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Biological elimination of a high concentration of hydrogen sulfide from landfill biogas

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ABSTRACT

Hydrogen sulfide (H_2S) is one of the main contaminants found in biogas which is one of the end products of the anaerobic biodegradation of proteins and other sulfur-containing compounds in solid waste. The presence of H_2S is one of the factors limiting the valorization of biogas. To valorize biogas, H_2S and other contaminants must be removed. This study evaluated the performance of a pilot-scale biotrickling filter system on H_2S removal from landfill biogas. The biotrickling filter system, which was packed with stainless-steel pall rings and inoculated with an H_2S-oxidizing consortium, was designed to process 1 to 10 SCFM of biogas and used to determine the removal efficiency of a high concentration of hydrogen sulfide from landfill biogas. The biofiltration system consisted of two biotrickling filters connected in series. Results indicate that the biofiltration system reduced H_2S concentration by 94% to 97% without reduction of the methane concentration in the outlet biogas. The inlet concentration of hydrogen sulfide, supplied to the two-phase bioreactor, was in the range of 900 to 1500 ppmv. The hydraulic retention times (HRT) of the two biotrickling filters were 3.9 and 0.9 min, respectively. Approximately 50 ppmv of H_2S gas was detected in the outlet gas. The maximum elimination capacity of the biotrickling filter system was found to be 272 g $H_2S.m^{-3}.h^{-1}$. During the biological process, the performance of biotrickling filter was not affected when the pH of the recirculated liquid decreased to 2-3. The overall performance of the biotrickling filter system was described using a modified Michaelis–Menten equation, and the K_s and V_m values for the biosystem were 34.7 ppmv and 200 mg $H_2S/L.h^{-1}$, respectively.

Keywords: Biogas; Hydrogen sulfide; Biotrickling filter; Landfill biogas; H_2S removal.

INTRODUCTION

Biogas is one of the end products of the anaerobic digestion process. This process involves a series of metabolic stages (hydrolysis, acidogenesis, acetogenesis, and methanogenesis), in which a consortium of microorganisms degrades the organic substances in the absence of oxygen. Landfill biogas is a renewable energy consisting mainly of methane (45-55% v/v), carbon dioxide (30-40% v/v), and some trace components such as volatile organic compounds, hydrogen sulfide (H_2S) and other sulfur-containing compounds (Rasi et al. 2007). H_2S should be removed from the gaseous stream to be used as an energy source (biomethane) for generating electricity and heat. H_2S concentration in landfill biogas usually ranges from 1000 to 2000 ppmv (Lastella et al. 2002). It is a toxic, flammable, and colorless gas with a characteristic odor of rotten eggs (Roth 1993). The main problems that occur as a result of high H_2S concentrations in biogas are (i) damages engines and other equipment due to its corrosive action, and (ii) H_2S combustion, which produces sulfur oxides (SO_x) and causes emissions of increasing environmental concern (Dumont 2015). As a result, H_2S in biogas must be removed to avoid treatment facility corrosion, unnecessary production of by-products, and possible public exposure and complaints (Namgung et al. 2012).

H_2S gas elimination processes include either physical or chemical oxidation treatments. These methods are rapid and effective but require the addition of chemicals which are unhealthy, costly and generate secondary wastes. To get control of these limitations, biological oxidation treatment process can be an alternative to a physico-chemical system due to its low energy consumption and cost, high efficiency, and environmental acceptability (Kennes et al. 2009). Biofiltration of H_2S has been extensively studied ((Dumont 2015); (Abatzoglou and Boivin 2009)) yet there is still a gap in the research that has been carried out regarding the treatment of landfill biogas and this is what this research investigates.

The biofiltration technique is the ability of microorganisms to degrade organic matter for their own metabolism. In biofilm, the contaminant air is transferred from a gas phase to a liquid phase (Eq. 1) where microbial attack occurs. For the biodegradation of H_2S, sulfur-oxidizing bacteria (SOB) are used to oxidize H_2S to sulfur product (sulfate and sulfur). The following overall biological reactions (Oyarzún et al. 2003) can be expressed by Eq. 2 to 4:

$$H_2S \leftrightarrow H^+ + HS^- \text{ (non-biological)} \quad (1)$$

$$HS^- + 0.5O_2 \rightarrow S^0 + OH^- \text{ (biological)} \quad (2)$$

$$S^0 + 1.5O_2 + H_2O \rightarrow SO_4^{2-} + 2H^+ \text{ (biological)} \quad (3)$$

$$HS^- + 2O_2 \rightarrow SO_4^{2-} + H^+ \text{ (biological)} \quad (4)$$

During biological oxidation of H_2S, the H_2S first dissolves, releasing a proton to the caustic liquid phase (Eq. 1). The biological conversion of H_2S to elemental sulfur (S^0) or sulfate (SO_4^{2-}) is directly related to the availability of oxygen, according to Eqs. (2) and (3). In the case of low oxygen concentration, the oxidation proceeds only to S^0. Consequently, due to the accumulation of S^0 over time, the biotrickfilter bed yellows and causes clogging. Generally, sulfide is oxidized to sulfate (SO_4^{2-}) (Eq. 4) by a mixed population of sulfur-oxidizing bacteria (SOB) when enough oxygen is provided; the overall reaction (Eq.4) occurs with no production of elemental sulfur as an intermediate. However, since oxygen is plentiful in air, most of the H_2S is converted into sulfate, causing media acidification. In this way, accurate control of the oxygen level is a key parameter for controlling reactor performance (approximately 5% vol depending on the H_2S inlet concentration) (Schomaker et al. 2000). For safety reasons and, it is necessary to control the concentration of oxygen because the lower and upper explosive limits for methane in biogas are 9.2 mol% and 23.2 mol%, respectively (Schroeder et al. 2014).

The type of packing material can control biotrickling filter performances. Different types of packing material such as organic, inorganic, and synthetic have been used. The organic materials include soil, peat, bamboo charcoal, pine bark, and ceramsite, which have been widely used because of their porosity and low costs and their capacity to adsorb contaminants (Chen et al. 2014; Jaber et al. 2014). Inorganic materials used in biofilters include, among others, lava rocks (Soreanu et al. 2009) and expanded schist (ben Jaber et al. 2016). The use of inorganic materials can be favorable because of their availability and excellent physical and mechanical properties which can limit pressure drops even during a long operating period ((Dumont et al. 2012). In general, packing materials must be inoculated with various microbial communities contained in activated sludge, which are responsible for H_2S removal in the biofilter. In addition, nutrient equilibrium plays an important role in microbial growth. Biofilters are usually irrigated by a nutrient solution to provide necessary nutrients for microbial growth and to maintain the bed moisture, in addition to the elimination of the by-products produced during the biological oxidation reaction.

Biofiltration has been widely used to remove H_2S from biogas (Dumont 2015). However, previous studies have mainly focused on the biofiltration of H_2S at concentrations of less than 200 ppmv (Jin et al. 2005; Tang et al. 2009). Only a few studies on biofiltration have been carried out at high H_2S concentrations, i.e., above 4000 ppmv in synthetic biogas (Fortuny et al. 2008; Rattanapan et al. 2009; Montebello et al. 2013), but even fewer have been done using real biogas with high H_2S as the inlet gas in a lab-scale reactor (Tomàs et al. 2009; Chaiprapat et al. 2011; Rodriguez et al. 2014). As a result of the production of sulfuric acid generated by the oxidation of H_2S, the biotrickling filter media

becomes substantially acidified. When the biofiltration system is operated with high H_2S inlet load, the pH decreases. This acidification reduces the solubilization of H_2S in water (Eq. 1). On the other hand, SOB is well adapted to low pH conditions (Robertson and Kuenen 2006).

The purpose of this study was to evaluate the performance of a pilot-scale biotrickling filter system for removing H_2S from landfill biogas (1000-2000 ppmv), while maintaining the energy quality of the purified biogas, using an immobilized sulfur-oxidizing bacteria consortium. Thiosulfate and bicarbonate were used to immobilize sulfur oxidation activity and microbial growth in a 1-L lab-scale reactor and were then applied into the biotrickling filter. The immobilization and operation processes of H_2S removal from the biogas by an SOB were evaluated to derive the optimum process. Moreover, the kinetic analysis of biological H_2S removal using immobilized cells was also investigated.

MATERIALS AND METHODS

Microorganisms and culture medium

Activated sludge from a local municipal wastewater treatment plant (Magog, Quebec, Canada) with a suspended material concentration of 3.5 g/L was used for the initial inoculation of the reactor. The latter was used to develop an H_2S-oxidizing bacteria consortium, which consists of chemolithotroph bacteria known for their ability to grow in sulfur-containing media (Schedel et al. 1975). The thiosulfate mineral medium (TMM) contained the following: 15g/L $Na_2S_2O_3.5H_2O$, 4g/L K_2HPO_4, 4g/L KH_2PO_4, 0.8g/L $MgSO_4.H_2O$, 0.4g/L NH_4Cl, 22g/L $NaHCO_3$, 50g/L EDTA, 22g/L $ZnSO_4.7H_2O$, 5.54g/L $CaCl_2$, 5.06g/L $MnCl_2.4H_2O$, 4.99g/L $FeSO_4.7H_2O$, 1.1g/L $(NH_4)6MO_7O_{24}.H_2O$, 1.57g/L $CuSO_4.5H_2O$, 1.61g/L $CoCl_2.6H_2O$ and 2g/L yeast extract. During the acclimation process, the inorganic carbon source used was sodium bicarbonate, while the source of sulfur used was sodium thiosulfate.

