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Article

Emission and Neutralization of Methane from a Municipal Landfill-Parametric Analysis

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Received: 30 October 2020; Accepted: 25 November 2020; Published: 27 November 2020



Abstract: An attempt was made to estimate the annual production of CH₄ at a municipal waste landfill site in Poland. As a matter of fact, the extent of the unorganized emission of CH₄ from the landfill surface was approached based on the adopted mathematical model. The Ward agglomeration method for cluster analysis and the Pearson coefficient were employed to evaluate the distance-based similarity measure and to optimize methods for estimating methane emissions from a landfill as well as to verify the input parameters for the model. In order to calculate the content of biodegradable organic parts in the waste, morphological tests of the landfilled waste were performed. Physical quantities, measurements and the actual amount of the landfilled waste as well as the volume of CH₄ neutralized in a collective flare were implemented in the model, respectively. The model-based findings and experimental outcome demonstrated stable gas production in the landfill with a high CH₄ content. On the other hand, a rather low efficiency of the landfill passive degassing installation indicated the necessity to design and develop its active counterpart with the prospective application of the generated biogas for energy production in a cogeneration system.

Keywords: renewable energy; landfill gas; degassing installation; municipal waste; landfill site; cogeneration

1. Introduction

In the last decade, approximately 2 billion people were deprived of waste collection services worldwide. Moreover, nearly 3 billion Earth inhabitants have no access to controlled waste disposal facilities. Unfortunately, it is expected that the waste-related issues will only worsen in line with urbanization growth, income rise and consumer-oriented economy expansion, especially in developing countries. Within the next 30 years, the overall amount of waste produced globally is foreseen to double from nearly 2 billion metric tons in 2016 [1]. Waste generation has been an inherent feature of human economic activity over the centuries, while its management is becoming a major problem for many societies and economies. The necessity of waste management results from the need to reduce the space indispensable for their neutralization/processing/storage and subsequently the protection of environmental resources. The generated wastes should be segregated and recycled including

the recovery of energy in the fermentation process of biodegradable waste—e.g., such as green waste and food [2,3]. In recent years, sequencing batch reactors (SBRs) are becoming more and more popular in the wastewater treatment due to their working flexibility and adaptability to the variable quality of raw wastewater [4]. Following European Union (EU) regulations, the priority of introducing a general obligation to separate biodegradable waste at the source, including households, restaurants and supermarkets, has been noticed. The waste separation procedures allow to extract such wastes from the mixed mainstream of waste passed to the landfill. It seems that a clean waste stream can be used as input material for the fermentation process and the subsequent production of biogas as an energy carrier [5–7]. Biowaste fermentation processes are also possible to implement without a centralized system—for instance, in rural areas, contributing to the energy needs of the local communities. Such a solution can improve the living standard of the inhabitants as well as reduce energy consumption [8]. In fact, the generation and disposal of municipal waste is becoming a progressively significant environmental, economic and social issue. It was scientifically confirmed that the mass of generated waste per capita is growing with the rise of economic growth, especially in developing countries [9]. The bioeconomy concentrates on the implementation of biomass/biowaste in fundamental production branches (e.g., agriculture, forestry, fisheries) and relevant valorization of raw materials. Undoubtedly, biomass processing can facilitate to accomplish the EU's renewable energy goals. Obviously, a noticeable linkage between the bioeconomy, circular economy, bioproducts, and bioenergy is observed globally [10]. Despite the applied segregation systems, municipal waste contains organic fractions with high energy potential for the bioeconomy [11]. On the other hand, the limited time and funding can hamper the achievement of the ambitious sustainable development objectives in mitigating climate changes. Hence, the use of the two-tier multi-criteria decision analysis (MCDA) seems beneficial in order to identify the promising bio-based value chains that are valid to the EU bioeconomy plans [12]. In many countries, the storage of municipal solid waste (MSW) in landfills is still the primary method of disposal. Hence, the EU has imposed restrictions on storage of biodegradable waste as well as the waste with a specific energy value on landfills—only residual wastes that cannot be used in any recycling processes are allowed to be stored. In reality, not only significant amounts of organic fractions are retained, but also wastes bearing signs of industrial waste. The wastes generated by mechanical treatment (having the energy value above 6 MJ/kg) can be potentially recycled with the generation of waste-based fuel [13–15]. As a matter of fact, there was no political will to reduce or prevent the waste production in the EU countries in the past—common practice was passing them to landfills. In the absence of appropriate policies, the deposition of the prevailing part of the municipal waste stream in landfills was affected by the growing costs incurred by the use of alternative methods [16].

The research performed on closed landfills indicated the presence of an increased content of polycyclic aromatic hydrocarbons (PAHs) and other organic compounds that contaminate the nearby groundwater. The noticeable relationship between the content of ammonium nitrogen in surface/groundwater and the increment in the concentration of $\text{NH}_4\text{-N}$ might suggest that landfill leachate can pollute the surrounding environment considerably [17]. In fact, heavy metals and other chemical compounds can migrate to the ground or surface water as a result of inappropriate sewage management or unintended installation breakdowns in a landfill, which pose a real threat to human health and life. Moreover, the operational problems in technological processes of water treatment at water intakes can endanger the quality of water overall [18,19]. In addition, the susceptibility of groundwater to contamination is higher the closer the river basin area is to the examined facility. Not surprisingly, this is correlated with the geological features and structure of a given area. In fact, a number of highly permeable formations grow in the phreatic zone, which negatively affects the class of groundwater—areas with high susceptibility to anthropogenic influence (including landfills) should be treated as high priority [20].

The uncontrolled emissions of landfill gas (LFG) to the air is another landfill-related threat to the nature exerted by municipal waste composed of the biodegradable fractions. On the whole,

rational water management and rational waste management are important mitigation and adaptation strategies in the circular economy era in order to counter climate change [21].

On the other hand, the increasing public awareness of human health hazards is among the major driving forces to reduce the LFG generation—mainly CH₄, CO₂ and O₂ (overall ≈90% of its content) in the transition to the circular economy (CE) model in Europe [22,23]. The main issue in the operation of municipal waste landfills is the emission of two gases—i.e., CH₄ and CO₂—which are also the basic greenhouse gases [24,25]. Current findings in the field of CH₄ emissions from landfills does not exhaust the topic due to their local conditions leaving a gap to fill—the undertaken research aims to supplement the knowledge in this area.

