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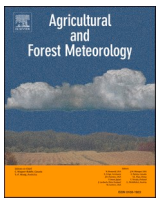

Determination of methane emissions from biogas plants, using different quantification methods

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ABSTRACT

Reliable and comparable quantification methods are needed for assessing the effectiveness of the biogas production and utilisation process in mitigating methane (CH_4) emissions as well as improving the database for emission inventories. The objective of this study was to compare and validate CH_4 emissions quantified at two agricultural biogas plants, for up to three days, using diverse on-site (two teams) and off-site methods (three teams), including differential absorption lidar (DIAL), tracer gas dispersion (TDM) and inverse dispersion modelling (IDM). For plant 1, with a constant combined heat and power (CHP) load, the average emission factor varied from 0.3% CH_4 (on-site approaches) to 1.2% CH_4 (off-site approaches). On-site approaches underestimated overall emissions due to many small (unquantified) CH_4 leakages. All methods observed comparable average emission factors for plant 2, ranging between 1.9 and 2.2% CH_4. In this case, the majority of emissions emanated from just a few sources. However, correcting the significant influence of the varying CHP load during the measurement campaign revealed significant differences between TDM and IDM (DIAL did not participate). It was demonstrated that TDM and IDM could recover the emission rate from a known point source (controlled release of CH_4 via a small diffuser) within an accuracy of 93 ± 15% (TDM) and 92 ± 17% (IDM) under favourable and similar conditions.

1. Introduction

Biogas plants employ the anaerobic digestion of highly calorific organic sources, residual waste streams and livestock manure to produce energy for electricity production, heating and transportation. In addition, digestate has the capacity to compete favourably with mineral fertiliser in agronomic plant production (Tampio et al., 2016). In Europe, about 19,000 biogas plants and 725 biomethane facilities produce 167 TWh biogas and 26 TWh biomethane, respectively, along with a significant portion of agricultural-based facilities (EBA, 2020).

The greenhouse gas (GHG) mitigation effect of biogas systems is twofold, namely the reduction of methane (CH_4) emissions from organic sources, such as organic waste and manure, and the substitution of fossil fuels through the production of renewable energy. When upgraded, biomethane is a renewable alternative to natural gas. However, CH_4 emissions from biogas production and utilisation reduce net GHG benefits. Data on the magnitude of these emissions is important for the environmental assessment of biogas production, the evaluations of potential and measures for GHG reduction as well as improving emission factors for national GHG inventories (Hrad et al., 2021; Scheutz et al.,

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2019). Furthermore, reliable quantification methods are relevant to fostering effective (self-)regulation, thus ensuring low direct CH_4 emissions from biogas plants.

In the last few years, different scientific studies have intensively investigated CH_4 emissions from biogas plants using different methods. Two major approaches have been characterised: on-site (component-scale) and off-site (facility-scale) methods.

On-site methods have been commonly applied to identify and quantify single (component) emissions from combined heat and power (CHP) or biogas upgrading units (BUU), open or not gas-tight storage of the digestate, leaks from biogas-bearing plant components and pressure release valves (Daniel-Gromke et al., 2015; Kvist and Aryal, 2019; Liebetrau et al., 2010, 2013; Reinelt and Liebetrau, 2020).

Whole-plant CH_4 emissions from biogas plants have been determined by means of concentration measurements in the surrounding areas, using off-site approaches such as inverse dispersion modelling (IDM) with open-path tunable diode laser absorption spectrometers (OP-TDLAS) (Baldé et al., 2016; Bühler et al., 2022; Flesch et al., 2011; Groth et al., 2015; Hrad et al., 2021) and tracer gas dispersion methods (TDMs) with cavity ring down spectrometers (Fredenslund et al., 2018; Jensen et al., 2017; Scheutz and Fredenslund, 2019). IDM has been used to quantify agricultural emissions for almost 20 years in various source configurations, including animal herds, ponds and entire farms in different countries worldwide (Bai et al., 2021; Bühler et al., 2022; Flesch et al., 2011; Laubach et al., 2014; McGinn et al., 2006). TDM is a well-documented method that has been applied to many different emission sources such as natural gas facilities (Lamb et al., 1995; Mitchell et al., 2015), landfills (Börjesson et al., 2009; Mønster et al., 2014) and wastewater treatment plants (Delre et al., 2017; Yoshida et al., 2014). However, only recently has the method been applied at biogas plants (Scheutz and Fredenslund, 2019).

In contrast to direct on-site methods, off-site approaches measure emissions from the entire plant and are able to monitor time-variant and/or operational emissions over a longer period, independent of plant size (Clauß et al., 2019; Reinelt et al., 2017). However, they depend on atmospheric dispersion and are limited by certain weather conditions as well as the topography or infrastructure of the surrounding area (Clauß et al., 2019; Denmead, 2008). In addition, they provide little information on the magnitude of emissions from individual emission sources and leakages.

Even though different quantification methods have gained acceptance in the scientific community, only a few have matured to a level where they are applied commercially and used for regulatory purposes. There is thus still a need for a common procedure that will harmonise CH_4 emission quantification at biogas plants (Reinelt et al., 2017). There is a basic guideline on the determination of diffuse emissions (VDI, 2005), in which off-site methods are considered. In addition, the European Committee for Standardisation (CEN) currently published a standard (EN 17628, 2022) on measurement techniques for the monitoring of diffuse emissions (of volatile organic compounds) from industrial sites, such as refineries. As part of this work, two field campaigns were carried out in a decommissioned area of a refinery for controlled releases, as well as at an active industrial site, to validate measurement techniques (CEN/TC 264/WG 38, Howes et al., 2018). A detailed review of available off-site approaches is provided in the EPA (U.S. Environmental Protection Agency) (2018), while Liebetrau et al. (2017) provide an overview of measurement methods for quantifying CH_4 emissions from biogas plants.

Very few attempts have been made to compare CH_4 emissions from biogas plants with several on-site and off-site methods. Of these studies, two (Fredenslund et al., 2018; Reinelt et al., 2017) suggested that on-site methods may not capture all emissions at a facility, unlike off-site methods. Reinelt et al. (2017) recommended future comparative studies in order to ensure simultaneous measurements, since on-site and off-site teams could not measure at the same time. In addition, both comparative studies lacked approaches validating emission results.

The remit of this study was to assess agreement between several on-site and off-site methods by comparing and validating quantified concurrent CH_4 emissions at two agricultural biogas plants. CH_4 emission rates were acquired by two on-site and three off-site teams applying TDM, IDM and differential absorption lidar (DIAL only at one plant). In terms of method validation, the study included on-site controlled CH_4 retrievals as well as off-site release tests. To the authors' knowledge, DIAL was used for the first time to quantify CH_4 emissions from an entire biogas plant as well as from different sources within the facility. However, DIAL has been used for more than 30 years and is routinely applied for measuring pollutants and GHGs from various emission sources (Innocenti et al., 2017; Robinson et al., 2011), including the regulatory monitoring of volatile organic compounds at industrial sites (EN 17628, 2022). A comparison of different on-site and off-site methods could improve decision-making in relation to suitable emission monitoring methods in the biogas sector.

2. Material and methods

2.1. Site descriptions

2.1.1. Biogas plant 1

Plant 1 produces biomethane through the digestion of energy crops (mainly maize silage) and subsequent biogas purification and/or upgrading. Fermentation takes place in two parallel main digesters and one post-digester (each 3400 m^3). Digestate is stored in three gas-tight tanks (each 6900 m^3), while biogas storage is implemented in the double-membrane domes of the digesters and storage tanks (see the schematic plant overview in Supplementary material SM 1 Section A). About 60 vol.% of the raw biogas (700 $m^3\ h^{-1}$) is upgraded to biomethane by chemical scrubbing, while the remaining gas portion (420 $m^3\ h^{-1}$) is utilised in the CHP unit (845 kW electric energy) along with subsequent thermal post-combustion. During the measurement campaign, an average of 554 $m^3\ CH_4\ h^{-1}$ was produced. The plant is situated in a very flat area surrounded by agricultural fields with only a few trees.

