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Optimizing Methane Recovery for Fuels: A Comparative Study of Fugitive Emissions in Biogas Plants, WWTPs, and Landfills

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Abstract: How accurate are current estimation methods for fugitive methane emissions in methane-producing facilities, and how do they vary across biogas plants, wastewater treatment plants (WWTPs), and landfills? Based on this, the hypothesis posited in this study is that current methods significantly underestimate methane emissions, particularly in WWTPs and biogas plants, due to limitations in accounting for recovered methane and the reliance on general parameters such as the oxidation factor. To test this, a comparative analysis was carried out involving 33 biogas plants, 87 WWTPs, and 119 landfills in the Iberian Peninsula, comparing officially recorded data with estimates derived from our own calculations. Our findings confirm the lack of precision in current emission estimation methods, particularly for WWTPs and biogas plants, where factors like the omission of recovered methane lead to underreporting. This study highlights that WWTPs emit the largest amount of methane due to their organic material processing, exceeding emissions from landfills and biogas plants. In contrast, methods for estimating emissions in landfills are found to be more reliable. The results suggest that improving calculation methodologies, especially for WWTPs and biogas plants, as well as enhancing leak monitoring and methane recovery systems, is crucial to reducing the environmental impact of methane-producing facilities.

Keywords: fuel production; methane emissions; biogas plants; wastewater treatment plants; landfills; methane recovery; greenhouse gases

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1. Introduction

Greenhouse gases (GHGs) are currently one of the most significant environmental concerns due to their involvement in global warming. Among the complete list of GHGs, carbon dioxide and methane are the most common and well known [1]. CO_2 is the most abundant, since it is the most oxidized form of carbon that can be found and is generated in oxidation reactions involving carbon. For its part, CH_4 has a global warming potential 28 times greater than that of CO_2 within a time horizon of 100 years [2].

Anaerobic digestion or decomposition (AD) is a waste management process for biodegradable materials, generating biogas, a combustible gas product consisting mainly of CO_2 and CH_4, and a digestate. This stabilized residue can be used as a soil amendment. On an industrial scale, this biological process can be found in waste biomethanation plants dedicated to biogas production. Thus, these are presented as potentially emitting methane leakage facilities [3]. CH_4 from this sector (B) accounts for around 3% of global anthropogenic GHG emissions [4].

In Europe, landfills are the second largest anthropogenic CH_4 emission source after natural events [5], mostly due to fugitive emissions, accounting for around 7% of the global anthropogenic GHG emissions [6]. These fugitive emissions from landfills (L) are expected to increase by 25% in the next nine years [5], and come mainly from the spontaneous AD of waste deposited in landfills. Managing L is one of the waste industry's priorities to reduce environmental impacts and evaluate the efficiency of gas recovery systems. Nevertheless, the analysis of L is enormously complex due to the large surfaces they occupy and the spatial and temporal variability; the flow data are punctual and often cannot be transported to other areas or sectors of the same landfill [7], so estimation models are essential when trying to evaluate L.

AD is also found in wastewater treatment plants (WWTPs), especially in the sewage sludge stabilization stage, where the organic matter (OM) of sewage sludge is converted into biogas, which, in some cases, is used for heat and electric production (co-generation). The sources of CH_4 fugitive emissions from WWTPs (W) can be found in both water and sludge lines. Up to 26% of the carbon footprint an entire WWTP (36 kg CO_2 eq/year) can be attributed to methane leaks from wastewater treatment and mainly from sludge treatment [8]. A study by Gärtner et al. confirmed that 75% of climate-relevant emissions from WWTPs come from W from sludge treatment, including 6% from raw sludge and 94% from digested sludge [9].

Regarding these three facilities, recent studies have determined that methane leaks can be the source of significant fugitive methane emissions from various locations [10]. Nevertheless, the surging growth of the biogas industry [11] creates new challenges regarding emissions monitoring, quantification, and reduction, especially their determination by means od simple and direct methods, which are based on experimental results but do not require complex and in situ measurements. It is important to study how accurate current estimation methods are for fugitive methane emissions in methane-producing facilities and how they vary across biogas plants, wastewater treatment plants (WWTPs), and landfills. This study reveals that existing methodologies markedly undervalue methane emissions, particularly from wastewater treatment plants (WWTPs) and biogas facilities.

To determine the impact and L, W, and B of different Spanish facilities, other calculation methods were discussed to select the one that best fits a precise estimate, and the appropriate parameters were set based on the context of each installation. To calculate these methane emissions, it was first necessary to make a list with all the input data for all the selected installations and describe their main characteristics. This study's hypotheses are as follows: it is expected that methane emissions from wastewater treatment plants will be higher than those from biogas plants and landfills due to inefficiencies in methane capture. Additionally, biogas plants are anticipated to implement more efficient recovery systems, resulting in lower fugitive emissions. A positive correlation is expected between methane emissions and economic factors such as gross domestic product (GDP) and population density. Furthermore, it is proposed that the estimation models for landfills are more accurate than those for wastewater treatment plants and biogas facilities. Finally, the efficiency of methane capture is expected to vary significantly between regions, regardless of the total number of installations.

2. Materials and Methods

In total, 239 installations across different Spanish regions, including 33 biogas plants, 87 WWTPs and 119 landfills in Spain, were studied. Those facilities that present data or have an appreciable capacity were chosen, thus ignoring those that are small in size.

2.1. Landfills

The measurement of methane emissions in landfills is highly complex and imprecise since it involves large areas with a large spatial and temporal variability. CH_4 measurements are made at points from the landfill's surface to several kilometers away and over different time scales, from minutes to months. Estimating emissions, rather than direct measurement, is a more robust and faster tool for assessing methane leaks. In fact, at present, CH_4 emissions reported for regulatory purposes are usually based on models and calculations [12]. The estimation model must consider several factors that affect CH_4 generation in landfills: the amount of waste dumped; the age of the dumped waste; the composition of the waste; the environmental physical-chemical conditions (humidity, temperature, pH, etc.); the efficiency of the gas collection system from the landfill; and the type of cover [13].

