MONITORING OF GAS EMISSIONS: A CASE STUDY IN ROMANIA

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This article is intended to present the main issues and problems regarding landfill gas collection systems at municipal waste landfills. First, aspects of the landfill gas collection system at the IRIDEX Chiajna – Bucharest landfill are presented together with the parameters and components of this gas. Also, the main problems posed by hydrogen sulphide (H_2S) existing in the composition of the landfill gas, collected for more than 11 years, are related.

Keywords: waste landfill, landfill gas, gas collection system, H_2S, gas parameters, energy aggregate

1. Introduction

Without any doubt, waste landfilling is the most inadequate waste management option for bio-waste and other types of waste. In many cases, however, the landfilling is a viable solution that depends on specific and local factors. These factors are: the collection system types, waste composition and quality, climatic conditions, the potential use of various products resulted from the waste management, such as electricity and heat, methane-rich gas or the compost [1].

All biodegradable waste stored in a compliant waste deposit generates landfill gas (LFG), a natural effect of anaerobic decomposition. LFG is a mixture composed predominantly of methane CH_4, carbon dioxide CO_2 and other decomposition gases. Usually, LFG is composed of 40-60% vol. CH_4 and 40-55% vol. CO_2, the rest consisting of oxygen, nitrogen, ammonia, hydrogen sulphide, and other volatile organic compounds.

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The main purpose of degassing in landfills that accept biodegradable waste is to prevent and reduce the emission of gas into the atmosphere due to the negative environmental consequences (greenhouse gas emissions).

Landfills that accept biodegradable waste generally produce LFG in large quantities, starting right from the current period of operation, with maximum amount in the period when it reaches almost maximum capacity and slowly decreasing in the post-closure period. It is produced continuously by microorganisms that act on disposed organic waste. Therefore, LFG should be collected from all landfills, treated and used. In addition to each landfill, it is mandatory to build and develop a system for collecting and safely burning of LFG. In some cases, it is recommended to use LFG to electricity generation at reasonable costs if this proves technically and economically feasible, [2].

The waste management regulations (especially regarding LFG) mainly describe waste management and control of LFG in general and include all the generated gas generated through waste biodegradation. Also, the gases resulting from chemical reactions and the volatilization of chemicals in the waste are mentioned [3, 4, 5].

Collection, treatment and use of LFG must be carried out in such a way as to minimize as much as possible negative environmental consequences, taking into account the protection of the population's health.

Despite methane is a greenhouse gas 21 times more harmful to the environment than carbon dioxide, methane is also considered a useful combustible gas for production of electricity and heat. Its elimination by controlled combustion is considered an environmental benefit. The benefits can increase if the carbon dioxide generated by burning methane is collected and valued. Methane from LFG is a useful fuel, and its potential energy recovery is a matter of good economic practice.

In any case, when an ecological landfill is built, it is assumed from the start that the bottom base sealing and covering system is sealed to gas and water, in both directions. Theoretically, it is impossible for the gas to escape from the landfill to the underground vicinity or to the atmosphere through the final closing system, [6].

Techniques for monitoring potential surface diffuse gas emissions include surface emission measurement, ground or low altitude imaging, and satellite and aerial imaging, [7, 8].

Surface emissions monitoring (SEM) is a technique that involves usage a portable methane meter near the surface of the landfill to measure concentrations on and near the site.

Ground or low altitude imaging can be done with infrared cameras (IR), which allow the observation of frequencies that the human eye cannot, including the frequencies in which methane is visible. Drone-mounted IR cameras can potentially monitor areas where existing personal are unavailable or cannot be accessed safely.

In the EU, 53% of anthropogenic methane emissions come from the agricultural sector, 26% from waste, and 19% from the energy sector. There are similar trends worldwide, with around 95% of total anthropogenic emissions from these three sectors. The EU Methane Strategy, therefore, focuses on these areas [14].

However, globally, it is estimated that about 140 billion cubic meters of gas are wasted annually by burning in flares, equivalent to $ 20 billion. Moreover, gas flaring annually adds to the atmosphere an amount of CO_2 equivalent to 200 million cars.

Globally landfilled waste of 1.5 billion tons annually produces about 75 billion cubic meters of landfill gas, the equivalent of harmful emissions into the atmosphere from about 423 million cars, [15].

