METHODS TO ASCERTAIN METHANE EMISSION OF LANDFILLS

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Abstract
This paper discusses the tools available to ascertain methane production and emission on landfills. In general very accurate activity data on waste amounts and waste composition of landfills are not available. Consequently modelling of landfill gas emission is rather inaccurate. The limitations of current methods are illustrated by providing an example of the application of different methods on three landfill sites in the Netherlands. Huge differences in results are observed. As long as we realise the limitations we can use the methods that are currently available. It is possible to establish a trend in the methane emission for a country or a landfill. It can not be considered useful or appropriate to compare countries or landfills or to try to determine the effectiveness of policies or measures between countries or landfills. Whole site methane emission measurement methods are not commercially available and expensive. It can therefore not be recommended to carry out whole site measurements as a standard procedure on every landfill. Further development of measurement techniques may provide more reliable and more accurate methane emission data on landfills.

1. Introduction

A landfill may or may not pose a threat to its immediate surroundings and/or the environment in general. One of the threats is associated with the production, migration and emission of landfill gas (Gregory et al., 1999; Bogner & Spokas, 1995). In specific cases methane in the landfill gas can cause health and safety risks (Christophersen & Kjeldsen, 1999; Jarre et al., 1997). Moreover methane emission from landfills is a major contributor to the greenhouse effect. In order to make decisions about the future destination and use of landfills an assessment of the risks associated with landfill gas production is expedient. This paper discusses methods available to ascertain landfill gas production and emission.

Regulators throughout the world are implementing waste management strategies, policies and regulations aimed at reducing of methane emission from landfills. In order to determine the effectiveness of reduction measures aimed at reducing of methane emission from landfills, quantification of the methane emission either per country or per landfill is essential. Guidance for the estimation of methane emissions has been developed in the framework of various initiatives. In May 2003, the United Nations (UN) adopted the Protocol on Pollutants Release and Transfer Registers (also known as PRTR's- or Kiev-protocol). Amongst others, this protocol requires landfills receiving more than 10 tonnes per day or with a total capacity of 25,000 tonnes to determine their methane emissions individually and make them available to the general public and their national government from 2007 onwards. The Commission of the European Communities (CEC) is currently considering adaptation of the European Pollutants Emission Register (EPER) into the E-PRTR's (CEC, 2004) to comply with the UN PRTR's-protocol. EPER requires that criteria such as accuracy, completeness, comparability, consistency and transparency are met for inclusion of emission data in a publicly accessible database. National governments also report to the United Nations Framework Convention on Climate Change (UN-FCCC) with respect to the Kyoto protocol. For EPER purposes several models to predict methane emissions originating from landfills have been proposed or are recommended by national governments. The most common type of models use single-phase or multi-phase first order kinetics that describe the decay of biodegradable waste and the production of methane.

It is obvious that one method that is suitable for reporting in all cases has advantages and will prevent confusion. The Intergovernmental Panel on Climate Change (IPCC) is currently updating its guidance on methane emission estimates for entire nations (Pipatti, 2005). IPCC have already gone through a process of harmonisation of modelling. In the new guidance only first order degradation models are recommended. IPPC guidance for estimation was never intended to be applied to individual landfills (Pipatti, 2005). A first order degradation model is considered sufficiently accurate for estimation of landfill methane emissions of an entire nation. In every country there are landfills with atypically low and atypically high emissions. When estimating national landfill methane emission these "outliers" will "statistically counterbalance" each other (Oonk, 2005). Consequently, we can not presume that first order degradation models are "sufficiently accurate" for the estimation of individual landfill methane emissions.

2. Landfill gas production and emission

Waste contains a certain amount of degradable organic matter. Inside the landfill microbiological degradation of that organic matter results in the production of landfill gas (LFG). The degradation of organic matter into LFG proceeds in four stages (Veeken et.al., 2000).

1. Hydrolysis. In the first stage complex solid organic material is solubilized by enzymes excreted by hydrolytic micro-organisms.
2. Acidogenesis. In the second stage soluble organic components including the products of hydrolysis are converted into organic acids and alcohols.
3. Acetogenesis. In the third stage the products of acidogenesis are converted into acetic acid, hydrogen and carbon dioxide.
4. Methanogenesis. In the fourth and final stage methane is formed mainly from acetic acid or from hydrogen and carbondioxide. Methane can also be formed directly from products of the acidogenesis such as formic acid and methanol.

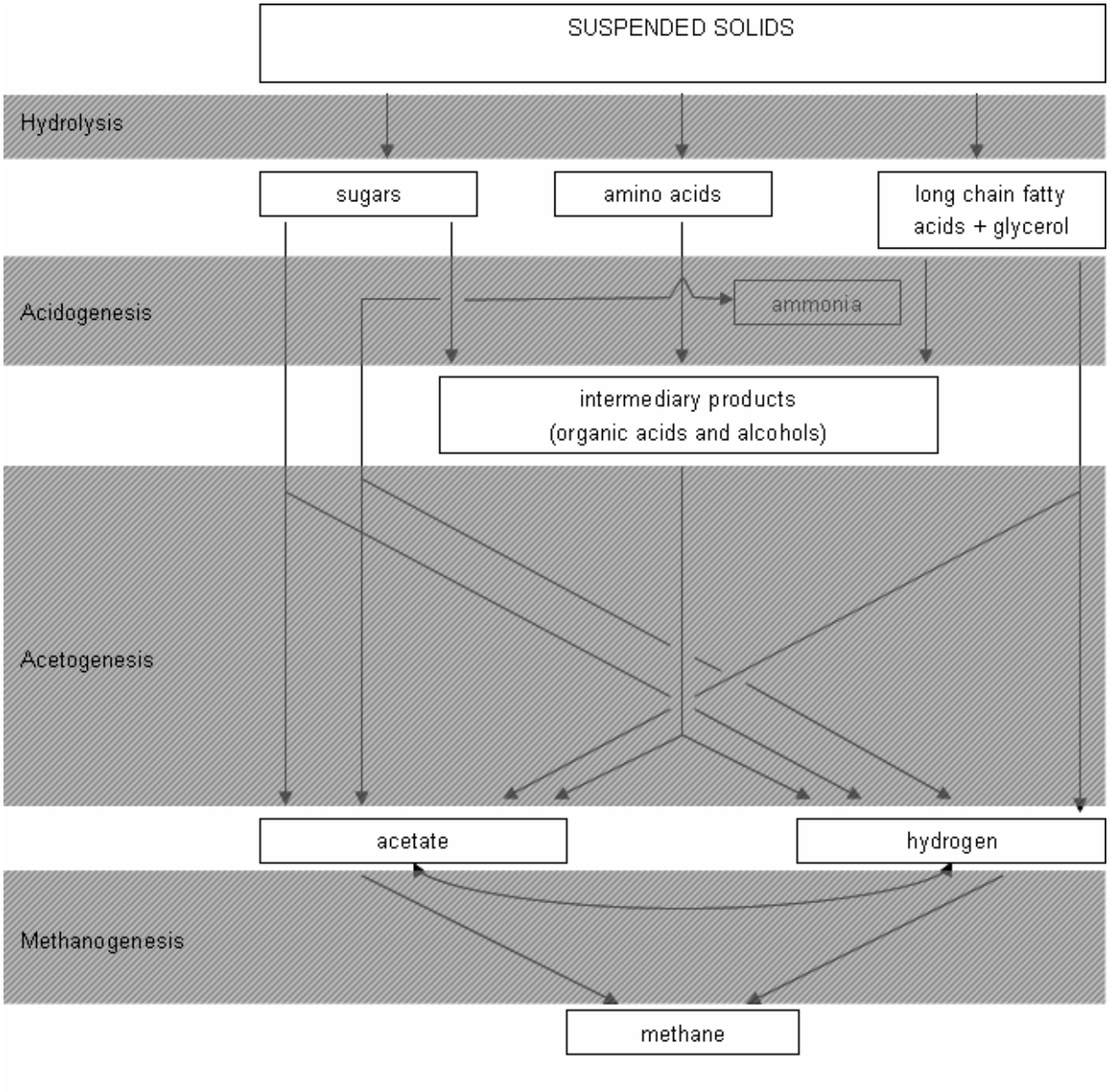

Figure 1. Stages in methane formation.

In reality, biochemical reactions do not proceed in a homogeneous way. Due to local variations in waste composition, moisture content, vicinity of inhibitors and nutrients and temperature, the reaction rates might differ significantly from place to place and all phases might occur simultaneously through the landfill.

Landfill gas predominantly consists of methane and carbon dioxide. From waste that mainly contains carbohydrates the composition of LFG is roughly 50% methane and 50% carbon dioxide. From waste that contains more fat and protein the composition is more likely to be 55% methane and 45% carbon dioxide. The methane concentration can even be higher in very wet landfills due to a higher solubility of carbon dioxide in water. In older landfills the production of landfill gas reduces and atmospheric air can enter the landfill. The landfill gas can contain significant amounts of nitrogen. And with the oxygen from the air methane can be oxidised. This results both in a reduction of the methane percentage and an increase of the carbon dioxide percentage. Therefore the quality of landfill gas in older landfills can vary considerably. Landfill gas can also contain traces of other ((poly) aromatic) hydrocarbons, halogenated hydrocarbons and sulphur compounds.

Different methods of estimation use different parameters. In comparing methods very often conversions have to be made between organic matter and organic carbon and between LFG and methane. For that purpose organic matter can be assumed to be predominantly cellulose. The biodegradation of cellulose can chemically be described by:

$$\underset{(180gOM/mol\,=\,72\ gC/mol)}{C_6H_{12}O_6} \rightarrow \underset{(48g/mol)}{3CH_4} + \underset{(132g/mol)}{3CO_2} \qquad \text{(Eq. 1)}$$

Therefore:

Methane and LFG production per kgOM degraded: $\frac{48}{180 \cdot 714} = 0.373\ m^3CH_4 = 0.75\ m^3LFG$

Methane and LFG production per kgC degraded: $\frac{48}{72 \cdot 714} = 0.933\ m^3CH_4 = 1.87\ m^3LFG$

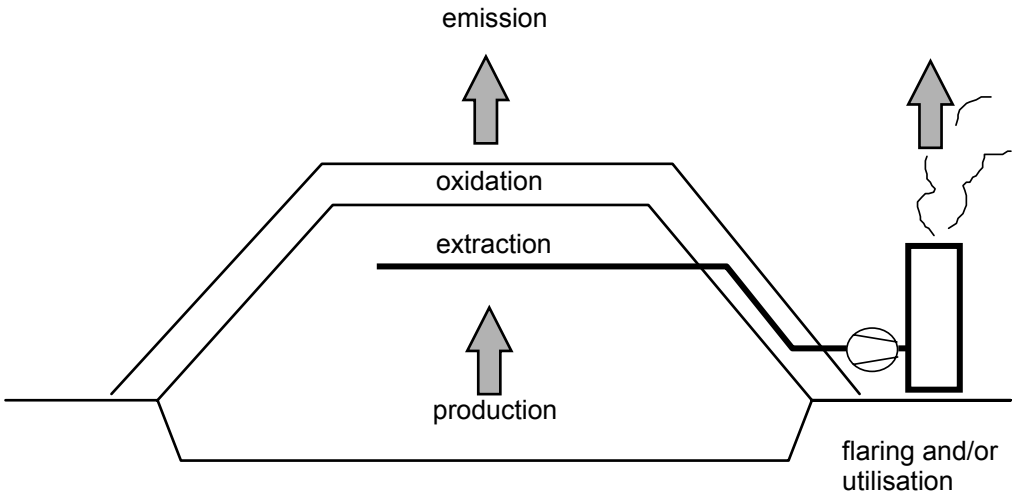

Figure 2. Landfill gas production, extraction, oxidation and emission.

