



## Removal of hydrogen sulfide in biogas from wastewater treatment sludge by real-scale biotrickling filtration desulfurization process

Sema Arıman <sup>a,\*</sup> and Serdar Koyuncu <sup>b</sup>

<sup>a</sup> Department of Meteorological Engineering, Samsun University, 55420 Ondokuz Mayıs/Samsun, Turkey

<sup>b</sup> Konya Metropolitan Municipality, Konya, Turkey

\*Corresponding author. E-mail: sema.ariman@samsun.edu.tr

 SA, 0000-0001-7201-9243; SK, 0000-0002-4757-4680

### ABSTRACT

The biological removal of hydrogen sulfide in biogas is an increasingly adopted alternative to conventional physicochemical processes because of its economic and environmental benefits. In this study, a real-scale biotrickling filtration (BTF) process packed with polypropylene carrier was used to investigate the removal of high concentrations of H<sub>2</sub>S in biogas from an anaerobic digester. The results show that H<sub>2</sub>S in biogas was entirely removed under different inlet concentrations of H<sub>2</sub>S from 2,923 to 4,400 ppmv, and the elimination capacity of H<sub>2</sub>S in the filter achieved about 52.71 g H<sub>2</sub>S/m<sup>3</sup>/h. In addition, the process efficiency was found to be independent of the inlet H<sub>2</sub>S concentration. The removal of high concentrations of H<sub>2</sub>S in biogas was accomplished by the BTF process with SOB (*Acidithiobacillus thiooxidans*), which is active in the acidic environment (pH 1.5–3.5). In addition, the process efficiency was found to be independent of the inlet H<sub>2</sub>S concentration. Consequently, a real-scale BTF process allowed the potential use of biogas and the recovery of elemental sulfur resources simultaneously.

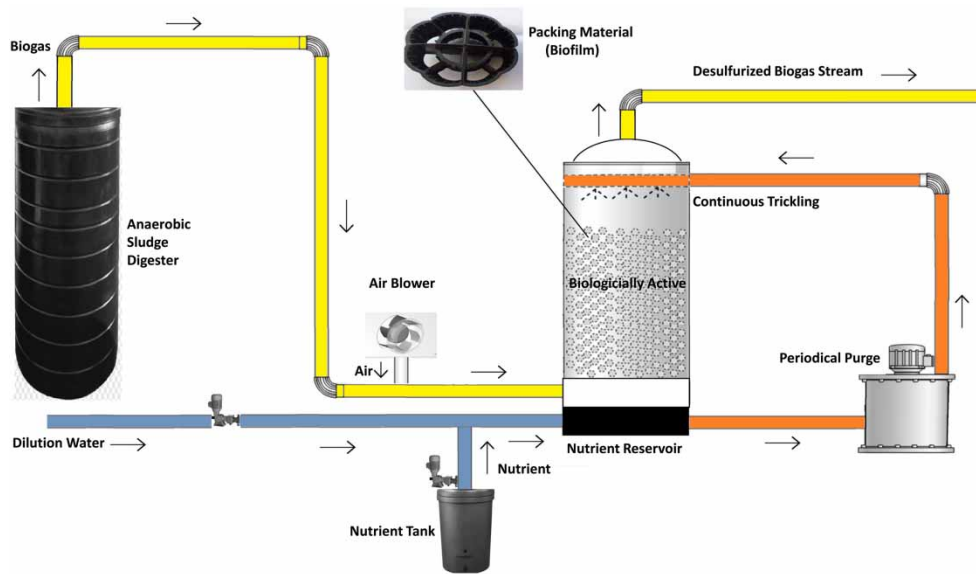
**Key words:** anaerobic sludge digester, biogas, biological desulfurization, biotrickling filter, removal of hydrogen sulphide

### HIGHLIGHTS

- H<sub>2</sub>S in biogas produced from WWT sludge was removed by a real-scale BTF plant.
- The process efficiency was found to be independent of inlet H<sub>2</sub>S concentration.
- The elimination capacity of the system reached a maximum of value.
- The real-scale BTF unit was found to provide sufficient removal efficiency for H<sub>2</sub>S in the biogas.

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## GRAPHICAL ABSTRACT



## 1. INTRODUCTION

Anaerobic digestion (AD) is commonly used in the treatment of organic waste, such as agricultural waste, sewage sludge and organic forms of municipal solid waste. During this process, approximately 95% of the organic matter and 95% of the energy present in the substrate are contained in the biogas (Guerrero *et al.* 2015). The most important ingredients in the biogas produced by the digestion of treatment sludges through anaerobic processes are: 60–70% methane (CH<sub>4</sub>), 30–35% carbon dioxide (CO<sub>2</sub>), 1–2% hydrogen sulfide (H<sub>2</sub>S) and 0.3–3% other gases (Al Mamun & Shuichi Torii 2015). The components of biogas can vary depending on the used substrate for their production (Rasi *et al.* 2007). If the substrate used for biogas production contains sulfur, the formation of H<sub>2</sub>S is inevitable (Chaiprapat *et al.* 2015; Dumont 2015).

The concentration of H<sub>2</sub>S in biogas varies from a few hundred to ten thousand ppm depending on the amount of bioavailable sulfur compounds in the feedstock and the outcome of the competition among sulfate-reducing bacteria, acetogens and methanogens for the organic substrates (Stams *et al.* 2005). A presence of a high concentration of H<sub>2</sub>S causes corrosion on equipment and increases the maintenance costs. In particular, due to the corrosive effect on the gas engines, engine life is shortened, the service/maintenance costs increase, and the conversion of biogas to electricity decreases (Rasi *et al.* 2011). For this reason, H<sub>2</sub>S must be removed from the produced biogas.

