# EMISSION MEASUREMENTS AS A TOOL TO IMPROVE METHANE EMISSION ESTIMATES

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Summary:

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

Keywords:

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

## 1. INTRODUCTION

### 1.1 General

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

### 1.2 Dutch waste policy

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

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

As a result it is expected that the amount of organic waste that is ultimately landfilled is going to decrease significantly (AOO, 1999). On top of that other important factors that influence biochemical processes in landfills will change as well. The humidity of the landfill will drop, since a large part of the water in the landfill comes in with the organic waste. The nature of the organic material will change from relatively rapidly degradable to a more woody, less well degradable nature. The sharp initial increase in temperature will be less due to the lack of rapidly degradable materials.

### 1.3 Dutch government

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

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

Another problem of the existing methodology for estimating methane emissions from landfills is that it is not able to monitor some specific measures that are proposed for furthergoing emission reduction. An example of this is the enhanced oxidation in top-layers. This option is widely recognised as a promising method to reduce methane emissions (VROM, 1999). Emission reductions achieved in this way can at present not be monitored, due to lack of a suitable monitoring methodology (either a measurement method or an accepted reduced emission factor) and the results obtained can not be incorporated in the national assessment.

### 1.4 Landfill operator

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

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

## 2. STRATEGIES TO IMPROVED EMISSION ESTIMATES

### 2.1. Current methodology

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

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

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

### 2.2 Two strategies for the national government

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

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

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

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

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

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

### 2.3. Landfill operators

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

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

estimated from the sum of methane and carbon dioxide emissions, it would be extremely useful when carbon dioxide can be measured as well.

## 3. VARIABILITY OF EMISSIONS

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

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

*Table 1: Some observations obtained from literature*

| Observation | Author |
|---|---|
| *Spatial variation:* | |
| Up to factor 1,000 difference between box measurements | Verschut et al., 1991 |
| No correlation between two box measurements >6m apart | Czepiel et al., 1996a |
| Up to factor 500 difference between measurements | Nozhevnikova et al., 1993 |
| *Hourly and daily variation:* | |
| Doubled emission with 30 mbar atmospheric pressure drop | Czepiel et al., 1996a |
| Reduced methane flux wit atmospheric pressure increase | Verschut et al., 1991 |
| 10-fold emission increase days after extraction system failure | Shorter et al.,1998 |
| *Seasonal variation:* | |
| From winter to summer 0 – 40% oxidation of methane | Boeckx et al., 1996 |
| Idem | Czepiel et al., 1996b |
| Maximum oxidation at 50% water holding capacity | Czepiel et al., 1996b |
| Higher oxidation at higher temperature and lower water content | Christophersen et al., 1999 |
| Complete oxidation except when soil was frozen | Maurice et al., 1997 |

## 4. LIMITATIONS OF MEASUREMENT METHODS

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

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

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

*Table 2: Comparison of measurement techniques*

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

When emissions are to be measured using closed chambers, the low spatial and temporal resolution requests a large number of relocations (more than 30 a day) on several days throughout the year (no experience how many measurement days are required here). This makes this method very labour-intensive and very expensive.
The mass-balance method seems to be better suited to measure emissions from larger surfaces during longer times. Its capability to measure $CH_4$ and $CO_2$ gives insight in the primary processes leading to emissions: methane formation and oxidation. For larger sites however, this method might bring about some problems and further developments are required to enable measurements from the whole of a larger landfill site. Developments might comprise the application of longer pylons (16 meter pylons are commercially available) and the application of more accurate $CO_2$-analysers. Drawback of the mass-balance method is that since there is not so much experience with the method, validation might be considered a requirement.
Plume measurements with TDL or FTIR technology can be considered the most accurate methods to measure emissions from an entire site. But the drawbacks of this method (complexicity, low temporal resolution and high costs of prolonged measurement campaigns) render this method not suitable to give a reliable impression of the annual emission. Since these plume measurements gives an indication of emissions at a single day, its costs will in practice reduce its temporal resolution. But since the method is generally accepted as being accurate it might be the best method for validation of other methods. The last drawback of the plume method may be avoided when a suitable stationary plume method based on gas sampling can be developed.
$^{13}CH_4$ -measurements are widely recognised for their applicability in quantifying the amount of methane oxidised in the top-layer, so this method might be the primary candidate to validate the suitability of the mass-balance or stationary plume method to get an impression of methane oxidation.

## 5. RECENT EMISSION MEASUREMENTS IN THE NETHERLANDS

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

An overview of the results is presented in table 4.

*Table 3: Afvalzorg landfills where emissions were measured*

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

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

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

At the Braambergen landfill a production estimate was calculated for November 1999. A level of 250 $m^3$ $CH_4.h^{-1}$ was obtained. Measurements using the plume method yielded an emission level of 240 $m^3$ $CH_4.h^{-1}$ , which is almost equal to the total production level. High peaks in methane emission were observed down wind from cells with relatively (1-2 years old) fresh waste. On the day of measurement the gas extraction system recovered 110 $m^3$ $CH_4.h^{-1}$ . So even with oxidation of 0% the sum of emission and extraction exceeds the production estimate. This discrepancy could be caused by the measurements being non-representative for mean emissions. But in December 1999, the extraction system was able to recover 245 $m^3$ $CH_4.h^{-1}$ (using extra wells). Therefore there is no doubt that the estimate of the production level is to low. Although this is the only landfill with recent deposits of municipal solid waste, a gasproduction prognosis that proved too low was not anticipated.

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

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

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

At Zeeasterweg landfill a gas production of 130 $m^3$ $CH_4$ $.h^{-1}$ was calcutated. Mass balance measurements during six weeks indicated a methane emission of 35 $m^3$ $CH_4$ $.h^{-1}$. Apparantly more than 60% of the methane was oxidized eventhough it was winter at the time. The explanation might be that the waste is relatively old and the average height is low, resulting in low fluxes per $m^2$. Furthermore the average day temperatures were not too low (between 9 and 12°C) to prevent microbiological activity.

## 6. CONCLUSIONS AND RECOMMENDATIONS

In general the findings of recent emission measurements in the Netherlands are in accordance with experiences in literature. Both suggest that in order to obtain an accurate annual emission estimate a measurement method with sufficient spatial and temporal resolution is required. Both national governments and landfill operators could benefit from such methods. National governments could obtain a more accurate description of the actual national methane emission and thus be able to define a more effective emission reduction policy. Landfill operators could obtain a more accurate estimation of the emission of each landfill and thus be able to make a more effective design of emission reduction measures. The measurement efforts of the landfill operators could contribute to the national estimate.

The mass balance method method can provide emission data of methane and carbon dioxide with a high spatial and temporal resolution and is therefore a good candidate. Combining methane and carbon dioxide emission data will also generate data on landfill gas production and methane oxidation. Another good candidate might be a low-cost system, derived from the stationary plume method. Both methods however are not internationally accepted and need further validation. Mobile plume measurements seem to be a good candidate for validation of emissions. Isotope measurements can be applied to estimate the average oxidation effect at the landfill.

It is recommended that national governments, research institutes and landfill operators with a genuine concern about landfill gas emissions team up to develop, validate and find acceptance for more accurate and affordable measurement methods.

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