





Quantifying methane emissions from anaerobic digesters

J. Tauber , V. Parravicini , K. Svardal  and J. Krampe 

ABSTRACT

In this research, sources of methane emissions of an anaerobic digester (AD) system at a municipal wastewater treatment plant (WWTP) with 260,000 population equivalent (PE) capacity were detected by a non-dispersive infrared (NDIR) camera. The located emissions were evaluated qualitatively and were documented with photographs and video films. Subsequently, the emission sources were quantified individually using different methods like the Flux-Chamber method and sampling from the digester's circulation pipe. The dissolved methane in the sludge digester was measured via gas chromatography–mass spectrometry (GC-MS) and 6.8% oversaturation compared to the equilibrium after Henry's law was found. Additionally, the residual gas potential of the digestate was measured using batch tests with 10 days' additional stabilisation time. The PE-specific residual gas production of the full-scale AD was calculated to 12.4 g CH₄/(PE · y). An extended chemical oxygen demand (COD) balance including methane emissions for the whole digester system was calculated. Also the measured methane loads were calculated and summed up. The total methane loss of the AD was calculated at 24.6 g CH₄/(PE · y), which corresponds to 0.4% of the produced biogas (4,913 g CH₄/(PE · y)). PE-specific methane emission factors are presented for each investigated (point) source like the sludge outlet at the digester's head, a leaking manhole sealing and cracks in the concrete structure.

Key words | anaerobic sludge stabilisation, greenhouse gases, methane

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INTRODUCTION

Anaerobic digestion (AD) is a cost-effective technique for sewage sludge stabilisation. Hereby, the organic matter of sewage sludge is converted into biogas, which is often used for heat and electric production. This conversion produces emissions of climate-relevant gases, mainly methane (CH₄), being the produced carbon dioxide (CO₂), mostly biogenic in origin. Unfortunately, part of the generated biogas is lost during the process due to leakages, entrained gas bubbles and residual gas potential in the digested sludge (Woess-Gallash *et al.* 2010; Daelman *et al.* 2012). Due to its high global warming potential, based on 100 years' timeframe (GWP₁₀₀) of 28 kg CO₂ equivalents/kg CH₄ according to IPCC (2013), these methane emissions can significantly affect the carbon footprint of the entire

wastewater treatment plant (WWTP) (Parravicini *et al.* 2016). According to this study, up to 26% or 9.5 kg CO₂e/(PE · y) of the carbon footprint of the whole WWTP (36 kg CO₂e/(PE · y)) can be attributed to methane emissions from wastewater treatment and mainly from sludge treatment. In contrast, maximum 17% (6.1 kg CO₂e/(PE · y) of the total carbon footprint of the WWTP could be compensated for by using the produced biogas as an energy source. Gärtner *et al.* (2017) report that 75% (27 kg CO₂e/(PE · y)) of climate-relevant emissions from WWTPs originate from methane emissions from sludge treatment, in which 6% arise through raw sludge and 94% through digested sludge.

There are many references about the methane emission sources of a WWTP in literature. Plant components such as the thickener, buffering tank, prior sludge dewatering, combined heat and power plant (CHP) and flare have been extensively studied and their emission factors are mostly known (Becker *et al.* 2012; Pinnekamp & Genzowsky 2012;

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Yoshida *et al.* 2014; Schaum *et al.* 2015). However, there is a lack of studies on direct methane emissions from anaerobic digestion reactors.

Schaum *et al.* (2016) compared emission data from 11 different authors and plants and summarised the emission factors of the entire wastewater treatment process in a range of 11–390 g CH₄/(PE · y), which corresponds to 0.2–7.9% of the biogas production. Moreover, Schaum *et al.* (2015) calculated methane emissions for the whole AD systems including residual gas production in sludge storage tanks, methane slip of combined heat and power plants, and dissolved methane in the digested sludge to 162 ± 87 g CH₄/(PE · y). For the digester(s) solely, there are only a few data on the gas emissions that are available, and these vary widely. Thus, there is a need to improve the data quality, in order to better assess the impact of these emissions on the carbon footprint of WWTPs and to identify proper emission reduction measures on-site.

