



Short communication

## Application of biofiltration to the degradation of hydrogen sulfide in gas effluents

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### Abstract

A laboratory scale bioreactor has been designed and set up in order to degrade hydrogen sulfide from an air stream. The reactor is a vertical column of 7 litre capacity and 1 meter in height. It is divided into three modules and each module is filled with pellets of agricultural residues as packing bed material. The gas stream fed into the reactor through the upper inlet consists of a mixture of hydrogen sulfide and humidified air. The hydrogen sulfide content in the inlet gas stream was increased in stages until the degradation efficiency was below 90%. The parameters to be controlled in order to reach continuous and stable operation were temperature, moisture content and the percentage of the compound to be degraded at the inlet and outlet gas streams (removal or elimination efficiency). When the H<sub>2</sub>S mass loading rate was between 10 and 40 g m<sup>-3</sup>h<sup>-1</sup>, the removal efficiency was greater than 90%. The support material had a good physical performance throughout operation time, which is evidence that this material is suitable for biofiltration purposes.

### Introduction

Biofiltration is one of the most promising clean technologies for reducing emissions of malodorous pollutants into the atmosphere. This technology, based on microbiological degradation of compounds from a gas stream, is considered to be an attractive alternative when compared to chemical and physical treatments, as it has advantages from the economic and environmental point of view. About biofilter economics, Williams (1995) discussed the technical merits and economics of using biofiltration for hydrogen sulfide control and found that total annual costs for the biofilter system were 46% lower than an activated carbon system and 40% less than a chemical scrubber. Biofiltration generates less residue than other technologies since the pollutants are decomposed. Other ad-

vantages include its simple operation and effectiveness for a wide range of compounds.

The design and operational parameters as well as microbial processes involved in biofiltration are not fully understood. Nevertheless, this technology proved to control effectively odours in dilute waste gas streams contaminated with sulfur compounds (Yang & Allen 1994; Smet et al. 1998; Ergas et al. 1995; Morton & Caballero 1998). One of these compounds is hydrogen sulfide (H<sub>2</sub>S) which is a toxic pollutant produced in association with industrial processes such as petroleum refining, waste water treatment, paper and pulp manufacturing, food processing and so on.

Most biofiltration applications rely on the microorganisms present in the packing material to degrade the contaminants of concern. The microorganisms capable of degrading H<sub>2</sub>S are ubiquitous and, thus, the nature

of the packing material is very variable. Hence, materials like vegetable compost (Yang & Allen 1994), sewage sludge (Degorce-Dumas et al. 1997; León et al. 1999), activated carbon (Webster et al. 1996) and peat (Hirai et al. 1990) have been described in literature for biofiltration purposes.

The parameters that must be carefully taken into account when selecting a suitable support or packing material are pH value, moisture content and the amount and quality of the biomass or microorganisms.

The pH value must always be higher than 3.2, as operating with lower pH values could lead to the destruction of the active biological population and, consequently, the removal efficiency (defined as the percentage of the contaminant compound that is degraded) will decrease progressively, especially when an organic support material is used (Yang & Allen 1994). High variation of the pH value can also result in a loss of diversity in the microbial population and even destroy the resident population (Lau et al. 1996).

The moisture content of the support material is very important as it is directly related to microbiological activity (Van Lith et al. 1997), although the operating range may be very wide. For instance, Yang & Allen (1994) used a compost as support material and they reported H<sub>2</sub>S degradation efficiencies of around 99.9% when the moisture content ranged between 30 and 62%. As far as biomass is concerned, certain studies have been carried out using cultures of pure microorganisms (Chung et al. 1996) but, as H<sub>2</sub>S is an easily biodegradable compound, biomass is usually supplied by such support materials as waste water sludge (Wani et al. 1999) or composts (Yang & Allen 1994) because both materials have suitable amounts of sulfoxidant bacteria populations.

In this work, a laboratory scale bioreactor has been designed and set up in order to study the removal efficiency of hydrogen sulfide when a new support material is used. This material consisted of a pelletized (with a cylindrical shape) mixture of pig manure and sawdust and was selected because it was able to supply the biomass and the nutrients required for microorganisms to grow properly and efficiently degrade the pollutant gas.

