Mitigation of Trichloroethylene Contaminated Air Streams Through Biofiltration: A Pilot-scale Study

Laura W. Lackey[1*], Johnny R. Gamble[2], and Jeffrey L. Boles[2]

Key words: biofiltration, trichloroethylene, propane-oxidizing consortium, bioremediation

ABSTRACT

As a result of abundant usage and improper disposal practices, trichloroethylene (TCE) is one of the most prevalent groundwater contaminants. Traditional cleanup methods of aquifers contaminated with TCE include pumping the water to the surface and treating with stripper technology, soil vapor extraction, and air sparging. As a result of each of these mitigation schemes, TCE is transferred from the aqueous to the gas phase. As regulations associated with air emission tighten, development of technologies both technically feasible and cost effective for remediating TCE laden gas streams becomes imperative.

This project demonstrated the use of biofiltration technology to mitigate TCE contaminated air streams. A pilot-scale biofilter system was designed, constructed, and subsequently installed at the Anniston Army Depot (ANAD), Anniston, AL. The system was inoculated with a propane-oxidizing microbial consortium that had previously been shown to degrade TCE as well as other short-chained chlorinated aliphatics and a variety of one-and two-ring aromatic compounds.

Critical process variables were identified and their effects on system performance analyzed. Results indicated that the process scheme used to introduce propane into the biofiltration system had a significant impact on the observed TCE removal efficiency. The inlet contaminant concentration as well as the loading rate also had an impact on observed TCE degradation rates. Results suggest that biofilter performance and economics are generally improved by manipulating a specific waste stream so as to increase the TCE concentration and decrease the volumetric flow rate of the contaminated air fed to the biofilter. Through manipulation of process variables, including the empty bed contact time, TCE degradation efficiencies greater than 99.9 percent were sustained. No microbial inhibition was observed at inlet TCE concentrations as high as 87 parts per million on a volume basis (ppmv).

INTRODUCTION

Trichloroethylene (TCE) has been widely utilized as a degreaser within a variety of industries over the past 30 years and as a result of this widespread use is now a prevalent groundwater contaminant. Sutfin et.al., 1996 reported that TCE and associated

[1] Mercer University School of Engineering, Department of Environmental Engineering, 1400 Coleman Ave, Macon, Georgia 31207-0001, lackey_l@mercer.edu, 478-301-4106
[2] Tennessee Valley Authority, Environmental Research Center, Muscle Shoals, AL 35662

chlorinated aliphatics contaminate groundwater and soil at approximately 400,000 sites in the United States. Between 1982 and 1999, the records of decision (RODs) indicate that cleanup technologies of contaminated aquifers identified as Superfund sites has varied. Technologies implemented include the traditional pump and treat scenario to contaminant containment, monitored natural attenuation, and insitu treatments such as air sparging and phytoremediation. During this 17 year period, pump and treat technology was employed at 71% of the sites (EPA-542-R-01-022). Once the contaminated groundwater is pumped to the surface, typical remediation technologies include adsorption and stripping. Stripping technology efficiently transfers the contaminant from the liquid to gas phase and is then discharged to the atmosphere. As new regulations are promulgated, it is anticipated that this type of emissions practice will be discontinued.

In 1985, Wilson and Wilson first demonstrated TCE cometabolic biodegradation in a methane enriched soil column. Since their discovery, others have used a variety of primary substrates including propane (Fliermans et.al., 1988; Wackett et.al., 1989), ammonia (Arciero et.al., 1989), phenol (Hopkins et.al., 1993) and toluene (Nelson et.al., 1987) to induce cometabolic TCE degradation.

Biofiltration technology is typically used to degrade contaminants from gas streams with low contaminant concentrations (e.g., 1-5,000 ppmv). Common applications include the treatment of waste streams generated by soil vapor extraction and air sparging systems and the treatment of industrial emissions points. Biofiltration processes are typically utilized to either convert organic contaminants in gas and air streams to simpler, less objectionable, compounds or to completely mineralize the contaminants to water, carbon dioxide, and salts. During biofiltration, a contaminated gas stream is passed through a packed vessel containing microorganisms. As the organic contaminants pass through the packing, they are sorbed into a biolayer on the packing where they are either consumed by the microorganisms or encounter enzymes which break down the contaminants. The type of packing used can vary and can be either synthetic or natural in origin.

