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## HYDROGEN SULFIDE REMOVAL IN BIOGAS USING A FULL-SCALE BIOTRICKLING FILTER: EVALUATING SPRAYING TIME AND NITRATE SOURCE

Thaís C. Piovezan<sup>1</sup>, Rúbia Mores<sup>2</sup>, Ricardo L. R. Steinmetz<sup>3</sup>, Airtón Kunz<sup>1,3\*</sup>

<sup>3\*</sup>Corresponding author. Embrapa Suínos e Aves/Concórdia - SC, Brasil.

E-mail: [airton.kunz@embrapa.br](mailto:airton.kunz@embrapa.br) | ORCID ID: <https://orcid.org/0000-0002-6818-6580>

### KEYWORDS

desulfurization,  
nitrate, biofilter and  
biogas.

### ABSTRACT

The hydrogen sulfide ( $H_2S$ ) present in biogas needs to be removed due to concerns about corrosion during transportation, storage, health and safety. One of the existing removal processes is biological, using a biotrickling filter (BTF). In this study, the performance of full-scale BTF for  $H_2S$  removal under different operating conditions was evaluated. The BTF system was operated for 300 days, during which two spraying regimes (constant and intermittent) and two sources of nitrate ( $NO_3^-$ ) as nutrient solution were evaluated (residual effluent from pig farming and synthetic solution prepared with commercial  $NaNO_3$ ). The performance was monitored by the following parameters: removal efficiency (RE), elimination capacity (EC), pH, dissolved oxygen (DO), empty bed residence time (EBRT) and nitrate concentration ( $NO_3^-$ ). The results showed an  $RE_{H_2S} = 36.3\%$  with an  $EC = 1.95 \text{ g}_{H_2S} \text{ m}^{-3} \text{ d}^{-1}$  for constant spraying,  $RE = 99.59\%$  and  $EC = 4.2 \text{ g}_{H_2S} \text{ m}^{-3} \text{ d}^{-1}$  for intermittent spraying with residual effluent from pig farming and  $RE = 99.26\%$  and  $EC = 4.13 \text{ g}_{H_2S} \text{ m}^{-3} \text{ d}^{-1}$  with synthetic solution prepared with commercial  $NaNO_3$  solution. The results indicate that intermittent spraying provides better efficiency in the removal of  $H_2S$  from biogas regardless of the nitrate source (effluent or synthetic medium).

### INTRODUCTION

Biogas has become an alternative source of energy, as it is sustainable and profitable (Nhut et al., 2020). In Brazil, there is an increase in the number of installations of biogas production systems, in 2010 the country had 33 biogas plants and in 2023, the number increased to 883 plants and of these 347 are located in the South region. Constituting 28% of total biogas production in the South region, this portion represents the second largest contribution, behind only cattle farming, which leads with a share of 53% (CIBiogás, 2023).

Biogas production in the southern region of Brazil is stimulated by the animal protein industry, with a production of 4.9 million tons in 2023 in Brazil, making the country the fourth largest producer of pork in the world (EMBRAPA, 2023). As a result, waste is generated from the process, making it essential to implement appropriate treatment

(Becker et al., 2022). The treatment of wastewater from pig farming can be carried out through anaerobic digestion (AD), which makes it possible to generate energy and, at the same time, reduce air and water pollution (Hollas et al., 2022).

The main composition of biogas is methane ( $CH_4$ ) (40-75%), carbon dioxide ( $CO_2$ ) (25-60%) and approximately 0.1-2% other components. Among these components are ammonia and hydrogen sulfide ( $H_2S$ ) (Rybarczyk et al., 2019). Depending on the organic substrate used in AD, the concentration of biogas components varies (Omar et al., 2019). The concentration of  $H_2S$  from swine wastewater varies from 0.1 - 0.5%, which corresponds to 1000-5000 ppm<sub>v</sub> (Hollas et al., 2022) and is produced in the anaerobic digestion process by the degradation of organic compounds and reduction of inorganic species ( $SO_4^{2-}$ ) present in the substrates (Ghimire et al., 2021).

<sup>1</sup> Universidade Federal da Fronteira Sul/Erechim - RS, Brasil.

<sup>2</sup> Universidade do Contestado/Concórdia - SC, Brasil.

<sup>3</sup> Embrapa Suínos e Aves/Concórdia - SC, Brasil.

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According to the resolution of the National Petroleum, Natural Gas and Biofuels Agency (ANP) No. 906/2022, the limit of  $\text{H}_2\text{S}$  concentration for use as a fuel source must be less than  $10 \text{ mg m}^{-3}$  and the maximum concentration of total sulfur of  $70 \text{ mg m}^{-3}$  for biomethane. Concentrations exceeding this limit will be responsible for increased operational costs, such as corrosion in compressors, engines and storage tanks, in addition to being harmful to human health (Ariman & Koyuncu, 2022).

Currently, there are technologies for the desulfurization of biogas, which are classified as physical (adsorption, absorption), chemical (addition of iron compounds or iron oxide to the substrate) and/or biological (biodesulfurization) (Almenglo et al., 2016; Dupnock & Deshusses, 2020; Das et al., 2022; Ravishankar et al., 2022). Biodesulfurization technology has advantages when compared to other methods, such as: low implementation and maintenance costs (Almenglo et al., 2023) and high efficiency (Nhut et al., 2020).