2.1.2. Biogas plant 2

Plant 2 processes energy crops (mainly maize and grass silage, as well as solid horse manure). Fermentation takes place in two parallel main digesters and one post-digester (each 1350 m^3), and the fermentation residues are stored in an open storage tank (4400 m^3, during measurement campaign: 960 m^3) once the capacity of the gas-tight storage tank equipped with gas collection (2500 m^3) is exceeded. Biogas collection is integrated into the double-membrane foil roof of the digestate storage tank. Additional storage capacity is provided by an external gas tank (see the schematic plant overview in SM 1 Section B). Generated biogas (during the campaign: 174 $m^3\ CH_4\ h^{-1}$) is utilised in three CHP units without thermal post-combustion in a flexible operation (installed capacity: 1363 kW electric energy). The plant is situated in a relatively hilly area surrounded by agricultural fields and high trees to the north. Elevation difference within a radius of 200 m around the plant is less than 10 m (see Hrad et al. 2021).

2.2. On-site methods

The on-site approach generally consists of two steps: (1) the initial identification of all unknown emissions sources and (2) the quantification of individual emission sources (see Sections 2.2.1–2.2.5). By taking the sum of the quantified sources, the whole-plant emission rate is provided – assuming the constancy of determined emission rates. The quantification methods used herein, as well as the applied measurement equipment, are described in detail in Clauß et al. (2019), Liebetrau et al. (2017) and Reinelt et al. (2017).

2.2.1. Leakage detection and quantification

At both biogas plants, the on-site teams A and B individually performed a leak search for one day during each measurement campaign, using an optical gas-imaging infrared video camera (GF 320, Co. FLIR, Wilsonville, USA) and a portable CH_4 detector (team A: LaserMethane mini gen2, Co. GROWCON, Abingdon, UK and BM 2000, Geotechnical Instruments Ltd., Leamington Spa, UK; team B: EX-TEC PM4, Co. Sewerin, Gütersloh, Germany). Team A quantified CH_4 emissions from leakages via a dynamic chamber system. Fixed chambers designed for flat emission sources (e.g. anaerobic lagoons) usually cannot be applied for leakages from gas pipes or membrane domes. The dynamic chambers used in this study were therefore adapted to the leakage source by using a flexible foil encapsulation (wind tunnel). A constant flow of outside air, using connected blowers (D 060, Co. Elektror Airsystems GmbH, Ostfildern, Germany), was maintained through the head space of the chamber, and any differences in concentrations between the in- and outlet were measured. Gas in the in- and outlet of the encapsulated point of measurement were sampled discontinuously via evacuated (20 ml) glass vials and then analysed in the laboratory by a gas chromatograph equipped with a flame ionisation detector (FID, Agilent 7890A, Agilent Technologies, Santa Clara, USA). At plant 1, two blowers were connected to the in- and outlet of the chamber, while at plant 2 the blower was only coupled at the suction side. Air flow was determined via either an orifice (DEBIMO, Co. MDUA® GmbH, Pulheim, Germany) with a pressure difference sensor (FDA 602 S2K, Ahlborn Mess- und Regelungstechnik GmbH, Holzkirchen, Germany) or a vane anemometer (Testo 416, Co. Testo, Lenzkirch, Germany). CH_4 emissions were determined according to Eq. (1):

$$E_{CH4} = Q_{air} \cdot \rho_{CH4} \cdot \left(c_{CH4,out} - c_{CH4,in}\right) \quad (1)$$

where E_{CH4} is the CH_4 emission mass flow (mg h^{-1}), Q_{air} is the volume air flow (m^3_{air} h^{-1} STP, dry), ρ_{CH4} is the gas density of CH_4 (mg ml^{-1}) and $C_{CH4,out}$ and $C_{CH4,in}$ are exhaust and background CH_4 concentrations, respectively (molar fractions). Team B directly quantified emissions from leakages, but only if they were ventilated via an air collection system (see Section 2.2.3).

2.2.2. Emission quantification of CHP units and BUU

Teams A and B quantified CH_4 emissions from CHP units and BUU by measuring CH_4 concentrations in the exhaust pipes according to the ISO 25139 (2011) (team A) and ISO 25140 (2010) (team B) standard methods. Team A determined CH_4 concentrations by discontinuous sampling with evacuated vials, whilst analyses were done with a gas chromatograph (see Section 2.2.1). Team B performed continuous sampling and analyses with an FID (JUM 3-900, J.U.M. Engineering GmbH, Karlsfeld, Germany). Volume flow could not be measured by flow velocity sensors due to short inlet zones for homogenising the flow field at the point of measurement. Instead, both teams calculated off-gas volume flows by using operational data. Uncertainty relating to these calculated off-gas volume flows cannot be specified, but it is expected to be smaller than values measured in insufficient conditions. For BUU, data on raw gas input and product gas (biomethane) output was used. Off-gas volume flow from the CHP units was calculated based on a combustion calculation according to Boie (Hellfritsch and Koppe, 2007), using electrical power, raw gas input and the (calculated) combustion-air ratio (Λ) during measurements (for more detailed information see Clauß et al. 2019). For the CHP units at biogas plant 2, operating times and conditions (full-load or partial-load) were additionally considered for calculating the off-gas volume. This was necessary, because the CHP units were operated discontinuously according to a schedule provided by the electricity grid operator.

2.2.3. Emission quantification of encapsulated sources at ventilation grids

CH_4 emissions from encapsulated sources, such as containers housing CHPs, biogas analysis units or compressor stations, were examined at the ventilation grids. Volume flows from the ventilation duct were determined by performing either grid measurements with a vane anemometer (PCE 007, Co. PCE, Meschede, Germany) related to the respective cross-sectional areas of the grids (team A: 16 points at the CHP container, four points at the biogas analysis units and compressor station) or fan data (team B: maximum flow from the characteristic curve of differential pressure and the volume flow of the fan). Team A measured exhaust air (ten samples at the grid) and fresh air (five samples at the air supply grid) CH_4 concentrations through discontinuous air sampling and laboratory analyses, while team B performed continuous gas measurements at a single point in the vented air flow (see Sections 2.2.1 and 2.2.2).

2.2.4. Emission quantification of air-inflated double-membrane domes

CH_4 emsissions from air-inflated double-membrane domes were quantified by measuring CH_4 concentrations in the outgoing air from the inflated outer layer (team A: discontinuous sampling; team B: continuous sampling; see Sections 2.2.1 and 2.2.2) as well as air volume flow. Team A determined volume flow through a pipe mounted on a chamber at the inflation-air outlet (according to the EN 15259, 2008 standard method), while team B directly measured air volume flow at the inlet of the inflation-air blower at biogas plant 1. At biogas plant 2, team B used fan data to determine volume flow.

2.2.5. Emission quantification from open digestate storage

Emission rate quantification from an open digestate storage tank at plant 2 was carried out by floating static (team A: 0.30 m^2, 0.19 m^3) and dynamic chambers (team B: 0.5 m^2; fixed volume flow of 15 m^3 h^{-1}). For the static chamber (team A), surface emissions (mg CH_4 m^{-2} h^{-1}) were determined by multiplying the linear increase (slope) in headspace CH_4 concentration inside the chamber (mg CH_4 m^{-3} h^{-1}; discontinuous gas sampling at defined time intervals after 0, 3, 6, 9, 12, 15, 20, 25 and 30 min) and the volume-to-area ratio of the chamber (m^3 m^{-2}). Since the chamber was usually not accessible for direct sampling, team A used two polyamide tubes (inner diameter 4 mm, length 10 m) connecting the chamber and a gas pump (VP 86, VWR International GmbH, Darmstadt, Germany). The sample tubing included a glass device with a butyl rubber septum for gas sampling by a syringe. Surface emissions quantified by the dynamic chamber (team B) were similar to quantifying encapsulated leakages and calculated according to Eq. (1) and then divided by the enclosed digestate surface (m^2). Team B used one polyamide tube (inner diameter 4 mm, length 20 m) connecting the chamber and an FID (see Section 2.2.2) for continuous sampling over 30 to 60 min per sampling point.

