Methodologies for measuring fugitive methane emissions from landfills – A review

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4 **Methodologies for measuring fugitive methane emissions from landfills – a** 5 **review**

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17

18 **Highlights**

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

22

23 **Keywords:** Landfill gas, qualitative methane reconnaissance techniques, surface flux

24 chambers, tracer gas dispersion, eddy covariance, mass balance, radial plume mapping, DIAL.

25 **Abstract**

26 Fugitive methane (CH₄) emissions from landfills are significant global sources of greenhouse
27 gases emitted into the atmosphere; thus, reducing them would be a beneficial way of overall
28 greenhouse gas emissions mitigation. In Europe, landfill owners have to report their annual
29 CH₄ emissions, so direct measurements are therefore important for 1) evaluating and
30 improving currently applied CH₄ emission models, 2) reporting annual CH₄ emissions and 3)
31 quantifying CH₄ mitigation initiatives. This paper aims at providing an overview of currently
32 available methodologies used to measure fugitive CH₄ emissions escaping from landfills. The
33 measurement methodologies are described briefly, and the advantages and limitations of the
34 different techniques are discussed with reference to published literature on the subject.
35 Examples are given of individual published studies using different methodologies and studies
36 comparing three or more methodologies. This review suggests that accurate, whole-site CH₄
37 emission quantifications are best done using methods measuring downwind of the landfill,
38 such as tracer gas dispersion and differential absorption LiDAR (DIAL). Combining aerial CH₄
39 concentration measurements from aircraft or unmanned aerial vehicles with wind field
40 measurements offers a great future potential for improved and cost-efficient integrated landfill
41 CH₄ emission quantification. However, these methods are difficult to apply for longer time
42 periods, so in order to measure temporal CH₄ emission changes, e.g. due to the effect of
43 changes in atmospheric conditions (pressure, wind and precipitation), a measurement method
44 that is able to measure continuously is required. Such a method could be eddy covariance or
45 static mass balance, although these procedures are challenged by topography and
46 inhomogeneous spatial emission patterns, and as such they can underestimate emissions
47 significantly. Surface flux chambers have been used widely, but they are likely to
48 underestimate emission rates, due to the heterogeneous nature of most landfill covers resulting

in sporadic and localised CH₄ 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 (CH₄: 55–60% v/v) and carbon dioxide (CO₂: 40–45% v/v), the production of which occurs through three initial sequential phases followed by a phase characterised by stable CH₄ and CO₂ 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 CH₄ 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, CH₄ 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. CH₄ 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 CH₄ 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 CH₄ 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 GtCO₂eq y⁻¹ in total anthropogenic GHG emissions in 2010, about 16% (~7.8 GtCO₂eq y⁻¹) came from CH₄ (Victor et al., 2014).

An important source of anthropogenic CH₄ 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 CH₄ emission source, while in the United States, they are the third largest (20%) (EEA 2016, USEPA 2016). On a global scale, CH₄ emissions from landfills are estimated to account for about 8% of anthropogenic CH₄ emissions (Blanco et al., 2014). Between 1970 and 2010, CH₄ 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 CH₄ generated in global landfills is captured at sanitary landfills (Themelis and Ulloa, 2007), and so controlling fugitive CH₄ 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 CH₄ annually, it should be reported to the European Pollutant Release and Transfer Register (CEC, 2006).

97 The amount of CH₄ emitted from landfills is often estimated using theoretical gas
98 generation models, applying standard conditions with respect to waste composition and surface
99 CH₄ oxidation. The first-order decay (FOD) model is generally recognised as being the most
100 widely used approach, as recommended by the Intergovernmental Panel on Climate Change in
101 the IPCC Waste Model (IPCC, 2006). The FOD model assumes that the degradable organic
102 matter in waste decays slowly over a few decades, during which time CH₄ and CO₂ are formed.
103 If conditions are constant, the rate of CH₄ production depends solely on the amount of carbon
104 remaining in the waste. The FOD models in the IPCC (2006) guidelines are multi-component
105 with individual values for CH₄ generation potential and CH₄ generation rate constants from the
106 degradable organic carbon contained in various waste fractions. In some countries other FOD
107 models have been developed to estimate CH₄ emissions from specific landfills – examples
108 being LandGEM (US) (Alexander et al., 2005), GasSim (UK) (Gregory et al., 2003) and
109 Afvalzorg (The Netherlands) (Afvalzorg, 2015; Scharff & Jacobs, 2006). The landfill CH₄
110 emission is obtained by subtracting the fraction of CH₄ that is collected and the fraction of the
111 uncollected CH₄ that is oxidised from the modelled CH₄ generation. At landfill sites with gas
112 collection, the amount of gas collected is often well-known, whereas the fraction of CH₄
113 oxidised is unknown. The IPCC (2006) suggests a default value of maximum 10%, which has
114 been highly debated (e.g. Chanton et al., 2009). Stable carbon isotopic analyses have been used
115 for in-situ landfill CH₄ oxidation quantification (e.g. Chanton et al., 2008a; 1999; Börjesson et
116 al., 2007); however, the validity of several assumptions in the methodology remains
117 unresolved. FOD gas generation models have been complemented with more recent, site-
118 specific process-based emission models that take into account local conditions such as soil
119 type, climate and CH₄ oxidation rates to calculate overall CH₄ emissions. One example in this
120 regard is the Californian process-based CH₄ emission model CALMIM, which includes site-

121 specific seasonal soil microclimate and CH₄ oxidation data (Spokas et al., 2015; 2011; Bogner
122 et al. 2014; Bogner et al., 2011; Spokas and Bogner, 2011). The CALMIM model assumes that
123 gas transport solely is governed by diffusion processes, while a similar model, the LandSEM
124 model, also includes advective gas transport (Abichou et al., 2015). The process-based cover
125 models estimate methane emission from the landfill cover using representative soil
126 characteristics, and are thus not able to include emissions from landfill infrastructures such as
127 leachate wells. Only few of the available models have been validated, and it is uncertain as to
128 what extent they describe actual CH₄ emissions (Cambaliza et al., 2017; De la Cruz et al.,
129 2016; Mou et al., 2015; Spokas et al., 2015; Bogner et al. 2014; Scheutz et al., 2011a; 2011b;
130 Scharff and Jacobs, 2006). In addition, input parameters such as deposited waste amounts and
131 compositions are often of low quality or even unknown, and no single model is applicable to
132 all landfills, due to the significant variations in landfill designs and operation (construction and
133 aftercare), meteorological conditions (precipitation, temperature, wind, etc.) and landfill cover
134 design and conditions. To assess landfill CH₄ emissions, and to evaluate the efficiencies of
135 mitigation initiatives such as gas collection, recovery systems and CH₄ oxidation installations,
136 measurement methods that can directly quantify CH₄ emissions are thus needed. Furthermore,
137 the quantification of CH₄ emissions based on measurements will provide more accurate
138 estimations and help evaluate the performance of currently used landfill gas generation and
139 emission models.

140 Several measurement techniques are currently available, but none of them has been
141 recognised as an international reference method. In addition, very few are commercially
142 available – and then only in a handful of countries. A few new methods have been developed
143 in the last few decades, albeit only in a small number of cases have the measurement
144 techniques been compared to existing techniques and/or to gas generation models used in

different countries. The main challenge in measuring CH₄ emissions from landfills is their temporal and spatial variability. Several studies have shown high spatial variations in landfill surface CH₄ 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 CH₄ 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 CH₄ 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 Gröngroft, 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, CH₄ 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).

169 Temporal and spatial emission variations, combined with the large area and challenging
170 topography of most landfills, make CH₄ emission quantification a challenging task.

171 In general, landfill emission measurement techniques can be divided into two
172 categories: surface emission factor techniques and mass emission techniques, also called
173 ‘bottom-up’ and ‘top-down’ techniques. Surface emission factor techniques are based on
174 quantifying emissions emanating from part of the surface of the landfill and obtaining an
175 emission factor, which then is applied to the remaining areas of the landfill. Mass emission
176 techniques, on the other hand, involve quantifying the landfill emission from a larger area or
177 the whole site. The objective of this paper is to provide a review of the different measurement
178 techniques by explaining the techniques as well as their applications, advantages and
179 limitations when quantifying an entire landfill’s CH₄ emission. A second objective is to review
180 studies on landfill CH₄ emissions, using one or more of the measurement techniques, and then
181 compare and reflect on these studies to enable the reader to choose the best method for his/her
182 application.

183

184 **2. Methane measurement techniques**

185 The different measurement techniques available for quantifying CH₄ emissions from
186 landfills all measure against different scales. Measurements are made from the landfill surface,
187 to several kilometres away (Fig. 1) and across different timescales, from minutes, to weeks or
188 months. Measuring at the surface of the landfill has the advantage of excluding interference
189 from surrounding CH₄ sources, but it also involves the challenge of having to extrapolate the
190 measurements to represent a large area with heterogeneous emissions, as it is practically
191 impossible to cover the whole landfill area. Methods measuring at elevated heights are able to
192 evaluate emissions from a larger area, thereby making extrapolation to full landfill size more

193 applicable, while methods measuring at a greater distance away are able to quantify whole-site
194 emissions, though they are more sensitive to potential surrounding sources. Table 1 provides
195 an overview of qualitative reconnaissance techniques used to locate CH₄ emission hotspots,
196 while Table 3 provides an overview of the various CH₄ measurement techniques employed for
197 emission quantification at landfills. In the following sections, each technique is described in
198 detail and its advantages and limitations are discussed.

199

200 **2.1 Qualitative reconnaissance techniques**

201 *Methodology.* A common reconnaissance technique involves screening the landfill surface
202 for CH₄, using a handheld gas analyser (such as a portable flame ionisation detector (FID), or other
203 analysers based on optical or electrochemical measurement principles, gauging concentrations in
204 ppm just above the landfill cover. A screening campaign can be performed by dividing the landfill
205 into sections, whereby CH₄ recordings are taken systematically at grid points or alternatively during
206 walkovers (e.g. Scheutz et al., 2011a; 2011c). A survey of CH₄ concentrations may give an
207 indication of certain hotspots or installations with CH₄ leakages. In the US, landfill operators are
208 legally obliged to carry out CH₄ surface screenings four times a year, and this is also mandatory in
209 the United Kingdom. Surface CH₄ screenings can be extended to include carbon dioxide (CO₂)
210 concentrations, for example by using a handheld infrared (IR) detector or a portable cavity ring
211 down spectrometer (CRDS), the latter of which has the advantage of being fast and able to measure
212 concentration changes in sub-ppm levels. Recent developments of the method have made it portable
213 (e.g. Juncher Jørgensen et al., 2015). Elevated CO₂ to CH₄ concentrations may indicate areas
214 experiencing CH₄ oxidation (confer section 2.3).

