

Biocover - Whole landfill methane emission

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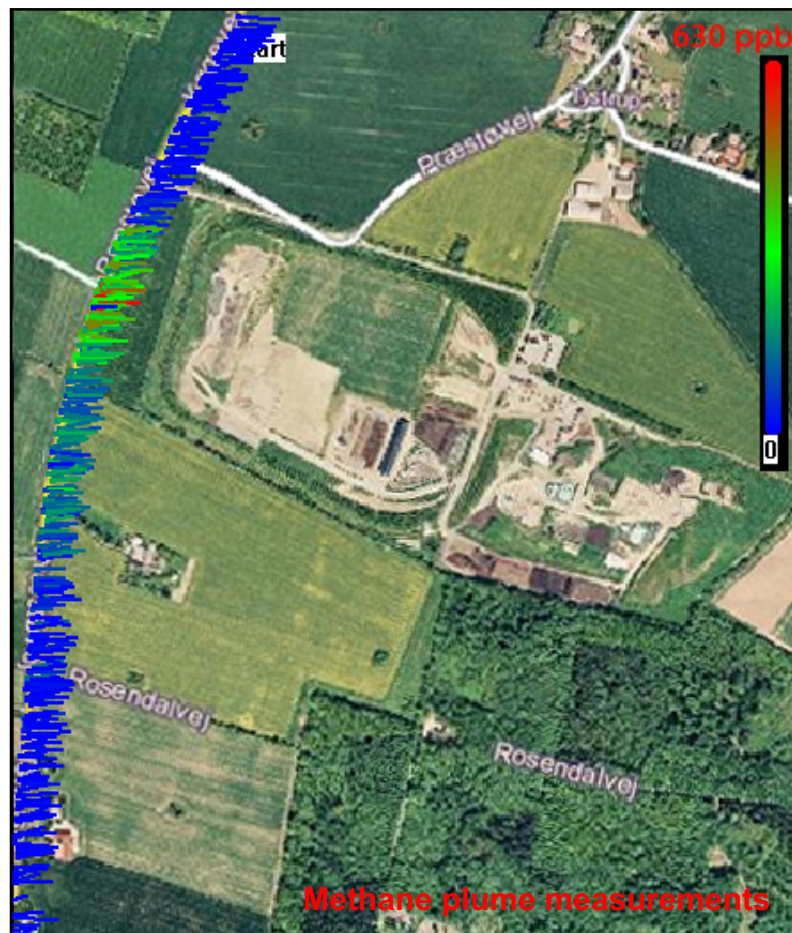
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BIOCOVER

Whole landfill methane emission



Institute of Environment & Resources
Technical University of Denmark

April 2007



INSTITUTE OF ENVIRONMENT & RESOURCES



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Preface

The full title of the BIOCOVER project is Reduction of Greenhouse Gas Emissions from Landfills by use of Engineered Biocovers. The project is funded by the LIFE III ENVIRONMENT programme, the Danish Environmental Protection Agency, and RENOSAM and runs from August 2005 to November 2008. This report presents the outcome of Action 3.2 Whole landfill methane emission (deliverable D.3.2.1b) as described in the project application (Biocover, 2005). Fakse Landfill serves as the demonstration landfill for the BIOCOVER project.

Action 3.2 was performed by E&R DTU in collaboration with the following partners:

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Summary

The whole methane emission from the disposal site was measured using a tracer technique, combining controlled tracer gas release from the landfill with time-resolved concentration measurements downwind the landfill using FTIR absorption spectroscopy. Initially to the release experiment a general leak search at the landfill was conducted with the main purpose to identify high emission areas for placement of the tracer release bottles. In parallel with dynamic plume measurements using the FTIR, a new and simpler method for whole landfills emission measurements was tested. This method is based on stationary sampling using evacuated canisters in the downwind plume. The advantage of this approach is that a sensitive on-site analyzer is not required. Instead the canisters can be sent to a certified laboratory for analysis. The drawback with this passive sampling approach is that there is no online information on the plume location and behavior during the sampling experiment. Furthermore the method has not been validated against other known emission measurement methods.

In all two field campaigns were performed; during October 11-12, 2006 and February 19-20, 2007. At both field campaigns an overall leak search showed that the methane emission from the old landfill section was localized to the leachate collection wells and some slope areas. In addition to the old landfill section a “new” methane source was identified west of a compost area close to the western border of new section of the landfill.

During the first campaign the methane emission from the old landfill section was estimated to be $31.2 \pm 6.8 \text{ kg CH}_4 \text{ h}^{-1}$, whereas the source at the new section was quantified to be $12.2 \pm 3.3 \text{ kg CH}_4 \text{ h}^{-1}$.

Measurements conducted at the compost area showed that no major fluxes of methane could be identified from the compost materials, and the overall methane emission was only $0.5 \text{ kg CH}_4 \text{ h}^{-1}$. The carbon dioxide flux from the compost material was determined to be $332 \text{ kg CO}_2 \text{ h}^{-1}$, corresponding to 91 kg h^{-1} of carbon being converted into CO_2 per hour. The nitrous oxide emission from the same compost was quantified to be $0.06 \text{ kg N}_2\text{O h}^{-1}$. A sludge pit located west of the compost area was quantified to have an emission of $2.4 \text{ kg CH}_4 \text{ h}^{-1}$, and $0.03 \text{ kg N}_2\text{O h}^{-1}$.

During the second campaign the methane emission from the old landfill section was estimated to be $30.5 \pm 2.2 \text{ kg CH}_4 \text{ h}^{-1}$, whereas the emission from the source at the new section was found to be $7.3 \pm 2.2 \text{ kg CH}_4 \text{ h}^{-1}$. Total facility traverses far downwind the site, showed an overall emission of $35.8 \pm 5.3 \text{ kg CH}_4 \text{ h}^{-1}$.

The new and simpler method for whole landfills emission measurements based on stationary sampling using evacuated canisters in the downwind plume gave an emission of $101 \text{ kg CH}_4 \text{ h}^{-1}$ from the landfill during the first field campaign, which is about 2½ times the emission found using the mobile FTIR-method ($43.4 \pm 7.5 \text{ kg CH}_4 \text{ h}^{-1}$). It is quite clear that the canister method overestimated the emission due to an unknown methane source very close to the road where the canisters were placed. In this situation, the tracer release did not simulate the methane emission very well, which is crucial for applying this method. During the second field campaign, the measured emission using the canisters ($39 \pm 5.0 \text{ kg CH}_4 \text{ h}^{-1}$) was very close to the emission measured with the mobile FTIR-method ($35.8 \pm 5.3 \text{ kg CH}_4 \text{ h}^{-1}$). Based on these results, the simpler canister method is promising for future emission measurements. However, this study also showed that a very good tracer simulation of the current methane emission as well as full coverage of the plume by the canisters is crucial for obtainment of accurate results.

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1 Introduction

Most landfills contain organic wastes which produce biogas, containing methane and carbon dioxide. Emission of methane from landfills is a serious environmental problem and is explicitly mentioned as a source for greenhouse gasses in the EU *Sixth Environmental Action Plan*. In a global perspective, landfills accounts for 7-20% of the anthropogenic methane emissions to the atmosphere.

