

Assessment of landfill gas composition dynamics in the operational and post-operational phases of municipal solid waste landfill sites

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ABSTRACT

This study aimed to conduct a comparative analysis of the gas compositions at two municipal waste landfill sites over the period 2018–2023. The novelty of the study was the comparative analysis of landfill gas dynamics at municipal waste landfill sites with different levels of organization and at different operational and post-operational stages. One site was unorganised, and the other one was organised. The unorganised site was used for approximately 10 years longer than the organised site, and over 15 years had passed since the end of waste disposal there. The landfill gas study involved measurements of CH₄, O₂, and CO₂, as well as the temperature and gas flow, from two landfill sites, primarily in H1 and H2. The unorganised site, which was still in the non-operational phase, exhibited an increase in oxygen (O₂) concentration, which peaked at almost 18%. By contrast, the methane (CH₄) concentration decreased to below 5%, indicating limited anaerobic conditions. On the other hand, high mean CH₄ content (~65%), temperature (~17 °C), and CH₄/CO₂ ratio (2.7) were observed at the organised landfill site, along with a negative correlation between CH₄ and O₂, confirming the dominance of methanogenic processes. The simultaneous increase in temperature, CH₄, and O₂ in the second half of the study period might depend on changing environmental conditions. The moderate correlation between CH₄ and CO₂ at both landfill sites reflects a similar trend in their biochemical processes, despite differences related to their operational and post-operational periods.

Keywords: landfill site, landfill gas, biogas composition, methane content, statistics.

INTRODUCTION

More than 2 billion tonnes of waste are generated worldwide each year, including in Europe, with the largest share coming from the construction sector and municipal waste. Thus, measures are required to increase the efficiency of waste management (Przydatek, 2016; Shah et al., 2023). Landfill sites often contribute to the deterioration of air, water, and soil quality and harm human health because they generate landfill gas (LFG) and leachate (Siddiqua et al., 2022; Vaverková, 2019). Furthermore, landfilled organic waste contributes to greenhouse gas emissions. Such waste is an important source of anthropogenic methane (CH₄) and represents one of the key sources of emissions in Europe (Mønster et

al. 2019). CH₄ is produced by the anaerobic decomposition of organic matter during waste deposition (Boëriessson et al., 1998; Scheutz et al., 2009; Porowska, 2021).

The amount of LFG produced in landfill sites depends mainly on the organisation of the landfill (i.e., its structure, the equipment used for environmental protection, and the type of sealing used), the amount and composition of the waste, and the operating conditions (Bouazza and Vangpaisal, 2006; Ersoy et al., 2013). Ozbay et al. (2021) differentiated landfill sites based on the type of waste deposited and the infrastructure used to minimise the environmental impact of a given type of waste. Oxygen (O₂) remains a key indicator of aerobic and anaerobic conditions of landfill sites. Nevertheless, aerobic and

anaerobic processes depend on the content of the biodegradable fraction (Christensen et al., 2020, Manheim et al., 2021).

After following waste deposition, the biogas composition (especially the proportions of O_2 , CO_2 , CH_4 , and N_2) changed from aerobic conditions in phase I to anaerobic conditions. Phases II–III are characterised by intense fermentation and an increase in CO_2 , whereas phase IV is characterised by a dominance of CH_4 and CO_2 , and phase V is characterised by a decrease in biogas emissions, except for N_2 and O_2 (Figure 1). In particular, the lack of surface sealing in landfills results in increasing gas emissions from waste decomposition (Borisova et al., 2023).

The novelty of this study includes the six-year comparative assessment of landfill gas composition and gas-generation dynamics at organised and unorganised municipal waste landfill sites located in close proximity and representing different stages of operational and post-operational.

According to Council Directive 999/31/EC (1999), LFG from sites where biodegradable waste is deposited must be managed and, where possible, used for energy purposes. Degassing using a passive or active system is a possible solution, both during landfill operation and after closure. Passive degassing involves burning LFG in an open or closed form to meet environmental protection requirements. By contrast, active degassing allows for the production of thermal and electrical energy during the combustion of LFG in a generator (Knutel et al., 2022).

