THE UPGRADING OF A VARIABLE QUALITY LANDFILL BIOGAS TO BIOMETHANE – FEEDBACK FROM THE OPERATION OF SEVERAL UPGRADING UNITS

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ABSTRACT: The upgrading of the landfill biogas to biomethane, is a challenge for the landfill operators. The WAGABOX® technology was designed specifically to upgrade the landfill biogas characterized by high variability in composition and in flow rate. It combines several upgrading technologies (based on membrane gas permeation and cryogenic distillation). This article analyzes field data from 10 landfill gas WAGABOX® upgrading units and focuses on deviations in the landfill gas composition that occur during 1 year of operation. More than five hundred thousand (500 000) samples have been analyzed for each unit. The analysis of data demonstrates that WAGABOX® units ensure the upgrading of landfill biogas irrespective of its great hourly, daily and seasonal variability. The biomethane produced has a stable composition and meets the specifications defined for injection into the natural gas distribution or transport grid. The uptime and recovery rate of the units are not affected either by the variability of the composition of the biogas nor by the nitrogen content reaching up to 25% in some cases.

Keywords: Biogas, Biomethane, Upgrading, Big data, Nitrogen, Oxygen, Methane, GHG, Monitoring, Meteorological factors

1. INTRODUCTION

The methane (CH_4) contained in biogas (also referred to as landfill gas) produced by the degradation of waste in landfills constitutes an abundant renewable energy, but also a powerful greenhouse gas contributor if not captured. The contribution of CH_4 direct emissions to global warming is 28 times that of carbon dioxide (CO_2) on a 100-year timescale (Stocker and Qin: Climate Change 2013). Over a 20-year timescale, methane's global warming potential (GWP20) is 81.2 times that of CO_2 (Smith et al., 2021, Contribution of Working Group I to the Sixth Assessment Report of the IPCC).

Even if the construction, the equipment, and the cover of the landfill are designed to reduce exchanges between waste mass and external environment, the landfill system is not fully airtight. It is assumed that the presence of air (N_2 and O_2) in the biogas, associated with an efficient vacuum applied on wellheads, is an indicator of capture efficiency. An efficient biogas collection system would capture at least 90%-95% of the biogas produced once final cover is installed.

"Landfill-gas methane content is usually from 45 to 55 %, carbon dioxide from 30 to 40 % and nitrogen from 5 to 15 %" (Jonsson et al, 2003). Even though available information is often within this range, the concentration profile of the landfill gas is wider, as shown in this paper, and higher concentrations in nitrogen have been frequently observed: up to 25% of the landfill gas associated with an oxygen concentration below 3%.

According to local standards and regulations, the dilution of landfill biogas with air (in case of high vacuum applied) can be used as a strategy to reduce CH_4 emission, comply with air permit and reduce potential odor nuisances. Additionally, the composition and flow of the landfill biogas is affected by other parameters such as atmospheric pressure, temperature, humidity, and wind that can vary over short time periods (sometimes, on an hourly basis). The effect of climatic variations has been studied by different research teams in different geographies (Meres et al., 2004; Janeckek et al., 1995)

Daily and seasonal variations have been documented by several studies confirming that the monitoring of meteorological parameters becomes a key indicator to manage properly the biogas collection and to reduce methane emissions into the atmosphere (Christophersen et al., 2001; Uyanik et al., 2012; Borjesson et al., 1997, Aghdam et al., 2019).

Considering these variables conditions, ensuring an efficient landfill gas capture to avoid methane emissions is challenging. The other challenge for the landfill operators is to upgrade landfill gas to biomethane (also referred to as Renewable Natural Gas or RNG), compatible with the local market requirements (grid injection, compressed or Liquefied Natural Gas).

Several technologies exist to upgrade landfill gas to Renewable Natural Gas. Waga Energy, patented technology, called the WAGABOX®, combines several upgrading technologies, based on membrane gas permeation and cryogenic distillation. The gas permeation unit, using hollow fiber membranes, has a large flexibility to remove most of the carbon dioxide. The design of the membrane unit must consider variations in flow and CO_2 concentrations with a dedicated compression unit providing additional flexibility under normal operation conditions to continuously adapt the performances to the specified outlet requirement. Cryogenic distillation is a well-known technology widely used to separate light molecules such as oxygen and nitrogen from methane. The design of this equipment considers the frequent variations in flow and composition guaranteeing achievement the target quality and contributing to improve the overall system performance.

The WAGABOX® unit is designed to accept a wide range of biogas quality, with no constraints on the CH_4, CO_2 and N_2 concentrations. The only limitation is on the O_2 content which must be maintained under 3.5 %, for process safety reasons.

This article analyzes field data from 10 landfill gas WAGABOX® upgrading units and focuses on deviations in the landfill gas composition will occur during the lifetime of the landfill. More than five hundred thousand (500 000) samples have been analyzed for each unit. Data relative to the biomethane production will be presented to highlight the flexibility and the stability of the process and to demonstrate the reliability of the WAGABOX® technology for upgrading current landfill gas into biomethane.

2. MATERIALS AND METHODS

2.1 Sites description

Data analyzed were collected from January to December 2022. The landfills studied are all located across France.

