

# **Methods for the Measurement of Methane Fluxes from Landfill Sites**

Final Report to  
The Environment Agency

M J T Milton and T D Gardiner

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**Methods for the Measurement of  
Methane Fluxes from Landfill Sites**

**Commissioned by  
The Environment Agency**

**National Physical Laboratory  
Centre for Optical and Environmental Metrology  
Teddington  
Middlesex  
United Kingdom  
TW11 0LW**

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National Physical Laboratory  
Queens Road, Teddington, Middlesex, TW11 0LW

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Approved on behalf of Managing Director, NPL  
by D H Nettleton, Head of Centre for Optical and Environmental Metrology

## “Methods for the Measurement of Methane Fluxes from Landfill Sites”

### ***Executive Summary***

Emissions of methane from landfill sites are difficult to monitor because they can be highly variable and occur through a variety of mechanisms including diffusion through the capping layers, escape through faults in any gas control system or migration through geological features prior to emissions beyond the site boundary. This study reviews five methods for monitoring such emissions. They vary in their cost, complexity and range of possible applications. The flux chamber and the flux survey methods are the two methods that are most likely to be of value in monitoring methane emissions for regulatory applications.



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# “Methods for the Measurement of Methane Fluxes from Landfill Sites”

## 1 INTRODUCTION

### 1.1 Background

Landfill gas is generated from the bio-degradation of the organic fraction of landfilled waste. Under the conditions prevalent in most modern landfill sites these bio-degradation processes are predominantly anaerobic and give rise to landfill gas which contains between 40 and 60 per cent methane with the balance being predominantly carbon dioxide, with trace concentrations of various non-methane hydrocarbon compounds (NMHC).

There is now significant concern about the environmental impact of methane and NMHC emissions from landfill which may include:

- contributions to global warming resulting from the greenhouse effect. Methane makes the second largest total contribution to global warming of the greenhouse gases (after carbon dioxide). National emissions of methane to the atmosphere are now regulated by the Framework Convention on Climate Change [IPCC 1995].
- risk of explosion from the accumulation of methane up to flammable levels. The escape and subsequent build up of landfill gas has led to increased concern about explosions in buildings situated close to landfill sites.
- unpleasant odour of some NMHC's and sulphur-containing compounds in landfill gas. One of the environmental nuisances that is often associated with landfill sites is the unpleasant odour due to the decomposition of certain materials.
- formation of asphyxiant atmospheres as a result of the very low oxygen composition of landfill gas.
- production of tropospheric ozone from reactions between NMHC's and nitrogen oxides (largely emitted by vehicles). Although the photochemical generation of ozone is a slow process which is not usually associated with local sources, emissions of NMHC's from landfill make a contribution to national emissions.
- possible carcinogenic effects of some NMHC's. Some studies of the trace components of landfill gas have identified low concentrations of carcinogenic compounds such as benzene [Scott 1985, Milton 1996].

A recent trend in landfill site management has been to exploit the energy potential of landfill gas through electricity generation schemes. Such schemes have been selected for support by the Government's NFFO (Non-fossil Fuel Obligation) programme. The development of sites for their methane potential has changed the way that sites are operated, and has imposed another constraint on site design in addition to minimising the environmental impact.

This report reviews possible methods for measuring methane in landfill gas. It is motivated by the responsibility of the Environment Agency to ensure that appropriate measurements are made of gases emitted from landfill sites.

### 1.2 Generation of Methane in Landfill Sites

Modern landfill practice is that industrial, commercial and domestic waste are placed together in a void space and compacted. The compacted waste is then covered with layers of soil or clay, usually at the end of each day. This process continues until the available space is full at which time the waste is capped with an impermeable layer of either natural (clay) or artificial (textile, HDPE) material.

Studies of the biological decay processes within a landfill site show that a series of different processes occur involving different bacteria. Initially, the waste decays anaerobically as a result of bacteria that hydrolyse complex biological polymers into smaller molecules. The products of hydrolysis then decay by the action of acetogenic bacteria into acetic and other fatty acids. In the anaerobic conditions within a landfill, these acids accumulate within the leachate and then decay through the action of methanogenic bacteria to form methane.

It may take several years before the decay processes reach the stage of methanogenesis. After, a further period of time, the increase in the decay processes will become limited by the supply of suitable substrate material, at which point the rate of generation of methane will decay exponentially for between 5 and 50 years.

### **1.3 Emissions and Extraction of Methane from Landfill Sites**

Preventing the escape of landfill gas is one of the most complex challenges in effective site design. Following its generation in areas within the site where bio-degradation is underway, landfill gas migrates through the site until it reaches the capping layer or the sides. It will then escape through any flaws in the cap or the sealing at the edge of the site. In the absence of any gas control system, any positive pressure that builds up will be relieved in this way. On sites where particular hazards due to gas migration have been identified, passive gas migration controls are often installed. These may consist of trenches or vent holes that limit migration in particular directions.

Since the early 1980's it has usually been a licence condition that landfill site operators install some form of gas collection system. This usually consists of wells sunk into the waste connected through pumps to a flare. In some cases, methane is used to generate electricity in which case any surplus gas, or any gas collected when the generation system is not in operation will also be flared.

Assessing the relative influence of different escape routes for landfill gas from a site is one of the most difficult aspects to monitoring emissions. As is highlighted in the review of different methods, some methods only measure gas escaping by a particular route. If such methods are used to estimate emissions from the site as a whole, they may give an unrepresentative picture of emissions as a whole.

### **1.4 Methods for Measuring the Concentration of Methane**

Methane is always present in the atmosphere at a concentration that varies between 1.6 and 2 parts-per-million. The higher concentrations are usually found in developed areas where there are a large number of methane sources. Since the atmospheric background is not constant, even at a fixed location, any methane emitted from a landfill site must be measured in the presence of these variations in the natural background. Hence, there is no benefit in using instruments to measure the concentration of atmospheric methane that have a sensitivity better than 1% of the expected background. (This does not apply to the eddy correlation method, which requires greater sensitivity as well as a faster response).

In Section 2 of this report we review possible methods for measuring fluxes of methane from landfill sites. All of the methods reviewed are capable of measuring not just the concentration of methane, but also its flux which is defined by:

$$\text{Flux [kg/m}^2\text{/s]} = \text{Concentration [kg/m}^3\text{]} * \text{Velocity [m/s]}$$

Each of the different methods combines a measure of concentration with a measure of gas velocity (or flow) in order to estimate flux.

