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

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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).

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 Eq. (1):

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

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

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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)):

$$GRE\ (\%) = \left(\frac{CH_4\ recovered}{CH_4\ generated}\right) \times 100 \quad (2)$$

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):

$$CH_4\ generated = CH_4\ emitted + CH_4\ oxidized + CH_4\ recovered + CH_4\ migrated + \Delta CH_4\ storage \quad (3)$$

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 ± 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 (ΔCH_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.

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