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Methodologies for measuring fugitive methane emissions from landfills – a review

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Highlights

- Several methods exist for quantifying fugitive methane emissions from landfills
- Temporal and spatial variations in methane emissions are a challenge for all methods
- Downwind methods are best for quantifying methane emissions

Keywords: Landfill gas, qualitative methane reconnaissance techniques, surface flux chambers, tracer gas dispersion, eddy covariance, mass balance, radial plume mapping, DIAL.
Abstract

Fugitive methane (CH$_4$) emissions from landfills are significant global sources of greenhouse gases emitted into the atmosphere; thus, reducing them would be a beneficial way of overall greenhouse gas emissions mitigation. In Europe, landfill owners have to report their annual CH$_4$ emissions, so direct measurements are therefore important for 1) evaluating and improving currently applied CH$_4$ emission models, 2) reporting annual CH$_4$ emissions and 3) quantifying CH$_4$ mitigation initiatives. This paper aims at providing an overview of currently available methodologies used to measure fugitive CH$_4$ emissions escaping from landfills. The measurement methodologies are described briefly, and the advantages and limitations of the different techniques are discussed with reference to published literature on the subject.

Examples are given of individual published studies using different methodologies and studies comparing three or more methodologies. This review suggests that accurate, whole-site CH$_4$ emission quantifications are best done using methods measuring downwind of the landfill, such as tracer gas dispersion and differential absorption LiDAR (DIAL). Combining aerial CH$_4$ concentration measurements from aircraft or unmanned aerial vehicles with wind field measurements offers a great future potential for improved and cost-efficient integrated landfill CH$_4$ emission quantification. However, these methods are difficult to apply for longer time periods, so in order to measure temporal CH$_4$ emission changes, e.g. due to the effect of changes in atmospheric conditions (pressure, wind and precipitation), a measurement method that is able to measure continuously is required. Such a method could be eddy covariance or static mass balance, although these procedures are challenged by topography and inhomogeneous spatial emission patterns, and as such they can underestimate emissions significantly. Surface flux chambers have been used widely, but they are likely to underestimate emission rates, due to the heterogeneous nature of most landfill covers resulting...
in sporadic and localised CH$_4$ emission hotspots being the dominant emission routes.

Furthermore, emissions from wells, vents, etc. are not captured by surface flux chambers. The significance of any underestimation depends highly on the configuration of individual landfills, their size and emission patterns.

1. Introduction

Landfill gas (LFG) is generated when organic material disposed in landfills decomposes anaerobically. The main components in LFG are methane (CH$_4$: 55–60% v/v) and carbon dioxide (CO$_2$: 40–45% v/v), the production of which occurs through three initial sequential phases followed by a phase characterised by stable CH$_4$ and CO$_2$ production (Christensen et al. 1996). LFG will continue to form until the majority of the organic material in the waste has been degraded, which can take several decades. Landfill gas that is not recovered or microbially oxidised in the landfill cover will be emitted into the atmosphere. Factors affecting the transport processes of LFG, leading to CH$_4$ migration and emission, can be divided into three classes: meteorological conditions (barometric pressure, precipitation, temperature, wind), soil/cover conditions (cracks, fissures, permeability, diffusivity, porosity, moisture content, organic content, CH$_4$ oxidation capacity) and waste and landfill conditions (gas production rate, internal barriers, gas vents, lateral migration area) (Kjeldsen, 1996). Of particular importance are spatial and temporal variations in the soil’s physical and chemical properties that influence soil gas transport and microbial activity (composition, depth, moisture, temperature, aeration status) (Scheutz et al., 2009).

The global emission and accumulation of greenhouse gases in our atmosphere is of great concern, and many countries have consequently committed themselves to reducing their emissions. Therefore, it is of the utmost importance to understand greenhouse gas sources and
sinks. CH$_4$ is a potent greenhouse gas with a global warming potential of 28 over a 100-year timeframe (without including climate change feedback) (Myhre, 2013). The global atmospheric concentration of CH$_4$ has increased from a pre-industrial value of about 722 to 1803 parts per billion (ppb) in 2011 (Hartmann et al., 2013), and it was about 1850 ppb in 2017 (Dlugokencky, 2018). The current atmospheric concentration of CH$_4$ far exceeds the pre-industrial range of the last 800,000 years (350 to 800 ppb) as determined from ice cores (IPCC, Masson-Delmotte et al., 2013). Of the approximately 49 GtCO$_2$eq y$^{-1}$ in total anthropogenic GHG emissions in 2010, about 16% (~7.8 GtCO$_2$eq y$^{-1}$) came from CH$_4$ (Victor et al., 2014).

An important source of anthropogenic CH$_4$ involves the disposal of organic waste in landfills. Of the total amount of municipal solid waste generated globally, about 67% is disposed in sanitary landfills or discarded in non-sanitary landfills or dumps (Fischedick et al., 2014). In Europe, landfills are the second largest (20%) anthropogenic CH$_4$ emission source, while in the United States, they are the third largest (20%) (EEA 2016, USEPA 2016). On a global scale, CH$_4$ emissions from landfills are estimated to account for about 8% of anthropogenic CH$_4$ emissions (Blanco et al., 2014). Between 1970 and 2010, CH$_4$ emissions from solid waste disposal almost doubled (Fischedick et al., 2014), and future gas emission as a result of waste disposal will depend on generated waste amounts, the success of diverting waste away from landfilling and the implementation of mitigation actions. Estimates indicate that annually about 12% of CH$_4$ generated in global landfills is captured at sanitary landfills (Themelis and Ulloa, 2007), and so controlling fugitive CH$_4$ emissions from landfills should be prioritised in waste management procedures. In Europe, for instance, landfills have to be registered nationally if they receive more than 10 Mg waste per day or have a capacity above 25,000 Mg (CEC, 2006). Moreover, if the emission is greater than 100,000 kg CH$_4$ annually, it should be reported to the European Pollutant Release and Transfer Register (CEC, 2006).
The amount of CH$_4$ emitted from landfills is often estimated using theoretical gas generation models, applying standard conditions with respect to waste composition and surface CH$_4$ oxidation. The first-order decay (FOD) model is generally recognised as being the most widely used approach, as recommended by the Intergovernmental Panel on Climate Change in the IPCC Waste Model (IPCC, 2006). The FOD model assumes that the degradable organic matter in waste decays slowly over a few decades, during which time CH$_4$ and CO$_2$ are formed. If conditions are constant, the rate of CH$_4$ production depends solely on the amount of carbon remaining in the waste. The FOD models in the IPCC (2006) guidelines are multi-component with individual values for CH$_4$ generation potential and CH$_4$ generation rate constants from the degradable organic carbon contained in various waste fractions. In some countries other FOD models have been developed to estimate CH$_4$ emissions from specific landfills – examples being LandGEM (US) (Alexander et al., 2005), GasSim (UK) (Gregory et al., 2003) and Afvalzorg (The Netherlands) (Afvalzorg, 2015; Scharff & Jacobs, 2006). The landfill CH$_4$ emission is obtained by subtracting the fraction of CH$_4$ that is collected and the fraction of the uncollected CH$_4$ that is oxidised from the modelled CH$_4$ generation. At landfill sites with gas collection, the amount of gas collected is often well-known, whereas the fraction of CH$_4$ oxidised is unknown. The IPCC (2006) suggests a default value of maximum 10%, which has been highly debated (e.g. Chanton et al., 2009). Stable carbon isotopic analyses have been used for in-situ landfill CH$_4$ oxidation quantification (e.g. Chanton et al., 2008a; 1999; Börjesson et al., 2007); however, the validity of several assumptions in the methodology remains unresolved. FOD gas generation models have been complemented with more recent, site-specific process-based emission models that take into account local conditions such as soil type, climate and CH$_4$ oxidation rates to calculate overall CH$_4$ emissions. One example in this regard is the Californian process-based CH$_4$ emission model CALMIM, which includes site-
specific seasonal soil microclimate and CH$_4$ oxidation data (Spokas et al., 2015; 2011; Bogner et al. 2014; Bogner et al., 2011; Spokas and Bogner, 2011). The CALMIM model assumes that gas transport solely is governed by diffusion processes, while a similar model, the LandSEM model, also includes advective gas transport (Abichou et al., 2015). The process-based cover models estimate methane emission from the landfill cover using representative soil characteristics, and are thus not able to include emissions from landfill infrastructures such as leachate wells. Only few of the available models have been validated, and it is uncertain as to what extent they describe actual CH$_4$ emissions (Cambaliza et al., 2017; De la Cruz et al., 2016; Mou et al., 2015; Spokas et al., 2015; Bogner et al. 2014; Scheutz et al., 2011a; 2011b; Scharff and Jacobs, 2006). In addition, input parameters such as deposited waste amounts and compositions are often of low quality or even unknown, and no single model is applicable to all landfills, due to the significant variations in landfill designs and operation (construction and aftercare), meteorological conditions (precipitation, temperature, wind, etc.) and landfill cover design and conditions. To assess landfill CH$_4$ emissions, and to evaluate the efficiencies of mitigation initiatives such as gas collection, recovery systems and CH$_4$ oxidation installations, measurement methods that can directly quantify CH$_4$ emissions are thus needed. Furthermore, the quantification of CH$_4$ emissions based on measurements will provide more accurate estimations and help evaluate the performance of currently used landfill gas generation and emission models.

Several measurement techniques are currently available, but none of them has been recognised as an international reference method. In addition, very few are commercially available – and then only in a handful of countries. A few new methods have been developed in the last few decades, albeit only in a small number of cases have the measurement techniques been compared to existing techniques and/or to gas generation models used in
different countries. The main challenge in measuring CH\textsubscript{4} emissions from landfills is their
temporal and spatial variability. Several studies have shown high spatial variations in landfill
surface CH\textsubscript{4} emissions (Lando et al., 2017; Scheutz et al., 2008; 2003; Ishigaki et al., 2005;
Czepiel et al., 1996a; Verschut et al., 1991); for instance, rates can vary by up to seven orders
of magnitude within a few metres, due to cracks or holes in the soil cover, thereby leading to
emission hotspots (Bogner et al., 1997). Rachor et al. (2013; 2009) investigated variations in
CH\textsubscript{4} emissions within just one square metre and found that even on this small scale, emissions
were very heterogeneous. Several studies have suggested that at some sites a substantial
proportion (50-75\%) of the total emission occurs from a very small part of the total surface
area dominated by hotspots (Gonzalez-Valencia et al., 2016; Scheutz et al., 2011a; 2011b;
Bergamaschi et al., 1998; Czepiel et al., 1996a). Furthermore, the temporal variability of CH\textsubscript{4}
emissions is often caused by changes in atmospheric conditions, and it has been reported how
both pressure changes and absolute pressure can lead to fluctuations within just a few hours
(e.g. Xu et al., 2014; Fredenslund et al., 2010; Gebert and Gröngröft, 2006; Czepiel et al.,
2003; Christophersen et al., 2001). Most of the studies reporting on temporal emission
variation were carried out at landfills without active gas collection or even at sites with landfill
gas venting (intended or unintended). It should be emphasised that operationally, at modern
landfills, wellfield vacuums are frequently adjusted relative to weather conditions, and at such
sites emissions could thus be expected to be less influenced by atmospheric pressure changes.
However, only a few studies have investigated the influence of barometric pressure changes at
landfills with gas collection. In three recent studies carried out at such locations, CH\textsubscript{4}
collection and emissions were found to vary significantly over time and to be related to
changes in atmospheric conditions such as barometric pressure changes and changes in wind
speed and direction (Rees-White et al., 2019; Aghdam et al., 2019; Fjelsted, et al., 2019).
Temporal and spatial emission variations, combined with the large area and challenging
topography of most landfills, make CH$_4$ emission quantification a challenging task.

In general, landfill emission measurement techniques can be divided into two
categories: surface emission factor techniques and mass emission techniques, also called
‘bottom-up’ and ‘top-down’ techniques. Surface emission factor techniques are based on
quantifying emissions emanating from part of the surface of the landfill and obtaining an
emission factor, which then is applied to the remaining areas of the landfill. Mass emission
techniques, on the other hand, involve quantifying the landfill emission from a larger area or
the whole site. The objective of this paper is to provide a review of the different measurement
techniques by explaining the techniques as well as their applications, advantages and
limitations when quantifying an entire landfill’s CH$_4$ emission. A second objective is to review
studies on landfill CH$_4$ emissions, using one or more of the measurement techniques, and then
compare and reflect on these studies to enable the reader to choose the best method for his/her
application.

2. Methane measurement techniques

The different measurement techniques available for quantifying CH$_4$ emissions from
landfills all measure against different scales. Measurements are made from the landfill surface,
to several kilometres away (Fig. 1) and across different timescales, from minutes, to weeks or
months. Measuring at the surface of the landfill has the advantage of excluding interference
from surrounding CH$_4$ sources, but it also involves the challenge of having to extrapolate the
measurements to represent a large area with heterogeneous emissions, as it is practically
impossible to cover the whole landfill area. Methods measuring at elevated heights are able to
evaluate emissions from a larger area, thereby making extrapolation to full landfill size more
applicable, while methods measuring at a greater distance away are able to quantify whole-site emissions, though they are more sensitive to potential surrounding sources. Table 1 provides an overview of qualitative reconnaissance techniques used to locate CH\textsubscript{4} emission hotspots, while Table 3 provides an overview of the various CH\textsubscript{4} measurement techniques employed for emission quantification at landfills. In the following sections, each technique is described in detail and its advantages and limitations are discussed.

