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On-site and ground-based remote sensing measurements of methane emissions from four biogas plants: a comparison study

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Abstract

Methods for quantifying methane (CH_4) emissions from biogas plants are needed, in order to ensure that emissions are within acceptable levels and to identify options for emission mitigation. Two emission measuring approaches were used at four biogas plants: an on-site approach, whereby emission sources were identified and subsequently quantified one at a time, and a ground-based remote sensing approach, which was applied to measure total CH_4 emissions. The emissions were between 5.5 to 13.5 kg CH_4 h^{-1} from the four plants, measured using ground-based remote sensing. Even though the measurements were performed on the same days at each facility, the sum of on-site emission rates varied between the remote sensing measurements (up to ~100%). Several factors may have caused this difference: emission sources not measured using an on-site approach and

short-time emission variation. On-site measurements showed that the majority of the emissions often occurred from just a few sources.

Keywords

Method comparison, anaerobic digestion, fugitive emissions, tracer gas dispersion method.

1. Introduction

Methane gas (CH_4) is an important fuel used for electricity production, heating and transportation purposes. Most of the CH_4 used for fuel purposes today originates from natural gas. In transitioning away from the use of fossil fuels, biogas plants may provide a valuable alternative, thereby reducing greenhouse gas (GHG) emissions through fossil fuel substitution. The anaerobic digestion of animal slurry also reduces CH_4 emissions from manure storage (Clemens et al., 2006; Sommer et al., 2004), and so biogas production has several GHG mitigation effects.

CH_4 emissions from biogas plants reduce net GHG benefits. Data on the magnitude of these emissions are sparse, though, so there is some uncertainty with regards to the environmental assessment of biogas production vis-à-vis global warming (Meyer-Aurich et al., 2012; Møller et al., 2009).

In one case study, using an inverse dispersion measuring technique over four campaigns, average CH_4 emissions from an agricultural anaerobic digester were 3.1 % of gas production (Flesch et al., 2011). The inverse dispersion method involves downwind, cross-plume measurements of atmospheric CH_4 concentrations, wind measurements and dispersion modelling, to estimate CH_4 emission rates. CH_4 emission rates from the digester were found to vary, seemingly according to operational conditions at the facility, where periods of relatively high emissions occurred during periods of biogas flaring (Flesch et al., 2011). Using a similar measurement methodology in another

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study, CH_4 emission rates measured for a German digester varied between ~7.2 and 57.6 kg CH_4 h^{-1} (or between 3 and 23% of gas production) over a period of little more than two hours (Groth et al., 2015). The highest emission rates coincided with opening the digester's pressure relief valve, while during normal operation, the CH_4 emission rate averaged 4% of production. Similarly, Reinelt et al. (2016) found pressure relief valves to be important sources of CH_4 emissions at two biogas plants. Furthermore, Liebetrau et al. (2013) monitored CH_4 emissions from 10 German biogas plants, where individual leaks were first identified and subsequently quantified using a variety of techniques, including chamber measurements. It was established that open digestate storage tanks and cogeneration units are important sources of CH_4 emissions – a finding supported by Baldé et al. (2016)'s study, in which CH_4 discharges from an open digestate storage unit were monitored over a two-year period, with emissions corresponding to 12 % of biogas production.

A comparative measurement campaign was performed at a biogas plant, using several on-site and remote sensing methods simultaneously to determine CH_4 loss, by four on-site measurement teams and two remote sensing teams (Reinelt et al., 2017). In that study, large but short-term variations in emission rates were observed, and CH_4 emission rates varied between 5 and 25 kg CH_4 h^{-1} (remote sensing measurements) and 5 and 17 kg CH_4 h^{-1} (on-site measurements). These variations were explained as resulting partly from changes in the operational state of the biogas plant, and the main sources of emissions were found to be an open digestate storage tank and a pressure release valve (Reinelt et al., 2017). Unfortunately, on-site and remote sensing measurements were not done on the same days, and so the paper recommended that future measurement studies comparing on-site and remote sensing measurements should be carried out simultaneously.

In this paper, we used on-site measuring methods to quantify CH_4 emissions from four Danish biogas plants, combined with measurements of total CH_4 emissions from the plants using a ground-

based remote sensing technique. At each plant, one measurement team measured one source at a time, while another team used a remote sensing method on the same day.

2. Materials and methods

2.1 Site descriptions

2.1.1 Biogas plant A

Biogas plant A is a centralised, agricultural-based plant utilising ~225,000 tons yr^{-1} biomass to produce ~3.4 million kg CH_4 yr^{-1} of biogas. Feedstocks include manure (~85% w/w) as well as organic, industrial surplus such as slaughterhouse waste. The biogas plant was constructed in 1989-1990, and a major reinvestment was executed in 2013-2014 to modernise the facility and to expand production capacity. The biogas plant has four continuously stirred reactors (CSTRs) with a combined volume of 12,950 m^3. All digestion units are operated at a thermophilic temperature (52°C) and are made of steel. There are also five biomass storage and pre-treatment tanks with a total volume of 3300 m^3. The biogas is used in a combined heat and power (CHP) unit at the site (1.35 MW) and in two CHP units (2 × 2.65 MW) at a power plant off-site (Table 1). All CHP units are gas engines. The digestate is stored in a 4300 m^3 closed concrete tank with gas collection and an integrated 3500 m^3 gas storage. Other parts of the biogas plant include a 200 m^3 closed mixing tank, a 1500 m^2 dry biomass storage building, two gas bag type, double membrane biogas storage units (combined volume: 4500 m^3, operated at slight overpressure), a gas flare and a180 m^2 biofilter for odour reduction with a capacity of 22,000 m^3 air h^{-1}. The mixing tank and dry biomass storage were covered, but not gas tight. No measurement of CH_4 emissions from the facility had been performed prior to this study. Leakage from the gas storage unit was suspected by the plant operator prior to the measurement campaign.

