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Closing the methane mass balance for an old closed Danish landfill

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Horizontal landfill gas migration; methane oxidation efficiencies; methane generation; gas recovery; tracer gas dispersion method; oxidation in landfill cover soil

Abstract

In this study, a methane (CH₄) mass balance was established for Hedeland landfill. CH₄ generation rates were modelled using a multiphase first-order decay model (The Afvalzorg model) and determined at between 57 and 79 kg h⁻¹. The CH₄ emission rate was quantified at between 2 and 14 kg h⁻¹, using the tracer gas dispersion method and the CH₄ gas recovery efficiency was between 8 and 21%. At three places along the perimeter of the landfill, gas remediation systems have been installed to protect the residential houses from any risk of migrating landfill gas. About 0.76 kg h⁻¹ of CH₄ was extracted from these three remediation systems. Using a carbon mass balance for the lateral migrating landfill gas showed a

fractional oxidation of about 78%, which corresponded to a CH₄ flux of 3.5 kg h⁻¹ from the three remediation systems, including the oxidised CH₄. The total lateral CH₄ flux (un-oxidised) from the total landfill perimeter was estimated at between 6.9 and 10.4 kg h⁻¹. CH₄ oxidation efficiency in the landfill cover soil, determined from stable carbon isotope analyses, was found to be between 12% and 92%. This resulted in an average CH₄ oxidation rate of 32 kg h⁻¹, using an average CH₄ emission rate of 8 kg h⁻¹. CH₄ surface screenings and surface flux measurements supported the hypothesis that oxidation efficiency was in the higher range and that oxidation could close the CH₄ mass balance.

1. Introduction

Landfilling waste containing organic matter will produce landfill gas (LFG), mainly in the forms of methane (CH₄) and carbon dioxide (CO₂). The greenhouse gas potential of CH₄ is 28 times greater than CO₂ over a 100-year period (not considering climate feedback) (IPCC, 2014), but it can also be explosive when mixed with air and might pose a risk to humans living near landfills. LFG can migrate to nearby houses and create an explosive atmosphere in confined spaces (Christophersen and Kjeldsen, 2001; Franzidis et al., 2008; Kumar et al., 2018; Williams and Aitkenhead, 1991). Furthermore, not only are landfills significant sources of CH₄ emissions into the atmosphere, but they are also a long-term sink for carbon, as a minimum of 50% of landfilled organic carbon is typically not converted into biogas carbon but remains in the landfill as lignin and cellulosic fractions, which decompose slowly (Bogner et al., 2008).

Methane generated in a landfill will have different pathways, as illustrated in Fig. 1. If CH₄ generation is significant, LFG can be collected and utilised for power and/or heat production, upgraded to biogas or flared for a thermal conversion to CO₂ (Stegmann, 1996).

Part of the LFG will migrate to the top cover, where it can be oxidised to CO₂ by methanotrophic bacteria in the cover soil. However, some of the migrating LFG may also escape into the atmosphere without any oxidation and add to anthropogenic CH₄ emission and accumulation in the atmosphere. LFG can also migrate laterally to surrounding areas, and finally some will be temporarily stored inside the landfill. This can be summarised in the following CH₄ balance for a landfill (Bogner and Spokas, 1993) (with all units in mass time⁻¹):

$$\text{CH}_4 \text{ generated} = \text{CH}_4 \text{ emitted} + \text{CH}_4 \text{ oxidised} + \text{CH}_4 \text{ recovered (flared or utilised)} + \text{CH}_4 \text{ migrated} + \Delta \text{CH}_4 \text{ storage} \quad (\text{Eq. 1})$$

Often, the only well-known term is the amount of CH₄ recovered, which is sometimes recorded based on flow and CH₄ concentration measurements. Alternatively the amount of recovered CH₄ can be calculated based on data (e.g. electricity produced) from the engine utilising the LFG. Generated CH₄ is often modelled based on landfilled waste amounts and waste compositions (Mou et al., 2015a), using different models for LFG generation (e.g. IPCC (IPCC, 2006) and Afvalzorg (Scharff et al., 2000; Scharff and Jacobs, 2006)), each of which has different advantages and disadvantages (Scharff and Jacobs, 2006).

Cracks and fissures in the top cover of the landfill and installations, such as leachate wells, can highly influence the magnitude of LFG emission. Also, the atmospheric pressure gradient will influence emissions (Rees-White et al., 2019; Xu et al., 2014). The emission of CH₄ can be quantified by using either remote methods, such as tracer gas dispersion, DIAL (Differential Absorption LiDAR) or radial plume mapping (e.g. Babilotte et al., 2010; Goldsmith et al., 2012; Mønster et al., 2015, 2014), or surface-based point measurements (e.g.

flux chamber measurements) that integrate total emissions (Mønster et al., 2019; Scheutz et al., 2009).

Potential CH₄ oxidation in the cover soil is influenced by the load of CH₄ to the cover, and soil conditions. CH₄ oxidised in the top cover can be estimated by measuring the fractionation of stable carbon isotopes in upwind samples and in the downwind plume, as methanotrophs prefers ¹²C over ¹³C (Börjesson et al., 2001; Chanton et al., 1999; Liptay et al., 1998).

The lateral CH₄ migration is influenced by different factors such as atmospheric conditions, the conditions of the bottom and side liners and the soil layers surrounding the landfill (Christophersen et al., 2001; Christophersen and Kjeldsen, 2001; Williams et al., 1999). Lateral migration is often calculated from diffusion through the bottom and side liners, under the assumption that the liner has not been compromised (Aghdam et al., 2018; Spokas et al., 2006). However, with a compromised liner, lateral migration can increase significantly, due to advective flow through cracks in the liner.

Change in CH₄ storage inside the landfill is probably the most difficult term to quantify. It can be calculated from changes in CH₄ concentration inside the landfill, the gas filled porosity of the waste body and the total landfill volume (Spokas et al., 2006). Changes in CH₄ storage are affected by many factors, such as fluctuations in the leachate level in the landfill, changes in void space from the decomposition of waste, compaction and settlement, changes in extraction efficiency and in the short term from changes in atmospheric pressure (Spokas et al., 2006).

