Removal Kinetic of Dimethyl Sulfide in a Biofilter with sugarcane bagasse as a packing material.

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Abstract :

The biological removal of Dimethyl Sulfide (DMS) was measured during 180 days in a biofilter inoculated with Hyphomicrobium VS and packed with sugarcane bagasse. During the operation of the biofilter the empty bed residence time (EBRT) was varied from 90 to 180 seconds and the inlet concentration of DMS from 12, 50, 100 and 200 ppmv operated at ambient lab conditions (T= 22^{o}C).

The removal performance data were collected and subsequently used in the determination of kinetics and modeling the DMS, a Michaelis- Menten type equation was applied and the half saturation parameter (Km), and the maximum volumetric elimination rate (r_m) were calculated.

The maximum elimination capacity (EC) of the biofilter was 5 g DMS/m^{3}h with a load of 10.40 g DMS/m ^{3} h, the maximum removal efficiency (RE) obtained was 97.6 % at 12 ppmv DMS inlet concentration, load of 0.62 g DMS/m ^{3} h and 180 s of EBRT.

Keywords: Dimethyl sulfide; *Hyphomicrobium* VS; biofilter; sugarcane bagasse

Introduction :

Dimethyl sulfide (DMS) is the most abundant biological sulphur compound emitted into the atmosphere and a major contributor to total sulphur emission in nature through biogenic processes [1]. It's characteristic unpleasant smell becomes highly odorous at higher concentrations, often when the source of the compound is the off-gases from pulp mills, oil refineries, manure and sewer systems, and wastewater treatment plants [2]. Since these volatiles have been identified as predominant odorants in the emission of a wide range of activities in the bio-industry. In the atmosphere, the photochemical oxidation of DMS promotes acid rain formation, bringing sulphate back to the earth [3]. Furthermore, DMS emissions also have important anthropogenic sources such as wastewater treatment plants and kraft pulp mills, which are notorious for their unpleasant odour.

Among the waste gas treatment technologies developed for VOSCs removal, biological technologies are gaining public attention owing to their low operational cost and absence of secondary waste stream. [4-9].

For gases containing mixtures of reduced sulphur compounds (RSC) , the preference of sulphur-oxidizing bacteria to use H_2S as an energy source over the rest of the RSC promotes low DMS consumption . A dual Biofiltration system constitutes an alternative for the treatment of RSC mixtures; the H_2S is degraded in the first biofilter and the remaining RSC, including DMS, in a second biofilter [6].

The use of bacteria in biotrickling filters (BTF) is a feasible way to eliminate low concentrations of reduced sulphur compounds in air streams such as those found in malodorous emissions (Odour threshold 1 ppbv). Biotrickling filtration include absorption of DMS into a biofilm where it is degraded by microorganisms.

Among the biotechnological waste gas techniques, biofiltration is the most common one. Contrary to bioscrubbers and biotrickling filters, biofilters are regularly used to treat off-gases in the bio-industry, e.g. in composting and rendering plants. In a biofilter, the gas to be treated is humidified and forced to flow through a bed packed with an organic carrier material, on which microorganisms are attached as a biofilm. Different research works done by previous investigators on the dimethyl sulfide biofiltration in laboratory scale show the feasibility of these methods [3],[10-12].

In case of biofiltration of DMS inoculation of the biofilter with specific cultures as been shown to increase the start up of the reactor significantly. A number of different microorganisms have been used for the inoculation of bioreactors removing dimethyl sulfide. These include mainly bacteria such as *Thiobacillus thioparus [7,9,10,13], AcidiThiobacillus [6], Pseudomonas fluorescens [14], Microbacterium [15] and Hyphomicrobium VS* [6,7,27]. The *Hyphomicrobium* species are able to utilize DMS as a carbon and energy source, *Hyphomicrobium* VS looks a promising organism for

application in biofiltration of air containing DMS. It can easily be cultured at a large scale on methanol, only a short period for adaptation to DMS was needed.

