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Protection of the Purification Station Thermal Engines Against the Effects of Hydrogen Sulfide by the Coupling of a Chemical and Biological Treatment of the Produced Biogas

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ABSTRACT

Among the various techniques used to reduce hydrogene sulfide in biogaz avoiding harmful effects on engines, the chemical and biological treatment appears particularly promising. The main objective of this article was to develop a new process to reduce the harmful effect of hydrogen sulfide (H_2S), contained in the biogas resulting from methanization, on the equipment of the wastewater treatment plant (WWTP) of FES city in particular the two cogeneration units. A multiple regime technique for biogas desulfuration, based on the chemical and biological treatment as well as internal micro-aeration of the digester was developed. Owing to the insights gained from this study, it was identified that reducing the concentration of H_2S in biogas and improving methane production (biogas production increased from 3.6 M Nm^3 in 2018 to 3.8 M Nm^3 in 2019, a saving of about 300,000 MAD); reduction of desulfurization tower downtime from 4 times/year to 1 time/year; increasing operating time of generating sets from 8800 in 2018 to 14 400 h in 2019; electricity production increased from 5.9 GWh in 2018 to 7.2 GWh in 2019.

In light of these findings, it can be affirmed that the study successfully achieved its objectives, presenting valuable avenues for future scientific exploration

Keywords: water treatment plant, anaerobic digestion (AD), biogas treatment, biological desulphurization, micro-aeration and thermal engines.

INTRODUCTION

Today, renewable energies represent a primary interest in the face of alarming energy consumption and environmental risks. Among the production techniques of these renewable energies, one can cite anaerobic digestion (AD), or methanization, which has many environmental, economic and social interests. Anaerobic digestion is a microbiological technique for converting organic matter into biogas in an oxygen-depleted environment, mainly in the presence of anaerobic microorganisms. This biogas can be recovered in the form of heat or electricity (Szczyrba et al. 2020). The positive environmental impacts of anaerobic digestion, such as the reduction of greenhouse gas emissions and odors, are widely used as strengths for the promotion of this process. Indeed, the biogas from AD is a mixture of methane (50–70%), CO_2 (30–50%), H_2 (1–5%), H_2S (0.1–3%), N_2 (2–7%) and sometimes NH_3 (Kapoor et al. 2019). These percentages depend essentially on the quality of the substrate and on several parameters of the operation digester (Id 2015). The upgrading of biogas CH_4 biogas in the production of electricity and heat remains limited due to the presence of pollutants, especially H_2S, which has harmful effects on equipment and heat engines (Okoro et al.2019). Anaerobic digestion or methanization is a microbial fermentation based on the degradation of organic matter in a reactor called a digester, in absence of oxygen and under fairly specific conditions (Quality of the substrate, T °, pH, length of stay, etc.) (Hajjaji et al. 2016). This microbial degradation releases a biogas composed mainly of methane (CH_4) and carbon dioxide CO_2 (Figure 1). It also contains compounds, such as hydrogen sulfide (H_2S), ammoniac (NH_3) and other volatile organic compounds at low concentrations (Kapoor et al. 2019).

This reaction process is classically divided into four biochemical steps (Ghouali et al. 2015, Meres et al. 2009):

- hydrolysis: organic macromolecules (polysaccharides, proteins, lipid compounds) are hydrolyzed into simpler elements, such as simple sugars, amino acids, short-chain fatty acids, glycerol;
- acidogenesis: the substrates resulting from the hydrolysis phase are transformed by bacteria into volatile fatty acids, alcohols, ammoniac, carbon dioxide and hydrogen;
- acetogenesis: volatile fatty acids (other than acetic acid) are converted by acetogenic reducing bacteria into acetate, hydrogen and carbon dioxide;
- methanogenesis: the substrates resulting from the hydrolysis phase are reduced by bacteria to methane and carbon dioxide.

