LITERATURE REVIEW: METHANE FROM LANDFILLS

METHODS TO QUANTIFY GENERATION, OXIDATION AND EMISSION

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CONTENT

SUMMARY AND CONCLUSIONS

Methane from landfills is considered a major source of greenhouse gases, both in EU and worldwide. Emission reduction from landfills is amongst the most feasible and cost-effective measures to reduce greenhouse gas emissions. The European Landfill Directive obliges European landfill operators to reduce methane emissions. The E-PRTR regulation requires landfill operators to report methane emissions annually to the competent authorities.

Landfill operators can quantify methane emissions using an emission model or an appropriate measurement method. Different emission models give very different results, even when the same data are entered. Emissions measurement methods are generally considered insufficiently accurate. This is no desirable situation, since it is hard to assess both the impact of measures taken by landfill operators and policies developed by regulators.

The Sustainable Landfill Foundation (SLF) is committed to minimization of landfill methane emissions. Since at the moment both model-approaches and direct measurement of emissions are not yet considered accurate enough, SLF commissioned OonKAY! to perform a comprehensive and critical review was performed of both available models and measurement methods with the following objectives:

- A literature review on methods for quantification of annual average methane emissions from an individual landfill.
- Evaluation of the methods, a.o. whether the methods meet the minimum standards as described in IPCC or E-PRTR guidance documents.
- Discussion of options for improvement and potential directions for harmonization.

CONCLUSIONS ON MODELLING

Methane emissions can be calculated from methane generation, methane recovery and oxidation.

There are several models available that describe **generation**, such as the IPCC-model, the TNO-model and GasSim. The French E-PRTR-model is much simpler and might be just as effective. These models will produce reasonable results for MSW dominated by household waste, landfilled in Western Europe. The accuracy of these models for other types of waste or in different regions in Europe is limited.

Oxidation is more difficult to describe, than methane generation, due to the scarcity of available information on actual oxidation under field conditions. The IPCC-default value of 10% seems a low-guess, leaving room for improvement. Modeled approaches to estimate methane oxidation, based on a.o. top-layer design and climate conditions are in development. However these approaches still lack full-scale validation.

In the end, modeled methane emissions are highly **uncertain**, due to a propagation of errors, which is highly unfavorable. Errors of 25-35 in methane generation result in errors of 65-85% in methane emissions.

An ideal methane formation or emission model doesn't exist and each of the models has their pro's and cons. Combining strengths of different models could be one way for harmonization (see below).

Conclusions on measurements

The main difficulty in measuring methane emissions from landfills is the spatial and temporal variability of emissions, in combination with the size of a modern landfill. Several methods are developed and tested to measure methane emissions from landfills. However at the moment there is no single method, that is widely recognized as the preferred method to measure annual average methane emissions.

Closed chamber methods are the most frequently applied measurement method. However, there is a growing agreement that they tend to underestimate emissions, even when prescribed procedures are followed for grid-wise measurements and application of geo-statistical methods for interpolation.

The 1D-mass balance method and both the mobile and static plume tracer measurements are methods that promise **acceptable accuracy at relative low-cost**.

Claimed accuracy of methods is in the order of magnitude of 25%, on the conditions that the measurement stays within the predefined constraints. However this claim is questionable. It requires more measurement intercomparisons and measurements in situations with controlled methane release, to confirm that this accuracy can be claimed with confidence.

For measuring **annual average emissions**, day-to-day and seasonal variations have to be dealt with and 4 to 6 one-day measurements will be required.

The most accurate method to quantify **methane oxidation** is measurement and interpretation of ^{13}C in the plume. Also this method is at discussion and most recent insights indicate that it might underestimate methane oxidation.

1D-mass balance measurements might be an alternative. However both the measurement of CH_4 and CO_2-emissions using this technique and the estimation of methane oxidation from a shift in CH_4/CO_2-ratio is not widely acknowledged as a reliable method.

Improving methods

In general, the quality of an emission inventory depends on the perspective for which the emission inventory is used. Quality criteria for national inventories of greenhouse gases to UN-FCCC differ from quality criteria for data on individual companies in the framework of E-PRTR. For data used in a legal context (e.g. to verify whether a company complies to its emission limits), again different quality criteria exist: in the end they have to be convincing in court.

Methods to quantify methane emissions might be improved by

- Model harmonization and validation. It is very well possible to take the strengths of every model and combine them into a harmonized version. The degree of complexity of such a model should be in balance with its expected accuracy. Harmonization doesn't necessarily imply more accurate models. For more accuracy, field validation is required;
- Improvement and validation of measurement methods e.g. by testing methods in situations with a controlled release of a known amount of methane. Improvement should also imply cost-reduction and proliferation, a.o. by taking methods away from the research phase and hand knowledge over to specialized companies.
- Definition of tiered approaches for quantification of emissions, allowing emission measurements, rather than modeling emissions. Landfill owners should be allowed to apply higher tiered methods to quantify emissions;
- Transfer of knowledge of both modeling and measuring methane emissions to landfill owners, national governments and local legislative authorities.

A harmonized model or a tiered approach will be acceptable for E-PRTR. For application in making national estimates and reporting them to UN-FCCC, such a model should be thoroughly validated. When methane emission limit values have to be enforced, models and their inaccuracies should be thoroughly validated. When emission limit values are to be enforced by measurement, methods should be accepted between peers and the accuracy should be well assessed. Testing methods in controlled release tests under varying conditions seems to be a strong tool in this.

CHAPTER 1: INTRODUCTION

When waste is landfilled, the organic fraction in the waste (all materials from a biogenic source, such as food and garden residues, textiles, paper) slowly decomposes. In this process, landfill gas is formed, a mixture of methane and carbon dioxide[1]. The emission of methane contributes significantly to greenhouse gas emissions. Total European emissions are estimated to be about 2% of total greenhouse gas of 5000 Mton per year (EEA, 2009). Landfill methane emissions are considered one of the main draw-backs of landfilling of solid waste and abatement of methane emissions from landfills is an important driver for current EU-Waste policy.

In the last decade, attention to methane emissions from landfills has grown significantly. Efforts of both nations and individual landfills are closely monitored. National authorities have the obligation to quantify landfill methane emissions and subsequently report emissions to UN-FCCC. Individual landfills have to report emissions within the framework of E-PRTR.

PROBLEM

To fulfill reporting obligations as described above, several methods are developed to quantify annual average methane emissions from landfills. Several models are developed in the framework of the reporting obligations to UN-FCCC and E-PRTR. However for an individual landfill different models result in emission estimates that are highly variable. So at a first glance, models don't seem reliable and accurate enough to enforce limit values for methane emissions. An alternative to modeling is measuring emissions. For this purpose, several methods are developed and tested in the past two decades. But at the moment there is no agreement on what methods are best applicable, and no single method is generally accepted as sufficiently accurate and still cost-effective.

The lack of proper tools for estimating methane emissions is no desirable situation. It is hard to assess both the impact of measures taken by landfill operators and policies developed by policy. As a result, local measures and national policies for reduction of landfill methane could still be more effective. The Sustainable Landfill Foundation (SLF) is committed to minimization of landfill methane emissions. Since at the moment both model-approaches and direct measurement of emissions are not yet considered accurate enough, SLF commissioned OonKAY! to perform a comprehensive and critical review was performed of both available models and measurement methods with the following objectives:

[1] Carbon dioxide emissions from landfills stem from a short carbon cycle. Upon growth of the organic materials carbon dioxide is sequestrated and the total cycle of growth, use and finally decomposition takes place over a interval of several months to maximum several decades. This is very short compared to the time interval of growth, use and decomposition of materials from fossil origin. Therefore CO_2-emissions from such short-cycle are 0 by definition.

PROJECT OBJECTIVES

- A literature review on methods for quantification of annual average methane emissions from an individual landfill.
- Evaluation of the methods, a.o. whether the methods meet the minimum standards as described in IPCC or E-PRTR guidance documents.
- Discussion of options for improvement and potential directions for harmonization.

THIS REPORT

This report contains the findings on the review. Chapter 2 gives information on modeled emission. Chapter 3 describes progress in developing measurement methods. Chapter 4 gives some information on yet another method to quantify emissions, based on recovered amounts of methane. Chapter 2, 3 and 4 discuss methods, irrespective of the context in which they are used. This context (UN-FCCC E-PRTR or enforcing emission limits) is of importance considering possible improvements in both models and methods in chapter 5. Differences in definition of the quality of a method result in slightly different ways ahead for each application.

CHAPTER 2: MODELING METHANE EMISSIONS

2.1 INTRODUCTION

Methane emissions might be obtained from models. Methane emissions are generally calculated from the methane mass-balance:

emissions = generation - recovery - oxidation (eq.-1)

Where methane generation is calculated as

methane generation = LFG generation * methane content (eq.-2)

When modeling methane emissions, most of the discussions are about modeling methane or landfill gas formation. There are numerous models around, most of the based on a first-order decay model or a multi-phase model. Modeling oxidation has received less attention. In most cases 10% of methane flux through the top-layer is assumed to be oxidized. However more recently other ways to deal with oxidation are being developed. This chapter gives an overview of all parts of the methane mass-balance as described in equations 1 and 2.

2.2 MODELING LANDFILL GAS FORMATION

2.2.1 GENERAL

When waste is landfilled, the organic matter in the waste is converted to landfill gas. Landfill gas is a mixture of methane (45-60%), carbon dioxide (40-55%) and trace components (H_2S, mercaptanes, organic esters and other volatile hydrocarbons, all of them giving landfill gas its characteristic smell).

Biodegradation of organic matter proceeds in a number of steps. A general description of consecutive steps was proposed by Farquhar and Rovers (1973). The degradation of organic material was by them as a sequential process of hydrolysis of the solid organic materials (e.g. hemicellulose, cellulose) into larger soluble organic molecules, subsequent fermentation of these materials, yielding organic acids and finally methanogenesis.

Organic material is not a single component, but consists of a broad spectrum of molecules with varying degradability. Smaller molecules, such as simple sugars and fats are easily degraded. Hemicellulose is also relatively easily converted, cellulose somewhat slower, as long as it is accessible for enzymes and bacteria. Lignin however is resistant to biodegradation under anaerobic conditions[2] and lignin can shield

[2] Anaerobic (no oxygen present) conditions are e prerequisite for methane formation. Under aerobic (oxygen-rich) conditions waste might biodegrade, but this process only yields CO_2.

cellulose, thus preventing it from biodegradation. According to Chandler et al. (1980) a relationship exists between lignin content and the maximum biodegradability of organic material under anaerobic conditions, as indicated in the figure below.

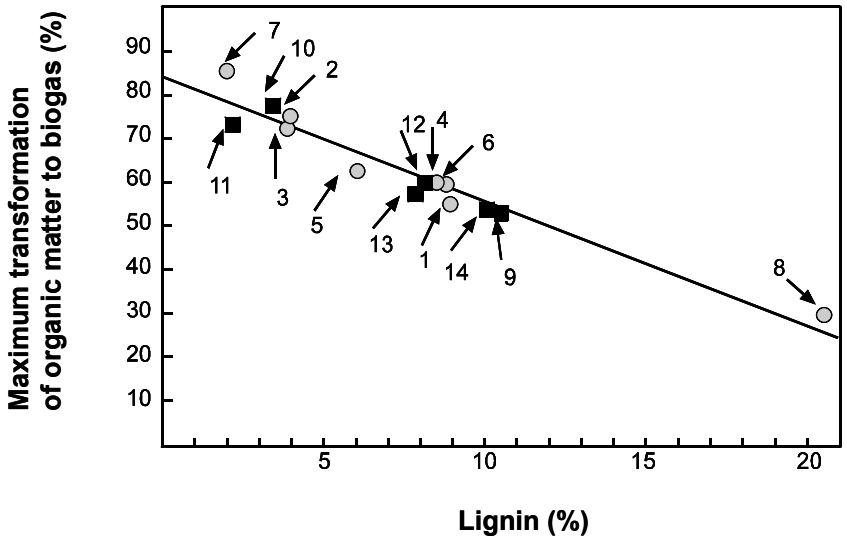

FIGURE 1: RELATIONSHIP BETWEEN FRACTION OF ORGANIC WASTE ULTIMATELY CONVERTED AND THE LIGNIN RATION OF THE WASTE (1 = WHEAT STRAW, 2 = CORN STALKS, 3 = CORN LEAVES, 4 = PURPLE LOOSESTRIFE, 5 = SEAWEED, 6 = WATER HYACINT, 7 = CORN FLOUR, 8 = NEWSPAPER, 9 = ELEPHANT MANURE, 10 = CHICKEN MANURE, 11 = PIGS MANURE, 12 EN 13 = COW DUNG; CHANDLER ET AL., 1980).

So not all organic material can be converted to landfill gas. And in practice not everything that can be converted will be converted, simply because conditions in parts of the waste inhibit biological activity. There are many possibilities why degradation is inhibited, e.g. because waste is locally too dry or because the waste was frozen upon landfilling and temperatures subsequently stay too low. It is also possible that the waste has excess water, leading to stagnant saturated zones in the waste, where the first two steps of biodegradation are fast and result in a drop of pH, thus limiting methanogenesis.

So the methane formation potential is generally based on the total amount of organic material, corrected for (i) the amount of organic material that does not degrade under anaerobic conditions and (ii) the amount that doesn't degrade because conditions are not favorable. The first amount is defined by the waste composition. The second part is determined by landfill design and operation and is most likely also influenced by climate conditions.

HISTORY OF LANDFILL GAS MODELING

Attempts to model landfill gas formation stem from the early '80's. In those days methane emissions was not yet recognized as a potential problem; however one was aware of the energetic potential of the landfill gas and eager to exploit this alternative energy source. So the first landfill gas formation models were made to help determine the size of landfill gas recovery projects: how much gas is formed, what are expectations for the next 10 years and which part of it can be recovered?

Since the mid-90's modeling emphasis shifts to quantification of methane emissions, first on a national scale (in the framework of obligation of countries to report greenhouse gas emissions to UN-FCCC) and afterwards as well on a landfill by landfill basis (in the framework of E-PRTR). The call for improved accuracy, transparency and the desire for benchmark emissions (comparison of methane emissions between nations or between landfills, comparison of UN-FCCC reported emissions by a country and emissions reported by individual landfills in this country) resulted in a number of emission models that can be considered state of the art.

2.2.2 Calculating methane generation

Determining methane potential

The methane potential, L_0, is the amount of methane that is produced throughout the lifetime of the waste. In most generation models L_0 is the amongst the most important parameters. As described above, landfill gas and methane are produced upon decomposition of organic parts of the waste. An often approach for determining methane generation from biodegradation is based on:

$$(CH_2O)_n \rightarrow ½\ n\ CH_4 + ½\ n\ CO_2 \quad \text{(eq.-3)}$$ [3]

In which $(CH_2O)_n$ is the approximate composition of organic matter in the waste. The methane potential or the landfill gas generation potential is generally described as proportional to the product of amount of waste landfilled (W) and the concentration of organic carbon (DOC[4,5]) in the waste. However described in chapter 2.1, not all organic material is converted. Part of it (lignin, cellulose covered by lignin) is not degradable under anaerobic conditions. Another part simply doesn't degrade because conditions in the waste are unfavorable for degradation. So when calculating L_0, a factor DOC_f is introduced that describes the part of DOC that ultimately is converted to landfill gas. The methane potential per ton of waste depends on the methane concentration in the landfill gas and L_0 is ultimately calculated as[6]:

[3] The reaction equation suggests a landfill gas composition of 50% methane and 50% CO_2. In reality methane concentrations are somewhat higher, due to biodegradation of components with a higher H/O-ratio. Part of the CO_2-produced is also dissolved and released as $CO3^{2-}$ in the water-phase in the landfill (the leachate).

[4] A full overview of symbols used is given at the end of this report.

[5] Please note the difference between organic carbon and dry organic matter. DOC generally refers to the amount of C in the $(CH_2O)_n$ and dry organic matter contains about 40% DOC.

[6] Model descriptions seem to differ in this aspect, but on a closer look they are all the same. E.g. IPCC calculates methane as F * 16/12 * DOC * DOC_f, which is similar to equation above. TNO calculates landfill gas formation potential (in $m^3\ hr^{-1}$) as 1,87 * DOC * DOC_f. Assuming a fraction F in the landfill gas and a density of methane of 0,72 kg/m3, L0 is obtained of F * 1,87/0,72* DOC * DOC_f. Afvalzorg doesn't base its calculation on organic carbon (DOC), but on dry organic matter (DOM) and calculates

$$L_0 = 1{,}33 * F * DOC * DOC_f \quad \text{(eq.-4)}$$

In most models DOC_f is considered a constant value in between 0,4 and 0,7, depending on the model. DOC is generally calculated on the basis of waste composition, either through its origin (DOC-values for household waste, industrial waste, etc.), or from its micro-composition (% putrescibles, % paper, % textiles, etc.).

SIMPLE FIRST-ORDER DECAY MODEL

Landfill gas generation is often described as a first order process or a variation of this. A first-order decay process implies a relative large amount of landfill gas being formed immediately after deposition, gradually being reduced in time. Characteristic of a first-order decay process is a fixed half-time of landfill gas generation. When a half-time of 7 years is assumed, methane generation after 7 years is 50% of the initial generation (in kg methane per year), after 14 years 25%, after 21 years 12,5% and so on. In a first-order model methane generation (CH_4-f) in time from a an certain amount of waste (W), landfilled in a single year, is described as:

$$CH_4\text{-}f = W * L_0 * k\, e^{-kt} \quad \text{(eq.-5)}$$

In which L_0 is the methane generation potential of the waste, k is the rate constant of biodegradation and t is the time elapsed since landfilling of the waste.

