

Recommendations for reliable methane emission rate quantification at biogas plants

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IMPRINT

Publisher:

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Funding:

DBFZ Deutsches Biomasseforschungszentrum gemeinnützige GmbH, Leipzig, an enterprise of the German Government with funding from the Federal Ministry of Food and Agriculture pursuant to a resolution by the German Bundestag.

With support from



by decision of the
German Bundestag

General Management:

Prof. Dr. mont. Michael Nelles
(Scientific Managing Director)
Daniel Mayer
(Administrative Managing Director)

DBFZ Report No. 33

Recommendations for reliable methane emission rate
quantification at biogas plants
Leipzig: DBFZ, 2019
ISSN: 2197-4632 (Online)
ISSN: 2190-7943 (Print)
ISBN: 978-3-946629-43-6

Pictures:

If not indicated on the image: DBFZ, Jan Gutzeit, Stefanie Bader (Map)

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Date of Publication:

June, 4 2019

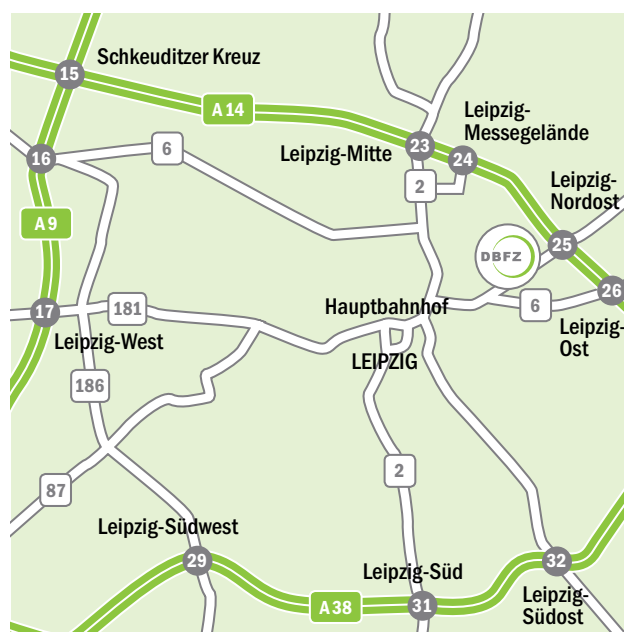
The German National Library has listed these Publications in the German National Bibliography; detailed bibliographic data can be found on the Internet at the address
www.dnb.de

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Funding institution

Project execution organisation

Involved project partner

With support from



by decision of the German Bundestag



Fachagentur Nachwachsende Rohstoffe e.V.



University of Natural Resources and Life Sciences, Vienna



Federal Ministry for Transport, Innovation and Technology



FFG
Österreichische
Forschungsförderungsgesellschaft



bioenergy2020+





Principal or grant authorities
(research funding)

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Date of creation:

June 2019

Funding institution:

Germany:
Federal Ministry of Food and agriculture; support codes: 22403215,
22405015
Austria:
Austrian Research Promotion Agency (FFG): project no.: 852046
Sweden:
Swedish Energy Agency, project no.: 41867-1
Denmark:
Technical University of Denmark

Total amount of pages
+ Appendix

115

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Lists of abbreviations and symbols

Abbreviations	Explanation
BAT	Best Available Techniques
bLs	backward Lagrangian stochastic
BUU	Biogas upgrading unit
CEN	Comité Européen de Normalisation (European Committee for Standardization)
CHP	Combined Heat and Power
CLC	Corine Land Cover
CRF	Controlled Release Facility
DC	Direct Current
DIAL	Differential Absorption Lidar
EGT	Exhaust Gas Treatment
EvEmBi	Evaluation and reduction of methane emissions from different European biogas plant concepts
FID	Flame Ionisation Detector
GHG	Greenhouse gas
GNSS	Global Navigation Satellite System
IDMM	Inverse dispersion modelling method
IR	Infrared
KM	Klug Manier
LASAT	Lagrange Simulation of Aerosol-Transport
lidar	Light detection and ranging
MetHarmo	European harmonisation of methods to quantify methane emissions from biogas plants
NPL	National Physics Laboratory
OGI	Optical Gas Imaging
OP-TDLAS	Open path tunable diode laser absorption spectrometer

OSP	Obukhov stability parameters
PED	Pyroelectric detector
PRV	Pressure relief valve
SCAQMD	South Coast Air Quality Management District
SNR	Signal to noise ratio
TDLAS	Tunable diode laser absorption spectroscopy
TDM	Tracer gas dispersion method
USA	Ultrasonic anemometer
VDI	Verein Deutscher Ingenieure (German Association of Engineers)
VMR	Volume mixing ratio
VOC	Volatile Organic Compound
WMO	World Meteorological Organization

Formula	Explanation	Unit
A_{ch}	Digestate surface enclosed by chamber	m^2
A_{EP}	Cross-sectional area of exhaust pipe	m^2
A_{ODS}	Surface area of the open digestate storage	m^2
A_s	Source area (IDMM)	m^2
$A_\lambda(r)$	Wavelength and range dependent absorption coefficient due to all atmospheric absorption excluding the target gas	m^{-1}
B_λ	Backscatter coefficient for the atmosphere at a wavelength λ from a range r	m^2
$C(r)$	Concentration of the target gas at range r	ppm
C_{CH_4}	(Downwind) concentration of methane	ppm, ppb, vol.%, $kg\ m^{-3}$

$C_{CH_4,BG}$	Measured background concentration of methane	ppm, ppb, $kg\ m^{-3}$
$C_{CH_4,ex}$	Exhaust methane concentration	ppm
$C_{CH_4,RC}$	Methane concentration in reference cell	ppm
C_{CO_2}	Carbon dioxide concentration	vol.%
$C_{O_2,offgas}$	Oxygen content in the off-gas of the CHP	vol.%
C_{target}	Measured downwind concentration of target gas (in this document mostly methane: $C_{target}=C_{CH_4}$)	ppb, ppm
$C_{target,BG}$	Measured background concentration of target gas (in this document mostly methane: $C_{target,BG}=C_{CH_4,BG}$)	ppb, ppm
C_{tracer}	Measured downwind concentration of tracer gas	ppb
$C_{tracer,BG}$	Measured background concentration of tracer gas	ppb
CL	Path-integrated gas concentration	ppm m
CL_{CH_4}	Path integrated methane concentration (at the downwind measurement path)	ppm m
$CL_{CH_4}-CL_{CH_4,BG}$	Difference of downwind concentration and background concentration of methane in units of $mg\ m^{-3}$	$mg\ m^{-3}$
$(C/Q)_{sim}$	Prediction of ratio of concentration at the sensor to the emission rate (IDMM)	$h\ m^{-1}, s\ m^{-1}$
D_λ	Range independent constant at wavelength λ	dimensionless
E_λ	Transmitted energy	W
h_A	Anemometer height	m
h_C	Height of laser path	m
N	Number of pulse pairs averaged (DIAL)	dimensionless
L	Obukhov length	m

L_{OP}	Open-path measuring distance	m
$L_{OP,BG}$	Open-path measuring distance for background concentration	m
$L_{OP,DW}$	Open-path measuring distance for downwind concentration	m
L_{RC}	Length of reference cell	m
M_{target}	Molar mass of target gas (in this document mostly methane)	kg mol ⁻¹
M_{tracer}	Molar mass of tracer gas	kg mol ⁻¹
N_{sim}	Total number of gas particles released at the measurement site in model simulation (IDMM)	dimensionless
P_{sim}	Number of points along measurement path in model simulation (IDMM)	dimensionless
$P_{\lambda}(r)$	Atmospheric return signal at wavelength λ (DIAL)	W
p	Atmospheric pressure	hPa
p_{H_2O}	Partial water vapour pressure	kPa
$p_{Off-gas}$	Static pressure from the off-gas volume flow	kPa
Q_{CH_4}	Methane emission rate	kg h ⁻¹ , mg h ⁻¹
$Q_{CH_4,spec}$	Surface specific methane emission rate	kg m ⁻² h ⁻¹ , mg m ⁻² h ⁻¹
Q_{target}	Target gas (in this document mostly methane $Q_{target}=Q_{CH_4}$) emission rate	kg h ⁻¹
Q_{tracer}	Tracer gas release rate	kg h ⁻¹
R^2	Coefficient of determination	dimensionless
R_m	Gas constant	8.3144598 kg m ² s ⁻² K ⁻¹ mol ⁻¹
R	Range	m
S	Received power after energy normalisation of the on- and off-resonant signals (DIAL)	dimensionless
T	Air Temperature	°C
T_{sonic}	Sonic temperature	°C

t	Time	s, h
t_{theo}	Theoretical time for complete air exchange	h
u^*	Friction velocity	m s^{-1}
u_1, u_2, u_3	Wind velocity components	m s^{-1}
u_3^0	Vertical velocity at touchdown in model simulation (IDMM)	m s^{-1}
\dot{V}	Air volume flow	$\text{m}^3 \text{h}^{-1}$
$\dot{V}_{\text{air,input}}$	Volume flow of the combustion air input to CHP	$\text{m}^3 \text{h}^{-1}$
$\dot{V}_{\text{biomethane,output}}$	Volume flow of upgraded biomethane	$\text{m}^3 \text{h}^{-1}$
V_{ch}	Chamber volume	m^3
$V_{\text{CH}_4,\text{STP}}$	Overall methane volume of all single release events under normal conditions (0 °C, 101325 Pa),	m^3
\dot{V}_{ex}	Exhaust volume flow under operational conditions	$\text{m}^3 \text{h}^{-1}$
$\dot{V}_{\text{ex,STP,dry}}$	Exhaust volume flow under normal conditions (0 °C, 101325 Pa), dry	$\text{m}^3 \text{h}^{-1}$
V_{HS}	Gas volume in head space	m^3
$\dot{V}_{\text{offgas,BUU}}$	Volume flow off-gas of the BUU	$\text{m}^3 \text{h}^{-1}$
$\dot{V}_{\text{offgas,CHP}}$	Volume flow off-gas of the CHP	$\text{m}^3 \text{h}^{-1}$
$\dot{V}_{\text{rawgas,input}}$	Volume flow raw gas fed to BUU	$\text{m}^3 \text{h}^{-1}$
\dot{V}_{STP}	Volume flow of air or carrier gas under normal conditions (0 °C, 101325 Pa)	$\text{m}^3 \text{h}^{-1}$
\bar{v}_{ex}	Average flow velocity at exhaust	m s^{-1}
x	Distance across the gas plume (TDM)	Arbitrary units
Z_0	Surface roughness	m
α_{OFF}	Absorption coefficient of the target gas at wavelength “off” (DIAL)	$(\text{ppm m})^{-1}$
α_{ON}	Absorption coefficient of the target gas at wavelength “on” (DIAL)	$(\text{ppm m})^{-1}$

α_λ	Absorption coefficient of the target gas at wavelength λ	(ppm m) ⁻¹
$\Delta\alpha$	Differential absorption coefficient ($\alpha_{OFF}-\alpha_{ON}$)	(ppm m) ⁻¹
$\sigma u_1, \sigma u_2, \sigma u_3$	Standard deviations of wind velocity components u_1, u_2, u_3	m s ⁻¹
θ	Off-gas temperature	°C
λ	Wavelength	m
Λ	combustion-air ratio	dimensionless
ρ_{CH_4}	Density of methane	mg ml ⁻¹
φ	Wind direction	°

1 Executive summary

Methane, which is a very important greenhouse gas (GHG), is a main component in biogas. The loss of methane within the biogas production and utilisation process should be minimised because of its impact on the environment, safety issues and the associated economic loss for the biogas plant operator.

Particularly with regard to the severe environmental consequences, it is important to investigate and mitigate the methane emissions. For the purpose of methane emission quantification, different methods have been established and are used at biogas plants. Identifying and quantifying emission sources at a biogas plant is a necessary first step towards the mitigation of GHG emissions from the plant. Additionally, by determining the amount of GHG emissions from a biogas plant, it is possible to assess the GHG emissions of the overall process or a certain plant technology, and to classify them in terms of environmental impact.

This document is a product of the project “MetHarmo – European harmonisation of methods to quantify methane emissions from biogas plants”, which aimed to compare different methods and intended to highlight their strengths and limitations. Since there are different methods available to quantify methane emissions, a need to compare the results of measured methane emissions from different measurement teams and/or different biogas plants is identified. The first step includes the improvement of the reproducibility of the results gained with one certain measurement method by giving recommendations in measurement procedures, protocols and documentation. In a second step, the differences by using the different methods will be identified concerning the purpose of the methane emission measurements, their strengths and limitations, the expense to apply the methods, and their measurement accuracy. From that, the document shall support the user to decide on a suitable measurement method. Additionally, the document shall transfer the knowledge about various measurement methods to the European biogas community and may give valuable information to policy makers.

An appropriate classification of the measurement methods mainly used within the biogas sector is the differentiation between on-site and remote sensing approaches. The on-site approach identifies and quantifies single emission sources directly. The remote sensing approach determines overall or component emissions (separately measurable parts) of the biogas plants by measuring from an adequate distance (usually more than 100 m). The on-site approach contains several sub-methods for the identification of unknown point and area sources and the determination of methane emission rates from different single sources, e.g. leaks, exhaust pipes, digestate storages. Remote sensing approaches also include a number of different methods all aiming for quantification of overall or component plant emissions by sampling atmospheric methane concentrations at points upwind and downwind of the emissions source. Within this document, three different remote sensing methods were considered: Differential Absorption Lidar (DIAL), Tracer gas dispersion method (TDM) and Inverse dispersion modelling method (IDMM). A short description of the methods can be found in Section 3.1 and a more detailed description can be found in Appendices A 1 – A 4.

The decision for a certain measurement method depends mainly on the purpose of the measurement, availability of a measurement team, applicability of the method to the specific site and expense and effort of the method. Each method has its individual strengths and limitations. By using the on-site approach, single emission sources can be identified and detected. Depending on the size of the plant or the

accessibility of the sources, not all sources can always be quantified. The detection limit of this method is quite low and the measurement method is not dependent on wind conditions. However, the effort to implement the on-site approach increases with the size of the plant, but depending on the purpose, the effort of the measurement can be adapted. Furthermore, overall methane emission rates obtained by remote sensing methods can be checked for plausibility by identifying the main emitters by using the on-site approach.

Remote sensing methods enable the quantification of overall plant emissions over a longer time period (several hours or days depending on the method) in comparison to on-site measurements, which normally measure the emission from a single point over 10-20 minutes, and the effort to measure the overall plant emissions is independent on plant size. However, remote sensing methods depend on transport processes in the atmosphere and are restricted by certain weather conditions and the surrounding area. For instance, there have to be enough space for the installation of measurement instruments like the truck of the DIAL system (see Figure 2 in Section 3.1). Furthermore, accessible roads for the measurement with the TDM and simple topographical conditions and accessibility of the surrounding area for the IDMM are required. Especially for the IDMM and TDM, the emission plumes of other emission sources in the area should not be mixed with the emission plume of the biogas plant, otherwise it is difficult or not possible to separate both plumes in the measurement.

The DIAL system is a relatively expensive technology for one off-site measurement, but less cost intensive when used for commercial measurement of several sites in the same country or region, e.g. 20-30 biogas sites measured over 1-2 months period. Currently, there are only a few measurement teams and systems available using this technology. The DIAL enables a fast quantification of different target gases with relatively low measurement uncertainties [1, 2]. Besides the determination of overall plant emissions, the DIAL system allows the direct investigation of certain component emissions of the biogas plant. Both DIAL and TDM visualise the emission plume in a two-dimensional way (with DIAL) and one-dimensional way (with TDM), respectively. Thereby, both methods are able to provide a differentiation between the emission from the biogas plant itself and other potential emission sources in the area. The data evaluation using the TDM is quite simple and is performed without dispersion models. In contrast to the other remote sensing methods, IDMM is a quite cost-efficient approach since long-term measurements (over some hours or days) can be performed without much additional expenses. However, the emissions rate calculation relies on dispersion modelling which brings additional uncertainty to this method.

In the document, concrete recommendations for the different methods are described in form of a measurement protocol. A thorough preparation of the measurements, including a description of the purpose of the measurement, the plant conditions and the selected method, and the communication with the operator and the adjacent land owners (only relevant for the remote sensing methods), is a crucial element for executing successful emission measurements at biogas plants. For all methods, a pre-visit of the plant is advised for optimal measurement preparation. For remote sensing methods, a prior inspection of the surrounding area is recommended. There must be enough space for the instrument installation, existence of accessible roads when using TDM or DIAL and simple terrain when using IDMM. Subsequently, the measurement protocols prescribe a number of steps to be followed for performing emission measurements using the respective method. The emission rate is dependent on constructive and operational details. For a meaningful interpretation of the measurement results and to relate the emission rate to a representative operation mode, it is important that the emission-relevant parameters of the biogas plant and the emission-relevant operational events and parameters during the

measurement are enquired and documented. Particularly, it is important to record all special and, in particular, other-than-normal events and the operational modes at the biogas plant during the measurements. This includes, for instance, full load or partial load operation of the gas utilisation, the gas storage filling level, stirring events at open digestate storage tanks, temperature and filling level of the open digestate storage tanks, and flare operation. It is recommended to use a questionnaire template to ask for the important facts concerning the biogas plant. Besides the documentation of the operational states of the biogas plant, it is advised to record the weather situation and the atmospheric conditions. The level of detail depends on the used measurement method. After the measurements, a measurement report should be prepared including all important facts about the biogas plant, the operational details and the details of measurements, surrounding area and weather conditions. A list including important information which has to be included in the measurement report is presented in Appendix A 5. A short summary of the results from the MetHarmo project is presented in Section 10.

In the following, the most important recommendations concerning both on-site and remote sensing approaches are listed.

1. On-site method:

Besides the use of a questionnaire and the pre-visits, the scope of the measurement (e.g., only leakage detection, quantification of the overall emission rate or certain emission sources) has to be clarified in detail. Prior to the determination of the emission rates, a leakage detection should be performed followed by the elaboration of a site map depicting identified leakages. For the leakage detection, the usage of both an optical gas imager (OGI) and a hand-held methane analyser device is recommended. A list of all identified emission sources should be developed and the decision which will be investigated or not have to be documented and measurements executed accordingly. There are several methods available for the quantification of methane emissions from different single sources. For channelled sources (e.g. off-gas from gas utilisation units, encapsulated emission sources connected to a biofilter, encapsulated emission sources with forced ventilation), it is referred to the existing International and European standards [3–6]. For leakages, which are not part of encapsulated emissions sources, the High Flow sampling method (equal to the dynamic chamber method) is recommended. Area sources, which can be divided in open storages and non-gas-tight covered storages, should be quantified with static or dynamic chamber methods and the air injection method, respectively. For emission quantification of pressure relief valves (PRV), permanent online monitoring is strongly recommended, which is described in more detail in the literature [7–10].

2. Remote sensing approach – DIAL system:

It is necessary to clearly define the scope of the measurement activities. It is important to identify all the areas to be measured and the ideal wind direction(s) to measure each area considering available parking locations.

1. Meteorological measurement:

- a) Identify the meteorological mast location in an undisturbed area. If site topography is complex, evaluate the possibility to deploy a second meteorological station.
- b) If possible, deploy a portable wind sensor along the DIAL measuring line-of-sight to assess the local wind at a relatively low elevation.

2. Concentration measurements:

- a) Ensure that the optical source is set-up at the correct wavelength according to the spectral scan test procedure.

- b) For a given wind direction, the areas that can only be measured with that specific wind direction should be prioritised.
- c) The measuring line-of-sight should be as clear as possible from obstacles.
- d) The highest scan elevation angle should be high enough to enclose a plume emitted from the highest point of the area under investigation.
- e) The scan total acquisition time should be less than 20 minutes, if possible.
- f) Carry out a set of at least four DIAL scans for each line-of-sight to minimise the uncertainty from the dominant DIAL uncertainties sources that have a random behaviour.
- g) Measure upwind sources, if present.
- h) Carry out all the necessary quality assurance measurements.

3. Remote sensing approach – TDM:

The tracer gas release must occur in the area(s) of methane emission at the biogas plant. It is highly important to ensure that measurements are unaffected by other sources of methane emission.

1. Methane concentration screening:
 - a) On-site methane concentration screening is used to locate main areas of methane emission, from where tracer gas should be released.
 - b) Off-site methane concentration screening (upwind + downwind) is used to identify nearby sources of methane emission (farms, wastewater treatment plants etc.) and to establish point of measurement of methane and tracer gas while releasing tracer gas.
2. Tracer gas release and traversing the plume:
 - a) The determination of tracer gas release rate must be accurate.
 - b) Concentrations of methane and tracer gas should rise, peak and fall at the approximate same times while traversing the plume. If this correlation is poor, the location of tracer gas release may need adjustment.
 - c) The plume must be traversed at least 10 times to reduce measurement uncertainty – the methane emission rate is calculated as an average value of performed measurements.

4. Remote sensing approach – IDMM:

1. Meteorological measurement:
 - a) Position of ultrasonic anemometer (USA): lee-ward side should be chosen to catch any turbulence induced by the plant.
 - b) Use of three-dimensional USA data (with a sampling rate of 10 Hz) is considered as the most accurate.
2. Concentration measurements for open-path devices:
 - a) Concentration should be measured far enough downwind of the plant to minimise sensitivity to assumptions about the source configuration.
 - b) Best results can be achieved when the whole emission plume of the biogas plant is caught with the chosen measuring paths.
 - c) The threshold distance for (downwind) concentration measurements should be more than 10 times the height of the dominant wind obstacle.

3. The accuracy of emission calculation can be affected by changes of background concentration during the measurement campaign, unless background and downwind concentrations are measured simultaneously.
4. Source configuration for dispersion model: a source area should be outlined in the dispersion model covering the whole plant site.
5. Terrain: when the biogas plant is located on hilly ground, the use of terrain for the modelling domain is recommended.

Additionally, there are still many open issues concerning the validation procedures, such as defining whether certain measurement conditions are suitable or not. Especially for the inverse dispersion modelling methods, it has to be proven if the certain weather situation, e.g. atmospheric stability, is suitable for modelling and how the data should be filtered and validated. Before using data from existing publications, it should be checked if validation concerning the applicability of dispersion modelling was applied.

2 Introduction

Anaerobic digestion plants produce biogas as an energy carrier. Nevertheless, a part of the produced biogas is emitted unintentionally to the environment. As methane is a very potent greenhouse gas (GHG), the environmental effects from the emissions of biogas plants are currently discussed. Relatively few studies have quantified methane loss from biogas production and utilisation, and the results published so far suggest that the quantity of emissions varies highly among biogas plants.

A number of heterogeneous sources of methane emissions can occur at a biogas plant. For instance, there are emissions from the CHP unit and leakages at biogas-bearing plant components such as gasholders, emissions from pressure relief valves (PRV) or from open or non-gastight storage of the digestate.

For the completion of the GHG inventory, and to meet the goal of a maximum global warming of 2 °C determined in the United Nations Framework Convention on Climate Change (UNFCCC), it is of strong interest for all countries to quantify methane emission sources including biogas plants in a reliable way.

Additionally, the acceptance of the biogas technology depends on a reliable determination of GHG balances, which facilitates a classification of the biogas technology with respect to its GHG emissions allowing a comparison of the GHG balance with other energy sources. As biogas production is often subsidised, it is in the interest of the biogas sector to be able to verify and ensure that the gas production is environmentally beneficial.

Not only for the environmental assessment of the biogas plant inventory, but also for single plants and concrete implementation of emission mitigation strategies, reliable emission quantification is important and can lead to an operation with fewer emissions. For the single biogas plant, the avoidance of emissions conduces also to safety and economic aspects.

The identification of low emission biogas plant concepts, the investigation of operational or seasonal effects and also the assessment of emission factors, requires reliable methods and the results of

emission measurements at different biogas plants made by different measurement teams should be possible to rank and compare. Otherwise, the comparison of the emissions from different plants, and also different operational modes is not useful and cannot produce credible and significant results.

So far, there is no European standard describing quantification measurements for the determination of the overall methane emission rate from biogas plants. Emissions can be quantified by several available methods, but it is not clarified whether the results from different measurement teams or obtained with different methods give equivalent results.

This guideline is a first step towards a standard by presenting available measurement methods and giving recommendations and measurement protocols for reliable measurement procedures as well as for an estimation concerning measurement uncertainties. However, it should be noted that it does not cover any valid regulations, which specify employment protection, explosion protection and plant safety. User of this document **must consider** the valid regulations and laws in their respective country concerning these issues when performing emission measurements according to this guideline.

The basis of this guideline are two comparative measurement campaigns, which were accomplished by the scientific partners using different methods for methane emission determination: on-site approaches directly determining the emissions from the single emission sources, and remote sensing approaches quantifying the overall emissions from the biogas plant from a proper distance of the plant, usually a few hundred meters. The detailed results of these measurement campaigns are not described within the present document. For more detailed information about the project's outcome, it is referred to [11, 12].

Upon completion of the measurement campaigns, an extensive process of method documentation and presentation and comparison of the measurement results followed including different evaluation and comparison steps for the individual methods (e.g. a DIAL system was used as reference, controlled methane release experiments, sensitivity analyses). As three different IDMM measurement teams took part in the measurement campaigns, a process of method harmonisation within this measurement approach was applied. The present document was elaborated based on the findings achieved during the international measurement campaigns and the subsequent documentation process within the MetHarmo project.

The present document aims at transferring the knowledge about the use of the methods to the European biogas community, and can lead to a more reliable determination of methane emissions from biogas plants and similar facilities in Europe. By considering the described measurement protocols, the guideline will reach the reproducibility of measurement results by different teams and/or at different biogas plants. The guideline aims to deliver information to measurement providers in order to achieve a common basis for the quantification of methane emissions from biogas plants. This is important for a higher precision of the results, a sound assessment of the biogas plants and a specific look at the different operational modes of the plants and the different technologies used within the biogas sector with respect to the GHG emissions. Furthermore, the document shall support service providers or plant operators to decide on a suitable method for reliable emission measurements. Measurement protocols for the different methods, a description of the individual approaches, the respective uncertainties and method comparison serve as a basis for the decision. The guideline refers in particular to the measurement of methane emissions on biogas plants. The transfer to other facilities or gases cannot be taken for granted.

Additionally, the information is given to support policy makers by informing about methods, their strengths and limitations, as well as recommendations for a reliable determination of methane emission. Based on this, given threshold values can be controlled and met. Consequently, this document is also a first step for a long-range reduction of the GHG emissions.

This document is derived from the experience of the MetHarmo project consortium, especially from the experienced measurement teams participating in the common measurement campaigns. Therefore, the present document is limited to the methods, which were used during the MetHarmo measurement campaigns, including the on-site approach and three different remote sensing methods: DIAL, TDM and IDMM. It gives recommendations for the determination of methane emission rates from biogas plants using the methods as listed above. Additionally, it provides guidance for the decision on a suitable measurement method for a certain purpose (see Section 3). The recommendations or measurement protocols for the different methods are listed in Section 4 (on-site approach), Section 5 (DIAL), Section 6 (TDM) and Section 0 (IDMM). A description of the various approaches can be found in the Appendices A 1 – A 4. Also a list including the important parameters, which should be recorded, is given in Appendix A 5.

Within the MetHarmo project and this document, the strengths and limitations, the effort, and the measurement uncertainties for the different measurement approaches are discussed. Additionally, by considering the recommended measurement protocols, it enables more precise and reliable measurements, and the results from different measurement teams and/or at different biogas plants are better to be interpreted. The comparison of the methods and the conclusions from the document are discussed in Section 0. Additionally, a summary of the results from the MetHarmo measurement campaigns and the comparison of the methods are presented in Section 10. However, there are still open questions and requirements for further research projects, presented in Section 11.

3 General aspects on selecting a measurement method

The decision for a certain measurement method depends primarily on the purpose of the measurement. It should be noted that every measurement method can only provide an estimate of the plant emission as found during the measurement period. The plant emission may vary over the measuring period (minutes, hours or days) due to changes in plant operation and conditions and due to changes in factors influencing the measurement method. For different operational modes or specific events, the emission situation of the plant can vary significantly.

Emission measurements can be executed in the following context: leakage detection, emission quantification for certification (e.g. GHG balances) or inventory purposes, scientific analysis, measurements for legislative purposes (e.g. emission standards of CHP units) and safety related control measurements.

Related to the purpose, the appropriate degree of detail of the results needs to be defined and consequently the methods need to be chosen. Additionally, the financial resources for the measurement as well as the availability of suitable measurement providers offering the favoured measurement method are crucial for the final decision.

One can differentiate between on-site and remote sensing approaches. With the on-site approach, the single emission sources are identified and quantified. In contrast, with the remote sensing approach, the overall plant emission rate or the emission rate of a certain plant area is quantified by measuring from a proper distance (e.g. several 100 m) of the plant.

3.1 Overview of measurement methods

The quantification approaches for determination of the methane emission rates included in the present document are described in detail in the Appendices A 1–A 4. Nevertheless, a very short introduction to the single methods will be listed here.

On-site Methods:

The on-site approach consists of two steps. First, an inventory of all known sources and the identification of so far unknown sources on the plant has to be performed. The identification of the unknown sources can be carried out by means of an OGI camera (see Figure 1) and a hand-held methane detector.



Figure 1: Leakage identification with IR-camera at a biogas plant (© Torsten Reinelt, DBFZ)

Second, the emission rate from every emission source needs to be determined. The specific quantification method depends on the type of the source. For calculation of the methane mass flow from each source, measurements of methane concentration and volume flow are needed. For channelled sources, the flow is measured directly with pitot tubes or anemometers within the pipe. The methane concentration can be measured on-site with mobile instruments (e.g. Flame Ionisation Detection – FID; Fourier Transform Infrared Spectroscopy – FTIR) or samples can be taken for later laboratory analysis. For area sources like open digestate storage tanks, dynamic or static chambers are usually used to determine the emissions from multiple small area parts of the source, and the total emission is then calculated via extrapolation to the whole surface area. For leakages, dynamic chambers are usually applied to quantify the emission rate. The investigation of the time-variant emissions from PRVs, long term studies using, e.g., flow velocity sensors have to be carried out. The emission rate of the plant is determined by adding all single sources together.

DIAL:

DIAL is a laser-based technique which enables range resolved gas concentration measurements along an open path. The measurement configuration is illustrated in Figure 2.

