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Research Paper

The Danish national effort to minimise methane emissions from biogas plants

Anders Michael Fredenslund [a,*], Einar Gudmundsson [b], Julie Maria Falk [a], Charlotte Scheutz [a]

[a] *Technical University of Denmark, DTU Sustain, Bygningstorvet, Building, 115, 2800 Kongens Lyngby, Denmark*

[b] *Rambøll, Hannemanns Allé 53, 2300 Copenhagen S, Denmark*

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ABSTRACT

In total, 69 biogas plants representing 59 % of Danish biogas production participated in a national effort to reduce methane (CH_4) emission. Measurements in terms of total plant CH_4 emissions, quantification of emissions from point sources, leak surveys and conceptual design plans to mitigate emissions were performed. Plant-level CH_4 emission rates varied between 1.3 and 81.2 kg CH_4 h^{-1}, and CH_4 losses expressed in percentages of production varied between 0.3 and 40.6 %. Agricultural plants generally had lower CH_4 loss rates compared to wastewater treatment plants. Biogas plants with a smaller gas production emitted a larger fraction of their production compared to larger plants, which was partly explained by the absence of gas collection from digestate storage tanks at smaller plants. A very commonly observed source of emission was pressure relief valves, where this source of leakage was observed at 53 % of the plants. A national emission factor (sum of CH_4 emissions/sum of CH_4 productions) was determined at 2.5 % for the Danish biogas production, whereof it was 2.1 % for agricultural biogas production and 6.7 % for biogas production at wastewater treatment plants. Measurements of total CH_4 emissions at six plants performed before and after implementation of mitigating actions showed that emissions were reduced by 46 % by carrying out relatively minor technical fixes and adjustments. An economic evaluation showed that, in some cases, mitigating actions could be economically beneficial for the biogas plant (positive net present value over a 10 year time frame), due to an increase in revenue.

1. Introduction

In recent years, biogas production has been incentivised in Denmark through state subsidies as part of a green transition initiative phasing-out fossil fuels in favour of renewable alternatives. In 2012, increase in subsidies for biogas production, aligned with the diversification of subsidies, were introduced, thereby helping in supporting Danish biomethane (bio-CH_4) production. This in turn led to a rapid expansion of biogas production in the country, exemplified by the share of bio-CH_4 in the Danish gas grid in relation to consumption increasing from 10 % in April 2019 to 30 % in September 2022 (Energinet.dk, 2022). Biogas production in Denmark is primarily based on manure, organic waste and sludge from wastewater treatment, while the use of energy crops is discouraged through the subsidy scheme (cap on share of energy crops – presently 8 % of biomass input). Organic waste for biogas production in Denmark includes source-separated household food waste, the amounts of which are increasing due to the implementation of the EU Waste Framework Directive, wherein anaerobic digestion and food waste composting are recognised treatment options enabling the recycling of nutrients (European Union, 2018).

The main components of biogas are the gases methane (CH_4) and carbon dioxide (CO_2), the former of which is a fuel that can be used for a variety of energy purposes. The production of biogas from manure and organic residues has several advantages in terms of environmental protection and climate change. The main advantages regarding climate change are the replacement of fossil fuels and the reduction of greenhouse gas (GHG) emission from manure storage, since digested manure has significantly fewer emissions when stored before field application compared to non-digested manure (Clemens et al., 2006; Kupper et al., 2020; Maldaner et al., 2018; Sommer et al., 2004; Vechi et al., 2022). Another very important advantage of biogas production in relation to climate change is that it can be accumulated in gas storage facilities. Storing biogas (upgraded to bio-CH_4) in a gas supply system helps to ensure security of supply in an energy mix with a high share of wind and solar power.

However, several studies have observed a high impact on the GHG balance of anaerobic digestion of the proportion of produced CH_4 emitted from production facilities into the atmosphere (“CH_4 loss”),

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[*] Corresponding author.

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since CH_4 is a potent GHG (Adams et al., 2015; Scheutz and Fredenslund, 2019; Delre et al., 2019). To ensure the potential GHG-mitigating benefits of increasing biogas production, it is thus important that the direct emission of CH_4 from biogas production is low. To date, only a few studies have documented total CH_4 emissions from biogas plants to an extent where national emission factors can be calculated (Scheutz and Fredenslund, 2019; Bakkaloglu et al., 2021). Recent developments in instrumentation and measurement methodologies have enabled such studies (Delre et al., 2017; Scheutz and Fredenslund, 2019; Hrad et al., 2022).

The objective of this study was to devise an inventory of measured CH_4 emission rates, comprising a large and representative list of biogas-producing facilities, including both agricultural (manure-based) and wastewater treatment plants. This paper describes the methodology behind this effort. It comprises the largest dataset to date regarding CH_4 losses from biogas production, sources of CH_4 emissions from biogas production and cost effectiveness of potential mitigation actions. Finally, it provides evidence of the effect of mitigation actions based on observations of CH_4 emissions at these facilities. The study was part of a Danish national effort to reduce methane loss from Danish biogas plants, which aimed at reducing CH_4 emissions resulting from biogas production as a part of a number of GHG reduction initiatives in the agricultural sector.

2. Materials and methods

2.1. Overview of sites and activities

Biogas production in Denmark is mainly done at two types of facilities: agricultural biogas plants and wastewater treatment plants. Agricultural plants typically utilise manure and organic waste – and to a lesser extent energy crops. Digester inputs are typically > 75 % manure (dry matter basis) with the remainder being mostly organic waste from industry and households. The digestate is used as fertiliser. Wastewater treatment biogas plants utilise sludge from wastewater treatment, and they may also include additional biomass such as source separated food waste. The digestate from wastewater treatment is sometimes used for fertiliser or incinerated, depending on pollutant content, including heavy metals and organic pollutants.

The plants in the study represented a wide range of Danish biogas plants in terms of plant age, plant type (agricultural plants, wastewater treatment plants), production capacity and the use of biogas in combined heat and power (CHP) units or biogas upgrade units. Overall, the study included 69 out of 144 existing plants, corresponding to a participation of 48 % (Table 1). Of the 69 plants, 44 were agricultural plants and 25 were wastewater treatment plants. Agricultural plants in the study represented 51 % of the nation's total (44 plants out of 87 in total) whereas wastewater treatment plants represented 44 % (25 out of a total of 57 plants) (Table 1).

