DTU Library

Determination of gas recovery efficiency at two Danish landfills by performing downwind methane measurements and stable carbon isotopic analysis

Fathi Aghdam, Ehsan; Fredenslund, Anders Michael; Chanton, Jeffrey; Kjeldsen, Peter; Scheutz, Charlotte

Determination of gas recovery efficiency at two Danish landfills by performing downwind methane measurements and stable carbon isotopic analysis

Ehsan F. Aghdam [a*], Anders M. Fredenslund [a], Jeffrey Chanton [b], Peter Kjeldsen [a], Charlotte Scheutz [a]

[a] Department of Environmental Engineering, Technical University of Denmark, 2800 Kgs. Lyngby, Denmark

[b] Department of Earth, Ocean and Atmospheric Science, 117 N. Woodward Avenue, Florida State University, Tallahassee, FL 32306-4320, USA

* Corresponding author: ehag@env.dtu.dk, Phone number: +45 4525 1498

Abstract

In this study, the total methane (CH_4) generation rate and gas recovery efficiency at two Danish landfills were determined by field measurements. The landfills are located close to each other and are connected to the same gas collection system. The tracer gas dispersion method was used for quantification of CH_4 emissions from the landfills, while the CH_4 oxidation efficiency in the landfill cover layers was determined by stable carbon isotopic technique. The total CH_4 generation rate was estimated by a first-order decay model (Afvalzorg) and was compared with the total CH_4 generation rate determined by field measurements. CH_4 emissions from the two landfills combined ranged from 29.1 to 49.6 kg CH_4/h. The CH_4 oxidation efficiency was 6-37%, with an average of 18% corresponding to an average CH_4 oxidation rate of 8.1 kg CH_4/h. The calculated gas recovery efficiency was 59-76%, indicating a high potential for optimization of the gas collection system. Higher gas recovery efficiencies (73-76%) were observed after the commencement of gas extraction from a new section of one of the landfills. A good agreement was observed between the average total CH_4 generation rates determined by field measurements (147 kg CH_4/h) and those estimated by the Afvalzorg model (154 kg CH_4/h).

Keywords:

Methane emission, methane oxidation, tracer gas dispersion method, landfill gas generation modeling

1. Introduction

The disposal of waste containing biodegradable fractions in landfills results in the generation of landfill gas (LFG) consisting of methane (CH_4: 55-60% v/v) and carbon dioxide (CO_2: 40-45% v/v). CH_4 is a potent greenhouse gas, 28 times more powerful than CO_2 in terms of global warming potential over a time period of 100 years (IPCC, 2013). In Denmark, the landfilling of organic waste has been banned since 1997. However, many landfills still continue to generate CH_4.

According to the European Pollutants Release and Transfer Registers (E-PRTR), landfills—excluding landfills of inert waste—receiving more than 10 tons of waste per day or with a total disposal capacity above 25,000 tons are required to report their CH_4 emissions (CEC, 2006). In many countries, including Denmark, CH_4 emissions are reported by modeling the CH_4 generation. These models are based on first-order decay (FOD) of organic matter, as shown by Equation 1:

$$m_t = m_0 \times e^{-kt} \tag{Eq. 1}$$

where m_t is the mass of organic carbon (g) after time t, m_0 is the mass of organic carbon (g) at t = 0; t is the degradation time (yr) and k is the FOD kinetic constant (yr^{-1}). These models often use waste quantities and compositions as their input. However, the uncertainty of these models can be significant, due to poor information about the waste composition and amounts of deposited waste, or changes in the landfilled waste due to implementation of new waste management strategies and technologies (Scharff and Jacobs, 2006). Therefore, field measurements are critical in order to achieve precise CH_4 emission rates from landfills.

CH_4 produced in landfills can be recovered and used as a renewable energy source to produce heat and electricity, which may reduce greenhouse gas emissions and can generate revenue (Spokas et al., 2006). Gas recovery is achieved through the construction of horizontal or vertical gas extraction wells. At landfills where the gas quality is too low to be utilized, the CH_4 is sometimes

flared or biocovers for oxidizing CH$_4$ are designed in order to mitigate CH$_4$ emissions from landfills. Determination of the gas recovery efficiency (GRE) is important in order to assess the potential for optimization of the gas collection system, aiming for higher CH$_4$ recovery and lower CH$_4$ emissions. Relatively few studies reported in the literature have determined the GRE from landfills using field measurements (Börjesson et al., 2009; Lohila et al., 2007; Mosher et al., 1999; Spokas et al., 2006). The GRE can be calculated by the ratio of the CH$_4$ recovery rate (kg/h) to the CH$_4$ generation rate (kg/h) as follows (Eq. 2):

$$GRE\ (\%) = \left(\frac{CH_4\ recovered}{CH_4\ generated} \right) \times 100 \qquad \text{(Eq. 2)}$$

$CH_4\ generated$ can be calculated as the sum of CH$_4$ recovered, CH$_4$ emitted to the atmosphere, CH$_4$ oxidized, CH$_4$ migrated laterally, and CH$_4$ internally stored in the landfill as follows (Bogner and Spokas, 1993):

$$CH_4\ generated = CH_4\ emitted + CH_4\ oxidized + CH_4\ recovered + CH_4\ migrated +$$
$$\Delta CH_4\ storage \qquad \text{(Eq. 3)}$$

In spite of gas flaring, utilization or oxidation systems, a portion of the generated CH$_4$ is emitted to the atmosphere from slopes, the leachate collection system, cracks in the landfill cover, or by diffusion through the cover. Different methods have been developed for the quantification of CH$_4$ emissions from landfills (Scheutz et al., 2009). A traditional method is using dynamic and static surface flux chambers (Barlaz et al., 2004; Christophersen et al., 2001; Kjeldsen and Fischer, 1995; Scheutz et al., 2011b). However, using field chambers is a laborious and expensive method, and it is more suitable for quantifying emissions from small-scale landfills, smaller sections of landfills or sources with homogenous emissions (Börjesson et al., 2000; Galle et al., 2001).

Other landfill CH_4 emission measuring methods include vertical radial plume mapping (Goldsmith et al., 2012; Thoma et al., 2010), micrometeorological measurements (Lohila et al., 2007; McBain et al., 2005), inverse dispersion modeling (Babilotte et al., 2010; Figueroa et al., 2009), and the tracer gas dispersion method. Some studies have compared different measurement methods and indicated the advantage and disadvantages of each method (Babilotte et al., 2010; Tregoures et al., 1999).

The tracer gas dispersion method is one of the most promising methods, which has been tested for quantification of CH_4 emissions from landfills (Börjesson et al., 2009; Czepiel et al., 2003; Mønster et al., 2015, 2014; Scheutz et al., 2011a). The tracer gas dispersion method is based on the assumption that the emitted CH_4 and tracer gas will disperse in the same way in the atmosphere. The CH_4 emissions from the landfill are quantified by the simultaneous measurement of CH_4 and tracer concentrations in transects across the CH_4 and tracer gas plume, downwind from the landfill.

A fraction of CH_4 is microbially oxidized to CO_2 by methanotrophs when passing through the landfill cover. According to the Intergovernmental Panel on Climate Change (IPCC) and the US Environmental Protection Agency (USEPA), the default value for CH_4 oxidation in the landfill cover is considered to be 0-10% of the CH_4 generated (IPCC, 2006; USEPA, 2004).

However, this value is challenged by some studies (Börjesson et al., 2007; Chanton et al., 2011, 2009). For instance, Chanton et al. (2009) reported a mean value of $35 \pm 6\%$ by reviewing literature results from 42 determinations of the fraction of oxidized CH_4 by laboratory and field measurements. Börjesson et al. (2007) obtained fractions of oxidized CH_4 between 6.0-24.8% for four active landfills and 36.7-42.8% for two closed landfills. The USEPA recently revised the default value for CH_4 oxidation in the landfill cover to a graduated oxidation efficiency depending

upon the loading of CH$_4$ to the cover (USEPA, 2013). According to the revised USEPA guidelines at low CH$_4$ flux (< 10 g CH$_4$/(m^2.d)) the landfill soil cover can oxidize up to 35% of the flux, while at higher fluxes (> 70 g CH$_4$/(m^2.d)) the oxidation efficiency is recommended to be 10%.

Site-specific determination of oxidized CH$_4$ is important in order to determine a reliable GRE. Stable carbon isotopic analysis can be used to quantify the CH$_4$ oxidation efficiency in landfill settings. The isotopic analysis is based on the preference of methanotrophs to oxidize ^{12}C instead of ^{13}C. Therefore, CH$_4$ becomes ^{13}C enriched after passing through the landfill cover (Börjesson et al., 2007, 2001; Chanton and Liptay, 2000; Chanton et al., 1999).