The biotrickling filter (BTF) is one example of the biodesulfurization process. In this technology, raw biogas enters the system in an upward and countercurrent flow while the nutrient solution is added (Jia et al., 2022), facilitating mass transfer through contact between the liquid and gas phases. The nutrient solution can be obtained by treating wastewater from pig farming after the aerobic nitrification process (Cândido et al., 2022), providing a reduction in operational costs (Cano et al., 2021).

Sulfur-oxidizing bacteria (SOB), as noted by Dupnock & Deshusses (2020), form a biofilm when added to the support material. The presence of nutrient solution, such as nitrate ( $\text{NO}_3^-$ ), as highlighted by Becker et al. (2022), is beneficial for sulfur oxidation reactions, as nitrate acts as an electron acceptor for  $\text{H}_2\text{S}$ . This contributes to stabilizing common fluctuations in  $\text{H}_2\text{S}$  removal during operation and relieving stress on the microbial population when dosing is intermittent.

Another factor for reducing process costs is the support medium for SOB microorganisms, which mutually

influences the desulfurization of  $\text{H}_2\text{S}$  (Almenglo et al., 2023). Synthetic material (polypropylene) stands out when compared to inorganic (porous ceramics, rubber) and organic (wood chips) (Hirai et al., 2001; Wu et al., 2018), as it has a high surface area and is stable for fixing the SOB in addition to withstanding long-term operating conditions, high concentrations of  $\text{H}_2\text{S}$ , high mass transfer capacity under pressure drop conditions, easy drainage and biofilm regeneration if necessary (Nagendranatha et al., 2019).

Monitoring operational parameters, such as pH, dissolved oxygen (DO), nitrate concentration and alkalinity in the nutrient solution are important factors to ensure desulfurization efficiency (Nhut et al., 2020). The result of sulfurization can be evaluated by these factors are  $\text{H}_2\text{S}$  concentration, removal efficiency (RE), empty bed residence time (EBRT) and elimination efficiency (ER) (Das et al., 2022).

In this context, this study evaluated a biodesulfurization system at pilot scale using a BTF by applying different nutrient solutions (synthetic and residual effluent from pig farming) and their spraying modulation (continuous and intermittent).

## MATERIAL AND METHODS

To design the biogas desulfurization system using a full-scale biotrickling filter (BTF), the parameters studied by Pirolli et al. (2016) were used. The vertical cylindrical reactor made of high-density polypropylene (5 m high and 0.82 m diameter, with a useful volume of  $2.6 \text{ m}^3$ ), closed at both ends and filled with corrugated polypropylene (PP) tubes of different sizes (Figure 1) was installed in the swine waste treatment plant (SWTP) at Embrapa Suínos e Aves located in Concórdia, State of Santa Catarina, Brazil ( $27^\circ 18' \text{ S}$ ,  $51^\circ 59' \text{ W}$ ) (Kunz et al., 2009).

The corrugated tubes were the support material for the growth of the SOB, occupying around 15% of the useful volume of the reactor. The BTF system was built and installed by the company Kemia – Tratamento de Efluentes.

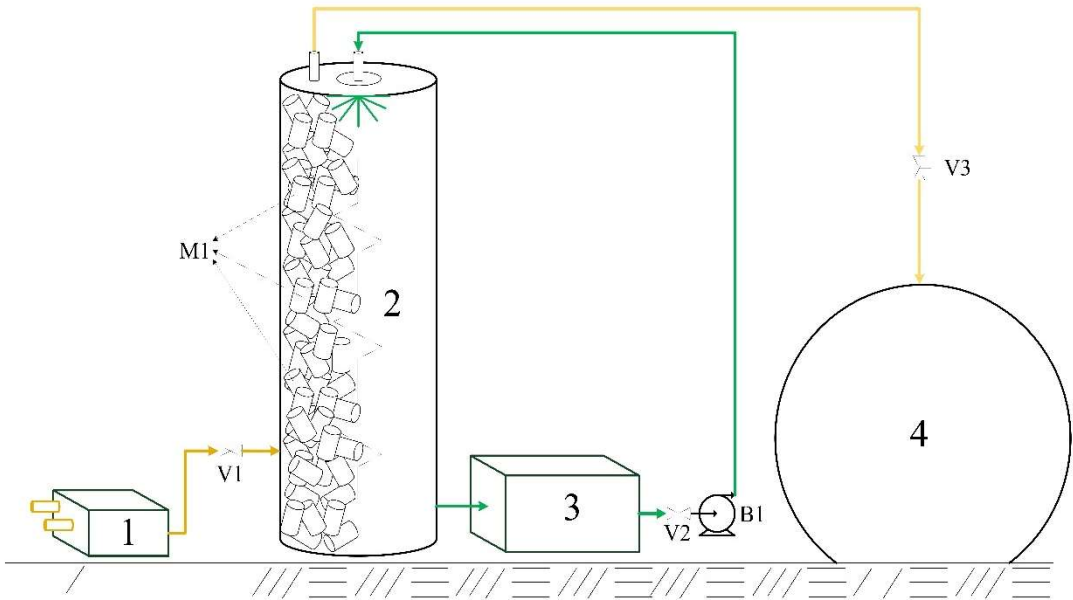


FIGURE 1. Simplified diagram of the system parts. 1= pressure equalizer; V1 = biogas inlet valve; 2 = biofilter; M1 = support medium; 3 = nutrient solution reservoir; B1 = 0.5 hp nutrient solution circulation pump; V2 = nutrient solution valve; 4 = desulfurized biogas reservoir; V3 = biogas outlet valve. 2 Food and nutritional medium; Yellow line= Biogas; Green line= Nutrient solution.