The biogas plants included in the study were categorized according to size ("small", "medium" and "large"), based on records provided by the Danish Energy Agency. Looking at the representativeness of the participating plants in terms of CH_4 production, CH_4 emission measurements were carried out on 39 % of all Danish biogas plants in the CH_4 production size category "small and medium" (<3.1 million Nm^3 CH_4/year corresponding to < 250 kg CH_4 h^{-1}) and 59 % for those classed as "large" (>3.1 million Nm^3 CH_4/year corresponding to > 250 kg CH_4 h^{-1}) (Table 1). In general, agricultural biogas plants in Denmark are larger than wastewater treatment plants. This is reflected in this study, where only one of the wastewater treatment plants was classed large, whereas the rest where either medium or small. Overall, CH_4 emission measurements were carried out at plants covering 59 % of Danish biogas production.

At each plant, at least two of the following five activities were undertaken: agreement of activities to be performed, and alignment of expectations (activity 1), preparation of a self-control programme, and instructions for reducing leaks at existing installations (activity 2), surveys of leaks and individual emission point sources and recommendations for emission reduction (activity 3), measurement of total plant CH_4 emissions and emissions from selected point sources (activity 4) and overview of potential plant specific CH_4 emission reduction actions (activity 5).

Activities 1 and 4 above were done at all participating biogas plants, whereas activities 2, 3 and 5 were voluntary. Table 1 lists the biogas plants' participation in these five activities, divided into three CH_4 production size categories.

If a leak survey was performed at a biogas plant (activity 3), total CH_4 emission measurement was done at least three weeks after the plant received the results of its leak survey. This enabled repairs/process adjustments to mitigate CH_4 emissions prior to quantification. The relatively short time did not allow for more significant actions such as replacing process units etc. At six biogas plants, total CH_4 emissions were measured both before and after the plants performed emission-mitigating actions, to evaluate the effect.

The supporting information (SI) provides more detail on the participating facilities, including gas production and on-site gas utilisation (CHP, biogas upgrade).

2.2. Survey of leaks and individual on-site emission sources

Surveys of CH_4 leaks and individual on-site CH_4 emission sources were carried out using an infrared gas camera (FLIR GF320), which is designed for natural gas leak detection (Zimmerle et al., 2020). Filters in the camera helped visualise CH_4 emissions, similar to plumes of steam from leaks. The biogas plant's process units were systematically reviewed with the camera, and any leaks or sources of CH_4 emission were documented by recording film sequences, thereby making it possible to identify from where the CH_4 emission originated (Fig. SI1). A leak was defined as an unintentional CH_4 loss (i.e. due to technical or human failure) from gas bearing pipes and other containment such as gas-tight biomass storage tanks (including both substrate receiving and digestate tanks), digesters, piping and gas storage. In addition, other on-site sources of CH_4 emissions not caused by defects were identified, such as the gas escaping from digestate storage units without gas collection, biogas upgrade units, process ventilation, etc.

After completing a survey, the biogas plant was informed about the

Table 1

Overview of activities undertaken in the study on different types and CH_4 production sizes of biogas plants.

Activity	Number of participating plants, divided according to plant type			Number of participating plants, divided according to CH_4 production million Nm^3 CH4 yr^{-1} (kg CH_4 h^{-1})			
	Agricultural plants	Wastewater treatment plants	Total	<0.6 (<50) "small"	0.6–3.1 (50–250) "medium"	>3.1 (>250) "large"	Total
1	44	25	69	23	17	29	69
2	18	21	39	19	13	7	39
3	28	22	50	20	16	14	50
4	44	25	69	23	17	29	69
5	7	11	18	8	8	2	18

identified CH_4 leaks and point sources and suggestions for repairing them or reducing emissions. Leak surveys were, in general, completed within a working day, and thus emission from each leak or point source was only observed over a few minutes.

A gas camera provides visual information to assess the size of CH_4 emission from an individual leak or point source, but it does not quantify CH_4 emissions as applied here. If figures are requested for how much CH_4 is leaking from a specific point source, an additional measurement with special measuring equipment has to be carried out. For leak surveys, it is recommended that the gas camera survey is supplemented with a "sniffer", i.e. an instrument that can measure the concentration of CH_4 (in ppm-levels) in the air close to the source. Experienced measurement technicians can find leaks and other emission point sources with a "sniffer" that would otherwise be difficult to detect with a camera, regardless of whether they have a very small or a very large CH_4 flow.

Although leak surveys using the described method do not quantify emission rates from individual leaks and other emission point sources, they were categorised as "small", "medium" and "large", derived from observations of camera recorded plume sizes and concentrations of CH_4 measured near the identified leak or point source. Each of these characterisations was done by the operator based on their assessments and experience, whereby these categories are qualitative in nature. This categorisation was used to assist the biogas plants in prioritization of leaks and point sources for emission mitigation.

2.3. Measurement of total plant methane emissions

All measurements of total CH_4 emissions were done using the tracer gas dispersion method, which applies a continuous release of a gaseous tracer (here acetylene, C_2H_2) at the facility, combined with downwind measurements of atmospheric concentrations of CH_4 and tracer gas (Delre et al., 2017; Fredenslund et al., 2018; Reinelt et al., 2017; Scheutz and Fredenslund, 2019; Yoshida et al., 2014). The instrumentation used in this study was the same as used in the validation study reported in Fredenslund et al. (2019), namely a cavity ring down spectrometer (G2203, Picarro Inc., USA) or an off-axis integrated cavity output spectrometer (CH_4-C_2H_2 analyzer, Los Gatos Research, USA). Measurements were performed in accordance with the best practise protocol described by Scheutz and Kjeldsen (2019), and included screening ambient concentrations of CH_4 at each biogas plant and in the surrounding area. Screening at the facility provided information on approximately where the primary CH_4 emission occurred, and screening of the surrounding area was performed to ensure that measured elevated levels of CH_4 were not affected by nearby sources such as farms, landfills, composting facilities etc.

