Chapter 5 Methane emission estimates from landfills obtained with dynamic plume measurements1

A. Hensen[1] and H. Scharff[2]
[1] ECN, Petten, the Netherlands
[2] Afvalzorg Deponie, Haarlem, The Netherlands

Abstract.
Methane emissions from 3 different landfills in the Netherlands were estimated using a mobile Tuneable Diode Laser system (TDL). The methane concentration in the cross section of the plume is measured downwind of the source on a transect perpendicular to the wind direction. A Gaussian plume model was used to simulate the concentration levels at the transect. The emission from the source is calculated from the measured and modelled concentration levels. Calibration of the plume dispersion model is done using a tracer (N_20) that is released from the landfill and measured simultaneously with the TDL system. The emission estimates for the different locations ranged from 3.6 to 16 m^3 ha^{-1} hr^{-1} for the different sites. The emission levels were compared to emission estimates based on the landfill gas production models. This comparison suggests oxidation rates that are up to 50% in spring and negligible in November. At one of the three sites measurements were performed in campaigns in 3 consecutive years. Comparison of the emission levels in the first and second year showed a reduction of the methane emission of about 50% due to implementation of a gas extraction system. From the second to the third year emissions increased by a factor of 4 due to new landfilling. Furthermore measurements were performed in winter when oxidation efficiency was reduced. This paper describes the measurement technique used, and discusses the results of the experimental sessions that were performed.

Keywords: landfills, methane emissions plume dispersion tuneable diode laser

[1] Reference: Hensen, A., Scharff, H., Methane emission estimates from landfills obtained with dynamic plume measurements, Water,Air and Soil pollution, Kluwer, focus1: 455-464, 2001.

5.1 Introduction

In the Kyoto protocol, the Netherlands committed themselves to a substantial reduction of greenhouse gas emissions. Within Europe the Netherlands have the highest emission density for methane of about 24 ton[2] CH_4 per km^2 per year. The Dutch 1997 emissions of methane from landfills of about 445 ktonne (Coenen *et al.*, 2000) counts for about 7% of the total national emission level in CO_2 equivalents. As a result of the current waste policy, methane emissions are expected to have decreased by a factor of 3 by 2010, thus contributing significantly to the total Dutch effort to reduce greenhouse gas emissions. Since landfills play such an important role in the Dutch effort to meet the Kyoto target, this source should be well monitored.

The mass balance of landfill gas is shown in Figure 5.1. The production inside the landfill is estimated from the total volume, composition and age of the stored waste. A part of the produced gas can be recovered using extraction systems, the remaining methane escapes to the atmosphere or is oxidised on its way out of the landfill body. In the emission inventories the oxidation level is set to 10% as an IPCC default, but in fact oxidation is both variable in space and time (Czepiel *et al.*, 1996).

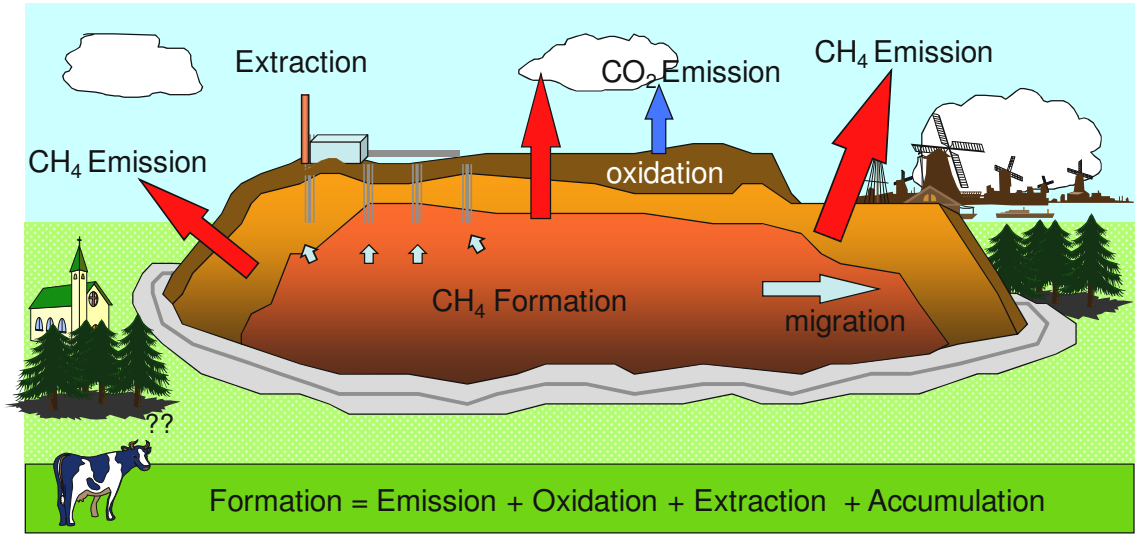

Figure 5.1 Methane emission as a function of production, recovery and oxidation.

