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COMPARING MODELLED, RECOVERED AND GENERATED GAS IN A MSW LANDFILL UNDER LEACHATE RECIRCULATION

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ABSTRACT: in-situ stabilization of waste bodies can be achieved by the infiltration of water or recirculation of leachate into the landfill, which is thought to enhance the microbial degradation of waste organics by (re-)moisturizing dry zones and flushing out metabolic products of organic matter decay. The success of in-situ stabilization should reflect in initially accelerated and thereafter reduced rates of anaerobic waste organic matter decay rates. This paper compares the methane generation that was modelled using the Afvalzorg multiphase model without the added effect of leachate recirculation with actually extracted methane in the landfill and gas generation on sampled wastes following five years of leachate recirculation on a Dutch landfill. Laboratory incubations revealed a methane potential between 0.03 kg CH4/t dw and 15.8 kg CH4/t dw for 365 days. Clear trends with respect to depth, moisture content, total organic carbon or share in hard plastics did not emerge as overall waste heterogeneity was high and likely obfuscated the correlation analysis. The results showed a recovery efficiency of 30.4% for 2021, with 0.07 kg CH4/t dw for the recovered methane and 0.23 kg CH4/t dw for the predicted methane in compartment 3. The average methane potential measured in the laboratory was almost twice as high as the remaining methane potential predicted for the period of 2021-2093. The discrepancy could be due to (i) enhanced waste degradability as a result of five years of recirculation, (ii) enhancing effects of material perturbation during sampling and/or (iii) impeded on-site methane generation and gas and water transport limitations due to presence of plastics. Overall, the laboratory incubations demonstrate a significant potential for waste biodegradation still residing in the waste.

Keywords: In-situ stabilization, leachate recirculation, landfill

1. INTRODUCTION

Landfills play an important role in waste management, presenting several benefits but also drawbacks (Zhang et al., 2018). The objective of sanitary landfilling is to minimize the release of emissions into the environment through waste isolation and by preventing the infiltration of water, which reduces the generation of leachate. However, the anaerobic environment typically prevailing in waste limits the biodegradation of waste (Zhang et al., 2019), resulting in a long period (possibly hundreds of years) before the waste reactivity subsides (van Turnhout et al., 2018). The point at which the landfill can be deemed stable is when the biodegradable materials within it have reached mineralization, resulting in minimal to no generation of gas or leachate, and the natural settling of the site's surface has come to a halt (Zhang et al., 2023). The success of in-situ stabilization should reflect in initially accelerated and thereafter reduced rates of anaerobic waste organic matter decay rates. Different techniques can be used to accelerate the stabilization of waste and reduce the aftercare period. One such technique is leachate recirculation, intended to moisturize dry zones and to flush components that may block reactivity (Lee et al., 2022; van Turnhout et al., 2018). However, a homogeneous distribution of moisture is still a challenge in landfills (Fei et al., 2016; Gebert et al., 2022).

The degradation of organic matter by microorganisms, under anaerobic conditions, releases methane (CH_4) and carbon dioxide (CO_2), resulting in a typical composition of landfill gas of 55-60% CH_4 and 40-45% CO_2. Methane can be recovered and used as an energy source (Kamalan et al., 2011), providing economic benefits for the operator of the landfill. One of the methods to assess the potential for recovering landfill gas is through a landfill gas model (Toha & Rahman, 2023). A model utilizes site-specific information on the waste inventory to estimate the generation of landfill gas over time, including methane (Afvalzorg, n.d.). Another method to estimate the gas potential in the waste is through laboratory experiments. Several studies have performed those experiments on fresh waste samples, but also natural materials such as sediments, quantifying the generated gas under aerobic and/or anaerobic conditions (Gebert et al., 2006, 2011, 2019; Zander et al., 2022).

In this paper, we focus on a municipal solid waste landfill in The Netherlands that has been treated by leachate recirculation since 2017 to stabilize the waste body. After five years of leachate recirculation, with another five years ahead, it was of interest to determine how far the landfill has already stabilized in terms of methane generation. This paper aims to compare the modeled methane generation with the actually recovered methane and with the methane generation by waste samples as measured in the laboratory. Further, we aimed to investigate the heterogeneity of the waste's methane potential and its relation to waste properties.

2. MATERIALS AND METHODS

This item is divided into four sections, starting with the description of the site and continuing with the explanation of the methane generation experiment, model estimates and recovered methane. Considering that the waste was sampled at the end of 2020 and the beginning of 2022, the year 2021 was selected as a reference to compare laboratory methane generation to modelled and recovered methane.