The chambers were placed at different positions on the digestate surface area near the border of the open storage tank. Team A performed four chamber measurements on a non-cracked surface area and five on a liquid surface area and weighted the digestate surface 1/3 (non-cracked): 2/3 (liquid) when extrapolating the measurements to the total surface area of the digestate tank (about 804 m^2). In contrast, team B only measured emissions from the liquid surface area, averaged over all three measurements.

2.3. Off-site approaches

2.3.1. Tracer gas dispersion method

TDM combines controlled tracer gas release (Q_T), e.g. acetylene (C_2H_2), over the area of the biogas plant with downwind concentration measurements of both a tracer (C_T) and CH_4 (C_{CH4}), using high-resolution analytical instruments mounted on a vehicle (also called "dynamic TDM"). The method assumes equivalent dispersion and the proper mixing of the emission and the tracer gas released into the atmosphere. Plant-integrated CH_4 emissions can be determined by a ratio calculation according to Eq. (2) (Scheutz et al., 2011; Mønster et al., 2014):

$$E_{CH4} = E_T \cdot \frac{\int_{plume\ start}^{plume\ end}(C_{CH4})dx}{\int_{plume\ start}^{plume\ end}(C_T)dx} \frac{MW_{CH4}}{MW_T} \quad (2)$$

where E_{CH4} is the emission rate (kg CH_4 h^{-1}) for each plume traverse, E_T is the release rate of the tracer gas (kg C_2H_2 h^{-1}), C_{CH4}/C_T is the measured downwind concentration of the CH_4 and tracer gas (ppb) above background levels, MW_{CH4}/MW_T is the molar weight of the CH_4 and tracer gas and x is the distance across the plume (m). Downwind plume concentrations were analysed with a cavity ring down spectrometer (G2203, Co. Picarro Inc., Santa Clara, USA, frequency ~1 Hz, precision of the CH_4 and C_2H_2 measurements 0.77 ppb and 0.06 ppb) by driving in several traverses perpendicular to the emission plume. A global navigation satellite system receiver was used for logging the measured concentrations to their geographical location (R330 GNSS receiver and A43 antenna, Hemisphere, Canada). Based on initial emission screenings for identifying representative tracer placements at the biogas plants, acetylene was released via high-precision 150 mm variable-area flowmeters (Sho-Rate, Brooks Instrument BV, Veenendaal, The Netherlands) from one or two locations and at total release rates ranging between 0.9 and 5.19 kg C_2H_2 h^{-1}. Locations of measurement transects and tracer releases are shown in SM 1 Figs. S1 and S4. More detailed information about the TDM method and applied instrumentation is given in Mønster et al. (2014). Measurement campaigns followed the standard procedures described in Scheutz and Kjeldsen (2019). The total uncertainty of emission quantification using TDM has been assessed by performance of controlled release tests and theoretical error budgets to be less than 20% (Mønster et al., 2014; Delre et al., 2017; Fredenslund et al., 2019).

2.3.2. Inverse dispersion modelling

IDM derives information on overall biogas plant CH_4 emissions from measurements of upwind and downwind concentrations, as well as micrometeorological parameters, using an atmospheric dispersion model. CH_4 emissions (E_{CH4}; kg h^{-1}) are determined according to the following Eq. (3) (Flesch et al., 2005):

$$E_{CH4} = \frac{(C_{CH4} - C_{BG})}{(C/Q)_{sim}} \quad (3)$$

where C_{CH4}/C_{BG} is the measured downwind and background concentration of CH_4 (g m^{-3}) and $(C/Q)_{sim}$ is the simulated ratio of the concentration rise above background levels to the emission rate by modelling backward trajectories of particles released from the concentration sensors into the source area.

In this case, IDM is based on a backward Lagrangian stochastic (bLS) model according to Flesch et al. (2004), implemented via WindTrax (version 2.0.8.9, Thunderbeach Scientific, Nanaimo, Canada) software.

Path-integrated CH_4 concentrations were measured with two OP-TDLAS devices (GasFinder 2.0, Boreal Laser Inc., Edmonton, Canada) and distant retro-reflectors positioned upwind and downwind of the biogas plants (height: 1.6 m, path length: 154–347 m, see SM 1 Figs. S2 and S5). Downwind measurements fulfilled distance requirements depending on the height of the largest wind obstacle (h_s) (plant 1: $h_s = 10$ m, plant 2: $h_s = 5$ m) and the separation distance between the source components (x_s) (plant 1: $x_s = 67$ m for east-west separation; plant 2: $x_s = 45$ m for north-south separation without substrate storage) in the along-wind direction – as suggested by Gao et al. (2010) (>0.5 x_s, >10 h_s). Due to organisational reasons, two approaches for measuring background concentrations were applied: (1) simultaneous measurement at plant 1 (except for 25.10.2016), using laser-specific correction factors to remove bias instrument errors, and (2) the assumption of a constant background concentration based on upwind measurements conducted prior to downwind measurements at plant 2. The latter one might have affected the accuracy of emission calculations due to potential changes in background concentrations during the measurement campaign.

The turbulence characteristics required for the bLS simulations were recorded with a 3D ultrasonic anemometer (Model 81000, R.M. Young Company, Michigan, USA) at a frequency of 10 Hz and a height of 4.5 m above the ground. The ultrasonic anemometer was placed on the leeward side of the plants, in order to catch turbulence on the downwind side.

In addition to concentration data and wind statistics (using relations between the mean product of the wind vectors u, v, w and temperature), the spatial dimensions and location of the biogas plants, the positions of the micrometeorological and concentration measurements were specified in the WindTrax project map. The entire area of each biogas plant was assumed to be spatially homogenous.

For each 10 min bLS simulation, 50,000 backward trajectories were released at 30 points along the open-path concentration measurement and analysed for touchdowns within the source area. Emission data were screened and removed when either (1) friction velocity (u*) fell below the threshold value u* <0.15 m s^{-1} (Flesch et al., 2014) or (2) the number of particle touchdowns within the source area covered less than 60% of it. Intervals were also removed when the concentrations measured by OP-TDLAS corresponded to low signal levels (return light level <4000 and >12,000 and the quality parameter R^2 <98%).

Based on various controlled release experiments with well-defined sources, uncertainty in relation to emission quantification using IDM was less than 20% (Gao et al., 2010; Ro et al., 2014).

2.3.3. Differential absorption light detection and ranging

The DIAL technique combines spatially resolved open-path measurements of CH_4 concentration paths over hundreds of metres with a vertical wind field profile to determine whole-site and area-specific CH_4 emissions.

A pulsed tuneable laser is used that operates alternately at one wavelength strongly absorbed by CH_4 (λ_{on}) while the second wavelength (λ_{off}) minimies absorption by CH_4 and other atmospheric components. CH_4 concentration is calculated from the ratio of the backscattered light intensity between the λ_{on} and λ_{off} wavelengths. The path-concentration integral (CL), calculated as function of the range (r) from DIAL, is:

$$CL(r) = \frac{1}{2\,\Delta\alpha} \log \frac{S_{off}(r)}{S_{on}(r)} - b(r) \quad (4)$$

where S_{on} and S_{off} are the return 'on' and 'off' signals (after normalisation due to laser power and signal offsets), $\Delta\alpha$ is the differential absorption coefficient in ppm m^{-1} and b(r) is a clean-air column. A non-zero clean-air column could be due to ambient levels of the target species, an interference species or to a range-dependent offset in the path-concentration integral column as a result of beam asymmetries and misalignments. In the case of CH_4, b(r) is typically a fixed value, due to the ambient CH_4 background, which is measured from the scan's last elevation angle, which does not contain emissions from any CH_4 source.

