Agro-industrial residues as biofilter media for biogas desulfurization: an alternative to activated carbon

Abstract: Biogas desulfurization is essential to prevent equipment corrosion and ensure its suitability for energy applications. Although several technically established desulfurization technologies are commercially available, their implementation may still be constrained by acquisition costs, operational expenses, and environmental impacts associated with the use of conventional materials such as activated carbon. In this context, this study investigates the potential of two abundant agro-industrial residues - *Luffa cylindrica* and *Carya illinoinensis* - as low-cost and environmentally favorable filter media for biofilters applied to biogas desulfurization. A pilot-scale system operated for 62 days treating biogas with H_2S concentrations above 5,000 ppm, and their performance was compared with that of a conventional activated carbon filter. Both biofilters demonstrated high initial removal efficiencies, reducing H_2S concentrations in the treated gas to below 20 ppm. Over time, the loofah-based filter showed a decline in performance due to sulfur accumulation, whereas the pecan shell medium exhibited greater operational stability and indications of self-regeneration, achieving performance statistically comparable to that of activated carbon. These results highlight the technical feasibility of using abundant agro-industrial biomass as alternative biofilter media, contributing to reduced operational costs and lower environmental impacts in biogas purification systems. The findings also reveal opportunities for integrating biomass waste valorization with biogas purification technologies, indicating promising pathways for more sustainable and economically accessible desulfurization strategies in decentralized and industrial biogas applications.

Keywords: Hydrogen sulfide removal. Biofiltration. Agro-industrial residues. Pilot-scale system. *Luffa cylindrica*. *Carya illinoinensis*.

Introduction

The large-scale use of fossil fuels has played an important role in industrial development but has also significantly increased greenhouse gas emissions and contributed to climate change (Piovani et al., 2025). In addition to environmental and economic considerations, the development of sustainable energy technologies is increasingly associated with broader social responsibility and societal impacts, including corporate sustainability strategies and public acceptance of energy innovations (Zvarikova et al, 2024a; Rehman et al, 2024). In response, renewable energy sources such as biomass have gained increasing attention due to their potential to convert agricultural and livestock residues into valuable energy carriers (Riva et al., 2025). Among biomass-based energy pathways, biogas production through anaerobic digestion has emerged as one of the most established technologies for converting organic residues into renewable energy.

The global biogas sector has experienced steady growth in recent years, reaching an installed capacity of approximately 120 GW worldwide and a market value of around USD 79 billion. Continued expansion toward 2030 is expected, particularly driven by biomethane production and upgrading technologies. Europe remains the most mature market, concentrating more than 70% of global production capacity, while countries such as China, India, Brazil, and the United States have shown rapid growth supported by renewable energy and waste-to-energy policies (Bórawski et al., 2024; Swinbourn and Wang, 2024). In parallel, technological developments have focused on improving feedstock conversion efficiency through biomass pretreatment and the use of agricultural residues such as straw and grass silage in anaerobic digestion systems, enhancing methane yields and expanding substrate utilization (Maroušek, 2013b; Maroušek, 2014). These advances highlight the importance of optimizing the entire biogas value chain, from biomass processing to gas purification and upgrading.

Studies have explored strategies to valorize anaerobic digestion by-products within a circular bioeconomy framework. One promising approach is the thermochemical conversion of digestate into biochar, which can accelerate methanogenesis by enhancing microbial activity and electron transfer processes. Due to its high porosity and sorption capacity, biochar can also act as an adsorbent for gaseous pollutants, including sulfur compounds, contributing to emission mitigation in biogas systems. In addition, its application as a soil amendment has been associated with improvements in soil structure, nutrient retention, and water holding capacity, highlighting its potential role in sustainable agricultural and waste management practices (Stehel et al., 2017; Vochozka et al., 2016; Strunecký et al., 2021).

Despite the technological progress and global expansion of the sector, biogas produced during anaerobic digestion may contain trace contaminants, among which hydrogen sulfide (H_2S) is particularly problematic due to its toxicity and corrosive properties (Horn et al., 2024; Vakili et al., 2024; Gupta et al., 2023). Its presence compromises the performance of engines and turbines, accelerates lubricant degradation, and, during combustion, leads to the formation of sulfur oxides responsible for acid rain (Karne et al., 2023). Therefore, desulfurization is essential to ensure the technical and economic viability of biogas (Mutegoa and Sahini, 2023).

H_2S removal systems must combine high purification efficiency, low cost, and operational simplicity. The removal of H_2S may occur either inside or outside the biodigester through physical, chemical, or biological methods (Cavaler et al., 2022).

Among physical methods, adsorption stands out as a gas-solid process in which biogas passes through a fixed bed of adsorbent material, promoting mass transfer and contaminant removal (Vuppaladadiyam et al., 2024). Materials such as zeolites, metal oxides, carbon-based materials, and polymers are commonly used as adsorbents (Pudi et al., 2022). The efficiency of the process depends on the surface area and porosity of the material, with activated carbon characterized by micropores (<2 nm), mesopores (2–50 nm), and macropores (>50 nm) (Paglini et al., 2022; Sultana et al., 2022). However, its commercial use is limited by high cost and rapid pore obstruction (Anastopoulos and Pashalidis, 2020). In this context, biological technologies such as biofilters emerge as more economical and sustainable alternatives (Agudelo et al., 2023).

Biological methods, such as biofiltration, employ porous materials that favor the formation of microbial biofilms capable of oxidizing H_2S into less toxic compounds, such as elemental sulfur and sulfate (Merouani et al., 2024). Biogas passes through a bed colonized by sulfur-oxidizing bacteria, under controlled conditions of humidity, temperature, and pH (Schirmer et al., 2023), including species such as *Thiobacillus*, *Sulfolobus*, *Thermothrix*, *Beggiatoa*, and *Thiothrix*, which exhibit high affinity for H_2S and O_2 (Huertas et al., 2020). The process consists of the transfer of H_2S to the liquid phase of the filter material, followed by microbial degradation (Aita et al., 2016; Agudelo et al., 2023), exhibiting stability, easy construction and simplified maintenance, suitable for small-scale operations and rural areas (Huang et al., 2024; Becker et al., 2022).

In biofiltration systems, hydrogen sulfide removal is mainly driven by biological oxidation mediated by sulfur-oxidizing microorganisms. At the molecular level, H_2S dissolves in the aqueous phase of the biofilm and dissociates into HS^- and S^{2-} depending on pH conditions. These reduced sulfur species are oxidized by bacteria such as *Thiobacillus* and *Beggiatoa*, transferring electrons to oxygen as the terminal electron acceptor. The process typically occurs in sequential steps, producing elemental sulfur (S^0) and, under higher oxygen availability, sulfate (SO_4^{2-}). These transformations convert toxic hydrogen sulfide into less harmful sulfur compounds, enabling its continuous removal in biofiltration systems (Muyzer and Stams, 2008; Janssen et al., 2009).

The selection of packing material is essential for the proper performance of biofilters, requiring high mechanical resistance, good moisture retention, large surface area, porosity, and the ability to support microbial growth (Pudi et al., 2022). In this study, the biological method of biofiltration was applied using organic materials such as loofah sponge (*Luffa cylindrica*) and pecan nutshell (*Carya illinoinensis*) as support media in the biofilter, selected due to their regional availability in Rio Grande do Sul, which reduces costs and adds value to local agro-industrial residues.

Loofah sponge stands out as an efficient support for microbial biofilms due to its renewable origin and properties such as high surface area, low density (0.82 - 0.92), good toughness, flexibility, easy processing, recyclability, low cost, and corrosion resistance - characteristics that make it a sustainable and economically viable option (Abdel-Fattah et al., 2022; Guevara-López and Buitrón, 2015; Akgül et al., 2013; Alhijazi et al., 2020).

Pecan nutshell, which represent 40–50% of the total mass of the fruit, are a widely available and underutilized agro-industrial by-product, commonly discarded by burning or exposure to the open air (Dunford et al., 2022; Ruiz et al., 2024). Studies indicate that this residue has physical properties suitable for use in biofilters for H_2S removal, with a specific mass of 752 kg m^{-3}, a total porosity of 77%, and an aeration space of 23% (Fermino et al., 2015). It also has potential for the production of high surface area activated carbon (400–1000 m^2 g^{-1}), expanding its applicability in adsorption processes and in the treatment of gases and effluents (Dunford et al., 2022; Mendoza et al., 2024).

Despite the growing number of studies on biological desulfurization of biogas, important knowledge gaps remain regarding the performance of alternative biofilter media derived from agro-industrial residues under pilot-scale conditions. Most existing studies focus on laboratory-scale systems or evaluate individual materials without direct comparison with conventional adsorbents such as activated carbon (Becker et al, 2022). Accordingly, this study investigates the purification of biogas using pilot-scale biofiltration systems packed with agro-industrial materials, specifically loofah sponge (*Luffa cylindrica*) and pecan nutshell (*Carya illinoinensis*), comparing their performance with the conventional adsorption method using activated carbon.