A landfill generally consists of waste deposited in a number of years. Methane generation from such a landfill is calculated as the sum of multiple equations (as in equation-5), each describing the methane being formed from the waste landfilled in one year of operation. Either a spreadsheet program is used for this calculation, or the calculation is part of a larger mathematical program.

N.B. Landfill gas generation is often *described* as a first-order process (or a variation of this). From a mechanistic point of view it is not. In a first order reaction (well-known in chemistry and physics) a reactant has a chance to react in the next hour, day or year, and this chance is independent of the amount of reactant still available. First order reactions can be characterized by their half-life, which is the time in which 50% of the original amount of reactant has reacted. An example of a first order reaction is radioactive decay. The chance that a molecule of plutonium (^{238}Pu) falls back to uranium (^{234}Ur) in the next year is a fixed one. The half-life of ^{238}Pu is about 88 years, independent on actual plutonium concentration. The half-time of biodegradation, $t_{1/2}$, can be calculated from k through:

$$t_{1/2} = 0{,}693/k \quad \text{(eq.-6)}$$

IPCC-REVISED EQUATION

A problem with first order models as described above is that it is an approximation. The method yields a methane generation for each year as a discrete value, rather than a continuous declining amount. Figure 2 illustrates this. As a result an underestimation of methane generation is obtained, compared to the continuous curve. This

landfill gas formation as 0,75 * DOM * DOC_f. However when assuming DOM contains 40% DOC, the Afvalzorg-model and TNO-model are in agreement.

underestimation is depending on the assumed value of k, is about 3,5% for k=0,1 y^{-1} and increases when k increases.

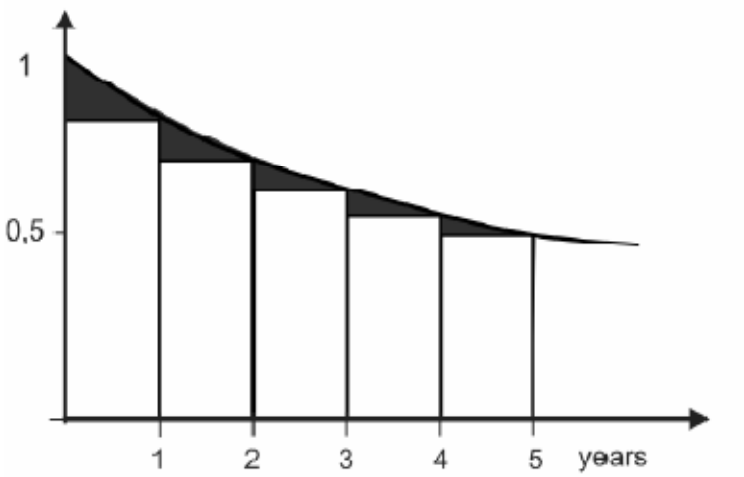

FIGURE 2: ERROR IN CALCULATED METHANE GENERATION IN A CONVENTIONAL FIRST-ORDER MODEL

The Landgem-model of US-EPA (Reinhart et al., 2005) minimizes the discrepancy by application of the conventional model per 1/10th of a year. The reason for this is that in the USA more and more landfill bioreactors are expected to be realized, where waste degradation is enhanced by leachate recirculation. Due to the high k-values encountered here (Reinhard et al., 2005, expect half-lives just in excess of 2 y) the error in the conventional first order model is increased. To accommodate Landgem for landfill bioreactors, Landgem is adapted to calculate per 1/10th of a year.

IPCC (2006) comes up with a more accurate equation, based on integrating the actual generation curve. The actual equation is more complicated as equation-1 and can be described as:

The discussion within IPCC was, whether the difference between both models is such a problem, because the error is relatively small in comparison to other model errors. But more important, first-order decay model parameters are validated (Oonk et al., 1994; Vogt et al., 1997) assuming the conventional first-order model. Upon validation a value for DOC_f was obtained that is 3,5% higher than the DOC_f that would have been obtained when a more accurate description was applied. So running a conventional first-order model, using model parameters validated for this model does result in an accurate estimation of methane generation. This is why IPCC considers conventional first-order models equivalent to the IPCC-revised equation.

MULTI PHASE MODEL

The multi-phase model is another elaboration of the first-order model (Hoeks, 1983). The multi-phase model describes that e.g. kitchen waste degrades much faster than wood or paper. Generally in multi-phase models three fractions are distinguished: fast, moderate and slow degrading waste, each with their own half-time of biodegradation.

The first approach assumes degradation of different types of waste to be completely dependent on each other. So the decay of wood is enhanced due to the presence of food waste, and the decay of food waste is slowed down due to the wood. The second approach assumes degradation of different types of waste is independent of each other. Wood degrades as wood, irrespective whether it is in an almost inert solid waste disposal site (SWDS) or in a SWDS that contains large amounts of more rapidly degrading wastes. In reality the truth will probably be somewhere in the middle. However the multiphase approach requires detailed information of carbon content and carbon quality of numerous waste categories. Usually reliable information is not or only partly available. Moreover there has been little research performed to identify the better one of both approaches (Oonk and Boom, 1995; Scharff et al., 2003) and this research was not conclusive.

Simplified models

A number of modeling approaches exist, that give less detail than the first order model described above. In general these are models developed in the early days of landfill gas generation modeling, developed for situations where little or no information was available on amount, age and composition of the waste. Since elaborated first-order and multi-phase models are now so readily available, these simpler models can no longer be considered 'state of the art' or ' good practice' (IPCC, 2000, 2006). In order to be complete, these models are only briefly mentioned here.

- Direct decay model. In a direct decay model, the whole methane potential of the waste landfilled is assumed to be released at one single moment (IPCC, 1996);
- In a zero-order model, waste is assumed to form a fixed amount of methane/landfill gas either for a fixed number of years or for eternity (e.g. Peer et al., 1992). This zero order model was until the midst 1990's frequently applied in design of landfill gas recovery (Vogt et al., 1997);
- A triangular model (Halvadakis, 1983) is similar to a zero-order model, but combined with a linear increase in generation in the first year and a linear decrease in the final years of landfill gas generation;
- A Scholl-Canyon model is a simplified first-order model (Emcon, 1980). Assuming annual amounts of waste deposited and waste composition to be equal throughout the exploitation period, a simplified equation is obtained.

More fundamental modeling Approaches

As described in chapter 2.1, generation of methane from organic matter in the waste actually progresses in a complex order of reaction steps, first enzymes break apart the solid organic macromolecules to smaller molecules, that are further processed microbiologically. A number of scientists are working on a more fundamental understanding of these processes and try to model the overall kinetics. An overview of activities is given by Lamborn (2005). However until now, these models do not produce reliable estimates of landfill gas generation from real batches of waste (Beaven, 2008). Most likely methane generation in actual landfills is governed by its heterogeneity and 'chance' plays an important role.

Lag time

In a simple first order decay model it is assumed that methane generation starts immediately after deposition of the waste. In reality this is however not the case. Most likely it takes several months to a year (Gregory et al., 2003; Bergman, 1995; Kämpfer and Weissenfels, 2001; Barlaz, 2004; IPCC, 2006) before all microbiological processes have started up and methane generation peaks.

Several approaches exist to describe methane generation in this initial period more accurately. The most simple approach is introduction of a lag-time. Methane generation is assumed zero during a certain time (e.q. 6 months) and afterwards methane generation is described as a normal first order decay process. In this way there is still a discontinuity at the moment when lag-time finishes and methane generation increases from zero to its maximum value in one day.

Therefore other approaches are developed as well, describing a slow increase of methane generation in the first months, after which first order decay gradually takes over (Findikakis et al., 1988; Keely, 1994; Van Zanten and Scheepers, 1995).

Methane correction factor (MCF)

Methane generation only occurs in parts of the landfill that are strictly anaerobic. In reality many landfills will not be completely anaerobic. Due to a.o. wind-activity and changes in ambient pressure parts of a landfill might contain oxygen, especially when a landfill is less well managed (no waste compactation, no daily covers, more thin or permeable temporary covers) and at older landfills where internal pressure due to gas production is reduced. In these parts methane generation is inhibited and aerobic decay of organic waste (not leading to methane) might take over. One way to deal with aerobic zones in the waste is the introduction of a methane correction factor (MCF), describing the part of the landfill that is not entirely anaerobic and from which no methane is generated.

Climate correction: ambient temperature precipitation

Climate in Europe is highly divers and methane generation from household waste in Finland will be different from generation in Italy. Methane generation is influenced by climate and mainly by temperature and precipitation and this has impact on both the decay rate of waste (the half-life) and amount of methane ultimately generated per ton of waste (L_0). Figure 3 describes climate zones in Europe and with respect to methane generation (and also methane oxidation, see chapter 2.4) at least 4 zones could be distinguished: (i) subarctic and highland, (ii) humid oceanic, (iii) humid continental and (iv) subtropical.

FIGURE 3: CLIMATE ZONES IN EUROPE[7]

It is known that *waste* temperature has an effect on the speed of methane generation (Hartz et al., 1982; Gendebien et al., 1992) but maybe also on the amount of methane being formed. However ambient temperature has little direct effect on the temperature of the deeper waste. But there might be an indirect effect. Initial stages of waste decomposition take place immediately after deposition and maybe even already in the bin and during collection and transport and ambient temperature in this period might have a long-lasting effect on methane generation in the landfill. For example, waste produced, collected and landfilled in the Nordic countries in winter will be largely frozen. As a result initial stages of waste decomposition will be seriously hampered and temperatures in the waste will remain low, compared to waste landfilled elsewhere in Europe.

The impact of moisture in the waste on waste decomposition is widely recognized. However its precise impact is still topic of scientific discussion. According to some, waste decomposition is enhanced by increased moisture content of the waste, until an optimum moisture content is reached. According to others movement of moisture in the waste is important (Klink and Ham, 1982). Too much stagnant moisture in the waste is even reported to inhibit waste decomposition (Oonk and Woelders, 1999; Wens et al., 2001). Moisture movement spreads methanogenic activity throughout the waste and avoids local build-up of inhibiting components. If the first

[7] From http://printable-maps.blogspot.com/2008/09/map-of-climate-zones-in-europe.html

is true, waste decomposition is favored by moisture content of the waste; if the latter is true, waste decomposition is favored by precipitation (Alexander et al., 2005) and a more permeable top-layer. Of course there is a correlation between moisture content of the waste and precipitation several models simply assume a correlation between the moisture content of the waste and the mean annual precipitation (Chian and DeWalle, 1979; McDougal and Pyrah, 2003).

2.2.3 Available generation models

The methods of calculating methane generation in the previous paragraph are mere descriptions how a certain potential of methane is released. For a practical application, these descriptions do not suffice. Therefore models are developed that enable calculation of methane generation in a specific year from landfilled waste. Input parameters in these generation models is the amount of waste landfilled in each year of exploitation and in most models also a specification of the waste. The model itself subsequently calculates DOC, DOC_f, L_0 and calculates the way this methane potential is released throughout the years.

In the past years a number of models have become available. Most of them consist of a spreadsheet program. Some of them are executables[8]. The most widely applied models are amongst the ones reviewed below. Some models (e.g. Calmin, the Finnish E-PRTR-model) are considered in this evaluation, because of they contain interesting features that might deserve follow-up.

The **IPCC-model** (to be obtained from IPCC, 2010) is developed by an international team of experts, and is intended to give guidance to national authorities in the quantification of methane emissions from all landfills in a country. But the model itself can also be used for individual landfills. The model itself is freeware and can be downloaded from the IPCC-website. Within the IPCC-process, transparency is of utmost importance and the method is described in detail by IPCC (2006). Input of the model is amount of waste per year and a classification of the composition of the waste in origin of the waste (household waste, industrial waste, etc.). Alternatively the model also allows for a waste-composition option, where waste can be defined in % food waste, % paper, % wood, etc. The choice exists between a first-order decay model (the IPCC-revised equation) and a multi-phase model (also based on the IPCC revised equation) and the default lag-time of 6 months can be adapted. The IPCC model accommodates for 4 different climate regions: wet boreal or temperate; dry boreal or temperate; wet tropical and dry tropical. The climate conditions chosen affect the chosen k-value.

The **TNO**-model (Oonk et al., 1994) is the first model, where model parameters were based on real data of landfill gas generation at a larger group of landfills. Methane and CO_2 emission measurements were used to validate the model (Oonk et al., 1995, Scharff et al., 2003). Both a first-order and a multi-phase model were made, that describe landfill gas generation as a function of amount of waste deposited from dif-

[8] an executable (file) causes a computer "to perform indicated tasks according to encoded instructions".

ferent origin (household waste, industrial waste, etc.). The model itself exists as a publication on paper, but a spreadsheet version is available on demand (Oonk, 2010).

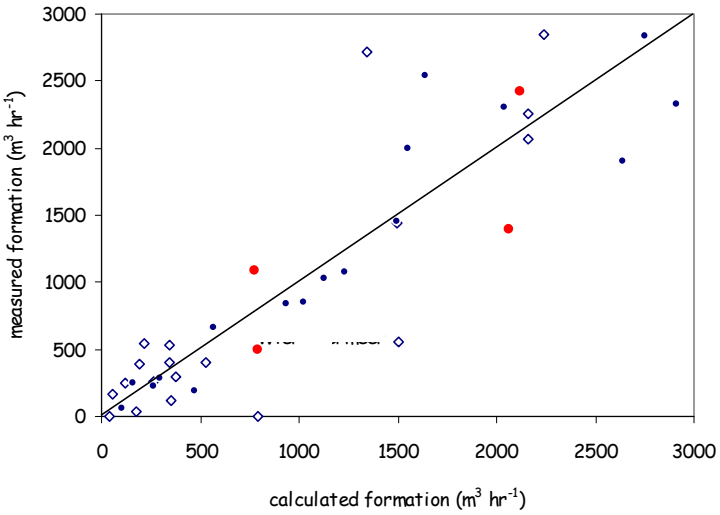

FIGURE 4: VALIDATION OF TNO-MODEL. COMPARISON OF CALCULATED AND MEASURED LANDFILL GAS GENERATION. BLUE DOTS ARE ESTIMATED FROM LANDFILL GAS RECOVERY (OONK ET AL, 1994), OPEN DOTS BLUE DOTS ARE FROM EARLY 90'S-MEASUREMENTS (OONK AND BOOM, 1995), RED DOTS ARE FROM EARLY 2000'S-EMISSION MEASUREMENTS (SCHARFF ET AL., 2003)

GasSim Lite is developed by Golder Associates (2010) for the Environment Agency of England and Wales. GasSim quantifies all landfill gas related problems of a landfill, ranging from methane emissions, effects of utilization of landfill gas on local air quality to landfill gas migration via the subsoil to adjacent buildings. At the moment (March 2010) GasSim 2.1 is the latest version and is commercially available; however a ' lite' -version 1.5 is available as freeware, and is designed to help operators with their pollution inventory,.
GasSim is an executable and default values used, algorithms applied and assumptions made are somewhat more hidden in the program. Information is however not confidential and staff of Golder Associates are willing to provide more information on demand (Gregory, 2010). GasSim is based on UK waste statistics and starts from hemicelluloses and cellulose content in the various waste fractions. For each waste fraction a DOC_f is assumed, based on research by North Carolina State University (Gregory, 2010).

Landgem is a model developed for and made available by US-EPA (2010). It is a first order decay model, with separate default values for k conventional regions, arid regions and for enhanced degradation cells[9]. The most recent version of the model is the 3.02-version, dated May 2005. The mathematics of Landgem is sometimes described somewhat confusingly as (note the W/10),

[9] In cells for enhanced biodegradation (bioreactors) landfill gas formation is accelerated by recirculation of leachate.

$$CH_4f_T = (W/\mathbf{10})* L_0 *f(T) \quad \text{(eq.-7)}$$

but in this way the model calculates methane emissions per $1/10^{th}$ of a year (for this purpose T is also expressed in tenths of a year, Reinhart et al, 2005). The reason for this is to avoid inaccuracies when k-values are used in excess of 0,1 y^{-1}.

The **Afvalzorg-model** is developed by NV Afvalzorg in the Netherlands. It is based on a combination of literature (as accumulated in the 2006-IPCC model) and own experiences with landfill gas generation and measured emissions at the Afvalzorg-sites at Nauerna, Braambergen en Wieringermeer. The model itself is a multi-phase model and is intended to give a more realistic prognosis of methane generation at landfills with little or no household waste deposited. The model itself is freeware and available on demand (Scharff, 2010).

The **French E-PRTR-model** (Ademe, 2003) is a simplified first order decay model. The model describes methane generation of 4.8 kg (6.6 m^3) per ton waste per year in the first 5 years after landfilling; 2.4 kg per ton waste per year the 5 years after, 1,3 kg per ton waste per year in the 2^{nd} decade and 0,6 kg per ton waste per year in the 3^{rd} decade after landfilling. For moderately decomposable waste (e.g. non-hazardous industrial waste; household waste that is milled or composted), methane formation is 50% of these values. The model is not available as a spreadsheet, but consists of a simple fill-in table.

