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MATHEMATICAL MODELS WHICH PREDICT CH₄ EMISSIONS FROM MSW LANDFILLS – COMPARATIONS

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Abstract: It is well known that municipal solid waste (MSW) landfilling according to legal provisions regarding environmental protection can generate landfill gas (LFG). An ecological landfill which is designed according to the current legislation cannot pollute the groundwater and at the surface of the soil has installed equipment for landfill gas collection. The landfills that are not in accordance with the legal provisions regarding environmental protection will pollute both the groundwater and the atmosphere, including also human agglomerations (cities, communes and villages). Landfill gas contains 50% methane (CH₄), 45% carbon dioxide (CO₂) and 5% nitrogen (N₂) and other gases including trace amounts of non–methane organic compounds. However, the landfill gas quality varies from time and degradation phase, location and gas collection. CH₄ and CO₂ are greenhouse gases being responsible for the global warming effect of the atmosphere, in proportion of 4–5% being emitted from anthropogenic activities. Because of CH₄ high global warming potential (21 times higher than CO₂) appear the need for collection and use as an economic resource. The CH₄ economic value has led to finding mathematical equations for quantification of the quantities emitted from the storage of the waste and moreover, to be able to predict the emitted quantities taking into account that waste generation is a continuous process. There is no relation (generally available) for CH₄ emissions estimation by calculus, thus every country has established as a function of quantities of waste generated and environmental conditions an equation for CH₄ prediction. In the present paper, there are discussed some mathematical equations which estimate the methane gas generation from municipal solid waste landfills, their limits and future applicability.

Keywords: MSW landfills, mathematical models, CH₄ prediction, CO₂

1. INTRODUCTION

CH₄ emissions from municipal solid waste landfills have a significant contribution to climate change phenomena and especially on the global warming of the atmosphere. Environmental authorities around the world are implementing strategies, waste management policies and are establish rules to reduce methane emissions at municipal solid waste landfills. Municipal solid waste landfills are not a punctual source of emission but a diffuse one [1],[2]. Moreover, the landfill gas emission containing greenhouse gases (CH₄, CO₂, N₂O and other gases) [3],[4] varies in time but also in space [1],[2]. Therefore, it is not easy to measure CH₄ emissions from a landfill. In Romania, in order to determine the effectiveness of measures aimed to reduce CH₄ emissions at landfills, it is necessary to quantify CH₄ emissions either at the national level or on a landfill. In 2017, in Romania, there were 43 municipal solid waste landfills in compliance with legislation regarding on environmental protection, of which only three do not have Integrated Environmental Authorization.

Romania, as a member of the E.U., adopted both Kyoto (2000) [3] and Kiev Protocol (2003) [4] regarding the pollutant release and transfer registers through which Romania has to make available to the general public and the government the emissions of CH₄ calculated from municipal solid waste landfills starting with 2007. Among other things, this protocol has imposed to landfills that receive more than 10 tons a day or have a total capacity of 25,000 tons / year to individually determine their CH₄ emissions to make them available to the general public and their national government since 2007. The Commission of the European Communities (CEC) has adapted the European Pollutant Emission Register (EPER) to the E–PRTR (CEC, 2004) in order to comply with the ONU PRTRs Protocol. National Governments also reports to the Intergovernmental Group regarding the climate change (IPCC) compliance with the provisions of the Kyoto Protocol [3]. Thus, a suitable method for estimating CH₄ emissions is needed. In the IPCC Guide, only first order kinetics models are recommended for estimating CH₄ emissions from landfills (MSW). Moreover, IPCC Guide has never intended to be applied to individual waste landfills [1],[2]. At the same time, the first order kinetics models, although are quite accurate, cannot be considered as applicable to any landfill. In Romania there is an obligation for economic operators (juridical or private persons) who manage landfills to transmit to the environmental authorities the amount of CH₄ emitted annually. Data from the database are compared by environmental authorities and the general public with information from other countries regarding the emission level. This will make it possible to compare which landfill is more environmentally friendly. To estimate CH₄ emissions from landfills there are several validated models.