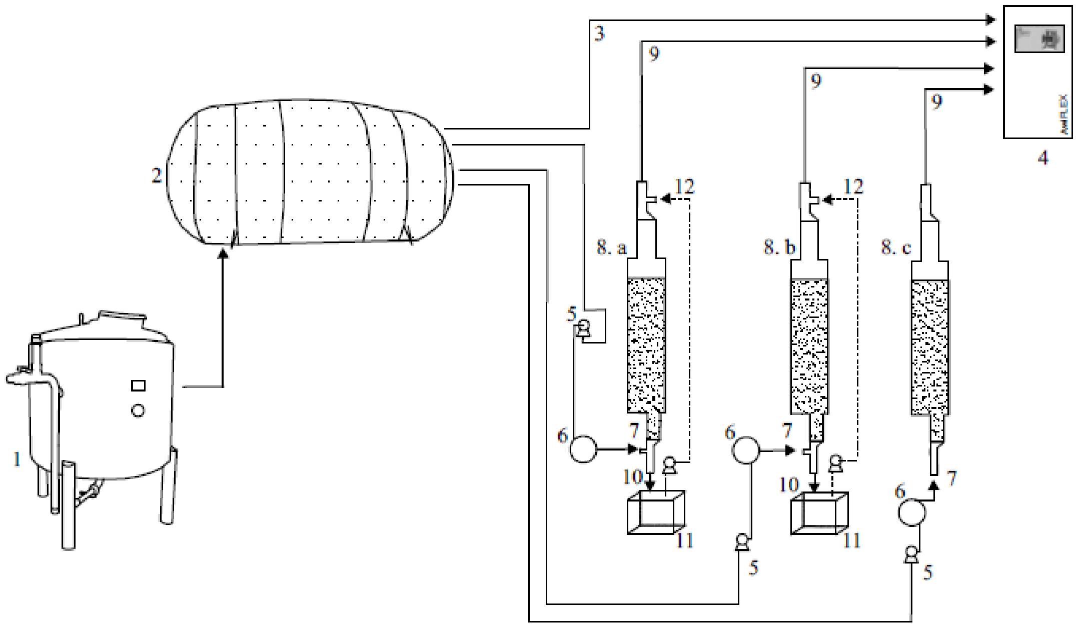
The central hypothesis of this study is that lignocellulosic agro-industrial residues with high porosity and surface area can serve as effective support media for sulfur-oxidizing microorganisms, enabling hydrogen sulfide (H_2S) removal efficiencies comparable to those obtained with commercial adsorbents. From an industrial perspective, identifying alternative packing materials derived from locally available agricultural residues may contribute to reducing operational costs and environmental impacts associated with conventional purification systems. By providing a comparative pilot-scale evaluation of these materials under the same operational conditions, this study contributes to the development of low-cost and sustainable desulfurization strategies for biogas purification. Given the rapid expansion of the global biogas sector and the increasing demand for efficient gas purification technologies, such approaches represent an important step toward improving the economic viability and environmental performance of biogas-based energy systems.

Methodology

Pilot Plant Configuration

The experiment was conducted at the institution's pilot plant. For the tests, three independent columns were constructed: two operated as biofilters, each packed with a different support material, and one operated as an adsorption column using activated carbon. All columns were packed with the same bed volume and operated simultaneously under identical operating conditions, including the same gas flow rate and inlet H_2S loading. The systems were operated predominantly under anaerobic conditions; however, small concentrations of oxygen were occasionally detected in the raw biogas, which is commonly observed in biogas systems due to minor air ingress or operational conditions during biogas production (Buffière et al., 2025; Jameel et al., 2024). These trace oxygen levels were not intentionally introduced and were inherent to the composition of the raw biogas stream. The system configuration is shown in Figure 1.

Figure 1. Configuration of the pilot plant used in the experiments



Legend: Schematic representation of the pilot-scale experimental system used for biogas desulfurization. The system consists of a stainless steel reactor (1) connected to a 20 m^3 gasometer (2) for raw biogas storage. The raw biogas outlet stream (3) is monitored using an AwiFLEX modular analyzer (4) and distributed to three treatment columns operating in parallel: (8.a) column packed with pecan nutshell (*Carya illinoinensis*), (8.b) column packed with loofah sponge (*Luffa cylindrica*), and (8.c) column packed with activated carbon, used as the conventional reference material. The biogas inlet stream to the columns (7) is controlled using Ritter flow meters (6), while RS-385 mini water pumps (5) promote digestate recirculation. The digestate recirculated within the columns (12) exits through the digestate outlet stream (10) and is stored in the digestate storage tank (11). After treatment, the purified biogas is discharged through the outlet stream (9). This experimental configuration enables the simultaneous operation of three columns under identical operating conditions, allowing a direct comparison between agro-industrial biomass materials and conventional activated carbon for H_2S removal from biogas. The system was designed to evaluate the technical feasibility of abundant agro-industrial residues as alternative biofilter media.

The biofiltration columns were constructed based on the study by Becker et al. (2024) using transparent polymethyl methacrylate, with a diameter of 0.1 m, a height of 1 m and a thickness of 0.002 m. The components were interconnected by means of polyvinyl chloride (PVC) connections (nominal diameter ½ inch). At the top of each column, the inlet points for the digestate and the outlet for the purified biogas were installed. At the base, the inlet for the raw biogas and the outlet for the liquid phase were positioned. Flow control for the liquid and gas streams was carried out by means of PVC ball valves ½ inch manual.

To ensure continuous circulation of the biogas, a 12 V RS-385 mini water pump was used, with an internal outlet diameter of 5.00 mm and an external diameter of 7.5 mm, and an estimated maximum flow rate of between 1 and 3 liters per minute was used. An identical pump was employed for digestate recirculation, ensuring adequate moisture in the filter bed. At the base of each biofilter, a reservoir was attached for storage and daily collection of digestate, enabling its monitoring and recirculation, as well as the control of parameters such as pH throughout the tests.

Each column was filled with a support material intended for microbial attachment and growth. One column was filled with a loofah sponge (≈2 cm cubes) selected due to its high porosity and large contact surface area (Adeniyi et al., 2023). The second column was filled with pecan nutshells, chosen for their physical structure, with a total porosity of 77% and an aeration space of 23% (Fermino et al., 2015). The shells were sieved and only those with a particle size greater than 6.3 mm were selected to optimize the gas and liquid flow in the column. The selection of materials took into account their wide regional availability: the loofah sponge was obtained from local producers and the pecan nutshells were supplied by a processing company.

Before the start of the tests, the columns were subjected to a microbial colonization phase with the aim of promoting biofilm formation on the support materials. Considering the Hydraulic Retention Time (HRT), the column containing loofah sponge was filled with 6 L of digestate and maintained under these conditions for 24 h, while the column with pecan nutshell received 5.3 L and remained for 50 h due to its lower water retention capacity. These differences are related to the distinct physical characteristics of the packing materials.

Subsequently, the recirculation of 2 L of digestate for 50 s every 24 h was initiated, controlled by a Programmable Logic Controller (PLC), without biogas flow, to promote microbial adaptation and stabilization. The microorganisms naturally present in the digestate served as biological inoculum and nutrient source, enabling the establishment of adapted communities capable of H_2S oxidation (Kunz et al., 2019).

During the operational phase, biogas was introduced gradually, allowing microbial adaptation to the new conditions, as recommended by Ningsih, Hasanudin and Roubík (2024). Injection began on the eighth day after inoculation of the filter medium in both columns, with an average flow rate of 1 L min^{-1}. From the eighth to the twelfth day, four daily cycles of 30 min were applied, followed by 30 min cycles every 3 h from the thirteenth day onwards. On the eighteenth day, the system began operating for 6 continuous hours per day, maintaining 30 min cycles every 3 h. This operating period was increased to 12 continuous hours on the twenty-fourth day and subsequently to 18 h on the forty-third day, remaining at this level until the end of the experiment.

The biogas volume was monitored using a Ritter TG meter (model TG 0.5, Ritter Apparatebau GmbH & Co. KG, Bochum, Germany), whose rotation corresponds to the displacement of 0.5 L of gas, with daily manual readings performed at the same time each day to ensure consistency in data collection. Considering the use of digested sludge as inoculum biomass, an average adaptation period of 10 days was estimated before continuous operation (Zhang et al., 2022b).

The third column, intended for adsorption, differs only in that it operates exclusively with gas flow. Raw biogas is introduced at the base and is collected at the top already purified. The choice of activated carbon as an adsorbent medium is based on its widespread use in the removal of sulfur compounds, due to its high surface area and adsorption capacity (Bhatnagar et al., 2013). This unit serves as a reference for evaluating the performance of the biofilters. The biogas flow, as well as the monitoring procedures and the evaluation of H_2S removal efficiency, followed the same protocol used for the biofilters. The operating time was increased stepwise, starting with short operating periods and progressively reaching 18 h of daily operation at an average flow rate of 1 L min^{-1}.

Data Collection and Analysis

The experimental period lasted 62 days. The quantification of the main constituents of biogas (CH_4, CO_2, O_2, H_2 and H_2S) was performed using the portable modular biogas analyzer AwiFLEX (Awite Bioenergie GmbH, Langenbach, Germany), with daily monitoring. Raw biogas measurements were taken twice a day, while treated biogas was automatically analyzed every 2 hours. The analyzer is designed for continuous and high-precision monitoring in biogas production and purification systems. Ambient temperature was continuously monitored using a FieldLogger datalogger (Novus Automation, Brazil).

pH readings were taken three times a week (Mondays, Wednesdays and Fridays), using a pH meter (model DM-320, Digimed Analytical Instruments, São Paulo, Brazil). This control is essential to maintain neutral conditions for the metabolic activity of microorganisms involved in the oxidation of sulfur compounds, such as Thiobacillus thioparus, whose growth is favored in a pH range of 6 to 8 (Souza and Rizzatto, 2022; Sundari et al., 2023). Furthermore, the oxidation of H_2S results in the formation of sulfuric acid, which can lead to acidification of the digestate and, consequently, reduce the solubility of H_2S in the aqueous phase under overload conditions (Ibrahim et al., 2022; Sáenz et al.; 2009). The digestate was recirculated once a day, ensuring adequate moisture conditions in the filter bed for the survival and activity of the microbial communities present in the system (Lenis et al., 2023).

The experimental data were organized in Microsoft Excel spreadsheets. The H_2S removal efficiency in the biogas was calculated according to Equation 1, with the result expressed as a percentage (%) (Cortés et al., 2021).