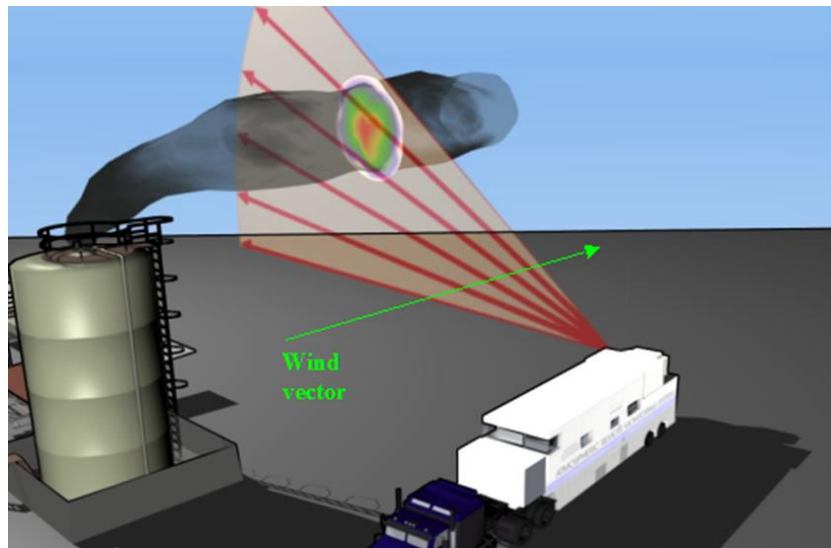


Figure 2: DIAL Measurement Configuration (©NPL).

The pulsed laser operates alternately at two adjacent wavelengths, which are chosen depending on the target gas. One of the wavelengths ('on') is tuned to an absorption line of the target gas, the other wavelength ('off') is chosen to minimise the absorption from the target gas. The range resolved target gas concentration is calculated from the difference of the two 'on' and 'off' backscattered signals. Multiple range-resolved concentration measurements are made along different lines in a vertical plane to determine the target gas concentration on a two-dimensional map over the area of interested. The emission rate from the sources in this area is calculated by combining the target gas concentration map with the measured vertical wind field profile.

TDM:

With TDM, a controlled release of a tracer gas, e.g. acetylene, is emitted on the area of the biogas plant to simulate the methane emissions from the biogas plant. The measurement configuration is illustrated in Figure 3.

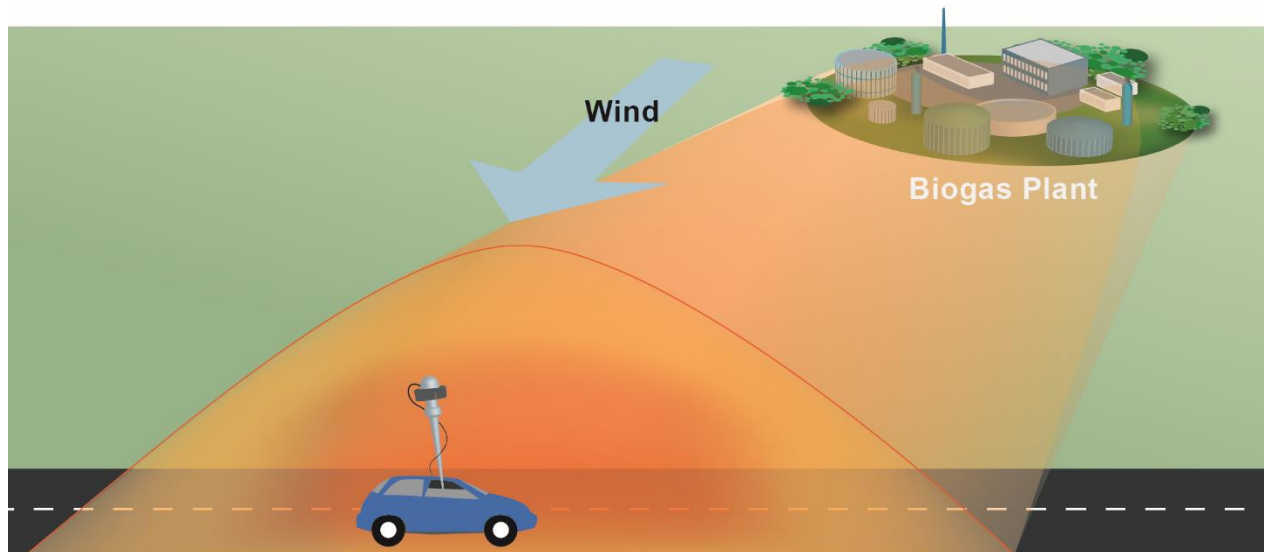


Figure 3: Measuring downwind concentrations of methane and tracer gases using a mobile analytical platform.

During the time of the continuous controlled gas release, methane and tracer gas concentrations are measured downwind of the plant, usually using a mobile analytical platform (e.g. vehicle carrying high resolution analytical instruments for gas detection) moving in the downwind area of the plant. With that, concentration plumes of the target gas and the tracer gas are determined. By cross plume integration, the target to tracer gas ratio is obtained. Based on that, the emission rates are determined.

IDMM:

With IDMM, usually the integrated methane concentration on an open path is measured on the lee and luv side of the biogas plant, e.g. by using an open-path tunable diode laser absorption spectrometer (OP-TDLAS, see Figure 4). The difference of both concentrations is used together with the data of an ultrasonic anemometer (USA), a temperature, and a pressure sensor to calculate the emissions from the plant via an inverse dispersion model.



Figure 4: Measuring methane concentrations close to a biogas plant using OP-TDLAS (© Tina Clauß, DBFZ).

3.2 Strengths and limitations of the methods

The identified strengths and limitations of the different measurement approaches are listed in Table 1.

Table 1: Strengths and constraints of the single methane emissions measurement approaches.

	Strengths	Limitations
on-site approach	<ul style="list-style-type: none"> • Identification of single emission sources • Low detection limit • The most common individual on-site methods are easy to implement except for long term measurement at PRVs or non-accessible sources • Execution of the emission quantification does not depend on wind conditions • Effort can be adjusted to the purpose • Specific components can be monitored by plant operator • Leakage detection included 	<ul style="list-style-type: none"> • Effort is proportional to size • Emission sources have to be identified and need to be accessible for measurement • Variety of methods for different source types is necessary • Leakage detection by OGI is dependent on weather conditions (in particular temperature and wind) and it can potentially fail to detect a leak in unfavourable conditions. • Precipitation and low atmospheric temperatures hinder the encapsulation and consequently the quantification of biogas leakages • Intrusive method, which may in some cases influence the emission condition
DIAL	<ul style="list-style-type: none"> • Possible to measure single emission sources • Non-intrusive method • Effort of determining emission rate of whole biogas plant is independent from plant size • High data capture rate, quickly quantify whole site emission • Minimal restrictions on meteorological conditions • Plume visualization • Proven track record (20+ years) of commercially available measurement service - routinely used for regulatory monitoring 	<ul style="list-style-type: none"> • Relatively expensive technology for one-off measurement • No data in the first 50 m to 100 m distance from DIAL • Currently only very few providers of the service available → limited availability of systems
TDM	<ul style="list-style-type: none"> • Non-intrusive method • Easy screening of plant emissions and local emission sources 	<ul style="list-style-type: none"> • Requires wind speeds of about 3-5 m s⁻¹ • Requires drivable roads around the biogas plants – potentially specific wind directions (depending on road orientation and other local methane sources)

	<ul style="list-style-type: none"> • Tracer gas can be used to assess if methane emission occurs from biogas plant or other possible sources (nearby farms etc.) • Effort is independent on plant size • Use of tracer gas avoids dependency of dispersion models • Simple data analysis • Plumes can be associated to emissions from different biogas plant parts in some cases • Not restricted by weather conditions such as fog, rain and snow • Not dependent on topography • Cost efficient method 	<ul style="list-style-type: none"> • Emissions are often measured over a relatively short time interval of 2-3 hours. Long-term measurements can be carried out using a stationary measuring approach. • Tracer gas placement needs to simulate plant emission to ensure accuracy
IDMM	<ul style="list-style-type: none"> • Relatively low expense and costs • Non-intrusive method • Effort is independent from plant size • Long-term measurements possible • Given a sufficient number of measurement paths, IDMM can be applied to determine emission from several (component) sources simultaneously • Determination of whole plant emissions with a small number of measurement points 	<ul style="list-style-type: none"> • Dependency on weather conditions • Limited application in complex topographical and infrastructural conditions (e.g. forest areas, hills, dense array of buildings) • Inevitable simplification of complex real-world processes due to modelling of dispersion

3.3 Effort of the different methods

The measurement approaches also differ in their effort, i.e. personnel, time and the amount and costs of the necessary equipment. A list of the differences in effort is listed in Table 2.

Table 2: Equipment, personnel and temporal expense of the different methane emissions measurement approaches.

	Necessary Equipment	Personnel	Time required for measurement
on-site approach	<p><u>Leakage detection:</u></p> <ul style="list-style-type: none"> • IR camera and hand-held methane analysers <p><u>Quantification measurement:</u> Basic equipment available from many measurement service providers:</p> <ul style="list-style-type: none"> • Gas analysers (e.g. FID, FTIR) • Devices for boundary conditions of the exhaust gas flow (flow velocity, temperature, pressure) 	<ul style="list-style-type: none"> • For performance of a leakage detection, measurement service providers with qualified personnel are available. • For quantification of channelled sources, measurement service providers with qualified personnel are available. • An additional qualification for the execution of a leakage detection and quantification of non- 	<ul style="list-style-type: none"> • Depending on the investigated emission sources, the plant configuration, and the measurement interval: at least one day for total site emission • In particular a high number of leakages increases the minimum time requirement

	<p><u>Special equipment for area sources:</u></p> <ul style="list-style-type: none"> • E.g., chambers, blowers, pumps etc. <p><u>Special equipment for PRV sources:</u></p> <ul style="list-style-type: none"> • Explosion-proof sensors • Data loggers • Check and approval of the setup by a technical expert 	<p>stationary sources is beneficial.</p> <ul style="list-style-type: none"> • Depending on the investigated emission sources, the plant configuration, and the measurement interval: 1-2 persons necessary. 	
DIAL	<ul style="list-style-type: none"> • DIAL system • 2 to 4 wind sensors at different elevations 	<ul style="list-style-type: none"> • 2 from service provider 	<ul style="list-style-type: none"> • Less than a day for total site emission
TDM	<ul style="list-style-type: none"> • High precision (< 1 ppb) an high sampling rate (>1 Hz) gas analyser for measurement of methane and tracer gases (e.g. cavity ring down spectrometer) • Vehicle for performing measurements and transport of gas bottles etc. • GNSS receiver and antenna • High precision flowmeters/regulators for tracer gas release • Weather station (optional) 	<ul style="list-style-type: none"> • 1 person with adequate training 	<ul style="list-style-type: none"> • 2-3 hours for total site emission and screening of methane concentrations on-site and in surroundings
IDMM	<ul style="list-style-type: none"> • Equipment for methane concentration measurement on an open path (e.g. OP-TDLAS) • Three-dimensional USA • Temperature and pressure sensors • Distance measurement equipment • Global Positioning System (GPS) to determine measurement locations • Dispersion model 	<ul style="list-style-type: none"> • Measurement equipment and personnel knowhow mainly at research institutions; no measurement providers at the moment • At least 2 persons 	<ul style="list-style-type: none"> • Few days including the modelling • Very dependent on weather conditions; at some conditions no measurement possible • Long-term measurements possible (over some hours or days)

4 Guidance and recommendations for the on-site approach

The on-site approach has been used to investigate methane emission rates from agricultural biogas plants including those with upgrading units for the production of biomethane as well as bio-waste treatment plants [7–10, 13–17]. It has also been used for many years in Sweden for measurements in the Voluntary Agreement system, on bio-waste treatment, waste water treatment and upgrading plants [18]. A handbook on how to perform these measurements and calculations has been published [19].

The on-site approach consists of two basic steps. First, the unknown emission sources of the plant need to be identified by means of a leakage survey to add those to the already known emission source inventory. Thereby, the following kinds of emission sources have to be considered:

1. Unknown point sources (leakages at biogas-bearing plant components)
2. Channelled sources
 - a. Off-gas (methane slip) from the CHP unit or its downstream exhaust gas treatment (EGT)
 - b. Off-gas (methane slip) from the biogas upgrading unit or its downstream EGT
 - c. Encapsulated emission sources (e.g. bio-waste pre-treatment halls) collected and transported (= connected) to a biofilter
 - d. Encapsulated emission sources with forced ventilation (e.g. CHP-container)
 - e. Methane diffusion from air-inflated double layer membrane domes (measured at the exhaust of the air cushion)
3. Area sources
 - a. Receiver-, mixing- or hydrolysis tanks
 - b. Open digestate storage tanks
 - c. Non-gastight covered digestate storage tanks
 - d. Aerobic post-composting of digestate
 - e. Biofilter
4. PRVs from the gasholders

After identification, the methane emission rate from each individual source is quantified using different measuring methods and analytical instrumentation. The quantification methods are described in detail in Appendix A 1.2. Finally all sources are added to give the overall emission rate.

4.1 Planning of the measurements and analysis of the measurement site

For the implementation of the on-site approach at a biogas plant, the following steps have to be considered before the emission measurement campaign starts:

1. The plant type (agricultural, biowaste treatment, wastewater) has to be evaluated and the plant specific particularities have to be investigated by means of a questionnaire. A recommended one with the focus on emission related questions is included in the Appendix. This provides the opportunity to evaluate the plant configuration and the possible methane emission sources as well as the mode of operation of the biogas plant.
2. The measurement service provider has to define the scope of the on-site approach.

- a. The sole performance of a leakage detection and tightness testing of membranes can be part of safety regulations. National regulations for the frequency of checks and required measurement equipment have to be considered.
 - b. If the determination of the (overall) methane emission rate is required, a source inventory, leakage detection and quantification measurements have to be carried out.
 - c. For specific investigations of a certain emission source (e.g. long term monitoring of open digestate storages or PRVs), an individualised measurement program has to be planned. Several planning details listed in Appendix A 1.1.1 concretise the planning of a measurement program.
3. Before the campaign starts, a pre-visit of the biogas plant in collaboration with the plant operator is recommended to identify and evaluate all known emission sources. Thus, the measurement institution knows which on-site measurement methods have to be applied and details as accessibility can be evaluated. Subsequently, adjustments concerning the measurement equipment and the plant components can be planned and implemented on-site. Furthermore, safety aspects for the emission measurements can be considered. A detailed description is included in Appendix A 1.1.2.

4.2 Execution of the on-site approach

For the performance of the on-site approach, the use of a gas detector as personnel protective equipment is required. In case of modifications at plant components like the refitting of PRVs with measurement equipment (see Section 4.2.6 and A 1.2.5), the use of respiratory protection is additionally required.

4.2.1 Leakage detection

For the implementation of the leakage detection, the combined use of an OGI method (infrared –IR– camera) and a hand-held methane analyser is recommended:

1. There are different OGI camera setups available. The main differences between the systems are the different real-time image presentation (grey coloured vs. false colour rendering) and the size (hand-held vs. stationary system with tripod). The systems are similarly suited for the leakage detection. Several instructions for use listed in Appendix A 1.2.1 should be considered.
2. The hand-held methane analyser has to pass the requirements from EN 15446 [20] or from the German Standard DVGW G 465-4 [21].

A precise leakage detection requires a complete documentation, including:

1. A documentation of the atmospheric conditions (daily average of atmospheric pressure, air temperature, wind speed, wind direction). For the documentation, data of the next meteorological station, a stationary weather station or a hand-held device can be used.
2. A site map with the detected leakages: The site map can be based on sketch or a GIS tool. The plan should include a colour-based differentiation between leakages and known emission sources as well as the OGI camera positions. A numbering, a short description and the measured methane concentration at the emission spot from every leakage should be included in the site map.

3. A list of not investigated biogas-bearing plant components with justification (e.g. area not accessible considering technical measures for safety and health).
4. In case an assessment of the safety-relevance of a leakage has to be performed, the approach of the German working group “QMaB” [22] (Qualitätssicherung Methanemissionsmessung an Biogasanlagen – Quality control for measurement of methane emissions at biogas plants) can be recommended.
5. A description of the operational state of the biogas plant including:
 - a. the average filling level of the investigated gasholders,
 - b. the capacity utilisation of the primary gas utilisation,
 - c. the flare operation.

In general, a leakage detection has to be differentiated to the proof of impermeability to gas, which is regulated by European and German standards for the proof of impermeability to gas of gas installations (EN 14291, [23] and DVGW G 469 A4, [24]). In the scope of this guideline a leak is defined as a release of methane in a process part due to technical or human failure. The process part will not release methane in its normal operation mode. A methane concentration measurement can give an indication of the size and relevance of the leak, but a definition of a limit for the measured concentration to estimate the size of the leak is not recommended. For precise measurement work it is recommended to investigate both the methane concentration at the leakage spot and the emission mass flow by doing measurements.

A complete measurement of all leakages in case of a large plant with a high number of minor sources might be not possible due to the effort of time for encapsulating and quantifying the emission rate (see Sections 4.2.3 and A 1.2.3). In such a case, the leakages with the potentially highest emission rates should be quantified. Consequently, a qualitative estimation of all the leakages should be carried out in order to identify the larger ones and the decision made which should be measured. For such an estimation different aspects need to be considered:

1. First indicator is the methane concentration measured at the leak. However, the dimension and size and the pressure difference of a leakage have large influence on the prospective emission rate as well as weather conditions on the measured concentration. Examples for the estimation serving as detailed information are included in Appendix A 1.2.1.
2. Images and videos from an OGI camera under consideration of the distance can support a qualitative estimation of the emission rate and consequently the relevance of the leakage when comparing sources located in similar places, for example two sources at a membrane dome. The smaller the emission rate of the leakage, the shorter is the maximal distance to detect it. However, OGI is only an optional estimation instrument to the mandatory and most important first step.
3. The accessibility of the leakage should be considered. This includes the effort, which is necessary for the quantification measurement setup (e.g. mobile platform).

The procedure of the leakage detection is based on three main plant components:

1. Detection of leakages occurring on exposed biogas bearing plant components

Firstly, screening measurements from different directions from all exposed biogas bearing plant-components along the biogas production and utilisation chain have to be carried out by means of an OGI camera. Subsequently, detected leakages have to be checked on plausibility with the

hand-held analyser. The measured methane concentration needs to be documented. Furthermore, a detailed investigation of typical leakage spots is strongly recommended including:

- a. the opening for ropes from the submerged agitators and also the PRVs (both components are usually accessible by a platform),
 - b. the connection (membrane fixation) between the membrane dome and the digester wall,
 - c. solid walls and concrete roofs.
2. Analysis of the natural methane diffusion or leakages from the air-inflated double layer membrane domes (if applicable):

In case of a leakage in the gasholder of an air-inflated double membrane dome, it has to be detected directly in the inflation air by means of a methane concentration measurement with a hand-held methane analyser. The concentration value does not allow a conclusion about the occurrence of a leakage. Please see additional information, in Section 4.2.5.

3. Detection of leakages occurring at biogas-bearing plant components (e.g. CHPs, gas pipes, compressors, etc.) placed in buildings with forced ventilation:

For this case, the previous use of a quantification method (see “Encapsulated emission sources with forced ventilation” in Section 4.2.2) is recommended. If this method confirms the occurrence of a leakage inside the building, it can be located manually by means of a detection instrument afterwards. If the quantification does not confirm a leakage, the manual detection can be omitted.

A proposed structure for a measurement report of the leakage detection is included in Appendix A 1.2.1.

4.2.2 Quantification measurements – channelled sources

Channelled sources are “source[s] whose emissions are conducted, for example, through stacks or tubes” [25]. Thereby, the determination of the methane emission rate usually is done by measurement of methane concentration and volume flow in the exhaust pipe. A detailed description of the measurement method and the valid standards are included in Appendix A 1.2.2.

Recommended specifications for different channelled sources occurring at biogas plants are listed below.

Off-gas from the gas utilisation units:

The point of measurement is the exhaust pipe from the gas utilisation unit (CHP or biogas upgrading unit). In case of an installed EGT downstream of the gas utilisation unit, which is able to reduce the methane concentration in the off-gas, the point of measurement is in the exhaust pipe of the EGT. For the execution of the emission measurement, it is recommended to use the existing European standards [3–5]. A detailed description is included in Appendix O. If the design of the exhaust pipe does not allow a flow measurement (e.g. too short inlet zone), the off-gas flow should be calculated from operational data (see Appendix O).

Encapsulated emission sources, air collection system, directed to a biofilter:

For this type of emission source, the same measurement procedure and methods [3–6], like described above for the off-gas from the gas utilisation units, is required. Since a biofilter does not reduce the methane concentration in the treated gas [13], the point of measurement can be located in both locations, prior to or after the biofilter. The measurement should be carried out directly in a pipe with a suitable point of measurement according to EN 15259 [5]. However, if both the raw gas and the off-gas have a suitable point of measurement, the emission measurement should be always carried out in the off-gas. Furthermore, the background methane concentration has to be subtracted from the methane emission concentration.

Encapsulated emission sources with forced ventilation (e.g. gas equipment room):

For this type of emission source, also the use of the existing European standards [3–6] is required. The point of measurement is the air vent where the ventilation air is released to the atmosphere. In dependence of the size of the air vent, measurement traverses have to be considered. Furthermore, the background methane concentration has to be subtracted from the methane emission concentration. The background methane concentration has to be analysed at the inlet air vent of the encapsulated emission source.

4.2.3 Quantification measurements – biogas leakages

In case that a leakage is a part of an encapsulated emission source with forced ventilation (e.g. gas equipment room), an additional determination of the emission rate from this single biogas leakage is not necessary. The emission rate is then part of the overall emission rate from the encapsulated source, which is measured like described above in Subsection “Encapsulated emission sources with forced ventilation” of Section 4.2.2.

Other leakages have to be investigated with the High Flow sampling method (equal to dynamic chamber method; see Appendix A 1.2.3). Following recommendations should be considered:

1. A qualitative estimation of all leakages in order to select the leakages, which should be investigated, is required (see Section 4.2.1). The estimation is not necessary, if all detected leakages are quantified.
2. An adequate air flow rate requires careful adjustment. A “suction” of the methane emission from the source should be avoided in order to reduce an impact on the emission rate of the source. Simultaneously, the air volume flow rate must avoid the formation of an explosive mixture in the off gas of the chamber. As limit a methane concentration of 20 % of the lower explosion limit of methane (about 0.9 vol% CH₄) is recommended.
3. For each leakage a quantification of the emission rate with two different air volume flows is recommended. This procedure supports the verification of the measured emission rate and gives an indication in case of a potential influence of the method to the emission rate.

4.2.4 Quantification measurements – area sources

Open digestate storage

The measurement of the emissions from a digestate storage gives a limited information about the emission situation. The rate is dependent on temperature, filling level and process parameters of the biogas plant. For a long term estimation of the emission situation, these factors need to be considered.

One option for the determination of the methane emission rate of open digestate storages requires the use of a static or a dynamic floating chamber. The chambers determine a surface specific emission rate, which can be extrapolated to the whole surface area of the storage tank. A detailed method description is included in Appendix O.

For the evaluation of the methane emission rate, the surface conditions should be considered. The emission rates from the liquid surface and if applicable the surface crust have to be differed and weighted according to emission behaviour and area in the calculation. Firstly, an optical estimation of the surface distribution should be carried out and documented by photos. The weighting of the emission rate should be determined by an estimation of the area of the digestate surface covered by liquid and crust. The recommended measurement procedure needs the following steps, which are described in detail in Appendix O:

1. A number of single chamber measurements have to be defined depending on the distribution of the digestate surface. Detailed information is given in Appendix O.
2. The chamber design and a sampling strategy have to be determined, which are given in Appendix O.
3. The use of static and dynamic chambers is similarly allowed for the determination of the surface specific emission rate.

Non-gastight covered digestate storage

For this emission source the use of the air injection method [14, 26] is required. With this method, the whole storage container acts as a dynamic chamber and a large fan blows fresh air into the headspace of the storage. A detailed description and recommendations for the method are included in Appendix O.

Aerobic post composting

In case of in house composting processes with air collection systems the emissions can be determined in the air collection system. Open windrow composting requires methods which equal the dynamic chamber method, which is applied for the investigation of open digestate storages. The sole difference is the larger scale of the used chambers, which are also called wind tunnels.

The calculation of mass based emission factors of measurements of open windrow composting requires the consideration of mass losses and retention time within the process. Emission rates will change during the process which needs to be reflected by the measurement set up.

4.2.5 Quantification measurements – methane diffusion and leakage quantification from air-inflated double layer membrane domes

The point of measurement is the outlet where the inflation air of the double layer membrane dome is released to the atmosphere. If the membrane dome has more than one accessible outlet, each one should be investigated.

In the ideal case, the outlet pipe meets the requirements of EN 15259 [5] as point of measurement regarding in- and outlet zone (at least 5 times of the hydraulic diameter as inlet zone and 2 times as outlet zone) for the volume flow measurement. Then the same measurement procedure and methods [3–6], like described above for the off-gas from the gas utilisation units, can be applied. In case the outlet is not an appropriate point of measurement according to EN 15259 [5], an alternative procedure is necessary. In dependence of the design of the outlet, an adopted chamber with a fixed pipe has to be developed and put on the outlet. With the chamber the outlet is converted into a channelled source and can be measured like described above. Anyway, in both cases the emission rate is calculated according to Equation 5.

During the emission measurement, the following basic conditions have to be documented. These factors have a direct influence to the methane diffusion/leakage:

1. Weather conditions (sunshine, clouds, rainfall): During the emission measurement, the atmospheric conditions should be stable. During fast changes of the atmospheric conditions (e.g. changes in sun radiation and temperature), which could potentially influence rapid changes in the filling level of the gasholder, an emission measurement should be avoided.
2. Air temperature, Air pressure
3. Filling level of the gasholder: An ideal filling level for the emission measurement is 60 to 80 %. This secures a defined moulding of the gasholder foil during the measurement.
4. Dimension of the membrane dome (diameter, height of the membrane dome centre) for an approximate calculation of the surface area of the inner membrane.
5. Methane concentration of the biogas in the gasholder

A defined limit value and/or a calculation method for the differentiation between apparent methane diffusion and leakage is currently not available and consequently not stated. Any assessment needs to consider the legally allowed diffusion rate for the membrane, the area of the membrane and the conditions (pressure and temperature at the plant and at the measurement for the membrane characterisation).

4.2.6 Quantification measurements - pressure relief valves

PRV are a very important safety device protecting the gastight covered digesters or gasholders from destructive pressure conditions. Consequently, these devices have a very time-variant and unknown emission characteristic since they only emit if the pressure limit is reached. Furthermore, the emission characteristic strongly depends on factors like weather (ambient temperature), mode of operation (biogas filling level) or the operational state (availability of gas utilisation). For a precise quantification, the use of the flow velocity and/or temperature method [7–10] within the outlet pipe of the PRV is required. Both methods allow a long term monitoring. The measurement of the flow velocity delivers precise quantitative

(emitted methane volume) and qualitative (number and duration of release events) results. The temperature in the PRV outlet gives number and duration of release events. In combination with a flow velocity sensor, the development of conversion factors for the temperature method is possible which can be used to estimate the emission rate. The technical background, safety issues as well as the available measurement methods are described in detail in Appendix A 1.2.5.

For an emission monitoring at PRVs, following issues should be considered, which are also described in detail in Appendix A 1.2.5:

1. The appropriate measurement method (flow velocity or temperature method) considering the purpose of the investigation has to be chosen.
2. The refitting of the PRVs with measurement instrumentation has to take place with personnel protective equipment (gas detector and breathing mask with fresh air circulating).
3. A final check and approval of the measurement setup by a technical expert for safety is recommended.
4. The installation of the measurement instrumentation depends on the design of the PRV to avoid spurious signals.
5. The uncertainties of both methods have to be considered when evaluating and interpreting the monitoring data.

Furthermore, the evaluation of the measurement data with respect to influences on the determined emissions needs a series of seasonal and operational data, including:

1. the ambient temperature as daily average, daily minimum and daily maximum. Data can be taken from a nearby meteorological station. If that is not possible, the ambient temperature should be measured by a temperature data logger, the probe should be not exposed directly to sun.
2. the daily availability of the primary gas utilisation, for instance as percentage of rated power. This is the main parameter, which describes the operational state of the biogas plant.
3. the operations diary where changes in the mode of operation, malfunctions and maintenance measures are documented.

4.3 Quality control for the on-site approach

General:

For a proper quality assurance for the on-site approach following aspects have to be considered:

- All analysis instruments should be calibrated, adjusted and maintained according to the typical instructions for quality management like ISO 9001:2015 [27] and to their specifications and/or the respective standard methods.
- Due to the variability of the emissions, the duration and point of time of the emission measurement has great influence on the determined methane emission rate. Time-variant source types (open digestate storage, PRV) have a difficult to predict behaviour. For a statistically proven emission rate, such sources need to be investigated continuously over a longer period of time, for instance several months or a whole year. Since this is hardly manageable for every plant, estimates of long term behaviour have to support the measurements. The emission measurement

is most often limited to a snap-reading method. In any case, to the determined emission rate as precise as possible, the duration of measurement, point of time and important basic conditions have to be stated (e.g. single measurement vs. permanent monitoring, technical specifications as pressure controlled flare operation).

- Very often the accuracy of the determined overall methane emission rate strongly depends on the quantification of a few main emission sources [15]. Concomitantly, the on-site approach is time consuming, in particular in cases where many minor sources have to be evaluated. Consequently, the precise quantification of the big emitters has priority compared to the determination of each individual small emission source:
 - The main emission sources which usually should be quantified are:
 - the gas utilisation which are usually the off-gas from the CHP or from the biogas upgrading unit (BUU)
 - open or non-gastight covered digestate storages
 - the methane diffusion/leakage from air-inflated double layer membrane domes
 - encapsulated emission sources, air collection system, directed to a biofilter and
 - encapsulated emission sources with forced ventilation
 - important biogas leakages according to a qualitative estimation of their emission potential
 - Emission sources which require special effort and are quantified in specific cases are:
 - Large numbers of small biogas leakages on biogas-bearing plant components like the wires to adjust agitators
 - PRVs due to their special time-variant emission characteristic.