Tracer gas cylinders were placed close to the primary CH_4 emission area(s) of the plant, and CH_4 and tracer gas concentrations were measured while traversing the plume along a drivable road that crossed the downwind plume during the continuous release of tracer gas. The road had to be at an appropriate distance (typically > 500 m) away from the source, and no other CH_4 sources should be present between the biogas plant and the measuring path. The distance to the measurement path depended on the physical layout and area of the biogas plant and how much CH_4 was emitted. The distance to the measuring path should be at least 4–5 times the width of the emitting area of the biogas plant and preferably further away, as this would result in a greater mixing of CH_4 and tracer gas – and thus a better simulation of CH_4 emissions. However, measurements could not be made so far away that the elevated concentration levels would not be measurable.

At each plant, a minimum of 10 plume traverses were carried out, with a correlation coefficient R^2 between CH_4 and the tracer gas>0.80. Each plume traverse took approximately 1–2 min. In addition, the signal-to-noise ratio (SNR) was set to at least 10, where SNR is the ratio between measured peak concentration above background level and variability when measuring background air. This data quality criterion is recommended in a major U.S. study on the measurement method (Foster-Wittig et al., 2015). If the desired data quality criteria are achieved, uncertainty relating to the total CH_4 emission results will typically be<20 % (Fredenslund et al., 2019; Delre et al., 2018; Mønster et al., 2014). The total uncertainty of a measured total CH_4 emission is calculated as the square root of the sum of the squares of the uncertainty of the method (~15 %) and the variability of the measurement. The measurement method uncertainty is estimated as the square root of the sum of the squares of individual uncertainties (Gaussian law of error propagation). The variability of a measurement is calculated as the sample standard deviation of the calculated emission rates from the individual transects divided by the square root of the number of plume transects. For more detail on the measurement method, we refer to Fredenslund et al., (2019), Mønster et al., (2014) and Delre et al., (2018).

2.4. Methane emission measurement from selected point sources

CH_4 emission rates were measured at selected point sources, namely exhaust from gas engines, CO_2 exhaust from biogas upgrade units and process ventilation. Due to the different nature of the point sources, there was a higher degree of freedom when measuring CH_4 emission rates compared to surveys of leaks and individual on-site emission sources and measuring total CH_4 emissions.

The measurement principle was to determine gas flow from the source, using a pitot tube or similar, combined with measurement of CH_4 concentration in the gas stream using flame ionisation detectors, photoacoustic gas analysers or similar. CH_4 emission rates were calculated based on measured gas flow and CH_4 concentration. Examples of instrumentation for these types of measurements included pitot tube/micro manometer (Mano-Air 100, Schiltknecht, Switzerland) for determination of air flow, and photoacoustic gas analyser (INNOVA 1412, Lumasense Technologies, USA) for determination of CH_4 concentrations.

We note that this approach quantifies emissions from biogas upgrade unit's CO_2 exhaust and gas engine exhausts, but not leaks from other parts of these systems. For budgetary reasons, the measurements were carried out to the extent that measuring points were available on the measuring day without the use of lifts or the like. Therefore, not all of the biogas plants' potential point source emissions were measured.

2.5. Plant-level methane loss calculation

CH_4 loss was defined as the fraction of CH_4 produced by a biogas plant that is lost to the environment (CH_4 emitted/CH_4 generated). CH_4 generated at the individual plants was calculated on the basis of measured total CH_4 emission and gas production recorded at the plant on the relevant day of measurement. CH_4 loss was expressed as a percentage and was calculated as shown in eq. (1):

$$Methaneloss(\%) = \frac{Q_E}{Q_P + Q_E} \tag{1}$$

where Q_E is the measured total CH_4 emission from the plant (kg CH_4 h^{-1}) and Q_P is the recorded CH_4 production at the plant on the day of measurement (kg CH_4 h^{-1}) – typically determined as bio-CH_4 to grid or gas flow to CHP and/or off-site utilisation. The total CH_4 emission measured using the tracer gas method include CH_4 emissions from all possible sources, such as leaks, tanks, biomass storage, sludge dewatering, gas engine and biogas upgrade units, etc. The measured total CH_4 emission was added to recorded production (Eq. (1)), as most emissions are usually not included in the plants' measured gas production. For example, typical causes of leakage are pressure relief valves on reactors and emissions from storage tanks without gas collection. In both cases, CH_4 leakage/emission is not registered at the plant as produced biogas as these emissions occur before the gas production recording unit.

2.6. National methane emission factor calculation

National CH_4 emission factors were determined for future use in the national inventory reporting of GHG gas emissions. CH_4 emission factors were calculated as production-weighted CH_4 emission averages, in that emissions were weighted according to CH_4 production at the plants (Eq. (2)):

$$Emissionfactor(\%) = \frac{Q_{E1} + Q_{E2} + Q_{E3} + \cdots + Q_{Ez}}{Q_{P1} + Q_{P2} + Q_{P3} + \cdots + Q_{Pz}} \quad (2)$$

where Q_E is measured the total CH_4 emission (kg CH_4 h^{-1}) at each plant and Q_P is the CH_4 production (kg CH_4 h^{-1}). We note that since reporting in the national inventory of GHG emissions is based on recovered CH_4 production (and not total CH_4 production, i.e. $Q_E + Q_P$), measured emissions from the plant are not included in the denominator when calculating the emission factor. National emission factors were calculated for the total biogas gas production in Denmark and for the total production in agricultural biogas plants and wastewater treatment plants, respectively.

2.7. Plant specific methane emission reduction actions

CH_4 emission reduction actions were devised for 18 of the participating facilities, consisting of one or more of the following actions (more detail in SI): Longer retention time and increased biogas production; collection of biogas from biomass/digestate storage tanks; destruction of CH_4 from gas engines, upgrading systems and process ventilation and changes to sludge/substrate management including pre-treatment.