Characteristics of the landfills taken into account in this study are: landfill “size” based on annual tonnage (Table 1), landfill cover composition (Table 2) and rainfall. Each site is described in (Table 3)

Table 1 Landfill size classification (based on annual tons stored)

Size	Annual tonnage
Small	< 90 000 t
Medium	90 000 t < X < 150 000t
Large	>150 000 t

Table 2 Landfill cover composition

Cover	Composition (bottom to top)
Semi-permeable	clay – drainage layer – vegetation
Impermeable	clay – membrane -drainage – vegetation
Mix	Part with semi-permeable and impermeable

Table 3 Landfills characteristics

Site ID	Size	Cover	Rainfall 2022 (mm)
X1	Large	***Impermeable*** and open zones with biogas capture	663
X2	Small	***Mix*** and open zones with biogas capture	771
X3	Medium	***Semi-permeable***	910
X4	Small	***Mix*** and open zones with biogas capture	429
X5	Small	**Mix- cover**	606
X6	Small	***Impermeable*** and open zones with biogas capture	806
X7	Small	***Mix – cover*** and open zones with biogas capture	625
X8	Small	***Impermeable*** and open zones with biogas capture	411
X9	Medium	***Impermeable*** and open zones with biogas capture	611
X10	Large	***Impermeable*** and open zones with biogas capture	468

2.2 Data records

Landfill gas data were obtained from 20 sensors installed on each WAGABOX® units. Data are sampled and recorded every 1-minute over the entire year 2022.

The monitoring of the raw biogas is implemented at the inlet pipe before and after the pretreatment step (mainly desulfurization and drying). The inlet biogas flow “Q” [Nm3/h], the temperature of the landfill gas “T” [°C] and the vacuum applied to the biogas collection system “P” [mbar] are sampled and recorded before pre-treatment equipment.

A fixed infrared analyzer, continuously measures and records the biogas quality including CH_4, CO_2, O_2 and H_2S. The N_2 concentration is obtained by a calculation of the balance component (N_2% = 100 %– [%CH_4] – [%CO_2] – [%O_2]).

The monitoring of biomethane produced is made before the grid station (operated by the natural gas grid operator). Parameters continuously measured are: CH_4[%], CO_2[%], O_2[%] and H_2S [ppm], temperature[°C], pressure[mbar], and flow rate [Nm^3/h]; the Higher Heating Value (HHV) is calculated.

Table 4 sampling points and Measurement the monitoring of the inlet and the outlet flows of the process.

Table 4 sampling points and Measurement associated.

Raw Biogas inlet		**Biomethane production**
Before pre-treatment	After pre-treatment (desulfurization and drying)	Outlet
Flow rate Qbiogas (Nm^3/h) Temperature (°C) Pressure (mbar)	CH_4 [%] CO_2 [%] H_2S (ppm) O_2 [%] N_2 [%]	CH_4 [%] Flow rate Qbiomethane (Nm^3/h) Temperature (°C) Pressure (mbar) HHV (kWh/Nm^3)

For this study, more than 500 thousand samples have been analyzed for each unit.

2.3 Data analytical methods

We focused, in this study, on the variation of parameters measured in the raw biogas (CH_4%, CO_2%, O_2%, calculated N_2% and flow rate).

The data sets have been processed and cleaned using different methods. To deal with missing values, two methods were used: replacing by the previous values and imputation based on linear regression.

Detecting and removing outliers from the raw data signal are crucial steps to prevent signal accuracy from being degraded. An outlier is an unlikely observation which is distinct or does not tally with other available observations in a dataset. Outliers are likely due to data corruption and measurement error. The z-score statistical evaluation was used to identify the outliers in the datasets.

After data processing, statistical analyses including regression analysis and correlation coefficients have been applied to the large pool of data collected in the year 2022 during the operation of the 10 units. The aim is to demonstrate the variation of the biogas inlet profile that the upgrading unit is dealing with and highlight relationships between meteorological parameters and the biogas composition.

Three time-scales were analyzed in order to highlight the ranges of variations and the correlations between parameters :

- Amplitude of variations occurred during a year for each site focused on CH_4, CO_2 O_2, N_2 and flow rate
- Monthly variations focused on the impact of atmospheric pressure on biogas quality.
- Daily variations focused on the impact of both temperature and atmospheric pressure on biogas quality.

3. RESULTS AND DISCUSSION

3.1 Variations per site

This analysis is focused on biogas quality and flow rate collected in the different sites, and the variation associated. The graphical representation used is the box plot.

Box plots provides a quick visual summary of values variability in a dataset. It shows the median, upper and lower quartiles, minimum and maximum values, and any outliers in the dataset (Figure 1.

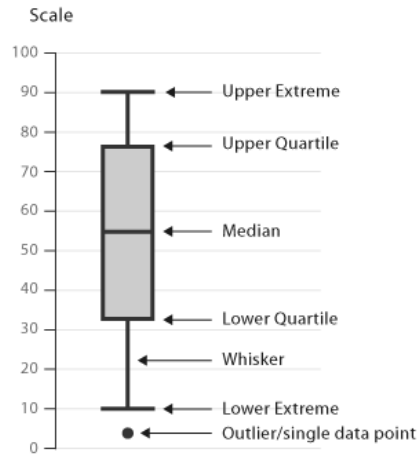

Figure 1 : Box Plot statistical analysis.

The operating range of biogas flow for the 10 studied units in this article is from 200 to 760 Nm3/h of inlet biogas. The operational ranges can be split in three unit sizes:

- 1: 200-400 Nm3/h
- 2: 300-600 Nm3/h
- 3: 460-760 Nm3/h

Larger upgrading units were commissioned in 2022 and are currently under construction (biogas flow rate of around 2000 and up to 3000 Nm3/h).

The analysis implemented in this study concerns the variability of biogas composition, regardless of the flow rate upgraded.

As we can see in Figure 2, the methane content in raw biogas is affected by important variations within each site and between different sites. The range of methane content in raw biogas currently upgraded by the WAGABOX® unit is between 37% and 54.5%. The most common range of operation is between 40% and 47% of CH_4. Regarding CO_2 concentration, the largest range is between 29.5% and 40.5% and the most common range is between 31% and 39% (Figure 3). In relation with the data often shown in the literature, as mentioned before, the CH_4 concentration range observed in this study is Lower. Considering the average range, (40 to 47%), methane concentration is 5 to 8% lower (in comparison with 45 to 55% mentioned in literature).