## Material and methods

The experimental equipment used in this work is shown in Figure 1. The bioreactor or biofilter itself consists of three interchangeable modules (Bed 1, 2

Table 1. Summary of initial packing material properties

Property	
Total organic matter (%)	72
Oxidisable organic matter (%)	40
Conductivity at 25 °C (mS cm <sup>-1</sup> )	1.3
Particle density (g cm <sup>-3</sup> )	1.24
Diameter range of the pellets (mm)	6.3–8
Moisture content (%)	42
pH	8.5
Total-N (%)	2.15
Total-C (%)	31.78
Total-H (%)	3.53
P (%P <sub>2</sub> O <sub>5</sub> )	1.65
K (%K <sub>2</sub> O)	2.60
Mg (%MgO)	1.30
Ca (%CaO)	0.73
Na (%Na <sub>2</sub> O)	0.60
Zn (µg g <sup>-1</sup> DW)	506
Fe (µg g <sup>-1</sup> DW)	6300

and 3 in Figure 1) made of PVC. The volume of the bioreactor itself is 7 litres and the dimensions are 1 m in length and 10 cm cylinder internal diameter. The empty bed residence time is 27 s. The volume of the support material is 5.9 litres. Sampling and measurement ports were located along the three modules.

The support material was supplied by the Spanish company SLIR (Specialized Engineering in Recycling Agricultural Residues) and its commercial name is ABONLIR. This material is a compost obtained by mixing pig manure and sawdust and the pellets are manufactured by mechanical compression without the addition of any chemicals. In order to ensure a minimum size of these cylindrical pellets, the compost was sieved so that only pellets whose diameter was between 6.3 and 8 mm were used as packing material. The overall length of the pellets was between 20 and 80 mm. The compost was stored in sealed plastic bags at room temperature to keep the material in its original moist condition. Table 1 shows certain chemical and physical properties of the pellets used as packing material.

The gas flow to be treated was obtained in the laboratory by mixing H<sub>2</sub>S (99.7% purity) and 80% water saturated air. The superficial loading in the biofilter was 100 m<sup>3</sup> m<sup>-2</sup> h<sup>-1</sup>. The H<sub>2</sub>S flow from the gas cylinder was controlled by a 5850S Brooks Mass Flow

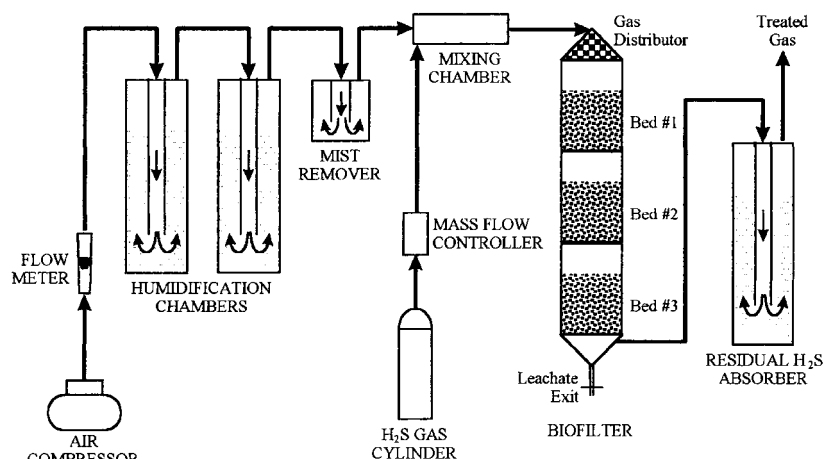


Figure 1. Outline of the experimental equipment.

Controller. The air stream from the compressor was regulated by a Platon Rotameter and, subsequently, this stream was bubbled into two PVC columns filled with deionized water in order to saturate the air stream until 80% water content. No nutritive solution for feeding microorganisms on the biofilter was used.