This paper analyses the landfill gas collection system and the structure of such a system, at the IRIDEX Chiajna - Bucharest landfill. The components and parameters of the landfill gas collected by fermentation of the biodegradable fraction from the landfilled waste, the landfill gas parameters in the main collection pipeline within the landfill, and data on the operation of the booster station during 01-31.12.2021 (average values per day), and for the entire period of operation of the energy system, from commissioning to the present (annual average values), are presented.

2. Materials and Methods

Landfill gas generation (LFG) models are an essential tool in the initiation and development of projects for the collection of gas from landfills because it can provide answers to the following problems:

- assessment of the feasibility of energy utilization of the LFG and the technical requirements of the system;
- estimating the amount of recoverable gas that is generated during the operation of the deposit;
- designing the gas collection system (extraction wells, in situ treatment system, top cover oxidation, etc.);
- evaluating the performance of existing collection and recovery systems;
- calculation of (national) greenhouse gas inventories (GES);
- predict residual environmental landfill impacts, [9].

Dimensioning of a gas collection system is done by using calculation programs, general or particular, which are not always applicable all the time and everywhere. For example:

- Land Gem (U.S. EPA, which estimates quantities emitted into the atmosphere, applicable mainly to U.S. conditions, but also most used and applied in almost all projects in Romania);
- Waste Reduction Model (WARM) (U.S. EPA which estimates emissions in the event of a reduction in the amount of waste stored);
- GasSim 2.5, applicable to U.K.;
- IPCC model - developed by UNFCC, model applicable, with some adjustments, worldwide.

To design and optimize a landfill gas collection system, it is necessary to quantify the gas generation over several years. [10, 11, 12].

The open/closed flaring of the LFG is generally a free combustion, in the presence of forced blown oxygen, i.e. with forced draft, inside a metal cylinder. During combustion, a temperature of 1100 °C is reached, and the gas burns inside for at least 0.3-0.6 s, according to the requirements of a complete and correct combustion. This ensures complete oxidation of CH_4 and other gas compounds, resulting in water vapour and CO_2. Furthermore, this is the simplest method of removing gas. In addition to the advantages mentioned above, most high temperature HT systems control heat loss in the environment and are closed type, the flame never exceeds the collar of the torch (cylinder).

Landfill gas processing and filtration, in those cases in which the LFG utilization option is chosen, involve:

- dust filtration - if the gas is to be used in various mechanical aggregates, to reduce their premature and accelerated wear;
- cleaning and elimination of certain compounds of chemical compounds, like H_2S, for environmental or air quality reasons, but also for reduce corrosion of the internal components of CHPs;
- gas filtration and purification - in the case of CNG (Compressed Natural Gas for Vehicle) and LNG (Liquefied Natural Gas) projects or injection into natural gas networks.

Increasing energy efficiency and reducing the environmental impact when using landfill gas are considered beneficial both in terms of electricity production and for reducing greenhouse gases, [10, 13].

In Romania, for the moment, there is only one functional project for using LFG for electricity production, integrated to the landfill. It is the case of the Chiajna landfill, near Bucharest, operated by IRIDEX GROUP, which collect, treat, process and use landfill gas as an energy source. The project is operational since March 2011. Its energy capacity is 3.6 MW_e (megawatt electrical) + 1.8 MW_{th} (megawatt thermal), having 3 CHP (Combined Heat and Power) aggregates and a controlled gas combustion plant with $T \geq 1100^{\circ}C$, for at least 0.3 sec.

For optimal operation of the plant, the LFG requires proper cleaning and filtration, to be used in internal combustion engines, as a fuel of the best quality. At the time when the plant started operation, activated carbon filters were used, which have good results from the point of view of reducing the concentrations of unwanted components, such as hydrogen sulphide. The activated carbon must be replaced periodically, when it becomes saturated with retained components or particles.

Activated carbon saturated and impregnated with retained components is considered hazardous waste, so it must be disposed in accordance with the relevant legal provisions, which leads to an increase in the maintenance and operation costs of the entire plant.

Since 2014, a filtering system of a different design, which uses certain types of bacteria to reduce the concentration of hydrogen sulphide, has been in operation. This concept of desulfurization is considered a bio catalytic process. Currently, the plant operates with this kind of bio catalytic desulfurization plant (BDS), [16]. But the active carbon filtration system was also preserved, kept in operation and therefore, can work in parallel with the bio catalytic system, as a safety and backup measure.

