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Applying guidance for methane emission estimation for landfills

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Abstract

Quantification of methane emission from landfills is important to evaluate measures for reduction of greenhouse gas emissions. Both the United Nations and the European Union have adopted protocols to ensure quantification of methane emission from individual landfills. The purpose of these protocols is to disclose emission data to regulators and the general public. Criteria such as timeliness, completeness, certainty, comparability, consistency and transparency are set for inclusion of emission data in a publicly accessible database. All methods given as guidance to landfill operators to estimate landfill methane emissions are based on models. In this paper the consequences of applying six different models for estimates of three landfills are explored. It is not the intention of this paper to criticise or validate models. The modelling results are compared with whole site methane emission measurements. A huge difference in results is observed. This raises doubts about the accuracy of the models. It also indicates that at least some of the criteria previously mentioned are not met for the tools currently available to estimate methane emissions from individual landfills. This will inevitably lead to compiling and comparing data with an incomparable origin. Harmonisation of models is recommended. This may not necessarily reduce uncertainty, but it will at least result in comparable, consistent and transparent data.

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1. Introduction

Methane emission from landfills is a major contributor to the Greenhouse Effect. Regulators throughout the world are implementing waste management strategies, policies and regulations aimed at reducing methane emission from landfills. Landfills are not a point source, but a diffuse source of methane. Moreover, the emission has a high temporal and spatial variability (Scharff et al., 2000). Therefore, it is not easy to measure methane emissions. In order to determine the effectiveness of measures aimed at reducing methane emission from landfills, quantification of the methane emission either per country or per landfill is essential.

In May 2003, the United Nations (UN) adopted the Protocol on Pollutants Release and Transfer Registers (also known as PRTRs- or Kiev-protocol). Amongst others, this protocol requires landfills receiving more than 10 ton per day or with a total capacity of 25,000 ton to determine their methane emissions individually and make them available to the general public and their national government from 2007 onwards. The Commission of the European Communities (CEC) is currently considering adaptation of the European Pollutants Emission Register (EPER) into the E-PRTRs (CEC, 2004) to comply with the UN PRTRs-protocol. EPER requires that criteria such as timeliness, completeness, certainty, comparability, consistency and transparency are met for inclusion of emission data in a publicly accessible database. The European Federation of Waste Management and Environmental Services (FEAD), representing the majority of the European waste management industry, supports reporting of emissions from landfills in accordance with E-PRTRs (FEAD, 2005). This is in accordance with FEAD policy for landfilling (FEAD, 2003) and more specifically with respect to open and readily available information.

National governments also report to the Intergovernmental Panel on Climate Change (IPCC) with respect to the Kyoto protocol. It is obvious that one method that is suitable for reporting in all cases has advantages and will

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prevent confusion. IPCC is currently updating its guidance on methane emission estimates for entire nations (Pipatti, 2005). In the new guidance only first-order degradation models are recommended. IPCC guidance for estimation was never intended to be applied to individual landfills (Pipatti, 2005). A first-order degradation model is considered sufficiently accurate for estimation of landfill methane emissions of an entire nation. In every country there are landfills with atypically low and atypically high emissions. When estimating national landfill methane emissions, these "outliers" will "statistically counterbalance" each other (Oonk, 2005). Consequently, we cannot presume that first-order degradation models are sufficiently accurate for the estimation of individual landfill methane emissions.

Several governments are considering or have already given guidance to landfill operators on determining their methane emissions. The recommended approaches all involve modelling of methane production. Data in a publicly accessible database will inevitably be compared both by regulators and by the general public. Typically, there is limited scope in a database to explain how the data were derived, and if there is room for explanation it might be disregarded. Different methods may result in different outcomes. The risk that regulators will base policy measures on a misconception of data is not purely hypothetical. It is also possible that members of the public object to a landfill permit application because the database seems to indicate that landfills in other countries are more environmentally sound. Negative effects of incomparable data are not hypothetical. In this paper, NV Afvalzorg, a landfill operator in the Netherlands, explores the effects of applying several methods currently available for the estimation of methane emission using waste acceptance data from three of its landfills. In that respect, comparability and transparency are of particular interest. It is not the intention to assess the suitability or accuracy of the methods discussed in this paper. A validation study requires a much larger number of landfills.

2. Background

Several models to predict methane emissions originating from landfills have been proposed or are recommended by national governments. The most common type of models use single-phase or multi-phase first-order kinetics that describe the decay of biodegradable waste and the production of methane. Methane production in combination with the oxidation capacity of the top cover and landfill gas (LFG) extraction data enables calculation of methane emission. LFG extraction can be measured accurately at low costs. Measurement of oxidation is complicated and expensive (Scharff et al., 2000). Research indicates a huge variation in oxidation as a percentage of LFG production (Scharff et al., 2000). Models in general use a default value for oxidation in the order of 10% of the methane transported through the cover of the landfill. Most methane production models are based on municipal solid waste (MSW).