The  $\text{H}_2\text{S}$  mass loading rate is defined as the amount of  $\text{H}_2\text{S}$  that is introduced to the system per unit time/per unit volume of the packing material ( $\text{g H}_2\text{S m}^{-3} \text{ h}^{-1}$ ). In this work, variable  $\text{H}_2\text{S}$  loading rates (from 10 to  $50 \text{ g H}_2\text{S m}^{-3} \text{ h}^{-1}$ ) were fed into the reactor for 140 hours in order to ensure a proper microbiological acclimation of the contaminant. After this acclimation period, operation time started (time 0). In order to ascertain the removal efficiency of the  $\text{H}_2\text{S}$ , runs were carried out beginning with an initial  $\text{H}_2\text{S}$  loading rate of  $10 \text{ g m}^{-3} \text{ h}^{-1}$  fed into the bioreactor (after the acclimation period) and increasing this concentration by  $10 \text{ g m}^{-3} \text{ h}^{-1}$  increments until degradation efficiency was below 90%. Consequently, we considered the failure mode or the end of operation when the degradation or removal efficiency was below 90%.

Three parameters were measured throughout operation time. The first parameter was temperature, which was continuously measured by using three thermocouples (controlled by a Hanna HI 92804 C meter) at each reactor module. Room temperature was within the  $20\text{--}25^\circ\text{C}$  range. The second parameter was moisture content and it was measured at the gas flow inlet and in the three modules by using a Testo 0636.9715C probe. This 12 mm diameter probe was connected to a Testo 400 register used in aeration and air conditioning equipment. The moisture content in the support

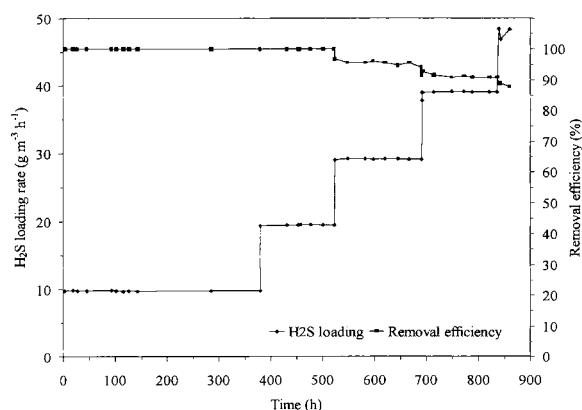


Figure 2. Removal efficiency and  $\text{H}_2\text{S}$  loading rate during operation time.

material was ascertained by thermogravimetry in a LECO TGA 500 device. The third parameter to be measured was  $\text{H}_2\text{S}$  content at the gas flow inlet and outlet by means of a Dräger Pac III electrochemical sensor. Pressure drop was determined with a Bioblock MP 310 differential pressure meter.

No source of carbon was provided to the microorganisms during the experimental phase and the only source of oxygen was provided by the air mixed with the  $\text{H}_2\text{S}$ .

## Results and discussion

The temperature measured in the three modules of the reactor varied slightly between  $20$  and  $22^\circ\text{C}$  during operation time. The temperature along the three modules only differed by  $1.5^\circ\text{C}$  and the standard deviation

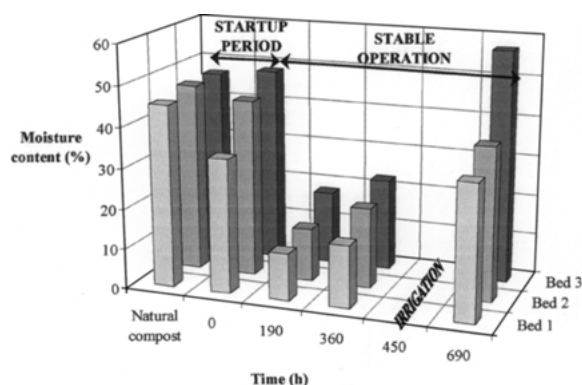


Figure 3. Evolution of moisture content.

for each module was  $0.32^{\circ}\text{C}$  during operation time. The first module or bed (at the upper part of the reactor) reached slightly higher temperatures than the other ones, as a consequence of higher mass content of  $\text{H}_2\text{S}$ , resulting in greater microbiological activity.