215 Theoretically, surface air concentrations recorded at a constant and short distance away from
216 the soil/atmosphere interface should be positively correlated with surface fluxes. However, this

217 correlation depends on interactions between the measuring device (e.g. pumping flow), the ground
218 (e.g. soil characteristics, vegetation) and the atmosphere (weather conditions) – and thus it is not
219 constant in time and space. Lando et al. (2017) found that landfill surface CH₄ concentrations
220 recorded using an open-path handheld laser detector were inversely proportional with wind speed,
221 and thus the authors recommended that surface concentration measurements should be performed at
222 wind speeds < 1 m s⁻¹. It has been suggested that CH₄ flux (after proper calibration) can be
223 quantified from simple measurements of CH₄ concentrations close to the landfill surface (Lando et
224 al., 2017; Gonzalez-Valencia et al., 2016; 2015; Park et al., 2016; Mackie and Cooper 2009).
225 However, CH₄ flux vs. surface concentration measurements often show relatively weak and varying
226 correlations, depending on the measuring time and location, and they are thus associated with large
227 uncertainties (Lando et al., 2017; Gonzalez-Valencia et al., 2016; 2015; Franzidis et al., 2008).

228 Other methods for identifying CH₄ emission hotspots include the use of thermal infrared
229 cameras that measure temperature differences (thermal anomalies) (Fjelsted et al., 2019; Tanda et
230 al., 2017; Battaglini et al., 2013; Lewis et al., 2003). Areas with elevated temperatures may be
231 caused by the leakage of landfill gas, which is warmer than the surroundings. Thermal infrared
232 photography is best carried out at times when there are large temperature differences between the
233 landfill gas and the surroundings, e.g. in the early morning when the air is cold. However, a number
234 of factors, other than emissions of warm LFG, can cause temperature differences and result in
235 measurement bias, which was thoroughly investigated in Lewis et al. (2003). Warmer areas in the
236 surface can be caused by high biological activity, e.g. soil or compost respiration or CH₄ oxidation.
237 Measurement bias can also be caused by variability in emissivity of different surfaces. Different
238 vegetation and soil conditions on the surface, having different emissivity, will cause temperature
239 anomalies in thermal infrared images. As an example, dark areas, such as soil surfaces without
240 vegetation or a dark plastic liner, will have higher heat absorption, and thus they will appear as

241 warm areas in an infrared image and result in a false indication of a CH₄ emission. Finally, a certain
242 minimum CH₄ flux can be expected before a temperature anomaly becomes detectable. Fjelsted and
243 co-workers (2019) concluded that the minimum observable CH₄ emission for the thermal infrared
244 camera to identify an emission hotspot was 150 g CH₄ m⁻² d⁻¹ occurring from an area of more than 1
245 m². In combination, these precautions most likely explain why studies presented in the literature
246 remain somewhat inconclusive on the usefulness of thermal infrared technologies for identifying
247 landfill CH₄ emissions (e.g. Fjelsted et al., 2019; Capodici et al., 2015; Beaumont et al., 2014;
248 Battaglini et al., 2013; Desideri et al., 2007; Ishigaki et al., 2005; Raco et al., 2005; Lewis et al.,
249 2003).

250 Both thermal infrared cameras and lasers capable of measuring CH₄ could be integrated with
251 unmanned aerial vehicle (UAV) technology to overfly the landfills (e.g. Fjelsted et al. 2019;
252 Tanda et al., 2017; Beaumont et al., 2014).

253 Finally, a visual inspection of the landfill surface is a simple and useful method, in that
254 cracks in the surface, vegetation damage and damage to the gas collection system are potential areas
255 of CH₄ emissions. Table 1 provides an overview of qualitative reconnaissance techniques that can
256 be used to study areas with CH₄ emissions.

257 *Advantages and limitations.* The advantages of the qualitative reconnaissance techniques
258 described herein are generally that they provide an opportunity for a relatively fast, easy and
259 inexpensive overview of areas or installations with CH₄ emissions that can subsequently be
260 repaired. Analytical devices are often easy to employ and the measurements can be performed by
261 landfill staff. Qualitative reconnaissance techniques are thus valuable tools in the daily operation
262 and maintenance of the landfill cover and of site installations such as the gas collection and
263 recovery systems. However, it must be emphasised that none of the reconnaissance techniques
264 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 CH₄ 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 CH₄ fluxes can thus not be determined based on above-surface CH₄ concentration readings. Where landfill surface CH₄ 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 (CH₄, CO₂, oxygen (O₂) 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 CH₄ 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, CH₄ and CO₂ concentration gradients of the cover layer can provide information on whether CH₄ oxidation is occurring and at what depth it takes place (Scheutz et al., 2009; 2008; 2003). CH₄ and CO₂ gas concentration profiles have also been used to quantify CH₄ 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).

289 It is important to be aware, however, that processes other than microbial CH₄ oxidation
290 contribute to CO₂ generation, e.g. soil respiration, and thus the method could lead to an
291 overestimation of the CH₄ oxidation. It is therefore advised that background measurements
292 (measurements performed in areas or soils without landfill gas exposure) are performed in
293 order to assess background soil respiration. In some settings such as compost-based landfill
294 covers, CO₂ generation caused by compost respiration can be a significant or even the
295 dominant CO₂ generating process, and the method will thus not be applicable for CH₄
296 oxidation evaluation.

297 Vertical soil gas concentration profiles can also be used in combination with soil
298 sampling at the respective cover depths, and following subsequent incubation in the laboratory
299 (by adding CH₄ and O₂ and subsequently measuring CH₄ turnover) the CH₄ oxidation potential
300 of the cover soil can be determined. Incubation tests can be performed in different conditions,
301 such as testing for temperature and moisture content, which can provide a better understanding
302 of the influence of weather and climate conditions on CH₄ oxidation (Scheutz et al., 2009;
303 2004).

304 Vertical soil gas concentrations combined with pressure gradient measurements in
305 landfill covers can be used to calculate the flux of CH₄ through the landfill cover (El-Fadel et
306 al., 1995; Jones and Nedwell, 1990; Young, 1990). The advective flux can be calculated using
307 Darcy's law, i.e. $u = -K \cdot (dP/dz)$, where u is the mass flux of gas (in m³ s⁻¹ m⁻²), K is the
308 medium's permeability for gas flow (in m² s⁻¹ Pa⁻¹) and dP/dz is the pressure gradient at depth
309 z (in Pa m⁻¹) (Young, 1990). Often, CH₄ flux will be governed by both advection and diffusion,
310 where diffusive flux is driven by concentration differences. In such cases, Fick's first law of
311 diffusion should be included, which requires knowledge of a number of factors such as
312 diffusivity, porosity, water content and gas viscosity (Kjeldsen, 1996; El-Fadel et al., 1995).

313 For a more correct description of the diffusion process, Stefan Maxwell equations may be
314 required instead of Fick's first law in case gas compositions vary significantly in the modelled
315 soil domain (Molins et al., 2008).

316 *Advantages and limitations.* Vertical soil gas concentrations combined with pressure
317 gradient measurements have only been used a few times for evaluating a whole landfill's CH₄
318 emissions (Bogner et al., 1995); however, the method has been used for evaluating the
319 influence of atmospheric pressure on CH₄ emissions (El-Fadel et al., 1995; Young, 1990). The
320 technique lacks spatial (small sampling area of < 0.5 m²) and temporal resolution (samples are
321 taken within 10-20 minutes), which is in conflict with the often heterogenic emissions escaping
322 from landfill surfaces. Emissions from hotspots, such as cracks in the top cover, or from
323 leachate wells cannot be captured using the gas concentration profile method. Additionally, the
324 method is very labour-intensive. Its main advantage, though, is the ability to obtain useful
325 information on mechanisms influencing the emissions, such as rapid changes in atmospheric
326 pressure, the effect of wind velocity and relative emissions as the result of convection and
327 diffusion. Soil gas profile measurements can also be useful in understanding CH₄ sources and
328 sinks, and they can therefore be useful for evaluating CH₄ oxidation (Röwer et al. 2011; Gebert
329 et al., 2011; Christophersen et al., 2001) and landfill gas migration (Christophersen and
330 Kjeldsen, 2001; El-Fadel et al., 1995). Christophersen et al. (2001) studied lateral gas
331 migration and oxidation at an old, completely and finally covered landfill, using gas profiles
332 combined with static flux chambers, finding seasonal variations with no CH₄ emissions during
333 summer, due to increased oxidation in the soil. They also found that atmospheric pressure,
334 temperature and especially soil moisture content had an influence on CH₄ emissions, while off-
335 site migrating CH₄ was oxidised before entering into the atmosphere.

336 In summary, vertical soil gas concentration profiles have limited applications as a
337 standalone method for quantifying entire landfill CH₄ emissions. The method has only been
338 used for CH₄ flux quantification in a few studies dating back to the 1990s, but it can
339 nevertheless provide valuable information about landfill gas migration, oxidation and
340 influencing factors (e.g. changes in weather conditions), and this supporting information could
341 be useful in combination with other quantitative emission methods.

342

343 **2.3 Surface flux chambers**

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

351

352 *2.3.1 Closed surface flux chambers*

353 *Methodology.* Closed flux chambers – also referred to as ‘static chambers’ – are
354 historically likely to be the most commonly used techniques for estimating CH₄ emissions
355 from landfills. These closed chambers can be set up with or without recirculating the analysed
356 gas flow. The recirculation mode is preferably used if the CH₄ analyser withdraws a significant
357 gas volume from the chamber, in order to avoid errors on the flux measurement by removing
358 gas as part of the sampling activity. The chamber technique was originally developed to
359 measure gas emissions from natural soils (e.g. Rolston, 1986), where surface emissions are

360 controlled by diffusion and happen more homogeneously, and so minimal emission variations
361 are expected over the emission area. The CH₄ emission flux from the surface covered by the
362 chamber is calculated from measuring changes in CH₄ concentrations inside the chamber:

$$Flux = \frac{dC}{dt} \cdot \frac{V}{A} \quad \text{Eq. 1}$$

364
365 where dC/dt is the change in concentration over time (mole m⁻³ h⁻¹), V is the volume of the
366 chamber (m³) and A is the surface area covered by the chamber (m²).

367 The closed flux chamber has been used widely for quantifying gas emissions from
368 landfill surfaces, where emissions are controlled by both pressure and concentration gradients
369 (e.g. Lucernoni et al., 2016; Rachor et al., 2013; Abichou et al., 2011; Bogner et al., 1995;
370 Kjeldsen and Fisher, 1995). Different chamber configurations and sizes have been used, from
371 small-size (chamber footage area less than 0.1 m² e.g. Scheutz et al. 2014; 2011) and mid-size
372 (0.4-1 m² e.g. Christophersen et al., 2001), to over-size (larger than 15 m² e.g. Geck et al.,
373 2016). A theoretically optimal chamber design was suggested by Lucernini et al. (2016) based
374 on computational fluid dynamics simulations. In many cases, the chambers have been equipped
375 with mechanical or electrical fans to assure the complete mixing of gases inside the chamber
376 during sampling. In order to take repeated measurements at the exact same locations, chambers
377 can be supplemented with a frame that is permanently installed in the landfill cover, topped by
378 a removable lid (Pratt et al., 2013; Schroth et al., 2012; Christophersen et al., 2001). In order to
379 minimise errors caused by increased pressure and concentration build-ups inside the chamber,
380 measurements are often done over a very short time period, and preferably no more than a few
381 minutes. A pressure build-up can be avoided by installing a vent consisting of a long small-
382 diameter tube, which can equalise pressure between the chamber and the surrounding air.

