Chapter 069

QUANTIFICATION OF FUGITIVE METHANE EMISSIONS FROM LANDFILLS:

AN OPEN ISSUE

**Abstract.** Methane fugitive emissions from landfills represent a significant source of climate-altering gases emitted into the atmosphere globally. The recent Communication COM (2020) 663 reports that in the EU, 53% of anthropogenic methane emissions come from agriculture, 26% from waste and 19% from energy.

Landfills therefore represent an important emission sector, on which it is necessary to continue investing in innovation and technology to limit fugitive emissions, especially of methane.

In Italy, Legislative Decree 36/2003 has imposed an obligation on landfill operators to quantitatively characterize biogas but does not indicate which technique should or can be used for monitoring. Quantifying landfill biogas flow is a complex exercise with varying degrees of uncertainty. This paper illustrates the main methods currently developed to perform this difficult task. For each method, measurement characteristics, instrumentation required, advantages and limitations identified for each are reported.

From the analysis of the results, it can be stated that at present there is absolutely no quantification method that is preferable to others; it is necessary to proceed with further research also considering the new instruments that technology makes available.

**Keywords.** Methane, landfill, biogas, flux measurement

# Introduction

The various techniques available for quantifying CH4 emissions from landfills operate on very broad spatial and temporal scales: measurements can be made from the surface of the landfill up to several kilometres away and take minutes, weeks, or months.

Regarding the spatial scale, directly measuring emissions on a portion of the surface allows the interference of any surrounding CH4 sources to be excluded, but makes the subsequent extrapolation step, which is necessary to represent a large area that is characterised by heterogeneous emissions, more difficult.

Methods that measure at greater distances are better than the previous ones for quantifying site-wide emissions but are more sensitive to potential surrounding sources.

The time scale, on the other hand, plays a decisive role on the feasibility and quality of the flux measurement, as landfill emissions are well known for their extreme temporal variability even in the very short term.

It follows that each technique is characterised by its own spatial and temporal resolution, with its own advantages and limitations: defining these aspects is of fundamental importance for understanding the status of the main existing quantification methods.

# Material and methods

The data was collected from a bibliographical search carried out online on the 'Scopus' bibliographical database, published by Elsevier, collecting material through specific keywords provided as input to the search engine, then selecting the sources according to both their year of publication and the techniques discussed to quantify emissions.

For each of the identified methods, data was collected on the spatial and temporal resolution, measurement characteristics, instrumentation required, and the advantages and limitations associated with each.

# Results

The table below shows the main flux quantification methodologies, sorted in ascending order according to spatial resolution and each described in terms of its special features.

Table 1. Spatial and temporal resolution of flux quantification methodologies

|  |  |  |
| --- | --- | --- |
| Flux quantification methodology | Spatial resolution | Temporal resolution |
| Concentration gradient measurement |  < 0.5 m2 | 10-20 minutes |
| Dynamic concentration measurement |  < 0.5 m2 | a few minutes |
| Flux chambers |  0.5 – 1.0 m2 | a few minutes |
| Eddy covariance method | < 1000 m2 | 30 minutes |
| Radial Plume Mapping (RPM) | h (10-50m); d (< 200m) | hours |
| Tracer gas dispersion method (static) | a few km | hours or days |
| Tracer gas dispersion method (dynamic) | a few km | 2-6 hours or days |
| Differential absorption LiDAR method | 400-800 m | hours or days |
| Inverse dispersion modelling method  | from 500 m to a few km | hours |

*Concentration gradient measurement:* It is a technique performed on small-volume samples, aspirated through probes inserted into the soil at varying depths and then analysed to derive a concentration value. Developed by Gebert et al. (2011), it has been used in various emission studies (Gonzalez-Valencia et al., 2016; Pratt et al., 2013) and to assess CH4 emissions from a landfill (Bogner et al., 1995).