MATERIAL AND METHODS

Two digesters at an Austrian municipal WWTP with a design capacity of 260,000 PE were examined. Figure 1(a) shows a scheme of the investigated digester including sludge in- and outlet, one of six installed gas lances for mixing (gas injection), the sludge riser and the position of the sludge shaft, which is covered with a gas-tight membrane and used as a Flux-Chamber as described later on. The balance quality (BQ) was calculated according to Formula (1), presented in Figure 1(b), where $F_{j,out}$ and $F_{i,in}$ are the in- and outflow COD-mass flows.

Methane emissions were measured online for 28 days at two different organic loading rates (OLR) of the AD (1.7 to

3.4 kg COD/(m³ · d)). The average hydraulic retention time (HRT) within the balance time was 42 d for both reactors, the average temperatures were 36.0 °C and 37.3 °C. The AD reactors with a volume of 5,000 m³ each were investigated using a non-dispersive infrared (NDIR) camera (FLIR Gas Find IR-320). With the NDIR-cameras, methane specific, deep cold optic, methane-emitting point sources were identified qualitatively. NDIR video films and photographs taken from the digesters were used also for documentation (Figure 2). Detected emission sources, such as manholes, concrete cracks and the sludge riser's top end at the digesters head were quantified with different methods as described.

With this method, point sources with emission rates below 1 g CH₄/h can be detected easily (Tauber 2018). In suitable conditions, emission rates down to 50 g/y (≈0.006 g/h) can be detected (FLIR 2016). Figure 2 shows an example of NDIR photographs, taken from two identified emission sources, (a) a leaking manhole sealing and (b) the sludge riser's top end at the digester's head, which were quantified later on as described below.

Flux-Chamber method

A reliable method for continuous gas emission measurement at the digester sludge outlet was developed. Based on the Flux-Chamber method according to Reinhart *et al.* (1992), a gas-tight membrane was used to collect the gas emissions from the sludge shaft at the digester's head (Figure 1(a)). The Flux Chamber was spilled with a known flow of scavenging air (120 m³/h) by using a compressor, 3.6 m³/h measuring gas was pumped out, filtered and dried. Methane and carbon dioxide concentrations were measured online by using two infrared photometers (SAXON JUNKALOR NDIR 5000 and 7000). Figure 3 shows the Flux-Chamber

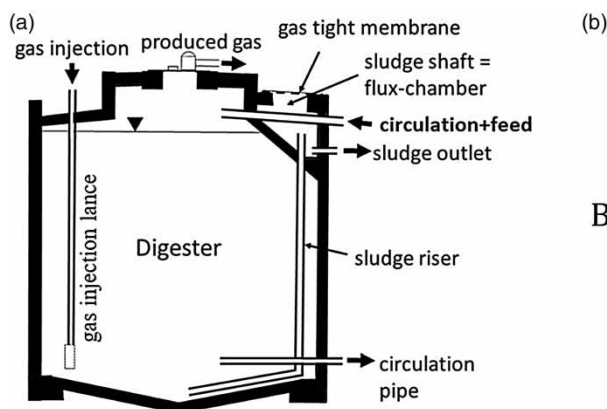


Figure 1 | (a) Scheme of the digester, gas injection, in- and outlet points (b) calculation of balance quality (BQ).