A fraction of the generated CH$_4$ can migrate laterally off-site (Christophersen et al., 2001). Lateral migration of CH$_4$ depends on many factors, including the type of top cover, weather events (changes in barometric pressure, etc.), and the geology of the landfill surroundings (Scheutz et al., 2009). Nevertheless, using geomembranes or geosynthetic clay bottom liners can minimize the lateral migration of CH$_4$ (Spokas et al., 2006).

Landfills can store a portion of the generated CH$_4$ ($\Delta CH_4\ storage$ in Eq. 3). The amount of the stored CH$_4$ within a landfill can be affected by changes in the barometric pressure and the moisture content of the cover (Scheutz et al., 2009). Apart from changes in barometric pressure and precipitation, many other factors can impact CH$_4$ storage within a landfill, including changes in leachate levels, changes in the amount of CH$_4$ dissolved in leachate, and changes in GRE (Spokas et al., 2006).

The overall objective of this study was to determine the recovery efficiency of the gas collection system installed at two Danish landfills. The objective was met by establishing a CH$_4$ mass balance for the two sites, determining the CH$_4$ generation, emission and oxidation. The CH$_4$

generation was modeled while the CH_4 emission and oxidation was determined by performing field measurements. The modeled CH_4 generation was compared to the results of the field measurements.

2. Materials and methods

2.1. Landfill sites description

The Odense Nord (55°27'16.15"N, 10°25'3.82"E) and Stige Ø (55°26'54.90"N, 10°25'39.41"E) landfills are located in Odense, Denmark. The Stige Ø landfill received several types of waste, including municipal solid waste generated in Odense, from 1967 to 1994. After 1994, the landfill received only soil (1.3 million tons) until its closure in 2005. The landfill contains around 7 million tons of waste and soil, and covers an area of 56 hectares. The Stige Ø landfill is covered by 1 m of soil and was converted into a recreational center after its closure.

The Odense Nord landfill received waste starting in 1994, and it is still in operation. The Odense Nord landfill receives different types of waste, including mixed waste, shredder waste, mineral waste, contaminated soil, garden waste and sludge. Until the end of 2015, around 3 million tons of waste and soil had been disposed in the landfill. In the northern part of the site, co-composting of park and garden waste, sewage sludge, and straw is carried out. The shredder waste is disposed in individual cells covering an area of 6.5 hectares and divided into two sections. Both sections are filled to their maximum capacity; however, the sections have not been finally covered, due to potential landfill mining of shredder waste in the near future. The cell with mixed waste covers an area of 12.6 hectares and is divided into three sections. Two sections are finally covered by around 10 m of soil, and one section is still in operation. The cells containing shredder waste and mixed waste are lined with a composite bottom liner (1 mm HDPE membrane on top of a 30 cm clay layer). A map of the Odense Nord (with its different sections) and Stige Ø landfills is presented in Fig. 1.
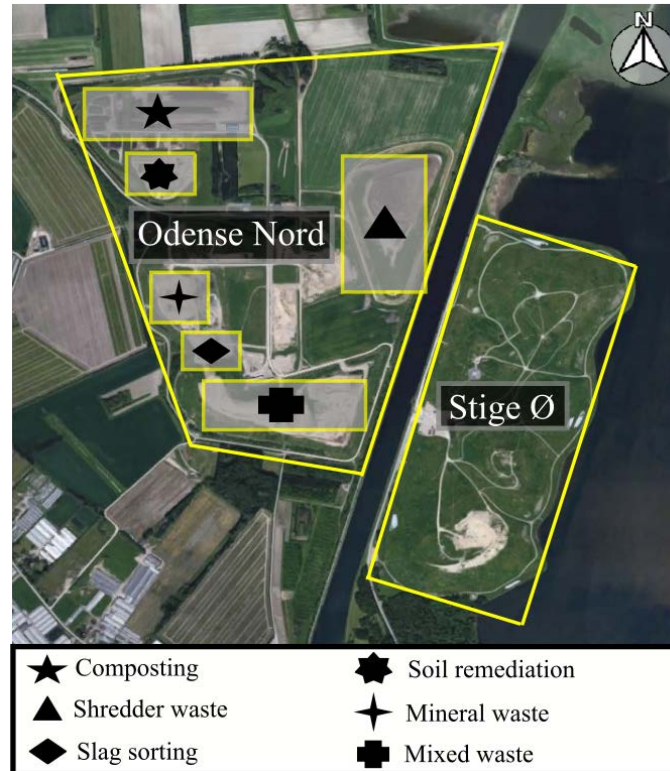
Fig. 1. Map of the Odense Nord (left) and Stige Ø (right) landfills (Imagery ©2016 Google, Aerodata International Surveys, Digital Globe, Map data ©2016 Google).

2.2. CH_4 recovery measurements

Gas recovery facilities are installed in the Stige Ø landfill, and in the cells with mixed waste and shredder waste in the Odense Nord landfill. There are 160 vertical gas wells in the Stige Ø landfill, which are connected to four measuring, pump, and regulation modules (MPR-module). These four MPR-modules are connected to two compressors. In the Odense Nord landfill, there are 19 vertical and 10 horizontal gas wells in the cell containing mixed waste, which are connected to a MPR-module and a compressor. In the first section of the shredder waste cell there are 13 vertical gas wells, which are connected to a MPR-module and a compressor. Moreover, in May 2016, 14 vertical gas wells were constructed in the second section of shredder waste cell, which were connected to the same MPR-module and compressor of the first section of shredder waste. In total

these six modules are connected to a local power plant where a gas engine produces electricity and heat.

The LFG flow (m³ LFG/h) at normal temperature and pressure (NTP; T=293.15 K, P=1 atm) and the CH₄ content of the LFG (v/v %) entering the gas engine was recorded every two minutes. The recovered gas flow entering the gas engine (in m³ LFG/h) was converted to m³ CH₄/h by multiplying the LFG volume by the recorded CH₄ v/v% entering the gas engine. Finally, the CH₄ volumes were converted to CH₄ mass by multiplying it by the density of CH₄ (0.668 kg/m³ at NTP). An average CH₄ recovery rate entering the gas engine, showing a sum of the total CH₄ recovery rate from both of the landfills, was calculated for the time interval when emission measurements were performed.

2.3. CH₄ emission measurements

The tracer gas dispersion method was used for quantifying total CH₄ emissions from the two landfills. CH₄ emissions can be quantified by performing several traverses downwind perpendicular to the plume from the landfill while measuring the atmospheric concentration of CH₄ and tracer gas. The CH₄ emission rate (E_{CH_4}) can be calculated as a function of the ratio of the integrated cross-plume concentration of the CH₄ emitted to the integrated cross-plume concentration of the tracer gas, according to the following equation:

$$E_{CH_4} = Q_{tracer} \times \frac{\int_{plume\ start}^{plume\ end}(C_{CH_4} - C_{CH_4 background})\,dx}{\int_{plume\ start}^{plume\ end}(C_{tracer} - C_{tracer\ background})\,dx} \times \frac{MW_{CH_4}}{MW_{tracer}} \qquad \text{(Eq. 4)}$$

where Q_{tracer} is the release rate of the tracer gas (kg/h), C_{CH_4} and C_{tracer} are the concentrations of CH₄ and the tracer downwind (ppbv), $C_{CH_4\ background}$ and $C_{tracer\ background}$ are the background concentrations of CH₄ and the tracer gas (ppbv), x is the distance across the plume, and MW_{CH_4} and

MW_{tracer} are the molar weight of CH4 and the tracer gas. Further details of the method can be found in Mønster et al. (2014).

Prior to each measurement campaign, a screening of the landfills was performed by driving on all roads onsite and in the vicinity of the landfill in order to identify major emission areas. Tracer gas bottles with acetylene were placed in these areas to simulate the occurring CH4 emission. The release rate of acetylene (Q_{tracer}) was known (typically around 0.6 kg/h from each bottle) and controlled with calibrated flow meters (Sho-rate, Brooks Instrument, Holland). Gas concentrations were measured by a C2H2/CH4/H2O analyzer (G2203, S/N DFADS2005, Picarro, Inc., Santa Clara, CA) based on Cavity Ring-Down Spectroscopy (CRDS). Background concentrations of CH4 and tracer gas were measured upwind from the source area.

The required distance from the source of the emissions to the plume integration transects depends on the size of the emission area, the atmospheric turbulence and how well the tracer and CH4 distribution match (Mønster et al., 2014). In this work, the distance from the source of the emission to the plume integration transects were between 1000 m to 2900 m. Driving speed was usually 30-40 km/h.