The conceptual design plans contained a short solution description, technical calculations, a description of expected GHG mitigation and economic assessment including calculation of capital investment and operating expenses and net present value (NPV) considering a 10- and 20-year time frame (NPV10 and NPV20). The result of the evaluation may yield a positive or a negative NPV, whereby a positive NPV is typically due to an increase in energy sales, which in some cases can finance both investment and running costs.

NPVs were calculated for the various actions using prices and technical data obtained from equipment suppliers, and the following input data: a discount rate of 2 %; a bio-CH_4 price of 8.5 DKK kg CH_4^1; an electricity price, sale of 1.25 DKK kWh^{-1}; an electricity price, purchase of 0.7 DKK kWh^{-1}; maintenance, engine: DKK 0.1 kWh^{-1}; and maintenance, other of 2 % of investment $year^{-1}$, where investment are costs associated with implementation of an action (construction, materials, planning etc.). We note that energy prices, and in particular gas prices rose significantly during finalisation of this study (spring 2022), whereby NPV for actions that increase energy sales can be higher at present energy prices.

For each conceptual design plan, a yearly GHG reduction estimate was calculated in ton CO_2-eq by considering a global warming potential of CH_4 of 28 ton CO_2-eq ton^{-1} CH_4.

3. Results and discussion

3.1. Observed leaks and individual emission point sources

In all, 473 individual leaks and point sources were identified at the 50 plants, where gas camera surveys were performed, varying between 0 and 38 leaks and point sources at each plant. The average number of leaks and point sources for both agricultural and wastewater treatment plants was 9.5, while it was higher for agricultural plants (13.5) compared to wastewater treatment plants (4.4). The reason for this difference may be that the agricultural plants in this study were, on average, larger (had a higher biogas production capacity) than the wastewater treatment plants with a higher number of digesters, tanks, etc. – and thereby the possibilities of leakage. Of the total number of leaks and point sources identified, 98 (equal to 21 %) were categorised as "large", 165 (equal 35 %) as "medium" and 210 (equal to 44 %) as small (further information including comparison of number of leaks to gas production and CH_4 emission/loss is provided in the SI).

Pressure relief valves were a commonly found cause of CH_4 emissions derived from the gas camera surveys, observed at 53 % of the facilities (Table 2). Pressure relief values have been found to be a common source of emissions at biogas plants and a study quantifying emissions from pressure relief valves found that these can be equivalent to 0.6–1.8 % of the CH_4 production (Reinelt et al., 2016; Reinelt and Liebetrau, 2020). These valves thus appear to be an important area of effort in terms of reducing CH_4 loss from biogas production.

Leaks were often observed at reactors (59 % of facilities) and at biomass tanks (incl. digestate tanks) (67 % of facilities) (Table 2). The cause of leakage varied, and included leaks at inspection hatches, pipe connections, in connection to wiring, leaks at soft covers and leaks near mixer axles. Leaks at other gas bearing components, as listed in Table 2 included leaks from gas engines, leaks from gas upgrade units, compressors, gas pipes, filters, pipe connections, and more, but does not include exhaust from gas engines or CO_2-exhaust from biogas upgrade units, as they were quantified separately.

Digestate tanks without gas collection ("open digestate tanks") were major sources of CH_4 emissions at many plants – especially at wastewater treatment plants and smaller agricultural biogas plants. Larger agricultural biogas plants have typically digestate storage with gas collection. Any detected leak in this regard is not due to defect, but is a consequence of the fact that the tanks are not designed for collecting gas. Very high concentrations of CH_4 (vol. % level) were measured near some of these tanks.

3.2. Total plant methane emission rates

Fig. 1 shows the measured total CH_4 emission rates for the plants included in the study where 44 were agricultural biogas plants and 25 were wastewater treatment plants (emission rates for all plants are listed in SI). Total CH_4 emission rates from the agricultural plants varied between 1.9 and 81.2 kg CH_4 h^{-1}, with average emissions at 14.4 kg CH_4 h^{-1} whereas total CH_4 emissions from the wastewater treatment plants varied between 1.3 and 28.2 kg CH_4 h^{-1}, with average emissions at 7.1 kg CH_4 h^{-1}.

The range of measured emissions (1.3 to 81.2 kg CH_4 h^{-1}) was larger than in a recent UK study reported in Bakkaloglu et al. (2021) at 0.02 to 58.7 kg CH_4 h^{-1}, and a study on Danish, Swedish and German facilities reported in Scheutz and Fredenslund (2019) at 2.3 to 35.5 kg CH_4 h^{-1}.

Table 2
Number and frequency of observed leaks/point sources grouped according to location. Times observed is the total number the type of leak was observed (several leaks were sometimes observed a single location (e.g. reactor). Frequency of observation is the fraction of the biogas plants, where the type of leak or point source was observed.

Location of observed leak/ point source of emission	Times observed	Frequency of observation among plants
Pressure relief valves	89	53 %
Reactors – other leaks[a]	100	59 %
Biomass storage tanks[b]	129	67 %
Gas storage units	7	12 %
Other gas bearing components[c]	137	65 %

[a] Including all leaks observed at reactors apart from pressure relief valves, like inspection hatches, pipe connections, leaks in connection to wiring, leaks at soft covers and leaks near mixer axles.

[b] Including digestate storage (with and without gas collection), mixer tanks and storage tanks for substrate.

[c] Including leaks from gas engines, biogas upgrade units, compressors, gas pipes, filters, pipe connections, and more, but not engine exhaust or CO_2 exhaust from biogas upgrade units.