2.1 Description of the landfill

De Kragge II is a municipal solid waste (MSW) landfill located near the town of Bergen op Zoom, in The Netherlands. It has four compartments and one of them (compartment 3) is under leachate recirculation since 2017. This compartment has a total surface of 5.6 ha, and an average height of 17 m. It has a bottom liner of 2 mm high-density polyethylene (HDPE) and a cover layer of soil and industrial debris.

De Kragge II has five waste cells that do not correspond exactly to the four compartments described before. Therefore, the information for the composition of waste for compartment 3 had to be recalculated (Meza, 2021). Figure 1 shows the composition of the landfilled waste in compartment 3, which is mainly household waste (29.2%), construction and demolition waste (21.4%) and commercial waste (21.3%).

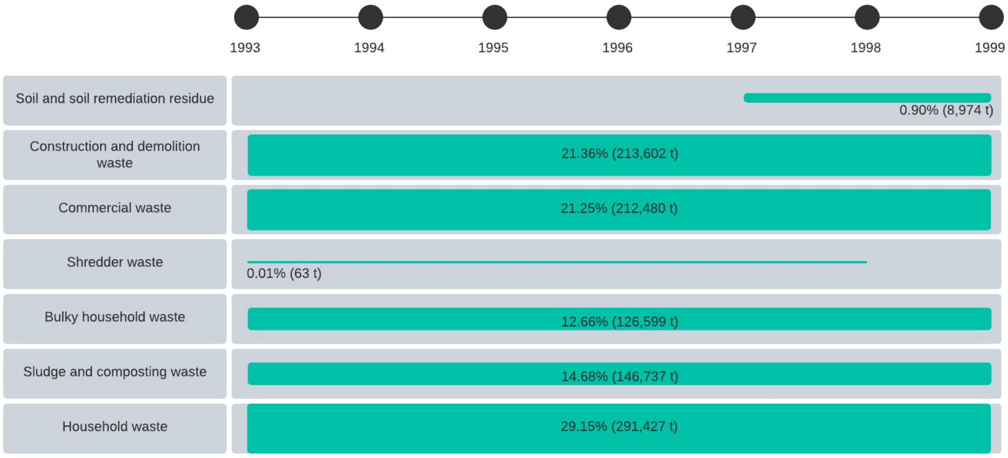
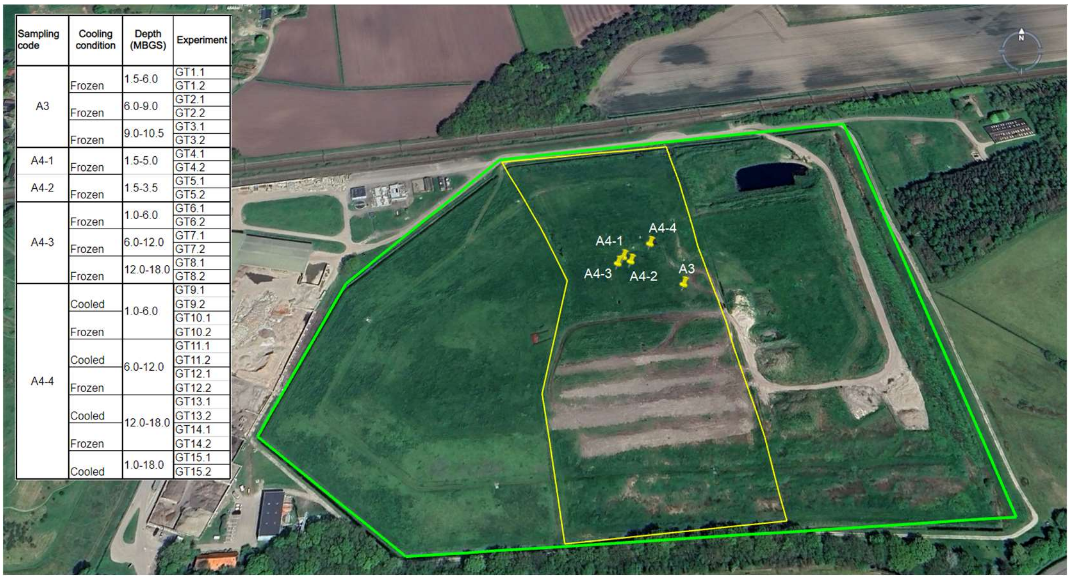


Figure 1: Waste composition over the landfilled period (1993-1998) in compartment 3.

The moisture content of the waste at the moment of landfilling is unknown. However, it is known that the moisture content in landfilled MSW waste typically ranges between 15% and 40% (Eck, 2000). Considering the waste composition and dry matter for different types of waste streams in the Netherlands (TAUW, 2011) and the type and amount of landfilled waste in compartment 3, a moisture content of 26% was assumed and all data recalculated to waste dry weight.