At the end of these successive reactions, biogas (composed of CH_4, CO_2, H_2S, etc.) as well as a stabilized and hygienized digestion residue, called digestate (sludge), are formed. H_2S is the result of the fermentation of sulfur compounds present in many organic materials (Aziza et al. 2012. It is produced by two types of sulfur-reducing bacteria (SRB). The latter use either acetic acid or propionic acid as organic substrate to reduce sulfates (SO_4^{2-}) to sulfides (H_2S and HS^-). The chemical reactions involved are represented by Equations 1 and 2, (Boivin et al. 2010):

$$H_2S0_4 + CH_3COOH\ H_2S + 2\ H_2CO_3 \quad (1)$$

$$H_2S0_4 + 4/7CH_3CH_2COOH\ H_2S + 12/7\ H_2CO_3 \quad (2)$$

From the point of view of the digester feed, the oxidized forms of sulfur are found in:

- minerals in the form of sulfates (for example : in many vinasses resulting from the regeneration of ion exchangers with sulfuric acid);
- organic matter: like proteins.

H_2S causes several problems when it is present in the biogas recovery circuit resulting from anaerobic digestion (Ramos et al. 2104). As expressed by equations 3 and 4, the combustion of H_2S produces sulfur dioxide (SO_2) which itself, when oxidized, produces sulfur trioxide (SO_3). The latter compound reacts chemically with water to form sulfuric acid, a strong acid which gives a corrosive potential to combustion biogas (Eq. 5) (Boivin et al. 2010):

$$H_2S + 3/2\ O_2 \rightarrow SO_2 + H_2O + 518\ kJ/mole \quad (3)$$

$$SO_2 + ½\ O_2 \rightarrow SO_3 + 99\ kJ/mole \quad (4)$$

$$SO_3 + H_2S \rightarrow H_2SO_4\ (gas) + 101\ kJ/mole \quad (5)$$

The presence of H_2S and water vapor in the biogas causes premature wear of combustion equipment and also alters the metal structures that surround the anaerobic digestion system. In the case of an internal combustion engine (Cogeneration), the

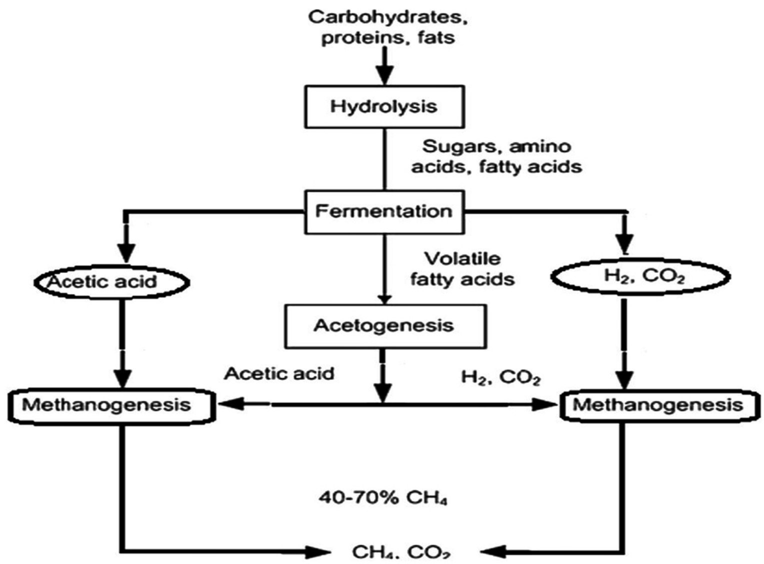

Figure 1. Flow diagram of the AD process

H_2S reacting with the water will attack the pistons and cylinders. In fact, the H_2S concentration should not exceed 500 ppm to limit damage to internal parts of the equipment (Zhao et al. 2010). Table 1 summarizes the consequences of H_2S contained in the biogas produced in the wastewater treatment plant.

In this context, and faced with all these effects of the H_2S component mentioned previously, and for a better valorization of CH_4 biogas, the authors worked on an important study for the treatment of H_2S, based on the combination between the biological method, the chemical method and a slight micro-aeration of the digester within the wastewater treatment plant in the city of Fez. The main objective of the study was to improve the quality and production rate of biogas, reduce the corrosive impact of H_2S on the equipment of the wastewater treatment plant, and optimize the operating life of the cogenerators and therefore the increase in the clean quantity of electrical energy produced.

MATERIAL AND METHODS

Biological treatment of H_2S in the WWTP-FES

For the treatment and valorization of biogas resulting from anaerobic digestion, within Fez city plant wastewater treatment (Morocco), the biological method (also called: Desulfurization of biogas) is used, which is carried out by biological washing on a biological purifier (Figure 2). This solution makes it possible to lower the H_2S content without producing polluting by-products, as a result, the maintenance frequency of the desulfurization tower will be considerably reduced, which will contribute to improving the efficiency of the station.

