

Modeling the final phase of landfill gas generation from long-term observations

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Abstract For waste management, methane emissions from landfills and their effect on climate change are of serious concern. Current models for biogas generation that focus on the economic use of the landfill gas are usually based on first order chemical reactions (exponential decay), underestimating the long-term emissions of landfills. The presented study concentrated on the curve fitting and the quantification of the gas generation during the final degradation phase under optimal anaerobic conditions. For this purpose the long-term gas generation (240–1,830 days) of different mechanically biologically treated (MBT) waste materials was measured. In this study the late gas generation was modeled by a log-normal

distribution curve to gather the maximum gas generation potential. According to the log-normal model the observed gas sum curve leads to higher values than commonly used exponential decay models. The prediction of the final phase of landfill gas generation by a fitting model provides a basis for CO₂ balances in waste management and some information to which extent landfills serve as carbon sink.

Keywords Gas sum potential · Log-normal modeling · Carbon sequestration

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Introduction

The fate of municipal solid waste plays a crucial role within the global carbon cycle. In the discussion about climate change, “waste to energy” and resource recovery were recognized as potential strategies to reduce carbon dioxide emissions (Hellweg et al. 2005). Regarding the landfill gas, there is a twofold goal, economic utilization of its energy content and reduction of negative impacts on climate (Ackerman 2000). According to Morris (2010) about 64% maximum of the landfill gas can be captured and converted. Due to the relatively low energy conversion efficiency and the long aftercare periods alternative concepts were aimed for. In accordance with the European multi-barrier concept Austrian and German

legislation stipulated a pretreatment of municipal solid waste prior to landfilling in order to prevent relevant emissions after final disposal. Besides incineration, the mechanical biological treatment (MBT) has become a main strategy in waste management (Tintner et al. 2010). The mechanical treatment focuses on the separation of plastics and big particles (metals, stones). The biological treatment of waste effects the degradation of waste organic matter under controlled aerobic conditions, the reduction of leachate and greenhouse gas emissions (Nosanov and White 1975), which leads to a shortening of the aftercare period. The output material of the biological treatment is characterized by low biological reactivity and a low calorific value (Binner 2003; Zach et al. 2000). For these parameters limit values were established. The reactivity of waste materials can be determined by biological tests. The microbial activity under aerobic conditions is determined by the respiration activity. Usually this activity is quantified using the oxygen uptake for 4 days. The gas generation under anaerobic conditions can be tested either in a liquid phase (fermentation test) or in a solid phase (incubation test) during a period of 21 days.

Indeed, MBT materials that comply with the limit values for landfilling, still feature residual reactivity causing weak degradation under anaerobic conditions in the landfill. The lean methane emissions can be abated by means of cover layers containing methanotrophic bacteria. It can be hypothesized that after a certain time under anaerobic conditions the microbial activity decreases to zero. The velocity depends on the composition of the input mixture (Hoeks 1983).

The questions arise, for what time span residual activity is relevant and which amount of organic matter remains in the landfill in the long term. There are different models to answer these questions. The classical approach (Tabasaran and Rettenberger 1987) uses an exponential decay model (first order chemical reaction) to explain the landfill gas generation quantitatively:

$$s(t, A, m) = A \cdot (1 - \exp(-t/m)) \quad (1)$$

In this equation “*s*” is the gas sum generation till day *t*, *A* is the maximum possible amount of produced gas (gas generation potential) and *m* is a constant of decay (mean value of time in days). The parameters *A*, and *m* are estimated from original data or taken from literature.

There are manifold derived modifications of this model, such as a considerable number of delayed exponential decay processes, as integrated in the IPCC Tier 2 model. However, the results do not differ much (Pipatti and Svardal 2006). Other approaches simulate the biochemical reactions and transportation processes within the landfill (Ricken and Ustohalova 2005; Ustohalova et al. 2006). These models require a sound knowledge of the genesis and the inner structure of the landfill. They are based on more detailed information on specific processes in landfills to model gas generation (Findikakis et al. 1988) or nitrogen dynamics (Mostbauer and Heiss-Ziegler 2005).

The first-order approach is useful to estimate the gas generation during the first phase of organic matter degradation in municipal solid waste. In terms of the long-term carbon storage in the landfill the final phase of the gas release is more interesting.

The objective of this paper is the prediction of the gas generation during the final phase of waste degradation. Based on long lasting gas generation tests for different MBT materials the best fitting model that describes the final tails of the gas sum curve adequately, should be identified. For this purpose the classical first-order approach was compared to alternative models.