383 Losses due to diffusion along the vent tube have been considered negligible (Hutchington and
384 Mosier, 1981). However, due to the venturi effect, vents may potentially introduce more errors
385 than they solve. Vented chambers have been used in a CH₄ emission study performed on an old
386 landfill (Rachor et al., 2013).

387 Due to its easy setup and inexpensive use, the chamber technique has been suggested
388 for measuring entire landfill emissions (UK Environment Agency, 2010). To obtain a whole-
389 site CH₄ emission rate, measurements are sometimes done by using randomly distributed
390 chambers or a systematic grid of single-point measurements, following which emissions for the
391 entire site are found by calculating an average of the measured emissions multiplied by the
392 landfill area, or by using geo-statistical tools (e.g. Gonzales-Valencia et al., 2016; Battaglini et
393 al., 2013, Ishigaki et al., 2005).

394 *Advantages and limitations.* The surface flux chamber method is very simple to employ
395 and does not require advanced analytical equipment; CH₄ fluxes, for example, can be measured
396 with an FID or an IR detector. The method is able to detect small fluxes of CH₄ – in most cases
397 positive (CH₄ release), though they can also be negative (uptake of atmospheric CH₄). The
398 surface flux chamber method is the only technique currently applicable for determining the
399 flux of trace gases contained in LFG (non-methane volatile organic compounds; NMVOCs)
400 (Scheutz et al., 2008; 2003). The flux chamber method, sometimes combined with soil gas
401 profile measurements, is a well-proven and appropriate method for quantifying CH₄ oxidation
402 at a specific location on the surface (using the carbon mass balance method developed by
403 Christophersen et al., 2001), or for studying environmental factors controlling CH₄ emissions
404 and oxidation. As mentioned in section 2.2., using CO₂ surface fluxes to quantify CH₄
405 oxidation could result in overestimations, and the CO₂ flux contribution from soil or compost
406 respiration should thus be determined either by performance of background flux measurements

407 or by laboratory soil/compost incubation studies (Scheutz et al., 2017). Alternatively, by
408 measuring the stable carbon isotope composition of CH₄ in the flux chamber, in-situ CH₄
409 oxidation can be determined (Chanton and Liptay, 2000; Liptay et al., 1998). However, the use
410 of stable carbon isotopes to quantify CH₄ oxidation should be interpreted with caution, as the
411 isotopic fractionation during microbial oxidation is influenced by a number of factors,
412 including temperature, soil moisture, CH₄ and O₂ supply (e.g. Gebert et al., 2017; Chanton et
413 al., 2008b). Additionally, processes other than microbial activities contribute to isotopic
414 fractionation, such as transport processes (Gebert et al., 2013). Several studies have used flux
415 chambers to observe, for example, the influence of changes in atmospheric pressure,
416 precipitation or temperature on the emission of CH₄ (e.g. Fredenslund et al. 2010; Gebert and
417 Gröngröft, 2006; Czepiel et al., 2003; Christophersen and Kjeldsen, 2001; Kjeldsen and
418 Fischer, 1995).

419 One of the limitations of the surface flux chamber is that it is intended for measuring
420 diffusional surface fluxes. At landfills, surface CH₄ emissions are governed by both diffusion
421 and advection, the most dominant of which is site-specific and can even vary between different
422 areas of the landfill, e.g. hotspots versus non-hotspots. At sites (and/or during specific time
423 periods) where the flux is controlled mainly by pressure differences over the landfill cover,
424 surface flux can be underestimated, due to a fast pressure build-up in the static chamber
425 (Senevirathna et al., 2007; Conen and Smith, 1998).

426 The main limitation of using closed surface flux chambers is that the measured
427 emission represents only the small area covered by the flux chamber itself (often 0.5-1 m²),
428 and it only captures the emission over a short period of time (minutes). In this way, the method
429 has the same spatial and temporal limitations as the pore gas concentration profiles mentioned
430 in the previous section. In an attempt to obtain a reliable estimate of the total CH₄ emission

431 from a landfill or section thereof, it is often proposed to apply a systematic testing strategy
432 (Gonzalez-Valencia et al., 2015; Battaglini et al., 2013; Bour, 2007; Spokas et al., 2006; Lang,
433 2004; Rosevaer et al., 2004; Savanne et al., 1997; Bogner and Scott, 1995). Such a sampling
434 strategy may be that the landfill is divided into grids of a certain size, followed by flux
435 chamber measurements performed in the grid points. Geo-statistic models can then be used to
436 estimate total CH₄ emissions from the landfill (Gonzalez-Valencia et al. 2015; Spokas et al.,
437 2003; Börjesson et al., 2000). Typical distances between the grid points are between 10 and 60
438 metres, and the more measurements performed, the more accurate a result can be expected.

439 Back in the 1990s, the UK Environment Agency developed a best practice guideline on
440 how to measure total CH₄ emissions from a landfill by creating a network of flux chamber
441 measurements and integrating individual flux measurements (UK Environment Agency, 2010).
442 Large numbers of measurements are needed in this regard, and the UK EPA suggests these
443 should be based on the size of the landfill area (or the area to be investigated), with a minimum
444 of six measurements and an additional number of measurement points equal to 0.15 times the
445 square root of the area (in m²) (UK Environment Agency, 2010). This results in 16
446 measurement points for a 5000 m² site, 53 points for 100,000 m² and 156 points at 1 million
447 m². However, even when performing a very large number of measurements, the measured CH₄
448 emission will only represent what has leaked out from a very limited area (<1%) of the total
449 landfill. In addition, a significant part of the emitted CH₄ occurs through small hotspots, as
450 demonstrated by Rachor et al. (2013), and the likelihood of capturing one of these hotspots
451 with a grid of chamber measurements is very small; thus, underestimating total landfill
452 emission is to be expected from the surface flux chamber method (Scheutz et al., 2011c;
453 2011b; Babilotte et al., 2011; Green et al., 2010; Fredenslund et al., 2010; 2007; Goldsmith et
454 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 CH₄ 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

480 the chamber comparable with ambient pressure, in order to obtain reliable emission rates.
481 Similar to the closed chamber, an open chamber can be equipped with a vent to equalise
482 pressure between the chamber and the surrounding air. Guidance on how to use both closed
483 and open flux chambers was published by Eklund (1992). Open flux chambers have also been
484 used in a few landfill emission studies as an alternative to the closed chamber approach, to
485 measure CH₄ emissions through landfill covers (Gallego et al., 2014; Park and Shin, 2001;
486 Reinhart et al., 1992).

487 *Advantages and limitations.* The advantage of the open flux chamber is that it is
488 continuously flushed with air, which limits the risk of pressure build-up and gas concentration
489 accumulation underestimating the measured flux. Pressure built up inside the chamber will
490 affect the flux through advection, whereas an accumulation in gas concentrations inside the
491 chamber will affect the diffusional flux. In both cases, this can result in non-linear gas
492 concentration accumulation curves, which will lead to underestimating the surface flux. In
493 addition, using an open flux chamber allows measurements over a longer period of time (e.g.
494 Huber-Humer and Lechner, 2001a; 2001b; Tregoures et al., 1999; Verschut, 1991). The flow
495 through the open chamber (also referred to as the ‘carrier gas’) can consist of ambient air, or
496 alternatively a synthetic gas can be used. If ambient air is used, it is important that the
497 background concentration of the gaseous compound being analysed is constant. At landfill
498 surfaces, CH₄ background concentration will often not be constant, which is why it may be
499 necessary to use a synthetic gas as a carrier when using open chambers. In comparison to
500 closed chambers, the set-up for open chambers is more demanding in terms of pumps required
501 to control the flow through the chamber and potentially synthetic gas containers, which will
502 also make it more time-consuming to perform a flux measurement. Regarding the temporal and
503 spatial challenge, open flux chambers have the same advantages and disadvantages as closed

504 flux chambers, and if employed at landfill sites with emission hotspots, one will likely
505 underestimate entire landfill site emission.

506 In summary, heterogeneous emissions from landfills make it difficult to quantify total
507 CH₄ emissions using the flux chamber approach. In spite of several studies using the approach
508 to quantify whole landfill site emissions, this approach remains unvalidated in terms of its
509 appropriateness for estimating accurately total CH₄ emissions, and thus there is a need for
510 further documentation (including appropriate numbers of measurements and statistical methods
511 for data interpretation) and testing against other measuring methods. Currently, flux chambers
512 are not recommended for whole landfill site CH₄ emission quantification. However, flux
513 chambers can be an important tool for obtaining emission rates for smaller areas, such as an
514 engineered biocover for CH₄ oxidation (e.g. Scheutz et al. 2017; 2011b; Geck et al., 2016) or
515 smaller scale landfill cover studies (e.g. covered vs. non-covered cells, slopes vs. none-sloped
516 area, etc.) (Wang et al., 2017; Di Trapani et al., 2013; Scheutz et al. 2008).

517

518 **2.4 The eddy covariance method**

519 *Methodology.* Micrometeorological eddy covariance measurement relies on the fact that
520 emitted gases are mixed vertically by turbulent eddies in the atmospheric inversion layer. The
521 method is often used for measuring emissions from larger surfaces such as fluxes of CH₄, N₂O
522 and CO₂ from agricultural soils (e.g. Kroon et al., 2010; Hensen et al., 1996), but it has also
523 been applied on landfills (Xu et al., 2015; McDermitt et al., 2013; Schroth et al., 2012; Eugster
524 and Plüss, 2010; Lohilla et al., 2007; Laurila et al., 2005). In a micrometeorological eddy
525 covariance setup, continuous measurements of the vertical gas mass fluxes of CH₄, CO₂ and
526 water are often carried out (Fig. 3). The vertical flux of CH₄ at this height takes place mainly
527 through convection, and any emissions can therefore be found from CH₄ concentrations and

528 local vertical wind velocities. Due to air turbulence, the wind will sometimes have an upward
529 component and at times a descending component. In the event that there is an emission from
530 the surface, the concentration of CH₄ through the plane will be slightly higher when the wind
531 is rising, and so an average positive CH₄ flux (in unit mass per time) can be measured. CH₄
532 concentrations and vertical wind velocities are measured continuously and at high frequency (~
533 10 times per second). The measured data are then stored and the average CH₄ emission is
534 calculated for time intervals, for example of 15 minutes.

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

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

543 *Advantages and limitations.* One of the advantages of micrometeorological eddy
544 covariance measurements is the possibility of having continuous measurements over longer
545 time periods, as well as measuring both CH₄ and CO₂, depending on the analytical device. The
546 method can run automatically for months, providing insights regarding temporal variability as
547 well as average emissions. Xu et al. (2015) measured the response of CH₄ emissions to change
548 in barometric pressure over a six-month period. Combined with other information such as
549 landfill gas composition and gas recovery, the method can provide an indication of total
550 landfill gas production and CH₄ oxidation (Xu et al., 2015; Lohila et al. 2007; Scharff et al.,
551 2003). However, a comparison of CH₄ to CO₂ 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 topography of a landfill is far from the flat surface that offers the ideal conditions for applying this method. Despite the challenges of topography 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.