Calculation methods for L are based on a balance of CH_4 in all processes that occur in landfills, considering that methane is generated, but part of it is oxidized. A large part is collected, recovered or eliminated [14], as shown in Equation (1):

$$CH_{4_{emitted}} = CH_{4_{generated}} - CH_{4_{collected}} - CH_{4_{oxidised}} \quad (1)$$

The most used models for L calculation are the Landgem model, GasSim, Level II (IPCC), and ADEME [14]. Table 1 compares the three methods regarding possible advantages and disadvantages to analyze which method best covers some essential features.

Table 1. Some advantages and disadvantages of the different first-order models.

	IPCC (Level 3)	ADEME	GasSIM
Is the fact that part of the biogenic carbon fraction is not degraded considered?	Yes	Yes	Yes
Is sequestered carbon calculated and a value presented?	Yes	No	No
Can the value be calculated accurately (fraction-by-fraction degradability parameters available)?	Yes	No	Yes
Can the value be calculated globally for the entire waste mass (for example, is the mean degradability parameter provided for MSW)?	Yes	No	Yes

Given the results obtained in Table 1, we decided to use the IPCC model with the amendments proposed by Vadillo-Abascal [15]. Each landfill has a different composition depending on the climatic zone and the type of population to which it supplies the service. According to the IPCC method, it was necessary to calculate the amount of degradable organic carbon from all waste deposited in the landfill, susceptible to decomposition, in a year T (DDOCmdT), according to Equation (2), which calculates it as the sum of the DDOCmdT of each fraction of residue (i):

$$DDOCmd_T = \sum_i (W_T \cdot DOC \cdot DOC_f \cdot MCF)_i \quad (2)$$

where WT represents the amount of residue fraction i deposited in a landfill during year T and expressed in tons. DOC is the degradable organic carbon of each residue fraction i during the year of deposit T measured in tons; in other words, it is the content of OM in each type of waste fraction. DOCf represents the fraction of DOC that can be decomposed under anaerobic conditions, and so it is dimensionless. Finally, the thermal MCF refers to the methane correction factor of each residue fraction i, which again is dimensionless. All parameters can be estimated following the IPCC standards and, in the case of compost rejection, the values determined in [15]. According to the MCF, this value varies depending on the type of landfill. Table 2 reflects different MCF values for each sort of landfill stipulated in [15].

Table 2. Value of the MCF parameter depending on the landfill type [15].

Landfill Type	MCF
Managed anaerobic	1
Managed semi-anaerobic	0.5
Unmanaged (>5 m depth)	0.8
Unmanaged (<5 m depth)	0.4

Next, the DDOCmaT was calculated, along with the DDOCmdT accumulated at the end of year T, considering what was generated, accumulated and transformed for the previous years. In the same way, it was calculated as the sum of the DDOCmaT of each fraction or type of residue i, and based on Equation (3):

$$\mathrm{DDOCma_T} = \sum_i (\mathrm{DDOCma_T})_i = \sum_i \left(\mathrm{DDOCmd_T} + \mathrm{DDOCma_{T-1}} \cdot e^{-k}\right)_i \quad (3)$$

where k represents the reaction rate constant (year-1), whose values depend on the type of waste (i) and climatic zone, which are specified in [15].

The next step was to calculate the parameter DDOCm_decompT, which is DDOCm decomposed during the year T, following Equation (4):

$$\mathrm{DDOCm\_decomp_T} = \mathrm{DDOCma_T} \cdot \left(1 - e^{-k}\right) \quad (4)$$

Methane generated in a landfill can be thus calculated with Equation (5):

$$\mathrm{CH_4}_{\mathrm{generated}_T} = \mathrm{DDOCm\_decomp_T} \cdot F \cdot \frac{16}{12} \quad (5)$$

where F represents the volumetric fraction of methane contained in landfill gas; according to [15], a value of 50% was assumed.

To generate the value of methane emitted into the atmosphere, it was necessary to return to Equation (1), which considers the recovered and oxidized methane. If the recovered value was unavailable, it was considered as zero. For its part, the oxidized methane value is regarded as 10% generically.

To understand the calculation mentioned above sequence, the block diagram from Figure 1 explains the steps followed with all the parameters and equations involved.

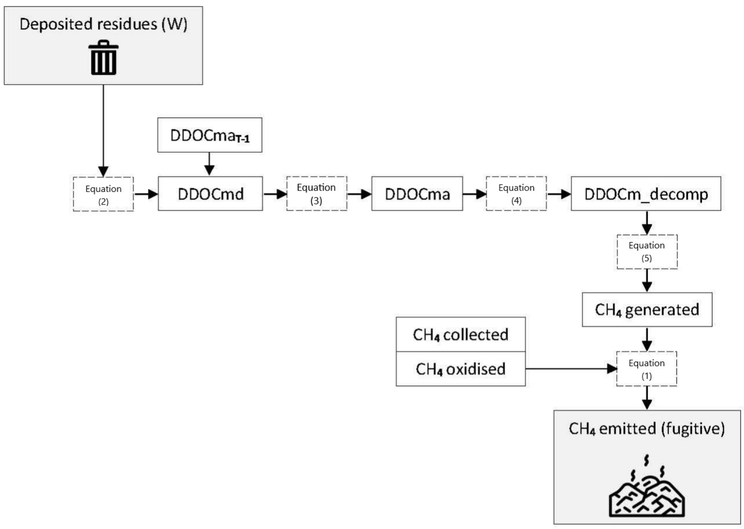

Figure 1. Block diagram of the calculation scheme for methane emitted to the atmosphere in a landfill, based on [15].

2.2. Wastewater Treatment Plants

In WWTPs, the phenomenon of AD also occurs, mainly in anaerobic digesters for sludge stabilization, where the OM of the sludge is converted into biogas. Unfortunately, part of the biogas is lost during the process due to leaks, the entrainment of gas bubbles, and residual gas being generated due to the remaining OM in the sewage sludge during its transfer and treatment [16]. Although studies on direct methane emissions from anaerobic digestion reactors are lacking, there are numerous research works on methane emission sources from WWTP components in the literature, such as sludge thickeners, intermediate storage tanks, and previous sludge dehydration [17].

