Waste Management xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

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

S.N. Riddick [a,*], B.R. Hancock [a], A.D. Robinson [a], S. Connors [a], S. Davies [b], G. Allen [c], J. Pitt [c], N.R.P. Harris [a,1]

[a] Centre for Atmospheric Science, Dept. of Chemistry, University of Cambridge, United Kingdom
[b] Viridor Waste Management Limited, Peninsula House, Rydon Lane, Exeter, United Kingdom
[c] Centre for Atmospheric Science, University of Manchester, United Kingdom

ARTICLE INFO

Article history:
Received 6 June 2016
Revised 2 December 2016
Accepted 4 December 2016
Available online xxxx

Keywords:
Methane
Landfill
Low-maintenance
Measurement
Emission
Meteorology

ABSTRACT

The chemical breakdown of organic matter in landfills represents a significant source of methane gas (CH_4). Current estimates suggest that landfills are responsible for between 3% and 19% of global anthropogenic emissions. The net CH_4 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 CH_4 emissions from landfills is therefore considerable. This study investigates a new method to estimate the temporal variability of CH_4 emissions using meteorological and CH_4 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 CH_4 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 $\mu g\ m^{-2}\ s^{-1}$ 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 CH_4 emissions. We also investigate the factors responsible for the large variability observed in the estimated CH_4 emissions, and suggest that the largest component arises from uncertainty in the spatial distribution of CH_4 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 CH_4 sensors could be used to measure a continuous CH_4 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 CH_4 emissions and their impact downwind by simultaneously monitoring plume advection and CH_4 concentrations.

1. Introduction

Atmospheric methane (CH_4) 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% CH_4 and is produced primarily by the anaerobic microbial breakdown of organic matter (Hegde et al., 2003). Sub-surface CH_4 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 CH_4 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 CH_4 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 CH_4 from the landfill

* Corresponding author at: Department of Civil and Environmental Engineering, Princeton University, United States.
E-mail address: sriddick@princeton.edu (S.N. Riddick).
[1] Present address: Centre for Atmospheric Informatics and Emissions Technology, Cranfield University, United Kingdom.

http://dx.doi.org/10.1016/j.wasman.2016.12.006
0956-053X/

into the air is reduced, decreasing the emission rate. Conversely, as the atmospheric pressure decreases CH_4 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 CH_4 emission is inversely related to ground temperature (Scheutz and Kjeldsen, 2004). This has been explained as the exponential increase in oxidation of CH_4 to CO_2 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 CH_4 emission, as more CH_4 is oxidised by bacteria.

To date, several approaches have been used to estimate CH_4 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 CH_4 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 CH_4 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 (C_2H_2) has been used to estimate CH_4 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 CH_4 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 to 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 C_2H_2 (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 CH_4 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 CH_4 concentration measurements to estimate the bulk net landfill CH_4 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 CH_4 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 CH_4 emissions from a landfill by measuring downwind CH_4 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 CH_4 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 CH_4 concentrations and produce CH_4 emission estimates from a landfill over a period of 2 months. We present bulk net CH_4 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 CH_4 concentration measurements: the

Please cite this article in press as: Riddick, S.N., et al. Development of a low-maintenance measurement approach to continuously estimate methane emissions: A case study. Waste Management (2016), http://dx.doi.org/10.1016/j.wasman.2016.12.006