Actually, H<sub>2</sub>S is produced under anaerobic conditions because sulfate (SO<sub>4</sub><sup>2-</sup>) acts as an electron acceptor while organic compounds are decomposed biologically (Yan *et al.* 2018). H<sub>2</sub>S is produced by anaerobic degradation of sulfur-containing compounds (mainly proteins) and reduction of anionic species (especially SO<sub>4</sub><sup>2-</sup>) in the feedstock of the digester (Ramos *et al.* 2013). Kuenen (1975) proposed the mechanism of H<sub>2</sub>S removal that occurs through a series of physico-chemical processes and biological reactions, summarized by Equations (1)–(4) below.

- (a) H<sub>2</sub>S<sub>(g)</sub> dissolution in water



- (b) H<sub>2</sub>S biological oxidation to SO<sub>4</sub><sup>2-</sup>



(c) H<sub>2</sub>S biological oxidation to S<sub>(s)</sub>



(d) S<sub>(s)</sub> biological oxidation to SO<sub>4</sub><sup>2-</sup>



For the removal of H<sub>2</sub>S in biogas, solid phase adsorption, liquid phase absorption, membrane separation, chemical, biological, and thermal methods are used (Rasi *et al.* 2011; Lin *et al.* 2013; Angelidaki *et al.* 2018; Peluso *et al.* 2019). The biological desulfurization of biogas can be performed in additional units mainly using biofilters and biotrickling filters during digestion process and by applying microaerobic conditions directly in anaerobic digestors (Ramos *et al.* 2013). This biological desulfurization treatment method for the cleaning of contaminated biogas is a relatively new trend and is of great interest. On the other hand other gas desulfurization methods have high operation costs and produce wastes that must be disposed of. The biological desulfurization method is economically more advantageous and more environment friendly than the other methods. In addition, this treatment method is also more useful because the gas stream contains biodegradable or biconvertible compounds (Tomas *et al.* 2009).

In bioreactor systems, the H<sub>2</sub>S is dissolved into the film, followed by the oxidation of H<sub>2</sub>S by sulfur-oxidizing bacteria (SOB) with oxygen in the liquid phase (Duan *et al.* 2006; Kobayashi *et al.* 2012; Nhut *et al.* 2020). High elimination capacity (EC) and stability in the presence of severe operating conditions are required for bioreactor systems to apply biological methods for the removal of hydrogen sulfide in a biogas stream. A large number of biodesulfurization processes are present, such as the biofilter processes (Montebello *et al.* 2014; Ramos & Fdz-Polanco 2014; Rodriguez *et al.* 2014), the bioscrubber processes (Valero *et al.* 2019), and the process using headspace of the digestors (headspace process) (Ramos & Fdz-Polanco 2012). The differences between these systems are the phase of the biomass (suspended or fixed), the state of the liquid phase (flowing or stationary) and the state of having or not having a carrier material (Ramirez *et al.* 2009). In real-scale biotrickling filtration (BTF), the waste airstream passes through a bed that is packed and has pollutant-degrading organisms immobilized in the form of biofilms. The contaminant either moves from the gas phase to the liquid phase and then to the biofilm, or it moves directly from the gas phase to the biofilm, where it is biologically degraded to harmless compounds (Gabriel & Deshusses 2003). Its major advantages are having low operation cost, requiring low-energy and chemicals and having high removal efficiencies (REs), mostly above 99% (Aita *et al.* 2016). Recently, BTFs have been widely applied to the treatment of H<sub>2</sub>S on both laboratory and industrial scales (Nhut *et al.* 2020; Khoshnevisan *et al.* 2018; Khanongnuch *et al.* 2019). However, limited studies are available regarding the removal of H<sub>2</sub>S in biogas from wastewater treatment (WWT) sludge by real-scale BTF process at acidic pH for highly loaded H<sub>2</sub>S gas streams in a real scale BTF. Thus, in this study, a real-scale BTF was used to investigate the removal of high concentrations of H<sub>2</sub>S contained in biogas from an anaerobic digester.

