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Control of Odor Emissions Using Biofiltration: A Case Study of Dimethyl Disulfide

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Abstract

A laboratory-scale dual-column biofilter system was used to study the biofiltration of dimethyl disulfide (DMDS). The gas flow rate and DMDS concentration to the biofilter were varied to study their effect on the removal of dimethyl disulfide. Operating parameters such as pH, temperature, and water content were monitored during the biofilter operation and necessary precautions were taken to keep these parameters within the acceptable limits. It was observed that the removal efficiency of DMDS was optimal at neutral pH values. After five month operation, the neutralization of the filter beds with sodium carbonate became necessary for the optimum operation of the biofilters. The microbial population already present in the compost mixtures was found to be adequate in treating DMDS. The compost mixtures were found to be similar in terms of biofiltration efficiency of DMDS. However, pressure drops observed in the first column compost mixture (compost/ peat mulch) was extremely high, making this compost economically not feasible. The second mixture (compost/bark) provided pressure drops within acceptable limits. A minimum residence time of 30 seconds at the optimal operating conditions appeared to be adequate for achieving high removal efficiencies (>90%).

Key words : biofilter, dulfur vompound, PH, pressure frop, eater content

1. INTRODUCTION

Public facilities such as wastewater treatment plants, solid waste composting plants and landfill sites are located either on the border of or within residential areas, due to the economics involved in transporting domestic wastes from the communities they serve. Consequently, these facilities are the source of increasing public complaints. Odor is one of the most noticeable forms of pollution, and is the main public perception problem that industrial and commercial facilities are required to face.

Due to the complex nature of odor, regulating odorous emissions has been a difficult issue for many environmental agencies. Concentrations even in the parts per trillion by volume (pptv) range for some chemicals can be detected by the human nose (Reed, 1990), whereas some of the most advanced analytical instruments are incapable of detecting these pollutants at low levels. The response of individuals to odors can also vary largely. Hyperosmic (high sensitivity), anosmic (unable to smell) and hyposmic (partial loss of smell) individ-

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uals are predicted to constitute 4% of the population (USEPA, 1992).

Dimethyl disulfide (DMDS), a highly odorous chemical substance, is generally found in emissions from wastewater treatment plants and solid waste composting plants. At these facilities, DMDS is released into the atmosphere as a by-product of anaerobic degradation. DMDS is also emitted from pulp and paper manufacturing industries utilizing the Kraft process. In the pulp and paper industries, DMDS is mostly present in the work atmosphere and is a source of interest for industrial hygienists. Although DMDS is considered to be highly toxic, its toxicology in the human body is not very well known. Odor threshold concentrations reported for DMDS vary in the range from 0.3 to 7.5 ppbv due to different odor measurement techniques and definitions of odor threshold values described in the literature.

Usually in wastewater treatment plants, DMDS is found together with other odorous sulfurous compounds, such as methyl mercaptan (MM), dimethyl sulfide (DMS), hydrogen sulfide (H_2S) and carbon disulfide (Ruokojarvi *et al.*, 2001; Hwang *et al.*, 1995; Allen *et al.*, 1987). In a wastewater treatment plants with 35 million gallons per day (gpd) capacity, DMDS concentrations averaging nearly 12 ppmv were measured at the plant headworks (Allen *et al.*, 1987). As a biosolid composting facility biofilter inlet (24,000 standard ft^3/min capacity), somewhat lower concentrations (270 ~490 ppbv DMDS) were observed (Amirhor *et al.*, 1994). Typical DMDS concentrations measured near Kraft paper mills can approach maximum values in the range from 0.85 ppmv to 3 ppmv (Goyer, 1990; Leach and Chung, 1982).

As with every new technique (Smet *et al.*, 1998), biofiltration, when used as an air pollution control device has many unknowns and requires extensive further study. The objectives of this study are to determine the pressure drop characteristics of the compost filter bed, and to assess the influence of biofilter operating conditions, such as compost water content, temperature and pH, on DMDS removal.

2. EXPERIMENATAL METHODS

2. 1 Biofilter system

In this study, a laboratory-scale dual-column compost biofilter system was used to study biodegradation of dimethyl disulfide (DMDS) in an air stream.

