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Control of Dimethyl Sulfide Emissions Using Biofiltration

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Laboratory scale experiments were conducted to evaluate the performance of a biofilter for eliminating dimethyl sulfide(DMS). A commercial compost/pine bark nugget mixture served as the biofilter material for the experiments. The gas flow rate and DMS concentration entering the filter were varied to study their effect on the biofilter efficiency. The operating parameters, such as the residence time, inlet concentration, pH, water content, and temperature, were all monitored throughout the filter operation. The kinetic dependence of the DMS removal along the column length was also studied to obtain a quantitative description of the DMS elimination. High DMS removal efficiencies(>95 %) were obtained using the compost filter material seeded with activated sludge. DMS pollutant loading rates of up to 5.2 and 5.5 g-DMS/m^3/hr were effectively handled by the upflow and downflow biofilter columns, respectively. The macrokinetics of the DMS removal were found to be fractional-order diffusion-limited over the 9 to 25 ppm range of inlet concentrations tested. The upflow column had an average macrokinetic coefficient(K_f) of 0.0789 $\pm$ 0.0178 $ppm^{1/2}$/sec, while the downflow column had an average coefficient of 0.0935 $\pm$ 0.0200 $ppm^{1/2}$/sec. Shorter residence times resulted in a lower mass transfer of the pollutant from the gas phase to the aqueous liquid phase, thereby decreasing the efficiency.

Key words : dimethyl disulfide, biofilter, residence time, inlet concentration, removal

1. Introduction

Odor is one of the most noticeable forms of air pollution and has been the subject of most complaints related to commercial and industrial effluent intrusions on surrounding communities. Organo-sulfur compounds are commonly identified in malodorous gases, and the more important odorous organo-sulfur gases include carbon disulfide (CS_2), carbonyl sulfide(COS), mercaptans(R-SH), and sulfides(R-S-R). Their major sources are from the paper and pulp manufacturing industry, waste water treatment plants, chemical manufacturing industry, and petroleum refineries[1,2]. Hydrogen sulfide, itself a very odorous gas, is generally emitted along with organo-sulfur emissions, yet in much higher concentrations, thereby often masking the latter[3]. However, in the absence of hydrogen sulfide, the strong odor of organo-sulfur gases can be distinctly recognized[4].

Conventional odor removal techniques include incineration, chemical oxidation, adsorption using activated charcoal, scrubbing, dilution using taller stacks, and masking with pleasant-smelling substances[5]. Although satisfactory removal efficiencies can be achieved using these techniques[6], they all have serious limitations, for example, high fuel, water, and chemical requirements, and high capital and operational costs. In some cases, instead of destroying the pollutant or generating by-products, these techniques actually condense or concentrate it, which can create environmental problems requiring further treatment. Moreover, methods such as dilution and masking do not provide a solution to the pollution problem. Re-

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cently, alternative control technologies using biological degradation have become attractive. In general, biological techniques, particularly biofiltration, involve lower capital and operating costs than their traditional counterparts, while still obtaining high removal efficiencies[7].

In Europe, biofilters are considered as standard odor control techniques[8]. Their use has recently been expanded to the treatment of dilute VOCs and toxic air pollutant emissions[9~11]. Biofiltration has been used sporadically in the United States since the mid-1950s and it has also been used occasionally in Korea since the mid 1990s. The reason for this lack of interest in the United States and Korea is the limited availability of systematic data concerning the performance of these systems. Most of the operating system designs are based on "rule of thumb" criteria, which makes the system performance irreproducible and uncertain. Another reason for their slow acceptance is suspicion about the effectiveness of "low-technology" systems. One remedy for this problem is to perform exhaustive studies to evaluate biofilter performance in controlling the emission of various types of air pollutants.