576 The method is also called the ‘1D mass balance method’, as it measures CH₄ flux that passes
577 the target tower (1D) and not through a larger vertical plane (2D) as in other mass balance
578 methods (see Sections 2.6, 2.7 and 2.9). It is important that the measured CH₄ concentrations
579 are corrected for any background concentration of CH₄, as the CH₄ flux will otherwise be
580 overestimated. This is done by measuring at the top of the tower, assuming that the background
581 concentration has the greatest influence on the measured CH₄ concentration at this point. The
582 height of the measurement point determines the source area contributing to elevated CH₄
583 concentrations, and when measurements are taken at a sufficient height above the landfill, in
584 theory, the whole CH₄ plume is measured and the total CH₄ emission flux is obtained (Scharff,
585 2003). Normally, however, this requires a very high tower, and in practice a lower tower is
586 used, thereby obtaining data from a smaller part of the landfill. By performing measurements
587 at a maximum height of 10 metres, emanations from an area with a radius of approximately
588 150 metres away from the tower will be covered (Scharff, 2003). The specific location and size
589 of the emission area contributing to the measured emissions is determined by dispersion
590 modelling, for which knowledge about atmospheric stability and the surrounding topography is
591 required. Emissions from all directions are measured as the wind direction changes, thus
592 enabling the method to provide emission rates from different areas of the landfill. It can be set
593 up for automated, continuous measurements over a longer period, thereby enabling long
594 emission data time series (weeks to months).

595 *Advantages and limitations.* The mass balance method shares many of the same
596 advantages and limitations as the eddy covariance method. It can be automated so that
597 emissions (CH₄ and other gases CO₂ and N₂O) from the landfill can be measured over a longer
598 period of time, providing information on the variability of the CH₄ emission over time and
599 partly providing a better determination of the average emission. Experiences with landfill

600 emission quantification using the stationary mass balance method are very limited. The method
601 was used for CH₄ quantification at landfills in the Netherlands (Oonk and Boom, 1995) and
602 was found to give similar emission fluxes in a comparison study with an inverse modelling
603 method, along with tracer gas calibration (Scharff, 2003) (Table 3).

604 A major drawback is that the method does not measure emissions from the entire
605 landfill but only from a small part thereof. In addition, it can be difficult to determine exactly
606 which part of the landfill has contributed to the measured emission, so any results thus depend
607 on assumptions about where CH₄ emissions take place. If emissions occur mainly closer to the
608 measuring tower than assumed, the overall emission will be overestimated. It is therefore
609 advisable to perform a qualitative CH₄ screening of the landfill surface, for example using an
610 FID (see section 2.1), to evaluate any relevant areas before a final interpretation of the
611 measured flux is made.

612

613 **2.6 Radial plume mapping**

614 Radial plume mapping (RPM) for measuring landfill CH₄ emissions has been under
615 development since the late 1990s. The method uses a combination of concentration
616 measurements and wind profiles to obtain a surface emission factor from an upwind area. It
617 can be used in two different configurations – horizontal (HRPM) or vertical (VRPM) (Fig. 4),
618 with HRPM providing qualitative information on the location of emission hotspots, while
619 VRPM is used for quantifying emissions.

620 *Methodology.* Quantifying emissions by VRPM is achieved by measuring the mass of
621 CH₄ crossing two vertical planes located upwind and downwind of an emission area,
622 respectively. CH₄ fluxes across the vertical planes are determined from the product of
623 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

648 from the entire landfill, several measurements must be taken, or alternatively it must be
649 assumed that the measured emission is representative of the entire landfill and can then be
650 summarised to determine total emissions. A methodological challenge in this regard is that
651 both wind speed and CH₄ concentration may vary over the measured plane. Thus, ideally,
652 measurements should be carried out simultaneously at different places in the upwind and
653 downwind planes, which require additional lasers, thereby adding considerably to the cost of
654 the instrumentation and the demand for staff. When using one laser, the area contributing to
655 the measured CH₄ flux through the vertical plane is determined by inverse dispersion
656 modelling, thus adding further to the uncertainty of the method. Determining the correct area
657 contributing to the measured emission can be a challenge on landfill sites with complex
658 topography (Abichou et al., 2010; Goldsmith et al., 2008), and at some landfills (or parts of
659 landfills) the method cannot actually be applied. In many sites, part of the waste is deposited
660 above ground, forming a small hill in the landscape. If measurements are carried out such that
661 the plane through which the CH₄ flux is measured is placed on top of the landfill, any emission
662 from the slope will not be included in the measurement, which may lead to a significant
663 underestimation of emissions, as hotspots are often seen on slopes, due to horizontal gas
664 transport and incomplete covers (Babilotte et al., 2009; 2008). Likewise, when reflectors and
665 lasers are placed on the downwind edge of a landfill, CH₄ emitted from an upwind part of the
666 landfill may be at an elevation higher than 10 metres (often the maximum position above
667 ground of the reflector) when it reaches the measurement points, which will add significantly
668 to the uncertainty of the method. An additional disadvantage is that measurements are usually
669 taken in hours, and thus any temporal emission variation can be a challenge to capture.
670 Another drawback is that the method (theory, equipment and application) is generally difficult

671 to understand for third parties, making it difficult for an independent body to assess the results
672 of emission studies performed by this method.

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

681 The method has been tested and used for quantifying 20 landfills in the USA, using a
682 double laser and reflector system (Goldsmith et al., 2012; Abichou et al., 2010). Abichou et al.
683 (2010) found that a change in wind direction could introduce an uncertainty factor of 20% and
684 that the area contributing to the measured flux is a function of the atmospheric stability class
685 with an uncertainty level of 10-30%. Goldsmith et al. (2012) pointed out that the OTM-10
686 method may have an application for the landfill industry, although only 31% of the collected
687 data in their comprehensive study of 20 landfills in the USA fulfilled quality requirements. The
688 method is challenged by the complex topography of a landfill, and there are sites where the
689 method is simply not applicable. In general, the output from RPM can be used to identify
690 emission hotspots, which makes this method useful for performing site surveys before
691 initiating remediation activities; however, it is highly important to assign the correct emission
692 area to the measured emission, which can be difficult at landfills with a challenging
693 topography (Goldsmith, 2008). The upshot of the disadvantages of the method is that it has

694 been used infrequently in recent years and replaced by other methods (tracer gas dispersion,
695 DIAL and most recently aircraft-based mass balance).

696

697 **2.7 Mass balance using aerial measurements**

698 *Methodology.* In recent years, the mass balance approach has been applied using
699 atmospheric CH₄ concentration measurements from aircrafts or UAVs. The method relies on
700 concentration measurements across the downwind plume at several heights, preferably
701 covering the total height and width of the CH₄ plume. The measured concentrations are then
702 used for creating a 2D concentration plane, which is combined with measured wind speed and
703 direction for calculating the flux of CH₄ through the downwind plane – and thus emissions
704 from the landfill. Fast measuring instruments are needed for performing aerial CH₄
705 measurements with sufficient temporal resolution, to create a 2D concentration plane. Such
706 instrumentation includes cavity ring down spectroscopy, quantum cascade laser spectroscopy
707 and thermal-infrared (TIR) imaging spectrometry (Cambaliza et al., 2017; 2015; Tratt et al.,
708 2014; Hirst et al., 2013; Peischl et al., 2013). Measurements using aircraft are usually carried
709 out several kilometres downwind of the landfill, as the aircraft can carry instrumentation
710 sensitive enough to measure concentration differences (plume concentrations above
711 background) at this distance. UAV-mounted instrumentation needs to be lighter in weight.
712 Such instrumentation currently has less sensitivity, and the UAV must therefore measure closer
713 to the landfill than aircraft (Allen et al., 2018). However, the fast development in
714 instrument/sensor technology might soon change this situation.

715 *Advantages and limitations.* One advantage of the method is that aerial measurements
716 do not require access to a landfill. When using an aircraft, each landfill is measured relatively
717 quickly, and so many landfills (and other CH₄ sources) in an area can be measured in one

718 flight. An advantage compared to VRPM and DIAL is that CH₄ concentrations at specific
719 points in the total cross-section of the CH₄ plume can be measured, resulting in a relatively
720 detailed 2D concentration plane. The advantage of the using aircraft for CH₄ plume
721 measurements compared to using UAVs is that measurements performed further downwind of
722 the landfill will give a more uniform and less changeable CH₄ plume, which is easier to model
723 correctly, as the measured concentrations can better be assumed to be constant in comparison
724 to when measurements are performed closer to the landfill, where local wind turbulence and
725 emission variations will result in larger concentration variations at individual x, y and z points
726 in the atmosphere. However, measurements taken far away from the landfill also introduce
727 possible errors, due to reduced CH₄ plume concentrations and to the risk of including CH₄ for
728 surrounding non-landfill emission sources.

729 One limitation is that it is not possible to quantify individual CH₄ sources relatively
730 close to each other, such as farms or wastewater treatment plants near to the measured landfill.
731 Additionally, the creation of the 2D concentration plane is associated with uncertainty, as well
732 as the wind field applied for calculating the flux. Ideally, the wind field (wind speed and
733 direction) should be measured at different heights above ground surface. Another limitation is
734 accessibility to an aircraft, sourcing fast and accurate instrumentation and associated costs.

735

736 **2.8 The tracer gas dispersion method**

737 *Methodology.* The tracer gas dispersion method uses simultaneous measurements of
738 atmospheric CH₄ concentrations and a tracer gas released at the emission source at a known
739 rate (Fig. 5). It is assumed that CH₄ and the tracer gas undergo the same fate (dispersion,
740 chemical/photochemical reaction) in the atmosphere within the time span of the campaign, so
741 for this reason, tracer gases with relatively long atmospheric lifetimes are used. SF₆ has been

742 used in many tracer gas studies, such as research on atmospheric dispersion (Martin et al.,
743 2011), ventilation (Timko et al., 1984) and the CH₄ emission of ruminants (Storm et al., 2012)
744 and gas facilities (Lamb et al., 1995), as it has a very long lifetime in the troposphere (3,200
745 years) and negligible background concentrations (Ravishankara et al., 1993). However, SF₆ is
746 a very potent GHG (23,500 times more potent than CO₂ on a 100-year scale) (Myhre et al.,
747 2013), and so it is not valid for environmental studies. A move has been made toward using
748 N₂O instead (Galle et al., 2001), as it is stable in the atmosphere and measurable, although it is
749 present in significant atmospheric concentrations, currently around 331 ppb (NOAA, 2018).
750 Nonetheless, N₂O is also a strong GHG (298 times more potent than CO₂ on a 100-year scale)
751 (Myhre et al., 2013), which in recent studies has led to the use of acetylene (C₂H₂) as a tracer
752 gas (Rees-White et al., 2019; Mønster et al., 2015; 2014; Foster-Wittig et al., 2015a; Green et
753 al., 2010). Although highly flammable, C₂H₂ is not a potent GHG, it is cheap and it is
754 relatively easy to release at a constant flow rate.

