.16	11.0 ± 0.6	–
20	CHP	262	10.0 ± 1.0	3.8 ± 0.38	42.5 ± 4.2	–
21[d]	–	525	33.5 ± 0.6	6.4 ± 0.12	142.3 ± 2.6	Biogas upgrade unit
22[c]	Biogas upgrade unit	83	10.0 ± 0.6	12.0 ± 0.78	42.3 ± 2.7	–
23[c]	Biogas upgrade unit	58	8.6 ± 0.4	14.9 ± 0.72	36.5 ± 1.7	–
Plant average CH_4 loss, **WWTP**: 7.5%						
Production weighted average CH_4 loss, **WWTP**: 5.8%						
All biogas plants						
Plant average CH_4 loss, all: 4.6%						
Production weighted average CH_4 loss, all: 2.5%						

[a] Results were partly (first measurement) reported in Fredenslund et al. (2018).
[b] Results were reported in Reinelt et al. (2017).
[c] Results were reported in Delre et al. (2017).
[d] Results were reported in Samuelsson et al. (2018).
[e] Considering an estimated revenue of 0.7 €/Nm^3 CH_4.
[f] About 20–30% of the gas is used in a CHP unit, while the remaining is transported off site.

the average agricultural biogas plant was thus not identified. In general, the CH_4 emission rate relative to production seemed to correlate with the size of the biogas plant (Fig. 1), in that units with the highest gas production emitted proportionally less CH_4 compared to plants with relatively low output. One reason for this finding may be that the larger facilities have more economical resources for maintenance, re-investment and employment of highly proficient plant operators. Another reason may be that the number of potential emission sources (number of process units, pipes, joints, valves, etc.) is not necessarily proportional to the rate of biogas production. There was also the tendency that the larger agricultural plants were built more recently and thus may better represent the most up-to-date technology. CH_4 emission from biogas plants is not regulated directly in Denmark, so no regulatory explanation for the difference in methane loss for small biogas plants compared to larger plants was found. CH_4 losses from plants built within approximately the last 5 years (plants 1, 2, 5, 6 and 8) were relatively low (0.4, 1.8, 0.6, 2.6 and 0.6%, respectively). Two of the 13 agricultural plants were solely energy plants, where the input was mainly crops grown specifically for energy production, and both had CH_4 losses lower than the average for all plants.

As mentioned previously, agricultural biogas plants rely mostly on revenue from energy production for their existence, whereas energy production is a secondary activity in the case of wastewater treatment biogas plants. The economic incentive to maximise energy production, and therefore minimise leaks, may therefore be stronger for agricultural biogas plants. Finally, it should be noted that in this study total CH_4 emissions from the plants were measured. Wastewater treatment plants are more complex in structure than agricultural biogas plants, as they also have a water treatment operation in addition to sludge management and biogas production. Therefore, CH_4 emission rates measured at wastewater treatment biogas plants could also encounter CH_4 emissions from the water treatment line and from the open storage of sludge, which is more common at WWTPs in comparison to agricultural plants. However, at WWTPs, the main CH_4-emitting source will be biogas activities, even though CH_4 emissions can also occur from the plant inlet and from aeration tanks. Samuelsson et al. (2018) quantified CH_4 emissions from various unit processes at a WWTP and found that overall, about 81% of the CH_4 emissions quantified on site were released from the sludge treatment line. Delre et al. (2017) came to a similar conclusion based on on-site screenings of atmospheric CH_4 concentrations, where the highest elevated intensities were seen in the vicinity of sludge treatment activities. Sludge (un-digested or digested) storage in open tanks or basins can be a potential source of CH_4, which is challenging to quantify due to the large open surface area. At some of the WWTPs, open digestate storage of sludge could explain (but only partly) the higher emission rates. As an example, the average CH_4 loss at WWTPs with open storage was 9.2% in comparison to plants without on-site open storage (6.1%).

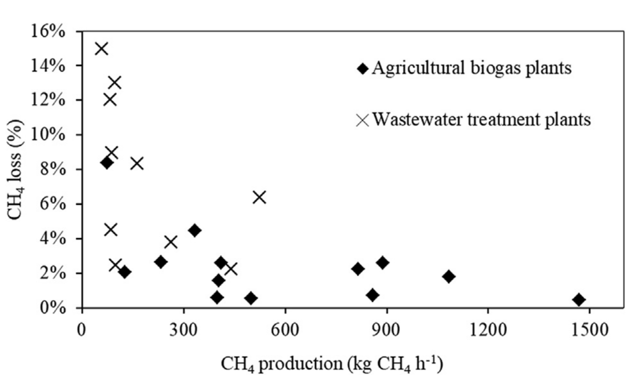

Fig. 1. Average CH_4 loss as a function of the average gas production at biogas plants.