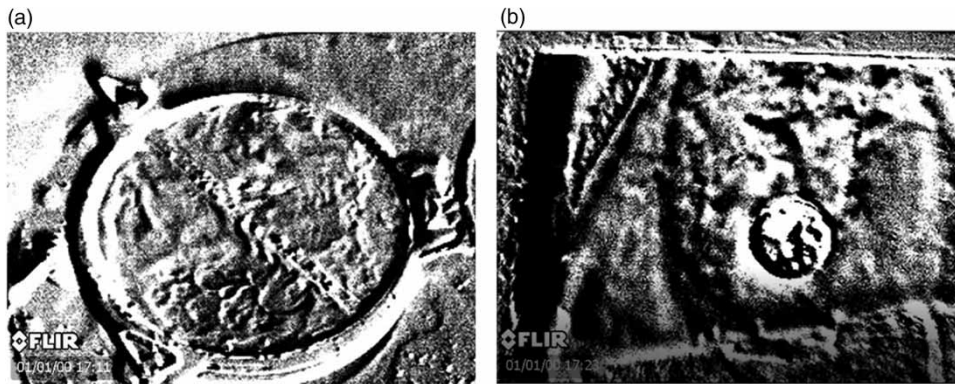


Figure 2 | NDIR images examples of investigated methane point sources (a) leaking manhole sealing and (b) sludge riser's top end at the digester's head.

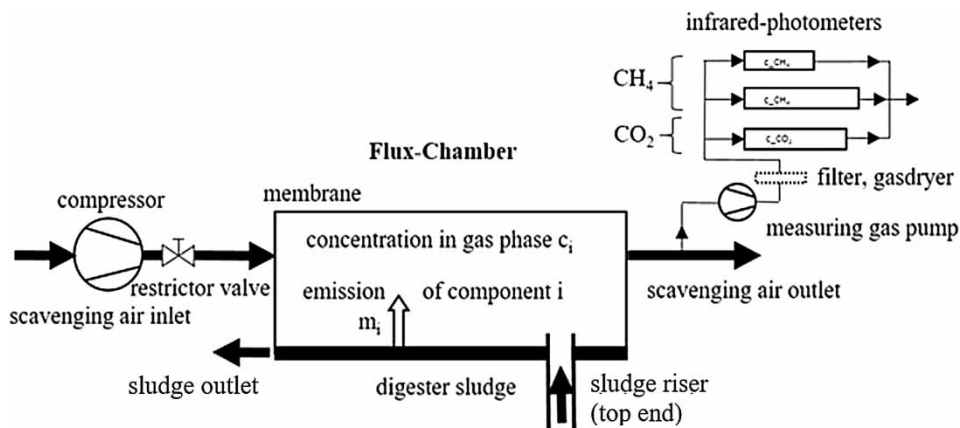


Figure 3 | Flow chart of the applied Flux-Chamber method.

method scheme. This method was applied for 28 days to continuously quantify the methane emissions from the digester at different organic loading rates.

Residual gas and dissolved methane

The residual gas potential in the digested sludge was investigated for an additional stabilisation time of 10 days. Using continuous stirred reactors with 3 L volume, the daily and total chemical oxygen demand (COD) and the specific gas production were measured. The tests were repeated four times. The temperature was kept at 37.0 ± 0.2 °C using a thermostatic bath (Julabo ED v.2) for all batch tests, while the average temperature in the full scale AD was between 36.0 °C and 37.3 °C. The COD and organic dry matter (oDM) in the digested sludge samples were measured according to DIN 38409-43 and DIN ISO 15705 for COD and DIN EN 12879

for oDM, using a minimum sludge sample size of 50 ml. The biodegradation of the COD and oDM were calculated as well as the COD balance. Using COD instead of volatile suspended solids (VSS) as usual for organic matter reduction, allows direct values for the extended COD balance to be obtained as described below. The COD of methane can be calculated using $1 \text{ g COD} = 0.35 \text{ L CH}_4$, while specific VSS values vary between $1 \text{ g VSS} \approx 1.21\text{--}1.61 \text{ g COD}$ (Contreras et al. 2002). Digested sludge from the observed WWTP and from the residual gas laboratory scale tests had a COD to VSS ratio in the range of 1.42–1.43 g COD/g VSS. The gas production was measured daily, using a drum gas meter (Ritter TG 05 PVC).