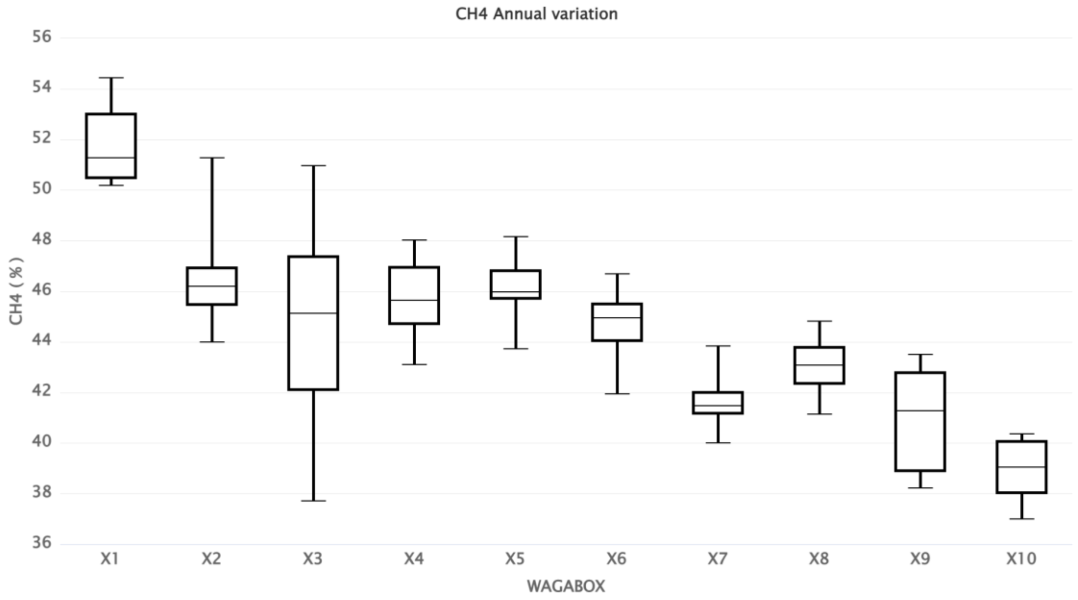

Figure 2 : Annual variation of CH4 concentration in the raw biogas.

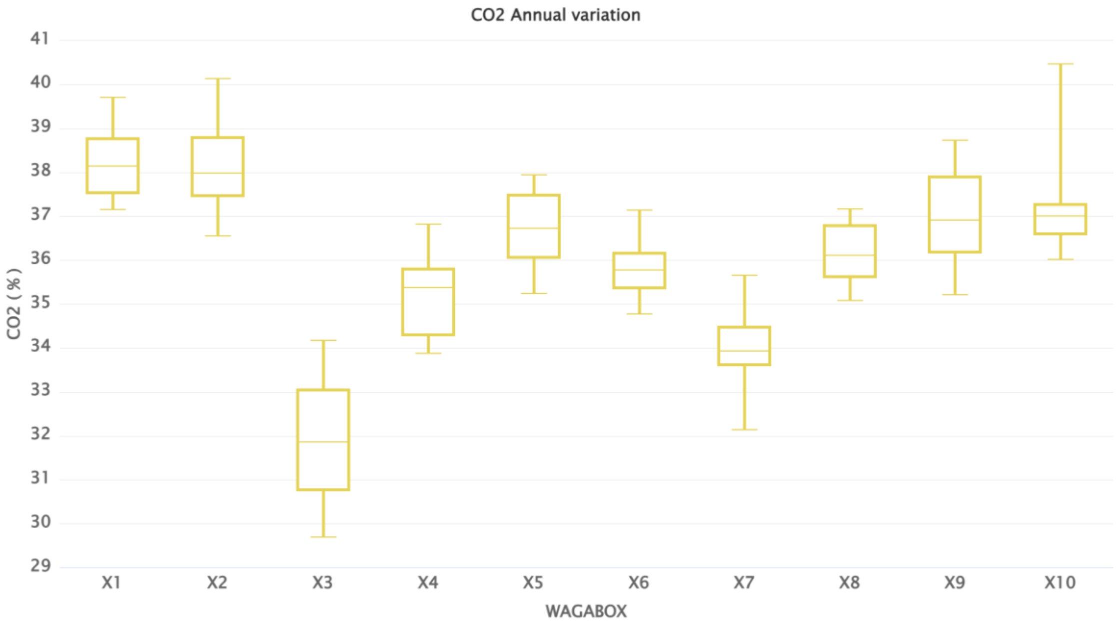

Figure 3 : Annual variation of CO2 concentration in the raw biogas

The range of N_2 content in the raw biogas currently upgraded by the WAGABOX® units is between 2.5% and 29% (Figure 4). The most common range of operation is between 12% and 25% of N_2%. The N_2 concentration in the raw biogas has a negative correlation with methane content, as N_2 is a good indicator of biogas dilution with air. The average concentration commonly used to characterize landfills (5 to 15%) is underestimated.

Figure 5 presents the range of variation of the O_2 concentration in the raw biogas currently upgraded by the WAGABOX® units. Over a concentration of 3.5% O_2 the unit stops for safety reasons, but concentrations profile on the range 0.4 – 2.5% occur.