The objective of this study was to establish a CH₄ mass balance for an old closed Danish landfill, namely Hedeland landfill. Lateral LFG migration and CH₄ emissions have been investigated for many years at Hedeland landfill, and the large amount of collected data

has been used to evaluate the different terms in a CH₄ mass balance and close this balance. A number of new site investigation methods were applied including vertical drilling and gas monitoring, whole-site CH₄ emission measurements and stable carbon isotopic analysis, to quantify CH₄ oxidation. The landfill site has neighbours less than 100 m away from the perimeter, and lateral gas migration is a potential risk. Therefore gas is collected outside the perimeter and vented into the atmosphere. Any future remediation needs to consider both the risk to neighbours and the environmental impact in terms of climate change. The establishment of the site's CH₄ mass balance forms the basis for designing and implementing a cost-efficient, long-term remediation strategy for the site.

2. Materials and methods

2.1 Site description

Hedeland landfill, near Roskilde, Denmark, is a controlled landfill with leachate and gas collection. The landfill covers an area of 115,000 m², of which approximately 100,000 m² has been used for landfilling. The site was established in 1979 and received approximately 2.9 million tonnes of mainly non-combustible waste and soil before after-care started in the middle of 2009. Hedeland was established in an old gravel pit and was constructed with bottom and side liners, which on the bottom and lower parts of the slopes consist of a polymer liner (no information about polymer type and thickness) on top of some refilled clay soil, and the upper parts of the slopes consist of at least one metre of clay, as shown in the cross-section diagram in Fig. 2. A leachate collection system was constructed with one pumping station draining leachate from all sections of the landfill. The side and bottom liners were not installed to prevent gas migration but to prevent leachate from entering the groundwater aquifer. In several places, the side liner was constructed without the polymer liner

overlapping; instead, a horizontal clay connection was installed (see Fig. 2). The final top cover consists of at least 1 m of soil with no polymer liner. In one section of the landfill, several metres of soil have been deposited at the top. The maximum waste filling height was approximately 40 m and the average filling height around 25 m.

The landfill has mainly received non-combustible waste with a low content of organic carbon. The main fractions consist of ‘soil’ (38%), ‘other waste’ (36%) not further specified in the available data, ‘other non-combustible’ (18%), waste incineration residues (‘bottom ash’, ‘fly ash’ and ‘flue gas cleaning residues’) (6%), ‘bulky waste’ (1%) and ‘waste water treatment sludge’ (1%). The remaining waste fractions make up less than 1% each.

The installed gas collection system consists of two parts. After establishing the permanent landfill cover, a gas collection system was installed in one section of the landfill with short vertical extraction wells (6-8 m deep). In another section of the landfill, a horizontal drainage system was installed on top of the waste, before the soil cover was installed. Knowledge about the landfill volume influenced by gas recovery is not available. The recovered gas is utilised in a dual-fuel diesel engine producing electricity.

Hedeland landfill is situated in a huge gravel deposit area. The gravel layer was originally between 10 and 15 m deep with an unsaturated zone of about 20 m deep. Within 100 m from the border of Hedeland landfill, seven residential houses are situated on top of a large unsaturated zone containing unsaturated gravel and sand, which is an easy route for lateral LFG migration and as such poses a risk for these residential houses. To stop the LFG from migrating to the houses, vertical remediation pumping wells and monitoring boreholes have been established outside of the landfilled waste.

2.2 Methane generation modelling

CH₄ generation in the landfill was modelled using the Afvalzorg model (Scharff et al., 2000; Scharff and Jacobs, 2006), which utilises first-order degradation kinetics and can accommodate eight waste categories with different biodegradable organic matter contents and degradation rates (rapid, moderate and slow) (Scharff & Jacobs, 2006). The model requires waste input data on an annual basis, distributed in maximum eight categories. Since 1997, there has been a ban in Denmark on landfilling waste suitable for incineration. Consequently, modern landfills in the country contain low-organic waste, which is also the case for Hedeland landfill. The Afvalzorg model contains datasets for low-organic waste categories, and previous studies have shown that this model represents Danish landfills best (Aghdam et al., 2018; Mou et al., 2015b; 2015a; 2014; Scheutz et al., 2011b). The net landfilled waste amounts, and their distribution into different waste types, are given in Table SM1 in the Supplementary Material (SM).

As part of an ongoing investigation at the landfill, four deep gas extraction wells were installed in the waste body, as part of a pumping test conducted from 2015 to 2017. The pumping tests were conducted to test if it was possible to prevent lateral migration of landfill gas to neighbouring plots by extracting gas in the waste body in the areas where lateral gas migration were observed. Locations of the four gas extraction wells are indicated in Fig. 3. Landfill gas was extracted from the waste with a pumping rate of 100-140 m³ h⁻¹ for at least two months from each well (albeit from one well at a time). After two months of pumping, CH₄ concentrations in the extracted gas seemed to have stabilised. Assuming that the stabilised CH₄ concentration was an indication of the level of CH₄ production in the waste body influenced by the extraction well, a CH₄ generation rate for the total waste body was calculated based on the applied pump rates, CH₄ concentrations measured in the extracted gas and an estimate of the influenced volume of the waste body from each extraction well using

literature data. A maximum influence radius around LFG extraction wells of about 75 m was found in both Ham (1996) and Lofy (1996). A rule of thumb is that the horizontal influence is ten times that of the vertical influence (Ham, 1996). This is supported by an observation in a field study, where the vacuum from an extraction well was found to extend to a maximum 9 m below the bottom of the screen (Lofy, 1996). The gas-filled porosity of the waste body was assumed to be $0.1 \text{ m}^3 \text{ m}^{-3}$ (Kjeldsen and Christensen, 2001), and it was further assumed that the influence of the extraction well was cylindrical around the screen – at a height that was indeed the height of the screen (in this case 4 m) plus twice the vertical influence (where the vertical influence radius is one-tenth of the horizontal influence (7.5 m) on both sides of the screen) resulting in a vertical influence of 19 m. From the estimated influence and CH_4 generation rate of the four pumping tests, a CH_4 generation rate for the total waste body of Hedeland landfill was calculated. This estimated rate from the pump test served solely as a comparison to the modelled CH_4 generation rate.

