RESEARCH ARTICLE

Modelling of the biofiltration of reduced sulphur compounds through biotrickling filters connected in series: Effect of H_2S

Javier Silva[1] ✉ **· Marjorie Morales[1] · Manuel Cáceres[1] · Paulina Morales[1] · Germán Aroca[1]**

1 Pontificia Universidad Católica de Valparaíso, Facultad de Ingeniería, Escuela de Ingeniería Bioquímica, Valparaíso, Chile

✉ Corresponding author: javier.silva@ucv.cl

Abstract

Background: The behaviour of two biotrickling filters connected in serie (BTF) inoculated with *Acidithiobacillus thiooxidans* and *Thiobacillus thioparus*, biodegrading hydrogen sulphide (H_2S) and dimethyl sulphide (DMS) simultaneously were studied. A model which considers gas to liquid mass transfer and biooxidation in the biofilm attached to the support is developed. Additionally, a fixed bed biotrickling filter where the microorganism is immobilized in a biofilm which degrades a mixture of H_2S and DMS is implemented. Validation of the model was carried out using experimental data obtained at different H_2S and DMS loads. **Results:** The inhibitory effect caused by the presence of H_2S on the DMS is observed, which is evidenced by the decrease of the DMS removal efficiency from 80 to 27%, due to the preference that *T. thioparus* has by simple metabolism. H_2S is not affected by the DMS, with removal efficiencies of 95 to 97%, but it decreases at high concentrations of the compound, due to the inhibition of metabolism by high H_2S input loads. The model which describes the BFT fits successfully with the experimental results and it has a high sensitivity to inhibition parameters. **Conclusions:** It is shown that the microorganism has a high affinity for H_2S, producing substrate inhibition when the concentration is high. The H_2S is able to inhibit the DMS biooxidation, whereas the DMS does not affect the H_2S biooxidation.

Keywords: biofiltration, DMS, hydrogen sulphide, mathematical model, *Thiobacillus thioparus*

INTRODUCTION

The Volatile Reduced Sulphur Compounds (VRSC) are composed mainly by hydrogen sulphide (H_2S), methyl mercaptan (MM), dimethyl sulphide (DMS) and dimethyl disulphide (DMDS) which are found frequently in processes where heating or anaerobic decomposition of organic matter occurs (Mudliar et al. 2010). In general, these compounds have adverse health effects, so their emissions are limited in many countries (Shareefdeen et al. 2002).

Biofiltration systems are an attractive alternative to remove these compounds from contaminated air streams, using the ability of aerobic sulphur oxidizing microorganisms to transform them into less-polluting compounds or to incorporate them into biomass. In these systems, the contaminant diffuses from the gas phase to a biofilm, where the contaminants are oxidized by the microorganisms and used as carbon or energy source.

The H_2S is one of the most studied VRSC in biofiltration systems. When it is present in a mixture of VRSC, it is degraded preferentially because it is more soluble and it is oxidized more easily that other

VRSC (Rappert and Müller, 2005). Furthermore, the degradation of MM, DMS and DMDS is more efficient at pH 7 and decreases at lower pH where the removal of H_2S is enhanced (Jin et al. 2007). It has been proposed that the degradation occurs preferentially in the order: H_2S, MM, DMS and DMDS (Cho et al. 1991). In addition, H_2S inhibits the biooxidation of VRSC, including DMS (Li et al. 2003).

Considering this pattern of biooxidation a system based on two biofilters operating in series has been proposed for treating a mixture of these compounds (Sercu et al. 2005). In the first biofilters at low pH most of the H_2S is biooxidized, while in the second biofilter, the others VRSC and the remaining H_2S are removed at neutrophilic conditions. This configuration had been tested in several works (Pinjing et al. 2001; Ruokojärvi et al. 2001). There are many models available in literature to describe systems of biofiltration where one substrate inhibits the biooxidation of another. In such models, a simultaneous removal of substrates is considered. Álvarez-Hornos et al. (2009) proposed a transient state model for the biofiltration of ethyl acetate and toluene individually and in mixtures; Ikemoto et al. (2006) developed a model that considers the inhibitory effect of nutrient stimulation and hydrophilic VOC inhibition. Dupasquier et al. (2002) developed a model for two substrates in which there is co-metabolism of methyl tert-butyl ether and pentane, this model considers that the ether needs the presence of pentane to be used as carbon source, modelling that effect using the kinetics described by Arcangeli and Arvin (1997), which considers the competitive inhibition and a stimulating effect of pentane. Zhang et al. (2008) studied the effect of methanol in the removal efficiency of DMS in a biofilter, where the methanol limits the biodegradation of DMS and it is described using a competitive model of inhibition on DMS. Recently, Ramirez et al. (2011) show that when the H_2S reaches 11.1 g·S $m^{-3}h^{-1}$, the biooxidation of DMS is inhibited over 97%, and when the inlet load of H_2S is up to 31.7 g·S $m^{-3}h^{-1}$ the removal capacity of the other VRCS does not decrease. On the other hand, a high input concentration of H_2S (over 94 g·S $m^{-3}h^{-1}$) produces a decrease in the removal capacity.