The BTF was continuously fed with raw biogas in an upward flow from the SWTP. The biogas was sent to an equalization box (0.1 m<sup>3</sup>) and went to the BTF system through the CONTECH-FT2 gas flow controller, Contechind. The desulfurized biogas was sent to a storage reservoir (50 m<sup>3</sup>).

The nutrient medium used as an electron acceptor in stage I and inoculation was collected at the outlet of the nitrifying reactor ([NO<sub>3</sub><sup>-</sup>] = 378 mg N L<sup>-1</sup> and Alkalinity = 390mgCaCO<sub>3</sub> L<sup>-1</sup>) and from the sludge settler located at the SWTP (Kunz et al., 2009). This residual effluent from pig farming was sent by a pump (BCR-2010 2P RT-128, Brazil) to the 480 L reservoir of the experimental system.

The synthetic solution was prepared using sodium nitrate (NaNO<sub>3</sub>) at a concentration of 400 mg NaNO<sub>3</sub> L<sup>-1</sup> (Synth) and sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>) at a concentration of 200 mg L<sup>-1</sup> to adjust the alkalinity (Quimica Moderna).

#### Reactor inoculation and system operation

The nutrient solution was stored in a reservoir (480L) and pumped with the aid of a centrifugal pump

(BCR-2010 2P RT-128, Brazil) to the upper end, which is sprayed onto the support material and comes into contact in counterflow with the biogas, after this process, the solution flows by gravity again to the reservoir (Figure 1).

#### Experimental Design

The work was divided into two stages, in stage I, the effect of the spraying time (constant and intermittent) and the H<sub>2</sub>S RE were evaluated using the residual effluent from pig farming as a nutrient solution at a flow rate of 2.8 m<sup>3</sup> h<sup>-1</sup>. In the intermittent experimental period, recirculation was evaluated over a period of 5 minutes divided equally into 8 cycles per day. In Stage II, the evaluation of the replacement of the recirculating nutrient solution (treated effluent from pig farming and synthetic solution) was carried out to remove the H<sub>2</sub>S present in the biogas, maintaining the spraying time with better performance according to the results of the first stage of the work. Table 1 presents the experimental configuration.

TABLE 1. Study of the impact of removal efficiency, with the nutrient solution being: residual effluent from pig farming (I) and synthetic (II); and evaluation of spraying modulation: constant (1) and intermittent (2).

Steps	Days of operations	Nutrient solution	Sprinkling regime
I-1	0-16	I- Waste effluent from pig farming	1- Continuous
I-2	17-200	I- Waste effluent from pig farming	2- Intermittent
II-2	201-300	II- Synthetic	2- Intermittent

## Biogas monitoring

Daily BTF measurements were taken at the inlet and outlet to quantify the H<sub>2</sub>S in the biogas. Concentrations were determined using the biogas analyzer (Geotech Biogás-5000, Geotechnical Instruments Ltd, United Kingdom) and flow measurement (CONTECH-FT2) and operational monitoring of the nutrient solution.

Nitrate determinations (NO<sub>3</sub><sup>-</sup> mg N L<sup>-1</sup>) were quantified by colorimetric methods in a flow injection analysis system (Fialab Instruments, Seattle, USA, model 2500), adapted from Rice et al. (2012). Alkalinity was determined using an automatic titrator (Titrino plus, Metrohm, Herisau, model 2500, Switzerland) and the results were expressed in mg<sub>CaCO3</sub> L<sup>-1</sup>, with these analyses being carried out twice a week. Daily monitoring of pH (pH, Hanna Instruments, Inc) and dissolved oxygen (DO) (YSI EcoSense DO 200A model) was carried out.

## Biogas quantification calculations

The evaluation of BTF with the aim of monitoring desulfurization was carried out through RE, elimination capacity (EC) in g m<sup>-3</sup> d<sup>-1</sup>, and empty bed residence time (EBRT) in h. The parameters were calculated according to eqs (1), (2) and (3).

$$RE = \frac{(C_{in} - C_{out})}{C_{out}} \times 100 \quad (1)$$

$$EC = \frac{C_{in} \cdot Q_{Biogas}}{V_{bed}} \quad (2)$$

$$EBRT = \frac{V_{bed}}{Q_{Biogas}} \times 24 \quad (3)$$

In which:

C<sub>in</sub> - input concentration, g m<sup>-3</sup>;

C<sub>out</sub> - outlet concentration, g m<sup>-3</sup>;

Q<sub>Biogas</sub> - Biogas flow input the BTF, m<sup>3</sup> d<sup>-1</sup>;

V<sub>bed</sub> - BTF reservoir volume, 2.4 m<sup>3</sup>;