The range-resolved concentration (C) can be calculated as the difference between two datapoints separated by the sampling spacing l:

$$C(r) = \frac{CL(r + l/2) - CL(r - l/2)}{l} \quad (5)$$

For this DIAL system, l = 3.75 m provides concentration datapoints every 3.75 m along the optical path. A vertical DIAL scan is achieved by recording several range-resolved path measurements at different elevation angles, thereby helping determine CH_4 concentration on a two-dimensional map. This direct measurement of the spatial distribution of the gas then helps to spatially separate different sources over the area of interest. Concentration data across the entire plume section (C_{plume}) are combined with the vertical wind field profile $\bar{v}$ (consisting of the average wind direction and logarithmic wind speed profile during the measurement period) to determine the DIAL emission rate (E_{CH4}) from each scan (Innocenti et al., 2017):

$$E_{CH4} = \sum_h C_{plume,h}\ \bar{v}_h = \sum_h C_{plume,h}\ v_h \sin\vartheta \quad (6)$$

where $C_{plume,h}$ and v_h are plume concentration and wind speed at elevation h, and ϑ is the angle between the average wind direction and the DIAL vertical measurement plane.

Wind data were collected from a fixed meteorological mast with four sets of wind sensors (Models A100K and W200P, Vector Instruments, UK) elevated 11.9 m, 9.0 m, 6.2 m and 3.4 m above the ground and located to the south of plant 1 (see SM 1 Fig. S3). The measurement paths, also called "lines-of-sight" (LOS), used for the DIAL scans conducted at plant 1 are illustrated in SM 1 Fig. S3. LOS were between approximately 30 and 150° in relation to the wind direction.

The DIAL measurements reported in this study are the average of a set of repeated scans made along each LOS, typically four, and they were conducted following a protocol that is part of a new European standard (EN 17628, 2022).

Based on a number of field comparison studies with known emission rates, the uncertainty of a DIAL measurement is typically between 5 and 20% (Barthe et al., 2015; Gardiner et al., 2017; Howes et al., 2018). The DIAL technique provides a better understanding of spatial emission distribution and therefore allows for a more comprehensive study of different emission sources at a biogas plant compared to TDM and IDM. More detailed information about DIAL can be found in Robinson et al. (2011) and Innocenti et al. (2017).

2.4. Measurement campaigns

The first comparative measurement campaign was carried out from 24.10.2016 until 28.10.2016 at plant 1, while the second campaign took place between 07.05.2017 and 12.05.2017 at plant 2. The measurement days are listed in Table 1 together with a short characterisation of the meteorological conditions as well as comments on the measurement procedures used by the off-site teams. DIAL was only used in the October campaign.

On 24.10.2016 and 08.05.2017, TDM performed an initial emission screening to identify the main emission areas for optimal tracer placement at the plant as well as potential hindering emission sources in the surrounding area (e.g. farms and manure tanks). On-site measurements were initiated by detecting leakages, while the remaining time was used to measure component emissions one at a time. In addition to whole-site emissions, DIAL was able to separate and quantify emissions from different source areas at the biogas plant, using variable wind directions during the measurement campaign. On individual days (24.10.2016, 28.10.2016, 07.05.2017), IDM checked the calibration of the used OP-TDLAS devices, using standard gases with known concentrations (500 ppm, 1000 ppm, 2000 ppm) as references. Furthermore, laser correction factors were determined when the two OP-TDLAS devices were set side-by-side (1 m separation). Due to restrictions relating to certain weather conditions (especially for IDM), the individual teams could not perform all concurrent measurements. However, the valid measurement periods overlapped with at least two off-site approaches (see SM 1 Table S1).

2.5. Methane retrieval test

Emission estimates acquired by TDM and IDM were validated using controlled on-site (plant 1: 26.10.2016, 27.10.2016; plant 2: 09.05.2017, 10.05.2017) and off-site (11.05.2017, 12.05.2017) releases of CH_4 with a known point source. At both biogas plants, CH_4 was released at the plant (on-site) in alternating (short) gas-on and gas-off periods, in order to separate the plant's CH_4 emissions from the known CH_4 release (Baldé et al., 2016; McGinn et al., 2006). The differences between the average gas-on and gas-off emissions were taken as an estimate of the CH_4 release recovery rate, assuming constant plant CH_4 emissions during the gas release experiment. The on-off release patterns were slightly adapted during the experiment (see Table 1) and varied between two or three intervals of 30 to 40 min CH_4 releases (3.7 ± 0.1 kg h^{-1}, gas-on phase) following a 30 min break (gas-off phase) at plant 1 and one long gas-on phase for 1 h (3.9 ± 0.1 kg h^{-1}) at plant 2.

CH_4 was released with a mass flow controller (FMA-2610A, Newport Electronics GmbH, Germany) via a diffuser unit (diameter 24 cm) at heights of 3.45 m (plant 1) and 0.8 m (plant 2) (see SM 1 Sections A and

Table 1
Overview of off-site measurement campaigns and meteorological conditions.

Date	Wind direction (°)	Ø Wind speed (m s^{-1})	Local stability	Ø Temp. (°C)	Controlled CH_4 release (kg CH_4 h^{-1})	Comment
Plant 1 (2016)						
24.10.	70	3.5	near-neutral	7.9	–	DIAL: whole-site emission measurement IDM: calibration check of used OP-TDLAS and laser correction factors TDM: initial emission screening (on-site)
25.10.	310-55	2.1	near-neutral	10.6	–	DIAL, IDM & TDM: whole-site emission measurements DIAL: measurements at gas upgrading & utilisation
26.10.	165-240	2.0	near-neutral	9.1	3.7 ± 0.1 (2 intervals for 40 min following a 30-min break)	DIAL, IDM & TDM: whole-site emission measurements DIAL: measurements in the digester tank area IDM & TDM: controlled on-site release test
27.10.	220-250	3.8	near-neutral	13.0	3.7 ± 0.1 (3 intervals for 30 min following a 30-min break)	DIAL, IDM & TDM: whole-site emission measurements DIAL: measurements in the digester tank & substrate storage areas IDM & TDM: controlled on-site release test
28.10.	no meteorological data available				–	TDM: whole-site emission measurements IDM calibration check of used OP-TDLAS and laser correction factors
Plant 2 (2017)						
08.05.	340-20	3.6	near-neutral	11.7	–	IDM & TDM: whole-site emission measurements TDM: initial emission screening
09.05.	320-40	2.3	neutral – unstable	8.4	3.9 ± 0.1 (for 1 h)	IDM & TDM: whole-site emission measurements and controlled on-site release test
10.05.	220-280	2.9	near-neutral	14.0	3.9 ± 0.1 (for 1 h)	IDM & TDM: whole-site emission measurements and controlled on-site release test
11.05.	120-200	2.4	near-neutral	20.8	3.9 (for 1 h)	TDM: whole-site emission measurements IDM & TDM: controlled off-site release test
12.05.	210-235	3.5	near-neutral	16.8	3.9 (for 2 h)	TDM: whole-site emission measurements IDM & TDM: controlled off-site release test

B; Hrad et al., 2021). At plant 2, additional CH_4 release experiments (3.9 ± 0.1 kg h^{-1} for 1-2 h via a diffuser unit) were conducted off-site in order to reflect the surrounding terrain of the biogas plant (see SM 1 Fig. S7).

2.6. Emission factor calculation

CH_4 emission factors (EF, CH_4 losses in % of produced/utilised CH_4) were calculated by relating CH_4 emission rates (kg h^{-1}) to either CH_4 produced in the digesters or the estimated utilised CH_4 of the CHP units. In the case of plant 1, average CH_4 production (weekly average) was used (information from the plant operator), while the amount of utilised CH_4 was estimated by assuming an electrical efficiency of 38% (based on manufacturer specifications minus a 5% tolerance adjustment according to ISO 3046-1 (2002) for the CHP units and an energy yield of 10 kWh m^{-3} CH_4 for plant 2.