In response to the growing demand for energy, a part of CH₄ generated in landfills is recovered in an organized manner and subsequently used for power production as a source of renewable energy—e.g., in cogeneration plants (Combined Heat and Power; CHPs). The remaining unrecognized amount of CH₄ accounts for fugitive emissions from the landfill surface to the atmosphere along with carbon dioxide in biogas [26]. It seems that the use of renewable energy sources (including landfill gas) is an alternative to improve the energy balance of individual countries with the depletion of fossil fuel resources [27]. The diversification of energy generation sources also applies to the other types of biogas (not only LFG), as part of energy policies adopted by individual countries. In terms of municipal management, biogas generated from municipal sewage sludge in a sewage treatment plant can be applied as a source of renewable energy, not only for the production of electricity and heat in the cogeneration process, but also, e.g., as a gaseous fuel (bioCH₄) after the conversion of biogas to bioCH₄—e.g., for motor vehicle engines [28]. Therefore, a justified need to search for highly effective methods of removing the contaminants from water and wastewater has been recognized [29]. Unfortunately, before the conversion to heat or electricity, the biogas must be cleaned of hydrogen sulfide (H₂S), a colorless, flammable and toxic odorant. The main issue related to high concentration of H₂S in biogas is its corrosive impact on some engine parts of cogeneration units and the formation of sulfur oxide (SO_x) in the result of the combustion of H₂S [30].

In order to avoid the negative impact of waste treatment and neutralization processes on the natural environment, the introduction of a waste flow management system with a life cycle assessment (LCA) as a support of decision-making processes is justified [31]. Hence, the emission of greenhouse gases (including carbon dioxide) is being monitored permanently. The observed increment of CO₂ emission enforced the search for process refinement, which not only reduces its emissions significantly, but also meets the economy-driven operational criteria of the installation [32]. Obviously, gas emission from landfill is related to both site conditions and environmental changes; however, few data are available that describe this linkage adequately [33].

The objective of the study was to perform a parametric analysis covering the production, emission and neutralization of CH₄ in a real facility. In order to estimate the potential CH₄ production in a waste deposit and the amount of the CH₄ emission, the First Order Decay Model was utilized, which is based on Intergovernmental Panel on Climate Change (IPCC) methodology. In fact, the results of tests, measurements and operational data of the landfill were implemented as the input model data. The model findings proved that gas is produced in the landfilled waste deposit, therefore its collection and neutralization are required. Unfortunately, the performed calculations showed a pretty low efficiency of gas recovery in the result of low capacity of the operating passive degassing installation in relation to the total CH₄ production in the waste deposit. The performed cluster analysis might be valuable in adapting the existing wells to their prospective use in the active degassing installations.

2. Materials and Methods

2.1. Case Study Description

The object of the research involves a municipal waste landfill located in the south of Poland, with an area of approx. 2.25 ha and a capacity of approx. 480 thousand m³. The landfill has a natural

sealing from the ground with an insulating material and additionally with Polyethylene High-Density (PEHD) foil. This is equipped with an under-foil drainage system for groundwater and an over-foil drainage system for leachate, respectively. The leachates from the landfill are treated and discharged into a sanitary sewage system located near the landfill. The landfill stores municipal waste after mechanical treatment in the amount of about 25 to 30 thousand Mg/year. During the research period, the landfill was in its ninth year of operation—it was filled to $\approx 50\%$ (percent) containing mainly wastes after mechanical treatment with average content (Total Organic Carbon) $\text{TOC} = 0.061$. In order to capture biogas, at the construction stage of the landfill, a passive degassing installation was made which consisted of 9 vertical degassing wells, each with an individual gas flare. Under its own pressure, landfill gas (LFG) is transported to each flare, where the thermal neutralization of CH_4 takes place without energy recovery.

The passive degassing installation was designed to capture CH_4 (an important component of landfill gas), which is formed spontaneously along with carbon dioxide in landfills via anaerobic decomposition of organic matter. The process of landfill gas formation is influenced by a number of internal and external factors, including waste quality and moisture, temperature, pH, anaerobiosis, nitrogen content, the degree of waste compaction, alkalinity and the presence of microorganisms, respectively, amongst others. In the processes of biochemical transformations in the waste deposit, organic components are subjected to hydrolysis, and subsequently the participation of initially aerobic bacteria (later mainly with anaerobic bacteria) waste is decomposed into organic acids, and finally into gases: CH_4 and CO_2 [34,35]. The mass of coal that can be biologically converted to CH_4 per ton of mixed municipal waste under the anaerobic conditions of a landfill is ≈ 160 kg.

The gas generated in the waste deposit is a mixture of CH_4 , CO_2 , O_2 , N_2 and other compounds, containing mainly carbon, sulfur and chlorine derivatives. The biogas density is lower compared to the air density ($1.293 \text{ kg}\cdot\text{m}^{-3}$ under normal conditions), which has a significant impact on fugitive emissions from the landfill. The monitoring performed confirms the presence of the above-mentioned gases within the following ranges: CH_4 36.2–49.2%, CO_2 24.3–34.5%, O_2 , 1.8–6.1% and N_2 14.1–32.8%, respectively. An active degassing system is currently a standard for this type of landfills, where gas recovered from the landfill deposit via a suction pipe is used for energy generation purposes [36,37].

2.2. Research on the Morphological Composition of Waste

The obtained test findings allow one to determine the content of organic carbon in the deposited waste. Moreover, the test results will constitute input data for the IPCC model in order to estimate the potential production of CH_4 in the waste deposit.

The laboratory sample (weighing approximately 5 kg) was collected in compliance with the requirements of the standard PN-EN 14899 (Polish norm in English version): 2006 with reference to waste sample collection for laboratory tests. Then, the sample was separated into two fractions using a sieve: fraction No. 1 with particle size < 10 mm and fraction No. 2 with particle size ≥ 10 mm.

Additionally, individual components were selected from the fraction remaining on the sieve including: food waste of plant origin, food waste of animal origin, paper and cardboard waste, plastic waste, waste textile materials, glass waste, metal waste, other organic and mineral waste as well. All selected components from fractions No. 1 and No. 2 were weighed with an accuracy of 0.5 g. The determination of the morphological composition of municipal solid waste was conducted following the standard PN-EN 15002: 2015-07. The percentage content of fraction No. 1 and the individual components of fraction No. 2 (X_n) are expressed as a percentage quantity according to Formula (1):

$$X_n = \frac{m_w}{m} \times 100\% \quad (1)$$

where:

m_w —mass of the individual waste components,

m —mass of the sample collected for examination.

The results of the tests on the morphology of landfilled waste are reported in Table 1.

Table 1. Morphological composition of waste.