2.2 Waste sampling and characterization

Several drillings were made to install piezometers and a pumping well. This occasion was used to retrieve waste samples. Removing the top layer (mostly landfill cover soil), the drilled waste was mixed across several meters of depth, expressed as meters below ground surface (MBGS, see table in Figure 2). A total of 12 samples from different layers and wells were collected in 10 L and 20 L buckets. Three of those samples were split into two subsamples, of which one was stored under freezing conditions and the other under cooling conditions. A total of 15 samples were stored in buckets under cooling or freezing conditions until the start of the experiment.



Sampling code	Cooling condition	Depth (MBGS)	Experiment
A3	Frozen	1.5-6.0	GT1.1
			GT1.2
	Frozen	6.0-9.0	GT2.1
			GT2.2
	Frozen	9.0-10.5	GT3.1
			GT3.2
A4-1	Frozen	1.5-5.0	GT4.1
			GT4.2
A4-2	Frozen	1.5-3.5	GT5.1
			GT5.2
A4-3	Frozen	1.0-6.0	GT6.1
			GT6.2
	Frozen	6.0-12.0	GT7.1
			GT7.2
	Frozen	12.0-18.0	GT8.1
			GT8.2
A4-4	Cooled	1.0-6.0	GT9.1
			GT9.2
	Frozen		GT10.1
			GT10.2
	Cooled	6.0-12.0	GT11.1
			GT11.2
	Frozen		GT12.1
			GT12.2
	Cooled	12.0-18.0	GT13.1
			GT13.2
	Frozen		GT14.1
			GT14.2
	Cooled	1.0-18.0	GT15.1
			GT15.2

Figure 2: Aereal photograph of De Kragge II landfill (green line), compartment 3 (yellow line), and the waste sampling locations.

To start with the experiment, the frozen samples were defrosted overnight at 10 °C. The amount of sample in each bucket varied between 1.2 kg and 10.2 kg. Two subsamples were taken per bucket. This made a total of 30 samples for the methane generation experiment.

The moisture content was analysed in approximately 100 g of the waste sample by drying the sample for 24 hours at 105 °C. The remaining waste was manually sorted into seven fractions: fines, soft plastics, hard plastics, wood, stones, metal, and rest, the weight of each fraction was recorded.

2.3 Methane generation experiment

The experiment aimed to quantify the cumulative microbial degradation of waste organic matter under anaerobic conditions, with CO_2 and CH_4 as the main products.

Around 250 g of waste were placed without further pre-treatment inside 1 L glass bottles which were closed with butyl rubber stoppers, all weights were noted. Due to the heterogeneity of the waste, two samples per bucket were taken. To establish anaerobic conditions, the bottle was flushed with 100% N_2 over a period of 5 minutes. Thereafter, samples were incubated in the dark at 20°C.

Methane generation over time was monitored by measuring the pressure and the composition of the bottle headspace. For both measurements, a syringe with a needle that passed through the rubber stopper was used. To analyse the gas composition, approximately 3 ml of headspace gas mixture was injected into a gas chromatograph (490-microGC, Da Vinci). The pressure was measured before and after gas sampling by connecting the needle to a digital manometer (LEX1, Keller) and total gas generation was corrected for the volume lost due to sampling. To prevent excessive pressure buildup inside the bottle, the pressure was released upon reaching 1200 hPa. Initially, the gas composition and pressure were measured twice a week, and thereafter, depending on the reactivity of the sample, the measurement frequency was reduced to up to twice per month.

Cumulative gas generation was calculated based on the ideal gas law:

$$PV = \mathrm{nRT} \text{ and therefore } n = \frac{PV}{\mathrm{RT}}$$

Where P is the pressure in Pascals (Pa), V the volume in litters (L), n the number of moles present in the gas phase, R the gas constant which for the units we have is equal to 8134.58134.5 L Pa / (K mol) and T is the temperature in Kelvin (K).

From the number of moles, the molar masses, the data on gas composition and sample dry weight (dw) the amount of methane produced per dry weight over the time were calculated. For each sample, the data points were fitted with a one-phase or two-phase exponential decay model using OriginLab software (Origin 2022), depending on which model delivered the higher coefficient of determination. All fits were statistically significant on the level of $p < 0.001$.

2.4 Estimation of methane generation using a model

The NV Afvalzorg Multiphase Landfill Gas Model(Afvalzorg, 2022) was used to estimate the amount of methane that the landfill would produce over 100 years. The model uses information such as the type and amount of waste that was landfilled over the years. It considers different decay rates (k-value) of degradable carbon for different types of waste (Scharff & Jacobs, 2006). Among the different parameters that the model considers is the factor of 0.5 for the fraction of methane generated in the landfill gas, 1 for the methane correction factor (IPCC recommended value for managed anaerobic landfill), and k-values of 0.030 year^{-1}, 0.099 year^{-1} and 0.187 year^{-1} for slow, moderate and fast rates. For this study, the methane generation estimated for the year 2021 was considered, as well as the remaining methane potential for the period 2021-2093.

