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Quantification of methane emissions from 15 Danish landfills using the mobile tracer dispersion method

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ABSTRACT

Whole-site methane emissions from 15 Danish landfills were assessed using a mobile tracer dispersion method with either Fourier transform infrared spectroscopy (FTIR), using nitrous oxide as a tracer gas, or cavity ring-down spectrometry (CRDS), using acetylene as a tracer gas. The landfills were chosen to represent the different stages of the lifetime of a landfill, including open, active, and closed covered landfills, as well as those with and without gas extraction for utilisation or flaring. Measurements also included landfills with biocover for oxidizing any fugitive methane. Methane emission rates ranged from 2.6 to 60.8 kg h^{-1}, corresponding to 0.7–13.2 g m^{-2} d^{-1}, with the largest emission rates per area coming from landfills with malfunctioning gas extraction systems installed, and the smallest emission rates from landfills closed decades ago and landfills with an engineered biocover installed. Landfills with gas collection and recovery systems had a recovery efficiency of 41–81%. Landfills where shredder waste was deposited showed significant methane emissions, with the largest emission from newly deposited shredder waste. The average methane emission from the landfills was 154 tons y^{-1}. This average was obtained from a few measurement campaigns conducted at each of the 15 landfills and extrapolating to annual emissions requires more measurements. Assuming that these landfills are representative of the average Danish landfill, the total emission from Danish landfills were calculated at 20,600 tons y^{-1}, which is significantly lower than the 33,300 tons y^{-1} estimated for the national greenhouse gas inventory for 2011.

1. Introduction

The disposal of waste results in landfill gas generation consisting of methane and carbon dioxide. The global warming potential of methane seen over a 100-year cycle is 28 times higher than carbon dioxide (IPCC, 2013). Landfills have been found to be a significant source of methane generation and emissions, and it has been estimated that worldwide emissions from the waste sector accounted for 18% of the global anthropogenic methane emitted in 2004 (Bogner et al., 2008). The significant emission from landfills has long been known and led to the implementation of a landfill directive in the European Union that sets targets for phasing out the landfilling of organic material and other combustible waste (EC, 1999). In 1997, Denmark as the first country in the European Union implemented a ban on landfilling of organic waste. However, many landfill sites continue to generate methane throughout their lifetime due to the slow anaerobic decomposition of various organic materials, which are present in even very old and covered landfills. At some landfills, methane collection systems have been installed to collect a fraction of the generated gas and to use it for electricity and/or heat generation. At sites where there is no need for the electricity/heat or where the gas quality is too low (too low methane content) the methane is flared to avoid it from entering into the atmosphere. At landfill sites generating lower amounts of methane, an alternative mitigation approach is to design biocovers consisting of a top layer, which is optimized to oxidize the methane as it passes through the cover. However, despite gas collection and oxidation systems, a proportion of the methane will still escape into the atmosphere and contribute to greenhouse gas loads.

Following the Intergovernmental Panel on Climate Change (IPCC) guidelines (IPCC, 2006), estimates of methane emissions are required for national greenhouse gas inventories. The European Pollutant Release and Transfer Register (E-PRTR) protocol requires landfill operators receiving more than 10 tons of waste per day, or with a total disposal capacity above 25,000 tons, to report their methane emissions (CEC, 2006). As in many other countries, reporting methane emissions from landfilling in Denmark is based

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on modelling methane generation, by using input parameters such as waste quantity and composition. These models are not well-validated, though, and uncertainty surrounding these estimations can be significant, due to a lack of historical information about landfilled waste as well as changes in the waste stream after the ban on landfilling of combustible waste (Scharff and Jacobs, 2006). As a result, using existing landfill gas generation models will often lead to overestimations of the methane emission, as the models take a conservative approach regarding the organic content of the deposited waste (Scheutz et al., 2011b). Methane that is not collected by gas extraction systems usually escapes from weak areas in the landfill cover (slopes, cell intersections, crack/fissures, etc.), leachate collection systems or leaks in pipe systems (Fredenslund et al., 2010; Lewis et al., 2003; Scheutz et al., 2011b), and identifying and quantifying these hotspots is a challenging task. Emissions from hotspots may range between 0.0004 and 4000 g m^{-2} d^{-1} (Bogner et al., 1997) or 0–9.7 mol d^{-1} (Rachor et al., 2013), while emission areas can move or change over time due to seasonal changes, changes in short-term weather conditions or operational changes (Börjesson et al., 2000). Quantifying methane emission from whole landfill sites is amongst others important for evaluating removal efficiencies for installed gas extraction systems, which can only be done with an acceptable level of accuracy if the emission is independently determined. One approach for quantifying whole landfill site emission is using a mobile tracer dispersion method (Scheutz et al., 2011a,b,c; Mønster et al., 2014).