## 2. MATERIALS AND METHODS

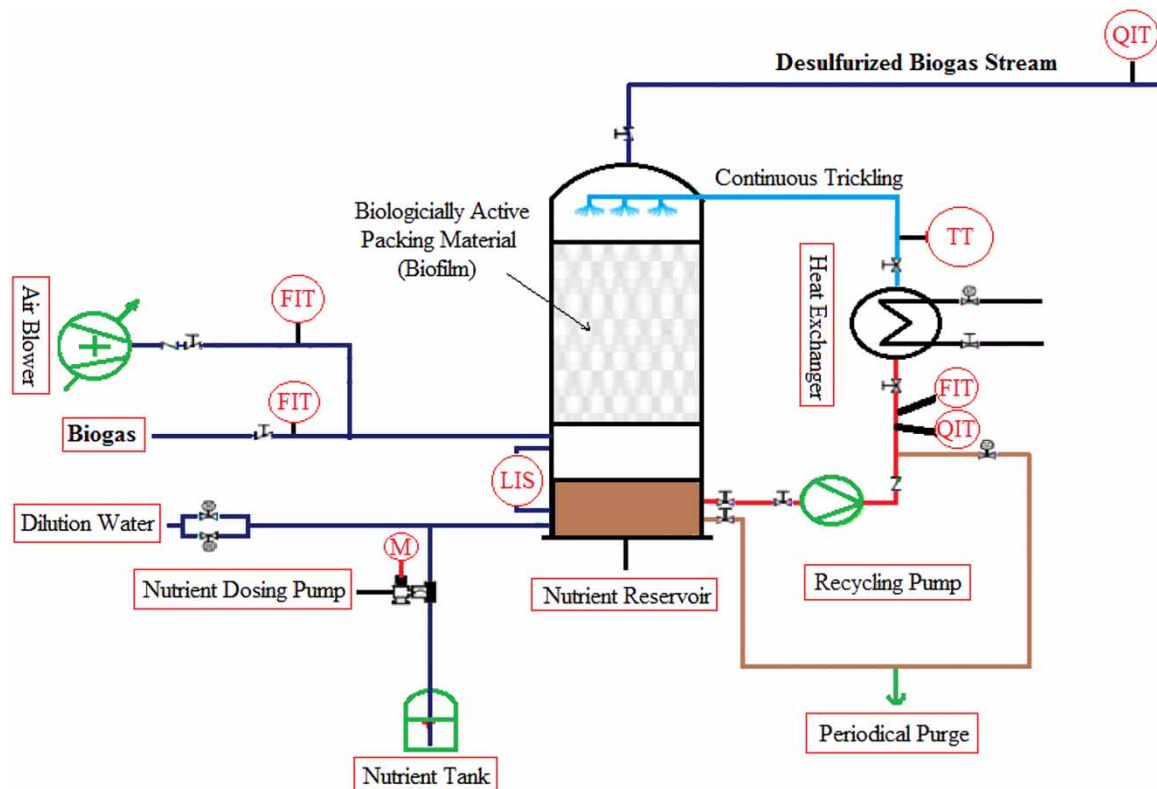
### 2.1. Real-scale BTF process

This study was performed at Konya advanced biological urban wastewater treatment plant (WWTP) with an equivalent population of 1,000,000 and 200,000 m<sup>3</sup>/day flow rate. Real-scale BTF was used for the purification of H<sub>2</sub>S in the biogas collected at the AD output used for sludge stabilization. In this process, the H<sub>2</sub>S is removed from biogas, the biogas is cooled to condense the moisture in it and the condensate is discarded. Biogas collected from anaerobic sludge digestors are transferred to the feeding chamber at the bottom of the closed tower where the BTF unit is located. The biogas moves from the bottom to the top and in the tower that contains layers of polypropylene media filling circles (Table 1) where desulfurization occurs. A complexed culture of SOB dominated by *Acidithiobacillus thiooxidans* acclimated from activated sludge was used as the bacterial strain and a biofilm was formed. To supply the substrate (nutrients) for the SOB, treated wastewater was fed to the feeding chamber at the bottom of the tower. The feeding water was heated in heat exchangers before adjusting the temperature to 35–36 °C, and it was sprayed onto the media material from the top of the tower. Some authors reported that for similar sulfide-oxidizing microorganisms, an optimum growth temperature at around 30 °C (Ravichandra

**Table 1** | BTP media material characteristics

Material	Polypropylene (PP)
Shape	Perforated rings
Size (D1-D2/L) (mm)	100–90/50–35
Colour	Black
Porosity	92%
Specific surface area ( $\text{m}^2 \text{m}^{-3}$ )	140
Weight (g)	39
Pieces per unit volume ( $\text{pieces m}^{-3}$ )	2,080
Density ( $\text{kg m}^{-3}$ )	80

*et al.* 2006; Sanchez *et al.* 2014). Yang & Allen (1994) reported 100% removal efficiency at 30–50 degrees Celsius but only 20% at temperatures below 10 °C. At the entrance point of the desulfurization unit 1.5–3.5% air was added to the biogas. In this process,  $\text{O}_2/\text{H}_2\text{S}$  ratio was 2/1. The end product of oxidation, sulfate (high  $\text{O}_2/\text{H}_2\text{S}$  ratio in biofilm) or elemental sulfur (low  $\text{O}_2/\text{H}_2\text{S}$  ratio), should vary depending on the availability of oxygen for microorganisms in the bioreactor. If the oxygen is more than the stoichiometric requirement, the formation of elemental sulfur decreases (Buisman *et al.* 1991). The treated biogas was passed through cooling units to decrease the temperature and moisture before it was fed into the gas conversion engines. The filtrate collected at the bottom of the unit was discharged into the sulphur fertiliser tank. The sludge layer accumulated on the polypropylene material was removed from the system by back-washing. The flow diagram of the BTF process is given in Figure 1.

**Figure 1** | Real-scale BTF process.

## 2.2. Real-scale BTF operational conditions

Real-scale BTF design criteria are given in Table 2. The biogas contains 65% methane ( $\text{CH}_4$ ), 34% carbon dioxide ( $\text{CO}_2$ ), 1%  $\text{H}_2\text{S}$ , and other gases. The process was designed for the average biogas to be 30 °C and the dilution

**Table 2** | Biological desulfurization process design criteria

Parameter	Unit	Value
Flow rate	Nm <sup>3</sup> /hour	1,500
Inlet H <sub>2</sub> S concentration	ppmv	5,000
Outlet H <sub>2</sub> S concentration	ppmv	200
Inlet biogas temperature	°C	30
Outlet biogas temperature	°C	5
Biogas pressure	mbar	15
Methane (CH <sub>4</sub> )	%	62
Carbon dioxide (CO <sub>2</sub> )	%	31
Nitrogen (N <sub>2</sub> )	%	5
Oxygen (O <sub>2</sub> )	%	2

water average temperature to be 15 °C. The H<sub>2</sub>S content in the BTF system is approximately 3,600 pmv. Air was supplied to the BTF process by blowers at 1% of the biogas flow rate. The inlet biogas temperature is 35–37 °C, and 15–20 of mbar pressure is given to the system. Treated wastewater was used as feedwater, and its flow rate is 6–8 tons/day. The feed water was sprayed into the tower at 14–16 m<sup>3</sup>/h flow rates, continuously circulating. The temperature of this water is 35–36 °C. Hot water was supplied with heat exchangers. The pH value in the BTF system was 1–2. The process discharged 6–8 m<sup>3</sup>/day of sulfurous water after treatment.

### 2.3. Monitoring and analytical methods

The pH is an important parameter affecting the process efficiency and the system was operated in the pH range of 1.5–3.5. The optimum pH should be in the range of 2–3.5 for activities of sulfide-oxidizing *A. thiooxidans* bacteria (Syed *et al.* 2006; Montebello 2013; Rodriguez *et al.* 2014). Kim & Deshusses (2005) reported that the biological activity of microorganisms was inhibited due to the low pH and high sulfate content (at pH 2 the sulfate content in the water was 1,900 ppm). To monitor and control the environment conditions of sulfur bacteria taking active role in the system, a full automation (SCADA) system was used. In this biological desulfurization process, biogas flow meters, air flow meter, circulation liquid flow meter, pH and temperature measurement devices, dilution (addition) liquid indicators, biogas oxygen analysis system, sulfur removal tower, tank level indicator, gas detector, pressure indicator, and other instruments were used. To compare the ability of biofilters on the same basis, the EC was used. It represents the ability in remove pollutants in gaseous form compared to the incoming pollutant mass, expressed as the mass of pollutant removed per unit time per bed volume. The parameters used in this study to describe the operating conditions and determine of the removal performances are given in Table 3.