By means of a landfill gas extraction system part or most of the landfill gas that is produced can be extracted (Figure 2). The remainder is potentially available for emission. Methane present in that remainder can be oxidised by methane oxidising bacteria present in a permeable top cover. Methane emission can therefore be calculated by:

$$CH_4\ emission = CH_4\ production\ -\ CH_4\ extraction\ -\ CH_4\ oxidation \qquad \text{(Eq. 2)}$$

It is apparent that the accuracy of the production model is an important factor in this type of approach. The extraction can be measured accurately at low costs. Measurement of oxidation is complicated and

expensive (Scharff et al, 2003). Research indicates a huge variation in oxidation as percentage of LFG production (Scharff et al, 2000). For different types of top covers oxidation rates of 10 to 100 $m^3CH_4.ha^{-1}.h^{-1}$ have been reported. To be on the save side, several regulators have issued guidance that requires no measures or an oxidising biocover when the emission rate is below 10 $m^3CH_4.ha^{-1}.h^{-1}$ (Bour et al., 2005; Etalla, 2003). This can be regarded a rule of thumb applicable to any landfill. Oxidation of methane is usually not known with any degree of certainty and in general an assumed value of 10% of the production minus extraction is used. Emission measurements can be used to establish the appropriateness of this value.

3. Variability of landfill gas emissions

Landfills are not a point source, but a diffuse source of methane. Moreover the emission has a high temporal and spatial variability (Scharff et al., 2000). Therefore it is not easy to measure methane emissions. Emission from landfills can be measured in various ways. Important aspect of the evaluation of techniques is the duration of the measurement. All techniques have typical durations: some give methane emissions as a point in time; others give an impression of emissions at a certain day, where a third category measures emissions during substantially longer times. The variability of the emission level in time provides constraints on the measurement methodology. Important question is: how representative are the emission levels obtained during the campaign for the annual average emission level? Over the last few years quite a number of emission experiments were performed at landfills. These different experiments showed that :

- 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 capacity 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: rainfall and pressure changes.

In the next paragraphs some results obtained from literature will be listed.

3.1 Spatial variation

Several researchers illustrate the high spatial variation in emissions, caused by the high heterogeneity of waste. Several examples: Verschut et al. (1991) measured emissions at three Dutch landfills using dynamic closed chambers and reported spatial variations of more than a factor 1,000. Czepiel et al. (1996a) studied the spatial variation at a landfill in New England and found no correlation between emissions from two points when the distance was larger than 6 meters.

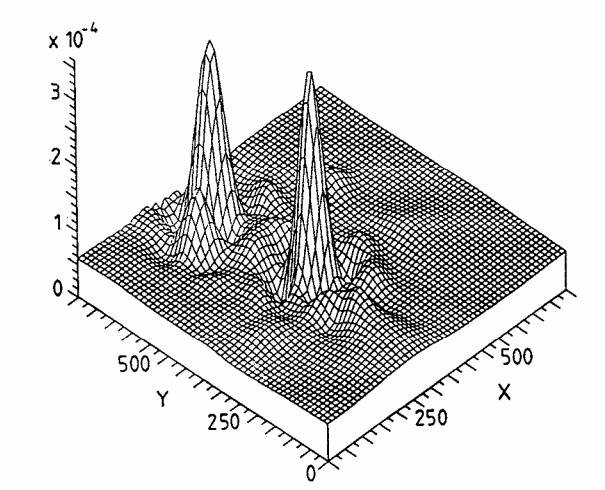

Figure 3. Methane emissions from the Kuchino landfill surface (Nozhevnikova et al., 1993)

3.2 Hourly or daily variations

There are several factors influencing landfill gas emissions on a relatively short time. Climatological phenomena are very important. E.g. (Czepiel et. al 1996b) show a correlation between the changes in atmospheric pressure and the emission of the landfill. Measurements of emissions at a landfill in the US show that a change of 30 mbar over a 5 day period gives a fluctuation in the emission level with a factor 2 (see Figure 4).

Other authors (e.g Verschut et al., 1991) mention pressure-variations to be of importance: increasing atmospheric pressure will lead to a reduced outflux of methane, and methane is accumulated inside the landfill. When the ambient pressure drops a significant increase in the emission can be observed. Variations

in wind can have the same effect, because an increase in wind locally causes a decrease in pressure (Bernoulli-effect). Rainfall also influences landfill gas emissions, since it reduces the permeability of the top-layer.

Emission events can be related with the gas extraction system as well: in the US a 10-fold increase in the emissions of methane into the atmosphere was found at a landfill site where the compressor unit of the extraction system failed for some days (Shorter & McManus, 1997; see Figure 5). When the compressor unit came into operation the old low emission level was reached again after 1-2 weeks. This kind of events can be evaluated when performance of the gas extraction system is well documented. It should be noted however that the gas extraction systems in the Netherlands on average are in operation during more than 90% of the year. Therefore the relative importance of emission events that are related to failure of the extraction system is probably low. When the average emission level at a site is to be evaluated in a measurement campaign however, it is necessary to know that the extraction system worked well (keeping a stationary extraction level) for a period of about 2 weeks before the measurements take place.

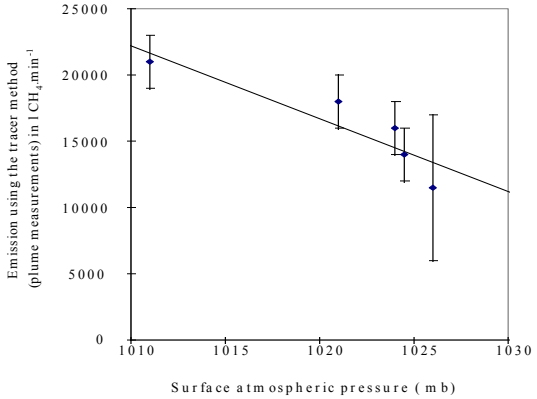

Figure 4. The CH_4 emission at a landfill in the USA, measured on 5 subsequent days. Higher emissions occur with a decrease in the atmospheric pressure. (After Czepiel et. al. 1996b)

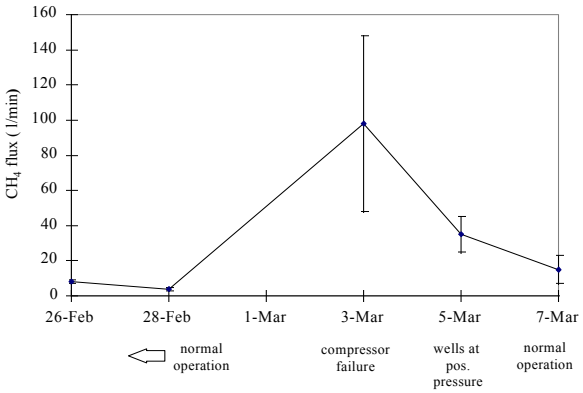

Figure 5. The emission of a landfill before, during and after a technical failure of a compressor unit in the gas extraction system. (Shorter& McManus, 1997)

3.3 Seasonal variation

The conditions (temperatures, humidity) for the methane producing bacteria inside the landfill most likely will not change during the season. So most likely formation will not fluctuate too much over the year. But there are several indications methane oxidation in top-layers is affected, resulting in a seasonal variation in the methane emissions from landfills. This phenomenon is reported in Denmark (Christophersen and Kjeldsen, 1999) , Sweden (Maurice and Lagerkvist, 1997), Belgium (Boekx et al., 1996) and in the US (Czepiel et al., 1996b). Figures 6 and 7 illustrate this seasonal influence.

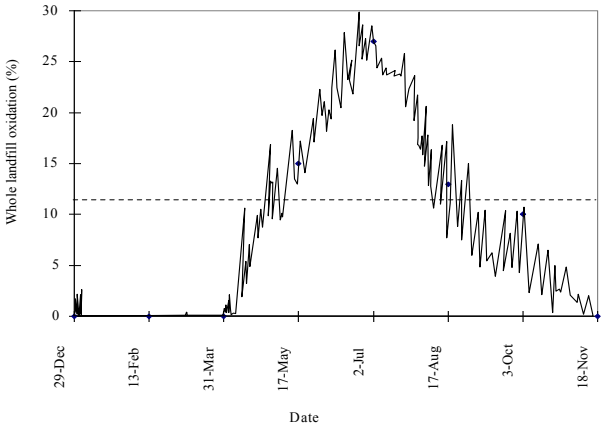

Figure 6. Emission estimated in different months at a landfill in Belgium show a clear seasonal variation caused by a variation in the oxidation in 30 cm top soil layer. (after Boekx et al., 1996.)

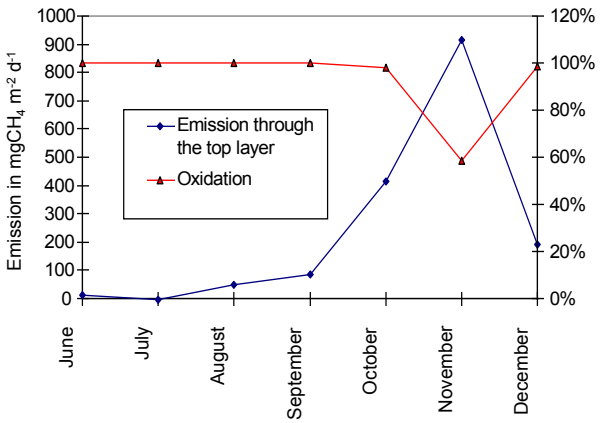

Figure 7. Model calculations of seasonal variation in the oxidation level is estimated based on changes in temperature and soil moisture. (free-style copy after Czepiel et al., 1996b)

The seasonal effect is caused by changes in both ambient temperature and rainfall. Ambient temperature is of great importance: all results show that oxidation efficiency is 0 below about 5°C and very slow

roundabout 10-15°C. Increased oxidation activity is found with increasing temperature and effective oxidation can take place at temperatures in excess of 20°C. Furthermore a clear dependence on the soil moisture level is observed. A maximum in the oxidation is obtained at a moisture level of about 50% of the water holding capacity (Czepiel et.al., 1996b). Increased moisture levels of the soil reduce the fraction of methane that diffuses homogeneously through the top-soil and will lead to an increased emission through cracks in the surface. So not all the bacteria are in contact with methane anymore and methane oxidation will be relatively low. Low moisture levels will decrease the activity of the bacteria as well.