Additionally, measurements at three different Austrian WWTPs with AD were performed to investigate the oversaturation of methane in the digester sludge. Table 1 gives an overview of the digesters' characteristics, such as plant design capacity, HRT, organic loading rate

Table 1 | Overview of four investigated WWTPs' characteristics and measured CH₄ oversaturation

WWTP [#]	Design capacity [PE]	Digester volume [m ³]	HRT [d]	OLR [kgCOD/(m ³ ·d)]	Co-substrate share [% COD input]	Mixing type	CH ₄ over-saturation [%]
1	260,000	2 × 5,000	42	1.70	45	Gas injection	6.8
2	200,000	2 × 3,000	59	1.22	20	Circulation pump	6.4
3	45,000	1 × 1,700	45	1.12	0	Circulation pump	8.0
4	80,000	2 × 2,000	43	1.35	0	Circulation pump	11.7

(OLR) and mixing type. The emissions from the gas dissolved in the digested sludge were measured with gas chromatography–mass spectrometry (GC-MS). A self-developed vacuum equipment for sampling the digested sludge from the circulation pipe was used according to Tauber (2018).

RESULTS AND DISCUSSION

Entrained gas bubbles

The measurement results show a strongly dynamic and load-dependent behaviour of the methane emission from the sludge outlet on the top of the digester. Figure 4 shows measured methane and carbon dioxide concentrations, specific biogas production and injection rates of the gas recirculation for reactor mixing. Two operation modes with different organic loading rates were considered for the evaluation. Figure 4(a) shows the measured data for an OLR of 1.7 kg COD/(m³ · d). When the gas injection is on, digested sludge is displaced from the digester into the sludge shaft at the digester's head. Dissolved gas is emitted through the pressure drop and gas bubbles are entrained with the sludge. Because of the short retention time in the sludge shaft, it is assumed that the gas bubbles are of greater relevance. The emission rate correlates with the sludge retention time in the sludge shaft and the amount of displaced digested sludge. Moreover, measurement data at an OLR of 3.4 kg COD/(m³ · d) are presented in Figure 4(b). The methane concentrations that occurred varied between 0 and 0.28%_{vol}. In contrast to the lower OLR, the emissions rise till the gas injection is stopped, and gas is still emitted afterwards.

The emission increased from 1.0 to 9.7 g CH₄/(PE · y) by doubling the OLR from 1.7 to 3.4 kg COD/(m³ · d). Further details about the measurements at the sludge outlet using the Flux-Chamber method are presented in Tauber *et al.*

(2017). With an organic load of 16,961 kg COD/d (169,000 PE including co-substrates) an emission due to entrained gas bubbles of 0.5–4.83 m³_n CH₄/d was calculated, where m³_n means volume (m³) at normal (n) conditions, with p_n = 101,325 Pa and T_n = 273.15 K.

Dissolved methane

In comparison to the state of equilibrium according to Henry's Law, a 6.8% oversaturation of dissolved methane was found in the digested sludge (2.1% for CO₂). But only 10% (168 kg CH₄/y) of the dissolved methane (1,889 kg CH₄/y) is released from the AD reactor during operation. Similar values for methane oversaturation in the digested sludge were measured at three other WWTPs with AD in Austria (6.4% – 11.7% CH₄ oversaturation). An overview of the plants' characteristics and the measured dissolved methane values are presented in Table 1.

Leakages

An investigated leaking manhole sealing (Figure 2(a)) contributed 176 kg CH₄/y or 1.1 g CH₄/(PE · y), while a crack in the top cover of the concrete reactor emitted 8 kg CH₄/y or 0.05 g CH₄/(PE · y).

The methane emissions from degassing sludge and other biogas losses due to container leakage amounted in total to 24.55 ± 2 g CH₄/(PE · y) and were low compared to the total biogas production (4,913 g CH₄/(PE · y)). Thus, the methane emissions from the digester correspond to 0.4% of the produced biogas.