This desulfurization of the biogas is done by biological washing in a counter-current contact tower. The biogas flow is ascending, the wash water flow (contains nutrients for the bacteria) to a descending flow. This solution reduces the H_2S content without producing polluting by-products. The biological elimination of H_2S is carried out in the presence of microorganisms and traces of oxygen (at < 25% of the allowed limit concentration) to oxidize the H_2S into sulfate. The final purge, which has a sulfate concentration of approximately 5 to 8% and a pH of around 1.5, will be pumped to the upstream side of the WWTP pretreatment. To maintain sufficient microbial activity, the temperature of the unit is maintained at 25°C using hot water from the cogeneration plant. Basic data of the desulphurization tower:

Table 1. The different harmful effects of H_2S (Zhao et al. 2010)

Effects of H_2S	Consequences
On human health	* 0.02-0.13 ppm H_2S: olfactory perception. * 250-500 ppm H_2S: headache, cyanosis, pulmonary edema. *>1000 ppm H_2S : Apnea, nervous system paralysis and death within minutes
On CH_4 biogas production	The presence of oxidized sulfur (SO_4^{2-}) consumes acetic acid and hydrogen and reduces the production of CH_4, according to the following reaction: $CH_3\text{-}COOH + SO_4^{2-} \rightarrow 2CO_2 + H_2S$
On cogeneration engines during combustion	* Sulphates (HSO^{4-}) form sulfuric acid which attacks aluminum alloy pistons and eventually pierces them. * The sulfur precipitates on the valves, decreases the compression and can eventually break the valve heads.

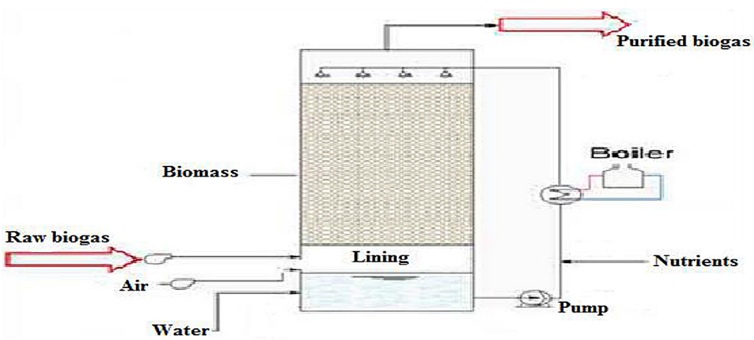

Figure 2. Synoptic diagram of the desulfurization tower (SCADA-STEP FES software)

- Raw biogas throughput to be treated: 1,440 Nm³/hour
- H_2S in content: Varies between 3,000 ppm and 7,000 ppm.
- H_2S out content: < 500 ppm
- Average removal capacity is: 6.5 kg H_2S/h.

However, it was necessary to ensure rigorous monitoring of certain parameters in order to guarantee the reliability of the study for improving the operation of the desulfurization tower and the micro-aeration of the digesters (Awe et al. 2017). Table 2 shows the monitoring schedule for the necessary parameters of the digester and the desulfurization tower operations.

Problem encountered at the desulfurization tower

The desulphurization tower, must reduce the concentration of H_2S from 4,500 ppmv (average value, in 2018) to less than 500 ppmv (recommended value) without producing dangerous by-products but, the major disadvantage of this method is the appearance of sulfur (S) according to reaction (Equation 6), which is a solid compound and causes a rapid clogging of the tower (Figure 3).

$$H_2S + O_2 \rightarrow 2S + 2H_2O \quad (6)$$

The cumulative effects of the clogging of the tower cause an internal increase in the pressure drops between the inlet and the outlet of the biogas, and significantly reduces the efficiency of H_2S removal. This makes the biogas more corrosive to the metal parts of the installation. Regulators, gas meters, valves and supports can quickly corrode. The combustion of biogas containing H_2S produces sulfur dioxide (SO_2). When SO_2 combines with water vapour, it produces sulfuric acid which corrodes the exhaust pipes of engines, etc. Gaseous SO_2 also dissolves in engine oil, causing the oil to become acidic and lose its ability to lubricate, damaging the engine and shortening the time between oil changes (Shab et al. 2013).