They are therefore not automatically suitable for situations with reduced amounts of organic waste. In most cases emission model validation has been carried out using LFG extraction data and assumptions for extraction efficiency and methane oxidation. Thus, major uncertainties were introduced. The authors are of the opinion that a proper validation of methane emission models requires comparison with whole site emission measurement data. Only two studies (Oonk and Boom, 1995; Huitric and Soni, 1997) have validated models using whole site methane emission measurements.

A problem that emerges immediately when a landfill operator applies one of the models is how to divide the different waste categories registered by the operator over the limited number of categories given in the models. The definitions of waste categories can differ between countries. For instance the category MSW is not used in the Netherlands. The Netherlands make a distinction between household waste and commercial waste. Within the category of commercial waste a subcategory exists called "waste from offices, shops and services". It seems that this category should be included to approximate with MSW in other countries. Residues from sorting plants are also considered to be commercial waste. Afvalzorg registers approximately 50 different waste categories. These categories are mainly based on tax regulations and on regulations related to hazardous waste. It is not possible to use these categories for modelling methane production and emissions other than based on a best guess. It is evident that it is impossible to control the best guess of all the landfill operators in the world. In Europe this problem will not be reduced by the introduction of the European Waste Catalogue (EWC). The EWC has approximately 800 different waste categories and they are not based on carbon content. Even if the carbon content of all these categories was known, it would not be easy to allow for so many entries in a straightforward simple model. In general there is very little information available on carbon content in waste. An illustration of this is that three of the six models taken into consideration in this paper refer to the same research investigating the composition of household and commercial waste that was carried out in the Netherlands some 15 years ago (Cornelissen, 1992).

In 1994 a study (Oonk et al., 1994) was performed at several landfills in the Netherlands. Both first-order and multi-phase models showed low mean relative errors in contrast to zero order models. This study resulted in the development by the Dutch research institute TNO (The Netherlands Organisation of Applied Scientific Research) of the single-phase first-order model used by the Dutch government to calculate and report national methane emissions as if the waste were deposited at one landfill. The Afvalzorg multi-phase model was developed by the Agricultural University of Wageningen based on TNO's model and the recommendations of the "Adviescentrum Stortgas" (Advice Centre on Landfill Gas) (Scheepers and van Zanten, 1994). LandGEM is recommended by the United States Environmental Protection Agency (US-EPA). The Anglo-Welsh Environment Agency prefers GasSim as the model for individual landfill operators to calculate and report their methane emissions. Recently, new models were developed in order to calculate landfill gas emissions in Germany and France.

3. Methods

3.1. Case studies

For EU landfill operators, guidance should preferably take into account that EU waste policy is aiming for diversion of organic waste from landfills and consequently landfills that contain small amounts of organic matter. In the last two decades, waste policy in the Netherlands resulted in a reduced amount of landfilled waste and a change in the composition of landfilled waste. In the province of North-Holland, household waste has been incinerated since the 1970s. The Nauerna landfill has a total surface of 72 ha. of which 68 ha is used to dispose of waste. The landfilling of waste was started in 1985, and the site is still in operation. From 1985 to 2004 a total of 9.4×10^6 Mg of waste was landfilled at Nauerna. The annual amounts of different types of waste are presented in Fig. 1. The abbreviations are explained in Table 2. Waste at the Nauerna landfill is not only characterised by a low content in organic matter, it also contains organic matter that is not readily biodegradable. This makes it a good case study for the future conditions of EU waste policy. Because of the low organic content of the waste, until 1995 it was believed that LFG extraction would not be cost effective. A low level of LFG production on a large site results in a methane emission that cannot be considered to be negligible. Therefore, LFG extraction was started in 1997 and its extent increased in

2000. The low rate of gas production, combined with waste with a very low porosity, makes it extremely difficult to reach a satisfactory extraction efficiency.

The Braambergen landfill was operated by a regional authority before it was taken over by Afvalzorg in 1997. LFG extraction was also started in 1997. The surface of the site is 46 ha, of which 30 ha are used for waste disposal. From 1982 to 2004 a total amount of 2.2×10^6 Mg of waste was landfilled at Braambergen (see Fig. 2). Landfilling of household waste was stopped in 2000. If household waste, coarse household waste and commercial waste, are assumed to be included in MSW, then more than half of the waste disposed on this site can be considered MSW. In that respect Braambergen is a more traditional landfill.