Landfill gas (LFG) is at some landfills extracted and utilized for energy purposes leading to methane emission reduction. However, it is not always feasible to extract and utilize the landfill gas. In these cases the gas is flared with risk of producing toxic combustion products, or is just escaping to the atmosphere.

A low-cost alternative could be to improve the top covering of the landfill in order to optimize the biological methane oxidation in the cover. Laboratory experiments have documented that a very high methane oxidation rate can be obtained in bio-covers, thereby reducing the methane emission significantly. The biological methane oxidation transforms methane into carbon dioxide, and since methane has a 21 times stronger global warming potential than carbon dioxide, a significant reduction in the source to global warming is obtained. Biocovers may also be a very cost-effective supplementary method at landfills with landfill gas utilization, since the efficiency of the gas extraction system often is in the range of 50-60 %.

The BIOCOVER project has the objective to perform a full scale implementation of engineered bio-covers and to document the methane reduction efficiency. Fakse Landfill in Southern Zealand, Denmark, serves as a demonstration landfill for the implementation of the technology.

Fakse Landfill is divided into two sections. The oldest section which was in use from 1981 until 1997 will be the focus of the project activities. This part of the landfill has an area of 12 hectares and has received mixed waste. Approximately 600,000 tonnes of waste has in total been disposed of at the older part of the landfill. The landfill is typical for Danish landfills of similar age.

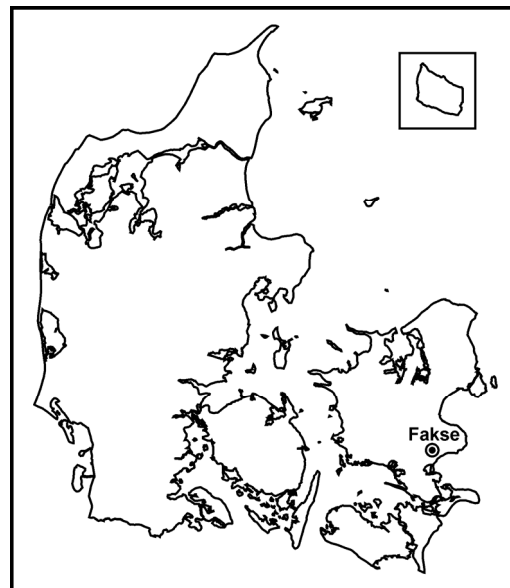


Figure 1. Map of Denmark showing the location of the study landfill, Fakse Landfill

This report concerns the baseline measurement of the whole landfill methane emission (Action 3.2). The main purpose of this report is to document the annual emission of methane (in tonnes year^{-1}) to be used as a baseline value in Task 6 in order to determine the oxidation efficiency of the installed biocover windows.

1 Objective

The overall objective of Action 3.2 was to determine the whole methane emission from the landfill. The expected result of Action 3.2 is an estimate of the annual emission of methane (in tones year⁻¹) to be used as a baseline value for comparison in Task 6. After installation of the biocover windows, the emission measurement will be repeated in order to determine the oxidation efficiency of the windows.

A secondary objective was to quantify the emissions from different areas and sources at the landfill. More precisely, the objective was to determine the emission from the two sections of the landfill (Section I: the old part of the landfill and Section II: the new part of the landfill) as well as the emission from a composting area and a sewage sludge storage unit.

The measurement campaign was carried out by E&R DTU in collaboration with FluxSense AB/Chalmers University of Technology using their automated method in comparison with a simpler manual method performed by E&R DTU using stationary sampling canisters. The stationary sampling canisters were provided by NV Afvalzorg, Netherlands, who also assisted in the field work. After sampling, the canisters were shipped for analysis to an analytical laboratory (ECN, Environmental Risk Assessment, Netherlands). The two methods were compared and evaluated. The advantages of using stationary sampling canisters is that the on-site use of advanced and expensive analytical equipment can be omitted reducing the cost of the field campaign significantly. Drawbacks of the method are the rather elaborate and time consuming sampling procedure as well as the fact, that data are not available in real time. For proper placement of the sampling canisters catching the total plume a rather detailed knowledge about emission sources and wind conditions is needed. Furthermore, the simpler method has not previously been validated why a comparison study is needed (Scharff and Jacobs, 2006).

The measuring times were chosen to represent stable weather conditions where the measured emission is believed to be representative for the whole landfill emission rate at the particular season. In all two field campaigns were performed; during October 11-12, 2006 and February 19-20, 2007.

2 Methodology and equipment

2.1 *Dynamic plume measurement using a mobile FTIR*

The very inhomogeneous surface gas emission from the site combined with several significant individual gas emission sources like the leachate collection wells prevented using traditional flux chambers for determination of the total landfill emission (Fredenslund et al., 2006). Instead the whole methane emission from the disposal site was measured using a tracer technique, combining controlled tracer gas release from the landfill with time-resolved concentration measurements downwind the landfill using FTIR (Fourier Transform Infrared) absorption spectroscopy (Galle et al., 2001). By the use of tracers, meteorological measurements and modeling can be omitted. In order to quantify the emission, tracer gas was released from the emitting areas, and the concentration ratio of the different gases relative to the emitted tracer was determined downwind the source. Both N₂O and CO were used as tracers, pinpointing different source areas. With the measured downwind concentrations expressed in a mixing ratio, M_T and M_M being molecular weights

of the respective gas, and knowing the emission rate of the tracer Q_T , the emission rate of the measured gas Q_M is obtained from:

$$Q_M = Q_T \cdot \frac{C_M \cdot M_M}{C_T \cdot M_T}$$

Provided that the tracer release simulates the source well a good estimate of the emission can be obtained.

Downwind concentration measurements of CH_4 , CO_2 , CO and N_2O were made with a mobile FTIR-instrument, operating with a time resolution of 21 seconds (Galle et al., 2001, Samuelsson et al., 2005). The FTIR technique used is an optical technique based on infrared absorption. Infrared light is transmitted a distance through a gas, and an absorption spectrum of the gas is recorded. The spectra of different gases are analyzed using multiregressing techniques e.g. CLS (Classical Least Squares). By the use of long optical path length, 96 m, sensitivities down to mixing ratios of a few ppbv are obtained. In the present application a medium resolution (1 cm^{-1}) FTIR spectrometer (Bomem MB104), connected to a closed multireflection cell (volume 12 L, optical path 96 m), was used (Galle et al., 2001). The concentrations were retrieved in the $3.1\text{-}5.5 \mu\text{m}$ wavelength range.

Initially to the plume measurements, the FTIR-instrument was driven around the site to identify potential leak sources. For determination of the whole site methane emission nitrous oxide was released as a tracer in four to five points at the site and concentrations measurements of methane and nitrous oxide was conducted in the downwind plume along a transect perpendicular to the wind direction. In order to determine both the emissions from the composting area and the sewage sludge storage pits carbon monoxide was used as tracer release at different time intervals from the different source areas. Depending on the wind direction the emissions from the two individual landfill waste sections (Section I and II) could be differentiated. The release rate of the tracers was determined setting the tracer flow rate from each tracer gas bottle with a two-stage regulator, clocking the release time, measuring the total weight of gas released with a precision scale, and also by integrating the number of liters released, as a backup. Figure 1A shows one of the N_2O tracer release units. Four to five N_2O tracers were used and one CO tracer. Figure 1B shows the measuring van with the FTIR built in, sampling air continuously from the roof. The position of the van was logged with GPS.