Accordingly, the current study evaluated a laboratory-scale compost biofilter for removing dimethyl sulfide(DMS), including the maximum elimination capacity(MEC) of the compost biofilter for DMS, macrokinetics of the biofiltration, and effects of the gas residence time on the removal efficiency. Two identical biofilter columns with an upflow and downflow configuration were operated simultaneously for comparison.

2. Experimental Methods

2.1 Laboratory Biofilter System

A dual-column laboratory-scale compost biofilter system was used in the current study. A schematic of the system is shown in Fig. 1. The main components of the system were an air blower, humidification chamber packed with 5 cm Pall rings, and two biofilter columns. One column was operated by flowing gas in an upflow direction, while the other column operated in a downflow direction. The columns were constructed of transparent acrylic cylinders with inner diameters of 0.15 m and lengths of 1.34 m. Each column was fitted with five gas-sampling ports, five compost-sampling ports, and five pressure ports. The transport of gas to and from the bioflilter columns was through 1-1/4" diameter SCH40 PVC pipes. The blower moved room air through each column at flow rates within a range of 17 to 36 L/min. The flow rates were measured using calibrated orifice meters.

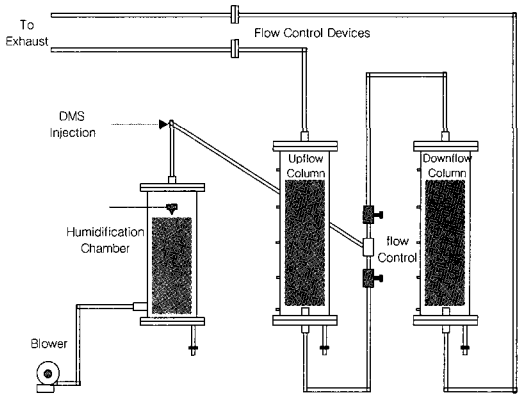

Fig. 1. Schematic Diagram of Biofilter System.

The injection of known quantities(9~27 ppm) of DMS was achieved using either pre-mixed cylinder gas(National Specialty Gases, Durham, NC) or a constructed system designed to control the evaporation of liquid DMS. The latter system consisted of a standard dilution flask inside a refrigerator that provided a controlled temperature of 5±2 ℃, which lowered the vapor pressure of the DMS to about 0.25 atm. The flask was also pressurized using a tank of nitrogen at 8 psig to further reduce the vapor pressure to about 0.18 atm. Known quantities of DMS vapor were then drawn off from the flask and fed into the biofilter system using a metering valve and calibrated small-bore rotameter.

2.2 Filter Material

The filter material was a commercial compost/pine bark nugget mixture(Atlas Peat and Soil, Boynton Beach, FL). The composition and physical and chemical properties of the mixture are presented in Table 1. The material was sieved prior to use to remove any particles smaller than 1 mm in diameter or larger than 20 mm in diameter. The

substantial weight fraction of the large particles in the mixture exhibited a very small pressure drop, yet still provided sufficient surface area for microbiological activity. The compost was seeded with return activated sludge from the Buckman wastewater treatment facility located in Jacksonville, FL.

Table 1. Physical and Chemical Properties of Biofilter Material

Property	
Organic Matter Content (wt%)	81.0
Initial pH	5.98
Bulk Density(g/mL)	0.485
Porosity(vol%)	52.1
Water Content (wt% dry basis)	72.4
Particle Size Distribution (wt%)	
>9.5 mm	49.7
4.8 ~ 9.5 mm	16.3
2.0 ~ 4.8 mm	11.4
0.85 ~ 2.0 mm	19.3
0.43 ~ 0.85 mm	3.3
< 0.43 mm	0
Composition	30% compost
	70% pine bark nuggets

Source : Atlas Peat and Soil Co., Boyton Beach, FL.

2.3 Gas Sampling and Analysis

2.3.1 Gas Sampling

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

The gas samples collected from the ports were slowly expanded into Tedlar bags using the slight positive pressure of the carrier gas in the columns. A staggered sampling method, incorporating a time delay, was used for filling the Tedlar bags at the different bed levels. This method allowed the same stream of pollutant gas to be sampled at each port based on considering the time taken for the gas to reach that port.