The flux-chamber method is a reliable and commonly used method for continuously measuring gas emissions at the digester sludge outlet: a gas-tight membrane collects gas emissions from the sludge shaft at the digester's head [18]. This method is commonly used in many studies for various WWTPs worldwide. However, to carry out the calculation, it is necessary to know all the flows that affect the digester. A method of calculating emissions emerged, developed by the IPCC, which only requires as the input data the total flow treated in the WWTP, calculated with Equation (6).

$$CH_{4_{emitted}} = Bo \cdot MCF \cdot (TOW - S) - R \quad (6)$$

where Bo represents the maximum CH_4 producing capacity (kg CH_4/kg BOD), which is usually set by default to 0.6; MCF corresponds to the CH_4 correction factor; TOW stands for the total amount of OM entering the treatment facility per year (kg BOD/year); S is the organic component removed as sludge per year (kg BOD/year); and R represents the amount of CH_4 recovered, collected, or flared per year (kg CH_4/year).

The value of MCF is critical for the correct calculation of CH_4 emissions, so it must be adjusted according to the type of facility. The values of the MCF, different and more accurate than those proposed in the IPCC, correspond to those established by [19] and are shown in Table 3. The MCF values used in this study were derived from the IPCC Guidelines for National Greenhouse Gas Inventories, as well as from recent studies that recommend adjustments based on specific waste types and environmental conditions. These values provide a standardized approach to estimating methane emissions, although we acknowledge that local conditions may vary.

Table 3. Summary of the proposed new entries and default MCF values for domestic wastewater treatment, based on Table 6.3 of the IPCC Guidelines [14].

Type of Treatment and Discharge Pathway or System	Comments	Proposed MCF	IPCC's MCF
Centralized, aerobic treatment plant	It must be well managed. Some CH_4 can be emitted from settling basins and other pockets.	0.06	0.0
Centralized, aerobic treatment plant with anaerobic sludge digesters	It must be well managed. Some CH_4 can be emitted from settling basins and other pockets. Fugitive emissions from digesters are considered.	0.32	-
Centralized, anaerobic (or anoxic) aerobic treatment plant	It must be well managed. Some CH_4 can be emitted from settling basins and other pockets.	0.08	-
Centralized, anaerobic (or anoxic) aerobic treatment plant with anaerobic sludge digesters	It must be well managed. Some CH_4 can be emitted from settling basins and other pockets. Fugitive emissions from digesters are considered.	0.34	-

2.3. Biogas Plants

The biogas sector in Europe has grown tremendously in recent years. At the end of 2018, more than 18,000 biogas plants were operating [20]. The IEA bioenergy report [21] states that most are associated with municipal wastewater treatment plants and landfill gas power units, with approximately 52 and 129, respectively. The primary uses of biogas are for electricity production, heat, and combined heat and power. Excess biogas is normally flared to reduce potential CH_4 emissions. Future opportunities exist for its application in the intensive livestock and food processing industries, driven by the readily available feedstock from process waste, higher electricity prices, and demand for on-site electricity, heat, or steam [22].

The most used technology in this type of installation is AD in concrete tanks with integrated membrane domes [23]. This type of digester is similar to that used in the AD of sewage sludge. Even its operation and maintenance are similar. Therefore, the method for calculating B is the same as that used for calculating emissions in treatment plants; that is, Equation (6) can be used for both facilities.

3. Results and Discussion

The raw results obtained for the fugitive methane emissions of each facility can be found in Appendix A. These results are displayed with L/W/B-XX-YY coding. L/W/B refers to whether the facility is a landfill (L), a wastewater treatment plant (W), or a biogas plant (B). The XX code refers to the region where the plant is located, which can be obtained by consulting Figure 1, and YY lists the different facilities within the XX region. The regions analyzed, numbered 61–77, correspond to the autonomous communities of Spain. This classification was chosen to simplify the aggregation and comparison of methane emissions across various areas, as each community may have distinct regulatory frameworks and environmental policies influencing emissions. The results highlight significant variations in methane emissions between these regions, indicating that factors such as local waste management practices, population density, and economic activities play a crucial role in emission levels.

The following figures and tables summarize the obtained data, which were calculated for 2020 as the latest official data available [24]. Figure 2 illustrates the facilities likely to emit methane by region. In contrast, Figure 3 quantitatively represents the emissions generated by each type of facility so that it is possible to obtain an idea of how each class contributes to the total emissions.

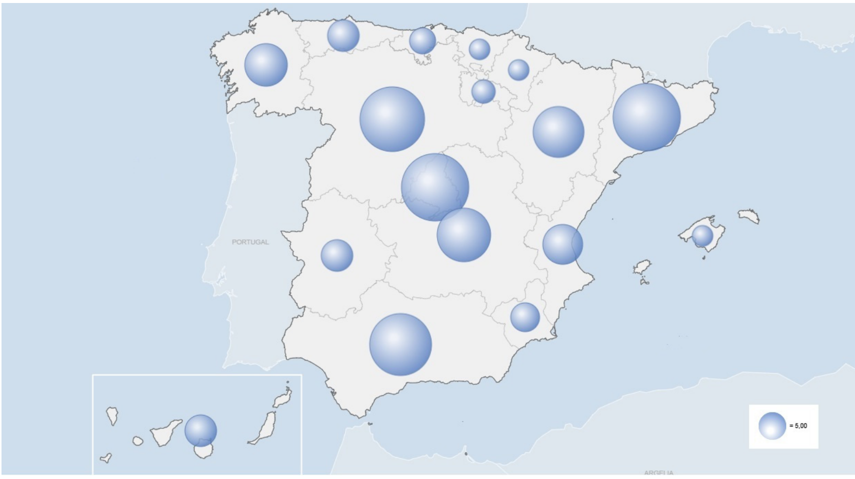

Figure 2. Installations susceptible to releasing CH_4 fugitive emissions.