Leakage detection:

A calibration of an OGI camera with an artificial biogas source under laboratory conditions with different distances and wind speeds is required [28]. As test gas a standard biogas mixture (e.g. 60% CH₄ and 40% CO₂) or pure methane can be used. The frequency of calibration is concern of the user, but a minimum of once every three years is recommended.

A calibration of the methane laser and the portable methane analyser with appropriate test gases is required. The frequency of calibration is concern of the user, but a minimum of once every year is recommended.

Gas concentration measurement instruments:

For FID instruments (online analyser) annual tests of response time, repeatability, lack of fit and interference are recommended according to EN ISO 25140 [3]. It is further necessary to annually determine the effect of oxygen, and if needed compensate for this effect when performing measurements in sources with an oxygen content other than 21 vol%. For discontinuous sampling and analysis with gas chromatography the requirements from EN ISO 25139 [4] have to be considered.

Volume flow measurement instruments:

The measurement instruments (e.g. pressure difference sensors, vane anemometers) should be calibrated once a year.

Additional possibilities of quality control for the single on-site methods are an essential part of the recommendations, which are described in the corresponding method sections and appendices. For determination of the measurement uncertainty an error propagation can be recommended.

4.4 Determination of the emission rate from the on-site approach

The overall emission rate is the sum of all individual emission rates which are assumed to be constant during the emission measurement campaign. The calculation of the individual emission rates is explained in the corresponding Subsections of Appendix A 1.2.

4.5 Advantages and limitations of using on-site methods

The on-site approach focuses on the identification and quantification of the individual emission sources of the biogas plant. This procedure contributes to the following main advantages:

1. The contribution of the individual emission sources to the overall emission rate can be determined. This allows an estimation which emission sources are the critical ones.
2. The implementation of emission mitigation measures can be deduced directly. Simultaneously, the possible effects of the measures can be verified.
3. Specific time-variant emission sources like PRVs can be investigated more precisely with an adopted on-site method and an appropriate measurement program [7–10].
4. By separately analysing the single emission sources, the detection limit for the single sources and the overall emission rate is low in comparison to remote sensing methods.
5. The execution of the emission quantification methods does not depend on the wind conditions.
6. The scope and focus of emission measurement with the on-site approach can be adopted individually. For instance, only a leakage detection or the investigation of a specific single emission source is possible.

The disadvantages of the on-site approach include the following.

1. The leakage detection and identification includes the risk that potential emission sources (biogas leakages) are not detected. Consequently, they cannot be quantified and integrated in the summation of the overall emission rate. Furthermore, the leakage detection by OGI is dependent on weather conditions (in particular temperature and wind) and it can potentially fail to detect a leak in unfavourable conditions.
2. Precipitation and low atmospheric temperatures might hinder the execution of emission quantification methods (in particular source encapsulation for the quantification of biogas leakages).
3. Time variant emission sources are difficult to quantify, since they first might not emit during the campaign, second the emission rate might not be representative for a longer period of time.

4. Some on-site methods (High Flow Sampling, chamber methods) require the encapsulation of the emission source. A certain influence on the measured emission rate due to changed temperature and pressure conditions is likely and can only be minimised but not completely avoided.
5. The effort for the implementation of the on-site approach increases with the size of the investigated biogas plant.

5 Guidance and recommendations for DIAL measurements

The control of the emissions of Volatile Organic Compounds (VOCs) to the atmosphere due to losses during storage or transport is covered by both EU legislation and international protocols. CEN/TC 264/WG 38 [29] is developing a new European standard method to determine fugitive and diffuse emissions of VOCs including methane. This standard covers several techniques, including DIAL, and it will specify the characteristics to be determined and performance criteria to be fulfilled by each technique. The following sections are extracted from the DIAL protocol that will be published in the standard.

For practical applications in order to be able to monitor hydrocarbons, specifically methane, the DIAL shall be able to operate in the 3 μm region. In order to characterise emissions from an industrial scale site, such as a biogas plant, the DIAL system shall:

1. Be mobile/transportable and transmit an eye safe beam, according to IEC 60825-1 ed3.0 [30].
2. Be able to scan with an absolute pointing accuracy better than one degree. The relative movement precision in a vertical and horizontal scan shall be better than 0.1° .
3. Be able to produce a narrow optical bandwidth in order to maximise sensitivity and reduce cross interference. The optical bandwidth should be equal or less than the gas absorption line.
4. Be able to ensure pulsed laser wavelength stability to avoid the selected differential absorption to drift. This can be achieved with stability better than the laser optical.
5. Be able to tune and control the on and off wavelengths.
6. Be able to ensure fast switching (>10 Hz) between on and off wavelengths to avoid atmospheric backscatter variation between the on and off return signals.
7. Be able to achieve a spatial resolution of 10-30 meters. This place requirement on the laser pulse length and detector bandwidth.
8. The transmitted beam energy shall be such that the system is able to record a vertical scan in 10 min – 20 min with an acceptable signal to noise ratio.

5.1 Planning of the DIAL measurements

Before Campaign:

It is necessary to clearly define the planned measurement activities. The following pre-campaign activity should be carried out:

1. Campaign logistics: this includes pre-visits of the site (if required) and checklists for ensuring the site operators are aware of the logistics of a DIAL measurement.

2. List and identification of all the areas to be measured as specified by the customer on a site map. Create a checklist of the areas to be measured and note:
 - a) Ideal wind direction/s to measure each area considering available DIAL parking locations and potential upwind sources.
 - b) Identify areas that can only be measured with a specific wind direction
 - c) Any information or requirement the customer may provide such as level of uncertainty or spatial resolution required.
3. Selection of measuring locations: identification of suitable measurement locations from a technical viewpoint (measurement range, upwind sources etc.) and also from a site logistics (parking and potential obstructions etc.) viewpoint.
4. Identification of likely emission sources: this requires information from the site, usually including site plans and information on existing emissions. Potential sources off-site should also be considered.
5. Assessment of wind field: pre-planning includes an assessment of the likely wind conditions, and what existing meteorological data are available.
6. Identification of the meteorological mast location in an undisturbed area. If site topography is complex, evaluate the possibility to deploy a second meteorological station.
7. Check the site magnetic declination angle during the measurement period.

Set-up and initial tasks:

The following should be carried out on site at the start of the campaign:

1. Site visit if a pre-visit was not carried out, check/agree with site personnel all the suitable measurement locations and update the checklist of the areas to be measured accordingly.
2. Set-up at the agreed meteorological mast locations 2 to 4 wind sensors at different elevations and check the functionality.
3. Assure that all the instruments and logger clocks are at the same time reference.
4. Set-up the lasers; for each species to be measured, the on and off DIAL wavelengths shall be selected such that:
 - a) The differential absorption between the wavelengths is of an appropriate level to achieve the required sensitivity. This requires pre-assessment of the likely emission rate and the system performance.
 - b) There are no absorption features from interference species, this requires knowledge of likely emission sources.
 - c) The on and off wavelengths should be as close as possible to minimise possible interferences.
 - d) A protocol shall be in place to ensure that the laser is tuned to the corrected wavelength and that the wavelength doesn't drift.
5. Set-up the detector and perform an acquisition test.
6. Check long-term weather forecast for the measurement period. This in combination with the checklist of the areas to be measured under certain wind conditions allows to determine a measurement plan. The plan should be updated daily with the short-term weather forecast. This in turn enables to:
 - a) Communicate each day to the site the possible DIAL locations and identify in advance possible locations for the portable wind sensor.

- b) Identify in advance the necessity to perform upwind scans for certain site areas.
- c) Quickly react if the actual wind direction during the day is not as forecast.

Daily Tasks:

At the beginning of the measurement period ensure:

- 1. Logging of the met data is started.
- 2. Optical source is set-up at the correct wavelength according to the spectral scan test procedure.

The following should be carried out for each measurement location:

- 1. Determine the measurement plan and move the DIAL to a location with clearest possible field of view.
- 2. If possible, deploy a portable wind sensor along the DIAL measuring line-of-sight to assess the local wind at a relatively low elevation.

While measuring record the following information:

- 1. Measurement locations, meteorological station locations and lines-of-sight on site map.
- 2. File name, time, scanner azimuth and elevation, other specific information of the scan.
- 3. Site information
- 4. The time when periodically measurements to check the detection and acquisition system response are made.
- 5. Update the notes on the checklist of areas to be measured, record:
 - a) If the measurement of a specific area is completed.
 - b) If the measurement should be repeated because it was made in a non-ideal condition or if more measurements are needed to achieve the required uncertainty.
 - c) If the upwind source is measured, is to be measured or is not required.

At the end of the measurement period ensure that:

- 1. All the meteorological data is downloaded.
- 2. All the data is saved and backed up.

5.2 Execution of the DIAL measurements

The selection of the area(s) to be measured depends on several factors driven by wind conditions, available parking locations and line-of-sight:

Step 1: Choice of the measurement location:

For a given wind direction, the area to be measured should be chosen in order of priority such as:

- 1. The areas that can only be measured with that specific wind direction are prioritised.
- 2. At low wind speed, it is preferable to measure, if possible, a relatively open area rather than an area with a potentially more complex wind profile.

3. Considering the range resolution of the DIAL system, depending upon the current meteorological conditions and the system sensitivity, a DIAL location is available to measure the area of interest.
4. The area under investigation is clear from upwind sources. If upwind sources are present, they can either be measured from the same location or from a different DIAL location under similar wind conditions.
5. When moving to a new location the measurement of the upwind contribution to the area that was just measured should be prioritised if such measurement is needed and possible.
6. If the site layout is such that several areas are downwind of each other, the first area to be measured should be (if possible) the one that has no upwind contribution from the rest of the site. The second area to be measured should be the one directly downwind of the first area and so on. In this way, for each area, the upwind contribution would be measured just before the area itself is measured.

Step 2: Choice of the scan line-of-sight:

1. At close distance from the emission source, the wind field could potentially have a complex behaviour due to the presence of buildings. If possible, a portable wind sensor should be deployed along the DIAL measuring line-of-sight.
2. The higher the wind speed the faster the emissions are diluted leading to a reduction in ambient concentrations. Higher concentrations are therefore obtained closer to the emission area.
3. The DIAL method usually cannot measure in the first 50 m to 100 m from the DIAL as consequence of the optical assembly.
4. The measuring line-of-sight is as clear as possible from obstacles.

Step 3: Set-up of the scan:

1. The highest elevation angle is high enough to enclose a plume emitted from the highest point of the area under investigation. Such an elevation can be estimated based on the convective conditions and observing the behaviour of other visible plumes on site.
2. If a plume is observed from the last elevation angle, extra lines should be added in the following scan to capture the whole plume.
3. If a broad slope above background is observed in the last elevation line, this could be due to a diffuse upwind source/background or to a diffuse source from the area under investigation. In the latter case, extra lines should be added in the following scan to capture the whole source. In the former case, there is no need to add extra lines and the slope above background can be used at the analysis stage to estimate the upwind background contribution in the measuring plane when a specific upwind measurement is not available. Which of the two cases is the most likely scenario should be assessed based on site layout and knowledge of potential upwind sources.
4. The selected DIAL scan rate is as fast as possible within the limit imposed by the detection limit (signal to noise) at the range where a plume from the area under investigation is expected and/or observed.
5. The scan total acquisition time is less than 20 minutes, if possible.
6. The step size between two consecutive lines should not be bigger than two degrees.

7. Considering the above restrictions, the number of measuring lines capturing a plume should be maximised.
8. If the signal to noise falls below an acceptable level for the required range during the measurement, either increase the averaging time or select a different measurement area where a shorter range is required.

Step 4: Achievement of measurements quality requirement:

1. Carry out a set of at least four DIAL scans for each line-of-sight to minimise the uncertainty from the dominant DIAL uncertainties sources that have a random behaviour.
2. Repeat one or two extra sets of measurements of the same area along different scan lines or from different locations or on a different day to decrease the uncertainty associated with that area, if specified by the measurement objective, by randomising any potential systematic effect due to a particular measurement configuration.
3. Measure upwind sources, if present.
4. Carry out all the necessary quality assurance measurements.
5. If the detector bandwidth can be varied, select the most appropriate value. The higher the bandwidth (i.e. higher spatial resolution), the greater is the noise. Higher spatial resolution can therefore be selected in favourable atmospheric conditions. In unfavourable conditions, when the DIAL signal to noise ratio is low, a smaller bandwidth may be selected.

5.3 Quality control for DIAL measurements

General:

Quality assurance of the emission measurements is necessary. These procedures require detailed project planning and progress monitoring with the project subject to regular internal reviews and quality audits at measurement institutions.

Care should be taken when measurements are made with wind speeds outside the 1 – 15 m s⁻¹ range and with variable wind direction.

Spectroscopic calibration procedures:

A crucial requirement for high quality DIAL measurements is the accurate knowledge of the actual differential absorption coefficients that are appropriate for a particular measurement. The following calibration procedures should be employed to ensure the spectroscopic quality, and therefore, the accuracy of the differential absorption measurement. The three key elements that need to be verified through these checks are that:

1. A suitable calibration reference cell prepared with a known (concentration*path length) parameter.
2. The laser source is operating with a suitably narrow linewidth to properly resolve the spectral feature of interest.
3. The wavelength of the laser source is fixed and stable on the appropriate on and off resonant wavelengths.

Calibration gases:

A standard gas mixture of the target gas (or an appropriate proxy - e.g., propane or pentane for the total hydrocarbon measurements) should be used to provide the reference for the spectroscopic measurements. These standards should be, where possible, gravimetrically prepared, internationally-traceable reference gas mixtures with absolute volume mixing ratio (VMR) accuracies of 0.5% or better.

Calibration cell:

Transmission is directly measured through a calibration cell filled to atmospheric pressure with the reference gas. This ensures that the pressure broadening, and therefore the linewidth, is the same for the calibration gas as in the ambient environment.

Spectral scans:

A spectral scan of the relevant absorption feature should be carried out on a daily basis. The measured absorption feature is compared to the expected one. This provides confirmation that the cell has been filled correctly and that the laser source linewidth is correct.

Continuous spectral monitoring:

Having established that a suitable reference cell is available and the laser source linewidth is correct, the on- and off-resonant wavelengths are set to their chosen values for the DIAL measurements. A system to check the wavelengths do not drift from the expected value should be in place and logged continuously.

Check of system performance:

In addition to verifying the spectroscopic performance of the laser source, a check that the full system is measuring correctly should be carried out at least once a day using a standard gas.

Meteorological sensors calibration:

The meteorological sensors should be calibrated once a year by the manufacturer. The calibration certificates may provide a calibration factor for the wind speed and wind direction readings. If data loggers are used to store the meteorological data, then analogue sensors, cabling and data loggers should be checked annually using a reference voltage generator. When known voltages are applied directly to the output terminal of the sensors and voltage readings are taken at the data loggers, a calibration factor is then obtained.

Methane Background:

For methane measurements, the concentration results are reported above ambient methane background. Therefore, a variable background may affect, if undetected, the emission rate. For each scan a check should be made during the analysis that the background level derived from the upwind and background measurements is not unrealistic. At each elevation angle during the analysis, a visual check of the integrated concentration (after background subtraction) should be carried out. In particular, at higher elevation angles, the concentration line can be expected to be flat (no emission). If the slope is

positive, it could be because of a real emission or because of an underestimation of the background level. If the slope is negative, it could only be because of an overestimate of the background since a negative emission is not possible. This visual check should therefore detect any background variation greater than 20 - 40 ppb.

5.4 Determination of the emission rate from DIAL measurements

The data acquired has to be analysed to give the range-resolved concentration along each line-of-sight. The data analysis process consists of the following steps:

Step 1: Background subtraction:

Any direct current (DC) background value is subtracted from the signals. This measured background takes account of any DC signal offset, which may be present due to electronic offsets and from incident background radiation. The background level is derived from the average value of the far field of the returned lidar signal where no significant levels of backscattered light is present.

Step 2: Normalisation for variation in transmitted energy:

The two signal returns are normalised using the monitored values of the transmitted energy for the on and off resonant wavelength pulses. The mean transmitted energy is used to normalize the averaged return signal. For this application, this has been shown to be equivalent to normalizing individual shots against transmitted energy and then averaging the normalised values.

Step 3: Calculation of path-integrated concentration:

The path-integrated concentration of the target species, out to the range r , is calculated. The absorption coefficients used in this calculation is derived from high-resolution spectroscopy.

Step 4: Calculation of methane background concentration:

Ambient methane background concentration is determined and subtracted from the path-integrated concentration using upwind scans and/or the concentration measured from the scan's last elevation angle when it does not contain emission from the target source.

Step 5: Derivation of range-resolved concentrations:

The integrated concentration profiles are piecewise differentiated with a selectable range resolution, to give the range-resolved concentration along the line-of-sight.

Step 6: Calculation of emission rates:

Range-resolved concentration measurements along different lines-of-sight are combined to generate a concentration profile. Due to the difference in data density at different ranges and due to the polar scanning format of the data, it should be taken care to reduce artefacts. The emission rate is then determined using the concentration profile together with meteorological data.

The emitted rate is calculated using the following mathematical steps:

1. The product is formed of the gas concentration measured with the DIAL technique at a given point in space and the component of the wind velocity perpendicular to the DIAL measurement plane at the same location, taking into account the wind speed profile as a function of elevation.
2. This product is computed at all points within the measured concentration profile, to form a two-dimensional array of data.
3. This array of emission rates is then integrated over the complete concentration profile to produce a value for the total emission rate.

A logarithmic wind profile can be used to describe the vertical distribution of the wind by using at least two wind speed sensors at different heights. It is advisable to use more wind speed sensors at different heights in order to calculate the variation of wind speed with height as a function of various parameters (such as the roughness of the terrain). At close distance from the emission source, the wind field could potentially have a complex behaviour due to the presence of buildings increasing the uncertainty in the determination of the emission rate. This can be reduced by deploying a portable wind sensor along the DIAL measuring line-of-sight to either use the wind speed for the wind profile determination or to scale the wind profile derived from the fixed mast sensors to match the portable wind speed at the portable elevation. Local terrain effects can be important and introduce systematic bias in emission rate determinations. The ground elevation where the wind measurement system is located needs to be checked to establish if it is similar to the ground level downwind of the source; if not, the ground elevation along the scan line where the plume is detected should be used as the reference point for establishing the wind profile. The calculated wind field is then combined with the measured gas concentration profile using the procedure described above.

5.5 Advantages and limitations of using DIAL

The DIAL technique provides a direct measure of the spatial distribution of the targeted gas species. From that, DIAL measurements can provide real-time qualitative gas concentration level information and near real-time quantitative two-dimensional concentration maps. As the DIAL technique has spatial coverage of hundreds of meters, it is suited to the measurements of fugitive emissions.

In contrast to path integrated remote sensing measurements, the direct measure of the spatial distribution of the targeted gas species enables the discrimination of localised, high concentration plumes from broad, low concentration plumes, and the spatial separation and independent quantification of emissions from different sources within an area. Thus, DIAL measurements not only provide operators and regulators with mass quantification of the emission rates, but also rich spatial information on the plumes and an indication of where the main emission sources are located.

The whole area of a typical biogas plant, as well as targeted smaller areas, could be measured in a day or less. Consequently, even if the DIAL is a complex technique and therefore it can be relatively expensive, the overall quantity and quality of the data collected (e.g. direct emission mass measurements and two-dimensional mapping of the emission plume) is cost effective when compared to other techniques.

Also, since the vertical distribution of an emission plume is referenced to a measured vertical wind profile (to calculate the emission rate) no assumptions about plume dispersion, lofting, or distribution have to

be made. This removes one major source of uncertainty from emission results and means that DIAL can be used to measure emissions when source distribution, topography, or meteorological conditions would lead to unreliable dispersion modelling.

DIAL is a single-ended remote measurement system, with both source and detector at the same location. This removes the need to access emission sources directly with no disruption to normal site operational activities, since measurements can be made more difficult to access areas at different elevations within the plant without requiring any direct access. From that, it is able to measure areas that would be inaccessible to the on-site approach due to location or safety issues, and it does not require assumptions about the specific location of the emission sources.

The DIAL system is typically mounted on a vehicle and it can only be parked at locations on roads of a site that do not affect site operations and safety. An advantage of the DIAL system is that the scanner unit can rotate 360° allowing different line-of-sight measurements to be taken from the same location.

All outdoor optical techniques like the DIAL are affected by different atmospheric conditions although DIAL measurements are not restricted to weather conditions. Fog has an impact on DIAL measurements by reducing the maximum working range but enhancing the signal to noise ratio in the working range. Light rain and snow not only enhance the signal to noise ratio but also the maximum working range. Heavy rain and snow may require stopping the measurements to avoid deterioration of the scanner mirrors. Clear atmospheric conditions with few particles would reduce the signal to noise ratio and the maximum working range. However, the DIAL measurements can be carried out under all meteorological conditions: during the day or night, clear or cloudy, under different atmospheric stability conditions, during precipitation and over a wide range of wind speeds.

Integrated quality assurance procedures ensure that every DIAL measurement is calibrated against a reference gas traceable to international standards. This ensures that no assumptions need to be made about spectral performance or stability, which are crucial to the accuracy of all optical remote sensing techniques.

The wind speed and direction are DIAL uncertainty sources and have an impact on planning DIAL measurements. With light wind speeds and variable wind directions, it is difficult to determine the sources contributing to the emission along a measurement line-of-sight. The higher the wind speed and the faster the gas concentration is diluted, the more an under-estimation of the emission might occur due to the concentration being below the detection limit of the DIAL.

In this situation, the DIAL scanner can be quickly reoriented in order to get closer to the emission area. If the wind direction varies significantly it might become impossible to measure the targeted area from where the DIAL is parked. An advantage of the DIAL compared to other optical techniques is that it can be quickly redeployed to a different location in order to carry out measurements of the targeted area. This capability ensures high data capture rates during measurement periods (six or more hours of valid data per day) and enables the emissions from spatially and temporally varying emission sources to be studied.

One DIAL constraint is that it does not provide data in the first 50 m- 100 m from the DIAL but this is also one of the main advantages of the technique as the DIAL does not have to be located in the emission source since it has to 'stand-off' by this much. This has the advantage that the DIAL usually does not need to be positioned in restricted site areas; therefore, it does not disrupt the routine operations on site.

6 Guidance and recommendations for tracer gas dispersion method (TDM)

The tracer gas dispersion method (TDM) has been used to measure fugitive emissions from area sources such as landfills [31–34], composting plants [35, 36], wastewater treatment plants [37, 38] and biogas plants [15]. It is the method of choice to quantify whole site landfill gas emissions in relation to the Danish biocover initiative, where the results are used to design biocovers to mitigate emissions as well as to document mitigation efficiency [39, 40].

The method relies on the continuous release of a gaseous tracer at a known, controlled release rate combined with cross plume measurements of methane and tracer gases using a mobile analytical platform. The use of a gaseous tracer distinguishes the method from the two other remote sensing methods described in this document. The use of a gaseous tracer removes the dependence on atmospheric dispersion models and measurements of wind speed etc., and the tracer can provide a confirmation that the observed measured plume originates from the biogas plant and not another, nearby source, such as a farm. The method requires accurate control in release rate of the tracer gas, as any inaccuracy will result in a corresponding inaccuracy in the determined methane emission rate. Also, the tracer gas needs to be released at the approximate location from where methane is emitted from the biogas plant. An exact methane source simulation becomes of less importance with increasing measuring distances. Any misalignment between plant methane emissions and tracer gas release will be visible from the real-time plume monitoring and the location of the tracer gas release points can be adjusted in the field.

The use of TDM requires a trained operator capable of adjusting to conditions at each measurement location according to observed nearby methane sources, wind speed and direction, available measurement locations and more. A correct use of the method has been proven to provide determination of release rates with errors generally below 10-15% in controlled release experiments [41, 42].

6.1 Planning of the TDM measurements

Before executing a TDM measurement at a biogas plant, it is recommended to perform a geographical survey of the area using a GIS tool such as Google Maps. The purpose of this analysis is to determine possible downwind locations for cross plume measurements using the mobile analytical platform. Two main criteria should be considered:

1. Existence of drivable roads in distances useful for the TDM measurements (approximately 500 m to 2000 m) – shorter measurement distances may be required if methane emissions are low (for example below $1 \text{ kg CH}_4 \text{ h}^{-1}$)
2. Low risk of interference from other sources of methane emissions (farms, wastewater treatment plants, landfills, etc.)



Figure 5: Geographical analysis of possible measurement locations as part of the planning of measurements. Green lines indicate possible, downwind measurement locations and red lines indicate locations, where interference from nearby, other sources of methane emission is considered a risk.

Several factors will affect the ideal measuring distance between biogas plant and measurement location. Long measuring distances assure mixing of methane and tracer gases. However, long measuring distances also increase the dilution of the gases, and at a certain distance, the uncertainty in the measured concentration will increase. For biogas plants, distances between 500 m and 2000 m are often useful, but the ideal distance may vary according to emission rate, wind speed, dispersion, and the precision of the used analytical instrument.

Figure 5 exemplifies a geographical analysis done prior to a measurement campaign. Here, two nearby farms with visible manure storage tanks were identified as potential interfering sources of methane emissions. From the map, potential roads, where it is possible to measure concentrations downwind unaffected by the other potential methane sources, are noted. From this analysis, usable wind directions were estimated to be from east, northeast, west and southwest. Wind directions from northwest and southeast are to be avoided due to likely interference from the nearby farms.

TDM measurements rely on the transport of methane and tracer gases from the facility to the measurement location by the wind. If there is no wind (a situation, which is very uncommon and normally does not last for many hours) at the time of measurement, the method cannot be used. Increasing wind speeds can increase the dilution of the gases whereby the concentrations of these gases in the plume may become too low. Other factors may affect the dilution of the gases such as atmospheric stability. The optimal wind speed is around 3-5 m s⁻¹. The authors experienced that methane emissions from biogas plants occur from much smaller areas than other sources such as landfills (which can cover several hectares), whereby methane and tracer gas plumes are well defined and concentrations are easily detectable, and the success rate is therefore relatively high even under sub-optimal weather conditions.

Another weather concern is increased vertical dispersion caused by sunlight. Under cloudy (or night-time) conditions, the downwind concentrations are therefore often higher. If the methane emission rate from biogas is low (a few kg CH₄ h⁻¹ or less), the chance of a successful measurement will be thus higher in cloudy weather.

Even though meteorological data such as wind direction and wind speed are not needed for emission rates calculations, it might be advisable to set up a weather station on-site and record meteorological conditions during the measurement campaign. Alternatively meteorological data can be retrieved from a local weather station after the campaign. Meteorological data can support the location of the observed plumes and the interpretation of the measured data, especially if the topography is complex and the average wind speed is below 3 m s⁻¹.

6.2 Execution of the TDM measurements

A measurement campaign (measuring total methane emission from a biogas plant using TDM) consists of three main steps:

Step 1: Area screening of methane concentrations:

Using the mobile analytical platform, drivable roads around the site are travelled while recording atmospheric methane concentrations. Background methane concentrations are taken upwind the biogas plant. The purpose of the area screening is to detect other potential sources of methane emissions from farms, manure storage, etc. in the area. There may be other sources of emission, which are not apparent in the geographical analysis described above, and potential sources identified in the analysis may turn out to be of low or no significance. Another purpose of this step is to locate a road where the downwind plume can be completely traversed (background concentrations of methane are observed on each side of the plume) in a distance of approximately 500-2000 m to the source.

Step 2: Screening of atmospheric methane concentrations at the biogas plant:

Similarly to Step 1, drivable areas are travelled using the mobile analytical platform on the grounds of the biogas plant. The purpose of this step is to determine where methane is emitted from the biogas plant, and thus where tracer gas is to be released. If other information regarding emission locations is available, this may also be taken into consideration when deciding upon tracer release locations. Such information could be previous measurement data, assessments of the plant operator or similar.

Step 3: Tracer gas release and plume traverse measurements:

Based on results from Step 2, one or more tracer gas bottles fitted with high precision valves/flowmeters are placed at the biogas plant, where the on-site screening indicated methane emissions. If the results from Step 2 are unclear (for example if many areas of the plant were inaccessible), it is recommended to place one or two tracer gas bottles centrally on the plant near digesters and open storage tanks or other typical sources of methane emissions from biogas plants. After placement of the tracer gas bottle(s), continuous tracer gas release is initiated. The release rate depends on the weather conditions, where high wind speed and/atmospheric instability and/or large distances require relatively high release rates due to dilution, whereas relatively low release rates are sufficient under optimal weather conditions

and/or measurements relatively close to the biogas plant. Release rates are thus depending on several conditions, which also include the sensitivity of the gas analyser used. A typical release rate for C_2H_2 is 1-2 kg h⁻¹.

Allowing for time for transport of tracer gas and methane from the biogas plant to the measurement location – typically 10 minutes or so, the plume is traversed using the mobile analytical platform at the location determined in Step 1, while measuring methane and tracer gas concentrations. As described in Appendix A 3.1, the plume should be traversed several times (no less than 10), whereby measurement error caused by random sources of uncertainty is reduced. To increase spatial resolution, it is recommended to perform the plume traverses at moderate speed – for example 30 km h⁻¹. Instruments with high detection frequencies may enable faster driving speeds. The recorded methane and tracer gas plumes should be assessed in the field to assure successful measurements. Figure 6 exemplifies a successful plume traverse, where both concentrations of methane and tracer are clearly distinguishable from background levels, where the plume from the biogas plant is traversed completely, and where tracer and methane concentrations are correlated. Correlation of methane and tracer gases is observed if concentrations of both gases rise, peak and return to background levels at the same time (Figure 6). Correlation indicates that the tracer gas release simulates methane emission well from the biogas plant, and that the gases are well mixed.