Materials and methods

Ten long-term gas generation tests were performed. The materials were taken from five different MBT plants. Table 1 compiles the materials and process operation in the plants. All materials were shredded wet to a particle size of <20 mm. The samples were selected according to input materials, process operation and stages of degradation.

The microbial gas generation of a solid waste sample under optimized anaerobic conditions (Binner and Zach 1999) is determined according to the specification of the Austrian Standard Institute (2004b), e.g. incubation test during a period of 21 days (GS₂₁). Therefore the water content is optimally adjusted. A constant temperature of 40°C is maintained which corresponds to optimal temperature conditions for mesophilic anaerobic microbial degradation (Schlegel 1992). Extreme pH values, high salt concentrations as well as toxic substances can cause inhibiting effects on microorganisms. Therefore the

Table 1 Input materials and process operation in the five plants

Plant	Input material	Rotting system	Sieving steps
A	MSW, SS	2 wk CS, 6–8 wk RP	80 mm CS, RP, 25 mm LF
B	MSW	4 wk CS, 7 wk RP	160 mm CS, 20 mm RP, LF
C	MSW	5 wk CS, 10–30 wk RP	25 mm CS, RP, LF
D	MSW, SS	3 wk CS	100 mm CS, LF
E	MSW, SS	3 wk CS, 11 + 5 wk RP	100 mm CS, 25 mm RP, LF

MSW municipal solid waste, SS sewage sludge, wk weeks, CS closed system, RP ripening phase, LF landfill fraction

electrical conductivity and pH were measured at the beginning and at the end of the test to identify such impacts. The respiration activity (RA_4) was measured as a control of the degradability of the material.

Lag-phases are not included in the 21-day period according to Austrian Standards Institute (2004b). The day, when 33 % of the maximum mean daily value is reached, is taken as the end of the lag-phase. The calculation of the gas sum during a period of 21 days (GS_{21}) starts the next day. The long lasting tests were carried out over several hundred days as described in the results section. The logging of the gas generation was done twice a day at the beginning of the measurement and once a week in the later phase.

Some parameters that describe the degradation of waste organic matter were measured as well, e.g. loss of ignition, total organic carbon, total nitrogen contents (Binner 1996). The parameters and the applied methods are listed in Table 2.

The data of the gas generation test are on a daily/weekly base, to recognize irregularities that may be incompatible with the model assumptions and long-term, to identify a sufficiently long segment of the final phase. For the main phase, the gas generation is not predictable. However, during the final phase (defined below), all samples showed a comparable pattern of gas generation (Fig. 1).

In this study the final phase of gas generation is defined by a small relative growth rate “ r ”. The final phase is reached at a moment, from which on the relative daily growth rate of the gas-sum-curve after the lag-phase is less than 1 % = 0.01 (days^{-1}). For the computations, “ N ” is defined as the number of measurements, the measurements are indexed by “ x_i ” for the gas sum at day “ t_i ”, where $i = 1, \dots, N$, and the growth rate “ r ” is computed by the formula (the index “ ii ” denotes the successor of i):

$$r = \frac{1}{x_i} \frac{x_{ii} - x_i}{t_{ii} - t_i} \quad \text{for } (1 \leq i \leq N) \quad (2)$$

Next, the data of this final phase were used to find optimal fits of the exponential and the sigmoid log-normal models. These models are easily applied, as also the log-normal distribution is available in MS Excel[®]. Therefore, all relevant computations for this model can be performed with spreadsheets. (Parameter fitting uses Excel’s Solver add-in.) If μ and σ are the mean value and the variance of a normally distributed random variable $y = \ln(t)$, then in Excel the distribution function of the log-normal distribution for x is defined as $f(t) = \text{LOGNORMVERT}(t; \mu; \sigma) = \text{NORMVERT}(\text{LN}(t); \mu; \sigma; 1)$. Here, “ $f(t)$ ” is defined by:

Table 2 Parameters and the corresponding methods of determination

Parameter	Method according to
Loss of ignition (LOI)	Combustion between 105 and 550°C
Total organic carbon (TOC)	Compost ordinance (BGBl. Nr. 292/2001)
Total nitrogen (TN)	Compost ordinance (BGBl. Nr. 292/2001)
Electrical conductivity (EC)	10 g dried sample in a ratio 1:10, elution time 3 h, elution medium: H ₂ O
pH-value	100 g fresh sample in a ratio 1:10, elution time: 2 h, elution medium: H ₂ O
Gas sum (GS_{21})	Austrian Standards Institute (2004b)
Respiration activity (RA_4)	Austrian Standards Institute (2004a)

