Waste Management 124 (2021) 82–93

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Quantification of methane emissions from UK biogas plants

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A R T I C L E I N F O

Article history:
Received 30 July 2020
Revised 21 November 2020
Accepted 8 January 2021

Keywords:
Biogas plant
Mobile survey
Fugitive methane emission
Gaussian plume modelling
Anaerobic digestion
Emission factor

A B S T R A C T

The rising number of operational biogas plants in the UK brings a new emissions category to consider for methane monitoring, quantification and reduction. Minimising methane losses from biogas plants to the atmosphere is critical not only because of their contribution of methane to global warming but also with respect to the sustainability of renewable energy production. Mobile greenhouse gas surveys were conducted to detect plumes of methane emissions from the biogas plants in southern England that varied in their size, waste feed input materials and biogas utilization. Gaussian plume modelling was used to estimate total emissions of methane from ten biogas plants based on repeat passes through the plumes. Methane emission rates ranged from 0.1 to 58.7 kg CH_4 hr^{-1}, and the percentage of losses relative to the calculated production rate varied between 0.02 and 8.1%. The average emission rate was 15.9 kg CH_4 hr^{-1}, and the average loss was 3.7%. In general, methane emission rates from smaller farm biogas plants were higher than from larger food waste biogas plants. We also suggest that biogas methane emissions may account for between 0.4 and 3.8%, with an average being 1.9% of the total methane emissions in the UK excluding the sewage sludge biogas plants.

1. Introduction

Methane (CH_4) is the second largest anthropogenic greenhouse gas (GHG) in terms of radiative forcing after carbon dioxide (CO_2). It is a substantial climate warmer because it has a 32 times larger heat-trapping capacity than CO_2 over a 100-year horizon (Etminan et al. 2016). According to Saunois et al. (2020), anthropogenic activities contribute approximately 60% of global methane emissions. Methane from the waste sector accounts for around 3% of global anthropogenic GHG emissions (Bogner et al., 2008), and about 12% of total global anthropogenic methane emissions for the 2008 – 2017 period (Saunois et al., 2020). Despite efforts taken in many countries to reduce emissions, CH_4 mole fraction is rising globally (Nisbet et al., 2020). According to Jackson et al. (2020), the global increases in anthropogenic methane emissions are equally distributed between agriculture and waste, mostly from Africa, Southern Asia and South America, and fossil fuel sources from China and North America. On the other hand, methane emissions are decreasing in Europe from agriculture, waste and fossil fuel sources (Jackson et al., 2020). The UK National Atmospheric Emissions Inventory (NAEI, 2020) suggests that in 2018 nearly 37% of methane emissions in the UK came from the waste sector, compared to an average proportion for European waste methane emissions of around 23% based on the 2020 European Environment Agency report (EEA,2020).

Anaerobic digestion is a waste management process for biodegradable materials which produces biogas and a stabilized digestate residue. Manure, food waste, organic industrial waste and sludge from sewage treatment are widely used in anaerobic digesters (AD) to generate biogas composed of 50 to 70% CH_4 and 30 to 50% CO_2, with traces of H_2S and NH_3 (UNFCC, 2017). Therefore, biogas production has numerous GHG mitigation impacts.

International Energy Agency (IEA) reported that biogas and biomethane production in 2018 could cover nearly 20% of worldwide gas demand (IEA, 2020). Europe is the largest biogas producer, followed by China and the Unites States accounting for 90% of global biogas production (IEA, 2020). According to World Biogas Association (2019), there are currently 132,000 small, medium and large-scale anaerobic digesters and 700 upgrading plants operating in the world. Production of renewable energy from biogas

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https://doi.org/10.1016/j.wasman.2021.01.011
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plants has grown in the UK. In 2019, 660 biogas plants were operating across the UK, with 955 megawatt electrical (MWe)-equivalent industrial capacity across both the electricity and biomethane-to-grid sectors, and there are plans for around 390 new plants with a combined capacity of 441 MWe-equivalent (ADBA, 2019). Generated biogas can be converted into electricity, heat or transport fuels. The Carbon Trust (2010) suggests that upgrading biogas to biomethane for use as a transport fuel or injecting it into the gas grid are the most efficient ways of carbon saving. Nevertheless, most biogas plants in the UK currently burn the biogas to produce electricity. Only 103 biomethane-to-grid biogas plants are in operation across the UK (ADBA, 2019).

Biogas plants can be the source of significant fugitive methane emissions. Recent studies have found that methane leaks may originate from various locations, including feedstock storage tanks, gas safety release valves from the digestion process, gas storage units, pipework, digestate storage tanks, flaring, foil roofs and wires and gas engine exhaust as shown in Table 1.

Table 1 summarizes the various approaches and methods adopted in scientific studies of methane emissions and their source at biogas plants in European and North American countries. There is a large variation in methane emission rates from different sources, but there is also a large variation in emission rates from similar sources in different countries, which makes it hard to draw general conclusions. Two main approaches have been used to quantify emissions rates: on-site and downstream quantification. An on-site approach can be used to identify and quantify single methane sources at a facility, whereas downstream measurements in combination with a source estimation based on mass balance methods or atmospheric transport modelling can help determine the overall emissions rates from biogas plant sources that are difficult to investigate or may be missed by on-site methods (Reinelt et al., 2017). Therefore, an approach with downstream atmospheric measurements was used in this study.

In recent years more waste has been diverted to alternative treatments such as biogas plants and less has been landfilled (see the supplementary information S1, NAEI, 2020). The rapid growth of the biogas industry raises new challenges regarding emissions monitoring, quantification and reduction. Fugitive emissions from biogas plants are not yet well quantified in the UK, and most are not yet included in the NAEI emissions inventory because they were built in the last decade (ADBA, 2019). Therefore, the objective of this study was to quantify methane emission rates from biogas plants in the UK fed by different materials, including food and farm waste to highlight the importance of the various feedstock types to biogas plant emissions. To the best of the authors' knowledge, this is the first biogas plant mobile measurement study to estimate methane emission rate in the UK. This paper quantifies the emission rates from various biogas plants and projects these rates for the next decade, emphasizing the need for better regulation and monitoring. Worldwide, very few wholly independent emissions studies have been carried out on biogas plants, and few studies have been mobile. Apart from the work of Scheutz and Fredenslund (2019), few have included a population of biomass facilities large enough to permit conclusions about national emissions.

2. Methods

2.1. Sampling locations

Mobile surveys of greenhouse gases in southern England were conducted between 2018 and 2020 to examine a wide range of sources. The prevalence of methane plumes originating from biogas plants resulted in the design of a focused study to quantify the emissions of biogas plants with suitable downwind access roads for plume transects under prevailing meteorological conditions. Most biogas plants started operating in the last few years. The official biogas map of the UK was used to locate biogas sources (NNFCC, 2019). Before each campaign, biogas plants to be included in the survey were selected according to accessible public roads and suitable prevailing wind direction. Most sites were visited at least twice to investigate whether the facilities produced sustained CH_4 emissions. During the first survey, ambient air samples were collected for isotope analysis which is beyond the scope of this paper and is the object of another research project. Data from specific visits were used for emission rate calculation.

