Performance evaluation of biofiltration in the removal of hydrogen sulfide from gas flue

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ABSTRACT. Hydrogen sulfide, the most common odorous reduced sulfur compound, has an odour threshold of 1-2 ppb and is emitted from variety of industrial air emissions. Many traditional technologies for H_2S treatment have already been developed. However, these technologies are expensive for treating dilute streams and consume more energy and generate secondary pollution. Biofiltration is a low-cost and highly effective air pollution control technology and an environmental amicable method. Performance of biofiltration for removal of hydrogen sulfide from waste gas stream was studied in a bench scale biofilter constructed from galvanized iron with inner diameter of 8 cm and height of 1.20 m. Bed material consisted mixture of municipal solid waste compost and PVC bits (1.0×1.5 cm) as a bulking agent in 1:1 volumetric ratio. Parameters that were evaluated included; elimination capacity, removal efficiency, effects of sulfate accumulation, gas retention time, pressure drop, bed water content and pH. Average removal efficiency of 98% obtained with retention time of 1.0 min. Accumulation of sulfate in the bed material showed reverse effect on biofilter efficiency and resulted increasing of pressure drop along biofilter column to the maximum level of 18 mm H_2O. Maximum elimination capacity of the biofilter was obtained about 22 g-s/m^3.hr during the experiment.

1 INTRODUCTION

Hydrogen sulfide (H_2S) is a toxic, colourless, flammable gas and is the most common odorous reduced sulfur compound, has an odour threshold of 1-2 ppb (ACGIH, 1991) and its emission is found in many industrial activities such as petroleum refining, pulp and paper manufacturing and food processing. Also its odour is major problem in the operation wastewater treatment plants and corresponding pump stations. Many physical and chemical technologies for H_2S treatment, including scrubbing, thermal oxidation, and adsorption, have already been developed (Leson and Winer, 1991). However, these technologies are expensive for treating dilute streams; they also consume more energy and have a greater tendency to generate secondary pollution than biological processes.

Biofiltration is a process that takes advantage of microorganism growing or immobilized on an organic porous support, the organic media act as a physical support

for active biomass and in some cases it provides nutrients for growth. The contaminated gaseous stream passes through the filter bed, the bed material absorbs biodegradable volatile compounds and the microorganisms degrade it into less harmful compounds (van Groenestijn and Hesselink, 1993). Biofiltration is attractive for H_2S removal, particularly in high volume, low concentration air emissions because it is potentially an efficient and inexpensive method for significantly reducing the mass loading of sulfur compounds to the environment. Because H_2S can cause odour problems at concentrations as low as 1 ppb, a number of researchers have investigated the effectiveness of biofiltration for streams containing very low concentrations of H_2S (Chung *et al*. 1997; Cho *et al*., 1992). Thus, it appears that biofiltration can effectively treat hydrogen sulfide emissions below 10 ppmv; however, the reported empty bed retention times required to achieve high removal rates are on the order of 40 - 80 s. However the most common range of inlet hydrogen sulfide concentrations addressed in the literature is 10-450 ppmv. At these concentrations, the effect of sulfate accumulation and pH decline over the reactor operation period is much more significant than for the low concentration studies. The lowest hydrogen sulfide removal capacity (14.9 g H_2S/m^3 bed.h) was observed by Zhang *et al*. (1991), while Cho *et al*. (1992) observed the highest removal capacities (108–315 g H_2S/m^3 bed.h). Chung *et al*. (1996) studied the biofiltration of hydrogen sulfide using a number of different bacterial strains, and they found that the maximum removal rates for their biofilters were 23–30 g H_2S/m^3bed.h. Very few studies have addressed the biofiltration of streams containing more than 450 ppmv of hydrogen sulfide (Yang and Allen, 1994; Cho *et al*., 2000). This is partly due to the existence of physical and chemical methods of treatment which become more economical, efficient, and reliable at higher hydrogen sulfide concentrations. Yang and Allen (1994) and Cho *et al*. (2000) also found the 60% of the H_2S was oxidized to sulfur, while only 40% was completely degraded to sulfate. Furthermore, both indicate that the proportion of sulfide degraded to elemental sulfur and sulfate may be an important indicator of biofilter performance capacities, particularly for long term operation.