This study utilized a biofiltration system inoculated with a propane-oxidizing consortium to degrade the vapor phase TCE emissions that result from pump and treat technology equipped with a stripper. The propane-oxidizing consortium had the ability to degrade TCE and a variety of other chlorinated aliphatics and 1- and 2-ring aliphatics (Lackey et.al., 1993).

PROCESS DESCRIPTION

The existing pump and treat system consisted of six extraction wells, a collection well, two strippers, a particulate filter system and a GAC filter system. Groundwater from the extraction wells was held in the collection well prior to treatment with air stripping followed by filtration of the aqueous phase. The demonstration biofilter was mounted on an open-bed tractor-trailer, was constructed of 304 stainless steel and contained 18m^3 of packing material. During testing, part or all of the groundwater flow from the collection well was pumped to a demonstration air stripper. The demonstration air stripper could be operated at various air and contaminated water flow rates allowing for more control over process variables (i.e. TCE concentration) that affect biofilter performance. The

contaminated air leaving the demonstration stripper passed through the blower, flowed through the biofilter as was discharged into the atmosphere. Water exiting the demonstration stripper was pumped to one of the existing strippers for further treatment. The demonstration unit was installed adjacent to the existing pump and treat system. Figure 1 shows a process schematic of the existing pump and treat facility as well as the demonstration biofiltration system.

The biofilter packing consisted of a mixture of pine bark, composted poultry litter, and pelletized dolomitic limestone. The pine bark served as a support matrix and a bulking agent to ensure that the air passed through the biofilter without an excessive pressure drop. The composted poultry litter served both as a nutrient source and a residence for the microorganisms. The pelletized dolomitic limestone was added to neutralize any acids that were produced during the course of experimentation. Throughout the demonstration, the biofilter packing was kept moist, by periodically pumping small amounts of water through a weeping type hose arranged over the surface of the packing. The packing was inoculated with a propane-oxidizing culture. After inoculation, the microorganisms were allowed to acclimate by passing an air-propane and TCE mixture through the biofilter for approximately a month. Propane was introduced into the system from a commercial 500-gallon tank. Propane flow was controlled with a mass flow controller equipped with control valve. A lower explosion limit sensor ensured that only non-flammable propane-air mixtures entered the biofilter.

The demonstration system was operated in two cycles: a TCE degradation cycle and a propane feeding cycle. During the TCE degradation cycle, contaminated water was pumped through the demonstration stripper while fresh air was drawn through the demonstration stripper and discharged into the biofilter. A single multifunctional controller (Honeywell UDC 3000) regulated the amount of time the system spent in each cycle, which provided flexibility in operation, such as automatically changing the air flow rate during the TCE degradation and propane feeding cycles.

During the propane feeding cycle, the flow of water to the stripper was discontinued and a stream of propane was fed into the biofilter. Two feeding cycle operating modes were tested during the demonstration: a single pass mode and a recycle mode (patent pending). When testing the single pass mode, a controlled amount of propane was injected into the biofilter influent air stream. The air/propane mixture flowed through the biofilter, where a portion of the propane was consumed, and the mixture was then discharged to the atmosphere via a 3-foot tall stack on top of the biofilter. (Note: The recycle loop shown in Figure 1 had not been installed when the single pass mode was demonstrated.)

When testing the recycle-feeding mode, the biofilter was isolated from the stripper and atmosphere using a series of dampers. Once these dampers were properly positioned, the air within the biofiltration system was continuously recycled between the blower and the biofilter and a controlled amount of propane was injected into the air just down stream of the blower.

On-line, real-time process monitoring was conducted by use of a gas chromatograph (GC), data loggers, a computer, and associated equipment located adjacent to the biofilter in a climate controlled portable trailer. The GC was equipped with both ECD and FID detectors as well as a Valco 10-way valve. Vacuum pumps and automated valves allowed for the inlet and outlet gases of the biofilter to be sampled each hour. Both TCE and propane were quantified during each sampling event.

Please refer to Lackey et.al., 2002 and AEC report SFIM-AEC-ET-CR-99045 for a more detailed description of the system.

