A comparison of measurement methods

to determine landfill methane emissions

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Samenvatting

Methaan uit stortplaatsen draagt aanzienlijk bij aan de emissie van broeikasgassen. In de laatste twee decennia heeft het afvalbeleid geresulteerd in een afname van de hoeveelheid gestort afval en een verandering van de samenstelling van het gestorte materiaal. Als gevolg van die veranderende samenstelling is de vorming van stortgas niet meer goed te prognosticeren, omdat modellen voor stortgasvorming mogelijk niet meer toepasbaar zijn. Als gevolg hiervan zijn de huidige emissie en de gerealiseerde emissiereductie van methaan moeilijk in te schatten. Voor een individuele stortplaats kan de onzekerheid in de CH_4 emissie oplopen tot 50 à 100% en deze onzekerheid maakt het moeilijk om maatregelen voor emissiereductie te dimensioneren.

Een meting van de emissieniveaus is moeilijk met name door de grote variatie van de emissie in de tijd en in de ruimte op zo'n grote stortplaats, met een steeds veranderend landschap van heuvels en dalen waarin zowel actieve stortdelen als afgedekte compartimenten aanwezig zijn. In de laatste jaren zijn er nieuwe meettechnieken beschikbaar gekomen waarmee mogelijk toch representatieve schattingen van de emissie kunnen worden uitgevoerd. In een eerdere evaluatie werden de massabalansmethode en de stationaire pluimmethode geïdentificeerd als veelbelovend. Doel van dit project was het verder ontwikkelen van deze twee technieken en ze te valideren in een vergelijking met de internationaal geaccepteerde mobiele pluimmethode.

De twee methoden zijn getest en gevalideerd na campagnes van vier tot acht weken op vier stortplaatsen in het westen van Nederland: Nauerna, Braambergen bij Almere, 3^e Merwedehaven bij Dordrecht en Wieringermeer. De datasets zijn gebruikt voor vergelijking van de meetmethoden onderling en vergelijking met de emissieschattingen op basis van de modelberekeningen voor stortgasproductie.

De belangrijkste conclusies van het project zijn:
- Zowel de stationaire pluimmethode als de massabalansmethode zijn bruikbaar voor emissie monitoring bij stortplaatsen. In het algemeen komen de emissieschattingen met de mobiele pluim metingen overeen.
- De massabalansmethode geeft inzicht in het tijd verloop van de emissie van de stortplaats. Bij twee stortplaatsen werd een duidelijke correlatie tussen de emissie en veranderingen in de luchtdruk gevonden. Deze methode heeft echter problemen met obstakels (bijvoorbeeld bomen of grote heuvels) op de stortplaats., waardoor de toepasbaarheid van de methode wordt beperkt.
- De stationaire pluimmethode werkt met name goed bij grote stortplaatsen. De methode is niet kritisch voor inhomogeniteit op de stortplaats, wel dient rekening gehouden te worden met andere CH_4 bronnen in de omgeving.
- Met de meetmethoden kan voor een individuele stortplaats een jaargemiddelde emissieschatting met een betrouwbaarheidsinterval kleiner dan 25 % worden verkregen.
- De methaanoxidatie op de vier gemeten stortplaatsen bedraagt naar schatting tussen de 20 en 40%. Voor het berekenen van de CH_4 emissies uit stortplaatsen op nationaal niveau wordt een gemiddelde van 10% voor de oxidate aangehouden. Indien de door ons gevonden oxidatie niveaus representatief zijn zou de emissie van stortplaatsen dus lager kunnen uitvallen dan nu wordt geschat. .
- De eerste resultaten lijken aan te geven dat de gasproductiemodellen nog steeds bruikbaar zijn.

De meetmethoden hebben een aantal toepassingsmogelijkheden:

- Het evalueren van de effectiviteit van maatregelen voor emissiereductie op een deponie.
- Het verkrijgen van een schatting van de CH_4-emissie ter validatie van het gebruikte gas productiemodel voor een individuele locatie en als basis voor de dimensionering van maatregelen voor emissiereductie.
- Het valideren van modellen voor stortgasvorming op basis van resultaten van enkele emissiemetingen. Dit kan de kwaliteit van de nationale emissie-inventarisatie sterk verbeteren.
- Directe monitoring van CH_4 emissies uit stortplaatsen voor nationaal klimaatbeleid. Metingen aan de 25 belangrijkste stortplaatsen in Nederland kan de onzekerheid in de nationaal geschatte CH_4 emissie aanzienlijk verkleinen.

Summary

Methane from landfills contributes significantly to greenhouse gas emissions. In the last two decades, waste policy resulted in a reduced amount of landfilled waste and a change in the composition of the landfilled waste. As a result, prognoses for landfill gas production become less reliable, , since existing landfill gas formation models might not be applicable anymore. As a result, current methane emissions and the established emission reduction since 1990 are less easily quantified. For a single landfill, uncertainties in the emission level make it hard to decide upon measures for emission reduction.

Evaluation of the emission levels with measurements is difficult because of the large temporal and spatial variability of the landfill source strength in a continuously changing landscape of hills and valleys, with active landfilling parts and covered landfill compartments. Fortunately measurement techniques have become available over the last years that can be used to get representative data on landfill CH_4 emissions. In a previous study, the mass balance method and the stationary plume method were identified as promising measurement methods. The objective of this project was the further development of these two methods and a validation of their accuracy in a comparison with the generally accepted mobile plume method.

These two methods were tested and validated in campaigns of 4-8 weeks at four landfills in the west part of the Netherlands: Nauerna, Braambergen, Wieringermeer and Dordrecht. The data sets provide an inter comparison of the techniques and a reference for the emission levels estimated from the landfill gas production model.

The main conclusions were the following:

- Both the stationary plume and mass balance measurement techniques are options for emission monitoring at landfills. In general the emission levels obtained with these techniques agreed with the mobile emission measurements.
- The mass balance method provides insight in the temporal pattern of the landfill emission. At two landfills a correlation was observed between decreasing atmospheric pressure and increasing methane emission. This method does have problems with obstacles on the landfill, for example large hills or tree lines.
- The stationary plume concept works especially well for large landfills. The method can deal with inhomogeneities on the landfill site. Sources outside the site should be taken into account.
- With the measurement techniques the annual emission level of a single landfill can be estimated with an uncertainty level below 25%.
- Methane oxidation on the four landfills is measured to be 20-40%. This is much higher than the 10% default value that is currently used in emission estimates.
- The first results seem to indicate that landfill gas production models are still applicable.

The measurement methods that are available now can be used for:

- Evaluation of emission reduction measures implemented on a landfill.
- Obtaining a measured emission level that can be used to assess the validity of the gas production model used at a specific landfill and therefore assist in the design of measures to reduce methane emissions.
- When several measurements are available, the results can be used to evaluate formation models that are applied in quantification of national emissions.
- Direct monitoring of the CH_4 emissions from landfills for national climate policymaking. Measurement at the 25 most important landfills in the Netherlands could be used to reduce the uncertainty in the national CH_4 emission level.

Contents

Chapter 1.

Introduction

✓ *Why monitoring methane emissions from landfills?*

Landfills are recognized as an important source of methane. Dutch 1990-emissions of methane from landfills are estimated to be approximately 570 ktonne (VROM, 2001). When expressed in CO_2-equivalents, this is a substantial part of total 1990 emissions of the greenhouse gases mentioned in the Kyoto protocol. Due to existing waste policy, methane emissions are expected to decrease autonomously. This autonomous development is however not incorporated in the substantial emission reduction the Netherlands have committed themselves to in order to comply with the Kyoto protocol. Therefore measures for an extra emission reduction are being sought for. Since landfills are responsible for a considerable greenhouse gas emission this source should be well monitored.

Table 1.1: Greenhouse gas emissions in the Netherlands (in 10^6 tonne CO_2-eq. y-1)

	1990	expected 2010*	objective 2010
methane from landfills	12	4	4
other methane	15	10	
other greenhouse gases	190	241	200
total	217	255	204

***** The 2010 prognose level for CH_4 emission from landfills in the Netherlands is subject to continuous changes due to the large uncertainty in this source. The values shown here were assumed at the time of writing this report. The large uncertainty in the prognoses is an important reason to carry out this project.

Afvalzorg and IGAT belong to the largest landfill operators in the Netherlands. Most landfill operators have an ISO-14001 environmental management system. This gives the obligation to monitor emissions and also to strive for a continuous improvement of environmental effects.

✓ *Changes in waste-treatment*

In the last few years waste policy in the Netherlands has led to major changes in amounts and composition of waste that is landfilled. A number of measures have been taken such as the effective implementation of separate collection of paper and organic materials; the capacity of municipal solid waste incineration was doubled in the last decade and the landfilling of combustible wastes is inhibited by landfill bans and landfill taxes. As a result, the amount of waste that is landfilled has decreased from 13,6 Mtonne in 1990 to 5,7 Mtonne in 2001. The organic content of the landfilled waste is reduced and the character of the organic waste will shift from rapidly decomposable vegetable waste to less decomposable materials. As a result the methane emissions from anoxic decomposition of organic matter will decrease.

✓ *Landfill gas formation models may not apply anymore*

So existing knowledge on methane generation in landfills and the subsequent capture, oxidation and emission might not be applicable anymore. This results in a number of problems: the Dutch Government (i.e. the Ministry of Housing, Spatial Planning and Environment - VROM) is less well able to quantify the national emissions and the emission reduction achieved in the period 1990-2010, while landfill operators such as Afvalzorg and IGAT have problems to quantify their emissions and assess the effects of measures for emission reduction.

✓ *Why emission measurements?*

There are several reasons for developing proper measurement methods. These reasons are summarized in table 1.1. In the definition phase of the measurement programme (Scharff et al., 2000) it was reasoned that emission measurements could provide information that is essential for improving current landfill gas formation models. Chapter 2 in this report gives a short summary of the findings of Scharff et al. on the prerequisites of the measurement methods and the sites selected.

Another reason for developing a measurement method is that the existing methodology for estimating methane emissions from landfills is not able to monitor some specific measures that are proposed for further going emission reduction. An example of this is the enhanced oxidation in top-layers. This option is widely recognized as a promising method to reduce methane emissions. Emission reductions achieved in this way can at present not be monitored, due to a lack of suitable monitoring methodology (either a measurement method or an accepted reduced emission factor) and the results obtained cannot be incorporated in the national assessment.

Table 1.1: Reasons for improved modelling – development measurement methods

Reasons for development of measurement methods	Stakeholders
▪ The landfill formation models that are currently used are expected to be no longer valid, because the composition of the waste has changed. Therefore gas emissions cannot be estimated properly and the implementation of emission control options is difficult. Methane emission measurement on a number of locations provides the basis for improved modelling.	Landfill operator, Government
▪ Direct measurement of the methane emissions from landfills contributes to the improvement of the quality of the environmental impact parameters, both for annual environmental reports for the operator to the authorities, and in the national reports to UN-FCCC.	Landfill operator, Government
▪ In a system with emission trading (either on a national scale or in the JI/CDM framework) an affordable and still accurate method to determine the emission estimate level is essential.	Landfill Operator, Government
▪ In test projects that aim to demonstrate emission reduction technologies the emission reduction must be quantified.	Landfill operator, Government

✓ *Objective of the project*

The aim of this project was to further develop and evaluate two potential emission measurement technologies that can provide information on the "whole-landfill" emission levels. Both the Mass Balance Method (MBM) and the Stationary Plume Method (SPM) were identified as promising options for emission monitoring (Scharff et al., 2000). The two methods were validated by comparing the methane emission levels obtained with a mobile measurement technique. This mobile technique can be considered as the internationally approved standard. The measurements of the CO_2 emission obtained with the MBM technique can be used in combination with the methane emission levels to estimate the methane oxidation in the top of the landfill. These measurements were compared in an experiment using ^{13}C-analyses.

The project was carried out in two phases:

- During the first phase both MBM and SPM were developed and applied at the landfill Nauerna. The emphasis of this part of the project is on the evaluation of the practical applicability of the methods. The results of both measurement methods were compared with

the results of three TDL campaigns. A ^{13}C-experiment was carried out as well but this failed due to problems with the GC analyses of the samples.

- During the second phase of the project measurements were carried out at three landfills near Almere, Dordrecht and Wieringermeer and the focus of the projects moved to the evaluation of the results of the measurements as well as the reliability of the emission estimates.

This report gives the results of the full project.

Chapter 2.

Emission measurements as a tool to improve national estimates of emissions

2.1 Strategies for improved emission estimates

In a study for the Dutch Government (Scharff et al., 2000) two strategies were identified to improve the emission estimates. Measurements of emissions at real landfills are an important part of both strategies:
In the first strategy, measurements are used to improve current methodology based on modelling landfill gas formation and insight in methane oxidation. Improving the existing methodology can imply both improving landfill gas formation models as well as improving oxidation factors.
The second strategy ultimately aims at an emission assessment based on frequent or continuous measurement of methane emissions at all relevant Dutch landfill sites. In 2010 a limited number of landfills will be responsible for the major part of Dutch methane emissions: over 95 % will be caused by a group of 25 landfills. This implies that such an approach to the inventory of Dutch methane emissions from landfills might be feasible, on the condition that an accurate and affordable measurement methodology is available.
An important prerequisite of the national emission estimate is that it is accepted in international negotiations. This means that the methodology and parameters used should meet certain requirements. Methodologies and default factors for model parameters are defined in the '1996 IPCC-Revised Guidelines' and the 'IPCC-guidelines on good practice'. Use of other model parameters compared to the defaults and even other methods than the methods defined by IPCC is possible on a few conditions:

- the result should be an improved, more accurate emission estimate;
- the definition of methodology or model parameters should meet certain standards of quality control: it must be based on a number of observations on real landfills; it must be able to withstand criticism of international experts; results should preferably be published in double-peer reviewed journals;
- attention should be paid to base-line correction: any change in methodology or model-parameters along the way must be accompanied by considerations about the necessity of adapting the methodology of estimating 1990-emissions as well.

2.2 Prerequisites for the measurement methods

Quite a number of emission measurements were already performed at landfills. Literature indicates that landfill gas is emitted with high spatial variability: emissions often take place through some preferential channels. Emissions from two spots, located a few meters away from each other, may differ three orders of magnitude. Due to the layer-wise construction of a landfill, horizontal gas migration is preferred and the slopes of the landfill are known to have relative high emissions compared to the top-surface. The inhomogeneity of emissions is increased due to rainfall or frost, when homogeneous diffusion is blocked and emissions through cracks in the surface are enhanced.

Emissions are also variable in time: emissions are a result of difference in internal and ambient pressure and an increase of ambient pressure temporally reduces emissions. When the gas production in the landfill is low, increases in pressure can even cause a net influx of air into the landfill. Also rainfall can cause daily variations in methane emissions.

Methane oxidation in the top-layer depends on temperature and soil-humidity and is relatively increased in the summer period. As a result, methane emissions show a seasonal variation with decreased methane e,emissions in summer periods compared to winter (e.g. Boeckx et al., 1996).

2.3 Limitations of existing measurement methods

In literature various methods for measuring methane emissions from landfills are described. Table 1 summarizes the various methods. The methods are:

- calculation from concentration profiles in soil cores (Bogner and Scott, 1995).
- using closed chambers sampling relatively small parts of landfill surface (e.g. Bogner and Scott, 1995);
- measuring concentration and wind-speed profiles on top of the landfill, enabling the calculation of fluxes with mass-balance or micrometeorological methods (Oonk and Boom, 1995; Savanne et al., 1997).
- determination of methane plumes on the landfill or further away from the landfill to obtain emissions from the entire landfill (e.g., Czepiel et al., 1996; Samuelson et al., 2001; Hensen and Scharff, 2001);
- $\delta^{13}C$ isotope measurements to determine methane oxidation in top covers (e.g. Boeckx et al., 1996). Isotope analyses might be performed both in samples obtained in closed chambers as from atmospheric samples obtained above or away from the landfill.

Table 2.1: Comparison of measurement techniques

technique	spatial resolution	temporal resolution	component	costs	experience[1)]
Soil concentrations	dm^2	hour	CH_4, CO_2 trace components	high	few
closed chambers	m^2	hour	CH_4	high	many
mass balance	few ha	continuous	CH_4, CO_2	moderate	few
stationary plume measurement	entire landfill	day	CH_4	moderate	very few
mobile plume measurement	entire landfill	hour	CH_4	high	some
$\delta^{13}C$ in closed chambers	m^2	hour	CH_4-oxidation	high	some
$\delta^{13}C$ atmospheric	entire landfill	hour	CH_4-oxidation	high	some

1) 'Few', 'some' or 'many' refer to the number od research groups applying this method. E.g. although at TNO there is a lot of experience with the mass-balance method, the experience with this method here is indicated as being few, since apart from a French test, this is the only group applying the method internationally.

When average emissions from the entire landfill are to be measured, flux boxes become very labour-intensive and expensive, considering the amount of data required to obtain a reliable estimate. There is significant improvement in this field using fast response sensors such as the TDL to do the box measurements. This enables a large number of box measurements to be taken on a single day still, for a big landfill like for example Nauerna a number of days will be needed to map the whole landfill. The box technology is particularly useful for process studies and for the evaluation of the effect on the emission level of an emission reduction measure implemented on a part of the landfill (e.g., Scharff et al., 2003).

For "whole landfill" emission measurements, spatial integrating methods like for example the mass-balance method (MBM) seem to be better suited. This technique measures emissions from larger surfaces during longer times and if methane concentration profiles are obtained until sufficient heights, emissions from the entire landfill can be measured. Drawback of the mass-balance method is the limited experience with the method, so validation might be considered a requirement.
Plume measurements with TDL or FTIR technology can be considered the most accurate methods to measure emissions from an entire site. Drawbacks of this method are need for expensive high accuracy detector systems and the relatively high costs when used in prolonged measurement campaigns. So simplification and cost-reduction of these methods are required in order to become practically applicable.
The stationary plume measurement method (SPM) also integrates over the complete landfill. It uses gas-sampling stations at 0.3-2 km away from the landfill to evaluate the emission level. This technique does not provide the full temporal pattern that the MBM method provides but does give estimates for the whole landfill, where the MBM method needs to up-scale one measured segment at a certain time to get the whole emission level. Like the mass balance method this method needs validation.

Chapter 3.

Development of improved methods

3.1 Project description

The project aimed at the development of a measurement method, which enables a reliable measurement of *average annual methane emissions from an entire landfill at reasonable costs*. Starting points in this development are both the mass-balance method and the plume method. Closed chamber methods were considered too labour-intensive and less capable for further automation.

The mass-balance method and plume measurements were improved or simplified as described below. Subsequently both measurements are tested in four field trials and compared with the results of several plume measurements, using N_2O as a tracer gas and as described by Hensen and Scharff (2001). The mass-balance method is in principle suited to measure carbon dioxide emissions as well, thus enabling the measurement of methane oxidation by comparing the emitted ratio of methane and carbon dioxide with the ratio in which methane and carbon dioxide are formed within the waste (obtained from extracted gas). This methane oxidation is compared with the methane oxidation as obtained from $\delta^{13}C$-analyses. These $\delta^{13}C$-analyses are performed by University of gent, who has vast experience in this topic.

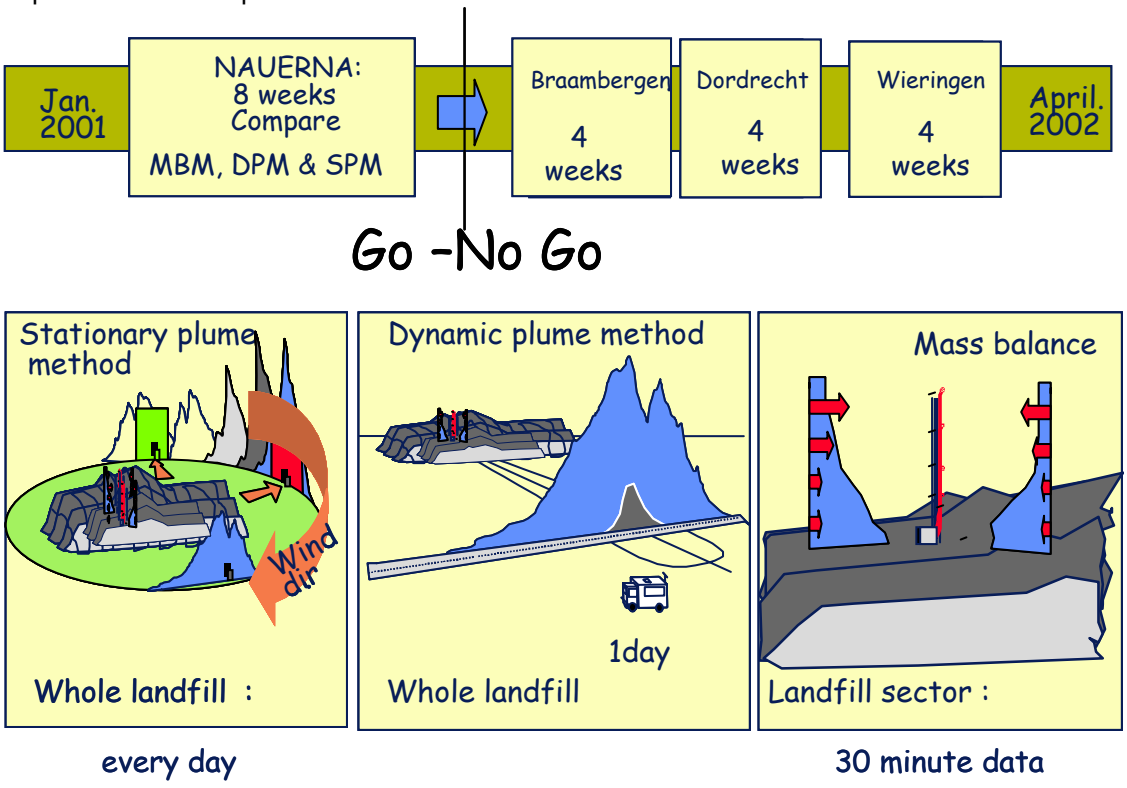


Figure 3.1: Overview of the measurement project

3.2 The Mass-balance method (MBM)

In the mass balance method, vertical methane and carbon dioxide concentration profiles are measured along with a wind-velocity profile, using sampling points in a pole up to 10, 15 or 25 meters high. Profiles are interpreted and emissions from the region upstream of the pole are obtained by:

$$J = \frac{\int_{z=0}^{z=l} u_z (c_z - c_l) dz}{x} \qquad (1)$$

In which J (in $l.m^{-2}.s^{-1}$) is the methane flux through the landfill surface; u_z (in $m.s^{-1}$) is the wind velocity at height z; c_z (in $l\ m^{-3}$, vol‰ or 1000 ppm) is the concentration at height z; c_l (in $l.m^{-3}$) is the background concentration of methane; l (in m) is the length of the pole; x (in m) is the fetch (the upstream length from the pole to the landfill slopes). The measurement of the concentration profiles both for CH_4 and for CO_2 was performed with an opto-acoustical multi gas monitor (Bruel & Kjaer 1402).

In the Netherlands, there is quite some experience with the use of this method for obtaining emissions from landfills (Oonk and Boom, 2000). The method has advantages above other methods: it is easily automated and emissions can be obtained for longer periods of time. Based on experience with the method in previous projects it was shown that approximately three weeks time is required to obtain consistent average emissions. Carbon dioxide emissions can be measured as well, though less accurate. There are also some limitations to the measurement method. For example the applicability of a 10 m high pole is restricted to situations where the distance to the sides of the landfill is less than 150 m. The result in terms of the emission estimate depends on assumptions on distribution of emissions over the surface.

In order to overcome these and other drawbacks, the method was improved, and in order to enable measurements over larger areas (larger landfills) a pole with the maximum measurement inlet at a height of 26 meters was used. A test was performed in which the opto-acoustical detector system that is normally used was compared with the measurements obtained with a CO_2 NDIR (Siemens Ultramat 5). This detector would provide a more accurate CO_2-concentration level. Finally it was foreseen to test, whether solid-state methane sensors can be used. This could significantly reduce costs for methane analysis and reduced the power consumption of the MBM set-up.

Figure 3.2: The mass balance set-up

✓ *Up-scaling of the MBM measurements.*

In order to assess the temporal variation of the landfill gas emissions and in order do evaluate the differences between the different measurement methods, the MBM dataset was used to simulate a "whole-landfill" emission time series. In order to do this, the percentile contribution of each sector to the total average emission was calculated. If a sector, averaged over the whole experimental campaign accounted for X% of the emission level of a landfill, the emission observed in this sector (Q_{sector}) at a time T is extrapolated to estimate the emission of the whole landfill ($Q_{landfill}$) at that time by:

$$Q_{landfill} = Q_{sector}(T) / X \qquad (2)$$

This extrapolation provides an emission time series that can be compared to meteorological parameters. The emission estimate will be most reliable when the sector used has a good over all data coverage and a significant contribution to the total emission. Larger scatter in the data can be expected when using data from a sector that has limited data coverage or a small percentual contribution to the total emission level.

3.3 The Stationary Plume Method (SPM)

The stationary plume method (SPM) and the mobile TDL plume measurements both use the plume of CH_4 that is observed downwind of the landfill. The SPM set-up used here has 4 fixed gasbag-sampling stations around the landfill. On the landfill a computer unit monitors the meteorological conditions. Using these data, the concentrations at the four-receptor stations are calculated. Whenever the predicted concentrations surpass a given threshold level, the computer activates the receptor station

by phone. At the station a battery operated electronic unit samples air in a gasbag for a 30-minute period. The central computer selects both a background sampling station and a station in the plume. In general 2 events are sampled each day, each box has 7 gasbags available. After one week the sets with gasbags are exchanged with empty sets.
The samples are analysed using GC-FID technique in the lab. This automated system analyses all 7 bags from one station subsequently with three analyses per sample. Cross contamination of subsequent samples in gasbags was shown to be below 1 % of the concentration difference. The GC system is calibrated using standards (CH_4 in air) with concentrations of 1,800 and 7,000 ppb. These working standards are calibrated versus NOAA station standards. Therefore comparison with other measurement stations is possible. This is done to enable the evaluation of the background samples in terms of contribution of other sources.

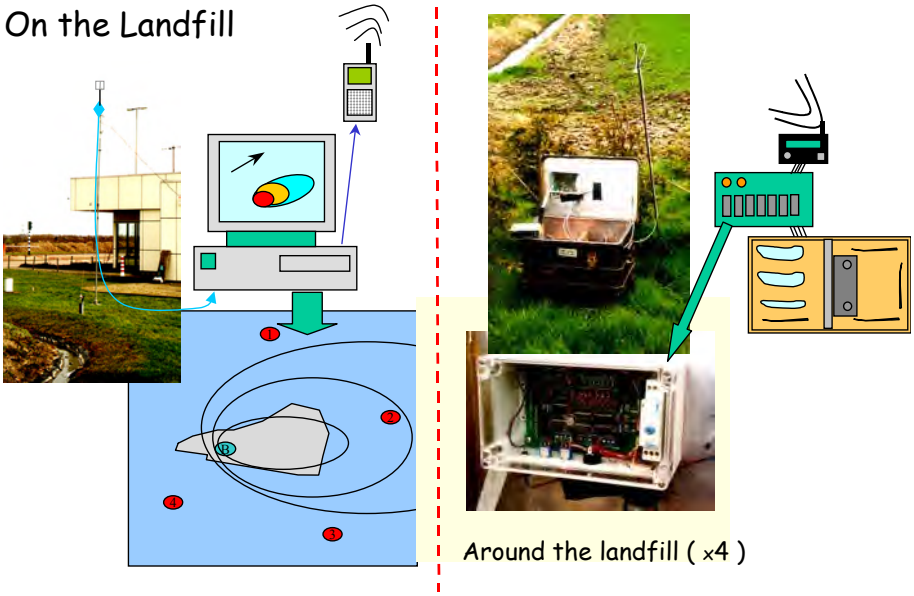


Figure 3.3: The SPM concept. The left part of the picture shows the meteo sensor at the Nauerna landfill site. The data wind-speed and wind direction data is used to run a dispersion model that predicts the concentrations at the 4 sampling stations. Whenever these concentrations exceed a threshold value the central station calls out to the sampling stations, shown on the right side in this picture. This station takes a 30-minute air sample in a 2 l gasbag. In each box a total of seven bags is available.