2.5 Recovery of landfill gas

To calculate the landfill gas recovered from compartment 3, data on gas composition and volumetric flow rates from the gas blower station were used. This was done on a weekly basis on the bulk extracted gas, which included the gas originating from compartments 1-4. Also, measurements of gas composition and gas velocity on individual gas wells in the landfill were performed every one or two months. For this study, we selected data from the dates on which individual wells in compartment 3 (the compartment under leachate recirculation) were measured. To derive gas flow rates from the velocities measured on individual wells, the recovered bulk landfill gas flow was divided proportionally to the gas velocities in all the measured wells in compartments 1-4. Finally, 13 gas wells located in compartment 3 were selected and the methane recovered was calculated using the data on gas composition for the year 2021.

3. RESULTS AND DISCUSSION

3.1 Waste characteristics and methane potential

The waste characterization included the sorting and moisture content measurement in the fifteen samples that came from the drilling (Figure 3). The sorting showed a significant amount of plastics in some samples, with values up to 50% in sample GT1. Samples GT5 and GT14 had the highest percentage of fines, with 81.8% and 69.3% respectively. The amount of plastic in the landfill may envelop parts of the waste, making it less accessible for degradation. Samples GT4 and GT8 showed free water, reflected in the highest moisture contents (126.8%dw and 115.6%dw, respectively). The lowest values were found in samples GT5 with 43%dw. Other studies have found moisture contents between 40 and 60% for a MSW landfill (Maciel & Jucá, 2011). The higher moisture content in the landfill may be caused by the leachate recirculation and/or by ponding of leachate in relation to impermeable materials such as sheets of plastics.

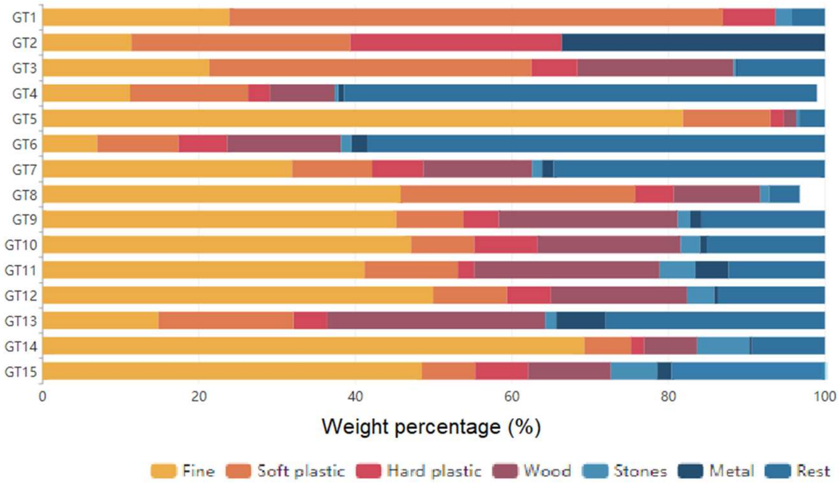


Figure 3: Waste composition of fifteen samples from compartement 3 in the landfill.

The CH_4 generation in all the samples followed a non-linear trend over time (Figure 4), indicating initial consumption of the most easily degradable organic matter at higher rates which then declined when the less degradable organic matter dominated the mixture over time. For more than half of the samples a 2-phase exponential decay function best fitted the data, indicating the presence of waste with different k values (see Figure 4, graph insertion bottom right). The high statistical significance ($p<0.001$) of the exponential decay fitting implied adequate capture of CH_4 generation kinetics. Compared to the k values of the predictive model (section 2.4) the k values for the laboratory tests were very high, indicating short half-lives of waste organic fractions, presumably due to waste perturbation upon drilling and sampling, enhancing surface accessibility. In line with the short half-lives, the CH_4 generation of most samples levelled off to very low rates or diminished completely within a year of laboratory testing, suggesting that the total CH_4 potential had been mostly exhausted within this period. Cumulative methane generation for 365 days of experiment varied between 0.03 kg CH_4/t dw and 15.8 kg CH_4/t dw. Also, previous studies have revealed a large variation in gas generation due to waste heterogeneity (Gebert et al., 2011; Hansen et al., 2004).