Emission reduction at landfill sites can be achieved using a gas recovery system or by improving the oxidizing capacity of the landfill cover. Apart from the CH_4 contribution to global warming, other landfill gas components can cause local environmental problems. Odorous emissions in particular can be an incentive to implement emission reduction measures.

5.2 Materials and Methods

5.2.1 Description of the measurement sited and methodology

Measurements were performed at Nauema (52°27'N, 4°45'E), a 72 ha landfill situated 15 km west of Amsterdam, at Hollandse Brug (52°19'N, 5°08'E), a 27 ha landfill situated 20 km south east of Amsterdam and at Braambergen (52°20'N,5°16'E) near the

[2] There was an error in the original paper with the unit kg instead of ton.

city of Almere. Characteristics of the locations are listed in Table 5.1. All these landfills are located in flat terrain at a height of 15-20 m.

Downwind of the landfills, the CH_4 concentration was measured on a transect through the plume that originates from the source. This was done using a Tuneable Diode Laser absorption spectrometer (TDLAS, Aerodyne Res. Inc. Billerica, Mass. U.S.A.) mounted in a van. Absorption of the light emitted from the diode laser occurs in a 30 m astigmatic multipass cell. The 1270 and 1271 cm^{-1} absorption lines are used for methane and nitrous oxide respectively. Ambient air is sampled from the roof of the van and lead through the cell. Each plume transect takes approximately 5-7 min. Figure 5.2 shows the measurement method. The TDL showed a 10 ppb resolution for CH_4 and 20 ppb for N_2O. In these campaigns 1 Hz concentration data was used. Calibration took place while driving, applying standards before and after each transect measurement. The position of the van was obtained using a GPS system (Survey II, Garmin Inc.).

Table 5.1 Overview of the measurement locations

Location	Waste Characteristics	Area (ha)	C-Content (kg/tonne)	Estimated formation in 2010 (Mm^3)
Hollandse Brug	Demolition waste	15	70	0.87
Nauerna	Polluted soil, industrial waste Sludge	60	50	5.25
Braambergen	Polluted soil, Domestic,industrial and demolition waste	30	80	3.5