In the present paper, a comparison will be made between the various models used in Romania but also in other countries regarding the estimation of CH₄ emissions from the landfills. At the same time, will be presented another way to estimate CH₄ emissions from landfills by calculation.

2. MATERIAL AND METHOD

– Working models & mathematical equations on estimating CH₄ emissions from municipal solid waste landfills

The most used models which predict methane gas generation over time from a mass of waste use single–phase or multiphase first order decay equation [1],[2]. By quantifying the amount of collected landfill gas with methane content it

is easy to establish the production of CH₄. The problems that arise are related to the emission of CH₄ through the storage cap as well as the migration through certain areas of the deposit. Researches have focused on establishing computational relations that include gas emissions through the collection system and rapid emissions. It was agreed that rapid emissions would be estimated by a pre-determined value of about 10% of the registered quantity. In order to validate a calculation model based on the organic material biodegradation equations, comparisons with emissions data across the entire area are required [1],[2].

Few studies [1],[2] validated mathematical models that use quantification of CH₄ emission on a landfill site based on the measurement of all emissions. In the case of a landfill, when applying a prediction model of landfill gas emission with CH₄ content, appears the problem of the division of waste types registered by the operator according to the waste disposal decision. The problem is to determine the percentage of carbon contained in the waste types stored.

– CH₄ EMISSION ESTIMATION MODELS FROM LANDFILLS

TNO, LandGEM (US–EPA), GasSim (UK Environment Agency and Golder Associates), Afvalzorg (in the Netherlands), EPER (mode France and model Germany), IPCC, and LFGEEN are the models used around the world for prediction of methane generation from landfills and all are first order decay models [1],[2],[5]–[8].

☐ First order model (TNO)

The effect of carbon depletion on waste in time is assessed using a first-order model [9]. Landfill gas formation with CH₄ content in a certain amount of waste is assumed to develop exponentially in time.

The first order model (TNO), used in Netherlands [1],[4], can mathematically be described by the equation (1):

$$a_t = \zeta \cdot 1,87 \cdot A \cdot C_0 \cdot k_1 \cdot e^{-k_1 t} \quad (1)$$

where: a_t – landfill gas formation at a certain time, [m³·year⁻¹]; ζ – dissimilation factor, ($\zeta = 0.58$); A – the amount of waste in landfill, [Mg]; C_0 – the amount of organic carbon in waste, [kg·Mg waste⁻¹]; k_1 – degradation rate constant [year⁻¹] ($k_1 = 0,094$); t – time elapsed since depositing [year].

In order to obtain the methane emissions based on the production prognosis, the following calculation is used (2) [1],[2]:

$$CH_4 \text{ emission} = CH_4 \text{ production} - CH_4 \text{ collected} - CH_4 \text{ oxidation} \quad (2)$$

This calculation can be and has been used in many approaches, such as: the first order model (TNO), multiphase model (Afvalzorg and Gas– Sim) and LandGEM model [1],[2],[6],[10]. It is obvious that the accuracy of the production model is an important factor in this type of approach. The recovery can be measured accurately. The amount of organic carbon in waste was established by laboratory tests.

☐ Multiphase model (Afvalzorg)

The different types of waste contain different fractions of organic matter that degrade at different rates. The advantage of a multiphase model is the typical composition of waste that can be taken into account. In the Afvalzorg multifunctional model there are eight categories of waste and three distinct fractions. For each fraction the landfill gas production is calculated separately. The waste categories, fractions and constant rates used in the multi-phase model Afvalzorg are listed [1],[2]. The multiphase model is a first order model used in Netherlands and can mathematically be described by the equation (3) [1],[2]:

$$a_t = \zeta \sum_{i=1}^3 1,87 \cdot A \cdot C_0 \cdot k_{1,i} \cdot e^{-k_{1,i} t} \quad (3)$$