Additionally, landfills should be monitored to ensure environmental protection. Monitoring should consider the qualitative and quantitative parameters of the waste, without specifying a final date for the site closure (Scheutz and Kjeldsen

2019). When a site shuts down, LFG is estimated by the cessation of waste deposit activity.

MATERIALS AND METHODS

Two municipal waste landfill sites located in Lower Silesia, in the Central Sudetes macro-region (Kistowski et al., 2018), approximately 10 km away, were selected for the comparative analysis of waste deposition. Landfill site A ($50^{\circ}48'51.40''$ N; $16^{\circ}18'36.55''$ E) was operated in the years 1974–2001, whereas landfill site B ($50^{\circ}45'30.90''$ N; $16^{\circ}15'36.02''$ E) was active from 2002 to 2019. These facilities were subject to LFG studies: landfill site A in the post-mining phase (2018–2023), and landfill site B in the operational (2018–2019) and post-mining phases (2020–2023) (Figure 2).

Localisation of research facilities

LFG testing was conducted in two degassing wells at landfill site A and at the combined intake point before disposal at landfill site B (Figure 2). Following Kopera et al. (2023), the tests included determining the content of methane (CH_4), oxygen (O_2) and carbon dioxide (CO_2), as well as measuring the temperature and flow of LFG. A calibrated portable GA 2000 (Geotechnical Instruments (UK) Ltd.) gas analyser was used for these measurements at the studied landfill sites. In addition, precipitation totals from the nearest (10 km) facility of the Institute of Meteorology and Water Management were used.

LFG composition was analysed once every six months (H1 and H2) each year from 2018 to 2023 at landfill site A and every month in year

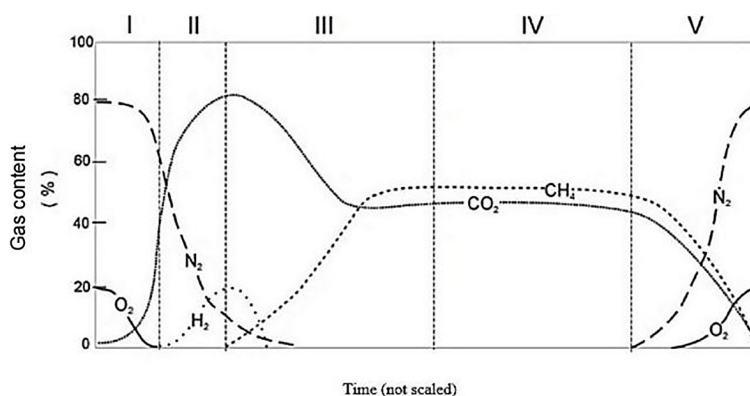


Figure 1. Waste degradation stages in a landfill, considering the change in landfill gas composition (Farquhar and Rover 1973; Kjeldsen and Christensen 2001)



Figure 2. Location of the research facilities in the southwestern part of Lower Silesia

from 2018 to 2019 and from 2022 to 2023 (January–June in H1, July–December in H2) and once every six months (H1 and H2) in each year from 2020 to 2021 at landfill site B, as part of an environmental monitoring program, and according to Regulation (2013). The administrator of the landfill sites provided the measurement data used in the analyses as part of a scientific cooperation.

Analysis of biogas composition

LFG composition was determined in situ, in two degassing wells at municipal waste landfill site A: S1, located on the outskirts of the eastern part of the landfill, and S7, located in its north-eastern part (Figure 3).

At landfill site B, biogas composition was measured before entering the biogas disposal installation (Figure 4).

- Municipal waste landfill site A – post-operational phase

The landfill site is located in a former ravine, between two hills, and is considered unorganised because of insufficient infrastructure. After 27 years of operation, the landfill was closed in 2001 and underwent technical and biological reclamation (i.e., the surface was sealed with mineral soil and a bentonite mat), which was also mentioned by Przydatek et al. (2024). The amount and type of waste deposited there have not been determined because of a lack of documentation, stemming from the facility's use since the 1970s (Table 1).

The surface of landfill site A is concealed under a mass of earth covered with low vegetation, and the biogas produced at the site is captured through a system of 17 gas wells.