Meteorological parameters (wind speed, wind direction, temperature, and barometric pressure) were recorded by a weather station (All-In-One weather sensor, model 102780, Climatronics, USA), which was mounted on top of the vehicle. Table 1 provides the date and time intervals of the measurements, wind direction, average wind speed, temperature, and barometric pressure for the time interval when emission measurements were performed, plus changes in barometric pressure during a period of 6 hours before each measurement, the number of tracer gas release points, and the number of accepted performed traverses.

Table 1. Overview of whole-site CH₄ emission quantification campaigns.

Date	Measuring time interval	Wind direction	Avg. wind speed (m/s)	Avg. temperature (°C)	Avg. barometric pressure (mbar)	Change in barometric pressure during the 6 hours before measurement (mbar/h)	Tracer release points	Number of plume traverses
January 14, 2016	16:30-22:00	E	5 ± 0.6	1.3 ± 0.2	1003 ± 0.3	-0.03	4	11
March 15, 2016	15:30-20:30	SE	3 ± 0.9	4.4 ± 1.4	1032 ± 1.1	+0.25	4	10
September 8, 2016	13:00-19:00	ESE	5 ± 0.8	24.0 ± 0.9	1009 ± 1.2	-0.33	4	9
October 6, 2016	12:00-17:00	NE	7 ± 0.5	11.3 ± 0.4	1027 ± 0.7	-0.20	4	12
October 7, 2016	08:30-13:00	ENE	6 ± 0.8	10.8 ± 0.1	1024 ± 0.5	-0.10	4	15
October 21, 2016	11:30-15:00	ENE	7 ± 0.4	8.0 ± 0.2	1015 ± 0.1	-0.03	4	11

2.4. CH₄ oxidation measurements

Stable carbon isotopic analysis was used to quantify CH₄ oxidation in the top layer of the landfills. The isotopic analysis is based on the preference of methanotrophs to oxidize ^{12}C at a slightly faster rate than ^{13}C. Thus, CH₄ becomes ^{13}C-enriched after the gas has passed through the cover and been exposed to CH₄ oxidation bacteria. Carbon isotopic composition, expressed as δ, is defined as follows:

$$\delta^{13}C = \left(\frac{R_{sample}}{R_{Standard}} - 1\right) \times 1000 \tag{Eq. 5}$$

where R_{sample} is the $^{13}C/^{12}C$ ratio of the sample and $R_{Standard}$ is the $^{13}C/^{12}C$ of a standard marine carbonate (PDB, 0.01124). The fraction of oxidized CH₄ (f_{ox}) can be calculated by (Börjesson et al., 2007; Chanton et al., 1999):

$$f_{ox} = \frac{\delta_{excess} - \delta_A}{1000 \ (\alpha_{ox} - \alpha_{trans})} \tag{Eq. 6}$$

where δ_{excess} is the excess of $\delta^{13}C$ in the plume corrected for background or upwind samples, δ_A is the carbon isotopic content of anoxic CH₄, α_{ox} is the fractionation factor associated with CH₄ oxidation by landfill cover and α_{trans} is the transport fractionation factor. The excess of $\delta^{13}C$ in the plume was calculated as:

$$[\delta \ CH_4]_{excess} = \frac{[CH_4]_{meas}.[\delta CH_4]_{meas} - [CH_4]_{amb}.[\delta CH_4]_{amb}}{[CH_4]_{meas} - [CH_4]_{amb}}$$ (Eq. 7)

where $[CH_4]_{meas}$ and $[\delta CH_4]_{meas}$ represent concentration and δ^{13}C values of CH$_4$ in the downwind plume and $[CH_4]_{amb}$ and $[\delta CH_4]_{amb}$ represent the concentration and δ^{13}C values of background air measured upwind of the landfills.

It was assumed that transport of CH$_4$ up through the soil was dominated by advection, which does not cause isotopic fractionation (Börjesson et al., 2007), thus the term α_{trans} was assumed to be 1. However, if diffusion was important, then $\alpha_{trans} > 1$. The effect of this assumption is that using the isotopic approach can lead to an underestimation of the CH$_4$ oxidation and thus the obtained results can be considered conservative, providing the minimum CH$_4$ oxidation (Chanton et al., 2009). The parameter α_{ox} was determined by the incubation of soil cover samples.

Gas samples for the determination of the carbon isotopic content of anoxic zones (δ_A) were taken from the four MPR-modules at Stige Ø and one MPR-module at the shredder waste cell of Odense Nord. The downwind samples were taken by two methods: samples were obtained in the downwind plume, and from surface air across the landfills. The plumes of CH$_4$ were located by the CRDS instrument. In addition, locations with elevated CH$_4$ concentrations across the landfill were identified by a hand-held analyzer (TVA-1000B, Thermo Environmental Instruments) equipped with a flame ionization detector (FID), where surface air samples were taken. Samples from the downwind plume (2-6 samples), and background air (2-3 samples) were taken in 120 mL evacuated glass serum vials, while samples from the surface air (4-5 samples) and anoxic zone (1-4 samples) were taken in 30 mL evacuated glass vials. The bottles were sealed with 1 cm thick butyl rubber stoppers and aluminum crimp-seals. Analysis of ^{13}C-content of CH$_4$ was performed at Florida State University, USA using the analytical set-up as describe in Chanton et al. (1999). Samples for determination of the CH$_4$ oxidation were collected on May 12, 2016 and February 10, 2017.

The rate of CH$_4$ oxidation, MO (kg/h) was calculated from the total CH$_4$ emission rate and fraction of oxidized CH$_4$ using the following equation (Scheutz et al., 2011a):

$$MO = f_{ox} \left(\frac{E}{1 - f_{ox}}\right)$$ (Eq. 8)

where f_{ox} is the fraction of oxidized CH$_4$ (%), and E is the total CH$_4$ emission rate (kg/h).

In order to determine the fractionation factor due to oxidation, α_{ox}, incubations of cover soil samples were performed. Three soil samples were taken from a 10-15 cm depth, including one sample from the shredder waste cell of the Odense Nord landfill, and two samples from the Stige Ø landfill: one from the northern part and one from the southern part. The sampling depth of 10-15 was chosen as CH$_4$ oxidation was expected at this depth interval due to overlapping gradients of CH$_4$ and oxygen (O$_2$) in the cover layer (Scheutz et al., 2009). After collection, the samples were transported to the laboratory and kept at 10 °C. In the laboratory, the samples were sieved (mesh size 4 mm) and 200 g of each sieved sample were placed in a 1.1 L glass incubation bottle, which were sealed with a rubber septum. Soil samples were incubated at room temperature (22 ℃).

CH$_4$ (100 mL) was added to each incubation bottle at time zero, resulting in an initial concentration of 10-11% CH$_4$. Immediately after the addition of CH$_4$, gas samples of 0.3 mL were withdrawn and analyzed for CH$_4$ using a trace gas chromatograph (TRACE 1310 GC, Q PLOT, 0.32 mm, 8 m, Thermo Scientific) equipped with a FID. Next, 8 mL gas samples were taken and stored in pre-evacuated glass vials for later isotopic analysis. Gas sampling was continued until the CH$_4$ concentration was lower than 1%. In total, six gas samples were taken from each incubation bottle. Analysis of ^{13}C-content of CH$_4$ was performed at Florida State University, USA.

The fractionation factors (α_{ox}) were calculated following the approach derived by Coleman et al. (1981) using their equation, as follows:

$$\delta^{13}C_t \cong 1000 \left(\frac{1}{\alpha} - 1\right) \ln\left(\frac{C}{C_0}\right) + \delta^{13}C_{t=0} \qquad \text{(Eq. 9)}$$

where $\delta^{13}C_t$ is the $\delta^{13}C$ value of the CH$_4$ remaining at time t, $\frac{C}{C_0}$ is the fraction of CH$_4$ at time t and $\delta^{13}C_{t=0}$ is the $\delta^{13}C$ value of CH$_4$ at the initial time. When $\delta^{13}C$ is plotted versus $\ln(\frac{C}{C_0})$, the slope of the line fitted to the data is $1000\left(\frac{1}{\alpha} - 1\right)$. The locations of the collected upwind and downwind gas samples, surface air samples, and soil samples are shown in the Fig. A1 in the Supplementary Material (SM).