where: a_t – landfill gas formation at a certain time (with CH₄ content), [m³·year⁻¹]; ζ = dissimilation factor; i – waste fraction with degradation rate $k_{1,i}$ [kg·kg_{waste}⁻¹]; 1.87 – conversion factor at [m³ LFG·kg_{degraded}⁻¹]; A – amount of waste in landfill, [Mg]; C_0 – amount of organic carbon in waste [kg·Mg waste⁻¹]; $k_{1,i}$ – degradation rate constant of fraction i [year⁻¹]; t – time elapsed since depositing [year]. For $\zeta=0.7$, $k_1=0.187$, $k_2=0.099$, $k_3=0.030$ for waste fractions rapidly, moderately, and respectively slowly, degradable [1],[2].

☐ LandGEM US–EPA model

The US EPA model (US–EPA, 2001) is based on LandGEM model. LandGEM model determines the mass of produced CH₄ using the methane generation capacity and the mass of residual waste. LandGEM model can be mathematically described by the equation (4) [1],[2]:

$$Q_{CH_4} = \sum_{i=1}^n k \cdot L_0 \cdot M_i \cdot (e^{-k \cdot t_i}) \quad (4)$$

where: Q_{CH_4} – methane emission rate [m³CH₄·year⁻¹]; k – the methane generation constant (AP42, $k = 0.04$) [year⁻¹]; L_0 – methane generation potential (AP42, $L_0 = 100$) [m³CH₄·Mg waste⁻¹]; M_i – the mass of waste in section i , [Mg]; t_i – the age of the section i [year]. For estimative calculations, the following values can be used: $k = 0.003$ – 0.32 (0.09 – 0.21) year⁻¹, $L_0 = 110$ – 170 m³·Mg waste⁻¹, $\rho_{msw} = 720$ kg·m⁻³ [11].

Sections were considered annual waste quantities removed. The protocols US–EPA (US–EPA, 2004, 2005) mention that the composition of waste used in the model reflects the composition of the waste in the USA. For a landfill the content of non-biodegradable waste can be lowered from waste acceptance rates. LandGEM recommends lowering the inert (non-biodegradable) materials only when the documentation is provided and approved by an environmental authority.

LandGEM ensures the generation of CH₄ at a constant rate both for compliance for CAA (Clean Air Act), and for AP42 (USEPA, 1998). It is recommended to use AP42 values for standard landfills (US–EPA, 2004) [1]–[5],[10]–[13]. Preset values have a high methane generation potential (L₀) of 180 m³CH₄·Mg waste⁻¹.

Once a model has run with the LandGEM program, the methane emission was determined by decreasing the amount of methane collected from the collection system and applying a standard oxidation factor of 10%.

☒ GasSim model

The GasSim model (version 1.00, June 2002) [1],[2] is equipped with two mathematical approaches to calculate a methane emission prognosis (GasSim Manual Version 1,00). The first approach uses the GasSim multiphase equation which is based on a multiphase model described by [1],[2]. The second approach to estimate the CH₄ formation is the LandGEM model which is similar to US–EPA model. The multiphase model requires the introduction of waste into Mg and the specific breakdown during the year on the types of wastes disposed. GasSim and GasSim LandGEM models are used in England, Northern Ireland and Wales.

☒ EPER model France

The French EPER model [1],[2],[6],[10],[12] offers two approaches to estimating CH₄ emissions from landfills:

- Estimates of CH₄ emissions for the landfill cells connected to a landfill gas (LFG) collection system by the landfill operator and the LFG collection efficiency.
- Estimates of CH₄ emissions for the landfill cells not connected to a LFG collection system using a multiphase operating system (ADEME version 15/12/2002) and the LFG collection efficiency.

The methane emission for landfill cells connected to the LFG recovery system can be calculated with the formulas (5) and (6):

$$A = F \cdot H \cdot [CH_4] \quad (5)$$

where: A – the LFG amount collected, [m³·year⁻¹]; F – the extraction rate of LFG, [m³·h⁻¹]; H – operating hours of the compressor every year, [h]; [CH₄] – methane concentration in LFG [%].