Finally, it should be noted that at four of the plants (5, 11, 14 and 21) all gas utilisation occurs off site and at three of the plants part of the gas utilisation (~70–80%) occurs off site (plants 3, 4 and 7). For these plants, any CH_4 emission from the off site utilisation was therefore not included in the measured total CH_4 emission and thus the total CH_4 emission from the combined production and utilisation could be higher than the values reported in Table 4. At two of the plants (WWTPs 14 and 21) the generated biogas is routed off site for biogas upgrading. CH_4 emission factors from biogas upgrading units vary depending on technology applied. An average CH_4 slip of 0.81% was recently reported based on measurements of nine biogas upgrading units located in Denmark (Kvist and Aryal, 2019). The highest (1.97%) CH_4 slip was detected in the water scrubber methane upgrading technology, while the lowest (0.04%) CH_4 loss was detected in an amine based chemical scrubber (Kvist and Aryal, 2019). At five of the plants (agricultural plants 3, 4, 5, 7 and 11) the generated biogas is used (or partly used) in a CHP located off site. Liebetrau et al. (2013) found biogas co-generation units to emit on average 1.74% of the utilized methane with losses ranging from 0.40 to 3.28% (based on measurements at 10 biogas plants).

3.2. Contribution of methane emissions to the overall CO_2 footprint

Applying the methodology described in Section 2.4, the importance of various levels of CH_4 loss on the environmental performance of an agricultural biogas plant was assessed (Fig. 2). The impact in terms of GHG emissions (reported in CO_2-eqvivalents) of the different levels of CH_4 loss was assessed for three scenarios. In scenario A, biogas is upgraded to biomethane and substitutes for natural gas. In scenario B, biogas is utilised in a CHP unit, whereby electricity is supplied to the grid, and heat is used for district heating. In scenario B, the average emission factor regarding the production and consumption of electricity was used, as described in Section 2.4. In scenario C, biogas is also used in a CHP unit, but here the marginal emission factor for the production and consumption of electricity (Section 2.4) was used, meaning in this case that electricity production replaces more fossil fuel.

In all scenarios, CH_4 losses from biogas plants had a significant effect on the overall CO_2 footprint (Fig. 2). At 5% loss, CH_4 emissions make a greater contribution to the CO_2 footprint burden (positive CO_2 emission) compared to the other individual positively contributing emissions, namely energy consumption and the transportation of feedstock and digestate in all scenarios.

In scenarios A and B, a CH_4 loss of 20% caused the net GHG emissions to be positive, meaning that the biogas plant can be considered a net emitter of GHG, despite the substitution of fossil fuels, the reduction of GHG from manure storage and the substitution of chemical fertiliser. This is seen similarly in Table 5, where emission factors are listed for the three scenarios and five levels of CH_4 loss. These emission factors are the calculated net GHG emissions of the biogas production per one tonne of feedstock (wet weight) derived from the calculation example described in Section 2.4. The emission factors vary significantly in cases where CH_4 loss is relatively low (1–2%), to cases where the loss is relatively high (10–20%). The results also show that the emission factors in scenario B (CHP, average) vary highly in comparison to scenario C (CHP, marginal). The cause of this difference is the much lower electricity emission factor in the average mix of electricity sources (0.053 kg CO_2-eq. MJ^{-1}) compared to the marginal emission factor (0.24 kg CO_2-eq. MJ^{-1}) (Table 3).

The average CH_4 emission from the 13 agricultural biogas plants equated to 2.4% of the daily plant production (Table 4). Comparing this average CH_4 emission to implications on the total CO_2 foot-