Fifty-six biogas plants were surveyed but emissions could be quantified for only 10 of them for this study. Fourty-six plants were excluded for several reasons. For instance, it was not possible to get close enough to eight of them to measure significant emission plumes, and for another seven the wind direction meant that the expected plumes could not be properly detected from public roads. Nine were not used in emissions rate calculations because they were close to other methane sources, such as landfill and composting facilities, and their plume shape was unsuitable for Gaussian plume modelling (see supplementary information S2). The remaining 22, where access and atmospheric conditions were favourable, may not have been emitting or operating at the time of the survey. Therefore, ten biogas plants with suitable plume emissions were selected for further research. CH_4 mole fractions (ppm) were measured across the CH_4 plume emitted by each site according to prevailing wind direction and speed.

Based on the biogas inventory, two main types of biogas plants were investigated (agricultural farm and food waste) which differed in size and feedstock material. The selected biogas plant sites and publicly available information on their facilities are listed in Table 2. Plants A to D are food waste biogas plants, and the remainder are agricultural biogas plants ordered by their electrical capacity. For nearly all biogas plants, some information is not in the public domain, so certain features of their facilities are unknown. Sewage sludge anaerobic digesters were not included in this study because most are installed in wastewater treatment plants, and without site access it would be challenging to distinguish whether their CH_4 emissions originated from the wastewater treatment or sludge anaerobic digestion process.

Four of the biogas plants (plants A to D) are categorised as "waste", which means that feedstock comprises mainly of the food waste from commercial and industrial processes and separated biodegradable waste from municipal sources. Plants A to D are called food waste biogas plants though biogas plant B receives additionally 30% of animal slurry as feedstock (NNFCC, 2019). All of the waste biogas plants were built in 2015 or later (Environment Agency, 2020). Plant A is the oldest one that got permission in 2015.

Six of the biogas plants (plants E to J) are classified as "farm". These plants utilise predominantly agricultural feedstock such as manures, slurries, energy crops and crop residues (NNFCC,2019). Three out of these plants (plants F, G and H) rely on energy crops such as maize silage, vegetable out grades and grass silage, respectively. The remaining three plants receive a combination of different feedstocks. Plant E receives 56% of potato waste, 42% of chicken manure and 2% of maize silage. Plant I is fed with 57% of cattle manure and 43% of maize silage. Plant J receives 49% maize silage, 33% animal slurries and 18% of grass silage. The age information is only available for plant E, which was constructed in 2019. According to the "Environmental Permitting Regulations" in England and Wales (Environment Agency, 2020), there is exemption for small-scale non-waste facilities permit, so the construction information of biogas plants F to J is not available on Environmental Agency permitting data (Environment Agency, 2020).

Table 1
Review of emissions measurements and methane emission factors determined from various biogas plant studies in Europe and North America.

Type of feedstock	Emissions sources	Location	Measurement method	Emission loss range	Emission factor range (%)	References
Pig manure	Around digester pits, gravity thickener	USA	Inverse dispersion modelling method with open path TDLAS and a bLs model	9.7 – 68.8 kg CH_4 h^{-1}		Harper et al. (2010)
Cattle manure, organic feedstock	Run-off pond, flaring, feedstock hopper, digester	Canada	Inverse dispersion modelling method with open path TDLAS and bLs model	0.7 – 32.7 kg CH_4 h^{-1}	0.5 – 25.0	Flesch et al. (2011)
WWTP sludge	Foaming event in anaerobic digester	Denmark	Trace gas dispersion method with controlled release gas tracer	4.99 – 92.3 kg CH_4h^{-1}	2.1 – 32.7	Yoshida et al. (2014)
Energy crops and liquid manure	Digesters, storage tanks, CHP units	Germany	Inverse dispersion method with open path TDLAS	7.2 – 57.6 kg CH_4 h^{-1}	3 – 23	Groth et al. (2015)
Food waste, food industry residuals, alcohol, thin stillage, fat, slaughterhouse	Leakages, diffuse emission from digestate storage tank, CHP units	Sweden	IR camera and high-volume sampler system/ open and closed chamber, portable detector	5.3 – 9.8 kg CH_4 h^{-1}	0.61 – 1.14	Holmgren et al. (2015)
Food waste, food industry residuals, alcohol, thin stillage, fat, slaughterhouse	Leakages, diffuse emissions from digestate storage tank, CHP units	Sweden	Inverse dispersion modelling method with open path TDLAS and bLs model and controlled release gas tracer	4.9 – 24.5 kg CH_4 h^{-1}	0.6 – 3	Holmgren et al. (2015)
Energy crops and pig manure	Filling and emptying of digestate storage tanks	Austria	Inverse dispersion modelling method with open path TDLAS and Lagrangian particle dispersion model	5.4 – 7.2 kg CH_4h^{-1}	3 – 4	Hrad et al. (2015)
Dairy manure, food waste	Digestate storage	Canada	Inverse dispersion modelling method with open path TDLAS and bLs model	0 – 97 kg CH_4 h^{-1}	12	Baldé et al. (2016)
Slaughterhouse, food industry and household waste	Open digestate storage and PRV	Sweden	Inverse dispersion modelling method with open path TDLAS and bLs model	5 – 25 kg CH_4 h^{-1}	0.6 – 3	Reinelt et al. (2017)
Slaughterhouse, food industry and household waste	Open digestate storage and PRV	Sweden	IR camera and a portable methane laser with IR sensor	5 – 17 kg CH_4 h^{-1}	0.6 – 2.1	Reinelt et al. (2017)
Integrated WWTP sludge	Sludge treatment and energy production units	Scandinavia	Tracer gas dispersion method with controlled tracer gas released with inverse Gaussian plume modelling	1.1 ± 0.1 – 18.1 ± 6.3 kg CH_4 h^{-1}	1.1 – 21.3	Delre et al. (2017)
Wet source-separated organic household waste	Anaerobic digester	Germany	Tracer gas dispersion method with controlled tracer gas released	28.5 ± 6.1 kg CH_4 h^{-1}		Jensen et al. (2017)
Manure, organic waste	Whole plant	Denmark	Remote sensing: tracer gas dispersion	3.3 and 9.5 kg CH_4 h^{-1}	1.4	Fredenslund et al. (2018)
Manure, organic waste	Pipe and PRV, leaks, gas engine, building ventilation	Denmark	On-site: optical gas imagining IR camera	3.4 kg CH_4 h^{-1}	0.8	Fredenslund et al. (2018)
WWTP sludge	Whole plant	Denmark	Remote sensing: tracer gas dispersion	13.5 ± 0.5 kg CH_4 h^{-1}	8.3	Fredenslund et al. (2018)
WWTP sludge	Open digestate storage, PRV, manhole cover, sludge pump tank, CHP units	Denmark	On-site: optical gas imagining IR camera	6.5 kg CH_4 h^{-1}	4	Fredenslund et al. (2018)
Manure, slaughterhouse waste	Whole plant	Denmark	Remote sensing: tracer gas dispersion	5 – 35 kg CH_4 h^{-1}	1.9	Fredenslund et al. (2018)
Manure, slaughterhouse waste	Water scrubber, biomass tank, digestate-gas storage unit air outlet	Denmark	On-site: optical gas imagining IR camera	27.8 kg CH_4 h^{-1}	4.1	Fredenslund et al. (2018)
Manure, straw, maize silage	Whole plant	Denmark	Remote sensing: tracer gas dispersion	13.4 ± 0.5 kg CH_4 h^{-1}	3.3	Fredenslund et al. (2018)
Manure, straw, maize silage	Gas outlets from receiving tank, mixing tank and ventilation from biofilter and gas building, leaks from PRV and caps	Denmark	On-site: optical gas imagining IR camera	15.4 kg CH_4 h^{-1}	3.8	Fredenslund et al. (2018)
Sludge	Whole integrated plant	Sweden	Mobile ground-based remote sensing method with tracer gas dispersion method	23.4 – 38.8 kg CH_4 h^{-1}	1.1 – 32.7	Samuelsson et al. (2018)
Sludge and industrial food waste	Ventilation of thickening and dewatering building	Sweden	Mobile ground-based remote sensing method with tracer gas dispersion method	1.6 – 4.8 kg CH_4 h^{-1}		Samuelsson et al. (2018)
Sludge and industrial food waste	Biosolids stockpiles	Sweden	Mobile ground-based remote sensing method with tracer gas dispersion method	3.7 – 28.9 kg CH_4 h^{-1}		Samuelsson et al. (2018)