2.1.2 Biogas plant B

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Biogas plant B utilises sludge from wastewater treatment to produce ~1.4 million kg CH_4 yr^{-1} of biogas, and it is a part of a wastewater treatment facility receiving wastewater from 265,000 people. The facility was constructed in 1965 and then expanded and renovated in 1995. It has four CSTRs made of concrete with a total volume of 24,000 m^3. Digestate is led to four 250 m^3 open storage tanks, and the digestate is de-watered and incinerated. The biogas is utilised in an on-site CHP unit (gas engine). Other parts of the biogas plant include a gas bag type biogas 1400 m^3 storage unit and a gas flare. Total CH_4 emissions had previously been measured at the facility, using the same measurement method and equipment as in this study (Yoshida et al., 2014). Emissions were found to vary significantly between the nine performed measurement campaigns. Short-term variations in emissions (i.e. variations during measurement campaigns) were also observed. Measured total CH_4 emissions were between 5 kg CH_4 h^{-1} and 92 kg CH_4 h^{-1}, with the highest emission rates observed during foaming events in the digesters. The biogas plant was found to be the most significant source of CH_4 emission compared to other parts of the wastewater treatment facility such as aeration tanks and primary/secondary clarifiers.

2.1.3 Biogas plant C

Biogas plant C is a centralised, agricultural-based biogas plant utilising ~240,000 tons yr^{-1} biomass to produce ~4.6 million kg CH_4 yr^{-1} of biogas, which is upgraded to bio-methane on the site, using a water scrubber. No post-treatment of the upgrading exhaust was present. Feedstocks include manure and slaughterhouse waste. The biogas plant was constructed in 2013-2014, and at the time of this study it was being expanded – including the installation of a new biogas upgrade facility, due to increased production. It has three continuously stirred 800 m^3 CSTRs (step 1) and a 2200 m^3 CSTR (step 2). The reactors are closed steel tanks. Digestate is stored in two closed concrete tanks with gas collection with a combined storage capacity of 2200 m^3. Other parts of the biogas plant include a mixing tank, a biomass hygienisation unit, a biological odour reduction unit and a gas flare. The

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upgraded bio-methane is injected into the natural gas grid. No CH_4 emission measurements had been performed prior to this study. Staff at the facility performs weekly visual inspections of the installations, to reduce gas leakage. The operator of the facility suspected the biogas upgrade facility to be emitting CH_4 at the time of measurement, since its production had exceeded the designed capacity of the upgrade unit.

2.1.4 Biogas plant D

Plant D is also a centralised, agricultural-based facility and utilises ~118,000 tons yr^{-1} biomass to produce ~2.9 million kg CH_4 yr^{-1} of biogas. Feedstocks include manure (~85% w/w), straw and maize silage. The biogas plant was constructed in 2014 to operate as a two-step system, where most of the gas production occurs in two CSTRs with a combined volume of 9200 m^3. From these tanks, the biomass is pumped into two secondary digesters, which also have a combined volume of 9200 m^3. All reactors are made of concrete and have double-walled, insulated soft covers. Digestate is stored in three storage tanks equipped with stirrers, with a collective total volume of 13,800 m^3. The digestate storage tanks are also made of concrete and have double-walled, insulated soft covers and gas collection. The biogas plant operates at a thermophilic temperature (54 °C), and biogas is upgraded to bio-methane, using chemical absorption in an on-site amine scrubber unit, which is then injected into the natural gas grid. Other parts of the biogas plant include walled bunker silos for maize silage, a mixing tank, a gas flare, an odour reduction unit (biofilter) and a biomass storage building. The biofilter is a two-stage system designed to treat 15,000 m^3 air h^{-1}. The first stage (69 m^2) contains calcareous materials to reduce hydrogen sulphide emissions. The second, main stage (115 m^2) contains a mixture of compost and lightweight clay aggregate. No CH_4 measurement had been performed prior to this study. Staff at the facility performs inspections of the installations to counter gas leakage.

2.2 Remote sensing – the tracer gas dispersion method

Total CH$_4$ emission rates from the biogas plants were determined using a tracer gas dispersion method (TDM) (Delre et al., 2018; Mønster et al., 2015, 2014; Yoshida et al., 2014). The TDM involves the continuous release of a gaseous tracer combined with downwind measurements of atmospheric CH$_4$ and tracer gas, performed while traversing the plume and using a mobile analytical platform. In the measurements reported herein, acetylene (C$_2$H$_2$) was used as tracer gas. The emission rate of the target gas (CH$_4$ in this case) can be determined using Eq. 1:

$$E_{target} = Q_{tracer} \times \frac{\int_{Plume\ start}^{Plume\ end} (C_{target} - C_{target,background})dx}{\int_{Plume\ start}^{Plume\ end} (C_{tracer} - C_{tracer,background})dx} \cdot \frac{MW_{target}}{MW_{tracer}} \quad (1)$$

where E$_{target}$ is the target gas emission (here CH$_4$), Q$_{tracer}$ is the release rate of the tracer gas (kg h^{-1}), C$_{target}$ and C$_{tracer}$ are concentrations of target and tracer gas (ppbv) above background level, MW$_{target}$ and MW$_{tracer}$ are the molecular weights of the target and tracer gas and x is the distance across the plume (m). The measurements were conducted by driving multiple times through the downwind plume, and emission rates were calculated using Eq. 1 for each plume traverse and then calculated as the average of all traverses in the campaigns, thereby giving a plant-integrated emission rate for the duration of the performed measurements. Uncertainties relating to the measured emission rates were estimated as the standard error of the mean (sample standard deviation of the CH$_4$ emission rates calculated for each plume traverse, divided by the square root of the number of plume traverses).