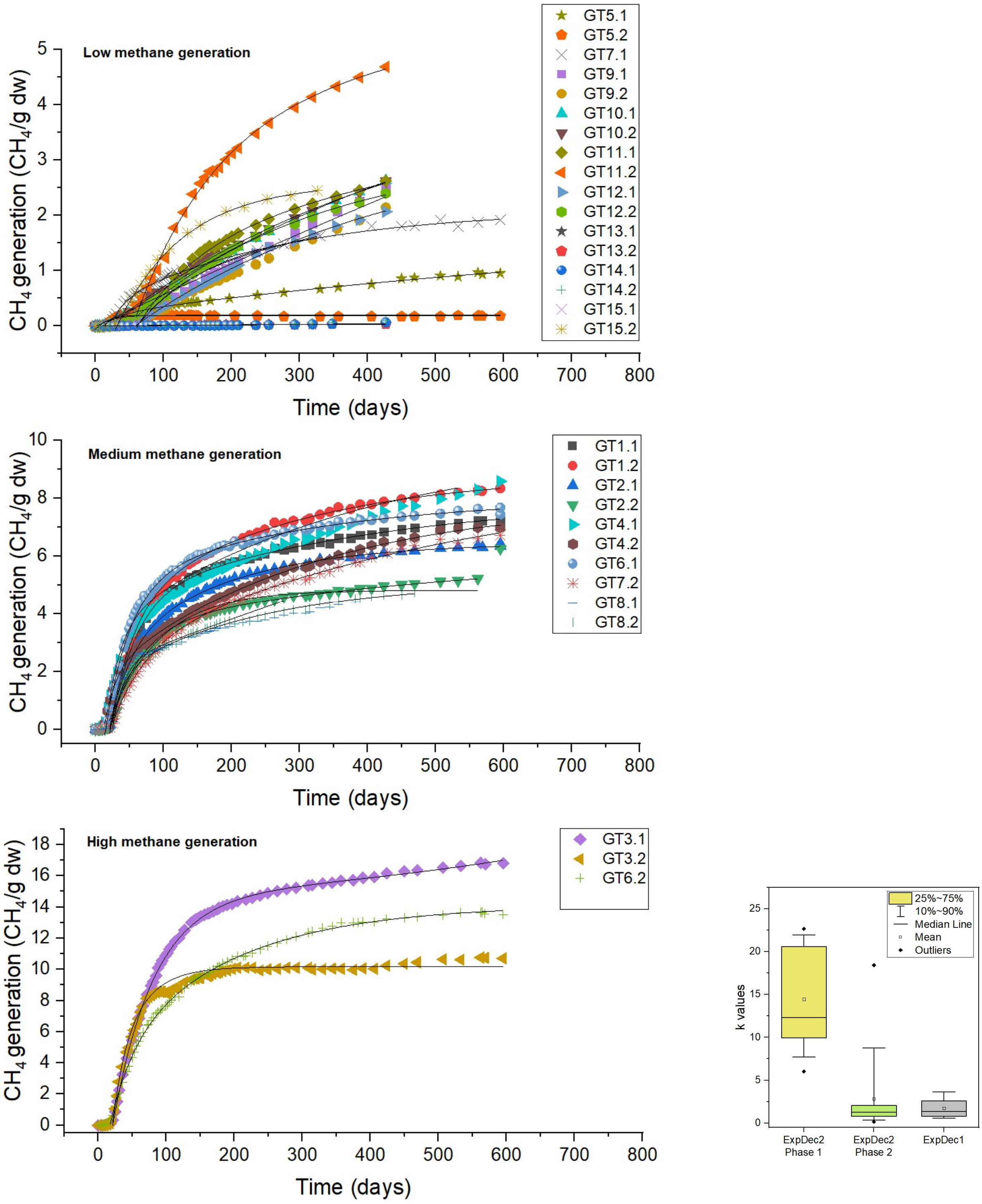


Figure 4: Course of CH4 generation over time in laboratory experiment. Samples with low methane generation (top); samples with medium methane generation (middle); samples with high methane generation (bottom). Symbols = measured data, lines = 1 phase or 2 phase exponential decay fits. Lower right graph: Boxplots of k

values derived from the exponential fits.

The results were clustered into groups of low, medium and high CH_4 generation, and a fitting curve without considering the initial lag phase was included in each sample, with the exception of samples GT14.1 and GT14.2, which showed a lag phase of > 200 days. The storage conditions (cooling versus freezing) did not seem to affect CH_4 generation, with values between 1.91 kg CH_4/t and 2.43 kg CH_4/t for samples and parallels stored under different conditions such as GT9-1, GT9-2, GT10-1 and GT10-2 (for storage conditions see Figure 2).

The cumulative methane generation within 365 days (1 year) was considerably higher in the laboratory than predicted by the Afvalzorg multiphase model and recovered by active gas extraction for 2021 (Figure 5, top). The drilling process and the perturbation during sample collection and processing may have enhanced available surface area and access to degradable organic matter and/or nutrients, leading to increased degradation of organic matter in the laboratory experiment. The difference is also influenced by the moisture content of the sampled waste in the laboratory experiment (between 43%dw and 126.8%dw) and the one considered in the model and the recovery data from the landfill in order to relate all data to waste dry weight. Waste dry mass at the moment of deposition was assumed with 26% dw, but no data are available, leading to uncertainty when relating modelled CH_4 generation and extracted CH_4 to the dry mass of the waste.