The modified biofilter system, illustrated in Figure 1, consists of an air blower, a humidification chamber, a syringe pump, and two identical biofilter columns that are operated in parallel. A half HP air blower (Model R3105-1, Gast Regenair) with a maximum air flow capacity of 1.5 m^3/min was used to move the room air containing pollutant vapor through the biofilter system. Before being introduced into the biofilter columns, the room air is first treated in the humidification chamber. The acrylic humidification chamber has an inner diameter of 0.15 m and a length of 0.86 m. The chamber houses a spray nozzle (Spraying Systems Co., Model 1/4J+SU1A) which atomizes tap water (using room air obtained from the blower). In order to increase the amount of contact between the counter-current air flow and the water streams, the humidification chamber is filled with 5 cm diameter Pall rings almost up to the nozzle level. The room air leaves the humidification chamber with greater than 95% relative humidity. Humidified air is transported to the biofilter columns via PVC pipes. Before entering the biofilter columns, liquid DMDS (CAS# 624-92-0, 99% purity, ACROS Chemicals, Pittsburgh, PA) is injected into the humid air flow using a syringe pump (Model 355, Sage Instrument, White Plains, NY).

The biofilter columns are made of transparent acrylic tubes of 0.15 m inner diameter and of 1.34 m length. The columns are connected to PVC pipes at the top and the bottom by acrylic flanges which are secured to the columns by bolts for easy access and dismantling. Contaminated air is fed into the biofilter columns from the bottom.

Several ports are located along the vertical columns to allow for measurement of gas and compost properties. The columns are packed with compost up to a hei-

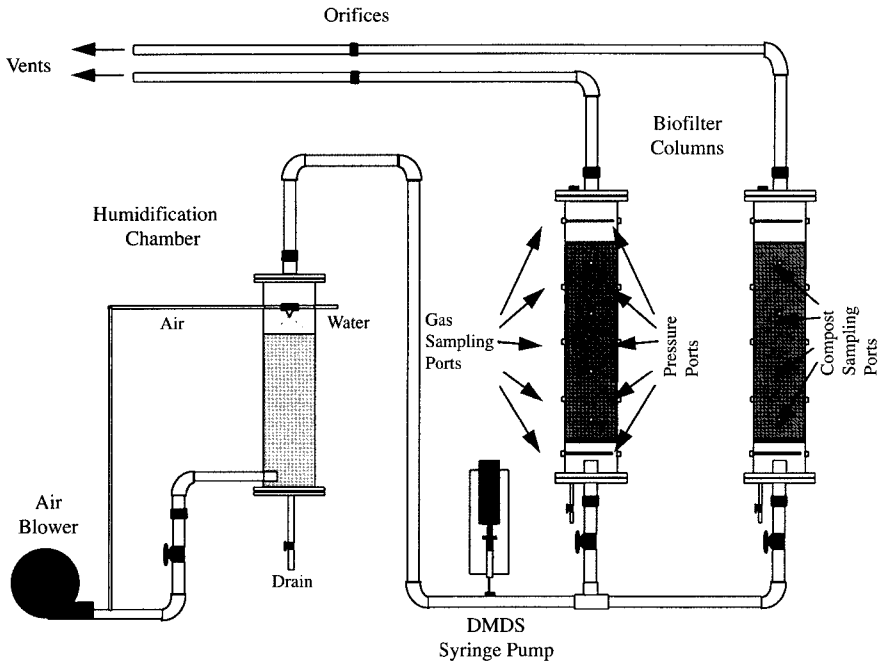

Fig. 1. Schematic diagram of the biofilter system.

ght of one meter. An acrylic perforated plate located above the column inlet is used to support the compost and uniformly distribute the incoming polluted air. A plastic screen placed between the compost and the acrylic plate, retains small compost particles in the compost bed. The cleaned effluent gas is discharged from the top of the biofilter columns via PVC pipes.

2. 2 Gas sampling and analysis

2. 2. 1 Gas sampling

Gas samples were extracted from the columns through five gas sampling ports. Each port consisted of a gas collecting assembly made from 6.35 mm diameter Teflon tube. The Teflon tubes were modified with fourteen equally spaced (1 mm diameter) holes and were packed with glass wool to prevent particles from contaminating the collected gas samples. The Teflon tubes extended diametrically across the biofilter column in order to collect a representative gas sample for that specific biofilter bed level.