Measurements of the target and tracer gas concentrations were performed using a cavity ring-down spectrometer (G2203, Picarro Inc., USA), installed in a vehicle, and from where atmospheric gas was pumped into the analyser from an intake placed on the roof of the vehicle. The measurement frequency of the instrument was ~1 Hz, and the precision of the CH$_4$ and C$_2$H$_2$ measurements was

0.77 ppb and 0.06 ppb, respectively (1σ, 1 hour). A monitor in the driver's cabin displayed real-time concentration data as they were recorded. A global navigation satellite system (GNSS) receiver was used to log the positions of the measurements (an R330 GNSS receiver and an A43 antenna, Hemisphere, Canada).

Initial emission screenings were performed in each case, to identify the main CH_4 emissions areas at the biogas plants as well as in the surrounding area, in order to establish from where tracer gas would be released and to evaluate if any other nearby CH_4-emitting sources (farms, wastewater treatment plants, etc.) could affect the measurements.

Tracer gas was released from one or two locations at the biogas plants at release rates of between 0.6 and 0.9 kg C_2H_2 h^{-1} in total. C_2H_2 flow was measured using 150 mm variable-area flowmeters (Sho-Rate, Brooks Instruments, Holland) calibrated for measuring between 0 and 1.65 kg C_2H_2 per minute and with an uncertainty of 5% on the maximum value.

2.3 On-site methods

2.3.1 Leakage detection

At each biogas plant, on-site measurements were initiated by performing a leak search. At biogas plants A, B and D, this was done with an optical gas imaging infrared (IR) video camera (GF 320, FLIR, Wilsonville, USA), which was used to inspect all biogas-bearing plant components (digesters, storage tanks, gas pipes, biomass pipes, valves, gas storage units, etc.). The IR camera visualises biogas emitted from a source as a grey-coloured cloud, and it can be used to sense leaks remotely from pipes, tanks, valves etc. and in hard to access areas. The camera was adjusted to wavelengths between 3.2 and 3.4 μm, thereby allowing it to detect a range of hydrocarbons – including CH_4. The IR camera has a detection limit of ~8-9 L CH_4 h^{-1} in laboratory conditions (pure

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CH_4, ~2 m s^{-1} wind speed, 3 m distance to source) (Reinelt et al., 2017). A potential advantage of this leakage detection method is the capability of detection of emission sources at a distance (and thus also at hard to access areas) without the need to sample the emitting gas.

At biogas plant C, the leak search was performed with a portable CH_4 analyser (EX-TEC PM4, Sewerin, Gütersloh, Germany) equipped with a semiconductor sensor detecting flammable gases, including CH_4 in the ppm range. A potential advantage of this leakage detection method is high portability and ease of use inside buildings. The CH_4 analyser leak search was supplemented with leak search using an optical gas imaging camera of the same make and model as described above.

The reason behind the different methodology used at biogas plant C was that the on-site measurements at that site were performed by a different measurement team with a partially different set of equipment's than the team at plants A, B and D.

2.3.2 Emission quantification, using a high-volume sampling technique

A high-volume sampling technique was used to quantify identified gas leakages on digesters, biomass storage tanks, gas storage units, etc. This method involved collecting the leaking gas, using a sampling hood modified to cover partly the leak in each case. The sampling hood was connected to an ATEX proof blower (MSX 200-3, GEOVENT A/S, Denmark) via a hose. CH_4 concentration in the air/biogas mixture was determined using either a photoacoustic gas analyser (INNOVA 1412, Lumasense Technologies, USA) or a tuneable diode laser CH_4 analyser (LGD F200A, Axetris, Switzerland). The flow rate of the air/biogas mixture was measured by analysing pressure differences with a calibrated orifice (FMU, Lindab GmbH, Germany) and a pressure sensor (2020P7, Digitron, UK). The flow rates were calculated using Eq. 2:

$$Q_{air} = C \cdot A_2 \sqrt{\frac{2(P_1 - P_2)}{\rho\left(1 - \left(\frac{A_2}{A_1}\right)^2\right)}} \qquad (2)$$

where Q_{air} is the flowrate of air ($m^3 h^{-1}$), C is an orifice flow coefficient (dimensionless), A_1 is the cross-sectional area of the hose (m^2), A_2 is the cross-sectional area of the calibrated orifice (m^2), P_1 is upstream pressure (Pa), P_2 is downstream pressure (Pa) and ρ is air density ($kg\ m^{-3}$).

CH_4 emissions were determined using Eq. 3:

$$E_{CH4} = Q_{air} \cdot \rho_{CH4}(C_{CH4,out} - C_{CH4,in}) \qquad (3)$$

where E_{CH4} is the CH_4 emission mass flow ($g\ h^{-1}$), Q_{air} is the volume air flow ($m^3 h^{-1}$), ρ_{CH4} is the gas density of CH_4 ($g\ m^{-3}$) and $C_{CH4,out}$ and $C_{CH4,in}$ are exhaust and background CH_4 concentrations, respectively (molar fractions).

2.3.3 Emission quantification from active ventilation units (ducts and pipes)

Quantifying CH_4 emissions from active ventilation units was done by measuring the air flow through the ventilation system as well as the concentration of CH_4 in the ventilation air. By multiplying the air flows with CH_4 concentrations above background levels measured near the biogas plants, CH_4 emission rates through these units were determined. The air flows were either measured by using a pitot tube/micro manometer (Mano-Air 100, Schiltknecht, Switzerland) according to the standard "DS/EN ISO 16911-1 Manual and automatic determination of velocity and volume flow rate in ducts – Part 1: Manual reference method" or were determined according to the ventilation units' technical specifications. CH_4 concentrations were measured using the photoacoustic gas analyser mentioned above, a tuneable diode laser or a flame ionisation detector (FID) equipped with a non-methane hydrocarbon cutter (3-900, J.U.M. Engineering GmbH, Germany).