The TMM medium was used as a recirculating liquid in the biotrickling filter system for the immobilization process, as well as for maintaining microorganisms. By adding 1 N NaOH or 1 N HCl, the final pH of this medium was adjusted to 7.

Acclimation and enrichment procedure

In 1-liter Erlenmeyer flasks, containing 500 mL of culture medium, 500 mL of activated sludge was added. The Erlenmeyer flasks were placed on a magnetic stirrer at room temperature. Air was continuously supplied with a stainless-steel diffuser to ensure aerobic conditions. The agitation was set at 100 rpm to avoid leading to shear forces of the biomass flocs. Bacterial growth (optical density), pH and dissolved oxygen were monitored daily. To guarantee

aerobic conditions, the O_2 concentration must be maintained between at least 2 mg/L and 4 mg/L (Wilen and Balmer 1999). During a decrease in the biomass concentration, as a result of a depletion of the nutrients, the culture medium was renewed at a rate of 30% by volume.

Inoculation of the lab-scale bioreactor

A volume of 500 mL of three-day acclimated bacterial suspension with an optical density equivalent to 4.5 (absorbance at 600 nm) was transferred to the laboratory-scale bioreactor (diameter = 10 cm & height = 17 cm); see Figure 1. For one month, the reactor, which was made of glass with a total volume of 1000 mL, was continuously supplied with a synthetic biogas composed of H_2S (0.07 to 0.2% v/v), CO_2 (20% v/v) and air (100% balance). The experimental conditions for these laboratory tests are summarized in Table 1. During the lab-scale tests, the initial H_2S concentration was initially 730 ppmv; then it was gradually increased to 1000 and 2000 ppmv at 3-day intervals. One part (300 mL) of the culture medium was renewed every 72 hours when there was a decrease in the rate of biomass growth (optical density) in the reactor.

Pilot-scale bioreactor

Packing materials

Stainless-steel pall rings (DN16) (Masaito Metal Company, Ltd., Hengshui City, China) were used as a support for the microbial population in this study. The pall rings are cylindrical in appearance, and their height is the same as their diameter (16 mm), as shown in Fig.2. The packing material had a high porosity of 95%. The physical characteristics of the stainless-steel pall rings are summarized in Table 2.

Pilot-scale set-up and operation conditions

The pilot-scale system for H_2S biofiltration is shown in Fig.3. It consisted of two biotrickling filters connected in series (BTF1 and BTF2) made of PVC. The first biotrickling filter (BTF1) has an inner diameter of 0.4 m and a height of 1.5 m. While the second biotrickling filter (BTF2) has an inner diameter of 0.2 m and a height of 1.3 m. The two columns were filled with the same packing material (stainless-steel pall rings DN16). The effective volume of the BTF1 was 110 liters, which corresponds to 0.9 m in height of DN16, while the effective volume of the BTF2 was 25 liters (0.8 m height). The real inlet biogas consisted of 52% v/v of methane, 39% v/v of CO_2, 11% v/v of CO, 1.2 % of O_2, and 1000-2000 ppmv of H_2S. The flow rate was controlled via a flow meter and regulator on the biogas pipe. The air flow in BTF1 was constant at 2% v/v of O_2 during the operation period. The inlet gas was fed to the bottom

of the BTF1 and BTF2 at a constant flow rate of 1 SCFM (28.3 L/min) and 0.9 SCFM (27.7 L/min), respectively. Part of the pre-treated biogas was exhausted after BTF1. Throughout the experimental period, the biogas compositions were measured periodically, and the percolate at the bottom of each biotrickling filter was evaluated to determine its pH, concentrations of sulfate and COD. In this experiment, the two biotrickling filters were first inoculated with the thiosulfate mineral medium (TMM) from the lab-scale reactor for 3 days. The medium prepared for the cultivation of the bacteria contained 5g/L $Na_2S_2O_2.5H_2O$, 1.5g/L KH_2PO_4, 1.5g/L K_2HPO_4, 0.4g/L NH_4Cl, 0.2g/L $MgCl_2.6H_2O$, and 0.01 g/L $FeSO_4.7H_2O$. This medium was added into Reservoir 1 and fed into the reactor via a peristaltic pump. After the immobilization process, $Na_2S_2O_3.5H_2O$ was removed from the liquid medium in the recirculation tank.

When the reactor system was operating, due to an increase of sulfate concentration in the liquid medium, which is induced by the biological oxidation of H_2S, a 30% volume of fresh medium was renewed every 7 days to maintain the level of SO^{-2}_4 buildup below 40 g/L. This nutrition solution was sprayed downward for the downflow mode of operation to maintain the bed moisture and provide necessary nutrients for microbial growth, in addition to the elimination of the by-products produced during the biological oxidation reaction. The nutrition solution flow rate was constant at 3.8 l/min, discharged from the bottom, and recycled. The operational conditions of BTF1 and BTF2 are summarized in Table 3.

Kinetic analysis

The removal rate of hydrogen sulfate was calculated using the equation below (Eq.4), which was derived from the Michaelis–Menten equation (Ma et al. 2006; Hirai et al. 1990; Chen et al. 2014; Chung et al. 2001)

$$\frac{1}{R} = \frac{K_s}{V_m} \times \frac{1}{C_{ln}} + \frac{1}{V_m} \quad (4)$$

Where R is the apparent removal rate in mg H_2S/L/h, K_s is the apparent half-saturation constant in ppmv, V_m is the maximum apparent removal rate in mg H_2S/L/h, and $C_{ln} = (C_{in} - C_{out})/\ln(C_{in}/C_{out})$, is the logarithmic mean of H_2S concentration in the biotrickling filter ; C_{in} (ppmv) is the inlet concentration of H_2S in the biotrickling filter; C_{out} (ppmv) is the outlet concentration of H_2S in the biotrickling filter. From the linear relationship between $1/C_{ln}$ and $1/R$ (Lineweaver–Barker method), the kinetic constants V_m and K_s of the biological degradation of hydrogen sulfate were calculated from the intercept and slope, respectively.

Analytical methods

Inlet and outlet concentrations of H_2S gas were continuously measured by using a colorimetric gas detection tube (Gastec corporation, Japan). The H_2S tube detection ranges were 2000 and 1-200 ppm for inlet and outlet H_2S gas concentrations, respectively. Methane, carbon dioxide, and oxygen were periodically measured with a portable gas analyzer (Landtec GEM5000) (QED Environmental Systems, Inc., United States). The spectrophotometric method (Hach-DR-2000) was used to determine the sulfate concentrations as described by APHA (1992). Sulfate samples were taken periodically from Reservoir 1 and Reservoir 2 (nutrition tanks). The pH of the nutrition solution was measured using a calibrated digital pH-meter (Mettler Toledo, Canada). The growth of the microorganisms was monitored by measuring the optical density of the growth medium at 600 nm with a spectrophotometer SpectraMax Plus 384 (Molecular Devices Corp., Sunnyvale, California, United States). After adequate dilution, depending on the turbidity of the suspension, the absorbance of the sample taken was measured against a blank, which consisted of distilled water. The final absorbance was the absorbance read for the diluted sample multiplied by the dilution factor. The chemical oxygen demand (COD) of the nutrient solution was determined by using the Hach-DR-2000 analyzer according to the standard method (Jirka and Carter 1975). The COD values were measured by using AccuSPEC reagent tubes (1500 ppm) (SCP SCIENCE Company, Québec, Canada).

The ICP-MS methodology was used for heavy metal quantification in wastewater (CEAEQ, 2012).

The removal efficiency (RE), the elimination capacity (EC), and the loading rate (LR) of the biotrickling filters were calculated as follows:

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

$$EC\ (g\ m^{-3}\ h^{-1}) = \frac{\mathrm{Q}}{\mathrm{V}}\ (C_{in} - C_{out})$$

$$LR\ (g\ m^{-3}\ h^{-1}) = \frac{\mathrm{Q}}{\mathrm{V}}\ C_{in}$$

Where C_{in} and C_{out} are the inlet and outlet gas concentrations ($g\ m^{-3}$) respectively, Q is the gas flow rate ($m.^3\ h^{-1}$) and V is the bed volume of packing material (m^3).

193 # RESULTS AND DISCUSSION

194 ## Immobilization and acclimatization of the consortium

195 The thiosulfate mineral medium (TMM) containing SOB was loaded into the lab-scale reactor with sodium
196 bicarbonates being the sole carbon source and thiosulfate being the sole energy source supporting SOB growth. Fig.4
197 shows the SOB growth and sulfate production when the pH of the medium was adjusted daily to 7 without the
198 replacement of fresh medium. The pH was in the range of 6 – 7. The acclimation process was performed for 45 hours.
199 This process reached a 4.5 optical density after 20 hours corresponding to a sulfate concentration of 2 g/L. After a 20-
200 hour operating period, the optical density decreased to 4 as the sulfate concentration increased to 10 g/L. The
201 concentration of the accumulated sulfate in the medium was 6 g/L. This value posed a toxic effect on the SOB growth
202 as indicated by the decreasing optical density and would subsequently affect the oxidation activity of thiosulfate in
203 the lab-scale system. To avoid the accumulation of sulfate in the medium, a 30% volume of the medium was renewed
204 when the growth rate of the biomass decreased.
205 In the biotrickling filter system, SOB was immobilized successfully on the packing bed by recirculating the liquid
206 which was inoculated from the lab-scale reactor. The recirculation liquid flow rate was 3.8 L/min. Throughout the
207 immobilization process, it was observed that the elemental sulfur was accumulating on the packing media (result not
208 shown). After 3 days, real biogas gradually flowed into the reactor system until 1 SCFM.