Media selection is critical in biofilter design. To operate efficiently, the media must provide a suitable environment for microbial growth and maintain a high porosity to allow air to flow easily. Critical properties of media material include porosity, moisture holding capacity, nutrient content, and slow decomposition [16]. In general, porous and non hydrophobic surfaces with high specific surface seem to facilitate or promote colonization by microorganisms and the subsequent formation of biofilms.

Moisture content of the packing has been identified as the most critical parameter to control in biofilters, the waste air is frequently humidified in packed towers before entering the biofilter. Most applications also have a sprinkling system for direct additional water supply onto the packed bed. Prehumidification in spray towers also removes particulate matter from the waste air, thus preventing clogging of the packed bed [17]

Packing materials used for biofiltration include polystyrene particles, peat, compost, granular activated carbon or porous inorganic matrix , sometimes coated with activated carbon. However, these supports pose problems of their disposal after utilization. An alternative lies in the use of agro-industrial by-products such as cassava bagasse or sugarcane bagasse, whose biotechnological valorization has been demonstrated. Numerous microorganisms able to grow on these natural supports [18].

Pure organic material like sugarcane bagasse is preferred instead of mixtures of organic and synthetic because then disposal is less a problem, looking for a locally available material which is now considered as “waste” using it as biofilters material is a way of upgrading and recycling the material. In addition, the possibility of using a waste as packing material in biofilters is particularly attractive from the environmental point of view. Sugarcane bagasse is available in a lot of countries such as Cuba, Brazil, Australia, Argentina and Mexico.

Sugarcane bagasse is an agricultural residue from industrial sugar extraction process. Although utilized in the sugar factories as fuel for the boilers, large quantities are accumulated in the mills, creating environmental problems. Recently, there is an increasing trend towards the utilization of sugarcane bagasse, as it represents a large and inexpensive source of raw material, which can be used as solid support also in several biotechnological processes [19]. Sugarcane bagasse is a residue composed approximately of 50% cellulose, 25% hemicellulose, and 25% lignin [20], therefore it is relatively resistant to biodegradation. There are some experiments with the use of sugarcane bagasse in biofiltration [18, 19], [21-26] but to our knowledge the use of this material for DMS biofiltration has not been reported yet.

The aim of this study was to evaluate the feasibility of using sugarcane bagasse as an alternative filter material for the biofiltration of air streams contaminated with dimethyl

sulphide and to determine the biodegradation kinetics, Km and *rm*, using the acquired experimental data

Methods :

Microorganisms and Media :

Hyphomicrobium VS [13,26] was grown using mineral medium (in g/L), containing K_2HPO_4 (3), KH_2PO_4 (3), NH_4Cl (3), $MgSO_4.7H_2O$ (0.5), and $FeSO_4.7\ H_2O$ (0.01) , all the compounds were dissolved in distilled water with the addition of 1% (v/v) methanol previous to sterilizing at 121^oC for 20 min at pH 7, with the addition of 1% (v/v) methanol. *Hyphomicrobium VS* was initially cultured by adding 50 µL of the strain (kept at – 80 ^{o} C in glycerol) to 5 mL of mineral medium containing 1% of methanol, and incubating the suspension for 5 days at 37 ^{o}C. For growing *Hyphomicrobium VS* on DMS, 20 mL of this pregrown culture was centrifuged and washed with sterile 0.9% NaCl twice and added to 1 L of mineral medium. This suspension was provided with 100 ppmv DMS in air (about 200 ml min^{-1}) until growth was visible. For starting the growth of *Hyphomicrobium VS* in a chemostat, 20 mL of the pregrown culture was added to 2 L of mineral medium. The chemostat was then aerated and supplied with fresh, sterile mineral medium at 75 mL / h and methanol at a 1% influent concentration.
The enrichment method for *Hypomicrobium VS* follow the experiences of Sercu et al [27].

Filter material :

Sugarcane bagasse, from Jose Marti Pilot Industry, Cuba, was used as support. It was sieved through 0.4 – 0.8 cm screens, washed with distilled water, dried at 80 $^\circ$ C for 24 h, and sterilized at 1MPa and 121 ^{o} C during 15 minutes.