The **Finnish E-PRTR-model** (Petäjä, 2010) is a multi-phase model with model parameters in line with the IPCC-model for wet boral or temperate regions. The model itself is completely in Finnish, which makes it tougher to evaluate here and difficult to apply for landfill operators outside Finland. The model itself however is interesting because the definition of waste-streams is based on the EWC-codes. This connects to the system of waste registration at landfills and reduces problems with waste definition.

Calmin is no generation model, however it serves a similar function and for reasons of clarity it is discussed in this chapter. Calmin is developed by researchers in USA by order of authorities in California, and quantifies methane emissions in a new and interesting approach. At the moment the beta-version is available on demand (Spokas, 2010). The model intends to provide an improved method for quantification of landfill methane emissions for the California greenhouse gas inventory. Calmin is not based on the methane mass-balance as described in equation 1. Instead it calculates methane diffusion through the top-layer and methane oxidation in the top-layer, ultimately yielding a methane emission. This methane emission is a function of the top-layers composition and the location of the landfill on the globe. For the latter purpose, the landfills coordinates are translated into climate conditions, processes are calculated for each day in the year and subsequently emissions are averaged. Weak point of the model is the assumption that emissions take place through diffusion. As a result the model applicability of Calmin might be limited on landfills where large part of emissions take place through preferential channels.
Calmin and its outcome are validated in a number of closed chamber measurements on two Californian landfills. However as indicated in chapter 3, closed chamber mea-

surements can not be considered a reliable measurement method, since they tend to miss methane emissions through preferential channels. In this respect, model and validation method seem to have similar flaws and the Calmin itself might give a good estimate of the part of methane that is emitted through diffusion.

2.2.4 Characterization of generation models

So there are numerous landfill gas generation models around. However all landfill gas generation model consist of two parts:

- a description of the total methane potential, L_0, which is the total amount of landfill gas which is formed during the lifetime of the landfill;
- a function f(t), that describes how this potential is released over time. So which fraction of the total methane potential is released in the 1st, 2nd, 3rd year and so on.

Total methane generation in year T (CH_4-g_T) can be described as follows:

$$CH_4\text{-}g_T = W^* L_0 * f(T) \qquad \text{(eq.-8)}$$

The function f(t) is in most models a first-order decay model, a multiphase model or a variation of this.

Rate constant of biodegradation (k), half-life of methane generation

In many evaluations of model parameters (e.g. Kühle-Weidemeier and Bogon, 2009), most attention is paid to the half-life of methane generation (or the rate constant of biodegradation, k). However, in many cases, the outcome is not that sensitive for assumed half-life or assuming multi-phase degradation, rather than first order degree degradation. This is illustrated in the model calculation in Figure 5. In this example, of k on methane generation is limited to about 20%, for k between 0,07 and 0,14 (half-lives of 5 tot 1 years). A change in k only results in a change of time when methane is assumed to be released. Shorter half-lives or higher values of k imply that the methane potential is released somewhat earlier, more during ad immediately after exploitation. Longer half-lives imply a shift in methane generation to the period after exploitation.

At the very low end of k (half-lives assumed in excess of 15 years), result in a methane generation that is both reduced during exploitation and also afterwards (compared to the assumed generation with k=0,1/y). Only on a very log term, this will be compensated by an increased methane generation. Studies indicating very long half-lives of methane generation (e.g. under arid conditions, Atabi et al., 2009) should be considered with great care. In these cases reduced methane gas formation can be caused by both a reduced rate of biodegradation as well as a reduced methane generation potential (L0, see below) and the difference between both can only be observed decades after closure.

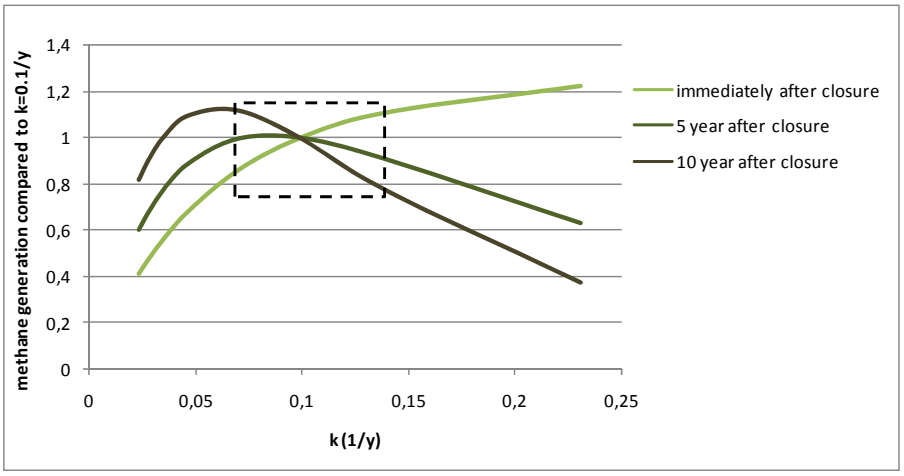

FIGURE 5: EFFECT OF ASSUMED RATE CONSTANT OF BIODEGRADATION ON METHANE GENERATION (METHANE GENERATION COMPARED TO GENERATION, CALCULATED ASSUMING K=0,1/Y). CALCULATION PERFORMED WITH THE AFVALZORG-MODEL FOR A LANDFILL WHERE SIMILAR AMOUNTS OF WASTE ARE DUMPED DURING 20 YEARS.

METHANE GENERATION POTENTIAL, L_0

Compared to k, L_0 (the methane generation over time from a ton of waste), has received considerably less attention. There are two ways to quantify L_0.

One way is a direct estimation of the methane or landfill gas generation potential in a field validation as the TNO-model and the model by Vogt et al. (1997. From a larger data set of waste characteristics at one hand, and methane generation at the other hand, L_0 can be obtained by linear regression. Assuming a value for DOC, DOC_f is subsequently estimated.)

The other method is to quantify it from equation 4, where DOC generally is estimated from waste analyses and DOC_f is obtained from literature. Most generation models in 2.2.2 are built this way.

$$L_0 = 1{,}33 * F * DOC * DOC_f \qquad \text{(eq.-4)}$$

VALUES FOR MODEL PARAMETERS

Table 1 refers to household waste or MSW. Most of the experiences with landfill gas generation comes from landfill recovery on this type of waste; the large-scale validation studies by Oonk et al. (1994) and Vogt et al. (1997) are performed on this type of waste; most of the emission measurements are performed on landfills with MSW. However in Europe, landfilling of MSW is more and more discouraged and as a result the non-municipal solid waste becomes more and more important for methane formation. Table 2 describes how methane emissions from industrial waste are handled by various models. It is well-known that industrial waste can contain a wide range of DOC. Examples of wastes without any significant DOC are wastes from the steel industry or asbestos wastes. In countries where biodegradable wastes are to a large extent banned from landfills an average industrial waste carbon content may no longer be appropriate. The Finnish approach defining DOC for each waste in the European Waste Catalogue cannot be presented in Table 2. The European Waste Cata-

logue has approximately 800 entries. This may nevertheless be a more suitable approach for landfills accepting wastes with low DOC.

Since they are based on extensive field-studies, the TNO-model and the model by Vogt et al (1997) might be considered as best guesses. The TNO-model can be considered representative for humid oceanic region and the model of Vogt et al. (obtained for landfills in California) as being representative for landfills in more subtropical conditions, where conditions are more dry, landfill generation might be inhibited, resulting in longer half-times and ultimately also a reduced conversion of organic material to landfill gas (hence a reduced DOC_f and L_0).

Table 1: Comparison of models for methane potential (kg methane per ton waste)and half-lifes for biodegradation for household waste or MSW

	L_0 (kg/ton)	half-life (year)	remark
IPCC-model	63[1]	12-23 (slow)[2,3] 7 (moderate)[2] 4 (fast degradable)[2]	MSW Europe
TNO-model	60	7	Dutch HHW
GasSim	51[4]	15 (slow) 9 (moderate) 6 (fast degradable)	HHW UK
Landgem	122 ('CAA')[5] 72 ('inventory')[5]	14 ('conventional')[6] 35 (' arid')[6]	MSW USA
Afvalzorg	39-45	23 (slow) 7 (moderate) 3 (fast degradable)	Dutch HHW
E-PRTR (Fr)	55	5-10	HHW France
E-PRTR (Fi)	65	23 (slow) 14 (moderate) 3,5 (fast degradable)	HHW Finland
Vogt et al. (1997)	44	17	MSW California

[1] value for bulk MSW

[2] values for wet boreal and temperate regions. For dry regions and tropical conditions other k-values are suggested;

[3] different half-lives specified for paper-like materials and wood-like materials;

[4] sum of methane emissions in the 1st 100 years after landfilling of 1 ton of 1980-2010 100% household waste, assuming no recovery and 0% oxidation, as calculated using GasSim Lite

[5] CAA defaults are based on requirements for US landfills, as specified in the Clean Air Act. Inventory-defaults are based on results of an inventory by US-EPA

[6] ' arid' refers to regions with less than 625 mm (25 inch) rainfall per year. 'Conventional' refers to non-arid regions.

Compared to the TNO-model, the IPCC-model and the Finnish E-PRTR-model has about the same L_0. The average half-time of the half-times of the IPCC model is about the same as the half-time of the TNO-model. Application of both models will

give about the same result, when applied to a landfill[10]. GasSim will give about 20% less methane generation. Afvalzorg about 30-35% less methane emissions. Landgem ('inventory' L_0 and ' conventional' half-time) however, has a higher L_0 but a substantially longer half-time. As a result initial methane generation might be comparable to IPCC, TNO or GasSim). On the longer term, Landgem will most likely overestimate emissions.

TABLE 2: COMPARISON OF MODELS FOR METHANE POTENTIAL AND HALF-LIFES FOR BIODEGRADATION FOR INDUSTRIAL WASTE

	L_0 (kg/ton)	half-life (year)
IPCC-model	50	12-23 (slow)[1,2]
		7 (moderate)[1]
		4 (fast degradable)[1]
TNO-model	50	7
GasSim	26[3]	15 (slow)
		9 (moderate)
		6 (fast degradable)
Landgem	not specified	not specified
Afvalzorg	36-39	23 (slow)
		7 (moderate)
		3 (fast degradable)
E-PRTR (Fr)	28	5-10

[1] values for wet boreal and temperate regions. For dry regions and tropical conditions other k-values are suggested;
[2] different half-lifes specified for paper-like materials and wood-like materials;
[3] sum of methane emissions in the 1st 100 years after landfilling of 1 ton of 1980-2010 100% industrial waste, assuming no recovery and 0% oxidation, as calculated using GasSim Lite

2.2.5 EVALUATION OF GENERATION MODELS

Apart from the aforementioned validation efforts of the TNO-model (Oonk et al., 1994; Oonk and Boom, 1995; Scharff et al., 2003) and the validation study of Vogt et al., (1997), there have been several other attempts to validate formation or emission models, for example:

- Ehrig and Scheelhase (1999) interpreted recovered amounts of methane at German landfill. On the basis of these data they suggest a methane generation

[10] It has to be noted that the IPCC-model ends up at as similar L0 but in a different way. IPCC combines a relative high value of DOC with a low value of DOC_f and F. In this way one ends up at similar methane generation potential. However the same DOC and DOCf are also at the basis of another important effect of landfilling: the amount of organic carbon that is sequestrated in the landfill. As a result of the same relative high DOC and low DOCf, IPCC ends up with a much higher amount of carbon sequestrated as e.g. the Afvalzorg model.

of about 2,5 kg methane per ton waste[11], 10 years after closure of the site. Kühle-Weidemeier and Bogon (2008) reinterpret these data and conclude a best fit is obtained assuming an L_0 of 80 kg methane per ton waste[12] and a half-life of biodegradation of 3,5 to 6 years.

- Fellner et al. (2003) compares modeled methane generation with actual methane generation for landfills described in literature. Unfortunately they only validate a model by Tabasaran and Rettenberger (1987) with a L_0 of 93 kg methane per ton waste and a half-time of biodegradation of 20 years. The conclusion was that the model overestimates generation in most of the cases.
- Scharff and Jacobs (2005) compared the outcome of a number of models (a.o. The TNO-model, the Afvalzorg-model, Landgem, GasSim and a zero-order models) with measured emissions at three Dutch landfills. For individual landfills, differences between models was enormous (difference between the lowest and highest estimation was more than a factor 10. In one case even a factor 20). According to Scharff and Jacobs, this is in indication that current models give no reliable methane emission. Methane emission measurements are also uncertain; however discrepancy between different measured emissions is much less as the difference between modeled emissions.
- Fredenslund et al. (2007) compare 4 generation models (Landgem, IPCC-model, GasSim and the Afvalzorg-model) at a landfill site in Denmark. Huge differences are observed between models, with highest generation in Landgem and lowest generation and lowest generation for GasSim and the Afvalzorg model. Also within a model, results are highly depending on specific assumptions. On basis of this comparison, Kühle-Weidemeier and Bogon (2008) conclude it is questionable whether generations models are reliable.
- Thompson et al. (2009) validated a number of generation models in a comparison with recovery at Canadian landfills. This article however has to be interpreted with care, since it appears to be erratic[13].

There are also some efforts to validate models in very arid or tropical zones. Although similar climates are not found in EU, these studies are of interest, since they illustrate the effect of climate on methane generation:

- Atabi et al., (2009) validate Landgem for a landfill in Iran in extreme arid conditions in a comparison with recovered amounts of landfill gas. Landgem, assuming a rate constant of biodegradation of 0,02/y (a half-time of biodegradation of 35 years) gave good description of landfill gas generation. As described in chap-

[11] Interpretation: Ehrig and Scheelhase conclude gas formation ten years after closure of the site $\leq$ 10 m^3/y per ton dry solids.

[12] Kühle-Weidemeier and Bogon estimate total gas potential of 196 m^3 per ton waste. L_0 is calculated assuming 57% methane in the gas and a density of methane of 0,72 kg/m^3.

[13] In some of the models reviewed landfill gas generation (in m^3/y) seems to be mistaken for methane generation (in kg/y). This results in a methane generation, which is about 2,5 times too high.

ter 2.2.3, it is likely that reduced landfill gas formation here is the result of both a reduced methane generation potential (L_0) and a reduced rate of biodegradation (k)

- Machado et al. (2009) measured the methane generation potential in waste samples of different age at a Brazilian landfill. Reduction in this potential could explained by a first order decay process with a L_0 of about 70 kg methane per ton waste and a half-life of biodegradation, of 3.5 years.
- Wangyao et al. (2009) describe rapid decomposition of waste on a landfill in Thailand. Methane emissions were measured in 2008 and 2009. Despite the high heterogeneity of emissions, a reduction in arithmetic mean emissions was observed, from which a half-life of biodegradation of about 2 years was estimated. This high rate of biodegradation is related to both the nature of the waste (containing a lot of putrescibles) and climate.

However interesting these validations may seem, they are all based on one to few landfills. are illustrative for problems encountered when trying to find a suitable model for the specific emission situation. As illustrated for the TNO-model in Figure 4, the uncertainty in a generation model is large, and depending on the quality of waste data, the chance exists that generation is over or underestimated by 25-50%. This makes it hard to draw conclusions on the basis of experiences at only one or a few landfills. For a proper validation of emissions a much larger set of observations is required, before one can conclude whether a model is on average a good predictor of methane generation.

In this report, evaluation of models is not only related to accuracy. Indicators as ‘scientific basis’, ‘transparency’ and ‘validated’ are also used. They indicate whether model-assumptions made are clear and in line with science. The evaluation is summarized in Table 3 and explained in more detail in the paragraphs below.

Availability: All models are freeware. This includes GasSim Lite, which is the freeware version of GasSim. GasSim Lite enables landfill owners to fulfill their reporting obligations in the framework of E-PRTR. In Table 3 a ‘++’ means that the model can be downloaded from the web. A 4 means available on demand. A ‘-’or ‘--’ means that users have to do considerable efforts to obtain a version of the model.

Ease of operation refers to the required expertise of the user with the specific model and the complexity of choices required by the user. Number of different manipulations/actions before a result is derived. In case of GasSim a ‘-’ is given, also because the model requires information that is not used in calculating methane emissions. The Finnish E-PRTR model get a ‘-‘, because it is set in Finnish and therefore less easy to operate for non-Finnish language.

Transparency refers to a proper description of the model, model parameters used, assumptions made and efforts done do validate the outcome of the model. A spreadsheet-based model is in itself more transparent than executables as GasSim and Calmin, since the method of calculation and default-values used can be traced back. For literature references related to transparency, see the description of the models chapter 2.2.3).

TABLE 3: SUMMARY OF EVALUATION OF METHANE GENERATION MODELS[1)]

	IPCC	TNO-model	GasSim	Landgem	Afvalzorg	Calmin	E-PRTR (Fr)	E-PRTR (Fi)
operational								
- availability	++	+	++	++	+	+	+	+
- ease of operation	+	+	-	+	+	0	++	-
- required input	0	+	+	0	+	0	0	+
performance								
- scientific basis	+	+	0	0	+	-	+	+
- transparancy	++	+	-	0	0	0	+	-
- validated	0	+		0	0	0	0	0
constraints								
- waste changes	+	0	+	-	+	--	-	+
- climate zones	0/+	-	-	0	-	+	-	-
accuracy	0	0	0	-	0	-	0	0

[1)] In this table '++' means very good and '--' means very poor. If a generation model scores '-' or less on one of the evaluation parameters, users of the model should be well aware of the limits of the model.