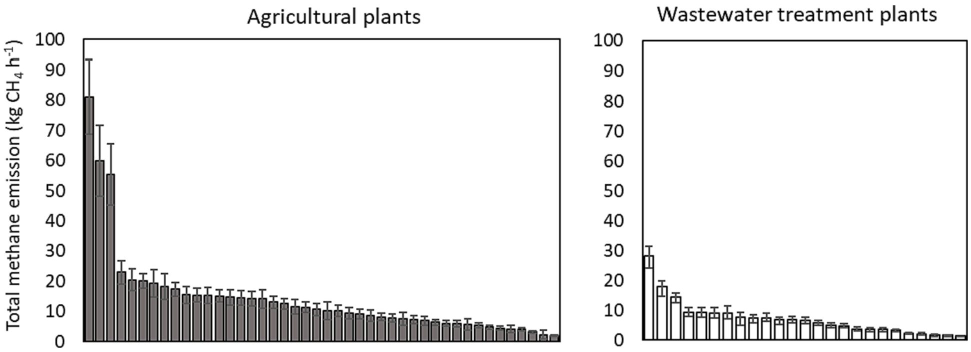

Fig. 1. Measured total CH_4 emission rates (kg CH_4 h^{-1}) at 69 Danish biogas plants including 44 agricultural plants and 25 wastewater treatment plants. Error bars show the total uncertainty of the quantification including method uncertainty and measurement variability.

One reason for these differences may be the relatively high number of facilities included here. Emission rates measured for wastewater treatment plants (1.3 to 28 kg CH_4 h^{-1}) were lower than measured at the wastewater treatment plant reported in Samuelsson et al. (2018), where an emission rate between 28.5 and 33.5 kg CH_4 h^{-1} was measured using several methods.

As shown in Fig. 1, total CH_4 emissions for three agricultural biogas plants were above 50 kg CH_4 h^{-1}, whereas for all other plants they were below 30 kg CH_4 h^{-1}. The highest total CH_4 emission rate was 81.2 kg CH_4 h^{-1}. This plant was newly constructed, the soft cover installation on the digesters was not tested and therefore not completed. It is likely that this caused higher than normal emissions, since a measurement performed later at the facility, showed a much lower emission (15.2 kg CH_4 h^{-1}). The second highest emission (60.0 kg CH_4 h^{-1}) was measured at the biogas plant with the highest gas production in this study, i.e. 2,583 kg CH_4 h^{-1}, which is more than five times the average gas production (478 kg CH_4 h^{-1}) of the studied plants. The third highest emission (55.5 kg CH_4 h^{-1}) was measured at the facility where the largest number of leaks and point sources (38 in total) was observed. At this facility, which is a relatively large, agricultural plant, digestate was stored in tanks without gas collection, which is atypical for a plant of this kind, and digestate storage without gas collection is a potentially significant source of CH_4 emission.

3.3. Point source methane emission rates and losses

At 11 plants, CH_4 emissions from the CO_2 exhausts from upgrading plants were measured. Five of these plants had amine scrubbers, three had water scrubbers and three had both types of biogas upgrade units. CH_4 emissions from the amine scrubbers varied between 0.2 and 1.8 kg CH_4 h^{-1} with an average emission of 0.6 kg CH_4 h^{-1}. At one location, influx (754 kg CH_4 h^{-1}) was measured, with emissions from this unit corresponding to 0.05 % of influx. Emissions from the water scrubbers varied between 6.6 and 16.1 kg CH_4 h^{-1} with an average emission of 8.9 kg CH_4 h^{-1}. Kvist and Aryal (2019) investigated CH_4 emissions from commercially-operating biogas upgrading plants, where CH_4 losses from water scrubbers were up to 1.97 % of influx, while from amine scrubbers they stood at 0.04 %, which is in line with our results regarding the amine scrubber.

CH_4 emissions were measured from 23 gas engines, and CH_4 influx was measured at 14 of these. CH_4 emission rates for the engines varied between 0.1 and 4.7 kg CH_4 h^{-1}, and CH_4 losses corresponded to an average of 1.4 % of influx from the 14 engines where influx was measured. The CH_4 loss range was 0.7 to 3.0 %. These values are comparable to results in Liebetrau et al. (2013), who found an average CH_4 loss of 1.74 % for gas utilization at 10 plants and a range of 0.40 to 3.28 %.

Quantifying CH_4 emissions from process ventilation was done at five plants. The highest emissions were measured at process ventilation from two sulphur removal units at biogas plants 9 and 11, recording 13.3 and 39.4 kg CH_4 h^{-1}, respectively. At both of these units, mitigation actions were taken, and total plant CH_4 emissions measured thereafter were lower than the sum of the point source emissions quantified before mitigating actions. At plant 11, where the emission rate as a result of sulphur removal was measured at 39.4 kg CH_4 h^{-1}, and emissions from biogas upgrade units were 10.6 kg CH_4 h^{-1}, total CH_4 emissions were later measured at 20.2 kg CH_4 h^{-1} indicating a reduction of about 50 %. Emissions from ventilation from gas storage was measured in three instances, with emission rates ranging between 0.1 and 4.0 kg CH_4 h^{-1}. Measured emission rates from the different point sources at the studied biogas plants are listed in the SI.

3.4. Plant-level methane losses

Plant-level CH_4 losses ranged between 0.3 % and 40.6 % where higher loss rates were observed for relatively small biogas plants compared to larger ones (Fig. 2). CH_4 loss appeared to increase exponentially with lower production rates, and the measured loss rates were related to production by exponential fits at $y = 0.120 \cdot e^{-0.003x}$ (wastewater treatment plants) and $y = 0.054 \cdot e^{-0.001x}$ (agricultural plants), where y is CH_4 loss in % and × is CH_4 production in kg CH_4 h^{-1}. Agricultural plants generally had lower loss rates (4.7 % on average) compared to wastewater treatment plants (11.3 % on average) (Fig. 2). Several reasons may explain these differences. Many of the larger agricultural biogas plants in Denmark are either newly constructed or are older plants that have undergone reinvestment and expansion. In general, digestate storage at these facilities is done in tanks with gas collection and utilisation, which is not normal practice at wastewater treatment and smaller agricultural plants. Many of the wastewater treatment plants are old and were built with the primary aim to treat wastewater. Only recently, it has become a goal for the plant operators to also become net energy producers and climate neutral.

Open digestate storage tanks are a known, significant source of CH_4 emission at biogas plants (Bakkaloglu et al., 2022; Baldé et al., 2016; Daniel-Gromke et al., 2015; Liebetrau et al., 2017, 2013; Reinelt et al., 2017). Eliminating this source of emission may be the cause of the relatively low CH_4 loss for the larger facilities shown in Fig. 2, as the larger facilities most often had digestate storage with gas collection. However, other factors may also contribute in this regard, such as higher staff levels, more frequent control and maintenance and more economic power to invest in new technology and mitigation actions at larger facilities.