To carry out the sizing calculations, methods based on the parameters established by Pirolli et al. (2016) were used. These analyzes were conducted considering a maximum H<sub>2</sub>S concentration (EC) of 4.8 g m<sup>-3</sup> h<sup>-1</sup>, together with the experimental values of the parameters, such as the initial concentration and nitrate concentration. The eqs (4)

to (9) going to to carry out the theoretical calculations of the BPF as described below:

$$CR = \frac{C_{in} \cdot Q_{biogas}}{V_{bed}} \quad (4)$$

$$[H_2S] = \frac{C_{in} \cdot MM}{V_m} \quad (5)$$

$$m_{H_2S} = \frac{[N-NO_3^-] \cdot V_r \cdot 6.07}{1000} \quad (6)$$

$$V_T = \frac{m_{H_2S}}{[H_2S]} \quad (7)$$

$$T_{sub} = \frac{V_{biogas}}{\Sigma Q_{biogas}} \quad (8)$$

$$Q_T = \frac{V_{biogas}}{T_{sub}} \quad (9)$$

In which:

CR = Removal Load, g m<sup>-3</sup> d<sup>-1</sup>;

[H<sub>2</sub>S] - H<sub>2</sub>S concentration, g<sub>H2S</sub> m<sup>-3</sup>;

MM - Molecular mass H<sub>2</sub>S, 34 g mol<sup>-1</sup>.

V<sub>m</sub> - Molar volume, 0.0224 m<sup>3</sup> mol<sup>-1</sup>;

m<sub>H2S</sub> - Mass of H<sub>2</sub>S, g<sub>H2S</sub>;

[N-NO<sub>3</sub><sup>-</sup>]: Nitrate concentration, mg L<sup>-1</sup>;

V<sub>r</sub> - Nutrient solution reservoir volume, 480 L;

6.07: coefficient, mg<sub>H2S</sub> mg<sub>N-NO3</sub><sup>-1</sup>

V<sub>T</sub> - Theoretical total biogas volume, m<sup>3</sup>;

T<sub>sub</sub> - Replacement time of nutrient solution for recirculation in BTF, d;

V<sub>biogas</sub> - Biogas volume, m<sup>3</sup>;

Q<sub>T</sub> - Theoretical Biogas flow input the BTF, m<sup>3</sup> d<sup>-1</sup>;

## RESULTS AND DISCUSSION

### Biotrickling filter (BTF) spray modulation

During the experimental period of the spraying regime study, the initial parameters in stages I-1 (continuous) and I-2 (intermittent) in the nutrient solution were similar, as shown in Table 2.

TABLE 2. Initial starting and monitoring parameters of 15 days (I-1) and after intermittent (I-2).

Initial Parameters	Units	I-1	I-2
Nitrate (NO <sub>3</sub> <sup>-</sup> )	mg <sub>N</sub> L <sup>-1</sup>	378	386
Alkalinity	mg <sub>CaCO3</sub> L <sup>-1</sup>	390	448
Hydrogen Sulfide (H <sub>2</sub> S)	ppm <sub>v</sub>	2080	1540
pH		7.18	6.63
Dissolved Oxygen (DO)	mg L <sup>-1</sup>	0.14	0.06



In Figure 2, the desulfurization activity in the BTF is shown according to the operational time of biogas purification using a biofiltration system at pilot scale. On the right, the results of  $\text{H}_2\text{S}$  in  $\text{ppm}_v$ , EBRT in hours, and pH are presented, and on the left, the results of RE in %, EC in  $\text{gH}_2\text{S m}^{-3} \text{d}^{-1}$ , and DO in  $\text{mg L}^{-1}$ . In Stage I-1, the system was operated with constant spraying of nitrate (real effluent) for 15 days. During this period, the system achieved a maximum RE of 36.3%

with an EC of  $1.95 \text{ gH}_2\text{S m}^{-3} \text{d}^{-1}$ . On the sixteenth day, the operation of the intermittent spraying system began, with a duration of 5 minutes eight times a day every 4 hours. When the system was modified to intermittent spraying, the RE in 24 hours was higher than 60% (RE = 67.33% and EC =  $0.8 \text{ gH}_2\text{S m}^{-3} \text{d}^{-1}$ ), and in 48 hours, it was >95% (RE = 98.04 and EC =  $1.27 \text{ gH}_2\text{S m}^{-3} \text{d}^{-1}$ ), demonstrating that the spraying time directly influences the RE.

