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MONITORING THE PROCESS OF BIODEGRADATION AND METHANE GAS GENERATION IN AN EXPERIMENTAL CELL OF MUNICIPAL SOLID WASTE IN BRAZIL

MONITORAMENTO DO PROCESSO DE BIODEGRADAÇÃO E GERAÇÃO DE GÁS METANO EM UMA CÉLULA EXPERIMENTAL DE RESÍDUOS SÓLIDOS NO BRASIL

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Abstract: Organic waste is particularly important as it influences the generation of leachate and gas in the landfill. This work aimed to monitor the process of biodegradation and methane gas generation in an experimental cell with urban solid waste in Brazil. Monitoring in the experimental cell was carried out for 1.100 days, using environmental measurement equipment and portable gas analyzers with measurements in the central vertical drain of the experimental cell. It was possible to monitor the methane gas generation curve and the composition of biogas during the solid waste biodegradation process. The results obtained from the research confirmed the explanation for the drop in the methane generation rate observed in several landfills after the completion of waste disposal.

Keywords: Waste, Biodegradation, Biogas, Methane.

Resumo: Resíduos orgânicos são particularmente importantes, pois influenciam a geração de lixiviado e gás no aterro sanitário. Este trabalho teve como objetivo monitorar o processo de biodegradação e geração de gás metano em uma célula experimental com resíduos sólidos urbanos no Brasil. O monitoramento na célula experimental foi realizado durante 1100 dias, utilizando-se equipamentos de medição ambiental e analisadores de gases portáteis com medições no dreno vertical central da célula experimental. Foi possível monitorar a curva de geração do gás metano e a composição do biogás durante o processo de biodegradação dos resíduos sólidos. Os resultados obtidos na pesquisa confirmaram a explicação para a queda na taxa de geração de metano observada em diversos aterros após a finalização da disposição dos resíduos.

Palavras-chave: Resíduos, Biodegradação, Biogás, Metano.

Introduction

Waste can be defined as any material that is discarded during human activities, which is in the form of solids, semi-solids, liquids and gases. Urban solid waste (MSW) is generated in homes, businesses, streets and various other urban activities. Urban solid waste is usually composed of: organic waste (food waste) and garden waste, paper and cardboard, plastics, wood, metals, glass and others [(1)].

The components of MSW are very varied and have very different physical and chemical properties. Organic residues, especially food, have a lot of moisture and biodegrade quickly. Paper and cardboard are drier residues and degrade less quickly. Plastics, on the other hand, are very dry and not very degradable. Variations that influence the biodegradation process, especially the biochemical processes, take place in the landfill [(2)].

Organic waste is particularly important as it influences the generation of leachate and gas in the landfill. The variation in the composition of urban solid waste is fundamentally due to the socioeconomic development patterns of a location[(3)]. In the case of Brazil, a developing country compared to more developed countries, it tends to generate higher levels of organic matter, and it is possible to observe that the percentage of organic matter in Brazilian cities is high, ranging from 50% to more. The moisture content is also very important as it influences the rate of biodegradation of the waste. Moisture varies with the composition of waste and local environmental and climatic conditions [(3,4)].

The biodegradation of waste in the landfill occurs through physical, chemical and biological processes [(5)]. This process involves different stages over time. According to Farquhar & Rovers [(6)], “these phases are divided into: aerobic, non-methanogenic anaerobic, non-stabilized methanogenic anaerobic and stabilized methanogenic anaerobic”. The initial phase, aerobic, is usually shorter, as the oxygen content is high in the waste that was recently deposited in the landfill. Gradually, oxygen is consumed, generating carbon dioxide, water and heat. As the temperature increases, organic matter begins to biodegrade. In this phase, the process becomes anaerobic, producing organic

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acids, and this phase is called acid anaerobic (non-methanogenic), which can last for months, significantly increasing the production of carbon dioxide [5,6,7,8].