*Advantages and limitations*: capable of providing information on mechanisms influencing emissions, such as rapid changes in atmospheric pressure, wind speed and emissions resulting from simultaneous convection and diffusion. It can identify point sources and estimate CH4 oxidation. The technique is characterised by a low spatial resolution (sampling area < 0.5m2), while the temporal resolution (sampling duration between 10 and 20 minutes) conflicts with the rapid variations in emissions from a landfill. It does not allow measurements at critical points (cracks in the cover or leachate pits) and the diffusion coefficient is complex to estimate. It is also labour-intensive.

*Dynamic concentration measurement*: proposed by Guerrieri and Valenza (1988), consists of a Cu probe (inserted inside a steel tube perforated in the end section), embedded in the soil, and connected by pump to an IR spectrophotometer. After a few minutes of pumping at constant flow, the gas concentration in the mixture stabilises and this concentration is directly proportional to the gas flow in the soil according to the equation

$$φ=MC\_{d}$$

where φ is the gas flux through the soil (g m-2 s-1); Cd is the dynamic concentration (g m-3); M is an empirical constant determined in the laboratory.

*Advantages and limitations*: much faster than the concentration gradient measurement, otherwise sharing the same advantages. The limitations are also the same, but this technique differs from the former due to its inapplicability when the layers to be traversed are not very thick (< 1.0 m). The empirical coefficient M depends not only on the geometry of the sampling system, but also on the permeability of the soil, and since it is estimated by means of laboratory tests, it is highly skewed and unrepresentative.

*Flux chambers*: It is a methodology that estimates the flux of CH4 from a surface by measuring the changes in CH4 within a chamber over a given time interval during which it is in contact with the soil:

$$F=\frac{dC}{dt}\frac{V}{A}$$

where F is the flux (g m-2 s-1), $\frac{ⅆC}{∂t}$ (g m-3 s-1) is the concentration change over the time interval, V the chamber volume (m3) and A the base (m2). Widely used to quantify gas emissions from landfills, (Lucernoni et al., 2016; Rachor et al., 2013; Abichou et al., 2011; Bogner et al., 1995), it is distinguished according to whether there is continuous insufflation of air inside it (dynamic) or not (static).

*Advantages and limitations*: very simple to use, concentrations can be measured with an FID or IR detector. The method can detect even modest fluxes of CH4 and is to date the reference technique for estimating the flux of trace gases contained in non-methane volatile organic compounds (NMVOCs) (Scheutz et al., 2008). It is time-consuming and labour-intensive, requiring appropriate geostatistical techniques. It does not lend itself to advective fluxes and surface perturbation can affect emission. It has a spatial resolution limited to the footprint surface of the chamber.

*The eddy covariance method*: It is based on the theories of turbulent transfer in the surface boundary layer of the atmosphere, applied in several landfill studies (Xu et al., 2014; McDermitt et al., 2013; Schroth et al., 2012). The measurement requires the determination of the mean vertical component of wind speed and the mean CH4 concentration from time series collected by anemometers and FTIR or TDLAS detectors, respectively. The gas flux is then calculated using the following equation:

$$F=w̅'c̅' $$

where F is the flux (g m-2 s-1), 𝑤̅’ the average vertical component of wind speed (m s-1) and 𝑐̅′ is the average gas concentration (g m-3).

*Advantages and limitations:* provide large-scale emission estimates and is suitable for flat sites with uniform emissions. It allows continuous measurements over long periods of time, providing information on temporal variability and average emissions. However, it requires assumptions of horizontal site homogeneity and atmospheric stationarity, which are only fulfilled if time windows of less than 60 minutes are considered (Kaimal & Finnigan 1994; Finnigan et al. 2003).