Equation 1

$$E = \frac{C_{H_2S\,in} - C_{H_2S\,out}}{C_{H_2S\,in}} \cdot 100$$

Where:

E = removal efficiency of the H_2S (%);

$C_{H_2S\,in}$ = inlet concentration of H_2S (ppm);

$C_{H_2S\,out}$= outlet concentration of H_2S (ppm).

Additionally, the elimination capacity (EC), inlet loading rate (ILR) and empty bed residence time (EBRT) were calculated to evaluate the operational performance of the biofiltration system, according to Equations 2–4 (Zhang et al., 2022a).

Equation 2

$$EC = \frac{Q}{V} \cdot \left(C_{H_2S\,in} - C_{H_2S\,out}\right)$$

Equation 3

$$ILR = \frac{C_{H_2S\,in} \cdot Q}{V}$$

Equation 4

$$EBRT = \frac{V}{Q}$$

Where:

EC = elimination capacity of H_2S ($g\ m^{-3}\ h^{-1}$);

ILR = inlet loading rate (g m^{-3} h^{-1});
EBRT = empty bed residence time (h);
$C_{H_2S\ in}$ = inlet concentration of H_2S (g m^{-3});
$C_{H_2S\ out}$= outlet concentration of H_2S (g m^{-3});
Q = gas flow rate (m^3 h^{-1});
V = packed bed volume (m^3).

The H_2S removal was calculated based on the difference between inlet and outlet concentrations, according to Equation (5).

Equation 5

$$H_2S_{removed} = \Sigma\ (C_{H_2S\ in} - C_{H_2S\ out})_1 + (C_{H_2S\ in} - C_{H_2S\ out})_2 + \ldots + (C_{H_2S\ in} - C_{H_2S\ out})_{62}$$

Where:
$C_{H_2S\ in}$ = inlet concentration of H_2S (g m^{-3});
$C_{H_2S\ out}$= outlet concentration of H_2S (g m^{-3}).

As the columns had identical dimensions and operated under the same gas flow rate, the EBRT was the same for all reactors. Graphs were developed to represent the temporal evolution of the main biogas components concentrations (CH_4, CO_2, O_2, H_2 and H_2S) and the H_2S removal efficiency throughout the experimental period. For statistical analysis, the non-parametric Kruskal–Wallis test was applied to verify significant differences in H_2S emissions among the three treatments. When differences were identified, pairwise comparisons were performed using the Mann–Whitney test. Additionally, Spearman's non-parametric correlation was employed to evaluate the relationship between ambient temperature and H_2S removal efficiency across the different treatments.

Results and Discussion

The system operated for a total of 1006 h after the start of biogas feeding into the columns, during which 139.5 L of biogas were treated. Among the systems evaluated, the columns filled with pecan nutshell and activated carbon processed similar volumes of raw biogas (51.185 L and 51.449 L, respectively), while the column with loofah sponge treated

36.911 L. These values correspond to average biogas flow rates of 0.96 L min^{-1} for pecan nutshell, 0.72 L min^{-1} for loofah sponge, and 0.97 L min^{-1} for activated carbon. The lower treated volume observed for the loofah sponge column is mainly associated with operational interruptions during the experimental period, including the replacement of pumps due to equipment malfunction, which temporarily interrupted the biogas supply to this system.

The difference in usable volume and hydraulic retention time (HRT) observed between the materials, 5.3 L and 50 h for the pecan shell column and 6 L and 24 h for the loofah sponge column, can be attributed to the physical characteristics of the packing materials. Highly porous materials tend to retain more liquid in their internal structure, increasing their liquid retention capacity, while more rigid and less absorbent materials retain less liquid, with flow occurring mainly through the interparticle spaces, which may require a longer contact time for adequate microbial adhesion (Rahman et al., 2021; Soares et al., 2025; Cui et al., 2024).

During the experiment, ambient temperatures ranged from −5.3 °C to 24.9 °C, with an average of 13.8 °C. This daily variation is characteristic of the region, which has a subtropical climate. According to Caron et al. (2024), during the winter, the average temperature in the state of Rio Grande do Sul tends to remain around 14.9 °C, considering the quarter in which the experiments were conducted.

To assess the influence of temperature on the efficiency of H_2S removal in each column, Spearman's non-parametric correlation was applied. The rs coefficient indicates the strength and direction of the relationship between two variables, ranging from −1 to +1, while the p-value indicates whether the correlation is statistically significant, following the criteria indicated by Baranhuk et al. (2024), the hypothesis is rejected when $p > 0.05$.

For the biological methods, the pecan nutshell column showed rs = 0.0084 and p = 0.1311, while the loofah sponge column showed rs = 0.00018 and p = 0.1863. For the activated carbon adsorption method, rs = 0.2504 and p = 0.0586. Although the positive rs in the activated carbon column indicates a correlation trend, the p-value is still above 0.05.

Therefore, none of the columns showed a statistically significant correlation, indicating that temperature variation did not influence H_2S removal efficiency.

Digestate Characterization

In the initial characterization of the digestate introduced into the biofiltration columns, the total solids were 0.59%, with a predominance of fixed solids (72.80%) over volatile solids (27.20%). However, it was necessary to replace the digestate on the 40th day of operation, due to an increase in the concentration of H_2S in the treated biogas, indicating a possible reduction in the microbial activity responsible for removing the contaminant.

The new digestate presented a distinct composition, with total solids of 0.87% and a higher proportion of volatile solids (51.56%) compared to fixed solids (48.44%). This increase in the volatile solids fraction suggests a greater availability of biodegradable organic matter (Pereira et al., 2024), which may favor the adaptation and growth of the microbial biofilm responsible for the oxidation of H_2S.

After recirculation in the columns, the digestate showed distinct changes depending on the packing material. In the column filled with pecan nutshell, total solids reached 0.94%, with 32.61% volatile solids and 67.39% fixed solids. In the column filled with loofah sponge, total solids reached 0.93%, with a higher proportion of volatile solids (37.95%) and 62.05% fixed solids. This behavior suggests greater retention of organic matter and, consequently, greater biomass accumulation in the loofah sponge column.

At the end of the experiment, further analyses of the digestate confirmed this trend. The pecan shell column showed 1.04% total solids, with 39.42% volatile solids and 60.58% fixed solids. The loofah sponge column showed 0.96% total solids, with 50.93% volatile solids and 49.07% fixed solids.

Throughout the operation, the continuous recirculation of the digestate and the development of microbial biofilm may contribute to a gradual increase in the volatile solids fraction, due to the accumulation of biomass and organic matter in the liquid phase. In biofilters and biotrickling filters, higher concentrations of organic solids in the recirculating liquid are commonly associated with increased biofilm formation. Highly porous and fibrous materials tend to enhance organic matter retention, microbial adhesion and the development of dense biofilms, which may explain the higher VS observed in the column packed with loofah sponge (Pineda et al., 2023; Feng et al., 2019; Tu et al., 2017).

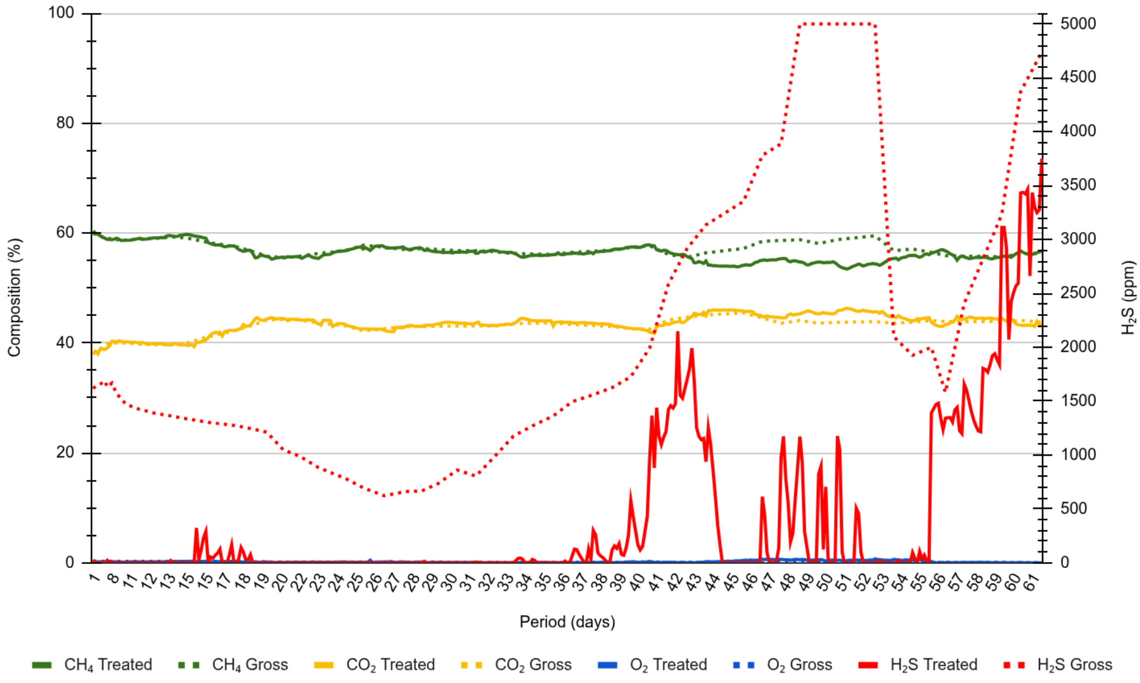
Additionally, lignocellulosic packing materials may undergo partial biodegradation during prolonged operation, releasing organic fractions into the liquid phase and potentially contributing to the increase in VS. Such processes can lead to structural changes in the packed bed, including reduced effective porosity and increased resistance to gas flow, which may limit H_2S mass transfer and reduce removal efficiency (Vela-Aparicio et al., 2021; Pu et al., 2018). These mechanisms have been reported as common causes of performance decline in biofilters treating H_2S and other gaseous contaminants. However, further microbiological investigations are required to better understand the interactions between microbial communities and natural support materials such as loofah sponge and pecan nutshell in biofiltration systems.

pH measurements were performed three times a week and, in both columns, the average value was 7.67, remaining within the range established in the methodology.