Residual gas potential

Figure 5 shows the cumulated residual gas production for 10 days of additional stabilisation time, while the average HRT in the full-scale digesters was 42 d and the average OLR was 1.7 kg COD/(m³ · d). The measured average residual gas

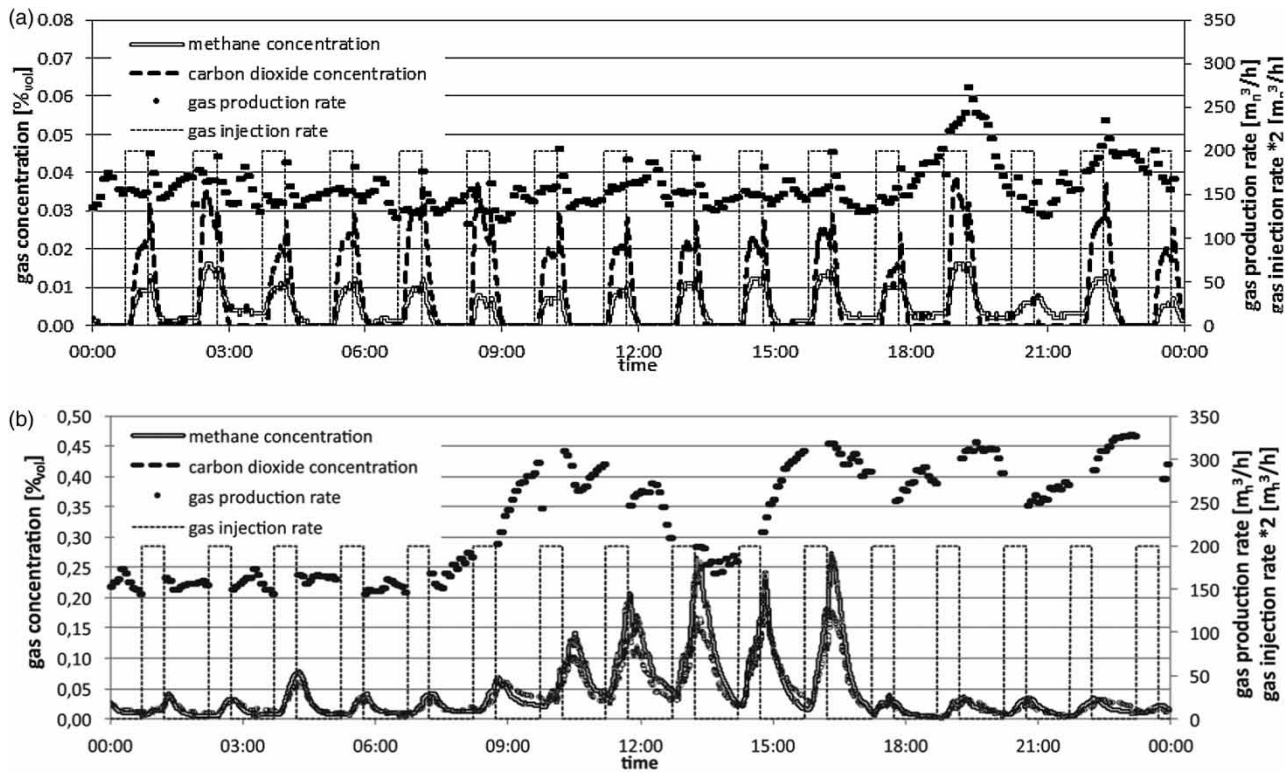


Figure 4 | (a) Methane and carbon dioxide concentration in the scavenging air against gas production rate and gas injection intervals for an exemplary day with 1.7 kg COD/(m³ · d) OLR and (b) an exemplary with 3.4 kg COD/(m³ · d) OLR.