The purpose of this work is to develop and validate a model of the simultaneous biooxidation of DMS and H_2S in a biotrickling filter, and to determinate the kinetics of the biooxidation of H_2S and the effect of H_2S on the kinetic of biooxidation of DMS. The model is validated using data collected from the second BTF in a biofiltration system composed by two BTF, the first inoculated with *A. thiooxidans* for the biooxidation of H_2S, and the second inoculated with *T. thioparus*, for the biooxidation of DMS and the remaining H_2S.

MATERIALS AND METHODS

Microorganisms and media composition

A. thiooxidans KS-1 and *T. thioparus* ATCC 23645 were used to inoculate the BTFs. The culture medium for *A. thiooxidans* was ATCC 290 and for *T. thioparus* was ATCC M290 both with 10 g/L of sodium thiosulphate which was used as energy source. Incubation conditions were 30ºC and 200 rpm. A volume of 0.4 L of culture of each microorganism growing in exponential phase was used for inoculation of the media support in the column by recirculating it through the packing material. The recirculating medium (2 L) was changed every two days.

Experimental setup and operation

The experimental system consists of two biotrickling filters connected in series, both made with acrylic tubes, diameter of 6.3 cm in diameter and 80 cm height, with a packed volume of 1.2 L. Polypropylene rings of 1 cm of height and 1 cm of diameter (density 280 kg/m^3) were used as packing material with a specific surface of 300 m^{-1}. Temperature was controlled by recirculating nutrient medium at 30ºC and 0.6 cm^3s^{-1}. The first column was inoculated with *A. thiooxidans* and operates at acid pH to remove most of the H_2S. The second column was inoculated with *T. thioparus* at neutral pH to remove the DMS and the remaining H_2S. The columns are provided with three sample ports with septa, separated by 10 cm.

H_2S was generated by mixing equimolar solutions of Na_2S and HCl. Humidified air (0.5 L min^{-1}) containing H_2S was fed at the bottom of the biotricking filter. DMS in gaseous phase at different concentrations was generated using a dynamic system proposed by Smet et al. (1998) (Figure 1). The diffusion system consist in a vessel that contains liquid DMS, connected by a capillary tube (i.d. 0.5 mm, length 5 cm) to a stainless steel spiral capillary tube (3 m length, id. 0.5 mm), submerged in a

thermo regulated water bath. A constant flow of 0.5 L min^{-1} of air is supplied through the capillary tube, the control of the temperature allows the control the DMS concentration in the gas stream.

Air containing H_2S and DMS at different concentrations was fed into the columns using an empty bed resident time (EBRT) of 120 sec. A solution of culture media (ATCC 290) without energy source was recirculated at 50 cm^3 min-1 at each BTF. H_2S was fed with input load until 11.1 g·S m^{-3}h^{-1} whereas DMS was fed with input load of 11.5 g·S m^{-3}h^{-1} at different H_2S input loads. The initial measured pH was 3.5 in the first column and 7.43 for the second column, whereas the sulphate concentration was 9 g L^{-1} for the first column and 3.1 g L^{-1} for the second column.

After reaching steady state; constant removal efficiency and elimination capacity of the biotrickling filter were determined, measurements of H_2S and DMS along the column were made during 3 days at steady state.

Analytical methods

A Gas Chromatograph Clarus 500 GC equipped with a flame photometric detector and an S-Supelpack column was used for the determination of H_2S and DMS, samples of 0.5 cm^3 of gas were used for the determinations. This equipment has a minimum detectable quantity of $1 \cdot 10^{-11}$ g S s^{-1}. Acidification of the medium due to the formation of sulphates was monitored by measuring pH in the recirculated solution using a model A20518 Hanna meter.

The amount of biomass in the rings was measured by taking the biomass from the ring by sonication for 10 min at 47 KHz. The live and death cells were counted by the following procedure: a staining solution was prepared using 1 cm^3 of ethidium bromide and 1 cm^3 of acridine orange, 750 μL of the cell suspension were mixed with 50 μL of the dye solution by using ultrasound, 10 μL of the sample was observed using a microscope of epifluorescence (Nikon model DS-brand Fi1) and the non-viable and viable cells were counted.

The concentration of sulfate was measured using the turbidimetric method with barium chloride (Clesceri et al. 1999).