The calculated recovered methane from compartment 3 was lower than the predicted methane generated from the same compartment (0.07 kg CH_4/t dw and 0.23 kg CH_4/t dw respectively). This would mean a recovery efficiency of 30.4%, which is consistent with the recovery efficiency found in other landfills (Duan et al., 2022), including de Kragge II (Vereniging Afvalbedrijven, 2015). Of course, model uncertainty based on uncertainties regarding the type and amount of landfilled waste (see section 2.1) has to be considered as well. The number of gas wells in the landfill also affects the amount of gas extracted with respect to the gas that is actually produced in the landfill (Themelis & Ulloa, 2007). Three out of the thirteen gas wells in compartment 3 showed no gas flow at all. The degradability of the waste in the landfill may be limited by the condition in the waste body such as moisture (Fei et al., 2016) and temperature, or inhibition by accumulation of metabolic products, but also by the intrinsic degradability of the waste organic matter itself.

The average methane potential for all the waste samples measured in the laboratory (6.1 kg CH_4/t dw) was in the same order of magnitude, but almost twice as much as the remaining methane potential predicted by the model for the period 2021-2093, hence after sampling (3.27 kg CH_4/t dw) (Figure 5, bottom). This was partly expected considering that the model does not take into account the stabilization process due to the leachate recirculation. These data show that although the reactivity of the waste is mainly low, there is still gas potential in the waste that is not tapped under the in situ conditions, or, at least, not within the same time frame as under laboratory conditions.