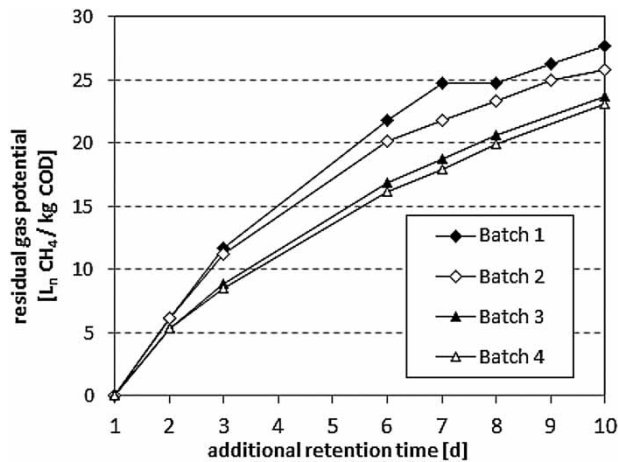


Figure 5 | Cumulated residual gas production at 10 days' additional retention time in the batch tests.

potential in the digested sludge was 25 L_n CH₄/kg COD (input).

Table 2 shows the residual gas production, additional COD and organic dry matter reduction, and the calculated balance quality for the four batch tests performed. COD

Table 2 | Additional COD and oDM reduction, residual gas production and balance quality for four batch tests

Batch test #	COD reduction [%]	oDM reduction [%]	Residual gas production [L _n CH ₄ /kg COD]	Balance quality [%]
1	8.9	8.9	28	91
2	7.4	7.4	26	91
3	8.0	8.0	24	115
4	7.6	9.0	23	120
Average	8.0	8.3	25	

reduction increased by 8%, while the organic dry matter reduction increased by 8.3% on average. The calculated balance quality of the performed batch tests was between 91% and 120%.

The measured residual gas production leads to a potential methane emission of 6.2 m³ CH₄/d or a specific emission of 12.4 g CH₄/(PE · y) for the full-scale plant. The load of CH₄ effectively produced and emitted on site will mainly depend on the temperature and HRT in the storage tank prior to dewatering.

Extended COD balance including methane emissions

The methane losses measured at the plant due to entrained gas bubbles, leakage at the manhole and the cracks in the tank, as well as the methane load dissolved in the digested sludge and the residual gas potential, were measured and calculated as shown above. The emissions were summed up and added to the usual COD balance of the anaerobic digester, which was performed for a balance time of 545 d. The inflow and outflow COD streams of the AD are shown in Figure 6. In addition, important input and output volume flows and the methane emission rates of the quantified point sources are represented in this figure.

The COD input flow consists of 9,261 kg COD mixed sludge (4,831 kg COD/d primary and 4,430 kg COD/d

surplus sludge from the WWTP) and co-substrates (7,700 kg COD/d, mainly glycol from biodiesel production), whereby the COD from the co-substrates makes up to 45% of the total COD load (16,961 kg COD/d). The figure illustrates the individual output streams, where approximately 53% of the COD is converted to biogas (9,100 kg COD/d). This corresponds to 5,000 m³ gas/d or 3,185 m_n³ CH₄/d. From this, 17 m_n³ CH₄/d is released from the digester as methane emissions, as shown in Table 3. This amounts to 0.52% of the daily produced biogas volume and a share of 0.39% of the daily reduced COD load. Respectively, this is 0.52% of the daily produced biogas and 0.51% of the reduced COD at doubled OLR. The difference results from the different methane concentrations in the emitted gas (average 36.0% CH₄) and in the produced gas (average