755 Based on downwind atmospheric concentrations of CH₄ and the tracer gas measured
756 during its controlled release, CH₄ emissions from the source can be calculated. The tracer
757 dispersion method is generally divided into either a stationary or a dynamic approach (Fig. 5).

758

759 *2.8.1 The stationary tracer gas dispersion method*

760 The stationary tracer gas dispersion method relies on CH₄ and tracer gas measurements
761 performed at fixed single or multiple points in the plume downwind from the emission/release
762 source. They have been used previously to study emissions from several different sources, such
763 as aromatic hydrocarbons in refinery wastewater (Howard et al., 1992), CH₄ from cattle barns
764 (Kaharabata et al., 2000; Marik and Levin, 1996), CH₄ from slurry tanks (Kaharabata et al.,
765 1998) and ammonia from manure spreading (Galle et al., 2000). The first landfill application of

766 the method was done by Czepiel et al. (1996), using SF₆ as a tracer gas. Before CH₄ and tracer
767 gas measurements were conducted, the downwind plume was located by using an analyser
768 placed in a vehicle. Subsequently, evacuated canisters were placed across the plume transect,
769 in order to sample air in the plume. Similar setups have been used in other studies using
770 evacuated canisters (e.g. Scharff et al., 2009; Jacobs et al., 2007; Scheutz et al., 2007;
771 Tregoures et al., 1999) or sampling bags filled by small pumps (Scharff and Hensen, 2009;
772 Babilotte et al., 2008). For landfills with poor access to downwind plume transects, Galle et al.
773 (2001) developed a method based on air concentration measurements performed at a single
774 point. CH₄ and tracer gas concentrations were measured for hours at a single point downwind
775 of the landfill, using N₂O as the tracer gas, and the method was used for a number of landfills
776 in Sweden (Börjesson et al., 2009).

777 Recent studies have shown that leachate wells can be significant point sources for CH₄
778 emissions (Scheutz et al., 2011a, 2011b; Fredenslund et al., 2010). Fredenslund et al. (2010),
779 for instance, suggested quantifying emissions from leachate wells, using a small-scale tracer
780 gas dispersion method (Fig. 6), whereby tracer gas was released at the bottom of the leachate
781 well and CH₄ and tracer gas were measured at a single point a few metres downwind of the
782 well.

783 *Advantages and limitations.* An important assumption is that the tracer gas should
784 simulate the CH₄ emission. In order to obtain a good mix of tracer gas and CH₄, measurement
785 or sampling campaigns have to be executed at a sufficient distance away from the landfill (e.g.
786 Mønster et al., 2014; Delre et al., 2018). Misplacing the tracer gas can result in significant
787 under- or overestimation of the emission, while the required measuring distance depends on
788 the physical size of the source, atmospheric conditions and the topography between the landfill

789 and the measurement/sampling point (e.g. Fredenslund et al., 2019b; Rees-White et al., 2019;
790 Delre et al., 2018; Taylor et al. 2016).

791 One drawback of the static tracer dispersion method is that it can be hard to locate the
792 CH₄ plume. Samples are thus taken blindly and the method relies on a stable wind direction
793 throughout the sampling period – a significant change in wind direction could move the
794 downwind plume away from the sampling point(s). If the CH₄ emission is not well simulated
795 by the tracer gas, sampling at the fringe of the CH₄ plume and tracer gas plume will result in
796 false emission rates, as the ratio between the gases is not representative of the plume.

797

798 *2.8.2 The dynamic tracer gas dispersion method*

799 The dynamic tracer gas dispersion method is based on performing transects of the
800 downwind plume and thereby measuring CH₄ and trace gas concentrations near to ground level
801 across the whole plume, along with the subsequent integration of the plumes. In order to use
802 this method, a sensitive and relatively fast analytical instrument is needed. A Swedish group
803 developed an FTIR instrument stable enough to be driven around in a van, enabling many
804 successful studies of CH₄ emissions from landfills (e.g. Scheutz et al., 2011b). This method
805 was then developed further to use two or more trace gases for quantifying CH₄ emissions from
806 multiple sources close to each other (Scheutz et al., 2011b).

807 The analytical instruments used for tracer gas dispersion measurements have often been
808 built specifically for this purpose and are large, heavy and power-demanding; consequently,
809 they are expensive to run and require highly skilled personnel to operate and maintain them
810 effectively. Recently, new instruments have been developed based on cavity ring down
811 spectroscopy (CRDS). These instruments are smaller, lighter and easier to mount in a vehicle.
812 The cavity ring down instrument developed for the tracer gas dispersion method focuses on

813 measuring CH₄ and C₂H₂ (tracer gas), and it has been used successfully for dynamic plume
814 measurements at two Californian landfills (Green et al., 2010), 15-30 Danish landfills
815 (Mønster et al., 2015; Fredenslund et al., 2019a), a number of landfills in the UK (Bourn et al.,
816 2019; Rees-White et al., 2019) and 15 US landfills (Foster-Witting et al., 2015a). Besides, it
817 was used in a model comparison study at a new landfill (de la Cruz et al., 2016) and a
818 horizontal stationary plume approach using gas sampling from a weather balloon, for
819 measuring in the plume centre, which was found to be situated at an elevated height (Han et
820 al., 2010) and used for exploring emission variations and possible bias in the tracer gas
821 dispersion method (Matacchiera et al., 2019; Delkash et al., 2015).

822 *Advantages and limitations.* The advantage of the dynamic tracer gas dispersion method
823 is its simplicity, because when the CH₄ and tracer gas plumes are mixed fully, analysis and
824 calculation are relatively straightforward. An important advantage of the method is that it
825 measures total emissions from the landfill, including those from hotspots and installations at
826 the site, and may be used independently of the landfill structure or topography. Moreover, it
827 can be applied at landfills of all sizes. When applied at larger landfills, it may be necessary to
828 release tracer gas at several points (Rees-White et al., 2019; Aghdam et al., 2018; Mønster et
829 al., 2014), but the method can also be used for improving the input data required for inverse
830 modelling. Piccot et al. (1996) and Hensen and Scharff (2003) used tracer gas measurements to
831 obtain accurate information about dispersion conditions during their measurement campaign,
832 thus improving the dispersion model in use. The advantages and limitations of the CRDS-
833 based instrument have been explored and compared recently with simultaneous FTIR
834 measurements (Delre et al., 2018; Mønster et al., 2014). Foster-Wittig et al. (2015a), for
835 instance, published a study involving tracer gas dispersion measurements on a large number of
836 US landfills, including suggestions for using data quality criteria for filtering measurements

837 for downwind traverses where tracer and CH₄ were not significantly mixed. Recent studies
838 recommend that a minimum 10 plume transects, but preferably 15, should be performed to
839 reduce the variability of a measurement (Fredenslund et al. 2019b; Mønster et al., 2014), while
840 learning how to establish the minimum detectable emission rate at individual sources has also
841 been suggested (Delre et al., 2017). In another recent study, Taylor et al. (2016) explored the
842 uncertainty of the tracer dispersion method by modelling downwind concentrations from
843 released tracer gas. They found that the main potential error was the correct placement of the
844 tracer gas bottles, while factors such as wind speed and direction had a much smaller influence
845 but could nevertheless be emphasised by the physical layout of the landfill and the emission
846 areas. Taylor et al. (2016) also concluded that the main uncertainties depend on the landfill and
847 thus are site-specific. The disadvantages of the technique are its dependence on the right
848 weather conditions (wind direction and wind speed) to ensure sufficient gas mixing, combined
849 with proper road access, which makes it difficult to measure at some landfills with limited
850 access and very few surrounding roads. Matarachiera et al. (2019) suggested that modelling
851 could be used as an initial step in planning a tracer gas dispersion measurement campaign
852 (tracer gas placement and measuring distance etc.), but the method alone cannot identify on-
853 site emission sources, and so care must be taken regarding the presence of other CH₄ sources
854 (manure storage tanks, farms, composting facilities) in the surrounding area, which can cause
855 extensive errors in site quantification. Furthermore, the instrumentation is expensive and
856 requires expertise to operate it effectively. An additional disadvantage is that measurements
857 are usually taken in hours (2-6 hours) or over a few days, and thus the temporal variation of
858 landfill gas emission can be a challenge to capture. Recent and current research focuses on
859 method validation (Fredenslund et al., 2019b; Matarachiera et al., 2019; Delre et al., 2018;

860 Mønster et al., 2014) and the establishment of quality criteria for data processing (Fredenslund
861 et al., 2019b; Foster-Wittig et al., 2015a).

862

863 **2.9 Differential absorption LiDAR method**

864 The differential absorption LiDAR (DIAL) method transmits pulsed laser radiation into
865 the atmosphere. Part of this radiation is then backscattered by atmospheric constituents and can
866 be measured by a detector to determine CH₄ concentrations by tuning the laser to the
867 appropriate absorption lines and comparing this with a laser at a slightly different wavelength
868 that is reflected in a similar way to the first laser but not absorbed by CH₄. The laser can be
869 pointed in any direction, and for landfill applications, measurements are taken along several
870 lines of sight, thereby obtaining a vertical concentration distribution downwind of the landfill
871 or sections thereof for which emissions are to be quantified (see Fig. 7). This allows for
872 capturing the entire vertical profile of the CH₄ plume, and emissions from the landfill (or
873 landfill section) can be calculated by combining the vertical concentration profile with wind
874 speeds at different heights (Innocenti et al., 2017; Robinson et al., 2011; Babilotte, 2011;
875 Babilotte et al., 2010).

876 Measurements are normally taken downwind and close to the landfill, in combination
877 with upwind measurements, to check for interfering sources and to obtain background
878 concentrations. Individual landfill sections can also be measured by ‘shooting’ the laser in a
879 vertical plane immediately downwind of the targeted section at the landfill (Babilotte, 2011). A
880 moveable DIAL system is a large setup mounted on a truck, and very few are currently
881 available worldwide, and it has been applied mainly for quantifying fugitive emissions caused
882 by petrochemical processes, while its use at landfill sites is still limited (Innocenti et al., 2017;
883 Bourn et al., 2019; Babilotte, 2011; Robinson et al., 2011; Babilotte et al., 2010).