Table 2. Planning for the required parameters of the digester and the desulfurization tower operations

Parameters	Digester		Desulphurization tower	
	Entrance	Exit	Entrance	Exit
Biogaz flow	Once a day	Once a day	Once an hour	Once an hour
pH	Once a day	Once a day		
temperature	Once a day			
Chemical oxygen demand COD_{total} (Sludge)	Once a day	Once a day		
$COD_{filtrée / after centrifugation}$ (Sludge)	Once a day	Twice a week		
Volatile fatty acids VFA (Sludge)	Once a day	Once a day		
Suspended matter (SM) / volatile Suspended matter (VSM) (sludge)	Once a day	Once a day		
Total nitrogen TN + NH_4^+-N (sludge)		Once a week		
total phosphorus + PO_4^{3-} P (sludge)		Once a week		
CH_4 et H_2S	once an hour	once an hour	Once an hour	Once an hour
Dwell time (sludge)	21 days minimum			

Figure 3. State of desulfurization tower clogging (WWTP Fez : 15-01-2018)

Corrective actions:

1. QSR (Quick Sludge Removal) operations that require between 4 and 8 hours of work: removes the sulfur deposit from the desulfurization tower by filling it with water and injecting compressed air.
2. Complete washing of the tower, which takes between 4 and 7 days.

For the year 2018, the periodicity of carrying out these two operations according to the value of ΔP, which must always be less than 5 mbar, is shown in Figure 4. After the analysis of Figure 4 it was noticed that the number of QSR operations and complete scrubbing of the desulphurization tower is still quite high, generating additional operating costs. In 2018, the following were carried out:

- Three (03) QSR operations.
- Two (02) complete washing operations of the tower.

Moreover, these operations cause a total stop of the desulfurization tower, which directly influences the purification of the biogas produced and consequently:

- A stoppage of both cogeneration units due to the unpurified biogas: Hydrogen sulfide H_2S in combination with water vapor in the raw biogas form sulfuric acid (H_2SO_4), which is highly corrosive to the cogeneration engine components.
- Very low self-production of electricity (cogeneration of stored biogas).
- Increase in invoiced electricity consumption (Table 3).