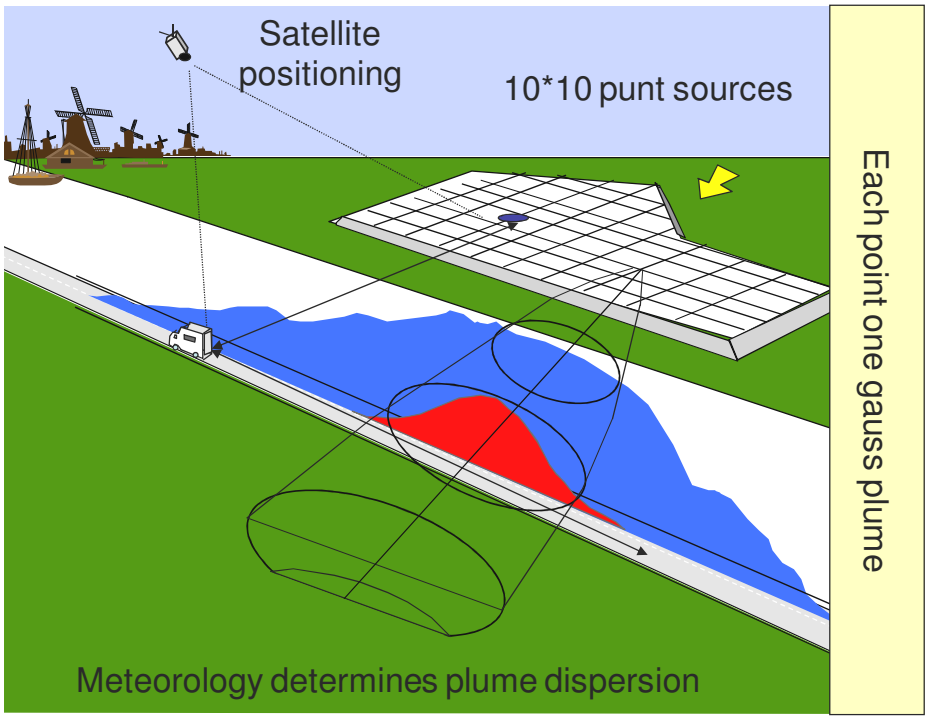

Figure 5.2 Measurement method, using the methane plume downwind of the landfill

5.2.2 Emission modelling

The measured concentrations in the plume transects were compared with the output of a multiple gauss plume model. This model uses a grid of 10 x 10 points over the landfill (Figure 5.2). For each grid point of the map a Gaussian plume is used, taking reflection of the plume at the ground level into account. No correction was applied for reflection at an inversion layer, the distance between source and receptor is not more than 2000 m and at that distance the effect of an inversion layer is not expected in daytime. For each source-receptor combination the receptor concentration is obtained:

$$C(X,Y,z) = \frac{Q}{2\pi \cdot u \cdot . \sigma_y \cdot \sigma_z} \cdot e^{-Y^2/(2\cdot\sigma_y)^2} \cdot \left(e^{-(z-h_s)^2/(2\cdot\sigma_z)^2} + e^{-(z+h_s)^2/(2\cdot\sigma_z)^2} \right)$$

with (2)

$$\sigma_y = A \cdot X^B \cdot z_o^{0.2} \cdot T^{0.35}$$

$$\sigma_z = C \cdot X^D \cdot (10 \cdot z_o)^{0.53\cdot E}$$

$$E = X^{-0.22}$$

Where X is the distance along the plume axis, Y the axis perpendicular to the plume axis, z the height above ground level, Q the source strength, u the wind speed measured on top of the landfill, and h_s the height of the emission (top of the landfill). σ_y and σ_z are dispersion parameters that depend on distance to the source, on the degree of turbulence of the atmosphere, the roughness length of the surface z_o, and on the timescale T used for averaging. A, B, C and D are dependent on the stability class (Pasquill, 1974).

The differences in source strength of the different parts of the landfill are accounted for by using a weighting index on different parts of the 10 x 10 matrix. Using this index map the total emission Q is distributed over landfill. The emission strength of the landfill is equal to the source strength needed in the model to obtain an agreement between the integral of the concentration along the transect for the modelled and the measured plume. In general, there are two extremes to consider when choosing the matrix. A minimum estimate for the emission strength is obtained when all sources are close to the measurement transect. With sources far away, a higher total source strength is needed to obtain the measured concentration levels at the plume transect. The shape of the modelled curve will show more spatial structure with sources close to the transect. The best estimate will be in between these two levels with source locations chosen to obtain a similar structure for the model and measured plume. Tests with individual plumes for the Nauema site showed that the minimum and maximum levels show a range of ±15% compared to the average level.

The meteorological data (heatflux, wind speed, cloud cover) indicate the Pasquill stability class, but a check on this choice is recommended. Therefore N_2O was released from a gas flask on top of the landfill and the TDL measured the N_2O and CH_4 plume simultaneously. For N_2O source, bath position and source strength are known, and the horizontal dispersion σ_y is obtained. The model calculation for this plume was used to check and if necessary to adapt, the dispersion parameters.

The uncertainty in the CH_4 concentration measurements is about 1-5% due to instrument noise, drift of the laser and uncertainty in the background concentration level. Changes in wind direction (on a timescale of 5-10 min) were the main cause for variation in the set of emission estimates. Furthermore, the emission of the landfill site will

not be constant with time and in order to obtain an annual average emission level, more measurements must be taken over the year.

5.2.3 The landfill gas production model

The landfill gas production model is based on a multi-phase first order degradation model used extensively in this field. In this case the model as described by the Dutch Landfill Gas Advisory Centre (Scheepers, 1994) was used. This model and its different parameters were optimised and validated on the basis of results from nine Dutch landfill gas projects (Coops et al., 1995).