Figure 3. Location of the vertical degassing well for determining the landfill gas composition at site A (50°48'51.40" N; 16°18'36.55" E, administrator's archive materials)

Table 1. Basic parameters of municipal waste landfill site A

Landfill site	Period of operation	Area	Number of quarters	Amount of deposited waste	Degasing wells
	years	ha	psc.	Mg	psc.
Unorganized	1974–2001	6.1	2	No data available	17



Figure 4. Location of the vertical degassing well and the point (Ph) for determining the landfill gas composition at site B (50°45'30.90" N; 16°15'36.02" E, administrator's archive materials)

Table 2. Basic parameters of municipal waste landfill site B

Landfill site	Period of operation	Area	Number of quarters	Amount of deposited waste	Degasing wells
	years	ha	psc.	Mg	psc.
Organized	2002–2019	3.55	2	152760	6

- Municipal waste landfill site B – operational and post-operational phases

The landfill site comprises two compartments: IA and IB, sealed with a double-layered bentonite mat and a 2.0-mm-thick geomembrane. This facility is classified as organised because its technical infrastructure primarily serves environmental protection.

During the 17 years of operation of the landfill site, 152,760 Mg of waste were deposited in two quarters with a total area of 3.55 ha (Table 2).

The field-degassing facility is an installation comprising six vertical degassing wells, which are used for capturing, purifying, and neutralising LFG in a flare.

Statistical analysis

Descriptive statistics were performed for each variable and measurement point, considering minimum and maximum values. Basic measures of central tendency (mean) and variability (standard deviation) were recorded to assess data distribution and enhance the analyses. The number of research results ranged from 12 to 51 between 2018–2023.

Results above the limit of quantification (LOQ) were used to calculate statistical parameters. If the

value for a given LFG component exceeded LOQ (understood as a multiple of the detection limit), the result was considered statistically significant and different from the blank sample (Helsel, 2012). Statistical significance was set at $\alpha = 0.05$.

Nonparametric correlation

The nonparametric Spearman rank correlation coefficient was used to assess the relationships between the studied LFG components. This method was justified by the failure to meet the normality assumption, which was verified by the Shapiro–Wilk test ($p < 0.05$). Correlation relationships between landfill sites A and B were examined, assuming that the observations from each landfill were independent, without taking the period into account. Statistical analyses were performed using the Statistica v13.1 software (StatSoft Polska, Kraków, Poland).

RESULTS

Landfill gas characteristics at site A

The LFG composition analysis at landfill site A, which was in use for 28 years and has been

closed for over 20 years, revealed parameters in only 2 of the 17 wells (S1 and S7). Well S1 exhibited the highest average O_2 concentration (16.95%, Figure 5). On the other hand, well S7 had a higher average temperature (15.70 °C); its average CH_4 and CO_2 contents were 8.30% and 5.40%, respectively, with a CH_4/CO_2 ratio of 1.49. In addition, a maximum O_2 content of 19.9% was recorded, 0.60% lower than that of well S1 (Figure 6). No LFG flow was detected during the measurements.

Landfill gas characteristics at site B

The LFG at the collective intake point at landfill site B, which was used for 17 years until its closure in 2020, indicated a maximum temperature of 33 °C and an average temperature of 14.80 °C. The landfill site recorded high maximum (>65%) and average (64.45%) CH_4 contents. The maximum and average CO_2 contents were 34.6% and 24.11%, respectively. The O_2

content ranged from <0.50 to 6.2%, with an average below 0.5%. In addition, the CH_4/CO_2 ratio reached a high value of 2.7. Despite the high CH_4 content (between 48.50 and >65%), no LFG flow was observed (Figure 7).

Comparative analysis of landfill gas parameters at sites A and B

Although landfill site A was used for 10 years longer than landfill site B, only the average O_2 content was higher by 16.2%. By contrast, higher average parameter values were recorded at landfill site B than at site A: the LFG temperature was higher by 0.9 °C, the CH_4 content by 58%, and the CO_2 content by 19.8%. The CH_4/CO_2 ratio at landfill site B was also higher by 1.3 (Figure 8).