209 ## Removal of hydrogen sulfide in the lab-scale reactor

210 The experiment was performed at the lab-scale level for 60 days using an average inlet H_2S concentration of 2400
211 ppmv. In the first phase (40 days of operation), the H_2S was generally removed by 38% to 80% for corresponding
212 outlet concentrations of 1500 to 500 ppmv, respectively.
213 In the second phase (between Days 40 and 60), the H_2S removal efficiencies were always higher than 99% and the
214 effluent concentrations were below the detection limit of 2 ppmv, as shown in Fig.5. The maximum EC was
215 determined as 50 g $H_2S.m^{-3}$ h^{-1} at the loading rate of 330 g $H_2S.m^{-3}$ h^{-1}. The results showed that the high removal
216 efficiency was due to the consortia adaptation from the first phase and subsequently the increasing bacterial growth
217 in the second phase. Moreover, the removal efficiency of H_2S increased with increasing operating times. This result
218 was consistent with other researchers on H_2S removal from biogas in a lab-scale reactor (Table 4).

Figure 6 shows the evolution of sulfate ions, biomass, and pH in the reactor. These figures show a good correlation with the whole H_2S removal process. However, the higher the biomass concentration, the more sulfate ions were produced, which acidified the medium. This is consistent with bacterial metabolism for hydrogen sulfide.

Biodegradation of H_2S in the pilot-scale bioreactor

The H_2S removal from landfill biogas was tested by using two biotrickling filters connected in series packed with stainless-steel pall rings. The H_2S inlet concentrations were between 900 and 1500 ppmv. Fig.7 shows the H_2S removal efficiencies in the two biotrickling filters packed with stainless-steel pall rings during 90 days of operation.

During the operation of BTF1, a 94% removal efficiency average for 900–1500 ppmv of H_2S was achieved, and approximately 50 ppm of H_2S gas was detected in the outlet of BTF1. BTF1 was found to be about 25% more effective in removing H_2S than BTF2 due to the different kinetics in BTF2 resulting from the lower inlet concentration of H_2S. Whereas, after adding BFT2 to the system, the removal efficiency of the whole system increased to 98%.

Furthermore, the maximum EC in BTF2 (81 g $H_2S.m^{-3}.h^{-1}$) was lower than BTF1 (272 g $H_2S.m^{-3}.h^{-1}$). Comparison of this result with previous studies shows that a maximum EC of 100 g H_2S m^{-3} h^{-1} was found at H_2S concentrations greater than 1500 ppmv using metallic pall rings as a filter bed, under mass transfer limiting conditions (Montebello et al. 2013). A maximum EC of 24 g H_2S $m^{-3}.h^{-1}$ was achieved at 100% RE in a laboratory biotrickling filter packed with plastic pall rings (Jin et al. 2005). Similarly, (Fortuny et al. 2008) showed that a 200 g H_2S $m^{-3}.h^{-1}$ elimination capacity was observed at H_2S concentrations of 900-10,000 in a biotrickling filter at an empty bed retention time (EBRT) of 180 s.

Therefore, results indicate that the biofiltration system, containing stainless-steel pall rings as a filter material, was suitable for H_2S removal with an average 94.2% efficiency for H_2S inlet concentrations in the range of 900 to 1500 ppmv H_2S and has potential for industrial application.

Effect of sulfate concentrations and changes of pH on H_2S elimination

The sulfate concentration in the recirculated liquid was evaluated in both BTFs because it is the final product of the H_2S biofiltration treatment process. In the presence of enough oxygen, the sulfide was oxidized to sulfate (SO^{2-}_4) by a mixed population of SOB, which is a water-soluble compound. The amount of sulfate that accumulated over a 90-day period is shown in Fig. 8a and 8b. The results indicate that the sulfate concentration reached 33 S-SO^{2-}_4 g/L, and there was 95% H_2S removal efficiency in BTF1. While the sulfate concentration was 3.7 S-SO^{2-}_4 g/L in BTF2, and its removal efficiency for H_2S was 67%.

According to this study and measurements, no significant effect was observed in the H_2S removal efficiency when the sulfate content was as high as 33 g/L (Fig.8). These results are consistent with (Fernández et al. 2014), who indicated that a sulfate concentration higher than 33 g/L must be avoided. Above this level, the activity of the microorganisms was significantly reduced due to the presence of sulfate at a toxic level.

After 5 days of continuous operation of the biotrickling filter, a slightly faster decrease in pH from 7 to 1.5 was observed in BTF1 as shown in Fig.8a. However, the significant changes in pH did not have a substantial effect on the H_2S removal efficiency, probably because SOB can live in acidic environments having a wide pH range (1 to 8) (Robertson and Kuenen 2006). These results are similar to the (Chen et al. 2014) study in which combinations of bamboo charcoal and ceramist were used for the removal of H_2S. In particular, it was found that the removal efficiency of H_2S did not change when the pH decreased from 7 to 3. As seen in the experiment of Sercu et al. (2005), the pH of the recirculated liquid did not affect the biotricking filter performance when decreased to 2-3 (Rodriguez et al. 2014). Such a low pH causes a drastic reduction in H_2S solubility in the liquid medium in BTF1 and hence some of the H_2S escapes without being treated. Therefore, the BTF2 with a pH of 7 captures H_2S and then oxidizes it to sulfate.

Methane, CO_2, and oxygen concentrations

Purified biogas is mainly used as feedstock to produce alternative fuels. Conversion of both gases, i.e., methane (CH_4) and carbon dioxide (CO_2), to alternative fuels such as methanol and syngas, will help to alleviate the greenhouse gas (GHG) release and global warming impact (Zain and Mohamed 2018). Therefore, it is very important to keep the CH_4 and CO_2 concentrations constant in the outlet gas from the biofiltration system.

During the biotrickling filter operation, the concentrations of the main biogas components, methane, and carbon dioxide showed minor variations (Fig. 9). The average inlet concentrations of methane and carbon dioxide were 45.5 ± 3.9 % (v/v) and 35.0 ± 2.9 % (v/v), respectively. While the treated biogas in BTF1 (Fig. 8a) had an average methane

concentration of 45.4 ± 3.7 % (v/v) and an average carbon dioxide concentration of 35.0 ± 2.7 % (v/v) of the biogas. The concentrations of methane and carbon dioxide in the biogas in BTF2 (Fig. 10b) showed the same behavior as in BTF1. The average concentrations of methane and carbon dioxide in the outlet were 46.5 ± 2.9 % (v/v) and 35.8 ± 1.9 % (v/v), respectively.

According to the results, there was no significant difference between the average concentrations of inlet and outlet methane and carbon dioxide in the BTF1 and BTF2. This indicates that the energetic content of the biogas was not reduced during the H_2S oxidation process in the biotrickling filter. The biotic process and the use of medium containing an inorganic carbon source did not affect the concentrations of methane and carbon dioxide in the biotrickling filters. Thus, the SOB is an autotroph organism capable of oxidizing such compounds as hydrogen sulfide from carbonate as an inorganic compound and does not use methane as a source of carbon and energy (Terrado et al. 2017).

There were no reductions in the concentrations of methane or carbon dioxide in the biogas. When compared to conventional aerobic biotrickling filters, maintaining the methane concentration is a major benefit. (Fernández et al. 2013). This fact demonstrated that the biofiltration system could be used efficiently for H_2S elimination while maintaining the energetic value of the biogas.

During the operation of an aerobic biotrickling filter system, air is supplied with the biogas stream to provide oxygen for biological oxidation and SOB microbial respiration. Oxygen control in the biotrickling filter is essential because oxygen deficiency causes a formation of elemental sulfur and subsequently increases clogging problems, while a high amount of oxygen means that the biogas is severely diluted and unwanted residual oxygen can be found in the outlet biogas. The landfill biogas does not contain enough oxygen to support complete H_2S bio-oxidation. Essentially, it requires an air/oxygen blower with security control to ensure that the air concentration does not exceed the lower explosive limit of methane in biogas of 9.2 mol% (Schroeder et al. 2014). The addition of 4-6% (v/v) air (0.8-1.2% (v/v) O_2) to biogas before biofiltration is to ensure proper sulfide conversion (Wellinger and Lindberg 2000). Soreanu et al. (2005) found that in a reactor with less than 3% (v/v) O_2, > 90% H_2S biological conversion was achieved. Schomaker et al. (2000) also suggested 5% (v/v) air as being a low enough concentration of O_2 in the inlet gas to biologically convert the H_2S to elemental sulfur instead of SO_4^{2-}, which requires a higher amount of O_2. This difference in sulfide oxidation species was also reported by (Duan et al. 2005) where 0.1mg/L was considered the oxygen limiting condition at which elemental sulfur would be the prominent product from sulfide oxidation. Sulfate would be the major product when sulfide was limited.

In this study, oxygen was supplied with an average concentration of 2.1 ± 0.9% (v/v) into the BTF1 inlet to support the biological oxidation of H_2S. As a result, the actual oxygen concentrations were found to be 1.8 ± 0.8% (v/v) and 1.6 ± 0.7% (v/v) in the BTF1 and the BTF2 outlets, respectively as shown in Fig.10.

Kinetic analysis of biodegradation

In this experiment, the kinetic process of biological H_2S degradation was studied using a pilot-scale biotrickling filter. Two percent oxygen was supplied to the biotrickling filter for SOB microbial activity and respiration. The H_2S removal in the biotrickling filter system could be regarded as a simple enzyme-catalyzed reaction with one substrate and without inhibitor product (Michaelis–Menten equation).