The range of plant-level CH_4 losses for wastewater treatment plants was 2.0 to 39.4 %. The upper range was higher than reported in Delre et al. (2017) and Yoshida et al. (2014) – 21 and 32.7 % of gas production, respectively, which may be due to the larger number of facilities in our study. The range of plant-level CH_4 losses for agricultural plants was 0.3 to 40.6 %. The upper range of plant-level CH_4 losses from

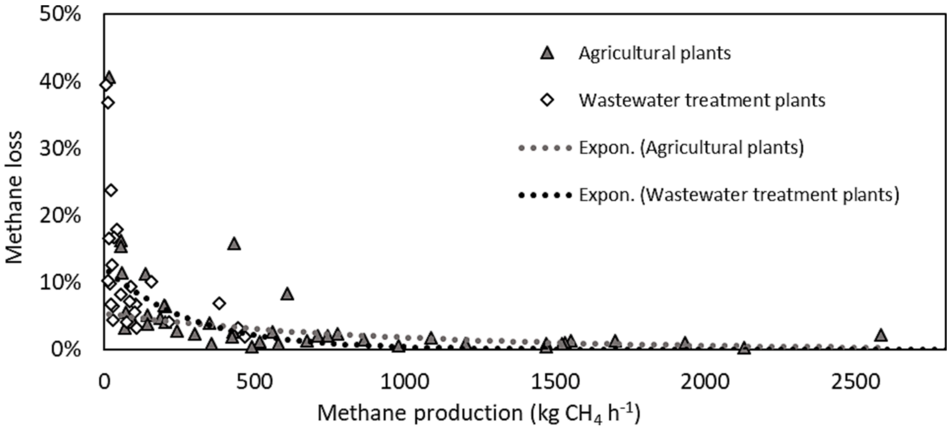

Fig. 2. CH_4 losses (%) plotted as a function of CH_4 production. Dotted black line shows exponential fit regarding wastewater treatment plants ($y = 0.120 \cdot e^{-0.003x}$, $R^2 = 0.38$), whereas dotted grey line shows exponential fit ($y = 0.054 \cdot e^{-0.001x}$, $R^2 = 0.42$) regarding agricultural plants.

agricultural plants was higher than reported in Scheutz and Fredenslund (2019) – 14.9 %, which again may be explained by a larger number of facilities here.

There were three plants (one agricultural and two wastewater treatment plants) where the measured CH_4 loss was higher than 35 %. Common to these three plants was that their gas production was low, namely<20 kg CH_4 h^{-1} (average production in this study: 478 kg CH_4 h^{-1}). The highest losses were thus measured at very small producers, which did not have gas collection on their digestate storage tanks. At one of the two wastewater treatment plants, a fairly high concentration of CH_4 in process ventilation from sludge storage, without gas collection, was observed (3.3 vol%), thereby indicating significant emissions from this source. At the second of the two wastewater treatment plants with particularly high emissions, very high emissions were detected at the gas storage unit, indicating defect. At the agricultural plant, where the highest CH_4 loss was measured, no leak search was carried out, and the cause of the relatively high CH_4 loss is therefore unknown.

In spite of the relative high CH_4 losses observed at many of the plants, the study showed that it is technical possible for biogas plants to limit plant-level CH_4 losses to<1 %. Low CH_4 losses were mainly seen for large centralised agricultural biogas plants with a CH_4 production higher than ~ 200 kg CH_4 h^{-1} (Fig. 2). This is most likely because most of these plants are newly constructed or renewed, driven very professionally with the focus of producing energy. Also, CH_4 loss from agricultural biogas plants has been in focus by the organization of agricultural biogas producers in Denmark as well as the Danish Energy Agency for several years. In comparison, wastewater treatment plants reacts mostly to government regulation, which does not include fugitive emissions of GHG from biogas production at this moment.

3.5. Short-term methane emission rate variation

At one wastewater treatment plant, total CH_4 emissions were measured repeatedly over six days to observe variations in the emission rate. The measurements were performed either once per day (midday between ~ 12 pm and ~ 2 pm) or twice per day (between ~ 8:30 and 11 am and later between ~ 6 pm and ~ 9 pm).

We observed some variations in the measured emission rate (Fig. 3). Average emissions during the period were 5.4 ± 1.6 kg CH_4 h^{-1}, while the maximum and minimum measured rates were 7.8 and 3.3 kg CH_4 h^{-1}, respectively. Even though our study on emission rate variation over time was performed within a week, the observed variation in emission was similar to what was observed in the longer term study in Hrad et al. (2015). Here, CH_4 emissions at an agricultural biogas plant varied

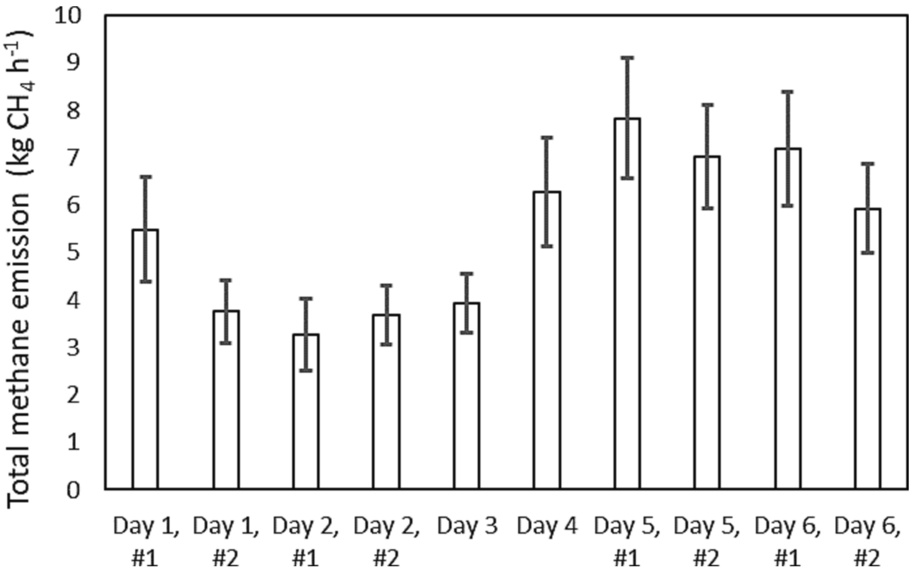

Fig. 3. Measured total CH_4 emissions at a wastewater treatment plant including both water treatment, biogas production and sludge storage over a period of 6 consecutive days during February 2021. Day 1 was a Thursday.