Table 1 (*continued*)

Type of feedstock	Emissions sources	Location	Measurement method	Emission loss range	Emission factor range (%)	References
	Biogas upgrading plants: amine scrubber	Denmark	On-site: sampling and analytical measurement		0.04 – 0.07	Kvist and Aryal (2019)
	Water scrubber	Denmark	On-site: sampling and analytical measurement		1.1 – 1.97	Kvist and Aryal (2019)
	Membrane technology	Denmark	On-site: sampling and analytical measurement		0.48 – 0.56	Kvist and Aryal (2019)
Agricultural: manure and energy crops	PRV	Germany	On-site: explosion-proof flow velocity and temperature sensor		0.5 – 9.9	Reinelt and Liebetrau (2019)
Agricultural	Whole plants	Denmark	Tracer gas dispersion	2.3 – 23.2 kg CH_4 h^{-1}	0.4 – 8.6	Scheutz and Fredenslund (2019)
WWTP	Whole plants	Denmark	Tracer gas dispersion	2.6 – 33.5 kg CH_4 h^{-1}	2.2 – 14.9	Scheutz and Fredenslund (2019)

bLS: backward Lagrangian stochastic model; IR: infrared camera; LN: normal litre; PRV: pressure relief valve; TDLAS: tunable diode laser absorption spectroscopy;WWTP: wastewater treatment plant

The biogas plants vary with respect to gas utilisation (Table 2). At most plants, all or some of the produced biogas is utilised in a CHP unit except for plants D and E. However, it is not publicly known whether all gas utilisation occurs off-site or on-site of the plants.

Plants with electrical capacity lower than 0.99 MWe were categorized as small plants, those in the range 1 – 1.99 MWe as medium-sized, and those higher than 2 MWe as large. In this study, CH_4 emissions from two large (A and B), three medium (C, D, and E) and five small biogas (F, G, H, I and J) facilities were monitored by mobile surveys.

2.2. Mobile surveys

The mobile monitoring campaigns used a Picarro G2301 CRDS (cavity ring-down spectroscopy) analyzer providing measurements of CO_2 and CH_4 dry mole fractions in ppm and water vapour (H_2O) in % every three seconds. The Picarro mobile module (A0941) included a hemisphere GPS receiver for a continuous record of location. Four 12 - volt, 110 Ah lead-acid batteries allowed the Picarro instrument to run for up to nine hours. The air inlet and the GPS were attached to a mast on the roof of the vehicle approximately 1.8 m above the ground. The GPS was synchronized with Picarro output data, allowing the data to be mapped during subsequent analysis. There was a seven- to nine-second time delay between 1 Hz (1 s) frequency GPS location and the delay from air intake to the actual air sample analysis, which allowed CH_4 spikes to be quickly pinpointed (see Zazzeri et al., 2015 for details). The Picarro was controlled by a laptop connected by wi-fi, so CH_4 was monitored continually while travelling.

The mobile instrument was calibrated to the WMO X2004A CH_4 scale once a month in the greenhouse gas laboratory at Royal Holloway University of London. Calibration-standard dry air cylinders were filled and measured by NOAA, and tertiary-standard cylinders were filled and calibrated by MPI-Jena Gaslab.

During the mobile surveys, the vehicle was driven along public roads close to the target biogas plants, typically at a speed of 25 – 30 km per hour (km hr^{-1}). When a plume was detected downwind of a given source, it was transected multiple (ideally five or more) times for later emission rate analysis. The wind speed and direction were measured by a hand-held anemometer between transects and the atmospheric conditions such as wind speed and direction were also confirmed by data from local meteorological stations. The dates and time interval of measurement used for Gaussian plume modelling, wind speed and directions, numbers of plume transects and measuring distance to biogas plant sites are given in Table 3. It should be noted that none of them are included in the 2018 NAEI methane emission point source inventory (NAEI,2020).

2.3. Data processing for Gaussian plume modelling

After each survey, the raw data were corrected according to calibration standards and inlet lag times to match the locations of spikes with measured mole fractions. Calculated excess CH_4 over the background (ppb) was used for emissions rate estimation. The excess CH_4 mole fraction was calculated by subtracting a moving background, defined as the lowest second percentile from a ± 10-minute moving average, to take into consideration changing background conditions in space and time. Typical variation of background mole fraction was ± 5 ppb during the plume transects.

In this study, the Gaussian plume model describing the mole fraction of a gas as a function of downwind distance from a point source (Seinfeld and Pandis, 2006) was used to estimate the emissions rates. Pasquill and Smith (1983) developed this model to predict the above-ambient ensemble average plume concentration C (x,y,z).

Table 2
Overview of main characteristics of investigated biogas plants (obtained from NNFCC, 2019 and Local Authority's web sites).

Name[a]	Type of feeding materials	Main substrate(s)	Total feedstock (tonnes)	Capacity (KWe)	Tallest height of facility (m)	Digestate storage (open/closed)	Gas utilization	Age of facility (year)
A	Waste	Food waste	45,000	2,600	19	N/A	CHP	5
B	Waste	Animal manures, food waste & organic fraction of MSW	50,000	2,000	N/A	N/A	CHP	1
C	Waste	Food waste	49,000	1,519	N/A	N/A	CHP	3
D	Waste	Commercial food waste	50,000	1,411	12.4	Closed	BtG & CHP	4
E	Farm	Poultry manure and energy crops	19,262	1,000	12.5	N/A	BtG & CHP	1
F	Farm	Maize sludge	20,000	989	6	Closed	CHP	N/A
G	Farm	Vegetable out grades	10,000	500	N/A	N/A	CHP	N/A
H	Farm	Grass silage	2,964	500	12.75	N/A	CHP	N/A
I	Farm	Maize silage & cattle manure	56,400	487	N/A	N/A	CHP	N/A
J	Farm	Animal slurries, maize and grass silage	12,800	485	N/A	N/A	CHP	N/A

AD: anaerobic digester; BtG: biomethane to natural gas grid; CHP: heat and/or power; MSW: municipal solid waste; N/A: not available
[a] We surveyed on public roads independently of the biogas plant operators so the sites will remain anonymous.