The linear relationship between 1/R and $1/C_{ln}$ in the biotrickling filter indicated in Fig.11. The maximum removal rate (V_{max}) and the half-saturation constant (K_s) of H_2S were calculated from the regression analysis as 200 mg H_2S/L/h and 34.7 ppmv, respectively. Generally, the lower K_s value, compared to the H_2S concentration range (900-1500 ppmv), is an indication of the affinity of the enzyme towards the substrate and the efficiency with which degradation will occur. Thus, the biotrickling filter showed a high capacity for H_2S oxidation. The kinetic parameters were within the range reported by other researchers. (Ma et al. 2006) reported a V_{max} value of 666.7 mg H_2S / (L.d) with a K_s value of 20.8 mg/L. (Chung et al. 2001) reported a V_{max} value of 1.11 g-S/day/kg bead) in the biofilter and K_s value of 34.6 ppm.

Characterization of the liquid phase of the biotrickling filter in treating landfill biogas

The liquid phase (nutrient solution) is an essential part in treating biogas. The nutrient solution is used to provide the necessary nutrients for the microorganisms, to control the pH of the reaction media and to solubilize the H_2S so that it can be bio-transformed. In this study, the average characteristics of the liquid produced are based on the measurement of pH, COD, sulfate concentration, and heavy metals of the percolate (Table 5). During the experiment period, since a large amount of sulfuric acid had accumulated in the medium, the pH was quite acidic. The mixed wastewater from the discharges of biotrickling filters 1 and 2 had a pH of 5. As a result, sulfate as a product of microbial oxidation of H_2S could be responsible for the pH decrease.

Based on the characteristics of the mixed wastewater produced from the biofiltration system, it was observed that high concentrations of chrome, iron, and nickel were leached to the liquid. These metals were probably chemically or biologically leached from the packing material (consisting essentially of stainless steel) at such acidic and oxidizing conditions (Montebello et al. 2014).

The COD of the initial nutrition solution before recirculation was around 20 mg/L. The COD of the liquid increased due to high concentrations of biomass released and organic content absorbed to reach 1520 mg/L after 6 months of operation. This value is higher than the COD concentration in the (Omri et al. 2013) study, where it was observed that the COD of the liquid increased from 20-30 mg/L to 350-680 mg/L at the outlet of the bed for 150 day of operation.

CONCLUSIONS

The results of this study show that a stable performance of a biotrickling filter system, packed with stainless-steel pall rings, for elimination of H_2S from landfill biogas, can be achieved in long-term operation. This system has the advantage of reaching relatively high removal efficiencies of H_2S with no reduction in methane concentration across the biotrickling filter. Removal efficiencies ranged between 94% and 97% for H_2S inlet concentrations in the range of 900 to 1500 ppmv. Approximately 50 ppmv of H_2S gas was detected in the outlet gas. The maximum elimination capacity of the biotrickling filter was found to be 272 g $H_2S/m^{-3}/h$. the performance of biotrickling filter was not affected when the pH of the recirculated liquid decreased to 2-3. Such low pH causes reduced solubility of H_2S in the liquid medium in BTF1, and hence some of the H_2S escapes without being treated. Therefore, the BTF2 with a pH of 7 captures H_2S and then oxidizes it to sulfate. As a result, biofiltration has proven to be a viable alternative to traditional treatment techniques for eliminating H_2S from landfill biogas. It was observed that high concentrations of chrome, iron, and nickel were leached to the liquid produced. These metals were probably chemically or biologically leached from the packing material (consisting essentially of stainless steel) at such acidic and oxidizing conditions.

ACKNOWLEDGMENTS

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DECLARATIONS

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and materials

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

All authors contributed to the study conception and design. Investigation, formal analysis and writing – original draft, were performed by **Rania Ibrahim**. **Abdessamad El Hassni** contributed to the investigation, methodology and data curation. **Shahram Navaee-Ardeh** contributed to the methodology, resources, and co-supervision. The supervision, project administration, funding acquisition and writing - review & editing were ensured by **Hubert Cabana**. All authors read and approved the final manuscript.

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List of Tables

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Table 1 The experimental conditions of the laboratory test.

Parameters	Value
Volume (L)	1
Flow rate (L/min)	0.5 – 1
Retention time	1 - 2 min
Reactor pressure (atm)	Atmosphere
Gas pressure before injection in the reactor (atm)	1.7
Concentration of H_2S (ppmv)	730 – 2000
Concentration of CO_2 (%v/v)	20
Air humidity	Balance 100%

Table 2 Physical characteristics of the stainless-steel pall rings.

Physical characteristics	
Nominal diameter (mm)	DN16
Diameter × Height × Thickness (mm)	16×16×0.4
Porosity (%)	95
Specific surface area (m^2/m^3)	362
Bulk density (Kg/m^3)	396

Table 3 The operational conditions of BTF1 and BTF2.

Parameters	BTF1	BTF2
Dimensions	1.5 m × 0.4 m	1.3 m × 0.2 m
Effective volume (L)	110	25
Hydraulic retention time @1scfm	3.9 min	0.9 min
pH	2	7
Concentration of inlet H_2S (ppmv)	900	100
Air humidity	< 4%	--

Table 4 Comparison of the H_2S elimination capacity of a lab-scale reactor with previous studies.

Biogas composition	Inlet H_2S concentration (ppmv)	EBRT	EC $gm^{-3}h^{-1}$	RE	Ref.
$H_2S + N_2$	10 - 100	12.71 - 31.77 s	19.24	98%	(Potivichayanon et al. 2006)
N_2 (65%) + CO_2 (35%) H_2S (traces)	1000	69 s	32.5	99	Bailón 2007
H_2S + Air	10 - 90	9-60 s	32.5	98.3%	Namini et al., 2008
Real biogas	1000 - 4000	10.29-72 min	14.58	99	(Soreanu et al. 2008)
$H_2S + N_2$	2,000 - 8,000	180	50	100	Montebello et al. 2010
Synthetic Biogas	0 - 2040	120 s	78.57	93.6%	(Vikromvarasiri et al. 2017)
Synthetic biogas	730- 2000	1-2 min	50	99	This study

527 **Table 5** Average characteristics of the liquid phase.

528

Parameters	Value
pH	5
Chemical oxygen demand (COD)	1520 mg/L
Total suspended solid (TSS)	143 mg/L
Volatile suspended solid (VSS)	47 mg/L
Sulfate ion (SO_4^{2-})	22200 mg/L
Chrome (Cr)	351 mg/L
Calcium (Ca)	42.9 mg/L
Iron (Fe)	1500 mg/L
Potassium (K)	440 mg/L
Nickel (Ni)	185 mg/L
Magnesium (Mg)	54.5 mg/L
Manganese (Mn)	22.9 mg/L
Phosphorus (P)	440 mg/L

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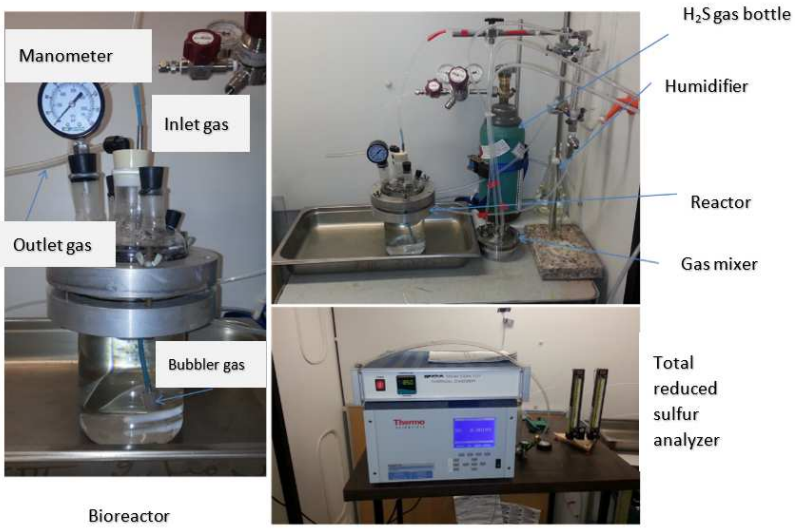

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531 **Figure 1** A laboratory scale bioreactor.

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534 **Figure 2** Stainless-steel pall rings (DN16).