Mathematical model

The bioxidation is described by a model that accounts for mass transfer and chemical degradation. The biotrickling filter is modelled as a fixed bed with a packing material that supports the microorganism in the form of biofilms. When air contaminated with H_2S and DMS flows throughout the column, H_2S and DMS are transferred from the gas phase to the liquid phase where they diffuse to the biofilm; there they are oxidized by the microbial activity.

Assumptions

The following assumptions for the model development were considered:

Steady-state operation; therefore the absorption of H_2S and DMS on the packing material is in equilibrium and should not be considered in the mass balance. Temperature and pH are constants.

The biomass accumulation rate in the reactor is small compared to the biodegradation rate of DMS and H_2S, therefore, mass balance for biomass will not be performed. No biofilm growth is assumed.

Oxygen is present in excess in relation to the DMS and H_2S, and the microorganism growth is not limited by this element.

The biofilm coating is formed in the surface of the packing. Due to the very small thickness of the coating, mass transfer is assumed perpendicular to the gas flow.

The concentration of H₂S and DMS in the interface is calculated using Henry's law, assuming the distribution coefficient similar to water's.

The effective diffusivity of the compounds in the biofilm is similar to the diffusivity of the compounds in water, thus the effective diffusivity can be calculated by applying a correction factor on the water diffusivity.

The thickness of the biofilm is relatively small in relation to the curvature of the media; therefore modelling can be performed using planar geometry.

The mixture of gases in the biotrickling filter can be described using a dispersion model.

The microbial activity of biooxidation of the suspended cells in the recirculating medium is negligible.

At the gas-liquid film interface, equilibrium is assumed to occur for H₂S and DMS using air/water partition coefficients.

Equations

The model used is based in the work of Spigno et al. (2004) that represent the convection-dispersion transport from the gas phase to the liquid phase and the subsequent biooxidation in the biofilm. The growth of biomass with time was considered negligible. However, it is considered that there is a profile of active biomass along the biotrickling filter. This profile was modelled using experimental measurements of the active biomass along the column (Silva et al. 2010). The dimensionless expression that fit those results for biomass profile along the column is:

$$f(\zeta) = \frac{n_V}{n_{max}} = -0.9\zeta^2 - 6.4 \cdot 10^{-2}\zeta + 1$$

[Equation 1]

This model was obtained by correlating the viable cell count (n_v) divided by the maximum cell count (n_{max}) determined at the input of the biotrickling filter, with the dimensionless length of the column. ζ is the dimensionless axial co-ordinate along the bed height.

Oyarzún et al. (2003) show that when the input concentration in the BTF is about 80 g·S m^{-3}h^{-1}, the elimination capacity decrease, therefore, the kinetics of biooxidation of H₂S was described by a Haldane type kinetics.

$$\kappa_{H_2S} = \frac{1}{Y_{x/H_2S}} \cdot \frac{C_{bH_2S}}{\sigma_{H_2S} \cdot \left(1 + \dfrac{C_{bH_2S}}{\iota_{H_2S}}\right) + C_{bH_2S}}$$

[Equation 2]

Due to the fact that H₂S inhibits the biooxidation of DMS (Li et al. 2003; Ramirez et al. 2011), a competitive kinetics models is proposed for the DMS biooxidation rate.

$$\kappa_{DMS} = \frac{1}{Y_{X/DMS}} \cdot \frac{C_{bDMS}}{\sigma_{DMS} \cdot \left(1 + \frac{C_{bH_2S}}{\iota_{DMS}}\right) + C_{bDMS}}$$

[Equation 3]

Where σ is the dimensionless Monod constant for each component, Y is the biomass yield coefficient for each component ι_{H_2S} is the substrate inhibition constant for the H_2S and ι_{DMS} is the inhibition constant for the H_2S over the DMS.

The dimensionless concentrations in the gas, in the biofilm and in the biomass are defined as:

$$C_{gi} = \frac{c_{g_i}}{c_{g_i}^{in}} \quad C_{bi} = \frac{c_{b_i}}{c_{b_i}^{in}}$$

[Equation 4]

Where c_{gi} is the concentration in the gas phase for the i compound (g m^{-3}), c_{bi} is the concentration in the biofilm for the i compound (g m^{-3}), c_g^{in} is the inlet concentration of the i compound in the gas phase (g m^{-3}) and c_b^{in} is the inlet concentration of the i compound in the biofilm (g m^{-3}).

The dimensionless axial axe systems along the biotrickling filter are defined as:

$$\zeta = \frac{z}{H}$$

[Equation 5]

Where z is the axial axe and *H* is the biofilter height (m).