The samples in the Tedlar bags were analyzed within the same day. The Tedlar bags were completely emptied after the analyses using a vacuum pump and cleaned by sequential flushing with nitrogen gas for reuse. A previous study on the sampling of reduced sulfur compounds, found that mixtures of reduced sulfur gases in moist air at low ppm concentrations could be kept in Tedlar bags for up to 7 days with no appreciable losses[12].

2.3.2 Gas Sample Analysis

The gas samples were analyzed for sulfurous compounds 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"x1/8" Teflon column packed with Super Q 80/100(Alltech Associates, Inc.). The temperatures of the detector and oven were maintained isothermally at 120 and 100℃, respectively. Nitrogen was used as the carrier gas at a pressure of 12 psig. The analytical system was calibrated with certified pre-mixed (DMS in N_2)cylinder gas using standard dilution methods. The detection limit of the analytical device was 0.5 ppm DMS. The overall precision and accuracy of the sampling and analysis were estimated at less than 5 %(relative standard deviation) and 10 %, respectively.

2.4 Compost Analysis

2.4.1 pH

pH measurements for both the compost and water samples were made using a calibrated pH meter (Model M245, Corning) that was accurate to ± 0.01 pH. The pH of the compost particles was determined using the method described by Robarge and Fernandez[13]. After being dried overnight at

a temperature of 105℃, the compost particles were mixed with deionized water to give a liquid to solid ratio of 10. Although this method does not provide the actual pH of the compost, it gives an indication of the hydrogen ions transferred to the water.

2.4.2 Water Content

The method suggested by Robarge and Fernandez[13] was used to determine the water content. A known amount of wet compost was weighed and then dried at 105 ℃ for 24 hours. After a constant dry weight was achieved, the dry compost was weighed again. The difference between the wet and dry weights divided by the initial wet weight of the compost provided the water content of the compost on a wet basis, expressed as a percentage.

2.4.3 Particle Size Distribution

The compost sample was first dried at 105℃ for 24 hours, then sieved using a series of standardized sieves(USA Standard Testing Sieve, A.S.T.M. E-11 Specification) with different mesh sizes. The compost retained on each sieve was weighed to determine the particle size distribution percentage.

2.4.4 Organic Matter Content

The organic matter content of the compost was determined using the Loss-On-Ignition(LOI) method, described by Robarge and Fernandez[13]. Accordingly, a known amount of oven-dried compost in a dish was ignited in a muffle furnace at a temperature of 450 ℃ for more than 12 hours. During this time period, the organic matter was completely oxidized to carbon dioxide and water and only the inorganic components of the compost sample remained. The ratio of the difference between the initial and final weights of the compost to the initial weight was then used to estimate the organic matter content of the compost sample.

2.4.5 Bulk Density and Porosity

A known volume of compost was weighed. The ratio of the weight to the volume provided the bulk density of the compost sample. As described by Hartenstein[14], porosity is the ability of the compost to hold water. An initially weighed sample of dry compost was thoroughly mixed with deionized water. After the excess water was drained by gravity, the wet compost was reweighed. The porosity was then estimated based on the ratio of the difference between the final wet and initial dry weights to the final wet weight.

3. Results and Discussion

One of the most important operating parameters for a biofilter is its moisture content. Therefore, since moisture can be stripped from the filter material when passing non-saturated gas through the bed, the filter was periodically washed to restore any lost water. Another important function of the biofilter washing in the current study was to remove the build-up of sulfuric acid, the end-product of DMS degradation.