✓ *The Gaussian model used for the SPM*

A simple Gauss model is used to calculate the expected concentration level at the receptor stations. This model uses a set of point sources over the landfill (Figure 3.3). For each point source a Gaussian plume is calculated, 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 2,000m (generally 600m) and at that distance the effect of an inversion layer only occurs at night under stable conditions. For each source-receptor combination the receptor concentration is obtained:

$$\text{Concentration } (x,y,z) = \frac{Q}{2\pi u \sigma_y \sigma_z} \cdot e^{-y^2/(2.\sigma y)^2} \cdot \left(e^{-(z-H)^2/(2.\sigma z)^2} + e^{-(z+H)^2/(2.\sigma z)^2}\right) \quad (3)$$

with $\sigma_y = A.x^B.z_o^{0.2}.T^{0.35}$, $\sigma_z = C.x^D.(10.z_o)^{0.53.E}$ and $E = x^{-0.22}$

With x 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, H 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 used for averaging. A, B, C and D are depending on the stability class. T is the averaging time, which equals the sampling time of 0.5 hour. The final model-concentration is obtained by adding all contributions from the different sources. The sources on the different parts of the landfill are scaled to get a total Q[model]=1 $gCH_4.s^{-1}$. The Emission in g $CH_4.s^{-1}$ is then calculated according to :

Q[landfill] = Q[model] * (Conc[plume]-Conc[background]) / Conc[model] (4)

At a distance of 1-2 km the plume of the landfill is generally well mixed and there is enough time for vertical mixing to obtain useful concentration levels at the 1.5 m sampling height. Sensitivity runs with the model were performed to assess the optimum sampling time. When sampling for a long time, for example 3 hours or longer, the actual plume of the landfill might have moved away from the receptor station. Over a short sampling period, however, the exact position of the sampling station in the plume is much more important. The averaging interval of 30 minutes will result in a plume that is smoothed compared to those obtained with the TDL plume method but still short enough to have a small standard deviation in the wind direction. An evaluation of longer sampling intervals was foreseen but since the number of samples obtained per campaign (4 weeks) is not very large it was decided to stick to the 30-minute sampling.

The model is run using the meteorological data obtained at the central computer station. In order to assess the sensitivity of the emission calculation to the model parameters, the model is run also at the wind direction +5° and -5°. When the three runs show a large difference the sample is taken on the slope of the plume and the result has a larger uncertainty. When the three model concentrations are close together the sample is taken in the middle of the plume and the uncertainty in the emission is smaller.

3.4 Mobile Plume Method with TDL

This method was used a number of times over the last years to evaluate emission of the Nauerna landfill and other locations (Hensen & Scharff, 2001). The method is accepted internationally to give a good estimate of the emission of a complete landfill site. In this method the CH_4 concentration downwind of the landfill is measured in a transect through the plume, that is composed by all the small methane emission locations on the landfill surface. The concentration measurements are performed using a Tuneable Diode Laser (Aerodyne Research Inst.). Absorption of the light emitted from the diode laser occurs in a 30-m astigmatic multi-pass 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. A plume-transect takes 5-7 minutes. The measurement method is shown in figure 3.4

The TDL system has a 10 ppb resolution for CH_4 and 20 ppb for N_2O and can measure at frequencies up to 20 Hz. In these campaigns 1Hz data was used. Calibration took place while driving, applying standards, before and after a transect measurement. The position of the van was obtained using a GPS system (Survey II, Garmin Inc.). The measured concentrations in the plume transect are compared with the output of a multiple gauss plume model described above.
The emission strength of the landfill is equal to the source strength needed in the model to get an agreement between the integral of the concentration along the measurement transect for the modelled and the measured plume. The meteorological data that is needed for the model calculations (wind speed, wind direction and turbulence) is obtained either from measurements on top of the landfill or from measurements at the van. These measurements indicate the Pasquill

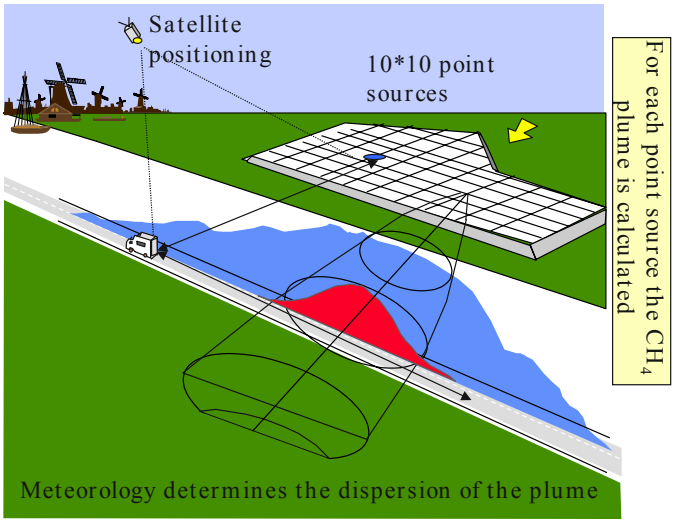


Figure 3.4: The mobile measurement method used to evaluate the emission of the landfill. Concentration measurements obtained on a transect downwind of the landfill are compared to the levels obtained using a Gaussian dispersion model.

stability class, but an extra check on this choice is recommended. Therefore N_2O was released from a gas flask on top of the landfill and the TDL measures the N_2O plume simultaneously with the CH_4 plume. For the N_2O source, both position and source strength are well defined. The model calculation for this plume is used to check, and if necessary to adapt, the parameters that determine the dispersion.

The uncertainty in the CH_4 concentration measurements is 1-5% due to instrument noise, drift of the laser and uncertainty in the background concentration level. The changes in wind direction (on a timescale of 5-10 minutes) are the main cause for variation in the set of emission estimates.

Furthermore the emission of the landfill site will not be constant in time. This measurement method gives an emission level for the whole location on the day of measurement.

3.5 ^{13}C isotope measurements

In nature two stable isotopes for carbon are present. In the atmosphere the ^{12}C isotope is dominant but approximately 1% of the carbon is present in ^{13}C. In natural processes that fix CO_2 from the atmosphere the ^{12}C is preferentially removed from the air and the $^{13}C/^{12}C$ ratio in this material is therefore different from the composition in air. Microbial CH_4-oxidaton also takes place with a slight preference for $^{12}CH_4$ compared to $^{13}CH_4$. Therefore the remaining CH_4 will become enriched with $^{13}CH_4$ compared to the original landfill gas as it flows through a layer in which methane oxidation

occurs. By measuring this enrichment of the emitted methane versus the original landfill gas the CH_4-oxidation can be estimated. (Bergamaschi et al., 1998)

The purpose of the ^{13}C experiment was to determine the average CH_4-oxidation in the cover soil of the landfill. The process of oxidation will have effect on the gas that is formed inside the landfill. Samples of this unoxidised gas were obtained from the gas extraction system. These samples were obtained in 250 ml brown glass flasks. In these samples the original $^{13}C/^{12}C$ ratio is determined. The gas that escapes to the atmosphere will be enriched with ^{13}C due to the preferential uptake of $^{12}CH_4$ by the microbes. To determine the average ^{13}C level of the emitted gas ambient air samples were collected both close to the landfill and at a distance of approximately 1 km from the landfill. For the air samples at different distances to the landfill the mixing ratio of the atmospheric background methane and the landfill gas changes. Plotting the methane $\delta^{13}C$ versus 1/[CH_4-concentration] will give a curve with a set of points. A linear fit to this set provides a line that at the intercept will give the estimate of the $\delta^{13}C$ value of the gas that exits in the landfill. This value is compared to the $\delta^{13}C$ of the landfill gas itself. The shift in isotope ratio is a measure for the oxidation level. The last step is to determine how much the $\delta^{13}C$ is shifted at a known amount of CH_4 oxidation. This is determined in the lab using soil samples that were taken at three locations on the landfill.

Al isotope analyses were performed using a PDZ isotope-ratio-mass-spectrometer (IRMS). The ^{13}C-contents of a sample is expressed as the $\delta^{13}C$-value:

$$\delta^{13}\mathrm{C} = \left(\left(\frac{R_{\mathrm{sample}}}{R_{\mathrm{standard}}} \right) - 1 \right) \times 1000 \qquad (5)$$

R_{sample} is the ratio $^{13}C/^{12}C$ of the sample, $R_{standard}$ is the ratio $^{13}C/^{12}C$ of an international standard (0,0112372).

The preference of the methanotrophic micro-organisms for $^{12}CH_4$ oxidation versus $^{13}CH_4$-oxidation is expressed using the α-factor:

$$\alpha = k_{12}/k_{13} \qquad (6)$$

with k_{12} the decay constant for $^{12}CH_4$-oxidation and k_{13} the decay constant for $^{13}CH_4$-oxidation. The level for α was determined with batch-experiments using the three soil samples. Each sample was put in a bottle that was sealed with ca. 1% of CH_4 added. The concentration and the delta-value of the remaining CH_4 in the bottle were monitored. The fractionation can then be determined by regression using the formula (Coleman et al., 1981):

$$\delta^{13}\mathrm{C} = 1000\left(\frac{1}{\alpha} - 1 \right) \ln\left(\frac{M}{M_0} \right) + \delta^{13}\mathrm{C}_{t=0} \qquad (7)$$

with M/M_0 the CH_4 fraction that remains in the flask at certain time. The CH_4 fraction oxidised in the cover soil can be determined using the formula (Liptay et al., 1998):

$$f_{\mathrm{ox}} = \frac{\delta E - \delta A}{1000\left(\alpha_{\mathrm{ox}} - \alpha_{\mathrm{trans}}\right)} \qquad (8)$$

with δE the delta-value of the CH_4 that leaves the landfill cover soil, and δA the delta-value of the produced CH_4. α_{ox} is the α-value described above, α_{trans} is the fractionation coefficient for transport through the cover soil. It is usually assumed that α_{trans} =1.

The soil samples and air samples were taken on October 3 2001 at the Braambergen landfill in Almere. Air samples were taken both up and downwind of the location. Landfill gas samples from the extraction system were sent to the University of Gent later.

Chapter 4.

Results

Whenever a m³ is mentioned in the next chapters, this is expressed as m³ STP, i.e. a cubic meter at Standard Temperature and Pressure: 20°C and 1.013 x 10^5 Pa.

4.1 Nauerna.

4.1.1 Description of the landfill

The first comparison of both methods with the TDL plume measurement took place on the Nauerna landfill in the period of April to June 2001. Nauerna (N 53°50' , E 4°55') is a landfill of approximately 1200m by 800m, located just north of the North sea channel and west of Amsterdam. In other directions, agricultural areas to the North and West surround the landfill. There is a small village with a marina to the east. South of the North Sea channel is a recreation area with forest.

The landfill was opened in 1985, and is still operational. At the moment of the emission measurement, in total 7.7 Mtonne of waste was deposited. Nauerna ultimately is designed to be integrated in the polder-landscape as four hills in a row, increasing in size from west to the east. At the moment of the measurement, the majority of the waste is landfilled in two larger hills, located in the darker-brown coloured area in figure 4.2.

The waste at Nauerna is characterized by its high inorganic content: the main waste streams are contaminated soil, demolition waste, shredder waste and sludge. Some organic material is fed into the landfill through industrial wastes, shredder waste and sludge. The average carbon content of the entire landfill is estimated to be 47 kg $tonne^{-1}$, which is approximately 40% of the Dutch average.

Despite the low organic content of the waste, landfill gas is extracted. In this period of measurement, landfill gas was extracted at a rate of approximately 155 m^3 hr^{-1}, containing on average 56% of methane. There are various prognoses available for landfill gas formation at Nauerna. Starting from different assumptions on e.g. the contribution of sludge to landfill gas formation, formation estimates range from 700 to approximately 2,000 m^3 hr^{-1}, indicating the difficulties that exist to make a proper prognosis of landfill gas formation in a relative inorganic landfill.

4.1.2 Emission measurements

The measurement period started April 5 and ended June 18, 2001. Average meteorological conditions during these weeks are listed in table 4.1. In the first weeks of the campaign there was a significant amount of rain and temperatures were generally low. At some nights in this period frost occurred. The first four weeks were characterised by relatively heavy rain, during which approximately 400 mm of precipitation was measured, which is half of the Dutch average annual precipitation.

Table 4.1: Average meteorological conditions at Nauerna

Week nr	Date Start	T_{avg} °C	T_{max} °C	T_{min} °C	U_{bar} m s^{-1}	Rain mm
12	2-4-01	9.9	21.5	3.9	6	157
13	9-4-01	6.1	11.4	-3.5	4.3	79
14	16-4-01	5.4	11.8	-1.3	3.8	112
15	23-4-01	10.3	16.5	3.4	4.4	80
16	30-4-01	10.7	20.5	4.8	4.5	20
17	7-5-01	15.9	24.9	4.1	3.8	0
18	14-5-01	12.6	20.2	3.5	4.5	17
19	21-5-01	15.1	22.9	6.3	3.8	0
20	28-5-01	13.9	21.8	5.4	4.6	19
21	4-6-01	13.1	20.4	3.3	3.7	5
22	11-6-01	14.7	22.4	5.2	2.6	45

Figure 4.1 gives an overview of all measurement activities: the four locations for static plume observations were located at both sides of the North-sea channel; the pole for the mass-balance method was located in the middle of the major waste locations. TDL-measurements were performed on three days, covering three different wind directions.

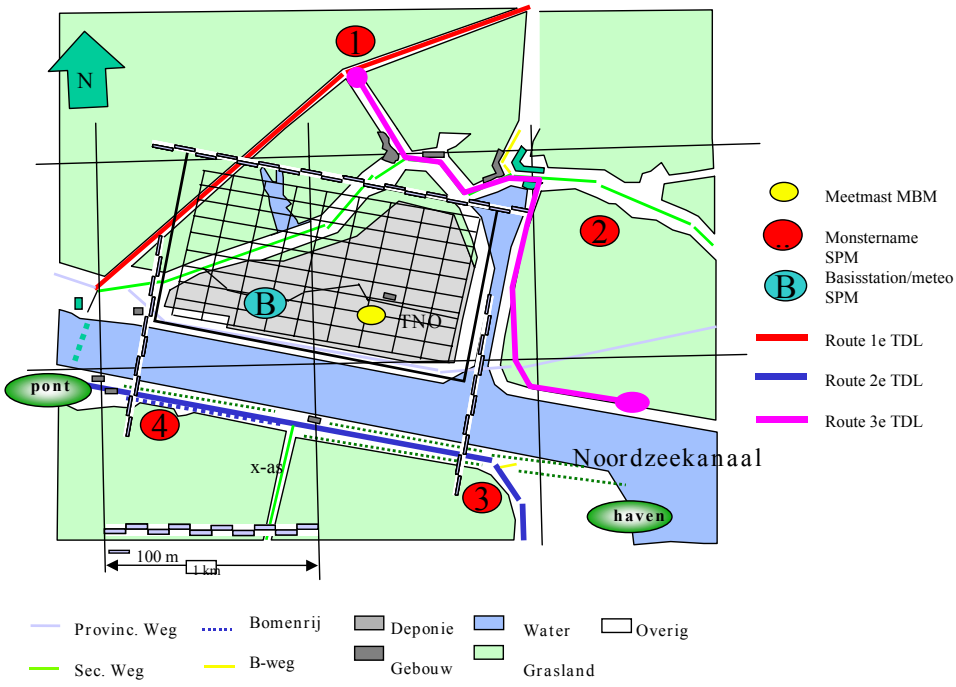


Figure 4.1: Overview of all emission measurements at Nauerna

4.1.3 Mass-balance method

✓ *Technical*

In week 12, 2001, the pole for the mass-balance measurement was erected at the eastern slope of the western waste hill. The pole was based on a concrete platform and the guy-ropes were attached to 2 by 2m concrete slabs at approximately 8 m distance from the pole.

Initially a few technical problems were encountered with breaking guy-ropes and a failure of data-acquisition. However after these problems were solved in week 13 the measurement worked without any further technical problems. Problems did exist with the Siemens Ultramat 5 NDR for CO_2-analysis. Those problems could not be solved and it was decided not to use the Siemens anymore in this project. All CO_2-analyses in this project are therefore obtained with the Bruel and Kjaer IR.

✓ *Determination of emitting area*

The emission that is calculated depends on the area that is assumed to be the source of emissions. The assumption that landfill gas emissions originate from the entire landfill area leads to other conclusions than the assumption that emissions originate from the hills where recently waste was deposited (see also the discussion in chapter 5). At Nauerna the majority of the waste is landfilled in the darker area in figure 4.2. The waste deposited here is also the more recent waste, so the most likely assumption is that the majority of emissions will stem from this region.

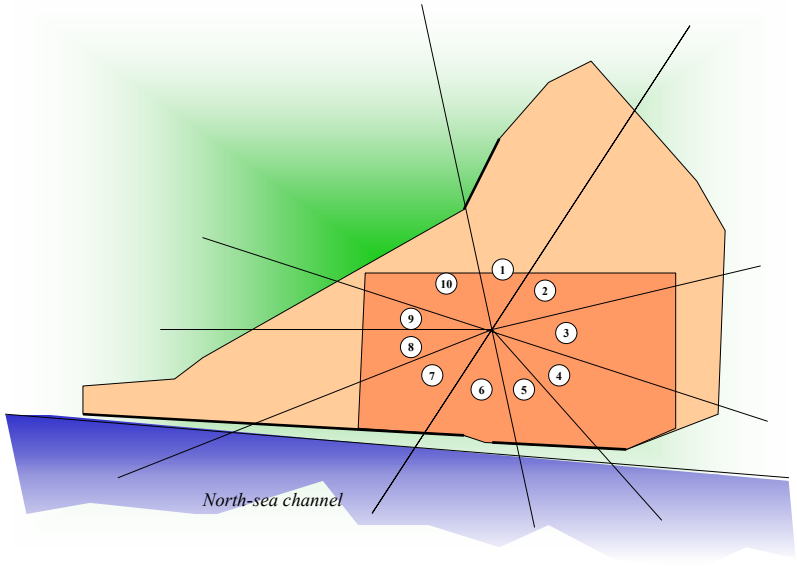


Figure 4.2: Sectors of the Nauerna landfill for the MBM-measurements

This assumption on emitting area is validated, by measuring methane concentrations at 30 cm above the surface, using a mobile CH_4-detector with a lower detection limit of approximately 10 ppm. In approximately 4 hours time, methane concentrations were measured on a 30 x 30m grid scale. The results are shown in figure 4.3. From these measurements it is clear that the darker area is the main emitting area, and the second assumption in table 4.3 is more realistic. In the conclusion chapter (5.3) a more in-depth discussion is held on the topic of location of emissions.

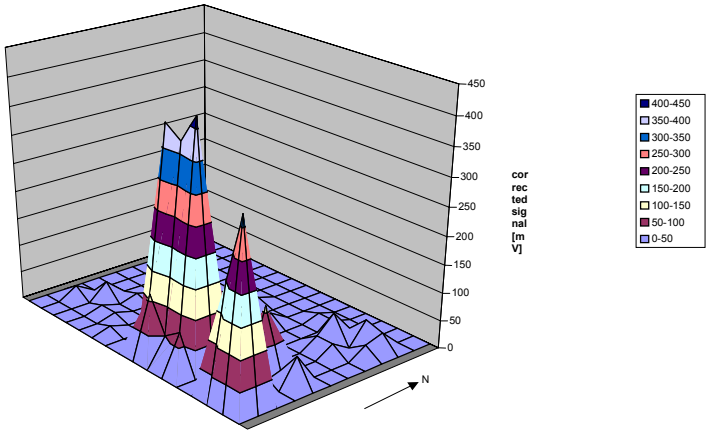


Figure 4.3: Distribution of methane concentrations on top of the Nauerna landfill

✓ *Results*

For interpretation of the mass-balance data, the landfill is divided into 10 sectors as indicated in the figure 4.2. Per sector methane and carbon dioxide emissions are determined, assuming the emitting area corresponds to the darker area in figure 4.2. The results are listed in tables 4.2 and 4.3.

Table 4.2 MBM results CH_4-emissions at Nauerna.

Sector	X (m)	angle (°)	area (m^2)	CH_4-flux (l $m^{-2}.hr^{-1}$) mean	SD	n	total flux (m^3 CH_4 hr^{-1})	95%-CI (m^3 CH_4 hr^{-1})
1	95	45	3,544	1.67	1.51	181	5.9	0.8
2	198	45	15,395	1.29	0.79	50	19.8	3.4
3	436	30	49,767	0.74	0.73	8	37.0	30.5
4	434	30	49,311	1.16	0.79	10	57.4	27.8
5	324	30	27,483	2.49	0.98	7	68.4	24.9
6	278	45	30,349	3.89	1.66	37	118.2	16.8
7	290	35	25,687	3.59	1.69	162	92.3	6.7
8	237	20	9,803	4.92	2.08	59	48.3	5.3
9	234	20	9,557	3.61	2.47	143	34.5	3.9
10	144	60	10,857	4.12	2.83	199	44.8	4.3
		360	**231,754**			**856**	**527**	

Table 4.3 MBM results CO_2-emissions at Nauerna.

sector	x (m)	angle (°)	area (m^2)	CO_2-flux ($l.m^{-2}.hr^{-1}$) mean	SD	N	total flux ($m^3CO_2.hr^{-1}$)	95%-CI (m^3 $CO_2.hr^{-1}$)
1	95	45	3,544	4.37	4.23	181	15.5	2.2
2	198	45	15,395	1.94	1.68	50	29.9	7.4
3	436	30	49,767	1.57	1.10	8	78.2	45.7
4	434	30	49,311	1.45	2.22	10	71.7	78.4
5	324	30	27,483	3.69	2.33	7	101.3	59.2
6	278	45	30,349	5.61	3.68	37	170.1	37.2
7	290	35	25,687	5.04	2.83	162	129.4	11.3
8	237	20	9,803	4.98	2.46	59	48.9	6.3
9	234	20	9,557	4.27	5.61	143	40.8	8.9
10	144	60	10,857	6.68	6.38	199	72.5	9.7
		360	**231.754**			**856**	**758**	

✓ *Duration of the measurement*

In the 8 weeks that the mass-balance method was operational, sufficient data are obtained from the sectors 1,2 and 7 to 10. In the whole period the occurrence of eastern wind was scarce and only few observations are obtained from the sectors 3 to 5. So the limited number of profiles from this direction in table 4.2 and 4.3 is because of the limited availability of raw data from these sectors and not because of some difficulties in interpreting profiles (as is the case, e.g. on Braambergen, see chapter 4.2). This indicates that under unfavourable weather conditions, a 4 weeks measurement period might be short.

✓ *Scaling the MBM measurements to a time-series of emissions of the entire landfill*

In order to assess the temporal variation of the landfill gas emissions and in order do evaluate the differences between the different measurement methods, the MBM dataset was used to simulate a "whole-landfill" emission time series. In order to do this the percentile contribution of each sector was calculated based on the whole data set. Then for each individual emission data-point the emission for the whole landfill is estimated by dividing the sectoral emission level by the percentage.
The results for Nauerna are shown in the discussion in figure 4.6.

✓ *Methane oxidation*

Upon emission, methane might be biologically converted to carbon dioxide in the top-layer of the landfill. The amount of methane that is oxidised, can be estimated by comparing the ratio of emitted methane and carbon dioxide and the ratio of methane and carbon dioxide in the landfill body, below the oxidising zone. The latter can be estimated from landfill gas that is extracted in the landfill gas recovery system at Nauerna.

Extracted landfill gas at Nauerna is periodically analysed for its CH_4, CO_2, N_2 and O_2-content. The mean composition of the extracted gas is given in Table 4.4. CH_4 and CO_2 are the product of anaerobic conversion of waste; N_2 and O_2 in the extracted gas are the result of air being sucked into the landfill due to the negative pressure in the extraction system. Due to this air intrusion, the composition of the extracted gas is not entirely representative for the landfill gas outside the spheres of influence of the extraction system, and this landfill gas outside this sphere of interest is more likely to be emitted to the atmosphere. So for determining methane oxidation from a comparison of the composition of extracted gas and emitted gas, the extracted gas composition should be corrected. This correction is also shown in Table 4.5. The method for correction is the following: every 4 vol% of

nitrogen was accompanied by 1 vol% of oxygen. The difference between the measured oxygen concentration and the calculated oxygen intrusion (1/4th of the N_2-concentration) is the oxygen consumption. The consumed oxygen is assumed to be reacted with the landfill gas (or the components in the leachate that for landfill gas). Since methane reacts with 2 molecules of O_2, the reduction in methane is half of the oxygen consumption; the increase in CO_2 is the same.

Table 4.4: Composition and CH_4:CO_2 ratio of extracted gas at Nauerna (corrected for oxidative processes due to air intrusion)

	extracted composition (vol%)	**corrected composition (vol%)**
CH_4	56.3	56.1
CO_2	32.8	31.2
N_2	10.3	10.1
O_2	0.7	2.6

oxygen consumption	1,8 vol%
CH_4:(CH_4+CO_2) in extracted gas (based on corrected values)	0.64

The ratio of emitted methane and total landfill gas emitted (sum of CH_4 and CO_2-emissions) at Nauerna is 0.40 -/-. Methane oxidation can be calculated from the concentration decrease of methane in landfill gas upon emission (so the difference in concentrations between the extracted gas and the emitted gas).

$$\text{Methane oxidation} = ([CH_4]_{extracted} - [CH_4]_{emitted})/ [CH_4]_{extracted} =$$
$$= (0.64\text{-}0.40)/0.64 = 38\%^{1}$$

4.1.3 Stationary Plume Method

✓ *Locations*

The SPM stations were located at remote houses at distances between 400 and 800 m in the four wind-directions around the landfill. The locations on the south side of the North Sea channel were on the edge of a recreational area. The station in the west was a farmhouse with no agricultural activities. The North location had some agricultural sources (a small barn and a manure heap) activity 80 m east of the sampling site.

[1] It has to be noted that this is an estimate of methane oxidation of unknown accuracy: at one hand the method for estimating emissions is not validated; at the other hand the methane oxidation is based a.o. on the measured carbon dioxide emission and this result has to be considered uncertain. See paragraph 5.4 for further discussion on using the MBM for estimating methane oxidation.

Figure 4.4: SPM station nr.3 at the south east side of the landfill. The box in the front is the sampling station. The landfill is 500m to the north behind the dike and the channel.

✓ *Technical experience*

During the first half of the campaign technical problems with the sampling units occurred. During the second half of the campaign the sampling system worked well. On average 2-3 samples were taken each day and a total of 100 samples was obtained. Since the method uses a combination of an upwind and a downwind sample approximately 30 complete events were obtained.

✓ *Results*

The scatter in the data obtained from the individual 30-minute emission estimates is significant. This scatter is a combination of measurement uncertainty and of the actual variation in the emission level. The scatter in the SPM data and the scatter in the time-series obtained from the MBM are similar and in the order of 50% of the emission estimate. The TDL campaigns that usually cover at least half a day indicate that the total emission level within a day is generally constant to within 10-15%. The results obtained per station are listed in table 4.5. The average of all (individual) measurements is 440 m^3CH_4/h.

Table 4.5: SPM Results Nauerna

Station	Emission ($m^3CH_4 .h^{-1}$)	Stdev	Nr	95% ci
Station 1	191	113	6	
Station 2	605	707	11	
Station 3	470	451	10	
Station 4	289	107	4	
All samples	440	506	31	180

4.1.4 Plume measurement with mobile TDL

For validation of the MBM and SPM measurements three 3 TDL-measurement sessions were performed at different wind directions examples of the plumes observed during these three campaigns are shown in figure 4.5.