2.1 Qualitative reconnaissance techniques

Methodology. A common reconnaissance technique involves screening the landfill surface for CH\textsubscript{4}, using a handheld gas analyser (such as a portable flame ionisation detector (FID), or other analysers based on optical or electrochemical measurement principles, gauging concentrations in ppm just above the landfill cover. A screening campaign can be performed by dividing the landfill into sections, whereby CH\textsubscript{4} recordings are taken systematically at grid points or alternatively during walkovers (e.g. Scheutz et al., 2011a; 2011c). A survey of CH\textsubscript{4} concentrations may give an indication of certain hotspots or installations with CH\textsubscript{4} leakages. In the US, landfill operators are legally obliged to carry out CH\textsubscript{4} surface screenings four times a year, and this is also mandatory in the United Kingdom. Surface CH\textsubscript{4} screenings can be extended to include carbon dioxide (CO\textsubscript{2}) concentrations, for example by using a handheld infrared (IR) detector or a portable cavity ring down spectrometer (CRDS), the latter of which has the advantage of being fast and able to measure concentration changes in sub-ppm levels. Recent developments of the method have made it portable (e.g. Juncher Jørgensen et al., 2015). Elevated CO\textsubscript{2} to CH\textsubscript{4} concentrations may indicate areas experiencing CH\textsubscript{4} oxidation (confer section 2.3).

Theoretically, surface air concentrations recorded at a constant and short distance away from the soil/atmosphere interface should be positively correlated with surface fluxes. However, this
correlation depends on interactions between the measuring device (e.g. pumping flow), the ground (e.g. soil characteristics, vegetation) and the atmosphere (weather conditions) – and thus it is not constant in time and space. Lando et al. (2017) found that landfill surface CH$_4$ concentrations recorded using an open-path handheld laser detector were inversely proportional with wind speed, and thus the authors recommended that surface concentration measurements should be performed at wind speeds < 1 m s$^{-1}$. It has been suggested that CH$_4$ flux (after proper calibration) can be quantified from simple measurements of CH$_4$ concentrations close to the landfill surface (Lando et al., 2017; Gonzalez-Valencia et al., 2016; 2015; Park et al., 2016; Mackie and Cooper 2009). However, CH$_4$ flux vs. surface concentration measurements often show relatively weak and varying correlations, depending on the measuring time and location, and they are thus associated with large uncertainties (Lando et al., 2017; Gonzalez-Valencia et al., 2016; 2015; Franzidis et al., 2008).

Other methods for identifying CH$_4$ emission hotspots include the use of thermal infrared cameras that measure temperature differences (thermal anomalies) (Fjelsted et al., 2019; Tanda et al., 2017; Battaglini et al., 2013; Lewis et al., 2003). Areas with elevated temperatures may be caused by the leakage of landfill gas, which is warmer than the surroundings. Thermal infrared photography is best carried out at times when there are large temperature differences between the landfill gas and the surroundings, e.g. in the early morning when the air is cold. However, a number of factors, other than emissions of warm LFG, can cause temperature differences and result in measurement bias, which was thoroughly investigated in Lewis et al. (2003). Warmer areas in the surface can be caused by high biological activity, e.g. soil or compost respiration or CH$_4$ oxidation. Measurement bias can also be caused by variability in emissivity of different surfaces. Different vegetation and soil conditions on the surface, having different emissivity, will cause temperature anomalies in thermal infrared images. As an example, dark areas, such as soil surfaces without vegetation or a dark plastic liner, will have higher heat absorption, and thus they will appear as
warm areas in an infrared image and result in a false indication of a CH$_4$ emission. Finally, a certain minimum CH$_4$ flux can be expected before a temperature anomaly becomes detectable. Fjelsted and co-workers (2019) concluded that the minimum observable CH$_4$ emission for the thermal infrared camera to identify an emission hotspot was 150 g CH$_4$ m$^{-2}$ d$^{-1}$ occurring from an area of more than 1 m$^2$. In combination, these precautions most likely explain why studies presented in the literature remain somewhat inconclusive on the usefulness of thermal infrared technologies for identifying landfill CH$_4$ emissions (e.g. Fjelsted et al., 2019; Capodici et al., 2015; Beaumont et al., 2014; Battaglini et al., 2013; Desideri et al., 2007; Ishigaki et al., 2005; Raco et al., 2005; Lewis et al., 2003).

Both thermal infrared cameras and lasers capable of measuring CH$_4$ could be integrated with unmanned aerial vehicle (UAV) technology to overfly the landfills (e.g. Fjelsted et al. 2019; Tanda et al., 2017; Beaumont et al., 2014).

Finally, a visual inspection of the landfill surface is a simple and useful method, in that cracks in the surface, vegetation damage and damage to the gas collection system are potential areas of CH$_4$ emissions. Table 1 provides an overview of qualitative reconnaissance techniques that can be used to study areas with CH$_4$ emissions.

**Advantages and limitations.** The advantages of the qualitative reconnaissance techniques described herein are generally that they provide an opportunity for a relatively fast, easy and inexpensive overview of areas or installations with CH$_4$ emissions that can subsequently be repaired. Analytical devices are often easy to employ and the measurements can be performed by landfill staff. Qualitative reconnaissance techniques are thus valuable tools in the daily operation and maintenance of the landfill cover and of site installations such as the gas collection and recovery systems. However, it must be emphasised that none of the reconnaissance techniques provides emission measurements in standard units for mass flux such as, for example, mass per area...
per time. At best, for instance using a field FID, they provide a semi-quantitative determination of atmospheric CH$_4$ concentrations near the ground surface. It should also be noted that relatively low readings in the air above the cover layer are not necessarily an indication that there is no emission, and so surface CH$_4$ fluxes can thus not be determined based on above-surface CH$_4$ concentration readings. Where landfill surface CH$_4$ concentrations are high, it may be possible to combine the field FID with a simple surface chamber apparatus for the direct measurement of fluxes (confer section 2.3). Similarly, thermal infrared photography can be coupled with surface flux measurements (e.g. Fjelsted et al., 2019; Capodici et al., 2015; Battaglini et al., 2013).

2.2 Vertical soil gas concentration profiles

Methodology. Vertical soil gas concentration profiles in the landfill top cover are obtained by inserting soil gas probes, with slits at the lowest part of the probe for the gas to enter, at different depths of the cover and, if necessary, in the upper part of the waste. Gas samples are withdrawn from the probes for determining pore gas composition (CH$_4$, CO$_2$, oxygen (O$_2$) and nitrogen) in the cover layer. Soil gas profiles can provide useful qualitative information on gas transport and distribution (Scheutz et al., 2008; 2003; Christophersen and Kjeldsen, 2001; El-Fadel et al., 1995), dominant gas transport processes (diffusive vs. convective transport) and CH$_4$ oxidation (Röwer et al. 2011; Gebert et al., 2011; Scheutz et al., 2011a; 2001c; Christophersen et al., 2001; Jones and Nedwell, 1990). As an example, CH$_4$ and CO$_2$ concentration gradients of the cover layer can provide information on whether CH$_4$ oxidation is occurring and at what depth it takes place (Scheutz et al., 2009; 2008; 2003). CH$_4$ and CO$_2$ gas concentration profiles have also been used to quantify CH$_4$ oxidation (Gebert et al., 2011). The methodology was developed and presented by Gebert et al. (2011) and has been used in a few emission studies (Gonzalez-Valencia et al., 2016; Pratt et al., 2013; Kjeld, 2013).
It is important to be aware, however, that processes other than microbial CH$_4$ oxidation contribute to CO$_2$ generation, e.g. soil respiration, and thus the method could lead to an overestimation of the CH$_4$ oxidation. It is therefore advised that background measurements (measurements performed in areas or soils without landfill gas exposure) are performed in order to assess background soil respiration. In some settings such as compost-based landfill covers, CO$_2$ generation caused by compost respiration can be a significant or even the dominant CO$_2$ generating process, and the method will thus not be applicable for CH$_4$ oxidation evaluation.

Vertical soil gas concentration profiles can also be used in combination with soil sampling at the respective cover depths, and following subsequent incubation in the laboratory (by adding CH$_4$ and O$_2$ and subsequently measuring CH$_4$ turnover) the CH$_4$ oxidation potential of the cover soil can be determined. Incubation tests can be performed in different conditions, such as testing for temperature and moisture content, which can provide a better understanding of the influence of weather and climate conditions on CH$_4$ oxidation (Scheutz et al., 2009; 2004).

Vertical soil gas concentrations combined with pressure gradient measurements in landfill covers can be used to calculate the flux of CH$_4$ through the landfill cover (El-Fadel et al., 1995; Jones and Nedwell, 1990; Young, 1990). The advective flux can be calculated using Darcy’s law, i.e. $u = -K \cdot \frac{dP}{dz}$, where $u$ is the mass flux of gas (in m$^3$ s$^{-1}$ m$^{-2}$), $K$ is the medium’s permeability for gas flow (in m$^2$ s$^{-1}$ Pa$^{-1}$) and $dP/dz$ is the pressure gradient at depth $z$ (in Pa m$^{-1}$) (Young, 1990). Often, CH$_4$ flux will be governed by both advection and diffusion, where diffusive flux is driven by concentration differences. In such cases, Fick’s first law of diffusion should be included, which requires knowledge of a number of factors such as diffusivity, porosity, water content and gas viscosity (Kjeldsen, 1996; El-Fadel et al., 1995).
For a more correct description of the diffusion process, Stefan Maxwell equations may be required instead of Fick’s first law in case gas compositions vary significantly in the modelled soil domain (Molins et al., 2008).

Advantages and limitations. Vertical soil gas concentrations combined with pressure gradient measurements have only been used a few times for evaluating a whole landfill’s CH$_4$ emissions (Bogner et al., 1995); however, the method has been used for evaluating the influence of atmospheric pressure on CH$_4$ emissions (El-Fadel et al., 1995; Young, 1990). The technique lacks spatial (small sampling area of < 0.5 m$^2$) and temporal resolution (samples are taken within 10-20 minutes), which is in conflict with the often heterogenic emissions escaping from landfill surfaces. Emissions from hotspots, such as cracks in the top cover, or from leachate wells cannot be captured using the gas concentration profile method. Additionally, the method is very labour-intensive. Its main advantage, though, is the ability to obtain useful information on mechanisms influencing the emissions, such as rapid changes in atmospheric pressure, the effect of wind velocity and relative emissions as the result of convection and diffusion. Soil gas profile measurements can also be useful in understanding CH$_4$ sources and sinks, and they can therefore be useful for evaluating CH$_4$ oxidation (Röwer et al. 2011; Gebert et al., 2011; Christophersen et al., 2001) and landfill gas migration (Christophersen and Kjeldsen, 2001; El-Fadel et al., 1995). Christophersen et al. (2001) studied lateral gas migration and oxidation at an old, completely and finally covered landfill, using gas profiles combined with static flux chambers, finding seasonal variations with no CH$_4$ emissions during summer, due to increased oxidation in the soil. They also found that atmospheric pressure, temperature and especially soil moisture content had an influence on CH$_4$ emissions, while off-site migrating CH$_4$ was oxidised before entering into the atmosphere.
In summary, vertical soil gas concentration profiles have limited applications as a standalone method for quantifying entire landfill CH$_4$ emissions. The method has only been used for CH$_4$ flux quantification in a few studies dating back to the 1990s, but it can nevertheless provide valuable information about landfill gas migration, oxidation and influencing factors (e.g. changes in weather conditions), and this supporting information could be useful in combination with other quantitative emission methods.

2.3 Surface flux chambers

The main idea behind the flux chamber is that gas escaping from the surface of a landfill site is captured inside the chamber, and changes in concentrations therein are then measured. Based on the rate of concentration change, the emission flux can be calculated. Many different variations of the flux chamber have been developed, but in general it has two basic setups, using either an open or a closed chamber. Fig. 2 shows the concepts of the open and closed chamber methods, including the basic equations used for calculating surface flux in each case. Table 2 provides an overview of the characteristics of the two types of chambers.

2.3.1 Closed surface flux chambers

Methodology. Closed flux chambers – also referred to as ‘static chambers’ – are historically likely to be the most commonly used techniques for estimating CH$_4$ emissions from landfills. These closed chambers can be set up with or without recirculating the analysed gas flow. The recirculation mode is preferably used if the CH$_4$ analyser withdraws a significant gas volume from the chamber, in order to avoid errors on the flux measurement by removing gas as part of the sampling activity. The chamber technique was originally developed to measure gas emissions from natural soils (e.g. Rolston, 1986), where surface emissions are
controlled by diffusion and happen more homogeneously, and so minimal emission variations are expected over the emission area. The CH\textsubscript{4} emission flux from the surface covered by the chamber is calculated from measuring changes in CH\textsubscript{4} concentrations inside the chamber:

\[ Flux = \frac{dC}{dt} \cdot \frac{V}{A} \]  \hspace{1cm} \text{Eq. 1}

where \( \frac{dC}{dt} \) is the change in concentration over time (mole m\textsuperscript{-3} h\textsuperscript{-1}), \( V \) is the volume of the chamber (m\textsuperscript{3}) and \( A \) is the surface area covered by the chamber (m\textsuperscript{2}).

The closed flux chamber has been used widely for quantifying gas emissions from landfill surfaces, where emissions are controlled by both pressure and concentration gradients (e.g. Lucernoni et al., 2016; Rachor et al., 2013; Abichou et al., 2011; Bogner et al., 1995; Kjeldsen and Fisher, 1995). Different chamber configurations and sizes have been used, from small-size (chamber footage area less than 0.1 m\textsuperscript{2} e.g. Scheutz et al. 2014; 2011) and mid-size (0.4-1 m\textsuperscript{2} e.g. Christophersen et al., 2001), to over-size (larger than 15 m\textsuperscript{2} e.g. Geck et al., 2016). A theoretically optimal chamber design was suggested by Lucernini et al. (2016) based on computational fluid dynamics simulations. In many cases, the chambers have been equipped with mechanical or electrical fans to assure the complete mixing of gases inside the chamber during sampling. In order to take repeated measurements at the exact same locations, chambers can be supplemented with a frame that is permanently installed in the landfill cover, topped by a removable lid (Pratt et al., 2013; Schroth et al., 2012; Christophersen et al., 2001). In order to minimise errors caused by increased pressure and concentration build-ups inside the chamber, measurements are often done over a very short time period, and preferably no more than a few minutes. A pressure build-up can be avoided by installing a vent consisting of a long small-diameter tube, which can equalise pressure between the chamber and the surrounding air.
Losses due to diffusion along the vent tube have been considered negligible (Hutchington and Mosier, 1981). However, due to the venturi effect, vents may potentially introduce more errors than they solve. Vented chambers have been used in a CH$_4$ emission study performed on an old landfill (Rachor et al., 2013).