In the methanogenic phase, which lasts for years, methane is produced in an accelerated way, however in the non-stable methanogenic phase, this production slows down substantially decreasing the rate of methane generation. At each stage, microorganisms act to metabolically transform many compounds[5]. The acid anaerobic step can be subdivided into hydrolysis, acidogenesis and acetogenesis. In hydrolysis, the action of water decomposes complex substances, dissolving them into smaller and simpler molecules, enabling the action of fermentative bacteria, which transform these simpler compounds into volatile fatty acids (main products, produced called acidogenic), alcohols, acids , carbon dioxide, hydrogen, ammonia and sulfides. Acetogenic bacteria oxidize these generated products, providing them with methanogenic bacteria that, in the methanogenic phase, produce, from the metabolization of these substrates, methane and carbon dioxide [1,5].

Estimating landfill methane generation is important for energy use and for selling carbon credits. Landfill gas (biogas), resulting from the mixture of gases resulting from the anaerobic biodegradation of organic waste, is essentially composed of methane and carbon dioxide. Biogas has a specific calorific value, which is directly related to the methane gas content. Landfill biogas typically contains about 50% methane gas [1,9].

The anaerobic biodegradation of organic waste in landfill can be represented in a simplified way by an equation, by which it is possible to determine the production of methane and carbon dioxide from an organic compound[10]. In the landfill there is a lot of waste and external factors, such as composition, age, humidity, temperature and others, that influence the biodegradation process. Modeling and estimating methane generation at the landfill is therefore very complex. However, methane generation can be approximated by a first order kinetic mathematical model [9,11].

There are many models to estimate methane generation in landfills. The INTERGOVERNMENTAL PANEL ON CLIMATE CHANGE (IPCC), UNITED STATES ENVIRONMENTAL PROTECTION AGENCY (USEPA) and WORLD BANK models are widely used [12] and relate the mass of waste that enters the landfill annually to the speed of methane generation. The potential for generating methane (L) depends on the content of organic waste and the environmental and climatic conditions of the landfill site. Also noteworthy is the temporal factor of this process, indicated by the rate of decomposition of organic matter, that is, the biodegradation constant (k), which is directly proportional to the local climatic condition (13).

Normally, in landfills, there are gas capture systems, formed by vertical drains, which capture these gases and send them for burning in flares (control of emissions) or internal combustion engines, which produce electrical energy(14). The systems for capturing gases in landfills are not 100% efficient, therefore, although gas emission rates are reduced, a portion of gas is still emitted into the atmosphere (15,16).

These emissions of methane gas (CH_4) in landfills contribute to warming, significantly impacting the environment. Methane emissions from landfills are highly dependent on climatic conditions. Estimating the temporal variability of methane emitted into the atmosphere can help us to reduce uncertainties in landfill emission estimates, especially in a closed landfill (17,18,19).

The landfill can be defined as a form of disposal of solid waste in the soil, particularly MSW, which based on engineering criteria and operational standards, allows safe containment for the environment. Landfills have soil protection systems, with the installation of geomembranes, which waterproof the soil and protect it (1,12).

The daily generation of solid urban waste in Brazil is of the order of 1 kg/day per inhabitant and the great Brazilian challenge is to dispose of waste properly in landfills, as approximately only about 50% is disposed of properly and practically 50% is destined inappropriately in the dumps (13).

The use of methane to generate electricity, mainly, has added economic potential to the operation of landfills. In this sense, monitoring the generation of methane in landfills has become very important, as there are currently several projects for the use of biogas energy in operation and being implemented in Brazil. Currently 22 thermoelectric plants are in operation in Brazil, totaling an energy potential of around 174.3 MW (20).

Thus, this work aims to monitor the process of biodegradation and methane gas generation in an experimental cell with urban solid waste in Brazil.