The Wieringermeer landfill was also operated by a regional authority and taken over by Afvalzorg in 1997. LFG extraction was started in 1996. The surface of the site is 44 ha, of which 32 ha are used for waste disposal. From 1985 to 2004 a total amount of 2.1×10^6 Mg of waste was landfilled at Wieringermeer (see Fig. 3). This waste is mostly commercial waste.

3.2. Methane emission models

In this study, six different models are used to calculate landfill methane emissions:

- First order model (TNO) (Oonk and Boom, 1995).
- Multi-phase model (Afvalzorg, developed in 1996).
- LandGEM (US-EPA) (US-EPA, 2001).
- GasSim (Environment Agency UK and Golder Associates) (Gregory et al., 2003).
- EPER model France (ADEME) (Budka, 2003).
- EPER model Germany (Umwelt Bundesamt) (Hermann, 2005).

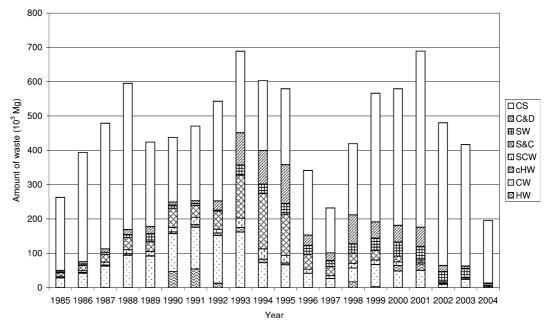


Fig. 1. Amounts of waste disposed at Nauerna landfill.

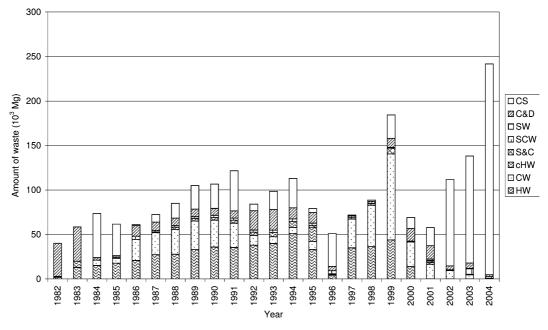


Fig. 2. Amounts of waste disposed at Braambergen landfill.

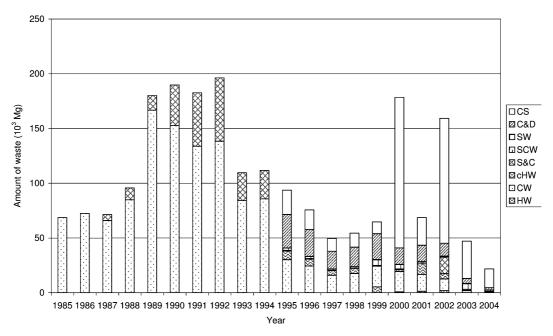


Fig. 3. Amounts of waste disposed at Wieringermeer landfill.

ς

In the following section each model is described individually. The units of the parameters and the default values used are given for each model.

3.2.1. First order model (TNO)

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

$$\alpha_t = \varsigma 1.87 A C_0 k_1 e^{-k_1 t} \tag{1}$$

 α_t landfill gas production at a given time $[m^3LFG \cdot y^{-1}]$

dissimilation factor 0.58 [-]

1.87 conversion factor
$$[m^3LFG \cdot kgC_{degraded}^{-1}]$$

- A amount of waste in place [Mg]
- C_{o} amount of organic carbon in waste [kgC · Mg waste⁻¹]
- k_1 degradation rate constant 0.094 [y⁻¹]
- *t* time elapsed since depositing [y]

The TNO model calculates LFG production based on the degraded organic carbon in the waste (Table 1). Gas-Sim multi-phase (see Section 3.2.4) uses carbon content

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

Waste category	Organic carbon content $[kgC \cdot Mg^{-1}]$
Contaminated soil	11
Construction and demolition waste	11
Shredder waste	130
Street cleansing waste	90
Sewage sludge and compost	90
Coarse household waste	130
Commercial waste	111
Household waste	130

to calculate methane production and emission. The Afvalzorg multi-phase model (see Section 3.2.2) calculates the LFG production with organic matter content. Other models provided a specific methane production per Mg waste. In this paper, conversions have been made between organic matter and organic carbon and between LFG and methane. All emission data were expressed in Gg methane per year. Organic matter was assumed to be predominantly cellulose. The biodegradation of cellulose can chemically be described by

Therefore

Methane production per kgOM degraded: $\frac{48}{180.714} =$ $0.373 \text{ m}^3 \text{ CH}_4 = 0.75 \text{ m}^3 \text{LFG}$ Methane production per kgC degraded: $\frac{48}{72,714} =$ $0.933 \text{ m}^3 \text{ CH}_4 = 1.87 \text{ m}^3 \text{LFG}$