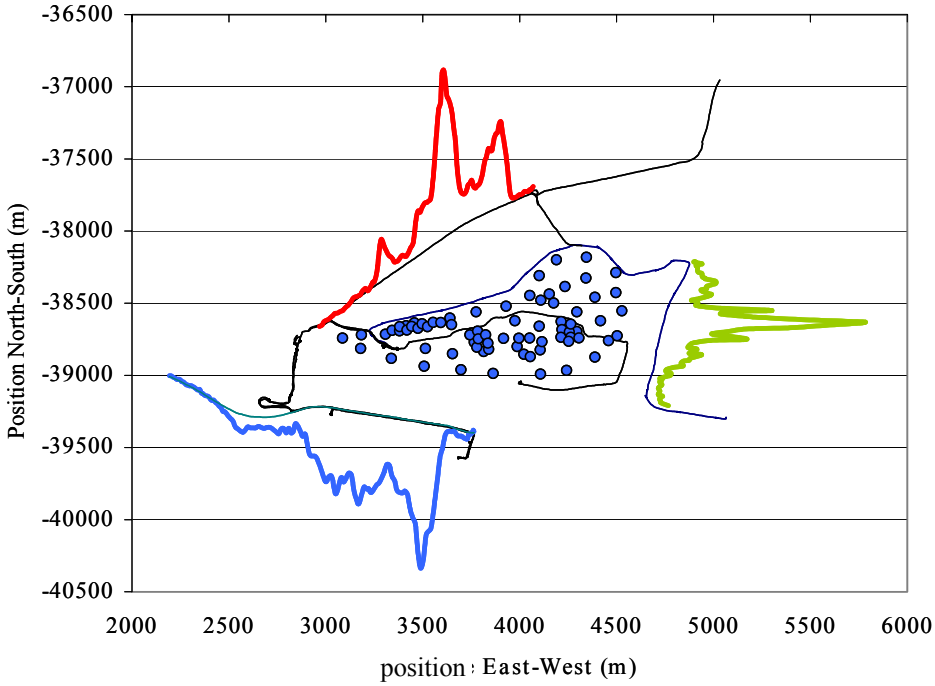


Figure 4.5: Emission measurements with the mobile TDL system obtained on three days. The concentration is plotted into the wind direction for each campaign

✓ *Campaign 1: April 24 2001*

Measurements took place on the North side of the landfill. Overall weather conditions were cloudy with wind-speeds of approximately 6 m/s. The observed plume showed 2 or 3 maxima along the transect indicating different active source areas on the site. N_2O was released from a part in the centre of the landfill. The main emission occurs around the two big hills on the site that were under construction. The active landfill site at the north side of the landfill also showed a substantial CH_4 emission. Finally it seemed that an emission takes place from the mid east part of the landfill where sludge compartments were emptied. Part of the top layer that normally inhibits the emission from the underlying waste was temporarily removed. Emission from this part of the landfill explains why in most plumes a peak is observed at the east end of the measurement transect.

✓ *Campaign 2: May 2 2001*

Measurements took place at the south side of the landfill, across the North Sea channel. Weather conditions were partially cloudy with 6m/s wind. The measurements that took place while driving on top of the landfill and along the southern border of the site provided important information to derive the source distribution. The measurements showed that there still was a significant emission occurring on the northeast sector of the landfill, the area with the sludge compartments. Also the emissions from the active landfilling part were observed. At the southern border of the landfill a semi constant concentration level was found except for a much larger peak in the concentration observed downwind of the passage between the two hills on the landfill. At the measurement transect the plume showed one maximum downwind of the set of two hills on the landfill.

Campaign 3: June 16 2001

In spite of the predictions of a southern wind, no wind at all was observed at the Nauerna when this campaign started. The wind started after a small period of drizzle, but came from the west.

Measurements therefore had to be performed at the east side of the landfill. This transect is not ideal, since the distance of approximately 200 m to the edge of the landfill is relatively small. The low wind-speed of approximately 3 m/s and the variable wind direction made the plume-shape very variable. Weather conditions were fair with approximately 1/8 cloud cover.

Table 4.6: The methane emissions form the Nauerna landfill measured using the TDL.

TDL-measurements 2001	Wind direction	T °C	U (m/s)	Stability Class	Nr. plumes	CH_4-emission ($m^3.hr^{-1}$)
April 24	South East		6	D	17	900 ±250 (=760+140)
May 2	North		6	D-C	13	550 ±90
June 15	West		3	B	16	540 ± 250

4.1.5 ^{13}C isotope measurements

During the third TDL campaign samples were collected for the ^{13}C isotope measurements. For this sampling small 250ml evacuated bottles were used. Unfortunately these sampled could not be analysed due to technical problems with the GC-MS system. The isotope experiment was repeated with success at the Braambergen site.

4.1.6 Discussion

✓ *Comparison of the measurement results.*

The average emission estimates and the 95% confidence interval for each method are shown in table 4.7. The mass balance method and SPM method agree within their confidence limits. The first TDL-measurement shows a significantly increased emission (which is partially attributed to incidental emissions due to excavations in a sludge compartment). The subsequent TDL campaigns show a slightly higher emission level for the second and third campaign. But, this difference is also within the confidence interval. The confidence level for the third campaign was relatively large because the meteorological conditions were not favourable with low wind speeds and variable wind directions. In spite of doubts about the small distance between the landfill and the transect used in the 3rd campaign and the meteorological conditions, the result in terms of emission level do compare well to the 2nd campaign.

Table 4.7: Comparison of emission levels obtained at the Nauerna Landfill

	CH_4-emission ($m^3.hr^{-1}$)	**CO_2-emission ($m^3.hr^{-1}$)**	**methane oxidation**
Mass balance method	530 ± 130	760 ± 200	38%
Static plume measurement	440 ± 180		
TDL	1st exp: 900 ± 112 (900 = 775+125) 2nd exp: 550 ±50 3rd exp: 540 ± 115		
Prognosis emission	220-900[3)]		

1) Indicated is the 95% confidence interval, based on the variations in measurement results. It has to be stressed that this is not the same as the inaccuracy of the method. Model uncertainties might contribute to the uncertainty as well.

2) The higher emission observed with the TDL was partially originating from an extra source on the North West part of the landfill. This contribution was estimated to be 15%.

3) Various prognoses are available for landfill gas formation, starting from different assumptions on fractions that contribute to total landfill gas formation, e.g. whether or not sludge contributed to landfill gas formation. The prognosis is the prognosis for methane emission and is corrected for extraction (85 m^3 hr^{-1} of CH_4 and 10% oxidation).

✓ *Time-series of emissions*

The agreement between methods becomes even more striking when the time-series in emissions is compared. This is done in figure 4.7. In this figure the small dots show the "up-scaled" emission levels using the MBM time series (see chapter 3.2 for the method of constructing them). The black line shows the 12-hour running mean curve using these data points. The blue line shows the -dp/dt, the inverted change of pressure over time. It turns out that the resulting time series showed a reasonable correlation with the change of pressure over time. Decreasing pressures lead to an increase in the emission level. The SPM and TDL results are also plotted in this graph. The error bars for the TDL experiments show the 95% confidence limit for the average emission level obtained from the set of measured plumes. The error bars for the SPM measurements show the difference in emission estimate obtained with wind-directions + or - 5 degrees compared to the actual wind direction. It is clear that the scatter shown by the SPM data does not seem remarkably different from the scatter observed for this modified MBM results.

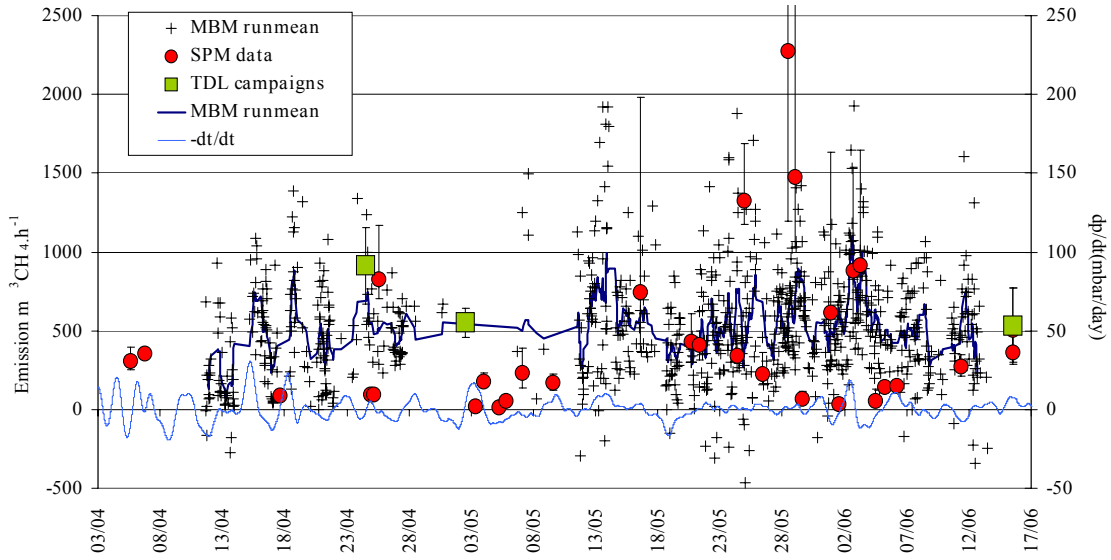


Figure 4.6: Temporal variation of the emission at Nauerna.

✓ *An incidental emission due to excavations after April 24th*

During the first campaign the TDL emission level was significantly higher compared to the average the other campaigns. The plume shape and the concentration levels indicated that additional methane originated from the Northeast part of the landfill. It turned out that indeed in this period two sludge compartments were emptied, removing the top layer of the underlying compartments in that section. This explains part (roughly 200 of the increase of 500 $m^3.hr^{-1}$) of the increased emission level. According to the TDL model runs, this could account for an increase of the emission of approximately 15-20% compared to "normal" situation. Without this extra source the emission on April 24 is still considerably higher than the emission on the other two days. The MBM data for that day also indicated an increase in the emission level (which is based on a extrapolation of the measurements in the wind-sector south of the mast; see figure 4.6 and the accompanying text). The increase in emissions is most likely caused by changes in weather condition. As explained in figure 4.6 changes in air-pressure seem to correlate well emissions from Nauerna. For the other landfills similar correlations are observed (see chapters 4.2 to 4.4).

The increased emission from the sludge-compartment part of the landfill was still observed on May 2 , during the 2nd campaign while driving on top of the Landfill.

✓ *Limitations to the measurement methods*

From the experiences with varying emissions at Nauerna, a number of things can be learned:

- Since the MBM-measurement depends on wind direction it might miss an event that occurs on the landfill site in a sector that is not measured at that moment. So the MBM does not measure the emission of the complete landfill at all times. The emissions from the sludge compartments around April 24 occurred downwind of the MBM mast and the emissions were not observed until May 1 when the wind-direction changed to the sector affected. After this day the MBM technique indeed showed elevated concentration at the top of the pole, suggesting high emissions. In the current measurement-set-up, concentrations at the top of the pole are considered to be background concentrations, and profiles with significantly increased background concentrations are neglected. So despite the profiles that were obtained after a change in wind-direction, still no methane-emission estimate was obtained from that sector;

- The TDL plume measurement does measure emissions due to incidents. However only part of the elevation in emission can be attributed to digging activities on the landfill. This confirms the conclusion from the definition study (Scharff et al., 2000) that a single TDL campaign is not applicable to estimate annual average methane emissions, due to the variability in emissions. Emissions from time to time might deviate 50% from the average value due to changes in weather. However, knowing that incidental emissions occur, a landfill emission estimate based on a single TDL campaign during an emission event will overestimate average emissions from the landfill, when the incidental emission is not taken in consideration.
- The SPM in principle combines the best of both worlds. It samples emissions from the whole landfill, so incidental emissions taking place on part of the landfill will be detected by the SPM. (In theory the SPM might miss events when the plume passes in between two sampling stations but with stations in four wind sectors this chance is small.). At Nauerna the system provided three emission estimates for April 24 & 25. Two of those events showed emission levels comparable to the MBM level, one estimate shows the same value as obtained with the TDL). On the other hand, the SPM must be capable of making a long time-series in emissions and deal with normal variations in emissions and incidents, provided that sufficient sample are taken.
- A general conclusion is that incidental emissions are important when understanding an emission measurement, therefore it is important to log all events (excavations in the landfill; top cover repair, repair and maintenance of gas extraction; amount and composition of extracted gas) for later interpretation.

4.1.7 Conclusion of the Nauerna measurements

Mass-balance method (MBM)

- The MBM proved to be well applicable. Power supply was not problematic here and was supplied using an extension lead. The out-of-pocket-costs of the method at Nauerna were approximately € 7.500-10.000 (for the 8 weeks measurement period)
- The methane emission estimate of the MBM is 527± 130 $m^3.hr^{-1}$ and is in agreement with the TDL-results. This is especially true, when the time-series in emissions is considered.
- The time-series in emissions as obtained from the MBM show a correlation with air-pressure. Temporal variations in the emission time series are in the order of magnitude of 50% of the average emissions. The timeframe in which changes of emissions might occur is about 6-12 hours, emissions might change considerably.
- The MBM measures emissions from a limited sector of the landfill and the method might miss incidental emissions that take place on a part of a landfill, located upstream from the pole.
- The CO_2-analysis of the MBM, made with the Bruel and Kjaer IR could not be improved due to failure of the Siemens NDIR-analyser
- The methane oxidation obtained from the mass-balance method could not be verified, due to a failure of the ^{13}C-analysis.

The static plume measurement (SPM)

- The SPM worked well especially considering that this system was not used before. After initial problems with sample triggering were solved, the equipment operated without many problems and a significant number of data could be gathered. Considering the variation in calculated emissions an even larger number of measurements and therefore a more robust triggering seems to be a prerequisite for application of the method. The out-of-pocket-costs for the SPM were € 15.000 and will probably be reduced to an estimated € 4.000 to 5.000 when more experience is gathered with the system.

- The emission level derived from the SPM data set of 440 $\pm$ 180 $m^3.hr^{-1}$ is somewhat lower compared to the other estimates, but in agreement when the uncertainty range is taken into account.
- The SPM stations are battery operated. Only a small meteo station is needed on the landfill site or on a location outside the landfill.

4.2 Braambergen

4.2.1 Description of the landfill

The Braambergen landfill is located near Almere. South of the landfill there are 3 farms at approximately 1km distance, with surrounding fields. North, east and west of the location is recreational area with grass fields and patches of forest. Further north of the landfill is a domestic area. The landfill is in operation since 1981. Until the year 2001 approximately 1.7 million tonne of waste was deposited on a total surface area of 10 ha. The eldest waste is deposited on a part in the south of the landfill. This area is lined with an HDPE-liner system and is expected not to leak methane anymore. In the period 1990-1999, waste was landfilled in the northwest region of the landfill. This area is currently covered with 1m soil. Since 2000, waste is landfilled at the northeastern part of the landfill. All landfill compartments are physically separated from each other. A road separates the southern and the northwestern compartment. In between the eastern and the western compartments a line of poplar trees is planted.

The waste deposited comprised of organic waste streams (as municipal solid waste and industrial wastes) and inorganic streams as contaminated soil and demolition waste. The mean content of organic carbon of the landfill is calculated to be approximately 79 kg C per tonne waste. A prognosis of landfill gas formation at the moment of measurement is 780 $m^3.hr^{-1}$.

There are several measures in place to reduce landfill gas emissions:

- approximately 470 $m^3.hr^{-1}$ is extracted in two separate systems for landfill gas extraction. This gas contains approximately 50% methane;
- since January 2001, a demonstration is in progress of the Smell-Well system to abate odorous gas and methane emissions (Scharff et al, 2002). This Smell-Well system aerates the slopes in the sectors 6, 7 and 8 in figure 4.9. The system comprises of lances that periodically inject and extract air from the slopes, as indicated in figure 4.8.

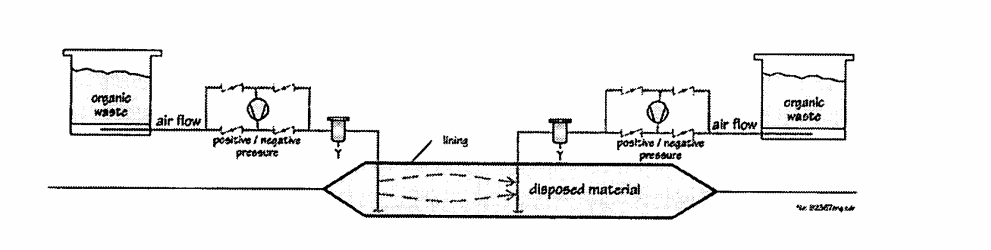


Figure 4.7: Air injection and extraction with the Smell-Well system

The system affects methane emissions in several ways:

- the microbial oxidation of methane to carbon dioxide in the top-layer of the system is enhanced
- the presence of oxygen inhibits further generation of methane; increasing amounts of oxygen result in aerobic conversion of waste, rather than anaerobic conversion;
- the major effect of the Smell-Well system, however is also a rather effective extraction of landfill gas from the surface.

The total emission reduction is estimated to be approximately 75 $m^3.hr^{-1}$. In October 2001 30-50 $m^3.hr^{-1}$ of methane was extracted from the slopes and fed to a biofilter. Since the biofilter is not that effective in reducing methane emissions, part (an estimated 15-30 $m^3.hr^{-1}$) of the emission

reduction from the slopes in sectors 6, 7 and 8 is relocated to sector 9, where the biofilter is located.

4.2.2 Emission measurements

Emissions were measured in October and November 2001. Figure 4.8 gives an overview of all measurement activities.

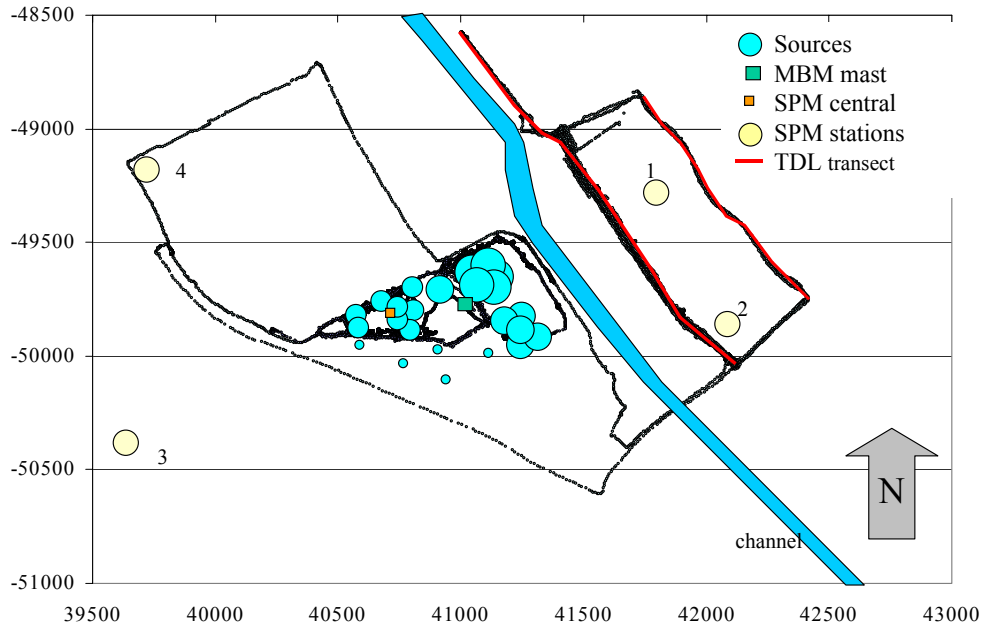


Figure 4.8: Braambergen measurement set-up

The meteorological conditions over at Braambergen are shown in table 4.8.

Table 4.8: Meteorological conditions at Braambergen

Week nr	Date Start	T_{avg} °C	T_{max} °C	T_{min} °C	U_{bar} m s^{-1}	Rain mm
37	3-9-2001	15.4	19.0	11.8	4.0	42
38	10-9-2001	14.8	17.5	11.4	3.4	37
39	17-9-2001	14.2	16.8	10.6	2.8	63
40	24-9-2001	15.9	20.3	10.2	2.5	16
41	1-10-2001	16.8	21.1	13.5	4.4	16
42	8-10-2001	15.3	17.1	14.7	7.1	0

4.2.3 Mass-Balance Method

The sample pole for the mass-balance measurement was located in the middle of the landfill area, on the northwestern compartment, as indicated in figure 4.9. One leaking sampling tube and one defect sampling valve hampered the measurement. So two out of 10 sampling points were defect and

concentrations were estimated through interpolation. For the rest, no technical problems occurred in performing the MBM.

✓ *Determination of emitting area*

Emissions will take place from the darker areas in figure 4.9. The southwestern part of the landfill is capped with an HDPE-liner and is considered leak-tight. In the past, a lot of attention is paid to emissions from the northwestern compartment and on basis of odour nuisance, vegetation damage and box-measurements it was concluded that emissions notably take place through the slopes. More recent box-measurements indicate, some emissions do take place through the top-cover. From the eastern compartments, emissions will take place from both the slopes and the top-surface.

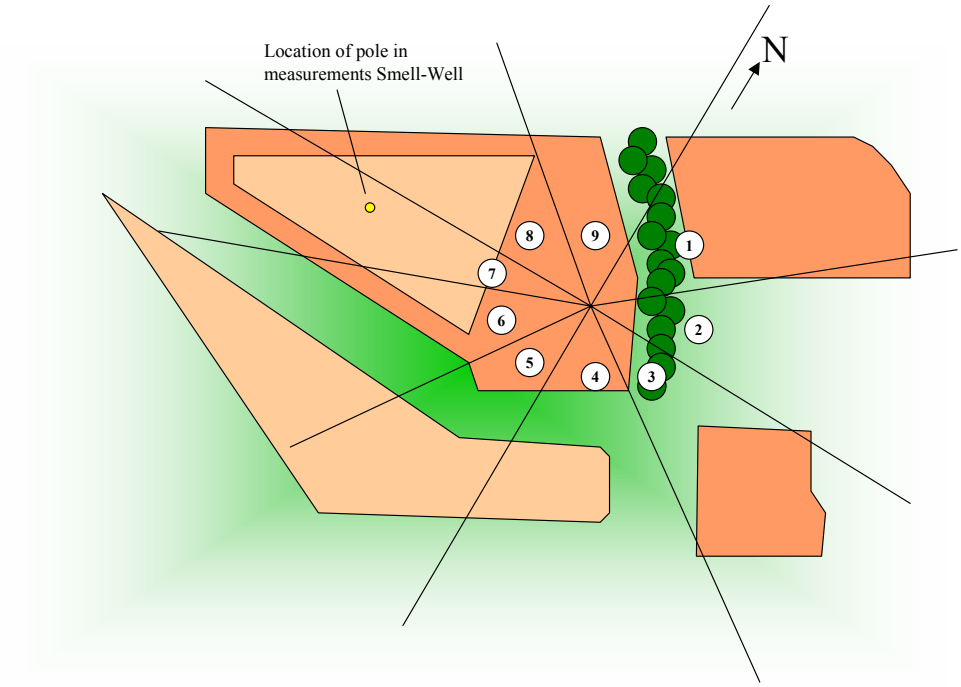


Figure 4.9: Sectors used for the MBM calculations

✓ *Results:*

Assuming emissions to be homogeneously distributed over surface area and slopes from the dark areas in figure 4.9, methane and carbon dioxide emissions are obtained and tabulated in Tables 4.9 and 4.10. When elaborating the results, a large fraction of the profiles from sector 9 to sector 3 could not be used, because the concentration profile was not smooth: background-concentrations sometimes were observed at the low part of the pole. In other cases at the top of the pole, concentrations were measured, significantly above background. As a result the amount of useful profiles from these sectors is very low and the emission estimate from these sectors has to be considered unreliable (in the discussion in chapter 4.2.6, more attention will be paid to the background of these problems).

Table 4.9: MBM results CH_4-emissions at Braambergen.

Sector	X (m)	angle (°)	area (m^2)	CH_4-flux ($l.m^{-2}.hr^{-1}$) mean	SD	n	total flux (m^3 $CH_2.hr^{-1}$)	95%-CI (m^3 $CH_2.hr^{-1}$)
1	233	50	36,354	0.29	0.28	24	10.5	4.4
2	48	40	812	3.10	0.81	3	2.5	1.6
3	184	35	19,743	0.35	0.35	20	6.9	3.2
4	95	55	4,376	1.18	0.93	142	5.2	0.7
5	126	35	4,899	0.80	0.45	126	3.9	0.4
6	30	35	6,000	5.21	5.17	27	31.3	12.3
7	30	20	8,400	3.03	2.75	39	25.5	7.5
8	30	40	7,200	2.77	4.23	96	19.9	6.2
9	141	50	8,763	0.38	0.43	9	3.3	2.9
		360	**96,547**			**486**	**109**	

Table 4.10: MBM results CO_2-emissions at Braambergen.

Sector	X (m)	angle (°)	area (m^2)	CO_2-flux ($l\ m^{-2}.hr^{-1}$) mean	SD	n	total flux (m^3 $CO_2.hr^{-1}$)	95%-CI (m^3 $CO_2.hr^{-1}$)
1	233	50	36,354	0.26	1.00	24	9.3	15.4
2	48	40	812	3.84	1.61	3	3.1	3.3
3	184	35	19,743	1.08	1.94	20	21.4	17.9
4	95	55	4,376	3.50	5.26	142	15.3	3.8
5	126	35	4,899	1.10	3.01	126	5.4	2.6
6	30	35	6,000	7.37	14.10	27	44.2	33.5
7	30	20	8,400	0.06	12.85	39	0.5	35.0
8	30	40	7,200	3.41	11.89	96	24.5	17.3
9	141	50	8,763	-0.21	1.84	9	-1.9	12.4
		360	**96,547**			**486**	**122**	

✓ *Scaling the MBM measurements to a time-series of emissions of the entire landfill*

Using the "up scaling" method described in section 3.2 the Braambergen MBM data was used to make a time series of the total landfill emission estimate. The results for Braambergen are shown in the discussion in figure 4.12.

✓ *Methane oxidation*

Prior to emission to the atmosphere, part of the methane is converted to carbon dioxide. From the ratio of methane and carbon dioxide emissions and the ratio in the landfill gas in the landfill body, below the oxidising top surface, an estimate of methane oxidation is obtained. For this purpose, the composition of the landfill gas that is extracted at Braambergen is used to estimate the composition of extracted gas. The method for correction is described previous for Nauerna in chapter 4.1.3.

Table 4.11: Composition and CH_4:CO_2 ratio of extracted gas at Braambergen (corrected for oxidative processes due to air intrusion)

	extracted composition (vol%)	corrected composition (vol%)
CH_4	50	50.0
CO_2	38	35.4
N_2	12	11.7
O_2	0	2.9

oxygen consumption	2.9 vol%
CH_4:(CH_4+CO_2) in extracted gas (based on corrected values)	0.59

The ratio of emitted methane and total landfill gas emitted (sum of CH_4 and CO_2-emissions) at Braambergen is 0.47 -/-. Methane oxidation can be calculated from the concentration decrease of methane in landfill gas upon emission (so the difference in concentrations between the extracted gas and the emitted gas).

$$\text{Methane oxidation} = ([CH_4]_{extracted} - [CH_4]_{emitted})/ [CH_4]_{extracted} =$$
$$= (0.59-0.47)/0.59 = 19\%^{2}$$

4.2.4 Stationary Plume Method

Two SPM stations were located on the North side of the landfill in two gardens in the domestic area. The station to the west of the landfill located in a garden of an employee of the Stichting AAP, a centre for exotic animals. The animal manure can affect measurements at this station with wind from Northern directions. The station in the South was located at the edge of a farm site. This site had local CH_4 sources too. This was accounted for when selecting background samples. A problem occurred with the communication and the phone that was used to call the stations from the central computer. Therefore the sampling was triggered by telephone from ECN Petten. A total of 84 gasbags were sampled in 21 events. After selection a set of 11 events remained. 6 of which used the results at station 1 for the plume measurements.

Table 4.12: SPM Results Braambergen

Station	Emission (m^3 CH_4 $.h^{-1}$)	Stdev	Nr	95% ci
Station 1	234	11	6	
Station 2	261	--	2	
Station 3	92	--	2	
Station 4	81	--	1	
All samples	277	121	11	71

[2] It has to be noted that this is an estimate of methane oxidation of unknown accuracy: at one hand the method for estimating emissions is not validated; at the other hand the methane oxidation is based a.o. on the measured carbon dioxide emission and this result has to be considered uncertain. See paragraph 5.4 for further discussion on using the MBM for estimating methane oxidation.

4.2.5 Plumes measurement with mobile TDL

The TDL plume measurements were performed on October 3 2001. Wind direction was southsouthwest and the plumes were measured on two parallel transects north of the landfill. The plume shape was remarkably constant over the whole experimental session showing a doublet shape. This is caused by the two compartments at the northeast end of the landfill that are separated by a non-emitting sand plane of approximately 120 m wide. N_2O was released at two locations on the landfill, and the plumes were well detectable. The N_2O model run and measurements are in agreement. For the "nearby" transect that runs through a strip of forest parallel to the channel shown on the map, 17 plumes were obtained. An example of a plume measurement is shown in figure 4.10. The emission result for this set of plumes was 315 $m^3CH_4.h^{-1}$ with a standard deviation of 75 $m^3\,CH_4.h^{-1}$. The 95% confidence limit was 33 $m^3CH_4.h^{-1}$.