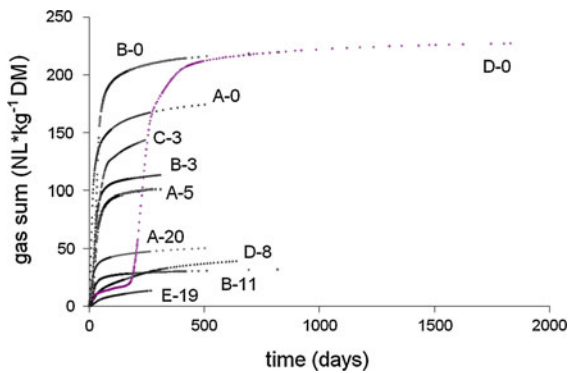


Fig. 1 Cumulative gas generation of ten long-term incubation tests; NL volume (L) indicated for standardized conditions (0°C , $1,013$ mbar)

$$f(t) = \int_{-\infty}^t \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{(\ln x - \mu)^2}{2\sigma^2}\right) dx \quad (3)$$

In Excel notation, the model function “ m ” for gas generation up to day t is

$$m(t; A; \mu; \sigma) = A \cdot \text{LOGNORMVERT}(t; \mu; \sigma) \quad (4)$$

Here, A , μ and σ are the parameters that are to be determined from the data. The parameter A is the maximum gas generation during the final phase. It has the same meaning, as in the exponential decay model, if this model is applied to the final phase. In waste management the log-normal distribution already was used for modeling the heterogeneity of landfills (Zacharof and Butler 2004a, b).

In addition we considered materials in their final phase (D-8 and E-19) without sigmoid characteristics. For these materials we rescale time; $t_0 \geq 0$ is the best estimation for the duration prior to observation. This leads to the following equation.

$$m(t; A; \mu; \sigma; t_0) = A \cdot \text{LOGNORMVERT}(t - t_0; \mu; \sigma) \quad (5)$$

Data fitting

Data fitting to obtain the model parameters uses a weighted form of the least squares method and weights the observations x_i by the time span, which they describe.

$$w_i = \frac{t_{ii} - t_i}{t_N} \quad (6)$$

The reason for the weights is the different frequency and importance of the measurement at the

main and later phases (of the considered final phase of gas generation). At the main phase measurements are more frequent, but the measurements are less important for the assessment of the final phase.

The parameters A , μ and σ (resp. first-order decay's A , k) and t_0 are found by minimizing the weighted sum of squares over the data x_i , using the above weights w_i :

$$\sum w_i \cdot (x_i - m(t_i; A; \mu; \sigma; t_0))^2 \quad (7)$$

with the constraint $t_0 \geq 0$. Fitting is done with the Solver in Excel.

For comparison, data were also fitted to the exponential model, using the same definition of the “final phase” and the same weights in order to effect better matching.

Results and discussion

Table 3 lists characteristics of the ten materials. The materials covered a wide range of reactivity.

Compared to the gas sum (GS_{21}) the initial respiration activity (RA_4) of the materials B-3 and B-11 were inhibited. The reasons were probably drying out of the material and a limited oxygen supply during the biological process in the MBT plant. Therefore aerobic degradation remains incomplete over the four-day-duration of the standardized test. This effect does not influence our result here as the anaerobic gas generation is not inhibited by lacking oxygen during the biological treatment. A lack of water during the MBT process does not affect the anaerobic test due to the long incubation period.

Compared to the respiration activity (RA_4) the gas sum (GS_{21}) of the material D-0 was too low. The material acidified strongly and the lag phase exceeded the test duration of 21 days.