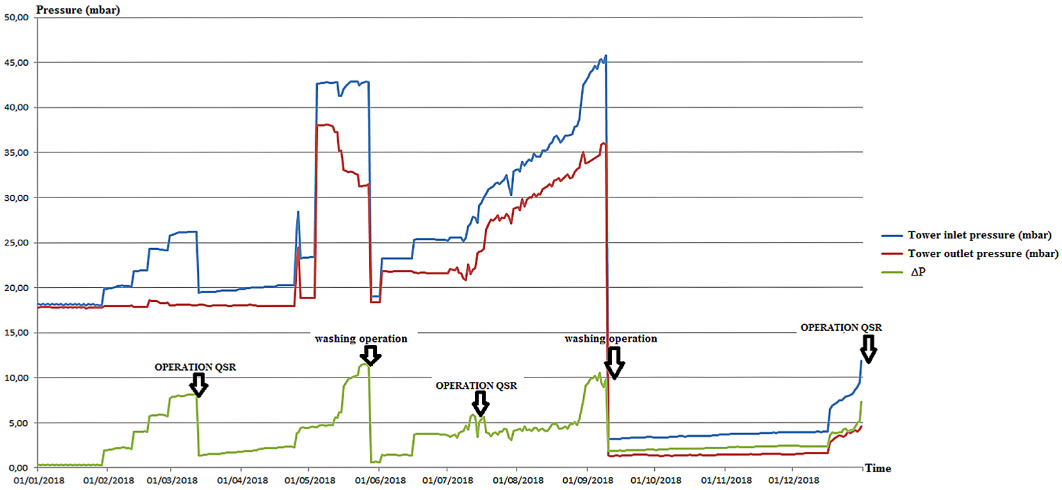

Figure 4. Periods of QSR operations and washing of the desulfurization tower

Table 3. Monthly electricity consumption of the WWTP for the year 2018

Month	Flow rate of biogas produced (m³)	Quantity of electricity invoiced (KWh)	Quantity of electricity cogenerated (KWh)	Cogeneration efficiency (%)	Total energy consumption of the WWTP (KWh)
January-18	29 836	330 225	0	0	330 225
February-18	81 559	352 276	0	0	352 276
March-18	99 292	414 372	51 416	11	465 788
April-18	119 203	295 769	388 931	57	684 700
May-18	206 088	184 982	565 784	75	750 766
June-18	290 618	504 902	314 723	38	819 625
july-18	562 105	607 215	923 541	60	1 530 756
August-18	550 102	221 520	1 033 751	82	1 255 271
September-18	534 672	699 160	527 659	43	1 226 819
October-18	471 860	302 520	886 233	75	1 188 753
November -18	365 849	263 760	760 222	74	1 023 982
December -18	282 287	499 472	412 393	45	911 865
Total	3 593 471	4 676 173	5 864 653		10 540 826

After analyzing the results, it was found that:
- Before each QSR (Figure 5), the self-production of electrical energy decreases although the electrical consumption billed increases, this is because of the reduction in the hourly rate of the operation of the two cogeneration units due to the reduction in the volume of biogas purified (shutdown of the desulfurization tower due to the increase in ΔP.
- After each QSR or washing operation, the desulfurization tower resumes his normal operation and the biogas can be purified and cogenerated.

The biological treatment of biogas from anaerobic digestion by a desulfurization tower remains a beneficial solution from an economic and environmental point of view, owing to the limited use of energy and chemicals. However, the major disadvantage of this process is the internal clogging of the tower due to the deposition of sulfur S (Eq. 6) and consequently an increase in the internal pressure at this tower, which has a negative influence on the purification efficiency of the biogas sent to cogeneration units for the production of electrical energy. A biogas that is not sufficiently purified can contain traces of H_2S which causes the degradation of the thermal motors of cogeneration units by two mechanisms: sulfurization where the sulfur attacks directly the metal components of the engine and corrosion where sulfuric acid formed with condensation water erodes metal surfaces (Maizonnasse et al. 2013).

Faced with this repetitive situation and following the technical and financial requirements, a technical design study for the desulphurization unit was established by combining the biological treatment of H_2S with a chemical treatment (chemical oxidation + a slight microaeration of the digesters) by injection external from oxygen to biogas upstream of the tower (Maizonnasse et. 2013).

Chemical oxidation study

The process for injecting oxygen upstream of the desulfurization tower is simple to set up and requires a small initial investment. It consists of the installation of an air injection pump coupled to a probe for measuring oxygen and biogas flow. The automatic regulation of oxygen flow prevents the creation of an explosive mixture (Diaz et al. 2015). Thereby, this process ensures at the same time, a microaeration of the digesters which will improve the production of CH_4 and reduce the appearance of H_2S (Polano et al. 2009).

Adopted solution

The proposed solution based on the injection of regulated air upstream of the desulfurization tower (Figure 6) allowed:

a) chemically oxidizing the H_2S to S sulfur. This solid element will be deposited on the biogas pipe upstream of the biological H_2S desulphurization tower (Figure 7), avoiding its frequent clogging and consequently the number of annual shutdowns of this tower will be decreased.

b) creating microaeration digester to improve the quality of biogas and reduce the production of H_2S is based on the biochemical oxidation of the sulfide to elemental sulfur (S0) or/and sulfate (SO_4^{2-}) (Dannesboe et al. 2019).

RESULTS AND DISCUSSION

After carrying out several oxygen injection tests upstream of the desulfurization tower, it was

