



Development of a low-maintenance measurement approach to continuously estimate methane emissions: A case study



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ABSTRACT

The chemical breakdown of organic matter in landfills represents a significant source of methane gas (CH₄). Current estimates suggest that landfills are responsible for between 3% and 19% of global anthropogenic emissions. The net CH₄ emissions resulting from biogeochemical processes and their modulation by microbes in landfills are poorly constrained by imprecise knowledge of environmental constraints. The uncertainty in absolute CH₄ emissions from landfills is therefore considerable. This study investigates a new method to estimate the temporal variability of CH₄ emissions using meteorological and CH₄ concentration measurements downwind of a landfill site in Suffolk, UK from July to September 2014, taking advantage of the statistics that such a measurement approach offers versus shorter-term, but more complex and instantaneously accurate, flux snapshots. Methane emissions were calculated from CH₄ concentrations measured 700 m from the perimeter of the landfill with observed concentrations ranging from background to 46.4 ppm. Using an atmospheric dispersion model, we estimate a mean emission flux of 709 μg m⁻² s⁻¹ over this period, with a maximum value of 6.21 mg m⁻² s⁻¹, reflecting the wide natural variability in biogeochemical and other environmental controls on net site emission. The emissions calculated suggest that meteorological conditions have an influence on the magnitude of CH₄ emissions. We also investigate the factors responsible for the large variability observed in the estimated CH₄ emissions, and suggest that the largest component arises from uncertainty in the spatial distribution of CH₄ emissions within the landfill area. The results determined using the low-maintenance approach discussed in this paper suggest that a network of cheaper, less precise CH₄ sensors could be used to measure a continuous CH₄ emission time series from a landfill site, something that is not practical using far-field approaches such as tracer release methods. Even though there are limitations to the approach described here, this easy, low-maintenance, low-cost method could be used by landfill operators to estimate time-averaged CH₄ emissions and their impact downwind by simultaneously monitoring plume advection and CH₄ concentrations.

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1. Introduction

Atmospheric methane (CH₄) has changed in concentration from 715 ppb in pre-industrial times to 1774 ppb in 2005 (IPCC, 2013), with this increase being attributed largely to anthropogenic activities (Prinn et al., 2000; Rigby et al., 2008; IPCC, 2013). Landfill gas typically comprises ~60% CH₄ and is produced primarily by the anaerobic microbial breakdown of organic matter (Hegde et al.,

2003). Sub-surface CH₄ diffuses through the soil, where it may be further attenuated by near-surface aerobic flora and may then be emitted to the atmosphere (Xu et al., 2014). The subsequent motion of the emitted methane within the Planetary Boundary Layer (PBL) is complex and depends critically on prevailing meteorology, time of day and physical properties of the surface.

As a result of CH₄ being produced below the surface of the landfill, environmental conditions at the surface do not readily affect the rate of production. However, Czepiel et al. (2003) and Xu et al. (2014) have measured an inverse correlation between CH₄ emission to air and surface atmospheric pressure from landfill sites at Nashua, New Hampshire using a tracer release method and Lincoln, Nebraska using an eddy covariance method, respectively. As atmospheric pressure increases, advection of CH₄ from the landfill

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into the air is reduced, decreasing the emission rate. Conversely, as the atmospheric pressure decreases CH₄ gas is drawn out of the landfill temporarily, increasing emission rates due to dynamic pumping. Observations made using a Tracer Release method also indicate that in dry soil conditions CH₄ emission is inversely related to ground temperature (Scheutz and Kjeldsen, 2004). This has been explained as the exponential increase in oxidation of CH₄ to CO₂ by methanotrophic bacteria as temperature increases between 2 and 25 °C in relatively dry soils (Maurice and Lagerkvist, 2004; Scheutz and Kjeldsen, 2004). An increase in temperature results in a decreased CH₄ emission, as more CH₄ is oxidised by bacteria.

To date, several approaches have been used to estimate CH₄ emissions from landfills, e.g. chamber methods, eddy covariance and co-advected proxy tracer plume measurements. Chamber and tracer approaches are only really suitable for short deployments, and so the site-wide temporal variability over weeks and months is not easily investigated. Chamber-based measurements are relatively easy to conduct as emissions can be estimated from the rate of change of CH₄ concentration in a chamber, the footprint area of the chamber and volume of the chamber. However, the main weakness of the chamber method when measuring emissions from a landfill is the typically heterogeneous nature of the landfill resulting in high spatial variability of emissions (Giani et al., 2002).

Eddy covariance (EC) methods have also been used by studies to estimate CH₄ emission from landfill over longer periods of time. Xu et al. (2014) used EC for seven months and Lohila et al. (2007) for six. Eddy covariance calculates a gas flux from the covariance between vertical wind speed and gas concentration, where both are measured at a high sampling rate, i.e. 10 Hz. The main advantages of this method are that it provides mean flux estimates over a larger area and it can be automated. One of the major shortcomings of EC for landfill applications is that there needs to be homogeneity in emission in the fetch, as prescribed by the height of the concentration and wind measurements. The EC method also requires relatively flat topography over the representative surface footprint to yield meaningful results. Neither of these prerequisites is typically expected for landfill sites. In addition, the need to get close to the edge of the area of the landfill actively emitting landfill gas, ensuring that the emissions are from the active area alone, makes measurements such as those conducted by Xu et al. (2014) difficult to perform.

A tracer release method can be used to address the emission heterogeneity issue. This method uses measurements of concentrations of a pollutant (in this case, methane) and a co-advected inert tracer gas downwind of a source of unknown flux, where the tracer is released at a known rate. The main assumption of the tracer release method is that the tracer gas and the pollutant share dispersion properties. The main advantages of the tracer release method are that micrometeorological and meteorological data are not required to calculate the emission, the calculation of the emission is relatively simple and measurement distances are only restricted by the detection limits of the gas analyser (Foster-Wittig et al., 2015). Tracer release of acetylene (C₂H₂) has been used to estimate CH₄ emissions from landfill (Czepiel et al., 1996; Mønster et al., 2014; Foster-Wittig et al., 2015) to derive instantaneous fluxes. However, even though this method been used effectively to measure CH₄ emissions from landfill, the tracer release method has limitations for use as a long-term, low maintenance measurement approach. A key logistical limitation of the tracer release method is that it requires a mobile measurement team to coordinate with the person releasing the gas and then traverse an accessible road perpendicular to the landfill plume in the time it takes for the plume to travel from the release site. This is not a trivial exercise. Another drawback is that the tracer release needs a continuous emission of tracer gas at a known rate and,

even though C₂H₂ (for example) is a relatively inexpensive gas, it is still highly flammable and subject to strict safety protocols (Foster-Wittig et al., 2015). A further difficulty with the tracer release method is in ensuring that the tracer gas is well mixed with the landfill methane as insufficiently mixed plumes can invalidate the co-advection assumption, result in large uncertainties in the emission estimate (Foster-Wittig et al., 2015).