Due to its easy setup and inexpensive use, the chamber technique has been suggested for measuring entire landfill emissions (UK Environment Agency, 2010). To obtain a whole-site CH$_4$ emission rate, measurements are sometimes done by using randomly distributed chambers or a systematic grid of single-point measurements, following which emissions for the entire site are found by calculating an average of the measured emissions multiplied by the landfill area, or by using geo-statistical tools (e.g. Gonzales-Valencia et al., 2016; Battaglini et al., 2013, Ishigaki et al., 2005).

Advantages and limitations. The surface flux chamber method is very simple to employ and does not require advanced analytical equipment; CH$_4$ fluxes, for example, can be measured with an FID or an IR detector. The method is able to detect small fluxes of CH$_4$ – in most cases positive (CH$_4$ release), though they can also be negative (uptake of atmospheric CH$_4$). The surface flux chamber method is the only technique currently applicable for determining the flux of trace gases contained in LFG (non-methane volatile organic compounds; NMVOCs) (Scheutz et al., 2008; 2003). The flux chamber method, sometimes combined with soil gas profile measurements, is a well-proven and appropriate method for quantifying CH$_4$ oxidation at a specific location on the surface (using the carbon mass balance method developed by Christophersen et al., 2001), or for studying environmental factors controlling CH$_4$ emissions and oxidation. As mentioned in section 2.2., using CO$_2$ surface fluxes to quantify CH$_4$ oxidation could result in overestimations, and the CO$_2$ flux contribution from soil or compost respiration should thus be determined either by performance of background flux measurements.
or by laboratory soil/compost incubation studies (Scheutz et al., 2017). Alternatively, by measuring the stable carbon isotope composition of CH$_4$ in the flux chamber, in-situ CH$_4$ oxidation can be determined (Chanton and Liptay, 2000; Liptay et al., 1998). However, the use of stable carbon isotopes to quantify CH$_4$ oxidation should be interpreted with caution, as the isotopic fractionation during microbial oxidation is influenced by a number of factors, including temperature, soil moisture, CH$_4$ and O$_2$ supply (e.g. Gebert et al., 2017; Chanton et al., 2008b). Additionally, processes other than microbial activities contribute to isotopic fractionation, such as transport processes (Gebert et al., 2013). Several studies have used flux chambers to observe, for example, the influence of changes in atmospheric pressure, precipitation or temperature on the emission of CH$_4$ (e.g. Fredenslund et al. 2010; Gebert and Gröngröft, 2006; Czepiel et al., 2003; Christoffersen and Kjeldsen, 2001; Kjeldsen and Fischer, 1995).

One of the limitations of the surface flux chamber is that it is intended for measuring diffusional surface fluxes. At landfills, surface CH$_4$ emissions are governed by both diffusion and advection, the most dominant of which is site-specific and can even vary between different areas of the landfill, e.g. hotspots versus non-hotspots. At sites (and/or during specific time periods) where the flux is controlled mainly by pressure differences over the landfill cover, surface flux can be underestimated, due to a fast pressure build-up in the static chamber (Seneviratna et al., 2007; Conen and Smith, 1998).

The main limitation of using closed surface flux chambers is that the measured emission represents only the small area covered by the flux chamber itself (often 0.5-1 m$^2$), and it only captures the emission over a short period of time (minutes). In this way, the method has the same spatial and temporal limitations as the pore gas concentration profiles mentioned in the previous section. In an attempt to obtain a reliable estimate of the total CH$_4$ emission
from a landfill or section thereof, it is often proposed to apply a systematic testing strategy (Gonzalez-Valencia et al., 2015; Battaglini et al., 2013; Bour, 2007; Spokas et al., 2006; Lang, 2004; Rosevaer et al., 2004; Savanne et al., 1997; Bogner and Scott, 1995). Such a sampling strategy may be that the landfill is divided into grids of a certain size, followed by flux chamber measurements performed in the grid points. Geo-statistic models can then be used to estimate total CH$_4$ emissions from the landfill (Gonzalez-Valencia et al. 2015; Spokas et al., 2003; Börjesson et al., 2000). Typical distances between the grid points are between 10 and 60 metres, and the more measurements performed, the more accurate a result can be expected.

Back in the 1990s, the UK Environment Agency developed a best practice guideline on how to measure total CH$_4$ emissions from a landfill by creating a network of flux chamber measurements and integrating individual flux measurements (UK Environment Agency, 2010). Large numbers of measurements are needed in this regard, and the UK EPA suggests these should be based on the size of the landfill area (or the area to be investigated), with a minimum of six measurements and an additional number of measurement points equal to 0.15 times the square root of the area (in m$^2$) (UK Environment Agency, 2010). This results in 16 measurement points for a 5000 m$^2$ site, 53 points for 100,000 m$^2$ and 156 points at 1 million m$^2$. However, even when performing a very large number of measurements, the measured CH$_4$ emission will only represent what has leaked out from a very limited area (<1%) of the total landfill. In addition, a significant part of the emitted CH$_4$ occurs through small hotspots, as demonstrated by Rachor et al. (2013), and the likelihood of capturing one of these hotspots with a grid of chamber measurements is very small; thus, underestimating total landfill emission is to be expected from the surface flux chamber method (Scheutz et al., 2011c; 2011b; Babilotte et al., 2011; Green et al., 2010; Fredenslund et al., 2010; 2007; Goldsmith et al., 2008).
One option for improving the chamber method is to combine flux chamber measurements with qualitative screening methods, in order to identify hotspots (see section 2.1) and subsequently perform more flux chamber measurements in these areas. Here, however, it is important that emissions from the hotspot areas are weighed correctly with the other measurements, in order to obtain a correct average. Even though measuring devices that can log CH$_4$ concentration over short time intervals (seconds) are currently available, it is very time-consuming to perform flux chamber measurements, due to the large number that need to be performed. A proper measurement often requires that vegetation is cut down and that the chamber’s contact with the surface is sealed with, for example, bentonite. If there is very high vegetation on the cover layer, it may in fact be a major challenge to perform flux chamber measurements, and so a campaign covering a whole landfill site will therefore often take from two to five days. During this period, changes in weather conditions (atmospheric pressure, temperature, precipitation, etc.) may cause significant changes in emissions, which may in turn complicate any interpretation of the results relative to what the measured emission represents.

Finally, surface flux chambers cannot be used to measure emissions from leaks in the gas and leachate collection system (risers, pumping stations, extraction and leachate wells), which can be significant routes for landfill gas leakages (Scheutz et al., 2011c; Fredenslund et al. 2010).

### 2.3.2 Open surface flux chambers

**Methodology.** The open flux chamber method – also referred to as the ‘dynamic chamber method’ – is similar to the closed method, albeit the chamber is continuously flushed with air, to avoid possible errors caused by pressure and concentration build-up as a result of surface emissions and to enable measurements over longer periods of time (e.g. Tregoures et al., 1999). The emission rate is subsequently calculated by using flow through the chamber and concentrations at the inlet and outlet, as shown in Fig. 2. It is important to maintain pressure in
the chamber comparable with ambient pressure, in order to obtain reliable emission rates. Similar to the closed chamber, an open chamber can be equipped with a vent to equalise pressure between the chamber and the surrounding air. Guidance on how to use both closed and open flux chambers was published by Eklund (1992). Open flux chambers have also been used in a few landfill emission studies as an alternative to the closed chamber approach, to measure CH$_4$ emissions through landfill covers (Gallego et al., 2014; Park and Shin, 2001; Reinhart et al., 1992).

*Advantages and limitations.* The advantage of the open flux chamber is that it is continuously flushed with air, which limits the risk of pressure build-up and gas concentration accumulation underestimating the measured flux. Pressure built up inside the chamber will affect the flux through advection, whereas an accumulation in gas concentrations inside the chamber will affect the diffusional flux. In both cases, this can result in non-linear gas concentration accumulation curves, which will lead to underestimating the surface flux. In addition, using an open flux chamber allows measurements over a longer period of time (e.g. Huber-Humer and Lechner, 2001a; 2001b; Tregoures et al., 1999; Verschut, 1991). The flow through the open chamber (also referred to as the ‘carrier gas’) can consist of ambient air, or alternatively a synthetic gas can be used. If ambient air is used, it is important that the background concentration of the gaseous compound being analysed is constant. At landfill surfaces, CH$_4$ background concentration will often not be constant, which is why it may be necessary to use a synthetic gas as a carrier when using open chambers. In comparison to closed chambers, the set-up for open chambers is more demanding in terms of pumps required to control the flow through the chamber and potentially synthetic gas containers, which will also make it more time-consuming to perform a flux measurement. Regarding the temporal and spatial challenge, open flux chambers have the same advantages and disadvantages as closed
flux chambers, and if employed at landfill sites with emission hotspots, one will likely underestimate entire landfill site emission.

In summary, heterogeneous emissions from landfills make it difficult to quantify total CH$_4$ emissions using the flux chamber approach. In spite of several studies using the approach to quantify whole landfill site emissions, this approach remains unvalidated in terms of its appropriateness for estimating accurately total CH$_4$ emissions, and thus there is a need for further documentation (including appropriate numbers of measurements and statistical methods for data interpretation) and testing against other measuring methods. Currently, flux chambers are not recommended for whole landfill site CH$_4$ emission quantification. However, flux chambers can be an important tool for obtaining emission rates for smaller areas, such as an engineered biocover for CH$_4$ oxidation (e.g. Scheutz et al. 2017; 2011b; Geck et al., 2016) or smaller scale landfill cover studies (e.g. covered vs. non-covered cells, slopes vs. none-sloped area, etc.) (Wang et al., 2017; Di Trapani et al., 2013; Scheutz et al. 2008).

### 2.4 The eddy covariance method

Methodology. Micrometeorological eddy covariance measurement relies on the fact that emitted gases are mixed vertically by turbulent eddies in the atmospheric inversion layer. The method is often used for measuring emissions from larger surfaces such as fluxes of CH$_4$, N$_2$O and CO$_2$ from agricultural soils (e.g. Kroon et al., 2010; Hensen et al., 1996), but it has also been applied on landfills (Xu et al., 2015; McDermitt et al., 2013; Schroth et al., 2012; Eugster and Plüss, 2010; Lohilla et al., 2007; Laurila et al., 2005). In a micrometeorological eddy covariance setup, continuous measurements of the vertical gas mass fluxes of CH$_4$, CO$_2$ and water are often carried out (Fig. 3). The vertical flux of CH$_4$ at this height takes place mainly through convection, and any emissions can therefore be found from CH$_4$ concentrations and...
local vertical wind velocities. Due to air turbulence, the wind will sometimes have an upward component and at times a descending component. In the event that there is an emission from the surface, the concentration of CH$_4$ through the plane will be slightly higher when the wind is rising, and so an average positive CH$_4$ flux (in unit mass per time) can be measured. CH$_4$ concentrations and vertical wind velocities are measured continuously and at high frequency (~10 times per second). The measured data are then stored and the average CH$_4$ emission is calculated for time intervals, for example of 15 minutes.

Measurements are taken on a tower with enough distance to the surface to cover the desired surface area. Typical heights will range between 0.5 and 10 metres, and the contributing emission area will (when the measuring time is long enough to capture wind directions from all angles) have a radius of approximately 100 times the height of the measurement point and form a drop shape with the tower located at the top of the drop (McDermitt et al., 2013).

The source area is estimated by using a dispersion model, and total emissions from the landfill are calculated by applying the obtained emissions per area to the total landfill area.

Advantages and limitations. One of the advantages of micrometeorological eddy covariance measurements is the possibility of having continuous measurements over longer time periods, as well as measuring both CH$_4$ and CO$_2$, depending on the analytical device. The method can run automatically for months, providing insights regarding temporal variability as well as average emissions. Xu et al. (2015) measured the response of CH$_4$ emissions to change in barometric pressure over a six-month period. Combined with other information such as landfill gas composition and gas recovery, the method can provide an indication of total landfill gas production and CH$_4$ oxidation (Xu et al., 2015; Lohila et al. 2007; Scharff et al., 2003). However, a comparison of CH$_4$ to CO$_2$ ratios obtained from raw landfill gas
composition analysis, with ratios based on surface flux measurements, should be done with caution, as soil respiration also results in CO₂ surface fluxes. Similar to microbial CH₄ oxidation, soil respiration depends on temperature and will thus vary over the season.

The main disadvantage of the micrometeorological eddy covariance method is the limited surface area it covers, meaning that it most likely does not provide representative emissions for the whole landfill site. In addition, the covered area changes in line with changing wind speed, and the often challenging typography of a landfill is far from the flat surface that offers the ideal conditions for applying this method. Despite the challenges of typography and heterogeneous emissions, the micrometeorological eddy covariance method is considered by some as a suitable estimation technique for measuring CH₄ emissions from landfills (Xu et al. 2015; Lohilla et al., 2007; Laurila et al., 2005), although Tregoures et al. (1999) found that it was not suitable for landfill gas emission quantification. From the comparison studies, it can be concluded that eddy covariance methods can be applied to landfills with relatively homogeneous emissions and flat surfaces. Lohilla et al. (2007) compared their eddy covariance measurements with closed chamber measurements and found good agreement, but closed chamber methods face the same challenges with heterogeneous emissions, and it is therefore questionable if the eddy covariance method can be validated by using chamber methods as a comparison.