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2.3.4 Emission quantification from open digestate storage

Biogas plant B was the only facility in this study with open tanks for digestate storage. Capacity was relatively low (retention time: ~12h), since the digestate from these tanks was dewatered and incinerated at the facility. Results from the leakage detection at biogas plant B suggested that the open digestate storage was a significant contributor to CH_4 emission, but the on-site measurement team was not equipped to quantify this source through measurement. This CH_4 emission was instead estimated by assuming that CH_4 formation in the open digestate storage units was similar to CH_4 formation (~6.7 g CH_4 m^{-3} digestate h^{-1}) in the digesters, due to the ~12h retention time, and that this CH_4 would emit from the digestate storage unit.

2.3.5 Emission quantification via the CHP units

CH_4 emissions from CHP units were not measured by the on-site teams, since they were beyond the scope of the project, which results are discussed here. However, to compare emission rates measured by on-site teams to the remote sensing results, we used typical emission figures for gas engines, to provide suitable estimates.

CH_4 emissions from the CHP units at biogas plants A and B were estimated by multiplying gas consumption from the engines with an assumed CH_4 emission of 1.5% of consumed CH_4 fuel. This value was converted from an emission factor of 300 g CH_4 GJ^{-1} for biogas engines listed by the Danish Energy Agency (Energistyrelsen and Energinet.dk, 2017), which is close to the average measured losses from biogas engines (1.7% of consumed CH_4 fuel) (Liebetrau et al., 2013). We note that CH_4 emissions from biogas engines are known to vary. In Liebetrau et al. (2013) emissions from eight gas engines were measured, and the emissions ranged from 0.4 to 3.3% of CH_4 consumption. Using a fixed factor to estimate CH_4 emissions from specific gas engines as done here may therefore be more uncertain compared to measurements.

2.4 Measurement campaigns

TDM and on-site measurements were performed at each of the four biogas plants on the same measurement days by two teams working independently between October 26, 2015 and Nov. 5, 2015. For biogas plant A, the measurements spanned over two working days, since the on-site measurements could not be completed on the first day, whereas measurements for plants B, C and D were completed over one working day. At each facility, the on-site team started their work by identifying leaks, and then they used the remaining time to measure emission rates from each of the identified leaks one at a time. Limitations due to weather conditions and the physical surroundings of the plants meant that it was only possible to perform TDM measurement part of the time while on-site measurements were performed.

Measurements and data analysis, including the calculation of emission rates and reporting, were done independently by the TDM and on-site teams. The results were reported by each team to the project leader, without one team knowing the results of the other team.

3. Results and discussion

3.1 Methane emissions from biogas plant A

At biogas plant A, measurements spanned over two days, since the facility was large and numerous sources of CH_4 emission were identified using an optical imaging leakage search. In total, 22 individual sources of CH_4 emission were found, some of which were quantified together; two leaks, for example, were found at two gas pipes (from a pipe joint and a valve) located inside a gas building, quantified by measuring the CH_4 flow (CH_4 concentration and air flow) through the building's ventilation system and thus resulting in a sum value. Two of the identified leaks located on the top of one reactor (at the biomass stirrer and pressure relief valve) were not quantified by the on-site team due to technical difficulties, and they were thus not included in the estimation of total

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CH_4 emissions using the on-site approach. The sum of CH_4 emission rates from on-site sources was 3.4 kg CH_4 h^{-1} (Table 2), with the majority (86%) originating from just four of the 13 identified emission sources (Fig. 1). The largest individual source of CH_4 emission from biogas plant A was the gas engine, comprising 45% of all sources (Table 2). The second largest source of CH_4 emission was building ventilation, led through a biofilter for odour control (26%). This source was quantified by measuring the CH_4 concentration at the inlet to the biofilter, and multiplying the concentration with the air flow, which was determined from the specifications of the blower leading air to the biofilter. The choice to measure methane flow at the inlet to the biofilter as opposed to the outlet was done by the measurement team for practical reasons. The biofilter may potentially have some CH_4 oxidation capability. However, due to low retention of the gas in biofilters for odour reduction, CH_4 oxidation is expected to be minor, and thus the choice of measuring before the filter inlet is not expected to have led to any significant overestimation of this emission source. The main sources of this CH_4 flow were found to be from two pipes located in the biomass-receiving building (inlets from manure transport), while two other sources were ventilation from the gas building and the compressor building, accounting for 6.9% and 7.0%, respectively, of the combined emissions identified by on-site measurements. Leaks at pressure relief valves on the digesters were also found to emit CH_4, albeit they only contributed to a relatively low proportion in this regard.

The sum of emission rates from biogas plant A, determined using the on-site approach (3.4 kg CH_4 h^{-1}), was lower than the total CH_4 emission rate determined using the TDM, where the average rate on the two measurement days was 5.5 kg CH_4 h^{-1}. One reason for this difference may be the two identified leaks on the top of the reactor that were not quantified, as well as other leaks not found during the initial search. The calculation method used to estimate the CH_4 emission rate from the gas engine was a contributor to uncertainty, in that the fraction of un-combusted CH_4 may vary from engine to engine (Liebetrau et al., 2013), and a default value of 1.7% was used herein, as

described in section 2.3.5. Another reason may be variations over time in the true emission rate. During the second day of measurement, total CH_4 emissions measured using TDM were considerably higher (9.5 kg CH_4 h^{-1}, an average of 21 plume traverses) than on the first day (3.3 kg CH_4 h^{-1}, average of 37 plume traverses). According to the plant operators, technical problems with one of the pumps at the plant may have caused gas pressure build-up, which may in turn have led to a higher emission rate on the second day due to emission through pressure relief valves.