*Radial Plume Mapping (RPM):* Developed in the late 1990s, it uses a combination of concentration measurements and wind profiles to obtain a surface emission rate from an upwind area. It can be used in two configurations: horizontal (HRPM) or vertical (VRPM), the former capable of detecting hotspots and the latter of quantifying flows. A laser beam is aimed at reflectors positioned at different heights (10-50m) and distances (up to 200m); each section provides an average CH4 concentration: a 2D concentration profile is thus modelled on the cross-section of the plume. By combining this data with wind speeds, the surface emission rate can be measured.

*Advantages and limitations*: capable of identifying point sources and estimating emission fluxes, lending itself to preliminary screening for remediation. Limitations are low spatial resolution (limited laser range) and high temporal resolution (several hours), incompatible with landfill flows. Upwind and downwind measurements would have to take place simultaneously (increasing the costs of the test) and the results are complex to interpret. Determining the precise area contributing to the measured emission is equally complex.

*The tracer gas dispersion method*: it consists of releasing a tracer gas from a position upwind of the emissive surface and measuring the CH4 and tracer concentrations downwind in each time interval. A distinction is made between stationary and dynamic, depending on whether measurements are made with instruments (FTIR or TDLAS detectors) placed at fixed points or equipped on a vehicle moving along a road.

*Advantages and limitations*: the main advantage is the simplicity of analysis (provided the CH4 and tracer gas are well mixed). It makes it possible to estimate the total emissions of a landfill (including point sources) and is not affected by errors related to site topography, as it can be applied to landfills of all sizes. Recent studies recommend measuring at least 10 points, recommending an average number of 15 samplings (Fredenslund et al., 2019b; Monster et al., 2014). The disadvantages of the technique concern its dependence on meteorological conditions (wind direction and speed), which are necessary to ensure proper mixing, as well as the need for suitable road access. It is unable to discriminate between the different sources contributing to the emission, leading to possible quantification errors. The instrumentation is expensive and requires specialised personnel. A further disadvantage is that measurements are carried out over hours (2-6 h) or a few days, so the temporal variation of the emission is a major disturbing factor on the quality of the data.

*Differential absorption LiDAR method (DIAL):* based on the analysis of the electromagnetic spectrum emitted by a surface. It uses different types of sensors and the data obtained are processed through image analysis, returning thematic maps where each colour is associated with a piece of information. Measurements are taken both downwind and upwind of the landfill: the flux emitted from the surface is obtained by difference.

*Advantages and limitations*: they provide total emissions from the entire site or portions. They also have a good spatial resolution (400-800 m), depending on atmospheric conditions. The main disadvantage of the DIAL method is the cost and computational burden of analytical modelling and data management. It requires suitable roads, and the measurements depend on the wind (it must be stable to measure the flow accurately) and present infrastructure. Measurements are conducted over hours or days.

With reference to a mobile version, very few studies are available to date on its use in landfills (Innocenti et al., 2017; Bourn et al., 2019; Robinson et al., 2011).

*Inverse dispersion modelling method*: By measuring CH4 concentrations on a leeward plane at the surface and combining them with meteorological data, the flux can be calculated using atmospheric gas dispersion models. Several models have been specially developed for this purpose, such as LASAT (Janicke Consulting, Überlingen, Germany) and WindTrax (Thunder Beach Scientific, Nanaimo, Canada).

*Advantages and limitations*: It can provide estimates of site-wide emissions even from critical points such as embankments and biogas collection systems, which are often difficult to intercept using traditional methods. On the other hand, it requires large quantities of high quality data as inputs for good emission estimates, such as atmospheric stability, surface turbulence and wind speed. Optimal conditions would require the landfill site to be in a flat area, with a road cutting through the downwind plume at a suitable distance.

# Conclusions

An analysis of the techniques used to date for the quantification of methane fluxes from landfills has shown that each method has its strengths and weaknesses, and that at present it is not possible to say that there is absolutely one quantification method that is preferable to others. This issue therefore remains open; however, technological innovations introduced in various sectors seem promising in the search for a more concrete solution, such as the use of drones equipped with appropriate sensors.

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