2.5 The stationary mass balance method

Methodology. In the stationary mass balance method, CH₄ concentrations are measured at different heights above the landfill surface (Fig. 3). By simultaneously measuring wind velocity and comparing it with the measured concentrations, the horizontal flux of CH₄ can be calculated, which represents emissions from the landfill area upwind of the sampling point.
The method is also called the ‘1D mass balance method’, as it measures CH$_4$ flux that passes the target tower (1D) and not through a larger vertical plane (2D) as in other mass balance methods (see Sections 2.6, 2.7 and 2.9). It is important that the measured CH$_4$ concentrations are corrected for any background concentration of CH$_4$, as the CH$_4$ flux will otherwise be overestimated. This is done by measuring at the top of the tower, assuming that the background concentration has the greatest influence on the measured CH$_4$ concentration at this point. The height of the measurement point determines the source area contributing to elevated CH$_4$ concentrations, and when measurements are taken at a sufficient height above the landfill, in theory, the whole CH$_4$ plume is measured and the total CH$_4$ emission flux is obtained (Scharff, 2003). Normally, however, this requires a very high tower, and in practice a lower tower is used, thereby obtaining data from a smaller part of the landfill. By performing measurements at a maximum height of 10 metres, emanations from an area with a radius of approximately 150 metres away from the tower will be covered (Scharff, 2003). The specific location and size of the emission area contributing to the measured emissions is determined by dispersion modelling, for which knowledge about atmospheric stability and the surrounding topography is required. Emissions from all directions are measured as the wind direction changes, thus enabling the method to provide emission rates from different areas of the landfill. It can be set up for automated, continuous measurements over a longer period, thereby enabling long emission data time series (weeks to months).

**Advantages and limitations.** The mass balance method shares many of the same advantages and limitations as the eddy covariance method. It can be automated so that emissions (CH$_4$ and other gases CO$_2$ and N$_2$O) from the landfill can be measured over a longer period of time, providing information on the variability of the CH$_4$ emission over time and partly providing a better determination of the average emission. Experiences with landfill
emission quantification using the stationary mass balance method are very limited. The method was used for CH$_4$ quantification at landfills in the Netherlands (Oonk and Boom, 1995) and was found to give similar emission fluxes in a comparison study with an inverse modelling method, along with tracer gas calibration (Scharff, 2003) (Table 3).

A major drawback is that the method does not measure emissions from the entire landfill but only from a small part thereof. In addition, it can be difficult to determine exactly which part of the landfill has contributed to the measured emission, so any results thus depend on assumptions about where CH$_4$ emissions take place. If emissions occur mainly closer to the measuring tower than assumed, the overall emission will be overestimated. It is therefore advisable to perform a qualitative CH$_4$ screening of the landfill surface, for example using an FID (see section 2.1), to evaluate any relevant areas before a final interpretation of the measured flux is made.

2.6 Radial plume mapping

Radial plume mapping (RPM) for measuring landfill CH$_4$ emissions has been under development since the late 1990s. The method uses a combination of concentration measurements and wind profiles to obtain a surface emission factor from an upwind area. It can be used in two different configurations – horizontal (HRPM) or vertical (VRPM) (Fig. 4), with HRPM providing qualitative information on the location of emission hotspots, while VRPM is used for quantifying emissions.

Methodology. Quantifying emissions by VRPM is achieved by measuring the mass of CH$_4$ crossing two vertical planes located upwind and downwind of an emission area, respectively. CH$_4$ fluxes across the vertical planes are determined from the product of concentrations in the air and wind velocity measured at each point in the two planes, and the
approach is thus considered a mass balance method. CH₄ emissions from the area are found by subtracting the flux measured through the upwind plane from the flux measured through the downwind plane. If the concentration in the upwind plane (i.e. background CH₄ concentration) can be assumed to be constant over both the plane itself and over the measurement period, the method can be simplified and the CH₄ flux can be calculated from the product of wind velocity and the difference between the upwind and downwind CH₄ concentrations. Multiple laser beam paths are used by pointing a laser beam at reflectors positioned at different heights (and at different distances to the laser across the plume) (Fig. 4). The laser and reflectors are placed as close to the landfill as possible or to the landfill area to be measured. Each laser beam path provides an average CH₄ concentration, representing the beam’s air package. Laser beam paths are typically up to 300 m long, and a typical maximum tower height for the vertical reflectors is 10 m. Based on the measured average concentration paths, a cross-section concentration profile of the plume is modelled and a two-dimensional (2D) concentration profile is obtained, hence the name ‘plume mapping’. This modelling step has a series of assumptions about the distribution of the plume, which adds to the level of uncertainty. Using the measured concentrations and modelling, the method provides information about the 2D distribution of CH₄ concentrations in the vertical plane of the plume (Goldsmith et al., 2012; Hashmonay et al., 2008; Hashmonay et al., 2001; 1999; Wu et al., 1999). The area contributing to the measured CH₄ flux is confined by the upwind and downwind measuring planes. If a setup with only one laser is used, the area contributing to the measured CH₄ flux across the vertical measurement plane is determined by reverse dispersion modelling.

**Advantages and limitations.** The limitations of the method are that the measured CH₄ flux normally represents only one part of the landfill (due to the limited range of the laser) and that it is difficult to determine the contributing area precisely. In order to determine emissions
from the entire landfill, several measurements must be taken, or alternatively it must be assumed that the measured emission is representative of the entire landfill and can then be summarised to determine total emissions. A methodological challenge in this regard is that both wind speed and CH$_4$ concentration may vary over the measured plane. Thus, ideally, measurements should be carried out simultaneously at different places in the upwind and downwind planes, which require additional lasers, thereby adding considerably to the cost of the instrumentation and the demand for staff. When using one laser, the area contributing to the measured CH$_4$ flux through the vertical plane is determined by inverse dispersion modelling, thus adding further to the uncertainty of the method. Determining the correct area contributing to the measured emission can be a challenge on landfill sites with complex topography (Abichou et al., 2010; Goldsmith et al., 2008), and at some landfills (or parts of landfills) the method cannot actually be applied. In many sites, part of the waste is deposited above ground, forming a small hill in the landscape. If measurements are carried out such that the plane through which the CH$_4$ flux is measured is placed on top of the landfill, any emission from the slope will not be included in the measurement, which may lead to a significant underestimation of emissions, as hotspots are often seen on slopes, due to horizontal gas transport and incomplete covers (Babilotte et al., 2009; 2008). Likewise, when reflectors and lasers are placed on the downwind edge of a landfill, CH$_4$ emitted from an upwind part of the landfill may be at an elevation higher than 10 metres (often the maximum position above ground of the reflector) when it reaches the measurement points, which will add significantly to the uncertainty of the method. An additional disadvantage is that measurements are usually taken in hours, and thus any temporal emission variation can be a challenge to capture. Another drawback is that the method (theory, equipment and application) is generally difficult
to understand for third parties, making it difficult for an independent body to assess the results of emission studies performed by this method.

The US-EPA has proposed the method for quantifying CH₄ emissions from landfills and given it the name ‘Other Test Method-10’ (OTM-10). In its development, more than 1000 individual tracer gas measurements were conducted across 61 experiments. Capture efficiency was determined as a function of wind speed (WS) and wind-adjusted release distance (WARD). Thoma et al. (2010) calculated a simple equation for the capture efficiency factor, namely CEF = 0.712 – 3.10×10⁻³(WARD) + 0.102(WS), and reported that the maximum upwind distance included in the measured area was 295 m at a wind speed of 2 m s⁻¹ and 460 m at 7 m s⁻¹.

The method has been tested and used for quantifying 20 landfills in the USA, using a double laser and reflector system (Goldsmith et al., 2012; Abichou et al., 2010). Abichou et al. (2010) found that a change in wind direction could introduce an uncertainty factor of 20% and that the area contributing to the measured flux is a function of the atmospheric stability class with an uncertainty level of 10-30%. Goldsmith et al. (2012) pointed out that the OTM-10 method may have an application for the landfill industry, although only 31% of the collected data in their comprehensive study of 20 landfills in the USA fulfilled quality requirements. The method is challenged by the complex topography of a landfill, and there are sites where the method is simply not applicable. In general, the output from RPM can be used to identify emission hotspots, which makes this method useful for performing site surveys before initiating remediation activities; however, it is highly important to assign the correct emission area to the measured emission, which can be difficult at landfills with a challenging topography (Goldsmith, 2008). The upshot of the disadvantages of the method is that it has
been used infrequently in recent years and replaced by other methods (tracer gas dispersion, DIAL and most recently aircraft-based mass balance).

2.7 Mass balance using aerial measurements

Methodology. In recent years, the mass balance approach has been applied using atmospheric CH$_4$ concentration measurements from aircrafts or UAVs. The method relies on concentration measurements across the downwind plume at several heights, preferably covering the total height and width of the CH$_4$ plume. The measured concentrations are then used for creating a 2D concentration plane, which is combined with measured wind speed and direction for calculating the flux of CH$_4$ through the downwind plane – and thus emissions from the landfill. Fast measuring instruments are needed for performing aerial CH$_4$ measurements with sufficient temporal resolution, to create a 2D concentration plane. Such instrumentation includes cavity ring down spectroscopy, quantum cascade laser spectroscopy and thermal-infrared (TIR) imaging spectrometry (Cambaliza et al., 2017; 2015; Tratt et al., 2014; Hirst et al., 2013; Peischl et al., 2013). Measurements using aircraft are usually carried out several kilometres downwind of the landfill, as the aircraft can carry instrumentation sensitive enough to measure concentration differences (plume concentrations above background) at this distance. UAV-mounted instrumentation needs to be lighter in weight. Such instrumentation currently has less sensitivity, and the UAV must therefore measure closer to the landfill than aircraft (Allen et al., 2018). However, the fast development in instrument/sensor technology might soon change this situation.

Advantages and limitations. One advantage of the method is that aerial measurements do not require access to a landfill. When using an aircraft, each landfill is measured relatively quickly, and so many landfills (and other CH$_4$ sources) in an area can be measured in one
flight. An advantage compared to VRPM and DIAL is that CH$_4$ concentrations at specific points in the total cross-section of the CH$_4$ plume can be measured, resulting in a relatively detailed 2D concentration plane. The advantage of the using aircraft for CH$_4$ plume measurements compared to using UAVs is that measurements performed further downwind of the landfill will give a more uniform and less changeable CH$_4$ plume, which is easier to model correctly, as the measured concentrations can better be assumed to be constant in comparison to when measurements are performed closer to the landfill, where local wind turbulence and emission variations will result in larger concentration variations at individual x, y and z points in the atmosphere. However, measurements taken far away from the landfill also introduce possible errors, due to reduced CH$_4$ plume concentrations and to the risk of including CH$_4$ for surrounding non-landfill emission sources.

One limitation is that it is not possible to quantify individual CH$_4$ sources relatively close to each other, such as farms or wastewater treatment plants near to the measured landfill. Additionally, the creation of the 2D concentration plane is associated with uncertainty, as well as the wind field applied for calculating the flux. Ideally, the wind field (wind speed and direction) should be measured at different heights above ground surface. Another limitation is accessibility to an aircraft, sourcing fast and accurate instrumentation and associated costs.

2.8 The tracer gas dispersion method

**Methodology.** The tracer gas dispersion method uses simultaneous measurements of atmospheric CH$_4$ concentrations and a tracer gas released at the emission source at a known rate (Fig. 5). It is assumed that CH$_4$ and the tracer gas undergo the same fate (dispersion, chemical/photochemical reaction) in the atmosphere within the time span of the campaign, so for this reason, tracer gases with relatively long atmospheric lifetimes are used. SF$_6$ has been
used in many tracer gas studies, such as research on atmospheric dispersion (Martin et al., 2011), ventilation (Timko et al., 1984) and the CH$_4$ emission of ruminants (Storm et al., 2012) and gas facilities (Lamb et al, 1995), as it has a very long lifetime in the troposphere (3,200 years) and negligible background concentrations (Ravishankara et al., 1993). However, SF$_6$ is a very potent GHG (23,500 times more potent than CO$_2$ on a 100-year scale) (Myhre et al., 2013), and so it is not valid for environmental studies. A move has been made toward using N$_2$O instead (Galle et al., 2001), as it is stable in the atmosphere and measurable, although it is present in significant atmospheric concentrations, currently around 331 ppb (NOAA, 2018). Nonetheless, N$_2$O is also a strong GHG (298 times more potent than CO$_2$ on a 100-year scale) (Myhre et al., 2013), which in recent studies has led to the use of acetylene (C$_2$H$_2$) as a tracer gas (Rees-White et al., 2019; Mønster et al., 2015; 2014; Foster-Wittig et al., 2015a; Green et al., 2010). Although highly flammable, C$_2$H$_2$ is not a potent GHG, it is cheap and it is relatively easy to release at a constant flow rate.

Based on downwind atmospheric concentrations of CH$_4$ and the tracer gas measured during its controlled release, CH$_4$ emissions from the source can be calculated. The tracer dispersion method is generally divided into either a stationary or a dynamic approach (Fig. 5).

2.8.1 The stationary tracer gas dispersion method

The stationary tracer gas dispersion method relies on CH$_4$ and tracer gas measurements performed at fixed single or multiple points in the plume downwind from the emission/release source. They have been used previously to study emissions from several different sources, such as aromatic hydrocarbons in refinery wastewater (Howard et al., 1992), CH$_4$ from cattle barns (Kaharabata et al., 2000; Marik and Levin, 1996), CH$_4$ from slurry tanks (Kaharabata et al., 1998) and ammonia from manure spreading (Galle et al., 2000). The first landfill application of
the method was done by Czepiel et al. (1996), using SF$_6$ as a tracer gas. Before CH$_4$ and tracer
gas measurements were conducted, the downwind plume was located by using an analyser
placed in a vehicle. Subsequently, evacuated canisters were placed across the plume transect,
in order to sample air in the plume. Similar setups have been used in other studies using
evacuated canisters (e.g. Scharff et al., 2009; Jacobs et al., 2007; Scheutz et al., 2007;
Tregoures et al., 1999) or sampling bags filled by small pumps (Scharff and Hensen, 2009;
Babilotte et al., 2008). For landfills with poor access to downwind plume transects, Galle et al.
(2001) developed a method based on air concentration measurements performed at a single
point. CH$_4$ and tracer gas concentrations were measured for hours at a single point downwind
of the landfill, using N$_2$O as the tracer gas, and the method was used for a number of landfills
in Sweden (Börjesson et al., 2009).