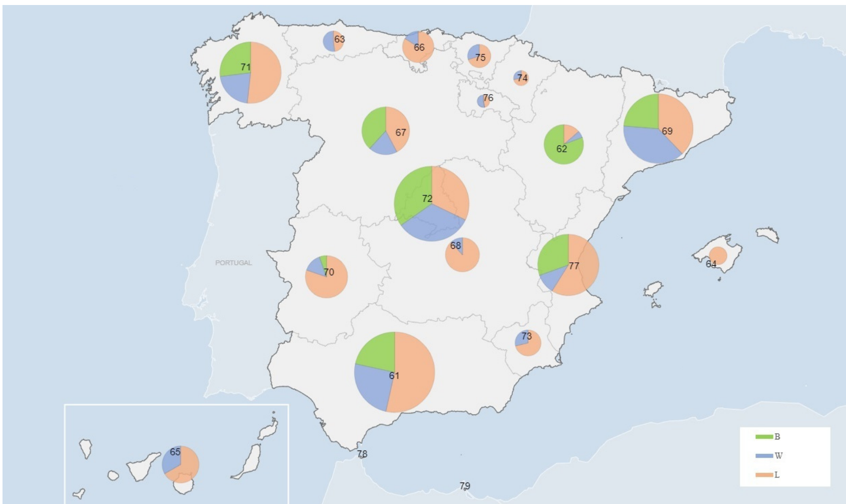

Figure 3. Fugitive methane emissions by region and type of facility.

This result can serve as a reference for studies of environmental impacts and how each facility contributes to greenhouse gas emissions. Table 4 shows numerical data on the contribution by region of each type of installation, indicating that, among all emissions, 51% comes from landfills, and the other half is divided equally between fugitive emissions from WWTPs and biogas plants. This is to be expected and also corroborates the stipulations of [7], since L occurs in large areas of the surface, that is, across the landfill, and its production is spontaneous, being more uncontrolled and difficult to capture; on the other hand, W and B are generated in delimited and controlled reactors, so, although the generation of CH_4 is enhanced and is the main objective, leaks are a minority, and are detected and corrected quickly.

Table 4. Summary of fugitive CH_4 emissions, calculated by region and facility type using the methods described in the manuscript, and parameters for each region in 2020.

Year	Region	N Inst	L	W	B	Total		Efficiency Level (0–10)	Extension [km²]	P [M hab]	GDP [MEUR]
				[kg CH_4/year]							
2020	61	28	179.44	83.48	72.48	335.40	(18%)	3	87,599	8.52	150,557
	62	20	11.38	4.65	67.30	83.33	(4%)	8	47,720	1.31	35,290
	63	8	11.89	12.95	0.00	24.84	(1%)	9	10,604	1.01	21,475
	64	3	19.53	0.00	0.00	19.53	(1%)	7	4992	1.22	26,789
	65	8	47.84	23.85	0.00	71.69	(4%)	5	7447	2.25	39,163
	66	5	46.10	8.79	0.00	54.89	(3%)	4	5321	0.58	12,867
	67	30	49.25	22.75	44.57	116.56	(6%)	9	94,224	2.38	55,401
	68	22	54.87	7.27	0.00	62.14	(3%)	9	79,461	2.05	39,573
	69	32	95.64	96.94	59.92	252.49	(13%)	6	32,113	7.68	212,931
	70	8	78.32	14.06	5.28	97.66	(5%)	3	41,634	1.05	19,386
	71	14	101.11	41.74	52.67	195.52	(10%)	2	29,575	2.69	59,105
	72	33	96.83	99.97	104.64	301.44	(16%)	5	8028	6.77	216,527
	73	6	26.17	10.54	0.00	36.72	(2%)	7	11,314	1.52	29,940
	74	3	9.52	3.70	0.00	13.22	(1%)	8	10,391	0.66	19,265
	75	3	20.81	8.94	0.00	29.76	(2%)	5	7234	2.18	66,558
	76	4	4.40	4.93	0.00	9.33	(0%)	10	5045	0.32	8129
	77	12	117.41	21.09	60.72	199.22	(10%)	0	23,225	5.07	104,724
2020	TOTAL Added	239	970.53 (51%)	465.65 (24%)	467.57 (25%)	1903.75 (100%)	(100%)		505,927	47.26	1,117,680

Regarding the analysis by region, it is observed that the region that produces the most fugitive emissions of CH_4 is region 61, while the one that emits the least is region 76. An analysis of the data shows that there is no clear relationship between total fugitive emissions and the number of installations since, for example, regions 61, 67 and 69 have a similar number of installations (28, 30, and 32, respectively). However, the fugitive emissions they emit are disparate, being 18%, 6%, and 13% of the total, respectively. This means that the efficiency of the facilities, and therefore the generation of fugitive emissions, does not depend on the total number of installations. To compare one region with another, no longer in gross terms of emission, but to know which one has facilities with less leakage, they were classified according to the efficiency of the facilities, rating them between 0 and 10. In this way, the least efficient region is 77, which, with 12 installations, emits 10% of the leaks; in descending order, the most efficient are 76, 63, 67, and 68.

Indicators that characterize the regions were obtained, such as their extension, population (P), and gross domestic product (GDP). It is observed in the analyzed data in Table 4 that there may be a correlation between fugitive emissions and these indicators. For example, region 61, which generates the most fugitive emissions, is the largest and one of the most populated and has one of the highest GDPs; region 72, which is another large emitter, has the highest GDP and population. By adjusting least squares and analyzing the correlation coefficient R2, it is observed that fugitive emissions of CH_4 are well adjusted with GDP or population but not with the extension of territory. This conclusion is logical considering that the larger the population and the greater the GDP, the greater the amount of waste and wastewater generated; thus, the methane emissions would be greater [25]. Table 5 shows the result of these correlations, where L, W, B, and total fugitive emissions (T) represent fugitive methane emissions per region, measured in kg of CH_4 per year and GDP and *p* values are entered in millions of EUR and millions of inhabitants, respectively.

Table 5. Arithmetical relations and correlation coefficient between fugitive methane emissions from landfills (L), WWTPs (W) and biogas plants (B), in $kgCH_4$/year, as a function of GDP, in MEUR, and population (P) in millions of inhabitants.