2.7. Statistical analyses

Statistical analyses were conducted using the statistical software R Development Core Team (2021). For plant 1, the different off-site methods (DIAL, TDM, and IDM) were compared through an analysis of variance (ANOVA) and a Tukey post-hoc test. Additionally, a non-parametric analysis, using the package for general factorial designs (Friedrich et al. 2017), and non-parametric pairwise comparisons, using the nparcomp package (Konietschke et al. 2015), were performed, with each yielding similar results. For plant 2, the two different off-site methods, namely TDM and IDM, were compared using an analysis of covariance (ANCOVA), considering the CHP load as a continuous covariate. The statistical analyses did not consider uncertainties and detection limits of the measurement methods.

3. Results

3.1. Comparison of on-site emission results

3.1.1. Biogas plant 1

Table 2 summarises the component emissions quantified by the two on-site teams at biogas plant 1. By taking the sum of the quantified sources overall CH_4 emissions ranged between 1.1 kg h^{-1} (EF = 0.3% CH_4, team A) and 1.7 kg h^{-1} (EF=0.4% CH_4, team B). CHP and BUU accounted for 16–26% of these total emissions. The majority of emissions escaped through various leakages, which could only be quantified in isolated cases due to temporal constraints. Although overall CH_4 emissions determined by both on-site teams fell into a similar range, small differences caused by undetected sources and deviating measurement approaches were identified. For instance, on-site team B quantified a CH_4 emission rate of 0.55 kg h^{-1} from the off-gas from the container housing the biogas analysis instruments (GU1), whereas team A did not investigate this source. In the case of CH_4 emissions from the air-inflated double-membrane domes, on-site team B directly measured air volume flow at the inlet of the inflation-air blower, resulting in a continuously higher volume flow than noted by on-site team A, which carried out measurements at the inflation-air outlet.

3.1.2. Biogas plant 2

Overall component emissions ranged between 2.3 kg h^{-1} (EF=1.9% CH_4, team B) and 2.7 kg h^{-1} (EF=2.2% CH_4, team A; see Table 2). The three CHP units were identified as the main sources accounting for 84-86% of total emissions. The emission rate of the open digestate storage tank, quantified by on-site team A, agreed well with the value determined by on-site team B (team A: 0.2 kg h^{-1}, team B: 0.2 kg h^{-1}). Only team A measured CH_4 leakages at the gas-tight covered storage tank and the post-digester (0.1 kg h^{-1}). However, they represented only a small part (5%) of the total emissions. In addition, the measurement procedure used for the air volume flow rate of the air-inflated double-membrane domes deviated. While on-site team A directly measured air volume flow at the outlet of the inflation-air blower, on-site team B used fan data, thus leading to a higher emission rate (team A: 0.02 kg h^{-1}, team B: 0.1 kg h^{-1}).

3.2. Comparison of off-site emission results

Emission data for valid time intervals/scans determined by the individual off-site methods for both biogas plants are given in SM 2.

3.2.1. Biogas plant 1

Fig. 1 shows CH_4 emissions from biogas plant 1, determined by the

Table 2
Summary of CH_4 emissions from component sources quantified by the two on-site teams at biogas plant 1 and 2.

Description component source	On-site A	On-site B	Emission rates (kg CH_4 h^{-1}) On-site A	Emission rates (kg CH_4 h^{-1}) On-site B	Emission factor (% CH_4) On-site A	Emission factor (% CH_4) On-site B
Plant 1						
Stationary/channelled sources	CHP, BUU	CHP, BUU	0.29	0.26	0.07	0.07
CH_4 diffusion from the air-inflated double-membrane domes	digester (D1, D2), post-digester (PD), digestate storage (DS 1-3)		0.50	0.82	0.13	0.2
Ventilation grids from encapsulated sources (buildings)	3 containers (for CHP, biogas analyses), 1 building (compressor station)	below detection limit	0.10	–	0.02	–
Leakages	4 leakages (GP4, GU2, GU3, GU4)	4 leakages (GU1, GU2, GU3, GU4)	0.21	0.60	0.05	0.15
Total plant 1			***1.1***	***1.7***	***0.3***	***0.4***
Plant 2						
Stationary/channelled sources	CHP 1-3	CHP 1-3	2.30	1.93	1.85	1.56
Area source – open digestate storage	$n = 9$ (08.05.17 – 10.05.17), 2/3 liquid surface + 1/3 surface layer (max. value)	$n = 3$ (10.05.17), liquid surface	0.23	0.23	0.19	0.19
CH_4 diffusion from the air-inflated double-membrane domes	gas-tight covered DS (outlet 1)		0.02	0.12	0.02	0.10
Ventilation grids from encapsulated sources (buildings)	not quantified	CHP container	–	0.02	–	0.02
Leakages	3 leakages (2 at gas-tight covered DS, 1 at PD)	detected but not quantified	0.11	–	0.09	–
Miscellaneous	CH_4 diffusion from gully between PD & gas-tight DS	CH_4 diffusion from gully between D1 & D2	0.01	0.002	0.01	0.002
Total plant 2			***2.7***	***2.3***	***2.2***	***1.9***

BUU (biogas upgrading unit), CHP (combined heat and power), D (digester), DS (digestate storage tank), GP4 (leakage at connection between foil roof and digester wall, GU1&2 (off-gas from biogas analyses), GU3 (leakage at the hydraulic valve at BUU), GU4 (leakage at a flange under a hood), PD (post-digester)

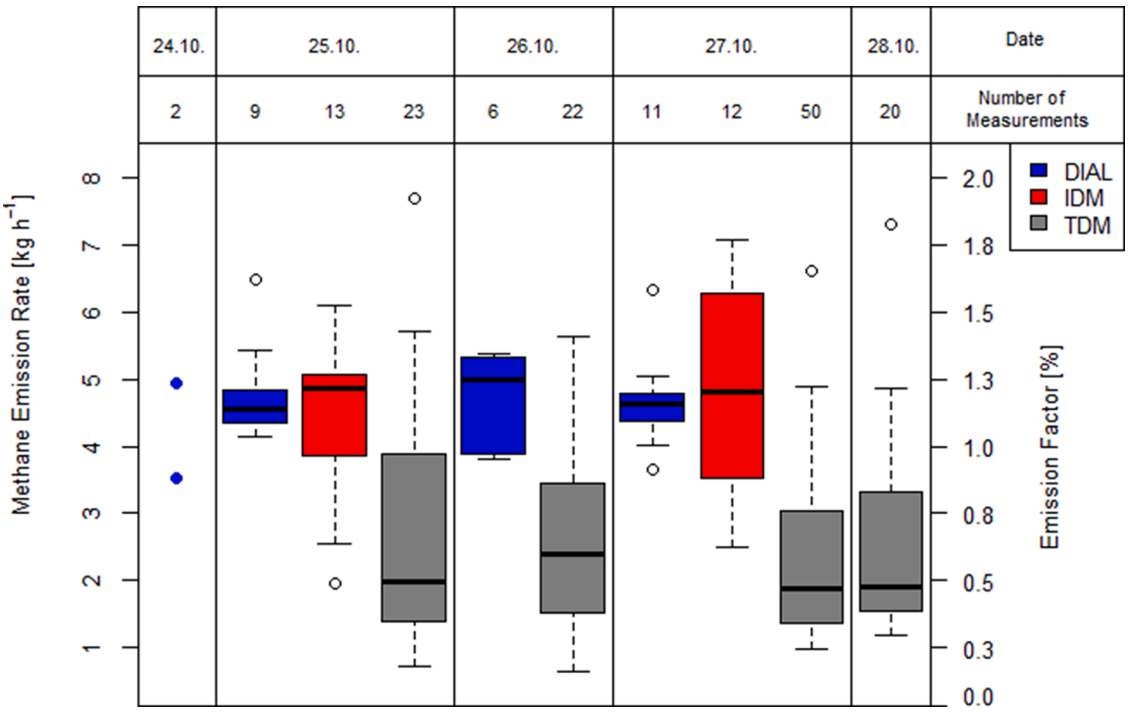

Fig. 1. CH_4 emission rates/emission factors from biogas plant 1, determined by three off-site teams (DIAL, IDM, TDM). If $n < 6$, individual measurements are plotted instead of boxplots (24.10.2016).

three off-site methods DIAL, TDM and IDM, separated by individual measurement days. The number of single measurements represents the number of downwind scans for the DIAL system (approx. 15 min per scan), the number of 10 min measurement intervals for IDM and the number of transects for TDM. We note that the measurements were not carried out exactly at the same time (see SM 1 Table S1), and they did not have the same time resolution.