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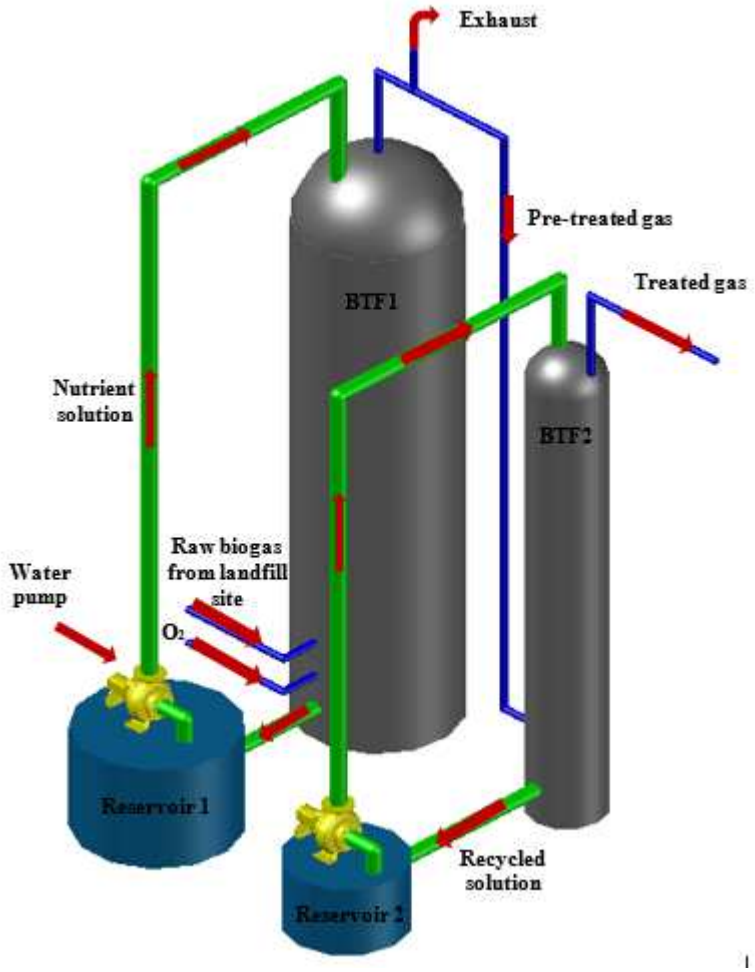


Figure 3 Pilot-scale biotrickling filter in landfill site packed with stainless-steel pall rings (DN16).

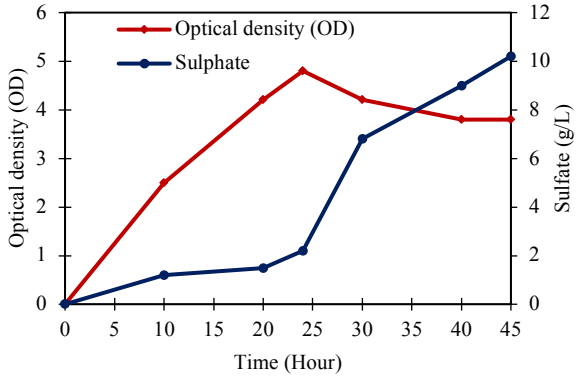


Figure 4 Evolution of sulfate and biomass as a function of time during acclimation tests (T =25° C, 60 rpm).

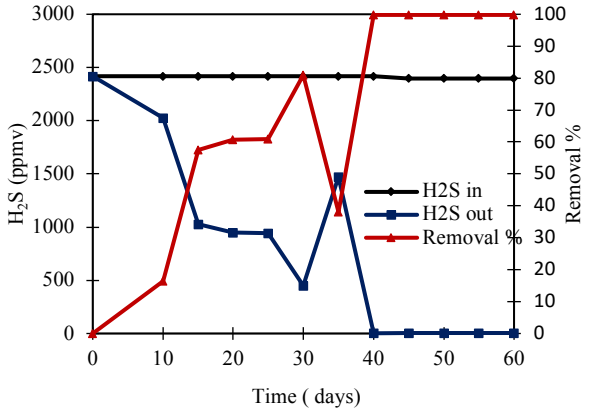
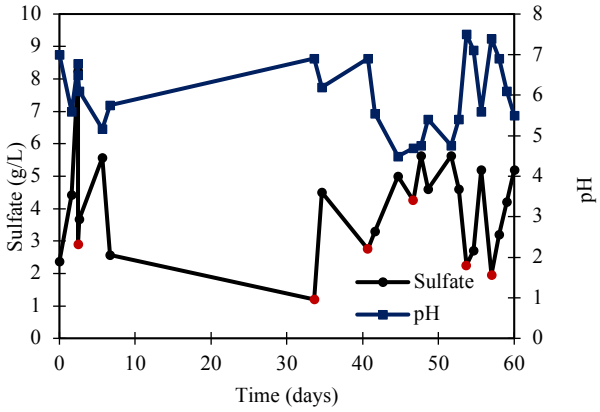
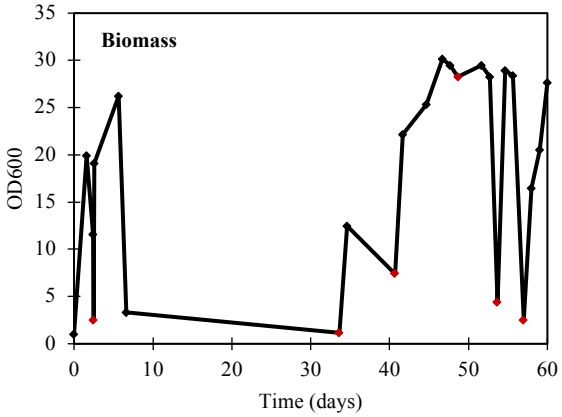


Figure 5 Monitoring the H_2S concentration at the inlet and outlet of the bioreactor and the rate of elimination. (T = 25 °C, 60 rpm, Initial biomass: OD600 = 3.75).

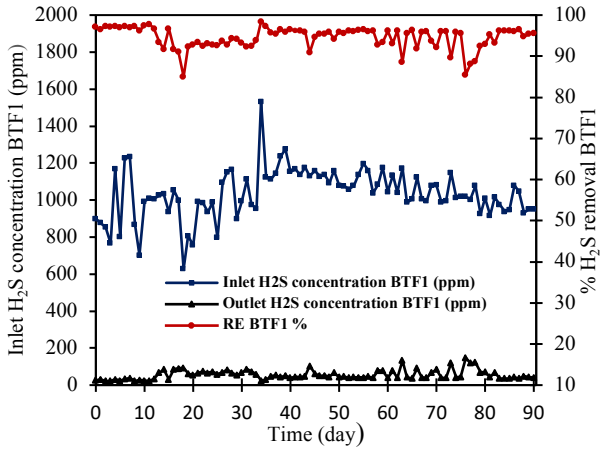


(a)



(b)

Figure 6 Evolution of pH, sulfate ion concentration and biomass concentration (T = 25 ° C, 60 rpm, Initial biomass: OD600 = 3.75). The points in red correspond to the times when the culture medium was renewed.



(a)

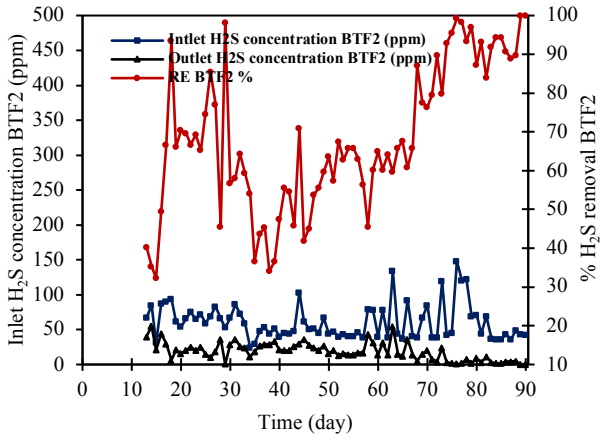
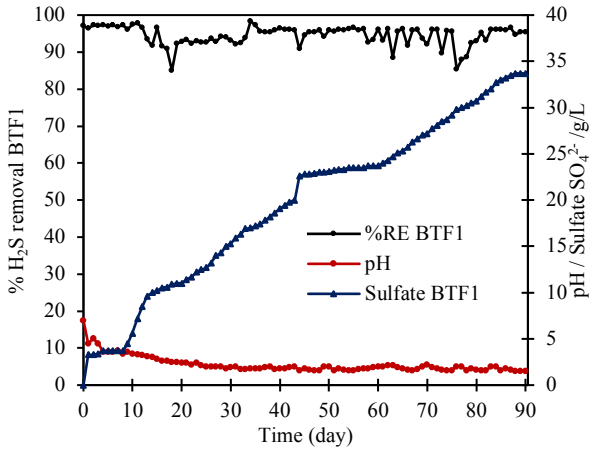
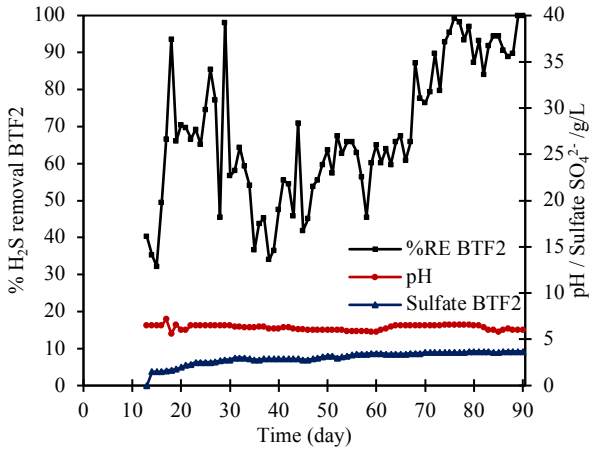


Figure 7 The removal efficiency of H_2S in the two biotrickling filters. (a) Biotrickling filter 1, and (b) Biotrickling filter 2.

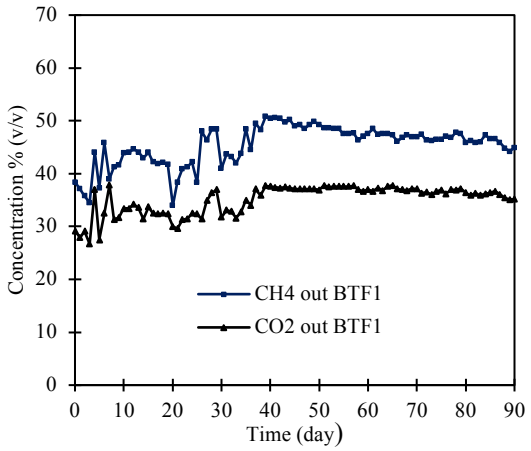


(a)



(b)

Figure 8 The removal efficiency of H_2S, cumulative sulfate SO_4^{2-} /g/L and pH changes in two biotrickling filters. (a) Biotrickling filter 1, and (b) Biotrickling filter 2.