Gas samples from the ports were slowly expanded into Tedlar bags using the slightly positive pressure of the carrier gas in the columns. A staggered sampling method, which incorporates a time delay was used for filling the Tedlar bags at different bed levels. This method allows for sampling of the same stream of pollutant gas at each port by taking into consideration the time for the gas to reach that port.

Samples in the Tedlar bags were analyzed within the same day. The Tedlar bags were emptied completely after the analyses using a vacuum pump and cleaned by sequential flushing with nitrogen gas for reuse. A study, conducted on the sampling of reduced sulfur compounds, showed that mixtures of reduced sulfur gases in moist air at low ppm concentrations can be kept in Tedlar bags for up to 7 days with no appreciable losses (NCASI, 1993).

2. 2. 2 Gas sample analysis

Gas samples were analyzed for sulfurous compoun-

ds using a Gas Chromatograph/Flame Photometric Detector (GC/FPD) (Model 250H, TRACOR Inc., Austin, TX). A sample pump connected to the sample valve effluent line of the GC/FPD was used to draw a gas sample from the Tedlar bag attached to the sample valve inlet line. The detector signals from the GC/FPD were processed using an electronic integrator (Model SP4270, Spectra Physics, San Jose, CA). The GC separation was carried out using a 30″ × 1/8″ Teflon column packed with Super Q 80/100 (Alltech Associates, Inc.). Characteristically, the response of FPD detectors to the organo-sulfur compounds are not linear. The signal varies approximately linearly with square of the sulfur concentration. Therefore, the log of peak area is linearly related to twice the log of the sulfur concentration. An instrument calibration curve was developed using several dilutions of a standard gas mixture containing 248 ppm of hydrogen sulfide (H_2S), 249 ppm of methyl mercaptan (MM), 509 ppm of dimethyl sulfide (DMS) and 248 ppm of dimethyl disulfide (DMDS) (Specialty Gases, Inc., Ocala, FL). The known dilutions were prepared by injecting known amounts of the standard gas into known volumes of nitrogen gas stored in Tedlar bags. Due to analytical difficulties in measuring DMDS, which resulted from peak spreading due to strong retention of the analyte by the column type and conditions employed, the limit of detection for the analytical method was only 1.0 ppmv DMDS. Therefore, the concentration range studied in this study was limited to values above 10 ppmv.

2. 3 Filter materials

For this study two different types of filter materials that were obtained from Atlas Peat and Soil, Inc. (Boynton Beach, FL) were used. The composition, physical and chemical characteristics of these materials are given in Table 1. Particle size distributions of the materials are provided in Table 2. The filter material used in the first biofilter column was a compost/pine mulch mixture, and the material used in the second column was a compost/bark nugget mixture. Due to the rather large size of the bark nuggets, the second column biofilter material had a higher percentage of large particles. In fact, more than 50% of the materials in the second column was larger than 9.5 mm (equivalent diameter). The larger particle size used in the second column may have had an advantage in terms of pressure drop but was at a disadvantage in terms of active surface area for microbial growth.

Table 1. Physical and chemical characteristics of the filter materials used.

Property	Column-1	Column-2
Organic matter content (wt%)	75.1	81.0
Initial pH	7.27	6.50
Bulk density (g/ml)	0.27	0.26
Porosity (vol.%)	50.5	37.4
Carbon content (wt.%)	32.54	39.81
Nitrogen content (wt.%)	0.146	0.792
Composition[a]	50% pine mulch; 50% compost	25% compost; 75% deco-pine bark nugget

[a]Source: Atlas peat and soil Co., Boynton beach, FL.

Table 2. Particle size distribution of the filter materials.

Particle size range[a]	Column-1	Column-2
>9.5 mm	7.3%	54.6%
4.8~9.5 mm	16.5%	9.1%
2.0~4.8 mm	23.6%	8.4%
0.85~2.0 mm	17.6%	7.6%
0.43~0.85 mm	13.7%	6.8%
<0.43 mm	21.3%	13.5%

[a]Equivalent diameter.