Figure 1. A: N_2O tracer release unit, including gas bottle, two-stage regulator, and volume integrator. B: The mobile FTIR instrumentation.

2.2 Stationary plume measurement using capillary sampling canisters

In order to evaluate the performance of the simpler stationary canister sampling technique, 14-15 canisters were placed for a 4 hour sampling period along the same transect where concentration measurements were performed using the mobile FTIR. The canisters sampled air continuously during the whole tracer experiment. To assure that the entire plume had been ‘caught’ the bottles at each far end of the plume needed to detect background concentration. One canister was located upwind of the landfill to obtain a background level. The canisters were standard refillable steel LP gas bottles (25 bar) (Sievert cylinder no. 2012, Belgium) equipped with M14 x 1.5 - BSP 3/8” LH cylinder valves (e.g. Sievert no. 700001, Belgium) (figure 2A). Before sampling the canisters were evacuated using a vacuum pump (Series Laboport N840 FT.18, N840.3 FT.18, KNF-lab, Netherlands) (figure 2B). The canisters were 4.8 liter in size and all filled up during sampling to 0.5 bar absolute pressure using critical capillaries. An absolute pressure of 0.5 bar assured a sufficient pressure difference for a continuous constant filling rate during the 4 hours sampling period.

The CH₄ and N₂O concentrations in the sampling canisters were shipped to a laboratory and analyzed by a QCL(Qunatum Cascade Laser)-Spectrometer (Aerodyne Research Inc., MA, USA) within 10 days after sampling. The average and standard deviation of the measurement were obtained from about 600 measurement obtained at 10 Hz for each sample. The QCL was calibrated after every 4-6 canister samples using 1700 and 5100 ppb CH₄ and 310 and 600 ppb N₂O. The QCL had a lower quantitative detection limit of app. 3 ppbv and 2 ppbv for methane and nitrous oxide respectively. The used QCL could not analyze CO₂ or CO like the FTIR.

Like for the dynamic plume measurements, the emission was calculated from the plume by integration of the excess concentration measured along the transect. This was done both for the N₂O and CH₄ plume. The methane emission is calculated from

$$Q_M = Q_T \cdot \frac{M_M \cdot \int C_M}{M_T \int C_T}$$

with $Q_T=2.7 \text{ gN}_2\text{O s}^{-1}$, $M_M=16$ and $M_T=44$.

A Gauss model was used to evaluate the N₂O-plume. Settings in the model were adjusted to reproduce the N₂O-plume. With the same settings the methane plume was calculated. With this procedure a best guess for the correction factor was obtained that accounts for the difference in source distribution for N₂O (four/five point sources) and CH₄ (the whole landfill).

On a high plateau in the center of the landfill wind speed, wind direction, air and soil temperature were measured with a meteorological unit. Barometric pressure was measured in the van.

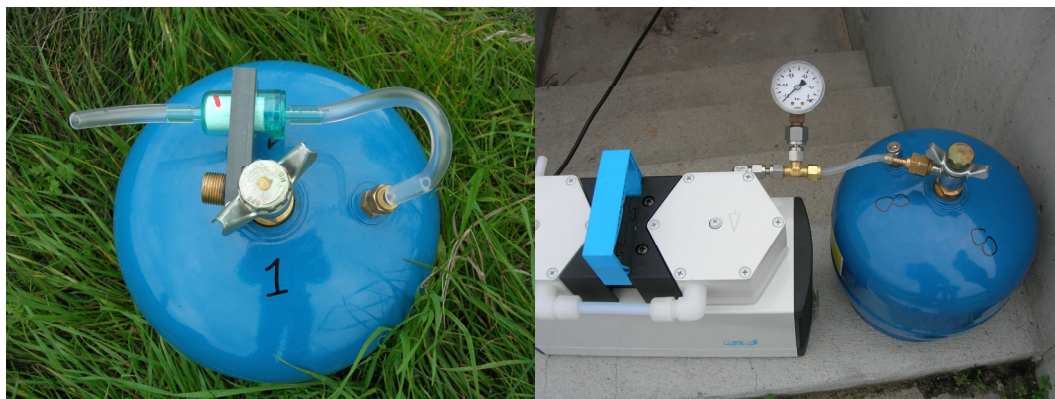


Figure 2. A: Canisters used for stationary sampling of downwind concentrations of methane and nitrous oxide. B: Vacuum pump used for evacuation of the canisters before sampling.

2.3 Tracer release and weather conditions during the two field campaigns

During the first field campaign tracer experiment with N_2O -release from 4 point sources on the old landfill was conducted between 12:00 and 16:30 on the 11th of October. In addition to this, a tracer experiment with CO release from one point source centered on a new methane source found at the new section of the landfill close to the main road in the western part of the landfill area was done between 15:30 and 16:30, and also between 17:10 to 18:00. A total N_2O tracer release rate of $9.7 \text{ kg } \text{N}_2\text{O } \text{h}^{-1}$ was used, whereas the release rate of CO was $2.1 \text{ kg CO } \text{h}^{-1}$. During the experiment the wind velocity was on average $4.4 \pm 0.7 \text{ m s}^{-1}$ (± 1 STD during the tracer experiment) coming from the east (87 ± 8 degrees). The weather was cloudy, and the air temperature was $16 \text{ }^\circ\text{C}$. During the 6 hour experiment the barometric pressure dropped linearly at a rate of $0.3 \text{ mbar per hour}$, going from 1015.7 mbar to 1013.7 mbar .

During the second field campaign a tracer experiment with N_2O -release from 4 point sources on the old landfill section and one point on the western source at the new landfill section, was conducted between 18:30 and 23:15 on the 19th of February 2007. A total N_2O tracer release rate of $11.6 \text{ kg } \text{N}_2\text{O } \text{h}^{-1}$ was used, with 80% released from the old landfill section, and 20% from the western source at the new landfill section. In addition to this, a CO tracer was centered also on the western methane source at the new section. The CO tracer release rate was $2.7 \text{ kg CO } \text{h}^{-1}$. During the experiment the wind velocity was on average 2.0 ms^{-1} coming from 252 degrees (WSW). The weather was cloudy, the air temperature was $1.9 \text{ }^\circ\text{C}$, and the barometric pressure was dropping with $0.2 \text{ mbar per hour}$, going from 1008.1 mbar to 1007.0 mbar during the 5 hour experiment. Plume measurements were conducted app. 1100 m downwind of the old section of the landfill, whereas the new section of the landfill was about 400 m upwind those measurements transects.

During the tracer release at both field campaigns, the plume from the landfill was continuously traversed from side to side at a driving speed of about 2.8 ms^{-1} . Each turning point was chosen observing the online evaluation, to make sure that the whole plume of methane and tracers respectively was traversed. For each traverse respectively plume was integrated over the distance traveled, and scaled directly with driving speed and the average wind speed for that traverse. The obtained value for methane was then divided with the corresponding value for the tracer, and multiplied with the mass flow of tracer.