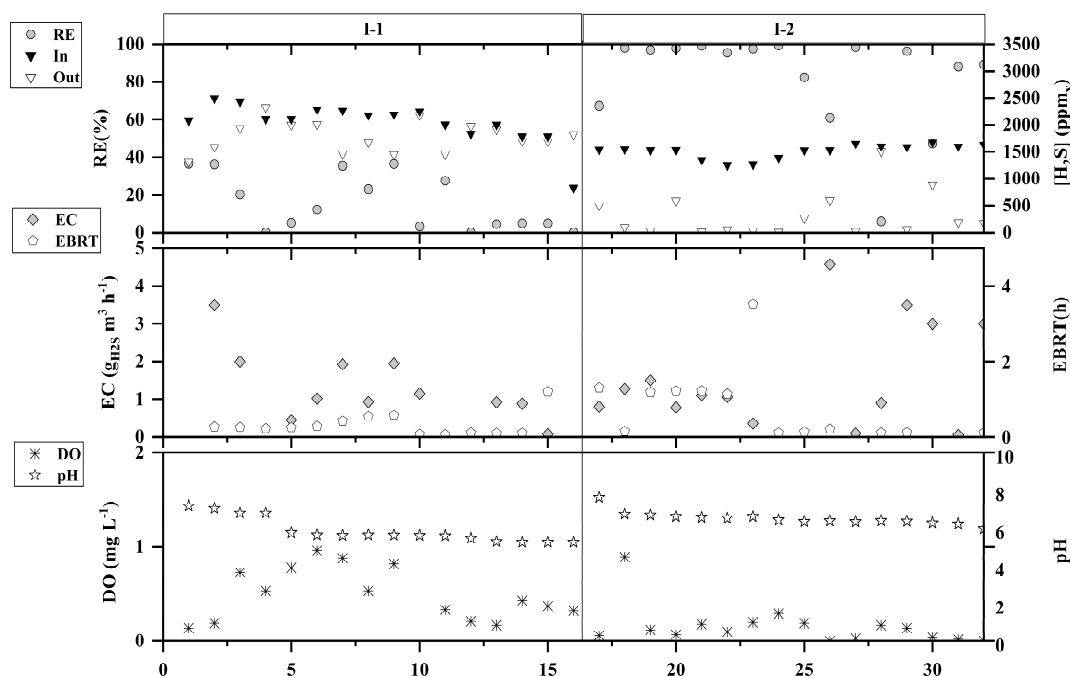


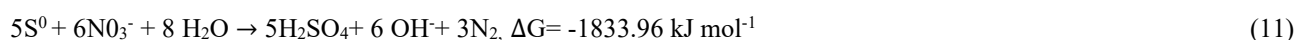
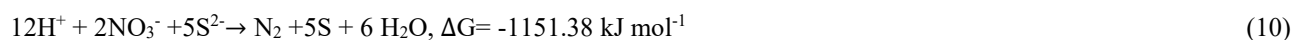
FIGURE 2. Profile of the beginning of desulfurization activity in biotrickling filter by modifying the recirculation time of the solution constantly over a period of 15 days (I-1) and after intermittent (I-2)

The spraying time of the nutrient solution plays a crucial role in the growth time kinetics of microorganisms in trickling biofilter systems. The relationship between these two elements is complex and directly influences the overall efficiency of the process (Becker et al., 2022). Adequate spraying time is essential to provide ideal conditions for the microorganisms present in the support medium (Cano et al., 2021). An insufficient spraying period can compromise the availability of essential nutrients, negatively affecting microbial growth kinetics (Becker et al., 2022). A lack of moisture and nutrients can result in lower-than-expected microbial growth, decreasing the system's ability to efficiently degrade gaseous pollutants (Almenglo et al., 2023). On the other hand, an excessively long spraying time can lead to an accumulation of moisture, adversely affecting microbial activity (Dupnock & Deshusses, 2020). Excess humidity can create unfavorable conditions, making the

environment less conducive to microbial growth (Almenglo et al., 2023).

The growth time of microorganisms, therefore, is intrinsically linked to the optimization of the spraying time of the nutrient solution. The search for an adequate balance between these factors is essential to ensure that microorganisms have sufficient access to nutrients, water and ideal environmental conditions for their growth and metabolic activity (Becker et al., 2022).

In Figure 2, the pH results, presented on the right axis, reveal a gradual decrease in the nutrient solution in both phases of the experiment. In stage I-1, during the first 15 days of operation with constant spraying, a faster reduction in pH was observed, going from 7.18 to 5.25. This phenomenon is directly associated with the oxidation of sulfide, which occurs in the denitrification process (10), followed by the subsequent oxidation of sulfur to sulfate (11). These chemical reactions are represented as:



These transformations lead to a reduction in the pH of the nutrient solution, as indicated in the results obtained (Nhut et al., 2020). Similar results were observed by Jia et al. (2022), where the pH started at 7.1 and reduced to 5.2, attributed to chemical reactions and the growth of sulfur-oxidizing bacteria (SOB) during the initial phase of BTF.

Regarding the relationship between  $\text{NO}_3^-$  and  $\text{S}^{2-}$  in BTF, it is essential to consider changing the spraying time. Changes in this parameter influence the kinetics of the reactions, affecting the presence of these ions in the nutrient solution (Becker et al., 2022). An optimized spraying time can favor sulfide oxidation, resulting in a higher concentration of nitrate ( $\text{NO}_3^-$ ) and affecting the system's efficiency in removing  $\text{H}_2\text{S}$ .

Other operational parameters, such as EBRT and dissolved oxygen (DO) concentration, can have an impact on system efficiency. However, the fluctuations that

occurred during the experiment remained within adequate ranges DO ( $0\text{--}1\text{ mg L}^{-1}$ ) (Becker et al., 2022) and EBRT  $> 120$  seconds (Nhut et al., 2020), showing resilience to pH fluctuations, but without affecting  $\text{RE}_{\text{H}_2\text{S}}$ .