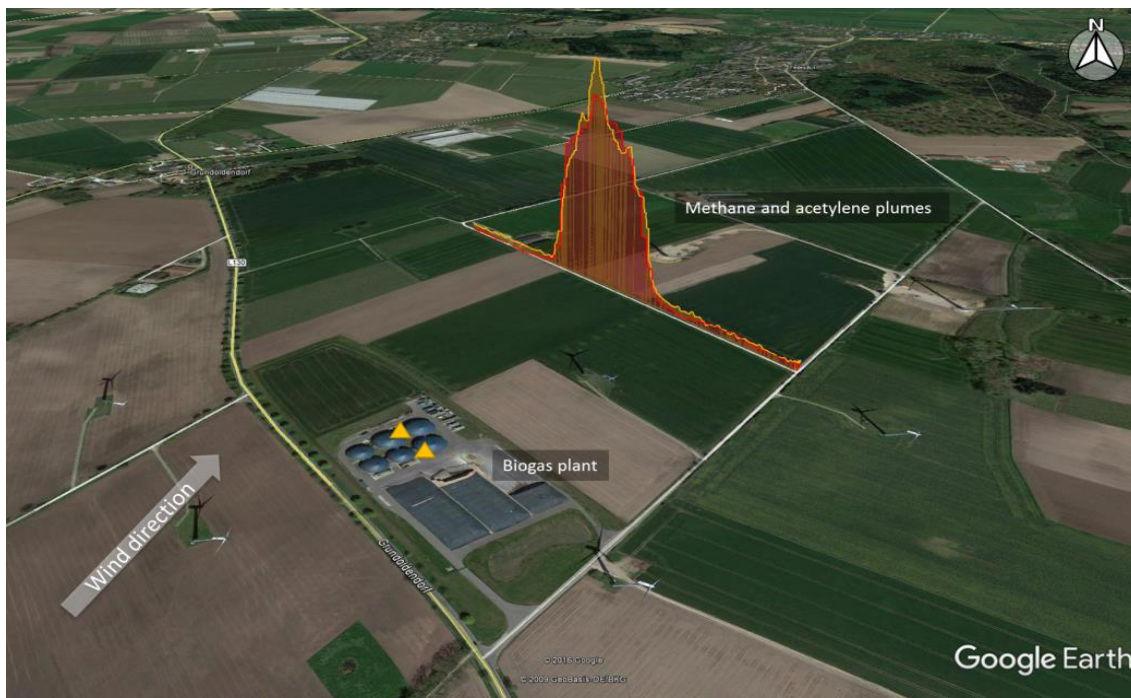


Figure 6: Example of plume traverse, where concentrations of methane and tracer gas are clearly distinguishable from background levels, and where plumes of the two gases correlate well. The height of the red line is proportional to measured methane concentrations above background level, and the height of the yellow line is proportional to measured tracer gas concentrations. The yellow triangles mark placements of tracer gas bottles.

Several corrective measures can be necessary and relevant. If both methane and tracer gas concentrations are hard to detect, it may be necessary to traverse the plume closer to the biogas plant. If only the tracer gas plume is hard to detect, it may be sufficient to increase the release rate of tracer gas.

Poor overlap/correlation of measured methane and tracer concentrations can be caused by “incorrect” placement of tracer gas release bottle(s). This can be corrected by adjusting the placement of tracer gas release using a trial and error approach.

6.3 Quality control for TDM

Ensuring equipment performance:

Accurate measurements using the TDM requires well-functioning, accurate analytical instruments and equipment used to control the release of tracer gas. Analytical error and error in calibration of the tracer gas release equipment will increase uncertainty in the determination of the rate of methane emission. It is therefore recommended that calibration frequencies and procedures specified by the manufacturers of the instruments are followed. In many cases, factory calibrations (instruments are calibrated by the instrument manufacturer) are needed.

It is highly recommended to perform regular controls to ensure the performances of the analytical instruments and the tracer gas release equipment. Analytical accuracy and precision of the gas analyser should be checked using a certified gas standard containing known concentrations of methane and tracer gases. Note some uncertainty (a few %) is to be expected regarding the actual concentrations of gases in gas standards. An assessment of this uncertainty is often provided by suppliers of gas standards.

Similarly, the performance of the tracer gas release regulating equipment can be checked by comparing flow rates obtained by readout of the flowmeter to flow rates obtained by other means such as weighing the gas bottle before and after a timed release of tracer gas.

Influence of other methane emission sources:

Interference from other methane emission sources should be avoided/minimised. As described above, each measurement campaign should include a screening of methane concentrations in the area to identify other sources (farms, wastewater treatment plants etc.), which may cause error. In cases, where an adjacent source of methane emission is suspected to have affected the measurement, and it is practically not possible to avoid separation of the sources of methane emission, this source of uncertainty should be described when reporting the results of the measurements.

In many cases, separation of a nearby methane emission source may be confirmed, if two clearly distinguishable plumes are detected at locations where they are expected from the locations of the biogas plant and other source and the wind direction.

Tracer gas release location(s) and source simulation:

The TDM measurement principle relies on a release of tracer gas, which simulates the emission of the target gas (here methane). It is thus important that the location of the tracer release is occurring in relatively close vicinity to the main sources of methane emission. To establish the location(s) of tracer gas release, it is recommended to perform a screening of methane concentrations at the biogas plant as described above. Also, “misplacement” of the tracer gas release bottles result in misaligned plumes of tracer gas and methane. If this is observed, the tracer gas release location may be changed until a

satisfactory correlation is seen. A quality plume selection criteria for optimal source simulation is the methane to tracer gas concentration scatter plot correlation factor – where a cut-off value of $R^2 > 0.8$ has been suggested (67).

Signal to noise ratio:

The determination of emission rate using TDM includes an assessment of background concentrations of methane and tracer gases. Low signal to noise ratio defined as the ratio of measured plume concentrations of methane and tracer gases above background level to random variations in measured concentrations, may lead to increased measurement uncertainty. The signal to noise ratio can be used as quality indicator to filter out low quality data. A signal to noise ratio larger than approx. 10 is desired, but lower values may be accepted in cases where emissions are low, thus causing only slightly elevated concentrations in the plume. A signal to noise ratio of minimum three is recommended.

6.4 Determination of the emission rate from TDM

As described in Appendix A 3.1, the methane emission rate is determined for each plume traverse, and the emission from the biogas plant is calculated as the average value using Equation 24.

$$Q_{\text{target}} = Q_{\text{tracer}} \times \frac{\int_{\text{Plume start}}^{\text{Plume end}} (C_{\text{target}} - C_{\text{target,BG}}) dt}{\int_{\text{Plume start}}^{\text{Plume end}} (C_{\text{tracer}} - C_{\text{tracer,BG}}) dt} \cdot \frac{M_{\text{target}}}{M_{\text{tracer}}} \tag{Equation 1}$$

Q_{target}	Methane emission rate in kg h ⁻¹
Q_{tracer}	Tracer release rate in kg h ⁻¹
C_{target}	Measured downwind concentration of methane in ppb
C_{tracer}	Measured downwind concentration of tracer in ppb
$C_{\text{target,BG}}$	Measured background concentration of methane in ppb
$C_{\text{tracer,BG}}$	Measured background concentration of tracer in ppb
M_{target}	Molar mass of methane in kg mol ⁻¹
M_{tracer}	Molar mass of tracer gas kg mol ⁻¹
t	Time of measurement in s

Figure 7 shows measured methane and tracer concentrations plotted as function of time during one plume traverse, where the cross plume integration of tracer gas is marked.

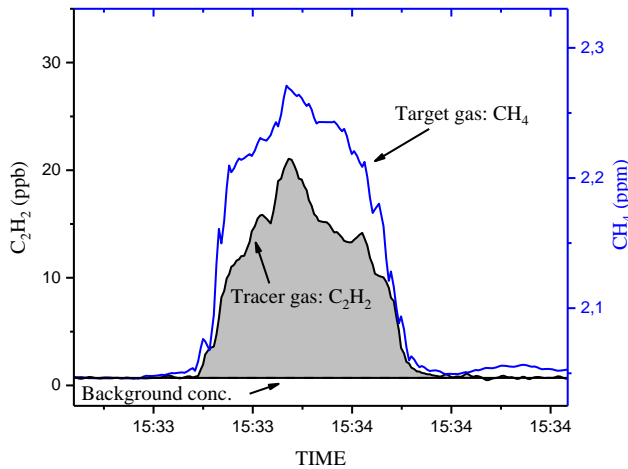


Figure 7: Concentrations of methane and tracer gases measured during a plume traverse plotted as function of time. The grey area indicates cross plume integration of tracer gas concentrations (background concentration subtracted).

The measured total methane emission from the biogas plant is calculated as the average value of the emissions calculated for the individual plume traverses. The variability of the emission rate may be estimated as the standard error of the mean (the sample standard deviation of the calculated emission rates of the individual plume traverses divided by the square root of the number of plume traverses).

Specific quality criteria have yet to be defined to either accept or reject measurements. As described above, correlated target and tracer gas plumes indicate that the tracer gas release simulate the methane emission well. A “correlated target and tracer gas plume” is where concentrations of the gases rise, peak and fall at approx. the same times during the plume traverse (Figure 7). Signal to noise ratio (SNR) is a concern. A SNR larger than approx. 10 is desired, but lower values may be accepted in cases where emissions are low, thus causing only slightly elevated concentrations in the plume.

6.5 Advantages and limitations of using TDM

The TDM was developed to determine total emission rates of methane from landfills due to a specific need to do this with high accuracy. Through this development, the method can be considered as a well-documented approach to determine fugitive emissions from area sources. The use of a gaseous tracer eliminates the reliance of wind measurements and modelling aspects necessary for other remote sensing methods to determine atmospheric dispersion. This thus eliminates the uncertainties those measurements and calculations may infer, and which may be of concern. However, the use of a gaseous tracer infers other uncertainties. Any difference in the assumed rate of the tracer gas release to the actual release rate will result in measurement error. It is therefore important to ensure that the release rate of tracer gas is accurately controlled. The potential error associated with the controlled release is reduced by using well-calibrated high precision flowmeters combined with weighing tracer gas release bottles before and after a release. It is also important to ensure that the tracer gas simulates the methane emission from the biogas plant well using the procedure described above. The uncertainty related to probe source simulations can be reduced by increasing the measuring distance.

The use of a tracer gas can also be useful to distinguish sources of methane emission. If, for example, a nearby farm is emitting methane, detection of tracer gas may help in determination if elevated methane concentrations are caused by emissions from the biogas plant or the farm. It is possible in many cases to observe separate plumes.

A disadvantage of the TDM using a mobile analytical platform is that it is impractical to use for long-term emission studies (for example continuous measurements over weeks/months), since the plume is traversed continuously using a vehicle during measurement. For longer-term emission measurements a stationary measuring approach (where tracer and methane concentrations are recorded at one location) can be applied.

7 Guidance and recommendations for the inverse dispersion modelling method (IDMM)

The IDMM is described in detail in the Appendix A 4. For the investigation of methane emissions via this approach, the methane concentration on an open path in luv and lee of the investigated site has to be determined. Together with the data of a USA, the methane emission rate from the source area can be derived by using an inverse dispersion model. There are several devices with different measurement principles to measure the methane concentration on an open path, one example is the use of an open path tunable diode laser absorption spectrometer (OP-TDLAS). The following recommendations are mainly addressed to users of inverse dispersion modelling method (IDMM) (either a forward or a backward Lagrangian stochastic model) in combination with OP-TDLAS. The recommendations for the measurement set-up will refer to the OP-TDLAS measurement method. However, the recommendations for the measurements of the meteorological parameters and a large part of the recommendation referring to the modelling set-up can be applied by users of other methane measurement methods in combination with IDMM.

7.1 Planning of the IDMM measurements

Before the execution of an emission measurement at a biogas plant with IDMM, it is recommended to look at the topographic and meteorological conditions at the site. Not all biogas plants are suitable for this method. The requirements to the measurement site, required meteorological conditions and the technical equipment are explained in the following.

Requirements to the measurement site

For the usage of IDMM, the topography, the building structure and the emission situation of the biogas plant (e.g. no or separable other emission sources) should be as simple as possible. The terrain should preferably be flat. It should be possible to install the measurement paths in luv and lee, so that no additional obstacles disturb the measurement. At least in the leeward side (depending on the wind direction), there should be enough free space to perform the emission measurements.

Since IDMM requires measurements in a proper distance to the plant, one should inform the landowners around the plant and ask for permission beforehand.

Analysis of the meteorological situation

Good practice requires a detailed meteorological analysis of the biogas plant area of interest. The dominant wind direction and wind speed at the measuring site can be checked in advance from e.g. a representative wind rose (for at least a year) from a local weather service. The frequency of stability classes is also necessary to know. In case of buildings, the representativeness of the meteorological station for the actual case has to be determined, since the wind field is strongly influenced by buildings. Important are the turbulence effects on the lee side of buildings. In case of no flat terrain, the influence of the topography on the wind distribution and the stability should also be taken into account, including the influence of, e.g., a nearby forest or single trees, lakes etc.

In addition, some reference scenarios with the dominant wind direction and wind speed could be conducted in a dispersion model. Such reference scenarios can be used as a support for the preparation and the planning of a measurement campaign. The day of measurement should then be chosen according to suitable meteorological conditions by examining the weather forecast (if a flexibility in deciding for a measurement day is possible).

The ideal weather conditions, at which the methane measurements can be well performed, are as follows: wind velocity at least 3 m s^{-1} , the friction velocity larger than 0.15 m s^{-1} , defined main wind direction, neutral stability class [43].

Technical Requirements

The following minimum requirements are needed for the IDMM measurement:

1. **Methane concentration detector** for open-path measurements, e.g. an OP-TDLAS device. The following measurement modes are possible:

	Simultaneous Mode	Semi-simultaneous Mode	Non-simultaneous Mode
Additional requirements	One or more methane concentration detectors	A pan-tilt unit	
Installation	Positioning of the corresponding measuring equipment for each desired measuring path (cf. Figure 10A)	Automatic, constantly recurring switch between several measurement paths in a predetermined time interval (software needed) from the same measuring position (cf. Figure 10B)	Manuel switch between several measurement paths by repositioning the measuring equipment serially (cf. Figure 10A)
Benefits	Enables scanning over two or more significant measurement paths at the same time	Enables scanning over several significant measurement paths with one measuring device	No additional equipment needed

A simultaneous or semi-simultaneous mode is highly recommended, especially to enable a simultaneous background measurement. The methane concentration measurement device should be able to record data over several hours in at least 1-Hz resolution.

2. An adequate number of **retroreflectors** (at least one per active measuring path), which enable a measurement path length of at least 300 m. It is recommended that the reflector is equipped with a 250- μm Lexan foil. With thicker Lexan foils interferences may occur and falsify the measurement result. It is recommended to use such a protection foil to avoid contamination of the reflectors.
3. A three-dimensional **ultrasonic anemometer** (USA), which is able to record wind and stability data over several hours (preferably with a resolution of 10 Hz).
4. **Air temperature and atmospheric pressure sensors** for the conversion of the measured concentration in ppm to units of mg m^{-3} . The sensors should be installed over a planted area and should be able to record measurement data over several hours, preferably with a resolution of 1 Hz.
5. **Distance measurement equipment** with a measurement sensitivity of 1-2 m should be used to determine the path lengths and the distance to distinctive points in the measurement area.
6. A **Global Positioning System (GPS)** device to determine the exact locations of the measurement devices, reflectors and other important points.
7. A **reference gas cell** for the calibration of the instruments. Additionally to the cell, calibration gas (e.g. with 500 ppm or 1000 ppm methane) is needed to execute the calibration measurements with the gas cell.
8. Software to run an **inverse dispersion model**.

7.2 Execution of the IDMM measurements

Step 1: Installation of the Ultrasonic Anemometer (USA) and additional meteorological sensors:

The following recommendations include requirements for USA measurements from WMO (World Meteorological Organization).

1. The **position of the USA** is very important. It is recommended that the USA is located in the lee-side of the biogas plant, as the USA will catch the turbulence induced by the biogas plant.
2. It should be ensured that the USA would **not be disturbed or influenced by obstacles** nearby, like trees or buildings.
3. The **measurement height** of turbulent flows is typically between 3 to 5 m above the displacement height [5, 44].
4. The USA stand should be **aligned** vertically to the ground.
5. The **measurement parameters** needed from the USA depend on the requirements of the used model. Usually, the wind velocity vector and the sonic temperature are measured. From these parameters, all other parameters like wind speed, wind direction, friction velocity, variance of the wind velocity vectors and the Obukhov length can be calculated [45].
6. In [45], 10 Hz or 20 Hz are stated as typical values for the **measurement frequency** of the USA used for turbulence measurement. If this is not possible, a configuration of the dispersion modelling program should be used where only wind speed, wind direction and Obukhov length have to be used as parameters.

7. **Additional meteorological parameters**, i.e. air temperature and pressure, are measured with respective sensors. These data are necessary for the conversion between ppm and mg m⁻³. As the measurement data from the instrument usually depend on air temperature and pressure, the air temperature and pressure values are used to correct the data by means of the correction curve. A measurement frequency of 1 s or even 1 min is sufficient for the air temperature and pressure data.

Step 2: Check influence of other methane emission sources:

The surroundings of the chosen biogas gas plant should be checked for possible other methane emitting sources approximately within a 1 km radius ahead of a measuring campaign. Secondary methane sources might be other biogas plants, farms with cattle or manure tanks. Potential sources that are further away might be of no interest due to the progressive dilution with ambient air.

The following recommendations apply for the considered measurement mode:

Simultaneous mode: Any methane emission sources and their temporal fluctuations are automatically considered when the measuring paths are set up appropriately. The quality of the measurement data is then assured at all times.

Semi-simultaneous mode: The coverage of potential secondary sources with the potential measuring path should be checked. In this case, the quality of the measured data is assured. This measurement mode is not suitable for a parallel background measurement when emission plumes of secondary sources cannot be covered.

Non-simultaneous mode: Only if the secondary methane source is emitting continuously with a known emission rate or does not influence the methane concentration within the measurement path, the quality of the measurement data can be assured. The occurrence of temporal fluctuating sources cannot be ascertained within intermediate periods where the focus is on other sources.

Step 3: Installation of Background Concentration Measurement Path:

The background methane concentration is very important due to the high sensitivity to the methane emission rate determination [46].

1. The background measurement path should be **positioned** upwind of a biogas plant and ideally perpendicular to the main wind direction (cannot be applied in a semi-simultaneous mode).
2. The **measurement height** of all involved reflecting and detecting devices (for background and downwind concentration) can be chosen in a user's favour but should ideally be the same in order to make the measured methane values comparable.
3. A **distance to the plant** should be chosen, so that there is no influence by the methane emission of the examined biogas plant itself or does not include secondary methane sources.
4. The **path length** should be identical to the cross section of the plant to cover the complete methane background of the biogas plant and not ascribe potential secondary sources to the plant emissions by mistake.
5. The methane background concentration can be measured with each of the three different **measurement mode** depending on the technical equipment.

- a. A simultaneous mode over the entire measuring period is recommended to avoid temporal fluctuations due to e.g. changing atmospheric mixing processes or the appearance of temporal secondary methane sources.
 - b) In case of a semi-simultaneous mode the background concentration is measured constantly recurring during the actual concentration measurement periods. Step 3a-d should be maintained as close as possible.
 - c) When only a non-simultaneous mode is realisable, the background concentration should be measured before and after the downwind concentration measurement of the biogas plant for at least half an hour. Consequently, the background concentration needs to be interpolated for the intermediate period
8. Depending on the designated length of the measuring path, a **fitting reflector** can be chosen. Reflectors can be distinguished in size and reflective surface. It should be ensured that the reflected signal is appropriate for the detecting device.

Step 4: Installation of Downwind Concentration Measurement Path

1. The **position** of the methane concentration measurement device should be chosen according to the known main wind direction. Best results for the later methane emission rate calculation can be achieved when the whole emission plume of the biogas plant is caught with the chosen measuring paths.
2. The **wind conditions** on site have to be re-checked frequently over the measuring periods and if necessary, the repositioning of the concentration measuring devices should be initialised.
3. The authors from [43] recommend a **distance from the plant** that equals 10 times the height of the highest plant unit. If the measuring instruments are set up too close to the source, the emission plume might pass above the measuring path. If the measuring devices are too far away, the dilution of the plume with ambient air is too high and the detected methane concentration will not be separable from the background concentration.
4. The **length of the measuring path** should be (similar to the background concentration measurement) identical to the cross section of the plant to cover all eventual methane sources plus 20 % to compensate minor fluctuations in the wind direction.
5. The **measurement height** of the downwind concentration should be chosen accordingly to the background concentration. If several OP-TDLAS devices take part at a measurement, they should all be installed in the same height.
6. A **fitting reflector** can be chosen depending on the designated length of the measuring path (similar to the installation of the background concentration).

Step 5: Documentation of the Measurement

1. Ideally, a **measurement protocol** is recorded during a campaign and a measurement report is prepared after the campaign.
2. Very important when using multiple devices (for e.g. concentration measurements, temperature, and wind information) is a proper **time synchronisation** of all devices.
3. The measurement protocol and the measurement report should **include the following information**:
 - Date, participants, purpose

- Information of used instruments
 - Type
 - Time stamps (primarily start and end, readjustments)
 - Positions (by GPS and/or distances to distinctive landmarks)
 - Measuring height
 - Measuring path lengths
 - Meteorological conditions (e.g. cloudiness, precipitation, fog, gust)
 - Significant plant operations (e.g. activated PRVs, maintenance, emptying or stirring of digestate)
4. The documentation with **pictures** of the sky (cloud cover information), pictures of the plant from different view angles, pictures of buildings or obstacles (trees, sees etc.) which can influence the dispersion might be very helpful to confirm the credibility of the dispersion modelling.

7.3 Determination of the emission rate with IDMM

The surface specific methane emission rate $Q_{CH_4,spec}$ of a biogas plant can be deduced from the measured (upwind and downwind) CH_4 concentrations ($C_{CH_4} - C_{CH_4,BG}$) and the dispersion model prediction of the ratio of concentration at the sensor to the emission rate $(C/Q)_{sim}$ according to the following equation [43]:

$$Q_{CH_4,spec} = \frac{(C_{CH_4} - C_{CH_4,BG})}{(C/Q)_{sim}} \quad \text{Equation 2}$$

$Q_{CH_4,spec}$	Surface specific emission rate of methane in $kg\ h^{-1}m^{-2}$
C_{CH_4}	Measured downwind concentration of methane in $kg\ m^{-3}$
$C_{CH_4,BG}$	background concentration of methane in $kg\ m^{-3}$
$(C/Q)_{sim}$	Prediction of ratio of methane concentration at the sensor to the methane emission rate in $h\ m^{-1}$

Depending on the selected dispersion model IDMM requires at least the following information/input in order to determine unknown source emission rates:

1. upwind and downwind gas concentrations in $\mu g\ m^{-3}$ or $mg\ m^{-3}$ ($C_{CH_4,BG}$ and C_{CH_4}); (if concentration is measured in units of ppm or ppb, specific ambient temperature and pressure data at the time of concentration measurement are needed to convert to absolute concentration (i.e. $mg\ m^{-3}$) (see Section 7.1 – Technical Requirements),
2. meteorological data (wind speed, wind direction, Obukhov length L , friction velocity u^* and wind statistics (standard deviations δu_1 , δu_2 , and δu_3 , of the wind velocity components u_1 , u_2 , u_3),
3. roughness height of the surrounding terrain (z_0),
4. source configuration (point, line, area or volume) as well as the source location, and
5. location and height of the concentration sensors (point or line-averaging).

The required model input parameters, the modelling setup as well as data processing and filtering for two exemplary dispersion models (LASAT – Forward Lagrange Simulation of Aerosol Transport and Windtrax - backward Lagrangian stochastic model). More information on the selected dispersion models are given in A 4.3.

Step1: Determination of Modelling input parameters

1. **Filtering of the concentration data (R^2 , light value):** In order to ensure good quality data, the measured CH_4 concentration should be checked to have a return light level between a minimum and a maximum light value specified by manufacturer, and a coefficient of determination (R^2) of the laser signal greater than 98%. The latter check eliminated periods when the spectrum from the laser reference cell did not match that from the gas sample spectrum.
2. **Conversion of concentration from ppm to mg m^{-3} :** The OP-TDLS techniques (see A 4.1) provide concentration in units of ppb or ppm over the measurement path ($\text{ppm}\cdot\text{m}$). Ambient temperature and barometric pressure for each observation period are needed to convert to absolute concentration (i.e. mg m^{-3}), which is used to determine the mass emission rate.
3. Application of **correction factors** between different devices, application of calibration factors.
4. **Averaging periods (10, 15, 30 min):** All observational data used in the dispersion models should represent an average over a reasonable period. Common averaging periods are 10, 15 to 30 minutes in length. Shorter averaging times may not capture an equilibrium state of the atmosphere (required for the application of the Obukhov similarity theory), while longer periods might be affected by changes in the gradual diurnal variation typical of the surface layer or reflect changes in plant operations.
5. **Determination of roughness height z_0 :** A way to harmonize the identification of the roughness length for the simulations is to determine the Corine Land Cover (CLC) class from the Copernicus Homepage: <http://land.copernicus.eu/pan-european/corine-land-cover/clc-2012/view>. The roughness length can then easily be detected from the CLC Class by using the paper from [47].
6. **Determination of meteorological parameters (φ , $|\bar{u}|$, u^* , L , σ_1 , σ_2 , σ_3):** The statistical properties of the wind in the surface layer are determined by a few key parameters: Wind speed, wind direction, the friction velocity u^* , which is determined by the vertical transport of horizontal momentum near the surface - and the Obukhov length L - which quantifies the stability of the atmospheric surface layer. All these parameters as well as wind statistics (standard deviations σ_1 , σ_2 , and σ_3 of the wind velocity components u_1 , u_2 and u_3) are typically determined with the use of a three-dimensional USA.

Step 2: Modelling Set-up

LASAT

In LASAT, the model prediction of the ratio of concentration at the sensor to the emission rate $(C/Q)_{\text{sim}}$ is determined by applying an arbitrary unity emission (e.g. $Q_0 = 1 \text{ g s}^{-1}$) and simulating the corresponding concentration value along the laser path (c_0). In LASAT, each laser path is covered with a certain amount of receptor points. Each receptor point records the simulated methane concentration in e.g. 10-minute means. The mean value of the receptor points for each time step along each laser path is used to calculate the methane emission rate with the inverse dispersion formula (Equation 2).

- **Horizontal/Vertical Resolution:** The vertical resolution should be as fine as possible (at least 1 m), as the (OP-TDLAS devices) measure at a certain height. The horizontal resolution should fit to the model domain. As the modelling domain for biogas plants is approximately 500 x 500 m, the horizontal resolution can be chosen also with 1 m.

- **Modelling domain:** It is important to use terrain for the modelling domain when the biogas plant is located on hilly ground. Model simulations with and without terrain have shown that, applying the terrain, leads to an improvement of the results (all other input parameters stay unchanged). Only for the flat terrain, when the surroundings of the biogas plant are flat too, no model terrain is needed.
- **Receptors along the laser path:** As many receptor points along the measurement path as possible should be set up to calculate the mean value.
- **Source configuration** LASAT runs are possible with different source configurations. The choice of the source configuration depends on a lot of factors (especially the building configuration of the plant) and should be planned in detail. Volume sources are useful as they spread the emission over a bigger volume which guarantees a better mixing of the plume. But this need not always be the case.

Windtrax

In Windtrax, the simulated ratio of concentration at the sensor to the emission rate $(C/Q)_{sim}$ (see Equation 2) is calculated from the number of points along the measurement path (P), the total number of gas particles released at the measurement site (N) and the modelled vertical velocity at “touchdown”, summed across all instances where a particle impacts (touch-downs) the ground within the emission source area (w_0 , m s⁻¹), as defined in Equation 3 [43, 48].

$$\left(\frac{C}{Q}\right)_{sim} = \frac{1}{P_{sim}} + \sum_{i=1}^{P_{sim}} \left(\frac{1}{N_{sim}} \sum \left| \frac{2}{u_{3,0}^0} \right| \right)$$

Equation 3

$(C/Q)_{sim}$ prediction of ratio of concentration at the sensor to the emission rate in s m⁻¹

P_{sim} number of points along the measurement path

N_{sim} the total number of gas particles released at the measurement site

$u_{3,0}$ vertical velocity at “touchdown” in m s⁻¹

- **Meteorological input (var1, var2, var3):** The use of three-dimensional USA data (measured a sampling rate of at least 10 Hz) in Windtrax is described as the most accurate. There are three relevant approaches to provide meteorological parameters measured by a USA in Windtrax (see A 4.3). If the wind components are measured at a frequency of at least 10 Hz, all three input approaches give similar results. Input options var 1 has the advantage of providing all necessary parameters (friction velocity u^* , roughness height z_0 , and Obukhov length L) from the measured raw data (relations of the mean product of u_1 , u_2 , u_3 and sonic temperature T_{sonic} : $\langle u_1^* u_1 \rangle$, $\langle u_1^* u_2 \rangle$, $\langle u_1^* u_3 \rangle$, $\langle u_2^* u_2 \rangle$, $\langle u_2^* u_3 \rangle$, $\langle u_3^* u_3 \rangle$, $\langle u_1^* T_{sonic} \rangle$, $\langle u_2^* T_{sonic} \rangle$, $\langle u_3^* T_{sonic} \rangle$,

$\langle T_{\text{sonic}} * T_{\text{sonic}} \rangle$). However, if meteorological parameters are measured at lower sampling rates (e.g. 1 Hz), it is recommended to use standard statistical relationships estimated by Windtrax (var3). Sampling rates lower than 10 Hz are not sufficiently high to observe the turbulent fluctuations of horizontal and vertical wind.

- **Source configuration:** A dispersion model (C/Q) calculation requires assumptions about the source configuration (e.g., point source, uniform area source, multiple source areas, etc.). A common assumption for anaerobic digestion plants is to outline a source area and assume a spatially uniform emission rate within that area (covering the whole area of the plant).
- **Number of points along measurement path P:** The particles are released from each point spaced evenly along the path length of the sensor. By default 30 points along the measurement path are used. An increase of P will decrease the uncertainty of predicted values, while longer calculation periods are required.
- **Total number of released particles (N):** Windtrax simulates turbulence dispersion by modelling the random movement of thousands of particles backward in time as they travel upwind from the concentration sensor (measurement path) being displaced by horizontal and vertical aerodynamic forces. By default, 50,000 particles are released at a time to shorten simulation time. By increasing the number of particles emitted the uncertainty in the predicted values will be reduced, while the simulation time gets longer. Based on a sensitivity analysis conducted by [49], the mean percentage difference in predicted concentration was less than 1% (maximum of 5%) when comparing particle number setting of N = 50,000 and 1 million.
- **Height of line-sensors:** In Windtrax, the variable heights of the line-concentration sensors are represented as best-fit linear lines with specified beginning and ending heights (only a linear height variation is allowed in the software).