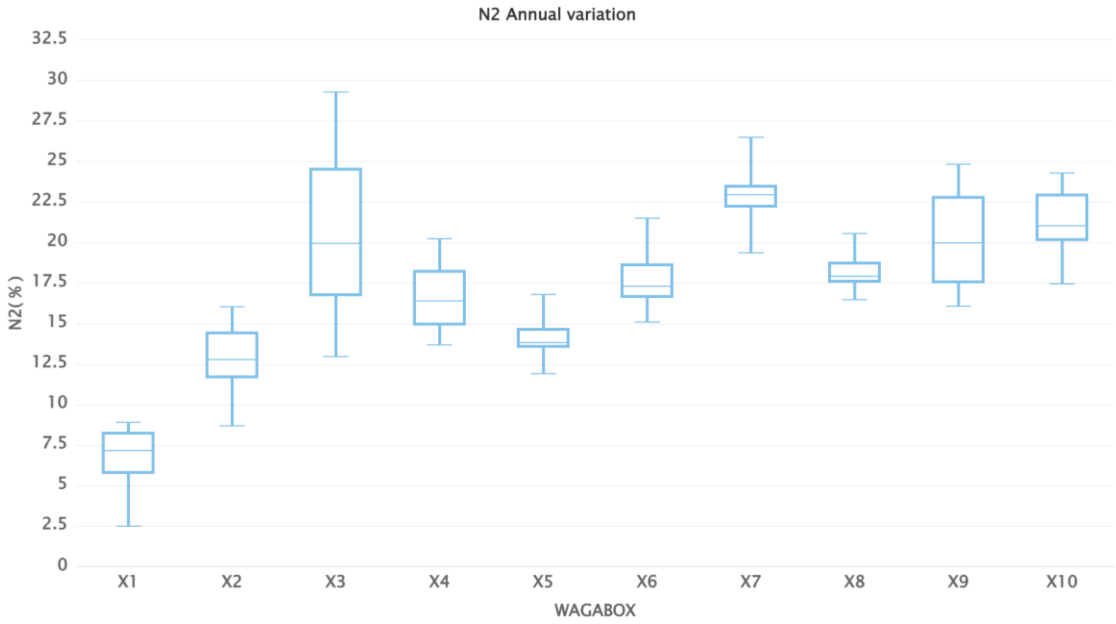

Figure 4 : Annual variation of N2 concentration in raw biogas (N2% calculated = 100 %– [%CH4] – [%CO2] – [%O2]).

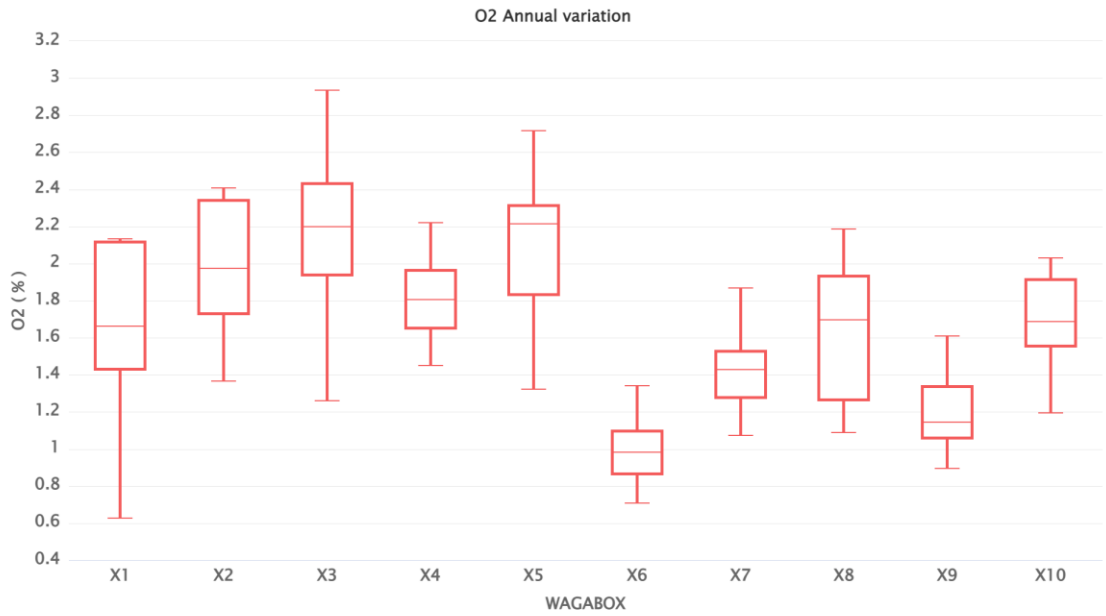

Figure 5 : Annual variation of O2 concentration in raw biogas.

The presence of N2 and O2 in the biogas is expected to be correlated with the impermeability of the landfill and the gas network. The expected effect of reducing air inlets linked to the presence of geomembrane is not obvious from the observation of the data. This particularity is likely due to the impact of capturing biogas on zones under operation (by vertical progressive wells or by horizontal trenches) which significantly increases the presence of air in the mixture with a visible effect of capturing N_2 and consuming O_2 in the body of the waste mass. For this reason, O_2 content is not a good indicator of air inlet and biogas dilution (Figure 5).

The ratio N_2/O_2 has been calculated to highlight different biogas capture strategies (Figure 6): on the left sides (sites X1 to X5), the vacuum applied at the well-heads is globally limited, CH_4 content in the mix flow is higher and N_2 content is generally lower than sites from X6 to X10. On the right side, capture efforts are more important often linked to odors nuisances mitigation strategies: air dilution is higher, and consumption of O_2 by superficial bacteria activity is more important, despite of the presence of geomembrane on closed parts of several of the landfills.