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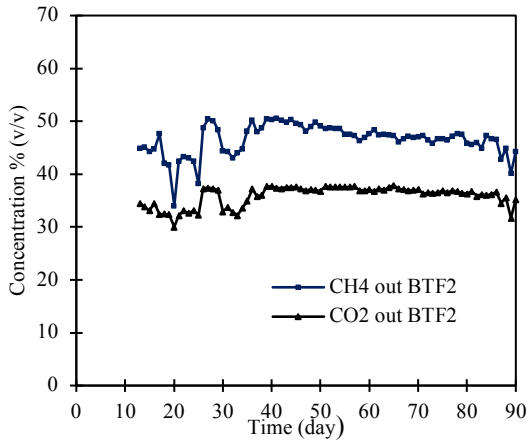
564 (a)



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566 (b)

567 **Figure 9** Methane and carbon dioxide composition in % (v/v). (a) Biotrickling filter 1, and (b) Biotrickling
568 filter 2.

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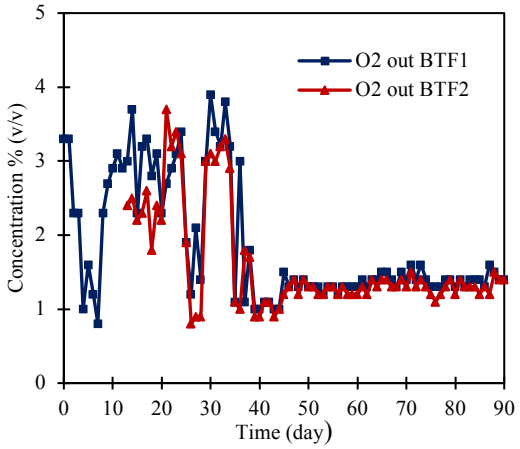

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571 **Figure 10** Oxygen concentration during operation of two biotrickling filters.

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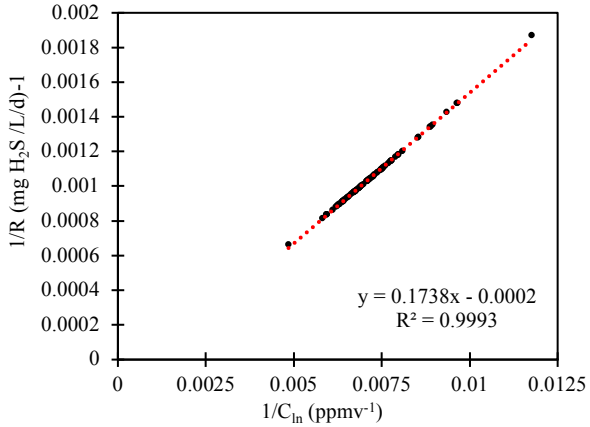

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574 **Figure 11** The linear relationship between $1/R$ and $1/C_{ln}$ for H_2S degradation in the pilot scale biotrickling

Figures

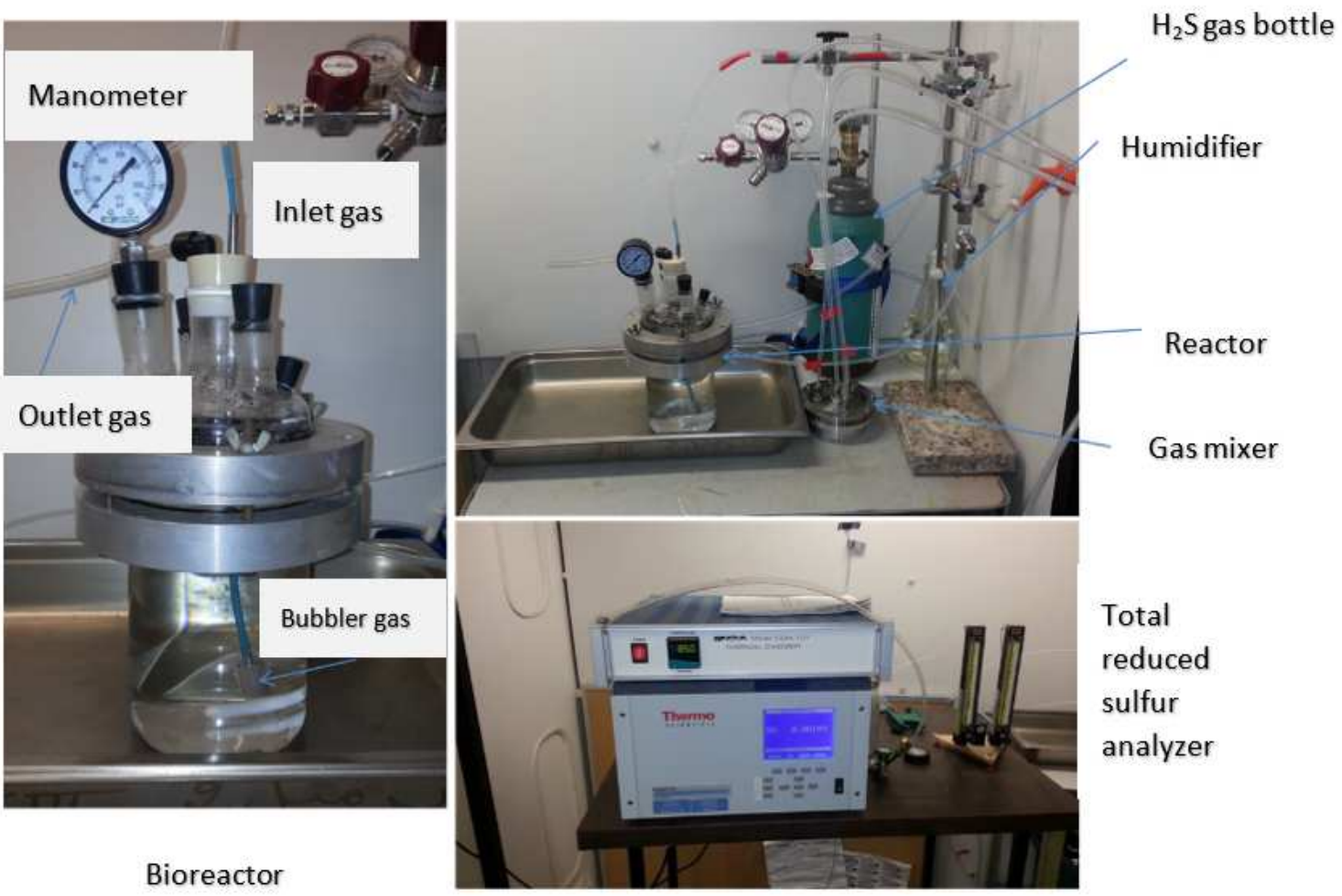


Figure 1

A laboratory scale bioreactor.

Figure 2

Stainless-steel pall rings (DN16).

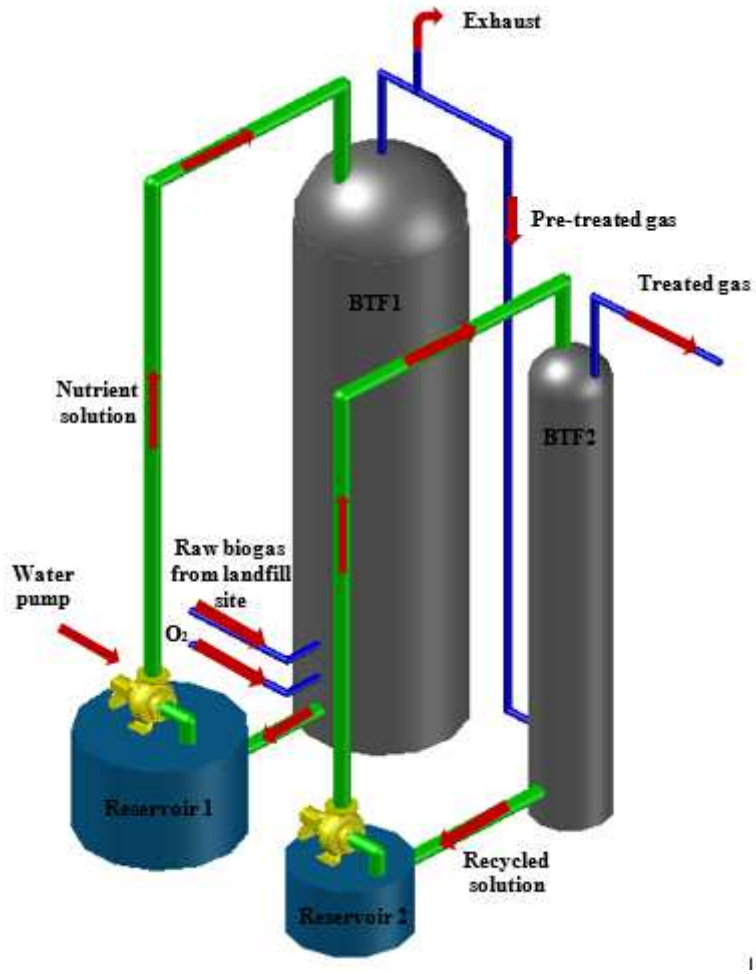


Figure 3

Pilot-scale biotrickling filter in landfill site packed with stainless-steel pall rings (DN16).