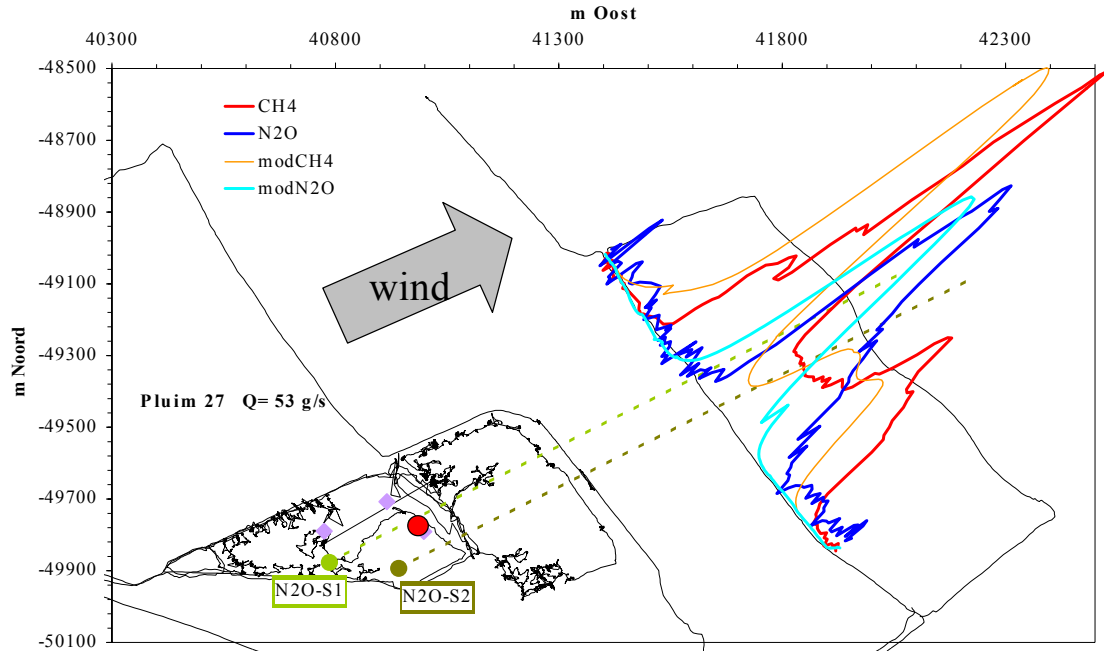


Figure 4.10: Example of the TDL plume measurements to the north east of the Braambergen landfill.

Running the model with the same settings for the transect further away, the three available plumes suggest a lower emission level of approximately 220 $m^3CH_4..h^{-1}$. The reason is that we use a roughness length (z_o=0.3m) in the model that represents the area between measurement and source. For the TDL Campaigns this estimate is adapted to make makes the right fit for the N_2O plumes. The plume at the transect further away passes over a domestic area for which the roughness length is bigger (z_o=1m for example). The houses will cause extra vertical mixing, leading to a decrease of the ground level concentration. The model used cannot separate the travelling plume into two parts with a different roughness. With a roughness length of 1 m the emission level is approximately 270 $m^3\,CH_4.h^{-1}$. So the emission still seems to be somewhat lower although both transects agree within the 95% confidence limit. For the evaluation we assume that the results of the 17 plumes at the first transect provide the best emission estimate. For this transect we also had the N_2O plumes available as calibration for the model.

Table 4.13 : Methane emissions from the Braambergen landfill measured using the TDL.

TDL-meting	**Wind direction**	**T °C**	**U ($m.s^{-1}$)**	**Stab.**	**Nr. plumes**	**CH_4-emissie ($m^3.hr^{-1}$)**
3 Oct	South West	17.8	6	D	17	315 ±33

4.2.6 ^{13}C Isotope measurement

Downwind of the landfill the TDL was used to find locations with a different concentration level. Gas flasks (20L) were pressurised to 20bar over a 10-minute interval. The concentrations in the flasks were obtained from the TDL. The $\delta^{13}C$-values obtained for these CH_4 air samples are shown in Table 4.14 Figure 4.11 shows the relation between the $\delta^{13}C$ and $1/[CH_4]$. The high levels of $1/CH_4$ show the atmospheric background data. The lower values of $1/[CH_4]$ indicate increased contribution of the landfill methane, leading also to lower $\delta^{13}C$ levels. The linear fit to this dataset has $r^2 = 0,78$. The cut off at the Y-axis provides the $\delta^{13}C$ source value of -47,66‰ for the emitted CH_4, with a standard error of 1,23‰.

Table 4.14: Isotope-ratios of the CH_4 air-samples.

Sample flask nr.	**$[CH_4]$ ($nl.l^{-1}$)**	**$\delta^{13}C$ (‰)**
131	3146	-43.81
150	3272	-43.40
136	2175	-40.44
104	2142	-41.95
112	2475	-43.28
139	3808	-45.01
106	1640	-40.73

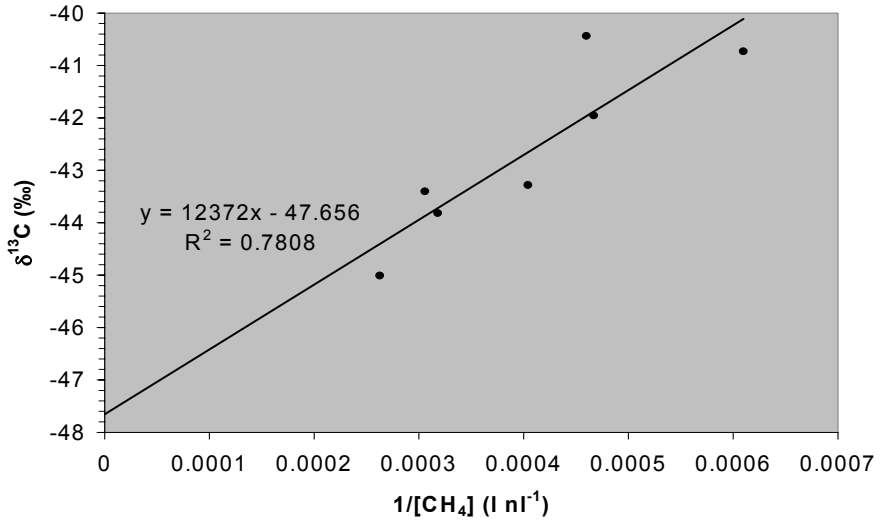


Figure 4.11: Isotope-ratios for CH_4 in the air-samples versus $1/[CH_4]$

The $\delta^{13}C$-value for CH_4 in the pure landfill gas was analysed four times. The average value was -53,42‰, the standard deviation 1,36‰. From that the standard error in the average was calculated to be 0,79‰.
These results indicate a difference $\delta E - \delta A$ of 5,76‰ with a standard error of 1,46‰.

The three measurements performed for the α factor gave resp. 1,01727; 1,01711 en 1,01887. The average of this set is 1,01775 with a standard deviation of 0,00097 and an estimated standard error of 0,00069. The resulting estimate for the CH_4 oxidation level is 32,5%, with a relative standard error of 0,256, which would mean an absolute standard error of 8,3%. From a statistical point of view we have 10 degrees of freedom, (5 for δE, 3 for δA and 2 for α). This results in a 95% confidence limit of [14% – 51%].

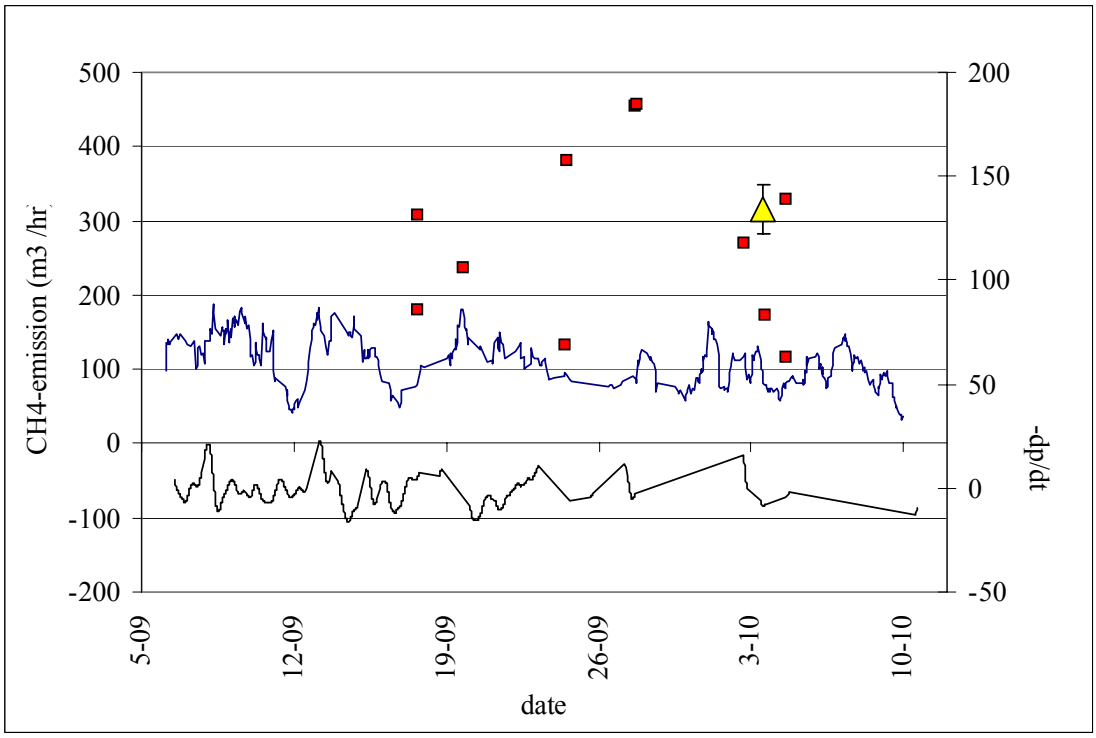


Figure 4.12: Time series based on the MBM data, individual SPM results and TDL results

4.2.7 Discussion

Table 4.15 summarises the results of the measurements at Braambergen.

Table 4.15: Comparison of results at Braambergen

	CH_4-emission ($m^3.hr^{-1}$)	**CO_2-emission ($m^3.hr^{-1}$)**	**Methane oxidation**
Mass balance method	109 ± 30	122 ± 40	19 %
$\delta^{13}C$ measurement			32.5%
Static plume measurement	277 ± 71 (n=11)		
TDL	315 ± 33 (n=17)		
Prognosis	20-195 *2		

1) Indicated is the 95% confidence interval, based on the variations in measurement results. It has to be stressed that this is not the same as the inaccuracy of the method. Model uncertainties might contribute to the uncertainty as well.

2) Based on a landfill gas production estimate of 780±195 $m^3.h^{-1}$, an extraction level of 480$m^3.h^{-1}$ and 75 $m^3.h^{-1}$ extraction by the Smell well system. CH_4 content=50% and 10% oxidation.

The experiments at Nauerna indicated that is that methane emissions are highly variable, with a range of approximately 50% around the average emission level. Therefore the TDL-emission estimate on a single day need not give a good representation of average emissions. At Braambergen a number of TDL experiments were performed over the last 3 years. The TDL emission levels obtained at Braambergen in this experiment can be compared to with the emission levels obtained in the other campaigns. This comparison indicates that the measurements in October 2001 are approximately 40% increased, compared to other measurements. There is no incidental reason known that might explain this increased emission, so it has to be contributed to natural variability of emissions. This raises the question if the TDL campaign is representative for the average emission level over the whole experimental period. The MBM data set shown in figure 4.12 clearly results in a lower emission level, The SPM data however suggest that emissions were indeed higher throughout the experimental period. The combination of these two data sets leads to the conclusion that the TDL campaign data is maybe 10% above the average emission level over the 6 weeks period, but can still be considered representative.

Table 4.16: Other TDL-campaigns

TDL campaign Year	**CH_4 Emission level & 95% confidence interval ($m^3.h^{-1}$)**	**Remarks**
1999 (Nov)	242 ±??	Measured at South side
2000 (Nov)	237 ± ??	Measured at North side 70 % from west part
2001 (Oct)	315 ± 33	Measured at North east
2002 (Oct)	227 ± 38	Measured at West & North West side 40 % from west part.

✓ *The mass balance method*

The MBM gives a relative low emission estimate, compared to the SPM and the TDL. Most likely the MBM was hampered by i) the line of trees at Braambergen, in between the eastern and the western compartments and ii) the point emission of the biofilter. In table 4.8 under 'n' the number of profiles is indicated on which the emission estimate from a sector is based. The emission estimate for the sectors 9 until 3 are based on a relative low number of profiles. A closer look at the raw data reveals this is not the result of adverse wind-direction, nor lack of wind. Most of the profiles could not be used, because either background-concentrations were observed at the low end of the pole or high concentrations were observed at the top of the pole. As a result 80 to 90% of all profiles were left unused in the interpretation of the measurement.

The emission measurement from the western part of the landfill seems to be more successful. The number of profiles available for interpretation was much higher and the resulting emissions seems to be more in line with the results of other MBM-measurements at Braambergen, and previous expectations with TDL:

- As a part of the Smell-Well-demonstration, MBM-measurements were performed, with the pole located more in the centre of the area of demonstration (see figure 4.9). Methane emission from the slopes in sector 6, 7 and 8 were measured to be 80 $m^3\,CH_4.h^{-1}$ (June 2001); 95 $m^3\,CH_4.h^{-1}$ (with smell-well temporarily shut down in January 2002) and 63 $m^3\,CH_4.h^{-1}$ (May 2002), which is in good agreement with what was measured here for those sectors (77 $m^3\,CH_4.hr^{-1}$).
- A TDL campaign in October 2002 indicated that approximately 40% of the total emission of the landfill comes from the western part of the landfill (approximately 125 $m^3CH_4.h^{-1}$). When the east sectors 1-3 are excluded, the MBM measurement yields an emission of 90 $m^3CH_4.h^{-1}$, which is in a reasonable agreement with the TDL estimate for that part of the landfill.

The MBM-emission measurement in this project and the emission measurement in the Smell-Well demonstration were performed at different places on the landfill, but lead to similar results. This might be an indication that the assumption that the slopes are the main emitting surface is correct. If the top-layer would be the main source of emissions, a false assumption on location of emissions would lead to completely different results for different locations of the pole.

✓ *Oxidation*

The oxidation as determined by the mass-balance method is much lower than the oxidation as determined from the ^{13}C-analyses. Of course, the inaccuracies in MBM-emissions estimates as described above will also lead to inaccuracies in the oxidation estimate derived from it. When considering only the emissions from the western part of the landfill (sectors 4 to 8), an analysis is disturbed by the negligible CO_2-emissions measured from sector 7. The CO_2-emission measurement in this sector is most likely hampered by the long distance from the pole to the slopes, where the majority of CO_2 is generated: 380 m. On this distance it is difficult to determine a small increase in CO_2-concentrations, against a background with an analyser that is not well suited for this purpose.

✓ *SPM*

The emission levels obtained with the TDL and SPM system agreed well in this case. The number of 11 successful sampling events with a total 21 collected sample sets 21 was disappointing. The mobile telephone communication between the central computer and the stations did not work properly and therefore we decided to collect the samples by calling the stations from ECN Petten.

4.2.8 Conclusions for Braambergen

✓ *MBM*

At Braambergen, limitations to the mass-balance method became very clear. The existence of large point-sources of methane, or objects that disturb dispersion of the methane plume make the mass-balance method less applicable. For the western part of the landfill where these limitations did not exist, good agreement was found between the mass-balance method, previous results of the mass-balance method and TDL-emission measurements from which methane emissions from that specific part of the landfill could be derived.

✓ *SPM*

The emission levels obtained with the TDL and SPM system agreed well in this case. Alike the TDL emission level, the SPM emission level of 277 ± 71 m^3 $CH_4.h^{-1}$ is also above the average emission level obtained in the TDL campaigns in other years. This suggests that the increased emission level is indeed real. It must be noted however that the number is based on a relative low number of successful sampling events.

✓ *Gas formation model*

The emission levels obtained with the TDL and SPM suggest that the landfill gas production model underestimates the gas production at this landfill.

4.3 3e Merwedehaven.

4.3.1 Description of the landfill

The 3e Merwedehaven landfill near Dordrecht is located in between the river Beneden-Merwede, the nature reserve 'Hollandse Biesbosch' and the industrial area of the city of Dordrecht. The area south of the landfill consists of forest and agricultural fields. Other industrial activities are located to the west of the landfill. The landfill is in operation since 1993. Until 2001 approximately 5.3 million tonne of waste was deposited on a total surface area of 35 ha. In this period no household waste was deposited. In the first few years some of the landfilled fractions contained organic material, such as industrial wastes, refuses derived fuel and demolition wastes. In later years the landfilling of organic wastes was phased out.

The mean content of organic carbon dropped from an estimated 94 kg C per tonne waste in 1993 to approximately 40 kg C per tonne waste in 1998. A further decrease of organic material is observed in the years after. In 2001, the average amount of organic waste is estimated to be approximately 60 kg tonne^{-1}, which is approximately half of the organic content of the waste landfill accepted before current waste policy came into force.

Prognoses of landfill gas formation estimate formation of landfill gas in 2001 to be approximately 2,120 m^3.hr^{-1}. Landfill gas extraction at 3e Merwedehaven extracts approximately 1,350 m^3.hr^{-1}, so a prognosis for landfill gas emissions is 770 m^3.hr^{-1}.

4.3.2 Emission measurements

Figure 4.13 gives an overview of all measurement activities:

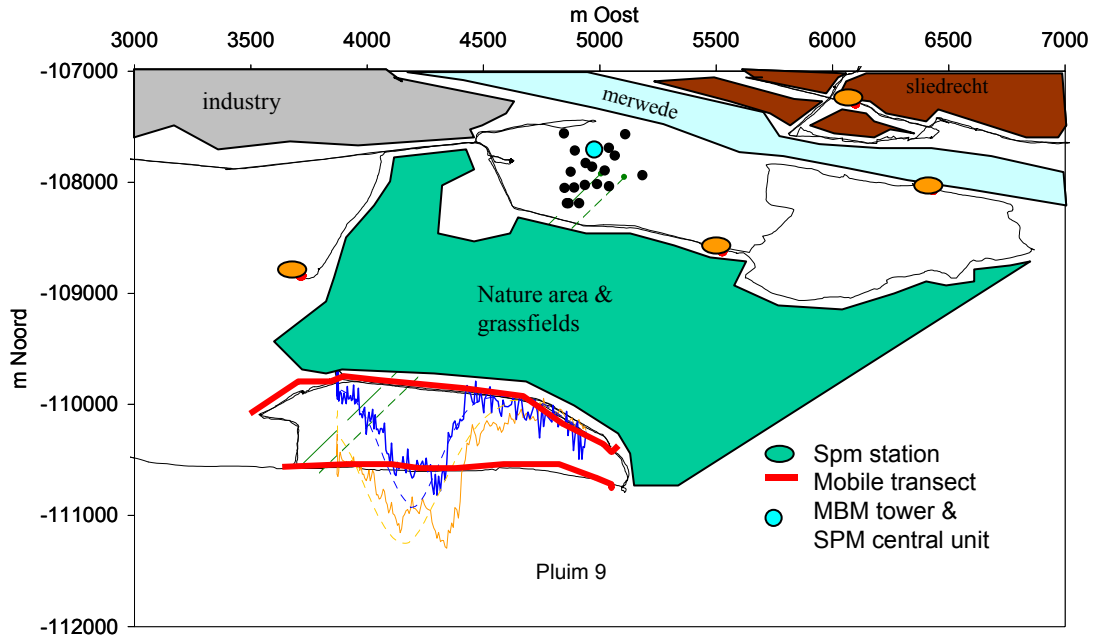


Figure 4.13: Experiment overview 3e Merwedehaven

Emissions were measured in October and November 2001. Meteorological conditions are listed in table 4.14.

Table 4.17: Meteo conditions at Merwedehaven

Week nr	Date Start	T_{avg} °C	T_{max} °C	T_{min} °C	U_{bar} $m.s^{-1}$	Rain mm
42	14-10-2001	12.7	16.6	7.3	3.4	3.4
43	21-10-2001	11.2	13.6	8.3	4.7	12.1
44	28-10-2001	8.8	12.8	2.5	4.5	6.4
45	4-11-2001	4.5	10.4	-3.0	4.4	22.0
46	11-11-2001	5.0	11.0	-4.1	2.4	5.2

4.3.3 Mass-Balance Method

✓ *Technical*

The location of the pole is in the middle of the landfill area, as indicated in figure 4.14. Prior to the measurements at 3e Merwedehaven, it was identified that the valve, connecting sample point 4 (sp4) to the analyser did not function properly. However due to a strict time schedule and the delivery time of the parts required, the valves could not be repaired in time. The effect of the leaking valve on the measurement was considered acceptable, but unfortunately a lot higher than anticipated:

- The methane concentrations obtained from sp4 proved to be rather high.
- Leaking air from sp4 diluted the samples from other sample points, resulting in increased background levels at the top of the pole and reduced concentrations at the bottom. Figure 4.15 gives a mean concentration and wind-velocity profile from one of the sectors. It is clearly seen that there is little or no concentration gradient above 10 m height; however concentrations converge to a limit of approximately 4 ppm, instead of background concentrations of approximately 2,5 ppm.

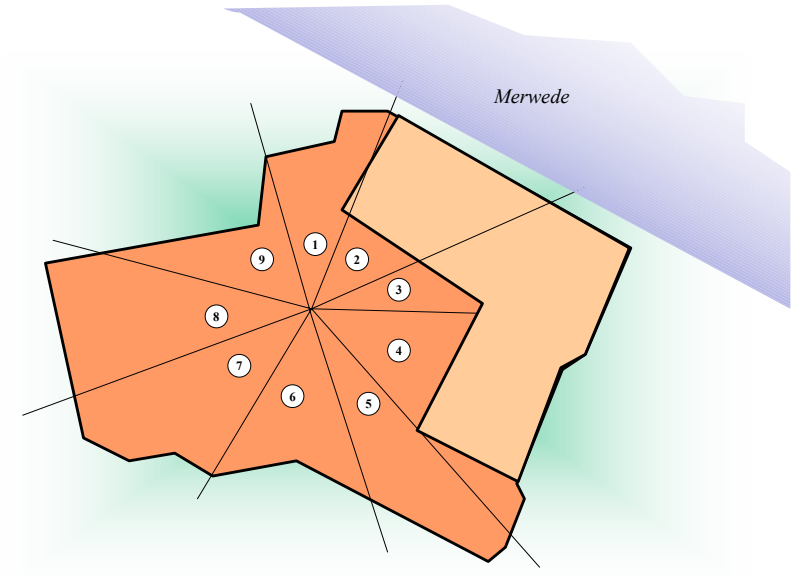


Figure 4.14: Sectors for the mass-balance measurement on the landfill 3e Merwedehaven

So all the measured methane and carbon dioxide concentration profiles were corrected, assuming the following procedure:

- Methane-background concentrations at approximately 0,1 ppm accuracy were obtained from ECN (measurement station Cabauw).
- The amount of dilution with the sample from sp4 is calculated from the measured concentration at sp4, the measured background concentration and the actual background concentration.
- All sample points, except sp4 were corrected, using the measured concentration at that point, the measured concentration at sp4 and the calculated dilution factor.
- sp4 was calculated from interpolation of sp3 and sp5.

In this procedure it is assumed that for a given profile, the degree of dilution for all sample points is the same. This assumption is justified, since for most profiles, the three highest sample points gave approximately the same concentrations, indicating that the degree of dilution of these three sample points is approximately the same.

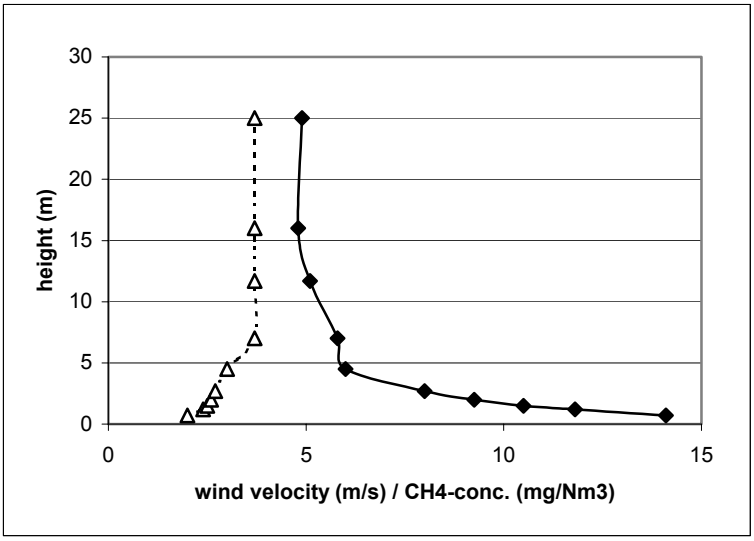


Figure 4.15: Raw wind-speed profiles and methane concentration profiles at 3e Merwedehaven.

Besides the leakage at sp4, the wind-direction measurement failed. Wind directions were obtained from ECN, except for the last two weeks of the measurement period, where wind directions were obtained from a weather station of the Province of South-Holland, located on the landfill site.

✓ *Determination of emitting area*

The dark area in figure 4.14 is the area that is assumed to be the source area. They lighter areas are area in which mud is deposited, and no or negligible emissions are expected from this part. At the other part of 3e Merwedehaven, attention is paid to avoid that the slopes are the main pathways of emissions. Therefore the emissions are assumed to be distributed homogeneously over surface area and slopes.

✓ Results

Tables 4.18 and 4.19 give the results of the MBM.

Table 4.18: MBM results CH_4-emissions at 3e Merwedehaven.

Sector	X (m)	angle (°)	area (m^2)	CH_4-flux (l $m^{-2}.hr^{-1}$) mean	SD	n	total flux (m^3 CH_4 hr^{-1})	95%-CI (m^3 CH_4 hr^{-1})
1	330	40	28,510	0.65	0.42	11	18.5	8.1
2	248	44	17,712	1.43	0.64	17	25.2	5.9
3	337	27	20,069	1.44	0.59	7	28.8	11.0
4	370	44	39,424	1.38	0.74	90	54.4	6.1
5	490	24	37,715	1.36	0.60	47	51.5	6.7
6	342	46	35,214	2.03	1.27	118	71.4	8.1
7	417	40	45,524	1.14	0.94	175	51.8	6.4
8	441	35	44,551	1.04	0.94	75	46.2	9.7
9	266	60	27,786	1.38	0.95	62	38.4	6.7
		360	**296,505**			**602**	**386**	

Table 4.19: MBM results CO_2-emissions at 3e Merwedehaven.

Sector	X (m)	angle (°)	area (m^2)	CO -flux (l.m .hr) mean	SD	n	total flux ($m^3CO_2.hr^{-1}$)	95%-CI (m^3 $CO_2.hr^{-1}$)
1	330	40	28,510	-0.10	6.18	11	-2.8	118.4
2	248	44	17,712	3.03	1.86	17	53.6	16.9
3	337	27	20,069	2.39	2.07	7	48.0	38.4
4	370	44	39,424	2.47	1.56	90	97.4	12.9
5	490	24	37,715	2.57	1.30	47	97.1	14.4
6	342	46	35,214	3.53	2.09	118	124.3	13.4
7	417	40	45,524	2.24	2.14	175	102.1	14.5
8	441	35	44,551	2.05	2.55	75	91.5	26.1
9	266	60	27,786	2.87	4.85	62	79.7	34.3
		360	**296,505**			**602**	**691**	

✓ *Scaling the MBM measurements to a time-series of emissions of the entire landfill*

In order to assess the temporal variation of the landfill gas emissions and in order to evaluate the differences between the different measurement methods, the MBM dataset was used to simulate a "whole-landfill" emission time series. In order to do this the percentile contribution of each sector was calculated based on the whole data set. Then for each individual emission data-point the emission for the whole landfill is estimated by dividing the sectoral emission level by the percentage. A 12 hour running mean curve is made, because an individual profile can not be considered as a reliable single measurement and to enable comparison with a TDL-measurement (which is in fact also derived from a number of TDL-measurements obtained at a single day).

✓ *Methane oxidation*

Upon emission to the atmosphere, part of the methane is converted to carbon dioxide. From the ratio of methane and carbon dioxide emissions and the ratio in the landfill gas in the landfill body, below the oxidising top surface, an estimate of methane oxidation is obtained. For this purpose, the composition of the landfill gas that is extracted at 3e Merwedehaven is used to estimate the composition of extracted gas. The method for correction is described previous for Nauerna in chapter 4.1.3.