2.5. CH$_4$ migration and changes in CH$_4$ storage

In the Odense Nord landfill the bottom liner consists of a HDPE membrane placed on top of a clay layer. The CH$_4$ flux through a HDPE liner with thickness of 1.5 mm has been estimated at 0.58 cm^3 CH$_4$/(m^2·d), corresponding to 4.2×10^{-7} kg CH$_4$/(m^2·d), at 1 atm (Pauly, 1989; Lim, 1995). The CH$_4$ migration through the bottom membrane of the Odense Nord landfill was calculated by multiplying this value by the area of the shredder waste and mixed waste cells of the Odense Nord landfill (19.1 hectares), assuming no compromise in the integrity of the membrane. This provides a conservative estimation of lateral CH$_4$ migration at the Odense Nord landfill, as the diffusional resistance of the clay layer was not considered. At the Stige Ø landfill, a drainage trench running along the edge of the landfill collects leachate and surface water. The drainage trench is unsaturated, as leachate is not allowed to accumulate and is continuously pumped away. CH$_4$ generated in the waste body is emitted to the atmosphere from the drainage trench and is thus accounted for in CH$_4$ emission measurements. Therefore, lateral gas migration from Stige Ø was assumed to be zero.

In theory, $\Delta CH_4 \, storage$ could be determined by shutting down the gas collection system for a period of time (e.g. one week) and monitoring the CH$_4$ concentration and LFG flow entering the

gas engine before and after the shutdown of the gas collection system. The amount of additional CH_4 collected after shutdown in comparison to prior shutdown is an approximation of $\Delta CH_4\ storage$. As shutting down the gas collection system could pose a risk to the health of citizens using the recreational center on the Stige Ø landfill, it was not possible to determine $\Delta CH_4\ storage$ in this study.

2.6. Gas generation modeling

The Afvalzorg model was used to estimate the total CH_4 generation from the two landfills, as this model was found to be the most suitable in a previous study comparing the three models LandGEM, IPCC, and Afvalzorg with measured CH_4 emissions (Mou et al., 2015a). The Afvalzorg model, which was developed by a Dutch waste management company, is a multi-phase model that calculates the CH_4 generation using a FOD model. This model operates with three degrees of degradability (fast, moderate and slow) and accommodates up to eight waste categories.

The annual deposited waste amounts were provided by the landfill operators. The amount of waste deposited at Stige Ø landfill during 1979, 1980, 1990 and 1994 was not available. For these years, the amount of deposited waste was assumed to be the same as their previous years. The amount of deposited waste was then fitted into the eight waste categories of Afvalzorg model, which can be found in Table A1 and Table A2 of the SM.

The total CH_4 generation was estimated by using the default values provided in the Afvalzorg model as well as by a revised version of the model using site-specific values for the degradable organic carbon contents (DOCC) and for the k-values of shredder waste, sludge and bulky mixed waste. The biochemical methane potential (BMP) and k-values of shredder waste, sludge, and bulky mixed waste for the Odense Nord landfill were determined in previous studies (Mou et al., 2015b,

2014). The BMPs of the above-mentioned fractions were used to calculate the DOCC using the following equation (IPCC, 2006):

$$BMP = DOCC \times F \times 16/12 \qquad (Eq. 10)$$

where F is the volume fraction of CH_4 and 16 and 12 are the molar masses of CH_4 and carbon, respectively. F was considered to be 50%. For waste categories other than shredder waste, sludge and bulky mixed waste, the default k-values and DOCCs provided by the Afvalzorg model were used in the revised model. The default and revised DOCCs and k-values can be found in Table A3 and Table A4 of the SM.

3. Results and discussion

3.1. CH_4 surface screening of the landfills

Fig. 2 shows the CH_4 screening results of the Odense Nord (2a) and Stige Ø (2b) landfills on March 15, 2016. The screening results for the other measurement campaigns can be found in the SM (Fig. A2-Fig. A5). At the Odense Nord landfill, elevated CH_4 concentrations were observed at the mixed waste cell and shredder waste cell. In addition, high CH_4 concentrations were seen at the composting facility (Fig. 2a). At the Stige Ø landfill, the majority of elevated CH_4 concentrations were observed next to the drainage trench around the landfill, especially in the northern section of the landfill (Fig. 2b). In order to quantify and potentially separate the CH_4 emissions from individual areas, four tracer gas bottles were placed at the identified main emission areas.
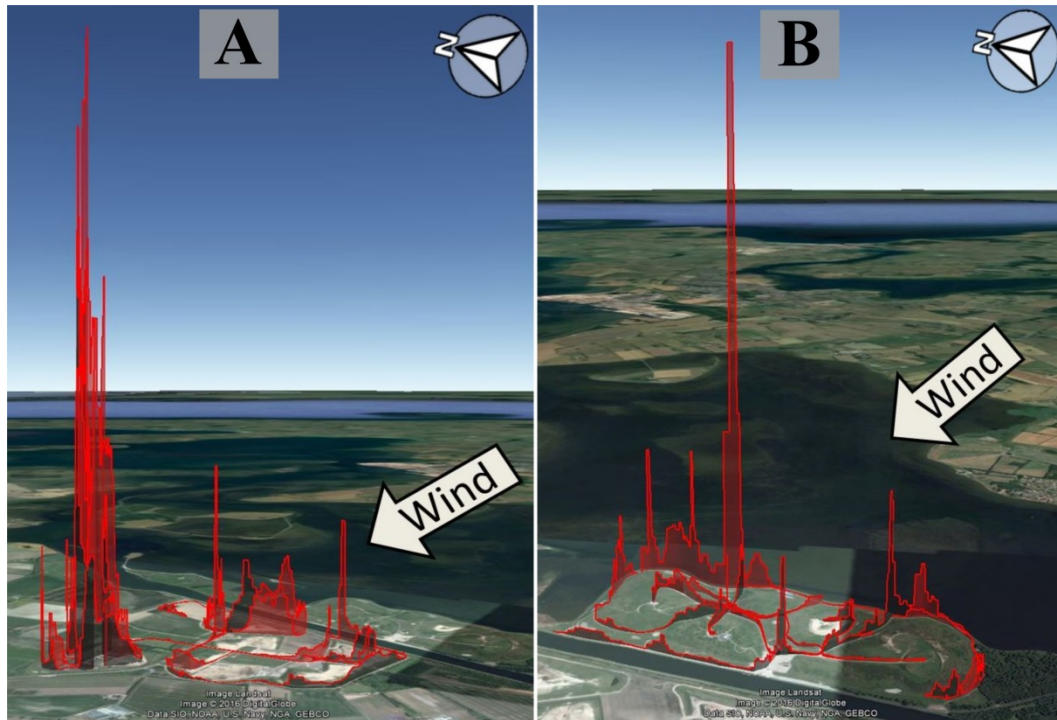
Fig. 2. Screening results of the Odense Nord (A) and Stige Ø (B) landfills on March 15, 2016. The red lines show the atmospheric CH_4 concentrations measured at 2 m height above the landfill surface. The background CH_4 concentration (1.9481 ppm) was subtracted. The highest CH_4 concentration measured was 13.2229 ppm above background (Imagery ©2016 Google, Aerodata International Surveys).