**Table 3** | Process control parameters used in this study

	Formula	Nomenclature
Loading rate (gH <sub>2</sub> S m <sup>-3</sup> h <sup>-1</sup> )	$LR = \frac{Q}{V} C_{in}$	C <sub>in</sub> = concentration of H <sub>2</sub> S in gas entering biofilter (ppmv)
EC (gH <sub>2</sub> S m <sup>-3</sup> h <sup>-1</sup> )	$EC = \frac{Q}{V} (C_{in} - C_{out})$	C <sub>out</sub> = concentration of H <sub>2</sub> S in gas exiting biofilter (ppmv)
Removal efficiency (%)	$RE(\%) = \frac{(C_{in} - C_{out})}{C_{in}} \times 100$	Q: Flow rate of mixed gas entering biofilter (m <sup>3</sup> h <sup>-1</sup> )
Empty bed residence time (min)	$EBRT = \frac{V}{Q}$	V: Empty packed bed volume (m <sup>3</sup> )

H<sub>2</sub>S removal efficiency on a real scale in the BTF system was monitored for 12 months between January 2017 and December 2017 and the performance of the process was evaluated. During this period, the flow rate of biogas produced in the anaerobic sludge digesters, minimum, maximum, and average values of H<sub>2</sub>S levels in the biogas and at the process outlet were monitored on a monthly basis to determine the H<sub>2</sub>S removal efficiency of the

process. During this study, biogas flow was measured by a flow meter (Drager) and H<sub>2</sub>S concentration was measured by H<sub>2</sub>S measurement tubes (Rea), and analyzed by colorimetric method (TS EN 1231: 2000).

Microorganisms are an important factor in evaluating the effectiveness of the BTF system because the other elements, such as biofilm and pH, depend on the development condition of microorganisms. Like this study, most studies, mainly in an SOB community, used wastewater for H<sub>2</sub>S removal. This study carried out subsequent denaturing gradient gel electrophoresis (DGGE) analyses to identify SOB.

Total solids (TS), chemical oxygen demand (COD), total Kjeldahl nitrogen (TKN), volatile fatty acid (VFA), protein, and alkalinity parameters were monitored in the anaerobic sludge digester, which affects the BTF process. These parameters were determined following the procedures described in Standard Methods (APHA, AWWA, WEF 1998). TS, COD, TKN, VFA, protein, and alkalinity were analyzed using the following methods: Gravimetric method SM 2540 D, COD; Closed Reflux Method SM 5520 C, Macro Kjeldahl method SM 4500 B, Capillary titrimetric method, Combustion, and Distillation method SM 4500 N B and titration method SM2320 B.

### 3. RESULTS AND DISCUSSION

#### 3.1. Production of anaerobic digester and biogas

The biogas flow rate and the H<sub>2</sub>S concentration in the biogas were measured for the efficient process operation. The operational parameters of the mesophilic anaerobic sludge digester (pH, organic loading rate, sludge feeding rate, ambient temperature, volatile organic acid concentration, sludge retention time) during the operation of the biological desulfurization process, were given in Table 4. The characteristics of sludge at the inlet and outlet of sludge digester (total solid material, COD, protein concentration, alkalinity) were given in Table 5. The most important indicator showing the efficient operation of the anaerobic sludge digesters is the biogas production. During the working period, the flow rate of biogas produced in anaerobic sludge digesters varied between 18,123 and 21,383 m<sup>3</sup>/day and an average of 19,519 m<sup>3</sup>/day (Table 6; Figure 2). However, the range of percentage composition of the biogas produced from AD processes is dependent upon several factors including the digestibility of organic matter, digestion kinetics, digester retention time, and the digestion temperature (Dobre *et al.* 2014).

**Table 4** | Anaerobic sludge digester operation parameters

Parameters	Value	Average value
pH	7.3–8.1	7.9 ± 0.1
Organic loading ratio (OLR) (kg day <sup>-1</sup> m <sup>-3</sup> )	1.2–1.5	1.3 ± 0.1
Sludge feed flow (m <sup>3</sup> h <sup>-1</sup> )	16–17	16.9 ± 0.1
Temperature (°C)	36–41	38 ± 1
Volatile fatty acid (VFA)/alkalinity	0.02–0.08	0.05 ± 0.02
Sludge retention time (days)	17	17 ± 0.01

#### 3.2. Hydrogen sulfide removal from biogas

The inlet H<sub>2</sub>S concentration was routinely measured per day to assess the variation of the inlet H<sub>2</sub>S load. The H<sub>2</sub>S concentration of biogas at the inlet of the BTF unit varied between 2,900 and 4,400 ppmv and an average of 3,632 ppmv (Table 6). The H<sub>2</sub>S concentration in biogas is consistent with the literature (Jenicek *et al.* 2008; Charnnok *et al.* 2013; Reddy *et al.* 2019). The biogas generated in AD facilities in WWTPs contains average concentrations of H<sub>2</sub>S in the range from 0.1 to 0.5 vol. % (1,000–5,000 ppmv) (Walsh *et al.* 1989). At the outlet of the BTF process, H<sub>2</sub>S concentration varied between 4 and 63 ppm and an average of 16 ppm (Figure 3). No relation was determined between the biogas flow rate produced in the anaerobic sludge digester and the H<sub>2</sub>S concentration in the biogas. It is thought that H<sub>2</sub>S is produced depending on the other factors (protein and sulfate concentrations in wastewater, etc.) completely independent from the produced biogas quantity.