We note that CH_4 emissions from the off-site CHP units receiving gas from biogas plant A were not quantified in this study, and that these emissions are therefore not included in the results listed in Table 2.

3.2 Methane emissions from biogas plant B

Relatively few leakages were found at biogas plant B, with the leakage search using optical imaging to identify nine sources of CH_4 emission, each of which was quantified (Fig. 1). These emission sources were open digestate storage tanks, pressure relief valves, near to the supply to the CHP unit, a manhole cover and a sludge pump tank. Emissions from pressure relief valves were not from pressure relief events, but leakages from closed valves. Additionally, CH_4 was emitted from the exhaust of the gas engine at the site, which the on-site team was not asked to include in their measurements but was subsequently estimated as described in section 2.3.5.

The largest source of CH_4 emission at biogas plant B was found to be the open digestate storage tanks at 53% of the total sum. This result is in line with previous studies on CH_4 emissions from facilities with open digestate storage tanks (Liebetrau et al., 2013; Reinelt et al., 2017). As described in section 2.3.4, quantification from this source was not done by measurement but was instead calculated by assuming that CH_4 formation and its subsequent emission were equivalent to CH_4 emissions in the digesters. The validity of this assumption was not tested, and so we consider

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this estimation to be potentially highly uncertain. Another major source of CH_4 emission was the CHP unit, from where 38% of the sum of quantified on-site sources originated. Other sources of emission from biogas plant B, including leaks from the pressure relief valves, were found to be of lesser importance (Table 2).

The sum of emission rates measured from biogas plant B (on-site approach) was 6.5 kg CH_4 h^{-1}. As for biogas plant A, the sum of quantified on-site sources of CH_4 emission at biogas plant B was significantly lower than that measured using TDM, which was 13.5 kg CH_4 h^{-1} (Table 2). The reasons for this difference may be quantification uncertainties, variations in the true emission rate and unidentified CH_4 sources not quantified by the on-site team, or a combination of these factors. Another possibility was interference from a wastewater treatment plant adjacent to the biogas plant. During screening of CH_4 concentrations in the atmosphere at the biogas plant, concentrations up to 5.25 ppm CH_4 above background level were observed, where the highest concentrations were measured near the open digestate storage tanks. At the area with wastewater treatment reactors, concentrations were up to 0.5 ppm above background level. The CH_4 concentration screening thus suggested that the emission of CH_4 from the biogas plant was higher than the emission from the wastewater treatment area. A previous study at the same biogas plant/wastewater treatment plant also showed that the most significant CH_4 emissions emanated from areas around the digesters (Yoshida et al., 2014). The emission rates quantified in this study were within the range reported for the same facility (5 to 32 kg CH_4 h^{-1} when excluding measurements performed during digester malfunction (92 kg CH_4 h^{-1})) (Yoshida et al., 2014).

3.3 Methane emissions from biogas plant C

The leakage search at biogas plant C, using a portable CH_4 gas analyser supplemented with optical imaging, found eight sources of emission that required quantification (Fig. 1). Four of those were at

the biogas upgrade unit at this site, whereas two were found to be ventilation air from a biomass receiving tank and a biomass mixing tank. The remaining two sources were leakages from a digestate/gas storage tank (safety vent and air outlet).

All identified emission sources at biogas plant C were quantified, with the highest emission rate measured at the CO_2 exhaust from the water scrubber used for biogas upgrading. According to the plant operator, gas production from this newly constructed biogas plant had exceeded the designed capacity for the upgrade unit, thus leading to excessive CH_4 loss through the CO_2 exhaust from the water scrubber, and the biogas upgrading unit itself requiring modification or replacement with a higher capacity element. The CH_4 emission rate from the biogas upgrade unit accounted for 75% of the sum of on-site source emissions.

Ventilation from the biomass mixing tank and biomass receiving tank comprised 22% of emissions, whereas the two leaks found at a digestate/gas storage tank were of lesser importance (3%).

The sum of the measured on-site sources of CH_4 emission was 27.8 kg CH_4 h^{-1}, whereas the average total from biogas plant C was measured at 12.6 kg CH_4 h^{-1} using the TDM. The fact that the sum of emissions using the on-site approach was higher than the TDM results was surprising. One possibility was that the results of the TDM measurement were lower than the true emission rate, due to incomplete tracer and CH_4 gas mixing. On-site measurements showed that a large part of leakage from the site occurred through a chimney, including CO_2 release from the water scrubber. Since the tracer gas was released near ground level, incomplete CH_4 and tracer gas mixing of in this case would have led to an underestimation of the CH_4 emission rate using the TDM. This possibility was evaluated by using a Gaussian plume model similar to an approach used in previous studies (Delre et al., 2017; Yoshida et al., 2014). The model results suggested that underestimation due to the emission height difference of CH_4 and tracer gasses was not likely to explain sufficiently

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differences in relation to weather conditions during measurement (Pasquill stability class B – slight

insulation), measurement distance (up to 1200 m) and the height of the chimney (~35m). Two roads

were used to perform the TDM measurements, and the distances between the chimney and the

observed peak concentrations were 400 m and 1300 m, respectively. Using the closer of the two

roads led to an underestimation of the CH_4 emission rate of 22%, according to the dispersion model,

whereas using the road farther away led to an underestimation of 3%. The average emission rate

using the road farthest away for TDM measurement was somewhat higher than the value for the

closer road (14.5 and 12.1 kg CH_4 h^{-1}, respectively), which is consistent with the more significant

underestimation using the TDM along this route. Another explanation for the observed difference

between on-site quantification and the TDM results was the variation in the true CH_4 emission rate

at biogas plant C. Fig. 2 shows total CH_4 emissions measured for each plume traverse at biogas

plant C using the TDM, suggesting that emissions varied between ~5 and 35 kg CH_4 h^{-1} on the

measurement day. Note that measurement uncertainty for the emission rates determined using TDM

for single-plume traverses should be considered higher compared to TDM measurements reporting

average values of several plume traverses (Fredenslund et al., 2018; Mønster et al., 2014).