4. Estimation of methane production and emission with models

Most methane production models are based on municipal solid waste (MSW). They are therefore not automatically suitable for situations with reduced amounts of organic waste. In most cases emission models are not validated to a larger number of real landfill data. Only two studies (Oonk et al, 1994; Huitric & Soni, 1997) have derived model parameters, while making a best fit using LFG extraction data and assumptions for extraction efficiency and methane oxidation. Only the model by Oonk et al.(1994) is validated using whole site methane emission measurements (Oonk and Boom, 1995). Thus major uncertainties were introduced.

A problem that emerges immediately when a landfill operator applies one of the models is how to divide the different waste categories registered by the operator over the limited number of categories given in the models. The definitions of waste categories can differ between countries. For instance the category MSW is not used in the Netherlands. The Netherlands make a distinction between household waste and commercial waste. Within the category commercial waste a subcategory exists called “waste from offices, shops and services”. It seems that this category should be included to approximate with MSW in other countries. Residues from sorting plants are also considered to be commercial waste. Afvalzorg registers approximately 50 different waste categories. These categories are mainly based on tax regulations and regulations with respect to hazardous waste. It is not possible to use these categories for modelling methane production and emission other than based on a best guess. It is evident that it is impossible to control the best guess of all the landfill operators in the world. In Europe this problem will not be reduced by the introduction of the European Waste Catalogue (EWC). The EWC has approximately 800 different waste categories and they are not based on carbon content. Even if the carbon content of all these categories was known, it would not be easy to allow for so many entries in a straightforward simple model. In general there is very little information available on carbon content in waste. An illustration of this is that three of the six models taken into consideration in this paper refer to the same research investigating the composition of household and commercial waste carried out in the Netherlands some fifteen years ago (Cornelissen, 1992).

There are many models to quantify LFG production. This paper focuses on those models that have either been validated with whole site emission measurements or are given as guidance by national authorities. In 1994 a study (Oonk et al., 1994) was performed at several landfills in the Netherlands. Both first order single-phase and multi-phase models showed low mean relative errors in contrast to zero order models. Single-phase models consider all organic carbon of the same quality and degrading at the same rate. Multi-phase models distinguish between different types of organic carbon degrading at different rates. The study resulted in the development by the Dutch research institute TNO (The Netherlands Organisation of Applied Scientific Research) of the single-phase first order model used by the Dutch government to calculate and report national methane emissions as if the waste were deposited at one landfill. The Afvalzorg multi-phase model was developed by the Agricultural University of Wageningen based on TNO’s model and the recommendations of the “Adviescentrum Stortgas” (Advice Centre on Landfill Gas) (Scheepers en van Zanten, 1994). LandGEM is recommended by the United States Environmental Protection Agency (US-EPA). The Anglo-Welsh Environment Agency prefers GasSim as the model for individual landfill operators to calculate and report their methane emissions. Recently new models were developed in order to calculate landfill gas emissions in Germany and France. The German EPER model is a zero order model. This means that the potential emission of an amount of waste is considered to occur in the year of disposal. On a landfill with a constant amount of waste of a constant composition this gives a good approximation. But even with a ten year average waste input, a zero order model can only give emission estimates up to nine years after closure. For this reason a zero order model is not suitable to estimate methane emission from former landfills. In the following section the models are described individually.

4.1 First order model (TNO)

The effect of depletion of carbon in the waste through time is accounted for in a first order model (Oonk et.al., 1994). LFG formation from a certain amount of waste is assumed to decay exponentially in time. The first order model can be described mathematically by:

$$\alpha_t = \varsigma 1.87 A C_0 k_1 e^{-k_1 t} \quad \text{(Eq. 3)}$$

Where:

α_t	= landfill gas production at a given time		[m^3LFG.y^{-1}]
ς	= dissimilation factor	0.58	[-]
1.87	= conversion factor		[m^3LFG.kgCdegraded^{-1}]
A	= amount of waste in place		[Mg]
C_o	= amount of organic carbon in waste		[kg C.Mg waste^{-1}]
k_1	= degradation rate constant	0.094	[y^{-1}]
t	= time elapsed since depositing		[y]

The TNO model is derived assuming the amounts of degradable organic carbon in Table 1. For the best results, preferably the same waste composition should be used when using this model to predict LFG production on other sites. It makes the model less dependent on errors in estimates of waste composition.

To calculate methane production, the LFG production obtained with the TNO model is multiplied with the methane concentration of 50% and volumetric mass of 714 $gCH_4.m^{-3}$. To obtain an emission estimate the recovered quantity of methane with the recovery system is subtracted from the calculated production and a standard oxidation factor of 10% is applied according to Eq. (2).

Table 1: Organic carbon content used in the TNO single-phase model.

Waste category	Abbreviation	Organic carbon content [kgC.Mg^{-1}]
Contaminated soil	CS	11
Construction & demolition waste	C&D	11
Shredder waste	SW	130
Street cleansing waste	SCW	90
Sewage sludge & compost	S&C	90
Coarse household waste	cHW	130
Commercial waste	CW	111
Household waste	HW	130

4.2 Multi-phase model (Afvalzorg)

Different types of waste contain different fractions of organic matter that degrade at different rates. The advantage of a multi-phase model is that the typical waste composition can be taken into account. In the Afvalzorg multi-phase model eight waste categories and three fractions are distinguished. For each fraction LFG production is calculated separately. The waste categories and fractions used in the Afvalzorg multi-phase model are described in Table 2. The multi-phase model is a first order model and can be described mathematically by:

$$\alpha_t = \varsigma \sum_{i=1}^{3} c A C_{0,i} k_{1,i} e^{-k_{1,i} t} \quad \text{(Eq. 4)}$$

Where:

α_t	= landfill gas production at a given time	[m^3LFG.y^{-1}]
ς	= dissimilation factor	[-]*
i	= waste fraction with degradation rate $k_{1,i}$	[$kg_i.kg_{waste}^{-1}$]**
c	= conversion factor	[m^3LFG.kgOM$_{degraded}^{-1}$]***
A	= amount of waste in place	[Mg]
C_o	= amount of organic matter in waste	[kgOM.Mg waste^{-1}]
$k_{1,i}$	= degradation rate constant of fraction i	[y^{-1}]*
t	= time elapsed since depositing	[y]

* : ς and k values for rapidly, moderately and slowly degradable waste fractions for Nauerna, Braambergen and Wieringermeer are presented in table 3.

**: Only rapidly, moderately and slowly degradable organic matter has been taken into consideration. The total organic matter content is higher than the sum of these three categories due to the presence of organic matter that is not considered biodegradable under anaerobic conditions. Examples are humic substances, lignin and plastics.

***: Minimum and maximum values of 0,7 and 0,74 m^3LFG.kgOM$_{degraded}^{-1}$ were used.

For some waste categories no organic matter or carbon content data were available. The division in phases was made by a 'best guess' of Afvalzorg. In order to express the uncertainty of this approach a minimum and a maximum amount of organic matter was attributed to each phase of each category. Including the minimum and maximum values for the conversion factor, this results in a 'best guess' of minimum and maximum methane production and consequently minimum and maximum methane emission. To calculate methane emission in Gg per annum the LFG production was multiplied with a methane concentration of 50% and 714 gCH_4. m^{-3}. To obtain an emission estimate the recovered quantity of methane with the recovery system is subtracted from the calculated production and a standard oxidation factor of 10% is applied according to Eq. 2.

4.3 LandGem US EPA

The US EPA model (US-EPA, 2001) is based on the LandGEM model. LandGEM determines the mass of methane generated using the methane generation capacity and the mass of waste deposited. LandGEM can be described mathematically by:

$$Q_{CH4} = \sum_{i=1}^{n} kL_0 M_i (e^{-kt}) \qquad \text{(Eq. 5)}$$

Where:

Q_{CH4}	= methane emission rate		[m^3CH4.y^{-1}]
k	= methane generation constant	(AP42 default = 0.04)	[y^{-1}]
L_0	= methane generation potential	(AP42 default = 100)	[m^3CH4.Mg waste^{-1}]
M_i	= mass of waste in i-th section		[Mg]
t_i	= age of the i-th increment or section		[y^{-1}]

US-EPA protocols (US-EPA, 2004 and 2005) state that the composition of waste used in the model reflects U.S. waste composition of MSW, inert material and other non-hazardous wastes. For a landfill containing non-biodegradable waste (i.e. inert material), such as ash from waste combustion, this portion may be subtracted from the waste acceptance rates. LandGEM recommends subtracting inert materials only when documentation is provided and approved by a regulatory authority. This is not recommended for sites that are typical MSW landfills containing a range of waste that may or may not be degradable. LandGEM provides methane generation constant and potential for both CAA (Clean Air Act) and AP42 (US-EPA, 1998) standards. It is recommended to use AP42 default values for standard landfills (US-EPA, 2004). CAA default values have a high methane generation potential (L0) of 180 m^3CH4.Mg waste^{-1}. After a model run with LandGEM the methane emission can be determined by subtracting the recovered quantity of methane with the extraction system and applying a standard oxidation factor of 10% according to Eq. 2.