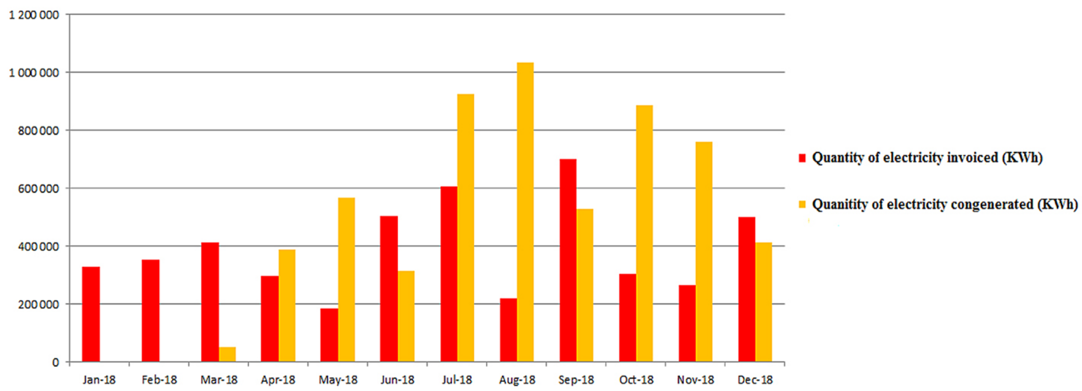

Figure 5. Monthly variations on the electricity consumption of the WWTP in 2018

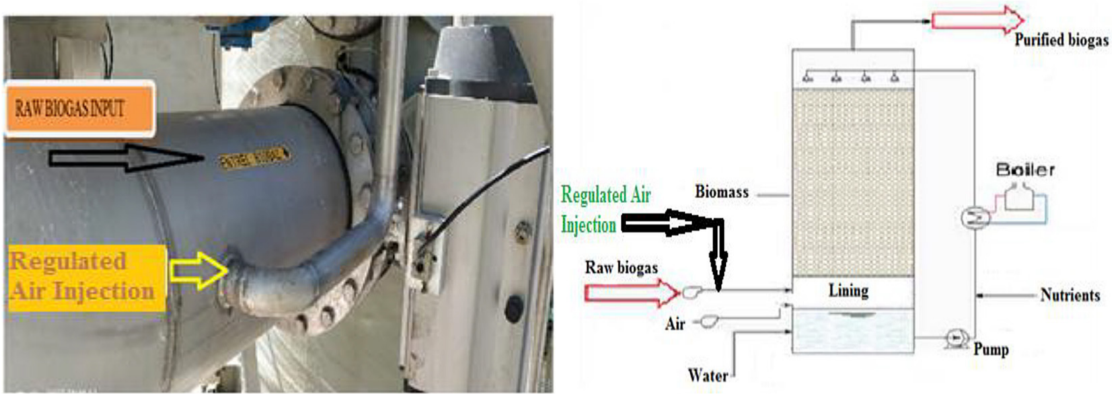

Figure 6. Regulated air injection point upstream of the desulfurization tower

Figure 7. Sulfur deposit in the Tower entry pipe

found that the optimal volume of oxygen injected is 1/20 of the volume of biogas produced, this allowed considerably improving the biological desulfurization of biogas within the Fez wastewater treatment plant (Figure 8). It enabled to chemically oxidize H_2S to sulfur (S), ensure microaeration of the digesters and significantly reduce the concentration of H_2S in the biogas mixture. On the one hand, according to the reaction : $H_2S + O_2 \rightarrow 2S + 2\ H_2O$, Microorganisms oxidize hydrogen sulfide with oxygen molecules and convert the rest of the elements into elemental sulfur and water, this solid compound (sulfur) will be deposited on the pipe before entering the biological desulfurization tower. This deposit allowed significantly reducing the rate of clogging and the variation of the internal pressure of the tower ($\Delta P < 7$ mbar), and consequently reducing the number as well as the time of the annual operations of the QSR and the washing (Figure 9). On the other hand, microaeration (Jenicek et al. 2010) allowed heterotrophic bacteria to inhibit SRB and minimize the appearance of hydrogen sulfide in the gas mixture inside the digester. The Figure 10 clearly shows this result and explains the reduction in the concentration of H_2S after the injection of air into the digester. (Jenicek et al. 2017).

The analysis of Figure 10 shows that the presence of oxygen in the digesters (by microaeration) oxidizes the sulfide present in the mixture, into

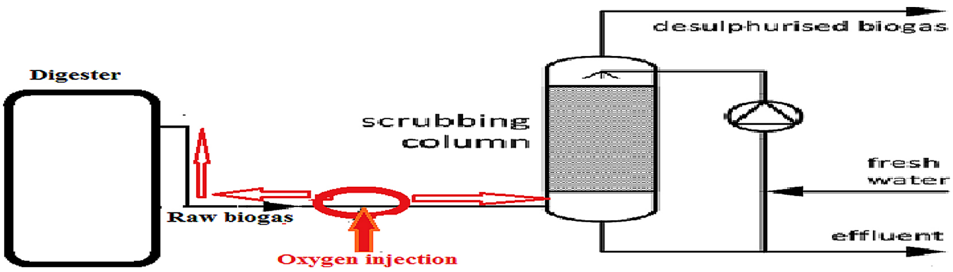

Figure 8. Synoptic diagram of oxygen injection upstream of the desulfurization tower