Required input. More detail of input is considered here an advantage. It allows a more accurate prognosis of landfill gas generation, since it might bring the flexibility to incorporate circumstances that are specific for this landfill. The model itself is evaluated in a more positive way, when the way the input parameters can be defined is in line with the type of information available at the landfill. So waste can be specified according to its source (household waste, offices waste, commercial waste, etc., as in the TNO-, Afvalzorg model) rather than its composition (putrescibles, paper, plastics, etc. as in the IPCC-model). Specification according to its source is preferred, since it connects to the way information is available at the landfill.
GasSim and the Finnish E-PRTR-model give the possibility to both change the amounts of waste per waste categorie, but on top also accommodates changes in composition of the waste streams. So a landfill operator can calculate the effect of both less household waste and a change in household waste composition, e.g. due to a reduced paper content.
In case of Landgem and the French E-PRTR-model, little or no room exists to specify waste composition detail of input is considered too low for an accurate model.

Scientific basis refers to whether a model can be considered ' state of the art' from a scientific point of view and **transparancy** refers to how clear assumption are. The IPCC-model, The TNO-model and the Afvalzorg model can be considered state of the art. GasSim seems to be state of the art as well, but is given a neutral value, because the scientific basis cannot be evaluated due to lack of transparency. The French E-PRTR model is very simple in comparison to other models. However its L_0 and half-life are in line with other models, and its outcome will be about the same. There is

no evidence that its prognosis of methane generation will be less accurate than other model prognoses. The scientific basis of Landgem is considered less, because of the high vales for L_0 assumed in the model. Calmin gets for the moment a negative evaluation, since the discussion on Calmins' approach is still pending.

Validated model – The TNO-model is most extensively validated. The model parameters themselves are determined in a comparison with landfill gas recovery at 9 landfills. The resulting generation is validated in a comparison with measurements of landfill gas emissions Dutch on 25 Dutch landfills, using a 1D mass balance method. The Afvalzorg model is validated in a more limited effort, using experiences from one Danish and three Dutch landfills. Landgem is based on the results of the validation of Vogt et al. (1997). Calmin is validated in a comparison with results of closed chamber measurements on 2 Californian landfills. However as concluded in chapter 3, closed chambers measurements cannot be considered a reliable method to measure methane emissions. The IPCC-model is not validated itself, but is for a large part based on the TNO-model and uses a comparable L_0 (although calculated in a different way). Validation of the other models is unclear.

Waste changes. The IPCC, TNO, GasSim and Afvalzorg-model can handle changes in waste composition. The default values in the TNO-model however seem to be a bit outdated. The approach in the IPCC and GasSim at one hand and Afvalzorg-model and TNO-model at the other hand differs: In IPCC and Landgem the composition of the waste can be defined (amount of putrescibles, paper, plastics, etc.). In the Afvalzorg- and TNO-model changes in origin of the waste can defined (e.g. amount of household waste, offices waste, commercial waste, etc.). As a result, the Afvalzorg-model is more suited to deal with changes in origin of the waste, where the IPCC-model changes in the composition of e.g. household waste. GasSim and the Finnish E-PRTR-model accommodate both changes in origin of the waste and changes in the composition of each individual stream. The French E-PRTR model does accommodate for changes in waste, but its assumption of a 50% reduction in methane generation is quite rough. Landgem does not accommodate for changes in waste composition. In Calmin waste composition is assumed not to be of influence on methane emissions.

Applicability to various climate zones. As described in chapter 2.2, climate has impact on methane generation and both the amount of methane generated per ton of waste, and the speed at which this is generated is influenced by climate. Most models however are made and validated for northwestern Europe (so the part indicated in Figure 3 as humid oceanic) and have to be considered less accurate when applied to other regions in Europe. Out of all models evaluated only the IPCC-model and Landgem distinguish somewhat between climate zones (these two models only the effect of 'wet' and 'dry' on half-life of methane generation, so methane generation in landfills in the south of Europe will be somewhat delayed).

The **accuracy** in Table 3 refers to the accuracy for types of waste and climate conditions for which the method is developed. Apart for the TNO-model, which is validated for waste landfilled in the Netherlands in the period up to 2000, there is little

or no information available on the basis of which methods mutually can be compared. In general most generation models are built on reasonable assumptions it is impossible to conclude that one set of reasonable assumptions yields a more accurate result than the other set.

Having said this, the IPCC-model, GasSim and the Finnish and French E-PRTR-model seem to be in fair to good agreement with the TNO-model for household waste/MSW. So for humid oceanic zone and for waste dominated by MSW these models won't be too far off. The Afvalzorg-model seems to underestimate methane generation from MSW, but the strength of this model lies in landfills with waste from other sources. On the other hand assumptions in Landgem and Calmin seem to be less reasonable. The methane potential, L_0, in Landgem seems rather high (also in comparison with the validation study of Vogt et al., 1997) and therefore Landgem will most likely overestimate generation at most landfills. Calmin only makes an implicit prognosis of landfill gas generation. This generation excludes methane emitted through short-cuts and therefore results in an underestimation of methane formation at landfills, where large part of methane is emitted through these short-cuts.

2.3 Methane content, recovery

The amount of methane recovered is generally calculated from the amount of landfill gas recovered and the methane content:

methane recovery = landfill gas recovery * methane content (eq.-9)

Landfill gas recovery

The most accurate way to obtain the amount of landfill gas recovered is by continuously measuring the flow of landfill gas to utilization and/or flare, by using a turbine meter. The measurement has to be corrected for temperature, pressure and moisture content, so pressure and temperature has to be measured and moisture content can be calculated from temperature, assuming full saturation of the gas.

When amount of gas is not metered, landfill gas recovery might be estimated from energy production, e.g. assuming 1,8 kWh produced per m3 of landfill gas extracted. However this estimate of landfill gas recovery is much les accurate as a metered recovery.

There is no accurate way to estimate the amount of landfill gas recovered. As indicated in chapter 4 in this report, 25% to 75% recovery efficiency can be expected, when the system of landfill gas recovery can be considered state of the art. IPCC gives a default recovery efficiency of 20% for systems without any further specification (IPCC, 2006).

METHANE CONTENT

Methane content of the landfill gas generated is a somewhat neglected parameter. Some emission models (as the IPCC-model and GasSim) assume a default value of 50%. However actual measurements indicate that average methane content in landfills will be more in the order of magnitude of 57% (Oonk and Boom, 1995).

Methane content are relatively easy measured using from time to time (daily to weekly) a simple FID-analyzer in the total amount of recovered gas. On basis of these measurements weighed annual average methane concentration can be calculated.

2.4 METHANE OXIDATION

2.4.1 PROCESSES OF METHANE OXIDATION

When methane passes through the top-layer, it enters an oxygen containing zone where bacteria can convert part of the methane to produce CO_2. This process, normally referred to as methane oxidation can be described as:

$$CH_4 + 2\ O_2 \rightarrow CO_2 + 2\ H_2O \qquad \text{(eq.-10)}$$

There are several factors that control the amount of methane being oxidized, the most important ones being:

- The homogeneity at which methane is emitted. At landfills, large part of the methane is released through short-cuts. These short cuts are all types of cracks and ruptures at the surface or subsurface, but also gas-wells or drainage pipes that are not well sealed or are leaking. As a result methane emissions are highly heterogeneous (see also chapter 3.1) and methane oxidation at hot-spots is most likely much less than oxidation methane that is emitted in a more homogeneous way;
- The flux of homogeneously emitted methane (the flow of methane from the bulk of the waste to the bottom of the top-layer). When this flux increases, diffusion of oxygen into the top-layer is reduced and methane oxidation itself as well (Scheutz et al., 2009a);
- The porosity of the top-layer. Increased porosity implies at one hand a more homogeneous methane emission. At the other hand, oxygen diffusion into the top-layer is enhanced. So increased porosity is advantageous to methane oxidation. Water-logging in periods with high precipitation decreases porosity (Gebert et al., 2009);
- The water-content of the top-layer. Bacteria need moisture to be active and bacteriological activity is favored by moisture. However too much water might block the pores. So there is an optimum water content of the top-layer (Börjesson et al., 1997, Cabral, 2004)
- The temperature of the cover-layer, which is closely connected to ambient temperatures. At higher temperatures bacteria become more active. Every 10 ^{o}C temperature increase means about 2-4-fold increase in methane oxidation (Gebert, 2007).

As a result of its moisture and temperature dependency, methane oxidation depends on average weather conditions. So it is climate-dependent. Methane oxidation is described to be at its maximum in temperate to warm conditions with limited excess rainfall. Methane oxidation is most likely less in colder climates and under warm but dry conditions (e.g. Abichou et al.,2010).

For a specific top-layer it is also depending on the season; methane oxidation is in winter is less than in summer. This is observed in Nordic countries, as Denmark (Christophersen and Kjeldsen, 1999) , Sweden (Maurice and Lagerkvist, 1997; Börjesson et al., 2007), Belgium (Boeckx et al., 1996) en in northern parts of USA (Czepiel et al., 1996b).

2.4.2 Methods for modeling oxidation

Application of simple defaults:

IPCC (2006) acknowledges the lack of reliable field measurements on oxidation and therefore propose a careful 10% default value for well managed landfills. This 10% default is meant to be a conservative first guess, leaving room for improvement. Since they were published, the IPCC default value drew a lot of discussion and proposals for improvement:

- Börjesson et al. (2007, see Figure 6) performed measurements at a few landfills in Sweden. Some landfills were still in operation while others were recently closed. The measurement method is based on 13C of the methane in the plume, a method which can be considered as one of the more reliable methods to quantify methane oxidation (see chapter 2.4.2). Börjesson et al. explicitly pay attention to improved default values for methane oxidation and propose 10% for active and 20% for closed landfills.

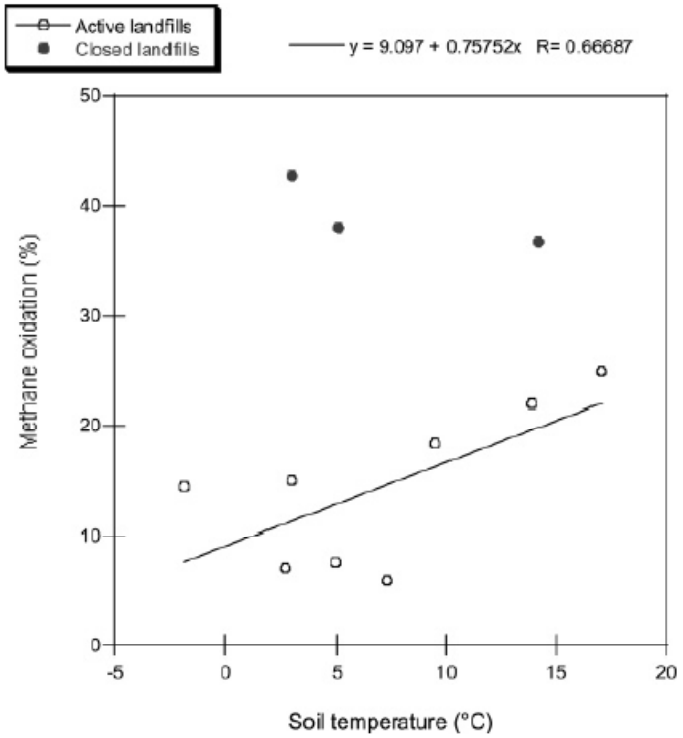

FIGURE 6: CORRELATION BETWEEN SOIL TEMPERATURE AND METHANE OXIDATION ON SWEDISH LANDFILLS (BÖRJESSON ET AL., 2007)

- Kühle-Wiedemeijer and Bogon (2008) review including methane oxidation on the basis of a literature survey in scientific journals and available grey literature on this topic (o.a. German research on methane oxidation at landfills for mechanically-biologically pretreated waste and an interpretation of the measurements of Oonk and Boom, 1995 and Scharff et al., 2003). They ultimately conclude there is no solid basis for the definition of more accurate default-values and therefore they propose to stay close to the IPCC default values (10% when methane flux is higher than 1,5 g CH_4 m^{-2} hr^{-1} and 15% oxidation when the flux is lower), but mention this is most likely an underestimation.
- Chanton et al. (2009) also put the IPCC-default value at discussion in a review that limits itself to peer-reviewed literature. They conclude that only 1 out of 10 measurements result in a value of less than 10%. Average of all available measurements is 35%. It has to be noted that Chanton et al. (2009) are not critical towards any of the measurements and simply make an average of all available measurements, both the reliable ones as well as the ones performed with less reliable methods (e.g. many measurements are performed using flux chambers, a method known for its inaccuracy on larger surfaces).
- GasSim gives the opportunity either to choose the 10% IPCC default, or to use your own value. When the latter is chosen GasSim produces a default of 25% oxidation, except for 10% of the methane that is emitted through preferential channels. It is unclear on what information this is based upon.
- Oonk (2010) reviews available literature. Important conclusion is that large part of methane is emitted through preferential channels and the percentage that is

emitted in a more homogeneous way determines methane oxidation. However there is a maximum oxidation capacity in per m^2 per year. He suggests a 10-30% oxidation for Dutch landfills in exploitation and 20-40% oxidation for closed landfills with a maximum of 5-10 kg $CH_4/m^2/yr$.

- Also in Australia the 10% IPCC-default is at discussion (Dever, 2010). Dever confirms the importance of short-cuts and indicates that actual oxidation will be significantly influenced by these short-cuts.

MODELED APPROACHES

- The most elaborated model of methane oxidation is performed in the framework of Calmin (Spokas et al., 2009; Abichou et al., 2010). Methane oxidation is determined on the basis of the composition of the top-layer and climate conditions. The model itself ultimately produces a maximum methane oxidation in $kg/m^2/yr$ and when the flux of methane to the top-layer is below this maximum, methane emission is assumed to be zero. Ultimately methane oxidation is much higher than the10-35% mentioned above and is in the order of magnitude of 75%. This high value is caused by the assumption in Calmin that all methane is emitted homogeneously. As described before, large part of methane is emitted through short-cuts and hot-spots and oxidation here is most likely low or negligible. For landfills where short-cuts and hot-spots play a role in emissions, Calmin will overestimate methane oxidation.
- The CLEAR group (an international group of leading experts on methane oxidation[14]) discusses improvement of quantification of methane oxidation. Some members of the group have proposed a draft model in which methane oxidation is either limited by the amount of methane that is homogeneously emitted or the maximum oxidation capacity of the top-layer. Both parameters are estimated as a function of methane flux, top-layer material, porosity, moisture content and ambient temperature and the lowest of both is actual methane oxidation. The draft model will be discussed, revised and defined in more detail by the whole CLEAR group during the next months (Scharff, 2010b).

2.4.3 EVALUATION OF MODELS FOR METHANE OXIDATION

Major problem in defining and evaluating models for methane oxidation is the lack of field data. Most of the measurements that are available are done using closed chambers and this method most likely overestimates methane oxidation (see chapter 3.4).

More recent evaluations of available information all yielded default values in the order of magnitude of the IPCC-default value of 10%. Most likely landfill sites in operation have less methane oxidation than closed landfill sites. Methane oxidation is generally expressed as a percentage of the methane flux from the bulk of the top-layer. But most likely, there is also a maximum methane oxidation, when expressed in $g\ m^{-2}\ hr^{-1}$. So beyond this maximum, the fixed percentage might lead to an underestimation. The principles outlined in the draft model for methane oxidation, being

[14] For more information on CLEAR, see http://ch4ox.lmem.us/

discussed by the CLEAR group (Scharff, 2010b), might result in the best guess for methane oxidation at the moment.

DIFFERENT CLIMATE ZONES

Although it is generally acknowledged that climate and season has impact on methane oxidation, this impact is not included in most of the evaluations for methane oxidation. An exception is Calmin. However Calmin assumes all methane emissions to take place homogeneously. It underestimates methane emissions from landfills, where short-cuts and hot-spots are important pathways for methane emissions. The draft CLEAR-oxidation model does correct for the impact of ambient temperature. The current version does not yet correct for the effect of precipitation.

ACCURACY

The accuracy of methane oxidation is unclear. E.g. IPCC (2006) doesn't give guidance on this topic. Interpretation of emission measurements from Oonk and Boom (1995) give values for methane oxidation in between 10 and 30% for landfills in exploitation and 10 to 60% for closed landfills. GasSim gives the opportunity to pick a realistic methane oxidation of 25% and a range of error from 10 to 40%.

Both uncertainty ranges are obtained for countries in a humid oceanic climate. Methane oxidation in subarctic or highland regions can be expected less, due to on average colder temperatures. Methane oxidation in subtropical regions can be expected less because of relative dry conditions of the topsoil. Methane in landfills in humid continental regions can be expected less as well, partially because of the longer and colder winters and more dry summers.

2.5 ACCURACY OF MODELED METHANE EMISSION

The accuracy of modeled methane emission is a function of the accuracy of modeled methane generation, the accuracy of methane recovery and the accuracy in methane oxidation. Since methane emission is obtained as a difference between generation and the sum of extraction and oxidation, the accuracy of the overall result is quite poor.