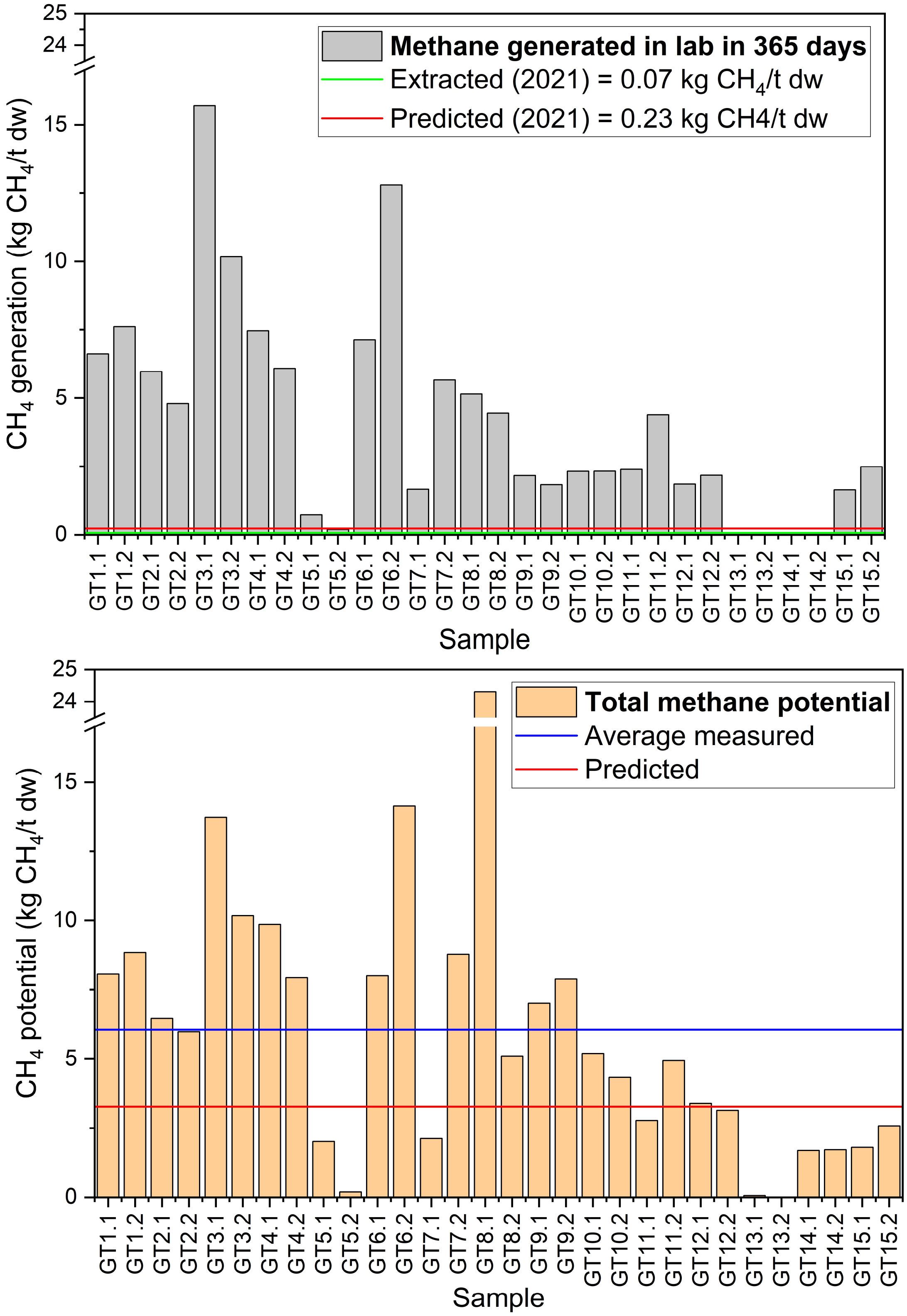


Figure 5: CH4 generation in the laboratory. Top: for 365 days and comparison with the predicted and extracted CH4 for 2021; bottom: total CH4 potential and comparison with the average of all samples and the predicted remaining CH4 potential, calculated by subtracting the CH4 potential predicted for 2021 from the CH4 potential predicted for 2093.

3.2 Correlation of methane potential with waste properties

The methane generation varied between 0.03 g CH_4/kg dw and 15.8 g CH_4/kg dw for 365 days of experiment. Data for cumulative methane generation over 365 days of incubation did not show a depth-related trend (Figure 6, top left). The mean went up towards the middle layer and decreased again towards the bottom. A lag phase at the beginning of the experiment can affect the data evaluation for short-term analysis, but less affects the long-term analysis (Gebert et al., 2011). As anticipated, the methane generation per drilling (Figure 6, top right) displayed the lowest variability for the drillings that reached the top layer and had only a couple of samples. Among the drills that went up to 18 MBGS, A4-4 showed the least variability and methane generation.

Figure 6, middle left showed a highly positive linear relationship between the methane generation over 21 days (MP_{21}) and 365 days (MP_{365}) after lag phase, suggesting that short-term tests are useful to predict long-term gas potentials. The gas generation in 21 days was below the limit of 20 l/kg for all samples, stipulated in the German landfill ordinance (DepV, 2009) as one of the limit values for the landfilling of wastes, even when considering that the methane generation in the GP21 test carried out according to DIN 38414-8 would be higher due a higher incubation temperature (35 °C).

The correlation between moisture content and methane generation showed that the sample with the lowest moisture content had one of the lowest methane generation rates (Figure 6, middle right). The moisture content may be a limiting factor for this sample, as the high activity of anaerobic bacteria requires a moisture content above 40% (Themelis & Ulloa, 2007) and no water addition was considered in the experiment. The relationship between moisture and methane generation over all samples was not so obvious, which may be due to heterogeneity in degradable organic carbon contents of the waste or different degrees of waste stabilization, either due to top-bottom stratification and/or due to differences in water transport properties. The highest methane generation was for samples GT3 with 81.2% moisture content, although one of the two parallels had almost half of the methane generation at 300 days (GT3.1: 40.1 l/kg dw; GT3.2: 24.7 l/kg dw), indicating high small-scale heterogeneity. The heterogeneity of the samples suggests to treat each parallel as an individual experiment.

Factors that may influence the reactivity in the waste are the amount of organic carbon and the proportion of non-degradable materials rich in carbon such as hard plastics (Gebert et al., 2011). Figure 6 bottom left and right, showed the relation between the methane generation and the TOC (%) and hard plastics (%), respectively. Although there is no strong relationship between them, it seems that samples with higher carbon content and lower content of hard plastics have a higher methane generation. If methane generation is related to the TOC content of the sample (data not shown), the correlation over the entire data set does not become more obvious; however, it is clearly seen that the sample with the highest share of hard plastics produces the lowest amount of methane per unit organic carbon. The heterogeneity of the waste is reflected in the results obtained in the methane generation of subsamples from the same sample. Therefore, it is planned to characterize the waste that is inside each bottle at the end of the experiment and to re-evaluate correlations between sample properties and methane generation.