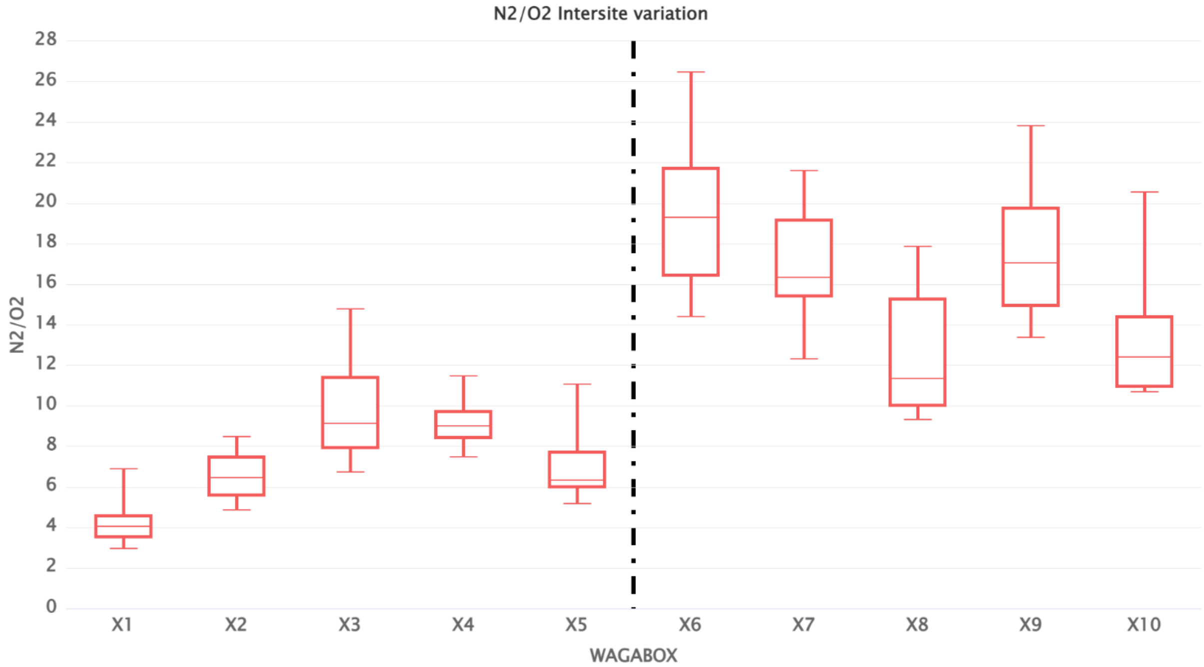

Figure 6 : Annual variation of N2/O2 ratio in raw biogas.

We do not dispose of surface methane emissions measurements, but we can assume that the risk of biogas leaks is higher for sites where the capture effort is light and where methane concentration in biogas wellhead is high.

3.2 Monthly variations

The impact of meteorological factors on the biogas composition was studied over a shorter time frame than the year to clearly identify and isolate the main influencing factors affecting the biogas capture and composition. Moreover, over a short period, external factors such as gas collection system expansion, maintenance activities or variations in the blower vacuum, can be avoided.

The influence of meteorological factors on bigas quality and on collected flow rate is known and documented (Christophersen et al. , 2001; Uyanik et al., 2012; Borjesson et al.,1997, Aghdam et al., 2019). The objective of this analysis, based on a large quantity of data, is to highlight the amplitude of variation which can occur over relatively short periods of time, and which requires significant adaptability of the landfill gas upgrading technology.

In Figure 7, specular variation of CH_4% and N_2% is mainly produced by pressure fluctuations. the capture of biogas is slowed down by the high pressure (drop in CH_4 content) and the entry of air is favored (rise in N_2 content).

N_2 content rises to 19.3% and CH4 content varies between of + and - 10% over a 24 hours period.