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Analytical Methods :

Influent and effluent DMS gas concentrations were determined using a Varian 3700 chromatograph equipped with a flame ionization detector and a 30 m CP-SIL 5CB column (Chrompack, internal diameter 0.53 mm, film thickness 5 µm). Helium was used as the carrier gas at a flow rate of 40 ml/min and the temperatures used in the injector, oven, and detector were 70ºC, 80ºC and 90ºC, respectively. The detection limit for this analysis method was 0.05 ppmv of DMS.

A Pressure-Lok Precision Analytical Syringe (Alltech, Deerfield, IL) was used for injecting 1 mL gas samples. The pH of the liquid was measured with an electronic pH sensor (Jenway 3310).

Biofiltration experiments:

Continuous experiments (over a period of 180 days) to study the removal of dimethyl sulfide were carried out in a lab scale biofiltration column inoculated with *Hyphomicrobium VS*, the biofilter were set up using Plexiglas columns of 45 mm of inner diameter and 945 mm of height for a packed volume of 1.5 L. The column was filled with 115 g of sugarcane bagasse. Characteristics of bagasse have been reported by Ramírez-López et al [22] as a specific surface of 10000 m^2 m^{-3}, void fraction of 76 % and more than 290 x 10^6 particles per cubic meter.

The moisture content of the filter material was maintained at the desired level (50–70%) either by bubbling the influent synthetic polluted gas stream in a humidification unit, or by periodically distributing a mineral salts solution, by means of a spray nozzle at the top of the packing material, flowing downwards counter currently with the gas flow.The relative humidity of the waste air containing dimethyl sulfide was 99%.

The biofilter was inoculated with Hyphomicrobium VS (7.9 ± 0.16 x 10^{11} cel/g of dry bagasse) if the pH was lower than 6.5 it was adjusted to 7 by adding 1M NaOH manually to the sugarcane bagasse.. The air flow (dry air) was provided in upflow mode at 0.5 and 1L min^{-1} providing an empty bed residence time (EBRT) of 180 and 90 seconds. DMS was dosed in the air stream by a capillary diffusion system, as described by [30]. The system consists of one or more 4mL vessels containing the liquid DMS, placed in a thermostatic water bath and each connected with the main air stream with a diffusion capillary. A concentration gradient between a vessel and the upper outlet of the diffusion capillary forces the compound to diffuse through the capillary. The DMS mass flux to the air stream is dependent on the capillary dimensions, water bath temperature and total pressure in the main air stream. Additional overpressure is provided by forcing the main air stream through capillary tubing before entering the reactor, to minimize the effect of varying atmospheric pressures. In this case the concentration of DMS in the air stream was regulated by the number of vessels connected to the air stream. Gas sampling ports were provided in the tubing before and after the biofilter.