Since the produced biogas is used in the production of electrical energy, H<sub>2</sub>S needs to be removed due to the corrosive effect of H<sub>2</sub>S on gas engines and other auxiliary equipment. As a result, before using biogas in gas



**Table 5** | Sludge characteristics in anaerobic sludge digester

Parameters	Feed sludge	Anaerobic sludge digester outlet
TS (mg L <sup>-1</sup> )	Min: 25,100 Max: 37,500 Mean: 33,568 SD: 3,480	Min: 21,000 Max: 30,100 Mean: 25,306 SD: 3,873
COD (mg L <sup>-1</sup> )	Min: 19,100 Max: 32,000 Mean: 26,243 SD: 4,336	Min: 9,400 Max: 9,000 Mean: 8,762 SD: 1,523
TKN (mg L <sup>-1</sup> )	Min: 1,500 Max: 4,150 Mean: 3,189 SD: 502	Min: 1,300 Max: 4,700 Mean: 2,460 SD: 571
Protein (mg L <sup>-1</sup> )	Min: 11,000 Max: 23,100 Mean: 17,992 SD: 2,854	Min: 7,400 Max: 22,000 Mean: 17,820 SD: 3,381
Alkalinity (mg L <sup>-1</sup> )	Min: 720 Max: 1,300 Mean: 1,218 SD: 144	Min: 2,450 Max: 3,900 Mean: 3,100 SD: 279
VFA (mg L <sup>-1</sup> )	Min: 450 Max: 1,450 Mean: 1,094 SD: 308	Min: 60 Max: 230 Mean: 128 SD: 42

**Table 6** | Operation parameters for BTF

Time	Biogas (m <sup>3</sup> /d)	Inlet [H <sub>2</sub> S] <sub>biogas</sub> (ppmv)	Outlet [H <sub>2</sub> S] <sub>biogas</sub> (ppmv)	H <sub>2</sub> S LR (gH <sub>2</sub> S m <sup>-3</sup> h <sup>-1</sup> )	EBRT (min)	EC (gH <sub>2</sub> S m <sup>-3</sup> h <sup>-1</sup> )	H <sub>2</sub> S removal efficiency (%)
January	19,627	2,923	63	33.47	7.34	32.74	97.84
February	19,581	3,233	14	36.93	7.35	36.77	99.57
March	19,673	3,200	23	36.72	7.32	36.46	99.28
April	19,714	2,900	12	33.35	7.30	33.21	99.59
May	19,526	3,253	17	37.05	7.37	36.86	99.48
June	18,233	4,103	6	43.64	7.90	43.58	99.85
July	18,123	4,000	20	42.29	7.95	42.08	99.50
August	18,683	3,900	4	42.50	7.71	42.46	99.90
September	20,583	4,400	10	52.83	7.00	52.71	99.77
October	21,383	4,133	4	51.55	6.73	51.50	99.90
November	18,617	3,433	11	37.28	7.73	37.16	99.68
December	20,483	4,100	12	48.99	7.03	48.85	99.71
SD <sup>a</sup>	945.53	504.69	15.16	6.48	0.36	6.57	0.53
<b>Average</b>	<b>19,519</b>	<b>3,632</b>	<b>16</b>	<b>41.38</b>	<b>7.39</b>	<b>41.20</b>	<b>99.55</b>

<sup>a</sup>Standard Deviation: SD.

engines, the H<sub>2</sub>S concentration should be reduced below the limit value. For this reason, the H<sub>2</sub>S concentration should be reduced below the limit value ( $\leq 260$  ppm). The recommended level of H<sub>2</sub>S in the produced biogas is in the range of 0.02–0.05% (w/w) (200–500 ppm) while H<sub>2</sub>S-free biogas is more desirable (Rodriguez *et al.* 2014). During the working period, the H<sub>2</sub>S removal efficiency ranged between 97.84 and 99.90% and an average of

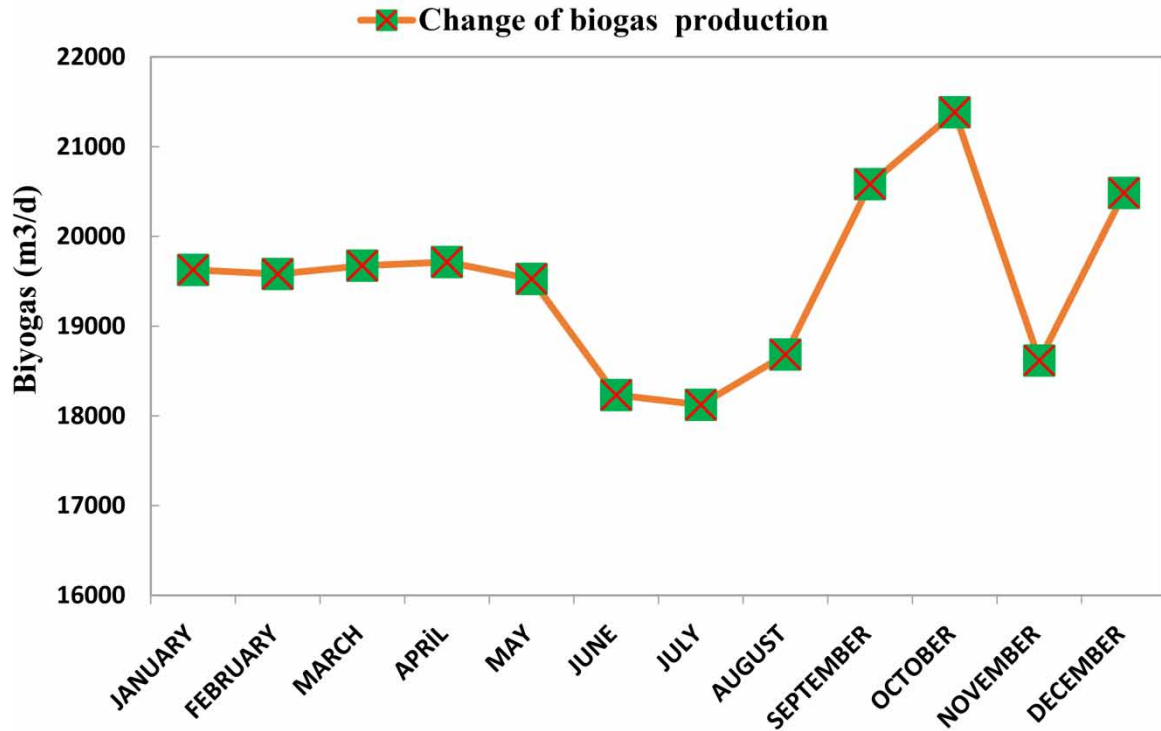


Figure 2 | Variation of biogas production.