Table 4.20: Composition and CH_4:CO_2 ratio of extracted gas at 3e Merwedehaven (corrected for oxidative processes due to air intrusion)

	extracted composition (vol%)	corrected composition (vol%)
CH_4	51.7	51.8
CO_2	42.2	40.9
N_2	5.9	5.8
O_2	0.0	1.5

oxygen consumption	1,5 vol%
CH_4:(CH_4+CO_2) in extracted gas (based on corrected values)	0.56

The ratio of emitted methane and total landfill gas emitted (sum of CH_4 and CO_2-emissions) at 3e Merwedehaven is 0.35 -/-. Methane oxidation can be calculated from the concentration decrease of methane in landfill gas upon emission (so the difference in concentrations between the extracted gas and the emitted gas).

$$\text{Methane oxidation} = ([CH_4]_{extracted} - [CH_4]_{emitted})/ [CH_4]_{extracted} =$$
$$= (0.56-0.36)/0.56 = 36\%^3$$

4.3.4 Stationary Plume Method

The SPM stations south and east of the landfill were situated at remote homes in the recreational area. The station to the south west was located at a municipal service station. The North station was located in the city of Sliedrecht on top of a roof. The problem with the communication between computer and the mobile phone system used to automatically call the SPM stations again occurred. So like at Braambergen sampling was triggered by direct phone calls from Petten. Since the area west and north of the landfill has a number of industrial activities we always sampled all four stations simultaneously to investigate the effect of possible other sources polluting the background levels. The result is 12 complete emission event sets that fulfilled the selection criteria. The average emission level obtained was 690 ± 230 m^3 CH_4.h^{-1} (230 is the 95% confidence interval for this data set) In the evaluation of the data it turned out that the effect of pollution on the background stations was not frequent. It would probably have been a better choice to have more samples with less "quality control" The emission level obtained with the SMP system is significantly above the levels obtained with the MBM system (see the discussion in 4.3.6)

Table 4.21: SPM Results at 3e Merwedehaven

Station	Emission m^3CH_4 .h^{-1}	Stdev	Nr	95% ci
Station 1	253	--	1	
Station 2	--		0	
Station 3	--		0	
Station 4	730	400	11	
All samples	690	400	12	230

[3] It has to be noted that this is an estimate of methane oxidation of unknown accuracy: at one hand the method for estimating emissions is not validated; at the other hand the methane oxidation is based a.o. on the measured carbon dioxide emission and this result has to be considered uncertain. See paragraph 5.4 for further discussion on using the MBM for estimating methane oxidation.

4.3.5 Plume measurements with mobile TDL

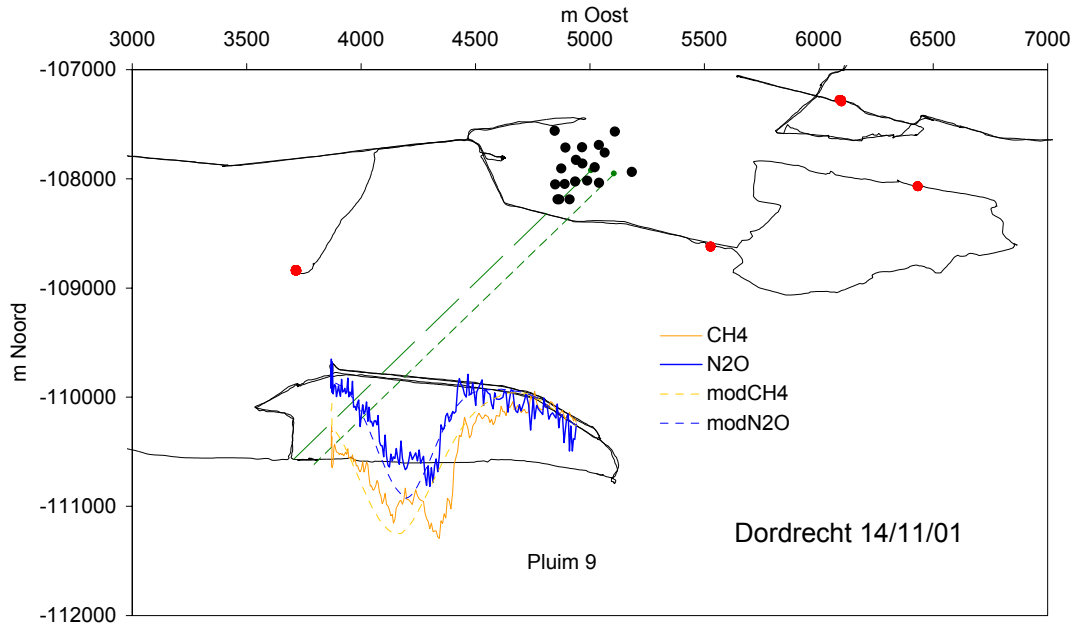


Figure 4.16: Example of a plume measurement at the 3e Merwedehaven Landfill

Table 4.22: The Methane emissions form the 3e Merwedehaven landfill measured using the TDL.

TDL-meting	Wind direction	T °C	U ($m.s^{-1}$)	Stab.	Nr. plumes		CH_4-emission ($m^3.hr^{-1}$)
Nov 14 2001	N		4	C	12		295 ±10

4.3.6 Discussion

Table 4.23 summarises the results of the measurement at 3e Merwedehaven.

*Table 4.23: Comparison of results at 3e Merwedehaven *1*

	CH_4-emission ($m^3.hr^{-1}$)	CO_2-emission ($m^3.hr^{-1}$)	Methane oxidation
Mass balance method	390 ± 100	690 ± 200	36%
Static plume measurement	690 ± 230		
TDL	300 ± 10		
Prognosis	70-700 *2		

1) Indicated is the 95% confidence interval, based on the variations in measurement results. It has to be stressed that this is not the same as the inaccuracy of the method. Model uncertainties might contribute to the uncertainty as well.

2) Based on the gas production was estimated to be 2,120 ± 530 $m^3\ hr^{-1}$ extraction level : 1,350 $m^3\ hr^{-1}$,50% CH_4 contents and 10% oxidation

✓ *MBM*

The MBM-measurement and the TDL are in good agreement, so the MBM-result seems to be quite accurate, despite the re-interpretation of raw data that had to take place to correct for air intrusion. Also when the time-series of the methane emissions is considered (reconstructed from the raw MBM-data is considered, see figure 4.17), a good agreement is observed between MBM and TDL.
It turns out that the resulting time series shows a remarkable good correlation with the change of pressure over time. In figure the small dots show the "up-scaled" emission levels using the MBM time series. The blue line shows the 12 hour running mean curve using these data points. The orange line shows the -dp/dt, the inverted change of pressure over time. Decreasing pressures clearly correlate with an increase in the emission level.

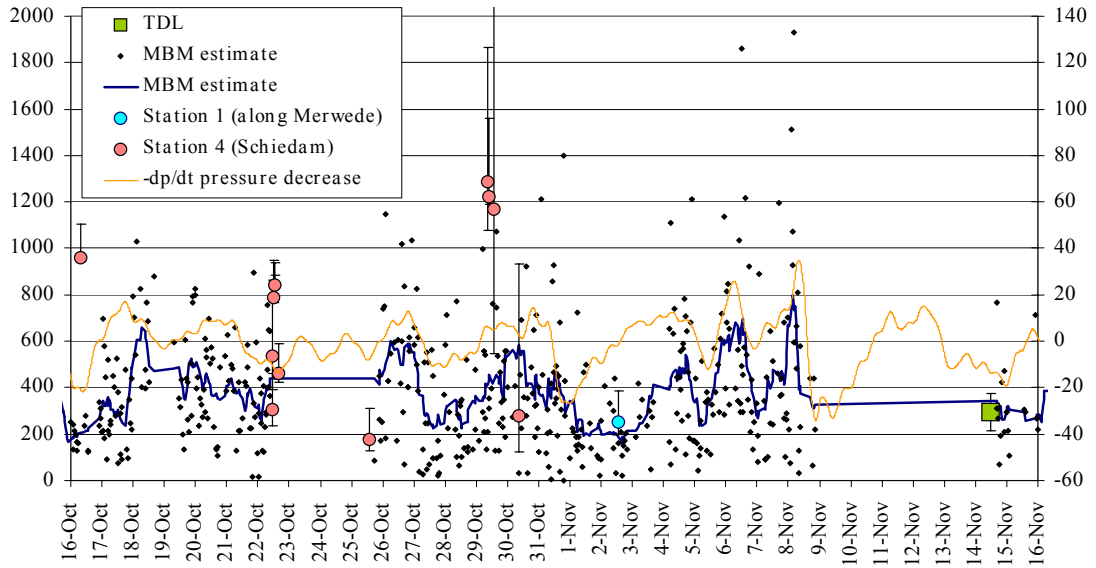


Figure 4.17: Emission results for the SPM station, the TDL campaigns and the time-series obtained from the MBM method. The orange line indicates the –dp/dt that correlates well with the emission level derived from the MBM up-scaling

✓ *SPM*

The SPM does not agree with TDL. A closer look at the results in figure 4.17 shows that the peaks in emissions roundabout October 29th influence the SPM-average. The peak in the emission that is observed on October 29 was evaluated further. In contrast with the event at Nauerna (see the discussion in chapter 4.1), where the MBM-data could not be interpreted when the wind came from the location of the event, the MBM did not give strange results from 3e Merwedehaven. Also for the 29th of October no strange things happened in the gas extraction system. So the reason for the peaks in SPM remains unknown.

4.3.7 Conclusions 3e Merwedehaven

✓ *MBM*

The MBM measurements showed a good agreement with the TDL campaign. There is a good correlation between the time-series of the pressure change (dp/dt) and the emission level obtained from the up-scaled MBM results. On average emissions on days with a pressure drop in the order of

20 mbar/day will show a 50% increase in the emission. This has to be taken into account when measurement takes place during a limited time, for example with the TDL.

✓ *SPM*

The SPM results are relatively high compared to the average level observed with the MBM measurements.

✓ *Gas production estimate*

The estimated emission level based on the gas production model has a large uncertainty range. The production level is believed to be accurate within 25%, the gas extraction level is very well defined. Since the extraction level is high, the relative uncertainty in remaining emission level is large as well. Nevertheless the calculated estimate agrees well with the TDL and MBM measured emission levels.

4.4 Wieringermeer

The fourth comparison of the mass-balance and the static plume method with the TDL plume measurement took place on the Wieringermeer landfill in the period of March and April 2002. Wieringermeer is a relatively small landfill of 18 ha, located in the northwest of the North-Holland province, near the IJsselmeer.

The landfill is in operation since 1985. At the end of 2001, in total 1.6 Mtonne of waste was deposited. At Wieringermeer, industrial waste, contaminated soil, demolition waste and sludges are the main waste inputs. Nonetheless there is significant methane potential, since industrial waste contains significant amounts of organic carbon. The average carbon content of the entire landfill is estimated to be 90 $kg.tonne^{-1}$.

Using the landfill gas formation model of Oonk et al (1994), landfill gas formation in 2001 is estimated to be 790 $m^3.hr^{-1}$. In this period landfill gas was extracted at a rate of approximately 145 $m^3.h^{-1}$, leaving a prognosis of 645 $m^3.hr^{-1}$ of landfill gas emissions. The extracted gas contains 57% methane.

In the west adjacent to the landfill, a biowaste composting plant and household waste transfer station are located. In 2001, approximately 60.000 tonne of biowaste were treated in the composting plant.

4.4.2 Emission measurements

In the period March 10 to April 24 2002 emissions were measured. Meteorological conditions are listed in table 4.23.. Figure 4.18 & 4.19 give an overview of all measurement activities: the four sites for static plume observations were located at distance between 400 and 1200 m away from the landfill. The pole for the mass-balance method was positioned just east of the major emitting waste locations. East of the MBM tower was a 300 m long covered compartment. Adjacent to the landfill on the west side are a composting facility and other wastes handling facilities. These locations will also have CH_4 emissions. At approximately 2 km to the West South-West of the landfill is a major natural gas pumping station. Plumes from this station were accounted for when evaluating the SMP data set. For the measurements at the landfill itself the contribution of the gas pumping station will be small compared to the local sources. TDL-measurements were performed on April 16. Figure 4.18 also shows the measurement transects used for the TDL measurements.

Table 4.24: Meteo conditions at Wieringen

Week nr	Date Start	T_{avg} °C	T_{max} °C	T_{min} °C	U_{bar} $m.s^{-1}$	Rain mm
10	1-3-2002	8.1	2.7	12.9	6	13
11	8-3-2002	9.5	4.8	12.7	6.5	6
12	15-3-2002	9.6	4.3	13.9	6	21
13	22-3-2002	8.1	6.3	9.7	3.4	0
14	29-3-2002	14.8	12.2	17.2	7.5	NA
15	4-4-2002	9	4.2	14.4	6.1	0.1
16	11-4-2002	9.5	7.7	11.7	4.3	16
17	18-4-2002	12.2	8	18	2.8	0.1

NA: not available

4.4.3 The Mass-Balance Method

✓ *Technical*

The pole was erected on top of the landfill surface, as indicated in figure 4.18 and close to a 2 m high heap of sand. After a first problem with electricity supply, the mass-balance method was performed without any problems.

✓ *Determination of emitting surface*

The emitting surface is shown in figure 4.18. Emissions are assumed to originate from the entire landfill, except for the eastern part which is capped with a 2mm HDPE-membrane.

✓ *Results*

The lower part of the methane profile showed an unusual shape, when the wind came from the direction of the sand-heap. But this did not give difficulties in interpretation of the MBM-results in tables 4.25 and 4.26.

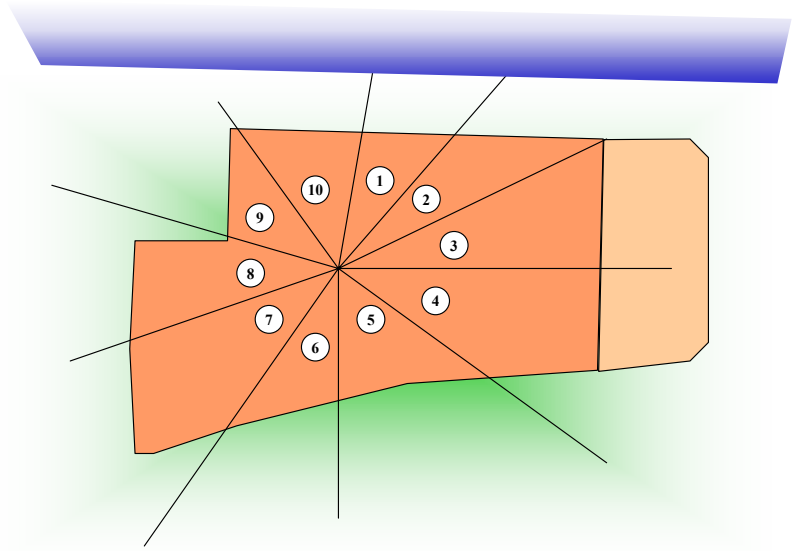


Figure 4.18: Sectors at the Wieringen Landfill used for the MBM calculations

Table 4.25: MBM results CH_4-emissions at Wieringermeer.

Sector	X (m)	angle (°)	area (m^2)	CH_4-flux ($l.m^{-2}.hr^{-1}$) mean	SD	n	total flux ($m^3CH_4.hr^{-1}$)	95%-CI ($m^3CH_4.hr^{-1}$)
1	192	30	9,796	0.50	0.45	64	4.9	1.1
2	285	25	17,986	0.23	0.29	39	4.2	1.7
3	343	25	26,052	0.01	0.12	33	0.3	1.1
4	308	35	29,409	0.05	0.05	125	1.4	0.3
5	190	55	17,587	0.22	0.25	74	3.8	1.0
6	201	35	12,525	1.20	1.43	52	15.1	5.0
7	291	35	26,252	0.49	0.42	75	12.8	2.6
8	231	35	16,543	0.98	0.68	32	16.1	4.0
9	91	40	2,934	3.49	3.01	57	10.3	2.3
10	64	45	1,633	1.02	1.26	17	1.7	1.1
		360	**160,716**			**568**	**70**	

Table 4.26: MBM results CO_2-emissions at Wieringermeer.

Sector	X (m)	angle (°)	area (m^2)	CO_2-flux ($l.m^{-2}.hr^{-1}$) mean	SD	n	total flux ($m^3CO_2.hr^{-1}$)	95%-CI ($m^3CO_2.hr^{-1}$)
1	192	30	9,796	1.09	1.55	64	10.7	3.8
2	285	25	17,986	0.67	0.99	39	12.0	5.8
3	343	25	26,052	0.23	0.99	33	6.0	9.2
4	308	35	29,409	0.52	1.24	125	15.2	6.5
5	190	55	17,587	1.49	2.15	74	26.3	8.8
6	201	35	12,525	1.54	2.16	52	19.2	7.5
7	291	35	26,252	1.37	1.62	75	36.1	9.8
8	231	35	16,543	2.36	4.55	32	39.0	27.1
9	91	40	2,934	6.92	7.96	57	20.3	6.2
10	64	45	1,633	4.13	5.40	17	6.7	4.5
		360	**160,716**			**568**	**191**	

✓ *Methane oxidation*

Upon emission to the atmosphere, part of the methane is converted to carbon dioxide. From the ratio of methane and carbon dioxide emissions and the ratio in the landfill gas in the landfill body, below the oxidising top surface, an estimate of methane oxidation is obtained. For this purpose, the composition of the landfill gas that is extracted at Wieringermeer is used to estimate the composition of extracted gas. The method for correction is described previous for Nauerna in chapter 4.1.3.

Table 4.27: Composition and CH_4:CO_2 ratio of extracted gas at Wieringen (corrected for oxidative processes due to air intrusion)

	extracted composition (vol%)	Corrected composition (vol%)
CH_4	57	56.9
CO_2	37	35.7
N_2	6	5.9
O_2	0	1.5

oxygen consumption	1,5 vol%
CH_4:(CH_4+CO_2) in extracted gas (based on corrected values)	0.61

The ratio of emitted methane and total landfill gas emitted (sum of CH_4 and CO_2-emissions) at 3e Merwedehaven is 0.36 -/-. Methane oxidation can be calculated from the concentration decrease of

methane in landfill gas upon emission (so the difference in concentrations between the extracted gas and the emitted gas).

$$\text{Methane oxidation} = ([CH_4]_{extracted} - [CH_4]_{emitted})/ [CH_4]_{extracted} =$$
$$= (0.61-0.36)/0.61 = 40\%^{4}$$

4.3.4 Stationary Plume Method

The SPM stations were equipped with extra data-loggers during this campaign in order to minimise the loss of samples. Unfortunately there were a lot of problems with the 220 V mains supply at the positions of the TNO tower and our meteo station. This is the reason that in spite of the extra effort we still ends up with a small data set. SPM station nr 1 showed some enhanced concentrations when operating as a background station due to a nearby stable. Also higher background levels were observed at position 3 south of the landfill. Station 3 was moved to a position more to the west in order to overcome this problem. Station 1 was kept in place since it had a good position for the in plume sampling.

Table 4.28 SPM Results at Wieringen

Station	Emission $m^3CH_4 .h^{-1}$	Stdev	Nr	95% ci
Station 1	371	33	2	
Station 2	124	110	2	
Station 3	117	145	5	
Station 4	211	110	3	
All samples	184	142	12	81

4.3.5 Plume measurements with mobile TDL

The TDL measurements took place on the road south of the landfill. This transect is not ideal because the distance to the landfill is relatively small. This is also visible in the TDL emission estimate that has a relatively large standard deviation, compared to the previous campaigns.

[4] It has to be noted that this is an estimate of methane oxidation of unknown accuracy: at one hand the method for estimating emissions is not validated; at the other hand the methane oxidation is based a.o. on the measured carbon dioxide emission and this result has to be considered uncertain. See paragraph 5.4 for further discussion on using the MBM for estimating methane oxidation.

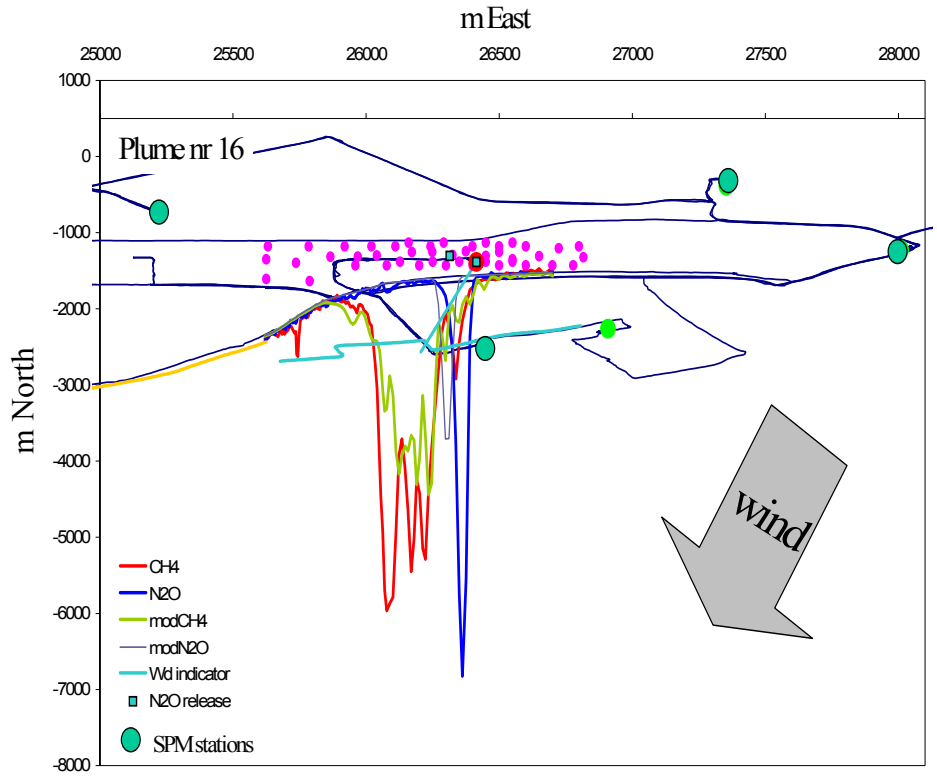


Figure 4.19: Example of a plume measurement at Wieringen .

Table 4.29: The Methane emissions form the Wieringen landfill measured using the TDL.

TDL-meting	**Wind direction**	**T °C**	**U ($m.s^{-1}$)**	**Stab.**	**Nr. plumes**	**CH_4-emissie ($m^3.hr^{-1}$)**	**95% CI**
April 16 2002	NE	10	3.5	D	18	135	32

4.4.5 Discussion

Table 4.30 summarises the results of the measurements.

Table 4.30: Comparison of results at Wieringermeer

	CH_4-emission ($m^3.hr^{-1}$)	**CO_2-emission ($m^3.hr^{-1}$)**	**Methane oxidation**
Mass balance method	70 ± 8	191 ± 35	40%
Static plume measurement	184 ± 81		
TDL	135 ± 32 [2]		
Prognosis Afvalzorg	330 (range 229-432)		

1) Indicated is the 95% confidence interval, based on the variations in measurement results. It has to be stressed that this is not the same as the inaccuracy of the method. Model uncertainties might contribute to the uncertainty as well.

2) The TDL measurements indicate that 25% of the emissions comes from the other activities on the Wieringen site. The emission from the landfill itself would then be 75% of the value reported above.

3) The emission prognosis is based on a production level of 790±200 $m^3.h^{-1}$, gas extraction level of 145 $m^3.h^{-1}$, CH_4 composition of 57% and 10% oxidation. At a 40% oxidation level an emission level of 200 $m^3.h^{-1}$ would be expected.

✓ *TDL*

The TDL plume measurements show that an important part of the emission from the source area does not come from the landfill itself but from the GFT composting plant adjacent to the landfill. In order to estimate the relative contribution of the sources on the landfill end the others we assume that the matrix with sources in the model that explains the plume shape is valid. Then the sources outside the landfill area are selected. This indicates that approximately 25% of the emitted CH_4 originates from the area were the other activities take place.
Assuming an emission factor of 2.5 kg CH_4 per tonne biowaste (DHV, 2000), composting of 60,000 tonne per year results in an emission of 150.000 kg CH_4 per year or 26 CH_4 m^3 hr^{-1}. This is approximately 19% of the total emission measured with the TDL and comparable to the 25% estimated with the plume shape. So methane emissions due to composting seem to be a reasonable explanation for the extra peak in emissions observed.

✓ *MBM*

The MBM is in fair agreement with the results of the TDL: 70 $m^3.hr^{-1}$ versus approximately 100 $m^3.hr^{-1}$ that is attributed to the landfill. Although the TDL measurements clearly show the emissions from area with the composting plant and the biowaste storage. The contribution of these sources is not visible in the MBM data set in the sectors 6 & 7. Apparently these sources lead to a similar and small increase in the concentration at all levels at the MBM tower.

✓ *SPM*

Like the TDL plume method the SPM stations also observe the total emission from the landfill plus the GFT storage and the composting facility. There is good agreement with the results of the TDL: 180 ± 80 $m^3.hr^{-1}$ versus approximately 135 ± 32 $m^3.hr^{-1}$. The implementation of small data-loggers in the SPM stations did work well. Unfortunately due to power supply problems at the landfill, the system worked only part-time at the number of data-points is limited which results in a large standard deviation and confidence interval for this data set.

✓ *Gas production estimates*

The estimated methane emission based on the gas production model, measured extraction and the standard level of 10% oxidation suggests a significantly higher emission level than was observed by the three measurement methods.

Chapter 5.

Discussion, Evaluation of the methods

5.1 Discussion mass-balance method

5.1.1 Practical applicability

The practical applicability of the mass-balance method is proven to be good. This is not such a surprise, since the method was applied in approximately 25 landfill emission measurements prior to this study. So most childhood-difficulties were already dealt with in the past.
There are a few possible drawbacks, but they all posed to be no problem:

- Energy supply was no problem on all four landfills. Electricity was provided either through a connection cable of max 300 m long or using a diesel-generator close to the unit.
- Having a 25 m high pole on a landfill was no problem. Foundation and construction problems were solved, and once erected the pole stood through heavy autumn winds.
- Due to frequent changes in weather in the Netherlands, four weeks mostly suffices to collect sufficient data from all sectors of the landfill.

5.1.2 Operational reliability:

The MBM-measurements at all four landfills was not without operational problems:

- Nauerna and Wieringemeer went relatively smoothly and only initial problems were encountered. As a result more man-hours were required as expected and the start of the measurement was delayed. Problems at Nauerna (breaking guy ropes and failing data-aquisition) can be attributed to the fact that the MBM-measurement is only performed occasionally and has to be rebuilt for every single occasion. Besides, part of the equipment was borrowed from elsewhere and TNO-employees had no experience with part of the equipment. Problems at Wieringen were caused by the poor electricity supply that had to operate both MBM and SPM. Initial problems of all sorts are difficult to avoid when the emission measurement is only performed once or twice per year. However they will be reduced when the method is perfomed more frequently.
- At Braambergen and 3e Merwedehaven, serious problems were experienced due to valve failure. Most likely due to the dusty atmosphere on top of the landfill, the membranes in the valves wear down more rapidly than expected. When the valves break down on the measurement location, this causes problems, because new membranes have several weeks delivery time and cleaning of valves and replacing the membranes is not easily done on-site. Valve failure can be prevented through a more frequent control and replacement of the (relative cheap) membranes in the lab.

The following recommendations are being made with respect to maintenance:

- check of the valves before every measurement;
- replacing the membranes in the valves every 3rd or 4th measurement campaign;
- one annual calibration of wind-speed and wind-direction measurement suffices;
- weekly or two times a week check, whether the measurement equipment is still operational. In this way, problems with the equipment do not lead to loss of too many data.