Porosity and bulk density of the bed media are important primarily for the effect they have on the gas phase pressure drop across the bed. On the other hand long term stability of H_2S biofiltration is a major concern due to the accumulation of hydrogen sulfide degradation by-products such as sulfate. The objective of this research was to evaluate the applicability of biofilter using municipal solid waste management compost based media as a microbial population provider and nutrient resource supplier mixed with shredded PVC as a bulking agent and for better air distribution to overcoming to the bed pressure drop due to sulfate accumulation.

2 MATERIALS AND METHODS

A three-stage bench-scale biofilter constructed from galvanized iron with an inner diameter of 8 cm was used in this study (Figure 1). The effective height of bed media was 120 cm. Perforated steel plates with pore diameter of 2 mm placed between sections acted as a support for the packing material as well as for gas flow redistribution. A 7-cm space in between the sections allowed for representative gas sampling. Provision of sampling ports at top, midpoint and at the end of each section allowed bed media access. The air stream was prepared by sending compressed air through a granular activated carbon canister to adsorb residual oil and remove particles and then sprayed through a 15-liter water container equipped with heated element for adjusting gas stream temperature and humidification. H_2S was prepared by drop-wise

introducing of hydrochloric acid feed from a burette into a plastic container containing sodium sulfide ($Na_2S + HCl \rightarrow H_2S + NaCl$). After humidification, the main air stream was mixed with the stream containing H_2S to generate feed air with the needed concentration. Changing water temperature in the humidifier controlled variation of humidity in the influent gas stream and biofilter material. Temperature control of the bed material was achieved by using a heated tape wrapped around the exterior of reactor wall that has been covered with glass wool. Temperature and bed water content of the biofilter were maintained at 30±1°C and 60-65%, respectively during all operation period

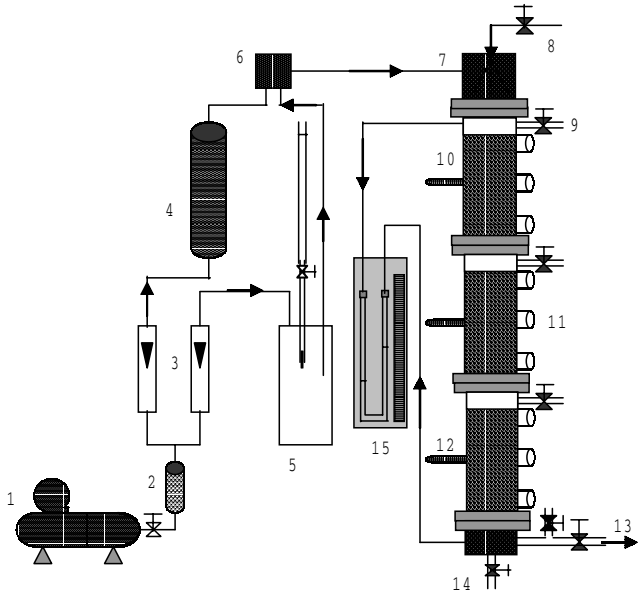

Figure 1. Schematics of biofilter system (1-compressor, 2-carbon filter, 3- rotameter, 4-humidifier, 5-H_2S vessel with HCL injector, 6-mixing chamber, 7-inlet, 8-nutrient, 9-gas sampling port, 10- biofilter bed, 11-bed sampling port, 12- thermometer, 13-outlet, 14-lechate, 15-manometer).

Bed Media was prepared by mixing yard waste compost and shredded high-density plastics (1.5 ×1.0 cm) as bulking agent to produce a 50:50 volumetric ratio of compost-bulk agent. In preparing the packing medium, thickened activated sludge obtained from municipal wastewater treatment plant (Tehran Water & Wastewater Co.) was added to this mixture to increase microbial density and developing compost particles attach on bulking agents. Mineral medium containing (g/L), KH_2PO_4, 0.54; K_2HPO_4, 0.05; NH_4NO_3, 0.5; NaCl, 0.26; $CaCl_2$. $2H_2O$, 0.025 and 1 ml trace element solution was added to the bed providing a C/N ratio of 30 for 15 days of biofilter operation (Ergas et al., 1994).