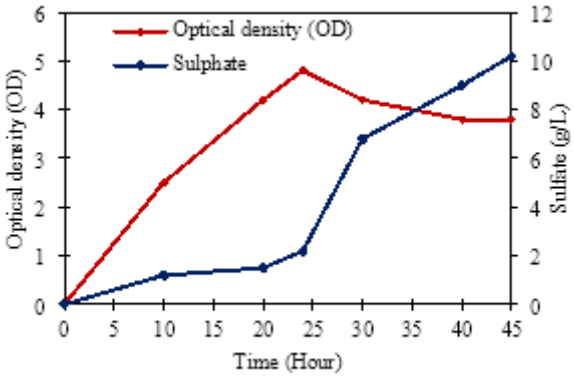


Figure 4

Evolution of sulfate and biomass as a function of time during acclimation tests (T =25° C, 60 rpm).

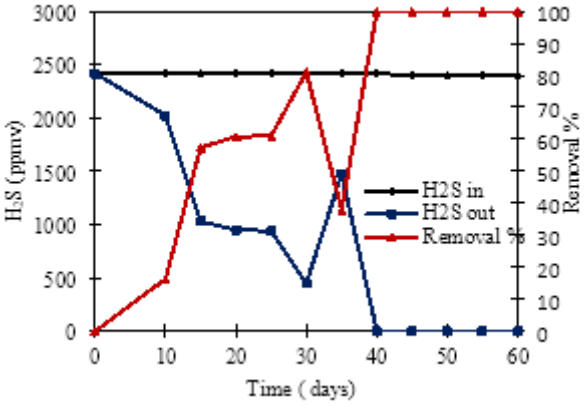
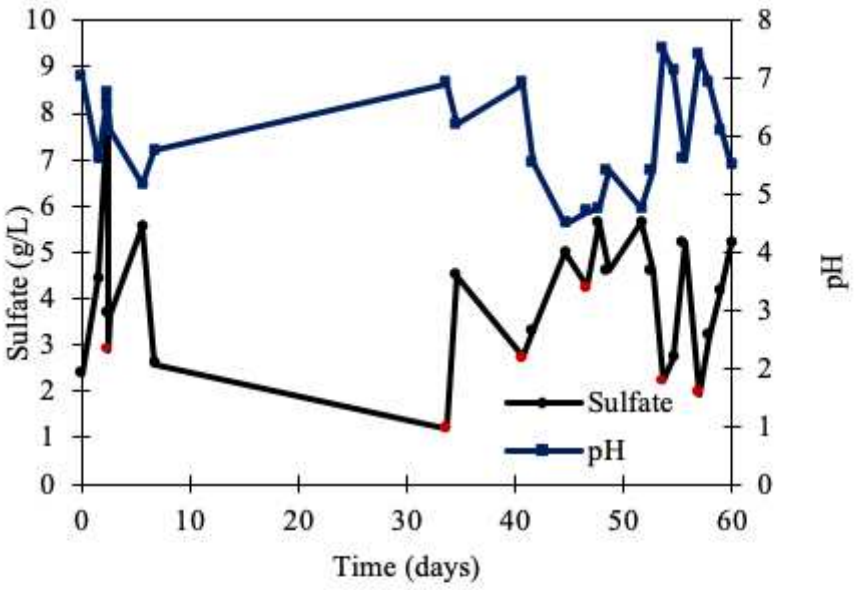
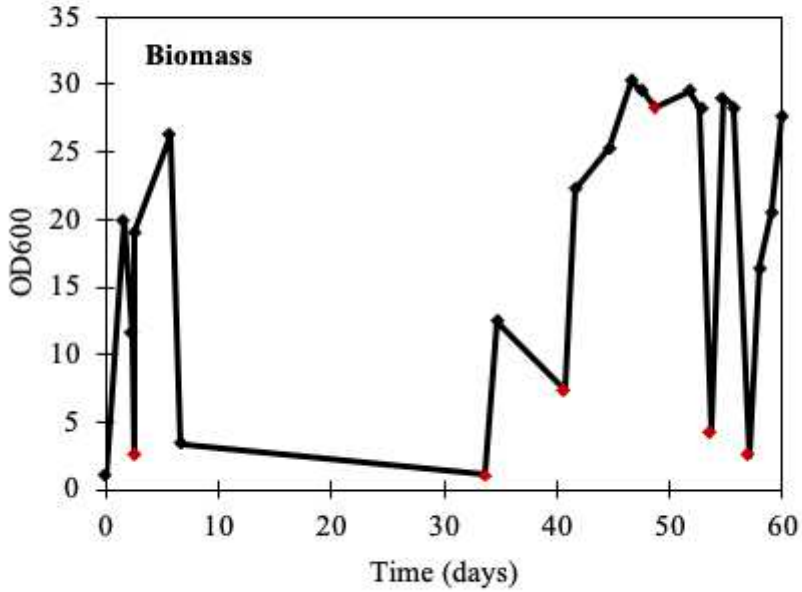


Figure 5

Monitoring the H2S concentration at the inlet and outlet of the bioreactor and the rate of elimination. (T = 25 °C, 60 rpm, Initial biomass: OD600 = 3.75).



(a)



(b)

Figure 6

Evolution of pH, sulfate ion concentration and biomass concentration (T = 25 ° C, 60 rpm, Initial biomass: OD600 = 3.75). The points in red correspond to the times when the culture medium was renewed.

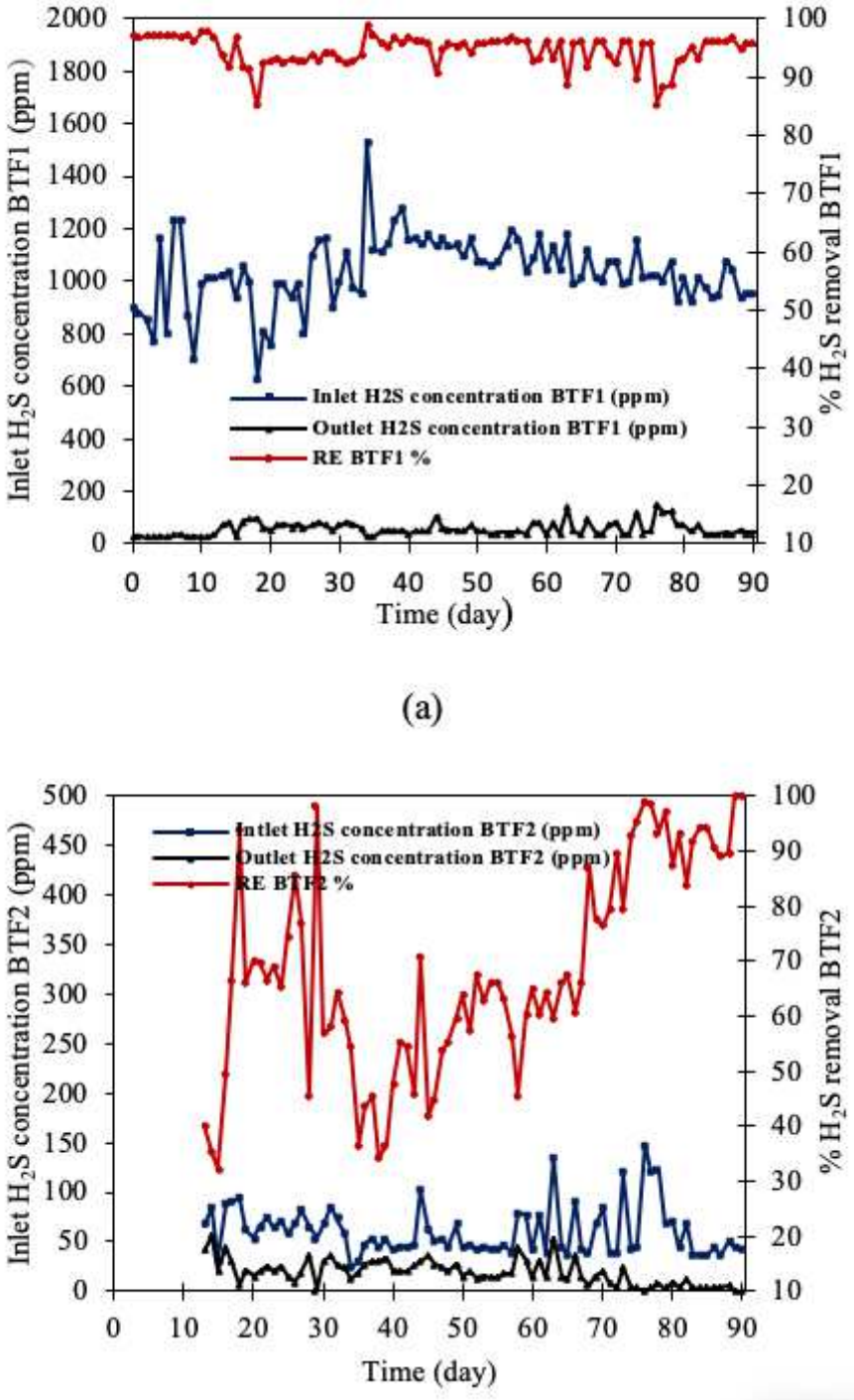


Figure 7

The removal efficiency of H2S in the two biotrickling filters. (a) Biotrickling filter 1, and (b) Biotrickling filter 2.