During the operation of the plant, advantages and disadvantages of both types of gas filtration systems were identified. Any of these systems can be used successfully for gas filtration, but the choice of one system over the other must take into account certain conditions.

First of all, must be analysed the filtering efficiency for the compounds that are considered harmful and therefore, must be eliminated.

There are also other parameters to be analysed, such as the predictability of electricity production, maintenance and replacement costs, location-specific weather conditions, availability of some consumables and materials.

In a future material, some data will be presented comparatively about the operation of both types of filtration systems, which will highlight the advantages and disadvantages of each system.

The ecological waste landfill within the Integrated Waste Management Centre (in Romanian Language – Centrul de Management Integrat a Deseurilor) IRIDEX Chiajna - Bucharest was established in 1999. CMID Chiajna is located north of Chiajna Railway Station, between the two railway lines, the Bucharest - Videle highway, and a secondary line serving the industrial units in the area. Access to the Chiajna Integrated Management Centre is made on the Chitila - Rudeni road [17].

CMID IRIDEX Chiajna occupies an area of 31.65 ha. The main beneficiary of the activity carried out within it is the General City Hall of Bucharest. CMID Chiajna includes the following structures: a waste landfill with seven compartments with perimetral dikes, compartmentation dikes, leachate drainage system, reverse osmosis leachate treatment modules, water supply and underground water monitoring drillings, wells for the extraction of the LFG, the transport gas pipes and the controlled combustion installation and recovery of the LFG. The installation has been in operation since 2011 and ensures the active degassing of the deposit through the depression applied to the gas capture system. Till now, a quantity of over 80 million m^3 of landfill gas with an average concentration of approx. 50% methane was captured and used for electricity production. This quantity of used landfill gas significantly reduces the amount of greenhouse gases emitted into the atmosphere. After the final closure of 5 of 7 compartments, the overview of the deposit is shown in figure 1, and the installation of controlled combustion and gas recovery in figure 2.

Fig. 1. IRIDEX Chiajna municipal landfill (overview), [16]

Fig. 2. Controlled combustion plant and landfill gas recovery with energy aggregates IRIDEX Chiajna

The landfill of CMID Iridex - Chiajna has an area of 25.74 ha, with a volume of approximately 4,400,000 municipal wastes at this time, compared to the projected capacity of 4,500,000 m^3. No waste has been received at the landfill since

May 2021. Now (July 2022), the part of the landfill still unclosed is in the process of temporary closure and consumption of settlements.

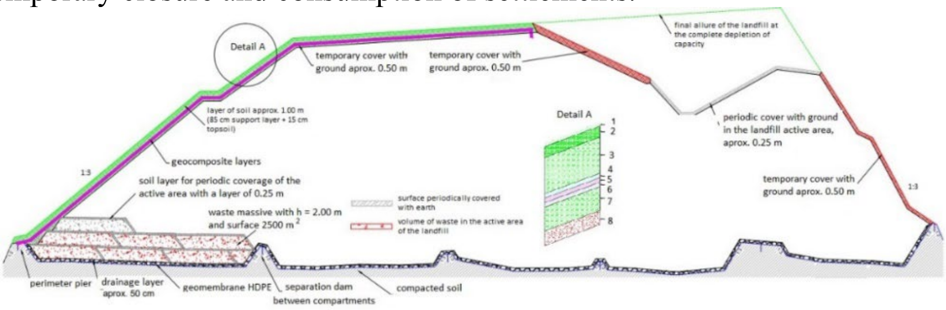

Fig.3. Vertical profile of the IRIDEX-Chiajna waste landfill, [16]
1.turf, erosion resistant vegetation; 2.layer of topsoil approx. 15 cm; 3.layer of clay soil with sand / gravel approx. 85 cm; 4.drainage geocomposite for rainwater; 5.bentonitic geocomposite; 6.drainage geocomposite for gas; 7.landfill body; 8.intermediate cover of waste with soil during usual landfilled of waste

The parameters that characterize the quantities of waste deposited, of leachate and landfill gas, were constantly monitored in order to protect the health of the inhabitants near the landfill and to respect the legislation related to environmental protection. The vertical structure of the IRIDEX landfill Chiajna complies with the international regulations for sealing the landfill body from the rest of the environment, as shown in fig. 3, [16].

There is a constantly monitoring of all the parameters of the extracted and recovered landfill gas, its composition, and the amount of electricity produced.

The energy recovery installation, shown in fig. 2, operates according to a functional diagram as shown in fig. 4, [16].