4.4 GasSim

The GasSim model (Version 1.00, June 2002) (Gregory et al., 2003) is equipped with two approaches to calculate an estimate of methane emissions (GasSim manual Version 1.00). The first approach uses the GasSim multi-phase equation, which is based upon a multi-phase model described by van Zanten en Scheepers (1994):

$$\alpha_t = \varsigma c \sum_{j=0}^{m} \sum_{i=1}^{n} A_j k_i C_{0,i,j} e^{-k_i (t-j)} \qquad \text{(Eq. 6)}$$

Where:

α_t	= landfill gas production at a given time		[m^3LFG.y^{-1}]
ς	= formation factor		[-]
c	= conversion factor		[m^3LFG.kgOM$_{degraded}^{-1}$]
m	= number of years of landfilling		[-]
j	= year of landfilling amount A_j		[y]
n	= number of fractions i		[-]
i	= waste fraction with degradation rate k_i		[kg_i.kg_{waste}^{-1}]
A_j	= amount of waste in year j		[Mg]
$C_{o,i,j}$	= amount of organic matter in fraction i	landfilled in year j	[kgOM.Mg waste^{-1}]
k_i	= degradation rate constant of fraction i		[y^{-1}]

t = time elapsed since depositing [y]

The multi-phase model requires waste input in Mg and the specific breakdown during the particular year of disposal. Each waste category in GasSim is made up of various fractions. Each fraction is assigned a degradability class and k value. The second approach to estimate methane formation is the LandGEM model, which is similar to the US-EPA model.

4.5 EPER model France

The French EPER model (Budka, 2003) gives two approaches to estimate methane emissions from landfills. The operator can select the most suitable approach.

- Methane emission estimates for landfill cells connected to an LFG extraction system using data of recovered LFG by the landfill operator and the LFG extraction efficiency.
- Methane emissions estimates for landfill cells connected or not connected to an LFG extraction system using a multi-phase model (ADEME version 15/12/2002) and the LFG extraction efficiency.

The methane emission for landfill cells connected to the LFG extraction system can be calculated with the following equations:

$$A = F * H * [CH_4] \quad \text{(Eq. 7)}$$

Where:

A	= recovered amount of methane	[$m^3CH4.y^{-1}$]
F	= extraction rate of LFG	[$m^3LFG.h^{-1}$]
H	= compressor yearly hours in operation	[$h.y^{-1}$]
$[CH_4]$	= methane concentration in LFG	[$m^3CH_4.m^{-3}LFG$]

A is then corrected to standard temperature and pressure ($m^3STP.y^{-1}$) by taking into account the ambient pressure and temperature at the moment of the gas quality sample. The surface area of cells connected to the LFG extraction system and the type of top cover present on that particular cell determine the extraction efficiency. For example a zone in operation which has no top cover and is connected to a LFG extraction system has an LFG collection efficiency of 35%. The remaining 65% of LFG will eventually be emitted to the atmosphere. The production of methane for cells connected to LFG extraction system is calculated by:

$$P = \frac{A}{\eta} \quad \text{(Eq. 8)}$$

Where:

P	= production of methane	[m3CH4.y -1]
η	= extraction efficiency	[%]

Methane emission is then determined similar to Eq. (2).

In the second approach the formation of methane is calculated with a multi-phase equation following the ADEME model:

$$FE_{CH_4} = \sum_x FE_0 * \left(\sum_{1,2,3} A_i * p_i * k_i * e^{-k_i t} \right) \quad \text{(Eq. 9)}$$

Where:

FE_{CH4}	= annual methane production	[$m^3CH4.y^{-1}$]
FE_0	= methane generation potential	[$m^3CH4.Mg\ waste^{-1}$]
p_i	= waste fraction with degradation rate ki	[$kg_i.kgwaste^{-1}$]
k_i	= degradation rate of fraction i	[y^{-1}]
t	= age of waste	[y]
A_i	= normalisation factor	[-]

The model describes three categories of waste and every category has a specific methane generation capacity per Mg of waste. The French model calculates with three fractions and three k values for each waste category. The model calculates an overall k value for each waste category. The distribution of the fractions is the same for Category 1 and 2. This results in the same k value. The k value for Category 3 is

zero. Therefore the French EPER multi-phase model can essentially be considered to be a single-phase model. The French EPER model assumes an oxidation capacity of the top cover of 10%. The total methane emission is calculated by:

$$CH4\ emission = P(1-\eta)\ *0,9 + FECH4\ *\ 0,9 \qquad \text{(Eq. 10)}$$

Where:

η	= extraction efficiency	[-]

4.6 EPER model Germany

The EPER model (Hermann, 2005) used in Germany is a zero order model and can be described mathematically by:

$$Me = M * BDC * BDC_f * F * D * C \qquad \text{(Eq. 11)}$$

Where:

Me	= amount of diffuse methane emission			[Mg $CH_4.y^{-1}$]
M	= annual amount of landfilled waste			[Mg waste.y^{-1}]
BDC	= proportion of biodegradable carbon		0.15	[MgC.Mg $waste^{-1}$]
BDC_f	= proportion of biodegradable C converted		0.5	[-]
F	= calculation factor of carbon converted into CH4		1.33	[Mg CH_4. MgC^{-1}]
D	= collection efficiency:	active degassing	0.4	[-]
		no recovery	0.9	[-]
		active LFG recovery and cover	0.1	[-]
C	= methane concentration		50	[%]

The model only takes “unconditioned residential or similar waste” into account. This category is not registered as such at Afvalzorg landfills. For the purpose of the emission estimate household waste, coarse household waste and commercial waste have been taken into account. The estimate will be considerably lower should an operator decide only to include household waste. The proportion of biodegradable carbon converted (BDC_f) can be compared to the dissimilation factor used in other models. The factor 1.33 for carbon converted to methane (*F*) is the molar weight of methane over the molar weight of carbon. The methane concentration in the landfill gas accounts for the amount of carbon that is converted to carbon dioxide. On all three sites compared in this paper the landfill gas extraction system is managed to maintain a methane concentration of approximately 50%. This value was used instead of the default value of 55%. The default value is proposed when the methane concentration of the landfill gas is unknown.

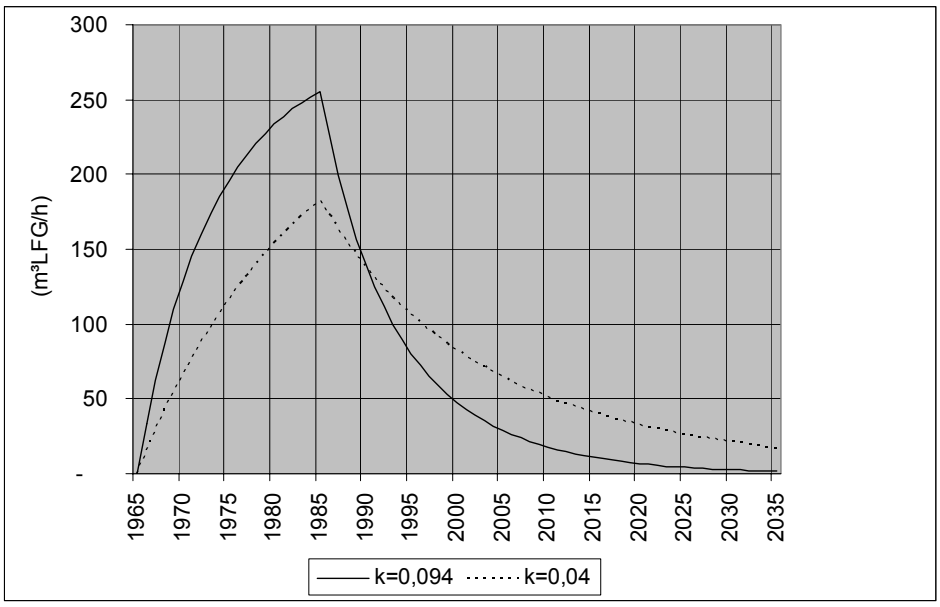

Figure 8. Typical development of landfill gas production in time

4.7 Applying models on former landfills

A typical development of the landfill gas production in time is given in Figure 8. Figure 8 was calculated with the TNO first order model for a hypothetical landfill in operation between 1965 and 1985. Former landfills in most cases are relatively small landfills, that only serviced the immediate surrounding area. The hypothetical landfill was assumed to have received 30.000 Mg of waste per year with an organic carbon content of 105 $kgC.Mg^{-1}$.

In this example twenty years after closure the landfill gas production has dropped significantly. Approximately 95% of the landfill gas formation has already occurred before 2005. Even if a much smaller k-value would be used, the conclusion would be that the remaining production potential is limited. This will be the case for the majority of former landfills. Consequently for many former landfills the environmental risks and impacts associated to methane emission are limited. This indicates that for many former landfill there is no need to have very detailed information on waste amounts and waste composition. Assuming that the hypothetical landfill has an average height (or depth) of 10 m and a surface of 6 ha, the production would be 5 $m^3LFG.ha^{-1}.h^{-1}$ or (at 50% CH_4 in LFG) 2.5 $m^3CH_4.ha^{-1}.h^{-1}$. With a k-value of 0.04 (instead of 0.094) 5.6 $m^3CH_4.ha^{-1}.h^{-1}$ would be produced. Assuming no extraction and 10% oxidation the emission would be far below the threshold level of 10 $m^3CH_4.ha^{-1}.h^{-1}$. In many cases measures other than a biocover are not necessary.

5. Measurement methods

5.1 Soil core measurements

Measurements in the top-layer may give useful mechanistic information about the fundamental steps leading to methane emissions: diffusion and oxidation. Methane and concentration gradients in the soil may give an indication of methane and carbon dioxide diffusion through the layer (Bogner et al., 1995). Landfill soil cores may be collected and transported to the lab for determining bacteriological activity of methanotrophes. The latter is done by exposing the soil sample to a high concentration of CH_4 and measure the decrease of the CH_4 concentration in time, thus giving an indication of the oxidation capacity of the soil. These experiments may be carried out for further study of oxidation process at different temperatures or soil moisture levels etc..

The advantage of soil-core measurements is that it gives insight in the fundamental steps leading to emissions. The method however also has some disadvantages: it is does not take into account emissions caused by convection and its spatial and temporal resolution are low (one gets an impression of emission and oxidation of a very small spot on a single moment). Besides, the methodology is very labour-intensive. Soil cores can also be investigated for porosity in order to supply information for potential migration of methane.

5.2 Gas concentration measurements

Methane concentrations can give an indication of the methane production in a landfill. Methane is oxidised in the upper part of the cover. A high methane production results in high methane concentrations close to the surface (or to the landfill perimeter). When the methane production is low, consequently the methane flux out of the landfill is low. In those situations atmospheric oxygen can more easily enter the cover and oxidise methane. Probes can be inserted and soil pore gas samples extracted and analysed to determine methane concentrations at several depths from the surface or distances from the perimeter (Rettenberger & Mezger, 1992). Probes can either be inserted temporarily or installed permanently. In the latter case they can be used for monitoring lateral migration of methane. The spatial resolution of probes is low. They can only give information about a very limited area around the probe (<1m). Assessment of an entire landfill requires a lot of probes and samples. A more fundamental limitation is that gas concentrations by themselves do not give quantitative data on gas emission. The flux component is missing. Concentration gradients in combination with soil gas pressure and porosity information (Rettenberger & Mezger, 1992; Christophersen & Kjeldsen, 1999) may result in an emission estimate.