Table 5.2 Information on the measurement campaigns

Location	Date	Wind speed ($m\ s^{-1}$)	Wind direction	Stability Class	Cloud cover	Distance to transect (m)	No of transects
Hollandse Brug	09/09/97	3	NE	B	7/8	750	7
Nauerna	15/04/97	7	N	D/C	2/8	780	8
	04/05/98	5	N	B	2/8	780	14
	19/11/99	4-6	N	D/E	6/8	780	10
Braambergen	10/11/99	5-4	NE	D	5/8	1000	13

Table 5.3 Emission levels during the measurement campaigns

Location	Year	No of transects	Emission $gCH_4.s^{-1}$	Landfill area (ha)	Remarks
Hollandse Brug	1997	8	11±4	15	No gas extraction
Nauerna	1997	7	62±12	60	No gas extraction
	1998	11	31±13	60	Gas extraction at 20 ha
	1999	10	109±33	60	Gas extraction at 20 ha
Braambergen	1999	13	48±8	30	Gas extraction at 14 ha

5.3 Results

Five campaigns were carried out in total at the three sites. Table 2 summarises the measurement conditions. Table 3 shows the emission levels that were derived for the different sites.

5.3.1 Experiments at Nauerna

Both the experiments in 1997 and in 1998 took place in spring. In 1997 the average windspeed was 7 m s^{-1}. Stability class D and z_o = 0.1 were used. During the experiment in 1998 the wind speed was lower (about 3 $m.s^{-1}$) and the wind direction was more variable. Stability class B and a roughness length z_o of 0.10 m were used in the model. Figure 5.3 shows an example of a plume transect as obtained at Nauerna in 1998. The line shows the measured excess concentration. The markers indicate the model calculation. For this plume the emission was estimated to be 70 $gCH_4\ s^{-1}$.

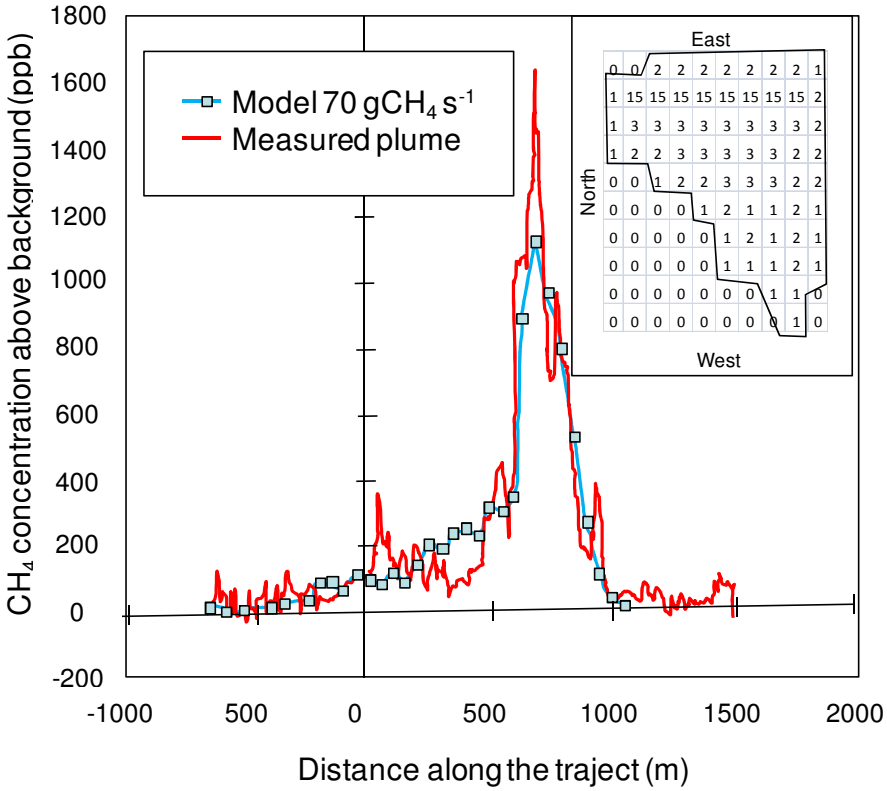

Figure 5.3 *Example of a transect measurement, the line shows the measured concentration, the markers the modelled concentration along the measurement transect.*