The dimensionless Monod constant and the constant of inhibition are expressed as:

$$\sigma_i = \frac{K_{si}}{c_{b_i}^{in}} \quad \iota_i = \frac{K_{li}}{c_{b_i}^{in}}$$

[Equation 6]

Where K_{Si} is the Monod constant for the i component and K_{li} is the inhibition constant for each component (g m^{-3}).

The model parameters are taken or derived from other works. The values of these parameters are shown in Table 1.

The specific surface, the porosity and the surface covered by biofilm were determined experimentally, the latest was determined experimentally measuring the cells over the packed material. The inhibition

constants were adjusted based on experimental data obtained in this work. The dispersion coefficients were calculated using the correlation used by Delgado (2006).

Numeric solution

Because the model includes two phases and two substrates inside the biotrickling filter, the solution was solved by finite differences for each interval simultaneously applying the Newton-Raphson method for multiple variables. Applying this method to the boundary conditions of the liquid phase yields a system of nonlinear equations that have the same number of dependent variable as the number of intervals. An algorithm written in MatLab 7.0 was developed. The solution is based on the work of Deshusses et al. (1995). In the present work, the biotrickling filter model was discretized using central second-order finite differences for dividing into n layers the column and the non-lineal system equations developed was solved simultaneously using the Newpton-Raphson method for multiples variables. At the gas-liquid film interface, equilibrium is assumed to occur for H_2S and DMS using air/water partition coefficients.

RESULTS

Using loads of 0.4 g·S $m^{-3}h^{-1}$ of H_2S and 0.5 g·S $m^{-3}h^{-1}$ of DMS, elimination capacities of 48% for both compounds were observed in the first BTF inoculated with *A. thiooxidans*. This work focuses on the behaviour of the second BTF inoculated with *T. thioparus*. Figure 2 shows the removal profiles of DMS, input load of 11.2 g·S $m^{-3}h^{-1}$ at different input loads of H_2S (0, 0.1, 0.4, 2.1, 2.8, 8.4 and 11.1 g·S $m^{-3}h^{-1}$) with a EBRT of 120 sec in the second BTF. Removal efficiencies of 25.8, 21.7, 15.2, 16.1 and 10.4%, were obtained respectively (Figure 3). When the H_2S is not fed in the BTF, the DMS elimination capacity is 9 g·S $m^{-3}h^{-1}$ (80.3% removal efficiency) and when the H_2S input load is 2.8 g·S $m^{-3}h^{-1}$ the DMS elimination capacity decreases to 1.8 g·S $m^{-3}h^{-1}$, reaching 1.1 g·S $m^{-3}h^{-1}$ at the highest load of H_2S tested.

Figure 4 shows the H_2S removal profiles along the column inoculated with *T. thioparus* with a EBRT of 120s and an input concentration of DMS of 0.4 g·S $m^{-3}h^{-1}$. It is possible to observe that the removal efficiency is almost complete when the concentration of H_2S in the inlet is up to 2.8 g·S $m^{-3}h^{-1}$, with removal efficiencies ranging between 95% and 98%. However, when we have high H_2S concentrations (> 8.4 g·S $m^{-3}h^{-1}$ or 210 ppm) the removal efficiency decreases due to inhibition by substrate (there is an important loss of the removal efficiency between 2.8 g·S $m^{-3}h^{-1}$ (98% removal efficiency) and 11.1 g·S $m^{-3}h^{-1}$ of H_2S (10% removal efficiency)). Previous works shows similar results (Aroca et al. 2007).

Figure 5 shows the values obtained from *T. thioparus* inlet cell counts using epifluorescence microscopy. It is possible to observe that there are no important variations in the inlet cell count due to changes in the H_2S inlet concentration and only axial biomass profiles are observed by Silva et al. (2010).

Low variations of pH were observed in the recirculating medium due to changes in the input concentrations of H_2S (Figure 6). The most important variation of pH and sulphate production was observed when high concentration of H_2S was fed into the column. It would indicate that there is no limitation for availability of H_2S from the gas phase and it also suggest that the biooxidation kinetics limits the biooxidation in the BTF. Silva et al (2010) had shown low Thiele modules for this system.

Figure 7, Figure 8, Figure 9 and Figure 10 show simulations of the BTF behaviour for different inlets concentrations of H_2S and DMS. In previous report similar elimination capacities were observed (Cáceres et al. 2010), however Ramirez et al. (2011) was able to achieved removal efficiency 67% feeding 12.8 g·S $m^{-3}h^{-1}$ of H_2S and 0.9 g·S $m^{-3}h^{-1}$ of DMS with EBRT of 59 sec. No reports are found of removal profiles of simultaneous biofiltration of H_2S and DMS. The inhibition constants estimated were $6 \cdot 10^{-3}$ g·S m^{-3} for the DMS and 0.05 g·S m^{-3} for the H_2S, obtaining a good fit between the model and the experimental data, also obtaining that the kinetics models describe correctly the BTF behaviour with a correlation coefficient of 0.88, 0.8, 0.82 and 0.9 for DMS and 0.84, 0.89, 0.85 and 0.92 for H_2S in Figure 7, Figure 8, Figure 9 and Figure 10, respectively.