884 *Advantages and limitations.* The main disadvantage of the DIAL method is the cost and
885 size of the analytical setup and the complexity involved in data handling. The truck
886 transporting the setup requires good, smooth roads for access, and measurements depend on
887 wind direction and infrastructure. In addition, stable and accurately measured wind conditions
888 are needed for precise flux quantification. Wind speed above ground level should be measured
889 at different heights for optimal modelling of the wind profile. As with other remote sensing
890 measurements, DIAL is sensitive to possible interfering sources in the vicinity of the measured
891 area. Measurements are conducted over hours or days, and long-term temporal variations can
892 therefore be difficult to capture. The advantage of the DIAL method is that it can measure CH₄
893 over a distance of 400 - 800 m, depending on atmospheric conditions, hence enabling
894 measurements of the total vertical concentration plane of small landfills and thereby
895 eliminating the effect of spatial emission variation. At larger landfills, it is necessary to
896 perform more measurements, due to the limited range of the laser. In such cases, the challenge
897 is to determine which area contributes to the CH₄ flux measured through the vertical profile.
898 The NPL (National Physics Laboratory) in England has two complete DIALs and has
899 conducted measurements at several landfill sites in the country (Innocenti et al., 2017; Bourn
900 et al., 2019). At a large landfill, a measurement campaign usually takes up to one week, with
901 the first day used to prepare the analytical equipment. In the following days, the truck is
902 moved around the landfill (or along its edge) and measurements are taken from different
903 sections to be added up at the end of the measurement campaign. The DIAL has the option of
904 measuring other atmospheric species by scattering light in infrared or ultraviolet spectra, such
905 as ethane, hydrogen, sulphur oxide, ozone and benzene, below ppm level (Babilotte et al.,
906 2010).

907

908 **2.10 Inverse dispersion modelling method**

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

914

915 *2.10.1 Inverse modelling – stationary*

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

emissions (Westbrook et al., 2007), CH₄ emissions from a Canadian landfill (Wyles et al., 2010) and suggested as a method to improve the planning of tracer gas dispersion measurements (Matacchiera 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 CH₄ 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 CH₄ 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

956 kilometres downwind of the site, depending on the size of and emission rate from the landfill.
957 The further the distance to the landfill, the more the spatial variation of the emission will be
958 minimised, and the landfill can be seen as a single-point source in the calculations of emission
959 rates, which is an important assumption when, for example, using a simple Gaussian model.
960 The analytical equipment used for these kinds of measurements needs to be able to measure
961 concentration differences of down to a few ppb, with a background concentration of CH₄
962 normally sitting between 1800 and 2000 ppb. Currently available and capable instrumentations
963 include quantum cascade laser spectroscopy, tuneable diode laser spectroscopy or cavity ring
964 down spectroscopy, each of which has its own advantages and disadvantages in terms of
965 temporal resolution and the number of measurable species.

966 *Advantages and limitations.* Mobile plume measurement with inverse modelling has the
967 advantage of measuring the emission plume from the entire landfill site, regardless of size. The
968 measured plume will also include emissions from the slopes of the landfill and the gas and
969 leachate collection systems, which are often difficult to measure using on-site methods.
970 Disadvantages include the need for specialised analytical equipment and a detailed emission
971 model of the landfill, which needs a number of input parameters such as atmospheric stability,
972 surface-induced turbulence and wind speed. For optimal conditions, the landfill should be
973 located in a relatively flat area with a road crossing the downwind plume at a suitable distance
974 away. In addition, accurately measured and relatively stable meteorological conditions are
975 desirable, such as stable wind direction and speed, minimal changes in atmospheric pressure
976 and stable atmospheric conditions in relation to turbulence. Hensen and Scharff (2001) used
977 inverse Gaussian modelling and calibrated their model using controlled emissions of N₂O as a
978 tracer gas, making it similar to a tracer gas dispersion method, albeit with subsequent inverse
979 modelling, as measurements were taken too close to the landfill for total mixing of the tracer

980 and CH₄ plumes. The authors measured three landfills in the Netherlands and found emissions
981 ranging between 40 and 392 kg CH₄ h⁻¹. Lan et al. (2015) measured various CH₄ emissions in
982 the Barnett shale area, combining dynamic plume measurement and inverse Gaussian
983 modelling. The emission quantifications included four landfills, with CH₄ emissions ranging
984 between 86 and 2086 kg h⁻¹ and with an estimated uncertainty smaller than 60%. Lamb et al.
985 (2016) performed inverse modelling on various CH₄ sources in Indianapolis, Indiana, to
986 estimate the whole city's CH₄ emissions, employing a tracer gas release approach for
987 calibration. The measurements were comparable with emissions estimated from the mass
988 balance modelling of aircraft measurements and inverse modelling from single-point (tower)
989 stationary measurements (Lamb et al., 2016). Fredenslund et al. (2019a) used a similar
990 approach, performing fast screenings of 91 Danish landfills, using downwind plume
991 measurements and inverse Gaussian modelling to find landfills suitable for mitigation
992 initiatives. CH₄ emissions were modelled to range from no detectable plume and up to 43 kg h⁻¹.
993 ¹. A good agreement between modelled emission rates with measured emission rates using the
994 tracer gas dispersion method was shown (a correlation coefficient of 0.965 based on 24
995 measurement campaigns). Emission rates based on inverse Gaussian modelling were very
996 sensitive to the choice of terrain and the Pasquill-Gifford stability class, which could affect the
997 emission rate by more than 100% (Fredenslund et al., 2019a). Downwind CH₄ plume
998 measurements combined with inverse Gaussian modelling was thus found useful as a fast
999 screening method but not appropriate for accurate landfill CH₄ emission quantification.

1000 In general, reverse modelling requires large amounts of high-quality input data to
1001 achieve a good emission estimate. Excellent atmospheric condition inputs (e.g. atmospheric
1002 stability, wind speed at different heights and surface-induced turbulence) from the exact area
1003 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 CH₄. 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.

1021

1022 **3. Methane emission comparison studies**

1023 The different methodologies used to quantify CH₄ emissions from landfills, in some
1024 cases, have been used in tandem to verify obtained emission rates (e.g. Lohila et al. 2007;
1025 Spokas et al. 2006; Börjesson et al. 2000; Mosher et al., 1999; Czepiel et al. 1996;). During the
1026 last couple of decades, a number of studies have evaluated available gas emission measurement
1027 methodologies by comparing them in field experiments, using controlled CH₄ 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 CH₄ 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 CH₄ 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-m² landfill test area, using closed flux chambers and eddy covariance. Over five measurement days, the 16 randomly distributed flux chambers showed an uptake of CH₄ (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

1052 same release rate, and releases at three points with different release rates (see Table 3)). In the
1053 controlled release test, Babilotte (2011) found that VRPM slightly underestimated emissions (-
1054 3 to -13%) measured at a distance close to the source (10 m) and by -42 to -48% at a distance
1055 100 m away. The dynamic tracer gas dispersion method (using FTIR instrumentation and N₂O
1056 as a tracer gas) overestimated emissions by between 4 and 19%. The highest error rate
1057 occurred when the source furthest upwind did not release CH₄, thus representing tracer gas
1058 misplacement and causing a higher dispersion of the tracer gas than the released CH₄, therefore
1059 resulting in an overestimation. Plume measurements for the tracer gas dispersion method were
1060 taken 400 m to 450 m away from the release area, which was a sufficient distance to obtain
1061 well-mixed plumes of CH₄ and tracer gas from a relatively small area, with 30 to 44 metres
1062 between emission spots. The DIAL method results showed both under- and overestimations,
1063 ranging from -21 to +19%, with no clear trends. Babilotte (2011) included closed flux
1064 chambers and eddy covariance during measurement campaigns at two landfills. It was
1065 concluded that limited credibility could be given to the closed flux chamber results, due to
1066 high variations (more than 100%) between the individual flux measurements; consequently,
1067 extrapolation to the entire landfill area would be very uncertain. Furthermore, Spokas et al.
1068 (2006) recommended caution when comparing chamber (static) and tracer gas measurements
1069 (static), due to difficulties in determining and comparing the flux footprint areas of the two
1070 methods. The other four methods in Babilotte (2011) showed results with emission rates for
1071 DIAL < Tracer < eddy covariance < VRPM.

1072 Comparison studies have also included inverse modelling. Scharff et al. (2003), for
1073 instance, compared the eddy covariance method with static and dynamic inverse modelling at
1074 four landfills in The Netherlands and found comparable results, although the eddy covariance
1075 method seemed to give a lower CH₄ flux. Tracer gas was used in inverse modelling to calibrate

1076 atmospheric stability parameters (Schaff et al., 2003). In addition, Babilotte et al. (2010)
1077 compared inverse modelling by using dynamic plume concentrations measured 300 m
1078 downwind of a landfill and 20 m downwind of the controlled release point. At the landfill,
1079 inverse modelling provided a significantly higher flux than for VRPM, tracer gas dispersion
1080 (static and dynamic) and DIAL (see Table 1). For the controlled release test, VRPM, both
1081 tracer gas methods (static and dynamic) and inverse modelling all seemed to overestimate
1082 emissions by factors of two to five, while DIAL underestimated by a factor of two. This study
1083 was carried out with a low release rate ($1.8 \text{ kg CH}_4 \text{ h}^{-1}$). Later research showed that DIAL
1084 could quantify down to $3.6 \text{ kg CH}_4 \text{ h}^{-1}$ with an accuracy rate of 20% (Babilotte, 2011).

1085 Recently, Cambaliza et al. (2017) applied four quantification methods at a landfill in
1086 Indiana, USA. Two of the methods (aircraft-based mass balance and tracer gas dispersion)
1087 were capable of measuring the entire site's emissions, whereas the other two methods (VRPM
1088 and static surface chamber) were used to measure emissions from individual parts of the
1089 landfill. The two whole-site methods showed similar CH_4 emission rates: aircraft-based mass
1090 balance between 400 and 980 kg h^{-1} and tracer gas dispersion between 460 and 750 kg h^{-1} ,
1091 which also agreed well with the modelled emission using CALMIM ($690 \pm 120 \text{ kg h}^{-1}$). These
1092 results, however, were not obtained through simultaneous measurements, and thus the
1093 comparison assumes that the performed measurements (20 measurement campaigns done from
1094 2009 to 2012, using tracer gas dispersion and two measurement campaigns for aircraft-based
1095 mass balance done in August 2012 and July 2014) represent the same, average emission from
1096 the landfill. The whole-site emission measurements were carried out over a period of almost
1097 five years, with the only overlap of the campaigns being in August 2012, when tracer gas
1098 dispersion measurements were done the day before and the day after an aircraft measurement
1099 campaign. In the measurements taken over these three days, the aircraft-based mass balance

1100 method showed approximately twice as high a CH₄ emission rate (980 kg h⁻¹) as the tracer gas
1101 dispersion method (480 ± 110 and 470 ± 40 kg h⁻¹). The comparison of CH₄ emissions
1102 measured using the static surface chamber and VRPM showed that the former was unable to
1103 capture the emissions observed with the VRPM method. Both methods were applied across the
1104 area with intermediate cover. The VRPM showed emissions ranging between 196 and 582 kg
1105 h⁻¹, while the static chamber method ranged between 0.3 and 1.2 kg h⁻¹.

1106

1107 **4. Conclusion and perspectives**

1108 Historically, methane (CH₄) emission measurements from landfills were initiated
1109 primarily for research purposes, to obtain emissions at a specific place and time, or to monitor
1110 temporal variability over time. Surface emission monitoring using simple reconnaissance
1111 monitoring, such as FID walkovers, sometimes combined with surface flux chambers, are used
1112 by landfill operators to determine the spatial variability of the emission and for cover
1113 maintenance. Annual CH₄ emissions reported for regulatory purposes are usually based on
1114 models, but when they are estimated solely from landfill gas generation models using waste
1115 data, these emissions are often overestimated due to a conservative approach and a lack of
1116 actual knowledge relating to deposited waste and CH₄ oxidation. More detailed process-based
1117 models such as CALMIM have been developed to overcome some of these challenges.
1118 However, only a few of the available models have been validated, and it is uncertain as to what
1119 extent they describe actual CH₄ emissions. For this reason, CH₄ emission measurements are
1120 important and can provide further knowledge on actual emissions at individual landfills, which
1121 can then be used in model evaluation/validation and for greenhouse gas emission reporting.