5.3 LFG test extractions

Test extractions involve the installation of several (2-4) temporary or permanent gaswells (Rettenberger & Mezger, 1992). The wells are hooked up to a small extraction unit and flare. The cost incurred is substantial. Very often a test extraction is only considered when it can be expected that installing a full-scale extraction is unavoidable. The test extraction serves the purpose of optimising the design of the full-scale extraction system. That is the number of wells per unit of surface, the size of pipes, the condensate removal, the precise capacity of compressors and flares, etcetera. Correct dimensioning is even more important when

utilisation of the gas is considered. Test extractions give information about the potential gas extraction on a limited part of the landfill. A test extraction does not supply information on emission. The gas production is usually estimated by assuming an extraction efficiency based on the nature of the cover.

5.4 Static closed chambers

Static closed chambers are the most simple method to measure fluxes through a surface and are most frequently used in literature. A sampling device consists of a box with a surface area of approximately ½ m^2 in which the increase in concentration of methane in time is measured. Methane fluxes through the surface are directly obtained from the rate in increase of concentrations. Closed chambers are simple, easy to understand and often applied world-wide. Chambers have a small sampling area. They are less appropriate to measure inhomogeneous emissions and landfills prove to be very inhomogeneous. Mosher et al. (1996) conclude that there is no correlation between emissions from two sampling points located further than 6 meters apart. The good correlation they observe, using the box sampling method is much to their surprise. In order to get this good correlation, a large number of measurements are required: more than 20-30 on a typical landfill according to Mosher et al. (1996). So the method requires continuous attention and is therefore very labour intensive. Upon measurement, concentrations in the chamber are being increased. Assuming diffusion to be the prime mechanism of methane transport, this build-up of concentration influences fluxes again, thus limiting the duration of a single measurement. To overcome this problem, Perera et al. (1999) propose a correction method for determining emissions. On surfaces with vegetation, CO_2-emissions can not be measured, since the closed chamber influences assimilation-dissimilation patterns of the vegetation. Although the sample area is small, a number of literature references mention 'good agreement' with other methods, provided that sufficient measurements are conducted on the landfill (Bogner et al., 1995; Mosher et al., 1996; Savanne et al., 1997). Such a good agreement is not obtained by Verschut et al. (1991), using dynamic closed chambers. Since it is often applied, the method is a good candidate for international acceptance. E.g., in the UK plans exist to base the British estimate of methane emissions from landfills on measurements using these closed chambers (Gregory et al., 2003).

5.5 Dynamic closed chambers

Dynamic closed chambers resemble static closed chambers, except for one aspect: in a dynamic chamber a continuous air-flow is maintained through the box, thus avoiding the build-up of concentrations and the influence of fluxes. In dynamic closed chambers, fluxes are obtained from the air-flow through the chamber, the inlet and outlet concentrations. Upon performing closed chamber measurements, maintaining the pressure in the chamber at comparable levels as ambient pressures is of utmost importance (Verschut et al., 1991). In order to have an optimum control of internal pressure, Verschut et al. (1991) therefore used inlet and outlet-fans to maintain the air-flow. Dynamic chambers have in general the same advantages and disadvantages as static closed chambers.

5.6 Mass-balance method

In the mass-balance method, methane and carbon dioxide emissions can be obtained from an interpretation of wind velocity and the methane concentrations at different heights over the landfill surface. At each level the product of concentration and wind velocity provides the horizontal flux and is subsequently related to the landfill area upwind of the sampling point. If the measurements are performed to a sufficient height over the landfill the whole methane plume is sampled and the emission flux is obtained. At varying wind directions, emissions from all part of the landfill are sampled, so this method also provides some spatial information about high and low emission areas. With this method automated, continuous measurement over a longer period is possible.

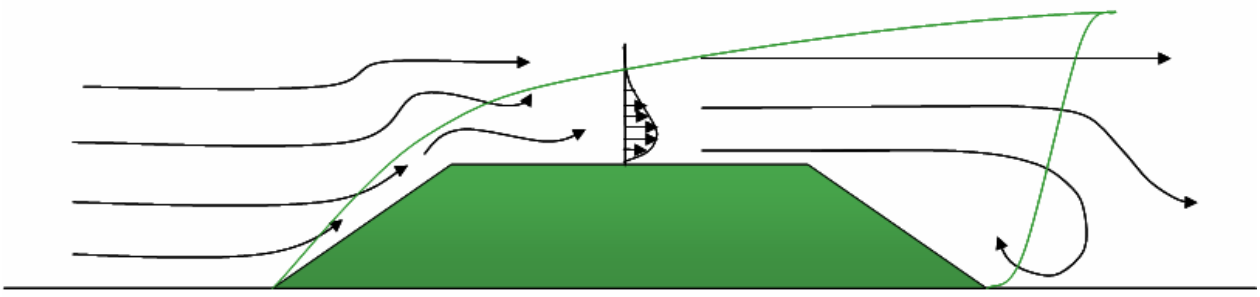

Figure 9. Schematic representation of the mass balance method.

Experience with this method exists in the Netherlands at almost 30 landfills (e.g., Oonk and Boom, 1995) and in France (Savanne 1997). CO_2-emissions can be measured as well on the condition that suitable analysing equipment is present. The combination of methane emission and carbon dioxide emissions give a better indication of landfill gas formation and methane oxidation, since formation can be obtained from the sum of methane and carbon dioxide emissions and methane oxidation from the ratio of methane and carbon dioxide emissions, compared to the ratio in extracted landfill gas. Compared to the box method only one measurement location is needed. The geometry of the landfill can limit applicability of the method. At larger landfills problems might occur, since the upper sample points have to be mounted at impractical heights on a mast. It is estimated that 3 to 4 measurements of 4 to 6 weeks per year would give an accurate estimate of the average annual methane emission. It is estimated that the costs of annual methane emission can be reduced from approximately €30,000 now to €15,000 in the future (Scharff et.al, 2003).

5.7 Micrometeorological methods

Micrometeorological methods are often proposed to measure emissions from larger surfaces (e.g. Fowler and Duyser, 1991). In these measurements concentration gradients are measured and recalculated as vertical fluxes using information about air transport and mixing at the scale of a few m^3 (this explains the name micrometeorology).These techniques however can only be applied on terrain that is rather flat over a large distance (several hundreds of meters), and where emissions occur in a rather homogeneous way. There are almost no landfills that are entirely flat and where the distance to the slope is in excess of 200-300 m. Moreover the slope itself will be a major source of emissions. According to some (Oonk and Boom, 1995, Savanne et al., 1997) this makes emission measurement with these techniques impossible and further discussions about advantages and disadvantages superfluous. The Finnish Meteorological Institute however considers the micrometeorological eddy-covariance method suitable for estimation of landfill gas emission (Ettala et.al., 2003).

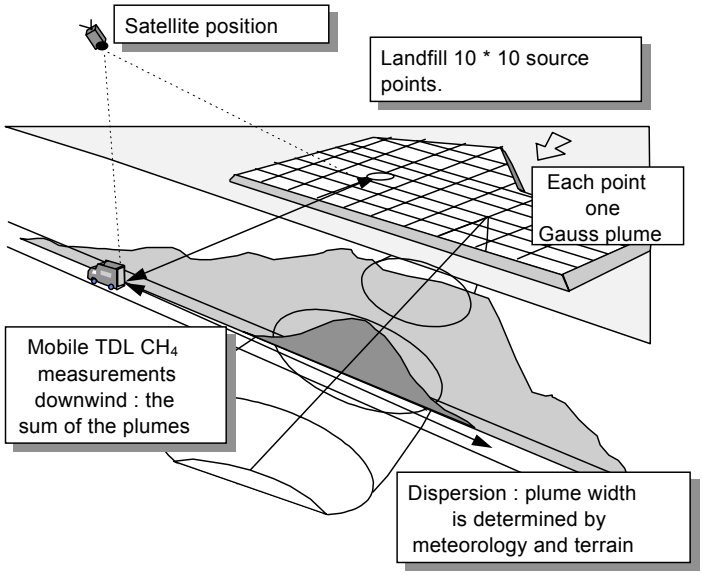

Figure 10: Schematic representation of a mobile TDL tracer measurement

5.8 Tracer measurements

In the tracer measurement method, emissions from the landfill are obtained from the difference in the methane fluxes through a transect screen downwind and upwind from the landfill. Using a fast response methane monitor (e.g. fourier transformed infra red spectroscopy or tunable diode laser) the concentration upwind of the landfill is determined. Downwind of the landfill a methane plume is found that originates from the landfill. In this plume the methane concentrations are higher than the background level. Using a dispersion model the concentration pattern can be used to estimate the emission level of the landfill. Simultaneous release of a tracer (e.g. N_2O or SF_6) at the landfill can be used to either calibrate the dispersion model, or to, directly, calculate the methane emission level by comparison of the plumes obtained from the landfill and the tracer. This type of measurement provides an integrated estimate of the emission of the whole landfill. The spatial differences of the emission level on the landfill automatically taken into account.

The method was used in the USA by Czepiel et al (1996) and by Shorter & McManus (1997). In Europe experiments were performed in France (Savanne,1997), in Sweden (Galle et al., 1999) and in the Netherlands (Hensen, 1997 & 1998 & 2000a & 2000b). With the tracer method an emission estimate for the whole landfill is obtained. Spatial inhomogeneity of the source is accounted for. Especially when using a tracer with a known source strength the interpretation of the data is straightforward. But the method can only be applied when a measurement transect is available downwind of the site. Tracer measurements can not be performed continuously (like for example the mass balance or box measurements). Since mobile measurement continuously needs manpower this type of experiments is relatively expensive. The costs for one measurement day are approximately €6,000. It is estimated that 10 to 20 measurement days are required for an accurate estimate of an average annual emission.

5.9 Stationary plume measurements

In stead of tracer measurements stationary plume measurements can be also be performed. Air samples are collected in canisters downwind of the landfill during a period of time. Experiments in the USA have shown a good agreement between these plume measurements and tracer measurements (Czepiel et al., 1996a) In the Netherlands this method was used to evaluate the emission of NH_3 from manured plots. Here also tracer measurements and stationary plume measurements were performed. A dispersion model is used to obtain the concentration variation versus time at the location of measurement. This modelled time series is averaged over the period in which sampling takes place. Comparison of the modelled and measured concentration levels provide the estimate of the source strength that causes the plume. The method was tested for NH_3 using an artificial source and the emission estimates compared well with the amount of NH_3 released. An emission estimate for the whole landfill is obtained, spatial inhomogeneity of the source is accounted for. The method does not need attendance while sampling so a monitoring system based on this method will be relatively low cost. But stationary plume measurement is relatively new and needs evaluation. Sampling locations outside the landfill are needed. Permission to use these locations is required. It is estimated that 10 to 20 measurements of an entire day would give an accurate estimate of the average annual methane emission. The costs are estimated to be approximately €20,000.