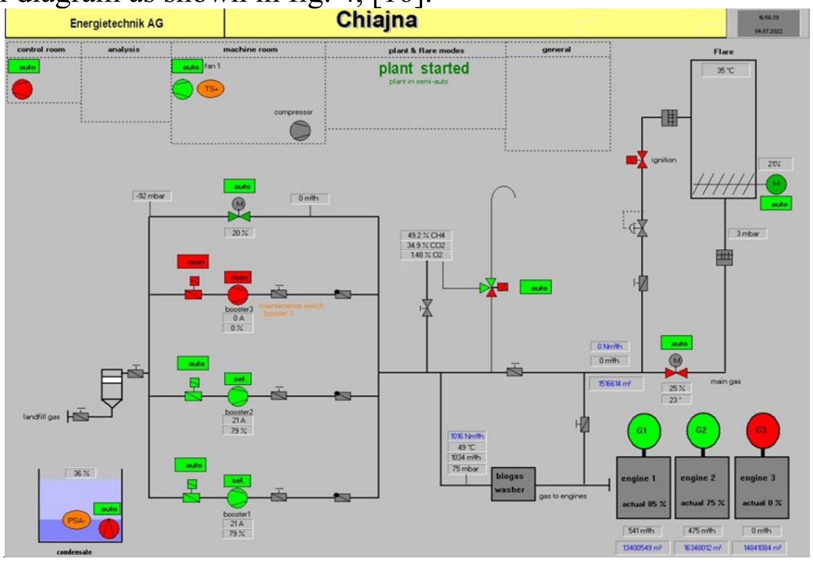

Fig.4. Functional and control scheme of the landfill gas energy recovery installation

3. Results and discussions

The quantities of gas extracted and recovered and the amount of electricity produced annually since the commissioning of the energy system of CMID Chiajna are presented in Table 1 and fig. 5.

Table 1

Quantities of landfill gas used and electricity produced annually at CMID IRIDEX-Chiajna

Year	LFG extracted, m^3	LFG used to for energy production, m^3	Electrical energy produced, MWh	Year	LFG extracted, m^3	LFG used to for energy production, m^3	Electrical energy produced, MWh
2011	6 399 424,5	6 396 904,5	12 976,16	2017	8 054 441,0	7 921 787,0	17 491,86
2012	7 500 303,1	7 444 500,0	15 784,30	2018	7 533 127,0	7 205 190,0	15 120,91
2013	6 921 790,9	6 910 508,0	15 435,30	2019	5 450 849,0	5 091 096,0	10 049,35
2014	7 482 189,4	6 172 386,5	14 401,80	2020	6 532 333,0	6 193 194,0	12 808,32
2015	9 721 035,9	9 626 389,7	20 673,68	2021	7 359 025,0	7 323 996,0	14 810,03
2016	8 077 989,8	7 780 265,8	17 082.89				
		TOTAL (2011-2021)			**81 032 508,55**	**78 066 217,48**	**166 634,59**

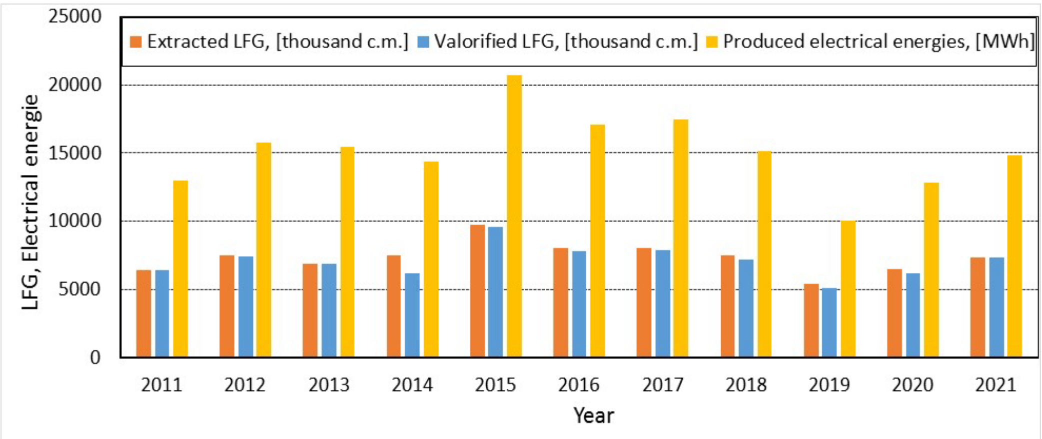

Fig.5. Quantities of landfill gas and electricity produced at CMID IRIDEX - Chiajna since the beginning of gas usage

It is worth mentioning that by recovering the LFG, a quantity of over 166 000 MWh of electricity was delivered to the National Energy System (SEN), thus substituting the consumption of fossil fuels from non-renewable sources.