It is the aim of this study to investigate the CH₄ mass flux from an operational landfill in Suffolk, to identify the magnitude of emission and to evaluate the uncertainties in the emission estimates. As a long-term monitoring solution, which is both less sensitive to spatial inhomogeneity in emissions than the EC method, and less resource intensive than the high-maintenance tracer release method, we propose the use of an inverse Lagrangian dispersion model in conjunction with continuous CH₄ concentration measurements to estimate the bulk net landfill CH₄ emissions. The atmospheric dispersion model back-calculates the advection of a pollutant, predicting a neutrally-buoyant particle's movement from a source to a receptor, where calculated horizontal and vertical winds dictate the pollutant's path in the atmosphere. An assumption of the model is that pollutant particles are inert on the timescales of advection between emission and measurement (Wilson and Sawford, 1995). The rate of emission can then be calculated from pollutant concentration and atmospheric turbulence measurements made at a point downwind of the source. The commercial software WindTrax (www.thunderbeachscientific.com), which we employ in this study (see Section 2.3), is based on an inverse dispersion model (Flesch et al., 1995, 2004) and developed to calculate the emission rate of a gas from an area source. Inverse modelling has been used in similar studies to estimate CH₄ emissions from waste materials (Zhu et al., 2013; Hrad et al., 2014).

The main advantage of WindTrax is its simplicity. To calculate the rate of emission from a source, WindTrax only requires input data on the location and size of the source, a measured gas concentration less than 1 km from the source, a background (assumed or measured upwind) concentration and 3D wind speed and direction. The model then outputs the mean emission rate and an explicit uncertainty in the calculation, which is expressed as the standard deviation of the mean emission rates. A major limitation of WindTrax is that it cannot calculate emissions if the aerodynamic turbulence is too great, i.e. the roughness height is >15 cm (Flesch et al., 2005, 2009; Laubach et al., 2008).

In the remainder of this paper, we present a pilot study investigating a new low-maintenance method of measuring long term CH₄ emissions from a landfill by measuring downwind CH₄ concentrations and meteorological data inverted using the WindTrax model. We present the data gathered from measurements at the Mason's Landfill in Great Blakenham, Suffolk between July and September 2014 where standard modelling methods used by the landfill operators have estimated the overall site CH₄ emission at 300 kg hr⁻¹ as an average over the year. The study presented here was part of a larger measurement campaign, the Greenhouse gas UK and Global Emissions (GAUGE). To our knowledge this is the first study to use an inverse model to continuously measure CH₄ concentrations and produce CH₄ emission estimates from a landfill over a period of 2 months. We present bulk net CH₄ emissions for this site and show how emissions vary with changing environmental conditions.

2. Materials and methods

2.1. Methane measurement instruments

Two instruments were deployed during this measurement campaign to make on-line CH₄ concentration measurements: the

Picarro Cavity Ring-Down Spectrometer (CRDS) and the Ellutia Gas Chromatograph – Flame Ionization Detector (GC-FID). The Picarro CRDS measures the gas concentration of an air sample by comparing the sample ring-down time to an absorption free ring-down time, as laser light is passed into a reflective cavity within the instrument. The Picarro CRDS reports CO₂, CO, H₂O and CH₄ concentrations every 5 s, with a precision for CH₄ of 15 ppb over an operating range of 0 to 5 ppm. Calibration of the Picarro CRDS was done daily for 10 min using low (1.93 ppm), target (2.03 ppm) and high (2.74 ppm) CH₄ gases calibrated on the World Meteorological Organization (WMO) scale. The Picarro CRDS was chosen as the “downwind” instrument as it has better precision. The Picarro CRDS was on-line via the Global System for Mobile Communications (GSM) network and real-time data could be observed from Lensfield Rd, Cambridge using TeamViewer Version 9 (TeamViewer GmbH, Uhingen, Germany).

The 200 series Ellutia GC-FID takes air to be assayed for CH₄ concentration mixed, via a 2 ml sample loop, with a carrier gas which passes through a 3 m long 1/8th in. O.D. column packed with HayeSep Q in an isothermal oven at 90 °C. As the gases exit the column they are pyrolyzed by a hydrogen/air mixture within the flame ionization detector. Ions formed during the combustion are measured to indicate the concentration of the gas species. The Ellutia GC-FID, as used here, has a detection limit of approximately 1.5 ppb and measures concentrations every 75 s. The instrument is calibrated every 30 min using a gas standard (2071 ppb) calibrated to the same scale as the Picarro instrument. To reduce the maintenance requirements of the GC, the carrier and fuel gases required were supplied by a Parker hydrogen generator and the air was supplied by a pressure controlled air pump that kept a low volume cylinder at 2 bar throughout the campaign. The GC was controlled and data was stored using an Arduino Mega (Arduino LLC, Somerville, MA, USA); the Arduino also sent hourly SMS message of the peak height to show the GC was still operational.

2.2. Field methodology

2.2.1. Masons Landfill Site, Great Blakenham

Between July and September 2014, Masons landfill was accepting unrecyclable waste on an open active area of approximately 70,000 m² (dotted area in Fig. 1). All methane emissions were coming from the active landfill region as shown in Fig. 1. To the north of the active area is ~120,000 m² of decomposing sub-surface waste permanently capped with a welded high-density polyethylene (HDPE) membrane and restored with at least two meters of soil (slanted lines in Fig. 1). An area of ~70,000 m² of waste temporarily capped with a welded HDPE was located to the east of the active area (horizontal lines in Fig. 1). Landfill gas was extracted operationally from the capped areas under suction using a network of pipes and wells and used to fuel on-site generators to produce around 2.5 MW of electricity from recovered CH₄. Surface emissions measurements using flux chambers by Ground Gas Solutions (Manchester, UK) showed that there were no measurable fugitive emissions of CH₄ from the capped areas and they do not contribute to the emissions source area.