	Extension	GDP		Population	
L	$R^2 = 0.204$	$R^2 = 0.772$	$L(GDP) = 0.4983{\cdot}GDP + 24{,}328$	$R^2 = 0.706$	$L(P) = 15{,}666{\cdot}P + 13{,}593$
W	$R^2 = 0.04$	$R^2 = 0.866$	$W(GDP) = 0.4698{\cdot}GDPP - 3498$	$R^2 = 0.838$	$W(P) = 11{,}745{\cdot}P - 5361.4$
B	$R^2 = 0.113$	$R^2 = 0.638$	$B(GDP) = 0.424{\cdot}GDP - 327.36$	$R^2 = 0.604$	$B(P) = 106{,}013{\cdot}P - 1965.7$
T estimation	$R^2 = 0.146$	$R^2 = 0.881$	$T(GDP) = 6{\cdot}10^{-25}{\cdot}(GDP)^6 - -4{\cdot}10^{-19}{\cdot}(GDP)^5 + +8{\cdot}10^{-14}{\cdot}(GDP)^4 - -8{\cdot}10^{-9}{\cdot}(GDP)^3 + +4{\cdot}10^{-4}{\cdot}(GDP)^2 + -6.1597{\cdot}GDP + +59{,}298$	$R^2 = 0.871$	$T(P) = 39.611{\cdot}P^6 - -943.62{\cdot}P^5 + +8539.4{\cdot}P^4 - -37{,}638{\cdot}P^3 + +85{,}864{\cdot}P^2 - -52{,}201{\cdot}P + +35{,}219$

While the R2 coefficient of 0.7 indicates a significant correlation between population density and methane emissions, it is essential to acknowledge that correlation does not imply causation. The production of CH_4 is influenced by various factors, including waste management practices, operational efficiencies, and the specific characteristics of each facility.

The results of these adjustments are presented in Figure 4. Each subfigure shows, through stacked areas, the estimates of fugitive emissions for each type of facility (L, B and W) and, with a continuous line, the estimation of total emissions (T estimation). The sum of the stacked areas L, B, and W gives the estimated value of total emissions T added. When comparing the results of the total fugitive emissions, either calculated by the sum of the stacked areas (T added) or by direct estimation from the mathematical regression (T estimation), the relative error between both options is found to be 0.00%, meaning they are practically the same, as shown in Table 6. An ANOVA analysis with a 95% confidence interval is developed with DMS and Tukey contrasts, between the estimates obtained by T added and those obtained by T estimation, and in any of the cases where the *p*-value is 1.00 (*p*-value > 0.05), as shown in the extract of the analysis shown in Table 6, this means that the initial hypothesis of equality of means between T added and T estimation can be assumed. It is then concluded that the total emissions can be estimated directly by means of mathematical regression since the result is practically the same (ε = 0.00% y *p*-value = 1.00), as in the case of estimating the fugitive emissions for each facility and adding them up.

The results indicate that CH_4 production is influenced by a combination of factors beyond merely the number of facilities or the total amount of waste processed. For instance, the efficiency of methane recovery systems, the specific technologies employed in waste treatment, and the operational practices within each facility play critical roles in determining emission levels. Additionally, regional differences in waste composition and local regulations can significantly impact the effectiveness of methane capture and overall emissions [26]. Therefore, while population density shows a correlation with methane emissions, it is essential to consider these multifactorial influences to fully understand the dynamics of CH_4 production.

Both suppositions, which means estimating according to GDP or P, adjust correctly. However, the evolution of GDP is more precise, in general and in total terms, compared to the population (as can be observed when analyzing the correlation factor R2). However, GDP fluctuates, and estimates will be tighter. Again, an ANOVA analysis was performed between the results of both estimations, with a 95% confidence interval, and accompanied by a DMS and Tukey contrast analysis. The *p*-value is higher than the significance level, so the equality of means between both estimations can be assumed. On average, the estimates based on GDP and P can be considered the same according to ANOVA analyses and can be assumed by the mean estimation of both. The mean estimation is represented as a dotted line in both cases in Figure 4, and, according to the low relative error between curves and the ANOVA analysis carried out, both shown in Table 6, it is concluded that both estimations can be considered the same, and assumed by the mean estimation (T mean estimation) between the estimation of T as a function of GDP and T as a function of P. Notably, since the estimates were developed based on the year 2020, for which the most recent official data are available to calculate them, the relative error was 0.00% in all cases.

According to this mean estimation and current GDP and P forecasts for the years 2023 and 2024, published in 2022 [27], as shown in the graphs in Figure 4 and results in Table 6, fugitive methane emissions were expected to fall in 2023 by 14.94 tons of CH_4—that is, −0.77%. However, by 2024, the path was expected to be increasing by 0.59%; that is, 11.41 tons of CH_4.

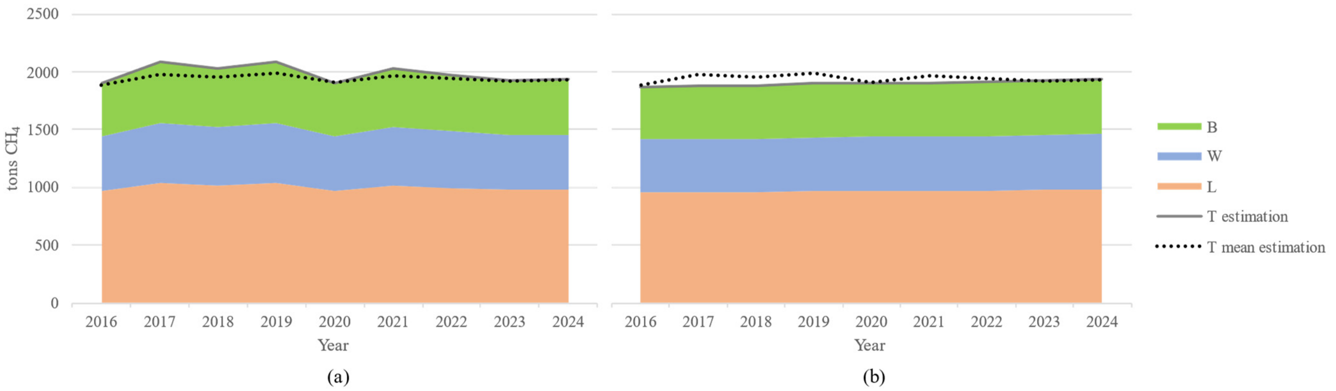

Figure 4. Arithmetical relations and linear correlation coefficient between fugitive methane emissions from landfills (L), WWTPs (W) and biogas plants (B) as a function of GDP (**a**) and population (**b**).