No.	Component Name of Waste Fraction	Characteristics of Component	Content of Individual Components, %
1.	Fraction < 10 mm	Remains from the mechanical treatment of waste	39.3
2.	Food waste of plant origin	Remains of plant substances generated during meal preparation	3.1
3.	Paper and cardboard waste	Remains of paper and cardboard products	6.6
4.	Plastic waste	Remains of plastic products	13.1
5.	Textile waste	Remains of products made of wool, cotton, linen and chemical fibers	4.8
6.	Glass waste	Remains of glass products and glass cullet	7.5
7.	Metal waste	Remains of products made from all types of metal and metal scrap	0.5
8.	Other organic waste	Organic waste remaining after the selection of components 1 ÷ 5	6.3
9.	Mineral waste	Mineral waste remaining after the selection of components 5 ÷ 6	18.8

2.3. Measurements of Landfill Gas

A passive degassing installation with 9 vertical wells was in operation during the research at the landfill site. Based on the natural pressure difference, the generated biogas was burnt in individual flares installed on each well without energy recovery. Each gas flare consists of a pipe supplying gas from the degassing well, manual shut-off/control valve, combustion chamber and gas sampling point, respectively. The measurements of biogas parameters were performed before the inflow to each flare four times a month for the period of one year (from January 1 to December 31) in the ninth year of landfill operation. In the course of the research, 434 samplings of biogas parameters were made for all wells. The measurements of the gas composition, the neutralization and CH₄ emission as well as the parameters of the degassing installation were subjected to statistical analysis using the Statistica program ver. 13.3 by TIBCOI Software Inc., Palo Alto, CA, USA [38]. Characteristic values were determined with reference to the above-mentioned indicators, and the correlations of the variables with scatterplots, variability plots and cluster analysis were performed as well. The concentration and flow of the landfill biogas were measured in line with the operative procedures based on the obtained accreditation. The emission of individual gases was calculated using the gas flow rate (made with an anemometer), the percentage contents of individual gases, the diameter of the degassing well, pressure, temperature and relative humidity of the biogas. The content of CH₄ and CO₂ was measured with a gas analyzer using the absorption of IR radiation, while the concentration of O₂ was specified using the electrochemical method.

The percentage content and the amount of CH₄ emissions are the input parameters to be implemented in the first-order decay model.

2.4. Model of Emission and Production of CH₄ in a Landfill

In order to estimate the potential CH₄ emission from the surface of a landfill in operation, the First Order Decay Model (IPCC Waste Model) was applied, based on the IPCC standard method for input parameters of a landfill [39]. The IPCC method allows one to estimate the annual amount of CH₄ generation in a waste deposit and the amount of the gas emission as the fugitive emission from the landfill surface to the air [40]. It is assumed that the decomposable organic components contained in the waste decompose slowly over many years, generating CH₄ and CO₂ emissions, respectively. In order to estimate the potential production of CH₄ L_O , Equation (2) was used based on the performed tests and measurements as follows:

$$L_O = MCF \times DOC \times DOC_F \times F \times \frac{16}{12} \left[GgCH_4 / Gg_W \right] \quad (2)$$

where:

L_O —potential CH₄ production [GgCH₄/Gg waste],

$MCF-CH_4$ —correction factor (depending on the type of landfill),

DOC_F —fraction of carbon that eventually undergoes decomposition,

$F\text{-CH}_4$ —content by volume in the landfill gas,

DOC —carbon content in the biodegradable organic in a given year [GgC/Gg of waste].

The content of carbon in the biodegradable organic part deposited in the landfill in a specific year (DOC) was calculated according to Formula (3), which takes into account the morphological analysis of the waste deposited in the landfill in percentage content by mass:

$$DOC = (0.4 \times A) + (0.17 \times B) + (0.15 \times C) + (0.3 \times D) \text{ [GgC/Gg waste]} \quad (3)$$

where:

A —waste such as paper, cardboard and textiles,

B —waste from parks, gardens and other organic waste, excluding food,

C —food waste,

D —wood and straw waste.

In order to estimate the potential volume of CH_4 emission from the landfill surface, Equation (4) was applied regarding operational data of the landfill.

$$E_{\text{CH}_4} = [(MSW_T \times MSW_M \times L_O) - R] \times (1 - OX) \text{ [Gg}\cdot\text{y}^{-1}] \quad (4)$$

where:

E_{CH_4} —potential volume of CH_4 emission [Gg $\cdot\text{y}^{-1}$],

MSW_T —total mass of municipal waste deposited in the landfill over a year [Gg],

MSW_M —a part of municipal waste collected from the landfill [Gg $\cdot\text{y}^{-1}$],

R —the volume of CH_4 from the landfill, recovered and neutralized in the landfill [Gg $\cdot\text{y}^{-1}$],

$OX\text{-CH}_4$ —oxidation index in soil or in the material covering the waste.

In the implemented IPCC Waste Model, the emission of CH_4 from the landfill is at its highest a few years after the deposition, and then it gradually decreases until the carbon contained in the waste is broken down by bacteria. The method properly estimates the annual emission for landfills, where the quantity and morphology of the deposited waste are known and the morphology changes insignificantly within a few years [41].

3. Results

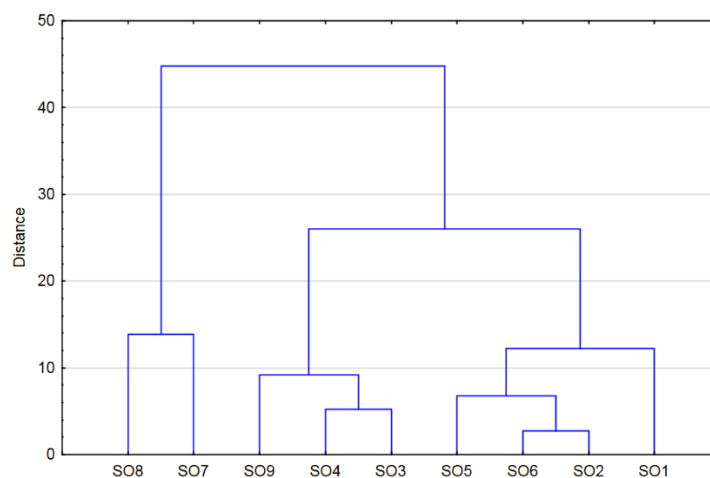
The morphology of the landfilled waste is examined once a year as part of the obligatory environmental monitoring. The morphology of the landfilled waste was scrutinized in the set the laboratory tests as reported in Table 1.