H_2S concentration in the gas phase was measured using samples collected with the flow rate of 0.5-2 liter/min by a rotameter from inlet and outlet of biofilter column in a series of midget impengers and absorbed in pH 3.0 cadmium sulfate ($CdSO_4$) solution to form cadmium sulfide (CdS). The latter compound was then measured iodometrically. An impinger containing hydrogen peroxide (H_2O_2) is included to remove SO_2 as an

interfering species (EPA, 1999). The lower limit of detection was approximately 8 mg/m^3 (6 ppm). The maximum of the measuring range was 740 mg/m^3 (520 ppm). Collaborative testing has shown the within-laboratory coefficient of variation to be 2.2 percent and the overall coefficient of variation to be 5 percent. Bed water content, pH and sulfate concentration in the material was analyzed using procedures published in standard methods for examination of water and wastewater (WEF *et al*., 1992). A U-tube manometer, 0 to 100 cm water column, was used for gas pressure drop monitoring along the biofilter column.

3 RESULTS AND DISCUSSION

Performance of the biofilter was evaluated during the start up period with the empty bed retention time (EBRT) of 1 min and inlet concentration of about 5-265 ppm (Figure 2). At first system was operated with inlet concentration of about 12 ± 5 ppm, then at lower than 10 days removal efficiency of biofilter reached to the maximum value of 97%. At the next days removal efficiency of the biofilter was decreased sharply. This may be due the fact that new biofilter medium acts as a very good adsorber at the start of biofiltration till the medium adsorptive capacity is occupied. This may be helpful in that it reduces the quantity of contaminant which escapes during microbiological acclimation period (Bishop and Govind, 1995). However the effects of adsorption on biodegradation rates depend on the medium, contaminant, and microorganisms (Alexander, 1994). After the acclimation period, gradually increasing of the H_2S inlet concentration to 265 ppm did not impact on the performance of biofilter that verifying the function of microorganisms in the degradation of H_2S as superior mechanism in the removal of this substrate

Maximum elimination capacity of the biofilter in the removal of H_2S was obtained about 22 $g\text{-}s/m^3$ bed.h. Figure 3 shows the accomplishment of biofilter at different H_2S loading rate with constant EBRT of 1 min. At H_2S loading rate of lower than 15 $g\text{-}s/m^3$ bed.h the removal efficiency of biofilter was nearby 100%, but increasing the loading rate was showed adverse effect on the biofilter performance. This capacity of the biofilter about two magnitudes is low in comparison with other studies on biofiltration of H_2S with pure cultures (Oyarzun *et al*., 2003). However our results identical with the other results reported in the literatures (Chung *et al*., 1996; Cho *et al*., 1992). The system was not more evaluated in the upper loading rates, but as the loading of contaminant increases, the removal rate will decrease from a maximum as biological growth is inhibited (Mcnevin and Barford, 2000).

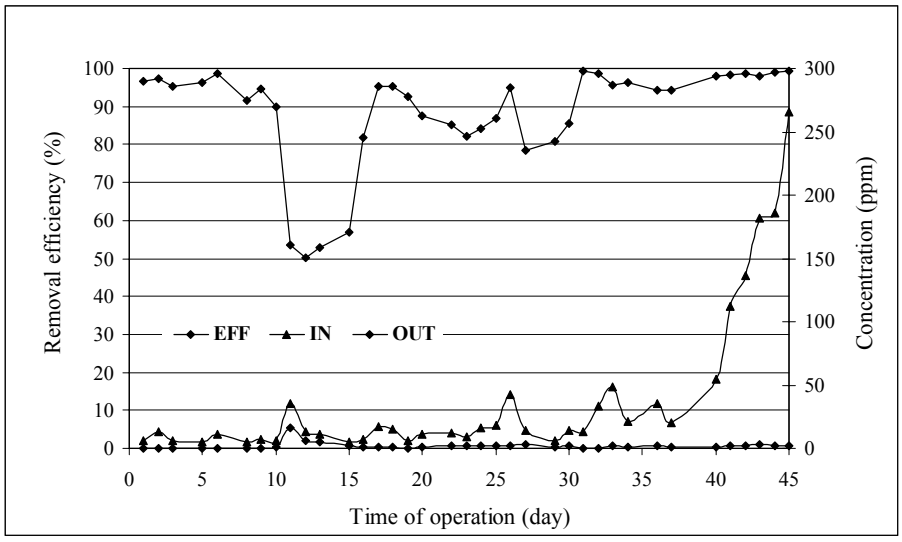

Figure 2. Performance of the biofilter in the start-up period.