5.10 Isotope measurements

Isotope measurements of CH_4 give insight in the fraction of methane that is oxidized in the top-layer (Bergamaschi et al., 1998). Methane that is formed inside a landfill has a specific isotope contribution ($^{12}C/^{13}C$) which is different from ambient air. When this methane passes the top-layer of the soil part of the methane is oxidized. In this process again the ($^{12}C/^{13}C$) ratio changes. This change is determined by the preference for ^{12}C of the oxidizing bacteria. The emitted CH_4 is “enriched” with ^{13}C. When the latter effect is determined (for example in the lab using cores-samples) the oxidation fraction can be obtained using the isotope composition measurements. The isotope composition measurement of the landfill gas itself is relatively easy to determine. More difficult is to determine the average isotope composition of the gas that is actually emitted into the atmosphere. By filling gas-flasks downwind of the landfill different mixtures of background air and emitted gas are obtained. From these samples the isotope composition of the emitted gas is calculated. Since this method also uses samples downwind of the landfill, the average of the oxidation over the whole site is obtained. The major disadvantage of the method is the cost for the sample preparations. An oxidation level for the whole landfill is obtained, spatial inhomogenity of the source is accounted for.

5.11 Comparison of measurement methods

The applicability, advantages and disadvantages of the various methods to measure methane emissions are summarised in table 2.

Emissions at different spots on a single landfill site may vary over three orders of magnitude. Temporal variation is also of great importance and emissions may range significantly over a range of weeks. On top of that the annual variation in oxidation activity plays a role. When emission measurements are used to get data about annual emissions of a whole landfill site, the surface area sample and the temporal resolution of the selected measurement method are of utmost importance. Taking this in account, the following observations can be made about the various methods. When emissions are to be measured using closed chambers, the low spatial and temporal resolution request 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 masts (16 meters masts are commercially available) and the application of more accurate CO_2-analysers. A disadvantage of the mass-balance method is that since there is not so much experience with the method, validation might be considered a requirement.

Tracer measurements can be considered the most accurate method to measure emissions from an entire site. But the disadvantages 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.

Table 2: Comparison of measurement methods

Method	Spatial resolution	Temporal resolution	Compo-nents	Costs	Expe-rience	Advantages / disadvantages
Soil core	m^2	hour	CH_4, CO_2	high	few	especially suited for mechanistic studies of oxidation, possible interference with normal landfill activities
Gas concentration	m^2	periodic	CH_4, CO_2	moderate	many	gives no direct information on emission
Test extraction	<1 ha	continuous	CH_4, CO_2	high	many	indicated to optimise design of extraction system
Closed chambers	m^2	hour	CH_4	high	many	many samples required to obtain emission from an entire landfill, possible interference with normal landfill 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	doubts about applicability
Plume measurement	entire landfill	hour	CH_4	high	some	considered most accurate, but expensive
Isotope measurement	entire landfill	hour	$^{13}CH_4$	very high	some	intended to measure amount of oxidation, expensive

Since a tracer measurement 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 tracer method may be avoided when a suitable stationary plume method can be developed.

$^{13}CH_4$ -measurements are widely recognised for their applicability in quantifying the amount of methane oxidized in the top-layer, so this method might be the primary candidate to validate the suitability of the mass-balance method to get an impression of methane oxidation.

6. Comparison on three landfill sites and discussion

6.1. Case studies

For EU landfill operators guidance should preferably take into account that EU waste policy is aiming for diversion of organic waste from landfills and consequently landfills that contain small amounts of organic matter. In the last two decades, waste policy in the Netherlands resulted in a reduced amount of landfilled waste and a change in the composition of landfilled waste. In the province of North-Holland household waste has been incinerated since the 1970's. The Nauerna landfill has a total surface of 72 hectares of which 68 hectares is used to dispose of waste. The landfilling of waste started in 1985 and the site is still in operation. From 1985 to until the end of 2004 a total amount of $9.4 \cdot 10^6$ Mg of waste was landfilled at Nauerna. The annual amounts of different types of waste are presented in Figure 11. The abbreviations are explained in Table 1. Waste at the Nauerna landfill is not only characterised by a low content in organic matter. It also contains organic matter that is not readily biodegradable. This makes it a good case study for the future conditions of EU waste policy. Because of the low organic content of the waste, until 1995 it was believed that LFG extraction would not be cost effective. A low LFG production on a large site results in a methane emission that cannot be considered to be negligible. Therefore LFG extraction was started in 1997 and its extent increased in 2000. The low rate of gas production combined with waste with a very low porosity, makes it extremely difficult to reach a satisfactory extraction efficiency.

The Braambergen landfill was operated by a regional authority before it was taken over by Afvalzorg in 1997. LFG extraction was also started in 1997. The surface of the site is 46 hectares, of which 30 hectares are used for waste disposal. From 1982 to until the end of 2004 a total amount of $2.2 \cdot 10^6$ Mg of waste was landfilled at Braambergen (see Figure 12). Landfilling of household waste was stopped in 2000. If the definitions of household waste. coarse household waste and commercial waste are taken as being MSW, then more than half of the waste disposed on this site can be considered MSW. In that respect Braambergen is a more traditional landfill.

The Wieringermeer landfill was also operated by a regional authority and taken over by Afvalzorg in 1997. LFG extraction was started in 1996. The surface of the site is 44 hectares, of which 32 hectares are used for waste disposal. From 1985 to until the end of 2004 a total amount of $2.1 \cdot 10^6$ Mg of waste was landfilled at Wieringermeer (see Figure 13). This waste is mostly commercial waste.

The results of model estimations and the different methane measurements are presented in Figures 14, 15 and 16. The measurement results were obtained from various projects with three techniques: plume measurements with TDL, mass balance measurements and static plume measurements. From 1997, whole site measurements were carried out at the Nauerna landfill (Hensen, 1997, 1998, 2000a). The first whole site measurement at the Braambergen landfill was carried out in 1999 (Hensen, 2000b). Measurements were carried out on all three sites in 2001 as part of a large project funded by the Dutch government (Scharff et al., 2003).

In 2001 within a period of eight weeks three TDL studies were carried out at the Nauerna landfill. Afterwards it became clear that the first TDL campaign was carried out during construction works for which the capping over several hectares had been removed. This result was a lot higher than the others. It was considered an outlier and omitted from the calculation of the average measured emission of the Nauerna landfill for 2001.

The comparison in this paper is based on three landfill sites. It is possible that these landfill sites are not representative. It can certainly not be used to assess or validate any of the models taken into consideration in this paper. On many occasions questions arose with respect to the application of a certain model. After discussions with experts, numerous corrections had to be made. This paper will probably not be the final word about the models applied to the three landfills, as this is a complicated area of study. The corrections did not however change the overall observation that there is a huge difference in methane emission estimates for the six different models applied on the three sites.

6.2 First order model (TNO)

The single-phase first order TNO model is a very straightforward model. It has a limited number of parameters and is therefore easy to use. The results followed a pattern that can be recognised in the other first order models as well. On all three sites the results were approximately in the centre of the range of all estimates. The TNO model estimated methane emission with the same waste categories as used in the Afvalzorg model. The TNO estimates were however higher than the Afvalzorg estimates. This can be explained by higher carbon contents in several types of waste and the assumption of a single carbon phase in the TNO model. All the organic carbon present in the waste is assumed to be potentially converted. Some of the carbon is however not converted because it is not anaerobically degradable or because conditions in the landfill do not allow biodegradation. This is accounted for by a dissimilation factor. This factor describes the percentage of carbon that is actually degraded. In the Afvalzorg multi-phase model the first is accounted for by excluding it from the calculation. The latter is accounted for by a dissimilation factor. The Afvalzorg dissimilation factor is higher than the TNO dissimilation factor. The combination of carbon content and dissimilation factor results in a higher assumption for methane generation potential in the TNO model than in the Afvalzorg model. The difference was most pronounced on the Nauerna landfill with the highest amount of inert waste. It was least pronounced on the Braambergen landfill with the lowest amount of inert waste.

6.3 Multi-phase model (Afvalzorg)

The results followed a pattern that can be recognised in the other first order models as well. The results from the Nauerna and Wieringermeer sites were at the lower end of the range of estimates. On the Braambergen site a different set of parameter values was used to try to compensate for the deviation from the measurement values. The maximum result was therefore more towards the centre of the range of estimates.