Table 3
Overview of the measurement surveys, which were utilized for emission rate calculation.

Site name	Date of measurement	Measuring time interval	Wind speed (m s^{-1}) and direction	Number of Gaussian plumes	Measured distance to biogas plant site (m)
A	30 – 01 – 2020	15:30 – 16:05	2.5 – 10, SE/S/SW	5	410 – 490
B	11 – 03 – 2019	14:45 – 15:20	3.5 – 14, W	3	110 – 230
C	23 – 05 – 2019	17:00 – 18:30	0.5 – 2, S	2	90 – 100
D	01 – 11 – 2019	14:25 – 15:00	2.3 – 9.2, SW	5	300 – 350
E	18 – 02 – 2020	15:40 – 16:05	3.7 – 15, WSW	6	120 – 200
F	01 – 11 – 2019	12:00 – 12:25	2.3 – 9, SW	3	200 – 400
G	01 – 11 – 2019	11:00 – 11:35	2.3 – 9, SSW	6	360 – 700
H	23 – 05 – 2019	14:25 – 14:55	0.5 – 2, WSW	3	160 – 200
I	21 – 01 – 2020	10:20 – 10:45	0.5 – 2, WNW	6	110 – 220
J	05 – 03 – 2020	14:40 – 15:15	3 – 12, NNE	9	220 – 270

$$C(x,y,z) = \frac{Q}{\pi\sigma_y\sigma_z u} \exp\left(-\frac{y^2}{2\sigma_y^2}\right)[\exp\left(-\frac{(z-h)^2}{2\sigma_z^2}\right) + exp\left(-\frac{(z+h)^2}{2\sigma_z^2}\right)] \quad (1)$$

where C is the concentration averaged over time t; x is the distance downwind; y is the distance crosswind; z is the height above ground level; Q is the source strength; σ_y and σ_z are the "dispersion coefficients" representing the crosswind and vertical mixing of the plume, respectively, depending on the atmospheric stability; u represents the vertically and time averaged wind speed; and h is the height of the release.

As the CH_4 is emitted, it disperses in y and z directions with respect to time. The concentration of methane (C, μg/m^3) depends on the source strength (Q, kg/h), the advective wind speed (u, m/s) and the rate of dispersion. Dispersion coefficients are calculated from the Pasquill-Gifford atmospheric stability classification.

When using the Gaussian plume model, it is assumed that the source emits at a constant rate, that diffusion in x direction is negligible, that methane mass is conserved, that there are no sinks or additional sources during transport, that the horizontal wind shear effect is negligible on a given horizontal elevation, and that the wind speed and vertical eddy diffusivity are constant with time, and that molecular diffusion is negligible compared to turbulent diffusion.

To calculate the emissions rate, the method first required each methane concentration plume to be identified. The individual plumes sampled for biogas plant A are illustrated in Fig. 1. The plumes closely follow a Gaussian shape. Second, the distance of each plume's centre to the source was calculated using the peak plume location and known source location. Mobile transects were usually not perfectly perpendicular to the wind; therefore, the angle between the road and the axis of each plume (i.e. the line connecting the source and the plume centre) was calculated using the synchronous GPS information converted into a Cartesian coordinate system in units of meters. Third, the flux through a control surface over the range [1, 2] meters and a horizontal axis spanning the width of the plume along the vehicle transect was computed considering the prevailing wind speed and direction. Fourth, the modelled flux was computed using input data on the source release rate, calculated distance to source, wind data from the survey day, and horizontal and vertical dispersion coefficients, $\sigma_y(x)$ and $\sigma_z(x)$, calculated following Briggs (1974) for different atmospheric stability classes according to each survey day's insolation and cloud cover conditions (Pasquill, 1974). Finally, the source strength of each peak was calculated from Eq. (2). A flow chart illustrating the methodology is shown in supplementary information S3. The reported emission rates were calculated as the average over all transects for the biogas sites' various source heights and wind data.

$$Q = \frac{\int measured, CH4}{\int model, CH4} \times input\ source\ strength\ (gs^{-1}) \quad (2)$$

The unknown variable was the height of the point of the release. The emissions may have come from the ground through a leaking pipe, pump or valve, or from a digestate storage tank, the dimensions of which are not publicly available for some biogas plants. Therefore, emission rates were calculated considering a range of release heights for each biogas plant site.

The uncertainty of the emission rate was derived from the unknown height of the source, the distance from the source, potential atmospheric stability class discrepancy, variability in wind speed and direction, and variation in each transect. Owing to limited information available on the biogas plant sites, a different range of heights was taken. The expected uncertainty in the wind speed data was taken as 50%, as recommended by Caulton et al. (2018). The propagation of uncertainty was computed as the square root of the squares of the uncertainty of individual parameters.