To calculate methane production, the LFG production obtained with the TNO model is multiplied with the methane concentration of 50% and volumetric mass of 714 gCH₄ \cdot m⁻³. In order to derive methane emission based upon the production estimate, a very straight forward calculation is used:

 CH_4 emission = CH_4 production - CH_4 recovery

$$- CH_4$$
 oxidation (3)

Table 2

Organic matter content used in the Afvalzorg multi-phase model

This calculation can be and was used in many approaches, e.g., the first-order (TNO), multi-phase (Afvalzorg and Gas-Sim) and LandGEM models. It is apparent that the accuracy of the production model is an important factor in this type of approach. The recovery can be measured accurately. However, the oxidation of methane is usually not known with any degree of certainty and in general an assumed value of 10% is used. Emission measurements can be used to establish the appropriateness of this assumed value.

3.2.2. Multi-phase model (Afvalzorg)

Different types of waste contain different fractions of organic matter that degrade at different rates. The advantage of a multi-phase model is that the typical waste composition can be taken into account. In the Afvalzorg multiphase model, eight waste categories and three fractions are distinguished. For each fraction LFG production is calculated separately. The waste categories, fractions and rate constants used in the Afvalzorg multi-phase model are described in Tables 2 and 3. The multi-phase model is a first-order model and can be described mathematically by

$$\alpha_t = \varsigma \sum_{i=1}^{3} cAC_{0,i} k_{1,i} e^{-k_{1,i}t}$$
(4)

where

- landfill gas production at a given time $[m^3LFG \cdot y^{-1}]$ α_t
- dissimilation factor [–]^a ς

waste fraction with degradation rate $k_{1,i} [kg_i \cdot kg_{waste}^{-1}]^b$ conversion factor $[m^3 LFG \cdot kgOM_{degraded}^{-1}]^c$ i

- с
- amount of waste in place [Mg] A
- $C_{\rm o}$ amount of organic matter in waste $[kgOM \cdot Mg waste^{-1}]$
- $k_{1,i}$ degradation rate constant of fraction $i [y^{-1}]^a$
- time elapsed since depositing [v] t

^a ς and k values for rapidly, moderately and slowly degradable waste fractions for Nauerna, Braambergen and Wieringermeer are presented in Table 3.

^b See Table 2.

 c Minimum and maximum values of 0.7 and 0.74 $m^{3}LFG\cdot$ kgOM_{degraded} were used.

Waste category	Minimum organic matter content [kgOM \cdot Mg ⁻¹]				Maximum organic matter content $[kgOM \cdot Mg^{-1}]$			
	Rap	Mod	Slow	Total ^a	Rap	Mod	Slow	Total ^a
CS: contaminated soil	0	2	6	40	0	3	8	42
C&D: construction and demolition	0	6	12	44	0	8	16	46
SW: shredder waste	0	6	18	60	0	11	25	70
SCW: street cleansing waste	9	18	27	90	12	22	40	100
S&C: sewage sludge and compost	8	38	45	150	11	45	48	160
cHW: coarse household waste	13	39	104	260	19	49	108	270
CW: commercial waste	13	52	104	260	19	54	108	270
HW: household waste	60	75	45	300	70	90	48	320

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

Table 3 Afvalzorg multi-phase model ς and k values per case study object

Landfill	Dissimilation factor ς	Rapidly degradable k ₁	Moderately degradable k ₂	Slowly degradable k ₃
Nauerna	0.7	0.187	0.099	0.030
Braambergen	0.8	0.231	0.116	0.030
Wieringermeer	0.7	0.187	0.099	0.030

For some waste categories, no organic matter or carbon content data were available. The division in phases was made by a 'best guess' of Afvalzorg. In order to express the uncertainty of this approach, a minimum and a maximum amount of organic matter was attributed to each phase of each category. Including the minimum and maximum values for the conversion factor, this results in a 'best guess' of minimum and maximum methane production and consequently minimum and maximum methane emission. To calculate methane emission in Gg per annum, the LFG production was multiplied with a methane concentration of 50% and 714 gCH₄ · m⁻³. The recovered quantity of methane with the recovery system was subtracted and a standard oxidation factor of 10% was applied according to Eq. (3).