Material and Methods

Study area: Caieiras Landfill

The Caieiras Sanitary Landfill is located in the municipality of Caieiras (23º21'51"S and 46º44'26"W) in the state of São Paulo, Brazil, and is part of a waste treatment center, with the objective of disposing of solid industrial waste of municipal districts of São Paulo and other neighboring cities (Figure 1).

Figure 1 – Caieiras Landfill in São Paulo state, Brazil.

This enterprise belongs to the company Solví Soluções Industriais. This operational unit has sanitary and industrial landfills, a biogas plant, a reverse logistics unit, a fuel waste plant, a slurry tank, a thermal and thermoelectric biogas desorption unit. The biogas thermoelectric plant, called Termoverde Caieiras, has an installed capacity of 29.5 MW [12] (Figure 2).

Figure 2 – The biogas thermoelectric plant - Termoverde Caieiras

The region has a subtropical climate controlled by the local relief. The average annual precipitation varies between 1.250 and 1.350 mm. Due to the high altitude of the area the average annual temperature varies between 16°C and 18°C, the average temperature during the summer varies between 19°C and 22°C, and during the winter, between 13.5°C and 15°C [(14)].

Experimental Cell

Figure 3 shows a scheme of the experimental cell (Figure 3a), which has dimensions of about 30x35 m^2, and a passive-capture vertical drain in the center for gas collection or measurements. The experimental cell has a central plateau with an almost rectangular flat surface and a lateral embankment on the periphery (Figure 3b). A 1.2 m high structure to contain the deposited MSW surrounds it. The base has a 0.5m compact silt soil layer covered by a 2mm thick high density polyethylene geomembrane and a geotextile to provide mechanical strength. Above it is a gravel system for slurry collection and in the center the passive vertical drain is made of a 0.6 m diameter concrete pipe surrounded by a 0.45 m thick layer of gravel. The deposited residue reaches a height of 4.5 m covered by a 0.5 m layer of silt soil on top. The vertical drain collects gas from the

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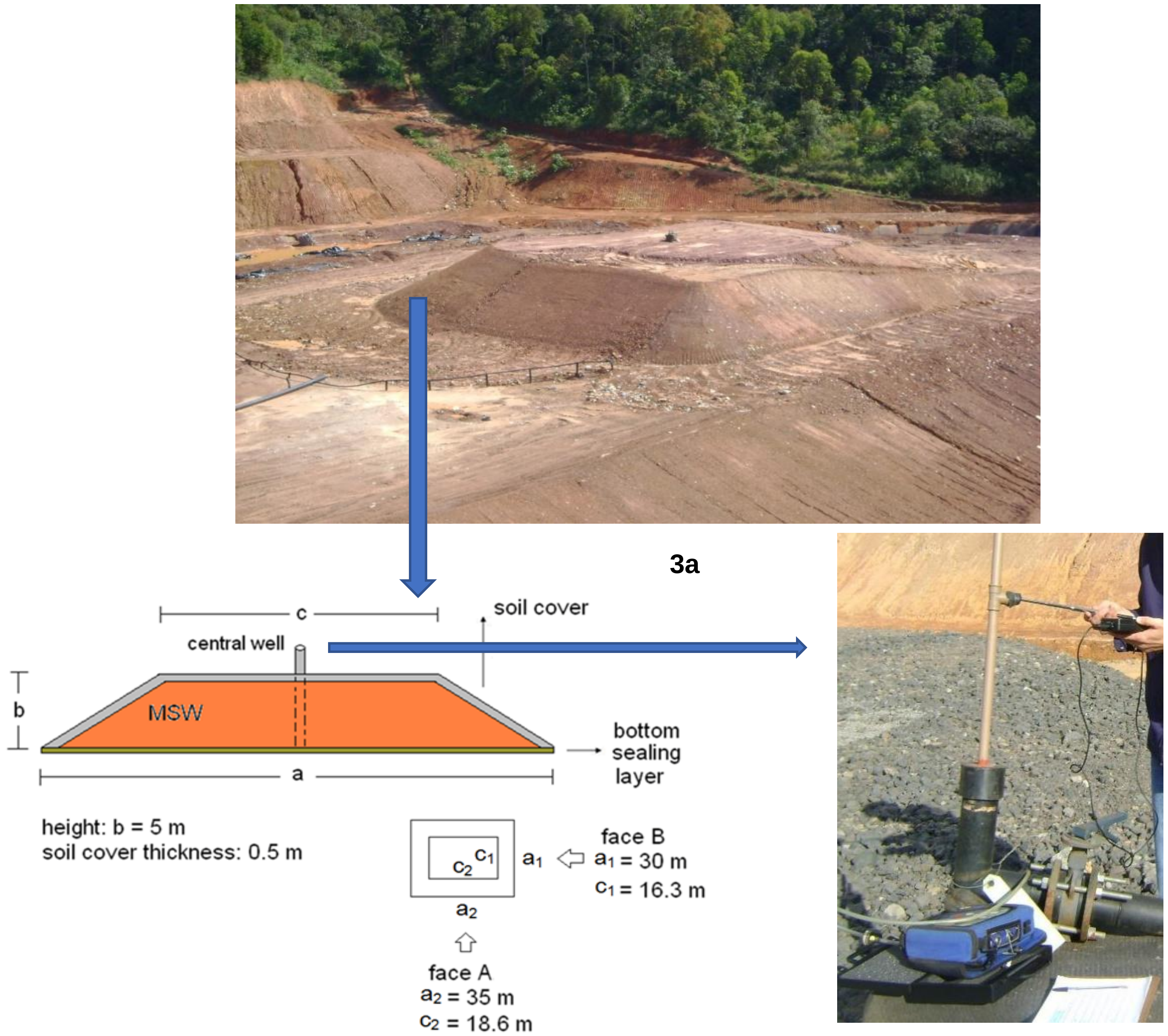
bottom of the landfill to the waste-ground cover interface. The side embankments have a slope of about 30° and are also covered by a 0.5 m silt soil layer[14].