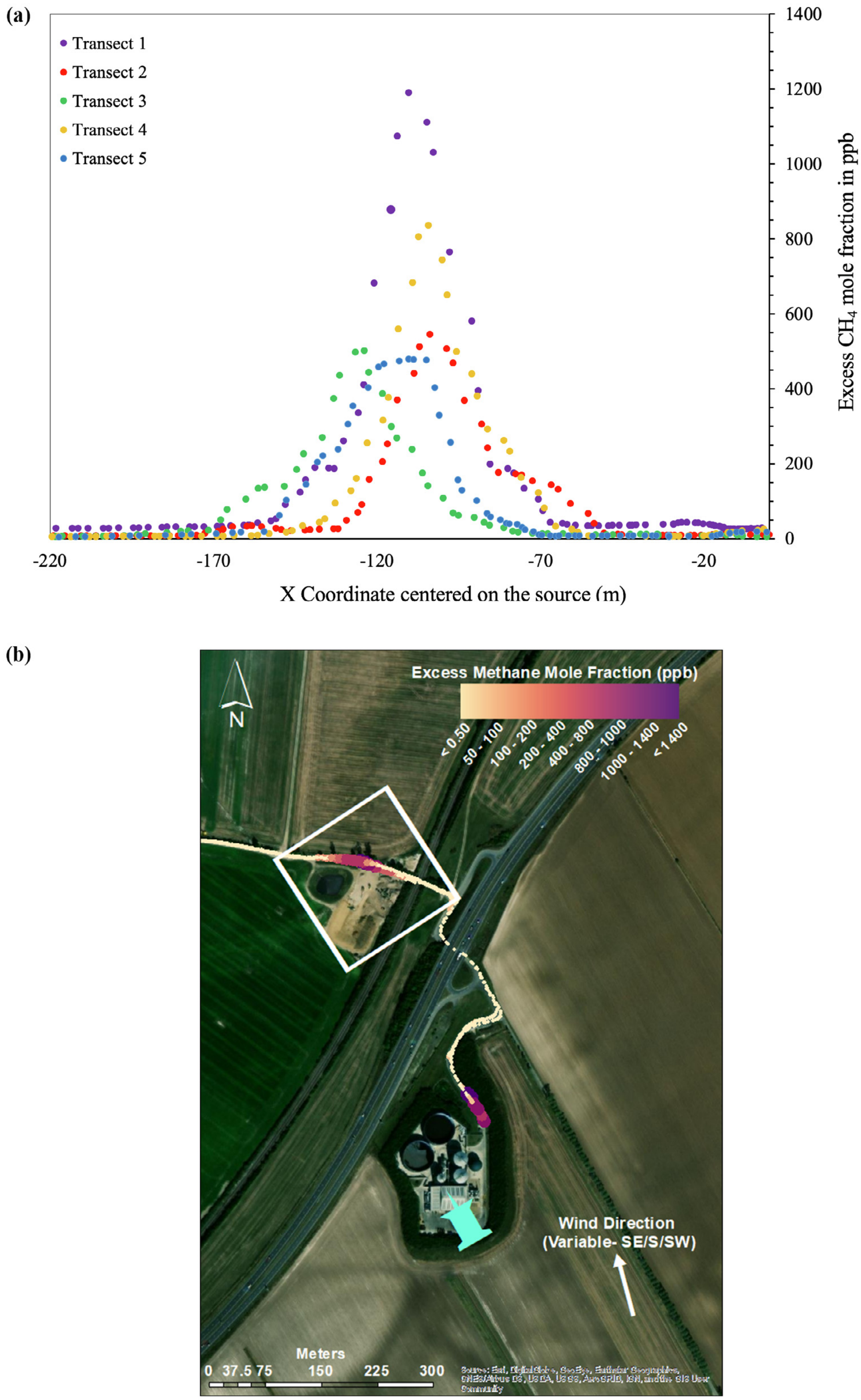


Fig. 1. **a)** Downwind plumes of biogas plant A sampled during a 1-hour period of a mobile survey on 30 January 2020. **(b)** ArcGIS map of excess CH_4 mole fraction above background in ppb, as a top view of downwind plumes of plant A. White arrows show the wind direction. White box represents the data used in emission rate calculation. Green pin indicates the site of biogas plant A. The surveyed road was reached after passing under the highway. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 4
Estimated methane emissions rates obtained from Gaussian plume modelling, methane losses relative to calculated production rates and emission factors calculated as annual emission rates divided by annual feedstock amount.

Name	Biomethane capacity (Nm^3/hr)	Calculated average CH_4 production rate (kg CH_4 h^{-1})	Estimated total CH_4 emissions (kg CH_4 h^{-1})	CH_4 loss relative to calculated production rates (%)	Emission factors (kg CH_4 emitted/ tonnes of feedstock)
A	N/A	970[b,c]	12.6 ± 3.8	1.3 ± 0.4	2.5 ± 0.7
B	N/A	861 [a,c]	58.7 ± 25	6.8 ± 2.9	10.3 ± 4.4
C	N/A	654 [a,c]	0.1 ± 0.02	0.02 ± 0.003	0.02 ± 0.004
D	990	709	2.8 ± 0.8	0.4 ± 0.1	0.5 ± 0.1
Plant average CH_4 loss and EF, **food waste**: 2.1% and 3.3, respectively					
Production weighted average CH_4 loss and EF, **food waste**: 2.3% and 3.4, respectively					
E	550	394	21.9 ± 6.2	5.6 ± 1.6	10.0 ± 2.8
F	N/A	425 [a]	14.3 ± 4.2	3.4 ± 1.0	6.3 ± 1.8
G	N/A	215 [a,c]	17.5 ± 3.7	8.1 ± 1.7	15.3 ± 3.2
H	N/A	198 [a]	0.5 ± 0.1	0.3 ± 0.1	1.5 ± 0.3
I	N/A	439 [a,c]	14.0 ± 3.9	3.2 ± 0.9	2.2 ± 0.6
J	N/A	209 [a,c]	16.6 ± 4.1	7.9 ± 0.02	11.4 ± 2.8
Plant average CH_4 loss and EF, **farm waste**: 4.8% and 7.8, respectively					
Production weighted average CH_4 loss and EF, **farm waste**: 4.5% and 6.1, respectively					
All biogas plants					
Plant average CH_4 loss and EF, all: 3.7% and 6.0, respectively					
Production weighted average CH_4 loss and EF, all: 3.1% and 4.4, respectively					

[a] results estimated by interpolation; [b] results found in public reports; [c] methane content of 60% and normal conditions (25 °C and 1 atm); CH_4 density = 0.7157 kg/Nm^3 at normal conditions (25 °C and 1 atm); EF: emission factor; plant average is equal to the sum of CH_4 losses divided by the number of the plants and weighted average is equal to the sum of the all estimated CH_4 emissions rates divided by the sum of calculated production rates.

3. Results and discussion

3.1. Emissions rate calculation

Table 4 presents the calculations of emission rates, losses and emission factors (EF) for the 10 biogas plants in this study, including their biomethane production capacity in Nm^3/hr. The biomethane capacities of plants D and E are recorded in the Official Information Portal on Anaerobic Digestion (NNFCC, 2019). However, most do not report daily production rates, so their production rates were estimated by linearly interpolating relative to known capacity (KWe) values from biogas plant D.

The emissions rates were calculated under different conditions because the facilities' properties are not reported publicly, and in situ measurements were not available. Therefore, emissions rates were estimated for a range of source release heights, from the tallest height of the facility where known and 15 m where unknown, and at 10 m, 5 m, 2 m and ground level to take into consideration any pipe, pump or valve leakages at lower levels. Also, each facility's CH_4 loss percentage was calculated taking into account its emissions rate relative to the interpolated CH_4 production rate. A higher uncertainty of emissions rates was observed at plant B than at other plants. This may be attributable to operational problems at the plant or atmospheric variability during the survey of ~ 35 min, or the venting process in the digester, which may emit small or very large amounts of CH_4 (Delre et al., 2017; Reinelt et al., 2015; Yoshida et al., 2014). According to Liebetrau et al. (2017), single large leaks or long-lasting pressure relief may also cause such large ranges of methane emissions. In addition, it was observed that the large uncertainty on the amount of emission rates was mainly due to the different emission heights for plant B.

Overall, the measured CH_4 emission rates varied between 0.1 and 58.7 kg CH_4 h^{-1}, and CH_4 losses ranged between 0.02 and 8.1%, with the average being 3.7%. These results are comparable with those of Scheutz and Fredenslund (2019), who found losses of 0.4 to 14.9% with an average of 4.6% of the CH_4 production gas at 23 biogas plants. The results are also comparable to Flesch et al.'s (2011) results, which estimated an average loss of 3.1% of CH_4 production for a Canadian biodigester. Baldé et al. (2016) also found a wide range of emission rates of 0 to 97 kg CH_4 h^{-1}. It is important to reiterate that we measured the total CH_4 emissions from the plants on the public road with the limited information about the processes. Therefore, it is hard to pinpoint the sources of the emissions in the biogas plants.