The analysis of the landfilled waste sample indicated the highest percentage of waste with a fine fraction <10 mm (39.3%) composed of the remains after the mechanical treatment of waste on the sorting line. Mineral waste was the second type of waste with a percentage of 18.8%. The remaining waste, including biodegradable waste, accounted for 41.9%. In the group of waste containing the biodegradable fraction, the greatest share is paper and cardboard waste (6.6%), then other organic waste (6.3%) and textile waste (4.8%). The percentages of these wastes show a low efficiency of the mechanical treatment process as well as the low level of source segregation in this region. The measurements of the landfill gas parameters collected from the operating passive degassing installation in nine vertical wells demonstrated that methanogenic processes are taking place in the waste deposit. The average monthly contents of the individual parameters of biogas measured at the inlet to the collective flare over a year are presented in Table 2.

Table 2. Parameters of landfill biogas in gas flares.

Number of Well/Flare	Percentage Content of Individual Parameters in Biogas				Stream of Biogas Volume in the Flare (Nm ³ ·h ⁻¹)	Biogas Neutralized in the Flare (Nm ³ ·y ⁻¹)	CH ₄ Neutralized in the Flare (kg·h ⁻¹)	Annual Volume of Neutralized CH ₄ in the Flare (Mg·y ⁻¹)
	CH ₄	CO ₂	O ₂	N ₂				
SO1	49.2	34.5	1.8	14.1	6.2	54,312	2.20	19.24
SO2	46.6	29.8	3.1	20.3	5.4	47,304	1.81	15.87
SO3	45.6	25.2	3.7	24.8	4.3	37,668	1.77	15.53
SO4	44.5	28.2	4.3	22.5	4.5	39,420	1.44	12.63
SO5	48.4	33.2	2.5	15.6	5.2	45,552	1.81	15.87
SO6	46.7	31.2	2.1	19.5	5.2	45,552	1.75	15.32
SO7	37.2	25.3	5.7	31.4	4.6	40,296	1.23	10.79
SO8	36.2	24.3	6.1	32.8	3.1	27,156	0.81	7.08
SO9	47.8	31.3	2.2	18.3	4.3	37,668	1.48	12.96

The measurements as well as calculations indicated that the annual average basic parameters of landfill biogas from vertical degassing wells were as follows: CH₄ 44.69%, CO₂ 28.85%, O₂ 3.71% and N₂ 22.47%, respectively. Landfill biogas with a CH₄ content of about 45% has an energy value ≈ 16 MJ/m³. The annual amount of landfill biogas from nine wells in the landfill in the year under consideration was 374.928 m³, and the amount of thermally neutralized CH₄ was 125.3 Mg. In order to group the wells according to the gas parameters and their efficiency, a cluster analysis was performed on data stored in Table 2. In the cluster analysis (hierarchical tree), Ward's agglomeration method was selected, which uses the variance approach to estimate the distance between clusters [42]. Ward's method aims to minimize the sum of the squared deviations of any two clusters, which may be formed at any stage. As a measure of distance, the Euclidean metric was used with the individual distances calculated on the basis of the raw data. Figure 1 illustrates the results of hierarchical grouping of the wells in the form of a dendrogram.

**Figure 1.** Dendrogram of cluster analysis.

The objects form clusters, where four main groupings can be distinguished. The first one contains a single object (SO1), the second and the third groups are composed of three objects, while the fourth one comprises two objects (SO7, SO8). The shortest distance in the second cluster is represented by the objects SO6 and SO2, mainly due to the fact that they are characterized by comparable parameters for all eight variables. A single SO1 object differs in parameters from the remaining ones—it is assigned to a binary tree with similar characteristics.

The generated biogas from the waste deposit wells is burnt in nine individual flares. Figure 2 illustrates the parameters of biogas in well SO1, which exhibits the highest parameters of CH₄ and biogas stream. The average annual content of CH₄ in the biogas was 49.2%, with the biogas yield of 54.312 Mg y⁻¹. Moreover, the SO1 well had also the lowest oxygen content (1.8%).

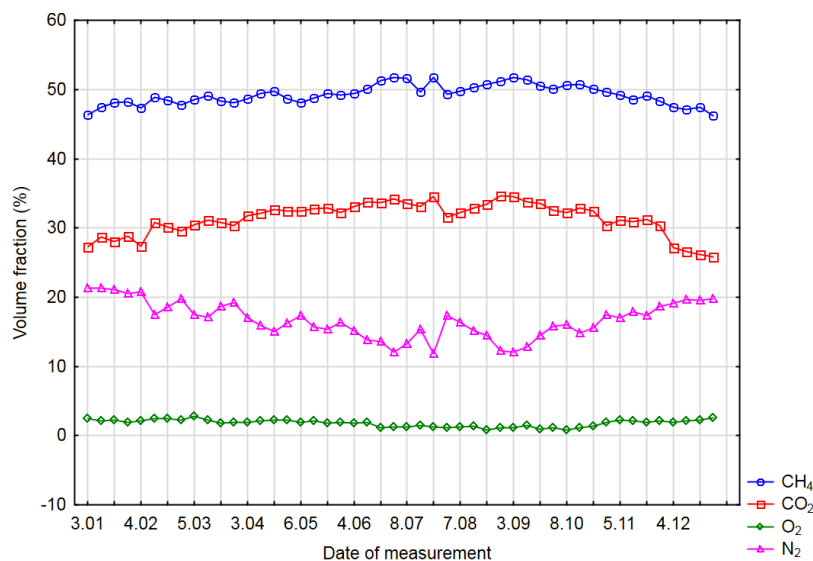


Figure 2. Biogas parameters in the single object (SO1) well.

On the other hand, the lowest values of parameters were found in the well SO8 (see Figure 2), with a CH₄ content of 36.2%, gas stream of 3.1 Nm³·h⁻¹ and oxygen content of 6.1%, respectively. The parameters of thermally neutralized biogas in individual gas flares indicate that the landfill is properly operated, including the degassing installation. The average annual capacity of passive degassing installation of the landfill is 4.76 m³ of biogas per hour.

Based on the morphology findings of the landfilled waste (see Table 1), the required parameters of waste reported in Table 3 were selected to calculate the content of carbon in the biodegradable organic part of the landfilled waste according to Formula (3).

Table 3. Percentage share of individual fractions of biodegradable waste.