In this Section, we describe some of the methods that are used when measurements of

methane concentration are required without any reference to the flux. The most important of these is the flame ionisation detector (FID). Additionally, we discuss dispersive and non-dispersive optical methods.

#### *1.4.1 Flame Ionisation Detection (FID)*

The flame ionisation detector (FID) is probably the commonest device used for making measurements of methane at concentrations below  $100 \text{ ugm}^{-3}$ . It works by passing the sample gas through a hydrogen flame mounted between the plates of an electrode. The volatile organic component of the gas is ionised which results in a current between the electrodes. The size of this potential is proportional to the concentration of covalent carbon in the sample gas. Therefore, the FID alone measures total organic carbon and has no capability to speciate different VOCs in the sample. Consequently, the FID is often used with gas chromatography which is able to separate different gases as they flow through a densely packed column of inert material. Alternatively, lower cost FID-based instruments may be used with a chemical filter to remove all VOCs apart from methane and are able to record the specific methane concentration.

FID systems have little or no cross-sensitivity to water, carbon dioxide or carbon monoxide at the concentrations encountered in landfill applications, although some older instruments show a response to oxygen. The measurement time is typically tens of seconds, and a calibrated system can have an accuracy of a few percent (at ambient methane concentration levels).

#### *1.4.2 Dispersive and Non-Dispersive Infrared*

Non-dispersive infrared (NDIR) monitors use an optical source together with some form of filtering to select a particular wavelength band which is then directed through the sample gas. The transmitted energy is then measured. This type of monitor is generally simple and robust, but does suffer from cross-sensitivity, particularly from water vapour. There are a number of different versions of the NDIR technique, including single beam, dual beam (signal and reference) and rotating filter. However, the sensitivity to methane is poor - even for the more sensitive versions the methane detection limit is typically 0.5 ppm. Therefore, the NDIR method is only suitable for applications where the methane level is at least an order of magnitude above ambient.

Dispersive optical monitors use some form of optical interferometer to separate light at different wavelengths. This enables them to distinguish those parts of the spectrum that are absorbed by methane from those that are not. The concentration of methane can then be calculated using standard values for the absorption coefficient of methane. Fourier Transform Spectrometers (FTS) are an example of this type of instrument. They have the capability to measure a very wide range of species at ambient concentrations including methane.

An alternative optical method with much greater sensitivity is direct optical absorption. An example of this type of instrument is the tunable-diode laser spectrometer (TDLS). It is capable of carrying out measurements sufficiently rapidly to be used in the eddy-correlation method as well as in open-path mode..

## 2 METHODS

### 2.1 Static and Dynamic Flux Chambers

#### 2.1.1 Principle of Method

The flux chamber (or “flux box”) captures gas escaping from the surface of a landfill site and measures the consequent increase in concentration within the chamber. The flux can then be calculated from [Czepiel 1996]:

$$E = \frac{V}{A} p \frac{\Delta C}{\Delta t}$$

where:  $V$  is the volume of the chamber,  $A$  is the enclosed surface area,  $p$  is the gas density within the chamber, and  $\Delta C/\Delta t$  is the rate of increase of concentration within the chamber. This is the simplest approach which is referred to as a static chamber since there is no flow of gas through the chamber apart from the small flux from the surface being measured. If measurements of methane are required, the concentration within the chamber can either be measured by taking a series of samples for analysis by GC-FID or by FID alone. Alternatively, the concentration within the chamber can be monitored continuously using an FID, in which case some method must be used to ensure that the pressure within the chamber is not reduced by the removal of the sample gas.

A more sophisticated version of the flux chamber uses a continuous flow of sweep gas through the chamber at a rate significantly larger than the emission rate from the surface. This is known as a dynamic chamber.

#### 2.1.2 Review of Applications of Flux chambers to Landfill Methane Measurement

Flux chambers have been used extensively for measurements of surface fluxes of methane and NMHC [Eklund 1992]. As the equation shows, the measured change in concentration is proportional to the area and inversely proportional to the volume, hence, the minimum detectable flux is only proportional to the height of the chamber. Generally, a cylindrical geometry is preferred over rectangular ones because mixing within the chamber is enhanced. Flux chambers should be constructed from metal rather than plastics, some of which emit hydrocarbons and even methane. Accurate measurements require a gas-tight seal between the chamber and the ground in order to prevent leakage or ventilation by wind.

The design developed by NPL (Figure 1) [Milton 1997] has the edges of the cylindrical chamber sharpened so that the chamber can be forced into the surface of the site to a depth of at least four centimetres. Of the different designs that have been discussed, the one most frequently used in the US [Reinhart 1992] consists of a stainless-steel dome 71 cm in diameter placed on top of a cylinder 30 cm high.

A novel design for a flux chamber that does not require clean sweep gas and does not disturb the surface being monitored has been patented [US Patent 5,355,739], but no applications have been reported yet.



**Figure 1** - Cylindrical flux chamber developed for monitoring methane emissions from the surface of landfill sites.

Difficulties that arise during the use of all designs of flux chamber which give rise to uncertainties in the results include:

- the need to mix gas within the chamber because emissions will not be homogeneous over the area being sampled. This is easily achieved in a dynamic chamber by the sweep gas, alternatively it can be achieved by the use of a mechanical stirrer.
- changes in the temperature, pressure and humidity at the surface being sampled as a result of the presence of the chamber.
- false negative fluxes being recorded as a result of high concentrations of ambient methane trapped in the chamber when it is placed on the site.

The largest single source of uncertainty associated with the flux chamber method is that associated with the sampling strategy used to ensure that the measured flux at selected points is averaged to give a representative estimate of emissions rates from the site as a whole. This aspect of the use of flux chambers can only be handled by establishing a sampling strategy relevant to the size and nature of the emissions and the objectives of the measurements themselves. This is usually done [Eklund 1992, Milton 1997] by dividing the site into areas that can be expected to be approximately uniform in their emission characteristics (for example they have the same capping material or surface vegetation and a constant depth of waste of similar age). Each area can then be divided into a grid which can then be sampled randomly, ideally with at least six surface flux measurements at each point. Since eight to ten measurements can usually be achieved during a day, two or three days work with each chamber may be required to measure each area of a site. However, even a well conceived statistical design will not be able to

deal accurately with a site for which emissions are dominated by gas escaping from the site through cracks or local sources (eg around wells).

#### *2.1.4 Recommended Applications*

Since the flux chamber method is very labour intensive when used to measure total site fluxes, it can only be recommended for use in the measurement of small areas of a site. It is well suited to studies of different types of capping since it is capable of measuring the very low fluxes expected through the cap. It might also be used for studies of long-term variations of emissions from a site in which repeated measurements are made at exactly the same location. It is unsuitable for monitoring in any situation where the emissions are highly inhomogeneous or where the surface is not uniform enough to form a gas-tight seal against the chamber.

