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Biofiltration of Wastewater Gas Emissions to Remove Volatile Organic Compounds (VOCs)

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Pilot plant tests were conducted at LACSD from 1993-1995, to evaluate the performance of conventional gaseous (vapor-phase) biofilters, and a modified biofilter, to scrub odorous gases and VOC emissions from municipal wastewater.

In the first phase, conventional biofilters with 3 feet (~ 0.92 m) of filter media consistently removed more than 95% of hydrogen sulfide gas, and reduced the Odor Units (Intensity) by 40-95%. The hydrocarbon gases, including benzene, toluene, xylene, butadiene, and some halogenated VOCs including chlorobenzene (CB), vinyle chloride (VC), carbontetrachloride (CT), dichloroethene (c-DCE), and dibromoethane (DBA) were removed up to about 95% with an overall average of about 65-90%.

Similarly, Dichlorobenzene (DCB), dichloroethane (DCA), and dichloroethylene (DCE) were removed between 25-80% with an overall average of about 50%. No effective removal was observed by the conventional biofilters for chloroform (CF), tetrachloroethylene (PCE), trichloroethylene (TCE), trichloroethane (TCA) and methylene chloride (DCM).

In the second phase, the bottom layer of the media was submerged up to 1.5 feet (~ 0.46 m) in tap water, and the overall filter depth was increased to 5 feet (~ 1.52 m).

The modified biofilters removed hydrogen sulfide between 50-99% with overall average of about 90%, and the Odor Units between 80-99%. Benzene (B), toluene (TOL) and xylene (XYL) were removed greater than 90%, with an overall average of about 95%.

Dichlorobenzene (DCB), dichloroethylene (DCE) and dichloroethane (DCA) were removed between 75-95%, with overall average of about 90%. Chloroform (CF) was removed between 20-95% with average of about 75%. Tetrachloroethylene (PCE) was removed 30-95%, with average of about 90%.Trichloroethylene (TCE), trichloroethane (TCA) and Methylene chloride (DCM) were removed between 25-95% with an overall average of about 75%.

Keywords: **Gaseous biofilters, Vapor-phase biofilters, modified biofilters withe submerged bottom layer, removal of hydrocarbon gases, removal of halogenated hydrocarbon gases, removal of carcinogenic gases.**

I. INTRODUCTION

Gaseous (vapor-phase) biofiltration is a simple, naturally self regenerative, and appropriate technology for both the developing and the developed countries, requiring relatively very little energy and chemicals, generates no secondary pollution, has low capital cost and low operation and maintenance costs, and provides effective degradation of most hazardous emissions, but could relatively be land intensive, particularly the conventional type with shallow filter depths.

Biofilters have been used extensively in Europe for several decades to control odor emissions from industrial facilities and from wastewater treatment plants (Winer et al, 1991). Biofilters are also adaptable to the breakdown of gas emissions from municipal and hazardous materials processing and landfills, as well as from the petrochemical and petroleum refining facilities, and the traffic tunnels. The capability of biofilters have not been fully appreciated to control the air pollution from most industrial activities, while these filters can easily adapt and treat the most dangerous air pollutants, including the highly carcinogenic emissions. However, due to the lack of sales incentives and difficulty in merchandizing, gaseous biofilters have been largely overlooked in the US.

Conventional gaseous biofilters consist of a damp soil/compost mixture media where the influent gases are uniformly distributed throughout the bottom of the filter and the gases are scrubbed as they percolate upward through the filter. Pomeroy patented and experimented with this simple and effective biochemical odor scrubbing technology in the

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US in 1957. However, in spite of the very good odor removal capability, conventional biofilters do not seem to remove many Volatile Organic Compounds (VOCs) consistently and effectively (Schroeder et al, 1992).

With the increasingly stringent control requirements on VOCs and odor emissions from wastewater facilities, this technology was revisited to determine it's potential effectiveness, particularly for the breakdown of highly carcinogenic gas emissions. Two types of biofilter pilot plants, the conventional, and a uniquely modified biofilter, consisting of a submerged zone at the bottom of the media were tested, to evaluate the removal of VOCs and odor compounds from municipal wastewater emissions in LA county.

II. LITERATURE REVIEW

According to Henry's law, at chemical equilibrium, gravity flow sewers could emit close to 300 parts per million by volume (ppmv) of hydrogen sulfide gas, for each milligram per liter (mg/l) of dissolved sulfate in the wastewater, into the sewer headspace. Hydrogen sulfide gas is a very serious lethal threat particularly to the sewer maintenance workers, and is also the precursor for sever concrete corrosion.

The oxidation of hydrogen sulfide to sulfuric acid by sulfide/sulfur oxidizing bacteria (SOB) that causes sewer corrosion is well documented (Parker & Jackson, 1965). The oxidation of sulfide evolves through a series of biotic and abiotic pathways and includes a complex microbial ecosystem (Islander, Devinny et. al. 1991). A microbial succession occurs and numerous species of the genus Thiobacillus dominate the environment as the pH is reduced (ibid.). Finally, Thiobacillus thiooxidans dominate the field that are strict autotrophic-aerobic bacteria, and thrive in the highly acidic environment of pH of about 1.0 (ibid), which corrodes concrete pipe and the reinforcement.