Waste Category	Waste Type	Value
A—paper and textiles	Remains of products made of paper, cardboard and textiles	0.114
B—organic waste except for food	Organic waste from parks and gardens	0.063
C—waste made up of food	Remains of plant substances generated during preparation of meals	0.031
D—wood and straw	Not found	0

Due to the fact that in the waste morphology tests wood or straw waste were not identified, category D was ignored in further calculations. The content of carbon in the biodegradable organic part landfilled in the investigated year (DOC) was calculated based on Formula (3), which takes into account the morphological analysis of the landfilled waste in percentage content by mass as follows:

$$DOC = (0.4 \times 0.114) + (0.17 \times 0.063) + (0.15 \times 0.031) = 0.061 \text{ [GgC/Gg waste]}$$

The findings of the waste morphological composition were implemented into the mathematical model. In consequence, the content of carbon in the biodegradable organic part DOC was calculated with a value of 0.061 [GgC/Gg of waste]. In order to estimate the potential amount of CH₄ produced in the waste deposit, Equation (2) was used. The following input values for the model were used:

- MCF-CH₄ correction factor (depending on the type of landfill); the value was adopted at 1. Following the IPCC guidelines, it is recommended that, for organized landfills with sealing and a waste transfer system, waste is thickened with a compactor,
- DOC_F—carbon fraction, which ultimately undergoes decomposition; the value 0.5 (weight fraction) was adopted following the IPCC recommendations,
- F—volumetric content of CH₄ in the landfill gas; the value adopted on the basis of actual measurements as an annual average, amounting to 44.69%—i.e., 0.447,
- DOC—carbon content in the biodegradable organic part of the landfilled waste in a given year was calculated as follows:

$$L_O = 1 \times 0.061 \times 0.5 \times 0.447 \times \frac{16}{12} = 0.018 [\text{GgCH}_4/\text{Gg}_W]$$

The potential production of CH₄ in the waste deposit from one deposited Gg of the waste was 0.018 Gg in the investigated landfill (1 kilogram [kg] = 1 × 10⁻⁶ gigagram [Gg]).

In order to estimate the potential emission of CH₄ from the landfill, Equation (4) was used and the following input values for the model were implemented:

- MSW_T—total mass of municipal waste deposited in the landfill during the year based on operational data; adopted at 31.21 [Gg],
- MSW_M—the waste collected or extracted from the landfill; 0 was adopted since such a case did not occur,
- R—the amount of CH₄ generated in the landfill and neutralized in a collective flare based on the measurements; value of 0.1253 [Gg·y⁻¹] was adopted,
- OX-CH₄ oxidation index in the soil or in the material covering the waste, which reflects the amount of CH₄ oxidized in the soil or in other material covering the waste. For surface-sealed landfills with a material enabling the migration of oxygen (e.g., soil, compost); set at 0.1 according to the IPCC guidelines.

$$E_{CH_4} = [(31.20 \times 0.018) - 0.125] \times (1 - 0.1) = 0.393 [\text{Gg}\cdot\text{y}^{-1}]$$

The estimated amount of CH₄ emissions E_{CH_4} from the landfill surface, reduced by the amount of CH₄ neutralized in the collective flare and by CH₄ oxidation in the soil was 393.1 Mg of CH₄ per year.

Based on the theoretical calculations, the annual amount of CH₄ production from landfilled waste was estimated at level of 561.78 [MgCH₄ y⁻¹]. The adopted parameters allowed us to calculate the efficiency of passive degassing of the landfill installation according to Formula (5).

$$\eta_{grs} = \frac{P_{CH_4} - E_{CH_4} - O_{CH_4}}{P_{CH_4}} \times 100\% \quad (5)$$

where:

η_{grs} —the efficiency of passive degassing installation [%],

P_{CH_4} —potential CH₄ production 561.78 [MgCH₄·y⁻¹],

E_{CH_4} —estimated CH₄ emission from the landfill 393.1 [MgCH₄·y⁻¹],

O_{CH_4} —CH₄ oxidized in the soil 43.38 [MgCH₄·y⁻¹],

$$\eta_{grs} = \frac{561.78 - 393.1 - 43.38}{561.78} \times 100\% = 22.3\%$$

The investigated landfill has the installation of passive degassing with an efficiency of 22.3%. The remaining biogas that was neutralized in the flare accounts for fugitive emission from the landfill surface to the air. In fact, the efficiency of the degassing installation is relatively low to ensure the collection and neutralization of the remaining biogas, including CH₄ produced in the landfill.

4. Discussion

The conducted tests on the morphological composition of waste deposited in the investigated year indicated the presence of a biodegradable fraction made up by food waste of vegetable origin at 3.1%, paper and cardboard waste at 11.4% and other organic waste (excluding food waste) at 6.3%, respectively. The content of organic fraction in the analyzed sample is typical for waste subjected to a mechanical treatment. Based on the research findings, the calculated carbon content in the biodegradable organic part deposited in the landfill in a specific year (DOC) was 0.061 [GgC/Gg]. Higher values can be found elsewhere, but mixed wastes are mainly under consideration. Similar results were reported previously with DOC = 0.08 and L_0 at the level of 0.05 compared to 0.018 in this study [43]. The carbon content in the organic fraction of the landfilled waste can be reduced by the application of an additional mechanical treatment prior to a biological treatment process, where 56% reduction in biological activity was observed after 4 weeks of treatment [44,45].

The storage of waste containing an organic fraction turns the landfill into a kind of bioreactor generating biogas with CH_4 . For CH_4 , which accounts for 44.69% of the biogas, anaerobic and methanogenic conditions prevail in the waste deposit [46]. The process of CH_4 generation results from the presence of an organic fraction in the landfilled waste left over after the mechanical process; however, it is also dependent on oxygen content, temperature and humidity. Such a state is conducive to the generation of CH_4 emissions from the landfill surface to the atmosphere, based on the implemented model and numerical simulations as presented previously [47]. A model to simulate the production of landfill gas and gas migration processes within and beyond the boundaries of landfills was proposed as well. It was proven that gas emissions from the landfill increase with the decrease in the atmospheric pressure—the numerical parameters confirmed the mechanisms of gas migration, as well as the other transfer-related processes. Obviously, the extraction of landfill gas and the subsequent neutralization of CH_4 is a priority in curbing the negative effects (greenhouse gas) of climate change [48]. The options of gas management at the landfill site were investigated as well [49]. The acquisition of gas and its application for energy generation in Combined Heat and Power (CHP) systems or the conversion to bio- CH_4 , or its employment for the synthesis of gas with high CH_4 through catalytic reforming were specified as the main goals [50]. The research on the content of biogas parameters conducted during the year demonstrated that the average annual O_2 values were at the level of 3.71% and CO_2 in the amount of 28.85%. Such results bespeak the ongoing methanogenic phase and partial leakage of degassing wells at their above-ground part. Unfortunately, the applied passive degassing system does not provide optimal conditions for the collection of biogas and the neutralization of CH_4 [51].