## 2.2 Flux Survey Methods

### 2.2.1 Principle of Method

The flux survey method works by measuring the concentration of methane in the atmosphere at a fixed height above the site along a series of paths both up- and down-wind of the emissions. The recorded concentrations are then processed to calculate the average concentration along each path. The flux is then calculated by reference to the wind speed and direction (measured simultaneously) and a model which calculates the expected profile of the plume at any distance from an area source.

### 2.2.2 Applications of Flux Survey Methods to Landfill Methane Measurement

The most detailed applications of the flux survey method have been developed by NPL [ref NPL Report QM 134] This implementation of the method uses an accurately calibrated portable FID (see Section 1.4.1) with automatic data logging, which is carried along the site at a constant height above the ground (Figure 2). Simultaneous measurements of the wind field are made with automated anemometers. The method also requires an estimate of the atmospheric stability, which is straightforward to obtain from standard meteorological observations. The wind and methane data are then combined with an estimate of the height (derived from the atmospheric stability) of the plume of methane above the site to give the flux. The method has the major advantage that it measures methane from a wide area of the site so it does not have the problem that flux chambers have with representativeness.



**Figure 2** - Portable flux survey equipment developed for cost-effective emissions monitoring.

The flux survey is similar in its implementation to published reports of methods used to fulfil the requirements of the US EPA's "Municipal Solid Waste Landfill New Source Performance Standard". This standard imposes the requirement on operators to measure the concentration of methane around the perimeter of sites and along a grid of lines spaced 30 metres apart. Published examples of work to fulfil this standard in Canada [Eade 1997] and the USA [Huitric 1996] are based on transportable FID's carried on a vehicle or manually. Neither of these methods involves combining the point concentration measurements with the wind profile to calculate the flux.

The use of a flux survey method called “Integrated Surface Sampling” (ISS) has been reported by California Air Resources Board, but no detailed information is provided about how the results are calculated from the measured data.

### *2.2.3 Recommended Applications*

The flux survey is the quickest and lowest cost method reviewed here. The equipment is straightforward to obtain commercially and costs less than £5,000. In general, the processing of the measurement data to produce results is not too difficult. Consequently, the flux survey method is recommended for rapid estimation of total site emissions and for screening sites to quantify the effectiveness of gas control systems. It is not suitable for estimating emissions from sites with highly complex topographies, for example, it might be difficult to achieve good results from a steep-sided land raising site.

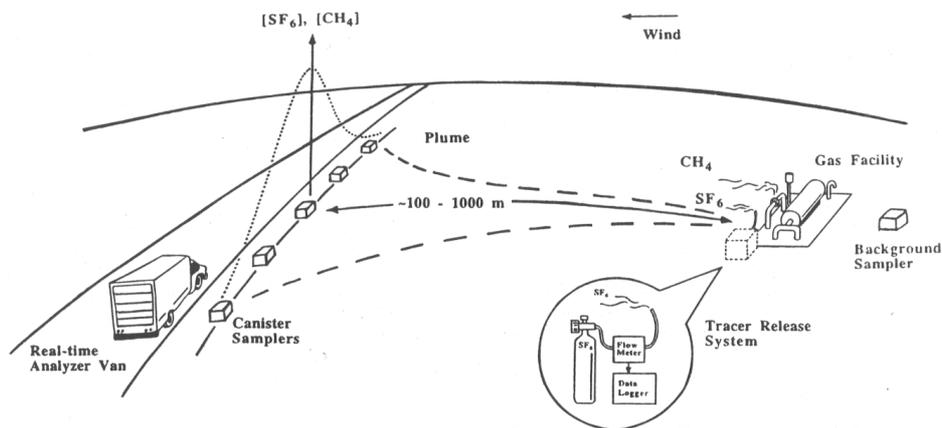
## 2.3 Tracer Gas Methods

### 2.3.1 Principle of Method

Tracer gas methods work on the basis that if a tracer gas can be mixed into the plume of emitted gas from a landfill surface, then measurements of the flux of the tracer gas (from its release points) can be used to calculate the flux of landfill gas from the surface using the equation:

$$Q_m = Q_t \frac{C_m}{C_t}$$

where  $Q_m$  is the flux of the gas being measured,  $Q_t$  is the flux of tracer gas,  $C_t$  is the concentration of the tracer and  $C_m$  is the concentration of the gas being measured (Figure 3). This equation is only accurate when the dispersion of the tracer gas in the atmosphere is the same as that of the surface emissions being measured. Hence tracer gas methods have principally been developed for the measurement of fluxes from point sources such as stacks. Their application to the measurement of large area emissions which are likely to be highly inhomogeneous is more difficult.



**Figure 3** - Schematic illustration of tracer gas. In this example it is used to measure methane emissions from a “Gas Facility”

The method is restricted to situations where there is no “natural” emission of the tracer gas. Consequently, species such as sulphur hexafluoride (SF<sub>6</sub>) which have very low ambient atmospheric concentrations are used as tracers.

### 2.3.2 Applications of Tracer Gas Methods to Landfill Methane Measurement

Tracer methods have been used by various researchers to study methane fluxes from a range of different sources [Howard 1992, Moriizumi 1996]. The main attraction of tracer methods is that it is theoretically possible to determine the methane flux from a single downwind measurement of the methane and tracer concentration. However, this principle only holds for a point emission source of methane with no interference from neighbouring sources. To apply the tracer technique to a diffuse source such as a landfill site a number of assumptions have to be made about the source distribution and the atmospheric conditions.

Czepiel *et al* (1996) describe the use of the tracer gas method to determine the methane emissions from a 60 hectare landfill site in the US. The tracer gas, sulphur hexafluoride (SF<sub>6</sub>), was released from three points along the upwind edge of the landfill. The tracer and methane

plume distributions were measured 1.2 to 3.5 km downwind of the landfill. The plumes were located using a vehicle fitted with fast response analysers which made repeated traverses across the plumes. Once a suitably mixed plume had been located a series of static gas-samples were taken across the plume and then analysed for methane and SF<sub>6</sub>. The methane flux estimated from the tracer measurement results was 17750 litres.min<sup>-1</sup>, which agreed reasonably well with the flux of 15800 litres.min<sup>-1</sup> determined from a series of 139 enclosure measurements. The agreement between the two results was improved when the effect of atmospheric pressure on the emission rate was taken into account.