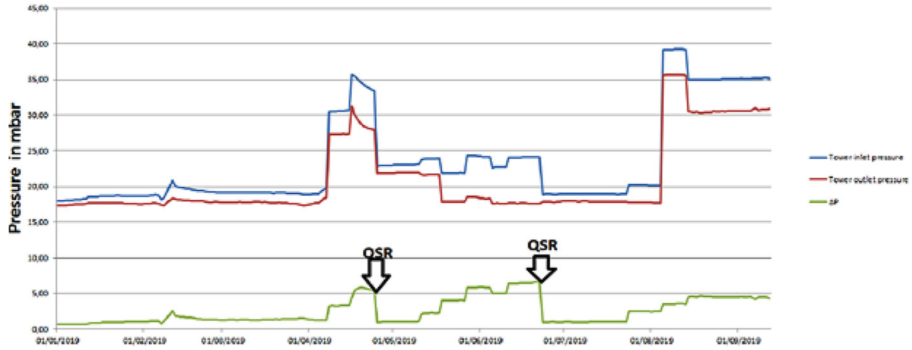

Figure 9. Periods of QSR operations and washing of the desulfurization tower (after the air injection study)

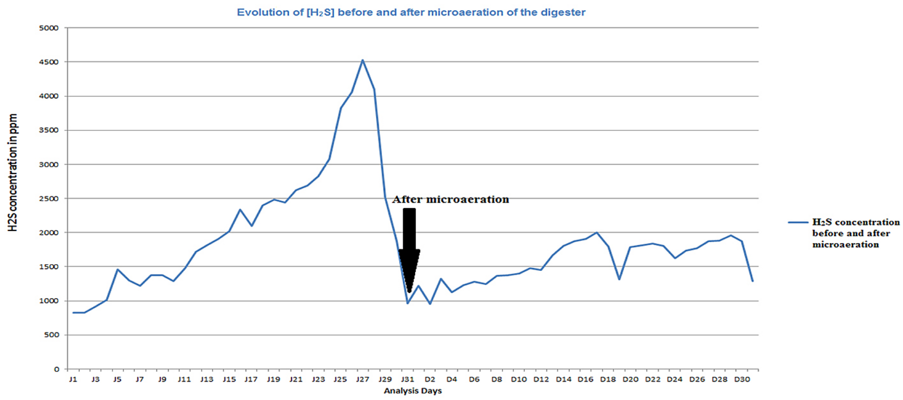

Figure 10. Evolution of [H_2S] before and after microaeration of the digester

elemental sulfur which will be precipitated and removed with the sludge, which means the fall of the concentration of H_2S after the technique of microaeration adopted in the study to improve the operation of the desulfurization tower. It is also noted that the microaeration improves the degradability of the COD and volatile suspended elements and enriches the quality of the biogas produced (Botheju et al. 2010). According to Table 4 and Figure 11, it can be said that the annual monitoring of the energy consumption of the WWTP, after the study of the new design of the desulfurization tower, showed a very satisfactory energy balance, because the availability of biogas streamlined allowed:

Table 4. Monthly monitoring of the electricity consumption WWTP electricity consumption for the year 2019

Month	Flow rate of biogas produced (m³)	Quantity of electricity invoiced (KWh)	Quantity of electricity cogenerated (KWh)	Cogeneration efficiency (%)	Total energy consumption of the WWTP (KWh)
Jan-19	274 053	111 080	428 180	79	539 260
Feb-19	275 503	120 000	565 676	82	685 676
Mar-19	432 987	136 440	958 795	88	1 095 235
April-19	391 868	253 725	826 601	77	1 080 326
May-19	299 758	712 840	526 978	43	1 239 818
June-19	268 015	673 701	687 293	50	1 360 994
July-19	423 988	563 982	498 957	47	1 062 939
August-19	387 538	264 040	653 413	71	917 453
Sept-19	482 823	330 280	887 254	73	1 217 534
Oct-19	477 125	388 320	907 916	70	1 296 236
Nov-19	404 325	187 360	296 781	61	484 141
Dec-19	389 541	166 480	0	0	166 480
Total	4 507 524	3 908 248	7 237 844		11 146 092