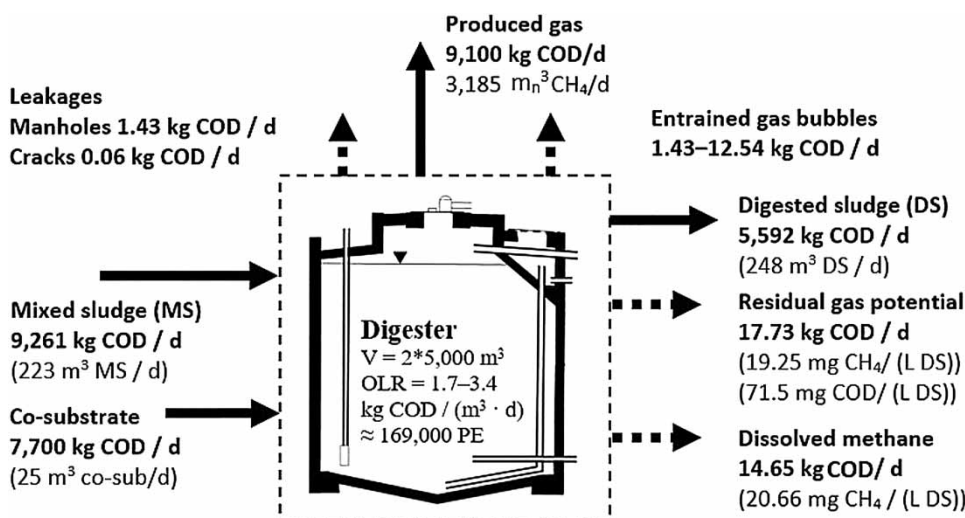


Figure 6 | Extended COD balance of the investigated full-scale anaerobic digester, including methane emissions.

Table 3 | Summary of the measured methane emissions from the anaerobic digester

Methane emission source	[m _n ³ CH ₄ /d]	[kg COD/d]	[g CH ₄ /(PE · y)]
Direct gas emissions through gas bubbles entrained in digested sludge	0.5	1.43	1.0
Direct gas emissions through gas bubbles entrained in digested sludge at double load (not considered in the total)	(4.83) ^a	(12.54) ^a	(9.7) ^a
Gas emissions from gas dissolved in digested sludge	5.12	14.65	10 ± 1
Emissions due to leaks in the digester (sum of manholes and cracks)	0.52	1.49	1.15
Residual gas potential of the digested sludge (10 days' additional stabilisation time)	6.2	17.73	12.4 ± 1
Sum of methane emissions from the digester	12.34	35.3	24.55 ± 2
Share of produced fermentation gas	0.39%	0.39%	
- at double organic load	(0.52%)	(0.51%)	

^aEmission due to gas bubbles at double load is not considered in sum and share.

63.7% CH₄). This is caused by the lower diffusion coefficient of CH₄ compared to CO₂ (Maharajh & Walkley 1973).

CONCLUSIONS

From the present study, it can be concluded that a non-dispersive infrared camera is well suited to detecting point sources of methane from AD reactors with emission rates below 1 g CH₄/h. The Flux-Chamber method showed conclusive results for quantifying the emissions over several weeks. A cross-check with references showed that the measured total methane emission rising directly from the digestion reactor is in the lower range compared to literature values for other plant components like the thickener, buffering tank, prior sludge dewatering, CHP and flare. In particular, methane emissions caused by entrained gas bubbles are very low. Specifically, the total methane loss of approximately 0.4% of the produced biogas (24.6 g CH₄/(PE · y)) is within the range of methane emissions published by Schaum et al. (2016), as well as Schaum et al. (2015), ranging 11–390 g CH₄/(PE · y) for entire wastewater treatment plants.

Considering the age of the investigated AD, which is more than 35 years old, methane emissions were lower than expected. Measured emissions from concrete cracks and permeable sealings are low, but should not be overlooked with respect to methane's high global warming potential, even if a renovation of existing plants is sometimes complex and not economical.

A short retention time of the sludge repressed from the digester led to low methane emissions. Therefore, design changes; that is, covering the sludge outlet, should be considered for future construction of digesters. A strong load-dependent behaviour of the digester's methane emission was found, which should be part of future research.

Except for dissolved methane, the presented measurements were carried out on one single Austrian wastewater treatment plant. However, other systems should be investigated in order to study the influence of different mixing systems (gas injection, circulation pump or screw shovel mixers), the organic loading rate, the overall condition and age of the plant and the design of the digester.

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