Waste was added to the active area during weekdays, Monday to Friday, between 0800 and 1700 and between 0800 and 1300 on Saturday. Deposited waste is (and was historically) mechanically compacted *in situ* to achieve the desired distribution and density. During non-working hours the actively emitting area was covered with soil to reduce animal activity and fugitive emissions. When uncovered, the trafficked areas of the active site were sprayed with water during dry conditions to prevent dust emissions.

2.2.2. Picarro CRDS - Chalk Hill Lane, Great Blakenham

The Picarro CRDS was deployed in a shelter on Chalk Hill Lane (52°06'53"N, 1°05'11"E) 300 m North-East of the perimeter of the landfill site (location denoted by the white cross in Fig. 1). The sampling inlet was 4 m above the Picarro and air was drawn down PTFE tubing (1/4 in. O.D., I.D. 3/16 in.). The inlet was protected from water incursion using an aluminium funnel and filtered using a 2 µm filter.

The location of this site was carefully considered before deployment and was especially chosen for the topography between the source and the detector. Care was made to ensure that there were no obstructions between the landfill and the inlet and that the inlet was as far as possible from the landfill to mitigate mechanical turbulence.

2.2.3. Ellutia GC-FID – Ingham's Farm, Little Blakenham

The Ellutia GC-FID was deployed at Ingham's Farm, Little Blakenham (52°06'16"N, 1°04'13"E), 300 m to the South-West of the landfill (location denoted by the white square in Fig. 1). This was used to measure the background concentration of CH₄ in air before it reached the landfill (Fig. 1). The farm is 700 m SW of the landfill site. The inlet tube (PTFE, 1/4 in. O.D., I.D. 3/16 in.) was attached to a 4 m mast, protected from water incursion using an aluminium funnel and filtered using a 2 µm filter.

All instruments were deployed from the 21st July 2014 to the 28th of September 2014.

2.2.4. Meteorological data – Weighbridge, Masons Landfill, Great Blakenham

Meteorological data were collected by a Skylink Pro instrument (Davis Vantage Pro System, Hayward, CA, USA) at the weighbridge of the landfill site. Data collected at the weighbridge included: wind speed (m s⁻¹), wind direction (°), air temperature (°C), humidity (%), rain rate (mm h⁻¹), air pressure (Pa), cloud height (ft) and solar radiation (W m⁻²). Wind data collected 2 dimensional, data is measured at 10 s intervals and averaged to 10 min.

2.3. WindTrax inverse dispersion model

Atmospheric dispersion models, such as WindTrax, mathematically model the path of pollutants in the atmosphere as they move from a source (Wilson and Sawford, 1995). WindTrax calculates an emission rate by modelling the random movement of thousands of pollutant particles as they are displaced by horizontal and vertical aerodynamic forces. The simulated ratio of concentration at measurement site to emission rate from the source, $(C/Q)_{sim}$, is calculated (Eq. (1)) from the total number of gas particles released at the measurement site (N) and the modelled vertical velocity of particles at “touchdown”, summed across all instances where a particle impacts the ground within the emission source area (w_0 , m s⁻¹) (Flesch et al., 2004, 2005).

$$\left(\frac{C}{Q}\right)_{sim} = \left(\frac{1}{N}\right) \sum \left|\frac{2}{w_0}\right| \quad (1)$$

The $(C/Q)_{sim}$ is used with the measured gas concentration (X_m , g m⁻³) and the background gas concentration (X_b , g m⁻³) to calculate the emission rate (Q , g m⁻² s⁻¹) (Eq. (2)).

$$Q = \frac{X_m - X_b}{\left(\frac{C}{Q}\right)_{sim}} \quad (2)$$

An advantage of WindTrax is that it only requires input data on the size of the emission source, the CH₄ concentration at the detector, background concentration and wind speed to calculate the emission from a source. More data describing the 3D motion of air can be used as input to reduce the uncertainty of the emission,

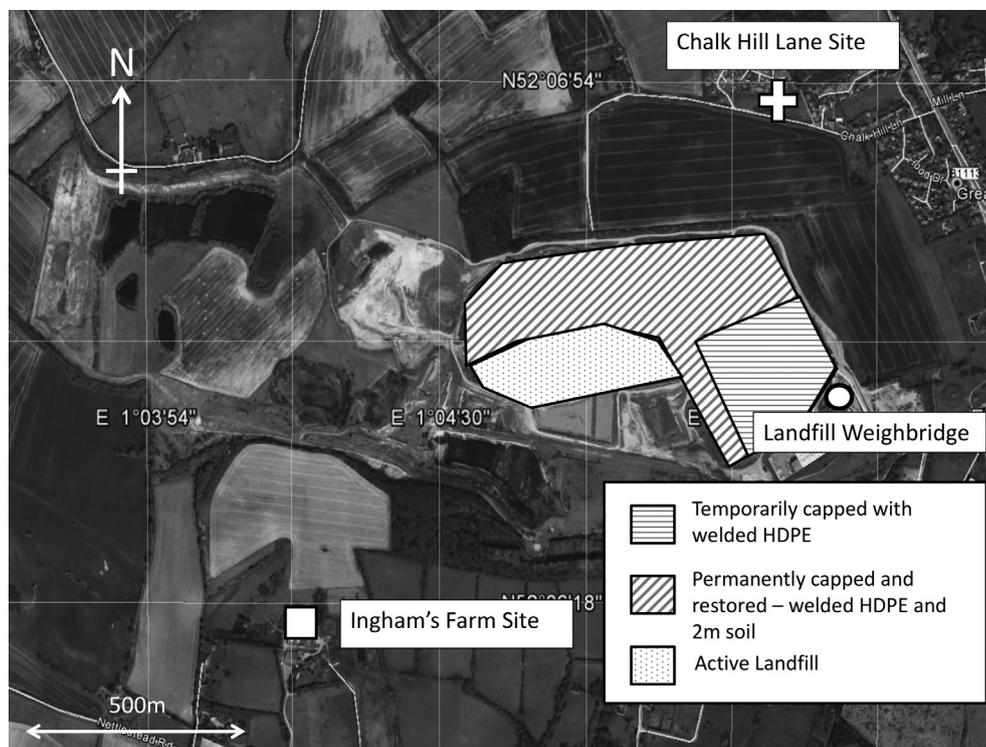


Fig. 1. Location of Chalk Hill Lane and Ingham's Farm measurement sites near Masons Landfill, Great Blakenham, Suffolk (52°06'37"N, 1°04'53"E). The coloured areas show the main areas within the landfill. The map was taken on 14th April 2015, courtesy of Google Earth. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

e.g. friction velocity and Monin-Obukhov length. These data were not collected because we could not guarantee the security of a sonic anemometer left at the side of a public road. The ideal terrain is an obstruction free surface, with maximum roughness length of 15 cm (Sommer et al., 2005; Laubach et al., 2008) and a maximum distance between the source and the detector of 1 km (Flesch et al., 2005, 2009). One important disadvantage of WindTrax is the uncertainty caused by mechanical turbulence (Denmead et al., 2008), as very few surfaces are perfectly flat. This was minimized in this study by selecting a measurement site where the fetch from the landfill to the inlet was free of obstruction >1 m tall.