The biogas plants selected for this study were of two types based on feedstock material: farm and waste (see Table 2). The farm types include slurry, manure, and purpose-grown crops such as maize, silage and grain. The waste types are mainly food waste. In general, CH_4 losses from farm biogas plants E, F, G, H, I and J were higher (0.5 to 21.9 kg CH_4 h^{-1} and 0.3 to 8.1% relative to calculated production rates) than from food waste biogas plants A, C and D (0.1 to 12.6 kg CH_4 h^{-1} and 0.02 to 6.8% relative to calculated production rate), yet the latter all had a higher capacity than the farm plants. At four of the ten biogas plants, the calculated CH_4 loss was higher than the average of 3.7% (Table 4). Of these four plants, three were farm biogas plants except plant B which can be considered as an outlier due to its largest amounts of emission rate uncertainty. The farm biogas plants E, G and J that emitted more than 3.7% had the lowest calculated biogas production (Table 4), capacity (KWe) and total feedstock amount (Table 2) excluding Plant H whose emission loss is also lower. Altogether, it seemed that there was a negative correlation between the CH_4 loss and the size of the biogas plant as Scheutz and Fredenslund suggested in 2019. The reason for this outcome cannot be determined owing to limited knowledge of the plants' operating conditions and properties. One reason may be that larger facilities are generally better maintained and that investment in modernization, operations and monitoring plans are higher. In addition, despite the very limited information on the age of the plants, it is known that 90% of plants in the UK were built after 2010, and many are similar in design (Ricardo Energy & Environment, 2017). The UK has no direct regulation to control biogas plants' CH_4 emissions. Environment Agency Standard rules SR2010 No16 (2012) applies to the total volatile organic compounds including methane emissions coming from engine stacks. Therefore, the difference in CH_4 loss from small plants compared with larger plants cannot be explained by regulations (Environmental Agency, 2012).

3.2. Emissions factor estimation

An emissions factor (EF) was calculated for each biogas plant assuming a wet weight basis (Table 4), taking into account its annual feedstock amount for which no drying process information was given in NNFCC (2019). Except for Plant B, the EF ranged from

Table 5
CH_4 production and emissions in the UK from the anaerobic digestion of food waste and from farm biogas plants in 2019 excluding sewage sludge treatment plants and landfill gas.

Biogas plant type	Food waste biogas plant	Farm biogas plants	Total
CH_4 production, kilotonnes (ADBA, 2019)	583.9	560.7	1,444.5
CH_4 emissions, kilotonnes Plant average; EF_{Food} = 2.1% EF_{Farm} = 4.8%	12.3	26.9	39.2
CH_4 emissions, kilotonnes Production weighted average; EF_{Food} = 2.3% EF_{Farm} = 4.5%	13.4	25.2	38.6
CH_4 emissions, kilotonnes EF_{IPCC} = 5%	29.2	25.3	54.5
Treated waste, million tonnes (NNFCC, 2019)	6.5	5.9	12.4
CH_4 emissions, kilotonnes Plant average[a] ; EF_{Food} = 0.003 EF_{Farm} = 0.008	19.5	47.2	66.7
CH_4 emissions, kilotonnes Plant weighted average[a]; EF_{Food} = 0.003 EF_{Farm} = 0.006	19.5	35.4	54.9
CH_4 emissions, kilotonnes (Ballinger and Hogg, 2015)[a] EF_{Food} = 0.002 $EF_{Garden\ waste}$ = 0.001	13	5.9	18.9

[a] Tonnes of pollutant per tonne of waste treated

0.02 to 15.3 kg of CH_4 per tonne of feedstock, with the average being 6 kg CH_4 per tone of feedstock for this study. It was found that the EF for farm biogas plants (average 7.8 kg CH_4 per tone of feedstock) was two times higher than the EF for food waste biogas plants (3.3 kg CH_4 per tone of feedstock). According to IPCC guidelines for Tier 1 (Doorn et al., 2006), the CH_4 EF for anaerobic digestion at biogas facilities is 0 to 8.8 kg CH_4 per tonne of waste treated on a wet weight basis. Based on the survey of ten biogas plants, it is difficult to determine a default EF in view of the broad emission range, but it can be proposed to use different emission factors for different types of biogas plants.

The NAEI (2020) figure for anaerobic digestion was 0.8 kg tonne^{-1} in 2018 in the UK, which is at the lower end of the range of EF values from our results. The CH_4 emissions factor from farm biogas plants (anaerobic digestion) are considered as a separate manure management section in the NAEI and EF is given as a factor of animal numbers which makes it hard to compare our results.

The 2019 IPCC Guidelines and NAEI (2020) classify biogas plants and anaerobic digestion emissions under the waste sector and manure management. Based on IPCC Guidelines, biogas plant emissions are generally between 0 and 10% of the amount of CH_4 produced. A default value of 5% should be used in the absence of sufficient information (Eggleston et al., 2006). Table 5 compares the CH_4 emissions in the UK from food waste and farm biogas plants estimated in this study with IPCC values. Estimated emission factors were obtained from two alternatives such as a plant average (3.7%) and a weighted production average (3.1%). The total UK annual CH_4 emission is estimated as 54.5 kilotonnes using the IPCC default emission factor which is higher than our prediction (~39 kilotonnes, Table 5). The total estimated CH_4 emission is calculated as 44.8 kilotonnes considering the weighted average emission factors for all biogas plants (see Table 4) and 53.4 kilotonnes by using the all plant average emission factor, which reveal slightly different results compared to IPCC.

Estimated emission factors were also calculated by the annual feedstock amount (Table 4). As expected, farm biogas plants have higher emission factor than food waste biogas plants with respect to both plant average and plant weighted average emission factors. Ballinger and Hogg (2015) reported CH_4 emissions factor from the anaerobic digestion process for both food waste and garden waste based on per tonne of waste treated in the UK (Table 5). They assumed that the treatment process itself, the digestate utilisation and combustion of the biogas during energy generation cause direct emission to air from anaerobic digestion systems. Their suggestion is lower than our estimated emission rate. These inconsistent results might be due to the disparity in the emission factors of farm and garden waste biogas plants. As referred to above, the garden waste emission factor might need to be used for specific feedstock materials.

3.3. Projection of fugitive methane emissions from UK biogas plants

As stated in section 1., the British anaerobic digestion market comprises 660 operational biogas plants, with 148 and 338 being waste and farm feedstocks, respectively (ADBA, 2019). The inventories are not all regularly updated for the biogas plants whose numbers have linearly increased since 2009 (ADBA, 2019). In this study, 31 of 148 waste and 25 of 338 farm feed biogas plants were surveyed as mentioned earlier. It is critical to estimate how much total CH_4 emission might come from biogas plants in the UK to underline the importance of fugitive CH_4 emission from biogas plants.

Upscaling of the results of this study to the impact of total fugitive CH_4 emissions of the UK was performed using two hypotheses. In hypothesis A, we assume that the 22 biogas plants, for which we could not detect a measurable emission, had in fact zero emissions. We thus use the ten biogas plants with noticeable emission rates (see Table 4) and 22 biogas plants with zero emission for upscaling of overall emissions, which we consider as the lower bound of CH_4 emissions from UK biogas plants. In hypothesis B, the emission rates of the ten biogas plants are upscaled for the total farm and waste biogas plants, which we consider as the upper bound of the total emission rate. The estimated emission rates in Table 4 were extrapolated to the UK anaerobic digestion market information given in Table 6 using a linear fit for the electrical capacity (KWe), feedstock amount (kilotonne), the number of biogas plants and biogas production amount (Nm^{3}hr^{-1}). The extrapolated inventory can give a guideline for assessing the different level of CH_4 emissions for both farm and waste type of biogas plants, and overall emission range.