Figure 3 – The experimental cell in Landfill Caieiras. 3a) Experimental Cell. 3b) Schematic Experimental Cell. 3c) Monitoring of methane generation in the experimental cell with portable gas analyzer



3a

3b

3c

The MSW material was deposited, spread to form a homogenous material and presented a uniform permeability to gas and leachate transport throughout the cell, compacted and covered with the soil layer.

The final compaction level used was high and care was taken to maintain the soil cover uniform during all measurements. Thus we can consider the experimental cell as a porous system with two regions: the MSW region and the topsoil cover region. The test cell generated gas through anaerobic decomposition of MSW, which migrated through the medium pores toward the central well and toward the atmosphere. The gas flow rate in the central well was used to monitor the gas generation in the cell, and was always kept open to the atmosphere. At its top, we installed a system for measuring the gas temperature, composition, and flow rate[(12)].

The MSW material was deposited in the experimental cell on September 4th, 2009 and, from that date, we carried out flow measurements in the central well of the cell until December 22th, 2012. The age of the RSU material was 1.100 days or about 3 years. The experimental cell was filled with 3.786,13±0.25 tons of solid urban waste (MSW) in a period of 12 days (CANDIANI & MOREIRA, 2015). Table 1 summarizes the experimental cell technical data and Table 2 presents the composition of the deposited MSW material, highlighting the significant presence (58.3%) of organic matter. The humidity of the residues deposited in the experimental cell was 60.9%[(14)].

Table 1 – General data from the experimental cell

Parameter	Data
Total deposited mass in 12 days (end of MSW deposition: Sept 4th, 2009)	3.786,13 t MSW
Total test cell area	1.050 m^2
Base liner materials and dimensions	Structural soil layer (0.5 m), High-density polyethylene geomembrane (2 mm thick) and geotextile
Thickness of topsoil cover (silt) Height of MSW column Total height of the cell	0.5 m 4.5 m 5.0 m
Central well dimensions	1.5 m diameter of gravel with concrete central tube of 0.60 m diameter
MSW specific mass	1.32 t MSW/m^3

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Table 2 – Composition of typical Brazilian and Caieiras Landfill MSW materials. The average moisture of the MSW is 60.9%.