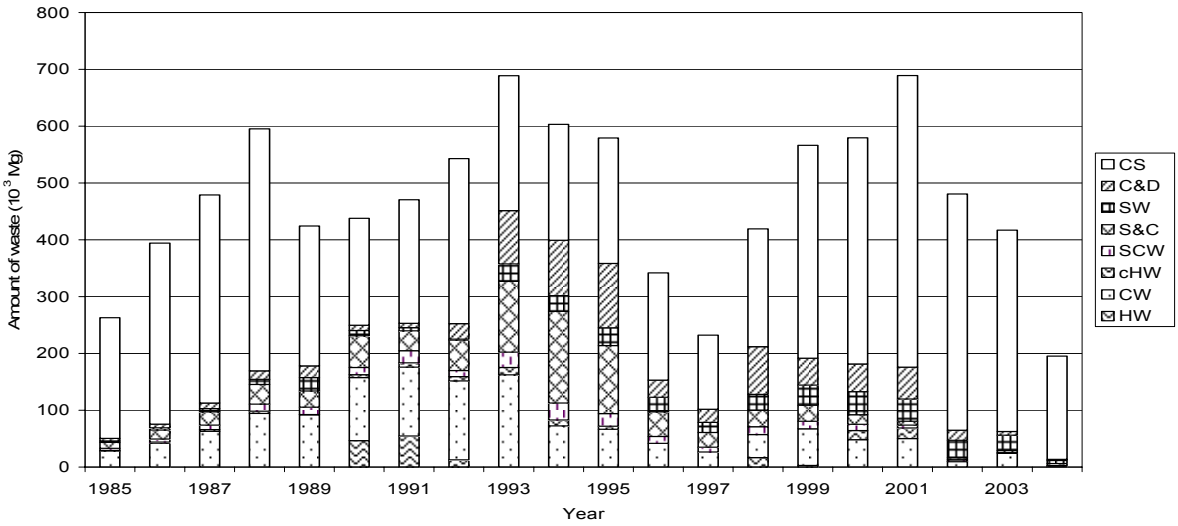

Figure 11: Amounts of waste disposed at Nauerna landfill

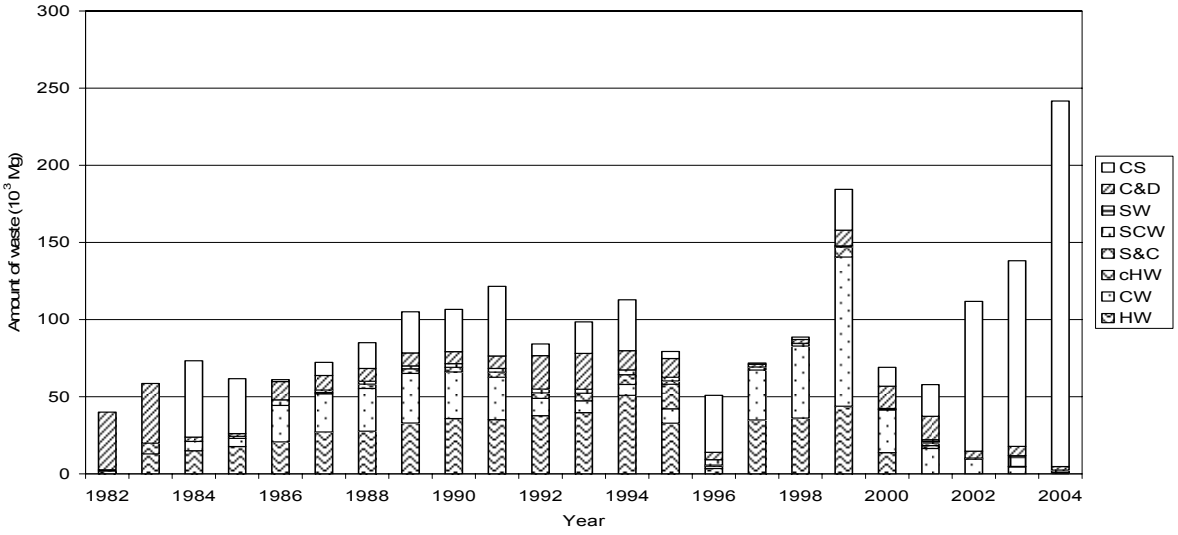

Figure 12: Amounts of waste disposed at Braambergen landfill

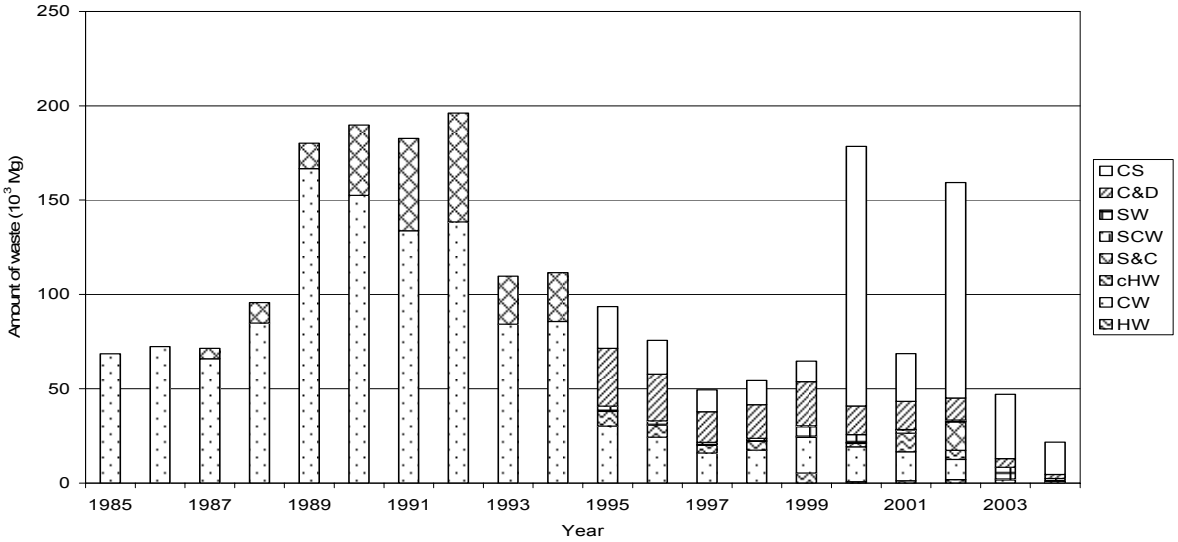

Figure 13: Amounts of waste disposed at Wieringermeer landfill

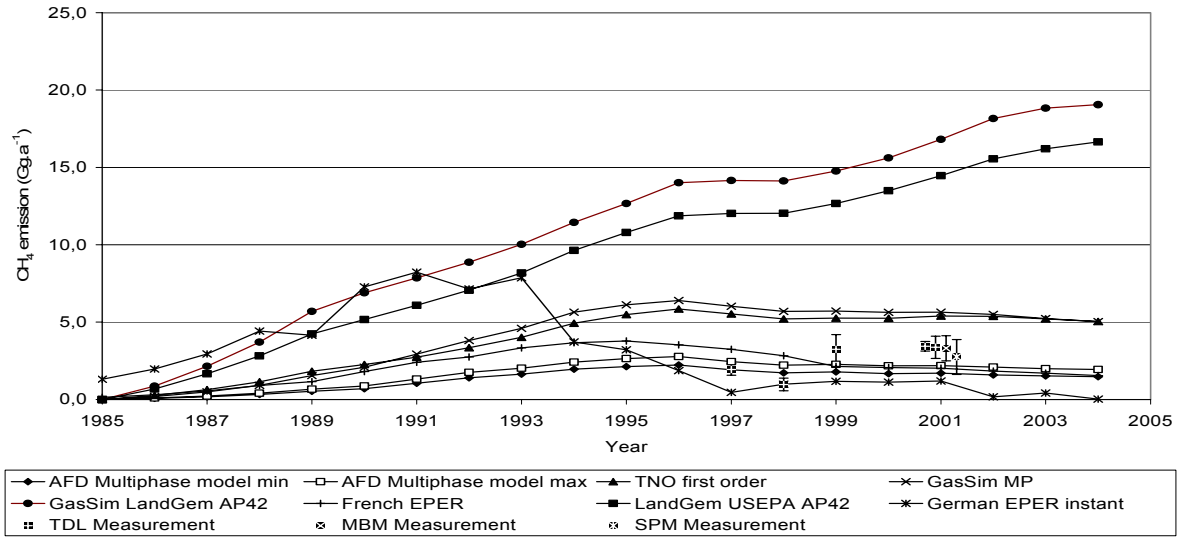

Figure 14. Modelled and measured methane emission at Nauerna landfill

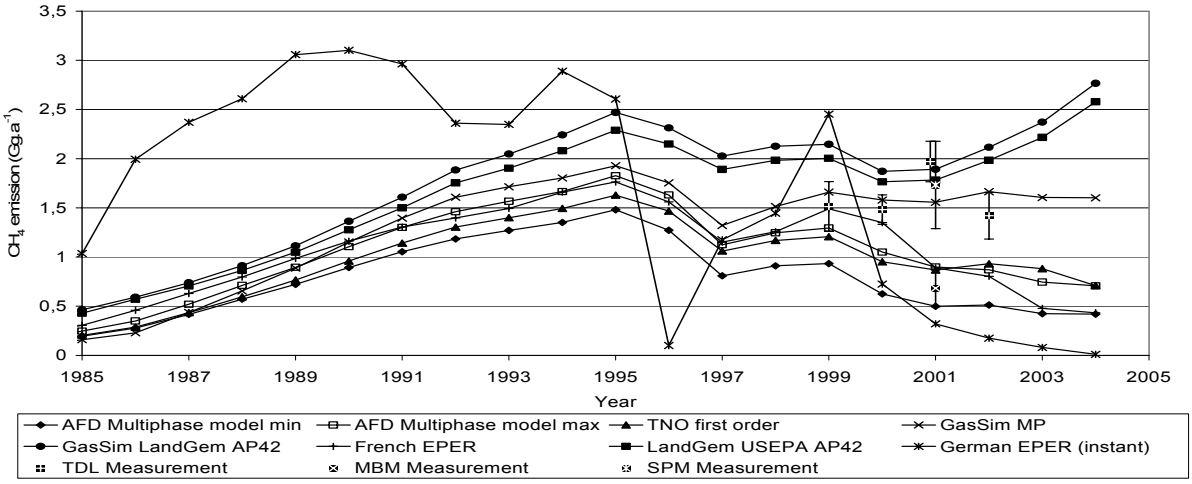

Figure 15. Modelled and measured methane emission at Braambergen landfill

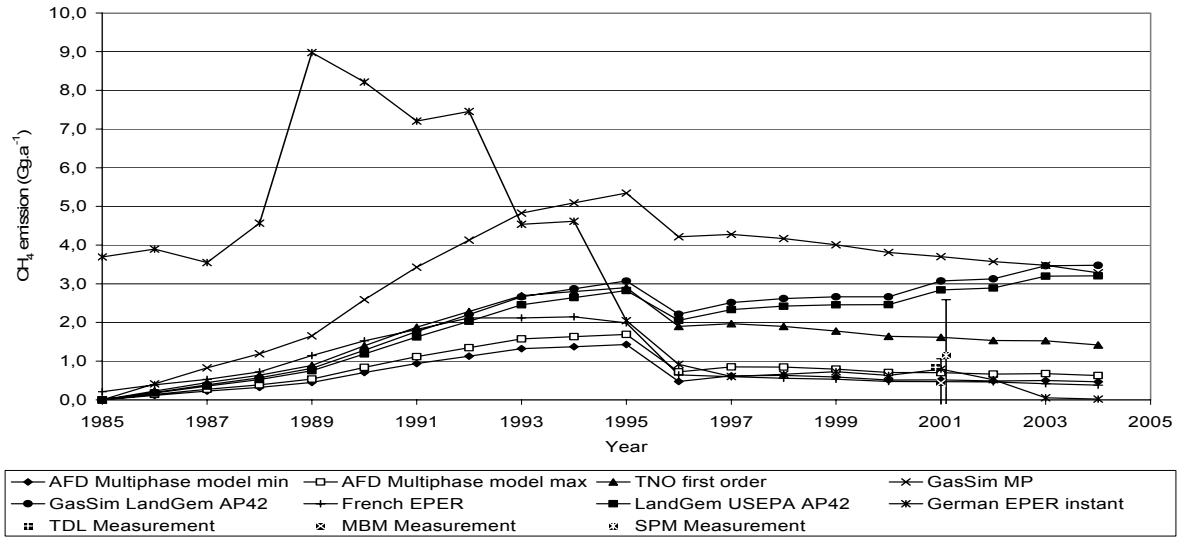

Figure 16. Modelled and measured methane emission at Wieringermeer landfill

6.4 LandGEM

A disadvantage LandGEM is that it can not allow for differences in organic matter content. LandGEM considers all waste to be MSW. It was therefore to be expected that the estimates would be among the highest of all the models. This applies both to LandGEM US-EPA and the GasSim LandGEM. The effect was strongest on the Nauerna landfill. For instance in 2001 the GasSim LandGEM estimate was 14 times higher than the lowest estimate. No explanation could be found as to why GasSim LandGEM gave slightly higher results than LandGEM US-EPA in all three cases.