In this study, the total CH_4 emission of ten biogas plants was estimated as 159 ± 27 kg CH_4 hr^{-1}, which varied between 1.1 and 1.7 kilotonnes annually. In particular, the overall estimated CH_4 emissions from four food waste biogas plants and six farm biogas plants ranged from 0.4 to 0.9 kilotonnes and 0.7 to 0.8 kilotonnes, respectively. The most recent published NAEI inventory reports that total annual UK CH_4 emission is 2,079 kilotonnes in 2018. In hypothesis A, the reported sum of all 486 biogas plants excluding the sewage sludge and other type biogas plants in the UK can be projected as between 9.3 and 31.3 kilotonnes (see Fig. 2.a) in various scaled-up categories to justify the extrapolation of emission range. Specifically, the overall projected CH_4 emission from 12 food waste biogas plants and 20 farm biogas plants range from 3.7 to 13.6 kilotonnes, and 5.5 to 19.3 kilotonnes, respectively. In hypoth-

Table 6
The current status of biogas industry in the UK (excluding sewage sludge biogas plants) and the 32 biogas plants investigated in this study.

Scale-up categories	Electrical capacity (MWe – e)[a]	Feedstock amount (million tonnes)[a]	Number of plants[a]	Biogas Production Amount (Nm^3hr^{-1})[b]
UK Waste Biogas Plants	235.6	6.5	148	93,176
Waste Biogas Plants (from this study)	17.2	0.4	12	10,729
UK Farm Biogas Plants	193.5	5.9	338	89,482
Farm Biogas Plants (from this study)	8.3	0.3	20	10,567

[a] data obtained from NNFCC, 2019;
[b] data obtained from ADBA, 2019.

esis A, biogas plants may account for 0.4 to 1.5%, with the average being 1% of the total CH_4 emission in the UK. In hypothesis B, the extrapolation of emissions from the ten biogas plants to the total number of biogas plants for the different upscaling parameters gives emissions in the range of 43.6 to 79.1 kilotonnes annually (Fig. 2.b). In hypothesis B, biogas plants may account for 2.1 to 3.8%, with the average being 2.8%, of the total CH_4 emissions in the UK. Note that the figure of 3.8% of the total UK emissions is an upper bound, as it is assumed that all plants resume operations and constantly emit in the same manner as the surveyed plants.

Hence, these hypotheses illustrate that biogas plant CH_4 emissions excluding sewage sludge biogas plants might be as low as 0.4% or as much as 3.8% of the total CH_4 emissions in the UK for 2018. On average, 1.9% of the UK CH_4 emissions can come from the biogas plants. We also highlight that CH_4 emissions from biogas plants may have intermittent emission patterns or highly unpredictable leaks, leading to an underestimate or overestimate of emission rates (Duren et al. 2019). This might be the reason for measuring no emission from 22 of the visited biogas plants (see the section 2.1).

The NAEI (2020) estimated CH_4 emission from anaerobic digestion processes as 7.7 kilotonnes in 2018 under the waste category rather than manure management using IPCC Tier 2 methodology, which is the 0.4% of total UK emissions. As observed in Fig. 2, the estimated lower range of CH_4 emissions from hypothesis A is very close to the NAEI (2020) inventory calculation. But it should be noted that anaerobic digestion from agricultural residuals are considered in the agricultural category. The CH_4 emissions from manure management were reported as 158.7 kilotonne by NAEI (2020) except excreta and waste of horses, goats and deer, but it is not clearly identified how much of these CH_4 emissions are coming from the anaerobic digestion process. Additionally, it has been recommended that landfilling of biodegradable wastes should be banned across the UK by 2025 and more food and garden waste will be diverted to the anaerobic digestion and composting facilities (CCC, 2020), which can cause more CH_4 emissions from anaerobic digestion in the coming years. The Committee on Climate Change's Net Zero report (CCC, 2019) predicts the rise in CH_4 emissions from anaerobic digestion will be to 9.0 kilotonnes by 2050. Thus, regular monitoring of biogas plant emissions is absolutely essential to quantify and reduce methane emissions and achieve the net zero aim by 2050.

3.4. Fugitive emissions and sustainability of biogas plants

Much of the organic waste now utilised in biogas plants, was previously deposited in landfills where it was producing methane that was partially oxidized in a layer of topsoil, partially released to the atmosphere, and partially captured and burnt in gas engines at the landfill site. The extent to which diverting organic waste to biogas plants helps to reduce GHG emissions thus depends on how much methane is released per mass of organic waste in comparison to the methane released by the same amount of organic waste deposited in landfills.

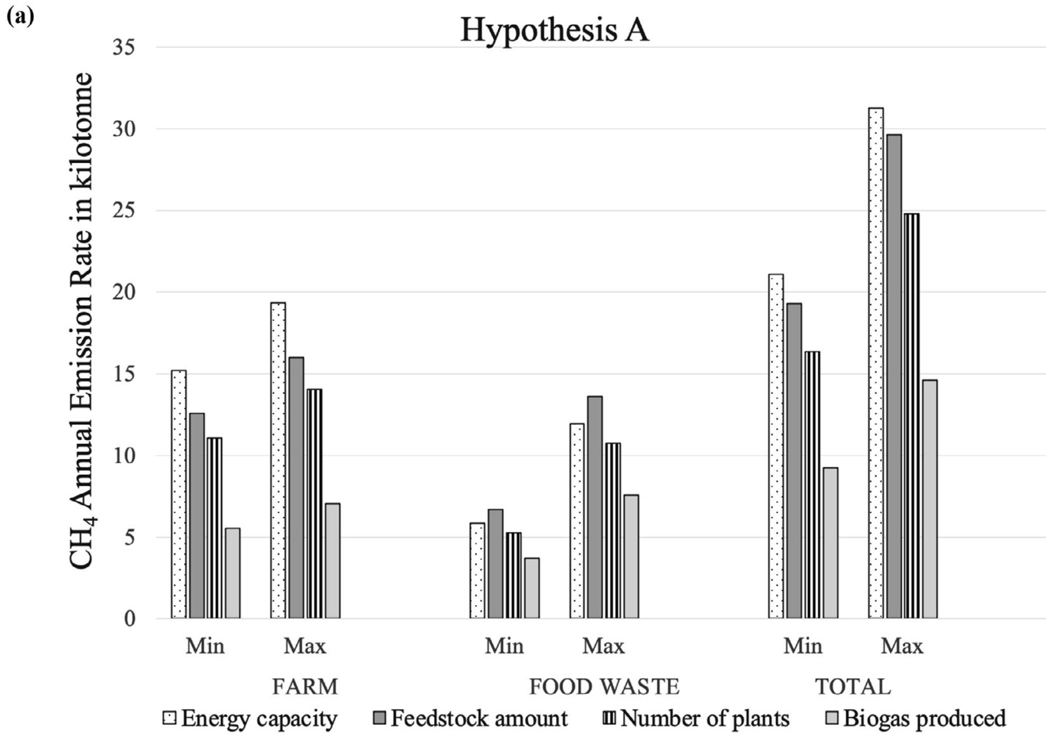
The UK biogas market comprises approximately 10% of the global biogas installation power capacity for 2018 (IEA, 2020). If the upper bound of emission estimate obtained from section 3.3 extends to the global scale, then anaerobic digestion emissions could account for 0.2% of the global anthropogenic CH_4 emissions based on the Saunois et al. (2020) average estimation of anthropogenic emissions for the 2008–2017 decade (359 Tg CH_4 yr^{-1}).