Even though WindTrax has been used with good effect to estimate trace gas emissions from various sources it does have limitations. One of the most important caveats of the model for this application is that the land between the source and detector must be uniform or relatively flat terrain. The effects of mechanical turbulence from topography may be overcome by careful selection of the detector such that it is located sufficiently far away from any aerodynamic obstacle. In this context, the site is much more of a landfill than a land-raise, being a back-fill of a sand/gravel excavation. As such the site stands relatively modestly above its surrounds (7 m max) and unlikely to be the dominant uncertainty in this case.

2.4. Methane emission calculations – input to WindTrax

The inversion function of the WindTrax atmospheric dispersion model version 2.0.8.8 (Flesch et al., 1995) is used to infer the CH₄ emissions from the landfill. Methane emissions are calculated using measured CH₄ concentration 300 m downwind, measured background CH₄ concentrations 700 m upwind, and the simulated ratio of CH₄ concentration to emission described above (Flesch et al., 2004, 2005).

To reduce any impact of mechanical turbulence while maintaining real changes to CH₄ emission caused by changing environmental

or atmospheric factors, both CH₄ concentrations and meteorological data are averaged over 15 min (Laubach et al., 2008; Flesch et al., 2009). Each of the 6600 15-min-averaged measurements are used as input data to back-calculate the CH₄ emission using 50,000 particle projections. Data used as input to WindTrax are: wind speed (u , m s⁻¹), wind direction (WD , °), temperature (T , °C), CH₄ concentration at 4 m (X , μg m⁻³), background CH₄ concentration at 4 m (X_b , μg m⁻³), the roughness length (z_0 , m) and the Pasquill-Gifford atmospheric stability class. The roughness length was estimated from observation and the option used was “tall grass” ($z_0 = 10$ cm) in the surface data sub menu of WindTrax. The Pasquill-Gifford atmospheric stability class (A–F) was assigned using wind speed and solar radiation data (Seinfeld and Pandis, 2006). Each 15-min average CH₄ concentration data value is screened for erroneous values and data are removed for any periods where wind did not come from the landfill, i.e. not between 170° and 260°, or for high atmospheric stability events, i.e. wind speed, $u < 0.15$ m s⁻¹.

2.5. Methane emission due to changing environmental conditions

To investigate potential relationships between environmental conditions and CH₄ emission, the following steps were conducted. Temperature measurements were rounded to the nearest 1 °C value and plotted against corresponding average CH₄ emission rates. Similarly, wind speed measurements were rounded to the nearest 1 m s⁻¹ value and plotted against corresponding average CH₄ emission rates. The travel time between the landfill and detection by the instrument was accounted for in the analysis.

The effect of changes in atmospheric pressure on CH₄ emission were also investigated. Studies have shown an inverse relationship between the methane emission and atmospheric pressure, where the emission decreases minutes after an increase in atmospheric pressure (Xu et al., 2014). To investigate the potential relationship between pressure change and emissions for the Suffolk site, we

include a box plot of the emission against the pressure change for each 15-min time measurement.

3. Results and discussion

The instruments operated in Great Blakenham from the 22nd July 2014 until the 28th September 2014 and were monitored and controlled from Lensfield Rd, Cambridge. As a low-maintenance instrument, the Picarro CRDS measured near-continuously and only stopped collecting data during power outages longer than the UPS lifetime of 15 min. Data collection from the gas chromatograph was interrupted more often, but overall the GC deployment still represented a success as it ran independent of gas cylinders, was controlled using Arduino technology and communicated with Cambridge via SMS for the duration of the deployment.

3.1. Time varying CH_4 concentrations and emission

Of the 6600 concentration measurements used as input data, WindTrax made 1800 inverse emission estimates. The number of inverse emission estimates was less than the number concentration measurements as data were filtered to only include only those that met the following criteria: (1) if $u > 0.15 \text{ m s}^{-1}$ (averaged over the 15-min window; 100% of data); (2) if $>5\%$ of particles intersected the active area of the landfill for each 15-min window (16% of data); and (3) wind direction between 170 and 260° (averaged wind direction for each 15-min window; 28% of data). No data were removed using criteria 1 and the removal of data using criteria 2 inadvertently fulfilled the action of criteria 3. In addition, similar to the method of Flesch et al. (2004), emissions calculated when less than 5% of modelled Lagrangian particles made contact with the surface within the source area were removed due to uncertainty resulting from unknown advection in the viscous surface boundary layer and from the underestimation of $(C/Q)_{\text{sim}}$ (Eq. (2)) caused by air being blown from the edges of the active area (Supplementary Material Section 2). Five percent of particles intersected the active area of the landfill was chosen as the limit to filter data as a trade-off between loss of data and the cost of bias caused by filtering in this way Figs. S.M.1 and S.M.2 in Supplementary Material Section 1 shows that the average CH_4 emission varies much less due to percentage of particles intersecting the active area after 5%. After filtering the data in this way, this study presents 620 CH_4 emission estimates between the 27th July and 28th September 2014 (Fig. 2), each representing 15 min each. The CH_4 emissions calculated by WindTrax varied from a minimum of zero to a maximum value of $6.21 \text{ mg m}^{-2} \text{ s}^{-1}$ on the 19th September.

3.2. Variability in emission

The mean emission rate was calculated as $709 \mu\text{g m}^{-2} \text{ s}^{-1}$ (median $388 \mu\text{g m}^{-2} \text{ s}^{-1}$; 5th percentile = $8 \mu\text{g m}^{-2} \text{ s}^{-1}$; 95th percentile = $2555 \mu\text{g m}^{-2} \text{ s}^{-1}$), equivalent to $61 \text{ g m}^{-2} \text{ day}^{-1}$, during August and September 2014 (Fig. 3A). Landfill CH_4 fluxes are dependent on many different factors (temperature, pressure and contents of waste), and so it is not always useful to compare emissions between landfill sites. However, the CH_4 fluxes estimated in this study are reasonably comparable to other landfill sites studied $2164 \mu\text{g m}^{-2} \text{ s}^{-1}$ (Czepiel et al., 1996), $1576 \mu\text{g m}^{-2} \text{ s}^{-1}$ (Goldsmith et al., 2012) and $250 \mu\text{g m}^{-2} \text{ s}^{-1}$ (Xu et al., 2014).