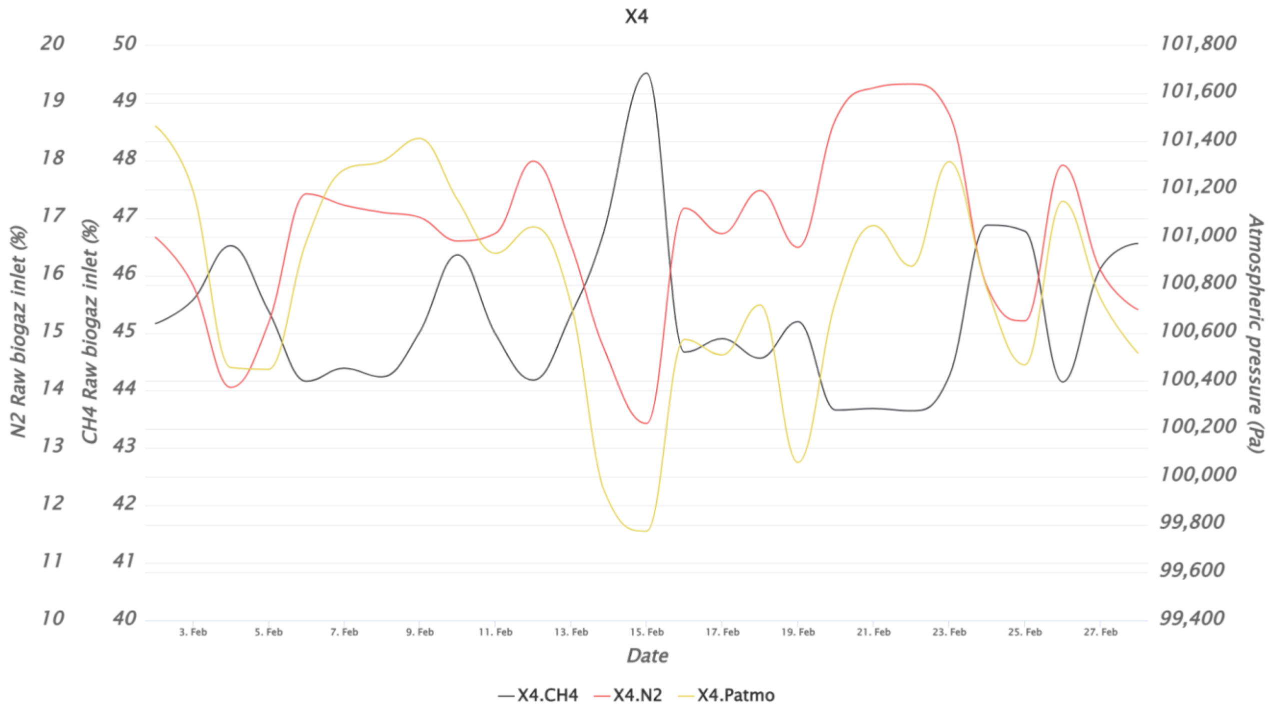

Figure 7 : Fluctuation of X4 biogas composition under pressure variation.

3.3 Daily variations

Daily cycles are strongly marked by variations in temperature and pressure. The impact of rainfall is complex to study because of its association with variations of two other two parameters. In the short term, the saturation of the covers (soil and clay) with water from rainfall increases impermeability and reduces air inlet. The effect is therefore cumulative with the reduction in pressure and the lowering of temperature generally linked to rainy episodes.

In Figure 8, CH_4 and N_2 (main indicator of air dilution) fluctuations are studied in relation to pressure and temperature variations.

Temperature have the main evident impact on the biogas composition : more biogas collection and less air dilution during the day (higher temperature period); the opposite being observed at night.

Pressure variation has a reinforcing effect: for equivalent temperature peaks, high pressure reduces the CH_4 content and amplifies the N_2 content.

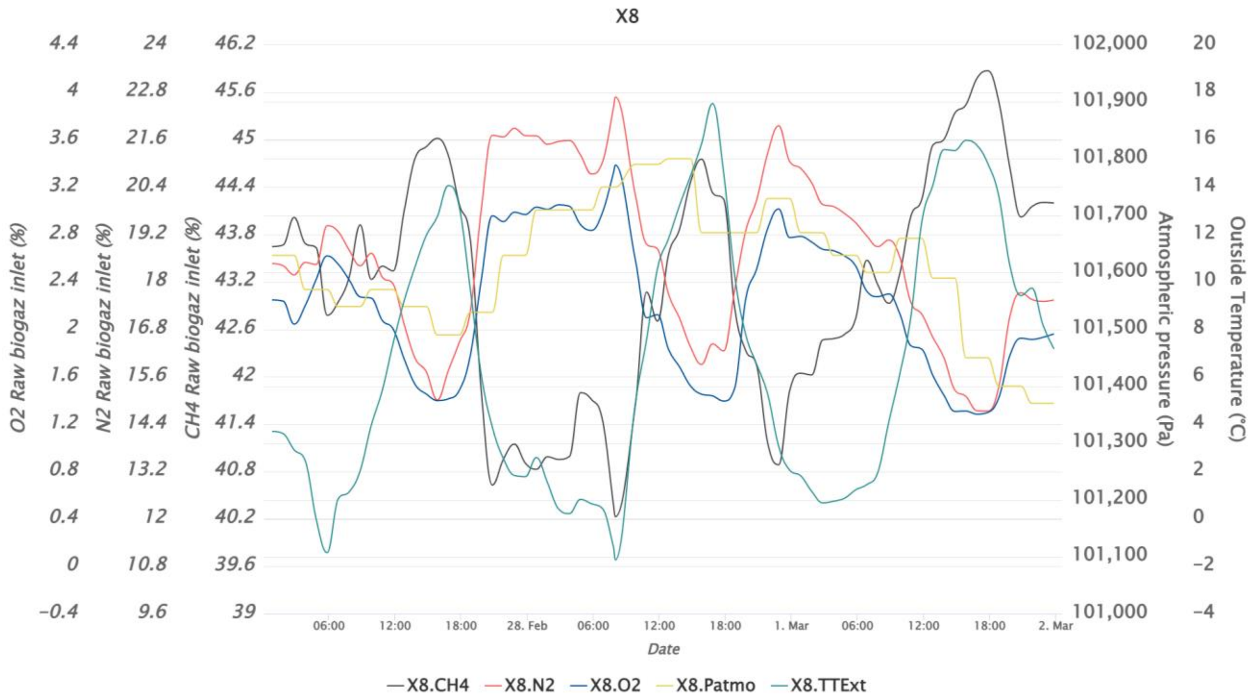

Figure 8 : Fluctuation of X8 biogas composition under temperature and pressure variation on a short period (3 days).

The effect of meteorological factors on the dilution of biogas by air is highlighted in Figure 9. In conditions of active biogas capture, some air is captured by the vacuum applied to the biogas collection system and travels through the cover and top waste layers. O_2 is consumed by microbial activity in these superficial layers. Higher temperatures generally lead to increase microbial activity and O_2 consumption. The ratio between N_2 and O_2 is strongly affected by this process : the N_2 concentration in biogas comes mainly form air and is not impacted by biological activity; oxygen is partially consumed by micobial activity. The ratio N_2/O_2 is thus strongly correated to temperature fluctuations.

In the UK Industry code of practise for biogas management (Brindley T., 2012), the "free nitrogen" is defined as the indicator of "how much air is being drawn directly into the waste mass, as it measures the level of oxygen being consumed by calculating the nitrogen left behind". The free nitrogen content considered appropriate for an optimized biogas capture is between 7.5% and 20%.