Recent studies have shown that leachate wells can be significant point sources for CH$_4$
emissions (Scheutz et al., 2011a, 2011b; Fredenslund et al., 2010). Fredenslund et al. (2010),
for instance, suggested quantifying emissions from leachate wells, using a small-scale tracer
gas dispersion method (Fig. 6), whereby tracer gas was released at the bottom of the leachate
well and CH$_4$ and tracer gas were measured at a single point a few metres downwind of the
well.

Advantages and limitations. An important assumption is that the tracer gas should
simulate the CH$_4$ emission. In order to obtain a good mix of tracer gas and CH$_4$, measurement
or sampling campaigns have to be executed at a sufficient distance away from the landfill (e.g.
Mønster et al., 2014; Delre et al., 2018). Misplacing the tracer gas can result in significant
under- or overestimation of the emission, while the required measuring distance depends on
the physical size of the source, atmospheric conditions and the topography between the landfill
and the measurement/sampling point (e.g. Fredenslund et al., 2019b; Rees-White et al., 2019; Delre et al., 2018; Taylor et al. 2016).

One drawback of the static tracer dispersion method is that it can be hard to locate the CH$_4$ plume. Samples are thus taken blindly and the method relies on a stable wind direction throughout the sampling period – a significant change in wind direction could move the downwind plume away from the sampling point(s). If the CH$_4$ emission is not well simulated by the tracer gas, sampling at the fringe of the CH$_4$ plume and tracer gas plume will result in false emission rates, as the ratio between the gases is not representative of the plume.

2.8.2 The dynamic tracer gas dispersion method

The dynamic tracer gas dispersion method is based on performing transects of the downwind plume and thereby measuring CH$_4$ and trace gas concentrations near to ground level across the whole plume, along with the subsequent integration of the plumes. In order to use this method, a sensitive and relatively fast analytical instrument is needed. A Swedish group developed an FTIR instrument stable enough to be driven around in a van, enabling many successful studies of CH$_4$ emissions from landfills (e.g. Scheutz et al., 2011b). This method was then developed further to use two or more trace gases for quantifying CH$_4$ emissions from multiple sources close to each other (Scheutz et al., 2011b).

The analytical instruments used for tracer gas dispersion measurements have often been built specifically for this purpose and are large, heavy and power-demanding; consequently, they are expensive to run and require highly skilled personnel to operate and maintain them effectively. Recently, new instruments have been developed based on cavity ring down spectroscopy (CRDS). These instruments are smaller, lighter and easier to mount in a vehicle. The cavity ring down instrument developed for the tracer gas dispersion method focuses on
measuring CH$_4$ and C$_2$H$_2$ (tracer gas), and it has been used successfully for dynamic plume measurements at two Californian landfills (Green et al., 2010), 15-30 Danish landfills (Mønster et al., 2015; Fredenslund et al., 2019a), a number of landfills in the UK (Bourn et al., 2019; Rees-White et al., 2019) and 15 US landfills (Foster-Witting et al., 2015a). Besides, it was used in a model comparison study at a new landfill (de la Cruz et al., 2016) and a horizontal stationary plume approach using gas sampling from a weather balloon, for measuring in the plume centre, which was found to be situated at an elevated height (Han et al., 2010) and used for exploring emission variations and possible bias in the tracer gas dispersion method (Matacchiera et al., 2019; Delkash et al., 2015).

Advantages and limitations. The advantage of the dynamic tracer gas dispersion method is its simplicity, because when the CH$_4$ and tracer gas plumes are mixed fully, analysis and calculation are relatively straightforward. An important advantage of the method is that it measures total emissions from the landfill, including those from hotspots and installations at the site, and may be used independently of the landfill structure or topography. Moreover, it can be applied at landfills of all sizes. When applied at larger landfills, it may be necessary to release tracer gas at several points (Rees-White et al., 2019; Aghdam et al., 2018; Mønster et al., 2014), but the method can also be used for improving the input data required for inverse modelling. Piccot et al. (1996) and Hensen and Scharff (2003) used tracer gas measurements to obtain accurate information about dispersion conditions during their measurement campaign, thus improving the dispersion model in use. The advantages and limitations of the CRDS-based instrument have been explored and compared recently with simultaneous FTIR measurements (Delre et al., 2018; Mønster et al., 2014). Foster-Wittig et al. (2015a), for instance, published a study involving tracer gas dispersion measurements on a large number of US landfills, including suggestions for using data quality criteria for filtering measurements.
for downwind traverses where tracer and CH$_4$ were not significantly mixed. Recent studies recommend that a minimum 10 plume transects, but preferably 15, should be performed to reduce the variability of a measurement (Fredenslund et al. 2019b; Mønster et al., 2014), while learning how to establish the minimum detectable emission rate at individual sources has also been suggested (Delre et al., 2017). In another recent study, Taylor et al. (2016) explored the uncertainty of the tracer dispersion method by modelling downwind concentrations from released tracer gas. They found that the main potential error was the correct placement of the tracer gas bottles, while factors such as wind speed and direction had a much smaller influence but could nevertheless be emphasised by the physical layout of the landfill and the emission areas. Taylor et al. (2016) also concluded that the main uncertainties depend on the landfill and thus are site-specific. The disadvantages of the technique are its dependence on the right weather conditions (wind direction and wind speed) to ensure sufficient gas mixing, combined with proper road access, which makes it difficult to measure at some landfills with limited access and very few surrounding roads. Matachiera et al. (2019) suggested that modelling could be used as an initial step in planning a tracer gas dispersion measurement campaign (tracer gas placement and measuring distance etc.), but the method alone cannot identify on-site emission sources, and so care must be taken regarding the presence of other CH$_4$ sources (manure storage tanks, farms, composting facilities) in the surrounding area, which can cause extensive errors in site quantification. Furthermore, the instrumentation is expensive and requires expertise to operate it effectively. An additional disadvantage is that measurements are usually taken in hours (2-6 hours) or over a few days, and thus the temporal variation of landfill gas emission can be a challenge to capture. Recent and current research focuses on method validation (Fredenslund et al., 2019b; Matachiera et al., 2019; Delre et al., 2018;
Mønster et al., 2014) and the establishment of quality criteria for data processing (Fredenslund et al., 2019b; Foster-Wittig et al., 2015a).

2.9 Differential absorption LiDAR method

The differential absorption LiDAR (DIAL) method transmits pulsed laser radiation into the atmosphere. Part of this radiation is then backscattered by atmospheric constituents and can be measured by a detector to determine CH$_4$ concentrations by tuning the laser to the appropriate absorption lines and comparing this with a laser at a slightly different wavelength that is reflected in a similar way to the first laser but not absorbed by CH$_4$. The laser can be pointed in any direction, and for landfill applications, measurements are taken along several lines of sight, thereby obtaining a vertical concentration distribution downwind of the landfill or sections thereof for which emissions are to be quantified (see Fig. 7). This allows for capturing the entire vertical profile of the CH$_4$ plume, and emissions from the landfill (or landfill section) can be calculated by combining the vertical concentration profile with wind speeds at different heights (Innocenti et al., 2017; Robinson et al., 2011; Babilotte, 2011; Babilotte et al., 2010).

Measurements are normally taken downwind and close to the landfill, in combination with upwind measurements, to check for interfering sources and to obtain background concentrations. Individual landfill sections can also be measured by ‘shooting’ the laser in a vertical plane immediately downwind of the targeted section at the landfill (Babilotte, 2011). A moveable DIAL system is a large setup mounted on a truck, and very few are currently available worldwide, and it has been applied mainly for quantifying fugitive emissions caused by petrochemical processes, while its use at landfill sites is still limited (Innocenti et al., 2017; Bourn et al., 2019; Babilotte, 2011; Robinson et al., 2011; Babilotte et al., 2010).
Advantages and limitations. The main disadvantage of the DIAL method is the cost and
size of the analytical setup and the complexity involved in data handling. The truck
transporting the setup requires good, smooth roads for access, and measurements depend on
wind direction and infrastructure. In addition, stable and accurately measured wind conditions
are needed for precise flux quantification. Wind speed above ground level should be measured
at different heights for optimal modelling of the wind profile. As with other remote sensing
measurements, DIAL is sensitive to possible interfering sources in the vicinity of the measured
area. Measurements are conducted over hours or days, and long-term temporal variations can
therefore be difficult to capture. The advantage of the DIAL method is that it can measure CH$_4$
on a distance of 400 - 800 m, depending on atmospheric conditions, hence enabling
measurements of the total vertical concentration plane of small landfills and thereby
eliminating the effect of spatial emission variation. At larger landfills, it is necessary to
perform more measurements, due to the limited range of the laser. In such cases, the challenge
is to determine which area contributes to the CH$_4$ flux measured through the vertical profile.
The NPL (National Physics Laboratory) in England has two complete DIALs and has
conducted measurements at several landfill sites in the country (Innocenti et al., 2017; Bourn
et al., 2019). At a large landfill, a measurement campaign usually takes up to one week, with
the first day used to prepare the analytical equipment. In the following days, the truck is
moved around the landfill (or along its edge) and measurements are taken from different
sections to be added up at the end of the measurement campaign. The DIAL has the option of
measuring other atmospheric species by scattering light in infrared or ultraviolet spectra, such
as ethane, hydrogen, sulphur oxide, ozone and benzene, below ppm level (Babilotte et al.,
2010).
2.10 Inverse dispersion modelling method

Inverse modelling relies on downwind concentration measurements. By combining these measurements with meteorological data, the emission rate from a source of known location can be calculated using the wind direction and the theory of gas dispersion in the atmosphere. The inverse modelling method can be divided into two approaches, using stationary or mobile measurements.

2.10.1 Inverse modelling – stationary

In stationary inverse modelling, one or several measurement points, or a path-integrated concentration, are used downwind of the landfill. Data are recorded either by taking continuous concentration measurements or by sampling for a fixed amount of time, aligned with subsequent analysis in the laboratory. Applying the concentration measurements in an appropriate model, together with information on atmospheric conditions and topography, provides an estimate of emission rates. Several different approaches to this method have been developed, using long-term sampling at one or multiple fixed points, by employing one or multiple continuous measurements or cross-plume-integrated concentration measurements (e.g. Riddick et al., 2017; Terent’eva et al., 2017; Foster-Wittig et al., 2015b; Zhu et al., 2013; Abichou et al., 2012; Figueroa et al., 2009). Different models have been developed based on the dispersion of gases, an example of which is the AERMOD steady-state Gaussian plume model, which simulates atmospheric dispersion in the boundary layer. It can handle both ground-level and elevated sources as well as different types of terrain, where the latter requires detailed information on elevation, surface structures, land use, etc. AERMOD was developed for regulatory purposes and to evaluate industrial emission, and it is thus not targeted specifically at landfills, though it has been used in various studies, for example to evaluate dust
emissions (Westbrook et al., 2007), CH$_4$ emissions from a Canadian landfill (Wyles et al., 2010) and suggested as a method to improve the planning of tracer gas dispersion measurements (Matachiera et al., 2019).

Additionally, inverse dispersion modelling has been introduced for area sources (e.g. Flesch et al., 1995), and specific models have been developed for path-integrated concentrations and for point concentration measurements, e.g. LASAT (Janicke Consulting, Überlingen, Germany) and WindTrax (Thunder Beach Scientific, Nanaimo, Canada). Common to these models is the need for information about the surface and the shape of the source, the location of measurements and wind speed and direction. Additional information on surface-induced turbulence can be used for improving the model output. The LASAT model, using path-integrating concentration measurements combined with anemometer measurements, has been employed for estimating CH$_4$ emissions from a landfill in Germany (Zhu et al., 2013) and a composting facility in Austria (Hrad et al., 2014). WindTrax has been used to estimate CH$_4$ emissions, using single-point measurements performed 700 metres from a UK landfill (Riddick et al., 2018) and 300 metres from another UK landfill to compare with two other model approaches (a simple Gaussian plume model and a Lagrangian dispersion model) using measurements taken at one point 7 km from the landfill (Gaussian plume) and from four measurement points ranging from 7 to 100 km away from the landfill (Lagrangian dispersion model) (Riddick et al., 2017).

2.10.2 Inverse modelling – dynamic

Dynamic inverse modelling relies on concentration measurements across a plume downwind of an emission source. The method is equivalent to the dynamic tracer dispersion method, but it is done without releasing a tracer gas. Instead, measured concentration profiles are used in combination with wind data, to fit into an inverse model and calculate emission rates from the source. At landfill sites, measurements are typically done from 500 m to several
kilometres downwind of the site, depending on the size of and emission rate from the landfill. The further the distance to the landfill, the more the spatial variation of the emission will be minimised, and the landfill can be seen as a single-point source in the calculations of emission rates, which is an important assumption when, for example, using a simple Gaussian model. The analytical equipment used for these kinds of measurements needs to be able to measure concentration differences of down to a few ppb, with a background concentration of CH$_4$ normally sitting between 1800 and 2000 ppb. Currently available and capable instrumentations include quantum cascade laser spectroscopy, tuneable diode laser spectroscopy or cavity ring down spectroscopy, each of which has its own advantages and disadvantages in terms of temporal resolution and the number of measurable species.