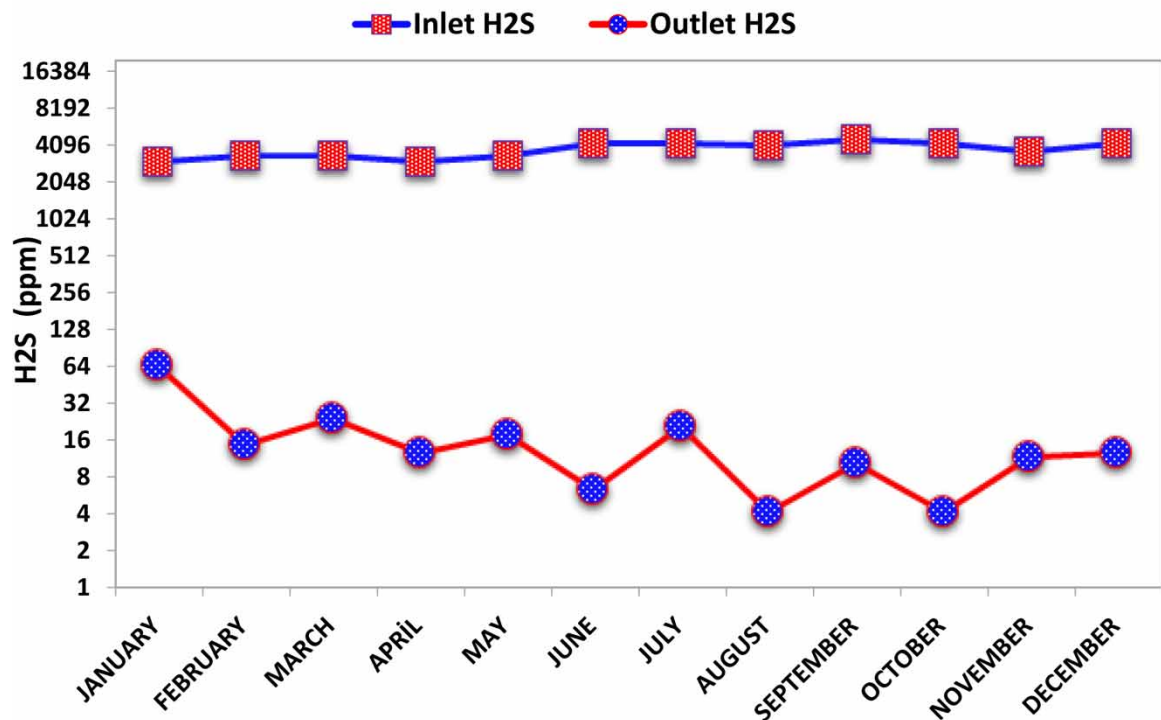
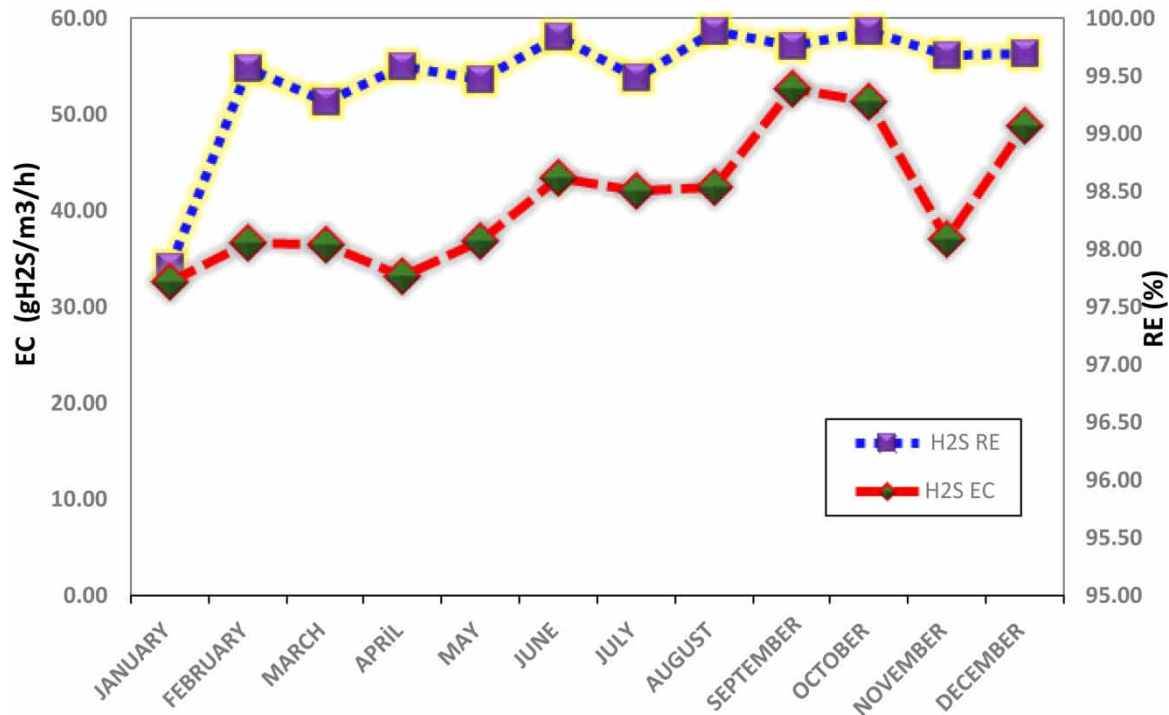


Figure 3 | Variation of H<sub>2</sub>S concentration at BTF inlet and outlet.