Step 3: Filtering of model output

The IDMM to estimate the source strength depends on a good description of atmospheric transport, which is known to be difficult in extreme stability conditions and/or low wind speeds. The criteria of [48, 50] should be used to exclude observation periods that might provide inaccurate emission calculations. Only the measurement periods that meet the requirements should be used to determine emission rates: $|L| \geq 10$ (strongly stable/unstable atmosphere) and $u^* > 0.15 \text{ m s}^{-1}$ (low wind conditions).

In addition, data filtering should be conducted on the simulated CH₄ emissions by removing emission values that are negative.

Depending on the applied dispersion model the following filtering criteria of the model output should be considered:

LASAT

The model output (emission plume) could be checked visually when the measurement paths were within a representative section of the source plume.

Windtrax

Windtrax reports unknown source strengths or concentrations as a mean with a standard error. In the backward mode, the fraction covered by touchdowns (“footprint”) is recorded indicating the fraction of the

source area where the emissions were “measured” by the concentration sensors (its values range from 0 to 1). For some observation, periods (e.g. change in wind direction) the emission plume may only “glance” the path of concentration sensors leading to uncertain emission estimates.

It is recommended to filter and remove emission rates calculated in Windtrax when the fraction covered by touchdowns <0.6 (unsuitable representation of the emission plume), or the modelled emission rate coefficient of variation > 20% (percentage of standard error divided by the mean), or unrealistic wind profile (surface roughness $z_0 > 0.25$ m, when input option var1 is used).

7.4 Quality control for IDMM

General:

To minimize uncertainties associated with the model input e.g. CH₄ concentrations, meteorological, emission source data), accurate and representative measurements of CH₄ concentrations and meteorological data are of crucial importance for IDMM purposes. For the acquisition of both concentration and meteorological data, two factors are critical: the resolution/sensitivity of the measurement instrument and the measurement location.

Like with all remote sensing methods, IDMM relies on meteorological conditions. Periods known to be problematic for inverse-dispersion calculations such as rapid atmospheric changes, low wind or extreme stability conditions may cause inaccurate emission estimates. Knowledge about atmospheric mixing is required to gain reliable results.

In the following, the main aspects of IDMM-quality assurance are described in more detail.

Ensuring equipment performance:

All used devices, especially methane concentration measuring instruments, have to be calibrated to ensure comparability of the measured methane values. In case of an open path methane concentration measuring device, a simple calibration can be made with a reference gas cell.

The reference gas cell should be continuously flushed using standard gases with known concentrations (e.g. 500 ppm, 1000 ppm, 2000 ppm) and inserted into the light beam of the measuring path. The calibration gas should have a high concentration to be differentiable from the background concentration. The measured concentration in the reference cell can be determined with the following equation:

$$C_{CH_4,RC} = \frac{CL_{CH_4} - CL_{CH_4,BG} \cdot (L_{OP} - L_{RC})}{L_{RC}} \quad \text{Equation 4}$$

$C_{CH_4,RC}$	Methane concentration in reference cell in ppm
CL_{CH_4}	Path integrated methane concentration in ppm m
$CL_{CH_4,BG}$	Path integrated background concentration of methane in ppm m
L_{OP}	open-path measuring distance in m
L_{RC}	length of reference cell in m

Compared to the actual calibration gas concentration the calibration factor is obtained for the current temperature. To assure temperature compatibility, the calibration needs to be performed at various temperatures.

For the meteorological sensors, calibration procedures recommended by the manufacturer should be followed for each meteorological instrument.

Quality assurance of measurement data:

The OP-TDLAS instruments have usually two parameters to control the accuracy of the reflected signal and thus of the measured methane concentration. The 'Light Value' gives the intensity of the back scattered light to the detector. The light value should be between a minimum and a maximum value corresponding to the manufacturer's data. Lower values indicate that the back scattered signal is too low and the output value is not reliable. Higher light values might irreparably compromise the detecting agent. At continuing low values, a different path or a bigger reflector should be installed. If the measured light value exceeds the permitted threshold, a smaller reflector should be considered (recommended). For short term light value increases (e.g. temporal clearing visibility conditions), several size varying masks are available. The use of these masks should be contemplated deliberately. In addition, an attenuation of the reflector with Lexan foils of different thicknesses is possible. The second control parameter is ' R^2 ', which can be understood as a signal to noise ratio. The higher the R^2 the less intercepted is the measured concentration value by noise. Low R^2 values indicate that the incidence angle at the reflector is inconvenient and the inclination of the reflector or the concentration measuring device should be adjusted.

Measured values should be screened based on the completeness of meteorological and concentration data for each observation period. In addition, input data should be reviewed for possible transcription errors. It is recommended to use a simplistic data set with known results to test the modelling software prior to the use of field data in order to verify the performance of the dispersion model.

By means of the inverse dispersion model, the measurement location of the methane concentration measurement device and the USA have to be evaluated, in order to decide if the measurement data are valid or not. Following questions should be answered positively: Does the emission plume cross the source region (covered fraction at least 50%)? Does the measurement path capture the whole emission plume? Is there only little influence of obstacles to the USA, and is the USA at the lee side of the emission source?

It should be mentioned that the dependence on the chosen source region in the inverse dispersion modelling program is much lower if the location of the measurement equipment is chosen thoroughly.

Possible validation procedures and tests:

An option of the validation of the measurements is an experiment using a controlled gas release. There are two possibilities of measurement validation by release tests: the release of methane from a synthetic methane source, and the release of a tracer gas.

In both cases, it should be tested by inverse dispersion modelling if the released amount could be reproduced. With that information, the applicability of the model and the calculated stability class can be

tested. When releasing methane, a known constant emission rate from the biogas plant has to be assumed to be subtracted from the overall result. Based on experiences the trueness of such an assumption is rather uncertain. For the release test with a tracer gas, an additional open path instrument measuring the gas concentration of the tracer gas is needed.

The release unit should be positioned at the biogas plant, so that the measuring device can catch the whole plume. For optimal positioning of the devices, a reference scenario can be considered, e.g. using the main wind direction, beforehand. The release height should reflect the height of the main emitting source.

Synthetic methane should be released for at least 2 hours (gas-on phase). The gas-off phase should be measured before the synthetic release to get the uninfluenced methane from the biogas plant. If the gas-off phase is measured after the controlled methane release, there might be an influence of the release in the first time steps. The difference between the average gas-on and gas-off emission can be used as the estimate of the synthetic methane release rate.

The dependence of the measurement result from the inverse dispersion model highly depends on the choice of the stability class (see also Appendix A 4.4). Therefore, one should check, e.g. with photographs, if the calculated stability class is reasonable or not. Further validation algorithms of the stability class or a validation of the model, should be an issue in further research on this topic.

7.5 Advantages and limitations of using IDMM

The flexibility of IDMM with OP-TDLAS opens up for a wide range of applications to obtain better knowledge of different GHG emissions from various sources. IDMM is a non-intrusive approach requiring only a small number of measurement points to determine fugitive emissions over a long time period. If extended measurement periods with changes in wind direction are planned, multiple measurement points (covering the biogas plant in every direction) might be required to cover all wind directions. Besides inferring whole plant emissions, IDMM can be also applied to simultaneously derive the emission characteristics of spatially distributed multi-component sources using appropriate instrument setup and a sufficient number of concentration sensors [51]. Also, whole plant measurements for sites with restricted access or improper layout could be suitable to partition component emissions by measuring e.g. before and after digestate removal or during normal operations and a downtime of the CHP unit.

In general, IDMM may be associated with uncertainties, which derive both from the inevitable simplification of complex real-world processes and from limitations of the available input data (e.g. on emission sources, concentration and meteorological data). Finding suitable measurement locations while meeting the placement criteria for IDMM (e.g. concentration sensors located at least ten obstacles height downwind) can be limited in complex topographical and infrastructural conditions (e.g. forest areas, hills, dense array of buildings). However, it has been demonstrated that IDMM could still be used in more complex terrain using a diagnostic wind field model.

Finally, a weakness of bLs models might be long computing times due to a large number of unique atmospheric trajectories required for producing statistically reliable results.

8 Documentation

For confirmability and the possibility to compare the results obtained by different methods or at different biogas plants, it is very important to document the measurements, the properties of the surroundings, and important parameters of the biogas plant (most important emission sources, general parameters, and parameters concerning operation during the measurement). Both the remote sensing teams and the on-site teams should document these parameters as precise as possible. This can be very helpful for a later interpretation of the results, and the comparison of emissions from different biogas plants. Also, for further investigations of emission factors of certain biogas plants, biogas plant concepts or whole biogas plant inventories, a precise documentation is needed, so that the emissions can be referred to certain biogas plant operation modes or on-site emission sources. A recommended outline including the most important parameters is given in Appendix A 5.

9 Conclusions

The choice for a certain measurement approach to quantify methane emissions at biogas plants mostly depends on the purpose of the measurement. With the on-site approaches, the identification and quantification of single emission sources are achieved. In contrast, the remote sensing approaches enable a quantification of the overall plant emission or well-delimited parts of the plant. Another important point for the choice of a method is the effort and the expense of the application of the methods or the availability of a measurement team capable of conducting the wanted method. To facilitate the decision for a suitable measurement approach, a table listing the strengths and limitations of the different methods is presented in Section 3.2, and a table summarizing the effort of the different measurement approaches is presented in Section 3.3.

For a better comparison among different teams using the same measurement technique, it is important to use similar measurement procedures and protocols. For that, recommendations for the usage of a certain method are proposed in this document in the form of a measurement protocol. This includes recommendations for a thorough preparation and the course of action of the measurements, the evaluation of the results, the quality control, a precise documentation of the measurement process, the weather situation, the plant parameters and the details concerning plant operation during the measurement period, in particular, all special and other-than-usual events. A list including important information, which has to be included in the measurement report is presented in Appendix A 5.

Within the MetHarmo project, two comparative measurement campaigns were performed [11, 12]. The two measurement campaigns showed that a very precise documentation is crucial for a useful interpretation of the results and the investigation concerning the differences between the emission rates of different biogas plants or the different measurement methods. A list of parameters, which should be documented, can be found in Appendix A 5. A short summary of the results from the MetHarmo measurement campaigns are presented in Section 10.

10 Summary and results from MetHarmo-Project

Within the MetHarmo project, two comparative measurement campaigns were performed [11, 12]. For the first campaign, the DIAL was chosen as the reference method. In previous validation studies, the DIAL measurements showed a deviation of 5 – 20 % from known emission sources [1, 2]. Due to the availability of these validation studies, the DIAL system was selected as reference method to be able to compare the methods. Nevertheless, TDM has shown similar deviation (10-20%) in previous controlled release test [31, 41, 52].

In the following, statistical results from the measurements are presented. It has to be mentioned that the measurements of the different teams did not cover exactly the same time periods. Furthermore, this analysis assumes a constant plant emission rate, which is a critical assumption as the emission of a biogas plant generally cannot be expected to be constant over 3-5 days. Any temporal variability in the emission rate could influence the comparison of different measurement methods if measurements are not performed simultaneously.

During the first measurement campaign, five remote sensing teams (DIAL, three IDMM teams, and TDM) and two on-site teams took part. The measurements were performed over a period of five days. Not all teams performed measurements on all five days and at the same time of the day. The results given as average emission rates and emission factors for all measurements performed during the five day campaign are summarized in Table 3. In the first measurement campaign, the DIAL instrument determined an emission rate of 4.7 kg h⁻¹ (Emission Factor EF=1.2 % CH₄, n=28) in average with a standard deviation of 0.7 kg h⁻¹ (EF=0.2 % CH₄). Two IDMM teams (excluding those measurements with not optimal distance from the plant and those with unfavourable wind conditions) measured an emission rate of 4.8 kg h⁻¹ and 4.9 kg h⁻¹ (EF=1.2 % CH₄, n=24 and n=10, respectively) with a standard deviation of 1.3 kg h⁻¹ (EF=0.3 % CH₄) and 1.6 kg h⁻¹ (EF=0.4 % CH₄), respectively. An additional third IDMM team (with possibly not optimal anemometer position) measured an emission rate of 3.4 kg h⁻¹ (EF=0.8 % CH₄, n=3) with a standard deviation of 2.7 kg h⁻¹ (EF=0.7 % CH₄). The TDM team measured an emission rate of 2.2 kg h⁻¹ (EF=0.5 % CH₄, n=150) with a standard deviation of 1.4 kg h⁻¹ (EF=0.3 % CH₄) during the first measurement campaign. The standard deviation of the DIAL measurements was much smaller than for the other remote sensing teams.

Table 3: Results given as average of all measurements performed by each team during the five day 1st MetHarmo measurement campaign.

	Emission Rate in kg h ⁻¹ and Standard Deviation in kg h ⁻¹	Emission Factor in % and Standard Deviation in %	Number of measurement scans/intervals/transects
DIAL	4.7 ± 0.7	1.2 ± 0.2	28
IDMM A	4.8 ± 1.3	1.2 ± 0.3	24
IDMM B	4.9 ± 1.6	1.2 ± 0.4	10
IDMM C	3.4 ± 2.7	0.8 ± 0.7	3
TDM	2.2 ± 1.4	0.5 ± 0.3	150
On-site A	1.1	0.3	
On-site B	1.7	0.4	

The Student's t-test was used to review if the mean value determined with the different remote sensing methods (IDMM and TDM) is the same as the mean value determined with the DIAL system using a level of significance of 5 %. The mean values of all three IDMM teams do not differ significantly from the mean of the DIAL instrument. However, the mean of the values determined by TDM differs significantly from the mean of the DIAL ($t=14.3$, $df_{unpooled}=74.8$; $t_{crit}=2.0$). It could not be clarified why TDM significantly underestimated the emission rate in comparison to DIAL. Anyhow, the true plant emission was unknown, therefore it is not possible to conclude which method provided the highest accuracy.

Using the formula from DIN ISO 5723:2 [53], the repeatability standard deviation including all valid (only with favourable distance and wind conditions) measurement intervals/scans/transects is 1.4 kg h^{-1} (EF=0.3 % CH₄) and can be interpreted as the variation of the measurement results by using the same method. The inter-method (inter-laboratory) standard deviation is 1.6 kg h^{-1} (EF=0.4 % CH₄). From that, the reproducibility standard deviation is 2.1 kg h^{-1} (EF=0.5 % CH₄), which is 53 % of the average emission rate (average of average from single remote sensing teams: 4.0 kg h^{-1}). The reproducibility standard deviation can be interpreted as the variation of the measurement results by using different methods measuring at a constant emission source.

During the first measurement campaign, the two on-site teams determined an emission rate of 1.1 kg h^{-1} (EF=0.3 % CH₄) and 1.7 kg h^{-1} (EF=0.4 % CH₄), respectively, with minimum and maximum values between 0.8 kg h^{-1} (EF=0.2 % CH₄) and 2.0 kg h^{-1} (EF=0.5 % CH₄), respectively. Hence, during the first measurement campaign, the on-site method most likely underestimates the overall emissions at the biogas plant. The biogas plant here was a quite large site using thermal post combustion after the CHP. At this plant, many small leakages at the gasholder membrane domes, which were not possible to quantify, accounted for the main emission sources at this plant. From that, the total emission rate of the biogas plant is underestimated in this case.

During the second measurement campaign, four remote sensing teams (three IDMM teams and TDM) took part. The results given as average emission rates and emission factors for all measurements performed during the five day campaign are listed in Table 4. Averaging all measurement intervals of the four remote sensing teams, an emission rate of 2.7 kg h^{-1} (EF=2.1 % CH₄) is determined (average of average of the single remote sensing teams is 2.4 kg h^{-1}). Team IDMM A measured an average emission rate of 2.4 kg h^{-1} (EF=1.9 % CH₄, $n=46$) with a standard deviation of 1.1 kg h^{-1} (EF=0.9 % CH₄), team IDMM B measured an average emission rate of 2.6 kg h^{-1} (EF=2.0 % CH₄, $n=37$) with a standard deviation of 1.4 kg h^{-1} (EF=1.1 % CH₄), and the measurements of IDMM C resulted in an emission rate of 1.5 kg h^{-1} (EF=1.2 % CH₄, $n=12$) with a standard deviation of 1.1 kg h^{-1} (EF=0.9 % CH₄). With TDM, an emission rate of 2.9 kg h^{-1} (EF=2.3 % CH₄, $n=123$) with a standard deviation of 1.8 kg h^{-1} (EF=1.4 % CH₄) was determined.

Table 4: Results given as average of all measurements performed by each team during the five day 2nd MetHarmo measurement campaign.

	Emission Rate in kg h^{-1} and Standard Deviation in kg h^{-1}	Emission Factor in % and Standard deviation in %	Number of measurement scans/intervals/transects
IDMM A	2.4 ± 1.1	1.9 ± 0.9	46
IDMM B	2.6 ± 1.4	2.0 ± 1.1	37
IDMM C	1.5 ± 1.1	1.2 ± 0.9	12

TDM	2.9 ± 1.8	2.3 ± 1.4	123
On-site A	2.9	2.3	
On-site B	2.6	2.0	

Student's t-test was used to test if the determined mean values of IDMM B, IDMM C and TDM differ significantly (with a level of significance of 5 %) from the mean value of IDMM A. It appears that the mean values of IDMM B and TDM do not differ from each other, whereas the mean value of IDMM C is significantly lower than the mean value of IDMM B. It is assumed that the difference is caused by the unfavourable anemometer position (luv side of the plant) combined with very low wind speeds, but it could not be clarified completely.

Using the formula from DIN ISO 5725:2 [53], the repeatability standard deviation including all measurement intervals/scans/transects is 1.6 kg h⁻¹ (EF=1.2 % CH₄), the inter-method (inter-laboratory) standard deviation is 0.4 kg h⁻¹ (EF=0.3 % CH₄), and following the reproducibility standard deviation is 1.6 kg h⁻¹ (EF=1.2 % CH₄). Therefore, the variation of the measurement results is the same by using either the same method or different methods.

Three different on-site teams took part in the second measurement campaign, whereas the third on-site team was only measuring the emissions from the leakages, and was therefore not able to determine the overall emission rate with its data only. On-site team A determined an overall emission rate between 2.7 kg h⁻¹ and 2.9 kg h⁻¹, on-site team B determined an emission rate of 2.6 kg h⁻¹. The minimum and maximum values of the emission rate using all on-site data are between 2.5 kg h⁻¹ and 3.1 kg h⁻¹.

During the second measurement campaign, the overall emissions from the on-site method were in the same range as for the remote sensing methods. Here, the emissions were determined at a smaller biogas plant, where the main methane emission sources were the CHPs. However, it needs to be mentioned that the emissions from the second biogas plant were not expected to be at a constant level during the campaign, since the CHP load changed during the measurement campaign due to a flexible operation and the CHP was identified as major contributor to the emissions in that case. Therefore the repeatability and reproducibility standard deviations are quite high (59 % of the average emission rate), which is caused by the variability of the emission rate and only partly by the methods themselves.

Besides the plant emissions, experiments installing an artificial methane emission source off-site the plant has been obtained during the second measurement campaign. The recovery rate (measured methane emission rate divided by released methane emission rate) was compared for all used measurement methods measuring. Here, four different devices for IDMM were used, and the TDM during the same time period. The mean values of the determined recovery rates of all methods varied between 0.9 (IDMM C) and 1.0 (IDMM D) with standard deviations between 0.2 and 0.3 [11, 12, 12]. From that, it could be shown that all remote sensing methods can recover the released methane within the one-fold standard deviation.

With that, it could be demonstrated that, under favourable and exactly the same conditions, the used remote sensing methods showed the same results. In order to reduce the variability range of the results, future investigations should look at a further specification of validation criteria for the measurements (e.g., suitable atmospheric conditions and same operational mode of the biogas plant). This might help to improve the reproducibility of the results and reduce the variation between different methods.

As a final outcome of the campaigns one and two, depending on the plant characteristics, there seems to be a tendency to underestimate the sum of all sources by the on-site methods, in case it is not possible to measure all emission sources, with a significant contribution to the overall emissions. From that, for the measurement of the overall plant emissions with on-site methods, the identification and quantification of the main emission sources of the biogas plant is very important.

The results from the measurement campaigns show that the emissions from a biogas plant can vary significantly over time. Since the comparison of the different methods under the varying conditions of a biogas plant is difficult, comparative measurements should be carried out with very precisely defined conditions, e.g. defined release rate, defined weather conditions (wind velocity, wind direction and stability class).

The measurements also show that a precise documentation of measurement and plant parameters is crucial for a further interpretation of the results, and the comparison between different biogas plants or between different measurement methods. Particularly, strongly varying emission relevant parameters should be documented during the measurements, e.g. filling level of the digestate storage. A list of important parameters, which should be reported is given in the appendix of this document.

11 Open issues and requirements on further research

The comparison of different methods to determine methane emissions within the MetHarmo project was a first step towards more reliable methane emissions measurement results. Within the project, the experiences between the measurement teams could be exchanged, and a direct comparison of the determined methane emissions from the same biogas plant at the same time by different methods was enabled. However, it was shown, how difficult it is to compare different methods with varying requirements (e.g. preference of diverse wind directions due to given topographical or infrastructural conditions) at a biogas plant. For example, it is very difficult to measure exactly at the same time (e.g. caused by organisational and technical challenges for on-site measurement teams or changing weather conditions for remote sensing teams), and the conditions are normally not reproducible. Thus, the differences between the measurement intervals are caused by the intrinsic measurement uncertainty, but also because of the high variability of the emission rate at the biogas plant or a change in measurement conditions, e.g. wind conditions.

Thus, there is still significant work to do concerning the uncertainty assessment of the different methods. Even if the results are within the same range referring to the double standard deviation, it should be aimed for a minimisation of the uncertainties by testing the individual methods, in particular the remote sensing approaches, under more restrictive validation procedures.

For a better assessment of the differences between the single methods, more comparison measurements are needed, preferably under reproducible conditions: defined emission rate, various controlled sources, reproducible wind and turbulence conditions, e.g. in a large wind channel. From that, a more systematic method comparison could be reached.

The knowledge from this guideline will be a basis for the subsequent “EvEmBi” project (funded via 11th ERA-NET call). Here, the focus will be on emissions from single emission sources on a biogas plant

depending on certain parameters. The protocols defined within this document can directly be used for further assessment of emission rates from different biogas plant concept, which will be used, among other data sets, for a statistical quantification system of emission factors to assess the emission factors of the different biogas plant concepts and the national biogas plant inventories.

Within the “EvEmBi” project, the emissions measurements at certain plants will be a basis to elaborate emission mitigation strategies at these plant and verify the success of the implemented measures. From the broaden knowledge about emission mitigation, concepts for operator workshops and national position papers will be elaborated to disseminate the knowledge.

A 1 Method description of the on-site approach and recommendations for the documentation of the measurements

Section 4.1 gives recommendations, which are complemented by the details given in the following sections of Appendix A 1.

A 1.1 Planning of the measurements and analysis of the measurement site

A suggestion for a general structure of a measurement report including the most important plant parameters which can be used in combination with all measurement methods is presented in Appendix A 5. A very detailed documentation of important plant parameters and events on the plant during the measurements might be important for a later interpretation of the results.

A 1.1.1 Planning details for specific investigations of a certain emission source

1. The implementation of the emission measurements on-site the biogas plant is the most important point.
 - a) Check the availability for connections of power supply and cable demand. Does an emergency power supply have to be available?
 - b) Is it necessary to install permanently measurement equipment at the biogas plant? Consider the possible rebuilding and technical acceptance of measuring points. The used measurement instrumentation has to pass the requirements of the corresponding explosion zone.
2. The frequency of the emission measurements should correspond to the time-variance of the emission source.
3. Does the emission measurement requires the sampling of substrate and/or digestate from the investigated biogas plant? The sampling procedure and sample transport have to be planned.

A 1.1.2 Pre-visits at the biogas plant

By undertaking pre-visits at the investigated biogas plant, the measurement institution will identify possible characteristics of the single emission sources. Important points are:

1. Biogas utilisation (CHP, biogas upgrading unit):
 - a. The point of measurement for off-gas sampling has to be localised. Which circuit points are necessary for the connection of the heated gas-sampling probe or the heated sample tube?
 - b. The point of measurement for the analysis of the off-gas volume flow has to be localised. Do the pipe conditions (at five times hydraulic diameter inlet zone before and three times outflow zone after the point of measurement) allow the measurement of the flow velocity? Elsewise the exhaust volume flow can be calculated from operational data with the assumption of full stoichiometric conversion of methane ($\text{CH}_4 + 2 \text{O}_2 \rightarrow \text{CO}_2 + 2 \text{H}_2\text{O}$), the

combustion-air ratio λ of the CHP engine and the fuel feed during the emission measurement (see Appendix O).

- c. From the dimensions of the exhaust pipe, it has to be deduced if measurement traverses (concentration and volume flow) are necessary [5].
2. Air-inflation of double layer membrane domes:
 - a. The design and position of the inflation air outlets on the membrane dome have to be checked. With it, an adapted chamber design can be developed.
 - b. When the outlet is not accessible from the edge of the dome, lifting platforms have to be scheduled.
2. Area sources – open digestate storage:
 - a) The design and position of submerged stirrers have to be checked. When a temperature and/or hydrostatic sensor shall be sunk in the storage tank, a secure distance to the stirrers have to be considered.
 - b) For the representative sampling of digestate, the sampling procedure has to be coordinated with the plant operator.
 - c) The accessibility of the storage tank should be checked. Are there any platforms allowing a positioning of the chamber on other places than the edge of the storage tank?
 3. Pressure relief valves
 - a) The dimension and design of the PRV have to be checked on-site. This includes the construction material (stainless steel or plastic), the cross-sectional area of the exhaust pipe (choice of a suited flow velocity sensor) and the connection of the exhaust pipe to the basic body of the PRV (modification of the PRV).
 - b) Which operational data can be provided for the evaluation of the measurement results? Is a readout of data sets like biogas filling level or biogas pressure possible? Which data periods are saved within the system control (one month, one year or other periods)? Relevant data sets are:
 - i) Operational state and downtime of the primary gas utilisation (CHP or biogas upgrading unit) caused by malfunctions and maintenance
 - ii) Point in time of activation and run-time of the secondary gas utilisation (mostly flare)
 - iii) Biogas filling level and biogas pressure during triggering periods of the PRVs
 - iv) Stirring intervals
 - v) Substrate input (intervals)

A 1.2 Execution of the on-site approach

The on-site approach considers the single emission sources of the whole biogas plant and consists of two basic steps. Firstly, the unknown emission sources on the plant need to be identified by means of a leakage survey to add those to the already known emission source inventory. Afterwards, preferably all single emission source are analysed separately. Based on the determined emission rates, the overall emission rate from the biogas plant can be summed up. Section 4.2 gives recommendations for the execution of the on-site approach, which are complemented by the details given in the following Appendices.

A 1.2.1 Leakage detection

A leakage detection is the sole possibility to identify the localisation of unknown leakage spots. There are different measurement devices for this purpose, including:

- OGI (IR) cameras
- Portable methane lasers (based on OP-TDLAS)
- Portable methane detectors (based on different measurement principles, such as IR, thermal conductivity or FID)

OGI cameras and portable methane lasers allow the remote sensing of emission spots, which are usually difficult to access. Then again, these devices cannot display a methane concentration value at the direct emission spot. An IEA report [54] contains a detailed description of possible measurement devices and their measurement principles. In general, the leakage detection at biogas plants usually requires the following procedure [15]:

- Preparation of a plant survey plan where the OGI camera positions and the detected leakages are documented.
- Scan of typical weak points with a remote sensing device and a portable methane analyser, including the following examples:
 - Wires to adjust agitators
 - Membrane fixation of the membrane domes
 - Biogas pipes
 - Bull eyes
 - Compressors, etc.
- Scan of detected leakages with a portable methane detector and documentation.
- Documentation of weather conditions (wind, temperature, air pressure, etc.).

Advices for the use of an OGI camera:

When using an OGI camera, following instructions for the use are recommended:

1. The OGI camera should not be pointed directly into the sun.
2. The use of the OGI camera as first leakage detection instrument is recommend for outdoor areas only. In closed rooms (e.g. CHP-container, gas compressor stations, etc.) the use of a portable methane laser or methane analyser is recommended.
3. An appropriate temperature range according to the measurement situation should be always adjusted.

Examples for the qualitative estimation of a leakage for the quantification process:

1. Typical leakages at agricultural biogas plants are the wires to adjust the submerged agitators. The leakage is usually caused by an insufficient lubrication. For this kind of leakage:
 - a) The methane concentration is high (10 to 50 vol.%) if the exact emission spot is detected by the portable methane analyser.
 - b) The exit face for the emission is very small.
 - c) The pressure difference between the gasholder and the atmosphere is small (usually about 2–5 hPa).

- d) Considering all three aspects the potential emission rate is presumably small and an emission measurement can be neglected.
2. Typical leakages at agricultural biogas plants occur at the membrane fixation of the gasholder on the digester wall.
 - a) the methane concentration is high (10 to 50 vol.%) if the exact emission spot is detected by the portable methane analyser.
 - b) The exit face for the emission can be high depending on the length of the leakage.
 - c) The pressure difference between the gasholder and the atmosphere is small (usually about 2 – 5 hPa).
 - d) Considering all three aspects the potential emission rate can be high and should be considered for a quantification measurement.
3. A possible leakage at the biogas distribution system can occur at a compressor for instance.
 - a) the methane concentration is high (10 to 50 vol.%) if the exact emission spot is detected by the portable methane analyser.
 - b) The exit face for the emission is usually small.
 - c) The pressure difference between the compressor and the atmosphere can be high depending on the pressure design data of the compressor.
 - d) Considering all three aspects the potential emission rate can be high and should be considered for a quantification measurement.