Similarly, specific microorganisms responsible for degradation of hydrocarbon solvent gases have also been identified mostly down to the genus level, and occasionally, down to the specific species. The microflora for degradation of a mixture of aromatic and chlorinated alkenes and alkanes in a biofilter experiment were identified and consisted of Actinomyces globisporus, Micromonospora albus, Micrococcus albus, and species of Penicillium (white), Cephalosporium, Mucor, and Ovularia (Schonborn, 1986). Besides the microorganisms, mites, collembolans, and nematodes were also found (ibid.).

Species of Pseudomonas, Alcaligenes, Methylocystis, and Nitrosomona, were also reported to degrade different VOCs (Young-Sook et al, 1994; Okada et al, 1995). Dehalospirillium multivorans, a strictly anaerobic bacteria dechlorinated PCE to cis-1,2-dichloroethene (c-DCE) using hydrogen gas or formate plus PCE as the sole energy source, and acetate as the carbon source (Neumann et al, 1995).

The metabolism of VOCs is accomplished by two different types of metabolic pathways. The primary pathway by some microorganisms, is when the VOCs are used as the sole source of carbon and energy. Alternatively, the VOCs could be cometabolized, that is the microorganisms use a primary substrate for growth, and induce non-specific enzymes such as monooxygenase or dioxygenase to fortuitously degrade the VOCs (Bielefeldt et al, 1995).

Two important features of cometabolic degradation, namely intermediate toxicity and competitive inhibition have been the subject of many researches. The former results from the toxicity of the intermediate metabolites, which could retard or terminate the viability of the degrading bacteria; while the latter occurs due to competition of the primary substrate, and the VOCs for the key enzymes (Ely et al, 1995; Anderson et al, 1995).

The reactive intermediate metabolites produced within the cell from TCE, consisted of epoxides or acyl chlorides that can covalently bond to enzymes causing long term enzyme inactivation (ibid). Enzyme inactivation and the ability of bacteria to produce new enzymes under adverse conditions, further add to the complexity of cometabolic degradation of the chlorinated solvents (ibid).

Degradation of many VOCs under a wide range of aerobic and anaerobic conditions have also been studied extensively. Trichloroethylene (TCE) was dechlorinated to 1,2-DCE and VC under anaerobic conditions (Major et al, 1995). Vinyl Chloride (VC) was degraded to ethene by a sulfate reducing, methanogenic and acetogenic microbial community that used methanol and acetone as the primary carbon source (ibid).

Perchloroethylene (PCE) is aerobically recalcitrant, however, PCE and many other chlorinated solvents undergo reductive dechlorination under anaerobic conditions (Govind et al, 1995). Dechlorination of PCE was linked to the anaerobic oxidation of toluene and its intermediate products such as benzoate, cresol, propionate and acetate (Sewell et al, 1995). The succession of reductive dechlorinations from PCE, consisting of PCE to TCE to c-DCE to VC to ethene, is often incomplete with the VC to ethene being the rate limiting step (Ballapragada et. al., 1995).

Addition of elemental iron enhanced the dechlorination of Carbon Tetrachloride (CT) and Chloroform (CF) by a methanogenic consortium (Weathers et al, 1995). Under anaerobic conditions elemental metals may serve two functions: (1) as a reducing agent, abiotically coupled to reduction of chlorinated methanes, and (2) as an ultimate electron donor for hydrogenotrophic methanogens (ibid).

Experiments with rotating biological contactors (RBC) and activated sludge (AS) reactors indicated that RCB provided much higher biodegradation rates by maintaining both aerobic and anaerobic conditions in the fixed film microbial community (Galil et al, 1994). Similar experiments indicated that competition for substrate, resulted in stratification of the biofilm and development of dynamic aerobic and anaerobic layers, which enhanced the degradation of substrates that are hard to remove (Zhang et al, 1994). The substrate would diffuse into the anaerobic sublayer, where some of the bonds are broken by the anaerobic and facultative microorganisms, and the metabolite fragments would diffuse back into the aerobic layers, where aerobic degradation occurs (ibid).

III. EXPERIMENTAL METHOD

Fig. 1 shows an schematic arrangement of the pilot plant units. The pilot units consisted of an air blower that forced

sewer gas through perforated pipes at the bottom of 12-inch (30 cm) diameter PVC columns packed with biofilter media. The units included a condensate collector, a flow meter, a throttling valve, a pressure gage, temperature probes, and sampling ports.

The biofilter media comprised of mixtures of composted sewage sludge, and seashells, perlite or lava rocks. The composted sewage sludge provides the main substrate compounds, and the seashells, lava rocks or perlite provide added minerals for microorganism growth. Lava rock in large enough sizes can also provide a stable structure to prevent the compaction of media over time.

