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BIOTREATMENT OF TCE-CONTAMINATED GROUNDWATER

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ABSTRACT

A trickle-bed bioreactor containing a consortium of microorganisms using methane as the primary carbon source was used to treat a synthetic groundwater containing trichloroethylene (TCE) and trans-1,2-dichloroethylene (DCE). With influent concentrations of TCE and DCE of 1 mg/L each and an average residence time of about 50 min, >50% of the TCE and >90% of the DCE was degraded. The reactor exhibited first-order kinetics with respect to TCE degradation.

INTRODUCTION

There have been several reports of the ability of methane using bacteria to co-metabolize short-chain chlorinated hydrocarbons such as TCE and DCE (1-5). Little, et al. (3) recently reported the mineralization of TCE by a pure culture of a methane-oxidizing organisms isolated from TCE-contaminated groundwater. The purpose of this study was to establish a methanotrophic bioreactor system and determine the process potential of such a system for remediating groundwater contaminated with TCE, DCE, and vinyl chloride.

MATERIALS AND METHODS

The bioreactor consisted of a 5 cm I. D. x 110-cm long glass column packed with 0.6-cm ceramic berl saddles. The microbial consortium was obtained from C. D. Little (Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee). A concentrated feed solution containing mineral salts (3) and TCE, DCE, or both was continuously bled into a stream of process water, and the mixture was distributed (generally at 10 mL/min) over the top of the packing. The influent concentrations of TCE and DCE were controlled by adjusting their concentrations in the feed concentrate and by varying the dilution with process water. A gas stream (25 mL/min) containing methane and air was also introduced at the top of the reactor.

Capillary gas chromatography using an electron capture detector was used to monitor TCE and DCE.

RESULTS AND DISCUSSION

As shown in Table 1, DCE was degraded more rapidly and to a much greater extent than TCE. During initial experiments with the bioreactor, there were indications that the rate of TCE degradation was depressed by DCE. However, at a later time, experiments to verify this effect were inconclusive.

Table 1. Degradation of TCE and DCE in a trickle bioreactor

	TCE			DCE		
Flow rate (L/min)	Influent (mg/L)	Effluent (mg/L)	Degradation[1] Rate (mg/d)	Influent (mg/L)	Effluent (mg/L)	Degradation[1] Rate (mg/d)
0.005	0.9	0.2	4.7	1.1	N.D.[2]	≥ 7.8
0.010	1.0	0.3	7.3	1.2	N.D.[2]	≥ 17.1
0.020	1.0	0.5	12.0	1.2	0.03	33.4
0.035	1.1	0.8	13.5	0.8	0.03	39.6
0.050	1.0	0.6	28.7	1.0	0.02	68.8

[1]Corrected for small losses of TCE and DCE in the off-gas (data not shown).

[2]Not detected. The detection limit was ~0.01 mg/L.

Although the reactor operated well at gas phase CH_4 concentrations up to 20% (v/v), 4% CH_4 was sufficient to maintain the microbial population and reactor activity. When the CH_4 concentration was decreased to 2%, activity was maintained for 4 to 5 days but then began to decrease.

Under the usual operating conditions (1 mg/L TCE, 10 mL/min), approximately 50% of the TCE was removed during a single pass through the bioreactor. To determine if the TCE concentration could be reduced

further, recycle of the reactor effluent was used to extend the residence time. The normal feed flow was stopped, and a fresh 1 mg/L solution of TCE was recycled through the reactor. After about 15 min, the TCE concentration was approximately 0.2 mg/L. However, after several hours the TCE concentration remained at 50 to 100 μ/L. This apparent lower limit also prevailed when methane was removed either from the onset of recycle or after the bulk of TCE had been degraded. The presence of DCE was shown not to be a factor. We have no explanation for this phenomenon.

Verification that TCE disappearance was, in fact, due to microbial action was threefold. Greater than 90% of influent TCE was accounted for in the effluent liquid and off-gas using both a blank column (without packing) and when the biological activity was virtually eliminated by shutting off the gas flow. Thirdly, shake flask experiments with the mixed culture from the bioreactor using ^{14}C-labeled TCE showed that in excess of 60% of the TCE was mineralized to CO_2. The rest of the label appeared in the cell mass (~25%) or in water-soluble products (5 to 10%).

No priority pollutants other than TCE and DCE could be detected in the liquid effluent from the bioreactor. In particular, no vinyl chloride could be detected. In a separate batch experiment, vinyl chloride was shown to be removed from an actual groundwater sample containing TCE, DCE, and vinyl chloride.

However, one other peak was prevalent during the course of bioreactor operation with DCE. The compound was also observed in batch-type DCE degradation experiments. It did decrease with time during recycle and batch experiments. Mass spectrometric analysis revealed that the compound had a mass of 112 or greater and likely contained two carbon atoms, two chlorine atoms, and possibly an oxygen atom. Although the compound was not identified, its characteristics are consistent with the DCE-epoxide that Janssen et al. (4) recently reported being produced by methanotrophs exposed to DCE. On occasion, particulary during recycle, other compounds appeared (as evidenced by chromatographic peaks) whose elution times were between TCE and DCE. These peaks also appeared to arise as a result of DCE metabolism. They were not noted when only TCE was fed into the system. During recycle, these peaks would gradually disappear.

There was a small amount (~10%) of stripping of TCE and DCE by the methane/air gas stream. In practice, this could necessitate some sort of treatment to remove these pollutants from the off-gas. Another possibility would be to recycle the reactor off-gas. Alternatively, we demonstrated that TCE/DCE degradation could be accomplished by presaturating the influent feed stream with methane and oxygen.

Average residence times (obtained by conductivity measurements combined with salt pulses) of 47 min, 17 min, and 10 min were determined at 11 mL/min, 32 mL/min, and 66 mL/min, respectively. Although these residence times were larger than expected, they are reasonable in view of

the local flooding that was seen in the reactor. The liquid holdup is, thus, about 500 to 650 mL, depending on the flow rate. The broad (in time) conductivity response obtained indicated a large degree of backmixing or relatively inert or inactive regions in the bioreactor. This suggests that the reactor may well be equally active with considerably less biomass in the column.

Reactor performance in terms of TCE and DCE degradation was measured at liquid flow rates of 5, 10, 35, and 50 mL/min (see Table 1). The average residence time at each flow rate was estimated from the residence time distribution studies. Reactor performance was consistent with first-order kinetics; that is, a first-order rate constant of 0.017 to 0.023 min^{-1} was found throughout this range of flow rates, except for a single high value of 0.048 min^{-1} at 50 mL/min. Although the TCE concentration in the effluent rose with increasing flow rate as expected, the total TCE degradation increased, presumably due to the higher average concentration of TCE in the reactor.

Future work will be directed toward improving reactor performance using alternative packings to eliminate local flooding. Also, we will attempt to gain an understanding of the apparent lower concentration limit of TCE achievable during recycle.

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