To maintain the pH of the biofilter beds at near-neutral conditions, the upflow and downflow columns were periodically washed with either deionized water or a weak alkaline solution of sodium carbonate(Na_2CO_3). The filters were washed from the top of the column, and any excess solution was drained from the bottom. Washing with water alone does not neutralize the bed acidity, but only dilutes and flushes any sulfuric acid from the filter. In contrast, chemical washing neutralizes the sulfuric acid and flushes the resulting residue. After several weeks, it was discovered that washing with just water was usually adequate to control the acidity in the upflow biofilter column, as the build-up of sulfuric acid tended to accumulate at the bottom of the bed. The chemical washing was distinctly effective in the downflow biofilter column as it created the desired alkaline conditions, and, since the acid build-up occurred at the top of the filter, the acid was immediately neutralized when washed. Whereas washing with water alone tended to push the acid downward through the column, creating acidification in the bed. As such, the use of the chemical washing with the downflow filter provided a distinct advantage when removing the organo-sulfur compounds.

3.1 Pollutant Loading

The flow rate at which the pollutant-laden gas stream enters the biofilter and concentration of the pollutant in the gas stream are often combined into a single quantity, the pollutant loading rate. The

pollutant loading rate is defined as the pollutant mass introduced into the biofilter per unit volume of filter material per unit time. There is a limit to the pollutant loading rate that can be treated by a biofilter before it begins to overload, i.e. the maximum elimination capacity(MEC) of the pollutant that can be degraded without inhibiting the normal microbiological functions. The MEC varies for different pollutants, filter materials, and operating conditions and thus needs to be assessed for every biofilter application.

The maximum elimination capacities for the upflow and downflow biofiter columns were experimentally determined. For the upflow column, the MEC was 5.2 g-DMS/m^3/hr, while the downflow column appeared to have a slightly higher value of 5.5 g-DMS/m^3/hr. A graph of the DMS Elimination Capacity vs. the DMS Loading Rate for the downflow column is shown in Fig. 2. These results compared favorably with those obtained in previous studies, which varied from 3.5 to 12.2 g-DMS/m^3/hr[15~18]. However, the comparative studies were conducted using laboratory-scale peat biofilters, where the lowest value was obtained using peat seeded with night soil sludge, and the highest value was obtained using two types of highly specific microorganisms, believed to symbiotically degrade DMS.

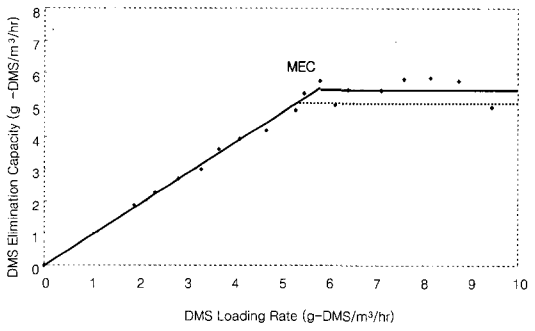

Fig. 2. DMS Elimination Capacity and MEC for Downflow Column.

3.2 Macrokinetics of Biofiltration

Ottengraf's biophysical model is represented in Fig. 3. According to this model a wet biolayer of thickness surrounds the filter particle and the following assumptions are made[19]:

- The interface resistance to mass transfer in the gas phase can be neglected and the biolayer pollutant concentration at the interface is assumed to be in equilibrium with the pollutant concentration in the bulk gas phase;
- In the biolayer, the pollutants(substrates) are transported by diffusion, which can be described by an effective diffusion coefficient D' ;
- The biolayer thickness, δ , is small compared to the diameter of the filter material particles;
- The microkinetics of the substrate elimination reactions in the biolayer can be described by Monod kinetics; and
- The flow of gas through the filter bed is a plug flow.