Removal Efficiency

The performance of the biofilters was evaluated through continuous monitoring of biogas composition before and after treatment. Figure 2 shows the biogas composition before and after treatment in the biofiltration column using pecan nutshell as the support medium.

Figure 2. Changes in biogas constituents after treatment in the column filled with pecan nutshell



Legend: The figure shows the temporal evolution of CH_4, CO_2, and O_2 concentrations in raw and treated biogas, as well as the H_2S concentration throughout the experimental period, after treatment in the column packed with pecan nutshell (*Carya illinoinensis*). Despite the variations in H_2S concentration in the raw biogas, the system achieved significant removal after treatment, while maintaining relatively stable levels of the main energetic constituents of the biogas. These results demonstrate the efficiency of pecan nutshell as a filter medium for H_2S removal without compromising the energetic quality of the biogas, reinforcing its potential as a low-cost alternative material for desulfurization processes.

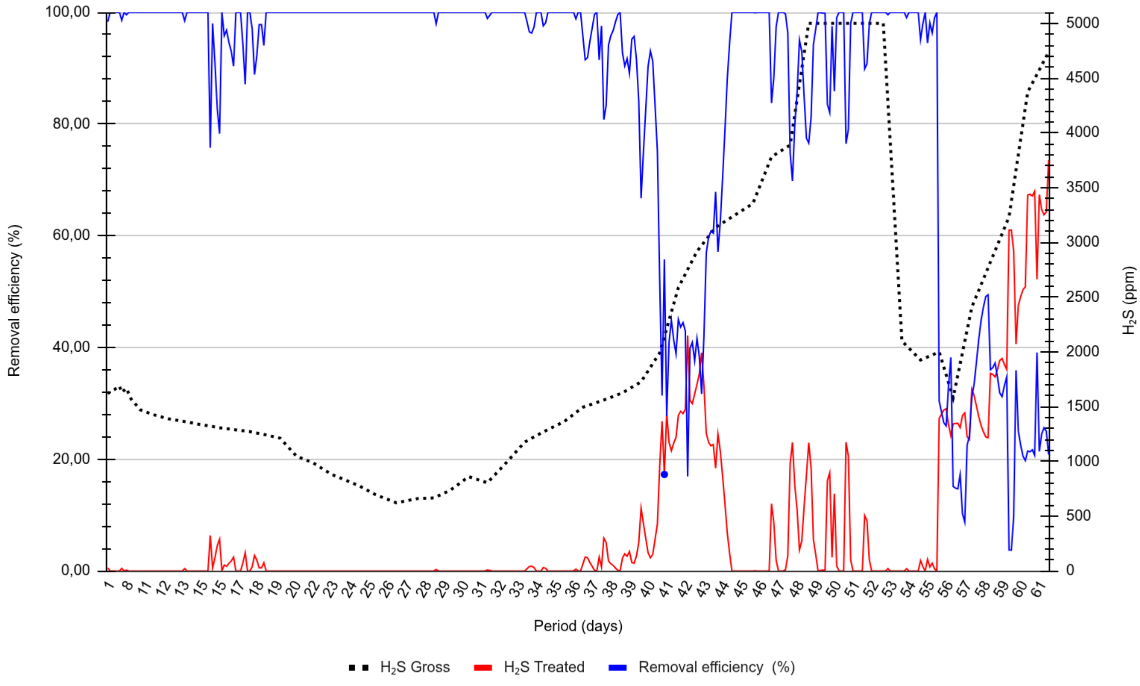
Figure 2 shows that, during the 62-day period, the composition of the raw biogas entering the system remained relatively stable with respect to its main components (CH_4 and CO_2), with average inlet concentrations of approximately 60% and 40%, respectively. It is important to note that the CH_4 and CO_2 outlet profiles exhibit an inversely proportional relationship, which is consistent with the stoichiometry of methanogenesis, where the variation in the production of one gas is compensated by the variation in the total volumetric composition of the other, a behavior similar to that reported by Wahid et al. (2019). Furthermore, a decrease in methane concentration at the outlet is observed from the 42nd to the 54th day of operation.

The concentration of raw H_2S showed significant variation over time, with a progressive increase, reaching peak values above 5,000 ppm after the 46th day of operation. In contrast, the H_2S concentration at the outlet remained close to zero until day 36, with fluctuations up to 324.16 ppm between days 15 and 18; after this period, the concentration at the outlet returned to zero. From day 37 onwards, an increase in the H_2S concentration at the outlet was again observed, reaching 1,440.16 ppm on day 40. This event indicated system saturation and the need for intervention, leading to the replacement of the digestate.

After the digestate replacement, the column showed fluctuations in the H_2S concentration at the outlet during the first few days, but subsequently re-established complete removal at various times. However, from day 56 until the end of the experiment (day 62), the system operated below optimal performance under the high input load, with H_2S concentrations at the outlet peaking at 3,749.85 ppm.

The efficiency of hydrogen sulfide (H_2S) removal was evaluated by comparing the gas concentrations at the inlet and outlet of the biofiltration system. Figure 3 shows the contaminant removal efficiency in the biofiltration column filled with pecan nutshells.

Figure 3. H_2S removal performance in the biofiltration column filled with pecan nutshell



Legend: H_2S removal performance in the biofiltration column filled with pecan nutshell (*Carya illinoinensis*). The figure shows the temporal variation of H_2S concentration in the raw biogas (H_2S Gross), the H_2S concentration after treatment (H_2S Treated), and the corresponding removal efficiency (%) during the experimental period. The inlet H_2S concentration exhibited significant fluctuations, reaching values close to 5,000 ppm, while the treated gas generally presented substantially lower concentrations during most of the operational period. These observations indicate that the biofiltration system operated with pecan nutshell packing was associated with substantial reductions in H_2S concentration under the tested conditions, while being subjected to variable inlet loads of hydrogen sulfide.

As shown in Figure 3, the column filled with pecan nutshell maintained an H_2S removal efficiency close to 100% until the 36th day of operation, with only minor fluctuations. During this period, the EC values followed the ILR trend, resulting in high removal efficiency and indicating stable operation of the biofilter. After this period, when the H_2S concentration at the inlet reached values close to 2,000 ppm, it was observed that the ILR exceeded the EC, which was associated with a decrease in removal efficiency.

In response to this condition, on the 40th day of operation, the column showed H_2S concentrations at the outlet exceeding 1,440 ppm, with a removal efficiency below 30%. Therefore, it was decided to replace the digestate while maintaining the previously cultivated biofilm in order to restore microbial activity and promote biofilter recovery.

After digestate replacement, between days 41 and 43 of the experiment, the column showed an average removal efficiency of 43.17%. From day 44 onwards, the system again showed satisfactory performance, with an average efficiency of 77.37%. Between days 45 and 47, it reached an average efficiency of 98.56%, operating stably until day 55, when the average removal efficiency was 94.83%. During this period, EC increased again and remained close to ILR for several days, indicating partial recovery of biofilter performance. Toward the end of the experiment, the increasing divergence between ILR and EC was accompanied by a reduction in removal efficiency, averaging 27.13% until the end of the experiment. In addition to these performance indicators, the column packed with pecan nutshells achieved an estimated H_2S removal of 142.3 g m^{-3} of treated biogas, highlighting the ability of the packing material to remove significant amounts of sulfur during the operational period. Biofilm development and the visual appearance of the column packed with pecan nutshell are shown in Figure 4.

Figure 4. Evolution of the biofiltration column filled with pecan nutshell

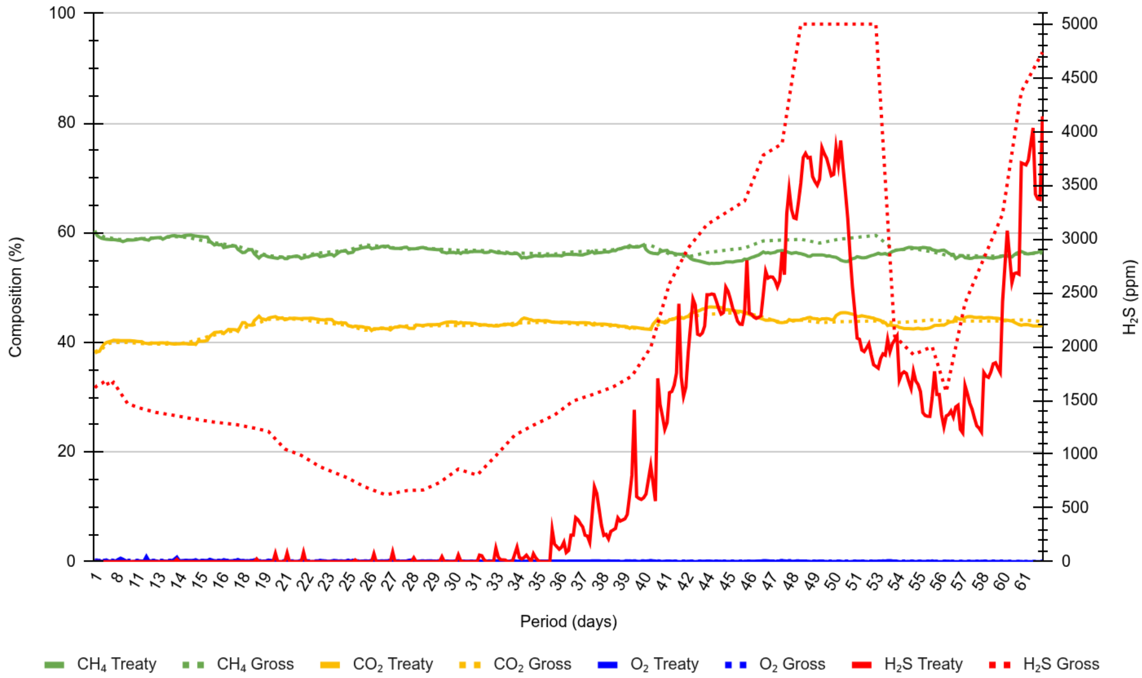
Legend: Images show the visual condition of the packing material at different stages of operation: (A) first day of operation, (B) 40th day of operation, and (C) 62nd and final day of operation. Progressive changes in the appearance of the packing material can be observed over time, including variations in surface coloration and the presence of deposits on the nutshell surface. These visual observations illustrate the physical changes occurring in the biofilter packing material during operation under biogas treatment conditions. Such changes may be associated with the interaction between the biogas stream, recirculated digestate, and the packing material over time, providing qualitative insight into the operational evolution of the biofiltration system.