Sewer gas was drawn from the headspace at the confluence of three major trunk sewers, considered to have high concentrations of industrial solvents and VOCs. Table 1 shows the composition of the influent wastewater VOC gases that were encountered, and the respective inlet concentration ranges. Influent and effluent gas samples were analyzed for odor intensity, hydrogen sulfide, and twenty-one VOCs.
The concentration of VOCs were measured by JWPC-Plant Air Laboratory, using a Varian 6000 gas chromatograph equipped with sample preconcentrator, photoionization detector, and electrolytic conductivity detector. Based on the recommendation of the Air Lab. supervisor, the inlet VOCs less than 10 ppbv were not included in the calculations.

Odor intensity was measured by a five member odor panel using the Standard Method for Measurement of Odor in Atmosphere, ASTM D-1391-57. The samples were diluted by adding measured volumes of fresh air, and dilution was continued until each member of the panel did not detect any odors. The results are plotted on probability scale, and the 50% threshold dilution is recorded as the odor intensity value.

Hydrogen sulfide gas was measured by "Dositubes" with detector tubes and a hand pump Model No. 488543 manufactured by MSA Corp., where gas concentrations in the parts per million by volume (ppmv) range were read on a graduated scale corresponding to the discolored portion of a chemical powder in the detector tubes. When the Dositube did not register a measurable amount, a Jerome Instrument gas detector Model 621 was used that covers a range of zero to 500 ppbv.

The primary objective in the first phase was principally to determine the odor removal capability of conventional biofilter units that consisted of 3 feet (~ 0.92 m) of a packed biofilter media, and were operated at empty-bed detention times of 20 and 30 seconds, and 1.0 minute. The detention time of 20 seconds did not show much removal, therefore the related data were not included in the removal calculations. The removal efficiencies of the VOC gases were measured throughout. The three conventional biofilter pilot units were operated continuously for about 2.5 to 4 months.

In the second phase of the study, the primary objective was to determine whether the required VOC removals by the SCAQMD regulations could be met by modified biofilter units, that included a submerged media at the bottom. The modified biofilters included up to 1.5 feet (~ 0.46 m) of the filter media at the bottom, submerged in tap water, and the total filter depth was increased to 5 feet (~ 1.52 m). The modified biofilters were operated at different empty-bed detention times between 1.3 to 7 minutes. The two modified biofilter pilot units were operated continuously for about 7 months.