5.1.3 Accuracy

✓ *Comparison with TDL-measurements*

When applicable, the comparability with the results of the TDL-measurement in general is good:

- At Nauerna, the MBM measured methane emissions to be 527 $m^3.hr^{-1}$. Three TDL-measurements on different days resulted in methane emissions of 900, 550 and 540 $m^3.hr^{-1}$. When the temporal variation of methane emissions is reconstructed from the MBM, the resemblance of results becomes even more striking. The increased CH_4-emission at the first day of TDL-measurement is accompanied by a peek in MBM-measured emission until approximately 750 m^3 $CH_4.hr^{-1}$. That day, approximately 200 $m^3.hr^{-1}$ of methane emissions seems to be caused by an activity upwind from the pole and was not detected by the MBM.
- At Braambergen, the results are less well comparable. The MBM gave an average emission level of 109 ± 30 m^3 $CH_4.hr^{-1}$, whereas the TDL data shows a level of 315 ± 33 m^3 $CH_4.hr^{-1}$. The MBM proved to be not applicable to assess emissions from the area north of the mass-balance method, due to the presence of a point source, close to the pole. The MBM is also not applicable to assess emissions from the eastern compartments, due to the presence of trees in between the location where emissions take place and where air is sampled. When results of MBM and TDL are compared for the section where MBM is accurate, MBM and TDL agree very well again: a mean CH_4-emission measured by MBM of 89 $m^3.hr^{-1}$ from this sector vs. approximately 125 $m^3.hr^{-1}$ for the TDL. It has to be noted, that the TDL and SPM-results for Braambergen seems to be high, compared to other TDL-measurements that are performed at Braambergen in the years 2000-2002.
- At 3e Merwedehaven, the result was good: a mean value for the MBM of 390 ± 100 m^3 $CH_4.hr^{-1}$ and 300± 10 m^3 $CH_4.hr^{-1}$ for the TDL. When the temporal variation in methane emissions is considered the agreement between MBM and TDL is even improved.
- At Wieringermeer the MBM resulted in 70 m^3 CH_4 $.hr^{-1}$, where the TDL yielded an emission from the landfill itself of approximately 100 m^3 $CH_4.hr^{-1}$. The TDL also identified a source west of the landfill of approximately 35 m^3 $CH_4.hr^{-1}$; most likely the composting plant adjacent to the landfill. This composting plant was not observed with the MBM.

✓ *Results depend on assumption of emitting surface*

The results of the mass-balance measurement depend on assumptions on the source region, located either further away or closer to the pole, as explained further in the box below. So this dependency contributes to the uncertainty of the mass-balance method. In practice the uncertainties on location of emissions is not such a big problem as suggested in the box below, because there are always indications on the location of emissions from:

- information in what parts of the landfill more recently larger amounts of organic waste were landfilled;
- information on the quality of covers of the top-surface and slopes;
- information on landfill gas recovery throughout the landfill;
- indications of local hot-spots in emissions as vegetation damages or odour nuisance.

In addition, indications of what area's emit methane can be obtained from other measurement methods, on the condition that they are available. When TDL-measurements or closed chamber measurements are available, the results also might give an indication on the location of emission. When no indications are available, a relative simple measurement of gas concentrations just above the landfill cover gives a distribution of location of methane emissions, as demonstrated at Nauerna (see chapter 4.1)

So concluding: uncertainties about the distribution of emissions over the landfill surface add to the uncertainty of the methane emission, measured with the MBM. However in general a lot of information on location of emissions will be readily available that can be used to improve the emission estimate. On top of that the emission measurement can be improved with a relative small and simple extra measurement activity.

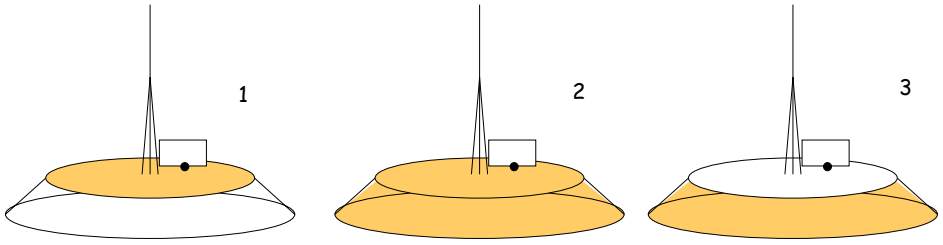
Box: measured emissions depend on assumed emitting surface

The reason for this dependency is the following: at the pole a sort of two-dimensional flux (Φ_{pole} in $l.m^{-1}.hr^{-1}$) of methane, stemming from the upwind part of the landfill, is calculated from the wind-velocity and concentration gradients. The surface flux (Φ_{surf} in $l.m^{-2}.hr^{-1}$) from the landfill is obtained by dividing Φ_{pole} by the length of the emitting area, upwind from the pole (L in m). The total flux from the sector is calculated from Φ_{surf} and the emitting area in the sector (A in m^2). So the total flux from a sector is calculated as:

$$\text{Total flux} = \Phi_{surf} \; A / L$$

and therefore the result depends on the area/length ratio of the assumed emitting area.

The consequences of the assumption are illustrated by the examples below. In situation 1, the top surface is the main source of emissions. In situation 2 both the top-surface and the slopes play a role. In situation 3, only the slopes are assumed to emit methane. The distance to the upper side of the slope is 150 m. The length of the slope is 40 m. The observed Φ_{pole} from the concentration and wind-velocity profiles is 200 $l.m^{-1}.hr^{-1}$. The calculation shows that the estimate of emission is unreliable, when emissions take place from a concentrated area, further away from the pole (e.g. mainly through the slopes) and when this is not incorporated in the calculations.



	1	2	3
Φ_{surf} ($l.m^{-2}.hr^{-1}$)	1,3	1,05	5
A (in m^2)	70 600	113 300	42 700
Total flux ($m^3.hr^{-1}$)	92	118	214

5.1.4 Measuring CO_2-emissions, using the MBM

An advantage of the MBM over other methods is that CO_2-emissions can be measured as well. However, measuring CO_2-emissions is more difficult than measuring methane emissions, because the plume of CO_2-emitted has to be determined against a relative high CO_2-background concentration. For both methane and CO_2, the increase of concentration is typically 50 ppm above background, at approximately 1 or 2 meters height, and a few tenths of ppm at the top of the pole. Measuring this for

CO_2 against a background of 400 ppm is much more difficult than measuring it for methane against a background of 2 ppm.

5.1.5 Limitations to the MBM

The measurements in this study revealed a number of limitations to the mass-balance method. All limitations are connected to phenomena as a result of which the methane concentration profile as observed at the pole is not ideal anymore and therefore rejected in the processing of results:

- Emission measurements are getting less accurate when emissions have to be measured from larger landfills. At larger landfills emissions originating from slopes might disperse to greater heights. As a result the methane plume from the landfill is higher than the pole with which emissions are measured. This introduces two errors, leading to underestimated emissions: (1) part of the methane flux is not detected by the pole and (2) the top-sampling points do not represent background concentrations but show an increased concentration, leading to decreased excess concentrations compared to background at the lower sampling points.
- Larger point-sources, further away from the pole result in concentration profiles observed by the pole that are not ideal. These non-ideal profiles generally have increased concentrations in the middle or at the high-end of the pole. In the current measurement method, such concentration profiles are being neglected and larger point-sources remain undetected in the current set-up.
- In the same way, objects that disturb dispersion lead to non-ideal concentration profiles and therefore unreliable emission estimates. An example of the latter is the line of trees at the Braambergen landfill.

5.1.6 Options for improvement

There are a number of improvements to the current mass-balance method. Improvements in methane concentration analysis make the method more accurate and might help overcome the limitations that are described above. The current measurement method does not produce very accurate absolute levels in concentrations. Differences in concentration, e.g. the difference between concentrations at the lower sampling points, compared to the concentrations at the higher ones are accurate. So when assuming the upper sampling points represents background concentrations, emissions can be calculated, despite the poor absolute quality of the analysis.
When the absolute accuracy can be improved, the accuracy of the measurement is improved and the problems indicated in chapter 5.1.3 are largely avoided, on the condition that background-concentrations of methane are registered separately. This is illustrated in figure 5.1. If the methane concentrations in 5.1 are relatively correct, background concentration is not known, and two errors are introduced: one represented by the area between real background and assumed background (on basis of concentrations in the top of the pole) and one because of part of the methane that is not caught by the pole.
If the methane concentration measurement is more absolutely correct and methane background concentrations are known, the methane flux from the landfill that passes the pole can be determined in a more accurate way and the part that super-passes the pole can be estimated through extrapolation of concentration profiles.

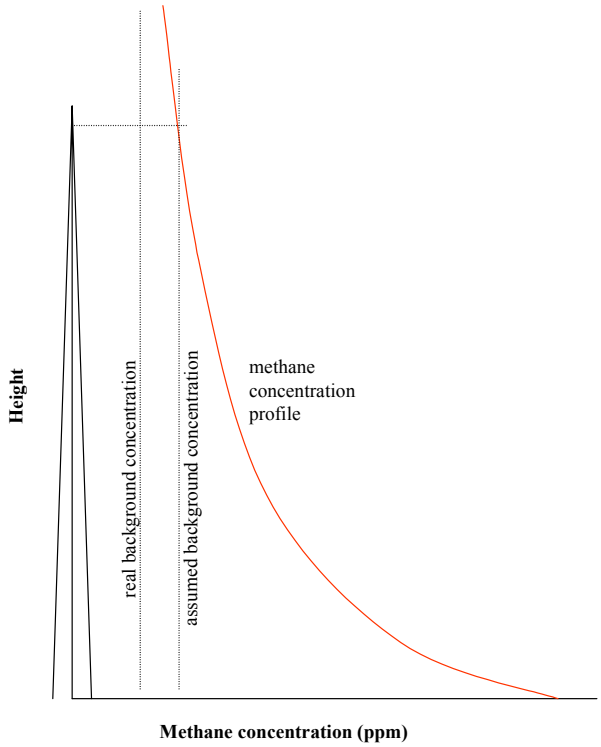


Figure 5.1: A methane concentration profile exceeding the height of the MBM-pole

There are a few options for improvement of the measurement method:

- Using the current Bruel & Kjaer, and introducing a calibration prior to every measurement cycle, rather than calibrating once in a few days.
- Application of improved measurement equipment. Recently accurate and relative cheap analysers (€40,000) for methane have become available, e.g. Gasfinder of Boreal Laser, which is an open path TDL, capable of measuring methane and/or carbon dioxide accurately (Boreal Lasers claims an accuracy in the ppb-range) at atmospheric concentrations.
- Application of sensors for methane (and carbon dioxide). Sensors with sufficient accuracy are not available yet, but sensor technology develops fast and it can be expected that cheap sensors might become available within a few years. At this moment it might be possible to custom-built methane-sensors with sufficient accuracy for the purpose of methane monitoring; however costs associated with custom-built sensors will still be high.

5.1.7 Costs

The costs of the MBM measurement are given in the table below. The table shows the costs of the current set-up, and it speculates about the costs when cheap sensors become available for methane analysis, and when the landfill owner himself is capable of doing large part of the activities associated with the measurement.

Table 5.2 Costs of the mass-balance method

	This study	Future
Investments equipment	€ 70.000,--	€ 35.000,--[1)]
er measurement[2)]	€ 1.900,--	€ 900,--
Hours landfill owner	20	45
Costs advisor	€ 6.000,--	€ 2.000,-- (first measurements); € 1.000,-- (sequential measurements)

[1)] This reduction in costs is based on the assumption that sensors become available for analysing methane concentrations

[2)] Assuming 8 measurements per year, during 5 years

5.2 Discussion SPM method

5.2.1 Practical applicability

The practical applicability of the SPM is proven to be good. The selection of the SPM sampling locations on private owned property did not give any problems. Of course the reward of one apple pie helped a lot. Selection was done in one day a week before the campaigns started. The acceptance of having a small monitoring station in the garden was facilitated significantly by the fact that the system is battery operated and therefore completely self-sufficient. Also the energy supply for the main station is small, this set requires 220V (< 0.2 kW) connection in the landfill site or at one of the sub stations. The handling of the gasbags and the subsequent analyses at ECN went without problems. The sets of gas bags were analysed semi automatically.

5.1.2 Operational reliability:

The SPM setup with wireless communication between the central station and the sampling stations gave a lot of problems during this project. In order to keep the system as low cost as possible, no dataloggers and low cost communication systems were deployed. We now think that investment of a few 100 euro more would have provided a lot of extra samples for this system. The stations worked well but needs the extra datalogger (used during the last session at Wieringen) to list the times that a phone-call actually reached the system. If one call failed now all 7 samples at a station were lost.
The meteo setup and central computing part is simple and robust with no moving parts so little maintenance (except from removing some spiders in autumn). The setup only showed problems at Wieringen when the 220V supply was interrupted frequently for long periods (to long for the UPS set to keep the power up). This was due to reasons outside the SPM system. This could be avoided using a solar-panel/wind turbine set that are available now but normally, providing a stable 220V supply line is not a problem.

5.2.2 Accuracy

✓ *Comparison with TDL-measurements*

The comparability with the results of the TDL-measurement in general is good. Of course both measurement techniques use the same plume concept. The main difference is that the SPM uses one point only in the plume to derive the emission level.

- At Nauerna, the SPM measurements indicated a methane emission of 440 $\pm$ 180 $m^3.hr^{-1}$. Three TDL-measurements on different days, resulted in methane emissions of 900, 550 and 540 $m^3.hr^{-1}$.

- At Braambergen, the SMP, with an emission level of 277 ± 71 m^3 $CH_4.hr^{-1}$ agreed well with the TDL estimates 315 ± 33 m^3 $CH_4.hr^{-1}$.
- At 3e Merwedehaven, the SPM gave a high average emission level compared to the TDL experiment. Mean value for the SPM was 690 ± 230 m^3 $CH_4.hr^{-1}$ and 300 ± 10 m^3 $CH_4.hr^{-1}$ for the TDL. Two high peaks that were observed with the SPM system mainly cause the discrepancy. There was no explanation found for these high peaks.
- At Wieringermeer there was good agreement with the results of the TDL. The SPM average level was 180 ± 80 m^3 $CH_4.hr^{-1}$ versus the TDL estimate of 135 ± 32 m^3 $CH_4.hr^{-1}$ for the whole Wieringen site. According to the TDL experiment approximately 35 m^3 $CH_4.hr^{-1}$ of this source is originating from the composting facility adjacent to the landfill. Because these sources are immediately adjacent, the SPM cannot be used to determine this source separately.

✓ *Results depend on assumed distribution of sources over the landfill.*

The results of the stationary plume method depend on the locations of the point sources in the model that is used. In general the SPM stations are between 300 and 1,000 m away from the landfill location. Sensitivity runs with the model show that in general the effect of a wrong assumption of the main emitting areas on the landfill will lead to an under or over estimation of the emission by 10% on a single station. Since the stations are in different wind directions, underestimation of the emission at one site will automatically mean an overestimation at the site in the opposite wind direction. So provided that enough data points are obtained the effect on the average emission should be smaller than 10%

✓ *Uncertainty due to the measurement of only one point in the plume.*

Whenever the SPM stations measure a plume that is on the edge/shoulder of the plume that comes from the landfill the uncertainty in the emission estimate increases. This means that in theory small landfills are more difficult to measure and large landfills easier. Here the situation is opposite to the situation with the MBM technique that has more problems measuring at large landfills.

5.2.3 Limitations to the method

The measurements in this study revealed a number of limitations to the stationary plume method.

- The main limitation for the SPM was the data availability. With the current performance of the system the datasets that are obtained are too small to resolve the temporal variation of the emission of the landfill.
- Large landfills are easier to measure than small landfills For small landfills in the order of 10-20 ha the distance to the landfill should be approximately 200-300 m in order to have the SPM stations in the plume during a significant time per month, but a smaller distance makes the method more sensitive to the distribution of sources within the landfill area.
- The sampling locations that fulfil the criteria for SPM have to be available. There should be no sources (i.e. cows, stables) in-between the landfill and the station, and no sources near to the station.
- The calculated emission levels become inaccurate when the samples are taken on the edge of the plume. More experience is needed with this method to optimise the sampling strategy.
- The risk of having instruments in the field outside the fence of the landfill is that they can get stolen.

5.2.4 Options for improvement

The SPM was developed within this project and we now have 25 weeks experience with this system. As a result a lot of small technical problems, varying from bursting bags and failing micro pumps at the start, to mains power supply problems at Wieringermeer, the datasets obtained were smaller than foreseen. At the start of the project we decided to keep the sampling stations as low cost as possible. This meant for example that the sampling stations do not have data-loggers on board. It was realised in advance that whenever a sampling trigger is not received, or whenever an extra trigger is received the whole set of 7 gasbags becomes unusable. When using the system for monitoring purposes in other projects the data-loggers are probably not needed. But for the aim of this project: to evaluate the potential applicability of the method, we must conclude that not having data loggers was not wise.

There are quite a number of improvements that can be made. In fact there are two possible ways to go. One way is further technological advancement. Improving the software and hardware of the system is required, so that less samples fail and location specific criteria must be taken into account when to sample and when not. The other option is to simplify the method. In that case sampling with simple self-filling containers at multiple points around the landfill during a day or a night is an option.

In these campaigns the central PC unit was set up on the landfill but for a monitoring set-up this station too could be placed elsewhere. This would save time when visiting the stations and thereby reduce cost.

5.2.5 Costs

Table 5.3: Costs of the SPM

	This study	**Future**
Investments equipment	€ 55.000,--	€ 33.000,--[1)]
Per measurement[2)]	€ 3.000,--	€ 1.500,--
Hours landfill owner	5	40
Costs advisor	€ 10.000,--	€ 4.000,-- (first measurements); € 2.500,-- (sequential measurements)

[1)] A GC is required for CH_4 measurements in the lab (estimated costs € 20,000), samples can also be analysed in an external lab. The sampling stations cost approximately € 2,500 each, the central pc & meteostation costs approximately € 3,000.

[2)] Assuming 8 measurements per year, during 5 years

5.3 Mass-balance method versus static plume measurement

✓ *Both methods are suitable to measure methane emissions*

In this project two methods were developed to measure methane emissions from landfills: the Mass Balance Methods (MBM) and the Stationary Plume method (SPM). The main conclusion is that both options seem to be suitable to monitor the emissions of CH_4 from landfills.

✓ *Specific advantages and disadvantages*

Although both methods are considered suitable to measure methane emissions, they both have specific advantages, disadvantages and limitations:

- *Availability*: although both methods can be improved further, e.g. to overcome certain limitations, both methods have to be considered as being developed, demonstrated and ready for implementation on other landfills.

- *Landfill geometry*: application of the current MBM seems to be limited to landfills with dimensions limited to approximately 400 by 400 metres. The MBM also seems to be less well applicable, when larger object are available on the landfill, that disturb the dispersion of emissions (e.g. the line of trees at Braambergen). Emission measurements from larger landfills seem to be possible, when the methane analysis is improved. This is not readily available, but needs further development and demonstration. In contrast, the SPM requires a certain minimum landfill size in order to be applicable.
- *Accuracy*: both methods are approximately 25% accurate. To achieve this accuracy, the MBM requires knowledge on the major locations of emissions throughout the landfill. When this knowledge is lacking, the MBM becomes less reliable. The result of the SPM is less dependent on landfill specific knowledge and more robust towards assumptions on location of emissions.
- *Additional insights*: when proper CO_2-analyses are available, MBM also yields the CO_2-fluxes. Combination of methane and CO_2-fluxes give valuable information on total landfill gas emission, efficiency of landfill gas extraction and methane oxidation. On top, MBM gives emission estimates from different parts of the system, so information can be gained e.g. from what parts of the landfill improvement of landfill gas recovery can best be achieved. The MBM can be up-scaled to give a time-series of the whole landfill emissions giving insights in temporal variability of emissions.
- *Costs*: the costs for the SPM option is likely to be 10-30% higher compared to the single station MBM set-up since more time is needed for handling of the gasbags etc. The current set-up for the SPM technique was designed for the 4-8 week campaigns in this project. To reduce the costs for an annual averaged emission monitoring, the system should be adapted to sample more plumes in one gasbag or to sample several wind sectors in different bags. This can reduce the handling costs significantly. Reducing handling means less temporal resolution for the SPM data but for annual averaged emission this need not be a problem.

5.4 Applicability of the TDL for measuring average emissions

✓ *A single day TDL measurement is not enough for an annual emission estimate*

The TDL plume method can provide accurate emission levels for a particular day for a complete landfill. Due to the day-by-day variability of the landfill emission level however more sessions will be needed to obtain an annual averaged emission level.

✓ *Specific advantages and disadvantages*

- *Availability*: the plume method is available and has provided emission data for a number of landfills already. The method requires well accessible roads at at least one side of the landfill at a distance between 100 and 1,000 m of the landfill.
- *Landfill geometry*: the method requires well accessible roads at least one side of the landfill at a distance between 100 and 1000 m of the landfill. The method works both for large and small landfills. Measurements can resolve different emitting areas within the landfill in a direction perpendicular to the wind direction Measurements within the landfill area can be used to further resolve the spatial pattern of emissions.
- *Accuracy*: the TDL emission data are 10-25% accurate depending on the location.
- *Additional insights*: the spatial information obtained by the TDL can be used to identify hotspots within the landfill area. Proper selection of the measurement days for a TDL campaign is required. The availability of personnel, the right wind direction, no significant pressure changes, no major disturbance at the landfill (like an extraction system failure) all have to be taken into account. This can sometimes require a significant stand-by period. For the campaigns in this project that lasted four weeks this was sometimes difficult. When the time window for the measurement days is more relaxed this will be less of a problem.

- *Costs:* The costs for a TDL campaign are approximately € 6,000 per campaign, when we assume that 6 campaigns are required to obtain an annual averaged emission level that is representative, this adds up to € 36,000 per location.

5.5 Predictability of landfill gas emissions

Landfill gas formation prognoses can be made using the well-known first order decay model:

$$\alpha = 1{,}87\ \zeta\ A\ C_o\ k\ e^{-kt}$$

In which a is the contribution of the waste landfilled t years ago to total landfill gas formation (in $m^3 \cdot y^{-1}$); 1,87 is the amount of landfill gas per kg organic carbon converted (in m^3.kg); ζ is the dissimilation, indicating the fraction of the organic carbon that is ultimately converted to landfill gas (dimensionless) factor; A is the amount of waste (in tonne); Co is the amount of organic carbon in the waste at the moment of landfilling (in $kg.tonne^{-1}$), k is the decay rate constant of decomposition (in y^{-1}) and t is the time elapsed since landfilling (in y).

In 1994, this model was validated by comparing the observed landfill gas formation with the amount and composition of the waste that is landfilled (Oonk et al., 1994). In a numerical regression, best guesses were obtained of the dissimilation factor (ζ = 0,58 -/-) and rate constant of biodegradation (k = 0,094 y^{-1}). In the validation, assumptions were made on the composition of the waste landfilled, and during application of the model, the same assumptions should be applied (see table 5.3).

Table 5.3: Assumed composition of waste in the first order model (Oonk et al., 1994)

Fraction	**C_{org} ($kg.tonne^{-1}$, wet)**
household waste	136
industrial waste	111
offices, shop and services waste	140
Sweeping waste	129
demolition waste	11
agricultural waste	135
Sludges	90
composting residues	125

The model was validated, comparing observed landfill gas formation with amounts, composition and age of the waste. Landfill gas formation was observed at 9 landfills, and for on average 2 years per landfill. The basis for this observation was the result of landfill gas recovery, using an expert judgement (by the engineering company Grontmij) to assess the collection efficiency. Besides landfill gas formation data were available from 20 emission measurements, performed in the years 1993 and 1994 (Oonk and Boom, 1995). At all landfills, household waste (municipal solid waste) was landfilled. The results of the validation are shown by the blue points in figure 5.3. It can be concluded that the formation model on average predicts landfill gas formation from the waste deposited before 1994 (so before new waste policy was implemented) rather well: in just as many cases the prediction overestimates and underestimates emissions. However the accuracy is limited: individual inaccuracies of 40% are no exception.

The collection of data of landfill generation from waste deposited before 1994 is also a frame of reference to assess whether landfill gas formation models still apply for landfills, since the new waste policy has come into effect. The four landfills where emission measurements were performed in this study are all landfills with little or no household landfilled and mean organic carbon contents significantly below the Dutch average of before 1994. For all four landfills, a prognosis is made of landfill gas generation, using this first order landfill gas formation model and compared to measured emissions and landfill gas recovery (see table 5.4). The resulting formation prognosis and observed formation (= extraction + CH_4-emission + CO_2-emission) are included in figure 5.2. The model uses the average concentrations of organic components in various wastes in table 5.3 and therefore this prediction is most likely not more accurate than the prognoses made by other bureaus that make use of site-specific estimates of carbon contents. In this chapter the results are used to assess, whether in average, waste decomposition has altered in the period 1990 until now. Since we have a prognoses and observed formation available from the period before 1994 derived with this model, this model also has to be applied here in order to come to conclusions.

As discussed in 5.1.3, carbon dioxide fluxes, as measured with the mass-balance method are inaccurate. In 5.1.2 it is concluded that the MBM-measurement at Braambergen is most likely low and is most likely in the order of magnitude of 230 m^3 per hr (interpolation of series of TDL-measurements); consequently carbon dioxide emissions will be at least in the same order of magnitude. Therefore, the comparison in Table 5.4 and figure 5.1 has to be interpreted with great care.

Table 5.4: Prognosis[1)], recovery and measured emissions at the 4 landfills ($m^3.hr^{-1}$)

	LFG-prognosis[1)]	**LFG-recovery**	**CH_4-emission[2)]**	**CO_2-emission[2)]**
Nauerna	2,060	113	527	758
Braambergen	775	465	230[3)]	230[4)]
3e Merwedehaven	2,123	1,350	386	691
Wieringermeer	789	145	100[5)]	191

1) The prognosis is based on a model, using average concentrations of organic components in various wastes (see table 5.3) and therefore this prediction is most likely not as accurate than prognoses that make use of site-specific estimates of carbon contents. In this chapter the results are used to asses, whether in average, waste decomposition has altered in the period 1990 until now; therefore this method is applied.
2) As measured with the mass-balance method; this implies that carbon dioxide emissions are considered to be inaccurate.
3) MBM emission measurement at Braambergen is also not accurate, therefore an interpolation of existing TDL-measurements is used.
4) CO2-emissions estimate on basis of CH4-emission estimate under footnote 2.
5) Based on TDL-measurement and assuming a source of 35 m^3 $CH_4.hr^{-1}$ outside the landfill

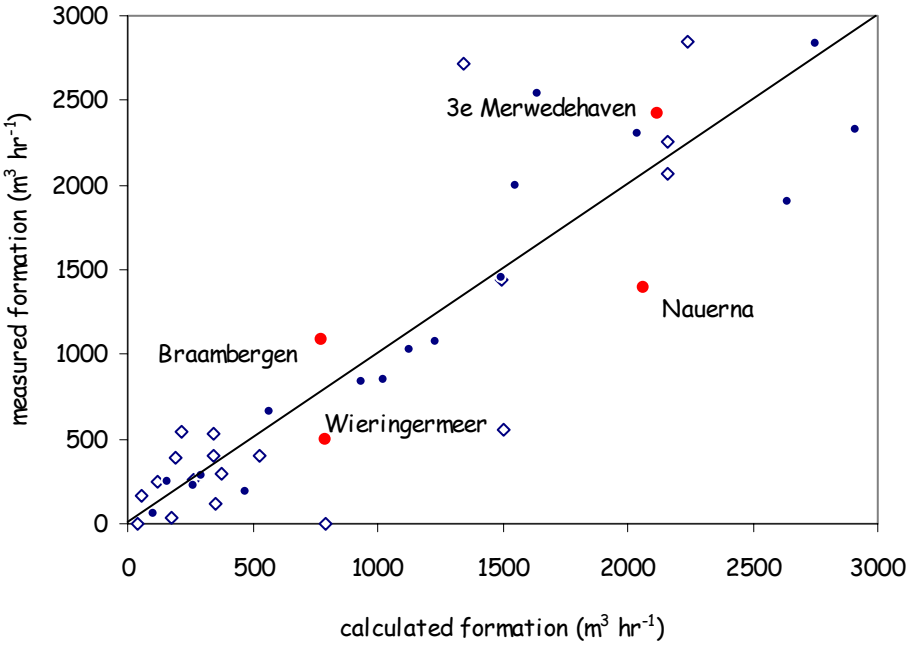


Figure 5.2: Landfill gas prognosis versus observed formation in current study (in red) and in 1994 (estimates from landfill gas recovery in solid blue; estimates from MBM-measurements in white-blue)

Considering the inaccuracies in carbon dioxide measurements, it is difficult to value the result in figure 5.2. However at a first glance, the new landfills (all filled with waste of lowered organic content) do not seem to behave that much different from the older landfills. Neither the mean value, nor the variation around the expected value differs much from the reference group of household waste dominated landfills in 1994. This is a surprising conclusion, since:

- The waste at the newer landfills differ significantly from the reference group in 1994: due to waste policy it contains less organic carbon, less readily degradable organic material, less moisture in the waste. The formation model does take into account changes in organic content, but does not consider changes in speed of degradation (k) or amount of organic material converted to landfill gas (ζ).
- The waste at newer landfills is dominated by industrial waste, where the elder landfills are dominated by household waste. Industrial waste can vary considerably throughout the country, so a larger variation in observed landfill gas formation can be expected for newer landfills. The results in this study do not confirm this.