Figure 4 illustrates the visual evolution of the biofiltration column filled with pecan nutshells throughout the experiment. Image A corresponds to the 1st day of operation, where the filter medium appears clean, maintaining the original characteristic color of the shell. In this initial phase, the system showed high H_2S removal efficiency, reaching 100%, as shown in Figures 1 and 2, indicating good adaptation of the biomass to the support material.

Image B corresponds to the 40th day of operation, when partial coverage by whitish elemental sulfur (S^0) is observed. Finally, image C, obtained on the last day of operation (62nd day), shows the filter medium largely covered by elemental sulfur, confirming the saturation of the system.

Next, the performance of the biofiltration column filled with loofah sponge as filling material was evaluated. Figure 5 shows the variation in biogas composition at the inlet and outlet of the system throughout the experimental period.

Figure 5. Changes in biogas constituents after treatment in the column filled with loofah sponge as packing material



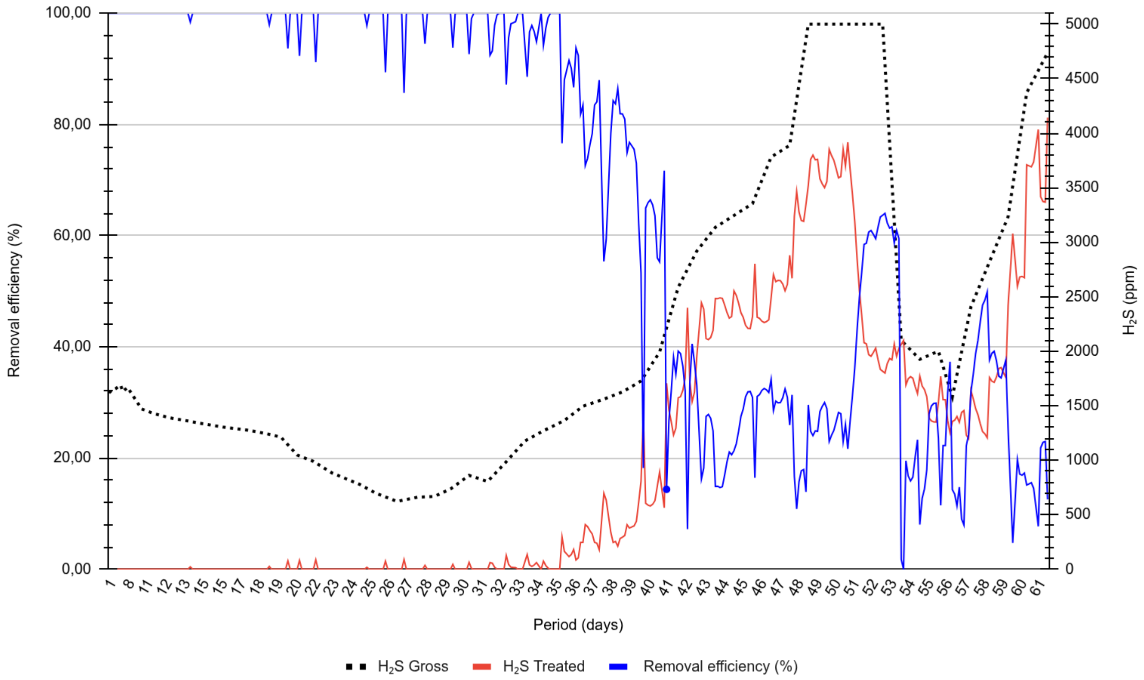
Legend: Changes in biogas composition after treatment in the column packed with loofah sponge (*Luffa cylindrica*). The figure shows the temporal evolution of CH_4, CO_2, and O_2 concentrations in the raw (gross) and treated biogas streams, expressed as percentage composition (left axis), together with the H_2S concentration in the raw and treated gas streams (ppm, right axis) during the experimental period. The dotted lines represent the raw biogas entering the system, while the solid lines correspond to the treated biogas after passing through the biofiltration column. The data illustrate the behavior of the biogas constituents during operation of the column packed with loofah sponge under variable inlet H_2S concentrations. The comparison between raw and treated streams allows the observation of changes in H_2S levels while the main energetic components of the biogas (CH_4 and CO_2) remained relatively stable throughout most of the operational period. These observations provide information about the operational conditions under which the loofah sponge packing material was evaluated for biogas treatment.

As shown in Figure 5, the CH_4 and CO_2 profiles in the treated biogas exhibit an inversely proportional behavior throughout the experimental period. In addition, a reduction in methane concentration at the outlet was observed between the 42nd and 54th day of operation, a pattern also identified in the column filled with pecan nutshells.

However, the H_2S concentration at the outlet remained close to zero until the 34th day, with small fluctuations during this period. From the 36th day onwards, an increase in H_2S at the outlet was observed, reaching values of up to 1,705.8 ppm on the 40th day, which motivated the replacement of the recirculated digestate in the column. However, unlike the result obtained with the pecan nutshells, the column filled with loofah sponge continued to show low performance after the intervention. The system did not show significant recovery, operating under fault conditions until the end of the experiment, on the 62nd day, during which time the H_2S concentrations at the outlet reached a peak of 4,143.31 ppm.

The evaluation of hydrogen sulfide (H_2S) removal efficiency is presented in Figure 6.

Figure 6. H_2S removal performance in the biofiltration column filled with loofah spong



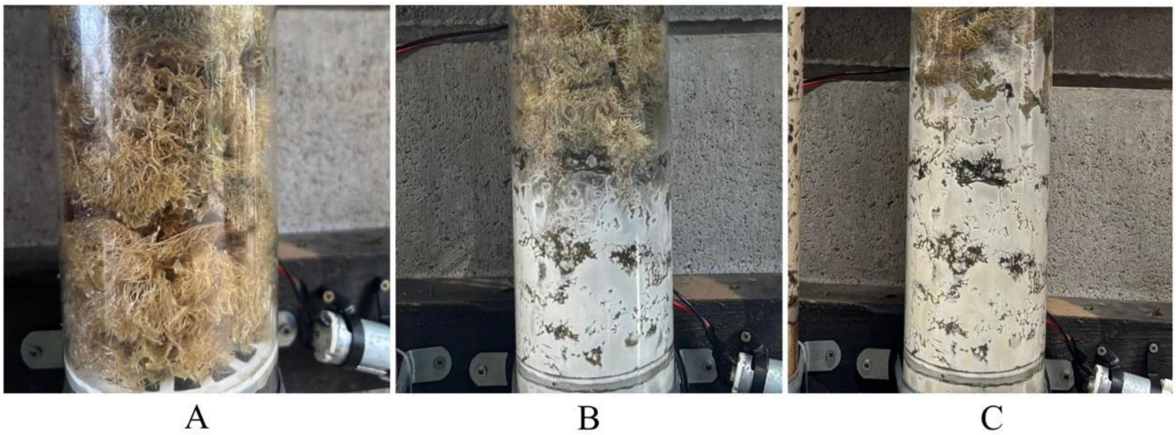
Legend: H_2S removal performance in the biofiltration column packed with loofah sponge (Luffa cylindrica) during the 62-day operational period. The dotted black line represents the H_2S concentration in the inlet biogas (H_2S gross), the solid red line corresponds to the H_2S concentration measured in the treated biogas at the outlet of the column (H_2S treated), and the blue line shows the resulting H_2S removal efficiency (%). The left vertical axis indicates removal efficiency, while the right vertical axis represents H_2S concentration (ppm). The figure shows the variation in removal efficiency as the inlet H_2S concentration increases throughout the experiment, including periods of high removal efficiency followed by performance decline under higher loading conditions.

As shown in Figure 6, the column packed with loofah sponge showed greater variability in performance after the acclimation phase. During the first 30 days of operation, the elimination capacity (EC) remained approximately equal to the inlet loading rate (ILR), indicating efficient H_2S removal. In this initial phase, the H_2S concentration at the outlet remained below 100 ppm, maintaining high removal efficiency. After this period, the H_2S concentration at the inlet reached values close to 2,000 ppm, and ILR began to exceed EC, leading to a decline in removal efficiency.

The point marked on the graph represents the day the digestate in the column was replaced, corresponding to the 40th day of operation. During this period, the column containing loofah sponge showed H_2S concentrations at the outlet above 1,700 ppm, with a removal efficiency below 20%. At this stage, the difference between the inlet loading rate (ILR) and the elimination capacity (EC) became more pronounced, indicating that the system was operating beyond its treatment capacity. After replacing the digestate, between days 40 and 43 of the experiment, the column showed an average removal efficiency of 29.05%. However, EC remained lower than ILR, indicating that the system did not recover its initial performance.