Compost mixtures were used in the biofilters without any preliminary size and microbial treatment. The filter materials may be sieved to remove small particles, in order to decrease pressure drop. The filter materials are generally seeded with activated sludge from wastewater treatment plants to introduce the microorganisms necessary for the removal of poorly biodegradable pollutants, such as xenobiotics (Ottengraf *et al.*, 1986). A disadvantage of using activated sludge is that it can clog the biofilter and create excessive pressure drops. It is also known that composts are rich in the number and variety of some but not all microorganisms are necessary for biodegradation. However, the com-

post mixtures used in this study were not seeded.

3. RESULTS AND DISCUSSION

3. 1 Effect of pressure drop on biofilter operation

Since pressure drop is one of the major conditions leading to greater energy usage and operating costs for biofilters, an assessment of the pressure drop for filter materials is necessary in order to determine the economic feasibility of using biofilters for air pollution control (Phillips *et al.*, 1995). Pressure drop in biofilters is caused by resistance of the filter materials to gas flow and by head losses in transporting the waste gas through the piping system. The pressure drop values measured in this study are primarily due to the resistance of the filter materials to gas flow.

Observed pressure drop values in a filter bed depend on several parameters including superficial gas velocity, water content, porosity and particle size distribution of the filter materials. It was observed that after a typical filter wash with distilled water, the resistance to gas flow in the filter bed increased. Most of the time, it became necessary to re-adjust the plastic ball-valves in order to maintain a constant flow rate to the biofilter.

The pressure drop variations determined as a function of volumetric surface loading rate for the two compost mixtures are shown in Figure 2. The volumetric surface loading rate can be defined as the gas flow rate applied per unit surface area of the filter material. Typical surface loading rates used to treat odorous gases from wastewater treatment plants are less than 100 $m^3/m^2/h$ for soil bed systems and uncontrolled biotowers (Hartenstein, 1987). However, surface loadings of 500 $m^3/m^2/h$ have been successfully applied in controlled biotowers (Dragt and Ottengraf, 1985).

The pressure drop values found for the first compost mixture were extremely high. This extreme was, somewhat, expected since the percentage of small particles present was quite high for this particular compost mixture. At a surface loading of 100 $m^3/m^2/h$ to the first compost mixture, a pressure drop of approximately 61.8 mmH_2O was measured. Typical values reported in the literature for biofilters of one meter depth are in the range of 5.2 to 25.8 mmH_2O (Hartenstein, 1987). For the same surface loading of 100 $m^3/m^2/h$ on the second compost mixture, the pressure drop was measured as only 10.3 mmH_2O.

3. 2 Effect of temperature on operating conditions

Temperature affects two major processes in the bio-

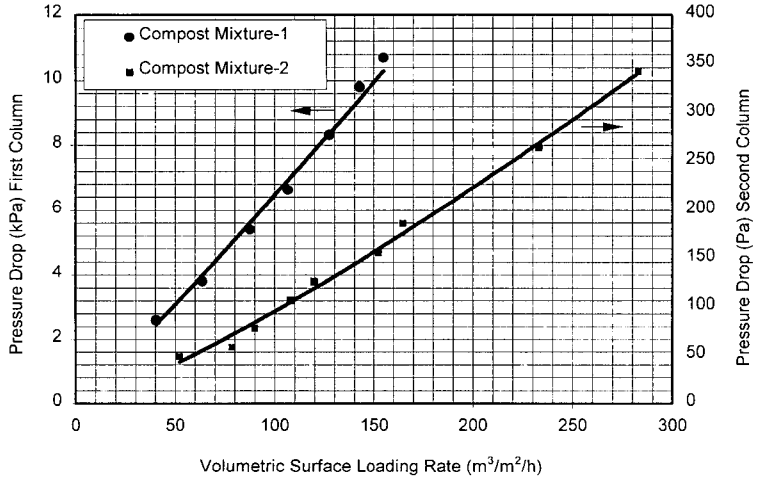

Fig. 2. Pressure drop as a function of volumetric surface loading.

filters; microbial degradation and mass transfer. Increase in temperature results in increased metabolic activity in many microorganisms; hence increasing the consumption of pollutants. However, higher temperatures may cause lower gas solubilities. The advantage, thus, gained by increasing the microbial activity may be offset by decreasing the mass transfer rates. Another effect of temperature that should be taken into consideration is the optimum temperature range for microorganisms. Biofilters, usually utilize mesophilic microorganisms, which have an optimum temperature range between 25 and 35°C. Also, the microorganisms reported in the literature that are known to degrade DMDS appear to have an optimum degradation temperature of 30°C.