Quantification of the largest point source of CH_4 emission (CO_2 exhaust from the water scrubber)

was done over a 20-minute period, one hour before the TDM measurements were initiated. This

emission occurred through a pipe, where the gas flow rate was determined using a pitot tube/micro

manometer, and the CH_4 concentration was measured using a FID (section 2.3.3). Since

measurement of this dominant, single source of CH_4 emission and the TDM measurements did not

occur simultaneously, emission dynamics may explain the difference in measured emission rates

between the on-site and TDM approaches. No further explanations such as change in operation of

the water scrubber, signs of erroneous measurement of the FID used to measure CH_4 from the water

scrubber or similar, were found to explain the difference between TDM measurements and CH_4 emission from this dominant, single source.

3.4 Methane emissions from biogas plant D

An IR camera leakage search found 19 sources of CH_4 emission at biogas plant D, four of which were ventilation air from a biomass receiving tank, a mixing tank, a biomass heating unit and a biogas upgrade unit leading to a biofilter set up for odour control. The quantification of these four sources was done collectively by measuring air flow and CH_4 concentration at the inlet to the biofilter. Individual measurements for these four sources were also performed, indicating that ~94% of the CH_4 leading to the biofilter was from the receiving and the mixing tanks. On-site measurements disclosed that CH_4 in the ventilation air leading to the biofilter constituted 84% of the total sum of quantified on-site sources, whereby ventilation air was the most significant contributor to CH_4 emissions. Other emission sources consisted of leaks from pressure relief valves (5% of on-site measured emission), various minor leaks from caps, edges of soft covers and other (9%) and ventilation from the gas building (2%). Two leaks at the top of a biomass storage tank that were identified using the IR camera were not quantified due to inaccessibility.

The sum of on-site quantified CH_4 emission rates was 15.4 kg CH_4 h^{-1}, which was close to the measured total CH_4 emission rate using TDM (13.5 kg CH_4 h^{-1}). Several reasons may explain the closer results of the on-site and TDM quantifications at biogas plant D. First, the digesters, tanks, etc. were much lower and wider compared to the other facilities, which may have increased the likelihood of detecting leakages, due to better accessibility. Second, all on-site quantifications were based on measurements, whereas the use of the calculation methods for estimating CH_4 losses from engines and open digestate storage at plants A and B may have increased the uncertainty of the on-site quantifications at these locations. Last, TDM measurements at biogas plant D suggested a

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comparatively lower variation in total CH_4 emissions from this site. The observed sample standard deviation was 2.5 kg CH_4 h^{-1} using TDM (or 19% of the average emission), while in comparison, for plant C, the rate was 6.0 kg CH_4 h^{-1} (or 48% of the average emissions). A more constant, actual CH_4 emission rate from biogas plant D may therefore explain the higher similarity between the on-site and TDM measured CH_4 emission rates.

3.5 Identified CH_4 emission mitigation options

Based on on-site measurements, emission mitigation options at the four biogas plants CH_4 were suggested. At biogas plant A, it was recommended that compressors and a blower unit in a gas building should be renovated to reduce leakage and that pressure relief valves should be checked regularly. Eliminating leakage from the gas compressor and the blower unit would result in a reduction in emissions of 0.47 kg CH_4 h^{-1} (~14%), according to the on-site quantification of these leakages. For biogas plant B, it was recommended that the open digestate storage should be closed off and fitted with a gas collection apparatus. Here, it was also recommended that a sludge pump tank be enclosed to reduce leakage. Eliminating these sources of emission would lead to a reduction of 3.6 kg CH_4 h^{-1} (~56%). For biogas plant C, it was recommended that a thermal oxidiser should be installed to reduce CH_4 emissions from the biogas upgrade unit, thereby eliminating emissions from this source and leading to a reduction in emissions of 20.9 kg CH_4 h^{-1} (~75%). The biogas upgrade unit was later replaced with a chemical scrubber with adequate capacity, which likely caused a significant reduction in CH_4 emissions. In addition, a hole in a pipe was found, which was recommended for repair. It was also recommended to perform maintenance on a safety vent and air outlet at a digestate/gas storage unit. Finally, for biogas plant D, it was recommended to look into utilising gas from the receiving and mixing tanks in the biogas upgrade unit. Eliminating emissions from those tanks would result in a reduction in CH_4 emissions of 12.7 kg CH_4 h^{-1} (~83%). It was

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also recommended to perform maintenance on various valves and fittings, to reduce minor leakages, and to replace a piece of piping, the type of which was deemed to be leading to CH_4 emissions.

3.6 Comparison of the on-site vs. the remote sensing approach

The sum of CH_4 emission rates from on-site quantification varied for total CH_4 emission rates measured using TDM on the same measurement days. The largest differences were observed at biogas plants B and C, where one rate was ~100% larger than the value of the other rate in both of these cases. As discussed in section 3.4, several factors may have caused these observed differences, apart from measurement uncertainties. These include sources of emission that were un-detected, and therefore not included in the on-site quantification, sources of emission that were difficult/impossible to quantify for several reasons and short-term dynamics in the true CH_4 emission rate.