On day 44, the average removal efficiency was only 17.16%, with a slight increase to 33.10 ± 17.84% between days 48 and 53. After this period, until the end of the experiment, the column showed an average removal efficiency of 22.98%. During this phase, ILR continued to exceed EC, which was consistent with the low removal efficiencies observed. Unlike the biofilter packed with pecan nutshells, the replacement of the digestate did not restore the initial relationship between EC and ILR, indicating persistent limitations in the system performance. Based on a simplified cumulative mass balance calculated from the difference between inlet and outlet H_2S concentrations throughout the operational period, the column packed with loofah sponge removed approximately 98.9 g m^{-3} of H_2S from the treated biogas. The development of the biofilm and the visual aspect of the column containing the plant sponge can be observed in Figure 7.

Figure 7. Evolution of the biofiltration column filled with loofah sponge

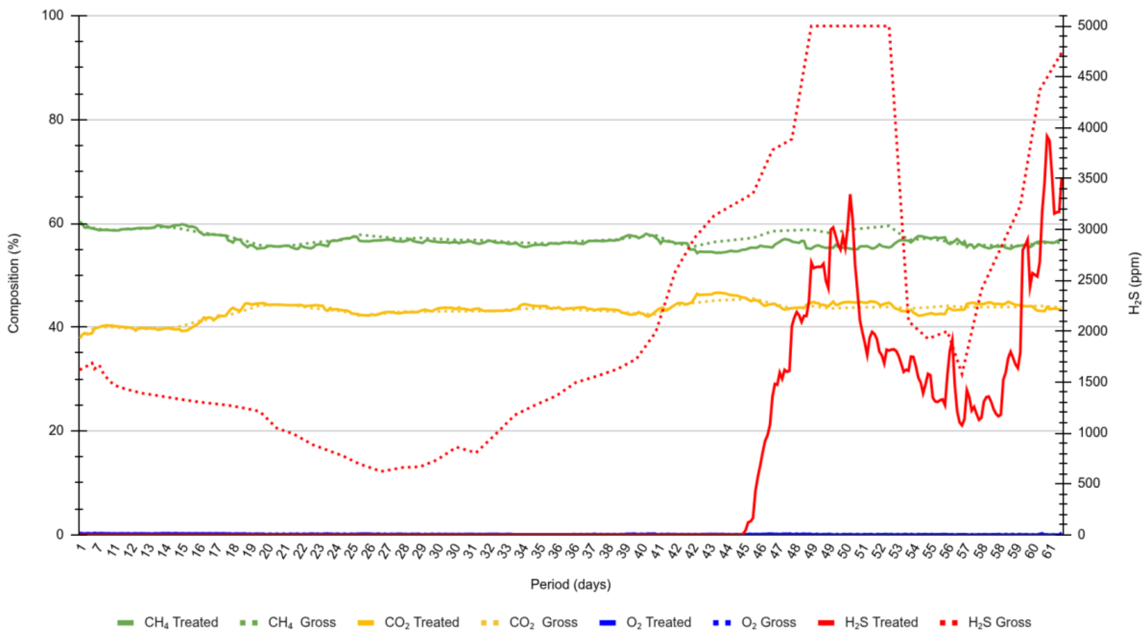


Legend: Evolution of the biofiltration column packed with loofah sponge (Luffa cylindrica) during the experimental operation. Images show the visual changes in the packing material throughout the biofiltration process: (A) initial stage of operation, with the clean loofah sponge before significant microbial colonization; (B) intermediate stage, where biofilm formation and initial deposits of sulfur compounds can be observed on the support medium; and (C) final stage of operation, characterized by greater accumulation of biomass and sulfur deposits along the column wall and packing material. These observations illustrate the progressive development of the microbial community responsible for hydrogen sulfide oxidation.

Figure 7 presents a comparison of the column filled with loofah sponge at different operational stages. Image A refers to the loofah at the beginning of the process, when it exhibited high removal efficiency. Image B shows the column on the 40th day of operation, when the digestate was replaced; at this point, sulfur accumulation can be seen at the base of the column. Finally, image C shows the column on the last day of operation, where a greater accumulation of S^0 is observed.

Similarly, the performance of the system filled with activated carbon, used as a reference because it is the conventional method for H_2S removal, was evaluated. This configuration allowed a comparison between the efficiency of the biological process and the physicochemical adsorption method. The inlet and outlet concentrations of the biogas constituents in this column are shown in Figure 8.

Figure 8. Changes in biogas constituents after treatment in the column filled with activated carbon



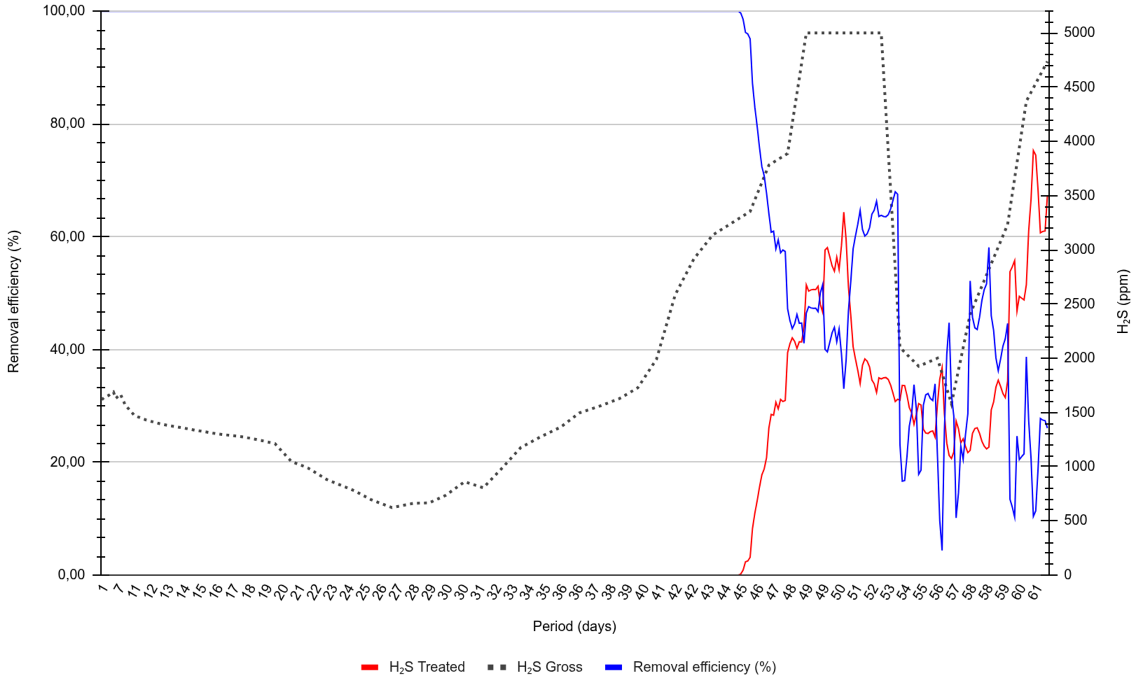
Legend: Changes in biogas constituents after treatment in the biofiltration column packed with activated carbon during the 62-day experimental period. Solid lines represent the composition of the treated biogas at the outlet of the column, while dotted lines correspond to the composition of the raw biogas at the inlet. Methane (CH_4), carbon dioxide (CO_2), and oxygen (O_2) concentrations are shown on the left axis (%), whereas hydrogen sulfide (H_2S)

concentrations are presented on the right axis (ppm). The figure shows that the main biogas components remain relatively stable during operation, while H_2S concentrations decrease significantly after treatment, demonstrating the effectiveness of the filtration process. This behavior is relevant to the research hypothesis because it indicates that the purification system can selectively remove H_2S without significantly altering the main energetic components of the biogas, allowing comparison between activated carbon and the tested agro-industrial biofilter media.

As shown in Figure 8, the activated carbon column operated with maximum H_2S removal until day 45. During this period, the H_2S concentration at the inlet varied from 622 to 3,245 ppm, demonstrating that the system maintained good performance even under different pollutant loads. At this stage, the elimination capacity (EC) remained equal to the inlet loading rate (ILR), indicating efficient H_2S adsorption. Furthermore, a decrease in methane concentration at the column outlet was observed between days 42 and 53, a behavior also observed in the biofiltration columns. During this period, the biogas feed to the system showed an average CH_4 concentration of 57.81%, while the average concentration at the outlet was 55.47%, representing an approximate loss of 4.05%. After the 45th day, the ILR began to exceed the EC, reflecting the progressive saturation of the adsorbent and the consequent reduction in removal efficiency. Furthermore, based on a simplified cumulative mass balance calculated from the difference between inlet and outlet H_2S concentrations throughout the operational period, the activated carbon column removed approximately 127.6 g m^{-3} of H_2S from the treated biogas.

The evaluation of hydrogen sulfide (H_2S) removal efficiency in the column filled with activated carbon is presented in Figure 9.

Figure 9. H_2S removal performance in the biofiltration column with activated carbon



Legend: H_2S removal performance in the biofiltration column packed with activated carbon during the 62-day operational period. The dotted black line represents the hydrogen sulfide concentration in the inlet biogas (H_2S gross), the solid red line shows the H_2S concentration in the treated biogas at the outlet (H_2S treated), and the blue line indicates the resulting H_2S removal efficiency (%). The left vertical axis corresponds to the removal efficiency, while the right vertical axis represents H_2S concentration (ppm). The figure illustrates the variation in removal efficiency as the inlet H_2S concentration increases throughout the experimental period, including periods of high efficiency followed by performance decline under higher loading conditions. These results provide a reference performance for a conventional adsorbent material and allow comparison with the tested agro-industrial residues.