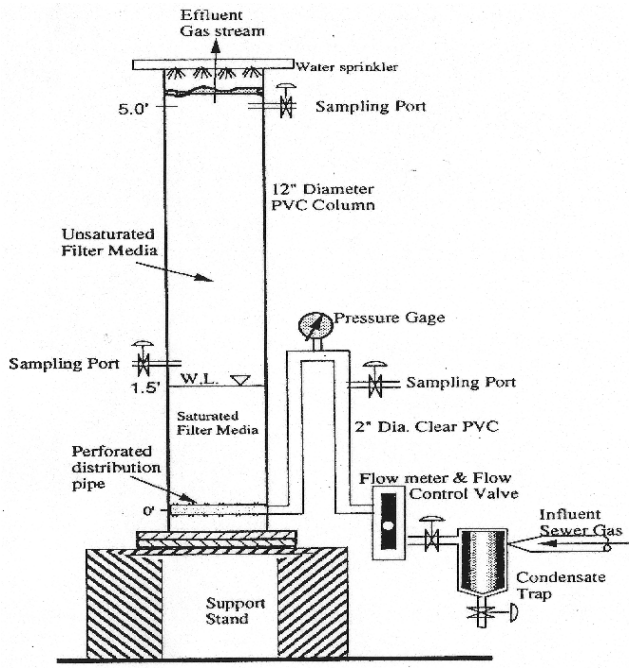

Fig. 1. Schematic presentation of the biofilter of the pilot plants

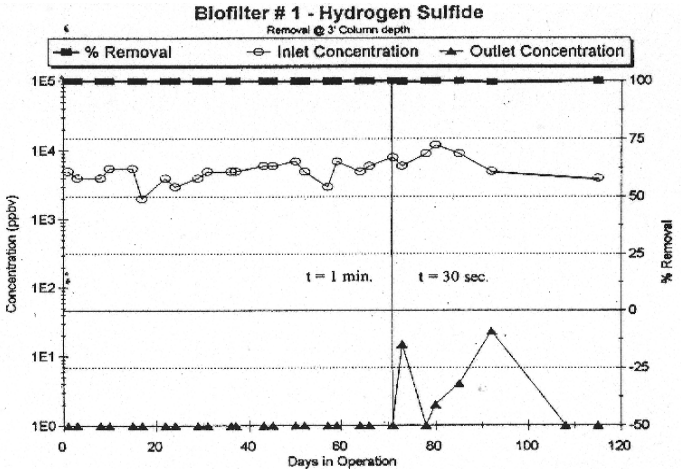

Fig. 2. Hydrogen sulfide removal by the conventional biofilter pilot plant

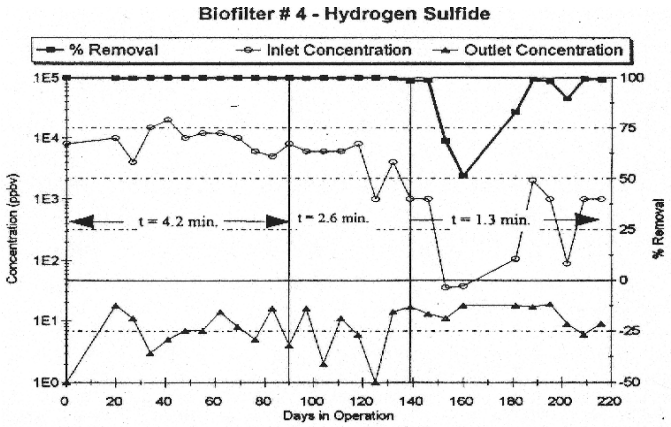

Fig. 3. Hydrogen sulfide removal by the modified biofilter pilot plant

Table 1

	Conventional Biofilter	Modified Biofilter
Measured Chemical Comp.	% Removal	% Removal
Hydrogen sulfide	>95%, Ave. ~99%	50-99%, Ave.~ 95%
Odor Units	40-95%	80-99%
Benzene, Toluene, Xylene, (BTX)	70-95%, Ave.~ 90%	>90%, Ave.~ 95%
Dibromoethane (DBA), Chlorobenzene (CB)	75-95%, Ave. ~ 80%	(No data, Low Conc.)
Carbontetrachloride (CT)	ditto	(No data, Low Conc.)
Dichloroethene (c-DCE), Vinyle chloride (VC)	0-95%, Ave.~ 65%	(No data, Low Conc.)
Dichlorobenzene (DCB)	25-80%, Ave.~ 50%	75-95%, Ave.~ 90%
Dichloroethylene (DCE), Dichloroethane (DCA)	ditto	ditto
Chloroform (CF),	-30% to +20%, Ave. ~0%	20-95%, Ave. ~75%
Perchoroethylene (PCE)	-50% to +25%, Ave. ~0%	30-95%, Ave. ~90%
Trichloroethylene (TCE), Trichloroethane (TCA)	-50% to +60%, Ave.~0%	25-90%, Ave. ~75%
Methylene chloride, (DCM)	-50% to +75%, Ave.~ 0%	ditto

Table 2- Concentration of inlet VOC gases to biofilter pilot units from the municipal sewer pipe

I. Aromatic Hydrocarbons	Abbreviation	Chemical composition	Inlet Conc. Range, ppbv
Benzene*	B	C_6H_6	5 – 1,500
Toluene	TOL	C_6H_5-CH_3	20 – 1,000
Total Xylene	XYL	C_6H_4-$(CH_3)_2$	30 – 1,800
Total Dichlorobenzene	DCB	C_6H_4-Cl_2	0 – 1,500
Chlorobenzene	CB	C_6H_5-Cl	0 – 25
II. Saturated Aliphatic Hydrocarbons			
Carbon Tetrachloride*	CT	C Cl_4	0 – 100
Chloroform*	CF	CH Cl_3	70 – 800
Methylene Chloride*	DCM	CH_2 Cl_2	30 – 4,000
1, 1, 1-Trichloroethane	TCA	CH_3-C Cl_3	20 – 1,000
1, 1-Dichloroethane	DCA	CH_3-CH Cl_2	0 – 80
1, 2- Dibromoethane*	DBA	CH_2 Br-CH_2 Br	3 – 400
III. Unsaturated Aliphatic Hydrocarbons			
Tetrachloroethylene (Perchloroethylene)*	PCE	C Cl_2=C Cl_2	20 – 5,000
Trichloroethylene*	TCE	C Cl_2=CH Cl	3 – 40
1, 1-Dichloroethene	1, 1-DCE	C Cl_2=CH_2	1 – 30
Cis-1, 2, Dichloroethylene	c-DCE	CH Cl=CH Cl	1 – 200
Vinyl Chloride*	VC	CH_2=CH Cl	1 – 50
1, 3-Butadiene*	BU	CH_2=CH CH=CH_2	2 – 50

- Denotes VOCs regulated by Rule 1401 of SCAQMD, Amended in December 1990.

IV. RESULTS

For ease of comparison, even numbered figures show the performance of the conventional biofilter pilot units, and the odd numbered figures, starting with Fig. 3 show the performance of the modified biofilter pilot units. Figs 2 and 3 indicate close to 100% hydrogen sulfide removal consistently by both types of filters without an apparent acclimation period, as the inlet H2S concentrations ranged from about 0.2 to 20 ppmv. It is also noted that the effluent concentration of H2S from the modified biofilter is slightly higher than that from the conventional biofilter.

Following an acclimation period, the conventional filters showed relatively fair removals for the aromatic hydrocarbons; but for many of the halogenated hydrocarbons, poor removals were observed. Benzene (Fig. 4), xylene and toluene showed overall removals of about 90%, and about 80% for some halogenated VOCs, including dibromoethane (DBA), chlorobenzene (CB) and carbon tetrachloride (CT). Dichloroethene (c-DCE) and vinyl chloride (VC) showed an overal removal of about 65%, and about 50% for dichlorobenzene (DCB) (Fig. 6), dichloroethylene (DCE) and dichloroethane (DCA).