As seen from the graph in Fig. 5, the highest production of LFG, extracted and recovered, was over 9.5 million m^3 in 2015. However, it is impossible to determine precisely whether this year was the most considerable amount of gas produced inside the landfill. In 2015, the landfill was in the current operation period, when it was decided to stop the landfilling on compartments 1-5 of the landfill (the first 5 compartments out of the 7 of the landfill). It is obvious that the largest amount of electricity was also in 2015.

At the same time, the gas losses without recovery, as shown in Table 1, had values in the range of 0.03-17.5%, with an annual average of 3.75% and a standard deviation of 4.64%, the highest loss being recorded in 2014.

For this specific year, this fact was due to an artificially inflated media context, mainly between February and October 2014. At that time, environmental authorities have imposed continuous accelerated degassing amid odor complaints, which directly involves flare-ups.

In 2014, for a certain period of time the entire electricity production facility was completely stopped. This temporary stopping was imposed by the installation and commissioning operations of the bio-desulfurizer. In 2014, over 1 300 000 m^3 of LFG was burned in the flare. It is an amount of gas that could have produced approx. 2 900 MWh, which was wasted.

The discrepancies between the annual values of extracted gas quantities and the recovered ones point out that some LFG quantities were burned instead of being recovered. Another cause of the different annual values of the extracted gas and the recovered one is the inherent failures of the thermal engines for transforming the gas into electricity, which led to their stopping for a while.

However, the differences noted may not be considered losses if we consider things differently. It is normal for a landfill to have periods when the LFG needs to be flared when the engines are defective or overhauled. It is a safety measure and, at the same time, a legal obligation, the landfill operator being obliged to ensure the continuous degassing of the landfill as much as is possible, regardless of the method of recovery or disposal.

In a classic gas recovery system (such as the one in Chiajna), it is practically impossible to capture and use all the gas generated by the landfill only for energy production because there are no storage tanks, and it is not legal and possible to have such storage tanks. There are such tanks in gas compression and processing plants for compressed natural gas (CNG) or liquefied natural gas (LNG).

The quantities of extracted landfill gas, the relevant gas quality parameters, and the quantities of electricity produced at CMID IRIDEX-Chiajna are recorded in the system. This values vary daily and even from hour to hour, as they depend on the atmospheric and meteorological conditions of the respective period. Therefore, it is needed a permanent adjustment, sometimes even dynamic of the system to the real conditions to simultaneously achieve, as much as possible, the two essential objectives: environmental protection and gas recovery.

For example, the average daily values of landfill gas parameters at the landfill booster station in December 2021 are shown in Table 2.

Table 2

Gas parameters at landfill waste IRIDEX Chiajna - Bucharest in period 01-31.12.2021