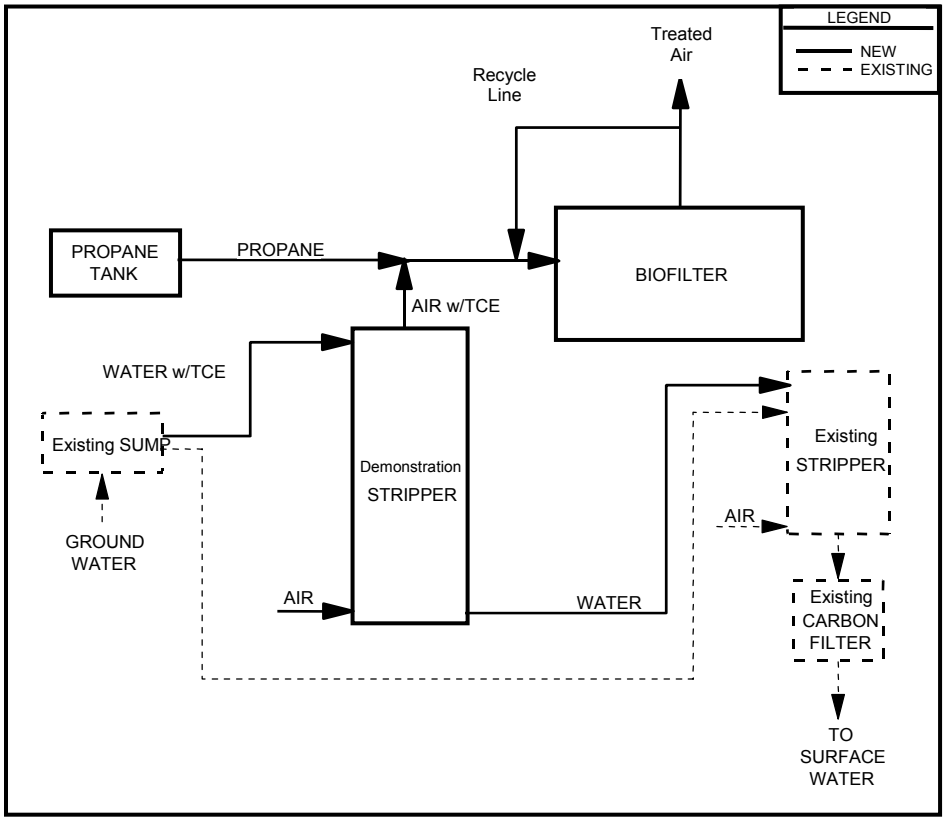

Figure 1. Process flow schematic of existing pump and treat system and the demonstration biofilter/stripper.

RESULTS AND DISCUSSION

The system was operated continuously (except for process changes and infrequent maintenance periods) for a 23-month period. Figure 2 shows the inlet and effluent TCE concentrations when the biofilter system was operated without recycle during propane feeding. During the operational period shown in the figure, propane and TCE were alternately introduced into the biofilter in a step-wise fashion. The EBCT was held constant at 45 min and the TCE and propane cycles were 6 hours each during the 3-day period represented in the figure. The average TCE loading rate was 0.5 g/d/m^3 and 7,791 L of propane were introduced into the system each day. The average degradation efficiency was 32% and the TCE degradation constant was 0.2 g/d/m^3. Note the