Since on-site quantification, in practice, should be done one source at a time, emissions from one source are only typically measured over a period of 10-20 minutes, which may cause the TDM to be less sensitive to short-term dynamics in emission rates, since the longer measurement time may include periods of both high and low emission. Both of our results, and previous studies, suggest that emission rates from biogas plants vary in the short term (Baldé et al., 2016; Groth et al., 2015; Yoshida et al., 2014). If, for example, ten leaks are found at a biogas plant which need to be quantified, and each of these leaks is measured over ten minutes, it would be possible to perform 100 minutes of measurement using TDM, even without considering the additional time required to move on-site equipment from one location to the next. Our results suggest that CH_4 emissions at biogas plants can have single emission sources, which contribute to 75% of the total emission rate, such as the CO_2 exhaust from the water scrubber at biogas plant C. If that source of emission varied in the short term, as the variation in the total emission rate using TDM, shown in Fig. 2, may

suggest, it may be beneficial to perform measurement over a longer period. The TDM is the most efficient option to measure the total plant emission over a longer period, but in the case of a single or few dominant sources (such as the water scrubber at biogas plant C) are identified, it may be beneficial to perform long-term measurement of the emission from that/those source(s), if the on-site approach is chosen.

At all four biogas plants, most of the CH_4 emissions occurred from a few dominant sources (Fig. 1). It is thus highly important that these sources are identified using the on-site approach, as well as to ensure that emission rates from these dominant sources can be measured in such a way to avoid reliance on uncertain estimates, such as for open digestate storage at plant B, where CH_4 potential for the digested sludge was used to estimate emissions.

Comparing measured emission rates to gas production for each facility (provided from each biogas plant's supervisory control and data acquisition SCADA system), relative methane losses (CH_4 emission/CH_4 production) were calculated. Measured losses using the TDM method ranged from 1.4 to 8.4% of production (Table 2). The summed average (sum of emissions, four plants/sum of production, four plants) was 2.7%, which is comparable to losses of 3.1% reported from an agricultural digester (Flesch et al., 2011), and 0.6 to 2.1% from a sludge digester (Reinelt et al., 2017).

One drawback of TDM is that it does not pinpoint or quantify individual sources of emission. If the purpose of measuring CH_4 emissions at a biogas plant is to find mitigation options, and thereby provide options to improve the environmental benefits of biogas production, on-site methods are needed. Conversely, a downside to the on-site quantification methods described in this paper is that quantification may be comparatively time-consuming – especially at large facilities, where

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numerous sources of emission are found and might be overlooked, with some technically unquantifiable points of leakage leading to an underestimation of the plant's total CH_4 emission.

Our results therefore suggest that on-site and remote sensing methods cannot substitute each other, if both an accurate assessment of the total emission rate and the identification of mitigation options are required. A strategy of combining quick, semi-quantitative on-site measurements and TDM or similar remote sensing methods may be effective in achieving both aims, which may be the subject of further development.

3.7 Allocating CH_4 emissions to gas production

Both gas production and gas utilisation occurred at all four biogas plants in this study. Gas utilisation is defined here as either use in CHP units or conversion to biomethane in biogas upgrade units. Since on-site measurements were performed, it was possible to allocate CH_4 emissions to either gas production or utilisation. At biogas plant A, the sum of quantified on-site sources was 3.4 kg CH_4 h^{-1}, of which 43% was estimated to be from the on-site CHP unit. Subtracting the gas engine emission (1.5 kg CH_4 h^{-1}) from the total plant emission determined by TDM measurements and sum of on-site sources, the emission related to gas production alone can be estimated at 1.9 kg CH_4 h^{-1} and 4.0 kg CH_4 h^{-1} respectively, if it is assumed that all other emission than from the CHP unit can be allocated to gas production. These emission estimates equalled 0.5% and 1.0% of the gas production, respectively. Similarly, emission from gas production from biogas plant B can be estimated at 4.0 kg CH_4 h^{-1} (on-site method) and 11.0 kg CH_4 h^{-1} (TDM) or 2.5% and 6.8% of the production. At biogas plant C, the gas utilisation (water scrubber) was found to be the dominant source (section 3.3), where the emission rate was found to be higher than the total emission from the biogas plant measured later using TDM. Allocating emissions to gas production by subtracting emission from the water scrubber from the sum of on-site emission sources, the emission from gas

production at biogas plant C is estimated to be 6.9 kg CH_4 h^{-1} equal to 1.0% of the gas production. At biogas plant D, the on-site measurement team did not observe CH_4 emission from the chemical scrubber, whereby all emission from that biogas plant can be allocated to gas production.

4. Conclusion

Methane (CH_4) emissions from four biogas plants were measured on the same days, using both on-site quantification methods and TDM. In general, CH_4 emission rates were between 5.5 and 13.5 kg CH_4 h^{-1}, using TDM corresponding to 1.4-8.3% of the CH_4 production. On-site measurements showed that the majority ($> \sim 80\%$) of the emissions often occurred from a few sources (between two and four sources). Our results suggest that on-site and TDM measurements cannot replace each other if accurate measurements of the total CH_4 emission rate as well as the identification of CH_4 emission mitigation options are necessary.

E-supplementary data for this work can be found in the online version of this paper.

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FIGURE CAPTIONS

Fig. 1. CH$_4$ emission rates measured on identified emission sources (or group of sources) at the four biogas plants (plants A, B, C and D), using the on-site approach.

Fig. 2. Total CH$_4$ emission rates from biogas plant C (single-plume traverses), determined using TDM.