3.2.3. LandGem US EPA

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

$$Q_{CH_4} = \sum_{i=1}^{n} k L_0 M_i(e^{-kt})$$
(5)

where

 Q_{CH_4} methane emission rate [m³CH₄ · y⁻¹]

- k methane generation constant (AP42 default = 0.04) $[y^{-1}]$
- L_0 methane generation potential (AP42 default = 100) [m³CH₄ · Mg waste⁻¹]
- M_i mass of waste in *i*th section [Mg]
- t_i age of the *i*th increment or section $[y^{-1}]$

In this paper, sections were considered to be annual amounts of disposed waste. US-EPA protocols (US-EPA, 2004, 2005) state that the composition of waste used in the model reflects US waste composition of MSW, inert material and other non-hazardous wastes. For a landfill containing non-biodegradable waste (i.e., inert material), such as ash from waste combustion, this portion may be subtracted from the waste acceptance rates. LandGEM recommends subtracting inert materials only when documentation is provided and approved by a regulatory authority. This is not recommended for sites that are typical MSW landfills containing a range of waste that may or may not be degradable. It was decided not to exclude any waste categories in this case study. LandGEM methane emissions were calculated with the total waste amounts given in Figs. 1–3 for all three case studies.

LandGEM provides methane generation constant and potential for both CAA (Clean Air Act) and AP42 (US-EPA, 1998) standards. It is recommended to use AP42 default values for standard landfills (US-EPA, 2004). CAA default values have a high methane generation potential (L_0) of 180 m³CH₄ · Mg waste⁻¹. It was decided to follow the recommendation and use AP42 standards exclusively in this paper.

After a model run with LandGEM, the methane emission was determined by subtracting the recovered quantity of methane with the recovery system and applying a standard oxidation factor of 10% according to Eq. (3).

3.2.4. GasSim

The GasSim model (Version 1.00, June 2002) (Gregory et al., 2003) is equipped with two approaches to calculate an estimate of methane emissions (GasSim manual Version 1.00). The first approach uses the GasSim multi-phase equation, which is based upon a multi-phase model described by Scheepers and van Zanten (1994). The Gas-Sim manual Version 1.00 (Golder Associates, 2002) does not provide the complete set of equations. Calculation modules in the program are protected. It was therefore not possible to reproduce the equations in this paper. The second approach to estimate methane formation is the LandGEM model, which is similar to the US-EPA model.

The multi-phase model requires waste input in Mg and the specific breakdown during the particular year of disposal. The waste categories and their carbon content used in the GasSim multi-phase model and the "translation" of Afvalzorg waste categories into GasSim categories are presented in Table 4. Each waste category in GasSim is made up of various fractions. The distribution of the fractions in each waste category is a table too large to reproduce in this paper (see Gregory et al., 2003). To give an impression, degradation rates, k values and fractions of the GasSim multi-phase model are presented in Table 5.

GasSim multi-phase methane emission calculations were performed with the model's average k values and a default oxidation factor of 10%. LandGEM methane emission calculations were performed with AP42 default values. Again CAA default values were not considered in this case study.

It is possible to include extraction efficiency of the LFG recovery system in the model and let GasSim calculate total surface emissions. However, this feature in GasSim only functions if waste in place is capped to a certain degree. This can be activated in the model by checking the checkbox and giving a percentage of waste capped. If the checkbox is not checked, but a recovery is operational and the efficiency is given in GasSim, the model does not take recovery into account. Large surfaces at all three landfills only had daily cover or were temporarily capped with a Table 4 Waste categories and organic carbon content of the GasSim multi-phase model

Waste categories	Carbon	Waste categories
GasSim	content	Afvalzorg
	$[kgC \cdot Mg^{-1}]$	-
Domestic	118	Household waste; coarse
		household waste
Civic amenity	71	
Commercial waste	182	Commercial waste
Industrial	0	Shredder waste; contaminated soil
Inert	0	Construction and demolition waste
Liquid inert	0	
Sewage sludge	36	Sewage sludge and compost; street cleansing waste
Composted organic material	51	-
Incinerator ash	4	
Waste sorted at MRF	0	
Recycling schemes	0	
Chemical sludge	0	
Industrial liq. waste	0	

thin layer of soil. Nevertheless, a LFG recovery system is in operation. Therefore, methane emissions were determined with Eq. (3). The removal of methane due to oxidation processes was excluded as this is already accounted for in both of the GasSim models.

3.2.5. EPER model France

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

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

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

$$A = F * H * [CH_4], \tag{6}$$

where

A	recovered amount of LFG $[m^3CH_4 \cdot y^{-1}]$
F	extraction rate of LFG $[m^3 LFG \cdot h^{-1}]$
Η	compressor yearly hours in operation $[h \cdot y^{-1}]$
$[CH_4]$	methane concentration in LFG $[m^{3}CH_{4} \cdot m^{-3}LFG]$

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

$$P = \frac{A}{\eta} \tag{7}$$

where

P production of methane $[m^3CH_4 \cdot y^{-1}]$

 η recovery efficiency [%]

Methane emission is then determined similar to Eq. (3).