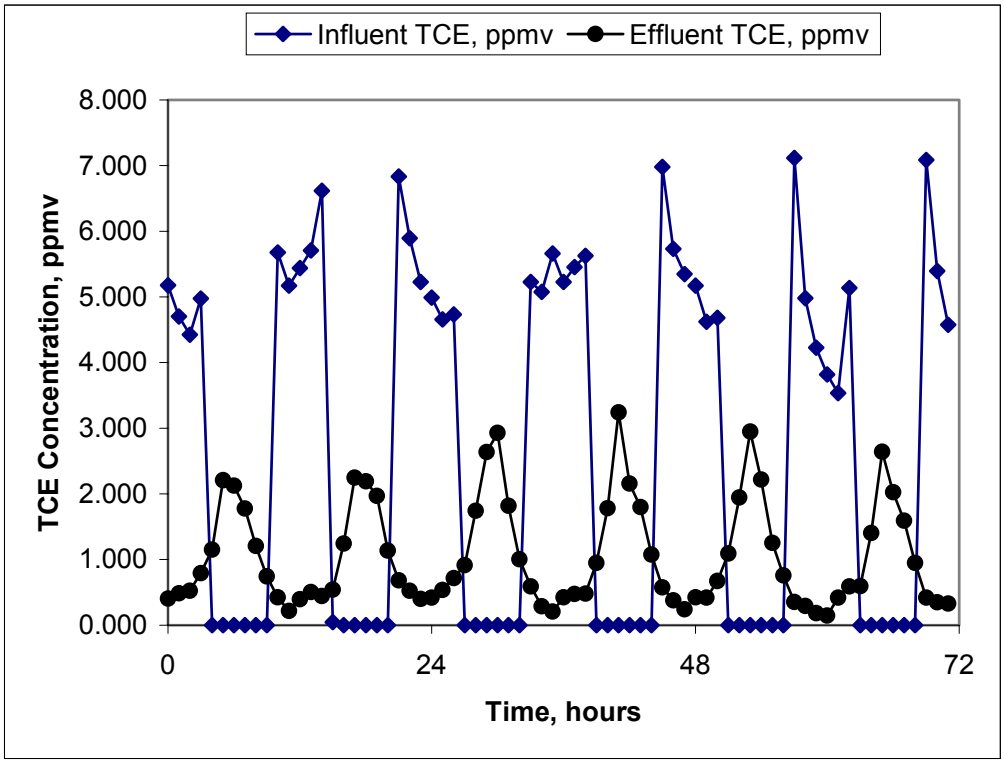

Figure 2. Biofilter Operation without Recycle Feeding at ANAD

relatively low effluent TCE concentrations during the 6-hr TCE cycle as compared to the large increase in TCE concentration in the biofilter effluent stream during the first 3 to 4 hours of the propane feed cycle. These results indicate that a significant portion of the TCE introduced to the system was adsorbed during the TCE degradation cycle and then desorbed during the propane feeding cycle (when the system was not receiving TCE). The apparent TCE degradation efficiency and rate during the TCE degradation cycle appeared to be very high; but when the TCE released during the propane feeding cycle was included in the overall material balance on influent and effluent TCE for the system, the realized TCE degradation efficiency (and rate) was much lower, as shown in Figure 3.

Some increase in TCE degradation was obtained by delaying the start of propane feed and/or by ceasing propane feed early during the feeding cycle. The delayed introduction of propane during the feed cycle (during the delay only ambient air was introduced into the system) decreased the quantity of TCE desorbed during the cycle. With the propane absent from the system during the delay period, the time available for the rapid desorption of TCE was reduced. Furthermore, the delay reduced the quantity of TCE available for desorption. The time delay allowed for TCE degradation to continue without propane available to create a competitive environment between primary and secondary substrate for the oxygenase enzyme. Similarly, by ceasing the introduction of propane early during the feeding cycle, propane was absent from the system at the start of the TCE cycle.