DIAL quantified relatively invariant average CH_4 emissions on the different measuring days, with an average emission of 4.7 ± 0.7 kg h^{-1} (EF=1.2 ± 0.2% CH_4, $n = 28$) over the week. However, short emission peaks over three different days – up to an average of 10.9 kg h^{-1} (EF = 2.7% CH_4, $n = 5$) – were observed. Unfortunately, the measurement times for emission peaks did not overlap with any other off-site method (see SM 1 Table S1). As these emission peaks were identified as specific short-term events, they were not included in the calculated average emission rate. In addition, the operational data for the biogas plant provided no indication of the cause of the emission peaks. IDM quantified comparable average CH_4 emission rates of 4.7 ± 1.4 kg h^{-1} (EF = 1.2 ± 0.3% CH_4, $n = 25$). On 26.10.2016, plant emissions could not be revealed due to the filtering criteria required to remove periods when u* falls below the threshold value (see Section 2.3.2). TDM determined a similar emission pattern, albeit with lower average CH_4 emissions of 2.5 ± 1.4 kg h^{-1} (EF = 0.6 ± 0.4% CH_4, $n = 115$). Although average emission rates for the individual days were comparable, plume transects clearly indicated emission variations during the measurement campaigns (<1 to 6 kg h^{-1}).

One-way ANOVA followed by a Tukey post-hoc test procedure indicated that DIAL and IDM significantly differed from TDM ($p < 0.001$). A non-parametric analysis yielded the same result.

Fig. 2 presents emissions quantified by TDM and IDM during the controlled on-site CH_4 release (gas-on values) at biogas plant 1. Although IDM determined significantly higher plant emissions compared to TDM both methods revealed reasonable recovery rates ranging from 100-119%. TDM quantified average emissions of 2.3-2.6 kg h^{-1} during off-gas and 6.1–6.3 kg h^{-1} during on-gas periods giving a recovery rate of 100–103%. IDM delivered a recovery rate of 119%

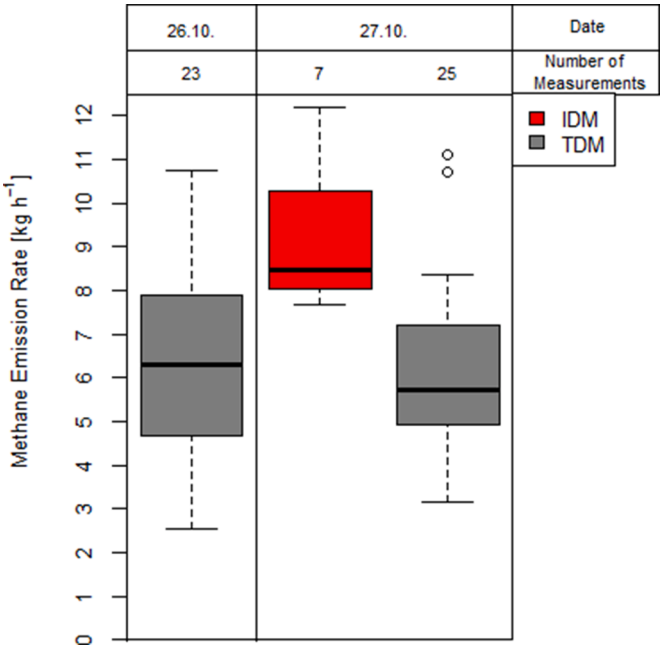

Fig. 2. CH_4 emission rates determined by IDM and TDM during the controlled CH_4 release (gas-on values) at biogas plant 1.

based on the differences between the average gas-on (9.3 ± 1.7 kg h^{-1}) and gas-off (4.9 ± 1.6 kg h^{-1}) emissions. Due to the relatively short period of the controlled releases, whole-plant emissions could not be determined with DIAL for the gas-on and gas-off phases respectively meeting the data quality requirements in the DIAL procedure.

3.2.2. *Biogas plant 2*

Fig. 3 illustrates emissions quantified by TDM and IDM at biogas plant 2, sorted according to CHP load and measurement day. In contrast

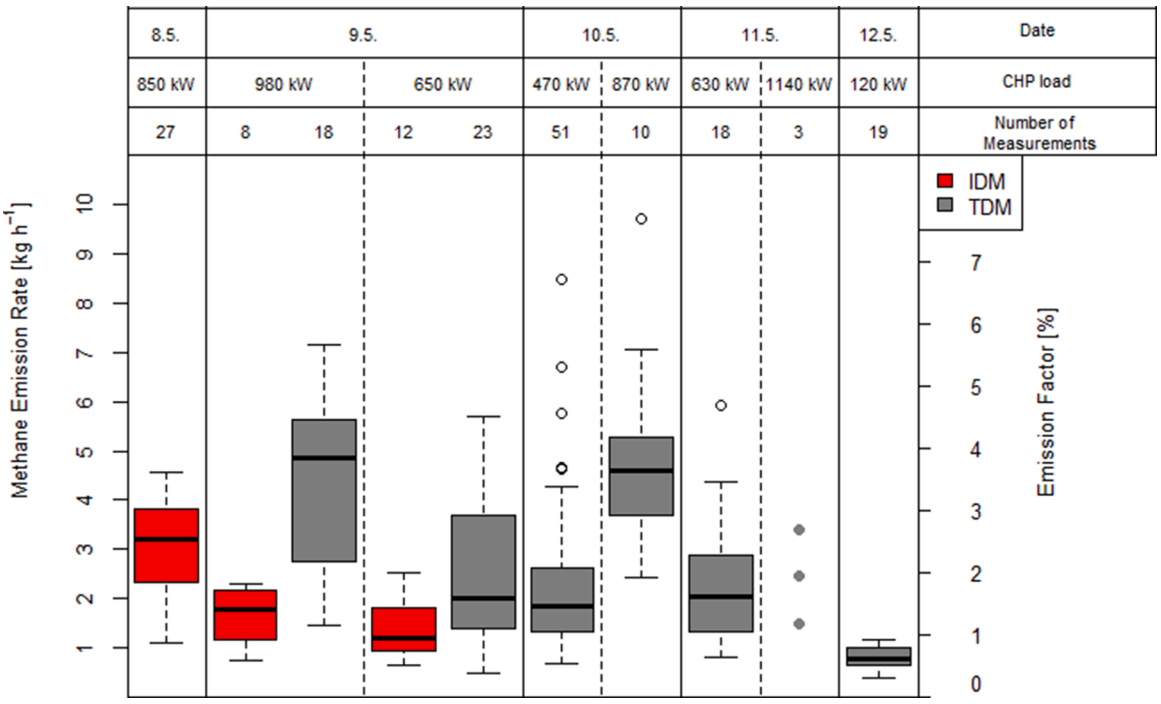

Fig. 3. CH_4 emission rates/emission factors from biogas plant 2, determined by IDM and TDM. If $n < 6$, individual measurements are plotted instead of boxplots (11.05.2017 for 1140 kW).

to biogas plant 1, overall CH_4 emissions from the second biogas plant were expected to change during the campaign due to the flexible operating mode of the CHP units (varying CHP load), which was also identified as the main emission source by on-site teams A and B (see Section 3.1.2).