Figure 9 shows a decline in efficiency beginning on the 45th day. From this point until the end of the experiment (62nd day), the average removal efficiency was 42.97%, while the inlet H_2S concentration reached peaks above 5,000 ppm, a factor that may be associated with the reduction in system performance. It is also observed that the activated carbon adsorption column did not exhibit any self-regeneration capacity, a behavior distinct from that observed in the biological method using the column filled with pecan nutshell.

Comparative Performance of the Three Columns

After presenting the results obtained for the three experimental columns, regarding the comparative performance between the biofiltration systems (pecan nutshell and loofah sponge) and the activated carbon adsorption system for hydrogen sulfide (H_2S) removal from biogas, this analysis aims to interpret the observed behaviors by relating them to operational conditions, characteristics of the packing materials, and evidence described in the literature.

The results indicate variation in the concentration of H_2S at the column inlet throughout the experiment. This variation is associated with changes in the composition of the biogas produced in the reactor, which was fed with different substrates that directly influence the H_2S content. This behavior is consistent with the observations reported by Das, Ravishankar and Lens (2022), who highlight that the composition of biogas depends on the type of substrate and the conditions of anaerobic digestion.

The initial behavior observed in the three columns, where the removal capacity (EC) equaled the input loading rate (ILR), indicates complete removal of H_2S by the system, which is consistent with removal efficiency profiles. As the experiment progressed, the divergence between EC and ILR showed that the system was being subjected to progressively higher pollutant loads than its removal capacity. When the difference between these values became more pronounced, it suggested that the system was approaching its operational limits, leading to saturation of the packing material. This behavior has been widely reported in biofilters and biotrickling filters for H_2S and volatile organic compounds (VOCs) in several studies (Reddy et al., 2019; Das et al., 2019; Wu et al., 2020; Malakar et al., 2018).

Based on a simplified mass balance calculated from the difference between inlet and outlet H_2S concentrations throughout the operational period, the cumulative H_2S removal reached approximately 142.3, 98.9, and 127.6 g m^{-3} for the columns packed with pecan nutshells, loofah sponge, and activated carbon, respectively. In biological desulphurisation systems, the removed H_2S is typically oxidised by sulphur-oxidising microorganisms into elemental sulphur (S^0) and sulphate (SO_4^{2-}), which may accumulate in the packing material or remain dissolved in the liquid phase of the system. This behaviour has been widely described in biofilters and biotrickling filters treating sulphide-containing gases, where the progressive accumulation of sulphur compounds may eventually contribute to reductions in system performance (Jaber et al., 2016; Zhang et al., 2022b; Huertas et al., 2020). In contrast, in adsorption systems using activated carbon, H_2S removal occurs mainly through adsorption and surface oxidation reactions, leading to the formation and retention of sulphur species within the porous structure of the adsorbent material, as discussed by Yang et al. (2020) and Tuerhong and Kuerban (2022).

The slight decrease in methane concentration observed at the system outlet may be associated with the dilution of biogas by small amounts of air entering the biofilter, corroborated by the increase in O_2 at the outlet (0.45 to 0.71%). This phenomenon was previously reported by Becker et al. (2024) and Nhut et al. (2020), who relate the dilution of CH_4 to the increase in H_2S load and air infiltration, influencing the final composition of the biogas.

When subjected to high H_2S inlet loads (≈2,000 ppm), the biofiltration columns showed reduced removal efficiency. This behaviour is associated with S^0 accumulation and microbial limitations under extreme conditions. Similar results were reported by Vela-Aparicio et al. (2022), who observed that increasing H_2S and NH_3 inlet loads led to a gradual decline in biofilter performance due to system overload and reduced microbial activity.

After the digestate replacement, the column filled with pecan nutshell showed a temporary decrease in efficiency, gradually recovering in the following days. Similar behavior was reported by Lenis et al. (2023), who observed transient efficiency losses after digestate replacement in a pilot-scale trickling biofilter, followed by progressive recovery associated with microbial readaptation. Despite operational differences, both systems depend on the activity of sulfur-oxidizing microorganisms (Qiu and Deshusses, 2017). This behaviour may therefore be related to the re-establishment of biological activity within the packing material. However, since no microbiological or sulphur balance analyses were conducted, this interpretation should be considered indicative rather than conclusive.

Although fluctuations occurred after the 40th day, even with inlet H_2S concentrations exceeding 5,000 ppm, the pecan nutshell column again achieved removal efficiencies close to 100%, demonstrating regenerative capacity. This behavior, rarely reported in the literature, reinforces the potential of this material as a sustainable biological medium, despite being subject to climatic limitations and prolonged operational stress.

In contrast, the column filled with loofah sponge maintained maximum efficiency for 34 days but showed no regeneration after the digestate replacement. It is believed that the system overload (H_2S > 5,000 ppm) caused irreversible pore saturation and microbial inhibition due to prolonged exposure to toxic concentrations. Because the material has a highly porous structure and high moisture retention, it favours internal sulfur deposition, which reduces gas diffusion and compromises microbiological activity (Woo et al., 2025).

Overall, both biological columns showed high initial performance, with average efficiencies of 99.23% (loofah sponge) and 98.58% (pecan nutshell). These results are consistent with those reported by Aita et al. (2016), Promnuan et al. (2023), and Vela-Aparicio et al. (2022), who observed efficiencies above 90% in biofilters operating for more than 30 days, reinforcing the feasibility of biofiltration for pilot-scale biogas desulfurization.

Visual inspection of the biofilm revealed the progressive accumulation of elemental sulfur (S^0) on the surfaces of the packing material, a phenomenon also reported by Jaber et al. (2017). On the 40th day, this accumulation coincided with the reduction in efficiency, indicating diffusion blockage and loss of biological activity. By the 62nd day, the material was extensively covered with sulfur, confirming system saturation, a behaviour similarly described by Vu et al. (2022).

Despite this, the environmental reuse of the packing materials is feasible, as the accumulated sulfur can be utilized in composting or as a soil conditioner, serving as a supplemental source of plant nutrients (Narayan et al., 2023). This possibility highlights the environmental benefits of using organic materials in biofilters compared to synthetic supports.

Activated carbon showed satisfactory initial performance, reaching maximum efficiency on the 45th day due to its high surface area and microporous structure (Bonatto et al., 2024). Subsequently, the average efficiency decreased to 42.97% as a result of saturation of the adsorption sites. According to Choleva et al. (2023), H_2S concentrations above 3,000 ppm accelerate saturation, requiring regeneration. However, common regeneration methods have sustainability limitations due to high energy demand and the generation of toxic byproducts, making landfill disposal a more viable option (Rehman et al., 2025; Choleva et al., 2023).

According to ABNT NBR 10.004:2004 (ABNT, 2004), activated carbon used in biogas desulfurization is classified as Class I (hazardous) waste due to the presence of sulfur compounds. Therefore, its disposal must take place in licensed industrial landfills, in accordance with the Brazilian National Solid Waste Policy (Law No. 12.305/2010) and its regulation (Decree No. 10.936/2022) which define principles and instruments for the environmentally sound management of hazardous waste. (Brasil, 2010; Brasil, 2022).

Between days 46 and 57, a decrease in methane concentration was observed at the column outlet. While the incoming biogas contained an average of 57.81% CH_4, the average at the outlet was 55.47%, corresponding to an approximate loss of 4.05%. This behavior is consistent with molecular modeling studies that show that CH_4 adsorption on activated carbon occurs mainly in micropores through weak and reversible Van der Waals interactions (Rugarabamu, 2025). The low affinity and reversible adsorption of CH_4 explains the small losses (~4%) observed experimentally in this study. Furthermore, during this period, H_2S concentrations at the inlet exceeded 5,000 ppm. Due to its higher polarity and adsorption energy, H_2S tends to occupy higher energy sites, altering the internal equilibrium of the bed and causing temporary retention of CH_4. Similar results were reported by Gao et al. (2024), who demonstrated that, although H_2S has significantly higher adsorption energy than CH_4, it exerts competitive effects on CH_4 and CO_2 in occupying active sites, resulting in partial inhibition of methane passage.

The column ceased to operate effectively after the 46th day, demonstrating the limited lifespan of the material under high H_2S loads. This behavior is widely described in the literature and was reported by Cavaler et al. (2022), who describe this process as semi-continuous, in which the progressive saturation of the adsorbent reduces the lifespan and makes regeneration economically and environmentally unfeasible due to the high energy demand and waste generation, thus limiting large-scale application.