A similar intercomparison of techniques was carried out by Savanne et al (1997). Two tracer methods were employed, one assumed a simple point source for the emissions and a second which made an attempt to reconstruct the source distribution. The two methods gave methane emission flux results of 67 ml m<sup>-2</sup> min<sup>-1</sup> and 55 ml m<sup>-2</sup> min<sup>-1</sup> respectively, which compared to flux chamber results of approximately 90 ml m<sup>-2</sup> min<sup>-1</sup>. Landfill measurements are also reported by Tregoures et al (1997). This work includes a number of modifications to the tracer technique which aim to improve the accuracy of the method when applied to diffuse inhomogeneous sources. However, most of these add significantly to the complexity of the method.

The review of the various tracer measurements has shown that, although the principle is relatively simple, the actual implementation of the technique can be complex and time consuming. In addition, the requirements for homogenous emissions, no interfering sources and particular dispersion conditions means that accurate results can only be obtained under very limited circumstances. However, the area where tracer measurements are useful is when they are combined with one of the other measurement techniques that requires dispersion modelling. The tracer measurements can be used to give accurate information about the dispersion conditions at the time of the measurement thereby improving the quality of the dispersion model, as demonstrated by Piccot *et al* (1996).

### 2.3.3 Recommended Applications

Tracer gas methods are best suited to research studies that are aimed at characterising the dispersion conditions at a site. Consequently, they are recommended for applications that involve quantifying the extent of dispersion from a site. Well planned tracer gas studies can be used to locate the origin of odorous or hazardous emissions and in most cases should be able to provide quantitative data for estimating environmental impacts. They could be used to do this many hundred metres from the source of the emissions but are ultimately limited by a lack of detection sensitivity.

## 2.4 Optical Integrated-Path Methods

### 2.4.1 Principle of Method

In the optical integrated-path method a collimated infrared beam is launched into the atmosphere and directed at a retro-reflecting mirror placed a suitable distance away (typically around 100 metres). The reflected beam is collected by a small telescope and the optical power is measured. The integral amount of target gas along the beam path is determined from the level of absorption of the infrared beam. This enables the integrated path concentration to be calculated from:

$$CL = \int c dl = \frac{1}{\alpha} \frac{I(l)}{I(0)}$$

where CL is the integrated path concentration  $c$  over a distance  $l$ .

Measurements are then made at different heights to determine the distribution of the gas in a vertical plane up- and down-wind of the site. These concentration measurements are combined with the wind velocity to determine the gas flux through the measurement plane.

### 2.4.2 Applications of Optical Integrated-Path Methods to Landfill Methane Measurements

The wide spatial coverage of open-path techniques means that they are often used in the measurement of emissions from large-scale diffuse sources such as landfill sites. The most common approach is to use a Fourier Transform Spectrometry (FTS) to carry out the measurement. Weber et al (1996) report FTS measurements of methane across a clay-capped landfill site. A site survey was performed in which concentrations were measured along a series of ground-level lines of sight. The maximum methane path-averaged concentration observed was 27 ppm. However, the results were not used to calculate emission fluxes.

One of the main advantages of the FTS technique is that many species can be monitored simultaneously. Haus et al (1995) have made measurements at a variety of different sites, including landfill, of numerous species including methane, carbon dioxide, carbon monoxide, ammonia, nitrous oxide and formaldehyde. Their measurements at a landfill included a site survey that showed methane concentrations varying between 4 and 20 ppm. They also measured the site flux along a 500 metre path 50 m downwind of the site boundary. The measured emission rate was  $0.36 \text{ mg m}^{-2} \text{ s}^{-1}$ . This rate was inferred from the path-averaged concentration along a single horizontal line-of-sight. A simple plume-dispersion model was used to extrapolate this measurement to give the integrated plume concentration.

The validity of using simple plume dispersion models to predict total emission rates from integrated path measurements has been studied by Piccot et al (1996). They set up a simulated diffuse methane source (30 metres by 7.5 metres) combined with a point source of tracer gas ( $\text{SF}_6$ ). Open-path FTS measurements were made in a vertical 'pie-slice' approximately 50 metres downwind of the source using four retro-reflectors mounted at different heights up to a maximum of 20 metres elevation. A plume dispersion model was used to predict the fraction of the emitted plume passing through the measurement area. The wind vector was measured at two heights (2 metres and 10 metres) and the results used to estimate the vertical wind profile. The two data sets were then combined to give the flux.

Compared to a landfill site, the experimental arrangement described by Piccot represents a relatively simple and small scale source, used for an extensive set of measurements. Despite this, the measured flux was consistently 20-40% lower than the known value. The authors trace this discrepancy to the calculation of Pasquill stability class used in the dispersion model. At the simplest level, the stability class can be determined through observation of the meteorological conditions at the time of the measurement. However, this method is somewhat subjective. A more prescriptive method relates the stability class to the standard deviation of the horizontal wind direction, and this method was used in the initial dispersion model runs. It

was found that the agreement between the measured and actual fluxes improved dramatically (2% variation) if a 'synthetic' stability class was used. In this case, the results of the simultaneous tracer measurements were used to determine the most suitable stability class on a run-by-run basis. The authors stress that this synthetic stability class does not represent the actual class, but instead the class that gives the best results in a dispersion model of a particular measurement. This work highlights the crucial importance of the dispersion model if the entire plume is not measured, and shows that simpler forms of the model can lead to significant errors in the flux calculation.

An alternative open-path measurement technique is described by Milton *et al* (1995) based on earlier work with an experimental system [Partridge *et al* 1986]. This system used a broadband light source and an oscillating optical filter to measure the IR absorption of various atmospheric gases including methane over open-paths up to 400 metres long (Figure 4). In order to simplify the mechanics of carrying out elevated measurements, a vertical slant path to a retro-reflector mounted on a portable mast (30 metres maximum elevation) was used. Where possible, measurements are made at high enough slant path angles to ensure that the entire vertical extent of the plume is measured, eliminating the dependence on the plume dispersion models discussed above. Model calculations show that with neutral stability (typical of over-cast UK conditions) 99% of the plume from a 600 metre site will be below 30 metres at the site edge. Measurements were performed at a landfill site using this technique resulting in a measured methane flux of  $1.7 \text{ mg m}^{-2} \text{ s}^{-1}$ .