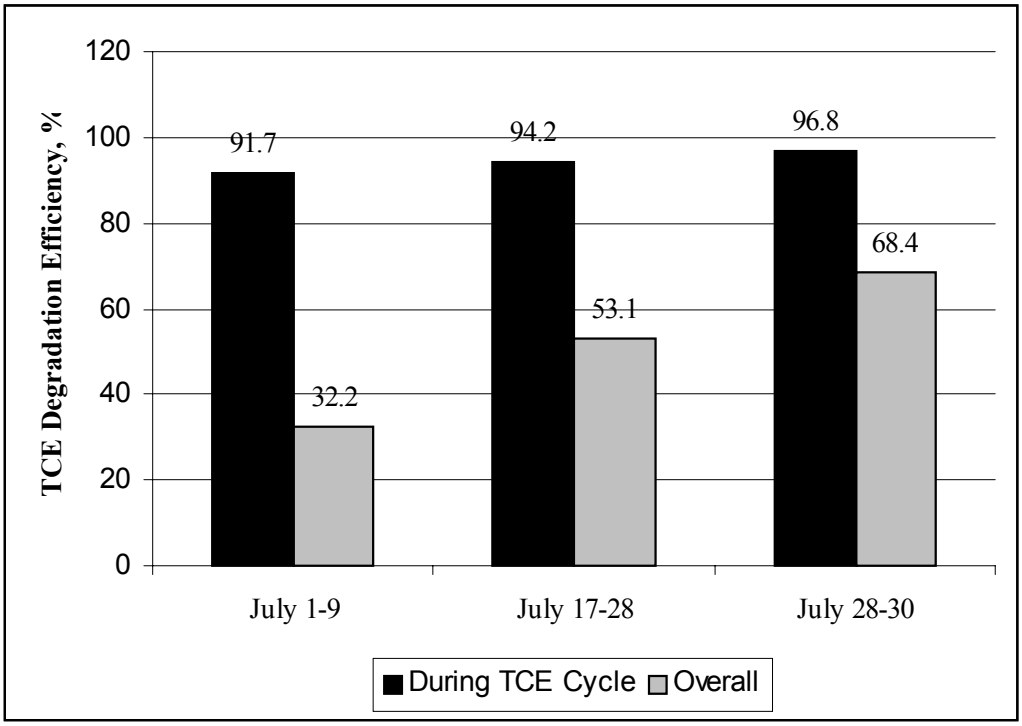

Figure 3. TCE degradation without implementation of the recycle feeding scheme.

As a result, depressed TCE degradation caused by competitive inhibition associated with the presence of primary substrate in the system was negated (Lackey et.al., 2002). However, this improvement was insufficient and still wasted propane. It was recognized that a change in process scheme was necessary to: (1) eliminate TCE emissions during propane feeding and (2) consume all propane fed to the biofilter.

A simple recycle scheme was implemented during the feed cycle that allowed charging the closed biofilter system with propane and then recirculating the in-system process gas until the propane was depleted. During the TCE feed cycle the biofilter was operated as an open system and the process gas was treated in a single pass. The recycle scheme was inexpensive and could be implemented quickly. Figure 4 indicates that the recycle system implemented during each feed cycle dramatically improved biofilter performance.

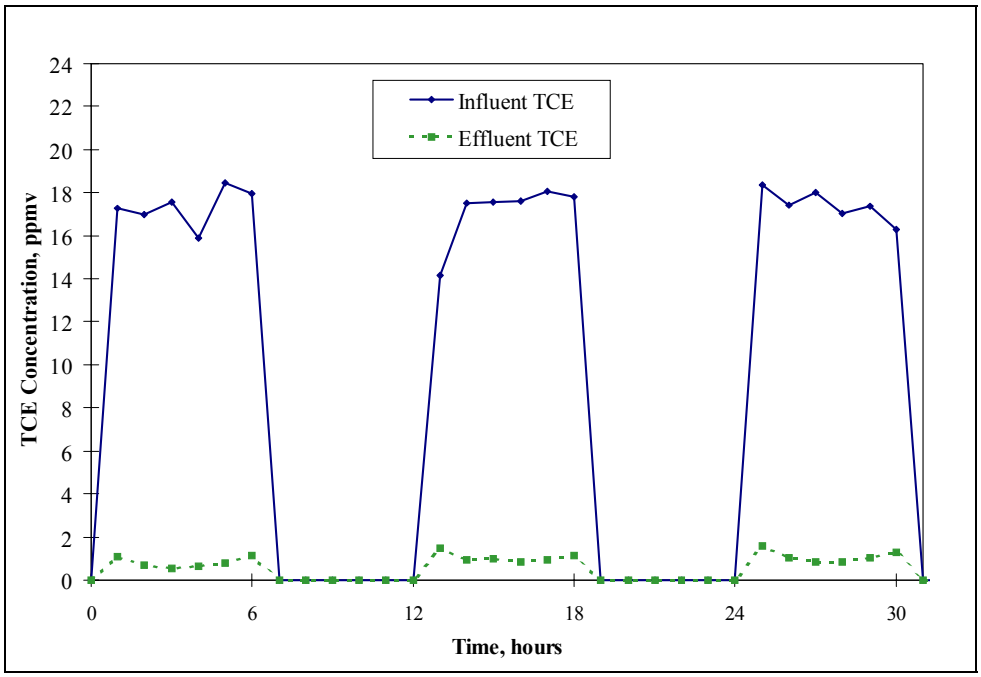

Figure 4. TCE inlet and outlet concentration while operating with recycle scheme implemented. No TCE was released during the feeding cycle and greater than 99.9% of propane was utilized.