TABLES

Table 1. Main characteristics of the four biogas plants

| Biogas plant	Feedstocks	Gas production (million kg CH_4 yr^{-1})	Gas utilisation	CHP unit(s), total capacity (MW)
A	Manure, organic waste	3.4	CHP on-site (~20%), CHP off-site (~80%)	1×1.35 (on-site) 2×2.65 (off-site)
B	Wastewater sludge	1.4	CHP on-site	2.55
C	Manure, slaughterhouse waste	4.6	Upgrade to bio-methane (water scrubber) on-site	-
D	Manure, straw, maize silage	2.9	Upgrade to bio-methane (chemical scrubber) on-site	-

Table 2. Measured CH$_4$ emission rates using TDM and on-site methods

Biogas plant	Total CH$_4$ emission – TDM measurement[a] (kg CH$_4$ h^{-1}; fraction of gas production)	Number of plume traverses (TDM)	CH$_4$ emission – sum of individual sources (kg CH$_4$ h^{-1}; fraction of gas production)	Number of individual sources identified, quantified	On-site measurement – emission sources (percentage of the sum of individual sources)
A[b]	5.5 ± 0.7; 1.4%	58	3.4; 0.8%	22, 20	Gas engine (45.1%); Leaks at pressure relief valves (6.8%); Ventilation from building through biofilter (26.4%), Leaks at biomass stirrers (1.4%); Gas compressor, gas measurement unit and other (20.2%)
B	13.5 ± 0.5; 8.3%	21	6.5; 4.0%	10, 10	Open digestate storage (53.1%); Gas engine (38.3%); Leaks at pressure relief valves (2.6%); Gas booster, ventilation from engine building and other (6.0%)
C	12.6 ± 0.9; 1.9%	49	27.8; 4.1%	8, 8	Biogas upgrade unit (75.3%); Ventilation from biomass mixing tank and biomass receiving tank (21.7%); Leaks at safety vent and air outlet at digestate/gas storage unit (2.5%)
D	13.4 ± 0.5; 3.3%	28	15.2; 3.8%	19, 16	Gas outlets from receiving tank, mixing tank and ventilation to biofilter (83.6%), Leaks at pressure relief valves (5.5%); Ventilation from gas building (1.7%); Leaks at caps and other (9.2%)

[a] Uncertainty is estimated as a standard error of the mean (see section 2.2)

[b] CH$_4$ emissions from off-site CHP units were not included
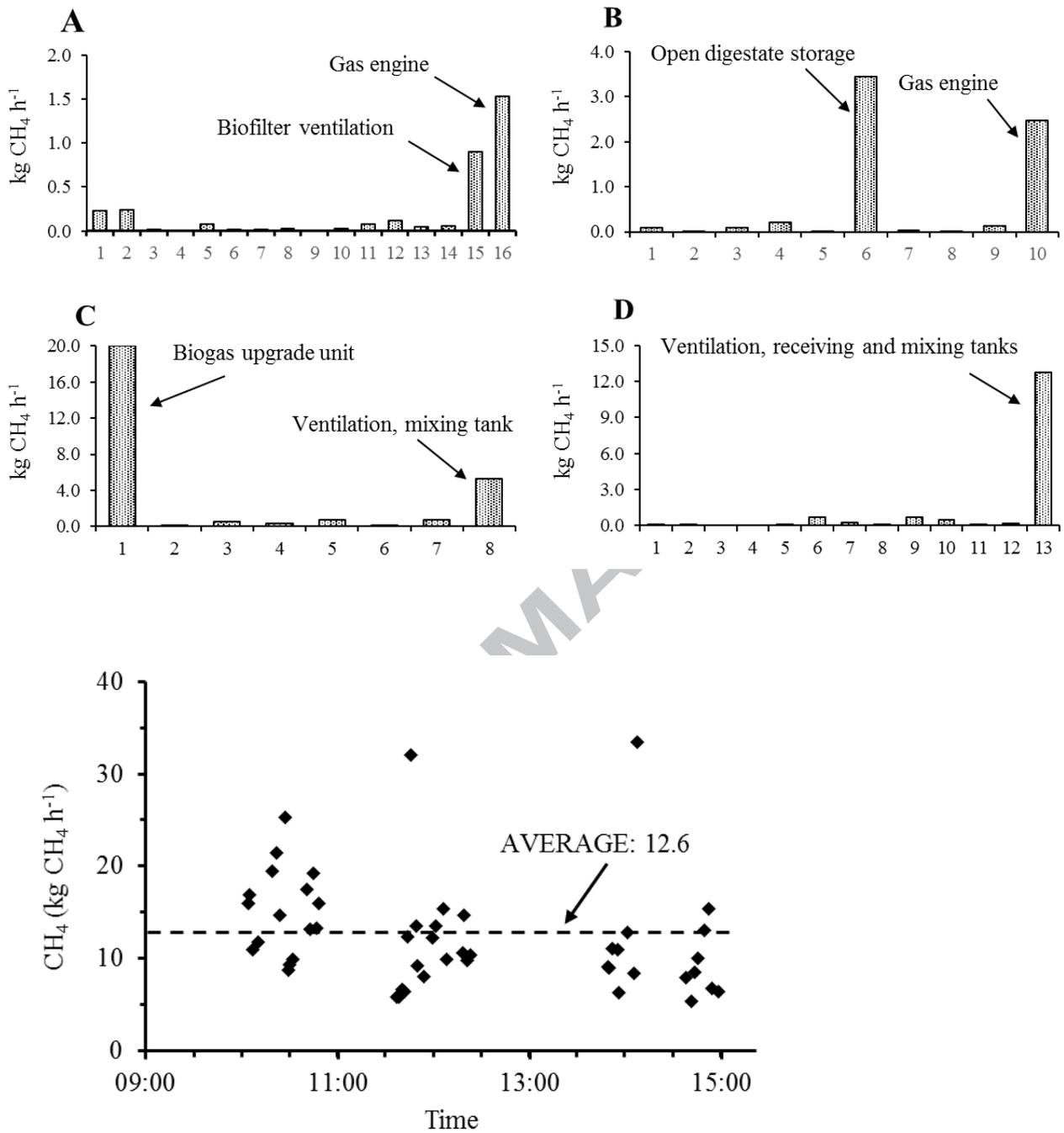
Highlights

- Same-day, on-site and remote sensing emission measurements for four biogas plants

- Remote sensing methane emission rates varied from summed on-site emission rates

- On-site methods can identify key points of leakage

- On-site measurements may not include all emission sources