Yang & Allen (1994) found that pressure drop values of the support material should be lower than 25 cm water column for stable operation. We measured pressure drop values as low as 3 cm water column, which was favoured by two facts. Firstly, the reactor was specially designed and it was divided into three modules (or beds) which prevented the support material from being degraded and compacted at the bottom of each module. The pressure drop increases linearly with packing height but its dependence on water content of the packing material is not very consistent (Yang & Allen 1994). On the other hand, the way the filter is packed should also be taken into account. Secondly, the microbial growth did not clog the system, which also favoured the low pressure drop.

The evolution of removal efficiency with different quantities of  $\text{H}_2\text{S}$  added by regular  $10\text{ g m}^{-3}\text{ h}^{-1}$  increments is shown in Figure 2. After the acclimation period, an initial  $\text{H}_2\text{S}$  loading rate of  $10\text{ g m}^{-3}\text{ h}^{-1}$  was fed into the reactor. The removal efficiency remained almost constant and greater than 90% until  $40\text{ g H}_2\text{S m}^{-3}\text{ h}^{-1}$  were added to the system. The moisture content and pH evolution were measured throughout operation time until removal efficiency was lower than 90% (Figures 3 and 4).

The moisture content decreased during the first 190 hours of operation, which is evidence of microbial degradation activity. After 190 hours, moisture content in bed 1 remained constant at a value of 12%, which is rather low, although no changes in removal efficiency were observed. During this operation period, the air

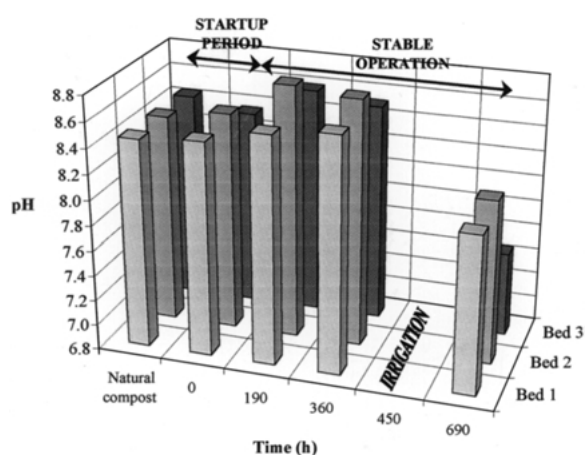


Figure 4. Evolution of pH.

supplied was 80% water saturated. In order to obtain a higher level of moisture content in the biofilter, deionized water was added to the humidification chambers subsequent to 250 hours of operation so that the air stream at the inlet was 100% water saturated. After this addition, the moisture content level increased to 17% in bed 1 and to 25% in bed 3 (data shown in Figure 3 after 360 hours of operation).

As the  $\text{H}_2\text{S}$  loading at the inlet was increased along time, and in order to ensure stable operation, irrigation of the system was carried out from the inlet of the first module (upper module). As a consequence of this irrigation, 35, 39 and 57% moisture content levels were obtained for modules 1, 2 and 3 respectively.

The pH evolution throughout operation time is shown in Figure 4. The oxidation of sulfur compounds can lead to the formation of acid intermediates which lower bed pH, with a subsequent reduction in removal efficiency. In our study (over 360 hours of operation), the pH value did not vary significantly (between 8 and 8.8), which indicated a good natural buffer capacity of the whole system. Consequently, no chemicals were necessary to buffer a possible acid production. Nevertheless, after the irrigation period, the pH value decreased in the three modules and the lowest value (7.5) was achieved in the third module, which was related to the dragging of acid compounds from the upper modules. This pH reduction, a consequence of the irrigation together with the fact that  $\text{H}_2\text{S}$  loading rate in the feeding flow was increased to  $40\text{ g H}_2\text{S m}^{-3}\text{ h}^{-1}$ , resulted in a decrease of the removal efficiency (lower than 90%), as shown in Figure 2. However, the limitation in the removal efficiency at the end of the experimentation period could also be related

to sulphate accumulation, which is known to inhibit hydrogen sulfide elimination (Yang & Allen 1994).

The packing material used in this preliminary study resulted in high removal efficiency of H<sub>2</sub>S under the experimental lab-scale conditions and showed a good performance; however, further research about this material is necessary, as for instance, physical, microbiological and chemical changes in the pellets throughout operation and long-term stability testing of the biofilter.

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