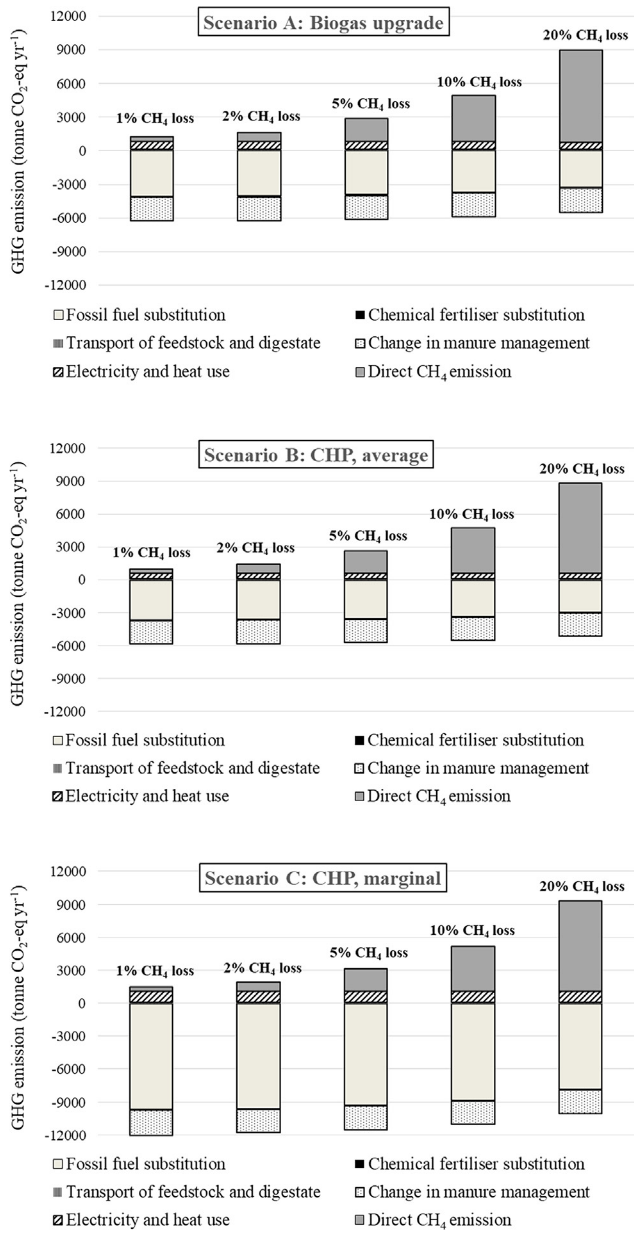

Fig. 2. Greenhouse gas (GHG) emissions calculated for an agricultural biogas plant, considering different biogas utilisation scenarios and five levels of methane (CH_4) loss. CHP: Combined heat and power.

print shown in Fig. 2, this relatively low loss indicates that the production of biogas is a net benefit with regards to GHG emissions. Since CH_4 emission rates compared to production varied greatly between biogas plants (Table 4), it is also likely that the CO_2 footprint of each individual plant will do so, too. Biogas plants, where the loss is particularly high (more than ~15%), may be net emitters of GHG, which underlines the importance of minimising CH_4 emissions from these facilities.

In this study, the carbon footprint of WWTPs was not determined, mainly because the primary purpose of a WWTP is not biogas generation but wastewater treatment, which implies that the services provided by the two types of plants are not comparable. Furthermore, not only CH_4 but also N_2O (another potent GHG) is emitted from WWTP,s primarily from the water treatment line, which needs to be included in footprint calculations. For an evaluation of the carbon footprint for biogas plants located at WWTPs, we instead refer to a recent study by Delre et al. (2019), which assessed carbon footprints for seven Scandinavian WWTPs, including some of the plants in this study. The study showed net carbon footprint values between 0.15 and 0.66 kg CO_2 eq. (Mg of input material)$^{-1}$, depending on the treatment facility. Direct CH_4 and N_2O emissions were the main contributors to the carbon footprint, accounting for between 44 and 71% of the total emission burden (Delre et al., 2019).

3.3. Fugitive methane emissions from Danish biogas production

Danish biogas producing facilities can be divided into four categories: agricultural (centralised and farm-scale) biogas plants (mainly treating manure), industrial biogas plants, wastewater treatment biogas plants (treating sewage sludge) and landfill gas. In total, 165 biogas-producing plants exist in the form of 82 agricultural (28 centralised and 54 farm-based), 51 wastewater treatment biogas, five industrial biogas and 27 landfill gas facilities (Danish Energy Agency, 2017b). The production of biogas has increased from 266 TJ (~5328 tonnes of CH_4) in 1990, to 7899 TJ (~157,985 tonnes of CH_4) in 2016 (Nielsen et al., 2018). In 2016, 86% of the generated biogas was based on manure/organic waste, 12% on sludge from wastewater treatment and only 2% came from landfills (Nielsen et al., 2018). Biogas production at the plants from which emissions were measured in this study represented about between 41% (agricultural) and 45% (WWTPs) of the annual Danish total (in 2016). National CH_4 emissions from Danish biogas production were estimated by applying the measured CH_4 emission factors to nationally generated CH_4 production, distinguishing between emission factors from agricultural and WWTP biogas plants, respectively. Two sets of CH_4 emission factors were used: a plant average and a weighted production average. The plant average was an average of CH_4 losses measured at the plants (sum of CH_4 losses divided by the number of plants), whereas the weighted production average was the sum of all CH_4 emission rates divided by the sum of all plants' CH_4 production rates (cf. Table 4). The plant average represents the biogas technology, whereas the weighted production average represents the combined biogas production in Denmark. Table 6 shows the estimated national CH_4 emissions (tonnes CH_4) for agricultural and WWTP biogas plants, and the total. The total estimated CH_4 emissions are between 3409 and 4683 tonnes, with emissions from agricultural biogas plants making up 6870%, while 30–32% originate from WWTPs (Table 6).