Table 6. Fugitive emissions estimates of CH4 for different years, by adding the estimates for each facility (T added), by direct estimation (T estimation) and the mean estimation curve (T mean estimation).

Year	T as Function of GDP				T as Function of P				T Mean Estimation	T as Function of GDP		T as Function of P	
	T Added	T Estimation	ε	*p*-Value	T Added	T Estimation	ε	*p*-Value		ε	*p*-Value	ε	*p*-Value
	[Tons CH_4/Year]		(T Added—T Estimation)		[Tons CH_4/Year]		(T Added—T Estimation)		[Tons CH_4/Year]	(T Estimation—T Mean Estimation)		(T Estimation—T Mean Estimation)	
2016	1904.13	1904.14	0.00%	1.00	1872.97	1888.55	0.00%	1.00	1888.55	−0.82%	1.00	0.83%	1.00
2017	2081.66	2081.66	0.00%	1.00	1876.01	1978.84	0.00%	1.00	1978.84	−4.94%	0.90	5.48%	0.90
2018	2023.67	2023.68	0.00%	1.00	1883.61	1953.64	0.00%	1.00	1953.64	−3.46%	1.00	3.72%	1.00
2019	2081.66	2081.66	0.00%	1.00	1897.67	1989.67	0.00%	1.00	1989.67	−4.42%	0.90	4.85%	0.90
2020	1903.70	1903.71	0.00%	1.00	1903.76	1903.73	0.00%	1.00	1903.73	0.00%	1.00	0.00%	1.00
2021	2027.82	2027.83	0.00%	1.00	1906.42	1967.12	0.00%	1.00	1967.12	−2.99%	1.00	3.18%	1.00
2022	1970.61	1970.61	0.00%	1.00	1910.22	1940.42	0.00%	1.00	1940.42	−1.53%	1.00	1.58%	1.00
2023	1927.04	1927.05	0.00%	1.00	1923.90	1925.47	0.00%	1.00	1925.47	−0.08%	1.00	0.08%	1.00
2024	1934.82	1934.83	0.00%	1.00	1938.96	1936.89	0.00%	1.00	1936.89	0.11%	1.00	−0.11%	1.00

3.1. Sources of Uncertainty

It is important to remember that studies using emission records always present high uncertainty [28], as these emission inventories are calculated, not taken in situ with special devices. In addition, several installations do not present these data, so they have not entered the comparison.

Regarding biogas plants, the scarcity of data on inlet flows for specific areas or installations further increases the uncertainty and is a considerable problem for analyzing methane emissions. This lack of information is due to the small number of biogas plants and the current lack of regulation.

For its part, in the case of landfills, it is vitally important to understand the large area they occupy and the complications related to obtaining a reliable value. Even with in situ measurements, the data are highly distorted due to spatial and temporal variability. Additionally, specific parameters, such as recovered methane (R), are not always suitable for every facility. Therefore, this also entails a certain degree of uncertainty in the results.

Regarding wastewater treatment plants and MCF values, the values used in calculations are based on established guidelines, and the accuracy of methane emissions estimations is inherently subject to uncertainty. Future research should aim to validate these calculations with direct measurements of methane emissions to provide a more robust understanding of emissions dynamics and improve the accuracy of the MCF values used in such estimations.

To obtain more accurate data, direct measurements of pollutant concentration by sampling should be used, which is not feasible in the case of landfills, for example. Parameters must be updated via experimental analyses and meta-analyses, and more interdisciplinary studies must be conducted to estimate future context [29].

3.2. Comparison and Discussion of Results with Previous Literature

This comparative analysis of fugitive methane emissions from biogas plants, WWTPs, and landfills shows significant differences in emissions behavior, which is key to optimizing methane recovery as a fuel. In the results obtained, landfills contribute to 51% of total fugitive methane emissions, an expected trend given their large surface area and the uncontrolled nature of methane generation. This is in line with previous studies such as those by Spokas et al., who noted that methane emissions in landfills are difficult to capture due to the spatial and temporal variability of emissions [7]. Biogas plants and WWTPs, which account for the remaining 49% of emissions, present more controlled environments where methane leaks are easier to detect and correct. Shen et al. indicate that less than 10% of WWTPs in the US fully utilize biogas, underscoring the opportunity to improve methane capture and utilization at these facilities [30].

The analysis by regions reveals that methane emissions are not necessarily related to the number of facilities in each region, but to their operational efficiency. Regions 61 and 77, despite having fewer installations, have higher fugitive emissions, reflecting inefficiencies in gas capture. This is in line with the findings of Schirmer and Crovador, who highlighted that many landfills in Brazil are not adequately equipped to recover energy from biogas, resulting in higher fugitive emissions [31]. In addition, the correlations between methane emissions, GDP, and population are remarkable. A study by Mingxi Du et al. highlighted that regions with higher GDP and population density tend to generate more waste and therefore more methane emissions, as also observed in our results [32]. This relationship is due to the fact that greater economic and urban activity generates more solid waste and wastewater, increasing the amount of methane produced.

The emissions estimate showed a slight decrease in 2023, followed by an increase in 2024, driven by economic and population growth. These fluctuations are consistent with studies such as that of Mroueh et al., who analyzed the economic feasibility of biogas capture projects in waste management facilities, noting that return on investment and technological efficiency can significantly influence projected emissions [33]. Despite progress in reducing emissions, uncertainty remains an important factor, especially in biogas plants, where data on input flows are limited. Spokas et al. also noted that the accurate measurement of landfill emissions is complicated due to spatial and temporal variability, suggesting that more studies are needed to improve emissions estimates [34].

4. Conclusions

Waste treatment facilities pose an environmental problem due to their high CH_4 generation. A methane recovery or storage system is significant in preventing large amounts of methane from being emitted into the atmosphere. However, leaks can always occur in lines, digesters, etc. For this reason, it is crucial to have not only measurement and monitoring techniques for possible leaks to correct them as soon as possible but also predicting techniques of future fugitive emissions, as their environmental impact is noticeable.