No removal was observed for some of the saturated and unsaturated aliphatic hydrocarbons. Methylene chloride(DCM) (Fig. 8), chloroform (CF) (Fig. 10), tetrachloroethylene (PCE) (Fig. 12), trichloroethylene (TCE) (Fig. 14), and trichloroethane (TCA) showed periodic cycles of adsorption and desorption by the conventional biofilters, with no apparent net removal.

The modified biofilters significantly increased the removal efficiency and consistency for the VOCs tested. Benzene (Fig. 5), xylene, toluene were removed by the modified biofilters with an overall average of about 95%. Dichlorobenzene (DCB) (Fig. 7), dichloroethylene (DCE), dichloroethane (DCA) and percholoethylene (PCE) (Fig. 13) were removed by the modified biofilters with an overall average of about 90%. Methylene chloride (DCM)(Fig. 9), chloroform (CF) (Fig. 11), trichloroethane (TCA) and trichloroethylene (TCE) (Fig. 15) were removed by the modified biofilters on an overall average of about 75%.

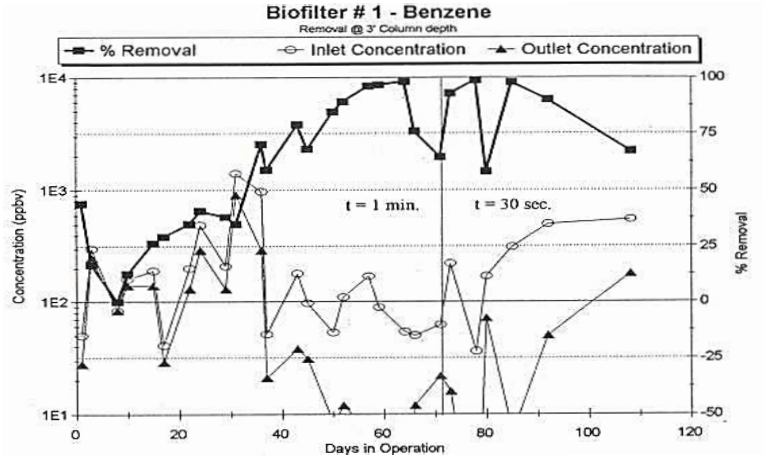

Fig. 4. Benzene removal by the conventional biofilter pilot plant

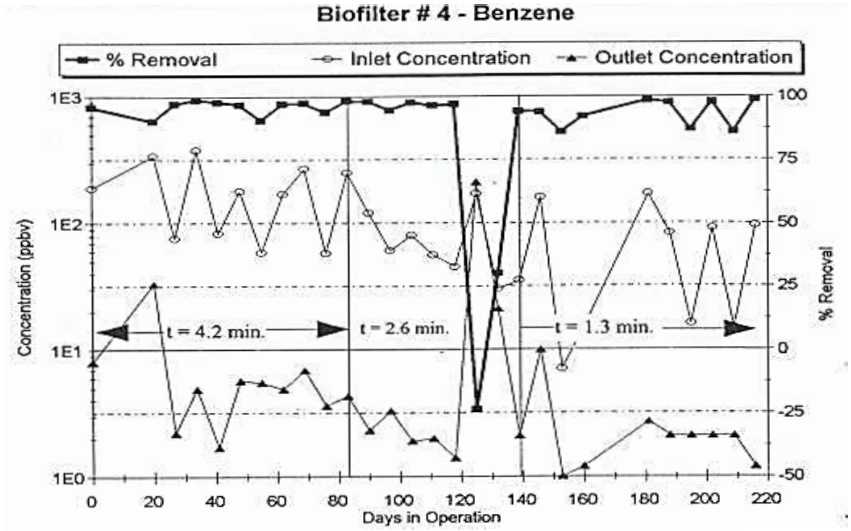

Fig. 5. Benzene removal by the modified biofilter pilot plant

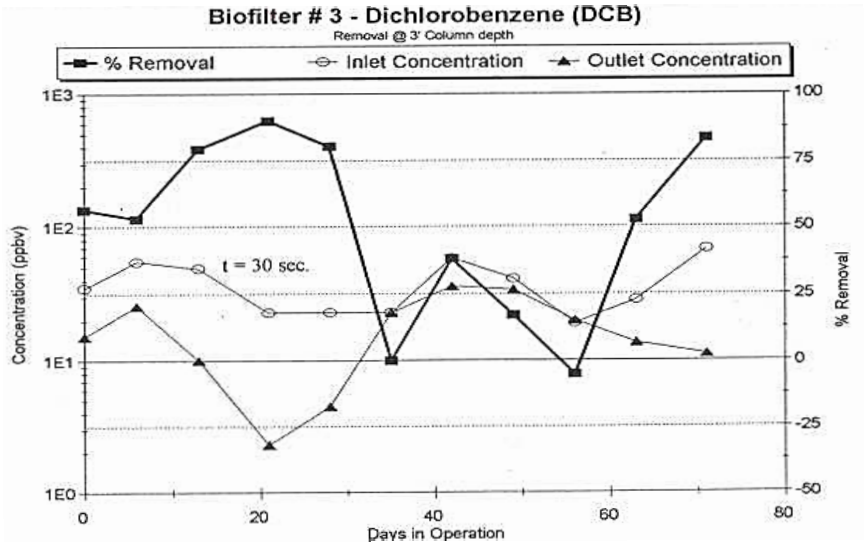

Fig. 6. Dichlorobenzene (DCB) removal by the conventional biofilter pilot plant

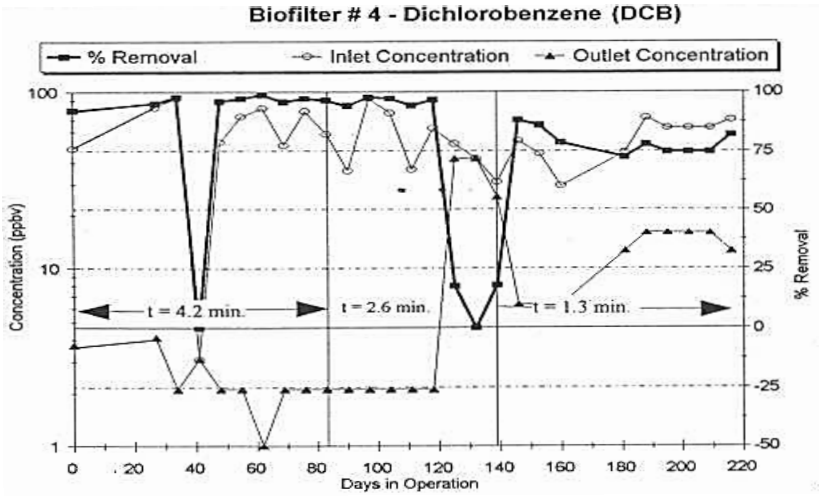

Fig. 7. Dichlorobenzene (DCB) removal by the modified biofilter pilot plant

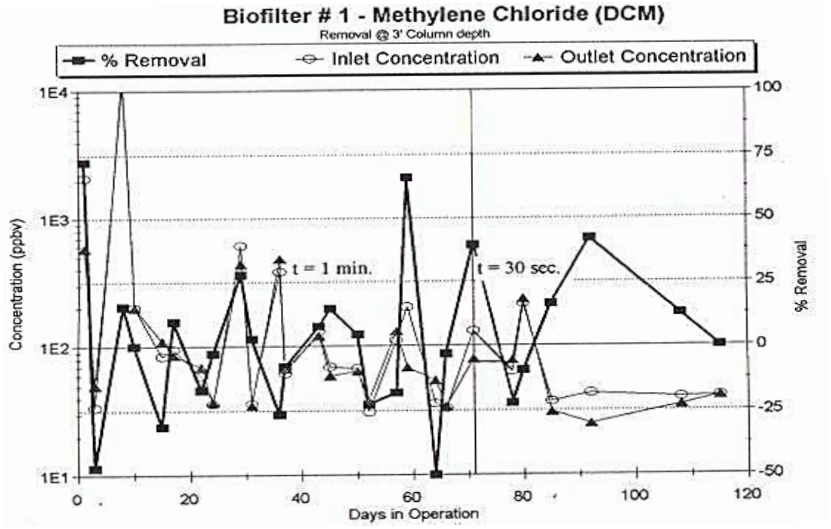

Fig. 8. Methylene Chloride (DCM) removal by the conventional biofilter pilot plant