At landfill site A, the highest average CH_4 (4.85%) and CO_2 (2.99%) values were recorded in the first half of the year. By contrast, the highest average LFG temperature (17.27 °C) and O_2

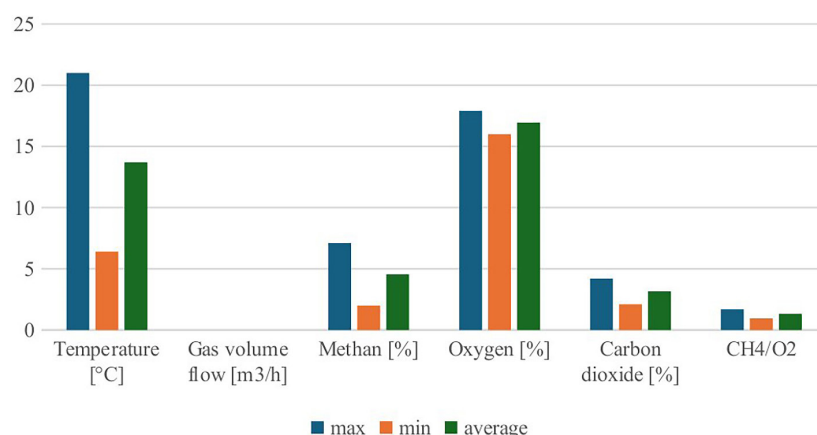


Figure 5. Landfill gas parameters at site A (S1 point) in the post-operational phase

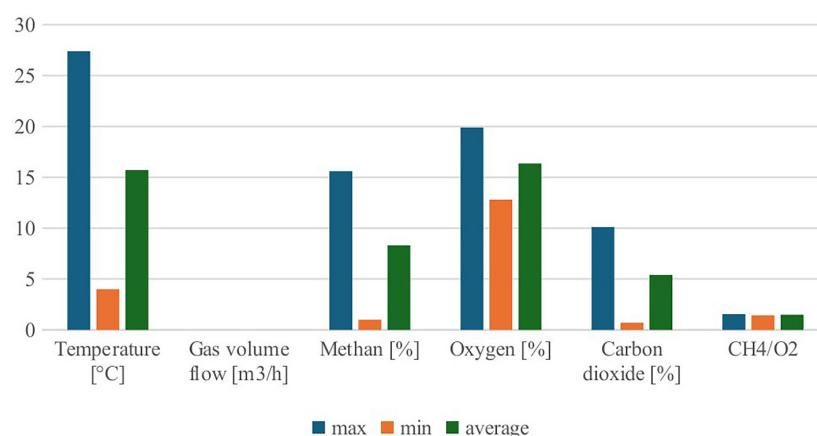


Figure 6. Landfill gas parameters at site A (S7 point) in the post-operational phase

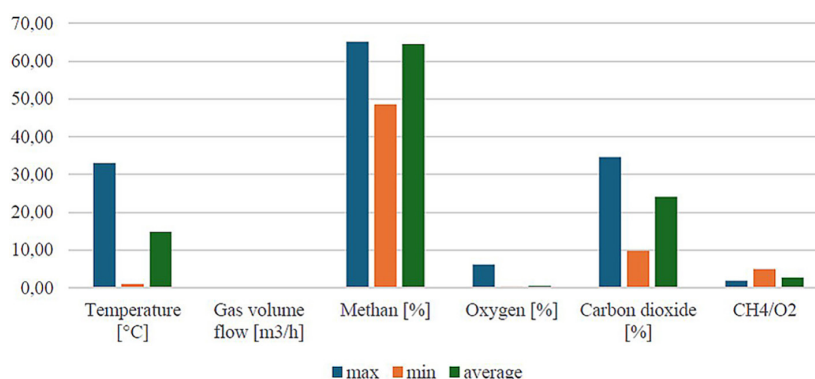


Figure 7. Landfill gas parameters at site B (Ph point) in the operational and post-operational phases

content (15.33%) occurred in the second half of the year. There was a noticeable increase in temperature (by 8.17 °C) and O₂ values. On the other hand, the average CH₄ and CO₂ contents decreased by 2.42%.