Atmospheric parameters		Gas parameters in the central collection pipe					Gas parameters to engine			
(°C)	(mbar)	Pressure (mbar)	CH_4, (%)	CO_2 (%)	O_2 (%)	H_2S (ppm)	Flow (Nm^3/h)	T, (°C)	Flow (m^3/h)	Pressure (mbar)
1.0	1007	-51	50.60	34.10	2.60	1520	878	29	874	65
0.0	1010	-48	50.80	34.00	2.58	1532	915	30	890	65
1.0	1005	-52	50.10	33.40	2.78	1516	940	28	920	65
0.3	1015	-52	49.60	33.20	2.94	1443	938	28	917	65
2.0	1011	-49	50.50	33.90	2.72	1434	917	26	894	65
1.0	1006	-53	49.70	33.50	2.90	1418	945	26	922	65
3.0	1002	-54	49.60	33.10	3.00	1450	956	28	942	61
3.0	1006	-54	49.50	33.20	3.05	1456	946	28	934	65
0.0	1013	-55	48.80	32.70	3.23	1334	960	28	934	70
4.0	1007	-57	49.70	33.20	3.07	1439	1010	29	1001	70
3.0	1008	-49	50.30	33.50	2.86	1430	927	28	905	70
11.0	1005	-58	50.20	33.50	2.94	1418	998	29	985	70
2.0	1018	-33	52.50	34.80	2.26	1494	743	25	727	70
3.0	1024	-49	50.60	33.60	2.90	1514	996	28	977	70
3.0	1024	-65	49.50	33.00	3.18	1368	1118	30	1095	75
2.0	1026	-63	48.20	32.00	3.61	1400	1143	30	1126	75
1.0	1023	-71	48.50	32.10	3.65	1398	1189	31	1172	75
0.0	1021	-97	50.10	33.60	2.75	1830	1170	34	1162	75
-2.0	1019	-88	51.80	35.00	1.96	1896	1118	33	1109	75
-1.0	1007	-93	50.40	34.20	2.52	1720	1152	34	1144	76
-3.0	1216	-102	48.40	32.10	3.05	1704	1175	35	1161	75
-8.0	1027	-86	48.30	31.10	3.40	1266	1194	32	1170	75
-5.0	1029	-69	45.70	30.80	3.80	1344	989	29	981	75
-2.0	1017	-79	49.00	33.00	2.35	1857	1129	31	1010	75
-1.0	1010	-91	45.00	30.50	4.27	1866	1218	33	1201	75
0.0	1013	-90	46.00	31.00	3.97	1792	1224	33	1216	75
-1.0	1015	-122	44.60	30.40	4.17	942	1254	37	1252	75
0.0	1014	-87	48.90	32.80	2.86	1428	1156	32	1141	75
0.0	1013	-89	48.70	32.60	2.89	1608	1158	32	1144	75
-1.0	1013	-89	47.90	31.90	3.14	1748	1170	32	1146	75
1.0	1022	-86	48.60	32.40	3.03	1282	1145	31	1131	75
Average	1020.8	-70.35	49.1	32.85	3.05	1511.2	1057.13	30.29	1038.2	71.35

As can be seen, the LFG flow in December 2021, after more than 11 years since the commissioning of the first energy aggregate, had average daily values of about 1038 m^3 / h (with a standard deviation of 128.03 m^3 / h), LFG that has been recovered for the production of electricity and heat. Therefore, in order to be able to be extracted from the landfill, an average depression of about –70.35 mbar had to be applied, the average temperature of the gases at the entrance to the energy aggregates being about 30ºC.

The composition of the LFG (in December 2021), presented in Table 2 shows an average percentage of methane (CH4) of about 49%, while the percentage of carbon dioxide (CO2) was on average 32.8%.

It is important to note that the LFG contain hydrogen sulphide (H_2S) whose concentration must be reduced for processing in energy aggregates. Therefore, its treatment and processing is currently done with the help of a biodesulfurizer (biocatalytic desulfurization).

It should be noted that the average daily value of hydrogen sulfide (H_2S in the collected landfill gas showed a value of about 1511 ppm at the entrance to the biodesulfurizer, a value which decreased to about 363 ppm after filtration in the biodesulfurizer, the pressure drop being in an average of about 0.9 mbar. The average efficiency of the biodesulfurizer in reducing the amount of H2S was about 76%.