1122 Current measurement methodologies do not allow continuous CH₄ emission
1123 measurements at all landfills worldwide, but some countries are starting to work toward

1124 relevant regulations based on measurements instead of models. The recent development in
1125 mass emission techniques, especially the dynamic tracer gas dispersion method, certainly
1126 allows for shifting to measurement-based regulation. Measurements are also essential for
1127 quantifying the effect of emission mitigation initiatives, such as CH₄ extraction and utilisation,
1128 or the engineered optimisation of CH₄ oxidation at the surface, which can significantly lower
1129 the waste sector's emanations of greenhouse gases. One example is the nationwide
1130 implementation of biocovers at old landfills in Denmark, with compulsory emission
1131 measurements at all involved sites for quantifying the CH₄ mitigation effect (Danish EPA,
1132 2018; Scheutz and Kjeldsen, 2019). Another example is the approach proposed by Bourn et al.
1133 (2019) where landfills with gas collection have to fulfil a minimum methane collection
1134 efficiency based on annual methane emissions measurements.

1135 The main challenges when quantifying landfill CH₄ emissions are high spatial and
1136 temporal emission variations. No single emission measurement method is perfect, and many
1137 years of research and tests have revealed some of the advantages and disadvantages of
1138 individual approaches. To quantify the CH₄ emission from an entire landfill, including a
1139 confidence interval, a mass emission method is required rather than a surface emission factor
1140 method. Both DIAL and tracer gas dispersion are mass emission methods, and by applying
1141 them accordingly, the total CH₄ emissions from the landfill are measured – and thus the
1142 challenge with spatial emission variability is circumvented. Both the DIAL technique and the
1143 tracer gas dispersion method have been demonstrated successfully in several landfill studies.
1144 However, the DIAL method uses very complicated and expensive equipment, whereas the
1145 tracer gas dispersion method takes an easier approach and uses more affordable apparatus. The
1146 relatively high cost of the DIAL equipment and the requirement for highly skilled personnel is
1147 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 CH₄ 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 CH₄ emission quantification, but they can be combined with soil gas concentration profiles to provide useful information about landfill gas migration, CH₄ 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 CH₄ 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

1172 covariance and the static mass balance method, can be applied. These methods can be set up
1173 for continuous measurements and thereby capture temporal emission variations. An optimal
1174 approach could involve combining continuous measurements with frequent short-term
1175 measurements, using the DIAL or tracer gas dispersion methods. As a result, this approach
1176 could provide longer-term emission rate time series, which could be validated or adjusted by
1177 more accurate, albeit more expensive and infrequent, measurements.

1178 In addition to quantitative methods, qualitative reconnaissance techniques exist. These
1179 are valuable tools in the daily operation and maintenance of the landfill cover and of site
1180 installations such as gas collection and recovery systems.

1181 Currently, there is a fast development of potentially improved methods based on the
1182 flexibility of UAVs and aircraft. Accurate and high-precision CH₄ instruments are currently
1183 too heavy to employ in UAVs, and aircraft-based measurements are still done mainly in the oil
1184 and gas sector. However, the fast development within both CH₄ sensors and UAVs could soon
1185 allow the use of UAVs at landfills for measuring CH₄ with sufficiently high accuracy and
1186 temporal resolution. In addition, satellites specifically measuring atmospheric CH₄
1187 concentrations are working on getting higher spatial resolution, in order to include area sources
1188 such as landfills.

1189 Although several measuring methods for landfill CH₄ emission quantification have been
1190 developed, the accuracy of many of these methods remains unknown. Determining this factor is a
1191 challenge, as accuracy and potential measurement errors stem from different influences extending
1192 far beyond the uncertainty of the analytical instrumentations applied. As an example, the choice of
1193 model input parameters, such as terrain conditions (open or urban, flat or elevated terrain, etc.),
1194 atmospheric conditions (mixing, stability, wind speed profiles, etc.) and spatial emission patterns
1195 (homogenous vs. inhomogeneous), will add to overall method uncertainty. This also means that

1196 uncertainty and potential errors are not only specific to each method and instrumentation applied,
1197 but also site-specific and depend on conditions on the measuring day. Moreover, some methods
1198 might not be applicable at all landfill sites and under all conditions, so their deployment in non-
1199 favourable circumstances can cause large errors. Future research should hence focus on establishing
1200 method accuracy and developing best practice in terms of monitoring, data processing and
1201 interpretation guidelines.

1202 With new technological capabilities for measuring CH₄ emissions from landfills, there is a
1203 need to develop measurement plans and strategies to comply with current and future legislation, as
1204 well as to document mitigation actions. For regulatory purposes, an average emission rate is of
1205 interest, which is why measurement strategies must pay attention to the dynamic nature of landfill
1206 emissions. Future research should thus focus on developing measurement protocols and strategies to
1207 obtain emission rates representative of normal landfill operations and environmental conditions.

1208

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1793 TABLES

1794 Table 1. Qualitative reconnaissance techniques for locating landfill ‘hotspots’ or determining

1795 surface emission patterns with potentially elevated CH₄ emissions. Reconnaissance techniques

1796 should be used in combination with the quantitative techniques described in Table 2, but they

1797 can be useful for establishing field experimental designs, checking the integrity of cover

1798 materials and planning site maintenance.

Application	Advantages	Limitations	Status and references
Portable CH₄ analyser			
Qualitative or semi-quantitative measurement of CH ₄ concentrations directly above the landfill surface. Applied as point measurements or random/systematic walkover surveys.	Rapid and simple to deploy in the field. Relatively inexpensive - only requires a portable instrument and a GPS for walkover surveys.	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.	A widely used technique as a tool to determine the need for cover remediation and maintenance.
Field infrared survey			
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.	Rapid and simple to deploy in the field, including aerial measurements with UAVs. Relatively inexpensive, depending on the scale and complexity of the field measurement programme. Best suited for identifying larger localised gas leakages.	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.	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).
UAV survey			
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.	A survey flight can be pre-programmed and performed much faster than traditional walkover surveys.	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).	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.
Visual inspection			
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).	Rapid and simple first approach for routine inspection procedures.	Subjective with many possible influences, including wind speed and direction, soil moisture, site topography and landfill cover maintenance.	A useful first step. Site personnel should be instructed to keep alert for odours and vegetation abnormalities indicating landfill gas emissions.

1801 Table 2. Characteristics of the closed and the open surface flux chamber.

Closed chambers	Open chambers
<ul style="list-style-type: none">• Also called a static chamber• No exchange of air inside the chamber with external air• 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• The chamber can have a vent to reduce the influence of internal pressure build-up	<ul style="list-style-type: none">• Also called a dynamic chamber• The chamber is continuously flushed with a flow of gas (a carrier gas) – this can be ambient air or a synthetic gas• 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• The chamber can have a vent to reduce the influence of internal pressure build-up

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1805 Table 3. Summary of quantitative techniques for the field measurement of CH₄ landfill emissions.
 1806 Arranged in ascending order from small-scale chamber measurements ($\leq \text{m}^2$ area) to large-scale
 1807 techniques ($> \text{km}^2$ area).

Application	Advantages	Limitations	Status and references
Vertical soil gas concentration profiles			
Single-point measurements of gas concentrations and pressure gradients in soil cover profiles, with a calculation of flux using equations for diffusional and pressure-driven flux.	Useful 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 CH ₄ sources and sinks.	Difficult to apply appropriately if emissions are associated with high spatial variability. The method is thus not useful for up-scaling for whole-cell/site emission quantification.	Limited to a few and often older landfill field studies but the basis for many transport models. Not appropriate for whole-site CH ₄ emission quantification (Glinski and Stepniewski, 1985; El-Fadel et al., 1995; Christophersen et al., 2001; Röwer et al. 2011; Gebert et al., 2011).
Closed surface flux chambers			
Single-point measurements, using a chamber placed on the surface combined with measurements of change in gas concentration within the chamber vs. time. Typically, a measurement takes less than 1 hour.	Good for process studies of emissions. Combined with vertical soil gas concentration profiles, CH ₄ oxidation can be accessed. The only technique that can measure both the emission and the uptake of CH ₄ . Can also be used for non-CH ₄ gases and isotopic composition analysis. Not sensitive to other CH ₄ sources. Simple to deploy with low limits of detection for fluxes (depending on analytical instrumentation).	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.	The method is historically the most applied for quantifying CH ₄ emissions from landfills. Not appropriate for whole-site CH ₄ emission quantification. Still used in many countries, due to lack of other and more appropriate methods (e.g. Rolston, 1986; Kjeldsen and Fisher, 1995; Tregoures et al., 1999; Conen and Smith, 2000; Christophersen and Kjeldsen, 2001a, Gebert and Gröngroft, 2006, Scheutz et al., 2011a; 2008; 2003).
Open surface flux chambers			
Single-point measurements using a chamber placed on the surface, which is continuously flushed with air or a synthetic carrier gas. Measures flux based on gas flow and the difference between incoming and outgoing chamber CH ₄ concentrations.	Same advantages as closed chambers except cannot quantify negative CH ₄ .	Same disadvantages as closed chambers. Not able to quantify very low fluxes because of dilution of chamber gas with carrier gas.	A technique commonly applied at landfills. Not appropriate for whole-site CH ₄ emission quantification (e.g. Rolston, 1986; Huber-Humer and Lechner, 2001a; 2001b; Hutchinson and Livingston, 2001).
Eddy covariance			
Measurements of the turbulent transfer of gases between the ground surface and the atmosphere. Need fast measurements of atmospheric CH ₄ concentration, wind speed and direction.	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.	Difficult to apply in variable topography and with interfering CH ₄ sources. Measured footprint is dependent on metrological conditions. Requires fast ($> 10 \text{ Hz}$) gas-specific instrumentation with high precision.	Widely used for landscape-scale fluxes from terrestrial ecosystems. Has not been used often for landfill CH ₄ fluxes (e.g. Fowler and Duyzer, 1989; Arya, 1988; Fowler et al., 1995, Hovde, 1995; Laurila et al., 2005; Schroth et al., 2012; Xu et al., 2014).
Stationary mass balance			
CH ₄ concentrations are measured at different heights above the landfill surface, and by combining these with simultaneously measured atmospheric conditions, the horizontal flux of CH ₄ can be calculated.	Same advantages as eddy covariance.	Same disadvantages as eddy covariance, but does not require as fast instrumentation.	Has rarely been used for landfill CH ₄ fluxes, but has been used in the Netherlands (e.g. Scharff, 2003).
Radial plume mapping			
Uses one or more lasers and mirrors to measure the downwind, cross-plume concentration. In combination with measured and modelled wind profiles, surface emissions	Gives an integrated measure of CH ₄ 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.	Rely on a relatively stable wind direction throughout the measurement. Two sets of instrumentation are needed if background (upwind) measurements need to be	Was developed in the late 1990s and made an "Other Test Method (OTM)" by the US EPA. Coverage increases with the development of better laser systems (e.g. Hashmonay et al.,