3 Results and discussion

3.1 Field campaign October 2006

General leak search

Initially to the release experiment a general leak search at the landfill was conducted with the main purpose to identify high emission areas for placement of the N₂O-tracer release bottles.

Figure 3 and 4 display the result of a leak search over the Fakse landfill incl. the compost area, with data from both the 10th and 11th of October 2006. Presented values are the average concentration of methane in ppb during 21 seconds. Roughly each plotted concentration dot represents the average concentration of the plume in the area halfway from the dot before, up to halfway to the next dot. The size and color of the dot is related to the concentration, going from blue (clean) to red (max), as indicated by the color scale. The lines emerging from the color dots point up in the wind, towards potential leak areas.

The overall leak search showed that the methane emission from the old landfill part was quite localized to the leachate collection wells and some slope areas, verifying the results found earlier by E&R DTU (Fredenslund et al., 2006). In addition to the old landfill section a “new” methane source was identified west of the compost area at the soil/waste-masses located close to the western border of the landfill area, seen as green lines/dots in figure 3. This was unexpected as the previous gas production modeling had shown insignificant gas production at the new landfill section in comparison with the gas production from the old landfill section (Lemming, G. and Kjeldsen, P., 2006). However, in order to quantify the emission from the old section, where the biocover windows will be installed) the emission from the new source area the new landfill section had to be determined.



Figure 3. CH₄ leak survey with FTIR across the Fakse landfill and composting area on the 12th of October 2006. Color scale and dot size indicate concentration of methane in ppb, sampled 2 m above ground. The scale is linear from 0 to 4111 ppb. The colored lines are pointing up in the wind, and thus indicate potential leak source area.

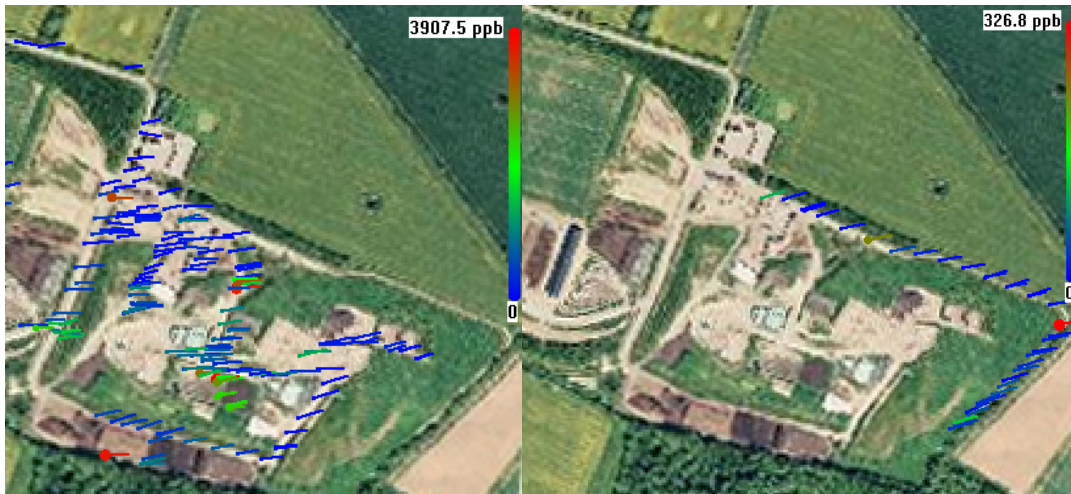


Figure 4. A: Methane concentration 2 m above ground for the central part of the landfill, with the scale going from 0-3908 ppb. B: Methane concentration 2 m above ground for the northeast corner and fence-line, but with the scale now from 0-327 ppb.

The whole landfill methane emission

Figure 5A and 5B show the concentration measurements downwind the landfill site. Table 1 summarizes the individual methane flux estimates. In table 1 three result columns are presented, reflecting the somewhat complex sampling situation, as seen in figure 3 and 4, and how that might affect the obtained result. The first estimate, named *best estimate*, was retrieved setting the border between the old landfill plume and the new source plume as good as possible based on wind direction, tracer plume and knowledge about the source distribution. Since the plume from the older part is much further away, and thus weaker, in comparison to the new source at the sampling position, it is difficult to decide exactly where the edge of the old landfill plume is. The *upper estimate* column in table 1, pushes the border used in the *best estimate* a bit more towards the north, possibly adding some of the new source plume for some of the traverses but making sure to include all of the old landfill plume.

The *best estimate* methane emission was $31.2 \pm 6.8 \text{ kg CH}_4 \text{ h}^{-1}$ ($\pm 1 \text{ STD}$, 27 traverses), while the *upper estimate* was $37.8 \pm 8.5 \text{ kg CH}_4 \text{ h}^{-1}$. This corresponds to $750 \pm 164 \text{ kg CH}_4 \text{ d}^{-1}$ and $907 \pm 205 \text{ kg CH}_4 \text{ d}^{-1}$ respectively.

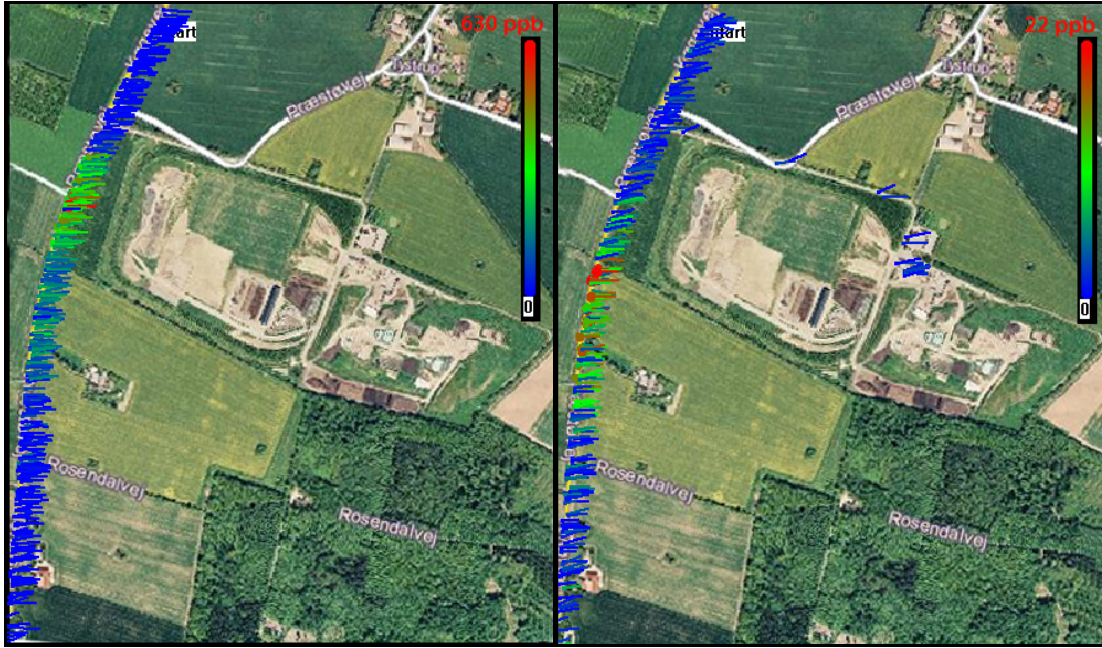


Figure 5. A: The CH₄-plume measured during multiple traverses downwind the landfill on the 11th of October 2006. B: The corresponding tracer plume (N₂O). The tracer plume matches the methane plume from the old part of the landfill, whereas a new methane source was found in soil and waste masses placed in the very west part, close to traverse road, giving rise to the highest concentrations due to the short dispersion distance.