2.3 Methane extraction and utilization

Gas collected at the landfill is utilised in a dual-fuel engine, which uses diesel oil as its support fuel and produces electricity. The engine is run with the aim of maintaining stable electricity production and a constant consumption of diesel oil per produced kWh. When production goes down because of a decrease in the CH_4 content in the collected gas, the engine is stopped, until the level of CH_4 is expected to have built up again (no information is available about time periods with no gas extraction). The amount of CH_4 collected was estimated by the operator of the engine based on the amount of electricity produced and the amount of diesel oil used. The used conversion rate was 0.235 m^3 of CH_4 kWh^{-1} including the

diesel use and the efficiency of the engine. Based on this information, CH₄ extraction rates were calculated as an annual average.

2.4 Methane emission quantification

Total landfill CH₄ emissions were quantified using the tracer gas dispersion method, which combines the controlled release of tracer gas (acetylene (C₂H₂)) from the landfill with concentration measurements downwind of the landfill, using a mobile high-resolution analytical instrument (Mønster et al., 2015; 2014). The tracer gas dispersion method is based on the assumption that a tracer gas will disperse into the atmosphere in the same way as CH₄ when emitted from the landfill (Mønster et al., 2015, 2014). The tracer method and the analytical equipment (based on Cavity Ring-Down Spectroscopy (CRDS)) are described in detail in Mønster et al., (2015; 2014) and Scheutz and Kjeldsen, 2019.

Table 1 provides an overview of measurement campaigns, showing dates, measuring distances, numbers of plume transects, wind speed and direction, numbers of tracer release points and tracer gas release rates for the CH₄ emission quantifications conducted at Hedeland landfill. All four measurement campaigns were conducted under stable atmospheric conditions and were assumed to be representative of the average CH₄ emission from the landfill.

2.5 Changes in methane storage

Any change in CH₄ storage (short-term or long-term) can be calculated from a change in the average CH₄ concentration in the waste, total landfill volume and the gas-filled porosity of the landfill (Spokas et al., 2006).

Examples of processes causing short-term CH₄ storage changes (hours or days) include fluctuations in atmospheric pressure or in LFG extraction rates. The change in CH₄ storage was evaluated against a decrease in atmospheric pressure and a drop in CH₄ concentration, simulating changes in the extraction rate using the ideal gas law and assuming an average gas temperature of 40°C.

Changes in CH₄ storage over the long-term (years to decades) could be caused by processes such as waste degradation and settlement causing changes in void space and the amount of material available for degradation and gas production. Long-term changes were not considered relevant, as the landfilled waste at Hedeland is very old (10 to 40 years) and no changes have been made to the landfill for many years since it was closed in 2009. Data for the mass balance thus represented a relative short time interval (about three years, from 2013 to 2015) in comparison to the lifetime of the landfill.

2.6 Lateral methane migration

To monitor the lateral migration of LFG, monitoring wells were installed and cased with two-metre screens placed in the upper part of the unsaturated gravel layer. In total, approximately 150 monitoring wells have been installed over the years, in order to monitor LFG content in the soil formation adjacent to the landfill. At three locations, remediation pumps have been installed on the wells, to pump any migrating LFG from the soil, before it reaches the residential houses. Gas compositions (content of CH₄, CO₂ and oxygen (O₂)) at each screen have been monitored several times a year for many years, thereby providing detailed information about laterally migrating LFG.

From the installed gas remediation pumps, the flow rate of the pumps and the recorded concentrations of CH₄ and CO₂ in the off gas, the lateral fluxes of CH₄ and CO₂ coming from

the landfill were estimated. Comparing the composition of the gas extracted from the remediation systems with landfill gas extracted from the gas wells installed in the waste indicated that a part of the CH₄ was oxidised during the lateral migration of the landfill gas. The soil compartment consisted of an unsaturated layer of mainly gravel and sand, thus it was assumed that the background level of CO₂ in the soil compartment was zero (assuming insignificant soil respiration), and then the initial amount of CH₄ migrating from the landfill, before oxidation, could be calculated from a carbon mass balance, using (Christophersen et al., 2001):

$$J_{CH_4,LFG} = C_{CH_4,LFG} \cdot (C_{CH_4,LFG} + C_{CO_2,LFG})^{-1} \cdot (J_{CH_4,RW} + J_{CO_2,RW}) \quad (\text{Eq. 2})$$

Where $J_{CH_4,LFG}$ is the flux of CH₄ migrating laterally through the landfill bottom and side liner (mol h⁻¹) before oxidation, $C_{CH_4,LFG}$ and $C_{CO_2,LFG}$ are the concentrations of CH₄ and CO₂ measured in the raw LFG (vol.%), respectively and $J_{CH_4,RW}$ and $J_{CO_2,RW}$ are the fluxes of CH₄ and CO₂ in the remediation wells (RW) (mol h⁻¹), respectively.

From the many years of investigations regarding laterally LFG migration, it is well documented where most of the lateral transport takes place. Unfortunately most of the transport occurs in the areas with residential houses and to protect the neighbours, remediation systems have been installed. The CH₄ flux for the remaining landfill perimeter was estimated based on recorded CH₄ concentrations in the monitoring wells located along the landfill perimeter. The perimeter of the landfill was divided in three groups: areas with remediation wells installed (corresponding to 25% of the total length of the perimeter), areas where CH₄ was detected (>1 vol.% CH₄ (between 1-27 vol.%)) in monitoring wells (not influenced by the extraction from the remediation wells) (25% of the total length of the

perimeter) and areas where no or very low CH₄ concentrations was detected (<1 vol.% CH₄) (50% of the total length of perimeter) (see Fig. 3). To assess a maximum and minimum CH₄ flux two approaches were used. The maximum flux of migrating CH₄ from the whole landfill perimeter, was estimated based on the assumption that the flux in areas where CH₄ was detected was the same as the flux detected in the areas with remediation systems, while it was assumed to be half in the areas where no CH₄ was detected (worst case, see Eq. 3). For estimating a minimum flux, the total flux of laterally migrating CH₄ was calculated based on the assumption that the flux in the areas where CH₄ was detected was half of the flux detected in the areas with remediation systems and only 25% in the areas where no CH₄ was detected (see Eq. 4). It should be noted, that a concentration of CH₄ below detection is not equal to no flux of LFG, due to oxidation of CH₄ in the soil compartment. The total maximum and minimum flux of CH₄ (un-oxidised); J_{CH₄,totalLM} migrating laterally from the landfill were given by:

$$J_{CH_4, totalLM, max} = J_{CH_4, LFG} + J_{CH_4, LFG} + 2 \cdot 0.5 \cdot J_{CH_4, LFG} = 3 \cdot J_{CH_4, LFG} \quad (\text{Eq. 3})$$

and

$$J_{CH_4, totalLM, min} = J_{CH_4, LFG} + 0.5 \cdot J_{CH_4, LFG} + 2 \cdot 0.25 \cdot J_{CH_4, LFG} = 2 \cdot J_{CH_4, LFG} \quad (\text{Eq. 4})$$

The CH₄ flux was thus assumed to range between the minimum and maximum flux with an average flux calculated as the average of the two.

2.7 Methane oxidation potential in the cover soil

The CH₄ oxidation potential in landfill cover soil can be estimated based on the analysis of the stable ¹³C isotope in upwind, downwind and raw gas samples (Chanton et al., 1999).

The method is based on methanotrophic bacteria's preference for oxidising ^{12}C over ^{13}C , so CH_4 becomes ^{13}C -enriched after exposure to methanotrophs (for a detailed description of the method, see e.g. Börjesson et al., 2007; Chanton and Liptay, 2000; Chanton et al., 2008, 1999; Liptay et al., 1998).

The CH_4 oxidation rate can be calculated from the CH_4 emission rate and the fraction of CH_4 oxidised in the cover soil, $f_{\text{ox,cover}}$, using the following equation (Börjesson et al., 2007; Scheutz et al., 2011a):

$$\text{MO} = f_{\text{ox,cover}} \cdot E \cdot (1 - f_{\text{ox}})^{-1} \quad (\text{Eq. 5})$$

where MO is the CH_4 oxidation rate (kg h^{-1}), f_{ox} is the fraction of CH_4 oxidised (%) and E is the CH_4 emission rate (kg h^{-1}). Next, $f_{\text{ox,cover}}$ can be calculated from (Börjesson et al., 2007; Chanton et al., 1999):

$$f_{\text{ox,cover}} = (\delta_{\text{excess}} - \delta_A) (1000 (\alpha_{\text{ox}} - \alpha_{\text{trans}}))^{-1} \quad (\text{Eq. 6})$$

where δ_{excess} is the excess of $\delta^{13}\text{C}$ in the downwind plume, corrected for the upwind or background samples, δ_A is the carbon isotopic content of raw gas CH_4 , α_{ox} is the isotope fractionation factor related to CH_4 oxidation by the landfill cover soil and α_{trans} is the isotope fractionation factor related to the transportation of gas through the cover soil.

Five raw gas samples were collected from both monitoring wells in the waste body of the landfill and from the gas extraction system. Downwind gas samples were collected in the plume, using CRDS to identify the plume. Three soil cover samples were collected from 10 cm below the surface and used to determine α_{ox} by incubation in the laboratory. The samples

were incubated at 22°C, following which gas samples were collected from the incubation bottles to analyse carbon isotope fractionation. The specific incubation test is described in more detail in Aghdam et al. (2018). All gas samples were analysed at Florida State University. Upwind of Hedeland landfill is another old landfill, which influences the fractionation of upwind samples from Hedeland, due to oxidation in its cover. As a result, fractionation (δCH_4) was obtained from another Danish landfill with upwind samples from the sea, as described in Aghdam et al. (2018).

3. Results and discussion

The years 2013-2015 were selected for the CH_4 balance, as these provided the most complete dataset. The CH_4 mass balance is given as an average for these three years.

3.1 Methane generation modelling

The result of the modelled CH_4 generation over the lifetime of the landfill can be seen in Fig. 4. The generation peaked in 1989-1991 and has decreased since, albeit with a small increase again in 2006-2010, where the annual landfilled waste amounts increased significantly before waste disposal was terminated in the middle of 2009. Gas generation was modelled at between 57 and 79 kg h^{-1} in the period 2013-2015, with an average of $67 \pm 8.6 \text{ kg h}^{-1}$ (average of maximum and minimum generation rates for the three years $\pm$ the standard deviation (STD)). CH_4 generation modelling was uncertain, as knowledge about the exact waste composition was limited. The main fractions of landfilled waste were unspecified or categorised in waste types, with no possibility of identifying potential biodegradable carbon content. Table SM1 and Table SM2 in the SM show the annual landfilled waste amounts

distributed in Afvalzorg waste categories and the translation of Danish waste categories and fractions to Afvalzorg waste categories, respectively.

In addition, CH₄ generation was estimated based on data from the pumping tests from four gas extraction wells in the waste body. Pumping rates and CH₄ concentrations in the extracted gas after stabilisation showed CH₄ generation rates of approximately 6, 11, 21 and 28 kg h⁻¹ for each of the four extraction wells, respectively. Summarising the CH₄ generation rates from these tests gave a total CH₄ generation rate of 66 kg h⁻¹ from the four extraction wells. Using a radius of influence of 75 m and a vertical influence height of 19 m, coverage of the total landfill body from the four pumping tests was estimated at about 71%. Assuming that the CH₄ generation rate was the same in the whole waste body, this corresponds to a total generation rate of 92 kg h⁻¹, based on the pumping tests. The assumed influence radius of 75 m was found valid based on observed concentration response curves in monitoring wells located about 65-75 m from one of the four wells (well A2 in Fig. 3) used in the pumping tests. Data from the three other pumping test wells, though, were not as clear about the influence radius. There were no monitoring wells installed in the waste body inside the expected influence radius of these extraction wells (wells A7, A8 and A11 in Fig. 3), thus observations were only possible in monitoring wells located outside the waste body. However, during extraction no responses were observed in the nearest monitoring wells located about 40 m to 60 m from an extraction well. This could indicate that the radius of influence could be lower than 40 m for these wells. Assuming that only one of the four pumping tests reached the maximum estimated influence, and the three remaining pumping tests only reached half of the maximum, then 66 kg h⁻¹ corresponds to 44% of the total landfill volume, in which case the total CH₄ generation rate would then be 148 kg h⁻¹, assuming the same generation rate for the total volume.