TDM quantified an average CH_4 emission rate of 2.6 ± 1.9 kg h^{-1} (EF=2.1 ± 1.5% CH_4, $n = 142$) over the week. Large fluctuations with single high emission rates were measured (see Fig. 3). Emissions increased instantaneously by a factor of six to 25 when stirring the open digestate storage tank (data not shown or included in average emissions, as these were considered other than normal operating conditions). Also, IDM observed increased emission rates up to 13 kg CH_4 h^{-1} during stirring events (data not shown). Average emission estimates by IDM (2.4 ± 1.1 kg h^{-1}, EF = 1.9 ± 0.9% CH_4, $n = 47$) were similar to the TDM results, albeit with lower values on 09.05.2017 (1.5 ± 0.6 kg h^{-1}, EF = 1.2 ± 0.5% CH_4, $n = 20$). Due to low wind conditions, IDM could not determine plant emissions on 10.05.2017. In contrast to TDM, no whole-plant measurements were conducted by IDM on 11.05.2017 and 12.05.2017 (see Table 1).

However, an ANCOVA indicates statistically significant differences between TDM and IDM ($p < 0.001$) as well as the significant influence of variances in the CHP load ($p < 0.001$). Both methods were able to observe and quantify temporal emission variations with increasing emissions during increasing CHP loading (see SM 1 Fig. S6). Following an ANCOVA, the regression coefficient of the covariate CHP load was estimated at 0.0038, which was significantly different from zero ($p < 0.001$).

Fig. 4 shows emissions determined by TDM and IDM during the controlled CH_4 release at biogas plant 2. While TDM recovered the CH_4 release very well (recovery rate 93–118%, gas-on phase: 5.9–7.4 kg h^{-1}, gas-off phase: 2.3–2.7 kg h^{-1}), IDM experienced lower recovery rates based on average gas-on (4.1 ± 1.1 kg h^{-1}) and gas-off emissions (1.5 ± 0.6 kg h^{-1}) of 69%. However, Hrad et al. (2021) revealed that the use of a terrain model might be more suitable to reflect the moderately complex terrain of biogas plant 2 in a dispersion model. Using a terrain model in LASAT (Lagrange Simulation of Aerosol Transport), the recovery rate increased to 112% (gas-on phase: 6.7 ± 1.7 kg h^{-1}, gas-off

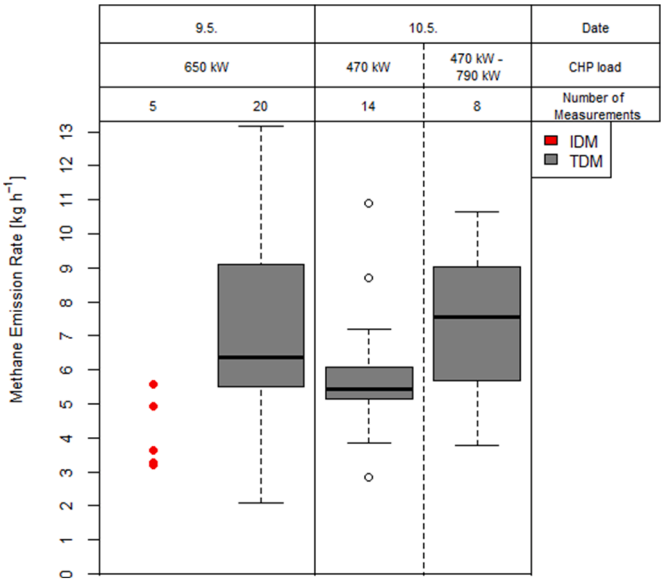

Fig. 4. CH_4 emission rates determined by IDM and TDM during the controlled CH_4 release (gas-on values) at biogas plant 2. If $n < 6$, individual measurements are plotted instead of boxplots (09.05.2017).

phase: 2.3 ± 1.5 kg h^{-1}; Hrad et al., 2021).

The controlled CH_4 release tests (3.9 kg h^{-1}) in the characteristic surroundings of plant 2 demonstrated that TDM and IDM were able to recover the released CH_4 with 8% uncertainty under favourable and similar conditions (Fig. 5). However, it is less problematic to determine emissions from a known point source (such as a gas bottle via a small diffuser unit) in comparison to a complex source with physical structures and different on-site emission sources. Release experiments on 11.05.2017 were not be determined by IDM, due to the filtering criteria not meeting the requests (see Section 2.3.2), while TDM could not record a sufficient number of plume transects (<10) according to Fredenslund

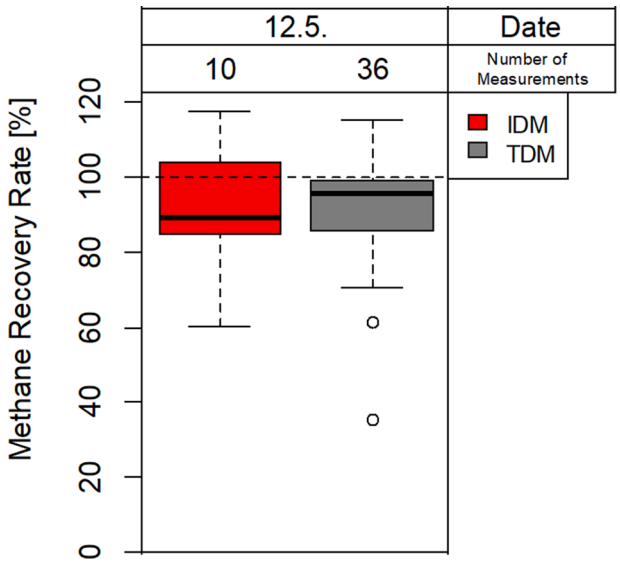

Fig. 5. CH_4 recovery rate during the controlled (off-site) release experiment.

et al. (2019).

3.3. Comparison of on-site vs. off-site emission results

At plant 1, the on-site teams quantified an EF of 0.3% CH_4 (1.1 kg h^{-1}, team A) and an EF of 0.4% CH_4 (1.7 kg h^{-1}, team B), both of which are lower compared to the values determined by the off-site methods (0.6–1.2% CH_4 or 2.5–4.7 kg h^{-1}). Hence, the on-site methods most likely underestimated overall emissions at this plant, which can be attributed to three main reasons. First, various small CH_4 leakages detected on the double-membrane foil roofs (gas-tight covered digesters) – most of which could not be quantified due to time constraints – accounting for the majority of emissions, Second, very low CH_4 slips in the exhaust gases of the biogas upgrading unit and CHP (0.3 kg h^{-1}; EF = 0.1% CH_4), due to the applied technology (chemical scrubbing; thermal post combustion after CHP). Kvist and Aryal (2019) also confirmed low CH_4 values by chemical scrubbers (0.04–0.1% CH_4). And third, leakages at other biogas plant locations (e.g. substrate storage, pipes, etc.) that could not be measured or were too small to detect.

During DIAL measurements of component emissions (see Table 1), four different areas could be differentiated: gas utilisation and upgrading, digester tank area, substrate storage and a combination of the digester tank and process areas (e.g. biogas analysis containers, compressor stations). While DIAL identified substrate storage as a minor emission source (0.3 ± 0.2 kg h^{-1}, EF 0.1 ± 0.1% CH_4), the remaining areas showed average emission rates in the range of 1.4–2.4 kg h^{-1} or 0.4 – 0.6% CH_4 (gas upgrading and utilisation: 1.4 ± 0.5 kg h^{-1}, digester tank area: 1.9 ± 0.4 kg h^{-1}; digester tank and process areas: 2.4 ± 0.3 kg h^{-1}). Except for substrate storage, emissions from the specific areas were even higher than the sum of all quantified sources of on-site teams A and B (1.1–1.7 kg h^{-1}, EF 0.3–0.4% CH_4). On-site results for the individual areas accounted for 21–58% of the emissions determined by DIAL. Differences in digester area emissions can be mainly explained by the many small (unquantified) leakages at the gas-tight covered digesters. However, the DIAL measurements were inherently consistent as the sum of area emissions (gas utilisation and upgrading, digester tank and process areas plus substrate storage) represented 87% of the whole-site emission rate.

For plant 2, the sum of the quantified emission rates from the on-site methods (2.3–2.7 kg h^{-1}, EF 1.9–2.2% CH_4) agreed well with the off-site ones (weekly average 2.4–2.6 kg h^{-1}, EF 1.9–2.1% CH_4). In this case, plant 2 represented a smaller biogas plant, with CHPs being the main CH_4 emission sources.