**Figure 4** - Optical integrated-path monitoring using a HAWK infrared monitor along one side of a landfill site.

#### 2.4.3 Recommended Applications

The different implementations of the optical integrated-path method are capable of giving accurate results, if carried out correctly. Under favourable wind conditions, they are cost-effective methods for producing accurate measurements of total site emissions. Unlike the other methods, they are not able to provide information about the distribution of high-emission points on the site. Since they are not straightforward to implement, they can be recommended for verifying the accuracy of lower-cost and more convenient methods. Current developments in diode laser technology offer the potential for lower-cost implementations with greater sensitivity.

## 2.5 Eddy Correlation Methods

### 2.5.1 Principle of Method

The eddy correlation method is a micro-meteorological method that is based on the fundamental assumption that the emitted gas is transported away from the surface by the vertical eddying motion of the atmosphere. This vertical transport can be measured by correlating the instantaneous concentration at a point with the instantaneous vertical wind velocity at the same location. Fast response detectors are required to carry out this type of measurement since the natural frequency of the atmospheric eddies can be as high as 10 Hz.

The mean flux density ( $F$ ) over any given measurement period is given by the mean of the product of instantaneous vertical wind speed ( $w$ ) and gas density ( $\rho_g$ ). This can be written as

$$F = \overline{w \cdot \rho_g} + \overline{\Delta w \cdot \Delta \rho_g}$$

where the bars denotes the mean value over the measurement period and  $\Delta w$  and  $\Delta \rho_g$  are the fluctuations about the mean wind speed and mean gas density respectively. In the simplest case the mean vertical wind speed is assumed to be zero, so the flux is given by the second term only. More detailed calculations can be used which take into account the effects of the net heat and water vapour flux .

### 2.5.2 Related Methods

#### Relaxed Eddy Correlation

If fast response monitors are not available for use in the standard eddy correlation technique described above, then the "relaxed" eddy correlation method can be used. In this method, gas concentrations are measured at one point, and the flux is estimated using the following relationship :

$$F = \beta(C_u - C_d)\sigma_w$$

where  $\beta$  is a dimensionless constant,  $C_u$  and  $C_d$  are the mean concentrations associated with upwards and downwards eddies, and  $\sigma_w$  is the standard deviation of the wind speed.

#### Gradient Diffusion

An alternative micro-meteorological method enables the vertical flux from a surface to be determined by measuring the gas density  $\rho_g$  at different heights,  $z$ , to determine the vertical gradient of the gas. The flux is then given by

$$F = -K_g \overline{\partial \rho_g / \partial z}$$

where  $K_g$  is the eddy diffusivity of the gas which depends on the meteorological conditions, the measurement height, and the surface roughness. The determination of  $K_p$  is the key factor in the application of this method and requires a number of meteorological and local site factors to be considered.

### 2.5.3 Applications of Eddy Correlation Methods to Landfill Methane Measurement

The key advantage of the eddy correlation technique is that it enables emissions from a wide area to be monitored from a single point. A number of authors have reported methane flux measurements using the eddy correlation technique, either on landfill sites or on other terrain types. Hovde (1995) reports the use of a near-infrared diode laser sensor to make 10 Hz methane concentration measurements at a clay-capped sanitary landfill site (with no gas recovery system). Rapid response measurements were also made of the 3-D wind vector, water vapour concentration and carbon dioxide concentration. A high level of correlation was observed among the four data sets. The average methane emission rate over a combined

period of three hours was  $0.076 \text{ mg m}^{-2} \text{ s}^{-1}$ . Some comparisons were made with other methane flux measurements (Fan 1992) but the results cover a very limited data set in terms of spatial and temporal coverage.

Relatively sophisticated equipment is required for the fast response measurements of methane and the 3-D wind velocity, but this equipment is becoming more generally available. Once the equipment is set up at a site it will generally not need to be moved and can be left to run automatically, reducing staffing requirements. Once the relevant procedures have been automated, the analysis of the results is relatively straightforward and the equipment does not require frequent calibration (Oonk 1995).

In their review of different measurement techniques for measuring landfill methane Savanne et al (1997) highlight one of the main issues with the interpretation of eddy correlation measurements. They quote an average emission density of  $0.019 \text{ mg m}^{-2} \text{ s}^{-1}$ , in a comparative study approximately 40 times lower than the other techniques reviewed. Savanne attributes this discrepancy to the position of the eddy correlation sensor which was located downwind of an area of the site known to have low emissions. The location of the sensor and the ground 'footprint' from which emissions are detected need to be monitored carefully. Fan et al (1992) show how this footprint can be estimated using simple plume dispersion models. In this work the methane flux from sub-Arctic tundra was measured using an eddy correlation sensor at the top of a 12 m tower. The calculations of the footprint area show that the up-wind footprint depends upon the atmospheric stability, varying from 200 to 300 metres for unstable conditions to more than 1000 metres for stable conditions. In comparison the cross-wind footprint is small - typically only 15% of the upwind footprint. Therefore, the location of the measurement footprint is strongly dependant on the wind direction, and this needs to be taken into account when analysing eddy correlation data.

One benefit of eddy correlation over the flux chamber or flux survey is that it is less effected by the micro-topography of the site. Clement et al (1995) conducted a long-term inter-comparison of methane flux measurements from an area of peatland using the eddy correlation and flux chamber techniques. For the first eighteen months of the inter-comparison the flux chamber technique gave results approximately 30% higher than the eddy correlation, and then 20% lower for the last six months (the latter period associated with a high water table level). The correlation between the results improved significantly when the flux chamber results were corrected for the effects of the micro-topographical spatial variations in the locations of the flux chamber measurements.

#### *2.5.4 Recommended Applications*

Eddy correlation methods have the potential to measure very low fluxes, but their accuracy is limited by the uniformity of the surrounding terrain and the uniformity of emissions. Since it is difficult to use and the data is difficult to process into measured flux results, it cannot be recommended for routine measurements of landfill methane emissions. It can be recommended for long-term studies of emissions from a single site with reasonably uniform emissions over an extended period of time.

## 2.6 Differential Absorption Lidar (DIAL)

### 2.6.1 Principle of Method

The Differential Absorption Lidar (DIAL) technique is a development of the optical integrated-path method described in Section 2.4. It works by measuring the small amount of light scattered by particles in the atmosphere when an intense pulse of radiation is transmitted through the measurement region. The exact distance to the scattering volume can be determined by timing the flight of the pulse from the transmitter to the scattering point and back. The measured signal energy is given by the Lidar (Light Detection and Ranging) equation which in simplified form gives the returned power  $P_x(r)$  from a range  $r$  as:

$$P_x(r) = E_x \frac{D_x}{r^2} B_x(r) \exp\{-2 \int_0^r [A_x(r') + \alpha_x C(r')] dr'\}$$

where  $C(r)$  is the concentration of an absorber with absorption coefficient  $\alpha_x$  and  $A_x(r)$  is the absorption coefficient due to all other atmospheric absorption,  $E_x$  is the transmitted energy and  $B_x$  is the backscatter coefficient for the atmosphere at wavelength  $x$ .  $D_x$  is a range-independent constant.