Both the modelled CH₄ generation rate and the CH₄ generation rate estimated from the pumping tests included a number of assumptions (e.g. deposited waste fractions, distribution in waste categories, carbon contents and degradation rates, influenced landfill volume from pumps, no overlap of influence from the four pumps, total gas-filled landfill volume). Both methods however, produced reasonable comparable results, thus supporting the findings and reducing some of the uncertainty. The modelled generation rate was used as the basis for the mass balance.

Mou et al. (2015a) compared modelled CH₄ generation rates, using the Afvalzorg model with standard values and a revised model using site-specific values for biochemical CH₄ potential (BMP) and the CH₄ generation rate constant (k-value) at four Danish landfills. They found that the Afvalzorg model using standard values overestimated the generation rate compared to field measurements using a CH₄ balance approach (considering CH₄ measured using the tracer gas dispersion method, recorded gas recovery rates and assuming oxidation of 10% of the emission rate), whereas using a revised model gave more comparable results. Other studies at Danish landfills, comparable to Hedeland landfill in terms of age and accepted waste categories, showed CH₄ generation rates using the Afvalzorg model comparable to CH₄ generation rates obtained by establishment of CH₄ mass balances based on field measurements (Aghdam et al., 2018; Scheutz et al., 2011b). Aghdam et al. (2018), for instance, found good agreement between estimated CH₄ generation by using a mass balance approach (including gas recovery, oxidation estimated from stable carbon isotope analysis and CH₄ emissions measured with the tracer gas dispersion method) and modelled using default values in Afvalzorg, compared to when site-specific data were used in the model. Scheutz et al. (2011b) compared gas generation rates obtained experimentally from waste sample incubations to modelled generation rates using the Afvalzorg and GasSim model. The results

showed that application of the Afvalzorg model gave the generation rate most comparable to the experimental rates.

3.2 Methane extraction and utilisation

Data from the operator of the LFG utilisation engine show that the recovered CH₄ rate varied between 6 and 12 kg h⁻¹, from 2013 to 2015, with an average recovery rate of 9 ± 2.9 kg h⁻¹, which corresponds to a recovery efficiency of between 8 and 21% of the total CH₄ generated (67 kg h⁻¹). The recovery rate is based on the average annual recovered CH₄. In three years, the amount of recovered CH₄ varied by a factor of two, mainly because the engine was out for service in autumn 2014 and also had some longer stops in 2015, thereby significantly decreasing annual recovery for these years.

Recovery efficiency at Hedeland landfill is low compared to other studies (e.g. Aghdam et al., 2018; Börjesson et al., 2009; Spokas et al., 2006). At three French landfills, for instance, recovery efficiencies of over 80% were found at seven out of eight landfill sites with a recovery system, and four had recovery efficiencies above 90% (Spokas et al., 2006). Börjesson et al. (2009) studied seven Swedish landfills, where recovery rates from 32 to 1006 kg h⁻¹ were found, resulting in recovery efficiencies of 14-78%. The low gas recovery efficiency at Hedeland landfill is due to relatively few – and shallow – gas recovery wells of only 6-8 m depth in a waste body at least 20 m deep.

3.3 Methane emission quantification

Four measuring campaigns were conducted over three years to measure total CH₄ emissions from Hedeland landfill in different conditions. For details about these measurement conditions, see Table 1. The measured total CH₄ emission rate ranged between 3.1 and 13.4

kg h⁻¹, with an average of 8 ±4.1 kg h⁻¹. Table 2 shows the results for all four measuring campaigns. The results of the emission quantifications show that operation of the gas recovery system highly influences the magnitude of CH₄ emissions from the site. The longer the recovery was stopped prior to a measuring campaign, the higher the emission rate. The lowest emission rate of 3.1 ±0.7 kg h⁻¹ was observed when gas recovery was in operation (recovering gas at an annual average rate of 12 kg h⁻¹), while the highest emission rate of 13.4 ±0.8 kg h⁻¹ was observed when the LFG recovery had been stopped for about 15 weeks prior to measurement. Measurements performed 2 and 8 weeks after LFG recovery had been stopped showed emission rates of 7.7 ±0.6 kg h⁻¹ and 9.8 ±1.0 kg h⁻¹, respectively. This suggests that there was some correlation between CH₄ emission rates and CH₄ recovery rates.

The emission rate at Hedeland landfill is lower than what has been found in previous studies at other Danish landfills. Quantifying CH₄ emission rates from 15 Danish landfills, using the tracer gas dispersion method, showed emission rates ranging from 2.6 to 60.8 kg h⁻¹, with Hedeland landfill in the lower half of the investigated sites (Mønster et al., 2015). Seven Swedish landfills investigated by Börjesson et al. (2009) showed emission rates between 12.8 and 441 kg h⁻¹, with the emission rate from Hedeland landfill comparable to the landfill with the lowest emission rate.

3.4 Changes in methane storage

Changes in CH₄ storage are affected by many factors, such as a change in the void space from waste decomposition, fluctuations in the leachate level, changes in gas recovery efficiencies or changes in temperature or atmospheric pressure. These factors result in fluctuations in the amount of CH₄ gas in the gas-filled volume of the landfill. Two of these change factors were evaluated herein, namely atmospheric pressure and gas recovery

efficiency, each resulting in a change in the CH₄ level. A change in atmospheric pressure of ± 1 mbar results in a variation in the mass of CH₄ in the waste of $\pm 0.6 \text{ g CH}_4 \text{ m}^{-3} \text{ LFG mbar}^{-1}$, using the ideal gas law and an average constant gas temperature of 40°C. At Hedeland landfill, with an assumed gas-filled porosity of $0.1 \text{ m}^3 \text{ m}^{-3}$, it resulted in a change in storage of 148 kg, and with an average CH₄ generation rate of 67 kg h^{-1} , it corresponded to approximately two hours of CH₄ generation per mbar change in atmospheric pressure. A change in recovery efficiency could result in a fluctuation in CH₄ concentration in the gas-filled volume of the landfill. In the evaluation of the effect, a change of 1 vol.% in the CH₄ concentration was assumed. From the total landfill volume, assumed gas-filled porosity and a change in the amount of CH₄ of 1 vol.% result in a fluctuation in CH₄ storage of approximately 1600 kg, corresponding to one day of CH₄ generation per 1 vol.% change in CH₄ concentration.