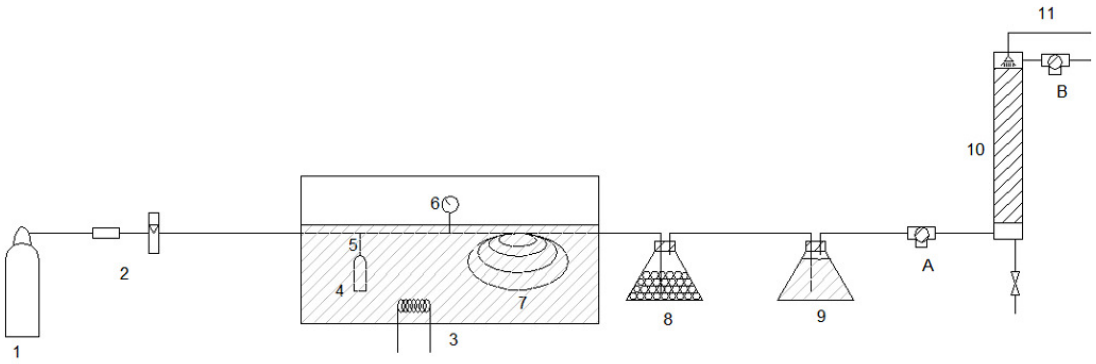

Figure.1 The set-up and design of the biofiltration system . **1** Dry air cylinder, **2** Mass Flow controller, **3** Constant temperature bath, **4** DMS vessel, **5** Capillary tube, **6** Pressure gauge, **7** Steel-tubing coils , **8** Mixed Chamber, **9** Humidifier, **10** Biofilter, **11** Water spray, **A** Inlet Sampling port, **B** Outlet Sampling port.

The performance of the biofilter was evaluated in terms of the removal efficiency (%) and the elimination capacity (EC) ($gm^3\ h^{-1}$) of the filter bed, which were estimated by the following equations:

Inlet loading rate, LR (gDMS m^3/h) = $Q*C_{in}$ / V *(1)*

Elimination capacity, EC (gDMS m^3/h) = $Q\ (C_{in} - C_{out}) / V$ *(2)*

Removal efficiency, RE (%) = $(C_{in} - C_{out})*100 / C_{in}$ (3)

where C_{in} and C_{out} are the inlet and outlet DMS concentration (gm^{-3}) obtained with the average of three measurements in each biofilter (n = 3) , V is the volume of packing material (m^3) and Q is the air flow (m^3h^{-1}).

Results and Discussion.

An overview of the LR, EBRT, EC, Cin, Cout and RE during the experiments is given in figures 2,3 and 4:

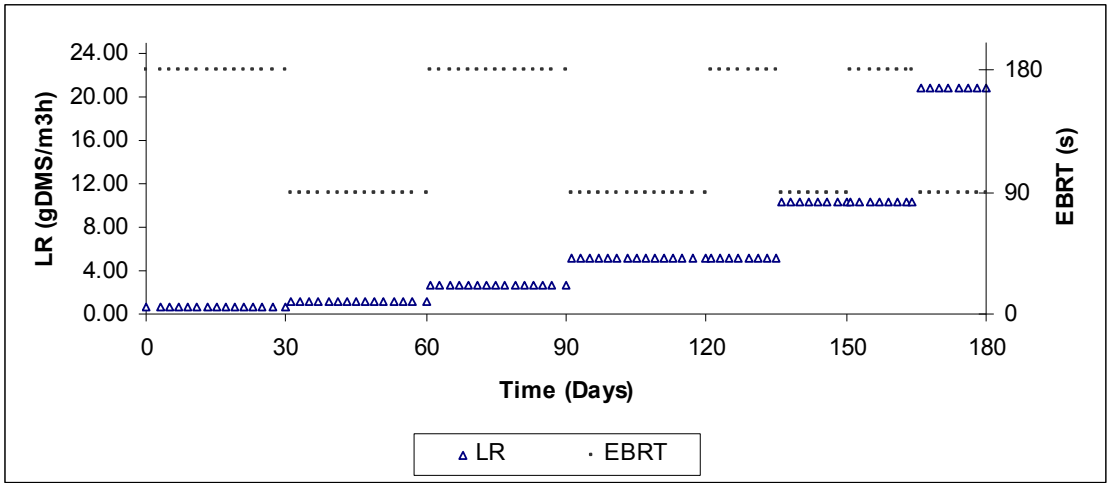

Figure 2.Operational conditions of the biofilter.