Structure for a measurement report of a leakage detection:

For the report of a leakage detection, a shortened version of the documentation from Appendix A 5 can be used:

1. Executive summary
2. Introduction
 - a) Client
 - b) Measurement institution
 - c) Date of measurement
 - d) Scope of the investigation (security check, certificate, research, etc.)
3. Site description
 - a) Plant picture
 - b) Description of the relevant biogas-bearing plant components
 - c) Summary of general plant parameters during the leakage detection (e.g. biogas production, substrates, etc.)
4. Method description
 - a) Used measurement instrumentation
 - b) Measurement method
5. Results
 - a) Site map with detected leakages
 - b) Description of every leakage type and probable cause

A 1.2.2 Channelled sources

Channelled sources always have a conducted exhaust pipe or an air vent where the emission mass flow is released to the atmosphere. This type of emission source needs the measurement of the methane concentration and the exhaust volume flow. Then the emission rate is calculated according to Equation 5.

$$Q_{\text{CH}_4} = C_{\text{CH}_4} \cdot \rho_{\text{CH}_4} \cdot \dot{V}_{\text{ex,STP, dry}} \quad \text{Equation 5}$$

Q_{CH_4}	Methane emission rate in mg h^{-1}
C_{CH_4}	Methane concentration in ppmv ($\text{ml}_{\text{CH}_4} \text{ ml}^{-1}_{\text{Air}}$)
ρ_{CH_4}	Methane density in mg ml^{-1}
$\dot{V}_{\text{ex,STP, dry}}$	Exhaust volume flow under normal conditions (0°C , 101325 Pa), dry in $\text{m}^3 \text{ h}^{-1}$

Measurement of exhaust gas concentration

The measurement of the methane gas concentration can be carried out by means of continuous and discontinuous analytical methods. For the determination of the methane concentration usually a FID is used [3, 4]. For a detailed description about possible analytical instruments as well as their correct use and calibration, this document recommends the use of the below stated standards. Depending on the dimension of the exhaust pipe (or air vent) point measurements or measurement traverses have to be performed.

Important standards:

1. Continuous measurement with FID and cutter:
EN ISO 25140:2010 [3]
2. Discontinuous measurement with gas chromatography and FID:
EN ISO 25139:2011 [4]

Furthermore, these standards are the base for the determination of methane concentration, which is necessary for all individual on-site methods (see Sections 4.2.2 – 4.2.5) except the on-site method for the quantification of methane emissions from PRVs.

Measurement of exhaust gas volume flow

For the determination of the exhaust gas volume flow, the measurement of the flow velocity and the cross-sectional area of the exhaust pipe is necessary and can be calculated by Equation 6.

$$\dot{V}_{\text{ex}} = \bar{v}_{\text{ex}} \cdot A_{\text{EP}} \cdot 3,600 \quad \text{Equation 6}$$

\dot{V}_{ex}	Exhaust volume flow under operational conditions	$\text{m}^3 \text{ h}^{-1}$
\bar{v}_{ex}	Average flow velocity at exhaust	m s^{-1}
A_{EP}	Cross-sectional area of exhaust pipe	m^2

The measured volume flows under operational conditions have to be converted to standard temperature and pressure by Equation 7.

$$\dot{V}_{\text{ex,STP, dry}} = \frac{(p_{\text{Off-gas}} - p_{\text{H}_2\text{O}}) \cdot 273.15 \text{ K}}{101,325 \text{ kPa} \cdot (273.15 + \vartheta) \text{ K}} \cdot \dot{V} \quad \text{Equation 7}$$

$\dot{V}_{\text{ex,STP, dry}}$ Exhaust volume flow under normal conditions (0 °C, 101,325 Pa), dry in m³ h⁻¹
 \dot{V}_{ex} Exhaust volume flow under operational conditions
 $p_{\text{H}_2\text{O}}$ Partial water vapour pressure from the off-gas volume flow in kPa
 $p_{\text{Off-gas}}$ Static pressure from the off-gas volume flow in kPa
 ϑ Off-gas temperature in °C

Important standards:

- Measurement of stationary source emissions: EN 15259:2007 [5]
- EN ISO 16911-1:2013 Stationary source emissions – Manual and automatic determination of velocity and volume flow rate in ducts – Part 1: Manual reference method [6]

Calculation of exhaust gas volume flow for the gas utilisation

In case the design of the exhaust pipe does not allow a quantification of the exhaust volume flow, it can be estimated from operational data. The off-gas flow from a BUU can be calculated considering the mass flows of the input (raw biogas) and output (separated carbon dioxide) stream according to Equation 8.

$$\dot{V}_{\text{offgas,BUU}} = \left((\dot{V}_{\text{rawgas,input}} \cdot C_{\text{CH}_4}) + (\dot{V}_{\text{rawgas,input}} \cdot C_{\text{CO}_2}) \right) - \left((\dot{V}_{\text{biomethane,output}} \cdot C_{\text{CH}_4}) + (\dot{V}_{\text{biomethane,output}} \cdot C_{\text{CO}_2}) \right) \quad \text{Equation 8}$$

$\dot{V}_{\text{offgas,BUU}}$ Volume flow off-gas of the BUU in m³ h⁻¹
 $\dot{V}_{\text{rawgas,input}}$ Volume flow raw gas fed to BUU in m³ h⁻¹
 $\dot{V}_{\text{biomethane,output}}$ Volume flow of upgraded biomethane in m³ h⁻¹
 C_{CH_4} Methane concentration in vol.%
 C_{CO_2} Carbon dioxide concentration in vol.%

The off-gas flow from a CHP can be calculated considering the current electrical power and the corresponding raw gas fed to the engine, the combustion-air ratio according to Equation 9 to Equation 12.

$$\Lambda = \frac{0.2095}{0.2095 - \left(\frac{C_{O_2, \text{offgas}}}{100}\right)} \quad \text{Equation 9}$$

Λ Combustion-air ratio
 $C_{O_2, \text{offgas}}$ O₂ content in the off-gas of the CHP in vol.%
 0.2095 O₂ content in air

$$\text{ratio}_{\text{CH}_4\text{-Air}} = \frac{2}{0.2095} \cdot \Lambda \quad \text{Equation 10}$$

$\text{ratio}_{\text{CH}_4\text{-Air}}$ Demand combustion air in m³ h⁻¹
 2 Stoichiometric O₂ demand for 1 m³ CH₄ in m³ Air
 Λ Combustion-air ratio
 0.2095 O₂ content in air

$$\dot{V}_{\text{air, input}} = (\dot{V}_{\text{rawgas, input}} \cdot C_{\text{CH}_4}) \cdot \text{ratio}_{\text{CH}_4\text{-Air}} \quad \text{Equation 11}$$

$\dot{V}_{\text{air, input}}$ Volume flow of the combustion air input to CHP in m³ h⁻¹
 $\dot{V}_{\text{rawgas, input}}$ Volume flow raw gas fed to CHP in m³ h⁻¹
 C_{CH_4} CH₄ concentration from the input of the CHP in vol.%
 $\text{ratio}_{\text{CH}_4\text{-Air}}$ Demand combustion air

$$\dot{V}_{\text{offgas, CHP}} = \dot{V}_{\text{air, input}} + \dot{V}_{\text{rawgas, input}} \quad \text{Equation 12}$$

$\dot{V}_{\text{offgas, CHP}}$ Volume flow off-gas of the CHP in m³ h⁻¹
 $\dot{V}_{\text{air, input}}$ Volume flow of the combustion air input to CHP in m³ h⁻¹
 $\dot{V}_{\text{rawgas, input}}$ Volume flow raw gas fed to CHP in m³ h⁻¹

A 1.2.3 Biogas leakages

For the determination of methane emission rates from leakages, the High Flow Sampling method (also called dynamic chamber or air flow method) is usually used. Instead of a fixed hood like used for area sources (see Appendix O), a leakage has to be encapsulated with a flexible material (e.g. foil) having an in- and output and a connected blower producing a constant air volume flow through the encapsulation. Also commercial products are available, e.g. the “HI FLOW® Sampler” [55]. The aeration can be done only from the suction side or from the pressure side and the suction side as well. The emitted methane from the leakage intermixes with the air inside the encapsulated leakage area. Then the methane/air mixture is conducted to the output of the encapsulation, where the sampling port is located. The methane concentration is analysed by sampling the gas in the in- and output stream of the encapsulation by continuous or discontinuous analytical methods (see Appendix O). The emission rate can be calculated according to Equation 13.

$$Q_{\text{CH}_4} = \dot{V}_{\text{STP}} \cdot \rho \cdot (C_{\text{CH}_4, \text{out}} - C_{\text{CH}_4, \text{BG}}) \quad \text{Equation 13}$$

Q_{CH_4} Methane emission rate in mg h⁻¹
 \dot{V}_{STP} Air volume flow under normal conditions (0 °C, 101325 Pa) in m³ h⁻¹
 ρ_{CH_4} Methane density in mg ml⁻¹
 $C_{\text{CH}_4, \text{ex}}$ Exhaust methane concentration in ppmv (ml_{CH₄} ml⁻¹ Air)
 $C_{\text{CH}_4, \text{BG}}$ Background methane concentration in ppmv (ml_{CH₄} ml⁻¹ Air)

A 1.2.4 Area sources

Open digestate storage tanks

The basic principle of the use of static and dynamic chambers for the quantification of area sources is described in VDI 4285 Sheet 1 [25]. Detailed descriptions are also included in scientific publications [14, 15].

General chamber method principle:

For the investigation of open digestate storages, floating and fixed chamber systems are used. At several different measurement points, they are put on the digestate surface to determine a surface specific emission rate. Repeated measurements on different points of the digestate storage are required, because typical chambers usually have very small surface areas (less than 1 m²) compared to a full-scale digestate storage (several hundred m²). The emission rate of the whole storage tank is calculated by extrapolating the measured surface specific emission rate to the whole digestate surface area according to Equation 14.

$$Q_{CH_4} = Q_{CH_4,spec} \cdot A_{ODS} \quad \text{Equation 14}$$

Q_{CH_4} Methane emission rate in mg h⁻¹
 $Q_{CH_4,spec}$ Surface specific methane emission rate in mg m⁻² h⁻¹
 A_{ODS} Surface area of the open digestate storage in m²

There are two different chamber types. The dynamic chamber works with forced ventilation of air or a certain carrier gas and the static chamber without.

Static chamber method:

The static chamber is a closed system after it is put on the digestate surface, which only has a little opening for the compensation of the air pressure. The measurement method uses the usually linear increasing methane concentration inside the chamber volume. A pump conducts the air inside the chamber to the sampling unit and then back into the chamber volume. After certain time intervals (e.g. 0, 10, 20 and 30 minutes), gas sample are taken into evacuated vials, which are then analysed in the laboratory. The gas concentration analysis with an online instrument like FID is also possible (see Subsection "Measurement of exhaust gas concentration" in Appendix O). If the methane concentration increase proceeds linear, the surface specific emission rate can be calculated according to Equation 15.

$$Q_{CH_4,spec} = \frac{\partial C_{CH_4}}{\partial t} \cdot \frac{V_{ch}}{A} \quad \text{Equation 15}$$

E_{spec} Surface specific methane emission rate in mg m⁻² h⁻¹
 $\frac{\partial C_{CH_4}}{\partial t}$ Linear methane concentration slope in mg m⁻³ h⁻¹
 V_{ch} Chamber volume in m³
 A_{DS} Enclosed digestate surface m²

Dynamic chamber method:

In contrast to a static chamber, a blower or pump forces a certain volume flow of air or carrier gas (e.g. nitrogen) through the dynamic chamber. For this purpose, the chamber has an input and output pipe and a connected blower/pump, which aerates the chamber from the pressure side. The emitted methane from the digestate surface intermixes with the air (or carrier gas) inside the chamber volume. Then the methane/air mixture is conducted to the chamber output, where the sampling port is located. The methane concentration is analysed by sampling the gas in the in- and output stream of the chamber by continuous or discontinuous analytical methods (see Section “Measurement of exhaust gas concentration” in Appendix O). The surface specific emission rate can be calculated according to Equation 16.

$$Q_{CH4,spec} = \frac{\dot{V}_{STP} \cdot \rho_{CH4} \cdot (C_{CH4,out} - C_{CH4,BG})}{A_{ch}} \quad \text{Equation 16}$$

$Q_{CH4,spec}$	Surface specific methane emission rate in $mg\ m^{-2}\ h^{-1}$
\dot{V}_{STP}	Volume flow of air or carrier gas under normal conditions (0 °C, 101325 Pa) in $m^3\ h^{-1}$
ρ_{CH4}	Methane density of methane in $mg\ ml^{-1}$
$C_{CH4,out}$	Exhaust methane concentration in ppmv ($ml_{CH4}\ ml^{-1}_{Air}$)
$C_{CH4,BG}$	Background methane concentration in ppmv ($ml_{CH4}\ ml^{-1}_{Air}$)
A_{ch}	Digestate surface enclosed by chamber in m^2

Non-gastight covered digestate storage tanks

At non-gastight covered digestate storage tanks, the digestate surface is not available for chamber methods. In that case, the whole digestate tank has to serve as a dynamic chamber [26]. For this purpose, a blower aerates the headspace of the digestate storage with a high volume flow of fresh air to replace the concentrated gas phase in the headspace until an equilibrium (constant) methane concentration is reached. The blower usually aerates the headspace on the pressure side. The gas concentration can be directly sampled from the headspace with a sample line. The methane concentration is analysed by continuous or discontinuous analytical methods (see Appendix O). Then the emission rate can be calculated according to Equation 5 (see Appendix O).

Aerobic post-composting of digestate

The determination of methane emission rates from the aerobic post-composting of digestate is also carried out by means of the dynamic chamber method. In contrast to the analysis of open digestate storage, the dynamic chamber (~wind tunnel) usually has a very large scale. The wind tunnel seals a section of the heap, which is aerated by a high volume flow of fresh air from the pressure side. Sampling, analytical analysis and the calculation of the emission rate are carried out like described above for dynamic chambers.

Recommendations for the quantification of digestate storage tanks

1. In case of one installed open digestate storage, the following number of single chamber measurements in dependence on the distribution of the digestate surface can be recommended:
 - a) 100 % liquid surface – six single measurements
 - b) 75 % liquid surface – 25 % surface crust – four and two single measurements

- c) 50 % liquid surface – 50 % surface crust – each with three single measurements on the corresponding part of the digestate surface. If two or more open digestate storages are installed on-site the biogas plant, the number of single chamber measurements per storage can be reduced to limit the effort of time for the quantification measurements.
2. An alternative option for choosing a number of single chamber measurements is based on the size of the area that is being sampled according to VDI 3880 [56].
3. One single chamber measurement should last 30 minutes (half hour average).
4. When the chamber is put on a surface crust, it has to be ensured that the crust does not crack, which potentially causes an overestimation of the emission. Simultaneously, the digestate surface, which is enclosed by the chamber should be widely sealed to the outer digestate surface which is not encapsulated by the chamber. This shall ensure that the emission does not escape from the chamber.

Static chamber method:

1. A cylindrical chamber design is recommended because of the better handling and stability on the digestate surface.
2. The following sampling strategy is required when using a discontinuous sampling method: The first sample has to be taken directly after the chamber was put on the digestate surface. Then, every three minutes one sample is taken until the 15th minute is reached. Then, every five minutes one sample is taken until the 30th minute is reached. This sampling strategy sums up to an overall sample number of nine per chamber measurement. Anyway, alternative sampling strategies or the use of an online instrument is also possible. The used sampling strategy has to be documented.
3. The volume flow of pump recirculating the air volume inside the chamber should be measured by a flow meter. The adjusted value should be documented.
4. Special requirements if a discontinuous sampling strategy is used:
 - a) After the end of one single chamber measurement, a last sample for quality control has to be taken and analysed with an online analyser (usually FID). The measured value has to be documented.
 - b) After the end of one single chamber measurement, the chamber, the connected hoses and the pump have to be flushed with fresh air. After flushing, a sample should be taken from the sampling port and analysed in an online analyser to ensure that no residual methane remained in the chamber system.

Dynamic chamber method:

1. A chamber design according to VDI 3880 [56] is required.
2. It is necessary to perform regular calibration of the volume flow of the internal pump.
3. To ensure that fresh air is injected to the chamber it can be necessary to attach a long flexible hose to the air inlet and put the opening of this hose in the windward direction.

Recommendations for the quantification of non-gastight covered digestate storage tanks

1. For this type of emission source the use of the air injection method is required.
2. The blower aerating the digestate storage should produce a volume flow that is large enough to give the wanted measurement time (Equation 17).

$$t_{\text{theor}} = \frac{V_{\text{HS}}}{\dot{V}}$$

Equation 17

t_{theor} Theoretical time for complete air change in h
 V_{HS} Gas volume in head space in m³
 \dot{V} Air volume flow in m³ h⁻¹

3. The aeration of the digestate storage should last until the whole headspace of the storage tank is replaced with fresh air.
4. To determine if the point of equilibrium has been reached, it is necessary to conduct repeated sampling of the methane concentration in the air outlet after the theoretical time for complete air change, e.g. every 30 minutes.
5. To calculate the emission mass flow, Equation 18 is used.

$$Q_{\text{CH}_4} = \dot{V} \rho_{\text{CH}_4} \frac{C_i}{1 - \frac{C_i}{C_0}}$$

Equation 18

Q_{CH_4} Methane emission in mg h⁻¹
 \dot{V} Air flow in m³ h⁻¹
 ρ_{CH_4} Methane density in mg ml⁻¹
 C_i Methane concentration, end value in percent
 C_0 Methane concentration, start value in percent

A 1.2.5 Pressure relief valves

Technical background and safety issues

PRVs are an important safety unit for the protection of gastight covered digesters and/or gasholders against pressure conditions exceeding the original design parameters. The functional principle of typical PRVs is explained in [8]. In any case, a PRV is a safety device in the end, which should not release biogas under normal process conditions. This is even more so as every release event also results in the emission of raw biogas and methane, respectively.

According to VDI 4285 [25] PRVs cannot be defined clearly as channelled or diffuse emission sources, since they have characteristics from both classifications. On the one hand, during a release event of a PRV, the emission of biogas is channelled by a fixed exhaust pipe. On the other hand, the emission characteristic of a PRV is extremely time-variant, which is a point for a diffusive emission source.

However, the fixed exhaust pipe can be used for the refitting with measurement instrumentation (see following subheading “measurement methods”) allowing a permanent online monitoring [7, 8, 10]. Please be aware that the operational capability of the PRV has to be ensured anytime. The refitting with monitoring instrumentation must not change the release event behaviour of the PRV. In particular, the pressure drop has to be considered, which can be caused by an installed sensor or by an extended exhaust pipe. Thus, the limit pressure of the PRV can increase. Additionally, PRVs are usually rated as a gas explosion zone. Consequently, the used sensors have to meet the demands of explosion category II at least. For legal security of the biogas plant operator and the measurement institution, the measurement setup has to be checked by a technical expert according to the legal regulations in the respective country. [9]

Measurement methods

To date, three single methods for the emission monitoring of PRVs are available. With the impulse counting [57] and the temperature method, the triggering of PRV and the related duration of the single events can be registered. The flow velocity method can additionally measure the methane emission rate released by the investigated PRV. This guideline only considers the flow velocity and temperature method which are described in detail in [8, 9].

Flow velocity method:

The monitoring of the flow velocity in the exhaust pipe of a PRV delivers the most exact measurement of the released methane emission rates from a PRV. A detailed description of the method is given in [8, 9]. When the PRV emits, the sensor registers the flow velocity of the released biogas in the exhaust pipe. The flow velocity is related to the cross-sectional area of the exhaust pipe to calculate the released biogas volume flow. Beside the flow velocity, following information has to be determined:

- Cross-sectional area (inner diameter) of the exhaust pipe
- Biogas composition in the respective gasholder (methane content)
- Normalisation of gas volume
 - Biogas temperature during release events and atmospheric pressure
 - Assumption of complete saturation of vapour of the emitted biogas

A detailed description of the calculation is given in [8, 9].

Temperature method without quantification:

This method is based on the temperature difference between the stored biogas in the gasholder and the ambient temperature [7, 9]. When the PRV emits, the released biogas causes a rising temperature flank. As long as the PRV actively emits, the temperature remains on an elevated level compared to the ambient temperature. When the release event ends, the cooling of the temperature to the ambient level causes a decreasing temperature flank. The signal strength depends on the seasonal ambient temperature level and the material of the exhaust pipe. For the determination of a single release event, the positive and negative slopes of the temperature flanks (first derivative) have to be used. Details are given in [9].

In contrast to the flow velocity method, the temperature method does not allow a direct quantification of the released methane volume. Only a qualitative monitoring including the number and the duration of release events is possible.

Temperature method without quantification:

Though, the temperature method does not allow a direct quantification of the biogas release events, an indirect estimation is possible. The condition is the development of a conversion factor to calculate an emission factor from the duration of the release events measured by the temperature method. A conversion factor can be developed under following conditions:

- At least one PRV is equipped with both sensor types, a flow velocity and a temperature sensor.
- From this PRV:

- The overall duration (t in hours) from all single release events is determined by the temperature method.
- The overall methane emission volume in normal conditions (V_N in m^3) is determined by the flow velocity method.
- Finally, the ratio of both values (conversion factor – CF) is calculated according to Equation 19.

$$CF = \frac{V_{STP}}{t}$$

Equation 19

CF Conversion factor in $m^3 h^{-1}$
 V_{STP} Overall methane volume of all single release events under normal conditions (0 °C, 101325 Pa) in m^3
t Overall duration of all single release events in h

- If all other PRVs are equipped with temperature sensors only, the conversion factor allows the estimation of emission factors by multiplying the release event duration with the conversion factor.

Detailed recommendations and notes for PRV quantification methods:

1. The choice of the measurement method should be carried out depending on the objective of the long term monitoring.
 - a) If the determination of methane emission factors is necessary (e.g. for GHG balances) the flow velocity method is recommended. The temperature method is basically also possible. The condition is the development of a conversion factor to calculate an emission factor from the duration of the release event.
 - b) If only the frequency and times of release events have to be registered, the temperature method is recommended.
2. A check and approval of the measurement setup by a technical expert according to the legal regulations in the corresponding country is strongly recommended. For instance, in Germany the Industrial Safety Regulation [58] and the Ordinance on Hazardous Substances [59] have to be considered. The two main points for the expertise are:
 - a) The measurement instrumentation has to pass the necessary explosion category, which is given by the explosion zone around the investigated PRV.
 - b) The functionality of the PRV as safety unit must not be changed by the measurement instrumentation. In particular, a possible pressure drop has to be considered.
3. Flow velocity method
 - a) The use of vortex sensors should be usually preferred compared to vane anemometers. A vortex sensor has a higher durability, because it has less free to move parts compared to a vane anemometer.
 - b) In case of exhaust pipes with a cross-sectional area higher than 0.15 m, the use of big vane anemometers with low detection limit ($0.2 m s^{-1}$) is recommended.
 - c) If a vane anemometer is used, the stopping time of the impeller has to be considered that could lead to an overestimation of the measured methane emission volume [8].
 - d) The installation of the flow velocity sensor has to be carried out depending on the length and the cross-sectional area of the exhaust pipe. For a length of 1 m and a cross-sectional area of 0.15 m of the pipe (typical design in Germany), the sensor should be installed in

a height of 0.5 m as a compromise between necessary inlet zone (3.3 times the hydraulic diameter) and wind protection (spurious signal).

- e) In case the PRV is connected to a service access with a submerged agitator, the vibrations during mixing intervals could cause spurious signals.
- f) For a general exclusion of spurious signals of the flow velocity sensor, the installation of an additional temperature sensor in the same PRV is recommended.

4. Temperature method

- a) The temperature sensor should be installed in the lower third of the exhaust pipe of the PRV.
- b) The temperature sensor and the resulting temperature flanks have to be calibrated depending on the construction material (Stainless steel or plastic (e.g. polypropylene)) and the colour of the exhaust pipe. Both characteristics affect the self-heating of the exhaust pipe (e.g. by sunshine) and hence the signal strength of the temperature flanks. For grey coloured plastic pipes, as used in [8, 9], slopes of $+ 1 \text{ K min}^{-1}$ (positive flank, beginning of release event) and $- 0.5 \text{ K min}^{-1}$ (negative flank, end of release event) can be recommended for temperature sensors with dip tubes.
- c) The duration of release events determined by the temperature method is overestimated compared to the flow velocity method due to the evaluation with slopes of the temperature flanks [9].
- d) For the quantification of the released methane volume, the use of the flow velocity method is always recommended compared to the temperature method with quantification. However, if the additional use of the temperature method is necessary, for instance due to expenses, the temperature method is also a recommended option.

A 2 Method description for DIAL

The Differential Absorption Lidar (DIAL) technique is a laser-based remote monitoring technique, which enables range-resolved concentration measurements to be made of a wide range of atmospheric species. Mass emission rates of various chemical species from a large area, such as an industrial site, can be obtained by combining DIAL and wind measurements.

The DIAL is able to make measurements of a wide range of species (one or two at a time) in the UV (benzene, toluene) and in the IR (total VOC, methane, ethane, ethene, ethyne, methanol). Other additional compounds could be measured (nitric oxide, sulphur dioxide, ozone, hydrogen chloride, nitrous oxide).

A scanner system directs the output beam and detection optics, giving almost full coverage in both the horizontal and vertical planes. From this data, concentration profiles and mass emission rates along defined vertical cross sections can be produced.

A 2.1 DIAL measurement principle

The DIAL technique uses pulsed tuneable laser radiation, which is launched into the atmosphere over the paths to be monitored. The gas concentration can be measured as a function of range from the laser source by tuning the laser wavelength on and off the spectral absorption feature of the target gas. The DIAL technique operates using these principles in the infrared, visible and ultraviolet spectral regions. This enables a wide range of gases to be monitored specifically and sensitively [60, 61].

The atmospheric return signal at the detector, P_λ , measured by a DIAL system from range r and at wavelength λ , is given by the Light Detection and Ranging (lidar) equation, a simplified form of which is given in Equation 20.

$$P_\lambda(r) = E_\lambda \frac{D_\lambda}{r^2} B_\lambda(r) \exp\{-2 \int_0^r [A_\lambda(r) + \alpha_\lambda C(r)] dr\} \quad \text{Equation 20}$$

$P_\lambda(r)$ Atmospheric return signal in W

r Range in m

λ Wavelength in m

D_λ Range independent constant (dimensionless)

$C(r)$ Concentration of the target gas in ppm

α_λ Absorption coefficient of the target gas at wavelength λ in (ppm·m)⁻¹

$A_\lambda(r)$ Absorption coefficient due to all other atmospheric absorption in m⁻¹

E_λ Transmitted energy in W

B_λ Backscatter coefficient for the atmosphere in m²

The equation has three basic components:

1. a backscatter term based on the strength of the signal scattering medium;
2. parameters associated with the DIAL system;

3. a term which is a measure of the amount of absorption of the signal which has occurred due to the presence of the target species.

In the DIAL technique, the laser is operated alternately at two adjacent wavelengths. One of them is chosen to be at a wavelength which is absorbed by the targeted species (λ_{ON}). The other (λ_{OFF}) is chosen to be at a nearby wavelength which is not absorbed significantly by the targeted species. They are also chosen so that any differential absorption due to other atmospheric species is minimised. Any measured difference in the returned signals is therefore due to absorption by the targeted species.

Pairs of on- and off-resonant lidar signals are then acquired and averaged separately until the required signal to noise ratio is achieved.

The two wavelengths used are close together, hence the atmospheric terms $A_\lambda(r)$ and $B_\lambda(r)$ in the lidar equation can be assumed to be the same for both wavelengths. These terms are then cancelled by taking the ratio of the two returned signals.

The path-integrated concentration (CL) may be derived (Equation 21) by multiplying the logarithm of the ratio of the signals by the ratio of the absorption of the two wavelengths by the target species.

$$CL(r) = -\frac{1}{2\Delta\alpha} \frac{1}{N} \sum_{i=1}^N \log \frac{S_{ON,i}(r)}{S_{OFF,i}(r)} \quad \text{Equation 21}$$

$CL(r)$ Path-integrated concentration in ppm m
 N Number of pulse pairs average (dimensionless)
 $\Delta\alpha$ Differential absorption coefficient ($\alpha_{OFF}-\alpha_{ON}$) in (ppm m)⁻¹
 S Received power after energy normalisation of the on- and off-resonant signals (dimensionless)

The DIAL measurement may be thought of as being comparable to a series of open-path measurements made with virtual retro reflectors at range, r , the spacing of which is defined by the range resolution of the DIAL system. The range-resolved concentration of the target species can then be derived by differentiating the path-integrated concentration. (Equation 22).

$$C(r) = \frac{dCL(r)}{dr} \quad \text{Equation 22}$$

$C(r)$ Concentration at range r along the line-of-sight averaged over the spatial resolution of the DIAL along its line-of-sight (typically 3.75 m) in ppm
 $CL(r)$ Path-integrated concentration in ppm m

Multiple range-resolved concentration measurements can be made along different lines-of-sight and combined to produce 2-D concentration distributions. These are typically made by scanning the line-of-sight in either azimuth or elevation, to produce horizontal or vertical scans. Horizontal scans are generally used to identify different sources across a sight. The typical DIAL measurement configuration is shown in Figure 2, with the mobile DIAL facility positioned downwind of the area being investigated. The DIAL laser beam is then scanned in a vertical plane and the distribution of the target gas in the measurement plane is mapped.

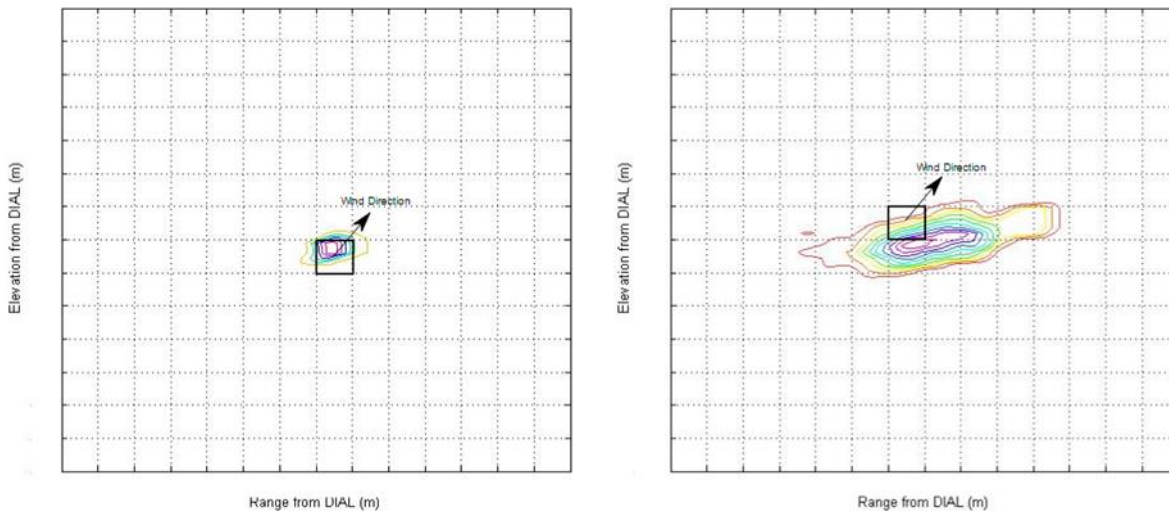


Figure 8: Illustration of the emission rate calculation approach (©NPL).