The biofilter columns used in these experiments did not have the means to control the filter bed temperature. However, the control of room temperature allowed the biofilter operating temperature to remain at a nearly constant level. During routine experiments the temperatures of the biofilter beds were observed to fluctuate between 22 and 28°C, with a value of 25°C being observed most frequently. The temperature profile along the biofilter columns was monitored during operation to observe any possible temperature increase due to exothermic reactions taking place. However, there were no observable temperature changes in different sections of the biofilter columns. The pollutant concentrations treated in the biofilters probably were not high enough to create significant changes in the temperature.

3. 3 Effect of water content on operating conditions

Water content is one of the most important operating parameters for biofilters, since microbial degradation apparently takes place in the aqueous phase. Control of water content, therefore, is vital for maintaining high treatment efficiencies. The optimum water content for compost-based biofilters is reported to be in the range from 40 to 60% (Ottengraf, 1986). Filter beds tend to dry out rather easily, however, due to the moisture stripping effect of unsaturated incoming waste gas. Usually the moisture is stripped from the inlet section of the biofilters.

In these experiments, the incoming gas was humidified in a humidification chamber to achieve relative humidity levels equal to or greater than 95%. The filter beds were also washed with distilled water once every two weeks for both moisture and acidity control. However, due to the observation of increasing acidity in the filter bed, the washing frequency was changed to once a week. Later this practice was abandoned due to an accumulation of excessive water in the filter bed. The drainage of wash water, especially in the first filter bed, was hindered due to counter-current flow of incoming gas. Excessive water in the filter material can be harmful to the operation of the biofilter by creating anaerobic zones. The water contents in both biofilter columns were maintained between 60 and 65%. Under these conditions no significant problems were encountered that resulted from water content variations in the beds during the entire period of operation of the biofilter systems.

3. 4 Effect of filter bed acidity on operating conditions

Acidification can be a major problem in biofilters that are used to treat sulfurous compounds. Sulfuric acid is formed as an end product in most aerobic biological degradation reactions involving sulfur. The decrease in pH, in turn, affects the microbial life existing in the filters. Some microorganisms are acidophilic and can resist acidification to pH values as low as 2. For example microorganisms treating H_2S are resistant to low pH levels (Yang and Allen, 1994). However, microorganisms involved in the degradation of methylated sulfur compounds require a neutral pH for optimum performance (Park *et al.*, 1993a, b; Shoda, 1993; Cho *et al.*, 1991, 1992a, b; Smith and Kelly, 1988a, b). Therefore, the filter beds, especially those used to treat high pollutant loads, should be continuously monitored for pH and chemically treated to increase the pH, if necessary.

Usually in biofilters, the inlet section of the beds is the location that becomes most acidic during operation

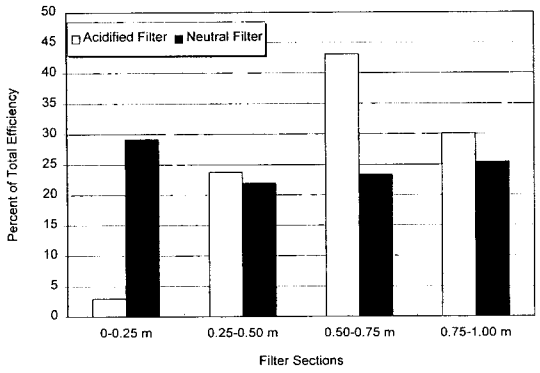

Fig. 3. Changes in removal efficiency corresponding to bed acidification for the first biofilter column after 5 months of operation.

due to the high microbial activity occurring in this section. If the acidity is not neutralized, the inlet section of the biofilter may lose its microbial activity. This specific situation is illustrated in Figure 3. The neutral filter depicted in this figure is the compost mixture in the first biofilter column in the early stages of operation, when the initial pH was about 7. After approximately five months of operation, the filter efficiency in the lower 25 cm of the bed had decreased drastically. The acidified filter at this stage of operation had an average pH value around 3.