Out of all available emission models, GasSim pays most attention to accuracy of the estimated emission. In GasSim an accuracy distribution of all input variables and model parameters can be introduced. In a Monte-Carlo analysis[15], 1 to 99% confidence intervals are calculated for methane emission. It has to be noted that such a Monte-Carlo analysis only quantifies the effect of *known* inaccuracies. There are also unknown model-inaccuracies, e.g. the inherent inaccuracy of the assumption that

[15] In a Monte-Carlo analysis all parameters are varied at random, within the defined distribution of accuracy. Subsequently methane emissions are calculated. This calculation is repeated 100 times, every time with a different random choice of parameters. Result is a probability distribution of methane emissions (1% chance methane emissions are less than x kg/yr; 5% that they are less than y kg/yr to 99% they are less than z kg/yr).

methane generation can be described through a first order or multi-phase model (even in the theoretical case that the exact amount, composition and rate of biodegradation of the waste is known, there is still uncertainty about methane generation because it is not clear whether the model is an exact description of how methane is being formed). Unfortunately GasSim does not specify any default-uncertainty, so it requires an experienced and knowledgeable user to make use of this knowledge.

In all other models, accuracy has to be calculated by hand and by propagation of errors. In general the minimum and maximum methane emission can be calculated as follows:

$$CH_4\text{-}e_{min} = CH_4\text{-}g_{min} - (R_{max} * F_{min}) - OX_{max} \quad \text{(eq.-10)}$$

$$CH_4\text{-}e_{max} = CH_4\text{-}g_{max} - (R_{min} * F_{max}) - OX_{max} \quad \text{(eq.-11)}$$

IPCC (2006) gives guidance to estimating the accuracy in methane emission through error propagation and this method can be applied to other models as well. Although the IPCC-methodology is made for estimation of methane emissions from all landfills in a country, guidance is also applicable to individual landfills. According to IPCC the error in landfill gas generation per ton of waste consists of

- the error in amount of organic carbon in the waste (20% when based on IPCC default values, 10% when based on regular sampling and analysis);
- fraction of organic carbon that actually decomposes (20% when based on IPCC default values, 10% when based on experimental data for real landfills over longer time periods) ;
- an error in the methane correction factor (10% for managed landfills) and
- an error in the assumed methane content of the landfill gas formed (5%);

Total sum of errors in methane generation per ton of waste, according to IPCC, ranges between 35% and 55%, depending on local information available. However as described in IPCC (2000), some of the parameters are mutually dependent and total error might be less than the one specified. E.g. organic carbon content is known with limited accuracy and the same goes for the fraction of organic carbon that actually decomposes and the methane correction factor. The product of the three is the amount of landfill gas that is produced per ton of waste, and this one is known more accurately than the sum of uncertainty of all three factors suggest. So actual uncertainty in amount of methane produced per ton of waste will be less than the 35 to 55% and might be 20 to 40%.

TABLE 4: INDICATION OF UNCERTAINTIES IN METHANE MASS BALANCE

	good case	bad case
amount of waste	1 à 2%: weighed waste	20%: estimated based on landfill volume
L_0	20%: accurate waste description, humid oceanic climate zone	40%: no accurate waste description, other climate zone
model uncertainty	5 à 10%	10%
methane recovery	10%: measured	25%: estimated
methane content	5%: measured	10%: estimated
methane oxidation	150%: other climate zone	250%: humid oceanic climate zone

Table 5 gives an example of the result of a simple error propagation of methane emissions from a landfill. Even with modest assumptions on accuracy of methane generation and other factors involved, the inaccuracy in methane emission in this example turns out to be 65%. As a rule of thumb, inaccuracy in methane emission increases when the efficiency of landfill gas recovery increases.

TABLE 5: EXAMPLE OF PROPAGATION OF ERRORS WHEN CALCULATING METHANE EMISSIONS FROM THE METHANE MASS BALANCE (LANDFILL IS CHOSEN IN SUCH A WAY THAT BEST GUESS METHANE EMISSION IS 100 KG/Y)

	minimum	mean	maximum
LFG generation (m^3/y)	300 (-30%)	428	557 (+30%)
methane content (vol%)	50	54	58
methane generation (kg/y)	108	167	232
methane recovery (kg/y)	61 (+10%)	56	50 (-10%)
methane oxidation (kg/y)	12 (25%)	11 (10%)	18 (10%)
methane emission (kg/y)	35	100	164

However on an individual landfill, knowledge of the local situation, e.g. on the quality of landfill gas extraction might improve the accuracy considerably. In this example, the minimum value would imply over 50% recovery efficiency, where the maximum emission would imply just over 20% efficiency of landfill gas recovery. An expert judgment of the quality of the recovery system, and efforts done in the past to optimize recovery should help to see what range in recovery value is realistic (see also chapter 4). Based on this, the error in modeled emission could be reduced.

2.6 CONCLUSIONS MODELLING

Modeling emissions of methane generally requires modeling of methane generation, measuring landfill gas recovery and assuming some methane oxidation.

In the last few years development of methane or landfill gas generation models have received most attention and seem to have developed. There are several models

available, such as the IPCC-model, the TNO-model and GasSim, all built-up from reasonable assumptions. However due to a lack of validation on real landfill data, these assumptions might result in only an apparent accuracy. The French E-PRTR-model is much simpler and might be just as effective.
All the aforementioned models might produce reasonable results for MSW dominated by household waste, landfilled in Western Europe. The accuracy of these models for other types of waste or in different regions in Europe is limited:

- The impact of climate on landfill gas formation is widely recognized. Climate will have impact on both the amount of methane that is ultimately released per ton of waste (L_0) and the speed at which methane is released (half-life values). The impact of climate on L_0 is until now neglected in formation models. The impact of climate on half-life is only described in the IPCC-model, but in a very rudimentary way. For an improved description, 4 climate zones in Europe could be distinguished: (i) subarctic and highland, (ii) humid oceanic, (iii) humid continental and (iv) subtropical;
- As a result of existing policy, landfilling of organic waste is more and more discouraged. This change in waste composition also requires improved default values for L_0. It is also possible that the speed and completeness will be affected at which organic waste that remains to be landfilled degrades;

Oxidation is more difficult to describe, than methane generation. Knowledge on oxidation is also limited by scarce information available on actual methane oxidation under field conditions. The IPCC-default value of 10% has to be considered as a low-guess, a conservative value, leaving room for improvement. Actual methane oxidation is again dependent on the design of the top-layer, the methane flux through the top-layer and climate conditions (precipitation and ambient temperature). Hot-spots and short-cuts for methane emission limit methane oxidation, since at many landfills large part of methane will escape without passing the oxidizing zone in the top-layer. Most likely is methane oxidation (expressed in %) somewhat higher at closed landfills, somewhat less at landfills in exploitation and becomes more or less a constant value in $kg/m^2/yr$ when methane flux to the top-layer is high (e.g. deep landfills, without state of the art landfill gas recovery).

In the end, modeled methane emissions are highly uncertain, even when methane formation and oxidation can be described relatively accurately. The reason for this is the propagation of errors, which is highly unfavorable. This is because methane emission is calculated as the difference of three uncertain parameters.

An ideal methane formation or emission model doesn't exist. Such an ideal model should have the transparency of IPCC-model, the level of validation of the TNO-model, a waste input module for non-household waste of Afvalzorg, an uncertainty analysis as in GasSim, a more reliable description of oxidation as a function of climate conditions as in Calmin, but then with more realistic assumptions on short-cuts as in the draft oxidation model of the CLEAR working group. However, as long as no additional validation efforts are performed, one should be aware that models with more sophistication, built on even more assumptions might only give an improved

apparent accuracy. So simplicity as in the French E-PRTR-model might also have its benefits.

CHAPTER 3: MEASURING EMISSIONS

3.1 Introduction

In the past decades there has been considerable interest in measurement methods for methane from landfills. In this period, various methods are proposed, developed, tested, improved. However at the moment there is no single method, that is widely recognized as the preferred method to measure annual average methane emissions.

The main difficulty in measuring methane emissions from landfills is the spatial and temporal variability of emissions, in combination with the sheer size of a modern landfill. The spatial variability of methane emissions is reported by various researchers. Emissions at one spot can be 1.000-fold of emission from a spot located a few meters away (Verschut et al., 1991). According to Czepiel et al. (1996) there is no correlation between emission at a spot at the landfill and the emission 6 meters away. They estimate that 50% of emissions is released at 5% of the landfill surface; Bergamaschi et al. (1998) estimate that 70% of methane emissions are released through short-cuts. Figure 7 gives a typical distribution of distribution of emissions at a landfill and similar patterns are published by Nozhevnikova et al. (1993), but similar distributions are reported by Oonk et al. (2004), Mackie and Cooper (2009) and Chanton et al (2010). Rachor and Gebert (2009) studied variation in emissions within the square meter and even at this small scale emissions proved to be highly heterogeneous.

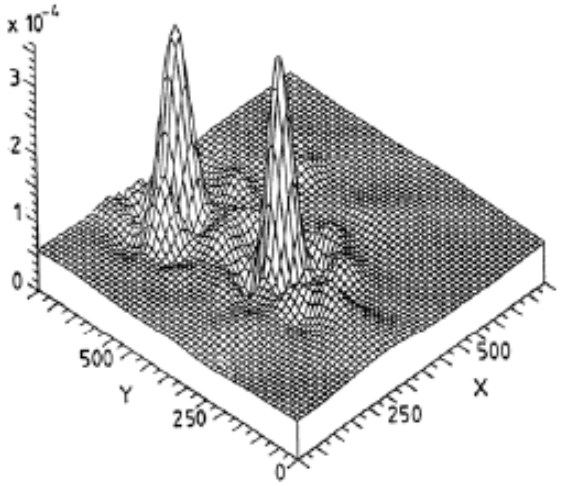

FIGURE 7: METHANE EMISSIONS FROM THE KUCHINO LANDFILL SURFACE (NOZHEVNIKOVA ET AL., 1993)

Changes in weather cause a temporal variability. Verschut et al. (1992) indicate pressure variations to be very important. Czepiel et al (1996) indicate that higher emissions are obtained during days with lower pressure. Also Scharff et al. (2003) report a correlation of methane emissions and changes in ambient pressure. Rainfall, wind and events in the gas-extraction system are other aspects that have impact on methane emissions.

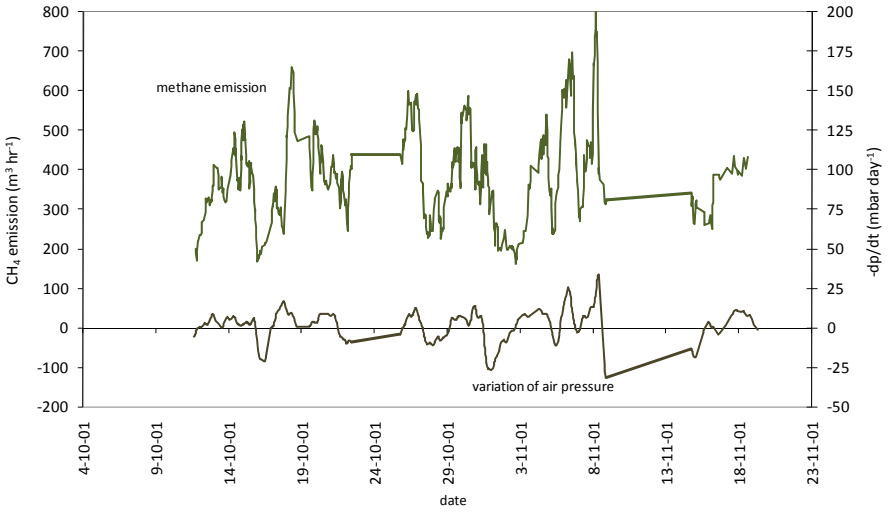

FIGURE 8: VARIABILITY IN EMISSIONS AND CHANGES IN PRESSURE DROP. RECONSTRUCTED FROM A 1D MASS-BALANCE METHOD MEASUREMENT (SCHARFF ET AL., 2003)

Based on day-to-day variabilities as in Figure 8, Scharff et al. (2003) estimate 4-6 day-measurements throughout the year are required to obtain an accurate annual average emission estimate.

On top of the day to day variation mentioned above, a seasonal variation in methane emissions is expected, due to a seasonal variation in temperature and moisture content. As described in chapter 2.4.1, average methane emissions in winter is somewhat higher than average emissions in summer, especially in Nordic countries.

So a method to measure annual average methane emissions should be able to deal with the temporal and spatial fluctuations as described above.

3.2 Available methods

3.2.1 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 carbon dioxide 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 at different temperatures or soil moisture levels etc. to study improve the mechanistic understanding of oxidation.

ADVANTAGES AND DISADVANTAGES

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

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 labor-intensive.

3.2.2 Closed chamber measurements

Application of closed chamber is most frequent applied to measure methane emissions from landfills. It is applied by many research groups around the world, both for monitoring methane emissions from smaller parts of a landfill (e.g. test-fields for enhanced methane oxidation) as well as estimating emissions from an entire landfill. An overview of selected applications of closed chambers is given by Scheutz et al. (2009).

In general terms, in a closed chamber measurement a flux box is put on the landfill, and the increase of methane concentrations in the box in time is measured. The methane flux is calculated from the increase of methane concentration in time, the volume of the box and the surface captured by the box.

The are a few pitfalls, when performing a closed chamber measurement:

- When landfill gas is collected within the box, the pressure in the box increases. So when the measurement is performed over a too long time, landfill gas emission from the surface encapsulated by the box might be affected and methane flux is underestimated;
- With vegetation on the landfill surface, sealing of the box to the surface is of importance. Any leakages will disturb the measurement. In case of excessive vegetation, mowing prior to the measurement might be an option. To improve sealing some research projects where the same spot is measured multiple times over time, use fixed collars which are mounted to the ground on which closed chambers can be positioned;
- When vegetation is present, the method is not suited for measuring carbon dioxide. This is because of dissimilation of carbon dioxide from the vegetation.

Some variations on closed chamber measurement are:

- Dynamic boxes, open channels through which a continuous air-stream is led. Using a matching pair of inlet- and outlet-ventilator the pressure in the box is kept ambient and landfill gas emission is not influenced (Verschut et al, 1991; Huber-Humer and Lechner, 2001ab);
- Fast box measurements, using analytical equipment that already can detect a few ppb increase in methane concentration. Using this box, a single measurement takes less than a minute. As a consequence the number of measurement that can be performed in one single day is significantly increased (Oonk et al., 2004).

Grid-wise measurements

The largest draw-back of closed chamber measurements is the small surface area sampled per measurement. In an attempt to obtain a reliable methane emission estimate, systematic sampling strategies are proposed (Bogner and Scott, 1995; Bour, 2007; Long, 2004; Rosevaer et al. (2004); Savanne et al., 1997; Spokas et al., 2006). Such a sampling strategy consists of sampling at points located on a systematic grid ,

sometimes followed by application of geostatistical models (Spokas et al., 2003). Typical distances between points on such a sampling grid are 10-60 meters; the more measurements made, the more accurate the result.

Another option to improve closed chamber measurements is to use qualitative surveys (see chapter 3.2.7) to identify hotspots and subsequently decrease the grid distance at places where hot-spots of emissions are suspected. This does however introduce the issue of weighing the hot-spots measurements and the other measurements correctly to obtain an overall landfill average.

ADVANTAGES AND DISADVANTAGES

Closed chamber measurements have a number clear advantages. To start with, the method is easy to understand, doesn't require analytical equipment beyond a common FID or a IR-analyzer. The method is able to detect small fluxes of methane and is not sensitive to topographic constraints or other sources of methane near the landfill.

The method itself also has clear disadvantages. The most important one is that on many landfills methane emissions take place in such a heterogeneous way, that closed chambers do not give a reliable average methane emission. There is a big chance that hot-spots of methane emissions are missed, resulting in an underestimation of emissions. (Perera, 2000, Perera et al., 2002, Pumpanen et al., 2004, Senevirathna et al., 2006, Babilotte et al., 2009; Gebert et al., 2009). The method does not measure emissions from leaks in the gas and leachate system (header pipes, extraction wells and leachate wells). The use of geostatistical models to interpret measurements is reported to be a large and non-quantifiable source of errors (Babilotte et al., 2008). Babilotte et al. (2008) mention the time needed to measure emissions at an entire site as a further disadvantage. This time is about a week, during which emissions will vary due to variations in weather conditions. This however might also turn out to be an advantage, since it might smoothen the impact of weather in measured average emissions a bit. The labor-intensity of the method and associated costs are mentioned to be a last disadvantage.

3.2.3 MICROMETEOROLOGICAL METHODS

Micrometerological measurements (also known as Eddy-correlation measurements) are a standard method to measure emissions from larger surfaces (e.g. fluxes of methane and nitrous oxide from agricultural soils, lagoons, etc. (Denmead, 2008; Laurila et al., 2005, Lohilla et al., 2007). In a micrometeorological measurement, the methane flux through an imaginary horizontal plane, about 0,5-3 meters above the landfill surface is measured. Flux of methane through this place takes largely place though convection and therefore the flux can be measured as the average of the product of methane concentration (C_{CH4}) and vertical component of the local wind-velocity ($v_{w,y}$).