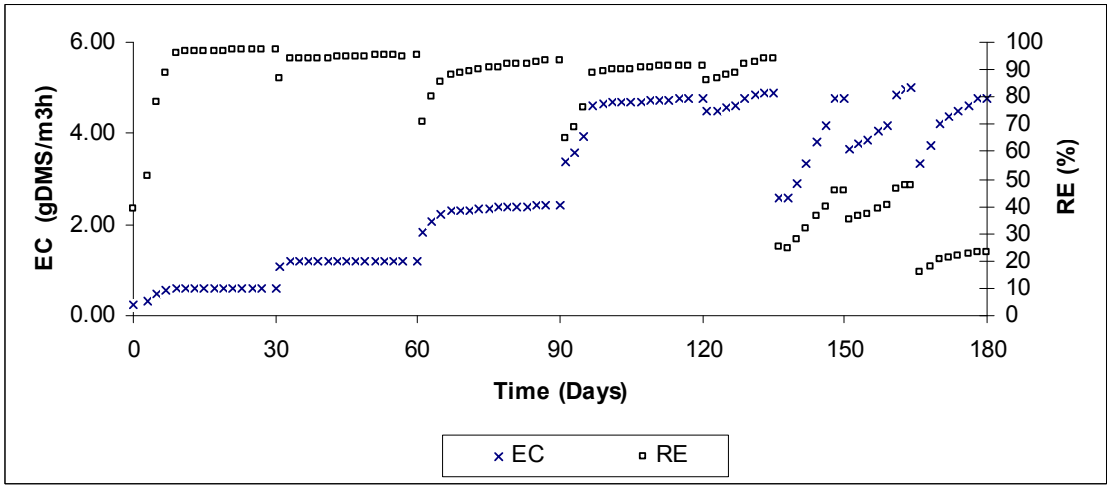

Figure 3.Performance of the DMS-degrading biofilter.

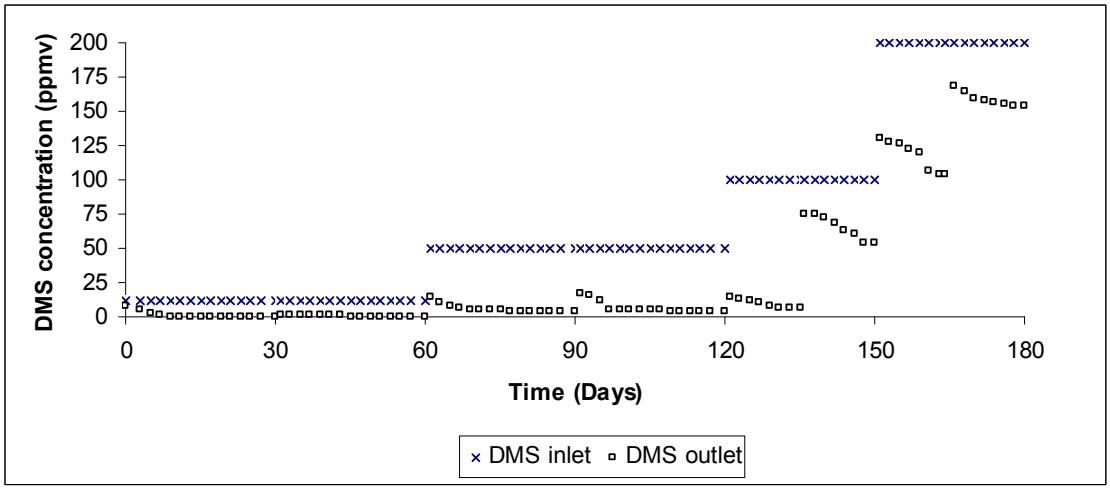

Figure 4 . DMS Inlet and outlet concentrations versus time.

The biofilter was operated for 180 days. The DMS mass loading rate was increased in six periods from 0.62 to 20.8 gDMS $m^{-3} h^{-1}$ (Figure 2). During each period the loading rate was kept constant. Three measurements of DMS inlet and outlet concentration were carried out every day.

The first day of operation, a removal efficiency of 39 ± 3.5 % was observed for the biofilter inoculated with *Hyphomicrobium VS.(Figure 3)*
The change in RE versus time shows that only 9 days are necessary as start up period for these microorganisms to adapt to DMS removal; a maximal removal efficiency for this biofilter of 97.6 ± 4.8 % is achieved in day 24. During a further increase of the DMS influent concentration, some temporary decreases of the removal efficiency were observed for the biofilter filled with sugarcane bagasse , a decrease in efficiency is linked to the increase in the loading rate of 1.25 gDMS $m^{-3} h^{-1}$ at 31 days of operation (Period 2). After 61 days of operation, LR was increased futher to 2.6 gDMS $m^{-3} h^{-1}$ by varying the inlet concentration. It is found that there is a noticeable decrease in the RE values for the biofilter (71 ± 4.6 %).