The removal efficiency of the biofilters after five months of operation started declining. The regular filter washes with distilled water did not seem to be very efficient in decreasing the acidity levels in the biofilter columns. It became necessary to neutralize the filter beds with chemicals. For this purpose sodium bicarbonate ($NaHCO_3$) or sodium carbonate (Na_2CO_3) were used. Although some researchers suggest the use of lime (Ottengraf *et al.*, 1984), this may result in clogging of the filter material due to the formation of insoluble residues of calcium sulfate. Each biofilter column was treated with 2 liters of the prepared solutions at specific times. Changes in the acidity of the biofilter columns were monitored by measuring the pH of leachate from the washings. The pH of the leachates after the application of neutralizing agents are shown in

Table 3. Effects of washing of filter beds.

No.	Day[a]	Washwater	pH of washwater	pH of leachate-1	pH of leachate-2
1	154	0.5 M $NaHCO_3$	8.13	2.75	6.80
2	156	0.5 M Na_2CO_3	11.35	3.43	9.30
3	162	0.5 M Na_2CO_3	11.35	6.66	10.38
4	176	0.1 M Na_2CO_3	10.60	6.05	9.73

[a]Biofilter columns started operation on day 1.

Table 3. As can be seen from this table, it was more difficult to neutralize the first compost mixture which had been allowed to become excessively acidic. Due to the high percentage of small particles, the washwater did not percolate easily through the bed and the upper parts of the filter bed were preferentially neutralized. This condition may be observed in Figure 4, where the pH values of the filter materials before and after treatment with sodium bicarbonate/sodium carbonate solutions are shown. After the third wash the upper portions of the biofilters became basic, which may also be harmful to the microorganisms. The utmost care, therefore, must be taken when neutralizing the accumulation of acidity on biofilter columns. Lower molarity solutions, such as 0.1 M or lower may be used in washing the filters. Also, it may be advantageous to use downflow biofilters for the treatment of sulfurous gases, where the waste gas is introduced from the top of the biofilter. In this case, when alkaline washwater is introduced to the system it will immediately contact the most acidified and microbially active section of the biofilter. However, if only untreated washwater is used in this configuration, premature acidification of the lesser used sections of the bed could occur.

As a consequence of decreased efficiencies due to extreme acidification of the filter beds, methyl mercaptan (MM), an intermediate oxidation product of DMDS, was observed in the gas samples collected. The MM concentrations observed were very low, probably in the ppbv levels. From the peak areas observed on the analytical gas chromatograms, it was concluded that the concentrations of MM decreased progressively along the filter bed, being totally removed at the bed outlet. The MM peak areas increased with inlet DMDS

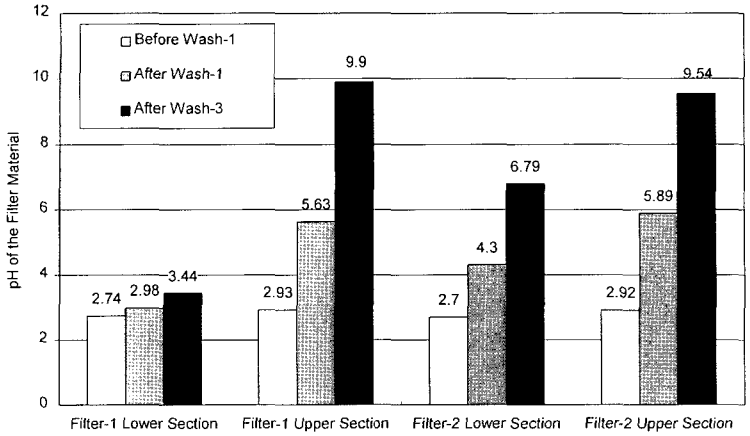

Fig. 4. Change of pH of filter materials with washes.

concentrations. This emergence of MM supports the degradation pathway proposed by Smith and Kelly (1988b) for T.thioparus, Strain E6. Hydrogen sulfide, which is also a by-product of DMDS degradation (Smith and Kelly, 1988b) was not observed in the gas samples. This is possibly due to the rapid degradation of H_2S on compost beds.