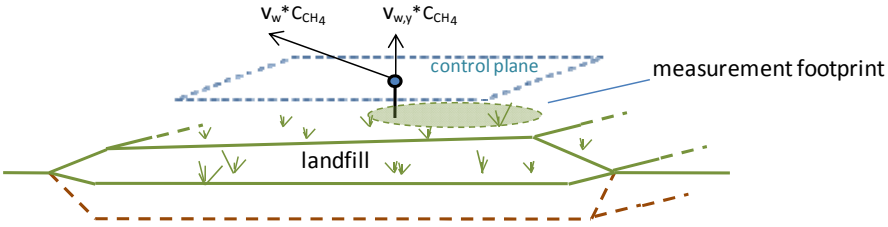

FIGURE 9: A MICROMETEOROLOGICAL MEASUREMENT

In a micrometeorological measurement both parameters are measured at a high rate (~10 times per second). Data are stored and every 15 minutes methane emissions are calculated. Due to air turbulences wind will be sometimes have an upward component and sometimes a downward. In case of an emission, the methane concentration at the plane will be slightly higher when wind is upward and an positive average methane flux is obtained.

ADVANTAGES AND DISADVANTAGES

An advantage is easy automation, which enables measurements over longer periods of time and the possibility of simultaneous monitoring of CH_4 and CO_2 emissions (IPCC, 2006). The method is able to run for weeks to several months, giving a good indication of both temporal variability and average emissions. Further advantages are the compact size of the required equipment its ease of operation (Babilotte et al., 2009).

A drawback of the method seems to be the limited footprint of the method, as a result of which it might not produce representative emissions from the entire SWDS. The footprint can be quantified from experimental data, changes during the measurement with changing wind direction and is most likely a circle around the measuring device with a radius of 50-100 meters (1 to 4 ha). Another disadvantage is the sensitivity of the method for the landfill topography. In general, the methods becomes more accurate[16] when the size of the landfill increases and more flat. According to Scheutz et al. (2009), the technique is expensive and requires specialized equipment. This might be true in comparison with closed chamber measurements, but the method seems to be relative simple in comparison with 2D mass-balance methods or plume methods. Moreover the required equipment is available at dozens of research groups around the world.

3.2.4 MASS-BALANCE METHODS/TRANSECT MEASUREMENTS

A) 2D MASS BALANCE METHOD

In a mass-balance measurement methane fluxes through two imaginary vertical planes before and after the landfill are measured. The difference between both is the methane emission from the landfill. Methane flux through a plane is measured as the product of wind velocity and methane concentration at each point in the

[16] Accurate refers here to the accuracy with which methane emissions are measured from the area, recognized as ‘ footprint’. It does not refer to how representative the footprint is for the entire landfill.

plane. When the methane concentration profile in the background is more or less constant, the method can be simplified and the emission from the landfill can be calculated from the product of wind velocity and difference between methane concentrations in the control plane and the methane background concentration.

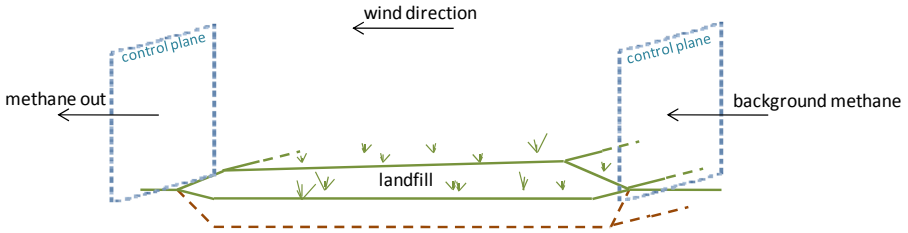

FIGURE 10: A 2D MASS BALANCE METHOD

Main problem is that both wind velocity and methane concentrations are not constant within the control planes. So the challenge of the mass-balance method is how to measure both at different positions in the control plane. Babilotte et al. (2008, 2009) tests two approaches. One approach (VRPM) consists of optical remote sensors in combination with mirrors, measuring average methane concentrations over a few lines (see Figure 11). From this the methane concentration distribution in the vertical plane is measured.

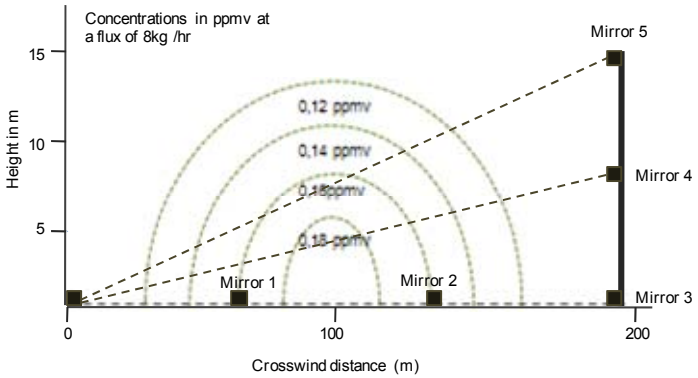

Figure 11: Measuring methane concentration distribution in a plane, using VRPM.

The second method (DIAL) uses the backscatter of pulsed tunable laser radiation to obtain a reconstruction of methane concentration profiles in the vertical control plane. In both cases, wind velocities are measured using one or two anemometers to measure wind velocity and consequently provide limited information on distribution of wind velocity at different heights.

ADVANTAGES AND DISADVANTAGES

An advantage of a mass-balance method is that it is based on a simple and robust principle. Apart from wind direction and velocity it doesn't require any meteorology or modeling of atmospheric dispersion to quantify methane emissions. A second advantage is that the footprint of a measurement is large. An advantage of DIAL over VRPM in this context is that the size of the vertical plane sampled is much larger. With VRPM the maximum width of the plane is about 200 meters, where DIAL can

measure methane at 400-800 m range. So depending on the tool used, part of the landfill cq. emissions from the entire landfill are measured. Also with increasing width of the control plane, the method becomes more robust with respect to landfill geometry.

Disadvantages of the methods are the size of the equipment, which makes it less flexible. Disadvantage is also the highly specialized equipment required, which means that the method is expensive (Equipment for DIAL costs over 1 million Euro) and only limited available. VRPM is a technique with limited width of the control plane, which yields only methane emissions from part of the landfill. So when (as was the case in the tests with VRPM, described by Babilotte et al., 2008, 2009) the control plane is located on top of the landfill (before the slope) emissions from the slopes are not measured. Since slopes are recognized as preferred emitters of landfill gas, methane emissions may be underestimated. Another disadvantage in general is that both science and equipment used is difficult to understand for third parties, which makes it difficult for an independent third party to judge the accuracy of technology.

B) Simplified 1D mass balance method

A simplified mass-balance method, uses a 1D control plane (a vertical line) on top of the landfill. This 1D control plane consists of a pole in which at different heights methane sampling points and anemometers are attached (Oonk and Boom, 1995; Scharff et al., 2003). The methane flux (corrected for background concentrations of methane) flowing through this 1D control plane can be related to methane emissions released at the line from the pole, wind upwards to the edge of the landfill. A measurement typically lasts 3-6 weeks, during which emissions from all wind-directions are measured, thus mapping the whole landfill.

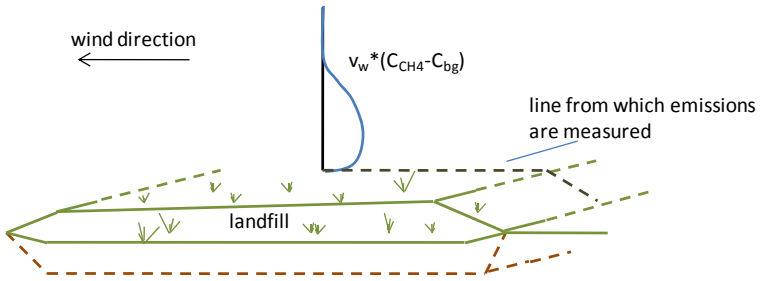

Figure 12: a 1D mass balance method

The result of the method depends on assumptions where methane is emitted. E.g. when actual methane emissions take place closer to the pole, total emissions might be overestimated. Therefore a qualitative survey of methane emissions is recommended to evaluate the location of emissions, prior to interpretation of the raw data.

Advantages and disadvantages

Advantages are its relative simplicity, relative simple measuring equipment required and relative low costs. The method can be automated and emissions from a landfill can be monitored for longer times, thus yielding at one hand information about va-

riability of methane emissions in time and at the other hand an average emission. Advantage of the method is, that it measures emissions from an entire landfill, on the condition that the landfill is not too large. The maximum distance from the pole to the side of the landfill is estimated to be 10 times the length of the pole and is 100-200 m. Additional advantage of this method is that is can simultaneously measure CO_2-emissions. Combination of CO_2, CH_4-emissions and information on landfill gas recovery and its composition gives also an indication of total landfill gas generation and methane oxidation (Scharff et al., 2004; see also chapter 3.2).

The disadvantage of this 1D mass balance method is that it is produces a less accurate methane emission than a mass-balance measurement with a 2D control plane. As described above, the actual location of methane release plays a role in this uncertainty. Disadvantage is that the method is less applicable at larger landfills (> 10 ha).

3.2.5 Tracer plume measurements

In a tracer plume measurement, a known amount of tracer (e.g. N_2O) is released on top of the landfill (Galle et al, 2001; Babilotte et al., 2008). Further away from the landfill, the ratio of tracer and methane is measured. When the plumes of the landfill and of the tracer are fully mixed, methane emissions from the landfill can be calculated as:

$$CH_4\text{-e} = \text{flux tracer} * (C_{CH4}/C_{trac})$$

A) Mobile tracer plume measurements

In a mobile tracer plume measurement, the analytical equipment is mounted on a car and driven through the plume. In this way a profile is obtained of methane and tracer concentrations across the plume. Analysis of this profile enables a check, whether plumes really have mixed sufficiently and whether methane emission is accurate. When the mix is considered insufficient, dispersion modeling of both the methane plume and the tracer plume can be used to correct for insufficient mixing. When doing this, the difference between a tracer plume measurement and a normal plume measurement (see 3.2.6) gets smaller.

Advantages and disadvantages

Biggest advantage of a tracer plume measurement is the simplicity of its approach. When a situation can be created where plumes of the landfill and of the tracer fully mix, analysis and interpretation is straightforward and without major model assumptions.

A disadvantage is the dependency of the method on favorable weather conditions, which means that the measurement-team has to remain stand-by for longer times. A limitation to the method is the availability of roads, enabling the measurement of a full plume. A second disadvantage is the relative distance between the landfill and the location of the measurement, as a result of which methane plume from the landfill is diluted somewhat more. As a result, methane due to landfill-emissions are more difficult to distinguish from background-concentrations and this gets worse, when other sources of methane are present in the neighborhood of the landfill.

B) Stationary tracer plume measurements

An alternative to the mobile tracer plume measurement is the stationary version (Babilotte et al., 2008; Scharff and Hensen, 2009). For this purpose In such several fixed sampling-bag filling points (typically 16) are located at various points around the landfill. These sampling points are used to sample concentrations in the plume. Interpretation of the method is comparable to the mobile tracer plume measurement.

Advantages and disadvantages

Advantages and disadvantages are comparable to the advantages and disadvantages of mobile tracer plume measurements. The stationary version has the advantage of being less dependent on available roads. Equipment can be carried in a back-pack and stationed off-road as well. Disadvantage is that only points in the plume are sampled, rather than a transect of the whole plume. So the method loses some of check on whether plumes of landfill methane and plumes of tracer have fully mixed.

3.2.6 Plume measurements

A) Mobile plume measurements

In a plume measurement, methane concentrations at various locations in the plume are measured, along with meteorological data. Subsequently inverse modeling[17] is used to determine the methane emission profile that fits the measured values best. A standard way to map the methane plume is to drive a car with analytical equipment through the plume at a distance of 500 to 1,500 m from the landfill. At that distance atmospheric dispersion is assumed to suffice to remove vertical concentration differences. In this way, a cross profile of the plume is obtained of methane at about 2 meters height. Analytical equipment used depends on the expected methane emissions and the distance to the landfill, where methane emissions are performed. Typically it is necessary to distinguish with sufficient reliability between 1,700 and 1,710 ppb. Measurement equipment has to be suited to detect methane at those concentrations and both TDL (Tunable Diode Laser), QCL (Quantum Cascade Laser) and OFCEAS (Optical Feedback Cavity Enhanced Absorption Spectroscopy, Babilotte et al., 2008) are proposed.

Advantages and disadantages

Advantages of the mobile plume measurement is that is provides methane emissions from an entire site. The method is applicable for landfills of all sizes, large and small landfills. The measurement also gives a best estimate of distribution of emissions

[17] In inverse modeling, an emission model of the landfill is made, methane dispersion is calculated and subsequently at measurement locations measured methane concentrations and calculated methane concentrations are compared. Depending on the outcome, the emission model is adapted (methane emission is increased reduced, location of emissions is changed), and methane dispersion is calculated and compared once more. This iterative process is repeated until the best 'fit' of emission model and measured data is obtained.

over the landfill, e.g. on a 10*10 m grid-scale. The measurement includes emissions from parts of the gas or leachate collection system.

Disadvantages are that the method requires a detailed emission model of the landfill. The measurement itself requires analytical equipment that goes beyond the standard FID or IR-analyzer. Interpretation of measurement data is based on dispersion of this emission model and is less straightforward as e.g. interpretation of results from mass-balance method. Besides the method has some prerequisites: the landfill has to be located in relatively flat terrain and well accessible roads have to be present at least at one side of the landfill at a distance between 500 and 1,500 m of the landfill. Weather and other conditions (availability of personnel, the right wind direction, wind-speed between 3 and 10 m/s, sufficient Pasquill stability, no significant changes in ambient pressure, no major disturbance at the landfill) have to be right for a measurement to be performed. Waiting for proper conditions can imply significant stand-by periods.

Accuracy: the TDL emission data are about 25% accurate depending on the location. and the meteorological conditions.

B) Stationary plume measurements

A stationary plume measurement (SPM) is intended and developed as a simplified, cheaper alternative to a mobile plume measurement (Scharff et al., 2004). In such a SPM, several fixed sampling-bag filling points (4 to 8) are located at various points around the landfill. When weather conditions are advantageous, a battery operated unit fills one sample-bag in a time interval of 30 minutes. Afterwards methane concentrations in the bags are analyzed in the lab and results are interpreted using reverse modeling.

Advantages and disadantages

Advantages compared to the mobile plume measurement are costs and size of the equipment. Also the automated sampling procedure implies that no personnel and equipment has to be stand-by for longer times, waiting for favorable conditions.

Disadvantages are the amount of data available for modeling, which is substantially less than in a mobile plume measurement. As a result, the outcome also seems to be less reliable. Also safe positioning of fixed sampling-bag filling points outside the landfill area is sometimes problematic. Filling points can not be stationed close to other sources of methane (e.g. cow stables).

3.2.7 Qualitative emission measurements

In a qualitative emission measurement, methane concentrations above the landfill surface is mapped. Such a map of methane concentration can not be correlated to methane emissions and also the absence of methane concentrations less than about 0,1% about 1 meter above the landfill surface is not an indication of absence of methane emissions. A map of methane concentration does give an indication where hot-spots of emissions exist and in some cases repair of such a hot-spot is possible, e.g. by repairing cracks in the top-layer or leaks in the gas-collection system. In USA

landfills are obliged to perform surface scans 4 times a year. A field survey can be done by walking a predefined grid with a portable FID or another field gas analyzer.

Other ways to identify hot-spots for methane emissions are application of thermal infrared surveys and simple visual field inspection. Thermal infrared surveys are preferably performed in the early morning is autumn, winter or spring and might detect spots in the landfill that are warmer than normal. Such a spot can be caused by landfill gas, coming out of the warmer deeper parts of the landfill. However warm zones might also caused by other phenomena, e.g. waste at the surface that is degrades aerobically. However simple field inspection, looking for cracks in the surface, damages to the gas recovery system and being alert to typical odor of escaping landfill gas is a very simple alternative to identify hot-spot of emitting landfill gas.

3.3 Evaluation of methods

3.3.1 General

As described above, there have been considerable efforts to develop methods to measure methane from landfills. Based on these developments, there have been a number of studies to mutually validate the methods:

- Verschut et al. (1991) compared dynamic closed chambers with a 1D mass-balance method at 3 Dutch landfills. Conclusion was that the spatial variation in emissions was too high to capture with closed chambers. Therefore 1D mass-balance method was used in a measurement campaign on 15 more Dutch landfills (Oonk and Boom, 1995);
- Trégourès et al. (1999) compared two closed chambers methods, a micrometeorological technique, a 1D mass balance method, two tracer gas methods and an airborne infrared thermography at a French landfill;
- Scharff et al. (2003) aimed at further development of relative cheap methods (1D mass balance method; static plume measurement) to measure landfill methane and to validate these low-costs method with a more accepted mobile plume measurement. Most important conclusion was that all three methods are in fair agreement. The 1D mass-balance method is the recommended low cost method at landfills below 10-15 ha; SPM is the recommended low-cost method at larger landfills.
- Jacobs et al (2007) compared static and mobile plume tracer measurements at a Dutch and a Danish landfill. Results of the low-cost static plume measurement were in good agreement with the dynamic method, with further verification of the method still being necessary;
- Babilotte et al. (2008) describe tests with two 2D-mass-balance, a micrometeorological test and a mobile and static plume tracer test at a landfill site in France. Conclusion is that there is no perfect method, that gives an accurate methane emission within limited time and budget. Most accurate emission measurements are expected from 2D-mass balance methods;
- Babilotte et al. (2009) compared two different 2D-mass-balance methods, a micrometeorological method, closed chamber measurements and a plume tracer

method at two landfill sites in USA. Conclusion is that whole landfill methods (whole landfill 2D-mass balance methods, plume tracer measurements) are preferred above methods that only measure parts of the landfill (closed chambers, a more localized 2D-mass balance method and micrometeorological methods).