Table 1. Methane emission from the old landfill section, section I.

Time (at center of plume)	CH ₄ emission, <i>best estimate</i> (kg h ⁻¹)	CH ₄ emission, <i>upper estimate</i> (kg h ⁻¹)	CH ₄ emission, <i>erroneous</i> <i>estimate</i> (kg h ⁻¹)
12:10	25.1	30.8	72.9
12:22	42.4	42.4	124.7
12:31	23.3	30.6	30.6
12:39	33.6	33.6	111.2
12:48	25.9	25.9	75.2
12:56	35.9	35.9	89.6
13:08	33.3	43.3	148.7
13:15	34.0	43.8	91.8
13:52	38.8	43.8	85.9
14:01	32.5	42.6	74.3
14:10	30.2	30.2	33.2
14:21	38.6	46.9	76.8
14:33	44.8	50.3	93.2
14:40	28.8	32.0	65.7
14:47	16.4	16.4	60.7
14:57	38.7	56.4	161.3
15:04	25.6	38.4	128.8
15:13	31.9	38.5	106.9
15:22	21.1	31.6	92.8
15:29	25.7	33.2	84.7
15:38	31.1	41.3	112.4
15:46	39.4	49.4	144.6
15:58	24.2	31.5	95.9
16:05	31.2	40.3	82.5
16:16	31.6	39.6	97.1
16:26	28.1	33.9	69.6
<i>Average</i>	<u>31.2</u>	<u>37.8</u>	<u>92.7</u>
<i>STD</i>	6.8	8.5	31.6
<i>STD/Average (%)</i>	21.8	22.6	34.1

Methane emission from the “new” source west of the compost area

In order to determine the methane emission from the “new” source located west of the compost area, and fairly close to the sampling road, a CO tracer was put in the center of the new source area. Between 15:55 and 17:35 seven traverses of methane and CO was conducted. Four of the traverses were done during the whole landfill emission experiment described in chapter 3, and three of them immediately afterwards. The resulting methane emission estimate from the close by source was 12.2 ± 3.3 kg CH₄ h⁻¹. The new source is about one fourth of the old landfill source. However, this source will severely affect the old landfill emission estimate if it is not taken into account that it is located much closer to the sampling road than the N₂O tracers placed on the old landfill.

Measurements at the compost area and the sludge pit

Leak search and quantification of localized emission sources of CH₄, N₂O and CO₂ was conducted at the composting area and a sludge pit at Fakse landfill. Figure 6A displays a picture of the compost materials, just being turned over. CO was used as a tracer in these experiments. Some CO might occur naturally from the compost environment due to oxidation processes, but this was observed to be negligible in comparison to the tracer signal (orders of magnitude). In general, low emissions of CH₄ was found at the compost facility, as seen in Figure 6B, and the methane plume emerging from the site was dominated by the landfill plume upwind of the compost area. The measurements at the

compost area were done fairly close to the source, and during a short time, with few repetitions. The uncertainty in the compost emission figures presented may therefore be as large as 50 %. Table 2 summarizes the results from the compost materials and the sludge pit.

Table 2. Emissions of methane, nitrous oxide and carbon dioxide from the compost area and the sludge pit.

Compound	Emission from the Sludge pit (kg h ⁻¹) (± 25 %)	Emission from the Compost materials (kg h ⁻¹) (± 50 %)
CH ₄	2.4	0.5
N ₂ O	0.03	0.06
CO ₂	-	332



Figure 6. A: Compost material being turned over. B: CH₄ leak search at the composting area at Fakse landfill 061011. As can be seen no major contribution to the methane plume comes from this part, as the concentration upwind and downwind is more or less the same.

A strong carbon dioxide plume was found at the compost materials, as seen in Figure 7. The derived CO₂ emission was 332 kg CO₂ h⁻¹. This corresponds to 91 kg carbon being emitted as CO₂ per hour. Table 2 summarizes the results from the compost area and the sludge pit close by. The detected sources of methane and nitrous oxide were small, and did not affect the landfill emission estimate significantly.



Figure 7. Concentrations of CO₂ at 2 m above ground around the compost area at Fakse landfill 061012. Color scale from 0 to 43 ppm CO₂ above ambient level.

A distinct methane source was found at the sludge pit, but the emission was only 2.4 kg CH₄ h⁻¹. The corresponding nitrous oxide emission from the sludge pit was 0.03 kg N₂O h⁻¹. The volume of stored sludge was about 40 m³, with an average depth of about 1 m. Figure 8A and 8B shows the correlation between the tracer CO and CH₄ and N₂O respectively, for the sludge pit measurements.

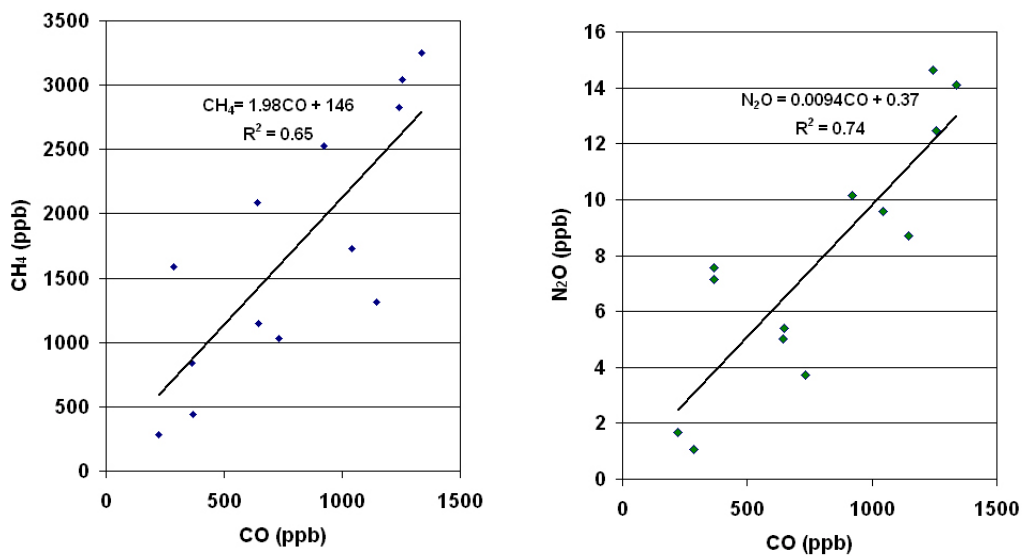


Figure 8. Tracer experiment with CO on the sludge pit at the compost area at Fakse landfill. The derived CH₄ emission (A) was 2.4 kg h⁻¹ and the N₂O emission (B) was 0.03 kg h⁻¹.

The whole landfill emission determined by the stationary canister method

Table 3 lists the concentrations measured in 12 canisters sampled along the downwind transect. Both the CH₄ and N₂O plumes were similar in shape as indicated by the calculated CH₄/N₂O-ratio listed in the 6th column of table 3.