The objective of this study is to quantify methane emissions from landfills in Denmark, using the mobile tracer dispersion method. Emissions were quantified at old closed landfills, landfills partly or fully in operation, landfills with gas extraction systems and landfills employing biocovers to reduce the escape of methane. The measured methane emissions were normalised by considering the area of the landfill, the amount of waste received, the age of the waste and the type of aftercare. This was done in order to evaluate emissions from different sizes and ages of landfill and to suggest contributory factors, where possible. By measuring at landfills representing the diverse range of Danish sites, the goal was to obtain estimations of overall methane emissions and then compare these with official estimates.

2. Landfill site descriptions

Fifteen Danish landfills were chosen to represent all 134 registered sites in Denmark (Danish Centre for Environment and Energy (DCE), 2013). The landfills were geographically distributed throughout the country, and Table 1 summarises geographical location, age, size, waste amount, main waste types received, aftercare and onsite activities. The landfills studied included old and covered landfills without aftercare (Eskelund and Uggeløse Section 1) and with leachate collection as aftercare (Frederiksværk and Uggeløse Section 2). Also included were more recent, but closed, landfill sites with leachate collection (Fakse and Ærø) or both leachate collection and gas recovery (Hedeland and Viborg). Landfills still in operation included five landfills with leachate collection but no gas recovery (AV Miljø, Audebo, Klintholm, Skovsted and Skårup) and three with both leachate collection and gas recovery (Feltengård, Glatved and Odense). Two of the sites employed engineered biocovers for methane oxidation (Fakse and Klintholm). Nine of the landfills had on-site composting facilities (see Table 1), eight of which composted garden/park waste in open windrows, while some sites combined garden/park waste with source-separated organic household waste (Klintholm), sewage sludge (Fakse) and manure (Odense). One landfill (Audebo) also had an onsite plant for anaerobic digestion of source-separated organic household waste.

Information about the composition and amounts of deposited waste varied greatly between the landfills. At the older sites, only an estimated amount of mixed waste could be obtained by assessing the approximate volume of the landfill and assuming a waste density. Further information on waste amounts and composition was available for the more recent landfills, as listed in Table 1.

3. Material and methods

3.1. Dynamic plume measurement using mobile analytical platforms

Total landfill methane emissions were quantified using a mobile tracer dispersion method that combines a controlled release of tracer gas from the landfill with concentration measurements downwind of the landfill, by using a mobile high-resolution analytical instrument (Börjesson et al., 2009, 2007; Galle et al., 2001; Scheutz et al., 2011a,b,c). The method has been used successfully in the last few decades, and with new developments in analytical technology it has become a powerful tool for quantifying methane emissions from landfills. A number of studies have compared different methods for quantifying total landfill methane emissions, and the tracer dispersion method (mobile or stationary) has been ranked among the best, both in terms of quantifying the emissions and of the uncertainty on the single measurements (Babilotte et al., 2010; Green et al., 2010; Tregoures et al., 1999). The tracer dispersion method in general is based on the assumption that a tracer gas released at an emission source, in this case a landfill, will disperse into the atmosphere in the same way as methane emitted from the landfill. Assuming that the wind direction is defined, the conditions in the air above the landfill are sufficiently mixed for the methane and tracer gas to be fully mixed, and the tracer gas release and methane emission are constant, the methane emission rate (E_{gas}) can be calculated as a function of the ratio of the integrated cross-plume concentration of methane emitted to the integrated cross-plume concentration of the tracer gas, as follows:

$$E_{gas} = Q_{tracer} \cdot \frac{\int_{Plume\ end\ 1}^{Plume\ end\ 2} C_{gas} dx}{\int_{Plume\ end\ 1}^{Plume\ end\ 2} C_{tracer} dx} \cdot \frac{MW_{gas}}{MW_{tracer}} \tag{1}$$

where Q_{tracer} is the release rate of the tracer gas (kg h^{-1}), C_{gas} and C_{tracer} denote cross-plume concentrations (ppbv) above the background, MW denotes molecular weight and x corresponds to distance across the plume (between 400 and 2500 m for all cross-plumes in this paper).

The tracer dispersion method for landfills was first reported by Czepiel et al. (1996) and later further developed to measure gas concentrations in a total cross-section of the methane plume downwind of the landfill using vehicle mounted instrumentation (Galle et al., 2001; Scheutz et al., 2011a,b,c). Previously, the analytical approach for this method has used Fourier transform infrared spectroscopy (FTIR), but in the current study a novel analytical instrument was used, based on cavity ring-down spectroscopy (CRDS), which improves the time resolution and sensitivity of the measurements (Mønster et al., 2014). Both analytical approaches were used in this study, and Table 2 shows for which campaigns each approach was used. The downwind measurements were done at suitable roads near the landfills, and distances varied from site to site, depending on the accessibility of roads, the size of the landfills and other methane sources interfering with the results, such as composting facilities. The optimal distance for measuring a site's total emissions is between 1 and 2 km, depending on the topography of the site and weather conditions such as wind speed and sun, leading to a higher dispersion of the methane plume (Mønster et al., 2014). All quantifications, where possible, were made within this distance range. At some landfills, other methane sources (composting facilities or anaerobic digestion plants) were present, but