4. Discussion

4.1. On-site and off-site emission estimates

By comparing on-site and off-site methods at various biogas plants, Fredenslund et al. (2018) and Reinelt et al. (2017) confirmed that, apart from measurement uncertainties, the sum of on-site emission rates in most cases was lower than the off-site approach, due to undetected emission sources, difficult/impossible quantification of certain sources and short-term dynamics in the true CH_4 emission rate. In the study of Reinelt et al. (2017) none of the four on-site teams was able to quantify all individual sources, due to differences in available measurement equipment and variable operational emissions. A study of 268 natural gas production sites also suggested that CH_4 emission estimates derived from on-site measurements and simulations of unmeasured emission sources may estimate lower whole-site emissions than off-site methods (e.g. TDM with dual tracers) conducted at the same facility (Bell et al. 2017). The sum of CH_4 emissions from on-site methods could therefore establish a lower limit for whole-site emissions (Bell et al., 2017). Fredenslund et al. (2018) showed that the majority of the emissions from four biogas plants emanated from just a few sources.

Reinelt et al. (2017) observed that TDM determined higher CH_4 emissions than IDM from a Swedish biogas plant, due to varying measuring periods and operating conditions. In contrast, controlled release experiments with a CH_4 point source comparing eight different off-site approaches, including TDM and IDM with OP-FTIR, proved successful in terms of emission quantification at 10-20% uncertainty (Feitz et al., 2018).

Total CH_4 emission rates measured using TDM at 23 biogas plants ranged between 2.3 and 33.5 kg h^{-1} (0.4–14.9% CH_4) with an average emission rate of 10.4 kg h^{-1} or 4.6% CH_4 (Scheutz and Fredenslund, 2019). CH_4 emissions from agricultural biogas plants based on IDM have been shown to range between 3 and 5% of total CH_4 production (Bühler et al., 2022; Flesch et al., 2011; Groth et al., 2015; Hrad et al., 2015). In comparison, emission rates determined at the two biogas plants in this study were rather low, thereby making emission quantification more challenging.

4.2. Comparative advantages and limitations of the on-site and off-site methods

On-site methods identify and quantify single (component) emissions, using several sub-methods for different source types (e.g. channelled point sources, diffuse area sources, leakages, etc.) with quite low detection limits and easy implementation. However, chamber methods used for leakages or open digestate tanks, for example, may influence the emission pattern of the source. In addition, a leakage search performed with optical gas-imaging infrared video cameras might be restricted during unfavourable weather conditions, e.g. limiting wind speeds, low temperature differences between the temperature of the emitted gas and the background (Zeng and Morris, 2019; Zimmerle et al., 2020). Combined leak detection with a handheld CH_4 detector proved useful in exactly locating the leakage point and analysing CH_4 concentration. As revealed during the measurement campaigns at plant 1, not all sources could be quantified within a reasonable time frame, due to many small leakages and limited accessibility. However, the effort of such measurements may be adapted to plant size and purpose (e.g. the identification and quantification of main emitters), thereby enabling the surveying of many facilities.

Although off-site methods mainly focus on the quantification of whole-plant emissions, they might also be useful in differentiating between individual sources. DIAL measurements helped to spatially separate different sources across differentareas of plant 1, with the sum of these emissions being comparable to whole-plant emission. The unique advantage of DIAL is thereby the spatial resolution of the concentration of single gas species in two or three dimensions. TDM enables

the easy CH_4 screening of plant emissions and local emission sources, thus indicating the relative strength of emissions in different plant locations. Under certain circumstances (e.g. sufficient numbers of measurement paths, suitable plant layout for laser alignment), IDM could also be applied to simultaneously determine emission rates from individual sources at a biogas plant (e.g. several open digestate storage tanks – as in Hrad et al. 2014).

In contrast to DIAL and TDM, IDM relies on Lagrangian stochastic models to determine emission rates, resulting in high computational demand, depending on the duration of the measurement program. The simulation of turbulent transport in complex terrains/topographies might be restricted when using dispersion models assuming idealised conditions such as bLS according to Flesch et al. (2004) and as revealed at plant 2. The use of a terrain model seems preferable in this case (Hrad et al., 2021). However, IDM is more appropriate for continuous emission monitoring compared to DIAL and TDM, although this comparison study (measurements over a few hours per day) did not take advantage of IDM's strength, namely its ability to observe changes in emissions that occur infrequently or seasonally.

In terms of instrumentation, IDM should be based on simultaneous background and downwind concentration measurements (best case). If two OP-TDLAS are used, regular cross-checks between the lasers are needed in order to remove biased instrument errors. In contrast, DIAL and TDM determine the background concentration for each scan (concentration measured from the scan's last elevation angle, which does not contain emissions from the target source), and plume traverse (background concentration equals the plume baseline), respectively. For IDM, the careful alignment of a laser emitter and reflector is important in order to minimise data loss, which could be challenging during long-term measurements (Prajapati and Santos, 2018). Conversely, closed-path gas analysers used for TDM, as well as the pulsed laser for DIAL, are less prone to data losses (less influence of rain, fog and dust). However, atmospheric conditions with a few dust particles and aerosols can decrease the backscattered laser light intensity and the maximum distance (line-of-sight) of DIAL measurements.

Based on the applied filtering criteria for low wind conditions and the resulting data losses during the two measurement campaigns, IDM exhibited more restrictions in the presence of certain meteorological conditions compared to DIAL and TDM. Depending on topographical and infrastructural conditions (e.g. driveable roads, forest areas, dense array of buildings, other local CH_4 sources), all applied off-site methods might require specific target wind directions. When determining positioning for downwind measurements, it should be noted that DIAL does not provide data for the first 50 – 100 m, while IDM needs to fulfil certain distance requirements (according to the height of the largest wind obstacle and the distance between the source components). Larger measurement distances are preferable for TDM, thereby reducing potential errors associated with tracer gas placement and the measurement of the tracer gas release rate.

5. Conclusions

Our comparison and validation study indicated that on-site (component-scale) and off-site (facility-scale) methods are complementary. On-site measurements were in good agreement with off-site methods if the majority of emissions emanated from just a few sources. On-site methods therefore play an important role in identifying – as well as quantifying – main emission sources and could establish a lower limit for whole-plant emissions. Off-site methods are more reliable in determining whole-plant emissions, especially in cases of undetected, unquantified sources and varying operating conditions, the latter of which is an important aspect of data interpretation and analysis (e.g. varying CHP load). For surveys of many facilities, DIAL and TDM might be better suited, while long-term emission observation at a facility is best suited for IDM.

This study revealed significant differences between TDM and IDM at both plants while correcting the significant influence of the varying CHP load at plant 2. A definitive explanation of this difference could not be extracted from the data. A possible explanation, relevant to plant 2, might be the restricted simulation of turbulent transport across relatively complex terrain for IDM, assuming ideal conditions. Both methods observed similar emission patterns, namely relatively standard daily emissions at plant 1 with a constant CHP load, and increasing emissions with an increasing CHP load at plant 2. As true plant emissions are unknown, it cannot be established, which methods and measurements yielded the best results, as on-site CH_4 retrieval tests also gave no clear indications in this regard. However, our findings are limited by sample size (measurements were taken over just a few hours per day during a one-week measurement campaign per plant) and the restriction of the study to two investigated plants. It should be noted that it was difficult to conduct measurements with the different methods at the same time, due to micrometeorological, infrastructural, organisational and technical conditions. Variations in true plant emissions may therefore partly explain differences in quantified average emission rates for the different methods. In contrast to the complex sources and physical structure of a biogas plant, emission quantification from a known point source showed very good agreement between TDM and IDM. Given that all investigated methods are commonly used for determining emission rates from biogas plants, method measurement protocols, in particular off-site approaches, should be tested with more restrictive validation procedures, or demonstrate equivalency to standardised methods.

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

The authors confirm that the data supporting the findings of this study are available within the article and its supplementary materials.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.agrformet.2022.109179.

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