During the 30 hours depicted in Figure 4, the EBCT was held constant at 43 min and the TCE and propane cycles were 6 hours each. The average TCE loading rate and inlet concentration was 1.5 g/d/m^3 and 17 ppmv, respectively and 840 L/day of propane were introduced into the system. The average degradation efficiency was 90% and the TCE degradation rate constant was 1.4 g/d/m^3. Notice that no TCE was released from the system during the propane feed cycle and all propane was consumed prior to the start of each TCE cycle. Also important is the reduction in propane required while operating in recycle mode compared to the single pass operational scheme. Assuming the cost of liquid propane to be \$0.33/L, the cost of primary substrate to degrade a unit of TCE decreased from 2.9 to 0.04 \$/g TCE degraded when the operational mode in the propane feed cycle was changed from single pass to recycle. Table 1 highlights the key process variables and results associated with the biofilter system operations depicted in Figure 2 and 4.

Table 1. Summary of process variables and results from both no recycle and recycle operation during the time periods plotted in Figures 2 and 4, respectively.

Mode	**Process Variables**			**Influent TCE**		**C_3H_8**	**Degradation**		**Effluent TCE**		**Propane Cost**	
	EBCT, min	TCE cycle time, min	C_3H_8 cycle time, min	Concen-tration, ppmv	Load, g/d/m^3	Influent rate, L/day	Effi-ciency, %	Rate, g/d/m^3	Avg ppmv	Max ppmv	\$ per day	\$ per g TCE
No Recycle	45	6	6	5	0.5	7,791	32	0.2	0.9	1.8	7.43	1.95
Recycle	43	6	6	17	1.5	840	90	1.4	1.7	3.1	0.80	0.04

Project results can be summarized as follows:

- Gas phase TCE can be degraded in a biofiltration system.
- TCE was adsorbed to the organic biofilter packing material and was then desorbed as propane was introduced into the system.
- The addition of the recycle scheme during the propane feed cycle increased the average TCE degradation efficiency.
- With the addition of recycle during the propane feed cycle, all propane introduced was degraded.
- The addition of the recycle scheme during the propane feed cycle improved system economics by decreasing the primary substrate cost associated with TCE degradation.

REFERENCES

Arciero, D.; Vannelli, T; Logan, M.; Hooper, A.B. Degradation of Trichloroethylene by the Ammonia-Oxidizing Bacterium *Nitrosomonas europaea*; *Biochem. Biophys. Res. Commun*. 1989, 159, 640-643.

Demonstration Results of the Biofiltration of a TCE-Contaminated Air Stream at the Anniston Army Depot, Anniston, Alabama. SFIM-AEC-ET-CR-99045, September 1999.

Fliermans, C. B.; Phelps, T.J.; Ringelberg, D.; Mikell, A.T.; White, D.C. Mineralization of Trichloroethylene by Heterotrophic Enrichment Cultures; *Appl. Environ. Microbiol*. 1988, 54, 1709-1714.

Groundwater Remedies Selected at Superfund Sites. EPA-542-R-01-022. January 2002.

Hopkins, G. D.; Munakata, J.; Semprini, L.; McCarty, P.L. Trichloroethylene Concentration Effects on Pilot Field-Scale In-Situ Groundwater Bioremediation by Phenol-Oxidizing Microorganism; *Environ. Sci. Technol.* 1993, 27, 2542-2547.

Lackey, L.W.; Phelps, T.J.; Bienkowski, P.R.; White, D.C. Biodegradation of Chlorinated Aliphatic Hydrocarbon Mixtures in a Single-Pass Packed-Bed Reactor; *Applied Biochemistry and Biotechnology*, 1993, 89/40, 701-713.

Lackey, L. W., Gamble, J.R., and Boles, J.L. Bench-Scale Evaluation of a Biofiltration System Used to Mitigate TCE Contaminated Air Streams, *Advances in Environmental Research*, In Print, 2002.

Nelson, M. J. K.; Montgomery, S.O.; Mahaffey, W.R.; Pritchard, P.H. Biodegradation of Trichloroethylene and Involvement of an Aromatic Biodegradative Pathway; *Appl. Environ. Microbiol*. 1987, 53, 949-954.

Sutfin, Julie A. How Methane Injection Attacks Chlorinated Solvents. 1996. *International Ground Water*, 2(4).

Wilson, J. T.; Wilson, B.H. Biotransformation of Trichloroethylene in Soil; *Appl. Env. Microbiol*. 1985, 49, 242-243.

Wackett, L. P.; Brusseau, G.A.; Householder, S.R.; Hanson, R.S. Survey of Microbial Oxygenases: Trichloroethylene Degradation by Propane-Oxidizing Bacteria; *Appl. Environ. Microbiol.* 1989, 55, 2960-2964.