3.2. CH4 emissions measurements

Fig. 3 shows CH_4 and tracer gas plumes measured downwind of the landfills, and Table 2 provides an overview of the measured emissions from the landfills, the composting facility and distinguished sections of the landfills, when possible. Overall, six measurement campaigns were performed from January to October 2016. Based on the screening results, four tracer bottles—one in the northern part of the Stige Ø landfill and three in the Odense Nord landfill—were placed, which are marked with orange triangles in Fig. 3.
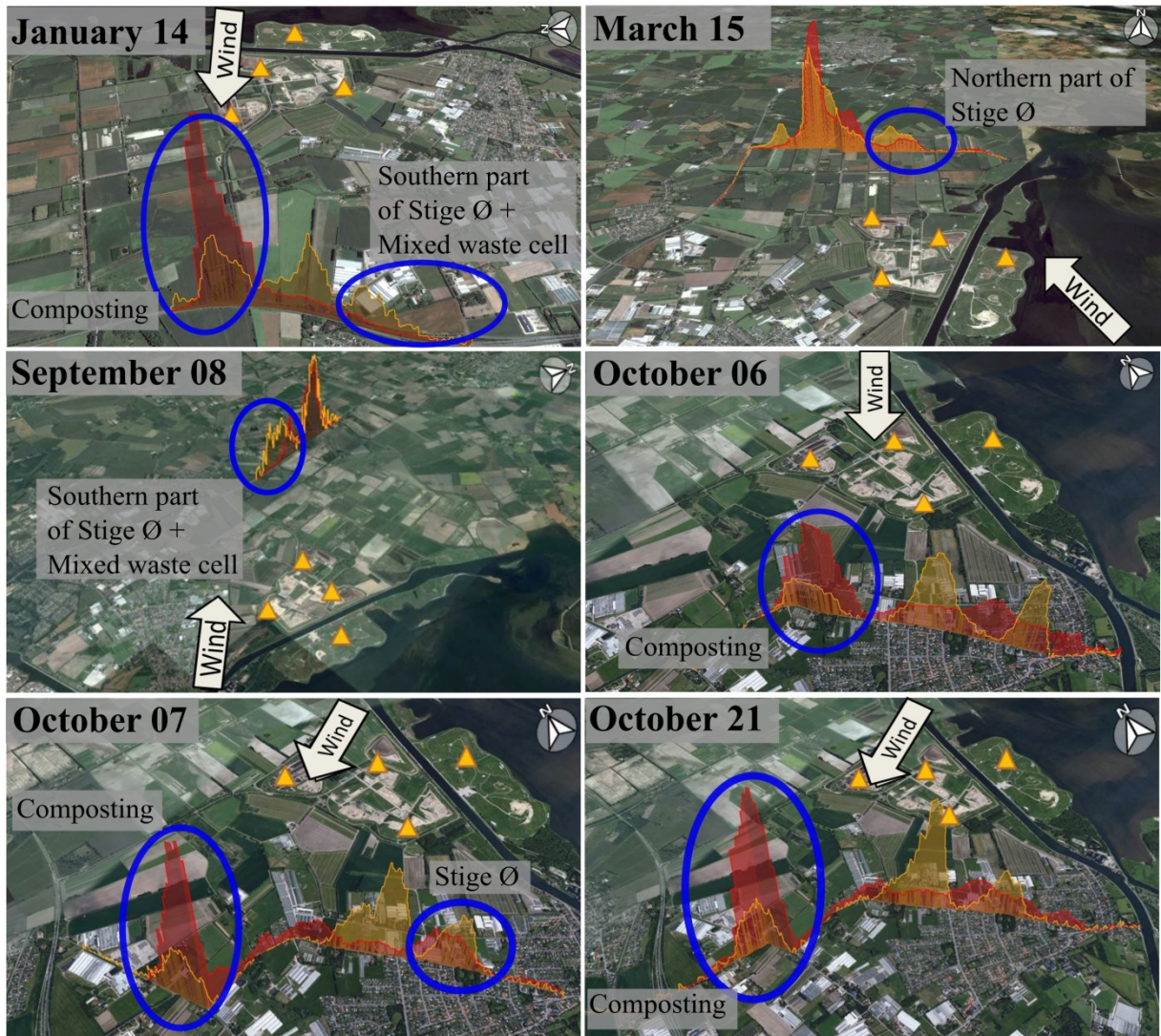
Fig. 3. CH_4 (red) and tracer gas (yellow) plumes measured downwind of the landfills. The yellow triangles indicate the tracer gas release locations. The blue circles show the emissions from the composting facility or distinguished sections of the landfills (Imagery ©2016 Google, Aerodata International Surveys).

The total emission from the two landfills ranged from 29.1 to 49.6 kg CH_4/h, corresponding to 0.93-1.58 g CH_4/(m^2.d) and 0.07-0.12 g CH_4/(ton waste.d). The lower emission rate from the landfills on March 15, 2016 (45.8 kg CH_4/h) in comparison to January 14, 2016 (49.6 kg CH_4/h) may be due to the increasing barometric pressure on March 15. Previous studies have shown that changes in barometric pressure can affect the emissions from landfills (Nastev et al., 2001; Poulsen et al., 2003; Xu et al., 2014; Young, 1992).

Table 2. CH$_4$ emission from the landfills and composting facility. Digits given in parentheses represent standard deviation.

Date	Emissions from both landfills + composting facility (kg CH$_4$/h)	Emissions from the composting facility (kg CH$_4$/h)	Emissions from both landfills (kg CH$_4$/h)	Emissions from the distinguished section of the landfills (kg CH$_4$/h)
January 14, 2016	68.5 (4.1)	18.9 (2.4)	49.6 (4.1)	Southern part of Stige Ø + Mixed waste cell of Odense Nord = 5.4 (1.1)
March 15, 2016	64.7 (9.1)	n.d.[a]	45.8[b]	Northern part of Stige Ø = 17.1 (1.2)
September 8, 2016	52.7 (4.3)	n.d.[a]	29.1[c]	Southern part of Stige Ø + Mixed waste cell of Odense Nord = 4.3 (0.7)
October 6, 2016	54.9 (7.0)	23.6 (3.8)	31.2 (4.1)	-
October 07, 2016	58.0 (5.6)	24.5 (5.1)	33.5 (3.8)	Stige Ø=12.7 (0.85)
October 21, 2016	61.0 (4.2)	28.0 (2.9)	33.0 (2.3)	-

[a]: Not distinguishable

[b]: As the emission from the composting facility could not be distinguished from the landfills, the composting emission rate of January 14 was subtracted from "Emissions from both landfills + composting facility" to calculate the emission from the landfills.

[c]: As the emission from the composting facility could not be distinguished from the landfills, the composting emission rate of October 6 was subtracted from "Emissions from both landfills + composting facility" to calculate the emission from the landfills.

Emission rates of 29.1, 31.2, 33.5, and 33.0 kg CH$_4$/h from the landfills on September 8, October 6, 7 and 21, respectively, were lower than emission rates of 49.6 and 45.8 kg CH$_4$/h on January 14 and March 15, respectively. This was most likely due to the commencement of gas extraction from the second section of the shredder waste cell in May 2016. Low CH$_4$ emissions were observed from the southern part of the Stige Ø landfill and the mixed waste cell of the Odense Nord landfill (5.4 and 4.3 kg CH$_4$/h on January 14 and September 8, respectively). This indicated that the majority of emissions from the two landfills came from the shredder waste cell of the Odense Nord landfill and the northern part of the Stige Ø landfill.

The measured emission of 18.9 kg CH$_4$/h from the composting facility on January 14, 2016 was comparable to the measured emission of 16.8 kg CH$_4$/h from the same composting facility on January 2012 (Mønster et al., 2015). The measured emission from the composting facility on October 6 (23.6 kg CH$_4$/h) was higher than the emission on January 14 (18.9 kg CH$_4$/h). This is

most likely due to higher amount of garden waste in October in comparison to January, and thus more composting activity in the facility.

The measured emissions in this study (29.1-49.6 kg CH_4/h) were higher than most of the 14 Danish landfills measured by Scheutz et al. (2011b, 2011c) and Mønster et al. (2015) using the tracer gas dispersion method, which most likely is due to higher waste amounts in Stige Ø and Odense Nord landfills (10.03×10^6 tons) in comparison to the other studied Danish landfills (0.23-2.20×10^6 tons). However, the measured emissions were significantly lower than the measured emissions of 167-1293 kg CH_4/h (L/min was converted to kg/h by using the density of CH_4 at STP (0.716 g/L)) from U.S. landfills measured by Mosher et al. (1999) using the tracer gas dispersion method. Moreover, the measured emissions per area of the landfill and per disposed waste mass in this study (0.93-1.58 g CH_4/(m^2.d), and 0.07-0.12 g CH_4/(ton waste.d) were significantly lower than the measured emissions of 23-130 g CH_4/(m^2.d), and 1.54-8.88 g CH_4/(ton waste.d) from the U.S. landfills measured by Mosher et al. (1999). The lower emissions from Danish landfills can be explained by the ban on landfilling of waste with high organic contents in 1997 in Denmark. However, apart from waste type, CH_4 generation and emission depends on many other factors, including the existence of a gas collection system or a biocover oxidation system, landfill cover type, and climate conditions.

3.3. CH_4 oxidation measurements

The fractionation factor α_{ox} obtained from soil incubations were 1.025, 1.015, and 1.024 for samples from the shredder waste cell, northern part, and southern part of Stige Ø, respectively. Plots of the $\delta^{13}C$ values versus $\ln(\frac{C}{C_0})$ can be found in the SM (Fig. A6). Table 3 presents a summary of the α_{ox} values used in the literature, with the associated temperatures and determination methods. The α_{ox} values measured for the soil collected at the shredder waste cell and southern part of Stige

Ø (1.025 and 1.024, respectively) were comparable to previous studies performed at 25 °C (1.022-1.023 in Table 3). However, the measured α_{ox} value for the northern part of Stige Ø was lower than values reported in the literature.

Table 3. Overview of reported CH_4 oxidation fractionation factors (α_{ox}) for landfill cover soils in the literature and their associated soil incubation temperatures.