On the basis of the developed gas measurements, diagrams of the correlation between CH_4 , CO_2 and O_2 are provided. The diagram in Figure 3 depicts the dependence between CH_4 and O_2 in the landfill biogas generated in nine degassing wells (as annual average values).

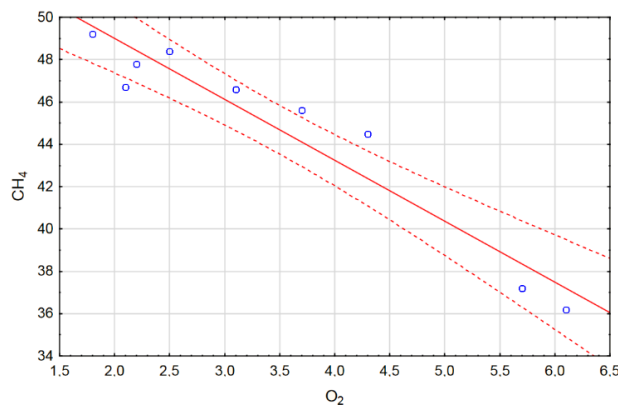


Figure 3. Scatterplot of the CH_4 and O_2 correlation.

The scatterplot illustrates the negative correlation between CH_4 and O_2 —the increase in the results of the variable Y (CH_4) is accompanied by the decrease in the second variable X (O_2), respectively. The least correlated points are the wells SO4 ($x = 4.3$; $y = 44.5$) and SO6 ($x = 2.1$; $y = 46.7$). The content of oxygen in the individual wells, ranging from 1.8 to 6.1%, indicates that oxygen is being sucked into the landfill from outside—i.e., the gas wells are leaking. A similar mechanism of action was noticed previously with attention paid to the quality of the surface structure of the landfill with some fissures as well as cracks resulting from uneven settlement of waste and improper compaction of waste [52].

In the process of biogas generation in the landfilled waste deposit, the most methanogenic substances are mainly carbohydrates, fats and proteins; therefore, biogas with high CH_4 content is generated. The higher the quotient CH_4/CO_2 the higher the energy value of the biogas. Figure 4 presents the graph of average annual values from nine wells with the correlation between CO_2 and CH_4 in the biogas collected at their outlet.

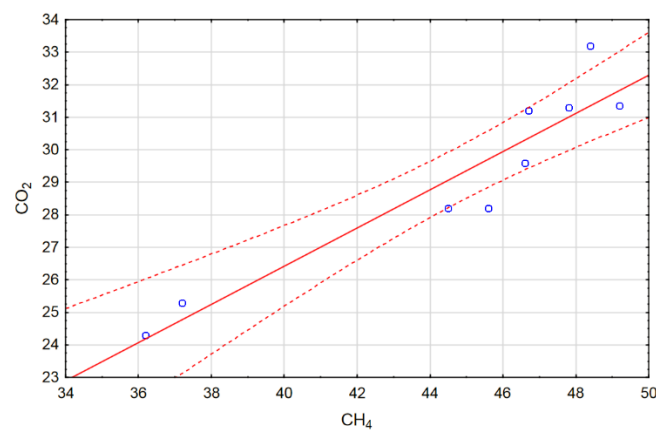


Figure 4. Scatterplot of the correlation between CO_2 and CH_4 .

The scatterplot in Figure 4 shows the positive correlation between CO_2 and CH_4 —the increase in the results of the variable Y (CO_2) is accompanied by the increase in the second variable X (CH_4). The least correlated points are wells SO3 ($x = 45.6$; $y = 28.2$) and SO5 ($x = 48.4$; $y = 32.3$). The average annual value of CH_4 in the wells ranged from 49.2 to 36.2%, while the content of CO_2 ranged from 34.5 to 24.3%, respectively. The percentage composition of main gas bespeaks the ongoing methanogenic processes in the waste deposit. Similarly, the previous findings provided the mechanisms of biogas generation in the landfilled waste content of CH_4 and CO_2 , indicating the large fluctuations that are related to the conditions of gas generation [53,54]. As a matter of fact, the following conclusions has been drawn: the composition of the produced gas may have large fluctuations, despite the fact that internal production rates are constant and that the extent of stoichiometric fluctuations will increase with the decrease in gas production [55]. This means that gas composition displays hysteresis, and it is hard to determine the degradation state of the object on the basis of gas composition measurements performed once or at rare time intervals. Methanogenic processes that take place in the waste deposit depend on both external and internal factors, respectively. Hence, the composition of the gas produced, in particular, in a landfill in operation will depend, inter alia, on the level of waste compaction that has a profound impact on the formation of anaerobic conditions as well as on the migration of water from precipitation.

It was found that the average annual stream of gas volume amounted to $4.76 \text{ m}^3 \cdot \text{h}^{-1}$ for the passive degassing installation composed of nine individual vertical degassing wells with gas burnt in the individual flares. Hence, it possible to generate 374.928 m^3 of gas from the waste deposit during the year, which enables the neutralization of 125.3 of $\text{Mg CH}_4 \cdot \text{y}^{-1}$. This allows the generation of approximately 525 MWh of electricity per year in the CHP system with an efficiency of about 40% [56]. Figure 5 presents the average annual CH_4 content in the gas and the capacity of the degassing installation.

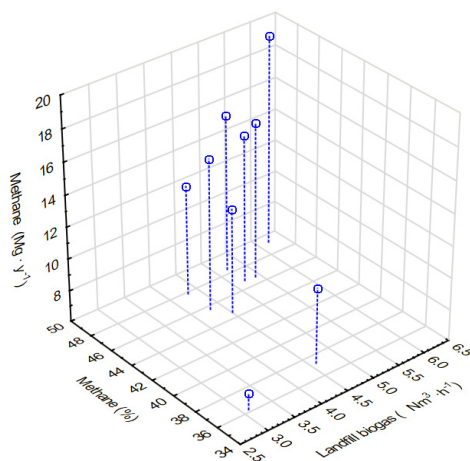


Figure 5. Capacity of the degassing installation.