between ~ 4.2 and 8.3 kg CH$_4$ h^{-1} in 14 measurements performed over more than a calendar year. Higher emission rates were measured during periods with full digestate storage tanks compared to empty tanks (Hrad et al., 2015). Short-term variations (variations over a few hours) in emission rates were also observed in studies by Hrad and co-workers, where spikes in emission rates coincided with the agitation of digestate (Hrad et al., 2022, 2015).

The causes of the short-term variations observed in our study were not determined. The biogas plant's gas engine was operating during all measurements, and the digestate storage tanks were not emptied during the six-day period. No operating conditions other than those normally carried out were reported by the biogas plant.

3.6. National methane emission factors

The national emission factor for agricultural plants was 2.1 % (Table 3), which is considerably lower compared to the emission factor for wastewater treatment plants of 6.7 %. This was primarily due to the relatively low CH$_4$ loss observed at the larger, agricultural plants as mentioned above. The national emission factor for all biogas producing plants was 2.5 %, which is close to that of agricultural plants, since most of the gas is produced at those facilities, both in the sector as a whole in Denmark and by the biogas plants in this study.

The emission factor for wastewater treatment plants (6.7 %) was higher than what was submitted under the United Nations Framework Convention on Climate Change and the Kyoto Protocol: 1.3 % (Nielsen et al., 2021), which is based on a COD mass balance approach to calculate emission. In contrary, the emission factor for agricultural biogas plants (2.1 %) was lower than what is currently applied in the Danish emission inventory: 4.2 % (Nielsen et al., 2021), where the emission factor has been determined from a previous study, where CH$_4$ emissions were measured at 9 Danish biogas plants (Kvist, 2016).

The Danish Biogas Association has set a target of 1 % CH$_4$ loss from the Danish biogas sector, which is ambitious but technically possible - at least at large agricultural biogas plants as shown in this study (section 3.4).

3.7. Observed effects of implemented mitigation actions

At six biogas plants, total CH$_4$ emissions were measured before and after mitigating actions to investigate the effect of these actions. The period between measurements was a few months, which allowed for minor technical fixes and adjustments, but not major reinvestments. All six plants were relatively large agricultural units (plant CH$_4$ productions of 204–2000 kg h^{-1}). Fig. 4 shows CH$_4$ losses determined before and after mitigating actions. At all plants, CH$_4$ losses were lower after mitigating actions. The sum of emissions (all plants) beforehand was 264 kg CH$_4$ h^{-1}, whereas thereafter it was 143 kg CH$_4$ h^{-1} corresponding to a reduction of 46 %. Converted into CO$_2$ equivalents, this difference in direct emissions amounts to approximately 29,400 tons of CO$_2$-eq., considering a global warming potential (GWP) of 28 ton CO$_2$-eq ton CH$_4^{1}$ for CH$_4$. The difference is also equivalent to approximately 1.5 million m^3 CH$_4$ yr^{-1}.

According to the owners of the facilities, some of the pressure relief valves (closing liquid types) were replaced with mechanical types with weight-loaded elements, which they had found to reduce leakage. Other actions were directed at leakages found on gas-bearing components, and the owners have subsequently invested in leak detection equipment to form of a regular self-control programme. The plant operator targeted the leaks identified during the camera survey. New leaks could have occurred after the leak survey and in this case, the reduction due to the repair of the located leaks is underestimated.

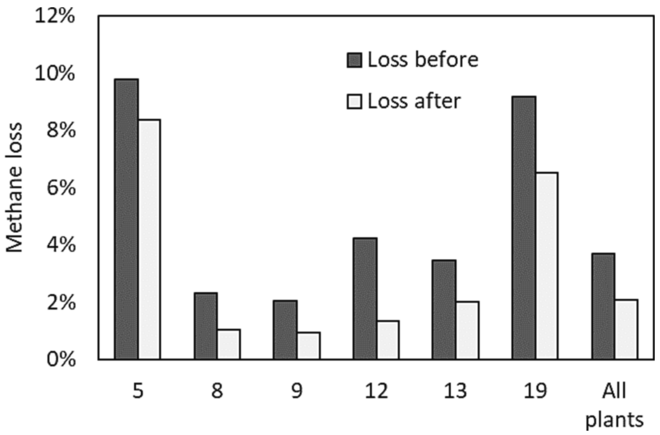

Fig. 4. Plant-level CH$_4$ losses, before and after mitigation actions. Numbers on the x-axis refer to plant ID, as listed in SI.

At plant 5, CH$_4$ losses were reduced, but they were still relatively high after mitigating actions at approximately 8 % (Fig. 4). This plant had digestate storage without gas collection, which is unusual for large agricultural biogas plants in Denmark. The owner of this facility has since decided to invest in gas collection at the digestate storage tanks, but this course of action had not been realised in the relatively short period between the two measurements. Similarly, plant 19 had digestate storage without gas collection, which was not changed in the period between measurements.

Table 3
National emission factors (sum of emission/sum of production) for agricultural plants, wastewater treatment plants and both types determined in this study.