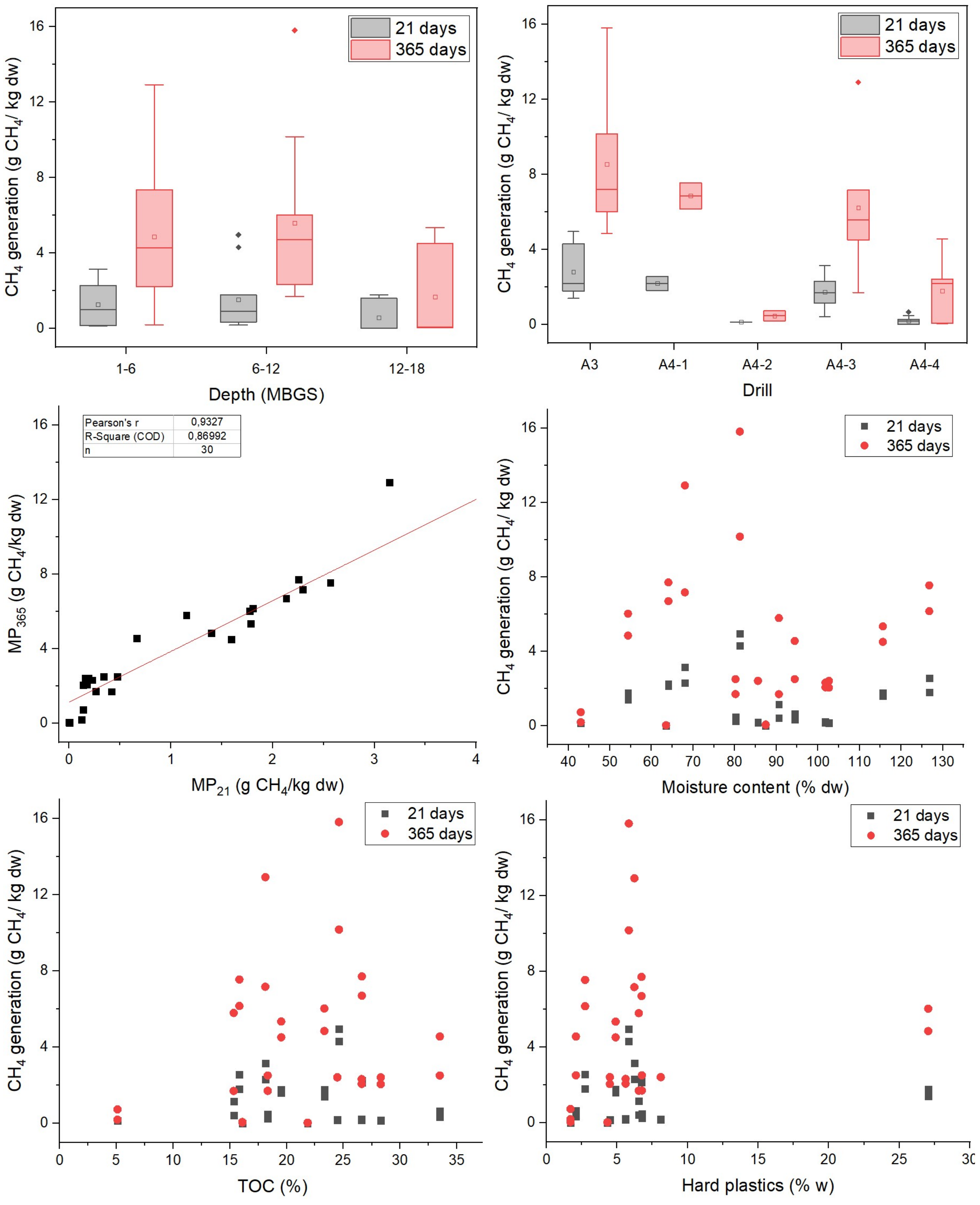


Figure 6: Methane generation per depth and time (top left); relationship between methane generation at 21 days and 300 days (top right); and relationship between methane generation and moisture content (middle right); elationship between gas generation and total organic carbon (TOC) and hard plastic percentage (bottom left and right respectively). MBGS = meters below ground surface.

4. CONCLUSIONS

Methane generation in laboratory incubation of sampled waste strongly exceeded modelled methane generation and methane extracted on site, likely due to material perturbation during sampling (causing increased organic matter degradation) in conjunction with low waste permeability due to plastics on site, resulting in lower accessibility of waste and impeded gas and water flow. Higher methane generation rates might also or additionally result from enhanced waste degradability as a result of five years of recirculation. However, the total methane potential, predicted from the exponential curve fitting of the long-term measurement data, was quite close to the methane potential predicted by the Afvalzorg multiphase model for the remaining period of 2021 to 2093. This suggests on the one hand a relatively good predictive quality of the model. The discrepancy, on the other hand, indicates that the amount of potentially biodegradable carbon in the landfill after five years of leachate recirculation was almost twice as much as predicted. The sampling depth, moisture content, carbon content, and percentage of hard plastics as materials with high non-biodegradable carbon partially seem to influence methane generation. However, the overall heterogeneity of the samples obfuscates trends over the entire data set and suggests re-analyzing these parameters on the individual subsets of samples used for the methane generation test.

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