Advantages and limitations. Mobile plume measurement with inverse modelling has the advantage of measuring the emission plume from the entire landfill site, regardless of size. The measured plume will also include emissions from the slopes of the landfill and the gas and leachate collection systems, which are often difficult to measure using on-site methods. Disadvantages include the need for specialised analytical equipment and a detailed emission model of the landfill, which needs a number of input parameters such as atmospheric stability, surface-induced turbulence and wind speed. For optimal conditions, the landfill should be located in a relatively flat area with a road crossing the downwind plume at a suitable distance away. In addition, accurately measured and relatively stable meteorological conditions are desirable, such as stable wind direction and speed, minimal changes in atmospheric pressure and stable atmospheric conditions in relation to turbulence. Hensen and Scharff (2001) used inverse Gaussian modelling and calibrated their model using controlled emissions of N$_2$O as a tracer gas, making it similar to a tracer gas dispersion method, albeit with subsequent inverse modelling, as measurements were taken too close to the landfill for total mixing of the tracer
and CH₄ plumes. The authors measured three landfills in the Netherlands and found emissions ranging between 40 and 392 kg CH₄ h⁻¹. Lan et al. (2015) measured various CH₄ emissions in the Barnett shale area, combining dynamic plume measurement and inverse Gaussian modelling. The emission quantifications included four landfills, with CH₄ emissions ranging between 86 and 2086 kg h⁻¹ and with an estimated uncertainty smaller than 60%. Lamb et al. (2016) performed inverse modelling on various CH₄ sources in Indianapolis, Indiana, to estimate the whole city’s CH₄ emissions, employing a tracer gas release approach for calibration. The measurements were comparable with emissions estimated from the mass balance modelling of aircraft measurements and inverse modelling from single-point (tower) stationary measurements (Lamb et al., 2016). Fredenslund et al. (2019a) used a similar approach, performing fast screenings of 91 Danish landfills, using downwind plume measurements and inverse Gaussian modelling to find landfills suitable for mitigation initiatives. CH₄ emissions were modelled to range from no detectable plume and up to 43 kg h⁻¹. A good agreement between modelled emission rates with measured emission rates using the tracer gas dispersion method was shown (a correlation coefficient of 0.965 based on 24 measurement campaigns). Emission rates based on inverse Gaussian modelling were very sensitive to the choice of terrain and the Pasquill-Gifford stability class, which could affect the emission rate by more than 100% (Fredenslund et al., 2019a). Downwind CH₄ plume measurements combined with inverse Gaussian modelling was thus found useful as a fast screening method but not appropriate for accurate landfill CH₄ emission quantification.

In general, reverse modelling requires large amounts of high-quality input data to achieve a good emission estimate. Excellent atmospheric condition inputs (e.g. atmospheric stability, wind speed at different heights and surface-induced turbulence) from the exact area of emission are very important and can be difficult to obtain. Stationary measurements are also
dependent on wind direction, and thus the correct placement of air sampling devices is of the utmost importance, as changes in wind direction can eliminate capturing CH$_4$. The dynamic approach needs specialised analytical equipment, a detailed emission model of the landfill site and access to downwind roads. In an ideal world, the landfill should be located in a relatively flat area with a road crossing the downwind plume at a suitable distance away. In addition, accurately measured and relatively stable meteorological conditions are required, such as stable wind direction and speed, minimum changes in atmospheric pressure and stable atmospheric conditions in relation to turbulence. Static inverse modelling makes long time series possible, thereby overcoming the temporal variations of emissions. The dynamic approach has the advantage of measuring the emission plume from the whole landfill site, regardless of its size. The measured plume will also include emissions from the landfill’s slopes and leachate collection systems. Generally, for the modelling methods, large numbers of input parameters are required, and the quality of these inputs influences significantly the outcome of the model’s calculations. Obligatory factors such as wind speed, wind direction and atmospheric turbulence are highly variable over time, and correct values are difficult to obtain for accurate model calculations. Additionally, factors such as atmospheric stability class and surface roughness need to be determined or estimated.

3. Methane emission comparison studies

The different methodologies used to quantify CH$_4$ emissions from landfills, in some cases, have been used in tandem to verify obtained emission rates (e.g. Lohila et al. 2007; Spokas et al. 2006; Börjesson et al. 2000; Mosher et al., 1999; Czepiel et al. 1996;). During the last couple of decades, a number of studies have evaluated available gas emission measurement methodologies by comparing them in field experiments, using controlled CH$_4$ release or at
actual landfill sites (Cambaliza et al., 2017; Babilotte, 2011; Babilotte et al., 2010; Green et al., 2010; Goldsmith et al., 2008; Tregoures et al., 1999). Table 3 provides an overview of reported comparison studies in which three or more methods were compared. In recent studies, the flux chamber method has significantly underestimated actual CH$_4$ emissions from landfill sites, while there has been better agreement between radial plume mapping and dynamic tracer gas dispersion (Green et al., 2010). The often underestimated emissions from flux chambers are most likely due to the fact that at many landfills a significant proportion of CH$_4$ is emitted from small hotspot areas, which are not represented in the chamber measurements. A similar result was observed by Schroth et al. (2012), who compared emission measurements from a 1500-m$^2$ landfill test area, using closed flux chambers and eddy covariance. Over five measurement days, the 16 randomly distributed flux chambers showed an uptake of CH$_4$ (negative emissions) most of the time, while the eddy covariance measurements showed an emission during all five measurement days. Both quantification methods showed a large daily emission variation ranging from -0.07 to 5.1 (chamber) and 1.6 to 52.7 kg h$^{-1}$ (eddy covariance). Tregoures et al. (1999) found slightly higher emissions, using static and dynamic flux chambers compared to static and dynamic tracer gas dispersion methods, and significantly higher results than using eddy covariance and static mass balance. Tregoures et al. (1999) concluded that the flux measured by eddy covariance and static mass balance was underestimated, as the setup and the dominant wind direction only allowed a small part of the landfill to be measured. The higher emission measured by chamber methods compared to tracer gas dispersion methods could be a result of tracer gas dispersion measurements being performed very close (100-200 m) to the landfill or that surface flux chamber measurements were performed at emission hotspots. Babilotte (2011) compared five methodologies at two landfills and three of the methods at a controlled release site at four different release configurations or rates (releases at one and two points with the
same release rate, and releases at three points with different release rates (see Table 3)). In the controlled release test, Babilotte (2011) found that VRPM slightly underestimated emissions (-3 to -13%) measured at a distance close to the source (10 m) and by -42 to -48% at a distance 100 m away. The dynamic tracer gas dispersion method (using FTIR instrumentation and N2O as a tracer gas) overestimated emissions by between 4 and 19%. The highest error rate occurred when the source furthest upwind did not release CH4, thus representing tracer gas misplacement and causing a higher dispersion of the tracer gas than the released CH4, therefore resulting in an overestimation. Plume measurements for the tracer gas dispersion method were taken 400 m to 450 m away from the release area, which was a sufficient distance to obtain well-mixed plumes of CH4 and tracer gas from a relatively small area, with 30 to 44 metres between emission spots. The DIAL method results showed both under- and overestimations, ranging from -21 to +19%, with no clear trends. Babilotte (2011) included closed flux chambers and eddy covariance during measurement campaigns at two landfills. It was concluded that limited credibility could be given to the closed flux chamber results, due to high variations (more than 100%) between the individual flux measurements; consequently, extrapolation to the entire landfill area would be very uncertain. Furthermore, Spokas et al. (2006) recommended caution when comparing chamber (static) and tracer gas measurements (static), due to difficulties in determining and comparing the flux footprint areas of the two methods. The other four methods in Babilotte (2011) showed results with emission rates for DIAL < Tracer < eddy covariance < VRPM.

Comparison studies have also included inverse modelling. Scharff et al. (2003), for instance, compared the eddy covariance method with static and dynamic inverse modelling at four landfills in The Netherlands and found comparable results, although the eddy covariance method seemed to give a lower CH4 flux. Tracer gas was used in inverse modelling to calibrate
atmospheric stability parameters (Schaff et al., 2003). In addition, Babilotte et al. (2010) compared inverse modelling by using dynamic plume concentrations measured 300 m downwind of a landfill and 20 m downwind of the controlled release point. At the landfill, inverse modelling provided a significantly higher flux than for VRPM, tracer gas dispersion (static and dynamic) and DIAL (see Table 1). For the controlled release test, VRPM, both tracer gas methods (static and dynamic) and inverse modelling all seemed to overestimate emissions by factors of two to five, while DIAL underestimated by a factor of two. This study was carried out with a low release rate (1.8 kg CH\(_4\) h\(^{-1}\)). Later research showed that DIAL could quantify down to 3.6 kg CH\(_4\) h\(^{-1}\) with an accuracy rate of 20% (Babilotte, 2011).

Recently, Cambaliza et al. (2017) applied four quantification methods at a landfill in Indiana, USA. Two of the methods (aircraft-based mass balance and tracer gas dispersion) were capable of measuring the entire site’s emissions, whereas the other two methods (VRPM and static surface chamber) were used to measure emissions from individual parts of the landfill. The two whole-site methods showed similar CH\(_4\) emission rates: aircraft-based mass balance between 400 and 980 kg h\(^{-1}\) and tracer gas dispersion between 460 and 750 kg h\(^{-1}\), which also agreed well with the modelled emission using CALMIM (690 ± 120 kg h\(^{-1}\)). These results, however, were not obtained through simultaneous measurements, and thus the comparison assumes that the performed measurements (20 measurement campaigns done from 2009 to 2012, using tracer gas dispersion and two measurement campaigns for aircraft-based mass balance done in August 2012 and July 2014) represent the same, average emission from the landfill. The whole-site emission measurements were carried out over a period of almost five years, with the only overlap of the campaigns being in August 2012, when tracer gas dispersion measurements were done the day before and the day after an aircraft measurement campaign. In the measurements taken over these three days, the aircraft-based mass balance
method showed approximately twice as high a CH$_4$ emission rate (980 kg h$^{-1}$) as the tracer gas dispersion method (480 ± 110 and 470 ± 40 kg h$^{-1}$). The comparison of CH$_4$ emissions measured using the static surface chamber and VRPM showed that the former was unable to capture the emissions observed with the VRPM method. Both methods were applied across the area with intermediate cover. The VRPM showed emissions ranging between 196 and 582 kg h$^{-1}$, while the static chamber method ranged between 0.3 and 1.2 kg h$^{-1}$.

4. Conclusion and perspectives

Historically, methane (CH$_4$) emission measurements from landfills were initiated primarily for research purposes, to obtain emissions at a specific place and time, or to monitor temporal variability over time. Surface emission monitoring using simple reconnaissance monitoring, such as FID walkovers, sometimes combined with surface flux chambers, are used by landfill operators to determine the spatial variability of the emission and for cover maintenance. Annual CH$_4$ emissions reported for regulatory purposes are usually based on models, but when they are estimated solely from landfill gas generation models using waste data, these emissions are often overestimated due to a conservative approach and a lack of actual knowledge relating to deposited waste and CH$_4$ oxidation. More detailed process-based models such as CALMIM have been developed to overcome some of these challenges.

However, only a few of the available models have been validated, and it is uncertain as to what extent they describe actual CH$_4$ emissions. For this reason, CH$_4$ emission measurements are important and can provide further knowledge on actual emissions at individual landfills, which can then be used in model evaluation/validation and for greenhouse gas emission reporting.

Current measurement methodologies do not allow continuous CH$_4$ emission measurements at all landfills worldwide, but some countries are starting to work toward
relevant regulations based on measurements instead of models. The recent development in mass emission techniques, especially the dynamic tracer gas dispersion method, certainly allows for shifting to measurement-based regulation. Measurements are also essential for quantifying the effect of emission mitigation initiatives, such as CH$_4$ extraction and utilisation, or the engineered optimisation of CH$_4$ oxidation at the surface, which can significantly lower the waste sector’s emanations of greenhouse gases. One example is the nationwide implementation of biocovers at old landfills in Denmark, with compulsory emission measurements at all involved sites for quantifying the CH$_4$ mitigation effect (Danish EPA, 2018; Scheutz and Kjeldsen, 2019). Another example is the approach proposed by Bourn et al. (2019) where landfills with gas collection have to fulfil a minimum methane collection efficiency based on annual methane emissions measurements.

The main challenges when quantifying landfill CH$_4$ emissions are high spatial and temporal emission variations. No single emission measurement method is perfect, and many years of research and tests have revealed some of the advantages and disadvantages of individual approaches. To quantify the CH$_4$ emission from an entire landfill, including a confidence interval, a mass emission method is required rather than a surface emission factor method. Both DIAL and tracer gas dispersion are mass emission methods, and by applying them accordingly, the total CH$_4$ emissions from the landfill are measured – and thus the challenge with spatial emission variability is circumvented. Both the DIAL technique and the tracer gas dispersion method have been demonstrated successfully in several landfill studies. However, the DIAL method uses very complicated and expensive equipment, whereas the tracer gas dispersion method takes an easier approach and uses more affordable apparatus. The relatively high cost of the DIAL equipment and the requirement for highly skilled personnel is a potential barrier to routine measurements. Radial plume mapping, eddy covariance and the
stationary mass balance method are all challenged by complex landfill topography and tend to cover only a fraction of the landfill, in which case extrapolation to the full landfill area is needed, thereby adding uncertainty to the measurements.

Methods measuring mass emissions from the whole landfill often result in higher emission rates in comparison to those measuring the emission rate from a given surface area followed by extrapolation to the entire landfill. Surface flux chambers have been used widely for landfill CH$_4$ emission measurements, but they are likely to underestimate emission rates, due to the heterogeneous nature of most landfill covers and the fact that emissions from wells, vents, etc. are not captured by flux chambers. Therefore, flux chambers are not recommended for whole landfill site CH$_4$ emission quantification, but they can be combined with soil gas concentration profiles to provide useful information about landfill gas migration, CH$_4$ oxidation and influencing factors (e.g. changes in weather conditions). Furthermore, this supporting information could be useful in combination with other quantitative emission methods.

Landfill CH$_4$ emissions fluctuate over time (short-term within hours as well as seasonally) due to changes in landfill operation, environmental conditions, etc. Most of the current measuring techniques are employed for a short period and thus measure the emission over a short time interval (hours). An obvious challenge therefore remains in relation to how to plan measurement campaigns (number and frequency) and inevitably to interpolate the measured emissions at discrete times to annual emission rates useful for reporting and/or evaluation of mitigation actions. The development of cheaper emission techniques would allow for more frequent measurements, which in turn could be used for tuning existing landfill generation models and for estimating emissions over longer timescales. For measurements over longer time periods, static plume methods combined with inverse modelling, or eddy
covariance and the static mass balance method, can be applied. These methods can be set up for continuous measurements and thereby capture temporal emission variations. An optimal approach could involve combining continuous measurements with frequent short-term measurements, using the DIAL or tracer gas dispersion methods. As a result, this approach could provide longer-term emission rate time series, which could be validated or adjusted by more accurate, albeit more expensive and infrequent, measurements.