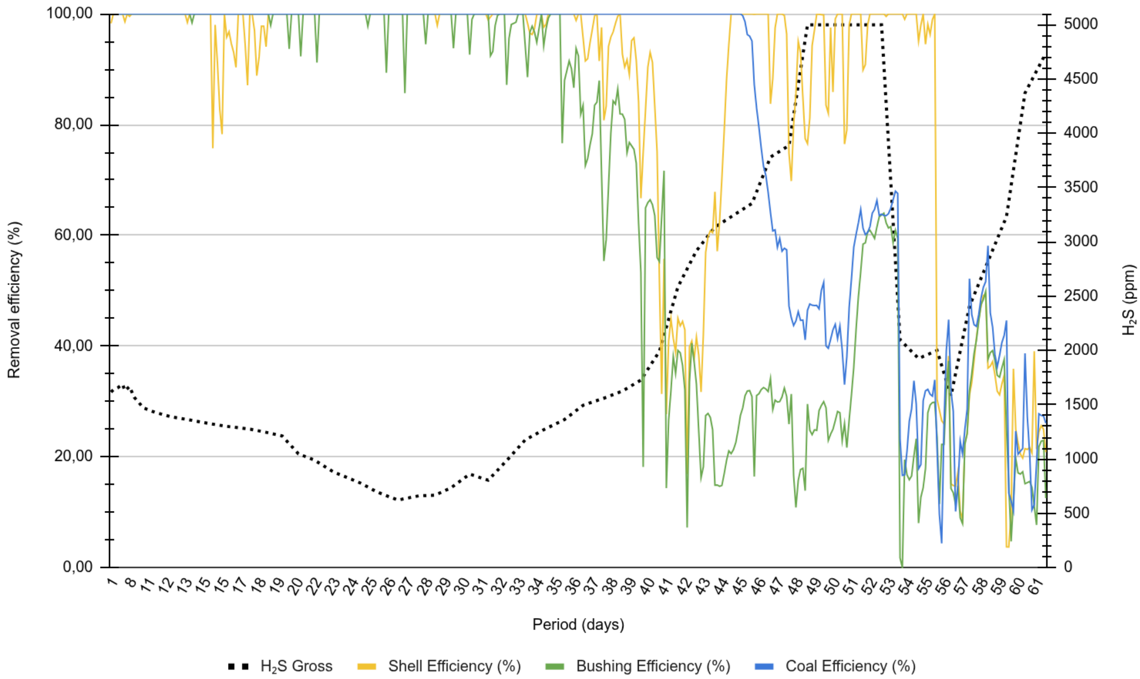
Statistical Analysis of the Data

Based on the results obtained, statistical analysis was applied to the experimental data. Initially, the non-parametric Kruskal-Wallis test was used, appropriate for comparing three or more independent groups when the data do not follow a normal distribution. The test presented a test statistic $H(\chi^2)$ of 48.91 and a tie-corrected value (Hc) of 58.59, with $p = 1.898 \times 10^{-13}$. This result, well below the significance level of 0.05, indicates a statistically significant difference between the groups and leads to the rejection of the null hypothesis (Almeida et al., 2022).

Pairwise comparisons were performed using the Mann-Whitney test. The pecan shell column differed significantly from the column filled with loofah sponge ($p = 7.23 \times 10^{-11}$), but did not show a significant difference compared to activated charcoal ($p = 0.3906$). In contrast, a significant difference was observed between the loofah sponge and the activated charcoal columns ($p = 8.68 \times 10^{-11}$).

These results demonstrate that the pecan nutshell exhibited statistically similar behavior to activated carbon, highlighting its potential as an alternative and low-cost material for biogas desulfurization. Figure 10 graphically illustrates these results, supporting the statistical analysis and showing the differences in removal efficiencies obtained for each material.

Figure 10. H_2S removal efficiency using different filtering materials



Legend: H_2S removal efficiency using different filtering materials during the 62-day experimental period. The dotted black line represents the hydrogen sulfide concentration in the inlet biogas (H_2S gross). The colored lines indicate the removal efficiency achieved by the different filtering materials: pecan nutshell (shell efficiency), loofah sponge (Luffa cylindrica) (bushing efficiency), and activated carbon (coal efficiency). The left vertical axis corresponds to the H_2S removal efficiency (%), while the right vertical axis represents the inlet H_2S concentration (ppm). The figure shows the variation in removal performance of each filtering medium as the inlet H_2S loading increases over time, highlighting differences in stability and efficiency among the materials.

Figure 10 illustrates the temporal variation in H_2S removal efficiency for the three evaluated columns and confirms the operational behavior previously discussed for each system. The column packed with pecan nutshells showed higher removal efficiency and greater operational stability when compared with the loofah sponge column, with the system recovering its performance after the replacement of the digestate. In contrast, the loofah sponge column exhibited lower overall efficiency and reduced stability over time. The performance of the activated carbon column also followed the expected behavior for adsorption systems, with a gradual decrease in removal efficiency during operation until saturation of the adsorbent material was reached.

Although activated carbon is widely used in biogas plants due to its high adsorption efficiency, its application is associated with high operating costs. Several studies, as reported by Estrada et al. (2012), Atelge et al. (2021) and Sosa et al. (2022), report that frequent replacement of activated carbon is often preferred, since regeneration after sulfur saturation requires high energy and water demand from the regeneration processes. These limitations highlight biological methods as promising alternatives for more sustainable and economically viable biogas desulfurization.

Furthermore, according to the results obtained, the lower cumulative volume of biogas treated by the loofah sponge column, mainly associated with operational interruptions during the experimental period due to pump replacement, did not compromise the comparative analysis of the systems. Under these comparable operating conditions, the loofah sponge column showed lower overall performance and operational stability than the pecan nutshell column, indicating that the characteristics of the packing material played an important role in the long-term efficiency of the biofiltration process.

Although the experiments were conducted at pilot scale, which provides a more realistic assessment of biofilter performance compared with laboratory-scale systems, some limitations should be considered when extrapolating the results to full-scale applications. Operational conditions in industrial biogas plants may vary significantly due to fluctuations in gas composition, temperature, and moisture, which can influence biofilter performance. In addition, parameters such as microbial community dynamics, sulfur accumulation within the packing material, and pressure drop across the bed were not investigated in detail in the present study. These factors may affect long-term stability and operational efficiency. Nevertheless, the controlled experimental conditions allow a reliable comparison between the tested materials, highlighting the potential of agricultural residues such as pecan nutshells and loofah sponge as alternative biofilter media. The concept of using locally available biomass residues for biogas desulfurization may be transferable to other regions, provided that suitable materials with similar structural properties are available.

Cost Competitiveness of Biological Methods for Biogas Desulfurization

Conventional technologies for hydrogen sulfide removal from biogas include adsorption using activated carbon, chemical scrubbing, and iron-based adsorbents (Georgiadis et al., 2020; Secco et al., 2023). These technologies are widely applied due to their high removal efficiency and operational reliability. However, they may involve relatively high operational costs associated with chemical consumption, energy demand, or the periodic replacement of saturated adsorbent materials.

In recent years, biological methods for H_2S removal, such as biofilters, bioscrubbers, and biotrickling filters, have attracted increasing attention as more sustainable and potentially cost-effective alternatives (Kasulla et al., 2021; Rouhollahi et al., 2024; Severi et al., 2025). These systems generally operate under milder conditions and require fewer chemical inputs, which may reduce operational costs and environmental impacts compared with conventional physicochemical technologies.

The use of low-cost filler materials derived from agricultural waste can further improve the economic viability of biological desulfurization systems, as they represent a low-cost resource that can be valued in environmental technologies. In this context, the use of locally available biomass waste as a filter medium can contribute to reducing treatment costs while simultaneously promoting the sustainable management of agricultural waste.

Implications and Future Research Directions

The overall behavior observed in the biofiltration columns can be explained by the interaction between biological sulfide oxidation and the structural characteristics of the packing materials. Hydrogen sulfide removal in biofilters is mainly driven by sulfur-oxidizing microorganisms that develop within the biofilm attached to the filter medium, converting H_2S into elemental sulfur and sulfate. The porous lignocellulosic structure of agro-industrial residues such as loofah sponge and pecan nutshell provides a favorable environment for microbial colonization by offering high surface area, adequate moisture retention, and sufficient void space for gas transfer. The results indicate three main findings. First, both agro-industrial materials demonstrated high initial removal efficiencies, confirming that natural lignocellulosic matrices can effectively support biological H_2S oxidation. Second, the progressive accumulation of sulfur and biomass within the packing material under increasing inlet H_2S loading conditions likely limited mass transfer and contributed to the observed decline in removal efficiency over time. Third, the comparative analysis with activated carbon showed that agro-industrial residues can achieve removal efficiencies within the same order of magnitude while offering potential advantages in terms of sustainability and resource valorization. Together, these findings highlight the potential of locally available agro-industrial residues as viable and environmentally sustainable alternatives for biofiltration-based biogas desulfurization.

These mechanisms and findings provide important insights for the design and optimization of low-cost biofiltration systems for biogas purification. However, further improvements in the overall efficiency of biogas systems may also depend on upstream process optimization. In particular, future research may explore the integration of gas purification technologies with optimized biomass pretreatment strategies during anaerobic digestion. Pretreatment methods such as steam explosion or lignocellulosic biomass conditioning have been reported to enhance substrate accessibility and improve biogas production performance, which may indirectly influence gas composition and the operational conditions of downstream purification systems (Maroušek, 2012; Maroušek, 2013a). Investigating the combined effects of feedstock pretreatment, digestion performance, and gas purification efficiency may therefore represent a promising direction for the optimization of integrated biogas production systems.

In addition, recent advances in artificial intelligence (AI) and data-driven modelling approaches may provide new opportunities to simulate complex industrial systems and evaluate the techno-economic impacts of emerging technologies under different economic conditions. Such tools, including AI-driven simulation and digital twin environments, could support future studies aimed at optimizing integrated biogas production and purification systems at industrial scale (Kliestik et al., 2024; Zvarikova, Gajanova, Horak, 2024b).

Conclusion

The results confirm the research hypothesis that lignocellulosic agro-industrial residues can act as effective support media for biological hydrogen sulfide oxidation in biofiltration systems. Materials such as loofah sponge and pecan nutshell demonstrated the capacity to sustain microbial activity responsible for H_2S removal, achieving performances comparable to conventional activated carbon under similar operating conditions.

Beyond the specific experimental outcomes, the study highlights the broader potential of agricultural residues as functional materials for gas purification technologies. Their natural availability, low cost, and compatibility with biological processes indicate that these materials represent an environmentally sustainable alternative for biogas desulfurization. The concept investigated in this study contributes to expanding current knowledge on the use of natural lignocellulosic materials in biological gas treatment systems and supports the development of more resource-efficient approaches within renewable energy and circular bioeconomy frameworks. Further studies are needed to evaluate operational limits, regeneration capacity, and microbial recovery mechanisms under overload conditions.

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