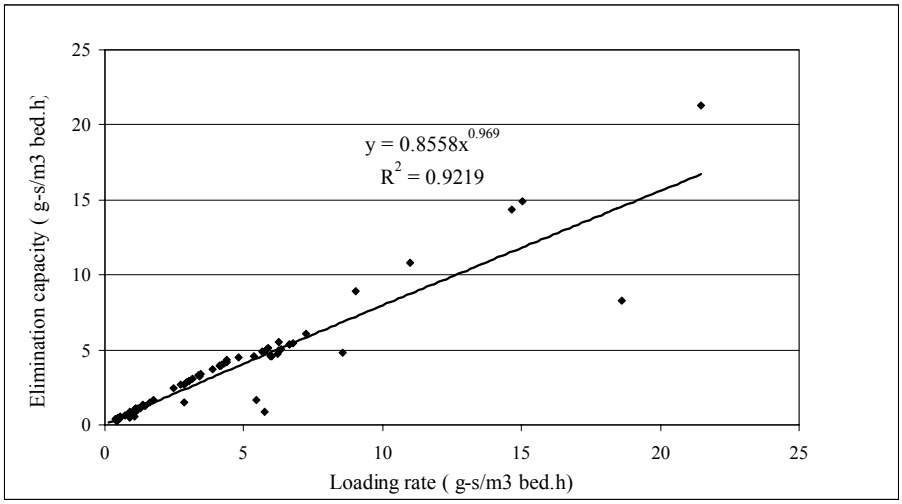

Figure 3. Elimination capacity of the biofilter as a function of inlet loading rate of H_2S.

The relation between H_2S loading rate on pressure drop is shown in Figure 4. The pressure drop ranged from non detectable amounts to 18 mm H_2O. The pressure drop across the biofilter is not increased gradually, but abundant of biomass or accumulation of sulfate resulted higher pressure drop along the biofilter column. However, increasing the loading rate on days 45 and 50 did not show direct relation between loading rate and pressure drop because of better performance of the used packing material. The biofilter was washed with water during the experiments and pressure drop was maintained in the lower quantities. Gas pressure drop through a biofilter bed increases with gas flowrate and diminishing particle size. Compaction of the filter bed over extended periods of usage and due to over watering, will also give rise to prohibitive pressure drops (Mcnevin and Barford, 2000). Any increase in pressure drop adds to the operating cost

of the biofilter as odorous air must be supplied at a greater pressure to achieve the same flowrate. Our results indicate that mixing the synthetic hard and porous material into the organic packing material reduces pressure drop across the biofilter.

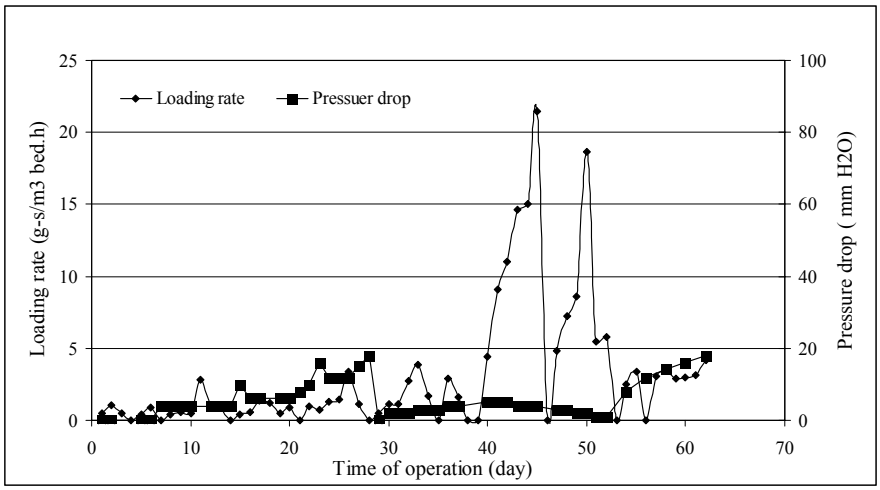

Figure 4. Effect of loading rate on the gas pressure drop across the biofilter.