A is then corrected at standard temperature and pressure (m³ STP·year⁻¹) taking into account the ambient pressure and temperature at the moment of the gas quality sample. The surface of the cells connected to the LFG collection system and the type of top cover present on that particular cell determine the collection efficiency. For example, an active zone that has no top cover and is connected to a LFG collection system has a collection efficiency of 35% LFG. 65% of LFG will be released into the atmosphere [1]–[9]. The methane production for the cells connected to LFG collection system is calculated by the equation (6):

$$P = \frac{A}{\eta} \quad (6)$$

where: P – methane production, [m³·year⁻¹]; η – recovery efficiency, [%].

The methane emission is then calculated using equation (3).

In the present paper it was used the second approach. The methane emission from landfill can be calculated using a multiphase equation according to ADEME model (7):

$$FE_{CH_4} = \sum_x FE_0 \cdot \left(\sum_{1,2,3} A_i \cdot P_i \cdot k_i \cdot e^{-k_i t} \right) \quad (7)$$

where: FE_{CH₄} – annual production of methane, [Nm³·year⁻¹]; FE₀ – LFG generated potential [m³CH₄·Mg waste⁻¹]; P_i – waste fraction with degradation rate k_i [kg·kg waste⁻¹]; k_i – degradation rate of fraction i [year⁻¹]; t – age of waste, [year]; A_i – normalization factor [–].

The French EPER model assumes an oxidation capacity of the top cover of 10%. The total methane emission is calculated by equation (8):

$$CH_4 \text{ emission} = P(1 - \eta) \cdot 0.9 + FE_{CH_4} \cdot 0.9 \quad (8)$$

where: η – recovery efficiency.

☒ EPER model Germany

The EPER model used in Germany [1],[2],[5–7],[12–14] is a zero order model and can be mathematically described by equation (9):

$$Me = M \cdot BDC \cdot BDC_f \cdot F \cdot D \cdot C \quad (9)$$

where: Me – amount of diffuse methane emission [MgCH₄·year⁻¹]; M – annual amount of stored waste, [Mg·year⁻¹]; BDC – proportion of biodegradable carbon [MgC·Mg waste⁻¹]; BDC_f – proportion of biodegradable carbon converted into LFG (BDC_f = 0.5); F – calculation factor of carbon converted into CH₄ (F = 1.33) [Mg CH₄·MgC⁻¹]; D – collection efficiency, (in collection active system = 0.4, without collection system = 0.9, with collection active system covered = 0.1); C – methane concentration in LFG [%] (C = 50 %).

3. CHARACTERIZATION OF PRESENTED MODELS

In table 1 there are presented the characteristics of the models from literature that estimate the landfill gas [1],[2],[5–7],[12–14].

Table 1. Characteristics of the models from literature that estimate the landfill gas

Model	Country where it is applied	Comment
TNO	Holland	The TNO single-phase first order model is a very simple model. It has a limited number of parameters and is therefore easy to use. The results followed a pattern that can be recognized in other first order models. However, TNO estimates were higher than the estimates of the multi-phase Afvalzorg model [1],[2].
Multiphase Afvalzorg	Holland	The results of the estimates on the deposits that participated in the experiment were lower than the TNO [1],[2].
GasSIM	England. Northern Ireland, Wales	The GasSIM multiphase model gave similar results to the one-phase TNO model on two MSW landfills. On another deposit, it calculated the highest estimates in comparison to all models [1],[2].
GasSIM LandGEM	England. Northern Ireland, Wales	Estimates of methane emission on MSW deposits in the study were higher than all the estimates obtained by other models [1],[2].
LandGem SUA-EPA	USA	Estimates of methane emission on MSW deposits in the study were higher than all the estimates obtained by other models [1],[2].
EPER ADEME	France	The French EPER estimates were the smallest and were comparable to those of the multi-phase Afvalzorg. This is mainly because much of the waste has been attributed to methane-free category 3. The French model mentions three fractions and three values – k – for each category of waste [1],[2].
EPER	Germany	Large fluctuations in methane emissions with the German EPER model were estimated. The model was applied to three landfills. The German model overstated methane emissions in the first 10 years of operation and underestimated methane emissions over the last 5 years of operation [1],[2].