Table 3. Concentrations of methane and nitrous oxide measured in 12 canisters sampled along the downwind transect.

Canister	Position East	Position North	CH ₄	N ₂ O	CH ₄ /N ₂ O-ratio	CH ₄ Above background	N ₂ O Above background
2	443	1184	2027	317	6	15	3
3	450	1153	2275	321	7	264	7
5	399	1054	2188	322	7	176	8
6	381	1011	2604	324	8	593	10
7	363	959	2488	324	8	476	10
8	349	912	2369	324	7	357	10
9	339	866	2418	332	7	406	18
10	314	770	2281	330	7	270	16
11	300	677	2806	344	8	795	30
12	291	584	2156	322	7	145	8
13	287	489	2170	315	7	159	1
14	284	331	2051	314	7	40	0
Background	1707	1082	2012	314	6		

Canister 1 was stolen during the field campaign. Canister 4 showed unrealistic results.

Table 4 lists the emission rates as determined by the stationary plume measurements using canisters. With the Gauss model an estimate for the N₂O emission of 2.9 g s⁻¹ was obtained, which is within 10% of the actual value of 2.7 g s⁻¹. For methane this model setting provides a total emission estimate of 30 g CH₄ s⁻¹ corresponding to 108 kg CH₄ h⁻¹. Correcting for the 10% difference shown with N₂O a final value of 28 g CH₄ s⁻¹ is obtained corresponding to 101 kg CH₄ h⁻¹. This is equal to the value obtained with direct ratio method.

Table 4. Whole emission as determined by the stationary plume measurements using canisters

Emission of N ₂ O	g N ₂ O s ⁻¹	kg N ₂ O h ⁻¹	Emission of CH ₄	g CH ₄ s ⁻¹	kg CH ₄ h ⁻¹
Reference N₂O	2.7	9.7	Direct ratio CH₄/N₂O	28	100.8
Model result	2.9	10.4	Model result	30	108.0
Model/Reference	109%	109%	Model corrected for N₂O	28	100.8

The estimated methane emission of 101 kg CH₄ h⁻¹ from the landfill with the canister method is clearly more than the 31.2 kg CH₄ h⁻¹ estimated by the FTIR measurements (or 43.4 kg CH₄ h⁻¹ including the western source at the new landfill section). This is explained by the “new” source located fairly close to the sampling positions, and much closer to the sampling position than the actual tracer release. The canister results treated the sampled methane molecules as originating from the same area as the tracer. This leads to an overestimation since the close by methane release is far less dispersed than the methane and tracer from the old landfill source. If the FTIR data were treated the same way, e.g. as if all

methane was released from the same area as the N₂O tracer gas, one would get an emission of 92.7±31.6 kg CH₄ h⁻¹, as given in the column *Erroneous estimate* in table 1. This is well comparable to the canister method result. The *erroneous estimate*, gives the estimated methane emission if one would not know about the close by methane source and thus anticipating that the methane from this source was released at the same location as the tracer, e.g. at the old landfill section. The *erroneous estimate* or the emission determined by the stationary canisters is almost three times higher than the *best estimate* measured by the FTIR measurements.

3.2 Field campaign February 2007

General leak search at the Fakse landfill on the 20th of February 2007

Figure 9 and 10 display the result of a methane leak search over the Fakse landfill and compost area, with data from the 20th of February 2007. The overall leak search showed that the methane emission from the old landfill part was quite localized to the leachate collection wells and some slope areas, verifying the results found in the October campaign and earlier emission measurements and surface screenings (Fredenslund et al., 2006).



Figure 9. CH₄ leak survey with FTIR across the Fakse landfill and composting area on the 20th of February 2007. Color scale and dot size indicate concentration of methane in ppb, sampled 2 m above ground. The scale is linear from 0 to 15500 ppb. The colored lines are pointing up in the wind, and thus indicate potential leak source area.



Figure 10. A: Methane concentration 2 m above ground for the central part of the landfill, with the scale going from 0-3290 ppb. B: Methane concentration 2 m above ground for the northeast corner and fence-line, but with the scale now from 0-13890 ppb.

The whole landfill methane emission

Table 5 summarizes the individual methane flux estimates from the old section, the new section and the whole landfill site, while figure 11 shows a comparison to the measurements done in October 2006.

The methane emission from the whole landfill area was estimated to be $30.5 \pm 2.2 \text{ kg CH}_4 \text{ h}^{-1}$ ($\pm 1 \text{ STD}$, 8 traverses), while the western source at the new section was found to emit $7.3 \pm 2.2 \text{ kg CH}_4 \text{ h}^{-1}$ (13 traverses). The sum of these two sources can be compared with the result from far downwind site traverses, integrating the emission from the whole site, as shown in Table 5. The result of $35.8 \pm 5.3 \text{ kg CH}_4 \text{ h}^{-1}$ (12 traverses) is well comparable to the two individual source areas, supporting the result from the leak search that methane emission from the compost area were negligible, compared to the emission from the other landfill areas. Compared to the emission survey in October 2006, the old landfill section emission was identical at this occasion, $30.5 \pm 2.2 \text{ kg CH}_4 \text{ h}^{-1}$ in February and $31.2 \pm 6.8 \text{ kg CH}_4 \text{ h}^{-1}$ in 2006.

Table 5. Methane emission (kg h^{-1}) from the old section, the new section and the whole landfill site based on 8, 13 and 12 plume transverses, respectively.

Old section		New section		The whole facility	
Time	Emission	Time	Emission	Time	Emission
18:42	28.6	18:37	4.7	18:54	30.9
19:20	27.7	19:38	4.8	19:00	33.4
19:47	29.2	20:35	5.8	19:07	39.4
20:29	31.7	20:47	9.0	19:14	31.5
21:27	29.1	20:55	6.3	19:56	31.1
21:34	31.8	21:00	6.5	20:07	27.6
22:15	32.0	21:06	9.0	20:15	40.0
22:34	34.1	21:11	10.6	20:21	39.5
		21:17	4.9	22:10	31.9
		22:20	5.9	22:44	38.7
		22:24	8.3	22:52	44.5
		22:30	10.7	23:00	41.0
		23:08	9.0		
Average	30.5	Average	7.3	Average	35.8
STD	2.2	STD	2.2	STD	5.3
STD/Average (%)	7.1	STD/Average (%)	30	STD/Average (%)	15