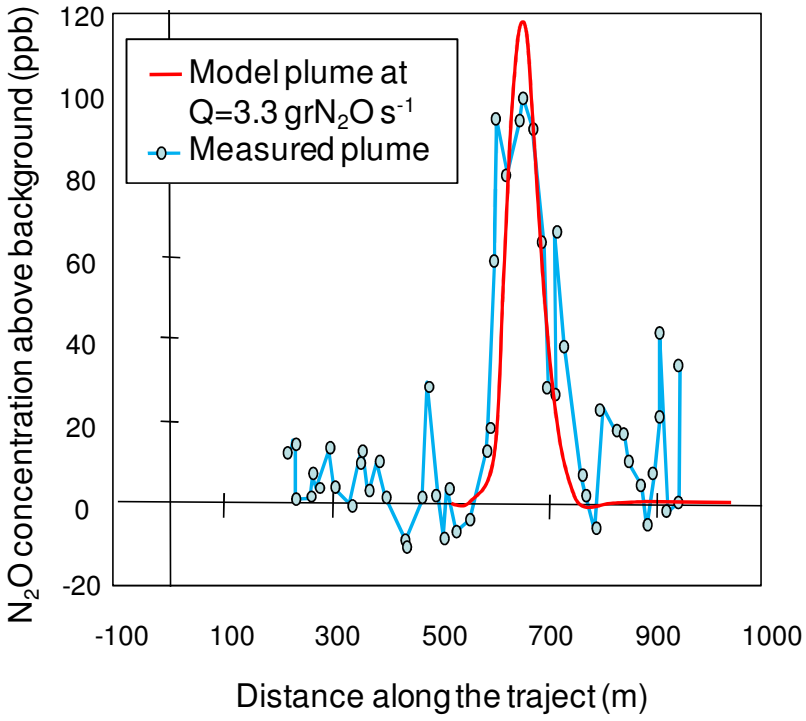

Figure 5.4 *The N_2O plume measured at the transect together with the modelled plume.*

The matrix in the graph shows the source distribution used. In April 1997 8 plume measurements were obtained. One of the plumes was rejected because it showed an emission level that was a factor of two above the average of the other plumes. The average emission level obtained using the 7 plumes was 62 ± 12 $gCH_4\ s^{-1}$. One year later, after the implementation of a gas extraction system on about 20 ha of the landfill, 11 plume transects were obtained. For this experiment the estimated emissions ranged from 19 g $CH_4\ s^{-1}$ to 40 g $CH_4\ s^{-1}$ with one outlier of 65 g $CH_4\ s^{-1}$ The average emission level was 31 ± 13 g $CH_4\ s^{-1}$.

In 1999 the average emission level was significantly higher 109 ± 30 g $CH_4\ s^{-1}$. Most of this emission originated from an area on the landfill where relatively fresh material was landfilled. Measurements look place in November at a temperature of about 2 °C, which will have reduced the oxidation efficiency of the landfill cover. The N_2O plume indicated that stability class E and roughness length z_o = 0.5 m give the best results for the model calculations. The N_2O release in 1999 was 8-10 higher than in 1998 which resulted in a peak concentration of 800 ppb and an improved S/N ratio compared to 1997 and 1998 (Figure 5.4). The standard deviation in the set of ten emission estimates was relatively large. Such large variation might have arisen due to the construction of a harbour, which meant that part of the measurement transect had to be carried out on a road parallel to the plume axis.

5.3.2 Experiments at Hollandse Brug

An old part of the landfill of about 7 ha is used as a recreation area. Because of the shape of the landfill, the south west part of the site was only 200 m away from the measurement transect, whereas the north east part was at a distance of 1500 m upwind. For a set of 7 transects a net emission level of 11 ± 4 $gCH_4\ s^{-1}$ was obtained. Meteorological data obtained on the site could not be used to derive stability due to disturbances from wind along the landfill slope. Stability data were obtained from a meteo station 25 km upwind of the site. This induces an extra uncertainty, which is reflected in the standard deviation reported above.

5.3.3 Experiments at Braambergen

At Braambergen, measurements took place on a road at about 1 km southwest of the landfill. Windspeed decreased during the experiment from about 6 to 3.5 m s^{-1}. Stability class D in combination with a relatively large z_o of 0.3 m showed the best agreement for the N_2O plumes. The plume data were corrected for the contribution of a dairy farm along this road that also showed a clear methane emission of about 1.8 g $CH_4\ s^{-1}$.