In this paper, the second approach was used. The formation of methane is calculated with a multi-phase equation following the ADEME model:

$$FE_{CH_4} = \sum_{x} FE_0 * \left(\sum_{1,2,3} A_i * p_i * k_i * e^{-k_i t} \right)$$
(8)

where

$$FE_{CH_4}$$
 annual methane production $[m^3CH_4 \cdot y^{-1}]$

 FE_0 methane generation potential

 $[m^{3}CH_{4} \cdot Mg waste^{-1}]$

 p_i waste fraction with degradation rate k_i [kg_i · kg⁻¹_{waste}]

 k_i degradation rate of fraction $i [y^{-1}]$

t age of waste [y]

 A_i normalisation factor [-]

The dimensions left and right in the equation do not match. It would seem that the amount of waste in $Mg \cdot y^{-1}$ is missing on the right-hand side. In the spreadsheet itself, the annual amounts of waste are used in the calculation.

Table 5	
Degradation rate, k values and fractions of the GasSim multi-phase mode	ĺ

Degradability	k values	k values $[y^{-1}]$		Fraction
	Dry	Average	Wet	
Rapid	0.076	0.116	0.694	Putrescibles, fines, garden wastes, sewage sludge, incinerator ash
Moderate	0.046	0.076	0.116	1/4 paper (excluding newspaper), nappies, miscellaneous combustible, composted organic material
Slow	0.013	0.046	0.076	3/4 paper (excluding newspaper), newspaper, textiles

Although a normalisation factor is given in the equation, it seems that this factor is not included in the spreadsheet. The model describes three categories of waste and every category has a specific methane generation capacity per Mg of waste. The three categories, their specific methane generation capacities and the "translation" from Afvalzorg waste categories are given in Table 6. Fractions and k values for each waste category are presented in Table 7. The French EPER model assumes an oxidation capacity of the top cover of 10%. The total methane emission is calculated by

$$CH_{4 \text{ emission}} = P(1 - \eta) * 0.9 + FE_{CH_4} * 0.9$$
(9)

where

 η recovery efficiency [-]

3.2.6. EPER model Germany

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

$$\mathbf{Me} = M * \mathbf{BDC} * \mathbf{BDC}_f * F * D * C \tag{10}$$

where

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

The model only takes "unconditioned residential or similar waste" into account. This category is not registered as such at Afvalzorg landfills. For the purpose of the emission estimate household waste, coarse household waste and commercial waste have been taken into account. The estimate will be considerably lower should an operator decide only to include household waste. The proportion of biodegradable carbon converted (BDC_f) can be compared to the

Table 7							
Fractions	and k	values	of the	ADEME	multi-p	ohase mod	el

Category	Fraction 1 ($k = 0.500 \text{ y}^{-1}$) (%)	Fraction 2 ($k = 0.100 \text{ y}^{-1}$) (%)	Fraction 3 ($k = 0.040 \text{ y}^{-1}$) (%)	Overall k value $[y^{-1}]$
1	15	55	30	0.120
2	15	55	30	0.120
3	0	0	0	0.000

dissimilation factor used in other models. The factor 1.33 for carbon converted to methane (F) is the molar weight of methane over the molar weight of carbon. The methane concentration in the landfill gas accounts for the amount of carbon that is converted to carbon dioxide. On all three sites, the landfill gas extraction system is managed to maintain a methane concentration of approximately 50%. This value was used instead of the default value of 55%. The default value is proposed when the methane concentration of the landfill gas is unknown.

3.3. Methane emission measurements

The measurement methods considered in this paper were compared with each other in 2001. An extensive measurement programme comparing different measurement techniques was carried out at various landfills. Several Dutch landfill owners cooperated in the framework of the programme "Reductie Overige Broeikasgassen" (Reduction of Other Greenhouse Gases) (Scharff et al., 2003). The project was aimed at developing simpler measurement techniques for methane emissions.

During the project three measurement techniques were used:

- Mobile Plume Measurement with Tuneable Diode Laser (TDL).
- Stationary Plume Measurement (SPM).
- Mass Balance Measurement (MBM).

The mobile plume measurement technique is a tracer method. A group of international experts concluded during a workshop that tracer measurements are the most reliable methods for a single moment analysis (one day in the year)