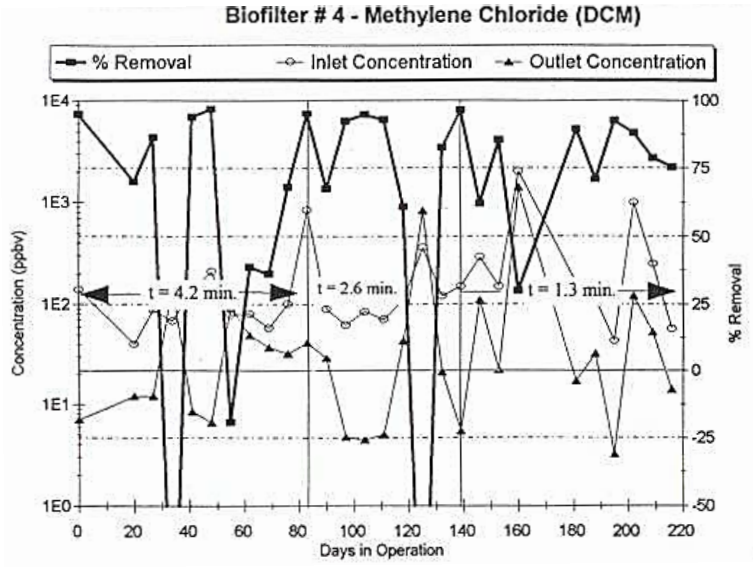

Fig. 9. Methylene Chloride (DCM) removal by the modified biofilter pilot plant

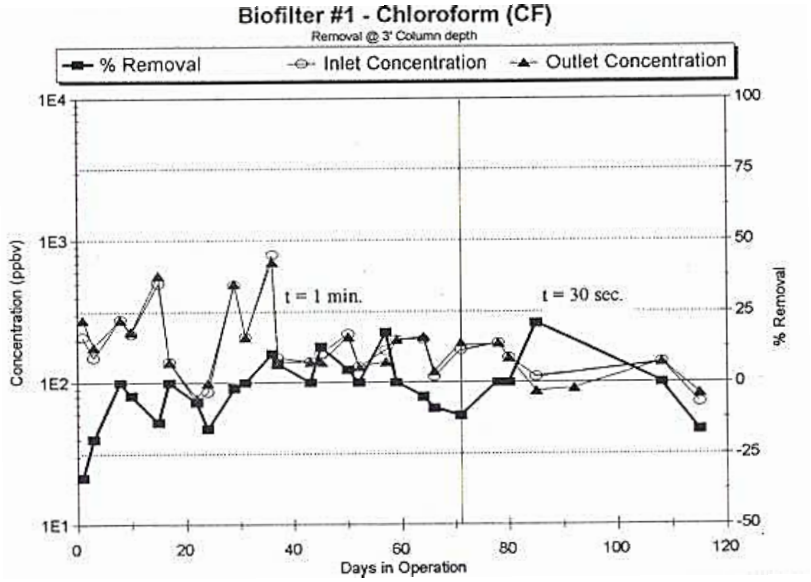

Fig. 10. Chloroform (CF) removal by the conventional biofilter pilot plant

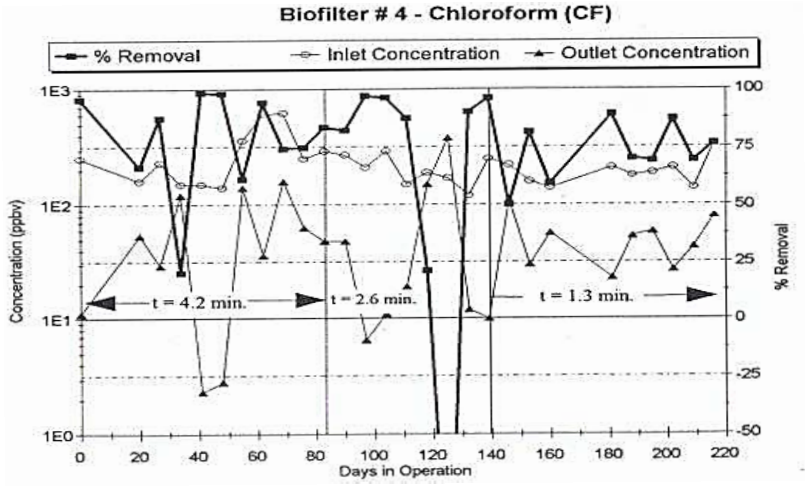

Fig. 11. Chloroform (CF) removal by the modified biofilter pilot plant

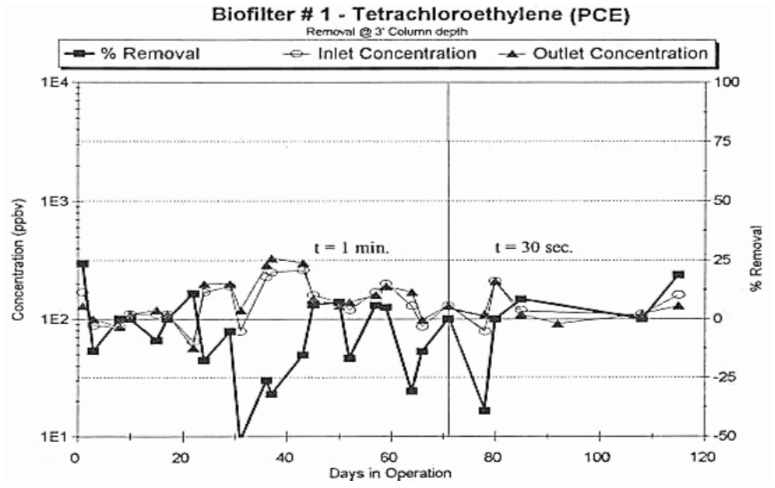

Fig. 12. Tetrachloroethylene (PCE) removal by the conventional biofilter pilot plant