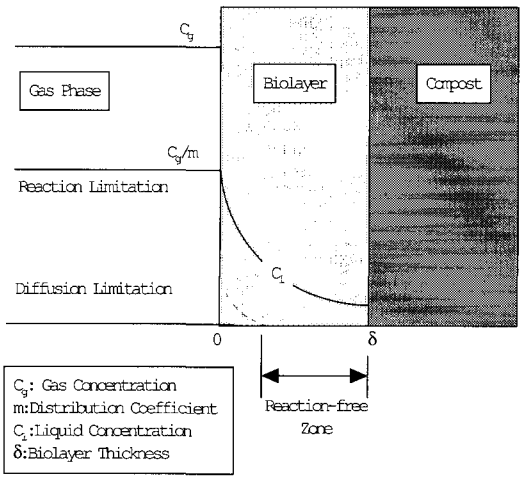

Fig. 3. Graphical Representation of Biophysical Model for Biofilter(Source : Ottengraf and Van den Oever[20]).

Using these assumptions, the following differential equation describing the pollutant concentration, C, inside the biolayer can be written:

$$D' \frac{d^2 C}{dx^2} - R = 0 \qquad (1)$$

where x is the distance along the biolayer and R is the microbial degradation rate. The microkinetics of the microbial degradation is described by the following Monod equation:

$$-\frac{dC}{dt} = \frac{V_{max} \cdot C \cdot B}{K_s + C} \qquad (2)$$

where V_{max} is the maximum reaction rate, K_s is

the half-saturation constant, B is the microbial population density, and t is the reaction time. The Monod kinetics equation can be simplified for certain situations: when $K_s >> C$, the first-order kinetics result and equation can be reduced to:

$$\ln\left(\frac{C_0}{C}\right) = k_1 \cdot t \quad (3)$$

where k_1 ($=V_{max} \cdot B/K_s$) is the first-order microkinetic coefficient, C_0 is the initial pollutant concentration. When $k_1 \cdot C$ is substituted for R in Equation 1, a macrokinetic equation similar to the microkinetic equation (3) can be obtained:

$$\ln\left(\frac{C_0}{C}\right) = K_1 \cdot t \quad (4)$$

where K_1 is the first-order macrokinetic coefficient.

When $C >> K_s$, the microbial reaction rate is said to be zero-order and the Monod equation reduces to:

$$C_0 - C = k_0 \cdot t \quad (5)$$

where k_0 ($=V_{ma} \cdot B$) is the zero-order microkinetic coefficient. Substituting k_0 for R in Equation 1 and solving the differential equation yields the corresponding macrokinetic equation. However, for zero-order microbial degradation, Ottengraf and Van den Oever[20] describe two situations. Referring to the biophysical model depicted in Fig. 3, these conditions are :

- Reaction limitation : There is no limitation in the diffusion of the pollutants(substrates) into the wet biolayer from the gas phase. The degradation of the pollutant is only controlled by the microbial degradation rate. In this situation the biolayer is fully active(solid line in Fig. 3).
- Diffusion limitation : The diffusion of the pollutants into the wet biolayer is limited. Thus, the pollutants can not penetrate through the whole biolayer. As a result, the biolayer is not fully active and there are reaction-free zones in the biolayer. In this case, the diffusion is the rate limiting step(dashed lines in Fig. 3).

For reaction limited conditions the macro-kinetic equation can be written in a manner similar to zero-order micro-kinetic equation 5 as:

$$C_0 - C = K_0 \cdot t \quad (6)$$

where K_0 is the zero-order reaction limited macrokinetic coefficient. The macrokinetics for diffusion limited degradation provides a fractional order dependence that is developed in a simple form:

$$(C_0)^{1/2} - (C)^{1/2} = K_f \cdot t \quad (7)$$

where K_f is the fractional-order diffusion limited macrokinetic coefficient and C_o is the inlet concentration.