Vertical scans are combined with wind information to derive the emission rate from the sources. This is done by determining a wind field, as a matrix of wind vectors, in the same plane as the vertical concentration data, and with the same spatial resolution. The emission rate in each cell is then determined by multiplying the wind vector for each cell with the average concentration in that cell. The plane of emission rates is then integrated to determine the total emission rate for that measurement. Figure 8 shows an example of how plume size affects the emission rate that is calculated. This figure shows two example plumes (the cell grids are for indication and are not to scale), one which has a small plume, and therefore a small integrated emission rate, and the other which has a larger plume, and therefore represents a larger emission rate, although the peak concentration in both is similar, and indeed may even be higher in the small plume than the large plume. Note that, in the case of methane measurements, the ambient background concentration is subtracted from the data prior to emission calculation. Ambient background concentration is determined using upwind scans and/or the concentration measured from the last elevation line of the scan. Each scan should be setup such as the last elevation angle does not contain emissions from the target source and therefore is a representative measure of the background concentration plus any further upwind diffuse source.

Figure 9 shows a schematic representation of two measurement plane configurations observing the same plume. One has a nearly perpendicular orientation to the plume, and the wind direction is therefore also perpendicular to the measurement plane. The other is at an angle through the plume, and therefore, the wind is not perpendicular to the plane of the measurements. If only the concentration profile were observed, the right hand measurement configuration would show a larger plume (as it cuts obliquely through the plume). However, when the wind direction is taken into account, the normal component of the wind vector is used, and this reduces the emission rate determined from this scan. This results in the same emission rate being determined for both measurement orientations.

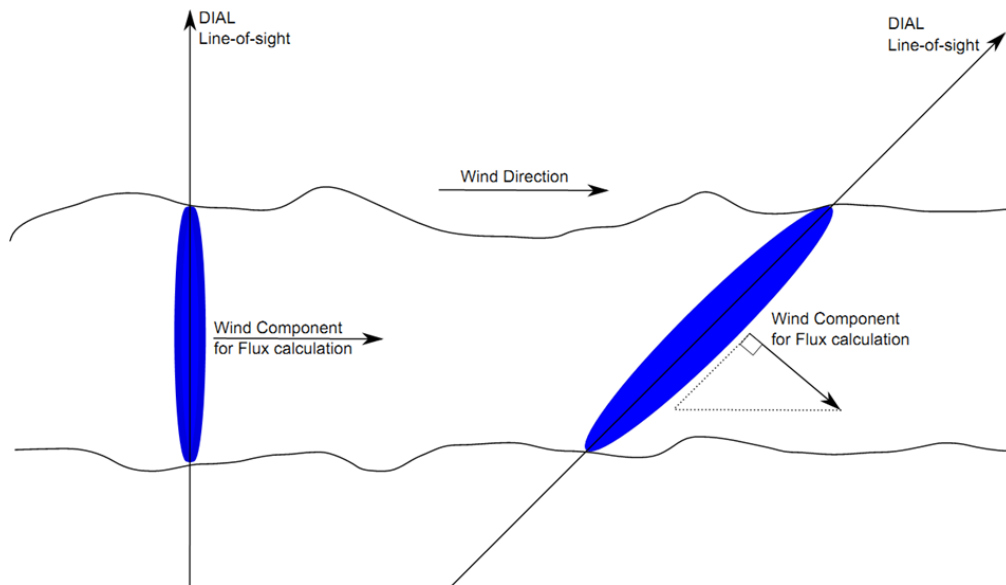


Figure 9: Schematic showing relationship between emission rate and wind direction (©NPL).

Emissions from other areas of the site may have been upwind of the measured target area and may therefore contribute to the measured emission rates. However, upwind sources can be excluded in three ways:

1. If the upwind sources to be excluded are close to the measured sources and produce localised plumes, these can be discriminated spatially from the measured rates by selecting the regions of the scanned region to integrate, in order to calculate the emission rate only from the area of interest.
2. Conversely, if the upwind sources cannot be spatially discriminated and the emissions from them have been measured separately in upwind scans, then these values are subtracted from the downwind emission rate.
3. If the upwind sources are further away (more than ~500 m) and relatively low, they normally would be diffuse and below the measurement noise or detection limit when the emission is relatively low. Larger emissions would appear as an increase of methane background and therefore are taken into account by changing the background level accordingly.

In general, the DIAL will be located so that a series of downwind scans can be obtained, and then it can be moved to a position to be able to monitor the upwind source, see Figure 10A. In many cases upwind and downwind emission rates can be obtained from a single location (Figure 10B).

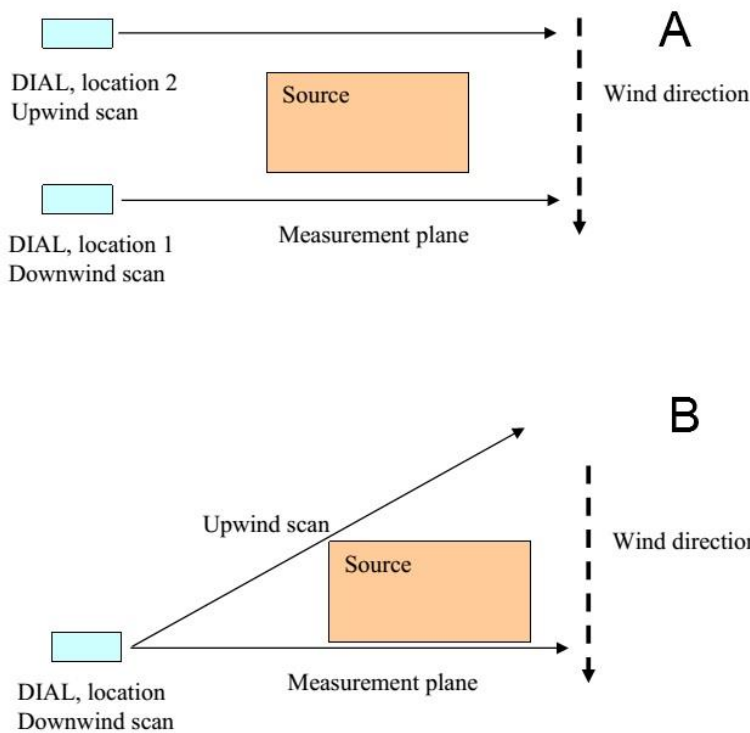


Figure 10: Measurements of upwind and downwind emissions from one (B) or two (A) DIAL locations (©NPL).

The vertical profile varies with atmospheric conditions, but is typically taken to be logarithmic assuming that the wind does not change direction with height and a non-adiabatic process is verified. In such conditions, the flow on the surface layer is defined by non-adiabatic wind and temperature profiles. The wind profile is obtained by fitting to point measurements made using a tall mast, usually deploying two to four wind sensors at different heights up to 12 m to 15 m. Meteorological data is then processed to provide vector averaged wind data for the periods of each DIAL scan.

The meteorological station is typically deployed in a clear area giving an unperturbed wind field. This is a main uncertainty in the determination of the emitted emission rate and it can be reduced by deploying a portable wind sensor along the DIAL measuring line-of-sight to scale the wind profile derived from the fixed mast sensors to match the portable wind speed at the portable elevation.

Local terrain effect can be important and introduce systematic bias in emission rate determinations. The ground elevation where the wind measurement system is located needs to be checked to establish if it is similar to the ground level downwind of the source; if not, the ground elevation along the scan line where the plume is detected should be used as the reference point for establishing the wind profile.

A 2.2 Calibration and validation

The NPL DIAL system has several in-built calibration techniques and procedures. The most important are the in-line gas calibration cells. The gas cells are filled with known concentrations of the target species, obtained from NPL standard gas mixtures, which are directly traceable to national standards. A fraction of the transmitted beam is split off and directed through a gas cell to a pyroelectric detector (PED), in the same way as with the beam for the transmitted energy monitors. This provides a direct measurement of

the differential absorption at the operating wavelengths by the target gas. The transmission through the gas cells is continuously monitored during the operation of the system to detect any possible drift in the laser wavelengths. The calibration cells are also periodically placed in the output beam to show the concentration response of the whole system is as expected.

A number of field comparisons have been undertaken to demonstrate the accuracy of the measurements obtained with DIAL. Examples of these carried out by NPL are summarised below:

Intercomparisons have been carried out on chemical and petrochemical plants where a large number of different volatile organic species are present. In these intercomparisons, the DIAL beam was directed along the same line-of-sight as a line of point samplers. The point samplers were operated either by drawing air into internally-passivated, evacuated gas cylinders or by pumping air at a known rate, for a specified time, through a series of absorption tubes which efficiently absorb all hydrocarbon species in the range C₂ - C₈. The results obtained for the total concentrations of VOCs measured by the point samplers and those measured by the infrared DIAL technique agreed within $\pm 15\%$. The concentrations of atmospheric toluene measured by the ultraviolet DIAL system agreed with those obtained by the point samplers to within $\pm 20\%$.

1. The ultraviolet DIAL system was used to monitor the emission rates and concentrations of sulfur dioxide produced from combustion and emitted by industrial stacks. These stacks were instrumented with calibrated in-stack sampling instruments. The results of the two sets of measurements agreed to within $\pm 12\%$.
2. DIAL Measurements of controlled releases of methane from a stack agreed with the known emission rates to within $\pm 15\%$.
3. South Coast Air Quality Management District (SCAQMD) in Southern California, USA, commissioned in 2015 a series of field tests at refineries and small retail petrol stations as part of a larger research programme. The NPL area source facility was used to separately validate several standard methods to determine fugitive and diffuse emissions at these sites. During the SCAQMD blind controlled release study, the DIAL was demonstrated to be very accurate [1]. The DIAL method was not affected by meteorological conditions and it was able to report emissions from all the test releases. For each release the difference between the released and DIAL rates was less than the standard deviation of the DIAL measurements, see Table 5. This includes Release 7 that was quite low and close to the DIAL detection limit that during this experiment can be estimated to be between 0.5 kg h⁻¹ and 1 kg h⁻¹. The emission rates measured by the DIAL were also linear as shown in Figure 11 with an excellent R² value.

Table 5: Summary of determined propane emission rates, for each controlled release the average emission and standard deviation are reported. Positive and negative % difference between measured and released rates corresponds to overestimation and underestimation of emissions respectively.

Notes	Average Emission	Standard Deviation	Scans	Controlled Release	Difference DIAL-Release
	kg h ⁻¹	kg h ⁻¹	#	kg h ⁻¹	%
Release 1 13:02-13:56 - 3m	4.8	1.3	6	5.8	-17.3
Release 2 14:00-14:55 - 3m	11.0	1.0	4	11.6	-4.8

Release 3 15:00-15:58 - 3m	15.2	3.0	3	17.4	-12.6
Release 4 10:44-11:43 - 3m	12.9	2.0	4	13.9	-7.2
Release 5 11:47-12:40 - 6.4m	5.1	1.3	4	4.6	10.0
Release 6 12:48-13:41 - 7.9m	17.5	3.9	4	18.0	-2.8
Release 7 13:43-14:38 - 7.9m	1.1	0.8	3	1.6	-31.1
Release 8 14:44-15:39 - 7.9m	9.5	1.0	3	9.3	1.9
Release 9 15:44-16:29 - 7.9m	25.0	1.3	3	25.4	-1.5
Release 10 16:34-17:16 - 3m	23.6	1.6	3	23.9	-1.4

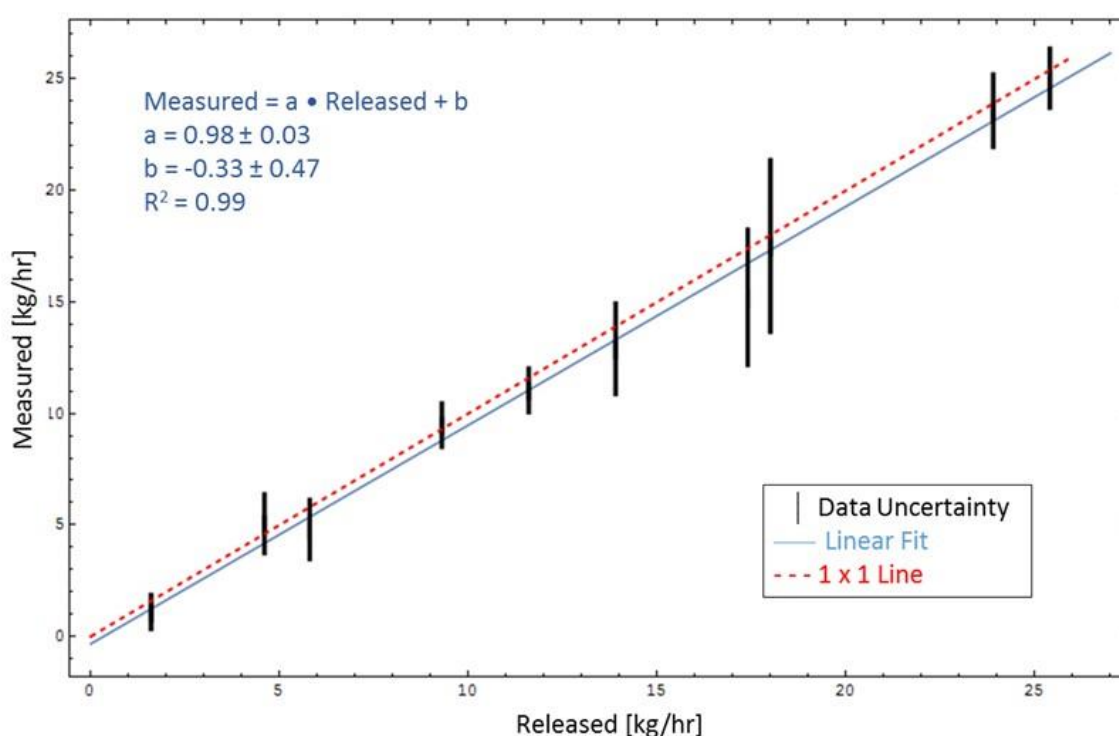


Figure 11: Comparison of actual propane release rates with DIAL measurements (©NPL).

- Validation of the DIAL technique during development of a European standard for VOC fugitive and diffuse emissions. The European Union has defined DIAL as one of the techniques suitable for monitoring fugitive emissions within its Best Available Techniques (BAT) Reference Document for the Refining of Mineral Oil and Gas [2]. The European Union mandated the development of a CEN standard for the determination of VOC fugitive and diffuse emissions, and as a part of that funded a series of validation studies. The results of these studies are still to be finalised and published but initial results of a controlled release study similar to the SCAQMD study again demonstrated the very good linearity, repeatability and lack of systematic bias of the DIAL system compare to other techniques. The standard deviation of a set of at least four DIAL scans was representative of the measurement uncertainty as expected when the dominant uncertainties sources have a random behaviour. The total DIAL measured emission rate for the 18 tests was 184.7 kg h^{-1} while the total controlled release emission rate was 184.6 kg h^{-1} with a difference of only 0.03%. This

confirms that the dominant DIAL uncertainties sources had a random behaviour and therefore by increasing the number of DIAL scans the uncertainty decrease accordingly. Each set of DIAL measurements has shown a behaviour consistent with a standard uncertainty of between 5% to 10% of the measured emission rate, which is lower than the protocol target of 20% to 30%, and with no significant systematic bias.

- Validation of the DIAL against controlled releases at NPL. As a part of research and development programmes NPL have a continuous development programme for the DIAL technology and have conducted a number of validation studies using the NPL controlled release facility (CRF). The results of a series of these have been assessed and published in the peer review literature [62]. These results confirm that the DIAL provides measurements of mass emission rates which are linear with release rate, fall within the stated variability for the measurements, and most critically, that the systematic biases in the DIAL measured emission estimates are less than 4% as illustrated in Figure 12.

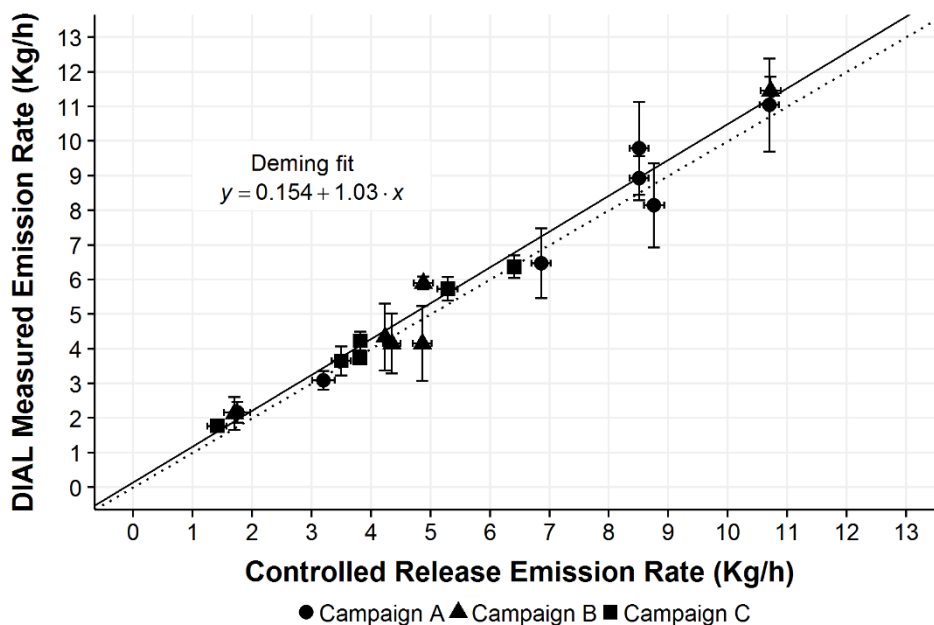


Figure 12: Results of three validation campaigns comparing the NPL DIAL emission rate measurements to 20 CRF release rates (Deming regression fit shown as solid line, 1:1 fit shown as dotted line) (©NPL).

A 2.3 Advantages and constraints

The DIAL system is typically mounted on a vehicle and it can only be parked at locations on roads of a site that do not impact on site operations and safety. An advantage of the DIAL system is that the scanner unit can rotate 360° allowing different line-of-sight measurements to be taken from the same location.

All outdoor optical techniques like the DIAL are affected by different atmospheric conditions although DIAL measurements are not restricted to weather conditions. Fog has an impact on DIAL measurements by reducing the maximum working range but enhancing the signal to noise ratio in the working range. Light rain and snow not only enhance the signal to noise ratio but also the maximum working range. Heavy rain and snow usually require stopping the measurements to avoid deterioration of the scanner mirrors.

Clear atmospheric conditions with few particles would reduce the signal to noise ratio and the maximum working range.

The wind speed and direction are DIAL uncertainty sources and have an impact on planning DIAL measurements. With light wind speeds and variable wind directions it is difficult to determine the sources contributing to the emission along a measurement line-of-sight. The higher the wind speed and the faster the gas concentration is diluted, the more an under-estimation of the emission might occur due to the concentration being below the detection limit of the DIAL. In this situation the DIAL scanner can be quickly reoriented in order to get closer to the emission area. If the wind direction varies significantly it might become impossible to measure the targeted area from where the DIAL is parked. An advantage of the DIAL compared to other optical techniques is that it can be quickly redeployed to a different location in order to carry out measurements of the targeted area.

Another DIAL constraint is that it does not provide data in the first 50 m to 100 m from the DIAL, but this is also one of the main advantages of the technique as the DIAL does not have to be located in the emission source since it has to 'stand-off' by this much. This has the advantage that the DIAL usually does not need to be positioned in restricted site areas (such as process units). Therefore it does not disrupt the routine operations on site.

The DIAL is a complex technique and therefore it is relatively expensive. However, in only a few measurement days the DIAL is able to quantify the emissions from a site of about a kilometre square area and to separate the emission from each of the site's several units. The DIAL has also a very short stand down time compared to other optical techniques as it can quickly react to variable atmospheric conditions. Overall, the quantity and quality of the data (e.g. direct emission mass measurements and three-dimensional mapping of the emission) collected in few DIAL measurement days is cost effective when compared to other techniques.

A 2.4 Uncertainty assessment

The DIAL emission rate is determined by multiplying the vertical concentration data with a matrix of wind vectors in the same plane as the vertical concentration data, and with the same spatial resolution. The wind profile across the measurement plane is a main uncertainty in the determination of the emission rate and the uncertainty in concentration varies with range and atmospheric conditions. The emission rate accuracy can therefore vary from a fraction of kg h^{-1} for emissions close to the DIAL and in favourable atmospheric conditions to a few kg h^{-1} for emissions far away from the DIAL and in unfavourable atmospheric conditions.

A set of at least four DIAL scans should be made in order to minimise the uncertainty from the dominant DIAL uncertainties sources that have a random behaviour. A DIAL measurement is the average emission rate calculated from these scans, which is expected to have an associated uncertainty of about 10% to 30%. The standard deviation of a set of scans will include effects of the source variability, DIAL measurement uncertainty and the influence of other factors such as the wind speed and direction variability during the course of individual measurements. From various DIAL validation studies, the DIAL estimated emission rate uncertainty of a single scan is about 20% to 40%. Some of this uncertainty will be included in the standard deviation of a set of scans.

The uncertainty associated with a set of scans can be further decreased by randomising any systematic effect due to a particular measurement configuration. To achieve this, one or two extra sets of measurements should be made under different wind conditions or along different scan lines or from different locations.

A 2.5 Performance characteristics

The detection limit values of a typical DIAL facility at industrial sites under typical conditions are about 0.1 kg h⁻¹ in the ultraviolet and 0.3 kg h⁻¹ in the infrared. These values are based on the actual levels of performance of existing DIAL systems obtained during field measurements and validation studies, rather than calculations based on theoretical noise performances. However, the actual sensitivity varies with atmospheric conditions, plume dimension and distance from the DIAL, wind speed and angle between the wind direction and DIAL measurement line-of-sight. The detailed performance behaviour of a DIAL system is therefore much more complex and there are a number of key points that should be noted:

1. The DIAL measurement is of concentration per unit length rather than just concentration. Measurements over a shorter path would have a lower sensitivity, and would be more sensitive over a longer path length.
2. For a fixed concentration sensitivity, the detection limit increases as the wind speed increases.
3. Since the backscattered lidar signal varies with range, generally following a (range)⁻² function, the sensitivity is also a function of range. The better sensitivity is obtained in the 100 m to 300 m region from the DIAL, and it will get poorer at longer ranges.
4. The maximum range of the system is generally determined by the energy of the emitted pulse and the sensitivity of the detection system, the atmospheric conditions, the pollutant being monitored and its concentration. It varies between 500 m to 2 km.
5. In all cases the performance parameters are based on those obtained under typical meteorological conditions. For the ultraviolet measurements the meteorological conditions do not have a great effect on the measurements as the backscattered signal level is predominantly determined by molecular (Rayleigh) scattering, and this does not vary greatly. However, in the infrared the dominant scattering mechanism is from particulates (Mie scattering). So the signal level, and therefore the sensitivity, is dependent on the particular loading of the atmosphere, and this can vary dramatically over relatively short timescales.
6. The optical configuration of the DIAL system means there is a minimum range between 50 – 100 m before measurements can be made.

The DIAL theoretical range resolution is determined by the detector bandwidth and the speed of the transient recorder used as data acquisition system. However, the actual range resolution is determined by the signal averaging used that depends on atmospheric conditions and the concentration of the measured pollutant, and may be of the order of 10 m to 30 m.

A 3 Method description for TDM

A 3.1 TDM measurement principle

The tracer gas dispersion method (TDM) is a remote sensing technique, which has been used to quantify fugitive emissions from various area sources such as landfills, composting facilities, waste water treatment plants and biogas plants [15, 31–36, 38]. Figure 3 illustrates the measurement set-up for the TDM. The TDM involves a controlled, continuous release of a gaseous tracer combined with downwind measurements of atmospheric concentrations of target gas (methane in this case) and tracer gas. As described here, the measurements are carried out using a mobile analytical platform consisting of a car fitted with a gas analyser, global navigation satellite system (GNSS) receiver, and a monitor enabling real-time observations of measured concentrations of target and tracer gases.

If the target and tracer gases are well mixed, the relationship between target gas emission rate and the known release rate of tracer gas can be assumed to be equal to the relationship between measured concentrations of target and tracer gases in the plume:

$$\frac{Q_{\text{target}}}{Q_{\text{tracer}}} = \frac{(C_{\text{target}} - C_{\text{target,BG}}) \times M_{\text{target}}}{(C_{\text{tracer}} - C_{\text{tracer,BG}}) \times M_{\text{tracer}}} \quad \text{Equation 23}$$

- Q_{target} Target gas emission rate in kg h⁻¹
- Q_{tracer} Tracer gas release rate in kg h⁻¹
- C_{target} Measured downwind concentration of target gas in ppb
- $C_{\text{target,BG}}$ Measured background concentration of target gas in ppb
- C_{tracer} Measured downwind concentration of tracer gas in ppb
- $C_{\text{tracer,BG}}$ Measured background concentration of tracer gas in ppb
- M_{target} Molar mass of target gas in kg mol⁻¹
- M_{tracer} Molar mass of tracer gas kg mol⁻¹

The target to tracer gas ratio is often established by measuring concentrations across the downwind plumes, and calculating the ratio of cross-plume integrations [42]:

$$Q_{\text{target}} = Q_{\text{tracer}} \times \frac{\int_{\text{Plume start}}^{\text{Plume end}} (C_{\text{target}} - C_{\text{target,BG}}) dx \cdot M_{\text{target}}}{\int_{\text{Plume start}}^{\text{Plume end}} (C_{\text{tracer}} - C_{\text{tracer,BG}}) dx \cdot M_{\text{tracer}}} \quad \text{Equation 24}$$

- Q_{target} Target gas emission rate in kg h⁻¹
- Q_{tracer} Tracer gas release rate in kg h⁻¹
- C_{target} Measured downwind concentration of target gas in ppb
- $C_{\text{target,BG}}$ Measured background concentration of target gas in ppb
- C_{tracer} Measured downwind concentration of tracer gas in ppb
- $C_{\text{tracer,BG}}$ Measured background concentration of tracer gas in ppb
- M_{target} Molar mass of target gas in kg mol⁻¹
- M_{tracer} Molar mass of tracer gas in kg mol⁻¹
- x Distance across the plume

To reduce the overall uncertainty of a measured emission rate the variability associated with performed plume measurements should be reduced. Therefore it is recommended that at least 10 but preferably

more transects are performed. The methane emission rate for each plume traverse is calculated using Equation 24 and the variability of the measured average emission is estimated as the standard error of the mean of the measurements (the sample standard deviation of the calculated emission rates of the individual plume traverses divided by the square root of the number of plume traverses) [41].

A 3.2 Equipment

To accurately quantify methane emissions using the tracer gas dispersion method, a high precision gas analyser is needed capable of measuring concentrations of methane and tracer gases (alternatively one instrument per gas) at a relatively high measurement frequency. Instruments with precision levels of below 1 ppb for CH₄ and tracer gas, and a measurement frequencies of app. 1 Hz or better are recommended. Examples of such instruments are cavity ring down spectrometers and instruments utilising off-axis integrated cavity output spectroscopy.

The method involves geographical evaluations of measurements, where it is recommended to include a GNSS receiver as part of the instrumentation. GNSS receiver with a horizontal positioning accuracy of 1 to 2 m is adequate. A data logger is necessary to record gas analysis data and positioning information, if a GNSS is included in the setup. Data logging capability may be a feature of the analytical instrument.

It is preferable (but not necessary) to monitor, and record weather information such as wind speed, wind direction, atmospheric pressure and temperature at the site. This can be done using a mobile weather station, hand held wind sensors, etc.

It is important to carefully control the release rate of the tracer gas, since any uncertainty in the tracer gas release rate will affect the overall uncertainty in emission quantification (Equation 24). The release rates can be monitored using calibrated variable area flow meters or mass flow controllers. The release rates may be double checked by timing tracer release, and weighing gas bottles before and after release.

Acetylene gas (C₂H₂) is often used as tracer at release rates of app. 1 to 2 kg h⁻¹ from 1 to 3 locations. Acetylene gas is favoured for several reasons: low cost, low background concentration, low risk of interfering sources and availability of suitable analytical instruments. Industrial grade purity of this gas (typically >99% purity) is sufficient.

A suitable vehicle is necessary to perform measurements. The vehicle must be able to transport the equipment listed above, as well as tracer gas bottles, etc. Note that transport of acetylene bottles poses safety concerns due to the flammability of this gas. Gas bottles should ideally be transported in an open storage compartment (i.e. a pickup truck), or alternatively in a separate, ventilated storage compartment (i.e. a van fitted with ventilation for the cargo compartment). Gas suppliers often provide guidance/training concerning safe transport of their products. Due to the often rural locations of biogas plants, and that the optimal measurement location often is not the nearest major road, it is practical to use a rugged vehicle capable of travelling on small country roads.

A 3.3 Uncertainty assessment

The overall uncertainty of TDM measurements is a combination of several uncertainties, some of which are instrument specific and some of which vary according to specific measurement conditions [41].

Factors, which may contribute to the overall uncertainty, the analytical uncertainties, uncertainties in the tracer gas release rate, data processing, and in tracer gas placement and source simulation [41].

Controlled release validation trials have been performed to assess the overall measurement uncertainty. In a recent field trial, two measurement teams from the Technical University of Denmark and University of Southampton, UK using two different analytical platforms performed simultaneous TDM measurements while a third team (National Physical Laboratory, UK) managed a controlled release system. This release system was designed to simulate an area source of methane emission from a number of release nodes (combined methane release rates between 5.3 and 10.9 kg h⁻¹). From these trials, rate differences between controlled release rate and measured emissions were generally below 10% and no higher than 18% [53]. These results were in line with an assessment of the uncertainty reported in [42]. By establishment of an error budget and comparison with the measured error based on the release test, it could be concluded that following best practice when performing measurements, the overall error of a tracer gas dispersion measurement is very likely to be less than 20% [41].