5.6 Accuracy of measured emission vs. predicted emission

When site specific information on composition of the waste is taken into account, an uncertainty in the landfill gas formation prognosis can be obtained of approximately 25 % for an individual landfill. The methane emission level for a landfill can be calculated from the formation figure using:

CH_4 emission = (CH_4 formation - CH_4 extraction) * (1 - oxidation factor).

Using simple error progression and taking into account the uncertainty in the different variables the uncertainty range in the error CH_4 emission level can be obtained. In general the relative error (in %) for the CH_4 emission estimate is larger compared with the % error in the production level.

Table 5.5: Uncertainty analysis of the CH_4 emission estimate for an individual landfill based on the gas production model.

	Example 1: 50% extraction 40% uncertainty in production model level	Example 2: 50% extraction 25 % uncertainty in production model level	Example 3: 10% extraction 25 % uncertainty in production model level
Prognosis CH_4 formation & uncertainty	1 ± 0.4 *	1 ± 0.25	1 ± 0.25
Extraction & uncertainty	0.50 ± 0.03	0.50 ± 0.03	0.10 ± 0.01
(1-Oxidation factor) & uncertainty	0.90 ± 0.1	0.90 ± 0.1	0.90 ± 0.1
CH_4 emission level & calculated uncertainty	0.45 ± 0.36	0.45 ± 0.23	0.81 ± 0.24
Uncertainty in %	81%	51%	30%

* The uncertainties in gas production modeling and extraction are based on based on available experience within the project team. The uncertainty in the oxidation level 10±10% is an assumption based on experimental data in literature.

The analysis in Table 5.5. indicates that:

- The uncertainty in the CH_4 emission level from an individual landfill is mainly due to the uncertainty in the gas production model especially to the uncertainty in the input data required for that model.
- The relative error (in % is smaller) when no extraction takes place. This does not apply to the absolute error. (And is not a good reason not to implement extraction systems).
- With extraction the uncertainty in the emitted CH_4 for an individual landfill is not likely to be below 50%.

The analysis furthermore shows that emission estimates with an uncertainty range below 25% based on the measurement techniques that are available now are indeed a significant improvement compared to the estimates based on gas production modelling only.

5.7 Methane oxidation

From the methane and carbon dioxide fluxes, an estimate of methane oxidation is obtained. The results of the methane oxidation are shown in Table 5.6.

Table 5.6: Methane oxidation estimates from the mass-balance method

	Estimated methane oxidation	Measurement period	Average temperature and rainfall
Nauerna	38 %	April - June 2001	15 ^{o}C; 50 mm week^{-1}
Braambergen	19 %	Oct.-Nov. 2001	12 ^{o}C; 29 mm week^{-1}
Braambergen ^{13}C	27 %	Oct 2001	
3e Merwedehaven	36 %	Nov.-Dec. 2001	8 ^{o}C; 10 mm week^{-1}
Wieringermeer	40 %	March - April 2002	10 ^{o}C; 8 mm week^{-1}

✓ *Reliability of methane oxidation*

The methane oxidation in table 3.5 is estimated from a) the methane emission, b) the carbon dioxide emission and c) the composition of landfill gas in the waste tip below the zone where oxidation takes place. All aspects are uncertain to some extent, but especially the resulting carbon dioxide emission have to be considered as uncertain (see the discussion of the results of the MBM under 5.2).

The inaccuracy of methane oxidation is not linear to the inaccuracy of carbon dioxide emission; the calculated methane oxidation is especially vulnerable for underestimations of carbon dioxide emissions. E.g. for Nauerna, carbon dioxide emissions that are 25% less, result in 30% less methane oxidation; carbon dioxide emissions that are 25% higher, result in an increase of 20% of methane oxidation.

In any case the levels obtained in this project all show oxidation levels in excess of the 10 % used in the IPCC and National inventory calculations.

5.8 How to reach a 5% uncertainty level in the national estimate

Within the Kyoto protocol it is possible to estimate the emissions of specific GHG sources using nation specific methods. However the method requires international acceptance. The main part of the contribution of landfill CH_4 to the national CH_4 emission could be estimated using actual emission measurements at the largest emitting sites Implementation of such a monitoring system is useful only when a the uncertainty level for the resulting emission estimate is sufficiently low. The ministry of VROM has an objective of estimating methane emissions from landfills with 5% accuracy.

In the coming decade approximately 25 landfills will account for the major part of the national landfill emissions. In order to estimate the sum of 25 sources with a 5 % accuracy we need the error in each if the individual estimates to be below 25%. In order to obtain a representative annual averaged emission level, a monitoring station should be able to provide enough data to cover the variability of the source. The total number of observations (emission estimates) needed over the period depends on the standard deviation of the emission over time. If we assume that the variability of the emission signal for a landfill is approximately 200% of its average emission level we need to have approximately 250 emission estimates in order to get a 95% confidence interval that equals 25 % of the average source level. If the variability of the source is smaller the number of observations needed, decreases. If we can assume that the variability is in the order of 50% 15 emission estimates would be enough to reach the 95% confidence limit of 25%.

Table 5.7: The number of measurements needed to reach a 95% confidence limit of 25%

Relative stdev. %	Measurements needed	95% CI
200	250	25
100	60	25
50	15	25

Based on the results obtained in this project we estimate that the standard deviation in the emission pattern over a whole year is approximately 100%. This would mean that a set of 60 samples taken over the year can provide an emission level of sufficient accuracy. Based on the results from this project we cannot claim to know the seasonal variation in the emission level. So preferably the 60 samples should be spread over the whole year.

Chapter 6.

Conclusions

Two methods are available to obtain annual averaged methane emissions
In this project two methods were developed to measure annually averaged methane emissions from landfills: the Mass Balance Methods (MBM) and the Stationary Plume method (SPM). The main conclusion is that both options seem to be suited to monitor the emissions of CH_4 from landfills.
In four campaigns of 4-8 weeks per location the methane (and CO_2) emission levels were determined at four landfills. The results of the campaigns are summarised in table 6.1.The conclusions were that the SPM and MBM results agreed on average with the TDL mobile plume measurements. Whenever disagreement was found the circumstances in the measurement period could explain the differences. The SPM results at Merwedehaven are not clear with emission levels that were far above the other two methods. At Braambergen the MBM results for a part of the landfill were not reliable due to disturbance of the measurements by a tree line.

Table 6.1: Overview of measurement results at the four landfill locations.

	CH_4-emission ($m^3.hr^{-1}$) *1	**CO_2-emission ($m^3.hr^{-1}$)**	**Methane oxidation**
Nauerna			
Mass balance method	530 ± 130	760 ± 200	38%
Static plume measurement	440 ± 180		
TDL	1st exp: 900 ± 112 *2 (900 = 775+125)		
	2nd exp: 550 ±50		
	3rd exp: 540 ± 115		
Prognosis emission	220-900 *3		
Braambergen			
Mass balance method	109 ± 30	122 ± 40	19 %
$\delta^{13}C$ measurement			32.5%
Static plume measurement	277 ± 71 (n=11)		
TDL	315 ± 33 (n=17)		
Prognosis	20 -195 *4		
3e Merwedehaven			
Mass balance method	390 ± 100	690 ± 200	36%
Static plume measurement	690 ± 230		
TDL	300 ± 10		
Prognosis	70-700 *5		
Wieringermeer			
Mass balance method	70 ± 8	191 ± 35	40%
Static plume measurement	184 ± 81		
TDL	135 ± 32 *6		
Prognosis	229-432 *7		

1) Indicated is the 95% confidence interval, based on the variations in measurement results. It has to be stressed that this is not the same as the inaccuracy of the method. Model uncertainties might contribute to the uncertainty as well.
2) Nauerna: The higher emission observed with the TDL was partially originating from an extra source on the North West part of the landfill. This contribution was estimated to be 15%.
3) Nauerna: Various prognoses are available for landfill gas formation, starting from different assumptions on fractions that contribute to total landfill gas formation, e.g. whether or not sludge contributed to landfill gas formation. The

prognosis is the prognosis form methane emission and is corrected for extraction (85 m^3 hr^{-1} of CH_4 and 10% oxidation).

4) Braambergen: Based on a landfill gas production estimate of 780±195 $m^3.h^{-1}$, an extraction level of 480$m^3.h^{-1}$ and 75 $m^3.h^{-1}$ extraction by the Smell well system.CH_4 content=50% and 10% oxidation.
5) 3de Merwede: based on the gas production was estimated to be 2120 ± 530 $m^3.hr^{-1}$ extraction level: 1350 $m^3.hr^{-1}$,50% CH_4 contents and 10% oxidation
6) Wieringen: The TDL measurements indicate that 25% of the emissions comes from the other activities on the Wieringen site. The emission from the Landfill itself would then be 75% of the value reported above.
7) Wieringen: The emission prognosis is based on a production level of 790±200 $m^3.h^{-1}$, gas extraction level of 145 $m^3.h^{-1}$, CH_4 composition of 57% and 10% oxidation. At a 40% oxidation level an emission level of 200 $m^3.h^{-1}$ would be expected.

SPM & MBM comparison conclusions:
The MBM method and SPM are complementary in the sense that The MBM method has increasing difficulty in evaluation of the landfill emission as the landfill site increases in size. The SPM works better for large sites and has increasing problems with smaller locations. The MBM can miss emission events when they occur in a part of the landfill downwind from the mast. The SPM does not have this problem. Compared with the SPM the MBM provides more insight in the temporal variation of the emission of the landfill site provides that the up-scaling technique that was used in this project is valid for the particular site.

A single day mobile plume measurement is not enough for an annual emission estimate.
Plume measurements with the mobile TDL measurement on a single day gives an accurate emission level for that day but cannot be used alone to obtain an average emission level estimate. This was demonstrated at Nauerna, when an event emission from one part of the landfill increased the total emission level significantly. At least 4-6 days in a year are required when this method is used.
The plume method still has the large advantage that all emissions from the site are taken into account at the time of measurement. For validation of the SMP and MBM measurement methods the data is useful and the only independent method available at the moment. It would have been better to have more mobile campaign data available which means that the per measurement costs should be reduced.

Oxidation level of 10 % used in the inventory calculations seems to be to low.
Although the accuracy of methane oxidation, as obtained with the mass-balance method, could not be validated, methane oxidation seems to be higher than the 10 % default value used in IPCC and national inventory calculations. All measurement, including the more accepted ^{13}C measurements showed CH_4 oxidation levels in the order of magnitude of 20-40%. Methane oxidation is not easy to assess and when attempts for a further increase CH_4 oxidation in the top cover of the landfills continue, development and validation of methods to assess actual oxidation level need further attention.

The landfill gas production models seem to be still valid.
For the four landfills in this study, no significant different relation was observed between modelled and measured landfill gas emission compared to the dataset obtained in 1993-1994: **on average** this model seems to describe landfill gas formation at these landfills well and the inaccuracy in results seems to be similar to predictions of landfill gas formation from waste, landfilled before 1994. This suggests that the landfill production models are still valid and can be used, in spite of the changes in waste composition and landfill management over the last decade.

Methane emission measurements are more accurate than predicted emissions
Despite the fact that landfill gas formation can be rather well predicted, the accuracy of a predicted methane emission is much less, due to error propagation when calculating emissions as formation minus landfill gas recovery minus oxidation. The uncertainty range in such an emission estimate is large and can range up to 100 %, so methane emission measurements can improve the assessment of emissions of a single landfill significantly.

An annual emission estimate for a single landfill with an uncertainty range below 25% can be obtained.
For an individual location approximately 60 emission measurements spread over the year are required to obtain an estimate of the annual average emission level with an uncertainty range below 25%.

A 5% uncertainty range for the national emission level is possible
Emission monitoring at 25-30 landfill sites in the Netherlands can cover the main part of the CH_4 emission and lead to an emission estimate that has an uncertainty range of approximately 5%.

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Appendix 1: Papers and abstracts

The progress and results of this project have been presented at several symposia: the First Intercontinental Landfill Research Symposium (December 2000, Luleå, Sweden), Sardinia 2001 (October 2001, Cagliari, Italy) and the Second Intercontinental Landfill Research Symposium (October 2002, Asheville, USA).

The presentations were invariably welcomed with enthusiasm. Some work on methane emission measurements is carried out in France, Denmark and USA. A measurement programme taking several years is carried out in Sweden. But in general there is an enormous lack of measurement data. This hampers understanding of processes that drive or may reduce methane emissions. The approach to compare and validate modelling and measurement data was considered very valuable. Also the idea to develop simple and easy to apply measurement methods that can be validated with more sophisticated plume methods was welcomed. It was encouraged to publish the results of the project in a peer reviewed journal to open the data for the entire scientific community.

In October 2002 in Asheville the CLEAR working group was established. CLEAR (Consortium for Landfill Emissions Abatement Research) is an international working group of the International Waste Working Group (IWWG). CLEAR coordinates interdisciplinary research on the quantification and mitigation of landfill gas emissions to the atmosphere. The participants in this project were invited to become members of the CLEAR working group.

First Intercontinental Landfill Research Symposium, December 2000, Luleå, Sweden

EMISSION MEASUREMENTS AS A TOOL TO IMPROVE METHANE EMISSION ESTIMATES

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Summary:

Application of IPCC-methodology most likely results in an overestimation of methane emissions from Dutch landfills. The Dutch government could benefit from better methods to quantify the national landfill methane emission. Two strategies are identified: 1) improvement of the present method; 2) estimates based on measurements. Deviation from IPCC-methodology is acceptable only if: a) based on a large number of real-scale observations; b) an accurate impression of the annual average is generated; c) base-line correction is applied. For an accurate annual average the authors recommend 3-4 campaigns of 1-3 weeks per year. Application of sophisticated methods (e.g. TDL) will be to expensive to determine a national annual average. Landfill operators could benefit as well and assist the government in obtaining emission data for both strategies. This however requires cheap and robust methods, possibly supervised by the landfill operator. The mass balance and stationary plume methods are proposed for further development to meet these requirements.

Keywords:

landfill; landfill gas; methane; emission; measurement; method; development

1. INTRODUCTION

1.1 General

In the Netherlands a lot of knowledge is gathered about landfill gas formation and design and operation of landfill gas extraction schemes. This knowledge has lead to the development of landfill gas formation models (Oonk et al., 1994), which proved to be a sound basis for the design of many landfill gas extraction schemes and are at the basis of the estimate of 1990 methane emissions from Dutch landfills. But all this knowledge is based upon the traditional municipal solid waste, landfilled before the waste policy affected amounts and composition of waste landfilled. Most likely under these changed conditions waste decomposes less rapidly and less complete, thus complicating the assessment of landfill gas generation rates. It is not clear whether existing landfill gas formation models still apply for the newer generation landfills.
Development of improved landfill gas formation models or other models to assess landfill gas formation and methane emission is not straightforward both for technical and procedural reasons. This paper describes some possibilities to improve current methods and depicts the role emission measurements can play in method development.

1.2 Dutch waste policy

Current waste policy in the Netherlands aims at both the reduction of amounts of waste to be treated as well as reducing the environmental effects of waste treatment. With respect to methane emissions of landfills, the most important developments are:

- the separate collection of paper and organic materials at households is almost fully implemented and quite effective at the moment;
- the capacity of municipal solid waste incineration was doubled in the last decade, so alternatives to landfilling are being developed;
- landfilling of combustible wastes is in principle forbidden and special allowances are required to continue the landfilling of this waste; on top of that landfill taxes are being superimposed thus rendering incineration price-competitive with landfilling.

As a result it is expected that the amount of organic waste that is ultimately landfilled is going to decrease significantly (AOO, 1999). On top of that other important factors that influence biochemical processes in landfills will change as well. The humidity of the landfill will drop, since a large part of the water in the landfill comes in with the organic waste. The nature of the organic material will change from relatively rapidly degradable to a more woody, less well degradable nature. The sharp initial increase in temperature will be less due to the lack of rapidly degradable materials.
In the Netherlands a lot of knowledge is gathered about landfill gas formation and design and operation of landfill gas extraction schemes. This knowledge has lead to the development of landfill gas formation models, which proved to be a sound basis for the design of many landfill gas extraction schemes and are at the basis of the estimate of 1990-methane emissions from Dutch landfills. But all this knowledge is based upon the old traditional household waste, landfilled before this new policy came into effect. It is not clear to what extent this knowledge still applies for the newer generation of landfills. Most likely under these changed conditions waste decomposes less rapidly and less complete, thus complicating the assessment of landfill gas generation rates.

1.3 Dutch government

The Dutch government under the Kyoto protocol is obliged to assess and report its emissions of greenhouse gases, a.o. methane emissions from landfills. Of special importance are the 1990-emissions, the 2010-emissions and the emission reduction achieved in the period 1990-2010. At the moment reliable forecasts of expected 2010-emissions are of utmost importance for policy definition.

The current estimate of 1990-emissions proceeds according to the first-order decay model as internationally accepted, described in the internationally accepted IPCC 1996 Revised Guidelines (IPCC, 1996) and can its result can be regarded as satisfactory. The effect of a decreased amount of organic materials to landfills is well monitored and its resulting methane emission is expressed in the Dutch emission estimate. The effect of changed conditions on landfill gas formation and methane emissions *is not*. The use of the existing methodology in quantifying current and 2010-emissions will most likely result in an overestimation. Simultaneously the emission reduction achieved in the period 1990-2010 will be underestimated.

Another problem of the existing methodology for estimating methane emissions from landfills is that it is not able to monitor some specific measures that are proposed for further going emission reduction. An example of this is the enhanced oxidation in top-layers. This option is widely recognised as a promising method to reduce methane emissions (ECN/RIVMVROM, 19981999). Emission reductions achieved in this way can at present not be monitored, due to lack of a suitable monitoring methodology (either a measurement method or an accepted reduced emission factor) and the results obtained can not be incorporated in the national assessment.

1.4 Landfill operator

Landfill operators with ISO-14001 certified environmental management system have taken the obligation to monitor their emissions and also to strive for a continuous improvement of their environmental effects. Afvalzorg has a detailed overview of the characteristics of the landfilled waste at most sites. Up to 60 different species of waste are defined and a 3-D database is kept up to date of what waste is where in the landfill. With these data making a fair prognosis of landfill gas formation should be no severe problem. But the waste composition at the landfills of Afvalzorg differ from what was landfilled in the Netherlands before 1990, so landfill gas formation models don't apply anymore and methane emissions are hard to predict. The results of a number of emission measurements show methane emission levels that deviate from the expected ones (see chapter 4). Besides for monitoring purposes, information on methane generation is also of importance for design and operation of landfill gas recovery schemes. Less accurate prognoses of landfill gas formation may lead to over- or under dimensioned extraction schemes and utilisation equipment and may render a project economically unfeasible.

Afvalzorg therefore seeks for ways to frequently or continuously monitor the emissions from its landfills. Besides, Afvalzorg is interested in improved prognoses of landfill gas formation in its sites to enable improved design and operation of their schemes for landfill gas recovery.

2. STRATEGIES TO IMPROVED EMISSION ESTIMATES

2.1. Current methodology

The current methodology for estimating Dutch methane emissions is based on the material balance:

$$\text{emission} = \text{formation} - \text{recovery} - \text{oxidation}$$

Formation is calculated using a first-order decay model as proposed by IPCC (1996), using the default rate-constants for biodegradation, and applying specific input-parameters for carbon-content of the waste and dissimilation. With regards to 1990-formation this methodology can be considered about as accurate as possible with existing knowledge. With regards to formation in waste, landfilled after 1990, due to the reasons described in chapter 1, existing models are not suited to make an accurate estimate of methane formation in waste landfilled after 1993.

The amount of recovered methane from the landfill is well monitored and accurate compared to the other factors.
The uncertainty in the oxidation efficiency of the landfill cover on the contrary is large. At the moment the methodology was made only little information was available on this topic and still it is difficult to extract a reliable oxidation factor from field data. One key problem is that no large large-scale measurement programme has been conducted. Estimated oxidation capacities on a larger number of landfills related e.g. to site specific parameters, or seasonal conditions are not available.

2.2 Two strategies for the national government

In a study for the Dutch Government (Scharff et al., 2000) two strategies were identified to improve the emission estimates. Performing measurements of emissions at real landfills is an important part of both strategies:

- In the first strategy, measurements are used to improve current methodology based on modelling landfill gas formation and insight in methane oxidation. Improving the existing methodology can imply both improving landfill gas formation models as well as improving oxidation factors.
- The second strategy ultimately aims at an emission assessment based on frequent or continuous measurement of methane emissions at all relevant Dutch landfill sites. In 2010 a limited number of landfills will be responsible for the major part of Dutch methane emissions: over 95 % will be caused by a group of 25 landfills. This implies such an apporach to the inventory of Dutch methane emissions from landfills might be feasible, on the condition that an accurate measurement methodology is available, that is affordable as well.

It must be stated however that whatever the choice is to obtain future estimates for CH_4 emission levels from landfills, a combination of both model evaluation and measurements will be needed. If is chosen to update the methane formation models in order to improve the emission inventory, new measurements are needed to see if the models are able to describe the current landfills now. If alternatively it would be decided to evaluate the emissions in 2010 using emission measurements at the individual landfills, the new emission data will provide information that can be used to improve the available models. This might lead to a revision of the emission estimates for 1990.

An important prerequisite of the national emission estimate is that it is accepted in international negotiations. This means that the methodology and parameters used should meet certain requirements. Methodologies and default factors for model parameters are defined in the ‘1996 IPCC-Revised Guidelines’ and the forthcoming ‘IPCC-guidelines on good practice’. Use of other model parameters compared to the defaults and even other methods than the methods defined by IPCC is possible on a few conditions:

- the result should be an improved, more accurate emission estimate;
- the definition of methodology or model parameters should meet certain standards of quality control: it must be based on a number of observations on real landfills; it must be able to withstand criticism of international experts; results should preferably be published in double-peer reviewed journals;
- attention should be paid to base-line correction: any change in methodology or model-parameters along the way must be accompanied by considerations about the necessity of adapting the methodology of estimating 1990-emissions as well.

So any attempts of the Dutch government to improve their emission estimate is subject to rather strict preconditions, which imply that every activity and result should be carefully communicated with e.g. international experts, IPCC and UN-FCCC.

2.3. Landfill operators

If an accurate method to measure emissions and estimate oxidation becomes available to landfill operators, emissions can be monitored directly and landfill gas formation (and the amounts of methane that might be recovered) can be calculated from emissions and oxidation. In case the method acquires international acceptance the results could be used by the national government for national annual emission estimates.

Sofar methane emission measurements have not been a widely applied tool to quantify landfill emissions. Therefore any method will be acceptable to inform the local authorities and the general public about landfill methane emissions, either direct or through an environmental annual report. Nevertheless scientific acceptance will be helpful.
Afvalzorg considers to use a low cost and robust monitoring system in order to monitor the methane emissions at their landfills for the years to come. This system should improve the evaluation of the annual emission levels for the different sites. Since formation might be estimated from the sum of methane and carbon dioxide emissions, it would be extremely useful when carbon dioxide can be measured as well.

3. VARIABILITY OF EMISSIONS

Over the last few years quite a number of experiments emission measurements were performed at landfills (see Table 1). Experiences in literature indicate that among others landfill gas is emitted with high spatial and temporal variability:

- The emission per m^2 on a single landfill shows a variability of three orders of magnitude.
- Emissions from landfills with comparable size can be different by about an order of magnitude.
- The oxidation of the top layer, and therefore also the CH_4 emission of landfills shows a seasonal variation.
- The amount of emitted methane is depending on meteorological conditions: temperature, rainfall and pressure changes.

Table 1: Some observations obtained from literature

Observation	Author
Spatial variation:	
Up to factor 1,000 difference between box measurements	Verschut et al., 1991
No correlation between two box measurements >6m apart	Czepiel et al., 1996a
Up to factor 500 difference between measurements	Nozhevnikova et al., 1993
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4. LIMITATIONS OF MEASUREMENT METHODS

In literature various methods for measuring methane emissions from landfills are described:

- Emissions can be calculated from concentration profiles in soil cores (Bogner and Scott, 1995).
- Static and dynamic closed chambers can be used to sample a relatively small part of the landfill surface (e.g. Bogner and Scott, 1995; Maurice and Lagerkvist, 1997; Perrera et al., 1999).
- Mass-balance or micrometeorological methods give concentration profiles on top of the landfill from which emissions can be obtained from a larger part of the landfill (Oonk and Boom, 2000; Savanne et al., 1997).
- On the landfill or further away from the landfill plumes can be determined to obtain emissions from the entire landfill (Czepiel et al., 1996a; Galle et al., 1999; Scharff and Hensen, 1999).
- $\delta^{13}C$ isotope measurements are proposed to determine methane oxidation in top covers (Boeckx et al., 1997)

For a description of the methods the authors would like to refer to the literature mentioned in this and the previous chapter. Sufficient temporal and spatial resolution in order to deal with the variability as described in chapter 3 can be considered a prerequisite for application of a measurement method. Table 2 indicates applicability, advantages and disadvantages of the various methods.

Table 2: Comparison of measurement techniques

technique	spatial resolution	temporal resolution	component	costs	experiences (world-wide)	other advantages/draw-back limitations
soil core	m^2	hour	CH_4, CO_2	high	few	especially suited for mechanistic studies of oxidation, possible interference with normal landfilling activities
closed chambers	m^2	hour	CH_4	high	many	many samples required to obtain emission from an entire landfill, possible interference with normal landfilling activities
mass balance	few ha	continuous	CH_4, CO_2	moderate	few	well-suited for automation
micro-meteorology	few ha	continuous	CH_4, CO_2	moderate	few	demonstrated not to be applicable
plume measurement	entire landfill	hour	CH_4	high	some	considered most accurate
isotope measurement	entire landfill	hour	$^{13}CH_4$	very high	some	intended to measure amount of oxidation

When emissions are to be measured using closed chambers, the low spatial and temporal resolution requests a large number of relocations (more than 30 a day) on several days throughout the year (no experience how many measurement days are required here). This makes this method very labour-intensive and very expensive.

The mass-balance method seems to be better suited to measure emissions from larger surfaces during longer times. Its capability to measure CH_4 and CO_2 gives insight in the primary processes leading to emissions: methane formation and oxidation. For larger sites however, this method might bring about some problems and further developments are required to enable measurements from the whole of a larger landfill site. Developments might comprise the application of longer pylons (16 meter pylons are commercially available) and the application of more accurate CO_2-analysers. Draw-backdrawback of the mass-balance method is that since there is not so much experience with the method, validation might be considered a requirement.

Plume measurements with TDL or FTIR technology can be considered the most accurate methods to measure emissions from an entire site. But the draw-backsdrawbacks of this method (complexity, low temporal resolution and high costs of prolonged measurement campaigns) render this method not suitable to give a reliable impression of the annual emission. Since these plume measurements gives an indication of emissions at a single day, its costs will in practice reduce its temporal resolution. But since the method is generally accepted as being accurate it might be the best method for validation of other methods. The last drawback of the plume method may be avoided when a suitable stationary plume method based on gas sampling can be developed.
$^{13}CH_4$ -measurements are widely recognised for their applicability in quantifying the amount of methane oxidised in the top-layer, so this method might be the primary candidate to validate the suitability of the mass-balance or stationary plume method to get an impression of methane oxidation.

5. RECENT EMISSION MEASUREMENTS IN THE NETHERLANDS

From 1997 onwards Afvalzorg has carried out methane emission measurements at several landfills. Apart from quantifying emissions an additional objective was to gain information on the methane balance (emission = formation - recovery – oxidation) and thus validate the formation model.
An overview of the results is presented in table 4.