Materials	Brazil (%)	Experimental cell (%)
Organic	65	58.3
Plastic	16	15.2
Paper and cardboard	13	14.6
Glass	2	2.5
Ferrous metals	2	1.8
Textile, leather and wood	-	3.7
Others (soil and rubble)	2	3.9
Total	100	100

Monitoring of gases in the experimental cell

The test cell generated gas through anaerobic decomposition of MSW, which migrated through the medium pores toward the central well and toward the atmosphere. The gas flow rate in the central well was used to monitor gas generation in the cell and was always open to the atmosphere. At the top, we installed a system to measure the temperature of the gas, composition and flow rate, using gas meters - portable gas analyzers. The molar fraction of CH_4, CO_2, and O_2 was measured utilizing an analyser that samples and analyses from a gas flow environment (Figure 3c).

Therefore, the variables needed to be measured in the well are the gas speed, temperature, and composition. The site ambient pressure during the measurements was 91,054 Pa. The speed and temperature measurements were conducted utilizing a thermal anemometer, which gives accurate results for the speed range between 0.2 to 20 m s^{-1} and temperature range between 273 and 323 K. At the top of the central well, a tube system was adapted to allow the connection of the measurement probes. The gas speed in the well is obtained out of a balance of mass flow rate[(10)].

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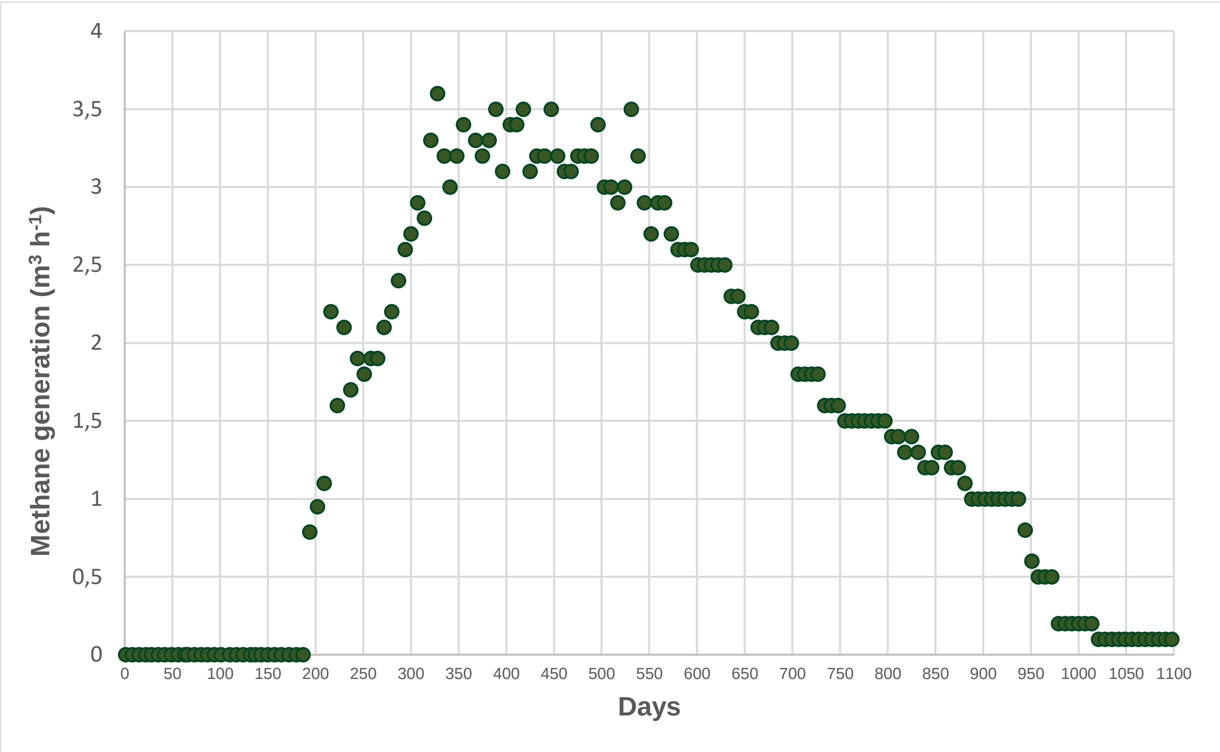
Results and Discussions