A 4 Method description for IDMM

Inverse dispersion modelling means to derive source strengths of emissions from measured concentrations at points upwind and downwind from the source combined with meteorological data using a dispersion model. Such models are a set of mathematical equations that attempt to simulate (model) the cause-effect link between source emissions and downwind concentrations (the „C-Q relationship“) at a given state of the atmosphere. The choice of the best appropriate model to be used depends on a range of factors: general application of the model, open source or license model, level of expertise of the user, nature of the available input information (location and terrain, building configuration, source configuration, meteorological data, etc.), practical consideration (accuracy of the results, temporal and spatial resolution etc.). Therefore, it is an important prerequisite for the determination of emission fluxes from the area of interest.

The inverse dispersion method can determine emission rates from defined source areas of any shape. The concentration measurement can be a point or a line average obtained with closed- or open-path (OP) analysers. Typically, OP sensors are used since they provide a better average over the source plume and reduce sensitivity to changes of the wind direction.

A 4.1 Methane measurement methods

Different measurement methods can be used to determine the methane concentration over an open path, which can be used in combination with IDMM for the determination of an emission rate (e.g. Open-Path Fourier Transform Infrared Spectrometer – passive or active). The application of these optical remote sensing methods are tested for emission rate determination, which is described in detail in [63].

A widespread and cost-effective method to determine path-integrated methane concentrations is the open-path Tunable diode laser absorption spectroscopy (OP-TDLAS). With the scanning over one gas specific absorption line, the detection of the specific gas is unambiguous and not influenced by other gases.

An OP-TDLAS device usually combines a light transmitting and a light receiving unit (monostatic configuration). The light beam, which is emitted by a laser diode, propagates through a gas mixture including a certain amount of the interested gas on an open path. In a proper distance, it is reflected by a retroreflector and can be detected by the light receiving unit of the OP-TDLAS device.

Depending on the gas of interest, a laser diode emitting light in a narrow wavelength range around a compound specific absorption line is chosen. The gas molecules absorb energy from the light beam, if the certain absorption wavelength is emitted. The detected absorption spectrum depends on the number of molecules of the interested gas on the open path. A qualitative measurement of the number of gas molecules on the open path is therefore possible and leads, considering air temperature and atmospheric pressure, to the path integrated gas concentration (usually in ppm).

The authors of this document used an OP-TDLAS, which used a wavelength of 1653.7 nm for the detection of methane. From that, the recommendations for IDMM are based on the authors' experiences

and measurements with this devices. However, most of the recommendations can be used for other gas concentration measuring devices as well.

A 4.2 Meteorological measurements

Meteorological measurements are crucial in order to obtain a simulation of emissions that is as realistic as possible to the natural dispersion in the atmosphere. Since the natural dispersion in the atmosphere is turbulent a USA with three-dimensional wind velocities (u , v , w) and a temperature sensor (T) is most fitting for the requirements. The wind vector and the temperature are derived based on the sound propagation along differently oriented paths.

Atmospheric stability can be determined from the Obukhov stability parameter (OSP) in m^{-1} , the inverse of Obukhov length L . Positive values of OSP indicate stable, negative values unstable atmospheric conditions; values near zero are classified as neutral. In general, the stability is the tendency to resist or enhance vertical motion (turbulence) in the atmosphere. While a stable atmosphere resists turbulence, the unstable atmosphere enhances thermal turbulence. Furthermore, the turbulence in the atmosphere greatly affects the concentration of a pollutant. The more unstable the atmosphere, the greater the dilution of a pollutant.

A 4.3 Dispersion models

For the simulations, two exemplary Lagrangian models will be introduced, which are described in the two following sections. Of course, other state-of-the-art dispersion models can be used. In general, there are a huge number of available models for dispersion modelling. The choice of the best appropriate model to be used depends on a range of factors: general application of the model, open source or license model, level of expertise of the user, nature of the available input information (location and terrain, building configurations, source configuration, meteorological data, etc.), practical consideration (accuracy of the results, temporal and spatial resolution etc.).

Lagrangian stochastic models can be employed in forward or backward mode to derive C-Q (sensor-source) relationships. In the forward case, individual particles are released from a prescribed source area, and transported with the mean and a stochastic turbulent flow field (e.g. LASAT - Lagrangian Simulation of Aerosol-Transport) [64]. In the backward case, the same formalism is applied, but the particles are modelled backward in time as they travel upwind from the concentration sensor. By analysing the backward trajectories, the locations, where the particles touch down within the source area, are recorded (e.g. Windtrax – backward Lagrangian stochastic [bLS] dispersion model [65]).

With a time series of concentration and wind measurements, the use of a dispersion model requires various inputs including the source type (point, line, area or volume), the source location and the related roughness characteristics of the surrounding terrain (roughness height z_0) as well as the location and height of the concentration sensors (point or line-averaging). In addition, meteorological data are required and should contain at least wind speed, wind direction and the stability of the atmosphere near ground (see Section 7.2). If concentration is measured in units of ppm or ppb, ambient temperature and barometric pressure are also needed. The measurement data (concentration and meteorological observations) is typically organised into suitable averaging intervals (e.g. 10 – 30 min).

LASAT

LASAT (Lagrange Simulation of Aerosol Transport), version 3.3 [64] is a model for the simulation of the dispersion of trace elements in the atmosphere. In the dispersion simulation, transport and turbulent diffusion are calculated for a large number of particles by means of a stochastic process (Lagrange simulation). LASAT is conform to the VDI guideline 3945 Sheet 3 (particle model; [66]). This is the official regulatory model in Germany as stated by the German Technical Instruction on Air Quality Control (TA Luft, [67]), and it was developed by Janicke Consulting on behalf of the German Federal Environmental Agency.

The model calculates the dispersion of passive trace substances in the lower atmosphere (up to about 2000 m altitude) in the local and regional surroundings (up to distances of about 150 km). The terrain can be plain or structured and it may contain buildings. For complex terrain, a diagnostic wind field model is integrated in the meteorological pre-processor. The diagnostic wind field model takes into account the flow around buildings; in this case, the recirculation and the increased turbulence developing on the lee side are parameterised.

Source term: For biogas plants, most of the sources (leakages) are usually not well known or difficult to define. LASAT can select a large number of emission sources (defined as point, line, area, raster, grid, or volume sources). Most of the parameters for the dispersion calculation – especially the emission strengths of the individual trace substances and the dynamic behaviour of the sources – should be defined in the time series (model input). The length of the averaging time can be freely chosen.

Meteorological input: Wind speed, wind direction and atmospheric stability are the minimum data required for the model set up. If the meteorological data are available from a weather station (as a time series over a year or as an annual statistics), both annual means and short-time values (percentiles and excess frequencies) can be calculated. The representativeness of such data for the site should be checked in advance. Available meteorological measurements directly in the vicinity of the plant should be preferred, since the assessment of the local turbulence has a major influence on the advection of the released substances. The atmospheric stability (defined in stability classes in the most of the models) has a major influence on the dispersion of the gases. LASAT uses the Klug-Manier stability scheme.

LASAT can use the Obukhov length L from three-axis USAs as a measure of the vertical mixing (turbulence) of the near-ground atmosphere. The Obukhov length L corresponds to the height above ground in which the mechanical turbulence production (on obstacles) and the thermal turbulence damping (by stable stratification) are balanced.

Influence of the terrain: Sensitivity studies with flat terrain and with including the topography have shown that in some cases the latter can result in a significant improvement of the results. If existing, topographical features have to be included when doing dispersion calculations with LASAT.

Windtrax

The backward Lagrangian stochastic model (bLs) is implemented by the free software product WindTrax (Thunderbeach Scientific, version 2.0.8.9), which assumes idealised wind conditions appropriate for simple undisturbed terrain (using Obukhov similarity theory) [65]. The software tool provides an easy-to-use graphical interface (simulation elements such as sources and sensors are represented by icons drawn on the screen) for predicting turbulent transport of pollutant gases over short range. A short

introduction guideline to WindTrax is available under <http://www.thunderbeachscientific.com/>. The method tends to be most reliable in open, flat terrain, for the short range distances (source-receptor separation <1 km), when non-ideal wind conditions are ignored and the source-sensor geometry (i.e., the positions of concentration sensors relative to the source) is optimal [43].

Source term: Windtrax determines emission rates from arbitrarily shaped area sources (ground based) and point sources, simultaneously;

Meteorological input: The use of three-dimensional USA data in Windtrax is described as the most accurate. There are three relevant approaches to provide meteorological parameters measured by a sonic anemometer in Windtrax:

- The first approach (var1) uses raw data (u_1 , u_2 , u_3 and sonic temperature T_{sonic}) measured by a three-dimensional USA. Raw averages are calculated (e.g. 10-min intervals) using relations of the mean product of u_1 , u_2 , u_3 and sonic temperature T_{sonic} : $\langle u_1 * u_1 \rangle$, $\langle u_1 * u_2 \rangle$, $\langle u_1 * u_3 \rangle$, $\langle u_2 * u_2 \rangle$, $\langle u_2 * u_3 \rangle$, $\langle u_3 * u_3 \rangle$, $\langle u_1 * T_{\text{sonic}} \rangle$, $\langle u_2 * T_{\text{sonic}} \rangle$, $\langle u_3 * T_{\text{sonic}} \rangle$, $\langle T_{\text{sonic}} * T_{\text{sonic}} \rangle$. Windtrax calculates friction velocity u^* , roughness height z_0 , and Obukhov length L from the provided data.
- The second approach (var2) uses a four-variable meteorological data set composed of wind speed, wind direction, Obukhov length L and roughness height z_0 as well as information on the wind statistics (σ_1 , σ_2 , σ_3 all given as a ratio to friction velocity u^*). Wind velocity, wind direction, u^* , L and wind statistics can be derived from the USA data (either provided by the USA software or own calculation necessary). z_0 has to be determined by the user (see Section 7.3).
- The third approach (var3) uses the same meteorological data set as the second approach (wind velocity, wind direction, L and z_0) but in this case the wind statistics are estimated by Windtrax using empirical relationships.

Concentration input: Windtrax handles both point and line-average (open-path) concentration sensors with specified beginning and ending heights. In the latter case the line-averaged concentration is assumed to be the average of P point concentrations spaced evenly along the path length. A set of predefined pollutant gases with the respective molecular weight (e.g. CH₄, NH₃, N₂O) is available for point and line sensors. Measured concentrations can be either entered in units of concentration over the measurement path (i.e. ppm*m), line-averaged or point concentration (i.e. ppm) or absolute concentration (i.e. mg m⁻³).

Windtrax can be employed in forward (to calculate particle trajectories for point sources) or backward mode (to calculate particle trajectories for area sources) in order to generate unknown emission rates and concentrations.

A 4.4 Uncertainty assessment

Sensitivity analyses based on artificial modelling exercises

Determining emission rates by IDMM are dependent on a number of measurement variables (e.g. concentration, wind velocity vectors) as well as input parameters for the dispersion model (e.g. source term, surface roughness). A simplified sensitivity analysis has been carried out to evaluate the robustness of IDMM, given the various uncertainties which affect the model input variables and parameters. Results

were stated as the changes in emission estimates to be expected from small changes in parameter values. It should be noted that the results of the study are not directly related to the IDMM accuracy and may not correspond to physical reality, but can identify the relative importance of the input variables and parameters. This is helpful in indicating how accurately the input variables must be measured or how assumptions about the source configuration or surface roughness affect the model output (=emission rate).

A base case was the model setup used for the assessment as shown in Table 6. The model setup was applied in Windtrax to determine line-average concentrations ($C - C_{BG}$) (measurement fetch: 100 m, measurement height: 1.5 m, path length: 300 m) downwind of a defined area source (5,000 m²) emitting 4 kg CH₄ h⁻¹ (= control emission rate) for neutral atmospheric conditions (Klug Manier (KM) Class III/1). A wind direction of 270° with a velocity of 3 m s⁻¹ and a surface roughness length of $z_0=10$ cm were considered. For each calculation in the sensitivity analysis, only one parameter was altered based on a realistic uncertainty budget (see Table 6). The other input parameters remain as per the baseline case. The parameters considered are the difference of the two concentration measurements ($C - C_{BG}$), sensor height (h_c), wind speed ($|\vec{u}|$), wind direction (φ), anemometer height (h_a), atmospheric stability class, source area (A) and surface roughness (z_0). Each measurement variable possesses a measurement uncertainty. Values for $C - C_{BG}$, $|\vec{u}|$ and φ were altered within a realistic range of typical measurement variations for 10 minutes averages (based on the MetHarmo project). Since concentration data was directly entered in absolute concentration (mg m⁻³), the total uncertainty in determining the path-integrated concentration was considered including the accuracy of the distance measurement, the resolution of the OP-TDLS system as well as the measurement variation of the temperature and pressure sensors for 10 minutes averages.

Table 6: Input parameters and their variations for sensitivity analysis

Input parameter	Base case	Scenario 1 (plus)		Scenario 2 (minus)	
		Absolute value	Variation in %	Absolute value	Variation in %
<i>Concentration measurement</i>					
Concentration ($C_{CH_4} - C_{CH_4,BG}$)	0.102 mg m ⁻³ based on $Q = 4$ kg/h	0.130 mg m ⁻³	28.2%	0.073 mg m ⁻³	-28.2%
Sensor height (h_c)	1.5 m	1.65 m	10%	1.35 m	-10%
<i>Meteorological parameter</i>					
Wind speed ($ \vec{u} $)	3 m s ⁻¹	3.45 m s ⁻¹	15%	2.55 m s ⁻¹	-15%
Wind direction (φ)	270°	297°	10%	243°	-10%
Stability class	Neutral (KM III/1)	Neutral (KM III/2)		Unstable (KM IV)	
Anemometer height (h_a)	5 m	5.5 m	10%	4.5 m	-10%
<i>Source parameter</i>					
Source area (A_s)	5,000 m ²	7,500 m ²	50%	2,500 m ²	-50%
<i>Surface parameter</i>					
Surface roughness (z_0)	10 cm	15 cm	50%	5 cm	-50%

The parameter $C_{CH_4} - C_{CH_4,BG}$ in units of mg m⁻³ is calculated from several measurement values by using the following formula:

$$C_{CH_4} - C_{CH_4, BG} = \left(\frac{CL_{CH_4}}{L_{DW}} - \frac{CL_{CH_4, BG}}{L_{BG}} \right) \frac{p \times 16.04 \times 0.1}{R_m \times (273.15 + T)} \tag{Equation 25}$$

- CL_{CH_4} Path integrated methane concentration at the downwind measurement path in ppm m
- $CL_{CH_4, BG}$ Integrated path concentration at the background measurement path in ppm m
- $L_{OP, DW}$ Length of downwind path in m
- $L_{OP, BG}$ Length of background path in m
- p Atmospheric pressure in mbar
- T Air temperature in °C
- R_m Gas constant $8.3144598 \text{ kg m}^2 \text{ s}^{-2} \text{ K}^{-1} \text{ mol}^{-1}$

Table 7 presents the error propagation of the standard errors of the measured variables to the absolute standard error of the variable $CL_{CH_4} - CL_{CH_4, BG}$ in units of mg m^{-3} . By using Gaussian error propagation, this leads to a relative standard error of 28.2%, which is used in scenario 1 and 2 (see Table 4). The associated error describes the sensitivity of the single measurement variables to the variable $CL_{CH_4} - CL_{CH_4, BG}$.

Table 7: Measurement variables needed for determining the difference between upwind and downwind CH₄ concentrations ($C - C_{BG}$) in mg m^{-3} and their assumed errors/variations for a realistic uncertainty budget.

Measurement variable	Base case	Unit	Variation (s) [Unit]	Variation [%]	$L_{OP, BG} + s$	$L_{OP, DW} + s$	$CL_{BG} + s$	$CL + s$	$p + s$	$T + s$
Path length ($L_{OP, BG}$) background	300.0	m	1.0	0.3	301.0	300.0	300.0	300.0	300.0	300.0
Path length ($L_{OP, DW}$) downwind	300.0	m	1.0	0.3	300.0	301.0	300.0	300.0	300.0	300.0
$CL_{CH_4, BG}$	600.0	ppmm	6.0	1	600.0	600.0	606.0	600.0	600.0	600.0
CL_{CH_4}	646.0	ppmm	10.0	1.5	646.0	646.0	646.0	656.0	646.0	646.0
Pressure (p)	1000.0	mbar	100.0	10	1000.0	1000.0	1000.0	1000.0	1100.0	1000.0
Temperature (T)	15.0	°C	1.5	10	15.0	15.0	15.0	15.0	15.0	16.5
$CL_{CH_4} - CL_{CH_4, BG}$ (Set point)	0.102	mg m^{-3}			0.106	0.097	0.088	0.124	0.112	0.101
% deviation from set point					4.4%	-4.7%	-13.2%	21.9%	10.0%	-0.5%
<i>associated error</i>					2.4%	2.8%	21.8%	60.5%	12.6%	0.0%
<i>absolute standard error</i>						0.03 mg m^{-3}				
<i>relative standard error</i>						28.2%				

Except for the stability class, the remaining key parameters used in the dispersion model were changed assuming reasonable degrees of variations from ±10% (for sensor and anemometer height) to ±50% (for surface roughness and source area). The source area was always a square and ranged from 2,500 to 7,500 m². The variation of the stability class is based on observed maximum divergence in L or OSP-values from three USAs placed on different locations at the same emission site (during the MetHarmo project).

Figure 13 shows the results of the sensitivity analysis as a percentage of variations in the model output compared to the base case (control emission rate $Q_{CH_4} = 4 \text{ kg h}^{-1}$). The results confirm that IDMM is sensitive to atmospheric stability, wind speed, surface roughness and concentration measurements. The defined plus or minus variation for $CL_{CH_4} - CL_{CH_4,BG}$ and wind speed is directly related to the percentage deviation in the model output. The inaccuracy in the spatial treatment of the emission source has a minor impact on the model in case the concentration is measured far enough downwind of the emission site. With these findings, it can be concluded that by obtaining high quality data (concentration and meteorological measurements) and careful selection of the surface roughness length, the variation bands can be decreased and thus improve the quality of the emission estimations by IDMM.

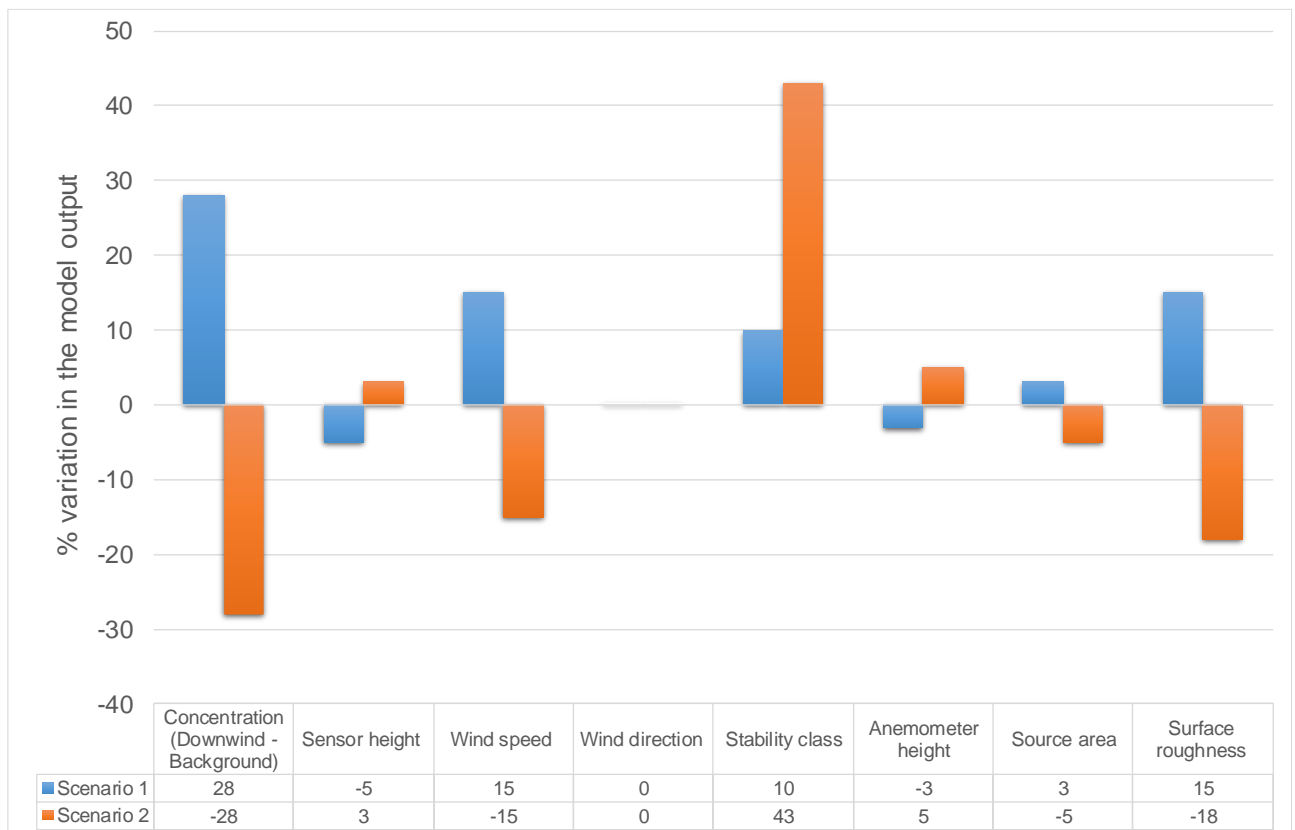


Figure 13: Expected variations in the model output due to variations in the input parameters according to scenario 1 (plus) and scenario 2 (minus).

Variations of IDMM results observed in the field

During the MetHarmo project three different teams using IDMM revealed methane emissions from two biogas plants. The uncertainty budget of IDMM with the dispersion model Windtrax is assessed by using real experimental variability measured with different OP-TDLAS and USAs during the measurement campaigns. The IDMM results of Team A are selected as a base case. As shown in Table 8, either the meteorological (scenario 1) or the concentration (scenario 2) input is altered, the other parameters remain as per the baseline case. The emission estimates are based on measurement days at two different biogas plants including synthetic methane release experiments on-site and off-site the plant. While different measurement locations (at the luv and lee side of the plants) of the USAs are considered, concentration measurements are performed at the same position. In Figure 14, results are depicted as a percentage change in mean emission estimates (according to the data of IDMM Team A = base case)

due to variations of meteorological and concentration input data measured by two other IDMM teams (B and C). Focusing on scenario 1, the variations of the meteorological input from two USAs led to a change in mean emission estimates in the range of 23% (maximum) and 5% (minimum). Scenario 2 revealed a variation of -16% (maximum) and -3% (minimum) by using different concentration input of the individual OP-TDLAS.

Table 8: Overview of the base case (IDMM Team A) and the variations of meteorological and concentration input measured by two IDMM Teams (B and C)

Base case	Scenario 1		Scenario 2	
	Meteorological input		Concentration input	
Team A: • Concentration data (OP-TDLAS Team A) • Meteorological data (USA Team A: lee side)	USA Team B (plant 1: disturbed luv side; plant 2: lee side)	USA Team C (luv side)	OP-TDLAS Team B	OP-TDLAS Team C

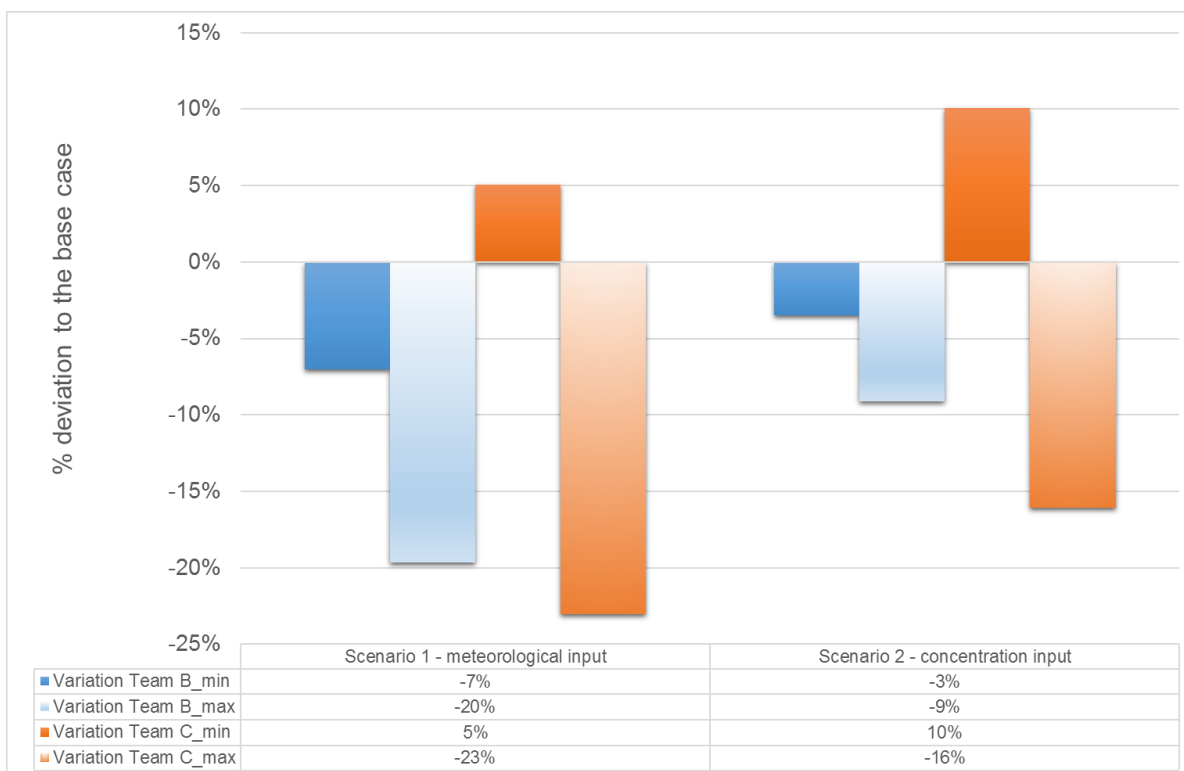


Figure 14: Minimum and maximum variations of IDMM results observed in the field

Uncertainty assessment based on synthetic gas release experiments

In previous projects, the efficiency of IDMM (using Windtrax and LASAT) was investigated using a synthetic area source (2 x 2 m) with known CH₄ release rates (5 l min⁻¹) at different meteorological conditions and laser path arrangements in an ideal surface-layer setting [68, 69]. The accuracy of IDMM is expressed as

the mean recovery rate of the known release rates and yielded $97 \pm 20\%$ (standard deviation) for IDMM using LASAT, and $91 \pm 34\%$ for IDMM using Windtrax. During the MetHarmo project, synthetic methane release experiments yielded a mean recovery rate of the known release rates $92 \pm 20\%$ for IDMM using Windtrax and $71 \pm 6\%$ for IDMM using LASAT considering measurement of four individual OP-TDLS systems.

From the overall results, it is clear that used dispersion models cannot be generalised. The performance of the models used depends on several factors like meteorology, emission scenarios, details of model input, source configurations, and so on. The fundamental difference between LASAT and Windtrax is the model philosophy: whereas the advantage of Windtrax is the direct backwards calculation (from concentrations to emissions), LASAT can do this only indirectly; but the strengths of this model are the selection of very different source configurations and the inclusion of terrain, if necessary. In this case, a diagnostic wind field is calculated which apparently refines the calculations.

A 5 Structure for measurement report including important plant parameters

In the following, a list of data and documentation which should be delivered from a measurement team after the measurement is presented. A detailed and precise documentation is a requirement to produce results which can be compared to results from other biogas plants and/or measurement teams. A precise documentation of all relevant plant parameters can also be useful for further investigation concerning the emission factor of a biogas plant in dependence of the used technologies or to compare different biogas plant concepts to each other, which can further be a first step in emission mitigation. Depending on the measurement approach, the availability of the data and the scope of the investigation, the description of certain parameters is optional. Please be aware that the following list is a suggestion only and does not claim to be complete.

1. Executive summary

2. Introduction

- a) Date of measurement
- b) Scope of the investigation (security check, certificate, research, etc.)

3. Site Description

- a) Biogas plant
 - i) Plant picture
 - ii) Plant type
 - iii) Specific particularities (e.g. construction year and commissioning, operation licence, type and number of digesters etc.)
 - iv) Description of the relevant biogas-bearing plant components
 - v) biogas production
 - vi) Used substrates
 - vii) Process parameters
- b) Biogas utilisation
 - i) Type of engine/upgrading unit
 - ii) Exhaust after treatment
 - iii) Combustion-air ratio of the CHP engine
 - iv) Maintenance intervals
- c) Digestate Storage
 - i) Open or covered (non-gastight or gastight covered)
 - ii) Dimension
- d) Gasholders
 - i) Integrated or external; respective storage capacity for each gasholder
 - ii) Type of membrane dome (single or double membrane, mechanically or pneumatically staked)
 - iii) Type of PRV (hydraulic, hydraulic weighted, producer, design)
 - iv) Measurement equipment for the determination of the gasholder filling level (type, automatic recording and integration in the system control)
- e) Mode of operations during the measurement period (plant diary)
 - i) CHP load
 - ii) periods of stirring of open digestate storage

- iii) periods of pumping from open digestate storage tank
- iv) periods of flare operation
- v) Filling level
- vi) Periods of PRV releases
- vii) Regulation of the flare (manually operated, automatically operated by pressure or biogas filling level)
- viii) Gasholder filling level in normal operation
- ix) Full load-/partial load operation of the gas utilisation (flexible biogas utilisation and/or production)
- f) Surrounding Area
 - i) Topographic particularities (e.g. card with altitude)
 - ii) Buildings and plantations (e.g. height of the plants) around the site
 - iii) Identification and description of possible other emission sources in the vicinity
- g) Weather Conditions
 - i) Picture of weather situation
 - ii) Air temperature and pressure
 - iii) Main wind direction and wind speed
 - iv) Sky cover
 - v) Atmospheric stability class

4. Method description

- a) Used measurement instrumentation
- b) Measurement method
- c) Description of validation procedure

5. Results

- a) Site map with measurement locations
- b) Important parameters of measurement instrumentation and set-up (e.g. height of open-path instruments etc.)
- c) Time (start and end) of measurement/ measurement intervals/ transects/ scans
- d) Number of valid measurement intervals/ transects/ scans
- e) Reason why other measurement intervals/ transects/ scans cannot be used
- f) Determined emission rate of single measurement intervals/ transects/ scans
- g) Average, Median, Standard Deviation of emission rate for all valid measurement intervals/ transects/ scans

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PUBLICATIONS

Previously published reports:

Most of the publications are in German language. The language of the title reflects the language of the publication.

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