The fugitive emissions data obtained in this study show how they reach 1.9 tons per year, with 51% from landfills. The remaining 49% is distributed, in equal parts, between treatment plants with anaerobic digestion processes and biogas plants. For each region, these results can be considered as a function of GDP and/or population in that area, with a correlation coefficient of R2 = 0.881. In any case, it has been shown that both correlations are valid, the errors are minimal, and can be considered statistically equal and assumed by the average of both estimates. Thanks to these relationships, it was possible to estimate a decrease of −0.77% in 2023 and then an increase of +0.59% in 2024.

The high amounts of fugitive emissions obtained in this study reveal the need for more research and design of climate change mitigation and energy use policies.

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Appendix A

Table A1. Raw fugitive methane emissions data, calculated for each facility using the methods described in this manuscript.

Codification	Fugitive CH_4 Emissions (Calculated) [kg CH_4/Year]	Facility	Region
L-61-1	3318.86	Landfill	61
L-61-2	4141.89	Landfill	61
L-61-3	38.87	Landfill	61
L-61-4	7560.81	Landfill	61
L-61-5	9028.67	Landfill	61
L-61-6	9018.03	Landfill	61
L-61-7	14,023.03	Landfill	61
L-61-8	9008.60	Landfill	61

Table A1. *Cont.*

Codification	Fugitive CH_4 Emissions (Calculated) [kg CH_4/Year]	Facility	Region
L-61-9	7209.19	Landfill	61
L-61-10	21,474.23	Landfill	61
L-61-11	7346.00	Landfill	61
L-61-12	11,309.19	Landfill	61
L-61-13	2654.32	Landfill	61
L-61-14	6321.28	Landfill	61
L-61-15	4.20	Landfill	61
L-61-16	16,365.20	Landfill	61
L-61-17	13,299.79	Landfill	61
L-61-18	9334.25	Landfill	61
L-61-19	20,718.43	Landfill	61
L-61-20	5780.65	Landfill	61
L-61-21	1481.66	Landfill	61
L-62-1	2577.40	Landfill	62
L-62-2	1629.98	Landfill	62
L-62-3	1356.20	Landfill	62
L-62-4	629.03	Landfill	62
L-62-5	1100.91	Landfill	62
L-62-6	1315.70	Landfill	62
L-62-7	1364.04	Landfill	62
L-62-8	1410.30	Landfill	62
L-63-1	11,891.02	Landfill	63
L-66-1	46,104.64	Landfill	66
L-68-1	28,315.51	Landfill	68
L-68-2	8322.62	Landfill	68
L-68-3	3364.64	Landfill	68
L-68-4	7239.85	Landfill	68
L-68-5	3233.16	Landfill	68
L-68-6	4395.62	Landfill	68
L-67-1	10,430.88	Landfill	67
L-67-2	3084.91	Landfill	67
L-67-3	582.72	Landfill	67
L-67-4	1332.61	Landfill	67
L-67-5	2227.99	Landfill	67
L-67-6	8116.72	Landfill	67
L-67-7	2973.66	Landfill	67
L-67-8	5998.44	Landfill	67
L-67-9	2570.76	Landfill	67

Table A1. *Cont.*

Codification	Fugitive CH_4 Emissions (Calculated) [kg CH_4/Year]	Facility	Region
L-67-10	1904.77	Landfill	67
L-67-11	1407.62	Landfill	67
L-67-12	8614.55	Landfill	67
L-69-1	3084.91	Landfill	69
L-69-2	5022.24	Landfill	69
L-69-3	21,835.84	Landfill	69
L-69-4	6351.28	Landfill	69
L-69-5	1445.75	Landfill	69
L-69-6	0.00	Landfill	69
L-69-7	11,263.91	Landfill	69
L-69-8	4913.06	Landfill	69
L-69-9	5458.96	Landfill	69
L-69-10	10,917.92	Landfill	69
L-69-11	701.80	Landfill	69
L-69-12	2456.31	Landfill	69
L-69-13	1637.69	Landfill	69
L-69-14	9369.10	Landfill	69
L-69-15	649.18	Landfill	69
L-69-16	473.73	Landfill	69
L-69-17	1368.56	Landfill	69
L-69-18	1315.94	Landfill	69
L-69-19	0.00	Landfill	69
L-69-20	982.61	Landfill	69
L-69-21	5458.96	Landfill	69
L-69-22	929.88	Landfill	69
L-77-1	666.76	Landfill	77
L-77-2	13,979.85	Landfill	77
L-77-3	12,688.81	Landfill	77
L-77-4	27,839.27	Landfill	77
L-77-5	8882.16	Landfill	77
L-77-6	8036.24	Landfill	77
L-77-7	8734.34	Landfill	77
L-77-8	15,438.05	Landfill	77
L-77-9	8882.16	Landfill	77
L-77-10	12,265.85	Landfill	77
L-70-1	35,461.73	Landfill	70
L-70-2	4574.31	Landfill	70
L-70-3	2997.38	Landfill	70

Table A1. *Cont.*

Codification	Fugitive CH_4 Emissions (Calculated) [kg CH_4/Year]	Facility	Region
L-70-4	32,697.76	Landfill	70
L-70-5	677.68	Landfill	70
L-70-6	1909.33	Landfill	70
L-71-1	2267.51	Landfill	71
L-71-2	83,987.77	Landfill	71
L-71-3	14,855.57	Landfill	71
L-64-1	3199.44	Landfill	64
L-64-2	14,805.08	Landfill	64
L-64-3	1526.97	Landfill	64
L-65-1	6903.77	Landfill	65
L-65-2	3614.47	Landfill	65
L-65-3	17,631.48	Landfill	65
L-65-4	9925.85	Landfill	65
L-65-5	6072.54	Landfill	65
L-65-6	349.38	Landfill	65
L-65-7	685.66	Landfill	65
L-65-8	2656.12	Landfill	65
L-76-1	28.31	Landfill	76
L-76-2	4367.17	Landfill	76
L-72-1	11,688.07	Landfill	72
L-72-2	41,707.27	Landfill	72
L-72-3	32,314.63	Landfill	72
L-72-4	11,121.94	Landfill	72
L-73-1	9036.57	Landfill	73
L-73-2	4169.71	Landfill	73
L-73-3	2801.66	Landfill	73
L-73-4	513.45	Landfill	73
L-73-5	9653.59	Landfill	73
L-74-1	3371.94	Landfill	74
L-74-2	2148.68	Landfill	74
L-74-3	4004.17	Landfill	74
L-75-1	15,944.39	Landfill	75
L-75-2	3409.21	Landfill	75
L-75-3	1461.09	Landfill	75
W-72-1	21,835.12	WWTP	72
W-72-2	12,525.25	WWTP	72
W-72-3	11,929.29	WWTP	72
W-72-4	7397.12	WWTP	72
W-72-5	6724.65	WWTP	72