Table 4 compares the material characteristics for MBT waste at the beginning and at the end of the tests. During the incubation test the pH-value generally rises, the electrical conductivity, LOI, TOC and TN decrease. The results of pH and electrical conductivity demonstrate the regular progress of the experiments. Depending on the initial TOC content and the reactivity of the samples their decrease during the anaerobic degradation is different. Furthermore the heterogeneity of the material has to be considered. The variability of MBT waste was measured by taking 34 samples of one MBT output charge ready for

Table 3 Age of the material at the test start, test duration and initial reactivity parameters (respiration activity RA_4 and gas sum GS_{21}); extreme values in bold characters; *DM* dry matter, *NL* volume (L) indicated for standardized conditions (0°C, 1,013 mbar)

Sample	Age (weeks)	Test duration (days)	Initial RA_4 (mg O_2g^{-1} DM)	Initial GS_{21} (NL ^a kg ⁻¹ DM)
A-0	0	502	51.9	109.8
A-5	5	310	20.0	53.0
A-20	20	502	21.2	29.5
B-0	0	819	55.2	93.7
B-3	3	309	16.0	67.4
B-11	11	819	3.9	16.9
C-3	3	244	Nd ^a	58.4
D-0	0	1,830	53.0	21.6
D-8	8	639	5.1	7.0
E-19	19	271	2.6	2.7

^a *Nd* not determined

landfilling. The standard deviation of the TOC covered a confidence interval between 0.83 and 1.36 ($\alpha = 0.05$).

Even starting at different levels of reactivity the final phase of the gas generation shows a similar pattern. The cumulative curves are displayed in Fig. 1. The process operation has the strongest impact on the slope of the curve during the main phase. Samples D-8 and E-19 were already in the final phase, when collected. This is reflected by higher gas generation ratios as the “observed main phase” is small compared to the actual initial generation and the final phase. For

the materials B-11 and A-20 with a comparable gas generation a main phase can still be distinguished in the measured data.

By modeling with Eq. 5 the gas generation of the final phase of all samples could be described. As the materials cover the wide range of MBT materials, we can assume that the model we found can be used for all kinds of these materials. In all samples the first order decay (exponential model) underestimated the gas generation. The lognormal function modeled the data much better. Figure 2 compares the optimal fit for two models of the final phase of sample B-11, log-normal

Table 4 Material characterization at the beginning and the end of the test (*DM* dry matter)

Sample	Unit	A-0		A-5		A-20		C-3		E-19	
		Start	End	Start	End	Start	End	Start	End	Start	End
pH-value		7.0	7.2	7.4	7.6	7.7	8.2	6.8	7.4	7.8	8.5
EC	(mS/cm)	3.0	2.5	3.7	1.9	3.0	2.3	1.1	0.5	3.2	2.4
LOI	(% DM)	52.7	40.3	41.9	28.5	37.4	32.7	43.3	29.1	34.8	31.4
TOC	(% DM)	29.2	22.9	23.1	16.0	21.0	16.6	23.3	16.2	20.1	16.8
TN	(% DM)	2.0	1.6	1.2	0.9	1.3	1.4	1.0	1.0	1.2	1.1
Sample	Unit	B-0		B-3		B-11		D-0		D-8	
		Start	End	Start	End	Start	End	Start	End	Start	End
pH-value		8.1	9.0	8.3	9.2	8.3	8.7	6.6	8.6	7.9	8.2
EC	(mS/cm)	3.7	1.8	3.6	2.6	2.8	1.8	4.1	Nd ^a	2.9	2.0
LOI	(% DM)	58.2	41.4	54.3	31.1	35.7	22.9	55.7	28.3	36.3	39.2
TOC	(% DM)	31.5	24.6	30.5	17.3	19.0	12.5	29.8	15.1	23.8	23.4
TN	(% DM)	1.2	1.3	1.1	0.8	1.1	1.0	0.9	0.7	1.1	0.8

^a *Nd* not determined

Table 5 Duration in days, carbon release in % DM, gas generation in NL*kg⁻¹ DM; DM dry matter, NL volume (L) indicated for standardized conditions (0°C, 1,013 mbar)