At landfill site B, the highest average temperature (16.72 °C), CH₄ content (65.10%), and O₂ content (0.65%) were recorded in the second half of the year. However, these parameters showed only a slight increase compared to the first half of the year. In turn, the average CO₂ content (24.30%) recorded in the first half of the year decreased (Table 3).

Descriptive statistics of landfill gas parameters

Low CH₄ (3.84%) and CO₂ contents (2.92%) were observed in the post-operational phase of landfill site A, concurrent with a simultaneous high O₂ content (17.52%). These conditions indicate that aerobic processes predominated, typical of phase V of organic waste decomposition

(Figure 1, Porowska, 2021). Substantial temperature data dispersion was observed relative to the mean of 13.94 °C (SD = 7.85 °C) (Table 4).

On the other hand, landfill site B was characterised by high mean LFG values, particularly for CH₄ (64.45%) and CO₂ (24.11%). However, the mean O₂ content was low (0.46%). Thus, anaerobic conditions and methane fermentation predominated, as is characteristic of phase IV (Figure 1, Porowska, 2021).

The low SD value (3.10%) of CH₄ confirms the small variation in results and the stable decomposition process of the deposited organic matter. The LFG temperature at landfill site B oscillated in both phases, fluctuating between 1 and 33 °C, with an average of 14.80 °C. The substantial dispersion of temperatures around the mean (SD = 9.27 °C) is responsible for the biogas flow stream.

The average rainfall quantity was 515 mm, with extreme values ranging from 81 mm to 954 mm. The standard deviation (361 mm) confirmed significant variation in values.

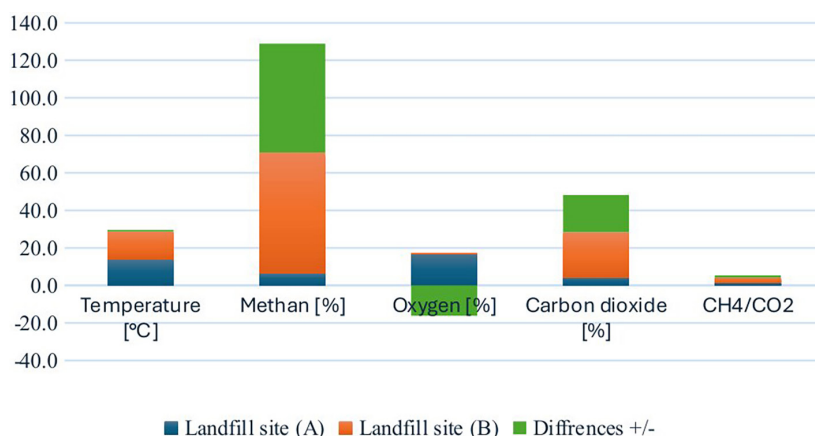


Figure 8. Average values of the landfill gas parameters at sites A and B and differences between them

Table 3. Average landfill gas parameters at sites A and B

Parameter	Average value			Differences +/-*
	Unit	Period		
		H1	H2	
Landfill site		A		
Temperature	°C	9.10	17.27	8.17
Methan	%	4.85	2.43	-2.42
Oxygen		14.70	15.33	0.63
Carbon dioxide		2.99	2.01	-0.97
Landfill site		B		
Temperature	°C	12.54	16.72	4.18
Methan	%	64.48	65.10	0.62
Oxygen		0.25	0.65	0.40
Carbon dioxide		24.83	23.28	-1.55

Note: *The differences between H2 and H1 cover the years 2018–2023; (+) represents an increase and (–) indicates a decrease.

Table 4. Descriptive statistics of landfill gas parameters for facilities A and B

Variable	Descriptive statistics					
	Unit	N important	Average	Min.	Max.	SD
Rainfall	mm	20	515	81	954	361
Landfill site	A					
Temperature	°C	12	13.94	4.00	27.40	7.85
CH ₄	%	12	3.84	1.00	15.60	4.24
CO ₂		12	2.92	0.70	10.10	2.49
O ₂		12	17.52	12.80	19.90	1.92
Landfill site	B					
Temperature	°C	51	14.80	1.00	33.00	9.27
CH ₄	%	51	64.45	48.50	65.10	3.10
CO ₂		51	24.11	9.80	34.60	8.29
O ₂		51	0.46	0.25	6.20	1.04

Correlation analysis

A Spearman rank correlation analysis showed clear relationships among LFG parameters. The nature of these relationships was similar in both analysed facilities.