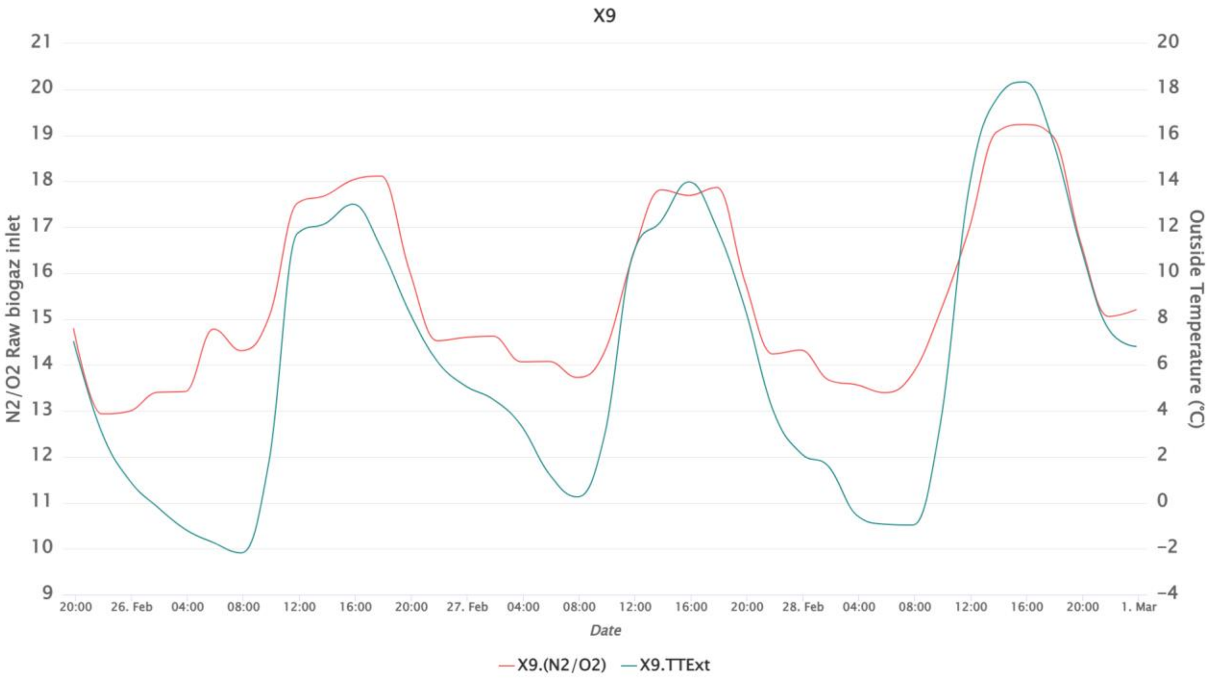

Figure 9 : variation of the ratio N_2/O_2 with temperature

As this data highlights, biogas capture is strongly affected by meteorological factors. Cyclical and non-cyclical weather factors affect air entry and, conversely, methane leakage. Maintaining active and efficient vacuum at all well heads of the landfill gas collection system, with a light but permanent air dilution, makes it possible to best control biogas leaks in areas of the landfill farthest from the vacuum points (limits of cells or portions between two wells).

3.1 Upgrading performance

Despite the great variability in the composition of biogas measured on the 10 sites analyzed, the mean uptime of the upgrading units is higher than 95% for the year 2022. For 8 units the uptime is higher than 95%, for 2 units it is about 93% because of unusual equipment failures. Relative to the energy recovery efficiency, it exceeds 87% on all the WAGABOX® and it reaches 90% on half of the units (Figure 10).

It is important to emphasize that the N_2 concentration can reach 29% (e.g. Site X3) without significantly affecting the operation of the recovery unit.

The quality of the biomethane produced and injected has a stable composition with a CH_4 content well above the threshold accepted by the natural gas pipeline in France (in Figure 11).