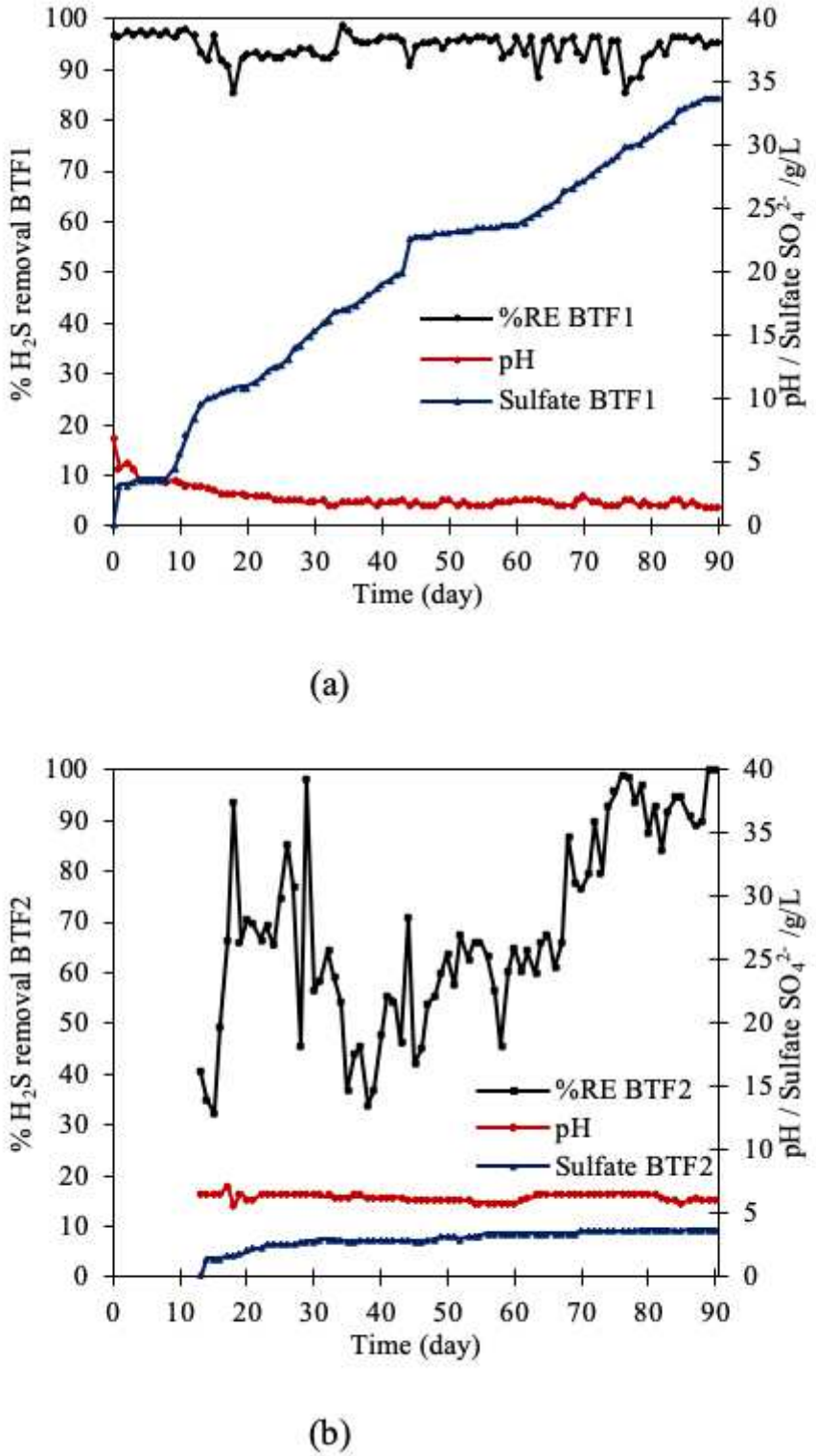


Figure 8

The removal efficiency of H2S, cumulative sulfate SO42- /g/L and pH changes in two biotrickling filters. (a) Biotrickling filter 1, and (b) Biotrickling filter 2.

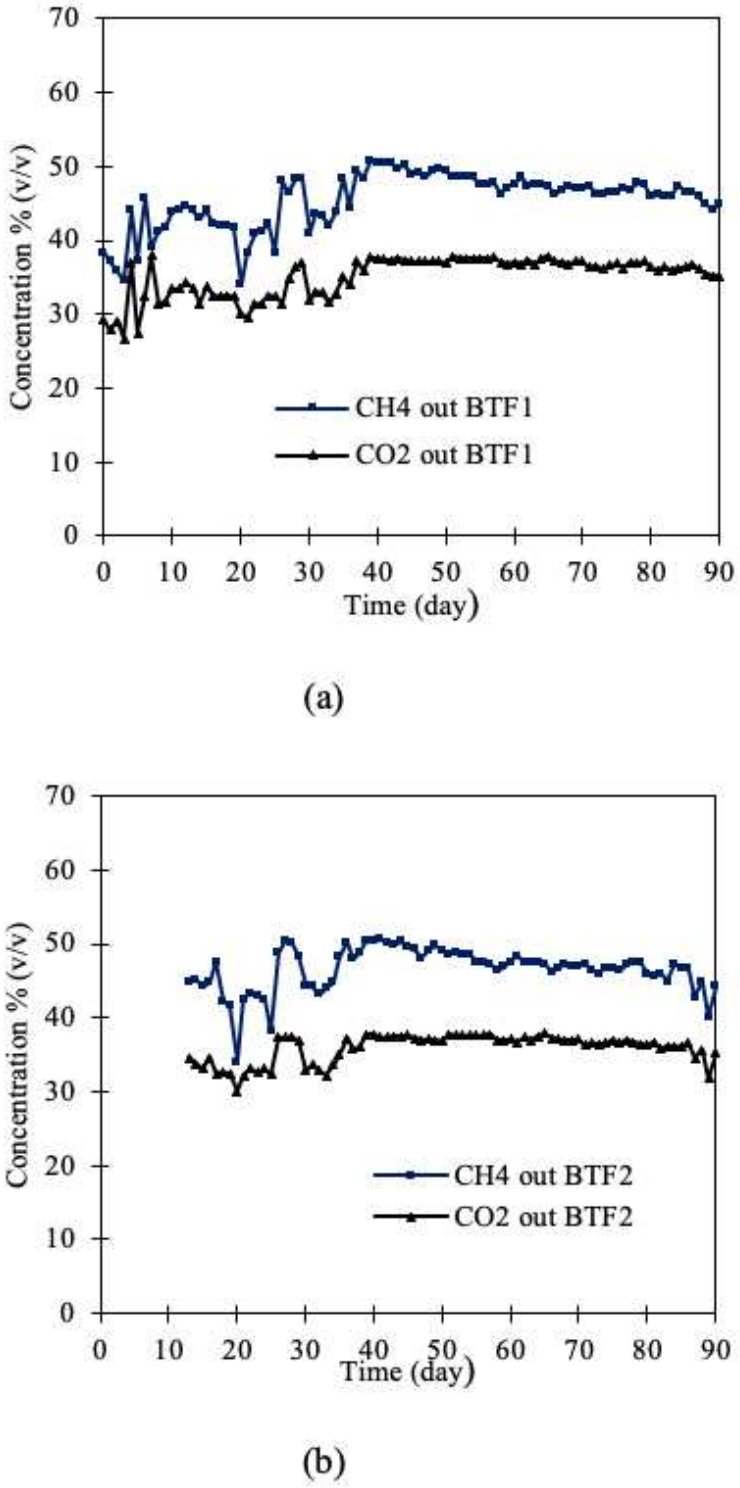


Figure 9

Methane and carbon dioxide composition in % (v/v). (a) Biotrickling filter 1, and (b) Biotrickling filter 2.

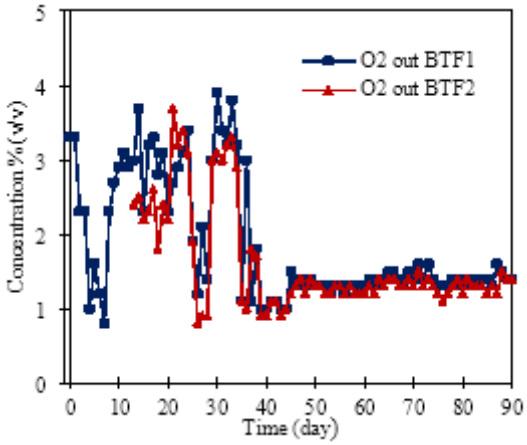


Figure 10

Oxygen concentration during operation of two biotrickling filters.

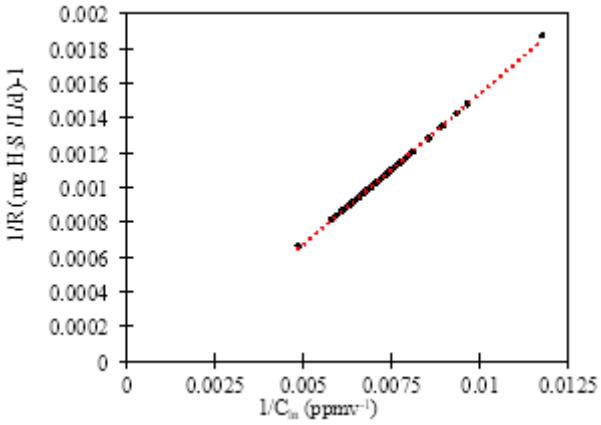


Figure 11

The linear relationship between ¹⁄R and ¹⁄C_In for H2S degradation in the pilot scale biotrickling