In reality, the passive degassing system with individual flares is widely used in landfills as an intermediary solution to biogas combustion with energy recovery in CHP. The prevailing belief that flares burn landfill gas with 100% efficiency was appealing, therefore further studies were conducted [57]. Hence, the numerical approach for emission specification was proposed to determine the gas composition in the flare, combustion conditions, throughput, wind speed, gas temperature, ambient temperature and atmospheric pressure, respectively. It was proven that the combustion efficiency drops sharply, while the content of CH_4 in the gases drops below 50%. For instance, with a 50% content of CH_4 in landfill gas, the efficiency of its combustion in a flare is about 80%, whereas when the CH_4 s content equal to 60% it approximates 92%. Regarding the previous findings, the efficiency of landfill gas combustion conducted in nine flares is foreseen to oscillate at approximately 80%. The use of passive carbon or biological lifting seems to be an appealing alternative to the use of individual flares for CH_4 neutralization. The project of furnishing selected French landfills with a biomitigation system for the oxidation of CH_4 has been described previously in detail [58].

The performed cluster analysis aims to distinguish the wells that have similar gas parameters and efficiencies in the context of building an active landfill degassing installation. This allows one to classify the wells according to their future operational applications, and to perform remedial actions (e.g., sealing) in order to limit the access of oxygen to the landfill gas. Moreover, the hierarchical grouping enables one to cluster the newly built wells with regard to the operational parameters and the parameterization of gas inlets, respectively.

The implemented IPCC first-order decay model is a supporting tool in the estimation of CH_4 production and the potential emission from the landfill area. It is important to determine the input tests/measurements parameters, including both the percentage of CH_4 in the biogas and the volume, that were neutralized (R). The practical application of the IPCC model confirmed that the actual data are important (not the estimated ones) [59]. For instance, the model demonstrates high sensitivity of DOC_F and DOC parameters. Even though, a multitude of properties determine the gas landfill production, these variables are connected with empirical relationships; therefore, it is tough to retrieve accurate information regarding gas generation. It should be emphasized that accessible landfill data also contain sources of uncertainty that make them almost insufficient [60].

Hence, some input data, e.g., DOC , R, MSW_T and F, were factual data—in this case the modeling results could be regarded as optimal. In fact, the parameter with a dominant impact on the final result is the content of carbon in the biodegradable organic part deposited in the landfill in a specific year (DOC). Component A (the residue of paper and cardboard products) was implemented in the model on the basis of the morphological composition of waste with a value of 11.4%. Thus, the model generated the total production of CH_4 at the level of $\text{MgCH}_4 \cdot \text{y}^{-1}$, while a change of component A's value by 1% upward will result in the increment of CH_4 production P_T by 9.8%, and the enhancement

of E_{CH_4} emission by 12.2%, respectively. Figure 6 illustrates the prediction of the quantity $A = 11.4\%$ implemented into the model—the forecast raised by 1%, i.e., $A = 12.4\%$.

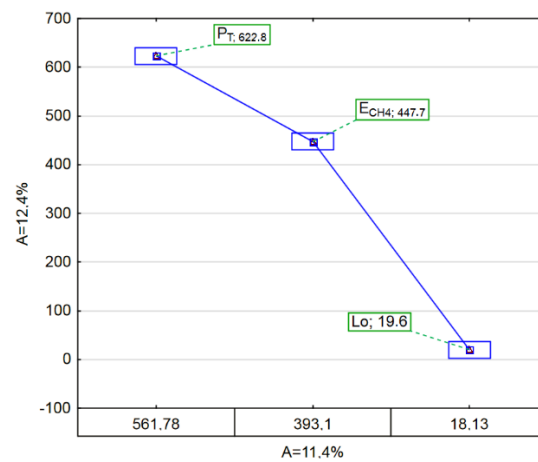


Figure 6. Dependence of modeling results on component A.

Moreover, a significant impact of DOC_F parameter (carbon fraction ultimately subjected to decomposition) on the final result was observed. In fact, the value of $DOC_F = 0.5$ was implemented in the IPCC model. It was foreseen (see Figure 7) that the growth of DOC_F by 0.1 results in the increment of CH_4 production by 19.9% and the increase in emissions by 24.2%, respectively.

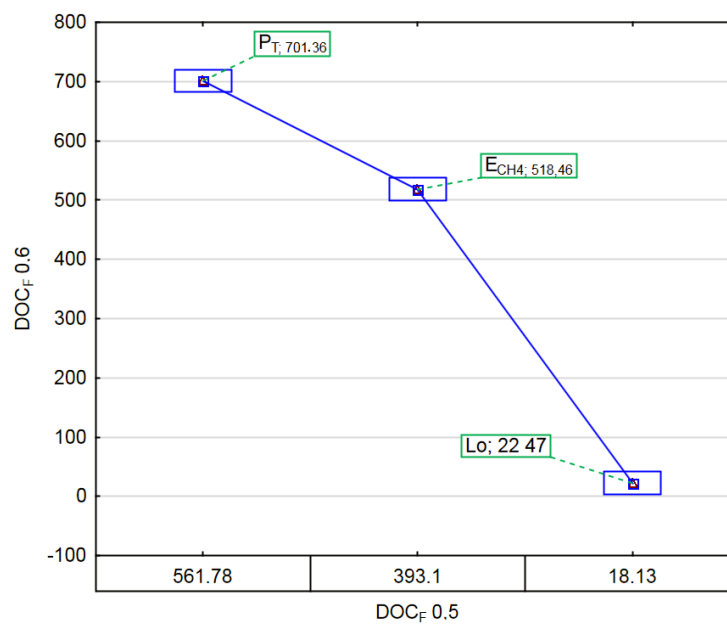


Figure 7. Prediction of DOC_F parameter variations.

Similar dependence was observed previously with the fluctuation of DOC_F in the range of $0.5 \div 0.77$. The former default value of $DOC_F = 0.77$ was replaced by the current default value of 0.5 (recommended now by IPCC) [61]. The Swedish findings demonstrated that the model does not cover the arguments for $DOC_F < 0.5$, while the DOC_F for different waste fractions does not change considerably over longer time horizons [41]. Moreover, the results of CH_4 emission modeling from landfills based on the variety of methodologies are available [62]. For instance, the comparison of CH_4 emission approximated with the IPCC methodology versus the Land-GEM (Land Gas Emissions Model) indicated that the final outcome of the IPCC methodology was higher by 13.1%, while the CH_4 production was lower by 5.5%, respectively. The estimation methods for the emission from the Legoli

landfill (Tuscany, Italy) based on three different models (LandGem v3.02, IPCC 2006 and GasSim Lite 1.54) were compared with the findings of spot measurements performed in the landfill with the application of an accumulation chamber and the use of the Kriging method [40]. Not surprisingly, the comparison of the model-driven parameters with the empirical measurements demonstrated that the calculated variables do not reflect real working conditions of the surroundings—direct changes occurring in the landfill should be taken into consideration as well. In fact, the most convergent findings to experimental data were specified using the LandGem and IPCC models, respectively. A release of CH₄, which is a potent greenhouse gas, can be reduced by in situ oxidation in the bioactive cover layer. Technically, the organic-rich porous materials (e.g., compost) are engaged in the process. Industrially engineered biocovers are characterized by the homogeneous particle sizes and an active microorganism consortium. In fact, the potential of CH₄ oxidation in biocovers is strongly related to their chemical/physical characteristics, the age and seasonal variations of the cover material as well as the actual concentration of CH₄ [36].