from the upwind area can be calculated.		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.	1999; Wu et al., 1999; Hashmonay et al., 2001; 2008; Goldsmith, 2008; Abichou et al., 2010; Thoma et al., 2010; Goldsmith et al., 2012).
Mass balance using aerial measurements			
CH ₄ 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 CH ₄ through the 2D plane can be calculated.	Gives an integrated emission from the whole landfill without having to access it.	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.	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 CH ₄ sensors and laser instruments the method has high future potential (e.g. Peischl et al., 2013; Krautwurst et al., 2016; Cambaliza et al., 2015; 2017; Allen et al., 2017).
Stationary tracer gas dispersion			
Simultaneous measurement of atmospheric CH ₄ and a tracer gas at one or more fixed points in the downwind plume. The tracer gas is released at a known rate and the CH ₄ emission rate can be determined from the measured concentration ratio.	Gives an integrated measurement of whole-cell or whole-landfill fluxes, including edge leakages and other on-site hotspots. Does not rely on measurements of atmospheric conditions.	Difficult to apply in the case of interfering CH ₄ 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 SF ₆ and N ₂ O have been used.	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).
Dynamic tracer gas dispersion			
Measurements of CH ₄ and tracer gas while driving across the downwind plume. Using the plume-integrated concentration ratio and known tracer release, the CH ₄ emission rate can be determined.	Same as static tracer gas dispersion. Gives a downwind plume, providing more details about the location of the emission compared to the tracer release. Does not rely on measurements or the modelling of atmospheric conditions.	Requires a road at a suitable distance downwind of the landfill and with no interfering CH ₄ sources in between. The most commonly used tracer gas is N ₂ O, which is a potent greenhouse gas but has been replaced with C ₂ H ₂ in recent applications.	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; Mønster et al., 2014, 2015; Foster-Wittig et al., 2015).
Differential absorption LiDAR (DIAL)			
A laser is shot into the atmosphere and the backscattered part is measured. DIAL is used for landfills by measuring the appropriate CH ₄ absorption lines. A 2D downwind concentration plan is modelled and combined with measured and modelled information on wind speed and direction.	Can give both whole-site and individual area emissions. Very little interference from other CH ₄ sources in the surroundings. Can give detailed information on emission patterns.	DIAL's analytical setup is large and expensive, and data handling is complex. The method relies on accurate wind measurement and modelling.	Mainly used for VOC emissions in the petrochemical industry, but has also been applied to landfills for CH ₄ emission quantification (e.g. Babilotte et al., 2010, 2011; Robinson et al., 2011; Bourn et al., 2019; Innocenti et al., 2017).
Inverse modelling - stationary			
Continuous concentration measurements or sampling in one or more downwind points, or cross-plume concentration, fed into an appropriate inverse atmospheric transport model, together with information on wind speed and direction.	Can give whole-site emission estimates and is good for long time series and thus able to capture emission variations.	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.	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).
Inverse modelling - dynamic			
Measurement of downwind plume and combined with accurate atmospheric condition measurements, the emission rate from the landfill can be estimated.	Measurements are fast to perform and can give whole-site emission estimates.	Instrumentation must be fast and precise. Good measurements of atmospheric conditions with high temporal resolution. Will often result in an emission with significant uncertainty.	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).
Inverse modelling - aerial			
Measurement of downwind plume and combined with	Measurements are fast to perform and can give whole-site emission	Instrumentation must be fast, precise and deployable in UAV	Has been applied at various sources in the US, including

accurate atmospheric condition measurements, the emission rate from the landfill can be estimated.	estimates. No access to landfill needed.	or aircraft. Good measurements of atmospheric conditions with high temporal resolution. Will often result in an emission with significant uncertainty.	landfills (e.g. Cui et al., 2015).
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1810 Table 4. Overview of published comparison studies of three or more methodologies for
 1811 quantifying CH₄, either from landfills or controlled CH₄ release imitating landfill emission.

Application	Methodologies	Emission <i>Unit: (m³ CH₄ d⁻¹)</i>	Comment
Landfill (Tregoures et al. 1999)	Closed flux chamber (no recirculation)	3750-14200	20 x 20 m grid
	Closed flux chamber (with recirculation)	10512-11375	
	Tracer gas dispersion method 1 (SF ₆)	7720±2650	Downwind plume sampled in two ways: static, multiple points (100 m downwind) and dynamic, multiple distances (115, 130 and 200 m downwind)
	Tracer gas dispersion method 2 (SF ₆)	6340±2070	Landfill divided into strips, each measured downwind at the edge of landfill
	Eddy covariance	11.2±3.4	Only one mast location. Wind direction enabled only a small part of the landfill to be measured
	Static mass balance	24±8	
	Remote sensing (airborne IR thermography)	No	Only used for finding hotspots
Four landfills (LF1-4) (Scharff et al. 2003)		<i>Unit: (m³ h⁻¹)</i>	
	Static mass balance	LF1: 530±130 LF2: 109±30 LF3: 390±100 LF4: 70±8	Sampling 10, 15 and 25 m above ground
	Inverse modelling (static)	LF1: 440±180 LF2: 277±71 LF3: 690±230 LF4: 184±81	Four sample bags collecting gas in different wind directions
	Inverse modelling (dynamic)	LF1: 540-900±50-125 LF2: 315±33 LF3: 300±10 LF4: 135±32	Using tracer gas for model calibration. At LF1, three experiments were done
12 landfill sections (P1 to P12) (Goldsmith et al. 2008)		<i>Unit: (g m⁻² d⁻¹)</i>	
	Closed flux chamber	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	Each landfill part was measured and compared individually
	Radial plume mapping (100 m path)	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	
	Radial plume mapping (200 m path)	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	
Landfill (Babilotte et al. 2010)		<i>Unit: (g CH₄ s⁻¹)</i>	
	Radial plume mapping	25.2±2.4	Individual cells were also quantified
	Tracer gas dispersion (static) (N ₂ O)	83±36	15 canisters sampled over 4 h across a plume 200 m downwind
	Tracer gas dispersion (dynamic) (N ₂ O)	41±17	Real-time TDL measurements across a plume 200 m downwind
	DIAL	12.1±2.4	
	Inverse modelling	163	Real-time measurements 300 m downwind using instrumentation able to measure ppb changes in CH ₄ concentration and applied to a Gaussian model software
Controlled release 0.5±0.04 (g CH ₄ s ⁻¹) (Babilotte et al. 2010)	Helicopter-borne spectroscopy	No	Able to find hotspots
	Radial plume mapping	0.89±0.05	Individual cells were also quantified
	Tracer gas dispersion (dynamic) (N ₂ O)	1.7±1	Measurements 100 m downwind
	Tracer gas dispersion (dynamic) (N ₂ O)	2.3±0.5	Model + measurements 100 m downwind
	DIAL	0.23±0.12	
	Inverse Modelling	1.5	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
Two landfills	Helicopter-borne spectroscopy	No	Able to locate release area
		<i>Unit: (g m⁻² d⁻¹)</i>	

(Green et al. 2010)	Closed flux chamber	0.018/0.125	26 points at each landfill
	Radial plume mapping	9.4/8.1	Area calculated on atmospheric stability class
	Radial plume mapping	16.7/10.5	Area calculated by multiple linear regression model
	Tracer gas dispersion (static) (C ₂ H ₂)	8.5/14.3	CRDS instrumentation
	Tracer gas dispersion (dynamic) (C ₂ H ₂)	7.9/13.1	CRDS instrumentation
Two landfills (#1/#2)		<i>Unit: (kg h⁻¹)</i>	
(Babilotte et al. 2010)	Closed flux chamber	290/868	1-3% of total area measured
	Eddy covariance	1005/1146	~30% of total area measured
	Radial plume mapping	1325/1244	10-15% of total area measured
	DIAL	659/314	Whole landfill
	Tracer gas dispersion (dynamic) (N ₂ O)	916/789	30-45% of total area measured
Four Controlled releases (Babilotte et al. 2011)		<i>Unit: g s⁻¹ (±SD)</i>	
	Release #1	1.095±0.005	One release point
	Radial plume mapping	1.04±0.22/0.71±0.23/0.62±0.20	10/50/100 m from source
	DIAL	0.86±0.27	10 m from source
	Tracer gas dispersion (dynamic) (N ₂ O)	1.16±0.10	450 m from source
	Release #2	1.095±0.005	Two release points
	Radial plume mapping	1.06±0.23/0.79±0.19/0.57±0.15	10/50/100 m from source
	DIAL	0.90±0.25	40 m from source
	Tracer gas dispersion (dynamic) (N ₂ O)	1.31±0.09	400 m from source
	Release #3	3.284±0.013	Three release points
	Radial plume mapping	2.98±0.65/2.65±0.73/1.92±0.45	10/50/100 m from source
	DIAL	0.90±0.25	40 m from source
	Tracer gas dispersion (dynamic) (N ₂ O)	1.31±0.09	400 m from source
	Release #4	1.642±0.007	Three release points
	Radial plume mapping	1.42±0.29/1.24±0.22/0.96±0.22	10/50/100 m from source
	DIAL	1.96±0.46	40 m from source
	Tracer gas dispersion (dynamic) (N ₂ O)	1.72±0.11	400 m from source
One landfill (Cambaliza et al. 2017)		<i>Unit: (mol s⁻¹)</i>	
	Closed flux chamber	0.005-0.02	Only intermediate cover measured
	Radial plume mapping	1.7-10.1	Only final and intermediate cover measured
	Aircraft-based mass balance	7-17	Whole landfill (from 2009 to 2012, n = 20)
	Tracer gas dispersion (dynamic) (C ₂ H ₂)	8-13	Whole landfill (in 2012 and 2014, n = 2)

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1814 FIGURES

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1817 **Fig. 1.** Overview of the most common methods used to identify and quantify CH₄ emissions
1818 from landfills.

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1822 **Fig. 2.** Schematic overview of open and closed flux chambers for quantifying CH₄ fluxes from

1823 landfills.

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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.

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1832 **Fig. 4.** Horizontal (HRPM) and vertical (VRPM) radial plume mapping with setup, as

1833 suggested in the OTM10 method (Hashmonay et al., 2008).

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1837 **Fig. 5.** Tracer dispersion method using the stationary or dynamic approach. Stationary can be
1838 done using sampling or direct measurements, while dynamic requires fast, direct
1839 measurements.

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1843 **Fig. 6.** Small-scale tracer dispersion method for quantifying CH₄ emissions through a leachate
1844 well. From Fredenslund et al. (2010).

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1849 **Fig. 7.** The DIAL method, using multiple 'line of sight' laser shooting with subsequent model
 1850 fitting to create a two-dimensional cross-plume concentration profile.

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