All models presented are included in computational programs. Besides the MSW quantities by types that arrived in the body of the deposit, further information is needed for calculations on CH₄ emission estimation.

4. A NEW CALCULATION METHOD FOR ESTIMATING THE CH₄ EMISSIONS FROM ROMANIA MSW LANDFILLS

This was published by Vieru D., ph.d. student at Politehnica University of Bucharest, in Atmospheric and Climate Sciences Journal, 2017 [15],[16]. The method is based on several findings such as:

- MSW are stored after sorting in the body of the random repository so that all types of waste come into contact;
- based on the recommendations of the IPCC expert group, six types of wastes have been identified that have a certain degradation rate expressed by the factor k;
- in the calculation year, CH₄ emission is due to the amount of MSW degraded to COD (organic dissolved carbon);
- it is possible to determine the percentage composition of the waste in the storage body with the data collected from the actors involved in the waste management system. The composition can be maintained for even 5 years or can be changed annually depending on socio-economic conditions;
- waste disposal is done in the 12-month calendar year;
- calculation year of the MSW amount degraded at COD is 6 months shorter compared to the calendar year;
- estimating the amount of MSW degraded in a calculation year that generates CH₄ emissions is made beginning with the end of the second year of waste disposal;
- a non-degraded waste remains in the landfill every year and it will be taken into consideration in the next year of calculation;
- CH₄-containing storage gas (LFG) will collect all types of gas in its incipient phase on its way to the landfill cap;
- a NOMOGRAM can be made for each waste landfill;
- landfills that collect CHF containing CH₄ must submit the information to the environmental authority;
- landfills that do not collect LFG's need to know when to install the collection system in order to stop paying environmental taxes.

The mathematical equation that estimates the CH₄ emission at landfills in Romania can be written as:

$$\text{CH}_4 = Q_{\text{MSWdegrad.T}} \times \text{TDOC}_{\text{dissolved.T}} \times \text{DOC}_f \times \frac{16}{12} \times F \times F_r \quad [\text{Gg/year}]_T \quad (10)$$

$$Q_{\text{MSWdegrad.T}} = (Q_{\text{MSW.T}} + Q_{\text{MSW.T-1}}) \times [1 - \exp(-Kt)] \quad [\text{Gg}] \quad (11)$$

$$Q_{\text{MSWdegrad.T}} = (Q_{\text{MSW.T}} + Q_{\text{MSWundegrad.T-1}}) \times [1 - \exp(-Kt)] \quad [\text{Gg}] \quad (12)$$

$$Q_{\text{MSWundegrad.T}} = (Q_{\text{MSW.T}} + Q_{\text{MSW.T-1}}) - Q_{\text{MSWdegrad.T}} \quad [\text{Gg}] \quad (13)$$

$$\text{TDOC}_{\text{dissolved.T}} = \sum(A + B + C + D + E + G) \quad [\text{Gg}] \quad (14)$$

$$A = Q_{\text{MSWdegrad.T}} \times \%Q_{\text{MSWbiodegrad.T}} \times k_o \quad [\text{Gg}] \quad (15)$$

$$B = Q_{\text{MSWdegrad.T}} \times \%Q_{\text{MSW(G+P)degrad.T}} \times k_1 \quad [\text{Gg}] \quad (16)$$

$$C = Q_{\text{MSWdegrad.T}} \times \%Q_{\text{MSW(H+C+text.)degrad.T}} \times k_2 \quad [\text{Gg}] \quad (17)$$

$$D = Q_{\text{MSWdegrad.T}} \times \%Q_{\text{MSW(wood+straw)degrad.T}} \times k_3 \quad [\text{Gg}] \quad (18)$$

$$E = Q_{MSWdegrad.T} \times \%Q_{MSWsludg.degrad.T} \times k_n \quad [Gg] \quad (19)$$

$$G = Q_{MSWdegrad.T} \times \%Q_{MSWwind.degrad.T} \times k_4 \quad [Gg] \quad (20)$$

$$TDOC_{dissolved.T} = \frac{TDOC_{dissolved.T}}{Q_{MSW \text{ taken into consid.T}}} \quad [Gg] \quad (21)$$

$$Q_{MSW \text{ taken into consid.T}} = Q_{MSW.T} + Q_{MSWundegrad.T-1} \quad [Gg] \quad (22)$$