In all studies described above, measurements were performed at landfills, where the actual methane emission is not known. So it is difficult to judge what method is more accurate. However Babilotte et al. (2008) did one additional test in the field, where a known amount of methane were released from a flask through a 10*10 meter grid at a landfill in preparation (see Figure 13).

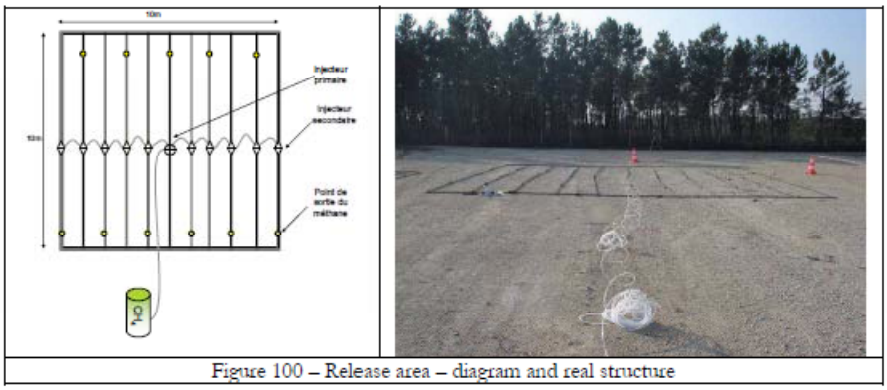

Figure 100 – Release area – diagram and real structure

FIGURE 13: CONTROL METHANE RELEASE AT INTERCOMPARISON OF MEASUREMENT METHODS (BABILOTTE ET AL., 2008)

This test is not entirely comparable with methane emissions at a landfill, since both scale (100 m2 rather than a few ha) and amount of methane released (about 2 kg/hr rather than several hundreds kg per hr) are not epresentative of what happens at an actual landfill. Moreover due to time pressure, the test was performed under unfavorable weather conditions. There was hardly wind, as a result of which the dispersion of the plume was minimal. Most measurement technologies were applied at conditions they normally would be considered not-acceptable. As a consequence, the overall results of the test, shown in Figure 14, can not be considered representative for the accuracy of various methods. However the strength of such a controlled release test in the evaluation of measurement technologies seems evident.

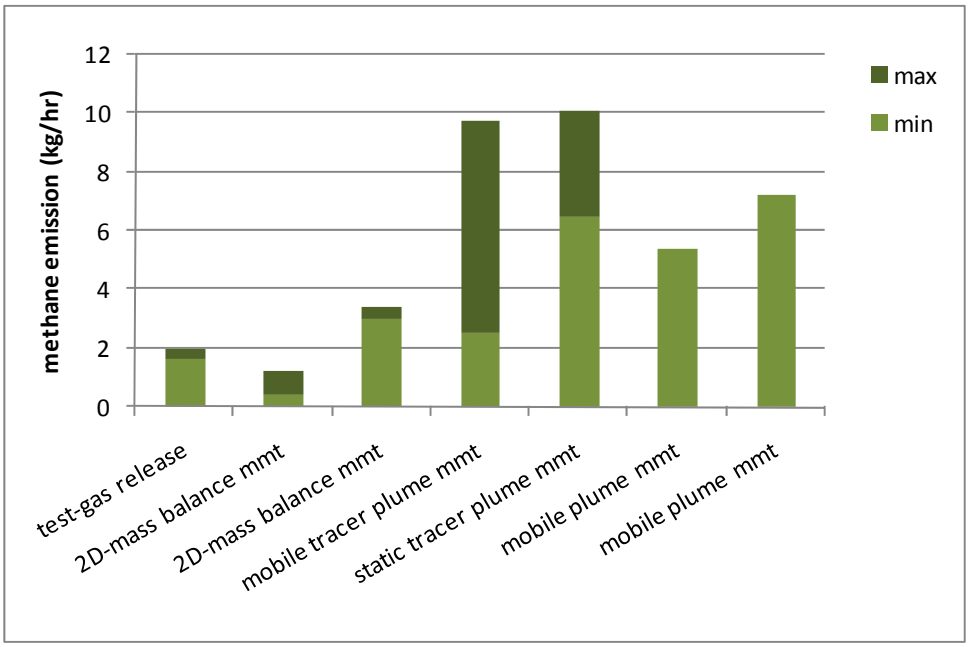

FIGURE 14: RESULTS OF MEASUREMENT METHOD INTERCOMPARISON AT CONTROLLED METHANE RELEASE. N.B. THE SAME MOBILE PLUME MEASUREMENT WAS PERFORMED TWICE IN THIS INTERCOMPARISON.

Based on the experiences in the studies listed above an evaluation is made of measurement methods. A summary of this evaluation is made in Table 6. Since accuracy is hard to quantify, the methods are evaluated on a number of parameters that all contribute to accuracy. Other parameters of importance are the effort required, the costs and also the constraints to the method. Further clarification of the evaluation is given further below. For the 2D-mass balance method an integral measurement is distinguished from a partial measurement. The evaluation of the integral measurement is based on DIAL, which enables a direct measurement of en emission of a whole landfill site. The partial measurement is based on VRPM, which in most cases only measures emissions from part of the landfill.

TABLE 6: EVALUATION OF MEASUREMENT METHODS

	soil-core method	closed chamber	micrometeorological method	2D mass balance method (integral)	2D mass balance method (partial)	1D mass balance method	plume tracer measurements	static plume tracer measurements	plume measurements	static plume measurements
performance										
-applicability										
temporal resolution	0	0	+	-	-	+	-	-	-	0
spatial resolution	--	--/-	0	++	0	0/+	++	++	++	++
-accuracy	0	++	+	-/0	-/0	+	+	+	0	0
-assumptions	-	+	-	++	-/0	0	+	0	0	-
equipment										
-equipment	+	++	+	--	-	+	0	+	0	+
-personnel	0	++	0	-	-	+	+	+	0	0
-maturity	0	0	0	0	-	+	0	+	0	0
constraints										
-large landfills	--	--	+	++	0	-	++	+	++	+
-small landfills	-	-	-	++	++	+	++	0	++	0
-complex shape	++	++	-	+	+	-	++	+	++	+
-wind	++	++	+	-	-	+	0	+	0	+
-accessibility	++	++	0	-	-	0	-	-	-	-
costs	-	-	+	--	-	0/+	0	+	0	+

[1)] In this table '++' means very good and '--' means very poor. If a generation model scores '-' or less on one of the evaluation parameters, users of the model should be well aware of the limits of the model.

3.3.2 ACCURACY

Applicability.

The ultimate objective is to measure the annual average methane emission for the whole landfill. How representative is the measurement of the sampled area on a specific day in this context):

- Spatial resolution. Soil core measurements and closed chamber measurements are performed at a very small scale. Even with a very tight grid (a measurement every 10*10 meters), less than 1% of the total surface area is sampled, and as indicated in chapter 2.2.3, this is considered not enough to get an accurate average methane emission; the micrometeorologial method and some 2D-mass balance methods (e.g. VRPM) sample part of the landfill and the question remains

to what extent this part is representative for the entire landfill[18]. The 1D mass-balance method enables measurement from the entire landfill, on the condition that the landfill is less than 10-15 ha. Otherwise only part of the landfill is measured. The other measurements directly yields a methane emissions from the entire site;

- Temporal resolution. Many measurements are performed in a few hours in single day. So they most likely do not catch all the hour-to-hour or day-to-day variation that occur in landfills and have therefore a temporal resolution that is too small to give an average emission for the season. Methods to estimate emissions from soil-core profiles, closed chambers, static plume measurements and static tracer plume measurements generally do take more than a day. Due to day to day variations they might underestimate emissions at one point, but overestimate emissions at another point. Some of the errors might level each other out. In any case the errors don't point the same way for all measurements, so the temporal resolution here will be somewhat better. 1D mass-balance methods and micrometeorological methods might be continued for weeks or months (in theory for the full year). A measurement of this duration has sufficient temporal resolution for the season it is applied. Due to seasonal variations, a measurement of one month is still not representative for the whole year

The **measurement accuracy** refers to whether the method accurately captures information required to calculate methane emissions. Closed chamber measurements only need to measure concentration increase in the box in time and this can be done very accurately. 2D measurements seem to give more problems and is valued '-/0. The '-' refers to the measurement of wind velocity seems. Wind velocity is not constant over the control plane (it might vary with height) and this variation get insufficient attention. The '0' in this refers to the accurate determination of the methane concentration distribution remains a challenge. With DIAL little experience exists worldwide and e.g. a cross-interference with moisture in the air might contribute to inaccuracy. Measurements in the 1D mass-balance method and micrometeorological methods seem to be straightforward, just as the various methods of plume measurements. Plume measurements have the drawback of measuring at larger distances, where methane concentrations are diluted and background-concentrations of methane can be problematic. This is described under ' constraints'.

Assumptions

The measurement principle of closed chamber based measurements, integral 2D mass balance methods and plume tracer measurements is very simple and has little or no assumptions. For a partial 2D-mass balance method, footprint becomes very

[18] An important difference between a 2D-MBM and a micrometeorological method is that the first one gives a methane emission in g/hr. An estimate of the footprint (in m^2) is required to calculate methane flux (in $g/m^2/hr$) and the result is highly dependent on assumed footprint area. A micrometeorological method gives a methane emission in $g/m^2/hr$. The footprint is only of importance to assess the area for which the measured flux is representative.

important and assumptions here have direct impact on measured methane flux. In case of VRPM, estimating the methane concentration distribution on basis of average methane concentrations along a few lines (see Figure 11) brings about uncertainties. When performing 1D mass balance methods, assumptions have to be made about the location of emissions on the landfill. The impact of these assumptions is limited, but the measurement itself will benefit from a more accurate identification of hot-spots of methane emissions. Plume measurements are based on emission models of the landfill and modeled dispersion of emission, and therefore depends on assumptions. Assumptions in micrometeorological methods and soil-core methods can have significant implications and might result in methane emissions that are significantly wrong.

3.3.3 Equipment

Availability

Soil cores, closed chamber method and 1D mass balance methods require analyzers that are readily available (FID or IR for measuring methane concentrations) and simple additional equipment. The micrometeorological method requires in addition meteorological equipment, that is quite specific. Plume measurements usually apply a TDL to measuring methane concentration. TDL's become more and more available, but are not as common as FID's or IR. 2D-mass balance methods however apply very specialized equipment for determining the distribution of methane concentrations in the landfill plume.

Personnel seems to be no limitation in most methods. Measurement and interpretation of results requires teams of people with laboratory or university degree. Required procedures are in general not that complicated and it doesn't take too much experience to perform a measurement. An exception might be 2D mass balance measurements, that might require specialists in relation to the specialized equipment used.

The **Maturity** is given a 0 in Table 6, when the technology is completely developed, and further improvements are not likely to take place in the next few years. Negative values are awarded when a technology needs further developments, before it can be used properly. Positive values are awarded when further improvements are still possible, resulting e.g. in cost reduction.

3.3.4 Constraints

Most measurement methods have their specific constraints. Problems with spatial heterogeneity of soil-core measurements and closed chamber measurements increase with increasing landfill size. 1D mass balance methods are best performed at smaller landfills (up to 10-15 ha), but becomes more problematic at larger landfills. Static plume measurements and maybe also micrometeorological methods become more applicable when the size of the landfill increases. For the rest all methods have specific requirements for suitable weather conditions, topography (including other sources of methane in the neighborhood) and accessibility. For this, see Table 6 and the description of measurement methods in chapter 3.2.

3.3.5 Costs

Costs are based on specification of costs by Babilotte et al (2008) and Scharff et al. (2004). In Table 6 a '++' refers to costs of about € 5000 per measurement; a '+' to about € 10.000; a '0' to € 20.000 per meting; a '-' to € 40.000 per measurement and a '--' to costs in excess of 50.000 per measurement. The costs refer to the costs per measurement. As described in chapter 3.1, emissions have a temporal variability: at one hand of a day-to-day nature; at the other hand a seasonal variation. To obtain an annual average, about 5-10 one-day measurements are required or about 3 measurements of greater temporal resolution.

3.4 Measuring methane oxidation

3.4.1 Introduction

As described in chapter 2.4, methane oxidation occurs, when methane passes through the top-layer. Methane oxidation is a biological process and depends a.o. on temperature and moisture. As a result methane oxidation is in winter less than in summer.

A number of methods exist to measure methane oxidation in the field. The methods can be characterized both by the way methane is sampled and by the way the sample is analyzed and results are interpreted.

3.4.2 Sampling methods

Soil core measurements and box measurements

As described in chapter 3.2.2 closed chambers are not well suited to determine methane emissions, due to the inhomogeneity at which methane emissions occur. The same is true for measuring methane oxidation with closed chambers. Moreover, a correlation can be expected between methane oxidation and methane flux, enhancing the sensitivity of the measurement for oxidation. Wherever methane emissions are low, a relative high methane oxidation (in %) can be expected. At hotspots for methane emission, methane oxidation will be low, and very often negligible. As described in chapter 3.2.2 closed chambers tend to miss hot-spots and therefore overestimate methane oxidation.

Plume measurements

Methane sampled in the plume above or the landfill can also be at the basis of a measurement of methane emission (Bergamaschi et al., 1998; Börjesson, 2007; Chanton et al., 2009). The 1D-mass balance method enables simultaneous measurement of CH_4 and CO_2 emissions. This opens up the possibility of estimating methane oxidation from the CO_2/CH_4-ratio (Scharff et al., 2003; Oonk, 2010a) as described in 3.4.3. Advantage of plume methods above soil core measurements and closed chamber measurements is that is gives information about methane oxidation from large part of the landfill or even the entire landfill.

GAS PUSH-PULL TEST

In a gas push-pull test (Streese-Kleeberg et al., 2009), a gas mixture consisting of methane and O_2 and a conservative tracer (e. g., argon), is injected into the soil. Subsequently a mixture of injected gas and soil air is extracted from the same location and periodically sampled. From the differences in the breakthrough curves of methane and the conservative tracer, an indication of biological oxidation of the top-soil is derived. Advantages of the push-pull test is that is a relative simple, cheap and robust method, with simple equipment required and little or no assumptions to be made upon analysis. Disadvantage is again the local nature of the measurement. Besides it doesn't really give actual methane oxidation at a certain spot at the landfill, but more the latent oxidation capacity.

3.4.3 ANALYSIS AND INTERPRETATION

Analysis and interpretation, resulting in a quantification of methane oxidation might be done by:

- ^{13}C-analysis. When methane is formed within the waste, it contains besides ^{12}C also a specific amount of the isotope ^{13}C. In the top-layer ^{12}C is oxidized more rapidly and as a result the ^{13}C-content in the emitted gas is increased. Analysis of the ^{13}C-content in the emitted methane, and subsequent comparison to the methane as formed results in an estimate of methane oxidation. ^{13}C-analyses are generally regarded as the most accurate method to quantify methane oxidation. The method itself contains a few assumption as a result of which the final result becomes more uncertain. However all uncertainties point in the same direction and consequently, the result of a ^{13}C-analysis can be regarded as the minimum methane oxidation (Chanton et al., 2008). A homogeneous conversion (in % of methane flux coming from the waste) of methane in the top-layer is one assumption. As a result of a by-pass effect, actual methane oxidation effect can be higher as calculated from ^{13}C-analyses.
- From a CH_4/CO_2-mass balance. Upon production, landfill gas has a certain CH_4/CO_2-ratio and oxidation of methane changes this ratio. Measuring emissions of CH_4 and CO_2 and the ratio in extracted landfill gas gives an estimate of methane oxidation. Results of methane oxidation however can be considered to be less reliable for a number of reasons (Scheutz et al., 2009):
  - Part of methane that is oxidized is not converted to carbon dioxide, but is used for growth of the bacteriological population, responsible for methane oxidation. As a result, the ratio of CH_4/CO_2 changes less than expected. The importance of this effect is unclear and depends on the fraction of methane that is used for growth of bacteriological population (the yield factor). After several years, a steady state condition of growth and decay can be expected. In that case the impact of growth can be considered negligible. From lab-experiments, yield factors up to 70% are reported (ref). However in the field hardly ever significant amounts of methane oxidizing bacteria are observed and the actual yield factor is most likely much less; 10% at maximum (Gebert, 2010);
  - The effect is also masked by other sources and sinks of CO_2, e.g. assimilation/dissimilation of CO_2 by the vegetation on the landfill. During the day

vegetation consumes CO_2, where during night-time part of the CO_2 is released again. The difference of both is used for biomass growth.

3.5 Conclusions measuring methane emissions and oxidation

3.5.1 Methane emissions

In the past decades, considerable effort are done to develop methods to measure methane emissions from landfills. However at the moment there is no single method, that is widely recognized as the preferred method to measure annual average methane emissions. The main difficulty in measuring methane emissions from landfills is the spatial and temporal variability of emissions, in combination with the size of a modern landfill.