To determine the kinetic dependence of the DMS removal through the biofilter columns, plots of $\ln(C_0)$-$\ln(C)$, C_0-C, and $(C_o)^{1/2}$ - $(C)^{1/2}$ versus the residence time were prepared according to Equations 5, 7, and 8, respectively. The data were then fit with corresponding least-square linear regression equations, and the plots with the highest correlations determined. For the experimental conditions studied, the DMS kinetics appeared to be fractional-order diffusion-limited. Typical graphs of the data plotted for first-order, zero-order, and fractional-order diffusion limited kinetics are shown in Figs. 4 and 5 for the upflow and downflow columns, respectively. Each line was plotted using a least-square linear regression analysis, where the error bars represent three standard deviations(99 % confidence level). The data were obtained for an inlet concentration of 20.6 ppm DMS into the upflow column and 22.8 ppm DMS into the downflow column. The empty-bed gas residence times were 45 seconds for both columns. The graphs in Figs. 4 and 5 clearly show high correlations for the fractional order macrokinetics. Summaries of the macrokinetic data over a range of inlet concentrations and gas retention times are presented

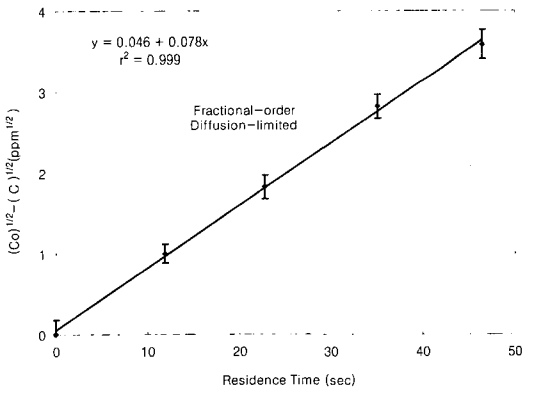

Fig. 4. Best Fit Kinetic Data for Upflow Column (@20.6 ppm of DMS Inlet Conc.).

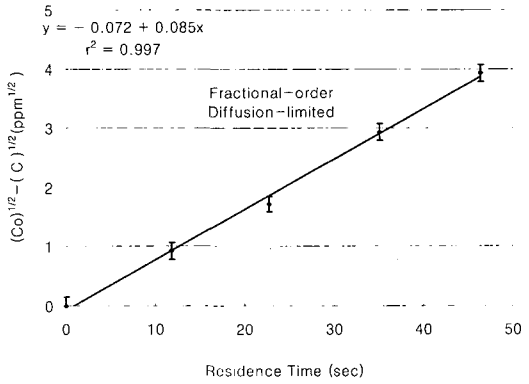

Fig. 5. Best Fit Kinetic Data for Downflow Column (@22.8 ppm of DMS Inlet Conc.).

in Tables 2 and 3 for the upflow and downflow columns, respectively. The upflow column had an average macrokinetic coefficient K_f of 0.0789 $ppm^{1/2}$/sec with a standard deviation of 0.0178 $ppm^{1/2}$/sec, while the downflow column had an

Table 2. Summary of Kinetic Data for Upflow Column

Inlet (ppm)	Zero-order Removal		Fractional-order Removal	
	K_0 ppm sec^{-1}	r^2	K_f $ppm^{1/2}$ sec^{-1}	r^2
8.8	0.228	0.989	0.0560	0.992
10.3	0.437	0.984	0.1111	0.995
11.4	0.324	0.966	0.0779	0.993
13.0	0.305	0.975	0.0580	0.990
13.2	0.490	0.992	0.0947	0.998
14.8	0.459	0.973	0.0964	0.994
15.1	0.328	0.948	0.0710	0.991
17.0	0.348	0.946	0.0680	0.992
18.0	0.562	0.991	0.1015	0.995
18.7	0.391	0.986	0.0730	0.994
19.4	0.582	0.988	0.0996	0.998
Mean	0.405	0.976	0.0825	0.994
Median	0.391		0.0779	
Std Dev	0.112		0.0189	
Max	0.582		0.1111	
Min	0.228		0.0560	

Note : A linear regression analysis of the first-order removal revealed average values for K_1 and r^2 at 0.0772(sec^{-1}) and 0.953, respectively.