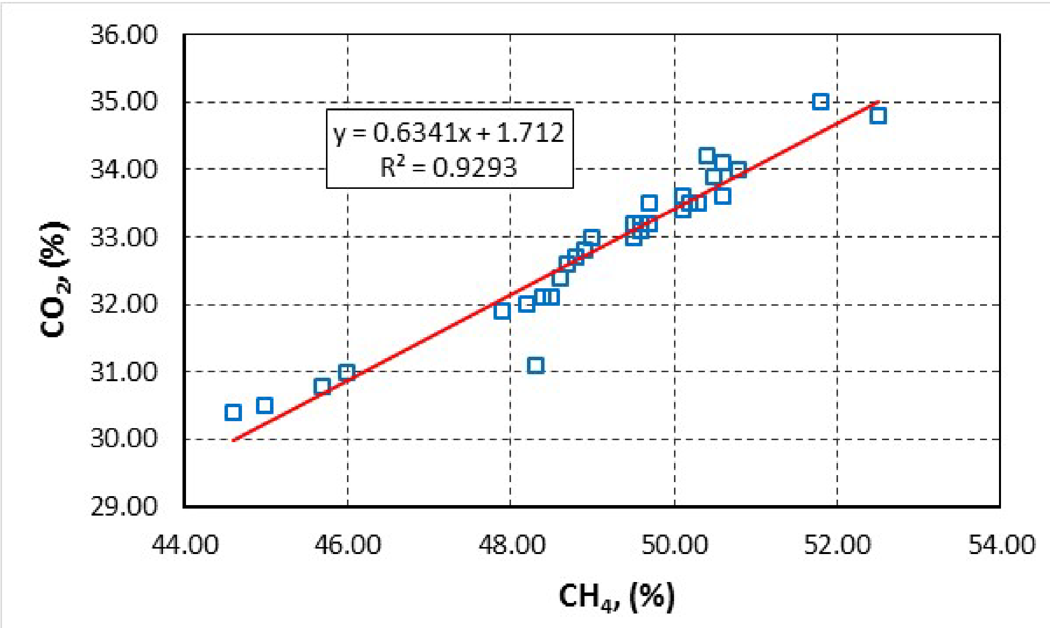

Fig.6. CO_2 variation with CH_4 in December 2021, 11 years after the start of gas usage

Overall, the average variation of CO_2 compared to CH_4 in December 2021 shows a linear distribution with a high correlation coefficient (R_2 = 0.929), with an ascending slope (n = 1.7). The percentage of methane in the landfill gas collected in December 2021 had an average value of 49.1%, compared to the percentage of

CO_2, which had an average value of 32.8%, the rest being represented by other landfill gas components.

Table 3

Quantities of landfill gas and its average composition during operation years

Year	Gas parameters in the central collection line (average values)			Active wells
	CH_4 (%)	H_2S (ppm)	O_2 (%)	
2011	53.40	687.70	1.24	60
2012	51.80	822.88	1.10	60
2013	52.66	1252.88	2.20	75
2014	56.40	1866.89	0.51	75
2015	54.70	1552.70	1.32	75
2016	53.41	1327.17	2.45	87
2017	51.86	1100.00	4.78	101
2018	49.40	1270.00	3.08	100
2019	51.19	1775.00	2.09	105
2020	48.76	1240.00	3.92	115
2021	49.03	1250.00	2.67	125

Table 3 shows that there is not necessarily a correlation between the number of wells and the amount of gas extracted, primarily because the gas is extracted mainly according to the coverage of the landfill by the radii of influence of the wells, as well as the degree of coverage of the deposit on certain areas, with soil or other materials.

4. Conclusions

Ecological municipal landfills are usually built on the premises of integrated waste management centres, taking special measures to isolate them from the rest of the environment.

When an ecological landfill is built, it is assumed from the start that the bottom base sealing and covering system is sealed to gas and water, in both directions. Theoretically, it is impossible for the gas to escape from the landfill to the underground vicinity or to the atmosphere through the final closing system, [6].

The insulation is made by a specific seal at the landfill's base so that the meteoric water that flows through the waste layers does not reach the groundwater, respectively, with a tight coating on the surface when the landfill capacity is exhausted. Under these sealing conditions, the landfill work as an anaerobic bioreactor.

In landfills, gas inevitably occurs as an effect of the degradation of the organic fraction under the action of bacteria and enzymes under anaerobic conditions. In most landfills in Romania, the landfilled waste contains a significant percentage of organic fraction from mixed household waste or gardens and parks.

To recover landfill gas from the decomposition of organic waste in the landfill body, a specific recovery system must be constructed with gas wells and HDPE pipes that lead the gas to filtration, pre-treatment, and treatment systems before being used for energy recovery.

Based on experience accumulated in more than 10 years of operation of the installation, it can be said that both gas filtration systems that are used have advantages and disadvantages.

It seems that activated carbon filtering systems are adequate and sufficient in the case of lower flows of LFG, with reduced concentrations of H_2S. But when the flow of LFG increases, simultaneously with the increase in the concentration of H_2S, a bio catalytic desulfurization system must be installed and put into operation, to ensure the correct operation of the energy production plant. These filtration systems can work in parallel or independently, concurrently or successively, depending on the actual operating conditions.

The methane content of the landfill gas depends on the waste's organic load, the landfill's age, the types of organic compounds in the waste, the moisture content and temperature of the waste, and other parameters that are difficult to control. Air temperature and its relative humidity are parameters influenced by the local climate specific to the location.

It is mandatory to take specific measures to protect the environment and avoid the release into the atmosphere the untreated or unburned LFG, which can lead to an increase of greenhouse effect.

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