Figure 11 shows a sensitivity analysis of the DMS inhibition constant when the system was fed with 1.2 g·S m^{-3}h^{-1} of DMS and 0.1 g·S m^{-3}h^{-1} of H_2S. The model is highly sensitive to inhibition constant and due to its low value this parameter describes the severe effect that H_2S produces over the DMS, however, in the simulation develops in this work, good fits were achieved for the inhibition parameter determined.

CONCLUDING REMARKS

The model describes the simultaneous biooxidation of DMS and H_2S in a biotrickling filter with a biofilm of *T. thioparus*. The kinetic expression proposed and the inhibition mechanism considered seems to be adequate to describe the inhibitory effect of the H_2S over the DMS. The model shows high sensitivity to the adjustment of the inhibition constant so that it describes the strong influence of the H_2S on the biooxidation of DMS.

The results showed the preference of *T. thioparus* for using H_2S as an energy source instead of DMS when it is present in the mixture. From a thermodynamic point of view, according to the Van't Hoff equation, the H_2S and DMS have oxidation free energies of -1.4 · 10^{-3} J x mol^{-1} and -2 x 10^{-4} cal mol^{-1}, respectively (Perry and Green, 2008) so the reaction energetically favours the first compound. Similar results were obtained by Kelly and Smith (1990), they proposed that in *Thiobacillus* species H_2S is oxidized preferentially than DMS.

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FIGURES

Fig. 1 System H$_2$S and DMS biofiltration through *A. thiooxidans* and *T. thioparus*: (A) air compressor; (B) manometer; (C) flow meter; (D) column-generating filler; (E) Na$_2$S drum; (F) drum HCl; (G) collecting the remaining liquid; (H) container lung; (I) container of recirculation; (J) biofilter inoculate *A. thiooxidans*, (K) current output first biofilter, (H) recipient; (L) pump; (N) current rich in H$_2$S; (O) air contaminated with H$_2$S and DMS; (P) sampling; (Q) thermocouple; (R) tank DMS; (S) biofilter inoculated with *T. thioparus.*

Fig. 2 Removal profiles of DMS for different H₂S inlet concentrations in the BTF inoculated with *T. thioparus*.
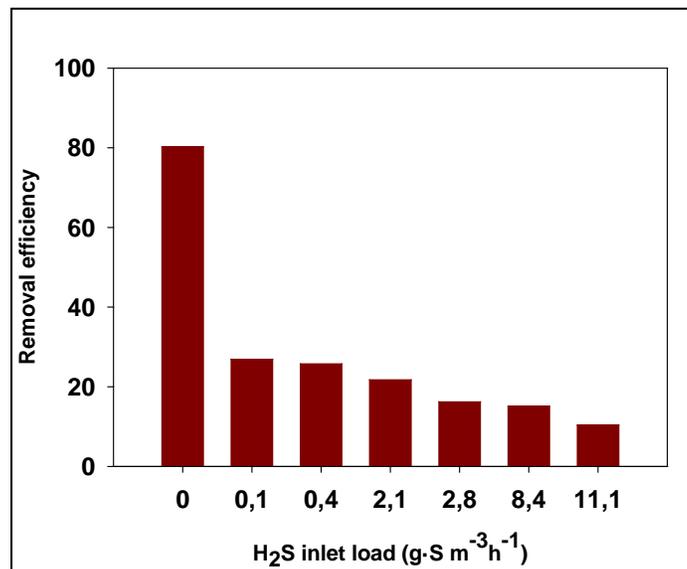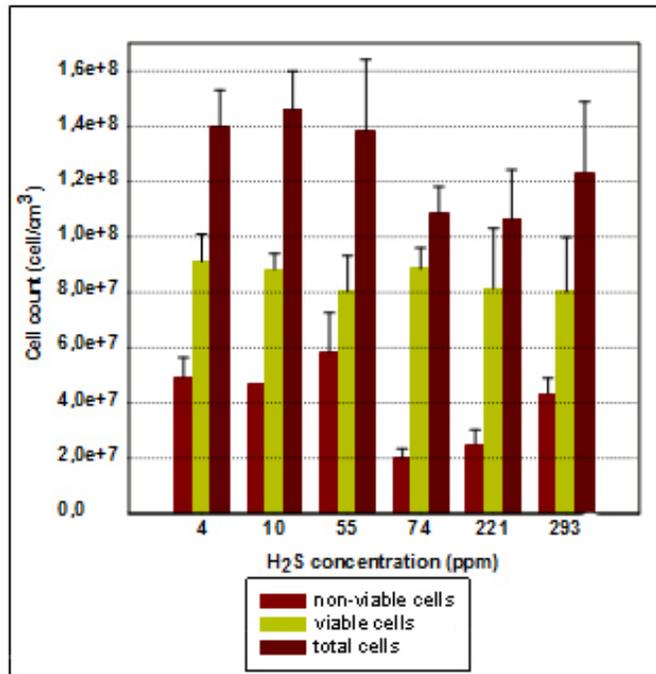