Table 3. Summary of Kinetic Data for Downflow Column

Inlet(ppm)	Zero-order		Fractional-order	
	K_0 ppm sec^{-1}	r^2	K_f $ppm^{1/2}$ sec^{-1}	r^2
9.5	0.263	0.977	0.0691	0.999
10.3	0.428	0.955	0.1090	0.989
11.5	0.332	0.963	0.0809	0.993
13.1	0.336	0.951	0.0717	0.983
13.4	0.534	0.990	0.1070	0.997
13.6	0.377	0.979	0.0809	0.996
15.2	0.444	0.969	0.0960	0.992
15.3	0.632	0.983	0.1210	0.994
17.7	0.463	0.929	0.0750	0.968
18.0	0.602	0.965	0.1160	0.994
18.2	0.525	0.956	0.1053	0.998
19.2	0.627	0.953	0.1210	0.990
20.0	0.547	0.964	0.1014	0.993
22.8	0.490	0.974	0.0870	0.996
25.1	0.494	0.959	0.0586	0.996
Mean	0.473	0.964	0.0935	0.990
Median	0.490		0.0960	
Std Dev	0.111		0.0200	
Max	0.632		0.1210	
Min	0.263		0.0586	

Note : A linear regression analysis of the first-order removal revealed average values for K_1 and r^2 at 0.0772(sec^{-1}) and 0.953, respectively.

average K_f of 0.0935 $ppm^{1/2}$/sec with a standard deviation of 0.0200 $ppm^{1/2}$/sec.

3.3 Effect of Residence Time and Concentration on Removal Efficiency

The effect of decreasing the overall gas flow rate on the removal efficiency of DMS was studied by maintaining a relatively constant pollutant loading on the biofilters. Decreasing the residence time resulted in a lower mass transfer of the pollutant from the gas phase to the aqueous liquid phase, thereby decreasing the efficiency. The empty-bed residence times and inlet DMS concentrations were varied to assess their effects on the biofilter removal efficiency. Note that at lower inlet concentrations(<15 ppm DMS), DMS concentrations of 0.5 ppm were assigned and the re-

sulting removal efficiencies were calculated. However, the actual removal of DMS may have been much higher(>95 %) for such data points.

The overall removal efficiencies were high (>95 %) with longer residence times, yet fell rapidly at high gas flow rates. At a residence time of 60 seconds, the removal efficiencies were >95 % for concentrations of up to 27 ppm DMS for both columns. At a 45-second residence time, similar high efficiencies were observed at concentrations of up to 20ppm DMS. For a 30-second residence time, high removal efficiencies >95 % were found at concentrations near 15 ppm DMS for the upflow filter and near 18 ppm DMS for the downflow filter.

4. Conclusions

High DMS removal efficiencies(>95 %) were obtained when using a compost filter material seeded with activated sludge. The maximum elimination capacity(MEC) that could be effectively handled by the upflow and downflow biofilter columns was 5.2 and 5.5 g-DMS/m^3/hr, respectively. The macrokinetics of the DMS removal were found to be fractional-order diffusion-limited over the range of inlet concentrations tested(about 9 to 25 ppm). The upflow column had an average macrokinetic coefficient of 0.0789 $\pm$ 0.0178 $ppm^{1/2}$/sec, while the downflow column had an average coefficient of 0.0935 $\pm$ 0.0200 $ppm^{1/2}$/sec. A shorter residence times resulted in a lower mass transfer of the pollutant from the gas phase to the aqueous liquid phase, thereby decreasing the efficiency.

Regular filter washing using either water or a weak alkaline solution(Na_2CO_3) effectively optimized both the moisture content and the pH in the biofilter beds during the experiments. However, in the downflow filter, the wash solutions were able to make quicker contact with the portions of the bed that had the lowest moisture content and highest acidification, which may have contributed to the slightly improved performance of the downflow column over the upflow column.

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