At the optimal condition of pH and water content, the inlet concentrations of DMDS were increased at approximately 20 sLpm of constant flow rates as shown in Figure 5 in order to see the performance of the dual column biofilters. At inlet DMDS concentrations near 45 ppmv, the removal efficiencies for both column filter materials decreased from about 95% to approximately 85%. When the inlet concentration was raised to about 50 ppmv, removal efficiencies were reduced to about 70%. Air flow rates were also varied from 17.4 to 53.4 sLpm to observe the effect of air flow rates on the removal efficiencies of DMDS. In general, a decrease in elimination efficiency was observed at low flow rates. A reduction in removal efficiencies was observed for residence times less than 29 seconds. The maximum elimination capacity for both compost mixtures was found to be in the range from 7.5 to 10 g-DMDS/m^3/h. The first column compost mixture was observed to have a slightly higher maximum elimination capacity than the second column mixture, probably due to

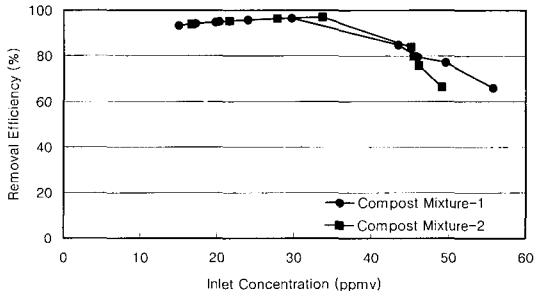

Fig. 5. Effect of inlet DMDS concentration on removal efficiency for the dual column biofilters.

the presence of a higher fraction of small particles.

4. CONCLUSIONS

The aim of the research described here was to understand the nature of the compost biofiltration of dimethyl disulfide, and to obtain qualitative and quantitative data on the influence of operating conditions on DMDS removal. The following results were obtained from this study:

(1) The first column filter material (compost/pine mulch mixture) displayed extremely high pressure drops due to the presence of small particles. It would not be economically feasible to use this compost mixture

in biofiltration even though the observed treatment efficiencies were quite high. However, by sieving and removing the small particles, the pressure drop may be optimized for this filter material. The second column filter material (compost/bark mixture) produced pressure drops that were within normal ranges reported in the literature. Also, removal efficiencies were similar to those obtained with the first column filter material. In a scaled-up biofilter application, the second column filter material should be chosen over the first column material, unless the latter is pretreated for optimum particle size distribution.

(2) The compost mixtures contained the necessary microorganisms for the degradation of DMDS without seeding. Preliminary inoculation of the compost mixtures using activated sludge was not necessary. Therefore, it can be stated that microorganisms capable of degrading dimethyl disulfide are abundant in nature and present in natural native wood (pine)/compost mixtures.

(3) The microorganisms responsible for the degradation of DMDS were found to be sensitive to pH changes. The optimum pH condition was found to be near neutral. This condition appeared to be generally the case with microorganisms known to degrade methylated sulfur compounds. Higher pollutant loading rates were achieved for less acidic filter beds.

(4) During high acidity conditions in the filter beds, methyl mercaptan (MM) was observed in the gas samples collected. Appearance of MM was probably due to decreased microbial activity in the lower portions of the biofilter. The presence of MM in the gas samples verified the findings of Smith and Kelly (1988b) that MM is an intermediate in the degradation of DMDS.

(5) Considering the neutral pH range required and the presence of MM, it is likely that the microorganisms present in the biofilters used in this research are similar to the T.thioparus species reported in the literature (Park *et al.*, 1993a, b; Shoda, 1993; Cho *et al.*, 1991, 1992a, b; Kanagawa and Mikami, 1989; Smith and Kelly, 1988a, b).

(6) In general, when the inlet concentration was increased at a constant flow rate, removal efficiencies were reduced. Air flow rates were also varied to observe the effect of air flow rates on the removal efficiencies of DMDS. In general, a decrease in elimination efficiency was observed at low flow rates. The maximum elimination capacity for the dual column biofilters was found to be in the range from 7.5 to 10 g-DMDS/m^3/h.

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