Table A1. *Cont.*

Codification	Fugitive CH_4 Emissions (Calculated) [kg CH_4/Year]	Facility	Region
W-72-6	6724.65	WWTP	72
W-72-7	5699.14	WWTP	72
W-72-8	2911.60	WWTP	72
W-72-9	2334.95	WWTP	72
W-72-10	337.40	WWTP	72
W-72-11	3127.00	WWTP	72
W-72-12	2023.62	WWTP	72
W-72-13	4423.95	WWTP	72
W-72-14	2918.69	WWTP	72
W-72-15	2689.86	WWTP	72
W-72-16	1011.81	WWTP	72
W-72-17	817.23	WWTP	72
W-72-18	583.74	WWTP	72
W-72-19	50.59	WWTP	72
W-72-20	120.64	WWTP	72
W-72-21	68.10	WWTP	72
W-72-22	778.32	WWTP	72
W-72-23	1416.07	WWTP	72
W-72-24	77.83	WWTP	72
W-72-25	1362.05	WWTP	72
W-72-26	25.03	WWTP	72
W-72-27	51.37	WWTP	72
W-62-1	1344.93	WWTP	62
W-62-2	801.67	WWTP	62
W-62-3	56.95	WWTP	62
W-62-4	933.98	WWTP	62
W-62-5	466.99	WWTP	62
W-62-6	33.08	WWTP	62
W-62-7	0.00	WWTP	62
W-62-8	0.00	WWTP	62
W-62-9	1011.81	WWTP	62
W-67-1	46.12	WWTP	67
W-67-2	3930.50	WWTP	67
W-67-3	6070.87	WWTP	67
W-67-4	163.45	WWTP	67
W-67-5	4572.61	WWTP	67
W-67-6	34.74	WWTP	67
W-67-7	1284.22	WWTP	67
W-67-8	1751.21	WWTP	67

Table A1. *Cont.*

Codification	Fugitive CH_4 Emissions (Calculated) [kg CH_4/Year]	Facility	Region
W-67-9	319.85	WWTP	67
W-67-10	1606.44	WWTP	67
W-67-11	101.18	WWTP	67
W-67-12	1867.96	WWTP	67
W-67-13	803.22	WWTP	67
W-67-14	194.58	WWTP	67
W-63-1	160.72	WWTP	63
W-63-2	537.97	WWTP	63
W-63-3	6.30	WWTP	63
W-63-4	5086.37	WWTP	63
W-63-5	1639.13	WWTP	63
W-63-6	175.12	WWTP	63
W-63-7	5346.10	WWTP	63
W-68-1	1751.21	WWTP	68
W-68-2	120.64	WWTP	68
W-68-3	933.98	WWTP	68
W-68-4	33.08	WWTP	68
W-68-5	46.70	WWTP	68
W-68-6	20.55	WWTP	68
W-68-7	34.21	WWTP	68
W-68-8	1926.33	WWTP	68
W-68-9	134.26	WWTP	68
W-68-10	54.48	WWTP	68
W-68-11	1400.97	WWTP	68
W-68-12	46.70	WWTP	68
W-68-13	116.75	WWTP	68
W-68-14	466.99	WWTP	68
W-68-15	159.55	WWTP	68
W-68-16	26.85	WWTP	68
W-76-1	4034.79	WWTP	76
W-76-2	895.06	WWTP	76
W-66-1	4256.69	WWTP	66
W-66-2	4421.46	WWTP	66
W-66-3	0.00	WWTP	66
W-66-4	108.96	WWTP	66
W-71-1	1362.05	WWTP	71
W-71-2	26,898.61	WWTP	71
W-71-3	5253.63	WWTP	71
W-71-4	3026.09	WWTP	71

Table A1. *Cont.*

Codification	Fugitive CH_4 Emissions (Calculated) [kg CH_4/Year]	Facility	Region
W-71-5	2977.06	WWTP	71
W-71-6	200.81	WWTP	71
W-71-7	2017.40	WWTP	71
W-71-8	4.67	WWTP	71
B-71-1	4669.90	Biogas plant	71
B-71-2	28,800.00	Biogas plant	71
B-71-3	19,200.00	Biogas plant	71
B-69-1	0.00	Biogas plant	69
B-69-2	5280.00	Biogas plant	69
B-69-3	5870.40	Biogas plant	69
B-69-4	24,960.00	Biogas plant	69
B-69-5	0.00	Biogas plant	69
B-69-6	8040.00	Biogas plant	69
B-69-7	0.00	Biogas plant	69
B-69-8	0.00	Biogas plant	69
B-69-9	15,768.00	Biogas plant	69
B-69-10	0.00	Biogas plant	69
B-62-1	67,200.00	Biogas plant	62
B-62-2	96.00	Biogas plant	62
B-62-3	0.00	Biogas plant	62
B-67-1	28,800.00	Biogas plant	67
B-67-2	15,768.00	Biogas plant	67
B-67-3	0.00	Biogas plant	67
B-67-4	0.00	Biogas plant	67
B-72-1	0.00	Biogas plant	72
B-72-2	104,640.00	Biogas plant	72
B-77-1	41,520.00	Biogas plant	77
B-77-2	19,200.00	Biogas plant	77
B-70-1	5280.00	Biogas plant	70
B-70-2	0.00	Biogas plant	70
B-61-1	14,880.00	Biogas plant	61
B-61-2	0.00	Biogas plant	61
B-61-3	0.00	Biogas plant	61
B-61-4	0.00	Biogas plant	61
B-61-5	28,800.00	Biogas plant	61
B-61-6	28,800.00	Biogas plant	61
B-61-7	0.00	Biogas plant	61
B-73-1	0.00	Biogas plant	73

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