Measurement of the differential Lidar signal at two wavelengths allows the range-resolved concentration to be calculated independently of the other parameters. This is the Differential Absorption Lidar (DIAL) method. Concentration measurements along different lines-of-sight are combined to generate a concentration profile across a vertical plane. The emitted flux is then calculated by combining the profiles of methane concentration in the atmosphere up- and down-wind of the measurement volume with the wind profile through the volume.

### 2.6.2 Applications of DIAL to Landfill Methane Measurements

The Differential Absorption Lidar method combines the wide spatial coverage of the open-path method with the fine detail of the point-sampling methods. DIAL systems can measure range-resolved methane concentrations at distances of up to one kilometre in any direction from the system. As a result, the entire plume distribution can be measured in almost all wind conditions, removing the need for any dispersion modelling and greatly simplifying the calculation of the emitted flux. In addition, the speed of the measurement means that the entire site flux can be measured in a few minutes. The fine scale range resolution also shows the structure of the plume which can give useful information on the emission distribution across the site.

Since a DIAL system is single-ended there is no requirement for carefully aligned retro-reflectors, leading to a considerable man-power saving over the open-path techniques. However, the main disadvantage of the DIAL method is that the equipment is large and expensive, and requires experienced operators.

Although there are a large number of DIAL systems operating around the world, measurements of methane emissions requires the use of laser transmitter sources that operate in the infrared spectral region. The number of such DIAL systems is extremely limited. Robinson et al (1995) describes the mobile DIAL system developed by NPL that has been used to measure hydrocarbon emissions (including methane) from a wide range of industrial sources. As part of the system calibration DIAL measurements have been made of the methane fluxes from a controlled emission source. The agreement between the measured and actual fluxes was within  $\pm 15\%$ . Diffuse industrial sources have also been measured, indicating that the technique can be applied to large scale sites with inhomogeneous emissions.

### *2.6.3 Recommended Applications*

The combination of wide area coverage and range-resolved measurements means that the DIAL method is well suited to carrying out detailed studies of site emissions; for instance, in the assessment of the quality of gas control measures. Since the DIAL method measures the entire emission distribution, flux calculations can be made without reference to dispersion models. This removes one source of uncertainty from flux results and means that DIAL can be used to measure emissions when topography or meteorological conditions would lead to unreliable dispersion modelling.

Another application area is in the measurement of emissions from sites where access is difficult or hazardous. Since the DIAL method is single-ended and has a range of up to one kilometre, the measurement locations can be several hundred metres from the site.

## **2.7 Other Methods**

### *2.7.1 Satellite Measurements*

There are a number of instruments operating on earth remote sensing satellites that are able to measure methane in the free troposphere and above. However, such instruments are not capable of measuring methane concentrations at distances corresponding to the typical plume height from a landfill site (typically no more than 50 metres).

### *2.7.2 Infrared Imaging*

Infrared images of landfill sites obtained using hand-held or airborne imagers are often used to provide information about the surface of sites. The images produced in the infra-red spectral region highlight variations in the temperature of objects as well as variations in their infrared emission characteristics. Although methane has a complex spectral signature in the infrared, thermal imagers do not have sufficient spectral resolution to discriminate between different causes of the variation in the amount of infrared received. Hence, they are not capable of distinguishing between variations due to the presence of methane in the atmosphere and variations due to the difference in the infrared emission characteristics of different surfaces.

### *2.7.3 Sub-surface gradient Methods*

Measurements of the gradient of the concentration of gases below the soil surface have been used extensively to study diffusion processes within the soil. These measurements are of particular interest in quantifying the influence of biological processes on the diffusion of gas through the subsoil. They can be time consuming to carry out, particularly if the soil is difficult to penetrate. Also they must be repeated a large number of times in order to develop a statistically representative result. Their accuracy depends upon the extent to which any concentration gradient measured can be interpreted by use of a simple diffusion law. These difficulties effectively limit the scope of applications of sub-soil gradient methods to scientific investigations of processes in cover soils.

### *2.7.4 Gas Bore Hole or Extraction Measurements*

It is routine operational procedure to measure the concentration of landfill gas (particularly the carbon dioxide and methane concentrations) in bore holes and gas extracted from all managed landfill sites. This information is primarily used to monitor whether subsoil conditions are anaerobic (leading to efficient methane generation) and the extent of methane migration from the site. Such "static" measurements of methane concentration do not readily give information about methane flux from the site as a whole. Even when they are combined with measurements of gas flow rates through the extraction system, they do not yield information about total methane emissions because they do not provide any measure of the amount of gas escaping through the surface or through defects in the surface or any pipework.

### 3 SUMMARY AND CONCLUSIONS

#### 3.1 Comparison of Methods

##### 3.1.1 Measurement Principles

It is difficult to make a true comparison of the methods reviewed in this study because they all attempt to estimate the same quantity (landfill methane emissions), but achieve this through monitoring different parameters that are combined on a different basis. The parameters measured by each of the methods are listed in Table A together with the principle used to combine them to calculate the flux. Each of the methods uses measurements made over a different length scale so that they yield results that are associated with different length scales. These vary from typically 1 to 2 metres for flux chambers to many hundreds of meters for the optical open-path methods. Since the total amount of methane emitted along a 100 metre path is larger than that along a 1 metre path, there is a tendency for the methods that measure over larger distances (or areas) to be more sensitive. The relationship between the minimum detectable flux and length scales of the methods is illustrated in Figure B.

**Table A: Basis for Flux Measurement for Each Method**

<b>Method</b>	<b>Parameters Measured</b>	<b>Basis of Flux Calculation</b>
<i>Flux Chamber</i>	Rate of change of concentration within enclosure.	Direct measurement of emission rate within enclosed area at a number of locations around the site.
<i>Flux Survey</i>	Point concentrations around perimeter of site (up- and down-wind)	Combination of measured concentrations and wind speed with estimated plume and wind profiles
<i>Tracer Gas</i>	Concentration of methane and tracer in plume downwind of source. Release rate of tracer.	Relate measured tracer and methane concentrations to known tracer release rate, assuming that tracer release and dispersion is representative of methane emission.
<i>Open-path Monitoring</i>	Integrated-path concentrations up- and down-wind of site.	As for flux survey, except plume profile can be measured directly.
<i>Eddy-correlation</i>	Short timescale fluctuations in methane concentration and vertical wind speed.	Correlation between vertical wind speed and methane concentration.
<i>DIAL</i>	Range-resolved methane concentration measurements in vertical planes up- and down-wind of source.	Combines measured concentration profile with measured (horizontal) wind speeds.