α_{ox}	T (ºC)	Method	Reference
1.022	25	Soil incubation	Chanton et al. (2008)
1.027 and 1.023	4 and 25	Soil incubation	Börjesson et al. (2001)
1.022	25	Soil incubation	Liptay et al. (1998)
1.049 to 1.025	8 to 35	Soil incubation	Chanton and Liptay (2000)
1.023	-	Estimated from literature	Cabral et al. (2010)
1.020	-	Estimated from literature	Rachor et al. (2011)
1.025	22	Soil incubation, shredder waste	This study
1.015	22	Soil incubation, northern Stige Ø	This study
1.024	22	Soil incubation, southern Stige Ø	This study

Table 4 shows the results of the stable carbon isotopic analysis and the calculated CH_4 oxidation efficiencies (f_{ox}). The $\delta^{13}CH_4$ values of downwind samples ranged between -59.94 and -48.45, and were comparable to the downwind $\delta^{13}CH_4$ found by Scheutz et al. (2011a) at a Danish landfill (-51.18 to -47.40), and at a number of Swedish landfills (-52.30 to -46.68) found by Börjesson et al. (2007). The upwind $\delta^{13}CH_4$ values, ranging between -49.43 and -46.68, were also comparable to upwind $\delta^{13}CH_4$ values found in previous studies (Börjesson et al., 2007; Scheutz et al., 2011a). The $\delta^{13}CH_4$ values of the anoxic zone, δ_A (-69.61 to -54.38), were lower than the excess $\delta^{13}CH_4$ values (-62.51 to -52.65), showing that the samples were enriched in ^{13}C after passing through the top layer of the landfills.

Table 4. The carbon isotope ratio of CH_4 in samples from the Odense Nord and Stige Ø landfills and the fraction of oxidized CH_4 during two measurement campaigns. Numbers in parentheses give the standard deviations. The numbers of samples are mentioned in section 2.4.

Landfill	Date	Plume / surface [a]	Downwind CH_4 concentration	Downwind $\delta^{13}CH_4$	Upwind CH_4 concentration	Upwind $\delta^{13}CH_4$	Excess $\delta^{13}CH_4$	Anoxic CH_4; δ_A	f_{ox}
Odense Nord	February 2017	Plume	2.14 (0.07)	-50.20 (0.76)	2.02 (0.01)	-49.43 (0.08)	-61.67 (10.33)	-69.61 (0.17)	0.32 (0.41)
	May 2016	Plume	2.48 (0.14)	-49.97 (0.64)	1.97 (0.02)	-47.99 (0.39)	-57.62 (1.00)	-66.80 (0.09)	0.37 (0.04)
	May 2016	Surface	24.23 (29.63)	-59.94 (6.98)	1.97 (0.02)	-47.99 (0.39)	-62.51 (7.17)	-66.80 (0.09)	0.17 (0.29)
Stige Ø	February 2017	Plume	2.34 (0.20)	-49.66 (0.28)	2.03 (0.03)	-48.90 (0.27)	-55.96 (3.69)	-57.21 (0.40)	0.06 (0.19)
	May 2016	Plume	2.74 (0.33)	-48.55 (0.25)	1.92 (0.01)	-46.68 (1.66)	-53.10 (0.98)	-54.38 (0.60)	0.07 (0.05)
	May 2016	Surface	3.98 (1.96)	-48.45 (3.86)	1.92 (0.01)	-46.68 (1.66)	-52.65 (7.15)	-54.38 (0.60)	0.09 (0.37)

[a]: This column shows whether the downwind samples were taken in the downwind plume or on the surface across the landfill.

The δ_A values at the Odense Nord landfill on both measurement campaigns (-69.61 and -66.80) were lower than the reported δ_A values of -62.33 to -53.81 found in previous studies (Börjesson et al., 2007; Chanton et al., 1999; Scheutz et al., 2011a). As the anoxic zone samples at the Odense Nord landfill were taken from the shredder waste MPR-module, this different isotopic signature could be related to the observed special gas composition—a high CH_4 content and very low or no CO_2—at shredder waste monofills in previous studies (Aghdam et al., 2017; Olsen and Willumsen, 2013; Scheutz et al., 2011b). A previous study (Aghdam et al., 2017) has shown that high CH_4 generation from shredder waste monofills is due to reduction of existing CO_2 in the produced LFG by H_2. The CO_2 reduction pathway produces more ^{13}C depleted CH_4 in comparison to the acetate fermentation pathway (Chanton et al., 2011) and thus a lower $\delta^{13}CH_4$ value.

Overall, the oxidation efficiency (f_{ox}) was higher in the Odense Nord landfill (17-37%) in comparison to the Stige Ø landfill (6-9 %). This could be due to a higher diffusion of atmospheric oxygen into the uncovered waste in the Odense Nord landfill, increasing CH_4 oxidation in comparison to the Stige Ø landfill, which has a 1-10 m compacted soil layer as the top cover. The significant difference in the measured CH_4 oxidation efficiency at the two landfills highlights the

importance of site-specific determination of CH_4 oxidation efficiency, rather than using the default value of 10% recommended by IPCC (2006) and USEPA (2004).

The downwind surface samples taken in May 2016 from the Odense Nord landfill showed a lower oxidation efficiency (17%) in comparison to the oxidation efficiency calculated based on downwind plume samples (32-37%). Moreover, a higher average CH_4 concentration (24.23 ppm) and a lower $\delta^{13}CH_4$ value (-59.94) was observed for surface samples taken in May 2016 from the Odense Nord landfill in comparison to the rest of the samples. This means that the lower oxidation efficiency obtained in surface samples at the Odense Nord landfill in May 2016 could be due to air samples taken from CH_4 emission hot spots, where CH_4 did not have enough retention time in the soil to be oxidized.

The average CH_4 oxidation efficiency was 18% and was used to determine the total CH_4 generation from both landfills. A very wide range of CH_4 oxidation efficiencies (0-94%) has been reported in the literature (Börjesson et al., 2001; Chanton et al., 2011; Scheutz et al., 2011a; Spokas et al., 2006). The average CH_4 oxidation efficiency in this study was comparable to the CH_4 oxidation efficiency of 16-21% determined by Scheutz et al. (2011a) at a Danish landfill before installation of a biocover.

The estimated CH_4 load to the landfill covers for Stige Ø and Odense Nord is 1.4 g $CH_4/(m^2 \cdot d)$, assuming that the CH_4 load is the CH_4 generated minus the CH_4 collected. According to the revised USEPA guidelines the oxidation efficiency at both landfills can thus be assumed to be 35% as the CH_4 flux to the landfill cover is < 10 g $CH_4/(m^2 \cdot d)$. Following USEPA guidelines the oxidation efficiency at Stige Ø will thus be overestimated.

3.4. Total CH_4 generation and gas recovery efficiency

Table 5 shows the CH_4 emission rate (E), CH_4 recovery rate (R), CH_4 oxidation rate (MO), total CH_4 generation rate (E+R+MO) reported in kg CH_4/h, and the obtained GRE (%). The calculated CH_4 migration was 0.003 kg CH_4/h, which was negligible in comparison to the CH_4 emission, oxidation and recovery, and thus was not included in Table 5. A composite bottom liner (a geomembrane on top a clay layer) is a common measure to effectively prevent the migration of CH_4 and the leakage of leachate (Bouazza and Van Impe, 1998) and thus the very low calculated CH_4 migration in this study was found to be reasonable.

Table 5. CH_4 emission, oxidation, recovery, and GRE of the landfills.

Date	E = CH_4 emissions rate (kg/h)	R = CH_4 recovery rate (kg/h)	MO = CH_4 oxidation rate (kg/h)	Total CH_4 production rate (E+R+MO; kg/h)	Gas recovery efficiency (%)
January 14, 2016	49.6	88.1	10.9	148.6	59
March 15, 2016	45.8	84.8	10.1	140.7	60
September 8, 2016	29.1	115.3	6.4	150.8	76
October 6, 2016	31.2	109.2	6.8	147.2	74
October 7, 2016	33.5	108.1	7.4	149.0	73
October 21, 2016	33.0	108.1	7.2	148.3	73
Average	37.0	102.3	8.1	147.4	69
Standard deviation	8.5	12.6	1.9	3.5	7

The lower CH_4 oxidation rates in September and October (6.4-7.4 kg CH_4/h), in comparison to January and March (10.1-10.9 kg CH_4/h), are a direct result of the lower CH_4 emission rate after installation of gas collection in the second section of shredder waste in May 2016 (see Eq. 8). The total CH_4 generation rate ranged between 140.7 and 150.8 kg/h with an average of 147.4 kg/h and a small standard deviation (3.5 kg/h). The CH_4 recovery rate and efficiency was 84.8-115.3 kg/h and 59-76 %, respectively. The higher CH_4 recovery rate and efficiency in September and October in comparison to January and March shows that commencement of gas extraction from the second section of shredder waste cell in May 2016 enhanced gas collection. The obtained GRE indicates that there is a quite high potential for the optimization of gas collection system, especially for Stige

Ø. An optimization of the gas collection system could lead to lower CH_4 emission to the atmosphere and higher CH_4 recovery, which may lower greenhouse gas emissions from the site and generate revenue. If the GRE can be improved by only 5% (7.4 kg CH_4/h higher recovery), this is equal to a reduction of 6.2 kg CH_4/h or 1520 ton CO_2-eq/year emissions to the atmosphere.