3.2.1. Diurnal variation

In general, the median CH_4 emission is relatively constant throughout any particular day, although emissions display a

greater variability from 0600 UTC until midnight compared to the early hours (Fig. 3). Indeed, the highest variability is observed at 0600 where the 75th percentile of emissions is in excess of $3500 \mu\text{g m}^{-2} \text{ s}^{-1}$. Between 0700 and 1700 the 75th percentile of CH_4 emission estimates vary between a maximum of $1500 \mu\text{g m}^{-2} \text{ s}^{-1}$ and a minimum of $500 \mu\text{g m}^{-2} \text{ s}^{-1}$ with only slight fluctuation and no discernible trend. The very low emissions at 0500 followed by a large change at 0600 may be caused by changing pressure gradients at dawn or it may coincide with the breakdown of the night time low-level inversion and what we observe is a pulse of night-time emissions which have pooled, i.e. collected over the landfill, and would not be accounted for in WindTrax.

3.3. Response of methane emission to changing environmental conditions

3.3.1. Temperature

When CH_4 emission is grouped and averaged by temperature (rounded to the nearest 1°C), CH_4 emission shows a weak inverse relationship to temperature (Fig. 4 Top Pane), though the variability is high. This is consistent with an increase in the activity of methane oxidizing methanotrophic bacteria with increasing temperatures, although this has only been observed during dry soil conditions (Maurice and Lagerkvist, 2004; Scheutz and Kjeldsen, 2004).

3.3.2. Wind speed

When CH_4 emission is averaged by wind speed (rounded to the nearest 1 m s^{-1}), CH_4 emission shows a weak positive relationship to wind speed (Fig. 4 Middle Pane). This may be explained by considering that an increasing wind speed decreases the aerodynamic resistance to methane emission from the surface of the landfill and acts to increase the net methane emission to the atmosphere. Also, high wind speeds tend to occur in more neutral convective conditions which tend to favour horizontal advection over vertical, leading to more favourable source-receptor transport characteristics for more accurate atmospheric dispersion modelling. Additionally, Poulsen and Moldrup (2006) observed that changes in wind turbulence at the surface of the landfill, caused by changes in wind speed, can cause pressure fluctuations in the soil resulting in vertical transport of landfill gas.

3.3.3. Changes in atmospheric pressure

This pressure-emission relationship is expected as a result of the gas extraction process, where gas is extracted by applying suction to the ground. The site takes time to respond to atmospheric changes and therefore lags the atmosphere: rising pressure leads to higher pressure gradients into the ground, reducing advective losses (Young, 1990; Czepiel et al., 1996, 2003; Poulsen et al., 2003; Gebert and Groengroeft, 2006; Xu et al., 2014). This was not statistically significant (Fig. 4 Bottom Pane; $R^2 = 0.0015$, p -value = 0.15) in our study, possibly because the sampling period used by our method could have been too long to observe this effect. Unfortunately, the length of sampling period is defined by the standard methods of the WindTrax model where a 15-min average is used to reduce any impact of mechanical turbulence while maintaining real changes to CH_4 emission caused by changing environmental or atmospheric factors.

The suppression and emission of CH_4 during high and low pressure events as observed by Xu et al. (2014) were not as obvious in our result (Fig. 5). This may be a result of several contributing factors; (1) landfill operations were on-going through the measurement campaign with landfill being moved around the active area contributing to a dynamic emission landscape and (2) as a result of a non-constant wind (direction and speed) resulting in data

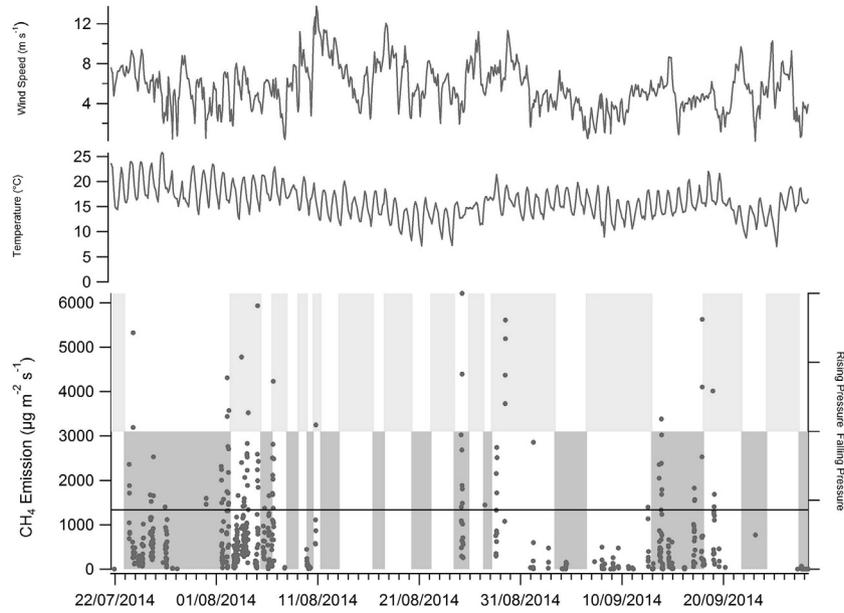


Fig. 2. Wind Speed (top), temperature (middle) and methane emission rate (black line; bottom) as calculated by the WindTrax atmospheric dispersion model from data collected at Masons Landfill, Suffolk between the 27th July and 28th September 2014.