The two factors assessed showed that associated changes in CH₄ storage would only be relevant on short-term (hours to days) and rather quickly be levelled out by the continuous gas generation. Most of the data (e.g. gas collected via the remediation system and gas collected via the gas collection system) used in the CH₄ mass balance were measured continuously over longer time periods (months to years). Using average data in the mass balance is thus expected to level out any short-term changes. Whole site CH₄ emission measurements were limited to four times. However, these were carried out during periods with stable atmospheric pressure and the emission rates should not be impacted by pressure changes. The results indicated that the CH₄ emission rate was influenced by the gas collection (see section 3.3). However, as measurements were carried out during different operational conditions the average emission rate can be expected to be a reasonably representative estimate of the

emission. In summary, the change in short-term storage was assessed as not being important for the overall CH₄ mass balance at Hedeland landfill.

3.5 Lateral methane migration

Several years of investigations at Hedeland landfill identified lateral CH₄ migration. Fig. 3 shows locations on the perimeter of the landfill where LFG has been detected in the unsaturated gravel layer. A concentration of CH₄ above 1 vol.% has been defined as not acceptable by the competent authorities at Hedeland landfill. The red areas indicate where CH₄ concentrations above 1 vol.% have been detected, while numbers in purple indicate the maximum concentration of CH₄ measured in 2014. Based on data from the remediation pumping system, the total rate of CH₄ captured by the three areas with a remediation system was determined at 0.76 kg h⁻¹ in 2015.

Data from the many monitoring wells at the landfill perimeter and at the neighbouring plots showed that for almost every measurement in the unsaturated zone around the landfill, the CO₂ concentration was higher than the CH₄ concentration – an indication of oxidation. From the carbon mass balance presented in Eq. 2 the flux of CH₄ laterally migrating from the landfill to the three areas affected by the remediation system, including the oxidised CH₄ was estimated to 3.5 kg h⁻¹ of this was 2.7 kg h⁻¹ oxidised. The total amount of CH₄ laterally migrating from the whole perimeter of the landfill can be estimated using Eq. 3 and Eq. 4 and was determined at 10.4 kg h⁻¹ and 6.9 kg h⁻¹, respectively corresponding to 9-18% of the generated CH₄ (assuming a CH₄ generation rate of 57-79 kg h⁻¹) giving an average lateral CH₄ flux of 9 ± 1.7 kg h⁻¹. Details about raw LFG composition and the off gas composition from the remediation well is given in Table SM3 in SM.

An oxidation rate of $0.1\text{--}0.3\ \mu\text{g CH}_4\ \text{g}^{-1}\ \text{h}^{-1}$ was found in a previous study, wherein gravel samples from the unsaturated layer at Hedeland landfill were incubated in a batch test (Pade and Paamand, 2008). This oxidation rate is relatively low in comparison to most CH_4 oxidation rates observed in landfill cover soils ($0.0024\text{--}173\ \mu\text{g CH}_4\ \text{g}^{-1}\ \text{h}^{-1}$ from batch studies of landfill cover soils (Scheutz et al., 2009)); however, due to the large unsaturated zone (10–20 m deep), even a low oxidation rate can become significant. Using an average oxidation rate of $0.2\ \mu\text{g CH}_4\ \text{g}^{-1}\ \text{h}^{-1}$, about $4\ \text{kg h}^{-1}$ would be oxidised during 1 m of lateral transport in the unsaturated zone ($\sim 15,000\ \text{m}^3$ in total, assuming a 10 m-deep unsaturated gravel zone and a 1500 m perimeter). This example assumes an even flow throughout the whole of the perimeter, which is not likely. However, considering the distance of 15–25 m from the liner to the screen of the monitoring wells used for remediation the example shows that landfill CH_4 might undergo significant oxidation during transportation in the unsaturated zone. Results from the batch test run by Pade and Paamand (2008) also showed very low respiration, with CO_2 production due to the respiration of only $0.0004\ \mu\text{g CO}_2\ \text{g}^{-1}\ \text{h}^{-1}$, which validates the assumption of negligible soil respiration for the use of Eq. 2.

An LFG plume was found in the unsaturated zone, stretching more than 100 m from the boundary of an old landfill in the United Kingdom, situated in an abandoned sand/gravel pit (Williams et al., 1999). (Williams et al., 1999) found changes in stable hydrogen and carbon isotopes, and a reduction in the CH_4 to CO_2 ratio, clearly indicating microbial CH_4 oxidation in the plume. Over a distance of less than 2.5 m, CH_4 concentrations decreased from approximately 20 vol.% to less than 1 vol.% at the eastern fringe of the plume. Two oxidation rates were found from the stable isotopes, namely a slow rate (half-life of 4.3 – 7.6 years) in the centre of the plume, where oxygen was limited, and a faster rate (half-life of 0.76–1.21

years) at the top of the plume and further away from the landfill (more than 60 m) (Williams et al., 1999).

Lateral LFG migration has been studied intensively at a Danish landfill similar to Hedeland by Christophersen et al. (2001), Christophersen and Kjeldsen (2001) and Kjeldsen and Fischer (1995). These studies found that changes in atmospheric pressure were an important factor driving lateral LFG migration. Seasonal variations in CH₄ and CO₂ concentrations with, in general, higher CO₂ surface fluxes in the summer were observed. On an annual basis, 89% of the laterally migrating LFG was oxidised (Christophersen et al., 2001).