Figure 4 shows temporal evolution of methane generation in the experimental cell.

Figure 4 – Temporal evolution of methane generation in the experimental cell



The mean molar fraction of methane (CH_4) was 45.6±1.6%, the mean fraction of CO_2 was 39.5±1.1% and the mean fraction of O_2 was 0.06±0.005%.

The central well flow rate was obtained from the gas speed in the well, and the corresponding methane molar fraction. The gas composition (molar fractions) in several dates is presented in Table 3, while the temperature and speed measured in the central well are presented in Table 4.

Table 3 – Gas composition (molar fraction) in the central well for several dates.

Year	CH_4 (%)	Average of values CO_2 (%)	O_2 (%)
2009	2.5	18.6	13.3
2010	33.1	27.3	3.45
2011	49.0	39.6	0.12
2012	42.2	39.5	0.00
Average of periods of greater stability in CH_4 generation (2011 and 2012)	**45.6±1.6**	**39.5±1.1**	**0.06±0.005**

In a period of 1.100 days, the mean gas speed varied between 0.0 to 1.6 m s^{-1}, however the average value was 1.3±0.05 m s^{-1}. The average methane flow rate was 2.15±0.10 m^3 h^{-1}, ranging from 0.8 to 3.6 m^3 h^{-1}. Biogas is considered to behave as an ideal gas, occupy the whole volume of the well and travel with average speed. The flow rate of the central well is 0.026±0.001 mol s^{-1}, and the gas temperature during the measurement is 273.15 K, and is the central well cross-section area (0.2835 m^2).

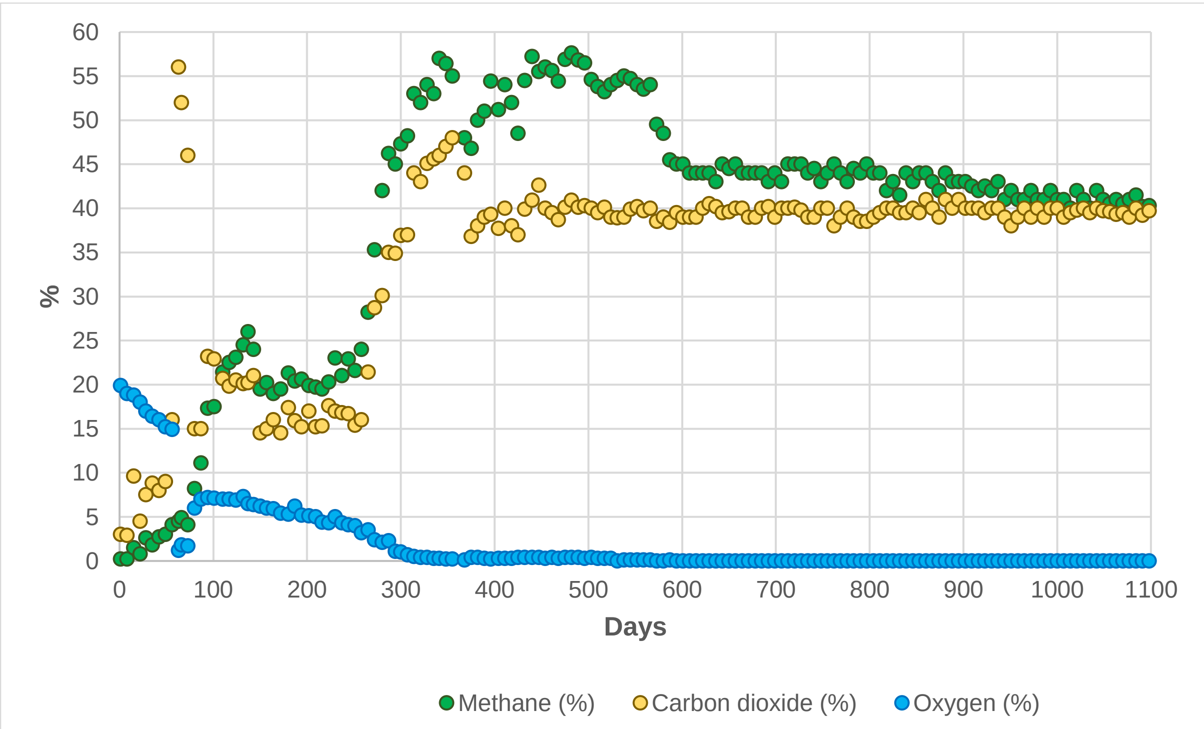
Table 4 – Gas temperature and speed measurements, and methane flow rate in the central well outlet