Plant type	Number of plants	Sum of measured total CH$_4$ emissions (kg CH$_4$ h^{-1})	Sum of gas production (kg CH$_4$ h^{-1})	National emission factor (%)
Agricultural	44	633.5	29,963	2.1
Wastewater	25	170.4	2,560	6.7
All	69	803.9	32,523	2.5

3.8. Potential plant specific methane emission reduction actions

Overviews of potential plant specific CH$_4$ emission reduction actions were developed for 18 of the participating biogas plants, considering different strategies to counter CH$_4$ emissions, as listed in SI (Table SI1 and SI2). Reduction actions involved investments varying in size – from zero to approximately 22.5 million DKK (~3.0 million Euro) (Table 4). For many of the plants, actions taken would increase revenue due to a rise in energy production. A further financial benefit for the wastewater treatment plants would be potentially reduced costs for sludge disposal. For these reasons, thermal hydrolysis (as a sludge pre-treatment), according to our analysis, would be financially viable over the long term (positive NPV20) in spite of high investment costs. We note that biogas production is subsidised in Denmark, and this may not be the case in all markets.

Open digestate storage without gas collection was, as mentioned, a frequently found source of CH$_4$ emissions from biogas plants. Five action plans included gas-tight coverings for digestate/sludge storage tanks along with gas collection and utilisation, which will generate income. In four of five cases, the NVP20 was positive, whereof NVP10 was positive in two cases, meaning that for most plants, implementing gas collection and utilization on digestate storage can pay for itself over a 20-year timespan according to these results. In the economic evaluations described in Agostini et al. (2016), it was similarly found that gas collection and utilisation from digestate storage can be economically beneficial for the biogas plant, due to an increase in revenue.

Since the finalisation of this study, the Danish as well as the European gas and electricity prices have increased (by almost three fold), which increase the calculated NPV10 and NPV20 values implying that

Table 4
Summaries of conceptual design plans developed for 18 biogas plants. Each row in the table concerns one biogas plant, where one more actions are proposed.

Plant type	Brief description of action(s)	Investment (million DKK)	NPV10 (million DKK)	NPV20 (million DKK)	Reduced GHG (ton CO_2-eq/year)
Agricultural	Installation of regenerative thermal oxidation (RTO) to eradicate CH_4 emissions from a water scrubber.	7.3	–	–	14,000
Agricultural	Gas pipe from the biogas upgrade unit to gas storage, which will reduce periods of flaring when raw biogas does not meet requirements on CH_4 content.	1.3	0.2	1.4	600
Agricultural	Increase in reactor volume, increasing hydraulic retention time from 62 to 80 days. Gas production is expected to increase by 5 %. Investment and operating costs not determined.	–	–	–	30,000
Agricultural	Increase in reactor volume by 4,000 m^3, increasing hydraulic retention time from 18 to 34 days + reinvestment in gas engine. Gas production is expected to increase by 6 % considering the mix of substrate and type of plant.	6.4	6.1	15.6	5,500
Agricultural	Replacing part of the substrate with pre-treated straw. Investment includes plant to pre-treat straw at 70 °C.	22.5	10.0	33.6	17,000
Agricultural	Gas-tight covering on digestate storage with gas collection and utilisation (CH_4 recovered: 8,500 kg CH_4/yr).	0.54	−0.067	0.063	230
Agricultural	Increase in digester volume to enhance process stability.	–	–	–	–
Wastewater	Gas-tight covering on sludge storage with gas collection and utilisation (CH_4 recovered: 10,300 kg CH_4/yr).	0.22	0.9	1.5	290
Wastewater	Conversion from mesophilic to thermophilic operation with resulting increase in gas production.	1.0	0.39	1.6	1,400
Wastewater	Thermal hydrolysis pre-treatment of sludge. Investment resulting in reduced costs for further treatment of sludge and an increase in gas production.	14	−9.1	−4.9	1,400
Wastewater	Thermal hydrolysis pre-treatment of sludge. Investment resulting in reduced costs for further treatment of sludge and an increase in gas production.	14	−3.3	0.7	4,100
Wastewater	Thermal hydrolysis pre-treatment of sludge and gas-tight covering on sludge storage with gas collection and utilisation.	15	−1.4	11.3	5,900
Wastewater	Decommissioning of old biogas plant at a small waste water treatment plant, and transport of sludge to nearby facility with better possibility of utilising CH_4 potential.	0.82	−0.14	0.33	170
Wastewater	Thermal hydrolysis pre-treatment of sludge. Investment resulting in reduced costs for further treatment of sludge and an increase in gas production.	14	−10	−7.0	3,700
Wastewater	Installation of solid separation at industry up-stream of wastewater treatment plant. Bio-solids sold to an agricultural biogas plant for better utilisation of CH_4 potential. The load to the wastewater treatment plant is reduced.	3.3	14.3	26.0	Not calculated
Wastewater	Gas-tight covering on sludge storage with gas collection and utilisation (CH_4 recovered: 1,500 kg CH_4/yr).	0.45	−0.30	−0.39	42
Wastewater	Decommissioning of old biogas plant, which was found to leak biogas, and transport sludge to a nearby facility with available reactor capacity.	0	1.1	1.9	Not calculated
Wastewater	Gas-tight covering on sludge storage with gas collection and utilisation.	0.10	0.11	0.53	220

reducing CH_4 losses from biogas could be even more economically favourable depending on the change in energy sales and energy consumption resulting from the actions.

4. Conclusions

Total methane (CH_4) emission rates measured at 69 Danish biogas plants varied between plant types (agricultural, wastewater) and within plant types, and we observed a tendency of smaller facilities to emit a larger fraction of gas produced compared to the larger ones. The national emission factor was 2.5 % of the Danish biogas production, where agricultural plants had an emission factor of 2.1 % and wastewater treatment plants had an emission factor of 6.7 %. Several factors may explain the difference between plant types – including a focus on CH_4 loss at the agricultural plants over several years. Measurements of total CH_4 emissions, leak searches and point source emission quantification can provide sufficient information for biogas plants to significantly reduce their CH_4 losses. This study provides evidence that six plants lowered their CH_4 emission 46 % through applying relatively simple mitigating actions. Further mitigating actions were outlined at 18 plants including calculations of costs and revenues. Many of these mitigation actions were economically beneficial, due to an increase in revenue. Those included gas tight covering of digestate storage with gas collection and utilisation, where 4 out of 5 cations had a positive NVP 20 years.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2022.12.035.

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