Lateral migration of LFG can be caused by diffusion, advection or a combination thereof. Assuming diffusion solely through a non-compromised polymer liner, a 1.5 mm HDPE polymer liner with a partial pressure decrease over the liner of 1 atm CH₄ will allow 0.58 cm³ CH₄ m⁻² d⁻¹ to pass through (Lim, 1995; Pauly, 1989). Information about the used polymer type and thickness of the bottom and side liners at Hedeland landfill was not available. Assuming a HDPE liner of 1.5 mm, the flux found at Hedeland landfill would result in a total diffusion of CH₄ through the bottom and side liners of 0.0024 kg h⁻¹, which is significantly lower than the migration rate found from the remediation systems, indicating that lateral migration at Hedeland is not caused purely by diffusion through a non-compromised liner. Results from the investigations along the perimeter of the landfill indicate that the side liner is compromised, possibly because it was constructed primarily to protect against seeping leachate and not specifically to prevent gas migration (Fig. 2), or because it has been damaged due to age and/or when the waste was landfilled.

3.6 Methane oxidation potential in cover soil

Samples for stable carbon isotopic analysis were collected in May 2016. The gas collection system was running during the sampling. Three downwind samples were analysed to estimate the CH₄ oxidation potential in the cover soil. Table 3 shows results for the upwind (collected at another Danish landfill, Stige Ø) and downwind CH₄ concentrations, and $\delta^{13}\text{CH}_4$, excess $\delta^{13}\text{CH}_4$, anoxic CH₄ and CH₄ oxidation efficiency, f_{ox} , from the stable carbon isotopic analysis. The downwind concentration of CH₄ ranged between 2.29 and 2.52 ppm, and $\delta^{13}\text{CH}_4$ ranged between -46.13 and -48.63. Compared to what had been found for other Danish landfills in Aghdam et al. (2018) and Scheutz et al. (2011), the $\delta^{13}\text{CH}_4$ was a little higher. The five samples of anoxic CH₄, namely δ_A , ranged between -51.46 and -58.00, which was also a little higher than found in Aghdam et al. (2018) and Scheutz et al. (2011). The three different downwind samples result in CH₄ oxidation efficiency in the cover soil of 12, 27 and 92%. In general, average efficiency is higher than the efficiencies found in Aghdam et al. (2018) and Scheutz et al. (2011), though they are comparable to what was found by Börjesson et al. (2007) at two closed Swedish landfills.

Using Eq. 6 with an average emission rate of 8 kg h⁻¹, and the three oxidation efficiencies found in Table 3, the CH₄ oxidation rate ranged between 1 kg h⁻¹ and 92 kg h⁻¹, resulting in an average oxidation rate of 32 ± 52 kg h⁻¹.

Oxidation rates determined by stable carbon isotopic analysis showed large variations and should therefore be interpreted with caution. First, the number of samples was limited, and second, the stable carbon isotopic method holds a number of uncertainties (e.g. the assumption that isotopic fractionation is only governed by bacteriological CH₄ oxidation, and gas transport through the cover is only governed by advection ($\alpha_{\text{trans}}=1$)) (Chanton et al., 2009; Gebert et al., 2011; Gebert and Streese-kleeberg, 2017). Recent field studies indicate that whole landfill site CH₄ oxidation efficiencies are underestimated when stable carbon

isotopic analysis is applied to upwind and downwind CH₄ plume samples, and the method thus provides minimum oxidation efficiency for CH₄ oxidation (Bourn et al., 2019; Sparrow et al., 2019).

The whole surface of the landfill was screened using a portable flame ionisation detector (FID) (TVA1000B, Thermo Scientific, USA), and relatively few hotspot areas were found with elevated concentrations of CH₄ (≥ 25 ppm) at the surface, thereby indicating limited CH₄ emissions. Results of the FID surface screening can be found in Fig. SM1 in SM. One of the hotspot areas with several elevated CH₄ concentrations has been investigated in four flux emission measuring campaigns (see Fig. SM1 in SM for location), with surface CH₄ fluxes only detected when atmospheric pressure decreased (Fjelsted et al., 2018). In that study, surface fluxes of CH₄ and CO₂ were measured at 100 points in a 100 m² area, and the average CH₄ and CO₂ flux was reported at between 0 and 0.28 mol CH₄ m⁻² d⁻¹ (average of 0.08 mol CH₄ m⁻² d⁻¹) and 0 and 0.57 mol CO₂ m⁻² d⁻¹ (average of 0.38 mol CO₂ m⁻² d⁻¹). Using Eq. 2 with the measured fluxes results in a fractional CH₄ oxidation in the landfill cover of 79%, which is comparable to the highest efficiency found using the stable isotopic method.

Many studies have been done through the years to determine the oxidation of LFG in cover soils, illustrating that the oxidation rate varies and depends on the cover type and conditions of the cover. In a review study by Chanton et al. (2009), 42 determinations of the fraction of oxidised CH₄ gave an overall average of 36% (a standard error of 6%) when including both laboratory column tests and field investigations. The highest oxidation efficiencies found in field measurements were 84% and 89% (the first from isotope fractionation, the latter from a CH₄ and CO₂ mass balance).

3.7 Closing the methane mass balance

The CH₄ mass balance for Hedeland landfill assuming a modelled CH₄ generation of 67 kg h⁻¹ is summarised in Fig. 5. The modelled generated CH₄ was 67 ±8.6 kg h⁻¹, while CH₄ emitted was 8 ±4.1 kg h⁻¹, CH₄ recovered was 9 ±2.9 kg h⁻¹ and lateral CH₄ migration was 9 ±1.7 kg h⁻¹ (including lateral oxidation). Individual terms in the mass balance have different uncertainties, but oxidation in the landfill cover was considered the most uncertain of these different terms. Using average rates for all terms other than oxidation in the cover leaves a deficit in the CH₄ mass balance of 42 kg h⁻¹, corresponding to 63% of the generated CH₄. Several observations supported high CH₄ oxidation in the landfill cover. Soil cover oxidation was supported by stable carbon isotopic analysis, indicating fractional oxidation of 12 to 92%. Carbon mass balance for the molar surface flux emission of CH₄ to CO₂ showed signs of fractional oxidation of about 79%. A low total CH₄ emission rate, found using the tracer gas dispersion method and the relatively few hotspots with elevated CH₄ concentration at the surface, also supported a high oxidation rate in the landfill cover.