GREs reported in the literature range from 14-94% (Börjesson et al., 2009; Lohila et al., 2007; Mosher et al., 1999; Spokas et al., 2006). The GRE depends on many factors, including the design and management of the system and the presence and type of top cover. The very low GRE of 14% measured at a Swedish landfill by Börjesson et al. (2009) was related to management problems, while the 94% GRE was measured at a French landfill with a final clay cover (Spokas et al., 2006). The obtained GRE in this study was comparable to the GRE of 69-79% measured by Lohila et al. (2007) at a Finnish landfill, which had a top cover consisting of a compost soil layer on top of a clay layer. However, it was lower than the 90% measured by Mosher et al. (1999) at a U.S. landfill with a geomembrane cover.

3.5. Modeling and comparison with field measurements

Fig. A7 and Fig. A8 in the SM show the minimum and maximum annual CH_4 generation rate at the Odense Nord and Stige Ø landfills, estimated by the Afvalzorg model using both default values and site-specific values. The modeled total CH_4 generation rate in 2016 for both landfills combined was 136.3-172.6 kg/h when using default values and 86.3-109.3 kg/h when using site-specific values. In general, a good agreement was observed between the modeled average CH_4 generation and the measured average generation (147.4 kg CH_4/h); however, the measured generation agreed best with the modeled generation when default values were used.

Mou et al. (2015a) observed the best agreement between field measurements and modeling results at four Danish landfills when using the Afvalzorg model, in comparison to using the IPCC

and LandGEM models. However, they found that using the default values of the Afvalzorg model resulted in an overestimation of the total CH_4 generation, while they observed a better agreement between the field measurements and the revised Afvalzorg model with calibrated BMPs and k-values. This is in contrast with our results, which could be due to the uncertainty related to waste sampling, and to differences in the factors governing gas generation at different landfills. Moreover, the CH_4 oxidation efficiency was assumed to be 10% in the study by Mou et al. (2015a), which may result in an underestimation of the CH_4 generation.

In a previous study by Börjesson et al. (2009), a good agreement was observed between field measurements (CH_4 emission measurements and determination of the CH_4 oxidation using stable carbon isotopic analysis) and the results of the IPCC model, when the fraction of degradable carbon dissimilated was set to 0.54, which is similar to the dissimilation factor of 0.58 used in our study.

4. Conclusions

The gas recovery efficiency from two Danish landfills was determined by a combination of field measurements, including the quantification of total CH_4 emissions by the tracer gas dispersion method and the quantification of the CH_4 oxidation efficiency in the top layer of the landfills by stable carbon isotopic technique. Moreover, the total CH_4 generation determined by field measurements was compared to the estimated total CH_4 generation using the Afvalzorg LFG generation model. The measured average total CH_4 emission from the two sites combined was 37 kg/h and the average CH_4 oxidation rate was 8.1 kg/h. The average CH_4 recovery rate was 102 kg/h and the gas recovery efficiency was 59-76%, indicating a further potential for gas collection. On-site CH_4 screenings showed areas with significant emissions, which can be used to develop a future plan for optimization of the gas collection system. The average total CH_4 generation rate determined by field measurements (147.4 kg/h) was in good agreement with the average modeled total CH_4 generation using the Afvalzorg model (154.4 kg CH_4/h).

Acknowledgments

Supplementary material

The Supplementary material contains the annual deposited waste amounts in the landfills (Tables A1 and A2), the default and revised DOCCs and k-values used in Afvalzorg model (Tables A3 and A4), locations of the collected gas and soil samples (Fig. A1), surface screening results (Figs. A2-A5), the soil incubation isotopic analysis results (Fig. A6), and the modeled annual CH_4 generation rate curves at the landfills (Figs. A7 and A8).

References

Aghdam, E.F., Scheutz, C., Kjeldsen, P., 2017. Assessment of methane production from shredder waste in landfills : The influence of temperature, moisture and metals. Waste Manag. 63, 226–237. doi:10.1016/j.wasman.2016.11.023

Babilotte, A., Lagier, T., Fiani, E., Taramini, V., 2010. Fugitive Methane Emissions from Landfills: Field Comparison of Five Methods on a French Landfill. J. Environ. Eng. 136, 777–784. doi:10.1061/(ASCE)EE.1943-7870.0000260

Barlaz, M.A., Green, R.B., Chanton, J.P., Goldsmith, C.D., Hater, G.R., 2004. Evaluation of a biologically active cover for mitigation of landfill gas emissions. Environ. Sci. Technol. 38, 4891–4899. doi:10.1021/es049605b

Bogner, J., Spokas, K., 1993. Landfill CH_4: rates, fates, and role in global carbon cycle.

Chemosphere 26, 369–386.

Börjesson, G., Chanton, J., Svensson, B.H., 2001. Methane oxidation in two Swedish landfill covers measured with carbon-13 to carbon-12 isotope ratios. J. Environ. Qual. 30, 369–376. doi:10.2134/jeq2001.302369x

Börjesson, G., Danielsson, A., Svensson, B.H., 2000. Methane fluxes from a Swedish landfill determined by geostatistical treatment of static chamber measurements. Environ. Sci. Technol. 34, 4044–4050. doi:10.1021/es991350s

Börjesson, G., Samuelsson, J., Chanton, J., 2007. Methane oxidation in Swedish landfills quantified with the stable carbon isotope technique in combination with an optical method for emitted methane. Environ. Sci. Technol. 41, 6684–6690. doi:10.1021/es062735v

Börjesson, G., Samuelsson, J., Chanton, J., Adolfsson, R., Galle, B., Svensson, B.H., 2009. A national landfill methane budget for Sweden based on field measurements, and an evaluation of IPCC models. Tellus, Ser. B Chem. Phys. Meteorol. 61 B, 424–435. doi:10.1111/j.1600-0889.2008.00409.x

Bouazza, A., Van Impe, W.F., 1998. Liner design for waste disposal sites. Environ. Geol. 35, 41–54. doi:10.1007/s002540050291

Cabral, A.R., Capanema, M.A., Gebert, J., Moreira, J.F., Jugnia, L.B., 2010. Quantifying microbial methane oxidation efficiencies in two experimental landfill biocovers using stable isotopes. Water. Air. Soil Pollut. 209, 157–172. doi:10.1007/s11270-009-0188-4

CEC (Council of the European Communities), 2006. Regulation of the European Parliament and of the Council of 18 January 2006 Concerning the Establishment of a European Pollutant Release and Transfer Register and amending Council Directives 91/689/EEC and 96/61/EC (No.

166/2006), Brussels, Belgium

Chanton, J., Abichou, T., Langford, C., Hater, G., Green, R., Goldsmith, D., Swan, N., 2011. Landfill methane oxidation across climate types in the U.S. Environ. Sci. Technol. 45, 313–319. doi:10.1021/es101915r

Chanton, J., Liptay, K., 2000. Seasonal variation in methane oxidation in a landfill cover soil as determined by an in situ stable isotope technique. Global Biogeochem. Cycles 14, 51–60. doi:10.1029/1999GB900087

Chanton, J.P., Powelson, D.K., Abichou, T., Fields, D., Green, R., 2008. Effect of temperature and oxidation rate on carbon-isotope fractionation during methane oxidation by landfill cover materials. Environ. Sci. Technol. 42, 7818–7823. doi:10.1021/es801221y

Chanton, J.P., Powelson, D.K., Green, R.B., 2009. Methane oxidation in landfill cover soils, is a 10% default value reasonable? J. Environ. Qual. 38, 654–663. doi:10.2134/jeq2008.0221

Chanton, J.P., Rutkowski, C.M., Mosher, B., 1999. Qunatifying methane oxidation from landfills using stable isotope analysis of downwind plumes. Environ. Sci. Technol. 33, 3755–3760.

Christophersen, M., Kjeldsen, P., Holst, H., Chanton, J., 2001. Lateral gas transport in soil adjacent to an old landfill: factors governing emissions and methane oxidation. Waste Manag. Res. 19, 595–612. doi:10.1177/0734242X0101900616

Coleman, D.D., Risatti, J.B., Schoell, M., 1981. Fractionation of carbon and hydrogen isotopes by methane oxidizing bacteria. Geochim. Cosmochim. Acta 45, 1033–1037.

Czepiel, P.M., Shorter, J.H., Mosher, B., Allwine, E., McManus, J.B., Harriss, R.C., Kolb, C.E., Lamb, B.K., 2003. The influence of atmospheric pressure on landfill methane emissions.