Table 6

Waste categories and methane generation capacity of the ADEME model

Category	Methane generation	Waste categories ADEME	Waste categories Afvalzorg
1	$100 [m^{3}CH_{4} \cdot Mg^{-1}]$	MSW	Household waste
		Sludges	Sewage sludge and compost
		Yard waste	
2	$50 [m^3 CH_4 \cdot Mg^{-1}]$	Industrial waste	Coarse household waste
		Commercial waste	Commercial waste
		Biologically pre-treated waste	
3	$0 [m^3 CH_4 \cdot Mg^{-1}]$	Inert waste	Contaminated soil
		Non-biodegradable waste	Construction and demolition waste
		e e e e e e e e e e e e e e e e e e e	Shredder waste

if the purpose is to measure emissions from the whole landfill (Christophersen and Oonk, 2001). The TDL method measures the methane concentration downwind of the site in a transect through the plume. The plume is composed of all of the small methane emission spots on the landfill surface. The concentration measurements are performed by driving a tuneable diode laser along a transect through the methane plume. A plume transect takes approximately 5–7 min; 8–12 transects are made in order to determine the methane emission for the whole landfill. The overall emission is estimated with meteorological data and a Gaussian dispersion model. The methane emission will not be constant through time. Since the measurement time is limited to one day, a TDL measurement only gives a good estimation of the emission level on the day of measurement.

There is a similarity between the SPM and TDL measurement methods. Both quantify methane emissions by use of the downwind plume originating from the landfill site. The SPM uses four fixed gasbag sampling stations around the landfill. Combining meteorological data and computer modelling, the four receptor stations can be activated whenever predicted methane concentrations surpass a certain threshold level. Once activated, air samples are taken for a 30 min period of time at two computer selected stations. One station is activated for background methane concentrations and the second station is activated to measure plume concentrations. The samples are analysed with a gas chromatograph. A Gaussian model is used to predict methane concentrations at the receptor stations.

In the MBM the vertical methane concentration profile is measured together with the wind velocity profile. The profiles are gathered by means of sampling points in a pole up to 26 m in height. In order to asses the temporal variation of the landfill methane emission, a landfill is divided in several sections. Usually, in a period of 4–6 weeks, wind directions change sufficiently to assess the contribution of each sector to the methane emission of the whole landfill.

Both SPM and MBM measurement techniques are more suitable to obtain an annual methane emission estimate for a particular landfill then the TDL technique. Both techniques have advantages and disadvantages and are suitable in different situations. In combination the SPM and MBM techniques are complementary to the TDL measurement technique and give more insight into the temporal variation of the emissions originating from the landfill.

4. Results and discussion

4.1. Results

The results of model estimations and the different methane measurements are presented in Figs. 4–6. The measurement results were obtained from various projects. From 1997, whole site measurements with the best available techniques were carried out annually at the Nauerna landfill (Hensen, 1997, 1998, 2000a). The first whole site measurement at the Braambergen landfill was carried out in 1999 (Hensen, 2000b). Measurements were carried out on all three sites in 2001 as part of a large project funded by the Dutch government (Scharff et al., 2003).

In 2001, within a period of 8 weeks three TDL studies were carried out at the Nauerna landfill. Afterwards it became clear that the first TDL campaign was carried out during construction works for which the capping over several hectares had been removed. This result was a lot higher than the others. It was considered an outlier and

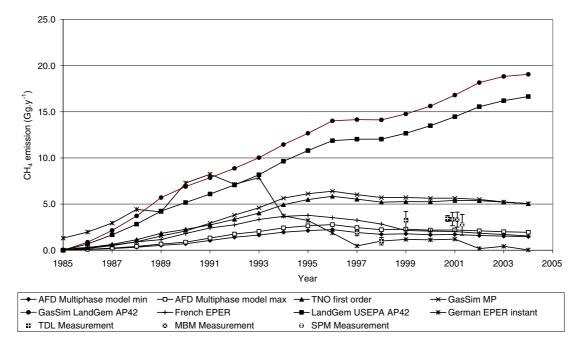


Fig. 4. Modelled and measured methane emission at Nauerna landfill.

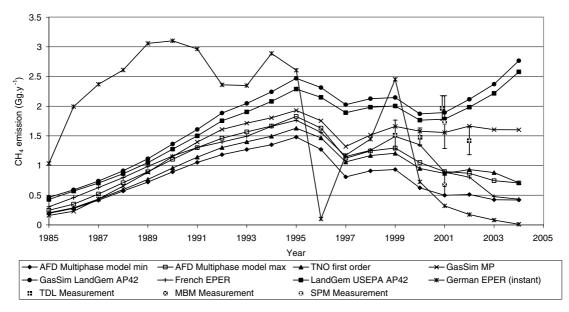


Fig. 5. Modelled and measured methane emission at Braambergen landfill.

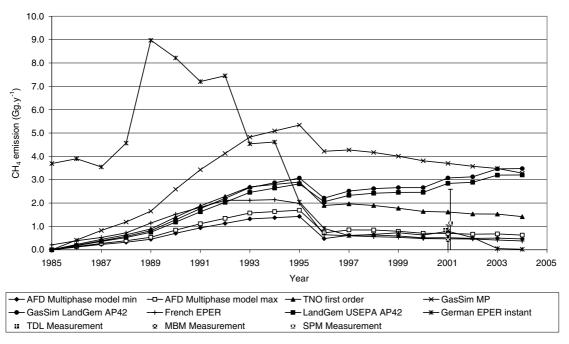


Fig. 6. Modelled and measured methane emission at Wieringermeer landfill.

omitted from the calculation of the average measured emission of the Nauerna landfill for 2001.