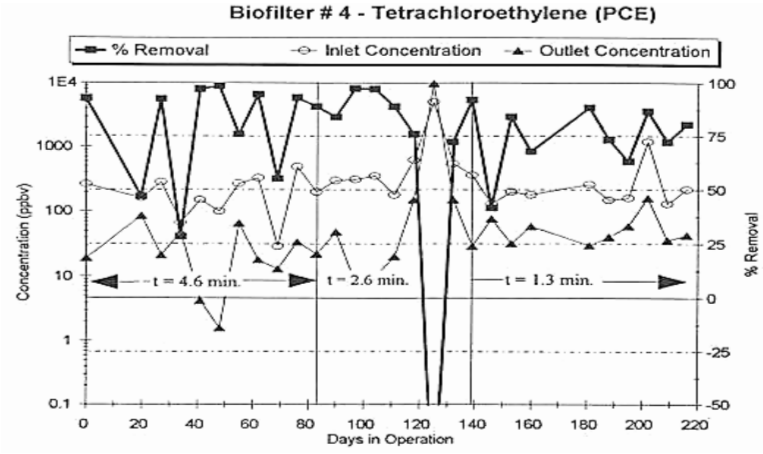

Fig. 13. Tetrachloroethylene (PCE) removal by the modified biofilter pilot plant

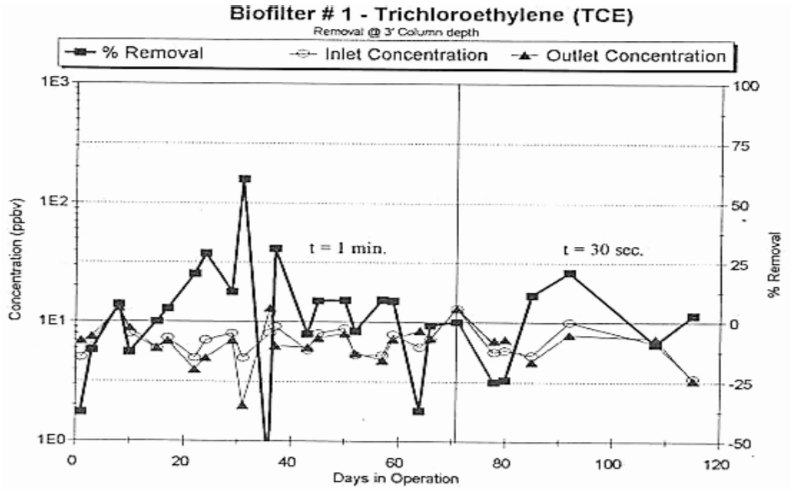

Fig. 14. Trichloroethylene (TCE) removal by the conventional biofilter pilot plant

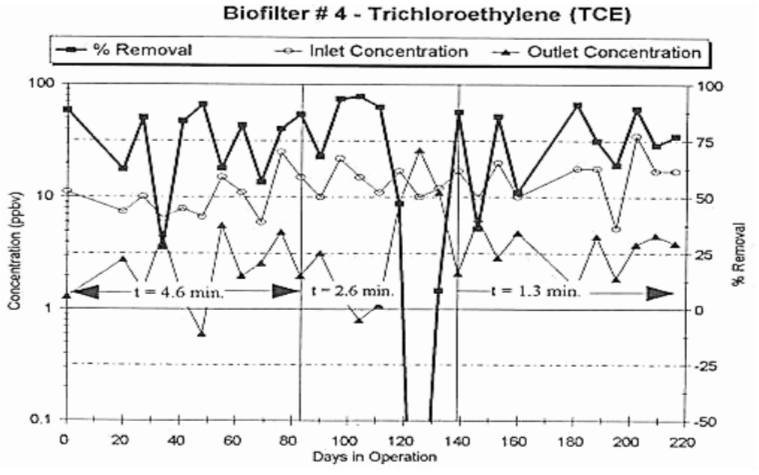

Fig. 15. Trichloroethylene (TCE) removal by the modified biofilter pilot plant

V. DISCUSSION AND CONCLUSION

The immediate effective removal of the hydrogen sulfide and the Odor Units by the conventional biofilter probably indicates the dominance of aerobic metabolism. It is speculated that the conventional gaseous biofilters, develop an aerobic to anoxic biofilm on the filter media that operate in predominantly aerobic to facultative metabolic pathways. Conversely, the relatively higher effluent H2S concentrations from the modified biofilters indicate a more reduced environment as compared to the conventional biofilter.

Some industrial discharges in municipal sewers, including some chlorinated VOC are not readily metabolized

aerobically, and are reductively dechlorinated initially by anaerobic metabolism. Conventional biofilters with the short detention time may be limiting the diffusion of VOC gases through the aerobic-facultative biofilm layers before the VOC could reach an anaerobic zone. The provision of a submerged zone at the bottom of the modified biofilter allows for development of both a fix biofilm as in a trickling filter, and suspended microorganisms as in activated sludge, that seems to operate predominantly in anaerobic to facultative mode. Thus, it is speculated that the submerged zone provides for an expanded growth environment for various types of microorganisms.

Furthermore, the submerged zone allowed for improved dissolution of gases in the aqueous phase, thereby providing more opportunity for the microbial degradation of the substrate. In addition, the submerged layer could potentially dampen the extreme fluctuations in the inlet gas concentrations, thereby maintaining a relatively more uniform substrate concentration for enhanced microbial uptake.

Finally, the increased removal efficiency by the modified biofilters indicates that deeper filter beds could improve the performance and increase the loading rate thereby reducing the required footprint. However, deeper filter beds would require higher capital and energy costs associated with a larger blower, thus offsetting some of the cost savings associated with the reduced land requirement.

Further research is needed to investigate the optimum ratio, or the size of the submerged layer as compared to the total biofilter media depth, the potential performance improvement with biofilters with increased depth to 10 feet (3 m) or more with increased loading rates, or other design & operation parameters. Also, further research is warranted to monitor the conditions by taking multiple samples throughout the depth of the biofilter media, particularly at the junction of the submerged layer with the unsubmerged section, for performance assessment and delineation of the removal stages, and to establish improved design and operation criteria.

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