Based on the conducted parametric analysis, the efficiency at the level of 22.3% was revealed for the passive degassing installation composed of nine vertical degassing wells with individual flares. It should be emphasized that, even in modern landfills, it is not possible to reach 100% of CH₄ efficiency in the degassing installation due to the fugitive emission [63]. On the other hand, an active degassing installation with gas used for energy generation can have an efficiency in the range of 67 ÷ 84%. Of paramount significance is the use of impermeable transfer layers in landfills. Moreover, it was suggested to split the recovery installations of CH₄ into two groups. The first one with an efficiency within the range of 14 ÷ 41%, and the second one ranging from 48 to 78%, respectively. Lower efficiency involves landfills, where the total production of CH₄ is in the range of 229 to 705 kg·h⁻¹ [41]. Unfortunately, the efficiency of presented gas recovery installation (22.3%) does not ensure the optimal reduction in the level of CH₄ emissions from the landfill since the remaining part of CH₄ is unorganized (fugitive emission). As a matter of fact, the modernization or expansion of the landfill degassing system should be considered as well. In the case of the active installation construction, which sucks in gas from a waste deposit with an efficiency of, e.g., 75%, and current CH₄ production amounting to 561.78 MgCH₄·y⁻¹, the foreseen amount of CH₄ neutralization should reach approximately 413.5 MgCH₄·y⁻¹. Hence, the optimal solution seems to be a use of CH₄ for energy production in the cogeneration system for generating electricity or heat.

5. Conclusions and Recommendations

The main objective of the study was to perform a parametric analysis covering the production, emission and neutralization of CH₄ in a real facility (a landfill). The results of the morphology studies of the landfilled waste demonstrated that the carbon content in the dissolved organic part deposited in the landfill was equal to 0.061 [GgC/Gg waste]. The measurements of gas parameters performed in the period of one year revealed that the average annual content of CH₄ was 44.69%, and CO₂ was 28.85%, respectively. Such gas parameters indicate that the landfill is in phase IV—the methanogenic one, where anaerobic (methanogenic) decomposition is dominant, characterized by a stabilized composition of transformation products. In order to estimate the potential of CH₄ production in a waste deposit and the amount of its emission, the First Order Decay Model was used, which is based on the IPCC methodology. In fact, the results of tests, measurements and operational data of the landfill were implemented as the input model data. Hence, potential production of CH₄ in the waste deposit was determined at the level of 561.78 [MgCH₄ y⁻¹] and its fugitive emission from the landfill surface to the air was approximated to reach the amount of 393.1 [MgCH₄ y⁻¹]. The negative correlation between CH₄ and O₂ was revealed, where the least correlated points are wells SO4 (x = 4.3; y = 44.5) and SO6 (x = 2.1; y = 46.7), respectively. On the other hand, a positive correlation between CO₂ and CH₄ was noticed, where the least correlated points are wells SO3 (x = 45.6; y = 28.2) and SO5 (x = 48.4; y = 32.3). The total production of CH₄ for an A value of 11.4% (the residue of paper and

cardboard products) is estimated by the model at the level of 561.78 MgCH₄·y⁻¹. A 1% increase in the A value will increase P_T CH₄ production by 9.8% and E_{CH₄} emissions by 12.2%.

Not surprisingly, it is better to use the actual parameters as the input data into the methodology—the results are more reliable. The model findings proved that off-gas is produced in the landfilled waste deposit, therefore its collection and neutralization are required. As a matter of fact, the passive degassing installation in the landfill neutralized CH₄ in the amount of 125.3 [Gg y⁻¹] in nine flares. Unfortunately, the performed calculations showed a pretty low efficiency of gas recovery (22.3%) in the result of the low capacity operating passive degassing installation in relation to the total CH₄ production in the waste deposit. In consequence, it is recommended to install an active recovery system of biogas in a cogeneration unit to use off-gas as a source of energy. The use of CH₄ neutralized in flares for energy purposes may result in the production of about 600 MWh of electricity, of which the landfill will use about 40% for its own needs, and the surplus will be transferred to the power grid. It has been proven so far that the replacement of a passive installation with one that sucks in gas from a waste deposit increases the CH₄ content in the landfill gas—this results in higher fuel value. Moreover, the performed cluster analysis might be valuable for adapting the existing wells to their prospective use in the active degassing installations. The produced energy from a renewable source can be used for the facility's own needs, and its surplus can be sent over to the external power grid or stored in the energy storage bank as well. Basically, the results indicate an unfavorable impact of the landfill on the air in consequence of mainly CH₄ emissions, therefore the implementation of technology limiting this phenomenon in the first place, and ultimately the need to limit the storage of waste containing biodegradable fractions is necessary.

The proposed methodological approach can be used in landfills with similar technical solutions and comparable parameters of CH₄ and gas stream, respectively. Moreover, the described procedure can be helpful to diagnose the current state of landfill and forecast the potential gas production as well as provide recommendations for more effective technical solutions. The research results are in line with the principles of sustainable development, including development through the exchange of scientific and technological knowledge, as well as the dissemination and transfer of new technologies. The proper functioning of the waste management system in the region must take into account the technical, environmental, economic and social objectives. The impact of waste neutralization technology on the natural environment is influenced not only by technological solutions, but also by the location and the so-called background conditions or the existing state.

The rapid intensification of cooperation between industry and academia is urgently needed in the field of emission and neutralization of methane from a municipal landfill.

Author Contributions: J.C., V.K., A.G., K.G., A.B., M.P. and K.B.—conceptualization, methodology, investigation, formal analysis, article writing and editing, data curation. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Acknowledgments: The authors would like to thank the anonymous reviewers for their helpful comments.

Conflicts of Interest: The authors declare no conflict of interest.

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