Year	Gas Temperature (K)	Gas speed (m s^{-1})	Average of values Methane flow rate (m^3 h^{-1})	Methane flow rate (mol s^{-1})
2009	300	0.0	0.0	0.0
2010	309	1	1.7	0.021
2011	310	1.6	2.6	0.032
2012	309	0.5	0.8	0.001
Average of periods of greater stability in gas speed (2010 and 2011)	**309.5±0.10**	**1.3±0.05**	**2.15±0.10**	**0.026±0.001**

Figure 5 shows the generation of biogas in the experimental cell. It is observed that the latency phase in the experimental cell can be characterized up to 194 days after the deposition of the wastes. It includes the hydrolysis, aerobic and non-methanogenic acid anaerobic phases, as in these phases there is no generation of methane gas.

It is noted that fluctuations in the production of biogas occurred over the period analyzed, an aspect probably explained by changes in the environmental conditions in the experimental cell arising from the action of the microorganisms and variations in the physical-chemical parameters (temperature, humidity etc.) caused by the conditions different climatic conditions throughout the observation period. At the beginning of the monitoring of the experimental cell, the climatic conditions must have influenced the process, as it was a very rainy period. Between 194 and 350 days, the experimental cell should be in the unstabilized methanogenic anaerobic phase. After 350 days to about 560 days of observation, methanogenic generation is intense and stable (Figure 4). It is possible to characterize the five stages of the biodegradation process of solid urban waste (MSW): hydrolysis, aerobic, non-methanogenic acid anaerobic, unstabilized methanogenic anaerobic and stabilized methanogenic anaerobic.

Figure 5 – Composition of biogas generated in the experimental cell



From Figure 4, it can be seen that the methane content was small and gradually increased, becoming more stable after 320 days and remains this way until about 560 days of observation. Regarding the carbon dioxide content, the production curve was practically similar to the methane production curve, noting that after 60 days a peak of carbon dioxide production occurred, then production fell and started to rise again. Analyzing Figure 5, it is possible to observe that the hydrolysis and aerobic phase in the experimental cell lasted approximately 56 days. After this period, the beginning of the acidogenic anaerobic phase is noted, with a significant reduction in oxygen (19.9 to 1.2%) and a significant increase in carbon dioxide (2.9 to 56%). This phase remained for up to approximately 80 days.