Strong relationships were found among LFG components in site A. There was a strong positive correlation between CH₄ and CO₂ ($r = 0.99$) (Figure 9), indicating their simultaneous transformations. Conversely, a strong negative correlation was found between CH₄ and O₂ ($r = -0.84$) (Figure 10). In addition, a moderate negative correlation was found between CO₂ and O₂ ($r = -0.79$). LFG temperature showed non-significant, weak positive correlations with CH₄ ($r = 0.36$) and CO₂ ($r = 0.40$), and a weak negative correlation with O₂ ($r = -0.36$).

Similarly, at site B, a strong, negative correlation was found between CH₄ and O₂ ($r = -0.82$). Furthermore, a non-significant correlation was found between CO₂ and O₂ ($r = -0.10$).

The corresponding correlations between the LFG parameters at sites A and B were low, implying independent gas productivity, except for a moderate positive correlation between CH₄ (A) and O₂ (B) ($r = 0.70$) (Sawilowsky, 2009) (Table 5).

DISCUSSION

Biochemical and anaerobic processes occur in municipal waste landfill sites, regardless of their size, and these processes contribute to LFG generation (Singh et al., 2023). Both the operational

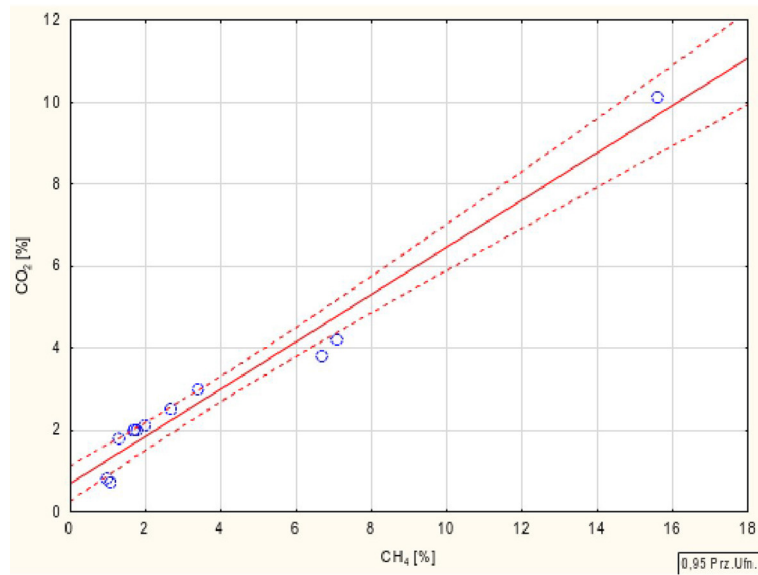


Figure 9. Relationship between CO₂ and CH₄ contents at landfill site A

Table 5. Spearman's rank correlation coefficient matrix for biogas parameters at landfill sites A and B

Variable	Spearman's rank order correlation							
	CO ₂ (A)	O ₂ (A)	CH ₄ (A)	Temperature (A)	CO ₂ (B)	O ₂ (B)	CH ₄ (B)	Temperature (B)
CO ₂ (A)	1.00	-0.79	0.99	0.40	0.75			0.30
O ₂ (A)	-0.79	1.00	-0.84	-0.36	-0.58			0.07
CH ₄ (A)	0.99	-0.84	1.00	0.36	0.70			0.27
Temperature (A)	0.40	-0.36	0.36	1.00	0.20			0.13
CO ₂ (B)	0.75	-0.58	0.70	0.20	1.00	-0.10	0.08	0.02
O ₂ (B)					-0.10	1.00	-0.82	-0.01
CH ₄ (B)					0.08	-0.82	1.00	0.06
Temperature (B)	0.30	0.07	0.27	0.13	0.02	-0.01	0.06	1.00

Note: Significant correlation coefficients are marked red, with $p < 0.05000$.