Figure 5 shows the effect of sulfate accumulation on the efficiency of the biofilter. The removal efficiency of biofilter dropped from 100% to lower than 40% by increasing sulfate concentration in the bed material up to the 30 mg/g bed. Yang and Allen (1994) found that sulfate concentrations of more than 25 mg-s/g dry bed material inhibited the performance of their hydrogen sulfide removing biofilter.

Performance of the biofilter was evaluated at different gas residence time (Figure 6). At empty bed retention time (EBRT) of 1.0 min the removal efficiency of biofilter was about 100%. Removal efficiency dropped from a maximum when retention time drops bellow about 0.5 min. Lower EBRT makes application of biofiltration economic. However a minimum residence time for mass transfer is required when designing gas flowrates for biofilters. When short residence time will be employed this will be insufficient for mass transfer to occur (Yang and Allen, 1994).

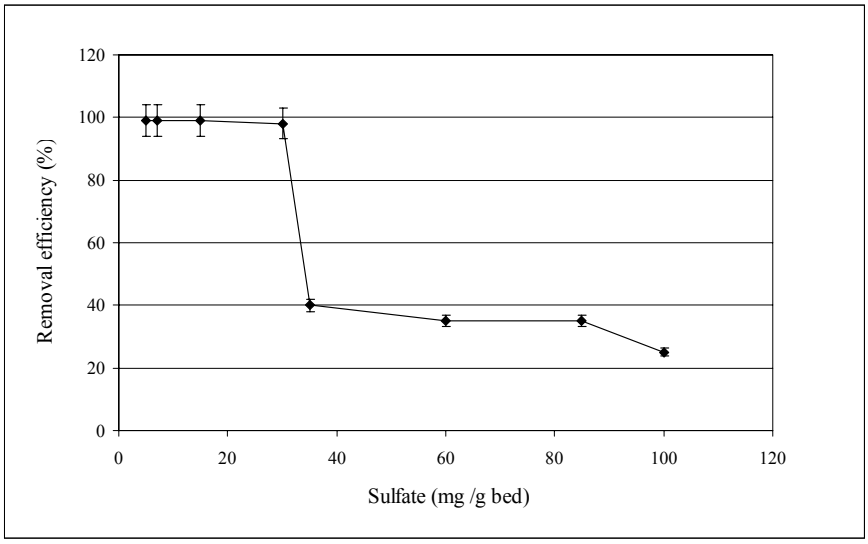

Figure 5. Effect of sulfate accumulation rate in the bed on the removal efficiency of H_2S biofiltration.

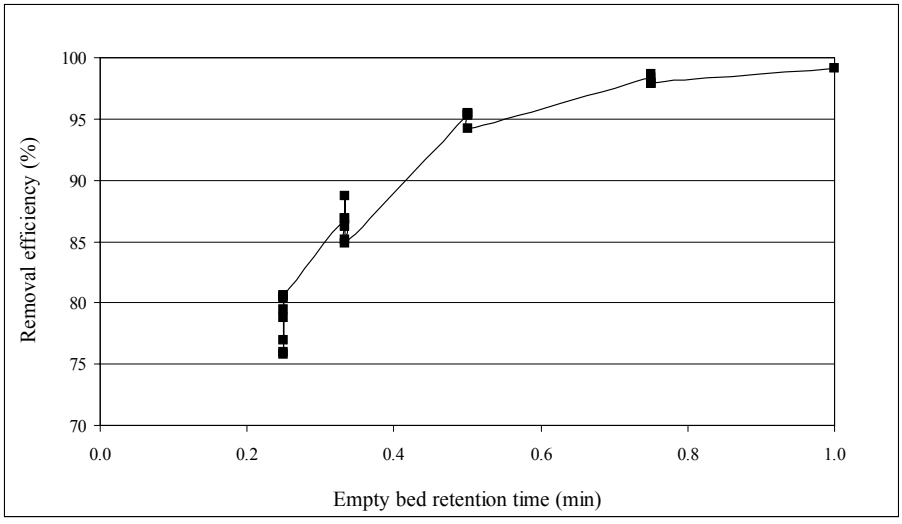

Figure 6. Performance of the biofilter at the different empty bed retention time (EBRT).

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