Table 3: Afvalzorg landfills where emissions were measured

Landfill	Period	Surface	Waste	Types of waste
Braambergen	1982 - 2004	30 ha	2.0 Mm^3	Municipal, commercial
Hollandse Brug	1970 - 2000	15 ha	1.3 Mm^3	Municipal, commercial
Nauerna	1985 - 2010	72 ha	4.5 Mm^3	Contaminated soil, commercial, industrial
Zeeasterweg	1980 - 1995	35 ha	1.5 Mm^3	Municipal

Table 4: Emission and production estimates (all values in m^3 $CH_4.h^{-1}$)

Landfill	Period	Method	Emission measured	Production modelled	Extraction measured	Oxidation estimated
Braambergen	Nov.1999	TDL	240	250	110	?
	Dec.1999	TDL			245	
Hollandse Brug	April 1997	TDL	60	100	0	40
Nauerna	April 1997	TDL	310	650	0	340
	April 1998	TDL	155	600	100	345
	Nov. 1999	TDL	600	550	50	?
Zeeasterweg	Dec. 1999	mass balance	35	130	0	95

At the Braambergen landfill a production estimate was calculated for November 1999. A level of 250 m^3 $CH_4.h^{-1}$ was obtained. Measurements using the plume method yielded an emission level of 240 m^3 $CH_4.h^{-1}$, which is almost equal to the total production level. High peaks in methane emission were observed down wind from cells with relatively (1-2 years old) fresh waste. On the day of measurements the gas extraction system recovered 110 m^3 $CH_4.h^{-1}$. So even with oxidation of 0% the sum of emission and extraction exceeds the production estimate. This discrepancy could be caused by some error in the emission measurements being non-representative for mean emissions. But in December 1999, the extraction system was able to recover 245 m^3 $CH_4.h^{-1}$ (using extra wells). Therefore there is no doubt that the estimate of the production level is to low. Although this is the only

landfill with recent deposits of municipal solid waste, a gas production prognosis that proved too low was not anticipated.

At Hollandse Brug a landfill gas production of 100 m^3 CH_4 $.h^{-1}$ was calculated. Measurements indicated a methane emission of 60 m^3 CH_4 $.h^{-1}$. This suggests that oxidation was around 40%.

At Nauerna landfill in 1997 the oxidation of CH_4 in the top layer appeared to be almost 50%. In 1998, with the extraction system recovering about 100 m^3 CH_4 $.h^{-1}$, a significant emission reduction of about 50% down to a level of 155 m^3 CH_4 h^{-1} was observed. The oxidation was almost identical to the 1997 level. The oxidation level is significantly different from the 10 % level used in the IPCC methodology. In November 1999 an emission level was found that seems higher than the production level. An oxidation level can of course not be negative. It should be noted however that the inaccuracy of both the production prognosis and the emission estimate is approximately 20%. In winter oxidation is likely to be zero. Matching levels of production and oxidation can be found within the range of inaccuracy. The small difference in oxidation between 1997 and 1998 and the large difference between 1998 and 1999 seem to indicate that the situation cannot be properly described by means of so few measurements. Also at Nauerna spatial peaks in methane emission could be related to landfill cells containing relatively fresh waste.

The measurements at Braambergen, Hollandse Brug and Nauerna were all carried out with the mobile plume method. The result is always an indication of the situation on a specific day. In general the observations with respect to spatial and temporal variability of the measurements are in accordance with observations from literature as mentioned in chapter 3. In all cases (and especially the experience at Nauerna) has shown that it appeared difficult to correlate such emission data with the formation and extraction. Methane emissions alone result in data that are hard to interpret in terms of formation and oxidation. It would be helpful to be able to measure carbondioxyde carbondioxide simultaneously.

At Zeeasterweg landfill a gas production of 130 m^3 CH_4 $.h^{-1}$ was calculated. Mass balance measurements during six weeks indicated a methane emission of 35 m^3 CH_4 $.h^{-1}$. Apparently more than 60% of the methane was oxidized even though it was winter at the time. The explanation might be that the waste is relatively old and the average height is low, resulting in low fluxes per m². Furthermore the average day temperatures were not too low (between 9 and 12°C) to prevent microbiological activity.

6. CONCLUSIONS AND RECOMMENDATIONS

In general the findings of recent emission measurements in the Netherlands are in accordance with experiences in literature data. Both suggest that in order to obtain an accurate annual emission estimate a measurement method with sufficient spatial and temporal resolution is required.
Both national governments and landfill operators could benefit from such methods. National governments could obtain a more accurate description of the actual national methane emission and thus be able to define a more effective emission reduction policy. Landfill operators could obtain a more accurate estimation of the emission of each landfill and thus be able to make a more effective design of emission reduction measures. The measurement efforts of the landfill operators could contribute to the national estimate.
The mass balance method can provide emission data of methane and carbon dioxide with a high spatial and temporal resolution and is therefore a good candidate. Combining methane and carbon dioxide emission data will also generate data on landfill gas production and methane oxidation. Another good candidate might be a low-cost system, derived from the stationary plume method. Both

methods however are not internationally accepted and need further validation. Mobile plume measurements seem to be a good candidate for validation of emissions. Isotope measurements can be applied to estimate the average oxidation effect at the landfill.
It is recommended that national governments, research institutes and landfill operators with a genuine concern about landfill gas emissions team up to develop, validate and find acceptance for more accurate and affordable measurement methods.

ACKNOWLEDGEMENT

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EMISSION MEASUREMENTS AS A TOOL TO IMPROVE METHANE EMISSION ESTIMATES

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SUMMARY: Application of IPCC-methodology most likely results in an overestimation of methane emissions from Dutch landfills. The Dutch government could benefit from improved methods to quantify the national landfill methane emission. Two strategies are identified:
1) improvement of the present method; 2) estimates based on measurements. Deviation from IPCC-methodology is acceptable only if: a) based on a large number of real-scale observations; b) an accurate impression of the annual average is generated; c) base-line correction is applied. For an accurate annual average the authors recommend 3-4 campaigns of 1-3 weeks per year. Application of sophisticated methods (e.g. FTIR, TDL) will be too expensive to determine a national annual average. Landfill operators could benefit as well and assist the government in obtaining emission data for both strategies. This however requires cheap and robust methods, possibly supervised by the landfill operator. The mass balance and stationary plume methods are proposed for further development to meet these requirements.

1. INTRODUCTION

Current waste policy in the Netherlands aims at both the reduction of amounts of waste to be treated as well as reducing the environmental effects of waste treatment. With respect to methane emissions of landfills, the most important developments are:

- separate collection of paper and organic materials is implemented and effective;
- the capacity of municipal solid waste incineration was doubled in the last decade;
- landfilling of combustible wastes is inhibited by landfill bans and landfill taxes.

As a result the amount of organic waste ultimately landfilled is going to decrease significantly (AOO, 1999). On top of that humidity of the landfill will drop and the degradability of the organic material will change. It is not clear whether existing landfill gas formation models still apply for the newer generation landfills.
The Dutch government under the Kyoto protocol is obliged to assess and report its emissions of greenhouse gases, a.o. methane emissions from landfills. Of special importance are the 1990-emissions, the 2010-emissions and the emission reduction achieved in the period 1990-2010. At the moment reliable forecasts of expected 2010-emissions are of utmost importance for policy definition. The current estimate of 1990-emissions proceeds according to the first-order decay model as described in the internationally accepted IPCC 1996 Revised Guidelines (IPCC, 1996) and its result can be regarded as satisfactory. The effect of a decreased amount of organic materials to landfills is well monitored and its resulting methane emission is expressed in the Dutch emission estimate. The effect of changed conditions on landfill gas formation and methane emissions is not. The use of the

existing methodology in quantifying current and 2010-emissions will most likely result in an overestimation. Simultaneously the emission reduction achieved in the period 1990-2010 will be underestimated.
Another problem of the existing methodology for estimating methane emissions from landfills is that it is not able to monitor some specific measures that are proposed for furthergoing emission reduction. An example of this is the enhanced oxidation in top-layers. This option is widely recognised as a promising method to reduce methane emissions (VROM, 1999). Emission reductions achieved in this way can at present not be monitored, due to lack of a suitable monitoring methodology (either a measurement method or an accepted reduced emission factor) and the results obtained can not be incorporated in the national assessment.
Landfill operators with ISO-14001 certified environmental management systems have taken the obligation to monitor their emissions and also to strive for a continuous improvement of their environmental effects. Information on methane generation is also of importance for design and operation of landfill gas recovery schemes. Less accurate prognoses of landfill gas formation may lead to over- or underdimensioned extraction schemes and utilisation equipment and may render a project economically unfeasible.

2. STRATEGIES TO IMPROVED EMISSION ESTIMATES

The current methodology for estimating Dutch methane emissions is based on the material balance:

emission = formation - recovery - oxidation

Formation is calculated using a first-order decay model as proposed by IPCC (1996), using the default rate-constants for biodegradation, and applying specific input-parameters for carbon-content of the waste and dissimilation. With regards to 1990-formation this methodology can be considered about as accurate as possible with existing knowledge. With regards to formation in waste, landfilled after 1990, due to the reasons described in chapter 1, existing models are not suited to make an accurate estimate of methane formation.
The amount of recovered methane from the landfill is well monitored and accurate compared to the other factors. The uncertainty in the oxidation efficiency of the landfill cover on the contrary is large. At the moment the methodology was made, only little information was available on this topic and still it is difficult to extract a reliable oxidation factor from field data. One key problem is that no large-scale measurement programme has been conducted. Estimated oxidation capacities on a larger number of landfills related e.g. to site specific parameters, or seasonal conditions are not available.
In a study for the Dutch Government (Scharff et al., 2000) two strategies were identified to improve the emission estimates. Performing measurements of emissions at real landfills is an important part of both strategies:

- In the first strategy, measurements are used to improve current methodology based on modelling landfill gas formation and insight in methane oxidation. Improving the existing methodology can imply both improving landfill gas formation models as well as improving oxidation factors.
- The second strategy ultimately aims at an emission assessment based on frequent or continuous measurement of methane emissions at all relevant Dutch landfill sites. In 2010 a limited number of landfills will be responsible for the major part of Dutch methane emissions: over 95 % will be caused by a group of 25 landfills. This implies such an approach to the inventory of Dutch methane emissions from landfills might be feasible, on the condition that an accurate and affordable measurement methodology is available.

It must be stated however that whatever the choice is to obtain future estimates for CH_4 emission levels from landfills, a combination of both model evaluation and measurements will be needed. Updating the methane formation models requires new measurements to see if the models are able to

describe current landfills. If alternatively it would be decided to evaluate the emissions in 2010 using emission measurements at the individual landfills, the new emission data will provide information that can be used to improve the available models. This might lead to a revision of the emission estimates for 1990.
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So any attempts of governments to improve their emission estimate is subject to rather strict preconditions, which imply that every activity and result should be carefully communicated with e.g. international experts, IPCC and UN-FCCC.
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Literature indicates that a.o. landfill gas is emitted with high spatial and temporal variability:

- Emissions per m^2 on single landfills show a variability of three orders of magnitude.
- Emissions from comparable landfills can be different by about an order of magnitude.
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- Static and dynamic closed chambers sampling relatively small parts of landfill surface (e.g. Bogner and Scott, 1995; Maurice and Lagerkvist, 1997; Perrera et al., 1999).
- Mass-balance or micrometeorological methods giving concentration profiles on top of the landfill from which emissions can be obtained from a larger part of the landfill (Oonk and Boom, 2000; Savanne et al., 1997).
- Determination of methane plumes on the landfill or further away from the landfill to obtain emissions from the entire landfill (Czepiel et al., 1996a; Galle et al., 1999; Scharff and Hensen, 1999).
- $\delta^{13}C$ isotope measurements to determine methane oxidation in top covers (Boeckx et al., 1996; Bergamaschi et al.,1998).

For a description of the methods the authors would like to refer to the literature mentioned in this and the previous chapter. Sufficient temporal and spatial resolution in order to deal with the variability as described in chapter 3 can be considered a prerequisite for application of of a measurement method. Table 2 indicates applicability, advantages and disadvantages of the various methods.

When emissions are to be measured using closed chambers, the low spatial and temporal resolution requests a large number of relocations (more than 30 a day) on several days throughout the year. There is no experience how many measurement days are required. This makes this method very labour-intensive and very expensive.
The mass-balance method seems to be better suited to measure emissions from larger surfaces during longer times. Its capability to measure CH_4 and CO_2 gives insight in the primary processes leading to emissions: methane formation and oxidation. For larger sites however, this method might bring about some problems and further developments are required to enable measurements from the whole of a larger landfill site. Drawback of the mass-balance method is that since there is not so much experience with the method, validation might be considered a requirement.

Table 2 - Comparison of measurement techniques

technique	spatial resolution	temporal resolution	com-ponent	costs	expe-rience	other advantages/draw-backs/ limitations
soil core	m^2	hour	CH_4, CO_2	high	few	suited for mechanistic studies of oxidation, possible interference with normal landfilling activities
closed chambers	m^2	hour	CH_4	high	many	many samples required for entire landfill emission, possible inter-ference with landfilling activities
mass balance	few ha	continuous	CH_4, CO_2	mode-rate	few	well-suited for automation
micro-meteorology	few ha	continuous	CH_4, CO_2	mode-rate	few	demonstrated not to be applicable
plume measurement	entire landfill	hour	CH_4	high	some	considered most accurate
isotope measurement	entire landfill	hour	$^{13}CH_4$	very high	some	intended to measure amount of oxidation

Plume measurements with TDL or FTIR technology can be considered the most accurate methods to measure emissions from an entire site. But the drawbacks of this method (complexity, low temporal resolution and high costs of prolonged measurement campaigns) render this method not suitable to give a reliable impression of the annual emission. Since these plume measurements gives an indication of emissions at a single day, its costs will in practice reduce its temporal resolution. But since the method is generally accepted as being accurate it might be the best method for validation of other methods. The last drawback of the plume method may be avoided when a suitable stationary plume method based on gas sampling can be developed.
$^{13}CH_4$ -measurements are widely recognised for their applicability in quantifying the amount of methane oxidised in the top-layer, so this method might be the primary candidate to validate the suitability of other methods to get an impression of methane oxidation.

5. RECENT EMISSION MEASUREMENTS IN THE NETHERLANDS

From 1997 onwards Afvalzorg has carried out methane emission measurements at several landfills (table 3). Apart from quantifying emissions an additional objective was to gain information on the methane balance (emission = formation - recovery – oxidation) and thus validate the formation model. An overview of the results is presented in table 4.

Table 3 - Afvalzorg landfills where emissions were measured

Landfill	Period	Surface	Waste	Types of waste
Braambergen	1982-2004	30 ha	2.0 Mm^3	Municipal, commercial
Hollandse Brug	1970-2000	15 ha	1.3 Mm^3	Municipal, commercial
Nauerna	1985-2010	72 ha	4.5 Mm^3	Contaminated soil, commercial, industrial
Zeeasterweg	1980-1995	35 ha	1.5 Mm^3	Municipal

Table 4 - Emission and production estimates (all values in m^3 $CH_4.h^{-1}$)

Landfill	Period	Method	Emission measured*	Production modeled*	Recovery measured	Oxidation estimated
Braambergen	Nov 1999	TDL	240 ± 40	250 ± 50	110	?
	Nov 2000	box	224 ± 100	2.0 ha	slopes of area B (fig.1)	
	Nov 2000	mass balance	185 ± 9	5.5 ha	80% of area B (fig.1)	
	Nov 2000	TDL	160 ± 16	7.0 ha	area B (fig.1)	
	Nov 2000	TDL	240 ± 24	380 ± 80	245	?
Hollandse Brug	April 1997	TDL	60 ± 25	100 ± 20	0	40
Nauerna	April 1997	TDL	310 ± 60	650 ± 130	0	340
	April 1998	TDL	155 ± 65	600 ± 120	100	345
	Nov 1999	TDL	520 ± 150	550 ± 110	50	?
	April 2000	TDL	270 ± 70	480 ± 100	80	130
Zeeasterweg	Dec 1999	mass balance	35± 5	130 ± 25	0	95

* 95% confidence interval

Several measurements in April point at oxidation levels of 40 to 60%. The measurements at Zeeasterweg landfill seem to indicate that also during the winter period (temperatures fluctuated around 10°C) high oxidation levels are possible. The levels are significantly different from the 10 % level used in the IPCC methodology. In November 1999 an emission level was found that seems similar to the production level even though recovery was effective. An oxidation level can of course not be negative. It should be noted however that the inaccuracy of both the production prognosis and the emission estimate is approximately 20%. In winter oxidation is likely to be very low. Matching levels of production and oxidation can be found within the range of inaccuracy. The difference in oxidation at Nauerna in different periods of the year indicates that the situation cannot be properly described by means of so few measurements.

The TDL measurements enable correlation of spatial peaks in methane emission to landfill cells containing relatively fresh waste. In November 2000 it was possible to assign 60 to 70% of the total landfill emission to area B of Braambergen landfill (figure 1). Area A is capped with a geomembrane. It is justified to assume this area does not contribute to the methane emission. Simultaneously box and mass balance measurements were carried out on the same area. Visual inspection indicated that the slopes of area B emitted most of the methane. The box measurements seem to confirm this. The results of TDL and mass balance methods were similar (see box in table 4). The results of the box measurements (38 locations on 2 ha sampled twice) seem to overestimate the methane emission. Because of the very large 95% confidence interval it can however not be stated that the results are in disagreement with the TDL and mass balance results.

In general the observations with respect to spatial and temporal variability of the measurements are in accordance with observations from literature. In all cases it appeared difficult to correlate such emission data with the formation and extraction. Methane emissions alone result in data that are hard to interpret in terms of formation and oxidation. It would be helpful to be able to measure carbondioxide simultaneously.

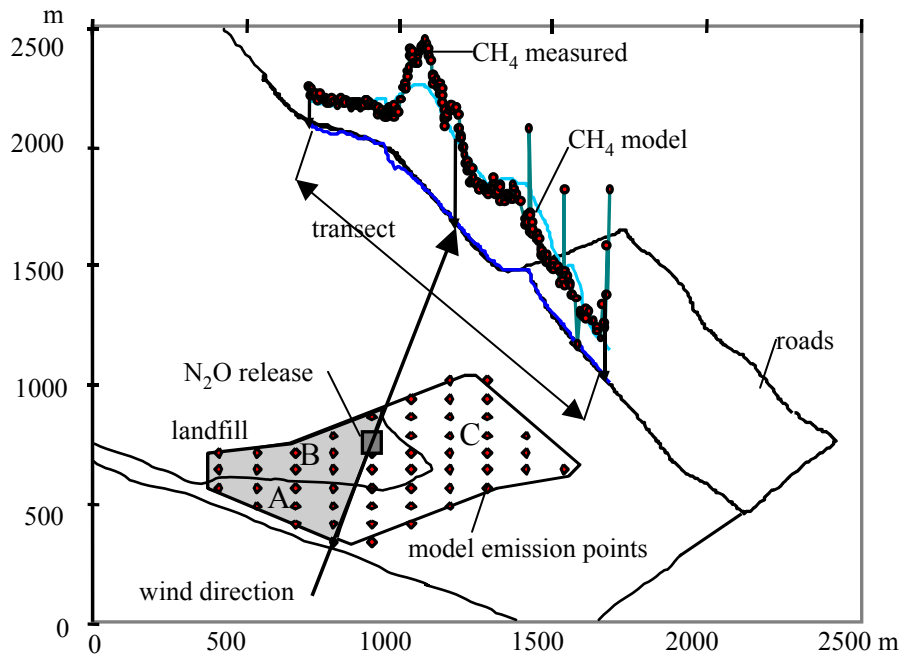

Figure 1. Braambergen landfill TDL emission profile November 2000.

6. IMPROVED MASS BALANCE METHOD

Methane and carbon dioxide emissions can obtained by measuring concentration and wind-velocity profiles, using sampling points in a pole of 10 to 15 meters high. Profiles are interpreted and emissions from the region upstream of the pole are obtained from:

$$J = \frac{\int_{z=0}^{z=l} u_z (c_z - c_l) dz}{x}$$

In which J (in g m^{-2} s^{-1}) is the methane flux through the landfill surface; u_z (in m s^{-1}) is the wind velocity at height z; c_z (in g m^{-3}) is the concentration at height z; c_l (in g m^{-3}) is the background concentration of methane; l (in m) is the length of the pole; x (in m) is the fetch (the upstream length from the pole to the landfill slopes). This method of interpretation is often referred to as the mass-balance method (Fowler and Duyzer, 1989).

In the Netherlands, there is quite some experience with the use of this method for obtaining emissions from landfills (Oonk and Boom, 1995, 2000) and the method has several advantages: it is easily automated and emissions can be obtained for longer periods of time (experience with the method indicated that about three weeks time is required to obtain consistent average emissions) at reasonable expenses; carbon dioxide emissions can be measured as well, though less accurate as the methane emission measurement. There are also some limitations to the measurement method: e.g., the applicability of a 10 m high pole is restricted to situations where the distance to the sides of the landfill is less than 150 m; the result depends on assumptions on distribution of emissions over the surface.

In order to overcome these and other draw-backs, an improved method is developed in this project, using a higher pole (up to 26 meters) for sampling larger areas; putting emphasis on more accurate CO_2-analysis; reduced costs of methane analysis, reduced power consumption and simplified power supply.

7. STATIONARY PLUME METHOD

The stationary plume method uses the same concept as the dynamic plume measurements (Scharff & Hensen, 1999). Downwind of the source a plume of CH_4 is observed. At 4 fixed measurement stations around the landfill, gas-bag sampling systems are deployed. On the landfill a computer unit monitors the meteorological conditions. Using these data the concentrations at the four receptor stations are calculated. Whenever the predicted concentrations surpass a given threshold level, the computer activates the receptor station by phone. At the station a small electronic unit selects a valve and actuates a pump for a 30 minute period. During this time interval air is sampled in the corresponding gas bag. The central computer selects both a background sampling station and a station in the plume. In general about 2 events are sampled each day. After one week the sets with gas bags are exchanged with empty sets. The samples are analysed using GC-FID technique in the lab. All 7 bags from one station are analysed subsequently using an automated system with three analyses per sample. The GC system is calibrated using standards (CH_4 in air) with concentrations of 1,800 and 7,000 ppb. These working standards are calibrated versus NOAA station standards. Therefore comparison with other measurement stations is possible. This is done to enable the evaluation of the background samples in terms of contribution of other sources.

The distance of the receptor points to the landfill and the sampling time at the receptor points was chosen using simple gaussian model simulations. At a distance of about 1-2 km the plume of the landfill is generally well mixed and there is enough time for vertical mixing to obtain useful concentration levels at the 1.5 m sampling height. The optimum for the sampling time will be evaluated in this experiment. When sampling for a long time, for example 3 hours or longer, the actual plume of the landfill might have moved away from the receptor station. Over a short sampling period however the exact position of the sampling station in the plume is much more important. The averaging interval of 30 minutes will result in a plume that is smoothed compared to those obtained with the dynamic plume method but still short enough to have a small standard deviation in the wind direction. Cross contamination of subsequent samples in gas bags was shown to be below 1 % of the concentration difference.

8. CONCLUSIONS AND RECOMMENDATIONS

In general the findings of recent emission measurements in the Netherlands are in accordance with experiences in literature. Both suggest that in order to obtain an accurate annual emission estimate a measurement method with sufficient spatial and temporal resolution is required.
Both national governments and landfill operators could benefit from such methods. National governments could obtain a more accurate estimation of the actual national methane emission and thus be able to define a more effective emission reduction policy. Landfill operators could obtain a more accurate estimation of the emission of each landfill and thus be able to make a more effective design of emission reduction measures. The measurement efforts of the landfill operators could contribute to the national estimate.
The mass balance method can provide emission data of methane and carbon dioxide with a high spatial and temporal resolution and is therefore a good candidate. Combining methane and carbon dioxide emission data will also generate data on landfill gas production and methane oxidation.
Another good candidate might be a low-cost system, derived from the stationary plume method. Both methods have been designed and constructed for field testing to take place in 2001. Parallel to conventional analytical equipment the two methods will be provided with recently developed sensors that could make the equipment a lot cheaper and easier to handle. Both methods however are not internationally accepted and need further validation. Mobile plume measurements seem to be a good

candidate for validation of emissions. Isotope measurements can be applied to estimate the average oxidation effect at the landfill. Verification of the applicability of the methods and validation of their results will be available before the end of this year.
It is recommended that national governments, research institutes and landfill operators with a genuine concern about landfill gas emissions team up to develop, validate and find acceptance for more accurate and affordable measurement methods.

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EMISSION MEASUREMENTS AS A TOOL TO IMPROVE METHANE EMISSION ESTIMATES

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Due to a successful national waste management policy the amount of organic materials processed on Dutch landfills has significantly decreased. At the same time the composition of the waste has changed. Landfills nowadays contain less biodegradable material and less moisture. This most likely reduces the amount of methane that is produced per tonne of organic material. Application of the IPCC-methodology to quantify national methane emissions from landfills probably will overestimate future emissions in the Netherlands. Consequently the emission reduction achieved in the period 1990-2010 as a result of current waste policy could be underestimated. This motivated the research effort to improve methods to quantify methane emission of Dutch landfills through emission measurements, as presented in this paper.

Two strategies were defined to improve national LFG emission estimates: either measure all landfills or measure enough representative landfills and validate the existing models. The Dutch government has chosen for model validation. The objective of an estimate determines the best measurement technique or strategy. National estimates of methane emissions from landfills require other types of measurements than determination of the effect of measures taken at a single landfill site. The objective of this research was to develop and compare measurement methods that would be useful both to landfill operators and the national government, ultimately resulting in methods that can measure emissions of a whole landfill, for longer times at acceptable costs. The methods selected to develop were the mass balance method (MBM) and the stationary plume method (SPM) (Scharff et al., 2001). Due to the small sampling area and the high spatial variability of emissions box-methods are considered less suitable (conclusions workshop on LFG emission measurements, Sardinia 2001). Plume measurements (FTIR, TDL) are considered the most reliable method to obtain whole landfill emission estimates at a specific moment in time, but too expensive for all-year monitoring. So plume measurements were used as reference.

Table 1. Production, extraction and emission in m³/hr

Landfill	Nauerna	Braambergen	Merwedehaven	Wieringermeer
Surface m²	720,000	296,000	350,000	180,000
Waste Mton	7.7	1.7	5.3	1.6
Emission MBM	527 ± 25%	109 ± 25%	386 ± 25%	83 ± 25%
SPM	750 ± 780	440 ± ?	820 ± 700	227 ± 194
1st TDL	1,400 ± 370	540 ± 108	390 ± 100	166 ± 43
2nd TDL	900 ± 150			
3rd TDL	496 ± 222			

* using parameter values obtained from the validation by Oonk et al. (1995).

On all landfills the emission was between 0.5 and 2.5 l/m².h. Plume measurements performed on different days gave different results: so emissions vary significantly from time to time. This confirmed our first preassumption, that more plume measurements are required to obtain a more reliable estimate, which makes the method rather costly.

The method of validation is comparison with other measurement methods and with prognosis of landfill

gas formation. It should be noted that information on amounts of waste, age and especially composition is often not accurately known. In this situation it is not useful to pursue very accurate measurements.

LFG formation models are hardly validated by sufficient field data. For validation whole landfill data should be available from a larger group of landfills. The only validations known to us that meet these requirements are the studies of Oonk et al. (1995) and Huitric et al. (1997) for the Netherlands and California respectively. These formation models are considered quite satisfactory and on average about 20-25% accurate. Both studies used the results of landfill gas recovery projects to obtain validated models. Oonk et al. performed emission measurements in addition for further evaluation of the applicability of the models.

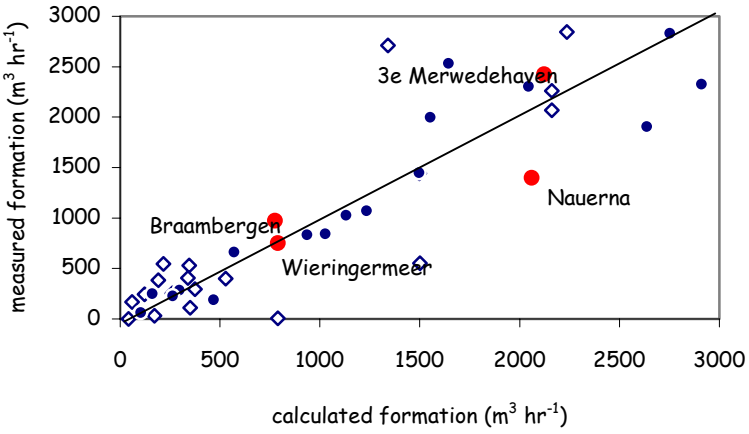

Figure 1. Data of this research in relation to validation of Oonk (1995).

Concluding remarks

The measurements and experience indicate that SPM and TDL measurements give results in the same order of magnitude; however the inaccuracy of both methods is large. MBM results in lower emissions and at the moment we are evaluating whether this is an artifact of this method, e.g. due to the central position of the equipment that never measures a possibly increased emission at the leeward side or due to inaccurate determination of background concentrations. The results of the MBM-measurements however suggest that LFG formation models are not that bad, also for the new situation.

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