### Replacement of the swine effluent nutrient solution with a synthetic solution

The total monitoring time of the system was 300 days, during which the EC varied from 0 to  $4.5\text{ g m}^{-3}\text{ h}^{-1}$ , as shown in Figure 3. This variation was directly linked to the following parameters:  $\text{H}_2\text{S}$  concentrations at the BTF inlet ranging from 500 to  $3500\text{ ppm}_v$  and the flow ranging between 0 and  $15\text{ m}^3\text{ d}^{-1}$ . These fluctuations can be attributed to changes in the composition of the raw material and the seasonality of the actual process at the treatment plant, as highlighted by Hollas et al. (2022).

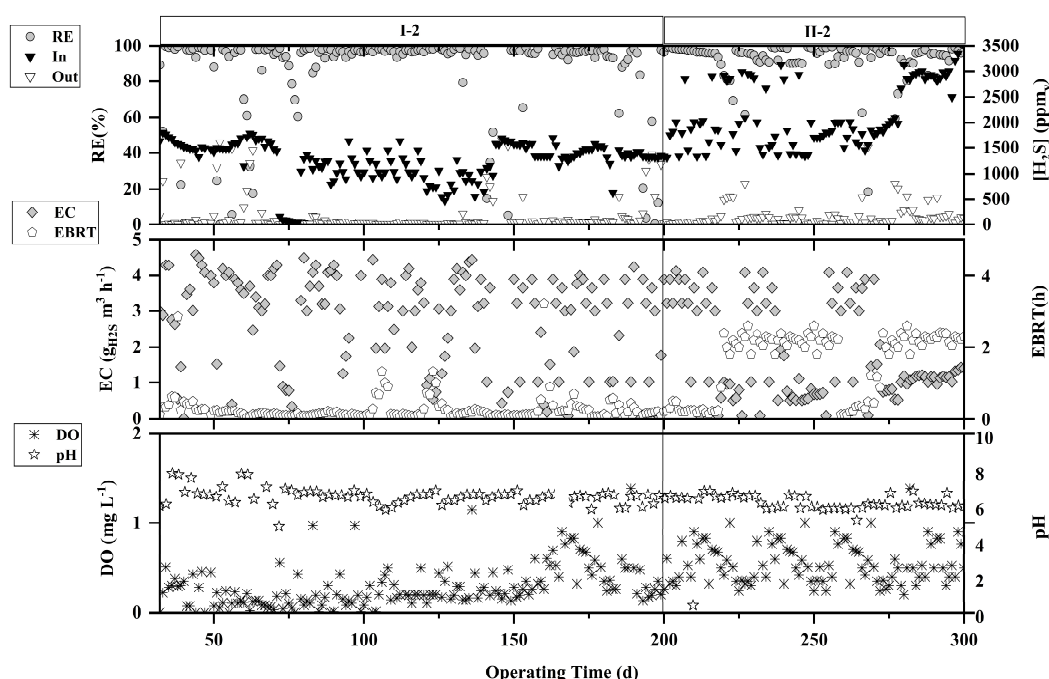


FIGURE 3. Hydrogen sulfide concentration measured at the biotrickling filter inlet and outlet along with removal efficiency, dissolved oxygen and pH.

Figure 3 details the evolution of desulfurization activity in BTF throughout the operation of the pilot-scale biogas purification system, using a biofiltration system in stages I-1 and II-2. Figure 3 presents the concentrations of  $\text{H}_2\text{S}$  in  $\text{ppm}_v$ , the EBRT in hours, the pH on the right axis, while on the left axis the results of RE in percentage, EC in  $\text{g}_{\text{H}_2\text{S}}\text{ m}^{-3}\text{ d}^{-1}$  and DO in  $\text{mg L}^{-1}$ .

During Stage I-2, the system operated with intermittent spraying of nitrate (swine waste effluent) for 200 days. During this period, a remarkable  $\text{H}_2\text{S}$  RE of 99.6% was recorded, accompanied by an EC of  $4.2\text{ g}_{\text{H}_2\text{S}}\text{ m}^{-3}\text{ d}^{-1}$ , EBRT of 0.16h, pH of 6.67 and DO of 0.29. The  $\text{NO}_3^-$  concentration was  $296\text{ mg L}^{-1}$ , and the alkalinity reached  $891\text{ mg CaCO}_3\text{ L}^{-1}$ . Similar results were observed by Pirolli et al. (2016) when using an anoxic BTF on a pilot scale with a nutrient solution from the same SWTP system, obtaining a RE of 99.8% and a higher EC of  $4.8\text{ g m}^{-3}\text{ h}^{-1}$ . These results highlight that the  $\text{NO}_3^-$  derived from the aerobic

biological process can be a viable alternative as a nutrient solution for the desulfurization of biogas using BTF.