In addition to quantitative methods, qualitative reconnaissance techniques exist. These are valuable tools in the daily operation and maintenance of the landfill cover and of site installations such as gas collection and recovery systems.

Currently, there is a fast development of potentially improved methods based on the flexibility of UAVs and aircraft. Accurate and high-precision CH$_4$ instruments are currently too heavy to employ in UAVs, and aircraft-based measurements are still done mainly in the oil and gas sector. However, the fast development within both CH$_4$ sensors and UAVs could soon allow the use of UAVs at landfills for measuring CH$_4$ with sufficiently high accuracy and temporal resolution. In addition, satellites specifically measuring atmospheric CH$_4$ concentrations are working on getting higher spatial resolution, in order to include area sources such as landfills.

Although several measuring methods for landfill CH$_4$ emission quantification have been developed, the accuracy of many of these methods remains unknown. Determining this factor is a challenge, as accuracy and potential measurement errors stem from different influences extending far beyond the uncertainty of the analytical instrumentations applied. As an example, the choice of model input parameters, such as terrain conditions (open or urban, flat or elevated terrain, etc.), atmospheric conditions (mixing, stability, wind speed profiles, etc.) and spatial emission patterns (homogenous vs. inhomogeneous), will add to overall method uncertainty. This also means that
uncertainty and potential errors are not only specific to each method and instrumentation applied, but also site-specific and depend on conditions on the measuring day. Moreover, some methods might not be applicable at all landfill sites and under all conditions, so their deployment in non-favourable circumstances can cause large errors. Future research should hence focus on establishing method accuracy and developing best practice in terms of monitoring, data processing and interpretation guidelines.

With new technological capabilities for measuring CH$_4$ emissions from landfills, there is a need to develop measurement plans and strategies to comply with current and future legislation, as well as to document mitigation actions. For regulatory purposes, an average emission rate is of interest, which is why measurement strategies must pay attention to the dynamic nature of landfill emissions. Future research should thus focus on developing measurement protocols and strategies to obtain emission rates representative of normal landfill operations and environmental conditions.

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Table 1. Qualitative reconnaissance techniques for locating landfill ‘hotspots’ or determining surface emission patterns with potentially elevated CH$_4$ emissions. Reconnaissance techniques should be used in combination with the quantitative techniques described in Table 2, but they can be useful for establishing field experimental designs, checking the integrity of cover materials and planning site maintenance.

<table>
<thead>
<tr>
<th>Application</th>
<th>Advantages</th>
<th>Limitations</th>
<th>Status and references</th>
</tr>
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<tbody>
<tr>
<td><strong>Portable CH$_4$ analyser</strong></td>
<td>Rapid and simple to deploy in the field. Relatively inexpensive - only requires a portable instrument and a GPS for walkover surveys.</td>
<td>Above-surface CH$_4$ concentrations are affected by interfering CH$_4$ sources, meteorological and soil variables and site topography. Recorded above-surface CH$_4$ concentrations are not universally correlated to CH$_4$ surface fluxes.</td>
<td>A widely used technique as a tool to determine the need for cover remediation and maintenance.</td>
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<td><strong>Field infrared survey</strong></td>
<td>Uses thermal infrared techniques to detect points or areas with elevated temperatures. Assumes elevated surface temperatures are caused by warm emitted landfill gas. Deployed either on site (inspection using handheld instruments) or on UAVs or aircraft, depending on the scale and required sensitivity.</td>
<td>Thermal anomalies may be due to processes other than the emission of warm landfill gas, e.g. microbial processes (respiration or CH$_4$ oxidation), or variability in the emissivity of different surfaces potentially causing sampling bias. In warm climates thermal anomalies due to emission hotspots may not be visible.</td>
<td>Thermal infrared cameras mounted on UAVs are used widely for leakage searches in the gas industry, but they are still to be proven useful at landfills, as multiple causes of observed thermal anomalies must be considered (e.g. Fjelsted et al., 2019; Tanda et al., 2017; Battaglini et al., 2013; Lewis et al., 2003).</td>
</tr>
<tr>
<td><strong>UAV survey</strong></td>
<td>Atmospheric CH$_4$ concentration measurements recorded by sensors or instruments in a UAV. Rotary UAVs are often preferred due to the possibility of slower flight speed and easier take-off and landing.</td>
<td>The extent of its use is limited by the safety regulations covering UAV application at the landfill site, battery time (rotary UAVs) and weather constraints (rain and wind).</td>
<td>A promising emerging technique. UAVs are getting cheaper and better (longer battery time, better navigation systems, etc.). CH$_4$ sensors are getting cheaper and CH$_4$ measuring instruments are getting smaller.</td>
</tr>
<tr>
<td><strong>Visual inspection</strong></td>
<td>Visual and olfactory inspection of the landfill cover. Emission hotspots can often be located by the odour of trace components in the landfill gas, by changes in vegetation and irregularities in the cover (cracks and fissures).</td>
<td>Subjective with many possible influences, including wind speed and direction, soil moisture, site topography and landfill cover maintenance.</td>
<td>A useful first step. Site personnel should be instructed to keep alert for odours and vegetation abnormalities indicating landfill gas emissions.</td>
</tr>
</tbody>
</table>
Table 2. Characteristics of the closed and the open surface flux chamber.

<table>
<thead>
<tr>
<th>Closed chambers</th>
<th>Open chambers</th>
</tr>
</thead>
<tbody>
<tr>
<td>• Also called a static chamber</td>
<td>• Also called a dynamic chamber</td>
</tr>
<tr>
<td>• No exchange of air inside the chamber with external air</td>
<td>• The chamber is continuously flushed with a flow of gas (a carrier gas) – this can be ambient air or a synthetic gas</td>
</tr>
<tr>
<td>• If gas concentrations are measured by pumping a gas flow through a detector, the analysed gas flow can be recirculated back to the chamber to avoid an internal pressure decrease</td>
<td>• The emission is determined by measuring concentrations in the inlet and outlet flows. If a synthetic gas is used, only gas concentration in the outlet flow needs to be analysed</td>
</tr>
<tr>
<td>• The chamber can have a vent to reduce the influence of internal pressure build-up</td>
<td>• The chamber can have a vent to reduce the influence of internal pressure build-up</td>
</tr>
</tbody>
</table>
Table 3. Summary of quantitative techniques for the field measurement of CH₄ landfill emissions.

<table>
<thead>
<tr>
<th>Application</th>
<th>Advantages</th>
<th>Limitations</th>
<th>Status and references</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vertical soil gas concentration profiles</td>
<td>Use for diffusive and convective flux studies, including the influence of pressure changes, passing storm fronts and changes in the wind. Can be used to understand the direction of the diffusional flux and the presence of subsurface CH₄ sources and sinks.</td>
<td>Difficult to apply appropriately if emissions are associated with high spatial variability. The method is thus not useful for up-scaling for whole-site emission quantification.</td>
<td>Limited to a few and often older landfill field studies but the basis for many transport models. Not appropriate for whole-site CH₄ emission quantification (Ginski and Stepniewski, 1985; El-Fadel et al., 1995; Christophersen et al., 2001; Röwer et al. 2011; Gebert et al., 2011).</td>
</tr>
<tr>
<td>Closed surface flux chambers</td>
<td>Good for process studies of emissions. Combined with vertical soil gas concentration profiles, CH₄ oxidation can be accessed. The only technique that can measure both the emission and the uptake of CH₄. Can also be used for non-CH₄ gases and isotopic composition analysis. Not sensitive to other CH₄ sources. Simple to deploy with low limits of detection for fluxes (depending on analytical instrumentation).</td>
<td>Time- and labour-intensive, and appropriate geostatistical techniques must be used for whole landfill site studies. Not an appropriate method for measuring convective fluxes. Often, emission hotspots will not be included in the measurements, leading to whole-site emission underestimation. Temporary disturbance of soil surface can influence the emission.</td>
<td>The method is historically the most applied for quantifying CH₄ emissions from landfills. Not appropriate for whole-site CH₄ emission quantification. Still used in many countries, due to lack of other and more appropriate methods (e.g. Rolston, 1986; Kjeldsen and Fisher, 1995; Tregouët et al., 1999; Conen and Smith, 2000; Christophersen and Kjeldsen, 2001a; Gebert and Gröngöft, 2006, Schuetz et al., 2011a; 2008; 2003).</td>
</tr>
<tr>
<td>Open surface flux chambers</td>
<td>Same advantages as closed chambers except cannot quantify negative CH₄.</td>
<td>Same disadvantages as closed chambers. Not able to quantify very low fluxes because of dilution of chamber gas with carrier gas.</td>
<td>A technique commonly applied at landfills. Not appropriate for whole-site CH₄ emission quantification (e.g. Rolston, 1986; Huber-Humer and Lechner, 2001a; 2001b; Hutchinson and Livingston, 2001).</td>
</tr>
<tr>
<td>Eddy covariance</td>
<td>Measurements of the turbulent transfer of gases between the ground surface and the atmosphere. Need fast measurements of atmospheric CH₄ concentration, wind speed and direction.</td>
<td>Gives an integrated measure of landscape-scale fluxes. Good for flat terrain with uniform emissions. Does not disturb the emission pathways. Useful for long- and short-term temporal studies under changing atmospheric conditions.</td>
<td>Difficult to apply in variable topography and with interfering CH₄ sources. Measured footprint is dependent on meteorological conditions. Requires fast (&gt; 10 Hz) gas-specific instrumentation with high precision.</td>
</tr>
<tr>
<td>Stationary mass balance</td>
<td>CH₄ concentrations are measured at different heights above the landfill surface, and by combining these with simultaneously measured atmospheric conditions, the horizontal flux of CH₄ can be calculated.</td>
<td>Same advantages as eddy covariance.</td>
<td>Same disadvantages as eddy covariance, but does not require fast instrumentation.</td>
</tr>
<tr>
<td>Radial plume mapping</td>
<td>Uses one or more lasers and mirrors to measure the downwind, cross-plume concentration. In combination with measured and modelled wind profiles, surface emissions are useful.</td>
<td>Gives an integrated measure of CH₄ emission from the area upwind of the laser and reflectors and includes edge leakages and other on-site hotspots. Method well described by the US EPA.</td>
<td>Rely on a relatively stable wind direction throughout the measurement. Two sets of instrumentation are needed if background (upwind) measurements need to be</td>
</tr>
<tr>
<td>from the upwind area can be calculated.</td>
<td>measured simultaneously. Laser coverage depends on visibility, and multiple measurements are needed for a whole landfill measurement. Method can be challenged by the topography of the landfill.</td>
<td>1999; Wu et al., 1999; Hashmonay et al., 2001; 2008; Goldsmith, 2008; Abichou et al., 2010; Thoma et al., 2010; Goldsmith et al., 2012).</td>
<td></td>
</tr>
<tr>
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</tr>
<tr>
<td>CH₄ concentrations are measured across the downwind plume from the landfill, using an aircraft. Based on the measured concentrations, a 2D concentration plane is modelled and combined with simultaneously measured atmospheric conditions, the horizontal flux of CH₄ through the 2D plane can be calculated.</td>
<td>Gives an integrated emission from the whole landfill without having to access it.</td>
<td>Has been applied at landfills in the US and UK. The methodology is still under development. With the development of new and light high-precision CH₄ sensors and laser instruments the method has high future potential (e.g. Perschl et al., 2013; Krautwurst et al., 2015; Cambaliza et al., 2015; 2017; Allen et al., 2017).</td>
<td></td>
</tr>
<tr>
<td>Mass balance using aerial measurements</td>
<td>Requires accurate weather data and an aircraft with high-precision instrumentation. Closely located sources can be difficult to separate. The whole plume should be covered.</td>
<td>Method is well proven and has been used in several older landfill studies. The method is being replaced by the dynamic tracer gas dispersion approach, as development in instrumentation allows this transition (e.g. Czepiel et al., 1986; Trégourès et al., 1999; Galle et al., 2001; Börjesson et al., 2007).</td>
<td></td>
</tr>
<tr>
<td>Stationary tracer gas dispersion</td>
<td>Difficult to apply in the case of interfering CH₄ sources. Proper source simulation (correct tracer gas placement) is challenged even by small changes in wind direction while performing measurements. Historically, potent greenhouse gases such as SF₆ and N₂O have been used.</td>
<td>A method of growing interest as better instrumentation has become available. US EPA is in the process of describing the method among their OTMs (e.g. Scheutz et al., 2011; Green et al., 2010, 2012; Mönster et al., 2014, 2015; Foster-Wittig et al., 2015).</td>
<td></td>
</tr>
<tr>
<td>Dynamic tracer gas dispersion</td>
<td>Requires a road at a suitable distance downwind of the landfill and with no interfering CH₄ sources in between. The most commonly used tracer gas is N₂O, which is a potent greenhouse gas but has been replaced with C₂H₆ in recent applications.</td>
<td>Mainly used for VOC emissions in the petrochemical industry, but has also been applied to landfills for CH₄ emission quantification (e.g. Babiiotto et al., 2010, 2011; Robinson et al., 2011; Bourn et al., 2019; Innocenti et al., 2017).</td>
<td></td>
</tr>
<tr>
<td>Differential absorption LiDAR (DIAL)</td>
<td>A laser is shot into the atmosphere and the backscattered part is measured. DIAL is used for landfills by measuring the appropriate CH₄ absorption lines. A 2D downwind concentration plan is modelled and combined with measured and modelled information on wind speed and direction.</td>
<td>Mainly used for VOC emissions in the petrochemical industry, but has also been applied to landfills for CH₄ emission quantification (e.g. Babiiotto et al., 2010, 2011; Robinson et al., 2011; Bourn et al., 2019; Innocenti et al., 2017).</td>
<td></td>
</tr>
<tr>
<td>Inverse modelling - stationary</td>
<td>Can give both whole-site and individual area emissions. Very little interference from other CH₄ sources in the surroundings. Can give detailed information on emission patterns.</td>
<td>Different model approaches have been suggested, but the method has yet to prove accurate for landfill emission rate estimates (e.g. Figueroa et al., 2009; Abichou et al., 2012; Zhu et al., 2013; Foster-Wittig et al., 2015b).</td>
<td></td>
</tr>
<tr>
<td>Inverse modelling - dynamic</td>
<td>Can give whole-site emission estimates and is good for long time series and thus able to capture emission variations.</td>
<td>Not a widely used approach but has been used as a fast screening tool for finding old landfills with significant emissions (e.g. Hensen and Scharff 2001; Fredenslund et al., 2019a).</td>
<td></td>
</tr>
<tr>
<td>Inverse modelling - aerial</td>
<td>Relies on good data on atmospheric conditions and large amounts of measurement data. Is sensitive to interfering sources and complex topography. Data treatment is complex. Will often result in an emission with significant uncertainty.</td>
<td>Different model approaches have been suggested, but the method has yet to prove accurate for landfill emission rate estimates (e.g. Figueroa et al., 2009; Abichou et al., 2012; Zhu et al., 2013; Foster-Wittig et al., 2015b).</td>
<td></td>
</tr>
<tr>
<td>Measurement of downwind plume and combined with accurate atmospheric condition measurements, the emission rate from the landfill can be estimated.</td>
<td>Measurements are fast to perform and can give whole-site emission estimates.</td>
<td>Not a widely used approach but has been used as a fast screening tool for finding old landfills with significant emissions (e.g. Hensen and Scharff 2001; Fredenslund et al., 2019a).</td>
<td></td>
</tr>
<tr>
<td>Measurement of downwind plume and combined with atmospheric condition measurements, the emission rate from the landfill can be estimated.</td>
<td>Instrumentation must be fast and precise. Good measurements of atmospheric conditions with high temporal resolution. Will often result in an emission with significant uncertainty.</td>
<td>Not a widely used approach but has been used as a fast screening tool for finding old landfills with significant emissions (e.g. Hensen and Scharff 2001; Fredenslund et al., 2019a).</td>
<td></td>
</tr>
<tr>
<td>Measurement of downwind plume and combined with atmospheric condition measurements, the emission rate from the landfill can be estimated.</td>
<td>Instrumentation must be fast, precise and deployable in UAV.</td>
<td>Has been applied at various sources in the US, including</td>
<td></td>
</tr>
<tr>
<td>accurate atmospheric condition measurements, the emission rate from the landfill can be estimated.</td>
<td>estimates. No access to landfill needed.</td>
<td>or aircraft. Good measurements of atmospheric conditions with high temporal resolution. Will often result in an emission with significant uncertainty.</td>
<td>landfills (e.g. Cui et al., 2015).</td>
</tr>
</tbody>
</table>
Table 4. Overview of published comparison studies of three or more methodologies for quantifying CH$_4$, either from landfills or controlled CH$_4$ release imitating landfill emission.