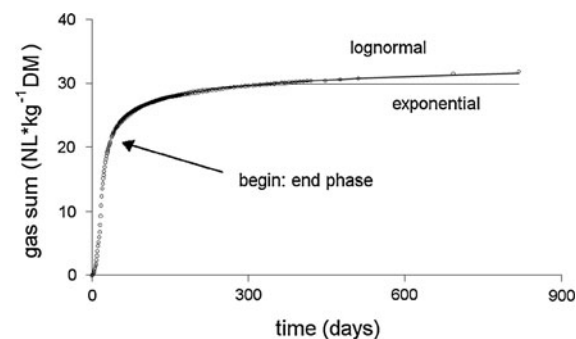
Curve number	A-20	B-0	C-3	B-3	A-5	A-0	B-11	D-0	D-8	E-19
Observed lag-phase (days)	4	14	8	11	10	2	9	184	8	6
Duration of main phase	33	52	62	42	52	30	40	262	58	63
Gas generation of lag phase	2.2	15.1	4.1	9.8	5.3	5.1	2.4	23.0	0.6	0.2
Gas generation of main phase	32.6	153.3	106.8	79.8	73.6	120.6	19.6	134.6	14.6	6.6
Total gas generation (including final phase)	60.6	228.1	193.0	117.7	103.9	189.3	33.7	229.0	43.9	17.0
Ratio: gas generation final phase/total phase	0.43	0.26	0.74	0.24	0.34	0.36	0.35	0.31	^a	^a
Parameters of the log-normal distribution										
A = gas generation during final phase	25.8	59.7	143.7	28.1	25.0	63.6	11.7	71.5	28.7	10.2
μ	5.5	4.3	3.4	3.7	3.6	4.6	4.7	4.4	5.4	5.2
σ	2.6	2.2	1.0	2.0	1.7	2.2	2.2	1.6	1.0	0.9
t_0 (days)	0	0	0	0	0	0	0	0	37	41
Parameters of the exponential model										
A = gas generation during final phase	14.7	49.1	140.3	22.2	21.3	46.5	7.9	66.4	26.8	5.5
m (days)	102	92	41	42	44	83	76	128	254	5

^a As the samples were taken after the beginning of the main phase ($t_0 > 0$), the ratio between main and final phase cannot be calculated

and exponential decay model. As can be seen, the log-normal model fits well to the data, even during the main phase. However, this main phase was not used for parameter estimation. The log-normal model results in the gas generation potential of $33.7 = A + 22.0$ ($22.0 =$ gas sum during the main phase), while the first exponential model leading to 29.9 underestimates it. This is even less than the last observed data.

The results of the modeling calculations are presented in Table 5.

The parameters of the log-normal distribution can be calculated from the samples: $\mu = 4.5$ (standard deviation 0.8) and $\sigma = 1.7$ (standard deviation 0.6).

**Fig. 2** Model fitting for the sample B-11.

The authors want to point out that this variability of the parameters refers to the final gas production of different materials under optimal conditions. There is still research to be done about the variability under sub-optimal conditions, which derives functional dependencies between e.g. temperature and the parameters of the log-normal model, or which explores the effect of imperfect mixing of materials. Such research needs additional data.

Falsification of the exponential model for the tail of the gas sum curve is observed for all data. The log-normal model thus amplifies the conventional exponential model that focuses on the early phase of the gas generation process. It can be concluded that a simple first order chemical reaction is inadequate to model the complicated interactions and transport processes within the matrix of landfilled waste.

The exponential model underestimates the gas generation potential of landfills. The traditional explanation for this effect is the poor choice of tabulated parameter values which could be corrected by the improvement of the parameters (Schachermayer 2006). However, according to this paper the underestimation is a systematic problem. It warrants the refutation of the exponential approach and the enhancement of all related models, including those of IPCC, by a log-normal approach.

Conclusion

The paper presents a new modeling approach to estimate the long term gas generation of landfilled MBT wastes and improves the conventional exponential model. Long term incubation tests enabled to model the maximum gas generation under anaerobic conditions. The investigation covered a wide range of waste materials out of biological treatment processes. The variety was intended and showed that the behavior of all materials leads to the same curve type. The log-normal model was found to fit the gas generation during the final phase, where daily gas generation falls below 1 % of the gas sum generated up to this moment. The parameters of the log-normal model are A , the asymptotic value of gas generation, μ and σ , whereby $\mu \approx 4.5 \pm 0.8$ and $\sigma \approx 1.7 \pm 0.6$. An important advantage of the log-normal model is the possibility to gain information about the long term gas generation even of samples, where the main phase of gas generation already happened before sampling ($t_0 > 0$) as it can be applied to all stages of degradation.

For MBT technology this modeling of the gas generation potential can be of further interest for estimating the amount of carbon fixed in the landfill under anaerobic conditions. The carbon sequestration potential of landfilled MBT material is remarkable. The log-normal model can be used for the quantification of this potential. Fitting of the curve to measured gas generation was the first step. Future research will concentrate on the question about the period of time that is necessary for the determination of μ and σ . It will provide the basis for the determination of the carbon fixation in any MBT sample.

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