Fig. 3 Removal efficiency of DMS for each inlet concentration of H₂S in the BTF inoculated with *T. thioparus*.
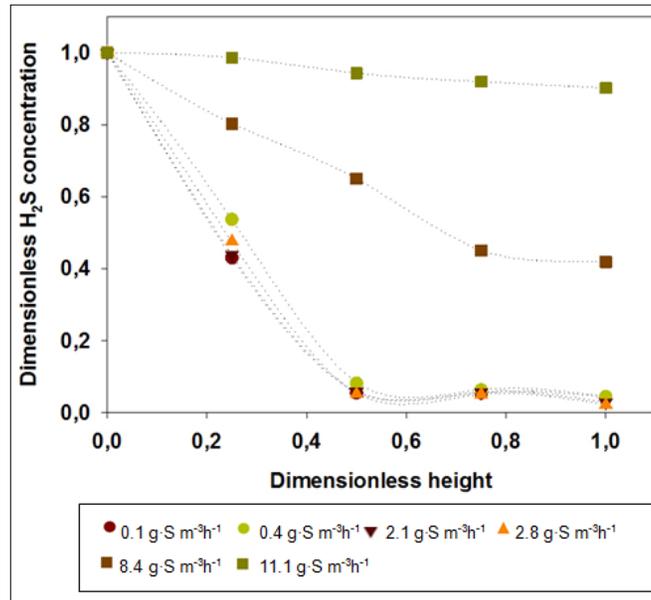
Fig. 4 H_2S Removal profiles for different H_2S inlet concentrations in the BTF inoculated with *T. thioparus*.

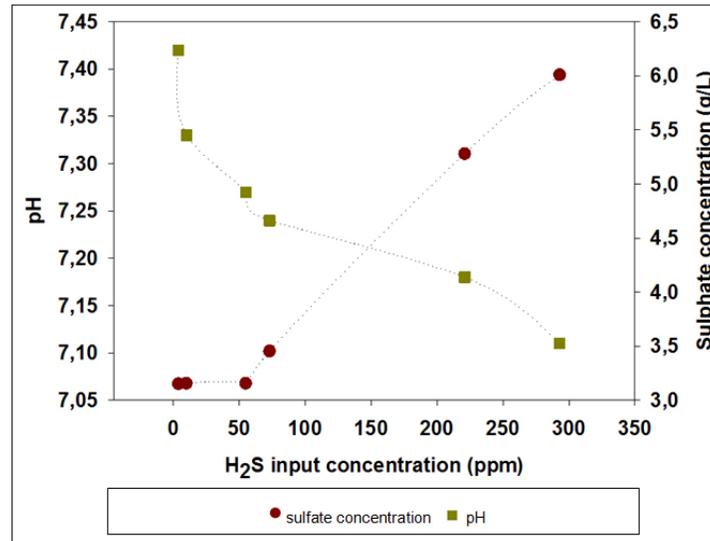**Fig. 5 Cell count for each input concentration of H_2S in the BTF inoculated with *T. thioparus*.**

Fig. 6 pH and sulphate concentration for each H₂S input concentration in the recirculating medium for the BTF inoculated with *T. thioparus.*
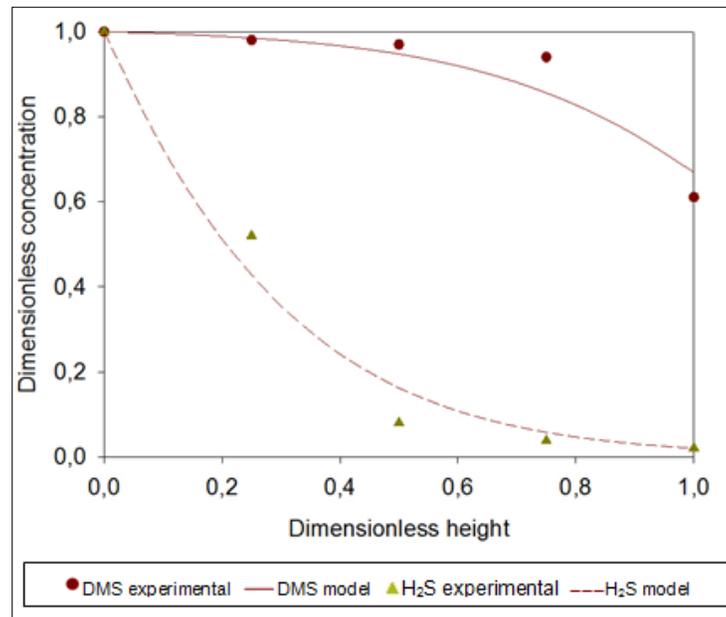