There are several methods available. Closed chamber methods are amongst the most frequently applied. However, there is a growing agreement that closed chambers are not able to catch spatial variability of emissions and tend to underestimate emissions, even when prescribed procedures are followed for grid-wise measurements and application of geo-statistical methods for interpolation.

Other methods applied are micro-meteorological methods, mass-balance methods and plume measurements. All methods have advantages and disadvantages and all method have their specific constraints with respect to landfill size, topography, accessibility and e.g. sources of methane adjacent to the landfill. However in many applications measurement costs will also be a factor of importance. So ultimately methods will prevail that are low-cost and still have acceptable accuracy. Best candidates seem to be the 1D-mass balance method and the mobile or static plume tracer measurements.

Claimed accuracy of methods is in the order of magnitude of 25%, on the conditions that the measurement stays within the predefined constraints. Intercomparisons of measurement methods and a measurement of a known methane release raise doubt, whether this accuracy is also met in actual field-situations. It will take more measurement intercomparisons and measurements in situations with controlled methane release, whether this accuracy can be claimed with confidence.

For measuring annual average emissions, day-to-day and seasonal variations have to be dealt with 4 to 6 one-day measurements will be required.

3.5.2 Methane oxidation

Most information on methane oxidation on actual landfills is obtained from closed chambers and interpretation of ^{13}C analyses of the methane captured in the box. This method however tends to miss hot-spots of methane emission and tends to overestimate methane oxidation. As a result most field results on oxidation is considered less reliable. Alternatives as measuring oxidation from soil core profiles, closed chambers in combination with interpretation of the CH_4/CO_2-ratio and gas

push-pull tests have a similar local nature and have the same disadvantage as the closed chamber method.

The most accurate method to quantify methane oxidation is measurement and interpretation of ^{13}C in the plume. Also this method is at discussion and most recent insights indicate that it might underestimate methane oxidation.

1D-mass balance measurements might be an alternative. However both the measurement of CH_4 and CO_2-emissions using this technique and the estimation of methane oxidation from a shift in CH_4/CO_2-ratio is not widely acknowledged as a reliable method.

CHAPTER 4. ESTIMATING EMISSIONS BASED ON RECOVERED AMOUNTS OF METHANE

4.1 GENERAL

The amount of methane recovered might be used as an indicator for methane generation. Methane emissions can subsequently be calculated from the methane mass-balance as given in chapter 2.1. However, methane recovery is only indicative of actual generation, when methane recovery meets certain prerequisites. And even then, the method will only give an rough indication of generation.

Application of methane recovery for estimating methane generation, implies that the recovery efficiency is know. In this case methane generation can be calculated as:

$$\text{methane generation} = \text{methane recovery}/(1-\eta)$$

in which η is the efficiency for methane or landfill gas recovery.

4.2 PREREQUISITES

The recovery efficiency can only be estimated when the system for landfill gas recovery is well designed and operated. So the amount of methane generated is an indication, only when a few prerequisites are met.

A very important prerequisite is that the amount recovered has to be limited by generation and not by e.g. the possibilities for utilization of the landfill gas. An example to clarify this: lets assume a landfill where 1000 m^3 of landfill gas is produced per hour. Technically it should be possible to recover 300-700 m^3 per hour. However the gas engine for landfill gas utilization can only handle 250 m^3 per hour. In such a case, there is no economic incentive to extract more landfill gas than the 250 m^3 per hour that can be utilized, and methane recovery is not an indication of landfill gas generation.

Another prerequisite is that landfill gas recovery is state of the art. This implies e.g. that sufficient recovery wells are used (when vertical wells are applied, a minimum of 2 ha), gas recovery also in parts of the landfill that are currently exploited, biweekly control and adjustment of under-pressure on the wells.

4.3 APPLICATION

When the prerequisites described above are met, amounts of methane recovered might give an indication of methane generation, assuming an efficiency of landfill gas recovery, which can vary between 10 and 80% (Oonk and Boom, 1995; Scharff et al. 2003). However measurements here were performed on a range of landfills and it is unclear whether extraction at landfills was state of the art. It can be assumed that

state of the art landfill recovery results in efficiencies between 25 and 75%. Further measurements are required to reduce this range in expected efficiency.

It is not recommended to base an estimate of methane generation solely on the amount of methane recovered. The methodology itself is still rather uncertain. However it can be used in combination with a prognosis of methane generation for a further reduction of uncertainties. Based on recovered amounts and assumed minimum and maximum recovery efficiency, either the low end or the high end of modeled methane generation will be less likely.

CHAPTER 5: IMPROVING QUALITY OF LANDFILL METHANE EMISSION INVENTORIES

5.1 QUALITY OF AN EMISSION INVENTORY

In general, the quality of an emission inventory depends on the perspective for which the emission inventory is used (Pulles et al., 2008). Quality criteria for national inventories of greenhouse gases to UN-FCCC differ from quality criteria for data on individual companies in the framework of E-PRTR. For data used in a legal context (e.g. to verify whether a company complies to its emission limits), again different quality criteria exist: in the end they have to be convincing in court.

EMISSION REPORTING TO E-PRTR

E-PRTR (European pollutant release and transfer register) is the Europe-wide register that provides key environmental data from European industrial facilities. The register aims to contribute to transparency in public participation and political decision making. It is used by scientists e.g. to calculate dispersion of pollutants in the environment and predict effects of specific measures. Quality within E-PRTR is determined by the legislating authority of the specific industry, but the overall aim is to be as accurate as possible. So best guesses are common practice, and everyone can make their own assumptions as long as the legislating authority agrees.

NATIONAL INVENTORIES TO UN-FCCC

National inventories of landfill methane emissions to UN-FCCC have to comply to the IPCC-guidelines (IPCC, 1996, 2000, 2006). The data are used to verify whether or not countries comply to international agreements on greenhouse gas emission reduction. The IPCC-methodology is defined and also made leading to ensure that efforts of all countries are measured in the same way. The methodology prevents that a country can just use another set of assumptions to quantify emissions. Comparability and transparency are keywords with respect to quality. It is not so bad if an emission estimate wrong, just as long as all estimates from all countries are just as wrong. Countries are allowed to use a higher tiered method, on the condition that the methods is validated, using sufficient field data.

ENFORCING LEGISLATION

In a legal context, presumption of innocence is the leading principle in EU-countries. A legal entity is innocent until proven guilty. In this context quality of an emission estimate is related whether it will stand in court. As a result there will be tough criteria defined with respect to quality of the methodology. And when enforcing emission limits most likely only the low end of the uncertainty range of a modeled or measured emission will be relevant. E.g. when emissions are measured to be 1.000.000 kg methane per year with an error of 50%, only the emission of 500.000 kg/yr may be considered proven and compared to emission limits.

5.2 Improving methods to quantify landfill methane

Stepping away from the legal context of an emission inventory as described in chapter 5.1, there are ways to improve current methods to quantify methane emissions from landfills. Four pathways can be distinguished:

5.2.1 Harmonise and improve methane emission models

Harmonization of methane emission models

As concluded in chapter 2.6, several models are available, none of them is perfect and they all have their specific strong points (see also Table 3). It is very well possible to take the strengths of every model and combine them into a harmonized version. Identify strengths and weaknesses of various models; collect, estimate additional parameters, build model. A few remarks can be made with respect to a harmonized model:

- A harmonized model for landfill methane emissions, valid for the entire EU will most likely not result in a more accurate prediction of methane generation or emissions. The uncertainty of the model will still be that large, that it will overlap uncertainty ranges of outcome of many other models. Biggest advantage of a harmonized model are an improved comparability of results for different landfills, and a framework for improved and validated models in future;
- In such a harmonized model, distinction between climate zones (effect on half-life, methane generation potential, L_0, and methane oxidation), and future changes in waste composition have to be prepared as much as possible, even when model-parameters can not yet be filled in. The model should also be kept open for other future technological developments, such as more sustainable landfilling technologies (e.g. aerobic landfills, landfill bioreactors and landfills with enhanced oxidation top-covers);
- At the other hand, one should avoid too complex models, based on too many 'reasonable' assumptions. The complexity of the model and its expected accuracy should be in balance. The way the model is built and the ' science' behind it should not suggest more accuracy as it can live up to. A model that is too complex is also more difficult or impossible to validate;
- Model default values should be chosen in a more conservative way (assumptions leading to a slight overestimation of emissions, rather than an underestimation), whenever those parameters can be easily measured by the landfill owner (e.g. it is state of the art to weigh the amount of waste landfilled and register the origin of each load. It is also state of the art to measure the amount and composition of recovered landfill gas). This is to avoid that bad monitoring practices at the landfill are rewarded, lower emissions are obtained and improved monitoring at a site is discouraged. Also when new insights can be expected on a relative short term, a slightly conservative approach might be a stimulus for individual landfills or national authorities for further research. An example of the latter is oxidation in top-layers, where the conservative IPCC-default of 10% initiated a lot of research. However conservative chosen defaults

should be selected with care. When all sorts of conservative assumption are piled up in one model, ultimately model accuracy will fall victim;
- The model should preferably include emission estimates of all components relevant in E-PRTR, including the emissions from utilization and flaring of landfill gas. Providing a ' one-stop shop' will stimulate landfill owners to make use of the specific model;
- The model should define uncertainty ranges and also specify what this 'uncertainty' means. Is it a 95% or 98% confidence interval, is it an average expected uncertainty, is it an absolute uncertainty, is it a best-guess of uncertainty. Uncertainty could be calculated in a Monte-Carlo analysis. However if a Monte-Carlo analysis has to be an effective tool, it requires well-quantified uncertainty ranges for the most important parameters;
- When developing a harmonized model, one should keep in mind the possibilities for future validation of either individual model parameters or the entire model.

In general three levels of validation can be distinguished, ranging from an effort that will be relatively easy to organize, to efforts that comprise major research. This goes for both a harmonized model as well as for the existing group of methane formation or emission models:
- a 0^{th}-phase validation might be based on existing sets of field data of at one hand amount of waste, waste age and composition and at the other hand methane generation. A few sets are available from data used elsewhere, e.g. in the Netherlands (Oonk et al., 1994, 1995; Scharff et al., 2004), in California (Vogt et al., 1997) and in Canada (Thompson et al., 2009);
- a 1^{st}-phase validation could be based on additional field data, to be collected from state of the art recovery projects throughout Europe. Collection of sufficient data will be a considerable effort, performed by people throughout Europe with sufficient experience in landfill extraction to judge whether a recovery project is state of the art and amounts of landfill gas extracted are representative for landfill gas formation;
- a 2^{nd} phase validation comprises full scale measurements on both methane emissions and methane oxidation at a larger number of well-described landfills throughout Europe. Such an effort would imply a major joint effort, by several European research groups and would also include further development and harmonization of accurate and affordable measurement methods.

5.2.2 Improve and validate measurement methods

In chapter 3 in this report a several measurement methods are described and intercompared. In the end a careful preference is expressed for three of the available methods. This preference is a conclusion in this project and this conclusion is certainly not (yet) accepted by scientists and measurement specialists worldwide. The preference is also based on today's knowledge. In the next years, new, better or cheaper methods or analyzers could become available.

A first step would be a further discussion on available measurement methods, followed by some convergence towards conclusions.

A second step would be a more thorough test of methods in various situations. Controlled release tests, as the one described by Babilotte et al. (2008) will ultimately be a very convincing tool to generate confidence in the methods and quantify its accuracy.

A third step might be a further cost reduction of the method, e.g. by use of cheaper analytical equipment (e.g. sensors) might play a role in it. Transfer of knowledge and writing standardized measurement protocols will also enable further cost reduction.

Ultimately further cost reduction can be expected when taking methods away from the research institutes hand and technology over to specialized labs throughout Europe. Prerequisite for these labs to take the effort and build up required knowledge is a market for the product. This requires either a willingness of waste treatment companies to perform voluntary measurements on a more structural basis or new legislation that enables governments to prescribe measurements in specific cases.

5.2.3 Define a tiered approach

Emission estimates might also be improved by allowing landfills to perform higher tiered methods to quantify their emission. Such a tiered approach could consist of (based on Scheutz et al., 2009):

Tier 1:
When little or no information is available on amounts of waste and the waste composition, simple models could be applied, such as zero-order models or a Scholl-Canyon model. The French E-PRTR-model could be an excellent alternative to this, since it combines simplicity and still can be considered state of the art with respect to its scientific basis;

Tier 2: state of the art
When more information is available, methane emissions can be calculated from methane generation, recovery and oxidation using the models and methods described in chapter 2. At the moment it is up to the legislative authorities to judge whether a model or an estimation method (e.g. for landfill gas recovery, when amounts of landfill gas recovered are not measured) is accepted. When landfill gas recovery can be considered state of the art, methane generation can also be estimated from the amount recovered. This estimate can be used to improve accuracy of an estimate. Chapter 2 of this report gives guidance in this.

In general it is recommended to use conservative estimates (leading to slight overestimation of emissions, rather than an underestimation), to prevent that landfills are rewarded (lower emission estimates) for inadequate monitoring;

Tier 3:
When a landfill incidental performs emissions measurements, results might be used to validate the modeled emission estimate. Methane generation or emission models might be used to extrapolate the emission measurement to other years.

TIER 4:

Frequent measurement of emissions enables an emission inventory entirely based on emission measurements. No modeling is required for quantification of methane emissions. Modeling can still be interesting to improve knowledge of behavior of the specific landfill. In general, these types of comparisons add to understanding of the models accuracy.

5.2.4 KNOWLEDGE TRANSFER

Improvement of quantification of landfill methane emission will also benefit from transfer of knowledge to member states governments, to the legislative authorities in the member states and to landfill owners. Transfer of knowledge can imply:

- knowledge on methane emission models, their pro's and cons and the impact of assumptions on the quantification of emissions;
- knowledge on at least the affordable measurement methods for landfill methane
- acceptance of higher tiered methods to quantify emissions

Making this report available to governments and legislative authorities can be a first step in knowledge transfer.

5.3 IMPACT OF IMPROVEMENTS

As explained in chapter 5.1 'quality of emission estimate' has a different meaning, depending on the context. It also depends on this context whether improvements will suffice to change methodologies.

IMPROVING EMISSION REPORTING TO E-PRTR

Most easy step is improvement of emissions reported to E-PRTR. Quality is determined by the legislating authority of the specific landfill. On the condition of sufficient transfer of knowledge to legislating authorities, both harmonized emission models and tiered approaches will be applicable here. Improvement will mainly imply an improved intercomparability of emissions between landfills. The method itself does not necessarily become more accurate upon harmonization. Improved accuracy will only be obtained after proper field validation.

NATIONAL INVENTORIES TO UN-FCCC

A harmonized model can not be considered an improvement in reporting of countries emissions to UN-FCCC. The current IPCC-methodology (IPCC, 2006) can already be considered as a model, that is harmonized in the framework of pending climate negotiations. The IPCC-methodology is made to prevent that individual countries make their own models, based on their own (but slightly differing from IPCC) 'reasonable' assumptions. IPCC does allow higher tiered methods, on the condition of validation in a sufficient number of measurements. A new harmonized model, thoroughly validated (see chapter 5.2.1) will be considered as such a higher tiered method and be acceptable to UN-FCCC.

ENFORCING LEGISLATION

When enforcing legislation, both the method to quantify methane emissions and their inaccuracy most likely has to be demonstrated. When modeling emissions, a harmonized model should be thoroughly validated and the method of defining accuracy should be beyond discussion. A Monte-Carlo analysis could be the preferred method to quantify inaccuracy, since the result of such an analysis is a specified accuracy distribution. When measuring emissions, the methodology (methodologies) should be accepted between peers and the accuracy should be well assessed. Testing methods in controlled release tests under varying conditions seem to be a strong tool in this.

SYMBOLS

In all available publications, there is little or no unity in symbols used for various parameters. For clarity, in this report symbols and units from the IPCC-methodology (2006) are used as much as possible. For several publications used this means that parameters had to be converted or recalculated to match the units below. Whenever this recalculation required assumptions, this is indicated in a footnote.

C_{bg}	background methane concentration	(g/m^3)
C_{CH4}	methane concentration	(g/m^3)
C_{trac}	tracer concentration	(g/m^3)
CH_4-e	methane emitted	(kg/y)
CH_4-g	methane generated	(kg/y)
$DDOC_m$:	mass of decomposable DOC deposited	(ton) [19]
DOC:	degradable organic carbon in waste	(ton C/ton waste)
DOC_f:	fraction of DOC that can decompose	(-/-)
F:	fraction of CH_4 in generated landfill gas	(volume fraction)
i:	waste category or type/material	
k:	rate constant of biodegradation	(1/y)
L_0:	CH_4 generation potential	(ton/ton waste)
MCF:	methane correction factor	(-/-)
OX_T:	oxidation factor in year T	(-/-)
R_T:	recovered CH_4 in year T	(ton)
t:	time after landfilling	(y)
T:	inventory year	
$t_{1/2}$:	half-time of biodegradation	(y)
v_w:	wind velocity	(m/s)
$v_{w,y}$	vertical component of wind veloxity	(m/s)
W:	mass of waste deposited	(ton)

[19] 1 ton = 1000 kg = 10^{-3} gG

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