For higher LR (5.20 gDMS $m^{-3} h^{-1}$) after 91 days of operation of biofilter the removal efficiency decrease notably to 55 ± 4.6 % in the biofilter inoculated with *Hyphomicrobium VS*, this biofilter retains a removal efficiency of 95.2 ± 4.9 % in day 8 of the period 4 indicating a strong ability to remove DMS.

At day 136 the LR was increased to 10.40 gDMS $m^{-3} h^{-1}$ and the removal efficiency declined until 25 %, 14 days later the RE was established at 46 % with an EC of 4.78 gDMS $m^{-3} h^{-1}$. At the day 166 the Load was increase again to 20.80 gDMS $m^{-3} h^{-1}$ but the EC remained constant (4.78 gDMS $m^{-3} h^{-1}$) during the last period of experiments because the biofilter was working in the maximum elimination capacity of the sugarcane bagasse.

The removal efficiency drops dramatically to loading rates higher than 5.2 gDMS $m^{-3} h^{-1}$ .This happens after day 136 of operation of the biofilter showing that Sugarcane bagasse has reached its maximum elimination capacity. During this period the pH was adjusted to 7 but it possible to see that bacteria are inhibited and they are not able to degrade DMS loading rates exceeding 5.2 gDMS $m^{-3} h^{-1}$. Bacteria are completely saturated and can not remove any more DMS.Smet [7] have previously shown that DMS concentrations can exert a toxic effect on a *Hyphomicrobium VS* enrichment culture, especially at high concentrations. In the experiments when the DMS inlet concentration is equal or above to 100 ppmv, the DMS outlet concentration tends to approach the inlet concentration showing a significant decrease of the removal efficiency (Figure 4)

Figures 3 show that the highest elimination capacities with *Hyphomicrobium VS* was 5 $gm^{-3} h^{-1}$ with *RE* = 48± 3.2 % and load of 10.40 gDMS $m^{-3} h^{-1}$ on day 164 .

Table 1 . The elimination capacities obtained for DMS in lab-scale biofilters with diferents organic supports.

Filter Bed Material	Inoculation	T (^oC)	EBRT (s)	EC ($gm^{-3}h^{-1}$)	Reference
Peat	Night soil sludge	d.n.a.	120	3.2	[11]
Peat	Thiobacillus thiop. DW44	25 ±2	33	4.0	[10]
Peat	Hyphomicrobium I55	d.n.a.	120	4.75	[12]
Bark	None	Ambient	31	0.42	[3]
Compost	None	Ambient	31	<0.42	[3]
Bark	Hyphomicrobium MS3	Ambient	31	1.25	[3]
Sugarcane Bagasse	Hypomicrobium VS	Ambient	90	5	This paper

d.n.a. data not available.

Table 1 show the results achieved in the lab – scale biofiltration of DMS using different packing materials.
For the biofilters inoculated with Hyphomicrobium better EC are obtained using sugarcane bagasse compared with the results reported by [12] and [3]. In the bagasse biofilter *Hyphomicrobium* VS (EBRT 31 s) obtained an elimination capacity 3.9 times higher than the EC obtained by [3] using *Hyphomicrobium MS3* with wood bark as support (EBRT 90 s). Using peat [12] reaches an EC of 4.75 g DMS / m^3 h when Hyphomicrobium I55 was inoculated at an EBRT of 120 s.

The best elimination capacity achieved when organic supports are used as a media for microorganisms in a lab-scale biofilter for the removal of DMS was obtained with *Hyphomicrobium VS* (EC = 5 g DMS / m^3 h) (Table 1).