Fig. 7 Behaviour of the BTF and the simulation for an input concentration of 0.4 g·S m-3h-1 of DMS and 0.1 g·S m-3h-1 of H2S considering biomass profile.
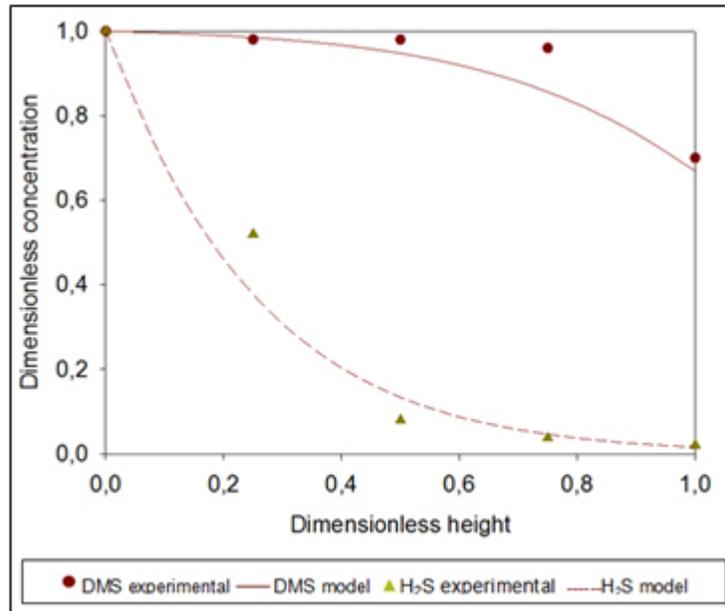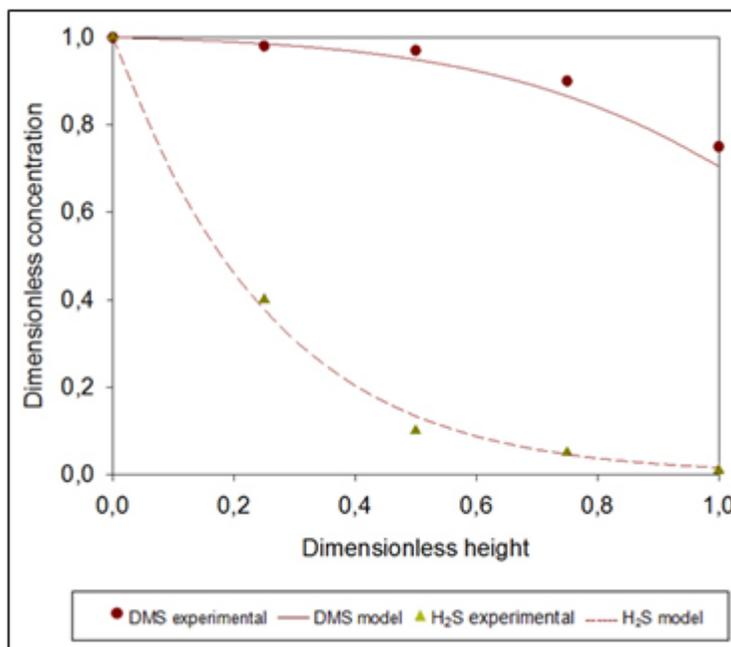
Fig. 8 Behaviour of the biotrickling filter and the simulation for an input concentration of 0.2 g·S m^{-3}h^{-1} of DMS and 2.1 g·S m^{-3}h^{-1} of H_2S considering biomass profile.

Fig. 9 Behaviour of the biotrickling filter and the simulation for an input concentration of 0.8 g·S m^{-3}h^{-1} of DMS and 0.1 g·S m^{-3}h^{-1} of H_2S considering biomass profile.