and post-operational phases, during which the changes related to the decomposition of deposited waste occur, pose an environmental threat, even 50 years after the site's closure (Reddy et al., 2017). Hence, emissions of the two main components of LFG (CH₄ and CO₂) can also occur in the post-operational phase (Singh et al., 2021; Chavan et al., 2022), indicating the need for extended landfill site monitoring. For instance, despite 2 of 17 vertical degassing wells operating at the inactive landfill studied, biogas emissions were detected at its edges. These emissions may be related to the almost three decades of use of the site, which is in its last phase, associated with a drop in biogas productivity (Barlaz et al., 1989; Tamru and Chakma, 2016).

The above-mentioned site exhibited a high average O₂ content, close to 18% and increasing,

and a low CH₄ content, below 5% and decreasing. These results suggest possible bed aeration as a result of fugitive surface emissions, as reported by Di Trapani et al. (2013). Some researchers (Hanson and Hanson, 1996; Huber and Humer et al., 2008; Pinheiro et al., 2019) concluded that an increase in O₂ leads to a decrease in CH₄ because of oxidation by metamorphic bacteria, especially on the surface of the landfill.

Furthermore, the landfill site that had been operated for longer exhibited an average O₂ content that was 16% higher than that in the site with a shorter operational period. Ma et al. (2023) demonstrated that the maximum contribution of aerobic processes to waste decomposition can reach 16%. The negative correlation between CH₄ and O₂ implies variability in the CH₄ generation process. Temperature remains a key

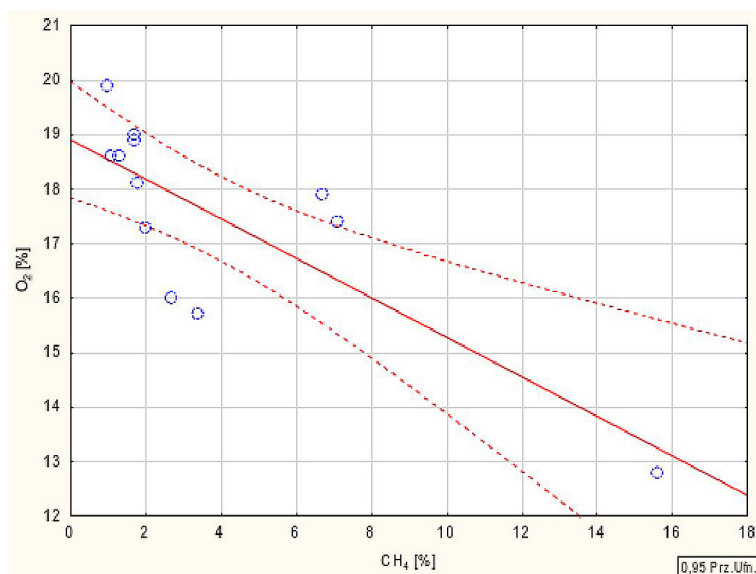


Figure 10. Relationship between O₂ and CH₄ contents at landfill site A

indicator in assessing the condition of a waste landfill. Lefebvre et al. (2000) demonstrated that O₂ diffusion and aerobic reactions increased the temperature of a waste landfill by up to 20 °C during the initial phase of decomposition. The high maximum temperature (33 °C), mean temperature (~15 °C), and standard deviation (~9 °C) in the organised landfill in this study indicate that anaerobic processes predominated, as demonstrated by Hanson et al. (2010).

Krause et al. (2023) reported LFG at high temperatures above 50 °C. Such high temperatures may be accompanied by destructive exothermic reactions (Reinhart et al., 2020). El-Fadel et al. (1995) demonstrated that temperature affects LFG transport and the rate of biochemical processes occurring in landfills, including biogas emissions. Here, the average temperatures at the two landfill sites differed by only 0.9 °C, indicating similar thermal conditions despite different closure dates. At the organised landfill, which was 2.55 ha smaller than the unorganised landfill, the highest average temperature (17.27 °C), CH₄ content (65.10%), and O₂ (0.65%) content were observed in the second half of the study period, during the operational and post-operational phases, with simultaneous increases in these parameters. This result may indicate the temporary variability of biogas generation depending on the different environmental factors, including the method of gas transport and the time scale (Gebert et al., 2011). Börjesson and Svensson (1997) and Przydatek et al. (2024) mentioned similar

relationships. Yentekakis et al. (2019) reported a CH₄ content ranging from 50 to 70% under anaerobic conditions, similar to the mentioned results. Additionally, Karanjekar et al. (2015) emphasised the importance of CH₄ in climate change, as it is a 25-fold more potent greenhouse gas than CO₂.