The unstable methanogenic anaerobic phase begins after this period, remaining for about 250 days. Note that when the oxygen content was below 5%, methane production increased, representing the end of the aerobic phase and the beginning of the anaerobic phase. After 280 days of monitoring, it was possible to verify that the methane concentration increased significantly (26 to 42%) and that after 360 days of monitoring the methane content continues to increase, reaching a value of 57%, an aspect that characterizes the anaerobic phase stable methanogenic. After 600 days of monitoring in the experimental cell, the methane content is decreasing. The monitoring of the gases produced in the experimental cell made it possible to characterize the different stages of biodegradation of the waste, as described by the analysis of Figures 4 and 5. The beginning of the monitoring in the experimental cell was marked by a very humid period, a condition that favored the percolation of water, and that may have contributed to increase the oxygen content in the first months of monitoring.

Table 5 summarizes the various phases identified in the methane generation biodegradation process in the experimental cell. According to Tchobanoglous, Theisen & Vinil[5] “all stages of biogas production in landfill together can take more than 40 years and each stage has a specific duration”. The duration of the two initial phases (aerobic and transition) together takes from 30 to 180 days, the acid phase takes from 180 to 1.100 days and the phase (methanogenic) takes from 3 to 40 years[5].

The methanogenic phase is characterized by the action of methanogenic microorganisms (bacteria of the genus *Methanobacterium*), which act to reduce gases (H_2 and CO_2) forming methane (CH_4) and water (H_2O) and transform acetic acid

(CH_3COOH) into methane and dioxide carbon (CO_2). In the methanogenic phase there is a production of 0 to 45% of methane (CH_4); however, this generation is very unstable.

The stable methanogenic phase is characterized by the production of methane in the proportion of 50 to 70% and CO_2 in the proportion of 30 to 50%[2,6]. According to Monteiro[21] point out that the most significant methane production, in an experimental cell at Aterro da Muribeca-PE, occurred after 300 days of monitoring.

The presence of excess moisture drastically reduces the production and quality of biogas in the landfill[22,23]. However, the humidity factor associated with the appropriate temperature, can favor the biodegradation process of the residues, anticipating the production of methane.

Table 5 – Stages of biogas generation in the experimental cell

Phase	Time interval (days)	Features
I - Aerobic (initial adjustment)	0 a 50	presence of oxygen (20%) - relatively short phase.
II - Anaerobic (transition)	50 - 100	presence of CO_2 - significant production of CO_2 (57%), reduction in O_2 content (0%), beginning of the most effective production of CH_4.
III - Acid	100 - 200	presence of water (hydrolysis) - characterized by the transformation of complex compounds, increased CH_4 production.
IV - Methanogenic	200 - 550	significant CH_4 production, CH_4 concentration stable for a long period at levels of 50 to 60%, the longest stage of the process.
IV - Maturation	550 - 1.100	the generation of biogas in this period begins to decline. This stage consists of the final stage of the MSW biodegradation process.

Conclusions

Each landfill has a useful life, which is its period of operation for reception of waste. At the end of the operational activities, the landfill closing period begins, including

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the monitoring of the composition and flow of biogas, mainly methane gas, which is used in the production of electricity. The idealized experiment with deposition of MSW in the form of a pulse allowed for the obtention of the temporal behavior of the biodegradation process for the generation of methane.

This result confirms the explanation for the fall of the methane generation rate observed in several landfills after completion of the deposition of waste. Several researches and practical experiences with data monitored in the field indicate that methane gas generation drops quickly after the closure of the landfill. With the results obtained from this work, it was possible to elucidate and verify this trend. Few Brazilian studies related to the biodegradation of solid waste in landfills are presented in the literature.

This makes it difficult to establish comparisons related to the stages of the biodegradation process. Knowledge of biogas production is a key aspect for the proper exploitation of this energy source, even in the post-closure period. For future research, it would be very important to quantitatively analyze the decay of methane generation, mainly determining the maximum deadline for the feasibility of using biogas in the landfil, and adjusting the landfill gas recovery projects from an economic point of view.

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