In stage II-2, which commenced after the initial 200 days of operation (Phase I-2) and extended for 100 days, the use of a synthetic nutrient solution was chosen. The attainment of high  $\text{H}_2\text{S}$  removal rates from the initial stages reflects the rapid adaptation of the system to the new solution. This phenomenon is attributed to the acclimation of the BTF with Sulfur-Oxidizing Bacteria (SOB) present in the biofilm adhered to the support medium (as illustrated in Figure 4). In this configuration, the peak of operational performance was reached on the 270th day of operation, recording an EC of  $4.13\text{ g m}^{-3}\text{ d}^{-1}$ ,  $\text{RE}_{\text{H}_2\text{S}}$  of 99.2%, EBRT of 0.30 hours, pH of 6.52, and DO of 0.69. Furthermore, the concentration of  $\text{NO}_3^-$  was at  $200\text{ mg}_\text{N}\text{ L}^{-1}$ , and the alkalinity was  $357\text{ mgCaCO}_3\text{ L}^{-1}$ , showing similar efficiency compared to the pig farming effluent solution.

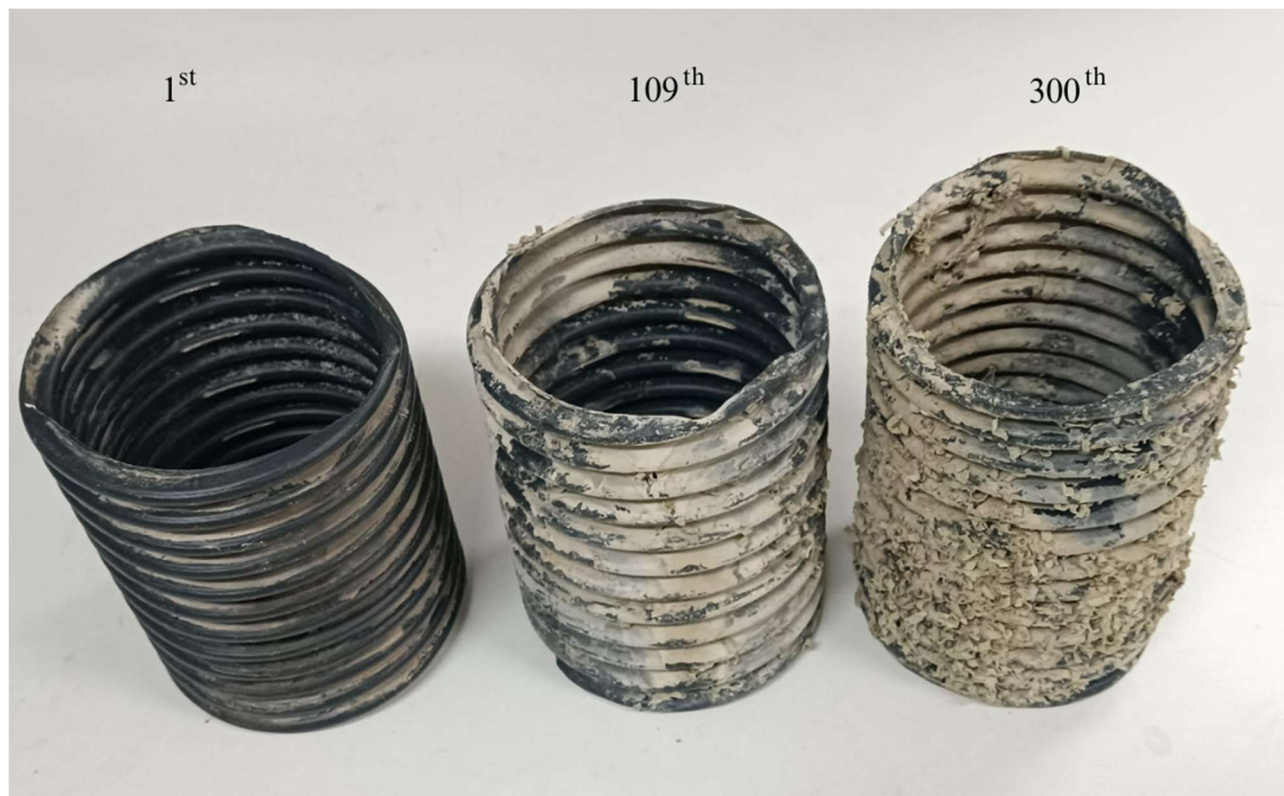
However, one of the factors that impacted the system was the variation in the nutrient solution replacement

period, which changed depending on the concentration and type of solution used. In Stage I-2, the replacement time was 5 days for the synthetic solution and 8 days for the solution coming from pig farming waste effluent. This difference was due to the longer time required for the solution to oxidize, as reported by Becker et al. (2022).

### Growth of the biofilm accumulated on the support material

Biofilm growth on a BTF plays a crucial role in the effectiveness of the gas treatment system. In this study, an increase in biofilm accumulation was observed over three distinct periods of operation: 1, 109 and 300 days (Figure 4).

FIGURE 4. Growth of accumulated biofilm on the support material on the 1<sup>st</sup>, 109<sup>th</sup> and 300<sup>th</sup> days of operation.



On the first day, the absence of biofilm suggests an initially clean surface, indicating the early stage of the project. After 109 days of operation, a notable increase in material adherence to the support was observed, indicating progressive biofilm development. At 300 days, even with the modification of the nutrient solution to a kinetic solution for a period of 100 days, the biofilm continued to grow, suggesting a persistent upward trend of the biofilm. The continuous increase can be attributed to the stability of the operating conditions, which provides a favorable environment for the gradual adaptation of SOB through a natural selection process (Nhut et al., 2020). The maintenance of the nutrient solution through pig farming effluent over time was a crucial factor for the continuous development of the biofilm, as stability in operational variables creates an environment conducive to sustained microbial growth (Pirolli et al., 2016).