We have applied the calculation steps for CH₄ emission estimation at 14 waste landfills on the Romanian territory and we have drawn up the chart on the greenhouse effect evolution. In the following figures are presented the greenhouse effect evolution at the Chitila – Rudeni – Iridex warehouse, between 2000 – 2016, and the greenhouse effect evolution at the Satu Nou – Baia Mare warehouse, Maramures County, between 1991 – 2011.

Table 2. Quantities of gas collected at the IRIDEX landfill – 25.11.2018

	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016
CH4 emitted, [Gg]	0	0.11	0.24	0.54	0.71	0.94	1.653	1.79	2.51	3.01	3.21	0.88	0.90	0.57	0.63	0.74	0.35
CH4 collected, [Gg]	0	0	0	0	0	0	0	0	0	0	0	5.64	5.51	5.09	5.36	6.97	5.79
CO2 equivalent, [Gg]	0	2.24	5.00	11.34	14.80	19.78	34.72	37.48	52.63	63.16	67.31	18.31	18.78	12.069	13.14	15.46	7.43

For landfill Chitila–Rudeni–Iridex: $[CH]_{4emitted} = [CH]_{4generated} - [CH]_{4collected} [Gg]$.

Table 3. Quantities of wastes for MSW deposited at Landfill (MSW) Chitila – Iridex, 8 environmental Region, Bucharest – Ilfov, Romania, for the period 2000÷2016

years of storage																
2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016
quantities of waste (MSW) stored, [Gg]																
43.5	361.2	361.7	309.4	349.5	384.5	368.0	245.5	448.7	434.9	425.5	361.0	371.6	338.30	306.3	272.6	310.5
m [number of months], values, according Nomogram deposit																
0	9.0	7.0	14.0	13.0	12.0	11.0	9.0	8.0	7.0	10.0	7.0	9.0	11.0	11.0	8.0	10.0
CH ₄ [Gg], collected																
0	0	0	0	0	0	0	0	0	0	0	5.640	5.513	5.086	5.363	6.968	5.790

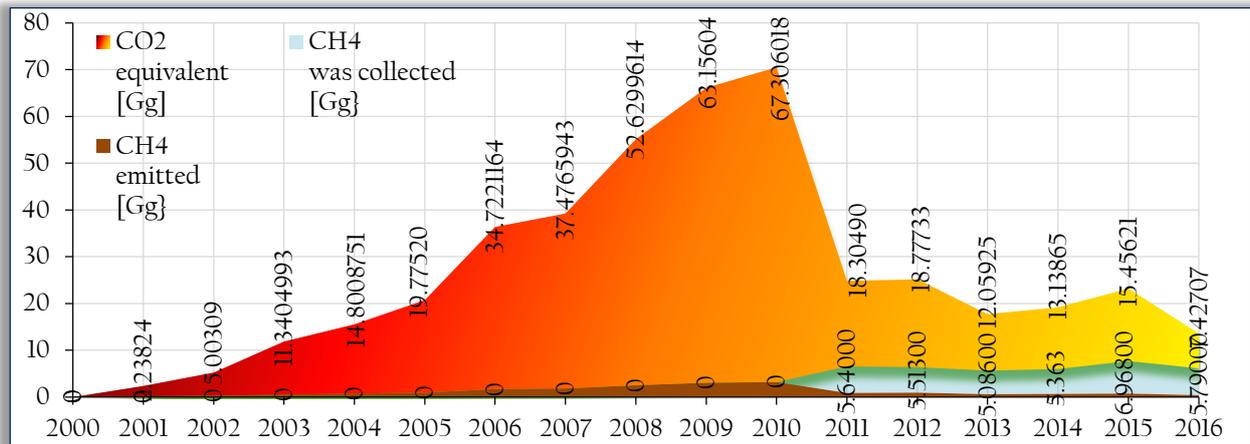


Figure 1 – Greenhouse effect evolution at the Chitila – Rudeni – Iridex warehouse in Bucharest between 2000 and 2016