<table>
<thead>
<tr>
<th>Application</th>
<th>Methodologies</th>
<th>Emission</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Landfill (Tregoures et al. 1999)</td>
<td>Closed flux chamber (no recirculation)</td>
<td>3750-14200</td>
<td>20 x 20 m grid</td>
</tr>
<tr>
<td></td>
<td>Closed flux chamber (with recirculation)</td>
<td>10512-11375</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Tracer gas dispersion method 1 (SF$_6$)</td>
<td>7720±2650</td>
<td>Downwind plume sampled in two ways: static, multiple points (100 m downwind) and dynamic, multiple distances (115, 130 and 200 m downwind)</td>
</tr>
<tr>
<td></td>
<td>Tracer gas dispersion method 2 (SF$_6$)</td>
<td>6340±2070</td>
<td>Landfill divided into strips, each measured downwind at the edge of landfill</td>
</tr>
<tr>
<td></td>
<td>Eddy covariance</td>
<td>11.2±3.4</td>
<td>Only one mast location. Wind direction enabled only a small part of the landfill to be measured</td>
</tr>
<tr>
<td></td>
<td>Static mass balance</td>
<td>24±8</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Remote sensing (airborne IR thermography)</td>
<td>No</td>
<td>Only used for finding hotspots</td>
</tr>
<tr>
<td>Four landfills (LF1-4) (Scharff et al. 2003)</td>
<td>Static mass balance</td>
<td>LF1: 530±130, LF2: 109±30, LF3: 390±100, LF4: 70±8</td>
<td>Sampling 10, 15 and 25 m above ground</td>
</tr>
<tr>
<td></td>
<td>Inverse modelling (static)</td>
<td>LF1: 440±180, LF2: 277±71, LF3: 690±230, LF4: 184±81</td>
<td>Four sample bags collecting gas in different wind directions</td>
</tr>
<tr>
<td></td>
<td>Inverse modelling (dynamic)</td>
<td>LF1: 540-900±50-125, LF2: 315±33, LF3: 300±10, LF4: 135±32</td>
<td>Using tracer gas for model calibration. At LF1, three experiments were done</td>
</tr>
<tr>
<td>12 landfill sections (P1 to P12) (Goldsmith et al. 2008)</td>
<td>Closed flux chamber</td>
<td>P1: 34.5, P2: 20.0, P3: -0.3, P4: 2.3, P5: 2.2, P6: 1.1, P7: 5.7, P8: 53.6, P9: 0.3, P10: 5.8, P11: 1.1, P12: 0.1</td>
<td>Each landfill part was measured and compared individually</td>
</tr>
<tr>
<td></td>
<td>Radial plume mapping (100 m path)</td>
<td>P1: 184.8 P2: 53.7, P3: 1.8, P4: 19.3, P5: 27.3, P6: 56.8, P7: 87.5, P8: 41.4, P9: 5.5, P10: 89.1, P11: 0.0, P12: 0.0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Radial plume mapping (200 m path)</td>
<td>P1: 92.4 P2: 26.9, P3: 0.9, P4: 9.7, P5: 13.7, P6: 28.4, P7: 43.8, P8: 20.7, P9: 2.7, P10: 44.5, P11: 0.0, P12: 0.0</td>
<td></td>
</tr>
<tr>
<td>Landfill (Babilotte et al. 2010)</td>
<td>Radial plume mapping</td>
<td>25±2.4</td>
<td>Individual cells were also quantified</td>
</tr>
<tr>
<td></td>
<td>Tracer gas dispersion (static) (N$_2$O)</td>
<td>83±36</td>
<td>15 canisters sampled over 4 h across a plume 200 m downwind</td>
</tr>
<tr>
<td></td>
<td>Tracer gas dispersion (dynamic) (N$_2$O)</td>
<td>41±17</td>
<td>Real-time TDL measurements across a plume 200 m downwind</td>
</tr>
<tr>
<td></td>
<td>DIAL</td>
<td>12±2.4</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Inverse modelling</td>
<td>163</td>
<td>Real-time measurements 300 m downwind using instrumentation able to measure ppb changes in CH$_4$ concentration and applied to a Gaussian model software</td>
</tr>
<tr>
<td></td>
<td>Helicopter-borne spectroscopy</td>
<td>No</td>
<td>Able to find hotspots</td>
</tr>
<tr>
<td>Controlled release 0.5±0.04 (g CH$_4$ s$^{-1}$) (Babilotte et al. 2010)</td>
<td>Radial plume mapping</td>
<td>0.89±0.05</td>
<td>Individual cells were also quantified</td>
</tr>
<tr>
<td></td>
<td>Tracer gas dispersion (dynamic) (N$_2$O)</td>
<td>1.7±1</td>
<td>Measurements 100 m downwind</td>
</tr>
<tr>
<td></td>
<td>Tracer gas dispersion (dynamic) (N$_2$O)</td>
<td>2.3±0.5</td>
<td>Model + measurements 100 m downwind</td>
</tr>
<tr>
<td></td>
<td>DIAL</td>
<td>0.23±0.12</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Inverse Modelling</td>
<td>1.5</td>
<td>Real-time measurements only 20 m downwind using instrumentation able to measure ppb changes in CH$_4$ concentration and applied to a Gaussian model software</td>
</tr>
<tr>
<td></td>
<td>Helicopter-borne spectroscopy</td>
<td>No</td>
<td>Able to locate release area</td>
</tr>
<tr>
<td>Two landfills</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Unit (m$^3$ CH$_4$ d$^{-1}$)**
<table>
<thead>
<tr>
<th>Method</th>
<th>Unit</th>
<th>Measurement Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Closed flux chamber</td>
<td>0.018/0.125</td>
<td>26 points at each landfill</td>
</tr>
<tr>
<td>Radial plume mapping</td>
<td>9.4/8.1</td>
<td>Area calculated on atmospheric stability class</td>
</tr>
<tr>
<td>Radial plume mapping</td>
<td>16.7/10.5</td>
<td>Area calculated by multiple linear regression model</td>
</tr>
<tr>
<td>Tracer gas dispersion (static) (C$_2$H$_2$)</td>
<td>8.5/14.3</td>
<td>CRDS instrumentation</td>
</tr>
<tr>
<td>Tracer gas dispersion (dynamic) (C$_2$H$_2$)</td>
<td>7.9/13.1</td>
<td>CRDS instrumentation</td>
</tr>
</tbody>
</table>

**Two landfills (#1/#2) (Babilotte et al. 2010)**

<table>
<thead>
<tr>
<th>Method</th>
<th>Unit</th>
<th>Measurement Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Closed flux chamber</td>
<td>Unit (kg h$^{-1}$)</td>
<td>290/868 1-3% of total area measured</td>
</tr>
<tr>
<td>Eddy covariance</td>
<td>1005/1146</td>
<td>~30% of total area measured</td>
</tr>
<tr>
<td>Radial plume mapping</td>
<td>1325/1244</td>
<td>10-15% of total area measured</td>
</tr>
<tr>
<td>DIAL</td>
<td>659/314</td>
<td>Whole landfill</td>
</tr>
<tr>
<td>Tracer gas dispersion (dynamic) (N$_2$O)</td>
<td>916/789</td>
<td>30-45% of total area measured</td>
</tr>
</tbody>
</table>

**Four Controlled releases (Babilotte et al. 2011)**

<table>
<thead>
<tr>
<th>Release #1</th>
<th>Unit: g s$^{-1}$ (±SD)</th>
<th>One release point</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radial plume mapping</td>
<td>1.09±0.005</td>
<td>1.04±0.220/7.1±0.23/0.6 10/50/100 m from source</td>
</tr>
<tr>
<td>DIAL</td>
<td>0.86±0.27</td>
<td>10 m from source</td>
</tr>
<tr>
<td>Tracer gas dispersion (dynamic) (N$_2$O)</td>
<td>1.16±0.10</td>
<td>450 m from source</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Release #2</th>
<th>Unit: g s$^{-1}$ (±SD)</th>
<th>Two release points</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radial plume mapping</td>
<td>1.09±0.005</td>
<td>1.06±0.230/7.9±0.19/0.5 10/50/100 m from source</td>
</tr>
<tr>
<td>DIAL</td>
<td>0.90±0.25</td>
<td>40 m from source</td>
</tr>
<tr>
<td>Tracer gas dispersion (dynamic) (N$_2$O)</td>
<td>1.31±0.09</td>
<td>400 m from source</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Release #3</th>
<th>Unit: g s$^{-1}$ (±SD)</th>
<th>Three release points</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radial plume mapping</td>
<td>3.28±0.013</td>
<td>2.98±0.65/2.65±0.73/1.9 2±0.45 10/50/100 m from source</td>
</tr>
<tr>
<td>DIAL</td>
<td>1.64±0.07</td>
<td>40 m from source</td>
</tr>
<tr>
<td>Tracer gas dispersion (dynamic) (N$_2$O)</td>
<td>1.31±0.09</td>
<td>400 m from source</td>
</tr>
<tr>
<td>Release #4</td>
<td>Unit: g s$^{-1}$ (±SD)</td>
<td>Three release points</td>
</tr>
<tr>
<td>Radial plume mapping</td>
<td>1.96±0.11</td>
<td>1.42±0.29/1.24±0.220/9 6±0.22 10/50/100 m from source</td>
</tr>
<tr>
<td>DIAL</td>
<td>0.90±0.25</td>
<td>40 m from source</td>
</tr>
</tbody>
</table>

**One landfill (Cambaliza et al. 2017)**

<table>
<thead>
<tr>
<th>Method</th>
<th>Unit: (mol s$^{-1}$)</th>
<th>Measurement Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Closed flux chamber</td>
<td>0.005-0.02</td>
<td>Only intermediate cover measured</td>
</tr>
<tr>
<td>Radial plume mapping</td>
<td>1.7-10.1</td>
<td>Only final and intermediate cover measured</td>
</tr>
<tr>
<td>Aircraft-based mass balance</td>
<td>7-17</td>
<td>Whole landfill (from 2009 to 2012, n = 20)</td>
</tr>
<tr>
<td>Tracer gas dispersion (dynamic) (C$_2$H$_2$)</td>
<td>8-13</td>
<td>Whole landfill (in 2012 and 2014, n = 2)</td>
</tr>
</tbody>
</table>
FIGURES

Fig. 1. Overview of the most common methods used to identify and quantify CH$_4$ emissions from landfills.
**Fig. 2.** Schematic overview of open and closed flux chambers for quantifying CH₄ fluxes from landfills.
**Fig. 3.** Simplified drawing of the eddy covariance and mass balance methods. Eddy covariance uses an ultrasonic anemometer to measure turbulence, while the mass balance approach uses a standard anemometer to measure wind speed and direction.
Fig. 4. Horizontal (HRPM) and vertical (VRPM) radial plume mapping with setup, as suggested in the OTM10 method (Hashmonay et al., 2008).
Fig. 5. Tracer dispersion method using the stationary or dynamic approach. Stationary can be done using sampling or direct measurements, while dynamic requires fast, direct measurements.
Fig. 6. Small-scale tracer dispersion method for quantifying CH₄ emissions through a leachate well. From Fredenslund et al. (2010).
Fig. 7. The DIAL method, using multiple ‘line of sight’ laser shooting with subsequent model fitting to create a two-dimensional cross-plume concentration profile.