### 3.1.1 Minimum Detectable Flux

In this section we calculate the minimum detectable flux and the measurement uncertainty of the six methods under typical conditions. Since five of the methods are based on measurements of the concentration of methane above the surface of the site, it is convenient to compare them on the basis of the same set of assumptions about the dispersion of methane from the site surface. These assumptions are used solely as the basis for a comparison in this report. They do not relate to any limit on the applicability of the methods themselves.

#### Baseline Conditions

The UK-ADMS atmospheric dispersion model has been used to model the profile of methane above a site [Milton, NPL 1997]. These calculations are a very straightforward application of dispersion modelling which characterises the atmospheric stability conditions according to Pasquill stability classes. In Figure D, we show the concentration of methane calculated at any point along the edge of a site emitting methane uniformly at a rate of  $1 \text{ unit m}^{-2} \text{ s}^{-1}$  under Pasquill C conditions which are typical of the UK. The measurement point is chosen to be on the upwind side of a site stretching 400 metres in the upwind direction, with a wind speed of  $4 \text{ ms}^{-1}$ . Figure D shows the profile of the methane concentration up to a height of 60 metres. If the data in Figure D are scaled appropriately, then it can be shown that if the surface emission rate is  $50 \text{ ug m}^{-2} \text{ s}^{-1}$  then the excess concentration of methane over the atmospheric background at a height of 1.5 metres will be approximately  $10 \text{ ug m}^{-3}$  (ie 0.2 parts-per-million). This calculated relationship between the excess ambient concentration and the surface emission flux forms the basis for calculating the minimum flux detectable by the different methods. This is applicable to the benchmark conditions of a  $4 \text{ ms}^{-1}$  wind on a site reaching 400 metres in the up wind direction. If the wind is lower, then the minimum detectable flux will be smaller. Similarly, if the site is larger, the minimum flux detectable will also be smaller.

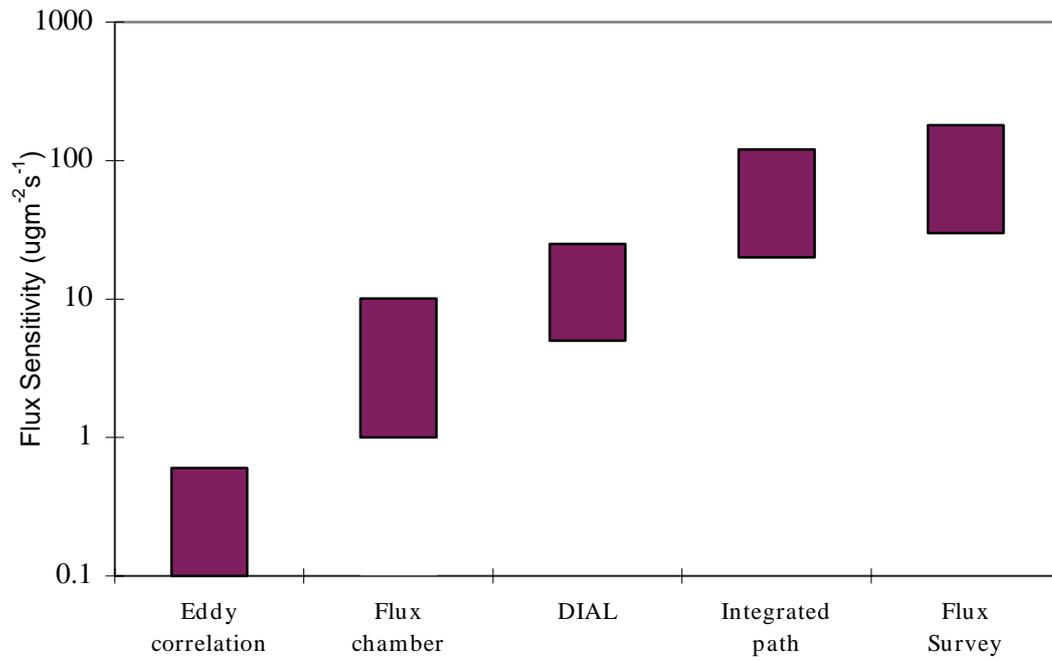
#### Optical Integrated-Path, Eddy Correlation, Flux Survey and DIAL

First we consider the optical integrated-path method. A typical minimum detectable point concentration for an optical instrument is 0.2 ppm. Hence we would expect it to be able to measure the baseline flux of  $50 \text{ ug m}^{-2} \text{ s}^{-1}$  under the conditions discussed. Taking account of a realistic range of site sizes from 200 to 600 metres, the associated range of minimum detectable fluxes would be 20 to  $100 \text{ ug m}^{-2} \text{ s}^{-1}$  [ref NPL 1995 paper]. The DIAL method is capable of measuring point concentrations of 0.05 ppm, hence the minimum detectable flux is four times lower. We estimate the minimum detectable flux for the eddy-correlation methods on the basis of using instrumentation that is capable of measuring correlated changes of methane concentration of  $1 \text{ ug m}^{-3}$  and changes in wind speed of  $0.1 \text{ ms}^{-1}$ . Allowing for a small range in each of these numbers leads to a minimum detectable flux in the range  $0.1$  to  $0.5 \text{ ug m}^{-2} \text{ s}^{-1}$ .

#### Flux Chamber

Finally, we consider the flux chamber method. Since this method involves direct measurement of the methane that has been emitted through an enclosed area of the site, the calculation is not based on the same assumptions as the other methods. If the instrument measuring the increase in methane concentration within the chamber is capable of monitoring a change of methane of the order of  $7 \text{ mgm}^{-3}\text{s}^{-1}$  (0.1 parts-per-million per minute) and it has a volume of the order  $1 \text{ m}^3$  and an area of the order  $0.5 \text{ m}^2$ , then its minimum detectable flux is of the order  $50 \text{ ug m}^{-2}\text{s}^{-1}$ .

The minimum detectable fluxes for each of the five methods are plotted in Figure 5. This illustrates that each of the methods has a different range of minimum detectable fluxes. In general, the methods can all be used to measure fluxes that are significantly larger than these minimum values



**Figure 5** - Flux sensitivities of the different measurement techniques. The range indicates the extent of variation of the sensitivity according to site and meteorological conditions.