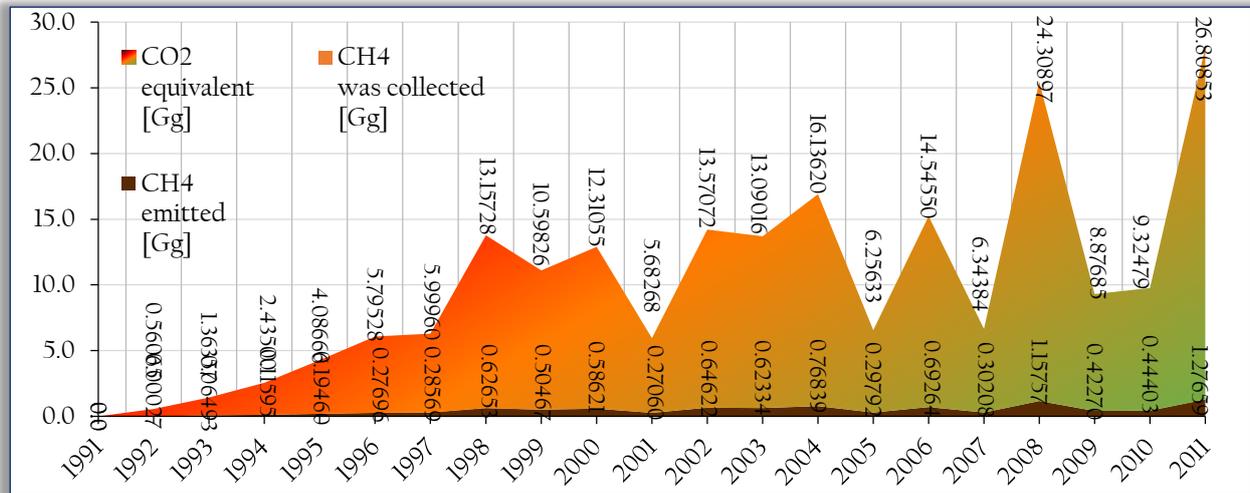


Figure 2 – Greenhouse effect evolution at Satu Nou – Baia Mare warehouse, Maramures County between 1991 and 2011

Table 4. Quantities of wastes for MSW deposited at New Village – Baia Mare, Maramures District, years of depositing period, 1991–2011

During storage (storage Baia Mare –New Village)																				
1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011
Quantity of waste (MSW) stored in the body of the deposit [Gg]																				
85.6	86.3	87.5	89.3	88.8	89.7	90.6	93.5	92.9	106.8	105.0	122.2	110.0	135.7	126.3	122.5	100.3	91.2	102.8	98.24	90.0
m – number of months fixed annual waste degradation, according nomogram																				
0	10	9	8	7	7	18	14	15	14	18	14	14	13	7	14	18	13	7	7	12

5. CONCLUSIONS

- CH₄ generated by MSW landfill is an energy resource. Its collection is beneficial for the protection of the environment and for human health. Figure 1 shows the spectacular decrease in the greenhouse effect by CH₄ source collection.
- The proposed calculation relationship can determine when the amount of CH₄ formed can be collected. It is beneficial information for investors.
- CH₄ collection investment costs are high but solutions can be found to achieve this goal. One thing is for sure that the state must be involved especially that the CH₄ collected quantities can generate electricity.
- The proposed relationship provides data that are reliable, comparable, consistent and transparent.
- Waste management needs to get new valences, especially as the tendency is to increase the amount of CH₄ generated. In addition, solutions for withholding odour-related smells at source will be found.

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