The 2006 IPCC Guidelines consider emissions from biogas plants (anaerobic digestion) as part of the waste sector. According to the IPCC Guidelines, emissions of CH_4 from biogas facilities, due to unintentional leakages during process disturbances or other unexpected events, will generally be between 0 and 10% of the

Table 5
Greenhouse gas emission factors (kg CO_2-eq tonne feedstock^{-1}) calculated for different biogas utilisation scenarios and five levels of CH_4 loss. A negative value implies an overall benefit to the environment, while a positive value implies an overall burden to the environment.

Scenario	1% loss	2% loss	5% loss	10% loss	20% loss
	(kg CO_2-eq tonne feedstock^{-1})				
Scenario A: Biogas upgrade	−44.6	−40.7	−29.0	−9.4	29.7
Scenario B: CHP, average	−42.1	−38.2	−26.5	−7.0	32.0
Scenario C: CHP, marginal	−89.7	−85.3	−72.0	−49.9	−5.7

Table 6
Estimated national CH_4 emissions from the anaerobic digestion of organic waste in agricultural biogas plants and biogas plants at wastewater treatment plants (WWTPs) in 2016 (excluding landfill gas). Numbers in brackets give the percentage out of total CH_4 emissions (excluding landfill gas).

Biogas plant type	Agricultural biogas plants	WWTP biogas plants	Total
CH_4 production, tonnes	135,867	18,958	154,825
CH_4 emission, tonnes (Plant average; $EF_{Agricultural}$ = 2.4% and EF_{WWTP} = 7.5%)	3261 (70%)	1422 (30%)	4683
CH_4 emission, tonnes (Production average; $EF_{Agricultural}$ = 1.7% and EF_{WWTP} = 5.8%)	2310 (68%)	1100 (32%)	3409

amount of CH_4 generated. In the absence of further information, a default value of 5% for the CH_4 emissions should be used (Eggleston et al., 2006).

CH_4 emissions from biogas production are reported in the Danish national greenhouse gas inventory as being a part of the waste sector's GHG emissions (Nielsen et al., 2018). CH_4 emissions were reported at 6635 tonnes in 2016, using an average adopted emission factor (EF) set equal to 4.2% for all types of biogas plants. This emission factor was based on a Danish project where CH_4 leakages were measured at nine biogas plants in Denmark, using on-site point measurement methods (Danish Energy Agency, 2015). Five of the plants were small, single-farm plants, while the other four were larger, centralised agricultural plants. The results were that the CH_4 losses varied from nil to 10% of production, resulting in a weighted average of 4.2%, which was adopted in the national inventory reporting for biogas production independently of the type of biogas plant. Our study shows a lower emission factor from agricultural plants, whereas the emission factor from biogas plants at WWTPs is higher than 4.2%. However, as the share of biogas generated at WWTPs is lower (12%) in comparison to agricultural plants (86%), the combined CH_4 emissions from these two types of facilities are almost comparable, resulting in a national emission of 3409 to 4683 tonnes of CH_4, which is close to the nationally reported figure.

The Danish Biogas Association is a trade organisation representing the Danish biogas sector, with members including plant owners, suppliers, agriculture and energy companies. Within the last few years, this organisation has initiated a voluntary measurement programme with the aim of keeping CH_4 loss at a minimum via a target of 1% loss for the sector. Our results indicate that some improvements are needed to reach this goal. However, the production weighed average loss was just 1.7% for the agricultural biogas plants, where most gas is produced and where production capacity is expanding, and thus the 1% target for the sector as a whole seems to be within reach. However, at plant level, emission rates are higher.

4. Conclusions

Methane losses were measured at 23 biogas plants and found to vary between 0.4 and 15.0% of the production total. Comparing those measured losses to an evaluation of the impact of methane loss on the overall carbon footprint of biogas production, it may be the case that methane loss is the largest positive contributor to greenhouse gas emissions for many biogas plants compared to other factors, such as energy use and the transportation of biomass.

Acknowledgements

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