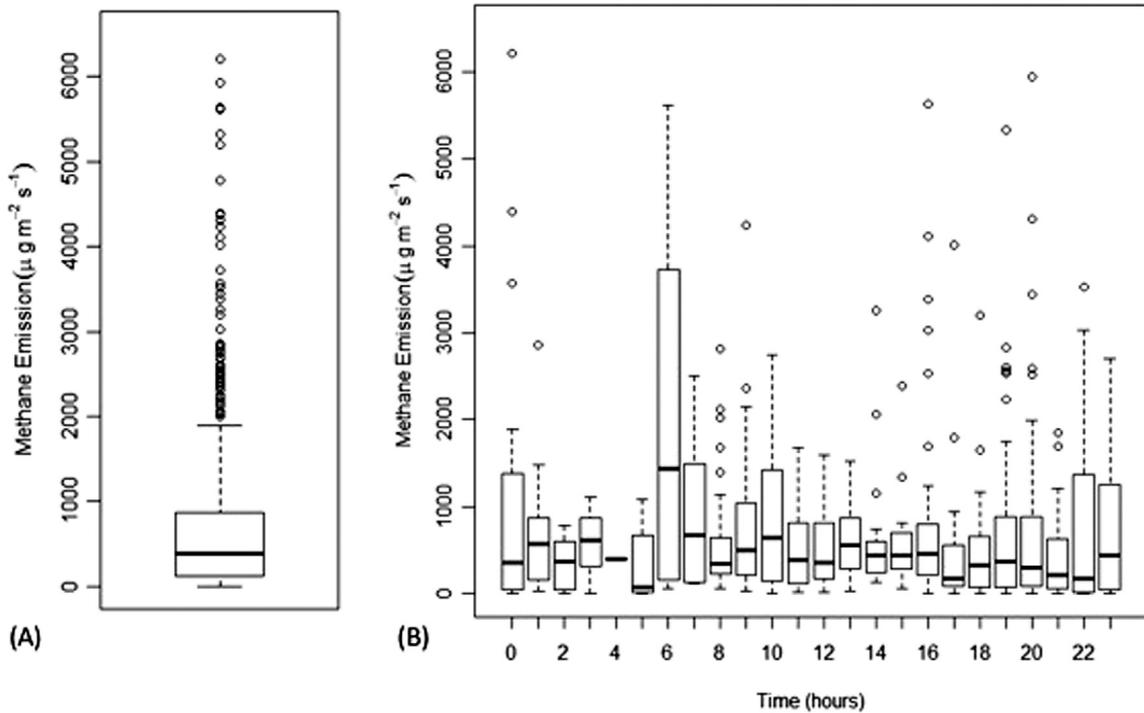


Fig. 3. (A) Methane emissions averaged 15-minutely between 21st July and 27th September. (B) Diurnal pattern of methane emissions averaged 15 minutely between 21st July and 27th September. For each boxplot, the middle line is the median and the top and bottom lines are the 75th and 25th percentile, respectively. The distance between the 75th and 25th percentile is the interquartile range (IQR). The top whisker shows either the maximum value or the 75th percentile plus 1.5 times the IQR, whichever is smaller. The bottom whisker shows either the minimum value or 25th percentile minus 1.5 times the IQR, whichever is smaller.

gaps, variability of CH₄ emissions caused by changes in air pressure are difficult to discern as air from one emission area is not measured for a significant amount of time. The measurement site of Xu et al. (2014) was in the centre of the landfill and emissions could be calculated from all wind directions. This study used a remote site that could only make emission estimates in certain wind directions.

3.3.4. Emission variability

Since temperature, wind speed and pressure are inter-dependent atmospheric quantities, a multivariate analysis was also performed using temperature, wind speed and atmospheric pressure in the linear modelling package in the R statistical environment (R Development Core Team, 2008). This shows that even though there is a positive correlation (Fig. 4), wind speed has the

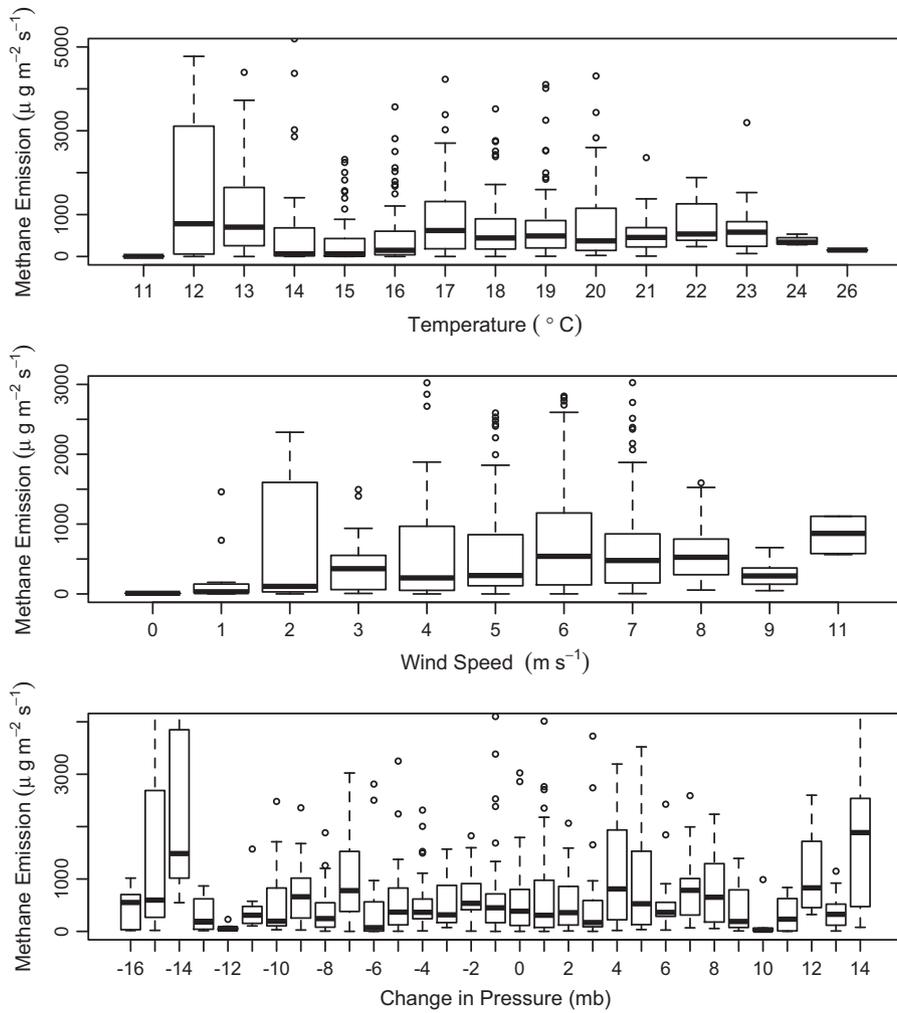


Fig. 4. Top pane: Temperature measurements rounded to the nearest 1 °C value and plotted against average CH₄ emission rates binned by temperature. Middle pane: Pressure measurements rounded to the nearest 200 Pa value and plotted against average CH₄ emission rates binned by pressure. Bottom pane: Wind speed measurements rounded to the nearest 1 m s⁻¹ value and plotted against average CH₄ emission rates binned by wind speed. For display purposes, the y-axis has been limited.