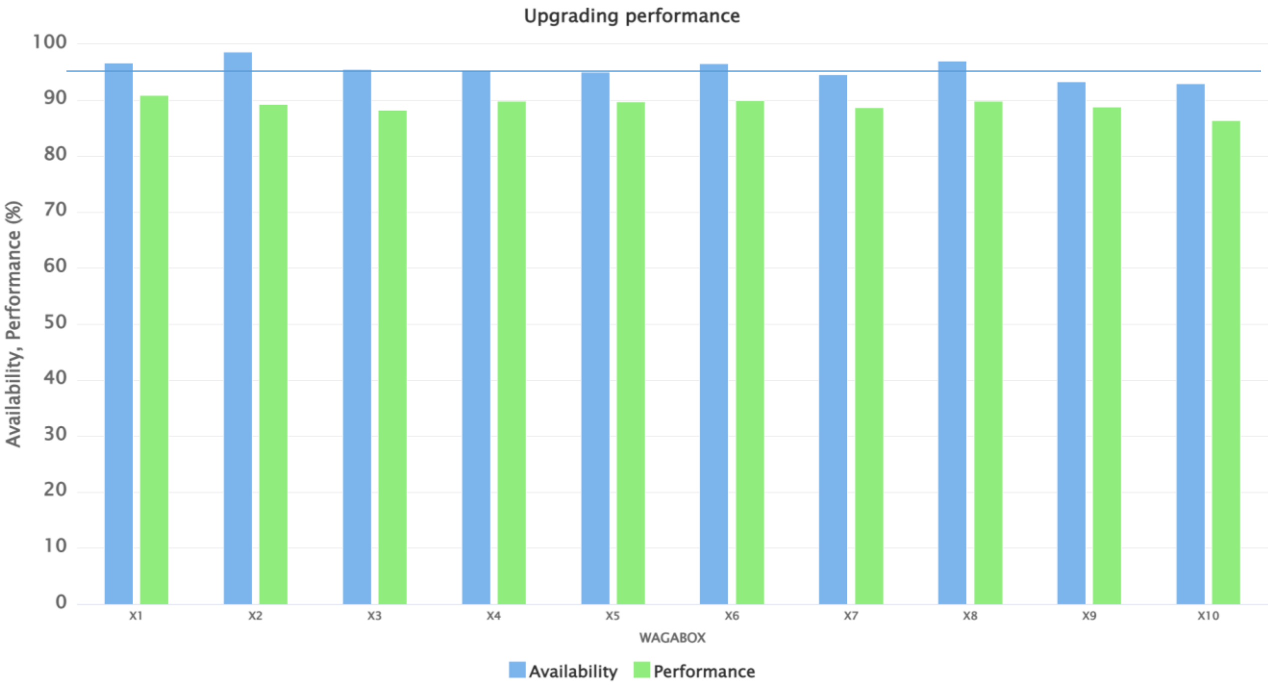

Figure 10 : Annual uptime and performance (CH4 recovery rate) of 10 WAGABOX®

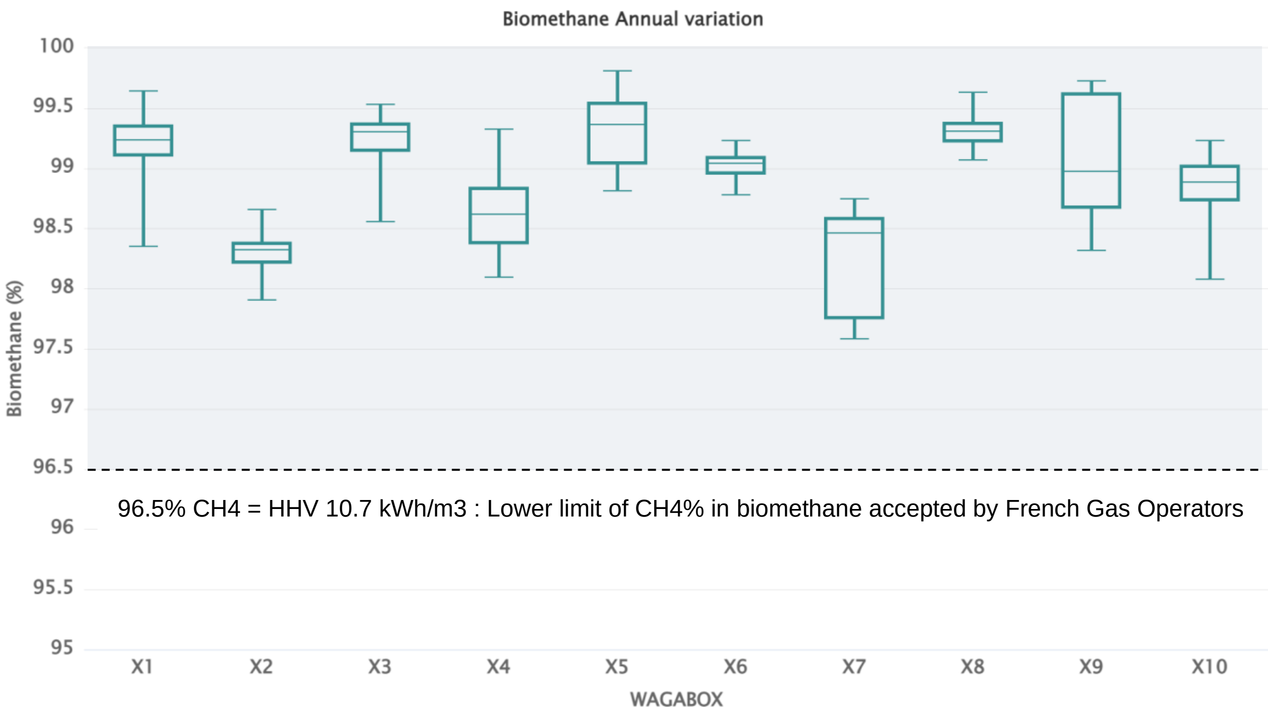

Figure 11 : variation of the biomethane quality over 1 year of operation of 10 WAGABOX®

4. CONCLUSIONS

Data collected from the operation of 10 upgrading unit (WAGABOX®) on a 1 min basis over 1 year of operation have been processed to observe the variation range of inlet biogas composition. The statistical analysis showed that the variation range is from 37% and 54.5% for CH_4 and from 2.5% and 29% for N_2, showing that the biogas concentration profiles are larger than commonly assumed.

Waste composition, landfill characteristics and operation have an impact on biogas collection, and meteorological aspects have a major influence on landfill biogas parameters. Temperature, precipitation, and atmospheric pressure variations are correlated and influence the biogas production, capture and composition. Understanding these relationships is crucial for optimizing gas collection systems, minimizing methane emissions, and ensuring landfill sustainability.

The analysis of data presented demonstrates that WAGABOX® units ensure the upgrading of landfill biogas irrespective of its great hourly, daily and seasonal variability. The biomethane produced has a stable composition and meets the specifications defined for injection into the natural gas distribution or transport grid. The uptime and performance of the units are not affected either by the variability of the composition of the biogas nor by the nitrogen content which can exceed 25%.

The next step to further improve the performance of the units is to implement advanced modeling techniques to predict biogas production based on meteorological forecasts and develop adaptive gas upgrading and biogas regulation strategies.

The ultimate objective is to maximize the capture of biogas to reduce greenhouse gases and to maximize the production of the biomethane, a local renewable and clean energy.

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