The CH₄/CO₂ ratio was 2.7 at the organised landfill site. This value exceeds the range of 1.0–2.3 reported by Zhao et al. (2020). Such a high value suggests a dominance of methanogenesis. By contrast, the CH₄/CO₂ ratio was lower by 1.3 at the unorganised landfill site. The high O₂ and low CH₄ contents indicate the development of aerobic conditions in the waste bed, along with the disappearance of the anaerobic conditions responsible for methanogenesis. Furthermore, the high correlation between CH₄ and CO₂ ($r = 0.99$) suggests a common link with waste biodegradation. Likewise, Przydatek et al. (2024) demonstrated positive correlations in the post-operational phase of the landfill site they evaluated. Here, a positive correlation ($r = 0.70$) was found between the CH₄ and CO₂ contents in the two landfill sites studied, suggesting similar dynamics in these parameters in the facilities located in close proximity. Yang et al. (2015) found similar positive correlations between these indicators at two landfill sites, but only under anaerobic conditions. Kjeldsen et al. (2002) concluded that the increase in CO₂ content may be due to increased O₂ consumption associated with the oxidation of organic matter.

Notably, although approximately 153,000 Mg of waste had been deposited and the CH₄ content

was high, ranging from 48.5% to over 65.0%, no gas flow was observed in the landfills, even several years after their closure. Zamorano et al. (2007) proposed that the amount of biogas produced depends on the amount of waste deposited, and suggested a unit value of $0.350 \text{ Nm}^3/\text{kg}$. Nonetheless, when LFG extraction is ineffective, there is a lack of pressure gradient to drive gas flow towards the degassing installation. Gebert and Groengroeft (2005) demonstrated that atmospheric pressure and particularly temperature drop in the landfill's vicinity. As a result, LFG may accumulate in the waste structure or migrate unevenly, increasing the risk of diffuse emissions. This is because the emission process is initiated by pressure gradients, which influence the advective flow, and concentration gradients, which lead to concentration diffusion (Nastev et al., 2001).

CONCLUSIONS

The following conclusions were drawn based on the analysis of the biogas composition in the operational and post-operational phases (from 2018 to 2023) at two landfill sites:

Both the operational and post-operational phases, during which changes related to the decomposition of deposited waste occur, contribute to the variability of LFG parameters.

The analysed landfill sites differed markedly in their operational period because the unorganised landfill site was active 10 years longer than the organised landfill site, and more than 15 years had passed since its closure.

The average O_2 content was approximately 18% and increasing, whereas the CH_4 content was low (below 5%) and decreasing in the post-operational phase of the unorganised landfill site. At the same time, a strong correlation ($r = 0.99$) between CH_4 and CO_2 was observed. The moderate correlation ($r = 0.70$) between the organised and unorganised landfill sites indicates similar dynamics in the biochemical composition of LFG.

The highest average temperature ($\sim 17^\circ\text{C}$), CH_4 content ($\sim 65\%$), and O_2 content (0.65%) were recorded at the organised landfill site in the second study period, with simultaneous increases in these indicators, which may suggest the influence of environmental factors on the temporary changes of biogas composition.

The high CH_4/CO_2 ratio (2.7) confirms the dominance of methanogenesis at the organised

landfill site. By contrast, the ratio at the unorganised site was lower by 1.3, implying the final phase of waste decomposition.

Higher temperatures at the organised landfill site mean that anaerobic processes predominated. However, the small average temperature difference (0.9°C) suggests similar thermal conditions between the sites, despite different closure dates.

Although the amount of waste deposited at the landfills exceeded 100,000 Mg and the CH_4 content was high, no gas flow was observed, suggesting low biogas capture efficiency.

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