99.55% (Table 6). In January 2017, when the performance of the process started to be monitored, H<sub>2</sub>S removal efficiency was observed to be 97.8% and increased during operation to 99% (Figure 4). It was determined that the H<sub>2</sub>S concentration at the outlet of the BTF process was well below the determined limit value.





**Figure 4** | Variation of H<sub>2</sub>S removal efficiency and EC.

The EC and RE as functions of the load supplied to the system were analyzed for the BTF reactor. Figure 4 shows the removal efficiency and elimination capacity of H<sub>2</sub>S monthly. EC changes as a function of empty bed residence time (EBRT) and loading rate (LR) values. In the BTF process, EBRT values were between 6.3 and 7.95 min, LR values were between 33.35 and 52.83 g H<sub>2</sub>S m<sup>-3</sup> h<sup>-1</sup>, EC values were between 33.21 and 51.71 g H<sub>2</sub>S m<sup>-3</sup> h<sup>-1</sup> (Figure 4). The average H<sub>2</sub>S removal was 99.9% at EBRT of 7.39 min (i.e., a LR of 41.38 g H<sub>2</sub>S m<sup>-3</sup> h<sup>-1</sup>). Also, this study shows that BTF process performance according to EC and H<sub>2</sub>S RE is better than in previous studies (Table 7).

### 3.3. Microbial community

In the effective removal of H<sub>2</sub>S, microorganisms have an important role. SOB are gram negatives that can use sulfide and thiosulfate as an energy source. Analysis of sequenced bands using DGGE in samples drawn from the BTF tower showed different SOB in the HS samples with a similarity of at least 94% of the closely related cultures, all belonging to the *Proteobacteria* division. Members found it belonged to *A. thiooxidans* (99% similarity), employed for the aerobic treatment of H<sub>2</sub>S in BTFs found in anaerobic enriched cultures for the anaerobic bio-oxidation of sulfide (Tang *et al.* 2009). This bacterium is thought to be an ideal inoculum for the biofiltration of H<sub>2</sub>S in biogas and it is the most acidophilic SOB (Aita *et al.* 2016; Caicedo-Ramirez *et al.* 2016). It has a pH range between 0.5 and 5.5 and an optimum at pH 2–3 for growth (Wang *et al.* 2019). This result is similar to the results of Lee *et al.* (2006), which showed that in degradation of H<sub>2</sub>S, *Thiobacillus thiooxidans* proliferating between pH 2 to 0.5 and *A. thiooxidans* AZ11 could grow at pH as low as 0.2. It was reported that it was still possible to reach high removal efficiencies of 99.9%, 98.0%, and 94.0%, respectively. In their study, Rodriguez *et al.* (2014) predicted that the dominant SOB species in wastewater might be *A. thiooxidans* and *T. thiooxidans*, thus requiring an acidic pH to promote bacterial growth. In acidic conditions, *Acidithiobacillus sp.* was reported to reach an H<sub>2</sub>S EC of 113 (Aita *et al.* 2016), 150.3 (Charnnok *et al.* 2013) and 113.5 gH<sub>2</sub>S m<sup>-3</sup> h<sup>-1</sup> (Chaiyaprat *et al.* 2011). Our EC 35 gH<sub>2</sub>S m<sup>-3</sup> h<sup>-1</sup> at the condition for H<sub>2</sub>S removal is similar to those reported in the mentioned *Acidithiobacillus sp.* predominant experiments. pH of the recirculating fluid was found to decrease rapidly and vary between 1.5 and 3.5. Aroca *et al.* (2007) proposed that RE was 100% when utilizing *A. thiooxidans* for H<sub>2</sub>S oxidation at pH 1.8–2.5. It can be seen from the results that the SOB culture in the BTF reactor has already adapted to the condition of inlet H<sub>2</sub>S concentration and removed H<sub>2</sub>S from biogas.

**Table 7** | Comparison of the performance of BTF reported in the literature on the treatment of biogas polluted by H<sub>2</sub>S