Oxidation of the mass balance deficit (42 kg h⁻¹) corresponds to a load to the soil cover of 20 g m⁻² d⁻¹ (assuming half of the landfill surface area is active, which equates to 50,000 m²). This load is significantly lower than most of the oxidation rates found in a review of 24 column experiments simulating landfill cover soil with CH₄ oxidation rates in the range between 22 and 230 g CH₄ m⁻² d⁻¹ (Scheutz et al., 2009). Moreover, Börjesson et al. (2007) found oxidation rates of 72-106 kg h⁻¹ at a closed landfill in Sweden containing household waste, but with an emission rate, recovery rate and generation rate significantly higher compared to what was found for Hedeland landfill. All in all, the results from Hedeland landfill supported by CH₄ oxidation rates reported in the scientific literature clearly show that CH₄ oxidation in the landfill cover is a significant pathway, and can account for 63% of the CH₄ generated at Hedeland landfill.

4. Conclusion and perspectives

A methane (CH_4) mass balance was established for an old closed Danish landfill containing waste with a low organic carbon content. CH_4 emissions, recovery and migration were estimated and compared to the modelled CH_4 generation. The study showed that in average 37% of the generated CH_4 (67 kg h^{-1}) was accounted for by emissions (12%), recovery (13%) and lateral migration (including lateral oxidation) (13%). Several observations including stable carbon isotopic analysis, surface CH_4 screenings and flux measurements supported a significant oxidation rate in the landfill cover soil, which could account for the deficit of 42 kg h^{-1} (corresponding to 63% of the generated CH_4 being oxidized or 84% of the methane load to the cover soil). CH_4 oxidation was unexpectedly high – much higher than the 10% of the cover load, which is suggested in the IPCC guideline for national inventory reporting.

Landfill bottom liners are constructed primarily with the aim of protecting groundwater and surroundings against seeping leachate, and they do not focus on protection against migrating LFG. Considering the CH_4 mass balance helps in designing and developing a permanent, cost-efficient solution for LFG at the site that considers both the risk of migrating LFG for neighbours and the consequences of climate change from CH_4 emissions. The relatively low recovery rate indicates that there is a potential for increasing gas recovery and utilisation, but this will require installing new and/or deeper extraction wells, which is an expensive undertaking. A thorough analysis of the economic and environmental impacts of increased recovery is required to determine the benefits in this regard. Off gas from the remediation system can be treated in a biofilter, which could also handle the dilute gas from the extraction system, when CH_4 concentrations fall to a level where utilisation is no longer

feasible. Moreover, the high oxidation rate in the cover could be utilised in the elimination of undesired emissions from the remediation system by re-infiltrating off gas from the remediation system in the wells already installed in the waste body.

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Supplemental Material

Supplemental material, with data on waste amounts, waste composition and the distribution of accepted waste categories into Afvalzorg categories, results of the surface screening, calculation of CH₄ oxidation in lateral migrating LFG, are available online. Calculation of the CH₄ mass balance deficit using both modelled and estimated (from pumping tests) CH₄ generation is also available online.

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Tables

Table 1. Overview of CH₄ emission measurement campaigns conducted at Hedeland landfill, using the tracer gas dispersion method.

Date	Measuring distance to the landfill [m] and total number of plume transects made (n)	Avg. wind speed [m s ⁻¹] and dominant direction	Number of tracer gas release points	Total tracer gas release rate [kg h ⁻¹]
14.05.2013	1000 (18)	5 S	2	0.9
21.05.2014	700 (8)	5.5 E-SE	2	1.3
27.11.2014	800 (45)	3 SE	3	1.4
20.11.2015	1100 and 4000 (11 and 10)	2.3 N-NE	2	2.1

Table 2. Overview of the measurement campaigns quantifying whole-site CH₄ emissions.

	14-05-2013	21-05-2014	27-11-2014	20-11-2015
CH ₄ emission [kg h ⁻¹]	3.1 ± 0.7	7.7 ± 0.6	13.4 ± 0.8	9.8 ± 1.0
Gas collection on/off	On	Off	Off	Off
CH ₄ recovery (Annual average) [kg h ⁻¹]	12	LFG recovery stopped for two weeks prior to measurement	LFG recovery stopped for 15 weeks prior to measurement	LFG stopped for 8 weeks prior to measurement

804 **Table 3.** Results from the stable carbon isotope analysis. Three downwind samples were
 805 analysed, and the results are presented in the table. Results from upwind samples and anoxic
 806 CH₄ are the average of three and five samples, respectively.

Sample no.	Downwind CH ₄ concentration [ppmv]	Downwind δ ¹³ CH ₄	Upwind CH ₄ concentration [ppmv]	Upwind δ ¹³ CH ₄	Excess δ ¹³ CH ₄	Anoxic CH ₄ ; δA	f _{ox}
1	2.52	-48.31	1.92	-47.79	-49.93	-55.07	0.27
2	2.30	-46.13	1.92	-47.79	-37.85	-55.07	0.92
3	2.29	-48.63	1.92	-47.79	-52.85	-55.07	0.12

807 ^a Data from Stige Ø (Aghdam et al., 2018).

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Figures

Fig. 1. Conceptual model for the landfill's CH₄ mass balance and CH₄ distribution. Modified from Scheutz et al. (2009).

Fig. 3. Areas along the perimeter of the landfill with elevated levels of LFG (>1 vol.% CH_4).

Areas in yellow boxes have remediation pumps protecting against migrating LFG, named I, II

and III. A2, A7, A8 and A11 indicates the location of the pumping test wells used in the

calculation of CH_4 generation rates and the CH_4 oxidation in the lateral migrating LFG.

Fig. 4. Gas generation modelling using Afvalzorg and the annual average CH₄ extraction rate for 2013-2015. Generation is based on disposed waste amounts and composition as shown in Table SM1 in the Supplementary Material.

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835 **Fig. 5.** Methane mass balance for Hedeland landfill. Lateral CH₄ migration includes CH₄
 836 oxidised in the soil compartment in the area between the side liner and the monitoring wells.

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