The support medium played a significant role in biofilm development, as the structured materials and various sizes provided a suitable surface for microbial adhesion (Becker et al., 2022). The absence of microbial analysis makes it difficult to identify the specific species

involved. However, in the study conducted by Pirolli et al., 2016, which used a nutrient solution from the same location, a group of predominantly hydrogenotrophic bacteria from the Methanobacteriales (MBT) group was obtained on their support material.

### System Sizing

For the sizing of the system, the values of maximum taxa  $4.80 \text{ g H}_2\text{S m}^{-3} \text{ h}^{-1}$ ,  $\text{RE} = 99.8\%$  and volume of 43 liters, are used as initial operational (Pirolli et al., 2016). The theoretical calculations covered the initial concentrations of  $\text{N-NO}_3$  in stage 1 of  $384.9 \text{ mg N L}^{-1}$ , in stage 2 of  $258.1 \text{ mg N L}^{-1}$ , in stage 3 of  $389.6 \text{ mg N L}^{-1}$  and in stage 4 of  $597.8 \text{ mg N L}^{-1}$  and  $\text{H}_2\text{S}$  concentration in stage 1 of  $1483 \text{ ppm}_v$ , in stage 2 of  $1590 \text{ ppm}_v$ , in stage 3 of  $1436 \text{ ppm}_v$  and in stage 4 of  $1023 \text{ ppm}_v$ .

The results are summarized in Table 3, which compares the  $\text{H}_2\text{S}$  removal efficiency of the BTF system with theoretical and experimental values, demonstrating biogas volume (V), nutrient solution replacement time ( $t_{\text{sub}}$ ), and treated biogas flow rate (Q).

TABLE 3. Comparison of removal efficiency of the biotrickling filter system with theoretical and experimental values.

System parts	Theoretical			Experimental				
	V <sub>T</sub> (m <sup>3</sup> )	t <sub>sub</sub> (d)	Q <sub>T</sub> (m <sup>3</sup> d <sup>-1</sup> )	V <sub>T</sub> (m <sup>3</sup> )	t <sub>sub</sub> (d)	Q <sub>T</sub> (m <sup>3</sup> d <sup>-1</sup> )	RE <sub>max</sub> (%)	EC (g <sub>H2S</sub> m <sup>-3</sup> h <sup>-1</sup> )
1	489.2	5	96.6	96.6	9	40.2	99.5	0.50
2	309.5	2	156.4	167.4	7	47.9	99.6	1.35
3	520.8	3	183.4	183.4	8	22.9	99.2	1.00
4	715.5	5	156.0	156.1	7	26.0	99.7	1.90

Which: V<sub>T</sub>- Total biogas volume m<sup>3</sup>; T<sub>sub</sub>- Replacement time of nutrient solution for recirculation in BTF ; Q<sub>T</sub>- Biogas flow input the BTF m<sup>3</sup> d<sup>-1</sup>; ER<sub>max</sub>- Elimination efficiency maxima; EC- Elimination capacity

During the monitoring periods of nitrate consumption, it was observed that the biogas volume remained lower than the theoretical volume, in contrast to the opposite behavior in nutrient solution exchange. These discrepancies can be attributed to fluctuations in H<sub>2</sub>S concentration, flow rate, and variations in microbial pathways throughout the experiment (Nhut et al., 2020). A significant factor is the flow rate, as evidenced by the results indicating that the system was operating at a lower load, even when using all the biogas production from the Swine Waste Treatment Plant (SWTP) at EMBRAPA, given that the theoretical flow rates supported by the system are higher.

It is noted that the load-bearing capacity is 10 times higher than the load to which the system was subjected, as evidenced also by the biomass growth discussed earlier. If subjected to a higher load, the system would have a greater amount of biomass until obstruction occurred in the passage of biogas from the BTF.

## CONCLUSIONS

Follow-up studies in relevant environments have demonstrated high RE (>90%) when applied to swine waste biogas with H<sub>2</sub>S concentrations of up to 3500 ppm<sub>v</sub> at the system inlet, with intermittent spraying modulation and a nitrate concentration exceeding 150 mg L<sup>-1</sup>, achieving only 20 ppm<sub>v</sub> of H<sub>2</sub>S at the outlet. BTF demonstrated robustness even with process changes and efficiency was related to nitrate availability.

The stability of operating conditions and the maintenance of an environment favorable to the development of microbial biofilm were essential to the success of the BTF. The use of a synthetic nutrient solution after 200 days also proved to be effective, allowing rapid adaptation of the system and maintaining high H<sub>2</sub>S removal efficiency rates.

This study provides experimental evidence that the pilot BTF can be an alternative for removing H<sub>2</sub>S in biogas in a pig farming scenario to enable biogas use projects with less investment capacity. In terms of sizing, the BTF capacity proved to be greater than the loads applied during the experiment, suggesting that the system could withstand greater loads without compromising efficiency.

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## Conflict of interest

The authors declare no conflict of interest.

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