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

4.2. First order model (TNO)

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

4.3. Multi-phase model (Afvalzorg)

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

4.4. LandGEM

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

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

4.5. GasSim (UK)

The GasSim multi-phase model gave similar results to the TNO single-phase model on two landfill sites. The Nauerna and Braambergen estimates were in the centre of the range of all the estimates. On the Wieringermeer landfill from 1993 onwards the GasSim multi-phase model gave the highest estimates of all models. This landfill was dominated by commercial waste, especially in the first period of operation. The high GasSim multi-phase estimate on the Wieringermeer landfill can be explained by the organic carbon content in commercial waste. GasSim multi-phase assumes 180 kg of organic carbon in commercial waste. If this organic carbon is assumed to be cellulose, it could potentially yield 168 m³CH₄ · Mg waste⁻¹. That is considerably higher than the assumption for methane generation potential in other models.

4.6. EPER model France

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

4.7. EPER model Germany

Large fluctuations in methane emissions were estimated with the German EPER model. In this paper, the zero order model was used as it was intended. That is, the methane production of an amount of waste landfilled in a certain year is instantaneous or in other words released in that same year. The fluctuations represent the fluctuations in household waste, coarse household waste and commercial waste disposed at the three landfills. If a constant amount of waste of constant composition is disposed in a landfill, the result is the same as for a first-order degradation model. If there is variation over the years, the German EPER model provides the option to estimate methane emissions with a 5 or 10 year average waste amount (Hermann, 2005). At all three landfills, the German EPER model overestimates the methane emission in the first 10 years of operation and underestimates the methane emission in the last 5 years of operation. This effect would have been reduced, but not completely eliminated, if the 10 year average option had been used.

Annex 1 no. 5 (d) of the proposal for an E-PRTR (CEC, 2004) states that "Landfills (excluding landfills of inert waste and landfills, which have been definitely closed before the 16 July 2001 or for which the after-care phase required by the competent authorities according to Article 13 of Council Directive 1999/31/EC has expired)" should report their methane emission. This means that the competent authority should determine that at a certain moment aftercare can be stopped. Only then the reporting obligation ends. A system for determining/deciding on landfill completion has not been established anywhere in Europe. Most EU Member States are considering applying 30-60 year aftercare periods. Guidance for determining methane emissions should therefore be able to estimate emissions up to at least 30 years after disposing of the last Mg of waste. Even with a 10 year average waste input, the German EPER model can only give emission estimates up to 9 years after closure.

4.8. Models versus measurements

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

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

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

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

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

5. Conclusions and recommendations

The intention of the UN-PRTRs and E-PRTRs is to disclose emission data to the general public and the regulators. Landfill operators throughout Europe welcome this as a positive development. We should, however, bear in mind that a number in a database represents the absolute truth in the eyes of the reader. There is no room for explanation as to what the number means or caution about its accuracy. It is good that criteria such as timeliness, completeness, certainty, comparability, consistency and transparency are set for admission of emission data in a publicly accessible database. However, one should check whether these criteria have been met and act accordingly. Otherwise this will inevitably lead to compiling and comparing data with an incomparable origin.

Disregarding extreme results, the highest estimates obtained with the models were five to seven times higher than the lowest estimates. This huge variation in results cannot be considered to be acceptable. It does not indicate that current methane emissions models meet the previously mentioned criteria. It cannot be concluded that the minimum accuracy has been achieved that might be considered necessary before admission in a database can be regarded useful. The authors do not consider it useful or fair to compile and compare data that have such an incomparable origin. It is recommended to build a feature in the publicly accessible database that accommodates reporting of the uncertainty. That should give the user a better impression of what the numbers mean.

The difference in results between the various measurement techniques seems smaller than between the different models. Further development of these techniques and subsequent use of them to tune models may provide a more reliable tool in the near future than current models.

The comparison has been made for three landfills only. These sites may not be representative. It is recommended that more data sets are compared before proposals are made for countries that do not yet have a guidance model. A larger data set may allow for a sensitivity analysis. Determining the most important parameters could lead to minimising the parameters in a guidance model.

It is also recommended that an attempt be made to harmonise the different existing models in order to enable a fair representation of the future methane emission data reported to UN- or E-PRTRs. Harmonisation of models may not necessarily solve uncertainty. However, it may at least result in comparable, consistent and transparent data.

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