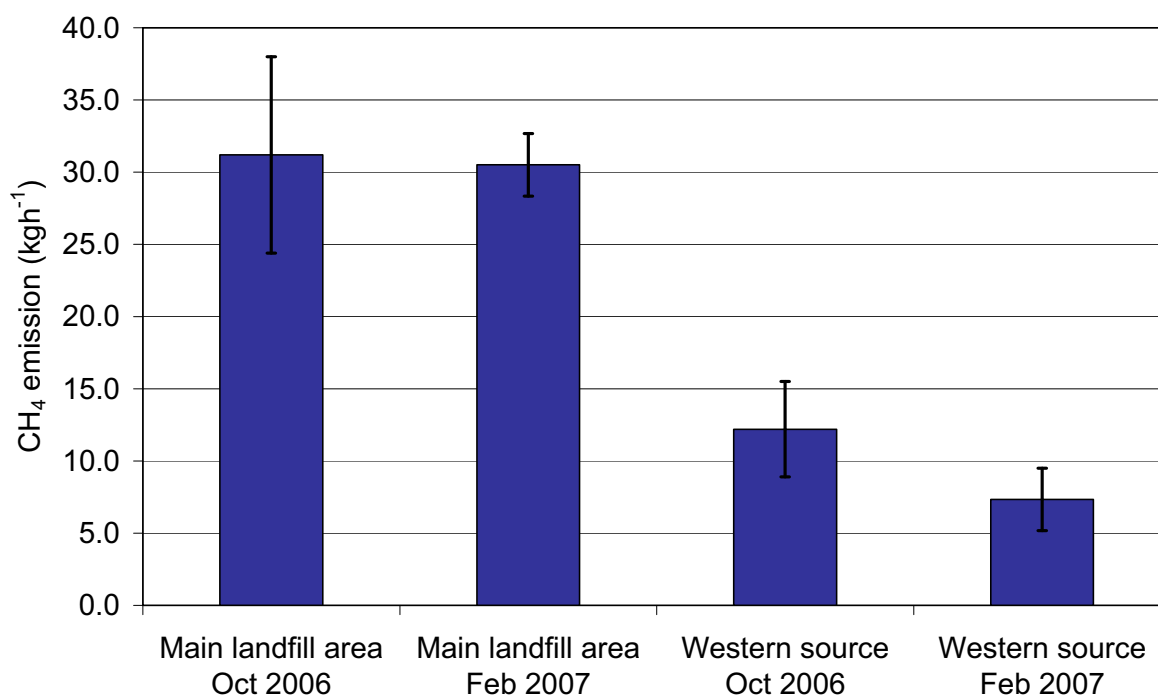


Figure 11. Comparison between methane emission measurements in February 2007 and October 2006. The error bars include variability in the emission itself, as well as in the measurement approach (± 1 STD).

Measurements at the compost area

Leak search of localized emission sources of CH_4 , N_2O and CO_2 was conducted at the composting area at Fakse landfill. CO was used as a tracer in these experiments for

quantification, where applicable. Some CO might occur naturally from the compost environment due to oxidation processes, but this was observed to be negligible in comparison to the tracer signal (orders of magnitude). In general low concentrations of CH₄ were found at the compost facility, as seen in Figure 12. Emission quantification was not possible since part of the source was too close to the sampling road. This was also the case for CO₂, as seen in figure 13. N₂O was more localized to the center of the composting area, see figure 14, and was quantified to be emitted at a rate less than 0.46±0.21 kg h⁻¹ (only two traverses, uncertain value).



Figure 12. CH₄ concentrations at the composting facility - scale from 0 to 1363 ppb above ambient concentrations.



Figure 13. CO₂ concentrations at the composting facility. Scale from the 0 to 54 ppm above ambient concentrations.



Figure 14. N₂O concentrations at the composting facility. Scale from the 0 to 91 ppb above ambient concentrations. N₂O compost emission quantified to be less than $0.46 \pm 0.21 \text{ kg h}^{-1}$.

The whole landfill emission determined by the stationary canister method

The preliminary results of the canister analyses are listed in table 6.

Table 6. Concentrations of methane and nitrous oxide (ppb) measured in 15 canisters sampled along the downwind transect.

Canister	Position East	Position North	CH ₄		N ₂ O		CH ₄ above background	N ₂ O above background
			avg	std	avg	std		
1	2174	618	3179	7	343	3	1048	21
2	2211	702	2259	5	347	3	129	24
3	2245	779	2281	4	354	9	151	32
4	2245	884	2251	3	348	0	121	26
5	2153	931	7252	51	402	5	5122	80
6	2067	965	2259	31	339	7	129	17
7	1968	1008	2332	26	345	3	202	23
8	1871	1036	2808	12	343	1	677	21
9	1786	1082	2227	0	341	0	97	19
10	1647	1079	2280	7	329	1	150	7
11	1552	1110	2130	0	341	1	0	19
12	1464	1141	2138	3	339	0	8	17
13	1363	1178	2230	6	345	2	100	23
14	1271	1206	2317	1	325	0	187	3
15	1151	1107	2189	3	334	1	58	12
Background	323	773	2263	9	322	2	132	0

Canister 5 showed unrealistic high concentrations why these results were disregarded.

A Gaussian dispersion model was used to evaluate the contribution of the different parts of the landfill to the measurement signal. The results showed that a part of the tracer plume was outside the measurement transect. This should be taken into account when calculating the emission from the landfill. This of course makes the valuation of the tracer plume more complex. Running the model using the measured wind direction (Wd) data (Wd average over 19:30-23:30=249 degrees) it seemed that the model predicted plume and the actual measurement have an offset indicating that the wind direction was off by about 10 degrees (259 instead of 249). The calculations show that with wind direction of 249 degrees 30 % of tracer released from point 2 is missed. With a wind direction of 259 this is reduced to 6 %. So depending on the wind direction, the source strength that should be included was 3.16 or 3.06 g N₂O s⁻¹ in comparison with the actual tracer release of 3.22 g N₂O s⁻¹.

Table 7 shows the calculated emission. All measurements (CH₄ and N₂O) and model runs were integrated along the transect using the distance between 2 sample locations multiplied with the average of the two concentration levels. For this purpose either the direct distance of the distance perpendicular to the wind direction can be used. It turned out that either way the same emission estimates were obtained. The emission calculation was done in three different ways.

1. The direct way dividing the integrated measurements of CH₄ and N₂O
2. By use of a dispersion model and the measurements for methane
3. By use of a dispersion model that also corrects for the ratio between measured and modeled N₂O

For the specific data set several extra choices can be made:

- A correction can be made for a part of the N₂O peak that is not covered with the measurements.
- Different meteorological settings in the dispersion model (stability class D (neutral) or E (stable)) and a possible shift in the wind direction (249 vs. 259 degrees).

The results of all combinations are shown in Table 7. The most likely emission factor is obtained with Wd=249 degrees correcting for missing 30% of one of the N₂O tracers and run the model for stability class E (nighttime stable conditions) indicated in gray in the table. The whole site emission is then $11 \pm 1.5 \text{ gCH}_4 \text{ s}^{-1}$ corresponding to $39.6 \pm 5.0 \text{ kgCH}_4 \text{ h}^{-1}$. The other calculations show that in general adjusting the wind direction 10 degrees have little effect on the result apart from when the calculations are done using only the model for CH₄ without evaluating the N₂O tracer release, which gives a 40 % lower estimate.

Table 7. Whole landfill emission as determined by the stationary plume measurements using canisters. For calculation of the emission three different integration methods were used.