Waste Manag. 23, 593–598. doi:10.1016/S0956-053X(03)00103-X

Figueroa, V.K., Mackie, K.R., Guarriello, N., Cooper, C.D., 2009. A Robust Method for Estimating Landfill Methane Emissions. J. Air Waste Manage. Assoc. 59, 925–935. doi:10.3155/1047-3289.59.8.925

Galle, B.O., Samuelsson, J., Svensson, B.H., Borjesson, G., 2001. Measurements of methane emissions from landfills using a time correlation tracer method based on FTIR absorption spectroscopy. Environ. Sci. Technol. 35, 21–25. doi:10.1021/es0011008

Goldsmith, C.D., Chanton, J., Abichou, T., Swan, N., Green, R., Hater, G., 2012. Methane emissions from 20 landfills across the United States using vertical radial plume mapping. J. Air Waste Manage. Assoc. 62, 183–197. doi:10.1080/10473289.2011.639480

IPCC, 2006. Guidelines for national greenhouse gas inventories. Available at http://www.ipcc-nggip.iges.or.jp/public/2006gl/index.html (accessed July 2016). Intergovernmental Panel on Climate Change, Geneva, Switzerland.

IPCC, 2013. Stocker, T.F., Qin, D.H., Plattner, G.K., Tignor, M.M.B., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P.M. Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Kjeldsen, P., Fischer, E. V., 1995. Landfill gas migration-Field investigations at Skellingsted landfill, Denmark. Waste Manag. Res. 13, 467–484. doi:10.1016/S0734-242X(05)80025-4

Lim, K.T., 1995. Mega-molecular dynamics on highly parallel computers: Methods and applications. PhD Dissertation. California Institute of Technology, Pasadena, California.

Liptay, K., Chanton, J., Czepiel, P., Mosher, B., 1998. Use of stable isotopes to determine methane oxidation in landfill cover soils. J. Geophys. Res. 103, 8243–8250.

Lohila, A., Laurila, T., Tuovinen, J.-P., Aurela, M., Hatakka, J., Thum, T., Pihlatie, M., Rinne, J., Vesala, T., 2007. Micrometeorological measurements of methane and carbon dioxide fluxes at a municipal landfill. Environ. Sci. Technol. 41, 2717–2722. doi:10.1021/es061631h

McBain, M.C., Warland, J.S., McBride, R. a, Wagner-Riddle, C., 2005. Micrometeorological measurements of N_2O and CH_4 emissions from a municipal solid waste landfill. Waste Manag. Res. 23, 409–419. doi:10.1177/0734242X05057253

Mønster, J., Samuelsson, J., Kjeldsen, P., Scheutz, C., 2015. Quantification of methane emissions from 15 Danish landfills using the mobile tracer dispersion method. Waste Manag. 35, 177–186. doi:10.1016/j.wasman.2014.09.006

Mønster, J.G., Samuelsson, J., Kjeldsen, P., Rella, C.W., Scheutz, C., 2014. Quantifying methane emission from fugitive sources by combining tracer release and downwind measurements - A sensitivity analysis based on multiple field surveys. Waste Manag. 34, 1416–1428. doi:10.1016/j.wasman.2014.03.025

Mosher, B.W., Czepiel, P.M., Harriss, R.C., Shorter, J.H., Kolb, C.E., Mcmanus, J.B., Allwine, E., Lamb, B.K., 1999. Methane Emissions at Nine Landfill Sites in the Northeastern United States. Environ. Sci. Technol. 33, 2088–2094. doi:10.1021/es981044z

Mou, Z., Scheutz, C., Kjeldsen, P., 2014. Evaluating the biochemical methane potential (BMP) of low-organic waste at Danish landfills. Waste Manag. 34, 2251–2259. doi:10.1016/j.wasman.2014.06.025

Mou, Z., Scheutz, C., Kjeldsen, P., 2015a. Evaluation and application of site-specific data to revise

the first-order decay model for estimating landfill gas generation and emissions at Danish landfills. J. Air Waste Manage. Assoc. 65, 686–698. doi:10.1080/10962247.2015.1008653

Mou, Z., Scheutz, C., Kjeldsen, P., 2015b. Evaluating the methane generation rate constant (k value) of low-organic waste at Danish landfills. Waste Manag. 35, 170–176. doi:10.1016/j.wasman.2014.10.003

Nastev, M., Therrien, R., Lefebvre, R., Gélinas, P., 2001. Gas production and migration in landfills and geological materials. J. Contam. Hydrol. 52, 187–211. doi:10.1016/S0169-7722(01)00158-9

Olsen, R., Willumsen, H.C., 2013. Characterization of Gas , Heat and Humidity Profiles in Landfilled Shredder Residue At Odense Nord Landfill , Denmark. Sardinia 2013, Proc. Fourteenth Int. Waste Manag. Landfill Symp. 30 Sept. 4 October, Sardinia, Italy.

Pauly, S., 1989, third ed.. In: Brandrup, J., Immergut, E.H. (Eds.), Permeability and Diffusion Data, Polymer Handbook Wiley, NY.

Poulsen, T.G., Christophersen, M., Moldrup, P., Kjeldsen, P., 2003. Relating landfill gas emissions to atmospheric pressure using numerical modelling and state-space analysis. Waste Manag. Res. 21, 356–366.

Rachor, I., Gebert, J., Grongroft, A., Pfeiffer, E.M., 2011. Assessment of the methane oxidation capacity of compacted soils intended for use as landfill cover materials. Waste Manag. 31, 833–842. doi:10.1016/j.wasman.2010.10.006

Scharff, H., Jacobs, J., 2006. Applying guidance for methane emission estimation for landfills. Waste Manag. 26, 417–429. doi:10.1016/j.wasman.2005.11.015

Scheutz, C., Fredenslund, A.M., Chanton, J., Pedersen, G.B., Kjeldsen, P., 2011a. Mitigation of methane emission from Fakse landfill using a biowindow system. Waste Manag. 31, 1018–1028. doi:10.1016/j.wasman.2011.01.024

Scheutz, C., Fredenslund, A.M., Nedenskov, J., Samuelsson, J., Kjeldsen, P., 2011b. Gas production, composition and emission at a modern disposal site receiving waste with a low-organic content. Waste Manag. 31, 946–955. doi:10.1016/j.wasman.2010.10.021

Scheutz, C., Kjeldsen, P., Bogner, J.E., De Visscher, A., Gebert, J., Hilger, H. a, Huber-Humer, M., Spokas, K., 2009. Microbial methane oxidation processes and technologies for mitigation of landfill gas emissions. Waste Manag. Res. 27, 409–455. doi:10.1177/0734242X09339325

Scheutz, C., Samuelsson, J., Fredenslund, A.M., Kjeldsen, P., 2011c. Quantification of multiple methane emission sources at landfills using a double tracer technique. Waste Manag. 31, 1009–1017. doi:10.1016/j.wasman.2011.01.015

Spokas, K., Bogner, J., Chanton, J.P., Morcet, M., Aran, C., Graff, C., Golvan, Y.M. Le, Hebe, I., 2006. Methane mass balance at three landfill sites: What is the efficiency of capture by gas collection systems? Waste Manag. 26, 516–525. doi:10.1016/j.wasman.2005.07.021

Thoma, E.D., Green, R.B., Hater, G.R., Goldsmith, C.D., Swan, N.D., Chase, M.J., Hashmonay, R. A., 2010. Development of EPA OTM 10 for Landfill Applications. J. Environ. Eng. 136, 769–776. doi:10.1061/(ASCE)EE.1943-7870.0000157

Tregoures, A., Beneito, A., Berne, P., Gonze, M.A., Sabroux, J.C., Savanne, D., Pokryszka, Z., Tauziede, C., Cellier, P., Laville, P., Milward, R., Arnaud, A., Levy, F., Burkhalter, R., 1999. Comparison of seven methods for measuring methane flux at a municipal solid waste landfill site. Waste Manag. Res. 17, 453–458.

USEPA, 2004. Direct Emissions from Landfilling Municipal Solid Waste, US Environmental Protection Agency, Washington, DC.

USEPA, 2013. Federal Register notice—78 F.R. 71925, 71968, 71971, November 29, 2013.

Xu, L., Amen, J., Lin, X., Welding, K., 2014. Impact of changes in barometric pressure on landfill methane emission. Global Biogeochem. Cycles 28, 679–695. doi:10.1002/2013GB004571

Young, A., 1992. The effects of fluctuations in atmospheric pressure on landfill gas migration and composition. Water, Air, Soil Pollut. 64, 601–616. doi:10.1007/BF00483369