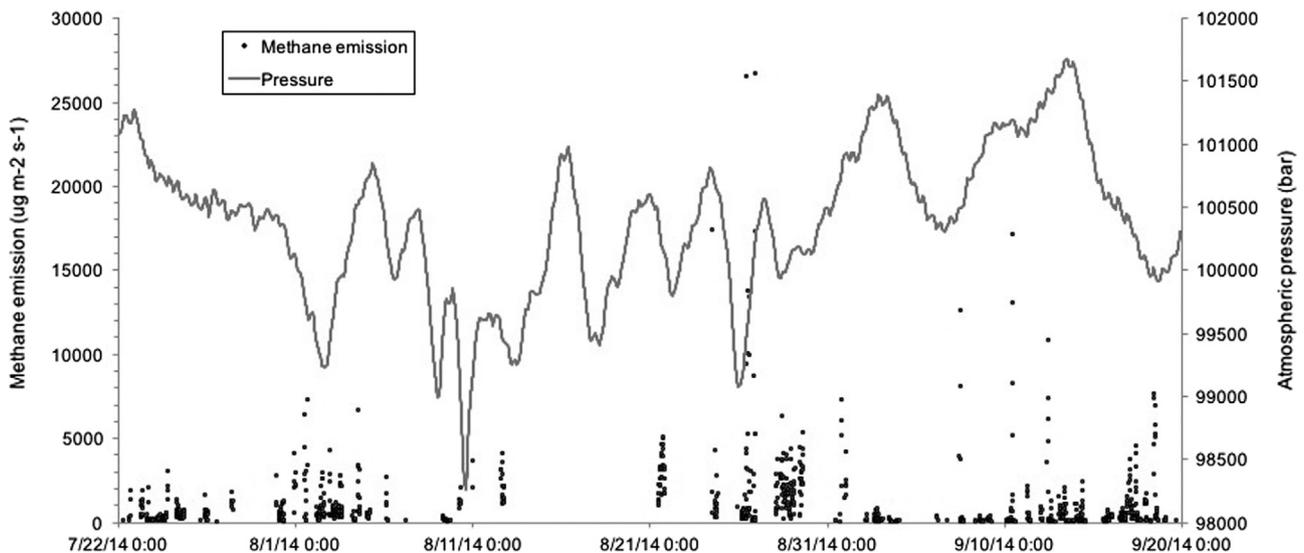


Fig. 5. Methane emission time series from Masons landfill (black squares) from 22nd July to 20th September 2014 and barometric atmospheric pressure (grey line line). Emission data points represent 15-min CH₄ emission rate and average barometric pressure. Data gaps were due to wind not from landfill (as described in Section 3.1).

Table 1

Uncertainty analysis conducted on the methane emission estimate as calculated within the WindTrax atmospheric dispersion model.

Variable	Value used	Average emission ($\mu\text{g m}^{-2} \text{s}^{-1}$)	Average emission (kg h^{-1})	Uncertainty (%)
Baseline	$z_0 = 10 \text{ cm}$, Area = 70,000 m^2	709	188	
Roughness length	10 cm	752	199	10
Emission area	140,000 m^2	376	189	1
PG stability class	+1 Stability class	964	256	36
PG stability class	−1 Stability class	518	138	−20

least significant effect on methane emission (p -value = 0.13). The effect of temperature on CH_4 emission is not statistically significant (p -value = 0.05).

However, these correlations do not explain much of the observed variability, and so we propose that the existence of discrete CH_4 emission hotspots may be responsible for the obvious peaks in emission (Fig. 2 bottom pane). We suggest the large peaks observed may be the result of wind blowing directly from the hotspot to the detecting instrument. Hotspot location may be expected to change as refuse and soil is moved around the site.

This observation is a limitation of the near-source method of estimating methane emission from landfill as the measured emission variability is not driven by environmental variables (wind, barometric pressure and temperature) but is driven by the spatial variability in location of the emission hotspots. We suggest that the greatest variability in CH_4 emission is a consequence of our sampling location, coupled with the non-uniform distribution of CH_4 emission throughout the source area. As we measure CH_4 concentrations relatively close (700 m) to the landfill site, large estimated emissions can be observed when air moves directly from CH_4 hotspots to the instrument without mixing. Similarly, lower estimated emissions can occur when these hotspots are under sampled by the air which reaches the measurement site. This may bias our overall emission estimates positively or negatively depending on the amount of time we are downwind of a hotspot. To remedy this in future we suggest moving farther from the landfill site, so that more mixing of the entire landfill plume can occur. In addition to this we recognize the shortcomings of WindTrax modelling plume rise in convective conditions at this distance from the source. Boundary layer dynamics not completely considered by WindTrax most obviously affect the modelled emissions in the evening/morning resulting in the greatest variability. This may suggest this method may be most useful when estimating emissions in neutral conditions and may be more uncertain during periods of greater atmospheric instability.

3.4. Uncertainty analysis

An uncertainty analysis was also conducted, where potential variant input values were used as an ensemble in re-run WindTrax scenarios to calculate the sensitivity of the calculated CH_4 emissions by changing one input variable at a time. These uncertainties were then combined as the square root of the sum of the squares of the instantaneous (15 min) uncertainties to give an overall uncertainty in emission estimate over the 2-month period (Riddick et al., 2014, 2016). Scenarios were run in WindTrax to reflect variability in roughness length, which propagates a 10% uncertainty in the CH_4 emission flux (Table 1; $z_0 = 10 \text{ cm}$). Varying the emission area to a maximum of 140,000 m^2 has a negligible effect on emission and resulted in $\sim 1\%$ increase in modelled CH_4 emissions. The uncertainty in estimating CH_4 emissions caused by the assignment of the Pasquill-Gifford stability class from meteorological conditions was the greatest source of uncertainty resulting in an uncertainty of the emission estimate of $\pm 40\%$. The overall uncertainty in CH_4 emission, calculated as the root of the sum of each component

squared, is estimated to be $\pm 42\%$, leading to a CH_4 emission estimate of $4512 \pm 1895 \text{ kg CH}_4 \text{ day}^{-1}$.