Fig. 10 Behaviour of the biotrickling filter and the simulation for an input concentration of 0.8 g·S m⁻³h⁻¹ DMS and 0.4 g·S m⁻³h⁻¹ of H₂S considering biomass profile.
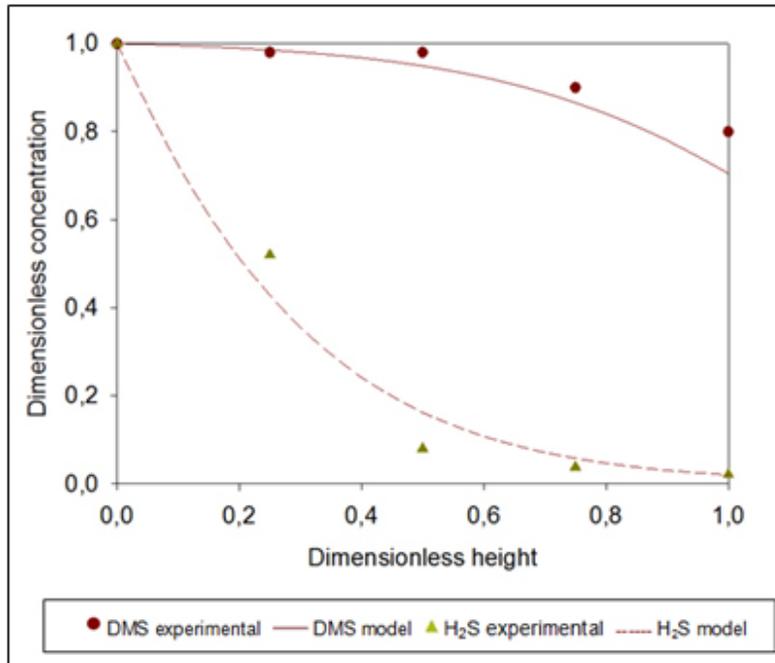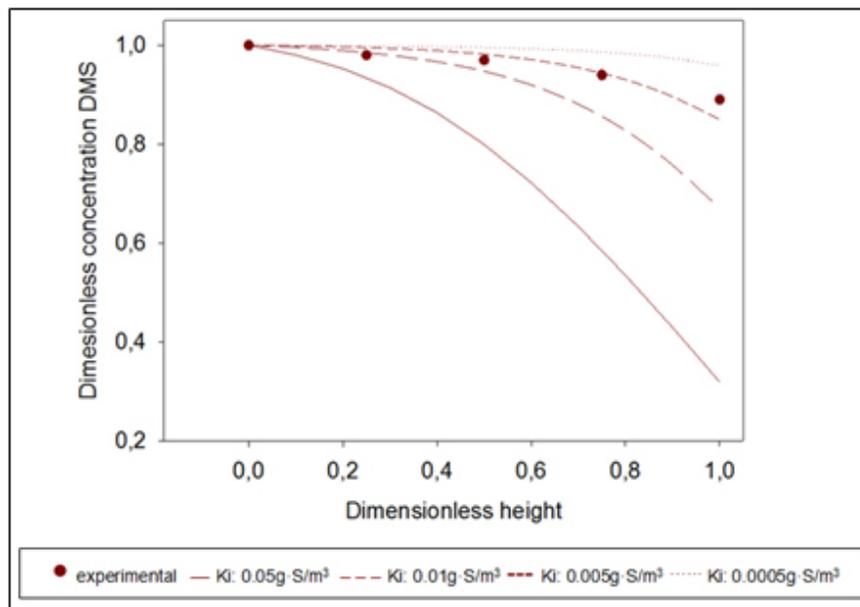

Fig. 11 Effect of the inhibition constant in the DMS removal.

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TABLES

Table 1. Parameters of the model for *T. thioparus* degraded H_2S and DMS.

Parameter	Value	Reference
Diffusion coefficient of H_2S in water (D_{H2S})	$1.93 \times 10^{-9} m^2 s^{-1}$	Tamimi et al. 1994
Diffusion coefficient of DMS in water (D_{DMS})	$1.51 \times 10^{-9} m^2 s^{-1}$	Nielsen et al. 2009
Partition coefficient air - water H_2S (m)	0.47	Dobryakov and Vitenberg, 2006
Partition coefficient air - water DMS (m)	0.07	Dobryakov and Vitenberg, 2006
Specific surface (a_s)	$300 m^{-1}$	Experimental
Porosity(ε)	0.3	Experimental
Surface covered by biofilm (α)	0.4	Experimental
Thickness (δ)	$30 \mu m$	Spigno et al. 2004
Maximum specific growth rate H_2S (μ_{max})	$0.045 h^{-1}$	Ramírez et al. 2009
Monod constant H_2S	$30.3 g m^{-3}$	Ramírez et al. 2009
Yield H_2S	0.03	Ramírez et al. 2009
Maximum specific growth rate DMS (μ_{max})	$0.015 h^{-1}$	Hayes et al. 2010
Monod constant DMS	$0.62 g m^{-3}$	Hayes et al. 2010
Yield DMS	0.05	Hayes et al. 2010