The amounts of inert waste increased between 2000 and 2004 at the Braambergen landfill. Considering this to be MSW explains why the LandGEM estimates increased in this period, whereas the other estimates do not. LandGEM allows for the exclusion of inert waste. It is recommended that this is only done with permission of the competent authority. This requires expert judgement by both the operator, who should take the initiative, and the competent authority. Many landfill operators will not bother or will not be very discriminating in applying a model.

6.5 GasSim (UK)

The GasSim multi-phase model gave similar results to the TNO single-phase model on two landfill sites. The Nauerna and Braambergen estimates were in the centre of the range of all the estimates. On the Wieringermeer landfill from 1993 onwards the GasSim multi-phase model gave the highest estimates of all models. This landfill was dominated by commercial waste especially in the first period of operation. The high GasSim multi-phase estimate on the Wieringermeer landfill can be explained by the organic carbon content in commercial waste. GasSim multi-phase assumes 180 kg of organic carbon in commercial waste. If this organic carbon is assumed to be cellulose, it could potentially yield 168 m^3CH_4.Mg $waste^{-1}$. That is considerably higher than the assumption for methane generation potential in other models.

6.6 EPER model France

In the French EPER model the landfill operator has the choice to select the approach. The first approach is based on LFG extraction data and fixed LFG extraction efficiencies for different types of cover. This approach is completely different from the other models. It might have led to incomparable results. It was decided to apply the second approach for this paper. The second approach resulted in estimates that follow a similar pattern as other first order models. The French EPER estimates were at the lower end of the range of results. They were comparable to the Afvalzorg multi-phase estimates. This is mainly caused by the fact that a major part of the waste was assigned to Category 3 producing no methane. The French model mentions three fractions and three k values for each waste category. The model calculates an overall k value for each waste category (Table 7). The distribution of the fractions is the same for Category 1 and 2. This results in the same k value. The k value for Category 3 is zero. Therefore the French EPER multi-phase model can essentially be considered to be a single-phase model.

6.7 EPER model Germany

Large fluctuations in methane emissions were estimated with the German EPER model. In this paper the zero order model was used as it was intended. That is the methane production of an amount of waste landfilled in a certain year is instantaneous or in other words released in that same year. The fluctuations represent the fluctuations in household waste, coarse household waste and commercial waste disposed at the three landfills. If a constant amount of waste of constant composition is disposed in a landfill, the result is the same as for a first order degradation model. If there is variation over the years, the German EPER model provides the option to estimate methane emissions with a five or ten year average waste amount (Hermann, 2005). At all three landfills the German EPER model overestimates the methane emission in the first ten years of operation and underestimates the methane emission in the last five years of operation. This effect would have been reduced, but not completely eliminated, if the ten year average option had been used.

Annex 1 no. 5 (d) of the proposal for an E-PRTR (CEC, 2004) states that "Landfills (excluding landfills of inert waste and landfills, which have been definitely closed before the 16 July 2001 or for which the after-care phase required by the competent authorities according to Article 13 of Council Directive 1999/31/EC has expired)" should report their methane emission. This means that the competent authority should determine that at a certain moment aftercare can be stopped. Only then the reporting obligation ends. A system for determining/deciding on landfill completion has not been established anywhere in Europe. Most EU Member States are considering applying 30 to 60 years aftercare periods. Guidance for determining methane emissions should therefore be able to estimate emissions up to at least 30 years after disposing of the last Mg of waste. Even with a ten year average waste input, the German EPER model can only give emission estimates up to nine years after closure.

6.8 Models versus measurements

The deviation between the single data obtained during the measurement campaigns was used to calculate a 95% confidence interval for the resulting methane emission estimates. For the emission estimates obtained with models it was not possible to determine or to estimate any confidence interval. Even disregarding an uncertainty in the result of each individual model, the results of the different measurement methods show a much smaller variation than the modelling estimates. Although there is only a limited amount of measurement data available, this could be considered an indication that further development and wider application of measurement methods would be useful for the estimation of methane emissions.

In comparison to the average measured methane emission of the three landfills the TNO first order, GasSim multi-phase, the GasSim LandGEM and LandGEM US EPA model seem to overestimate the methane emission in the period 1999-2002. The Afvalzorg multi-phase, German EPER and French EPER models seem to underestimate methane emissions in the same period. It is not a surprise that the results of the Afvalzorg multi-phase model for the Nauerna and Wieringermeer sites are very close to the measurement results. Afvalzorg has used the measurement results to try to "tune" its model. This was more difficult for the Braambergen landfill.

The largest set of measurement data is available for the Nauerna landfill. The results for 1997 to 2001 indicate that the emission in those years is constant. Five of the models also estimated a rather constant emission in that period. On top of that the different measurement methods gave similar results. Based on measurements we can therefore be confident about an approximate emission of 3 Gg $CH_4.a^{-1}$ in 2001. The model estimates range from 1.2 to 17 Gg $CH_4.a^{-1}$ for 2001. This is between 40% to 570% of the measurement result. Should we disregard the LandGem AP42 results, as the model estimates range from 1.2 to 6.2 Gg $CH_4.a^{-1}$, this is between 40% to 205% of the measurement result.

The second largest set of measurement data is available for the Braambergen landfill. This landfill is surrounded by trees. It might therefore be expected that measurement methods relying strongly on straightforward flow of atmospheric air over the surface of the landfill are impaired by the local conditions. This is the case for MBM measurements. Both TDL and SPM measure at a distance from the landfill where mixing may be expected to have resulted in an evenly distributed methane concentration. These methods can therefore be expected to suffer less from the local conditions at the Braambergen landfill. The MBM result is considerably lower than the other results. Based on measurements, the emission from the Braambergen landfill was approximately 1.5 Gg $CH_4.a^{-1}$ in 2001. In the same year the model estimates for the Braambergen landfill range from 0.3 to 1.9 Gg $CH_4.a^{-1}$. This is between 20% to 125% of the measurement result. In this case the LandGem AP42 results are closer to the measurement results than results with other models. Being a landfill dominated by household waste and commercial waste it would not be logical to disregard the LandGEM AP 42 results.

Measurement data are only available for the year 2001 for the Wieringermeer landfill. The results of the different methods indicate that the emission in 2001 was approximately 0.7 Gg $CH_4.a^{-1}$. In 2001 the model estimates for the Wieringermeer landfill range from 0.5 to 3.7 Gg $CH_4.a^{-1}$. This is between 70% to 520% of the measurement result. Should we disregard the GasSim multi-phase and LandGem AP42 results, as the model estimates range from 0.5 to 1.7 Gg $CH_4.a^{-1}$, this is between 70% to 240% of the measurement result.

7. Conclusions and recommendations

There are huge differences in methane emission estimates of different models. The highest estimates obtained with the models can be five to seven times higher than the lowest estimates. This huge variation in results cannot be considered acceptable for reporting purposes. Current models are highly uncertain, mutually incomparable and inconsistent. The minimum accuracy that might be considered necessary for comparison between different models and countries is not met. Harmonisation of models may not necessarily solve uncertainty. However, it may at least result in comparable, consistent and transparent data.

With respect to landfills modelling is hampered because of a lack of data on waste amounts and composition. The introduction of the EWC and more stringent waste reporting obligations is not going to solve this problem.

Due to the development of the landfill gas production in time in many cases the methane emission on older landfills has reduced considerably. In these cases modelling with worst case assumptions can show that the methane emission is very low. Consequently the associated risks will also be very small. Only in those cases where this conclusion can not be drawn a further assessment of the methane emission would be recommended.

Methane emission measurements are not commercially available and very expensive. Tracer measurements are considered to be accurate for the short time span of measurement. A single tracer measurement does however not provide a reliable average annual emission. It would therefore at the moment not be advisable or feasible to carry out such measurements as a standard procedure on landfills. Further development of measurement techniques especially focusing on cost reduction is required. The current lack of such a method should however not induce a decision to do nothing. Already with the current methods and limited use of them it is possible to "tune" models. This may provide a more reliable tool to estimate methane emissions in the near future than current models.

In case measurements are considered to be applied on the short term, the best option to obtain a proper estimate for an annual emission level at a certain location will be a method with a good resolution in time, in combination with spatial integrative techniques. 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. 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. Tracer measurements seem to be a good candidate for validation of emission measurement methods. Isotope measurements can be applied to estimate the average oxidation effect at the landfill.

In the mean time the reporting obligations are a fact. We will have to apply the methods and use the activity data that are currently available. That should not be a problem as long as we realise the limitations. For instance in continuing to use one particular model it is possible to establish a trend in the methane emission for a country or a landfill. When the same model is used for both individual landfills and national totals it is also possible to compare a national estimate with the sum of all individual landfill estimates. This enables us to identify and subsequently reduce the error margins. Emission estimates will continue to be inaccurate. But when the same model is used, at least they are more comparable, more consistent and more transparent. If it is unclear which methods are used, it can not be considered appropriate to compare landfills. Even if the method is known, comparing different landfills or different countries may not reveal useful information. In these cases it is therefore not easy and straightforward to determine the effectiveness of policies or measures between countries or landfills.

Therefore we recommend harmonisation. Individual landfill estimates within a region have to be made with the same model and preferably based on the one that the national government uses for reporting to UN-FCCC. This allows comparison between landfills and the national total with the sum of the individual estimates. IPCC has already gone through a process of harmonisation. We recommend that a similar process of harmonisation is also carried out with respect to guidance for landfill operators. For further improvement of models we recommend to initiate an integrated European research programme. An important aspect is to validate the models with sufficient whole site methane emission measurements. Such an improved model has to allow for differences in landfill gas formation due to climatic conditions within Europe (eg. Nordic and Alpine, North Sea Region, Central Europe and Mediterranean regions). This has to be taken into consideration when defining such a research programme.

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