Biodegradation kinetics

From the well known Michaelis–Menten expression (see Equation 4) with Km, the half saturation parameter, and r*m*, the maximum volumetric elimination rate, as parameters, the Equation (5) can be derived [29], which was applied to determine the biodegradation kinetics, Km and r_m, using the acquired experimental data (Cin, Cout and EBRT):

$$r = r_m * \frac{C}{Km + C} \qquad (4)$$

$$C_{in} - C_{out} - Km * \ln\left(\frac{C_{out}}{C_{in}}\right) - r_m * \frac{V}{Q} = 0 \qquad (5)$$

This model is based on the Elimination capacity in function of the Inlet loading rate and it is used making the following assumptions :

- steady state conditions were reached for each applied inlet load
- the biomass activity was evenly distributed throughout the biofilter bed
- The DMS removal rate followed the Michaelis–Menten kinetics.

Plotting $\beta = \frac{C_{in} - C_{out}}{\ln\left(\frac{C_{out}}{C_{in}}\right)}$ versus $\alpha = \frac{EBRT}{\ln\left(\frac{C_{out}}{C_{in}}\right)}$,

resulted in a linear regression with r_m and Km the corresponding slope and intercept.

This regression resulted in Km= 0.057±0.11 g m^{-3} and r_m = 4.79 ± 0.72 g m^{-3} h^{-1} for an EBRT of 180 s. Analysis of the experimental data corresponding to an EBRT of 90 s gave Km = 0.027 ± 0.002 g m^{-3} and r_m = 4.26 ± 0.49 g m^{-3} h^{-1} . Indeed, an increase in EBRT, giving rise to a longer contact time between the contaminated air and the biofilm on the packing material, is known to give a higher value for r_m [30-33].

Equation (6) was obtained from Equation (5) after substitution of Equations (1) and (2).

$$IL = \frac{EC}{1 - \exp\left[\frac{(EC - r_m) * EBRT}{Km}\right]} \qquad (6)$$

By applying the obtained r_m and Km values in Equation (6) the data at an EBRT of 90 s and 180 s could be modeled sufficiently.

EC	

Using Equation (6) it could be calculated that at an EBRT of 180 s and IL =3.g m^{-3} h^{-1} RE of 85 % will be obtained (EC= 2.62 g m^{-3} h^{-1}) , at the same IL and EBRT= 90 s will be obtained and RE of 81 % (EC= 2.49 g m^{-3} h^{-1}) .

Table 2 . Elimination capacities real and estimated for differents Inlet loading rate at EBRT of 180 s

IL(g m^{-3} h^{-1})	EC_{Real} (g m^{-3} h^{-1})	$EC_{Estimated}$ (g m^{-3} h^{-1})
0.62	0.61	0.61
2.6	2.31	2.3
10.4	4.27	4.2

Table 2 shows the comparison of the Elimination capacity calculated using the experimental data acquired in the biofilter (EC_{Real}) and the Elimination capacity calculated with the data obtained by the model of the Equation 6 ($EC_{Estimated}$). These results show that the model adequately represents the behaviour of the biofilter to the operating conditions.

Conclusions

Sugarcane bagasse inoculated with Hyphomicrobium *VS* as packed material obtain good results in the biofiltration of air stream contaminated with DMS. The highest elimination capacities reached was 5 g DMS m^{-3} h^{-1} with *RE* = 48± 3.2 % and load of 10.40 g DMS m^{-3} h^{-1} and EBRT of 90 s.

A mathematical model based on the Michaelis–Menten theory was fitted to the experimental data in such a way that the half saturation parameter Km and the maximum volumetric elimination rate *rm* could be calculated. For an EBRT of 90 s, Km = 0.027 ± 0.002 g m^{-3} and *rm* = 4.26 ± 0.49 g m^{-3} h^{-1}, and for an EBRT of 180 s, Km= 0.057±0.11 g m^{-3} and *rm* = 4.79 ± 0.72 g m^{-3} h^{-1}.

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