The inverse dispersion method for estimating methane emissions from a landfill in this study has a higher uncertainty when compared to uncertainties in other currently used measurement methods, $\pm 32\%$ and c. $\pm 25\%$ for the eddy covariance (Xu et al., 2014) and tracer dilution methods (Czepiel et al., 1996; Mønster et al., 2014; Foster-Wittig et al., 2015), respectively. However, this uncertainty may be acceptable when considering inverse dispersion as a method for long term monitoring of landfill emissions using lower cost methane measurement sensors. It would also be reduced by including additional sensors.

3.5. Comparison to other measurements

Mønster and Scheutz (2015) measured CH_4 emissions from Masons Landfill at the same time as our campaign. The only time that both Mønster and Scheutz (2015) and this study measured at the same time was the 12th August 2014 between 1630 and 1830 h, where they estimated an emission of $247 \text{ kg CH}_4 \text{ h}^{-1}$ (S.E. 20) and our estimate at the same time was $365 \text{ kg CH}_4 \text{ h}^{-1}$ (S.E. 78).

Sonderfeld (in preparation) used an FTIR at the edge of the active area to measure CH_4 concentrations and applied a CFD model to estimate the CH_4 emission at 76 kg h^{-1} as an average between the 9th and 12th August. The measurements of Sonderfeld (in preparation) were taken very close to the active area, our measurements were taken 700 m away and Mønster and Scheutz (2015) were measured between 1.5 and 6 km downwind of the landfill.

4. Conclusions

The data presented in this paper give the first near-continuous and autonomous measurement-based CH_4 emission flux from an active landfill using the WindTrax atmospheric dispersion model. Continuous measurements of methane and meteorological parameters were made from July to September 2014 on north-east and south-west sides of the Masons landfill in Suffolk, United Kingdom. This approach was found to be valid when air flows from the source to the measurement site (between 170 and 260° for the averaged wind direction for each 15-min window) and meets the following criteria: (1) if $u > 0.15 \text{ m s}^{-1}$ (averaged over the 15-min window); and (2) if $> 5\%$ of particles intersected the active area of the landfill for each 15-min window, which in our study occurred 10% of the time during a measurement period of 69 days. We found that mean methane emissions over this period were $709 \mu\text{g m}^{-2} \text{s}^{-1}$ on average, with a maximum value of $6.21 \text{ mg m}^{-2} \text{s}^{-1}$. The largest values are found in the morning and evening and these may be caused by planetary boundary layer effects. Scaled up to the area of the Suffolk site, we estimate the total net CH_4 emissions at Masons Landfill to be 188 kg h^{-1} ($61 \text{ g m}^{-2} \text{ day}^{-1}$) which is similar to the landfill owner's estimates of 300 kg h^{-1} generated by waste industry-standard landfill gas resource modelling methods (Davies et al., 2011). The CH_4 emissions estimated at Masons Landfill during August and September are similar to measurements

made by Mønster and Scheutz (2015) at the same time and to previous studies for other active landfills made elsewhere. For example, CH₄ emissions of 187 g m⁻² day⁻¹ were measured at active landfill sites in New Hampshire, USA (Czepiel et al., 1996), 84 g m⁻² day⁻¹ in Colorado, USA (Goldsmith et al., 2012), and 117 g m⁻² day⁻¹ in Wisconsin, USA (Goldsmith et al., 2012).

One highlight of this measurement campaign is the range of emissions to a maximum of 6.21 mg m⁻² s⁻¹ from the landfill showing the heterogeneous and time-varying emission landscape within the active landfill area. We cannot confirm previous findings that changes in atmospheric pressure influence the magnitude of CH₄ emissions. Contrary to previous studies, our data show no clear relationship between CH₄ emission and changing atmospheric pressure and any relationship is not statistically significant. This may be a result of the sampling period being too long to observe the changing emission, i.e. emission changes from the landfill may occur less than 15 min after the change in atmospheric pressure. Another reason could be that the inverse dispersion method is just not that accurate for short-term whole-landfill methane emissions as the landfill emission landscape has many hot spot emission regions. With longer term measurements this may be aggregated to give an emission estimate from the entire active area.

A positive correlation between wind speed and methane emission, possibly due to a decreased aerodynamic resistance as wind speed increases, is observed but is not statistically significant. The observations indicate CH₄ emissions could be inversely related to ground temperature, suggesting methanotrophic bacteria are more active than methanogenic bacteria at higher temperatures. However, statistical analysis suggests that the effects of both wind speed and temperature on methane emission are insignificant in this case.

Our high-resolution CH₄ data allow investigation of underlying processes in formation and subsequent CH₄ emission from a landfill. The temporal variability in emissions may be caused by topographical effects, with the high peaks in the late evening or early morning being associated with boundary layer transitions, e.g. a “drainage flow” regime, as the downwind monitoring location is at a lower elevation than the site. Drainage flow has been observed by other studies and has been explained as the flow of CH₄, pooled overnight, moving *en masse* on early morning winds resulting in higher concentrations measured downwind (Foster-Wittig et al., 2015). Alternatively, the variability could be caused by the operational practice of covering the active part of the site in the evening and uncovering it in the morning.

The use of a fixed site used to capture plume measurements downwind of the landfill successfully allowed for emission estimates to be made in a range of conditions over the 69-day measurement period. A drawback in this methodology is that the instruments were measuring background air for most of the time and only measured the landfill plume when the wind direction was favourable, approximately 10% of the measurement period.

This study describes the novel use of low-maintenance instruments and an inverse dispersion model to estimate long-term landfill CH₄ emissions. It is our intention to develop this methodology to make near-continuous landfill plume measurements using additional methane sensors deployed around the landfill, where at least one gas analyser will measure the landfill emission at any time. This method has the advantage over other measurement technologies, such as tracer release, as it essentially passive sampling over long periods of time which, after initial deployment, does not require significant attention from an operator. In fact, given the range of concentrations measured and the use of a 15-min-averaged concentration measurements, many cheaper gas analysers with lower specifications, such as the low-cost solid state sensors which have been used to make long term methane concentration

measurements (Eugster and Kling, 2012), could be used in a network surrounding the landfill to collect CH₄ concentration data from all wind sectors and allow for continuous emission estimates to be made. When coupled with continuous meteorological measurement this could be used for longer periods and used to monitor landfill emissions for prolonged periods. In addition, analysis of the variability in the measurements would improve the understanding of the relationship between meteorology and CH₄ emission and provide a method for predicting the response of CH₄ emissions.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.wasman.2016.12.006>.

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