5.3.4 Comparison between measured emission levels and production estimates

The emission levels obtained from mobile measurements were compared with those based on the landfill gas production models. Gas extraction levels are monitored continuously (Table 5.4). In theory the sum of recovered and emitted methane must be below the production level estimate. The difference between these two levels is an estimate of the oxidation.

The gas production model predicted a methane production of 110 ± 10 $m^3\ CH_4\ hr^{-1}$ at Hollandse Brug in 1997. The measured emission is equivalent to about half that value. This also happened at Nauerna in 1997 where the gas production model predicted a methane production of 590 ± 60 $m^3 CH_4\ hr^{-1}$. In 1998 the gas production estimate at Nauerna decreased to 540 ± 54 $m^3\ CH_4\ hr^{-1}$.

This decrease, combined with the gas extraction system that was in operation lead to a 50% emission reduction compared to the 1997 level. In both years the oxidation levels during the experiments must have been 50-60% in order to account for the relatively low emission levels. This is high compared to the 10% IPCC default value. It was a surprise to find that the gas extraction system on 20% of the landfill reduced the emission level by 50%. We assume this is caused by an increase in oxidation efficiency. Also under-pressure induced by the gas extraction will reduce gas emission through cracks in the top cover.

The 1999 production level estimate of 500 ± 50 m^3 CH_4 h^{-1} at Nauerna was lower compared to the sum of recovered CH_4 and the measured emission level. We do not have a good explanation for this. Probably fresh waste material that was landfilled shortly before the measurement day was not accounted for well enough in the production estimate. A production estimate that lies below the sum of measured emission and extraction was also observed at Braambergen in 1999. One month after the emission measurements the landfill gas extraction system was extended from 14 to 24 ha of the total landfill (30 ha). It was found that the extraction system recovered an amount of CH4 that was equal to the estimated production level. Since a 100% efficiency of the system over the whole site is not expected, the production level estimate must have been too low.

Table 5.4 Emission data and production model compared (m^3 hr^{-1})

Location	Date	Production	Recovery	Emission	Oxidation
Hollandse Brug	1997	110	0	55	50%
Nauerna	April 1997	590	0	310	50%
	April1998	540	110	155	60%
	Nov.1999	500	110	450	??
Braambergen	Nov. 1999	500	200	500	
	Dec. 1999	500	490(!!)	See text	??

5.4 Conclusions

With favourable meteorological conditions the TDL plume measurements only showed a standard deviation of 20% between individual estimates over one day. The TDL measurement technique is convenient for the landfill operator since it does not interfere with the landfill operation.

Experiments in 1997 and 1998 at Nauerna and Hollandse Brug showed that emission levels obtained from the measurements correspond with those obtained from the landfill gas production model when an oxidation level in the top soil of 50% is assumed. This is a factor of 5 above the 10% level used in the national emission inventories in the Netherlands. This finding is not unique Bergamaschi et al. (1998) also showed 30-40% oxidation. At Nauerna a 50% reduction of the emission occurred between 1997 and 1998. This emission reduction was larger than expected from the model calculations. An increased oxidation efficiency of the landfill cover probably caused this. It showed however that even assumptions for oxidation efficiencies based on previous measurements at the same site need not be representative when conditions at the landfill have changed or when measurements are carried out in a different season. The experiments in 1999 showed that both for Braambergen and for Nauerna, the produc-

tion model leads to under predicted emission levels. The emission level of a landfill is variable both in space and time. Czepiel *et al.* (1996) report an increase in emission of almost a factor of 2, associated with a decrease in the atmospheric pressure over 5 days. For the campaigns reported here air pressures were similar and stable at about 1025 mbar. This confirms that emission levels reported here can be considered representative for the measurement day. More measurements in a year are needed to obtain an annual emission level. Landfills world-wide are considered to be an important anthropogenic methane source. The estimated emission levels however have a significant uncertainty. Most landfills do not have the detailed documentation on the composition of the deposited waste which is available in the Netherlands. Furthermore crude assumptions are used with respect to top cover oxidation. The importance of more measurements at more landfills of different types is therefore obvious. For the Netherlands, in 2010 a limited number of landfills will be responsible for the major part of the methane emissions: over 95% will be caused by a group of 25 landfills. This implies that an inventory of Dutch methane emissions from landfills might be based on direct measurements from this group, on the condition that an accurate and affordable measurement methodology is available.

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