Measure		Integral ppb*m	±Std	±Std (%)		
N ₂ O		27395	2943	11		
CH ₄		270860	12091	4		
Direct	QN ₂ O (g s ⁻¹)	QCH ₄ (g s ⁻¹)	±Std	QCH ₄ (kg h ⁻¹)	±Std	
N ₂ O source	All	3.21	12	1.6	43.2	5.7
	Wd 249 miss 30%	3.05	11	1.4	39.6	5.0
	Wd 259 miss 6%	3.16	11	1.5	39.6	5.4
Model only		Integral CH ₄				
	Stability D Wd 249	24921	11	0.5	39.6	1.8
	Stability E Wd 249	43416	6	0.3	21.6	1.1
	Stability E Wd 259	33278	8	0.4	28.8	1.4
Model + N ₂ O correction		Integral N ₂ O with Q = 3.21g s ⁻¹				
	Stability D Wd 249	28193	11	1.1	39.6	4.0
	Stability E Wd 249	48341	11	1.1	39.6	4.0
	Stability E Wd 259	45194	13	1.4	46.8	5.0

Table 8 provides an overview of the emission measured during the two field campaigns using the two different methods. The measured emission using the canisters during the first field campaign ($101 \text{ kg CH}_4 \text{ h}^{-1}$) is about 2½ times the emission found using the mobile FTIR-method ($43.4 \pm 7.5 \text{ kg CH}_4 \text{ h}^{-1}$). It is quite clear that the canister method overestimates the emission due to the unknown methane source very close to the road where the canisters were placed. In this situation, the tracer release did not simulate the methane emission very well, which is crucial for applying this method. During the second field campaign the measured emission using the canisters ($39 \pm 5.0 \text{ kg CH}_4 \text{ h}^{-1}$) is very close to the emission measured with the mobile FTIR-method ($35.8 \pm 5.3 \text{ kg CH}_4 \text{ h}^{-1}$). Based on these results, the

simpler canister method is promising for future emission measurements. However, this study also showed that a very good tracer simulation of the current emission as well as full coverage of the plume is crucial for obtainment of accurate results.

Table 8. Whole landfill emission (kg h^{-1}) as determined using two different methods; a dynamic plume measurements method using FTIR and a stationary plume measurement method using canisters.

Field campaign	October		February	
	FTIR	Canister	FTIR	Canister
Whole landfill emission	43.4±7.5	101	35.8±5.3	39.6 ± 5.0
Old landfill section	31.2±6.8		30.5±2.2	
New landfill section	12.2±3.3		7.3±2.2	

The total gas production for 2005 from the whole site was estimated using three gas production models to 25-52 $\text{kg CH}_4 \text{ h}^{-1}$ (range using three different gas production models) (Lemming & Kjeldsen, 2006). The gas production from the two landfill sections was estimated to between 23-49 and 1.5-13 $\text{kg CH}_4 \text{ h}^{-1}$ for the old and the new section respectively. At the old section the total emission was assessed to be 22 $\text{kg CH}_4 \text{ h}^{-1}$ (Fredenslund et. al, 2006). This value was established using a tracer release technique (emission through leachate collection and recirculation systems) and flux chamber measurements (emission through soil cover). From this study it was concluded that the methane emission at the time of measurement occurred primarily through the leachate collection system (65 %), while methane emission through the soil cover was focused in emission “hot spots”, which were located on slopes of the low permeable soil cover used at the site. It was also concluded that methane oxidation in the soil cover was negligible at the time of measurement (Fredenslund et. al, 2006).

In comparison the whole landfill emission based on downwind plume measurements shows is within the range of the total gas production predicted by the models. This is the case for both the total site as well as for the old section of the landfill. The measured gas emission from the old section of the site is higher than the emission measured from the leachate collection wells and the surfaces from the old section (22 $\text{kg CH}_4 \text{ h}^{-1}$). It is likely that the assessed emission based on measured surface emission and measurements from the leachate system is underestimated due to the high spatial variation of surface emissions and the high dynamic of the gas flow in the leachate system making the emission measurement more uncertain. Also measurements of local methane emissions focused on the most important sources; the leachate collection system and the “hot spots” on the soil cover. More diffuse emissions were not quantified, and thus an underestimation of the actual total emission in this study is likely.

The measured emission from the new section was in the higher end (7.3 – 12.2 $\text{kg CH}_4 \text{ h}^{-1}$) of what was expected based on the gas models (1.5-13 $\text{kg CH}_4 \text{ h}^{-1}$). As the emission from the new section contributes significantly to the total emission it is important that the emission from this source is determined also in the future measuring campaigns in order to determine the efficiency of the installed biocover windows on the old section.

4 Conclusion

Based on the two conducted field campaigns the following can be concluded:

At both field campaigns the overall leak search showed that the methane emission from the old landfill section was localized to the leachate collection wells and some slope areas. In addition to the old landfill section a “new” methane source was identified west of the compost area at the soil/waste-masses located close to the western border of new section of the landfill.

During the first campaign the methane emission from the old landfill section was estimated to be $31.2 \pm 6.8 \text{ kg CH}_4 \text{ h}^{-1}$, whereas the source at the new section was quantified to be $12.2 \pm 3.3 \text{ kg CH}_4 \text{ h}^{-1}$.

Measurements conducted at the compost area showed that no major fluxes of methane could be identified from the compost materials, and the overall emission was only 0.5 kg h^{-1} . The carbon dioxide flux from the compost material was determined to be $332 \text{ kg CO}_2 \text{ h}^{-1}$, corresponding to 91 kg h^{-1} of carbon being converted into CO_2 per hour. The N_2O emission from the same material was quantified to be $0.06 \text{ kg N}_2\text{O h}^{-1}$. The sludge pit located west of the compost material was quantified to have an emission of $2.4 \text{ kg h}^{-1} \text{ CH}_4$, and $0.03 \text{ kg h}^{-1} \text{ N}_2\text{O}$.

During the second campaign the methane emission from the old landfill section was estimated to be $30.5 \pm 2.2 \text{ kg CH}_4 \text{ h}^{-1}$, whereas the source at the new section was found to be $7.3 \pm 2.2 \text{ kg CH}_4 \text{ h}^{-1}$. Total facility traverses far downwind the site, showed an overall emission of $35.8 \pm 5.3 \text{ kg CH}_4 \text{ h}^{-1}$.

In parallel with dynamic plume measurements using the FTIR, a new and simpler method for whole landfills emission measurements was tested. This method is based on stationary sampling using evacuated canisters in the downwind plume. Using this method an emission of $101 \text{ kg CH}_4 \text{ h}^{-1}$ from the landfill was found during the first field campaign where as the second field campaign showed a total emission between $39 \pm 5.0 \text{ kg CH}_4 \text{ h}^{-1}$. During the first field campaign the canister method overestimated the emission due to an unknown methane source very close to the road where the canisters were placed. In this situation the tracer release did not simulate the methane emission very well leading to erroneous results.

During the second field campaign a very nice fit between the measured emissions using the two methods were obtained. For the simpler canister method to be replaceable with the mobile FTIR-method a very detailed knowledge about on-site emission sources is required in order to obtain a good tracer simulation. Proper placement of the canisters is also crucial in order to cover the whole plume from the landfill. Without on-site measurements available this can be a challenge. Gas dispersion models can support the interpretation of the measured data based on the canister samples. However, this requires accurate measurements of wind directions etc. Based on the comparison study of the two methods it is concluded that the canister method has a potential for future whole site emission measurement. However, due to the limited experience with the method and the complex emission pattern observed at Fakse landfill the following field campaigns after installation of biocover windows will be performed using the mobile FTIR-method.

5 References

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