Scale	Type bed	Packed bed volume	Inlet [H <sub>2</sub> S] (ppmv)	Empty bed residence time (EBRT)	Elimination capacity (EC) g H <sub>2</sub> S m <sup>-3</sup> h <sup>-1</sup>	H <sub>2</sub> S RE (%)	Reference
Laboratory-scale	Polypropylene pall rings	1 L	170	36 s	20	100	Cox & Deshusses (2001)
Laboratory-scale	HD-QPAC	2 L	2,000	3 min	55	99	Maestre <i>et al.</i> (2010)
Laboratory-scale	Polypropylene rings	1 L	5,415	5.5 min	89.4	100	Zhou <i>et al.</i> (2015)
Laboratory-scale (bench)	Calgon AP460	6.4 L	20–100	4–16 min	22.1	90	Duan <i>et al.</i> (2005)
Laboratory-scale (pilot)	Plastic pall rings	5.15 m <sup>3</sup>	1,954 ± 454	180 s	50 ± 11	94	Rodriguez <i>et al.</i> (2014)
Laboratory-scale	HD Q-PAC	2.15 L	2,000–8,000	180 s	50	100	Montebello <i>et al.</i> (2010)
Laboratory-scale (pilot)	Metallic Pall rings (AISI 316)	2 L	2,000	180 s	100–140	95–100	Montebello <i>et al.</i> (2012)
Laboratory-scale	Polypropylene Pall rings	2 L	2,000	131 s	50–100	35–100	Montebello <i>et al.</i> (2013)
Laboratory-scale	Metallic Pall rings	2.4 L	2,000–10,000	130 s	100–140	70–80	Montebello <i>et al.</i> (2014)
Laboratory-scale	HS- Q-PAC	2.15 L	900 – 10,000	180 s	200	84	Fortuny <i>et al.</i> (2008)
Laboratory-scale	HD-Q-PAC	2 L	2,000	167–180 s	84	97 ± 0.3	Fortuny <i>et al.</i> (2011)
Laboratory-scale	Polypropylene pall rings	2.4 L	850–8,500	2.4–3.5 min	99.8–130	99	Fernandez <i>et al.</i> (2014)
Laboratory-scale (pilot)	Polyurethane foam	600 m <sup>2</sup> m <sup>-3</sup> surface area and 35 kg m <sup>-3</sup>	5–25	15–40 s	15–95	99	Gabriel & Deshusses (2003)
Laboratory-scale (pilot)	Ceramic granules Volcanic rocks	1,177 L	2.84 ± 1.76 mg m <sup>-3</sup>	5–20 s	2.82–2.85	60–100	Li <i>et al.</i> (2012)
Full-scale	Polypropylene pall rings	–	3,000	180 s	170	90	Tomas <i>et al.</i> (2009)
Laboratory-scale (pilot)	BioSulfidEx	2.21 m <sup>3</sup>	500–600	84	32.3	98	Naegele <i>et al.</i> (2013)
Laboratory-scale	Polyethylene (HDPE)	1 L	0–2,040	120	78.57	100	Vikromvarasiri <i>et al.</i> (2017)
		12 L	1,000–4,000	10.29–72 min	14.58	100	Soreanu <i>et al.</i> (2008)

(Continued.)

Table 7 | Continued

Scale	Type bed	Packed bed volume	Inlet [H <sub>2</sub> S] (ppmv)	Empty bed residence time (EBRT)	Elimination capacity (EC) g H <sub>2</sub> S m <sup>-3</sup> h <sup>-1</sup>	H <sub>2</sub> S RE (%)	Reference
Laboratory-scale (pilot)	Commercial polyester fibers						
Laboratory-scale	Schist	7.85 L	1,100	300 s	30.3	100	Jabera <i>et al.</i> (2017)
Laboratory-scale	3D-printed honeycomb monolith	0.2 L	2,000	41 s	122	95	Qiu <i>et al.</i> (2017)
Laboratory-scale	K1 packing material	0.5 L	200	40–100 s	-	92.27 ± 10.30	Zhuoa <i>et al.</i> (2019)
Laboratory-scale	HDPE Plastics	1 L	2,000	120 s	82.98	99.5	Juntranaporn <i>et al.</i> (2019)
Laboratory-scale (Semi-pilot)	Polypropylene Pall rings	4,000 L	2,000	15 min	29.5	94.6–99.6	Reddy <i>et al.</i> (2019)
Laboratory-scale	Bamboo charcoal	643 L	5–20	10.9–28.9 s	6.58	99.8	Chen <i>et al.</i> (2019)
Laboratory-scale	Polyurethane foam	3 L	1,246–305	1.6 min	98	95–99	Tayar <i>et al.</i> (2019)
Laboratory-scale	Polypropylene pall rings	2.8 L	2,000	118 s	120	100	Lopez <i>et al.</i> (2019)
Pilot scale	Polypropylene spheres	440 L	1.2 g m <sup>-3</sup>	40 s	122	100	Xia <i>et al.</i> (2019)
Real-scale (WWT)	Polypropylene pall rings	100,000 L	2,900–4,400	6–8 min	33–53	98–100	This study

#### 4. CONCLUSION

In this study, the removal of H<sub>2</sub>S from biogas produced at a real-scale anaerobic sludge digester by the BTF process was investigated. The average biogas flow rate produced in the mesophilic anaerobic sludge digester varied between 18,123 and 21,383 m<sup>3</sup>day<sup>-1</sup> and H<sub>2</sub>S concentrations varied between 2,923 and 4,400 ppmv. The H<sub>2</sub>S concentration in the produced biogas is completely independent of the biogas flow rate. The removal of high concentrations of H<sub>2</sub>S in biogas was accomplished by a real-scale BTF process with SOB (*A. thiooxidans*) which are active in an acidic environment (pH 1.5–3.5). The BTF process was operated at pH:1.5–3.5, O<sub>2</sub>/H<sub>2</sub>S:1/2, EBRT:6.3–7.95 minutes, LR:33.35–52.83 g H<sub>2</sub>S m<sup>-3</sup> h<sup>-1</sup>. The H<sub>2</sub>S RE varied in the range of 97.84–99.90 and the H<sub>2</sub>S EC varied in the range of 33.21–52.71 gH<sub>2</sub>S m<sup>-3</sup> h<sup>-1</sup>. The process efficiency was found to be independent of the inlet H<sub>2</sub>S concentration. The average H<sub>2</sub>S values in biogas desulphurized by the BTF process ranged between 4 and 63 ppm. As a result, the BTF process regardless of the biogas flow and the inlet H<sub>2</sub>S concentration was found to be an effective and efficient process for the removal of H<sub>2</sub>S from biogas produced in the real-scale anaerobic sludge digester.

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#### ETHICS APPROVAL

Not applicable

#### CONSENT OF PARTICIPATE

Not applicable

#### AVAILABILITY OF DATA AND MATERIALS

Not applicable

#### COMPETING INTERESTS

We declare that they have no conflict of interest.

#### FUNDING

No funding was received for conducting this study.

#### CODE AVAILABILITY (SOFTWARE APPLICATION OR CUSTOM CODE)

Not applicable

#### CONSENT FOR PUBLICATION

Not applicable

#### AUTHORS' CONTRIBUTIONS

All authors contributed to the study conception and design. Material preparation and data collection were done by SK. The study of analysis was performed by SA. The first draft of the manuscript was written by SA and SK commented on previous versions of the manuscript. All authors read and approved the final manuscript.

#### DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

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