## **3.2 Recommendations**

### *3.2.1 Costs and Availability*

It is not straightforward to compare the costs of the five methods reviewed here, because they would be used to study different qualities of problem and would yield different qualities of data. For example, one day of operation with a DIAL facility would generate many tens of thousands of accurate methane concentration measurements, but one day of operation with a flux chamber would produce no more than ten measurements of surface flux. Consequently, the following estimates of cost should be interpreted with care.

Typical implementation of the flux survey and flux chamber methods would involve two people working on a site for one day using a portable monitor, calibration gases, meteorological equipment (for the flux survey) and flux chambers (for the flux chamber method). The total cost of this equipment would not exceed £5,000 and is available from a number of UK suppliers. The results from such day-long measurements would be an estimate of total site emission flux (by the flux survey method), or several tens of surface emission measurements (using the flux chamber). Additionally the flux survey method produce detailed information about the distribution of high emission points across the site.

The eddy correlation and optical integrated-path methods would typically require a team of two people working for a week on a single average-sized site. The capital cost of the equipment would be in the range £10,000 to £30,000 for optical integrated-path. Both of these methods would yield a representative estimate of total site emissions at the end of such a survey.

The DIAL method is uniquely able to characterise a complete site within one day, including locating high emission points and producing a representative estimate of total site emissions. Such a survey would cost between £5,000 and £10,000.

### *3.2.2 Presently Available Technology*

A recommendation to use one of the methods reviewed in this study can only be made when considering a particular application. In Table B, the major advantages and disadvantages of each of the methods is summarised together with some of the applications for which the method may be most suitable.

### *3.2.3 Future Technology Developments*

The flux chamber and flux survey methods both rely upon a flame-ionization detector (FID) instrument. This is a very well developed technology that would not be expected to change in performance or cost in coming years. Hence, we would not exceed the costs or availability of these methods to change significantly in the future.

The two optically-based methods (DIAL and optical integrated path) are likely to reduce in cost and laser and optoelectronic systems become more sophisticated and compact. The eddy-correlation method relies upon fast-response anemometers and methane monitors. This might also benefit from advances in optical-based methane measurement instruments.

**Table B:** Advantages, Disadvantages and Applications for Six Different methods of Measuring Fluxes of Methane from Landfill Sites

Method	Advantages	Disadvantages	Applications
<i>Flux Chamber</i>	<ul style="list-style-type: none"> <li>- accurate measurement over small area.</li> <li>- high sensitivity if required (with long averaging time)</li> <li>- can measure NMHC as well as methane</li> </ul>	<ul style="list-style-type: none"> <li>- labour intensive</li> <li>- time consuming</li> <li>- requires large number of samples to give representative results</li> <li>- intrusive measurement technique</li> <li>- requires full access to site</li> </ul>	<ul style="list-style-type: none"> <li>- landfill cover studies</li> <li>- localised emission measurements</li> <li>- long-term studies</li> </ul>
<i>Flux Survey</i>	<ul style="list-style-type: none"> <li>- cost-effective</li> <li>- simple equipment</li> <li>- locates significant emission sources around site</li> </ul>	<ul style="list-style-type: none"> <li>- inaccurate on complex terrain or in difficult wind conditions</li> <li>- requires dispersion modelling to give total site flux</li> <li>- no measurement of vertical plume profile</li> </ul>	<ul style="list-style-type: none"> <li>- rapid, cost-effective estimation of total site emissions</li> <li>- screening of sites for quality of gas control installations</li> </ul>
<i>Tracer Gas</i>	<ul style="list-style-type: none"> <li>- no assumptions about wind profile required</li> </ul>	<ul style="list-style-type: none"> <li>- assumes point release of tracer is equivalent to diffuse methane emission</li> <li>- large number of sophisticated sample collection and release systems required</li> </ul>	<ul style="list-style-type: none"> <li>- confirming dispersion characteristics</li> <li>- source identification for odour/hazard impacts at downwind locations</li> </ul>
<i>Optical integrated-path</i>	<ul style="list-style-type: none"> <li>- large area coverage (up to 500 m)</li> <li>- vertical plume profile can be measured.</li> <li>- good accuracy</li> <li>- limited dependence on dispersion modelling</li> </ul>	<ul style="list-style-type: none"> <li>- time consuming installation of equipment (future advances may improve this)</li> <li>- provides no information about localised emissions</li> </ul>	<ul style="list-style-type: none"> <li>- verification of other methods.</li> </ul>
<i>Eddy correlation</i>	<ul style="list-style-type: none"> <li>- single point measurement with large area coverage</li> <li>- long-term (automatic) operation possible</li> <li>- measurements have high sensitivity</li> </ul>	<ul style="list-style-type: none"> <li>- initial installation complex.</li> <li>- complex data processing</li> <li>- accurate results require numerous parameters to be measured.</li> <li>- accuracy depends on surface conditions</li> </ul>	<ul style="list-style-type: none"> <li>- long-term monitoring of site emissions</li> </ul>
<i>DIAL</i>	<ul style="list-style-type: none"> <li>- large area coverage (up to 1 km)</li> <li>- measurement of complete plume distribution</li> <li>- rapid flux measurements</li> <li>- localised sources can be identified</li> <li>- no requirement for dispersion modelling.</li> <li>- no site access required</li> </ul>	<ul style="list-style-type: none"> <li>- large, complex equipment</li> <li>- reasonable road access required</li> </ul>	<ul style="list-style-type: none"> <li>- detailed studies of sites</li> <li>- screening sites for quantity of gas control installations (particularly where site access is not possible)</li> </ul>

### **3.3 Conclusions**

This review highlights the advantages, disadvantages and applications of different methods for monitoring methane emissions from landfill sites. Between them, these methods should be able to address most of the measurement problems associated with methane emissions from landfill sites.

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### **4.3 List of Acronyms**

DIAL	Differential Absorption Lidar
EPA	US Environment Protection Agency
FID	Flame Ionisation Detector
FTIR	Fourier Transform Infrared
FTS	Fourier Transform Spectrometry
GC	Gas Chromatography
HDPE	High Density Polyethylene
Lidar	Light detection and ranging
NDIR	Non-Dispersive Infrared
NFFO	Non-Fossil Fuels Obligation
NMHC	Non-Methane Hydrocarbon
SF <sub>6</sub>	Sulphur Hexafluoride
TDLS	Tunable Diode Laser Spectroscopy
VOC	Volatile Organic Compound

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