The Renewable Energy Directive (2009/28/EC) requires a threshold of 35% of GHG emission saving for biofuels and bioliquids compared to fossil fuels as a minimum set of sustainability criteria (EC, 2009). The sustainability of biogas plants depends on the land requirement and GHG accounting (OFGEM, 2018). The UK Solid and Gaseous Biomass Carbon Calculator (BCC) tool is used to calculate the carbon intensity and GHG emission saving of solid biomass and biogas used as heat and electricity generation (E4tech, 2014). This tool contains the UK defined default emission factors to calculate GHG emissions from supply chain with hidden assumptions (Adams et al. (2015). They applied this tool to highlight the importance of using actual data when accounting the GHG emissions from biogas facilities. Liebetrau et al. (2010) also point out the uncertainty of methane emission rates owing to difficulties in measurement and changes in operations.

Most biogas operators use default values because there is no clear guidance on how to measure fugitive CH_4 loss precisely (Ricardo Energy & Environment, 2017). Germany, as a good example, utilises country specific EFs to estimate anaerobic digestion emissions. These EFs were specified for each technology considering the changes in atmospheric conditions by measuring the same plant emissions in summer and winter (UNFCC, 2019). Therefore, to eliminate the uncertainty coming from changes in meteorological conditions and operation in the facilities, daily emissions should be monitored at the site. It is also recommended to perform internal site surveys to detect leaks in the facility, and to conduct emission measurement at least once every three years by external consultants as performed in Sweden under The Voluntary Agreement (Holmgren et al., 2015).

4. Conclusions

Methane emissions were measured at ten biogas plants and were found to vary between 0.1 and 58.7 kg hr^{-1}, and the percentage of losses relative to the calculated production rate ranged from 0.02 to 8.1%. It can be generalized that fugitive losses of whole site farm and waste biogas plants are estimated to be a maximum loss of 9%, when considering previous work (Scheutz and Fredenslud, 2019, and Samuelsson et al. 2018) and the new estimations. This study was also suggested that biogas plant methane emissions may account for up to 3.8% of total UK methane emissions. Comparing those estimated losses to the default values of GHG emission calculators, the measured emission rate could be significantly larger than the inventories. The sustainability of biogas plants and the UK Net Zero Commitment may be jeopardised unless robust, consistent emission measurements and legal requirements are put into practice in the near future.



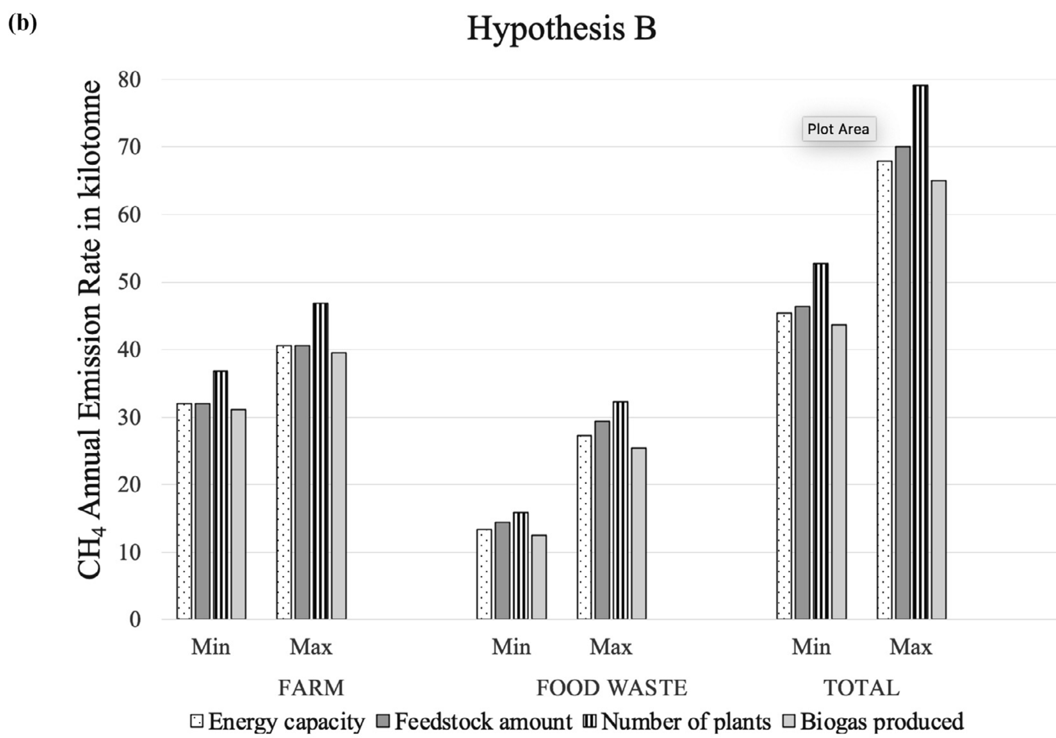


Fig. 2. a) Extrapolation of farm, food waste and total annual fugitive emissions from surveyed biogas plants to all UK biogas plants, based on four parameters: energy capacity, feedstock amount, number of biogas plant and produced biogas amount. Total mean emission rates of 10 biogas plants and zero emission rates of 22 biogas plants were scaled up to observe the possible emission range in the UK. The standard error was taken into consideration to estimate minimum and maximum values of emission. The maximum and minimum extrapolated emissions were obtained from the energy capacity and amount of biogas production, respectively. (**b**) Emission estimation based on hypothesis B. The maximum and minimum emissions were based on the number of biogas plants and the amount of biogas production, respectively.

The measured emissions were associated with high and unknown uncertainties due to not only methodological and meteorological conditions but also temporal fluctuations in emissions from various sources. It is recommended to use 2D-3D anemometers at fixed sites in the plumes for future studies to minimize the uncertainties.

There are no publicly available data on daily or seasonal variation of CH_4 emissions from biogas plants in the UK. This study demonstrated that biogas plants can emit a considerable amount of CH_4. In the next decade, CH_4 emissions from the biogas plants are expected to increase as more waste is diverted from landfills. Therefore, we strongly suggest that biogas plant emissions should be monitored on a daily basis to capture the emission and dispersion pattern due to site activities or meteorological variations on diurnal, weekly and seasonal basis, and emission reduction should be achieved through better regulation.

The mobile technique has enabled quantification of emissions from a selection of UK biogas plants. Repeated surveys with site access and good cooperation with biogas plant operators are recommended for more detailed future studies.

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.

Acknowledgements

This study is funded by MEthane goes Mobile: MEasurement and MOdeling (MEMO^2) project from the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 722479. We would like to thank the U.K. Natural Environment Research Council for current grant NE/P019641/1, New methodologies for removal of methane, for laboratory and field support. We are obliged to Mathias Lanoisellé for help with driving the survey vehicle. We are grateful to Jerry Morris for maintenance of the survey vehicle. Special thanks go to Dr. Peter Nisbet-Jones, Dr. Aalia Al-Shalaan, Julianne Fernandez and Barbara White supporting this study.

Input files necessary to reproduce the model are available from the authors upon request (semra.bakkaloglu.2018@live.rhul.ac.uk)

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2021.01.011.

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