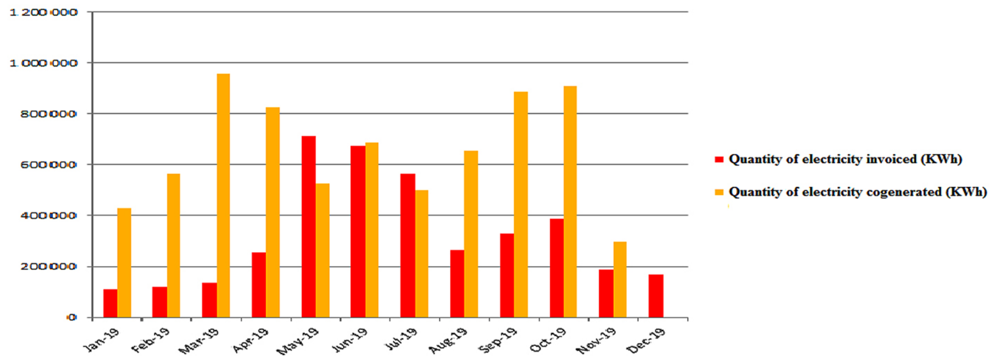

Figure 11. Graphic representation of the monthly monitoring of the electricity consumption of the WWTP for the year 2019

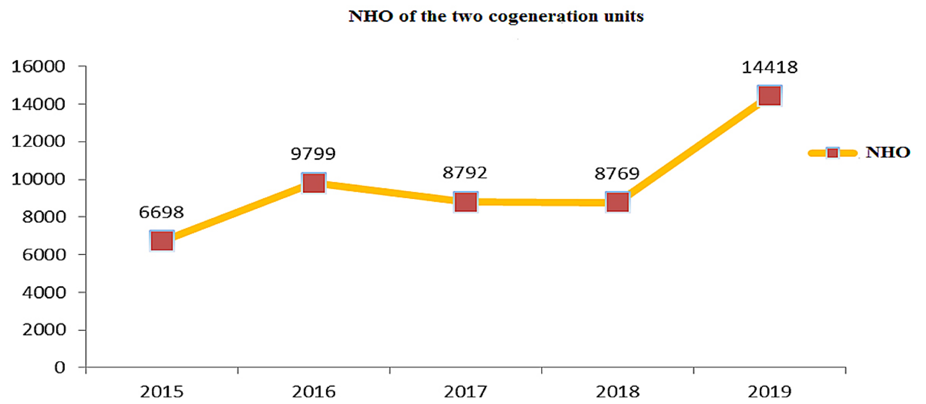

Figure 12. Annual monitoring of the number of hours of operation (NHO) of the 2 cogeneration units

- the recovery of a large quantity of CH_4 biogas;
- the cogeneration of a large amount of electrical energy;
- minimization of the amount of electricity purchased;
- The reduction in the rate and frequency of maintenance of biogas circuit equipment and especially the heat engines of cogeneration units. This has a positive influence on the number of hours of operation of these two cogeneration units, because it was possible to go from 8,763 hours of operation in 2018 to 14,418 hours of operation in 2019 (Fig. 12).

CONCLUSIONS

The combination of hydrogen sulfide oxidation upstream of the desulfurization tower with digester micro-aeration showed the following results: protection of cogeneration units and heat engines against the harmful effects of H_2S; reduction of the clogging rate of the desulfurization tower; improvement of methane CH_4 production; increase of electricity production; increase of equipment service life.

Moreover, the study of the oxidation of hydrogen sulfide upstream of the desulfurization tower with microaeration of the digesters has enabled to obtain promising results, especially in terms of: flow of biogas produced of 4,507.524 m^3 in 2019 instead of 3,593.471 in 2018; quantity of electricity invoiced of 3,908.248 kWh in 2019 instead of 4,676.173 kWh in 2018; quantity of cogenerated electricity of 7,237.844 kWh in 2019 instead of 5,864.653 kWh in 2018; annual gain can reach an amount of 300,000 MAD.

It was also noted that anaerobic digestion assisted by partial aeration can serve as a beneficial treatment strategy for the simultaneous treatment of waste and energy production.

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