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In Situ Bioremediation Via Horizontal Wells

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Abstract

The U.S. Department of Energy, Office of Technology Development, has been sponsoring full-scale environmental restoration technology demonstrations for the past three years. The Savannah River Site Integrated Demonstration focuses on the clean-up of soils and groundwater contaminated with chlorinated volatile organic compounds (VOCs). To optimize resources, the project is simultaneously evaluating and testing a large number of drilling, monitoring, characterization, and remediation technologies developed by SRS, other DOE sites, national labs, industry and universities. During fiscal year 1992 alone, more than 44 different technologies were tested at the site. The principal remediation technology being tested during 1992 was **in situ** bioremediation. **In situ** air stripping was the first remediation technology demonstrated at the test site during 1990 using parallel horizontal wells (one below the water table and one above). This first very successful demonstration provided the impetus and the characterization and monitoring data to serve as an excellent control for the **in situ** biostimulation demonstration. Several laboratories including our own had demonstrated the ability of methanotrophic bacteria to completely degrade or mineralize chlorinated solvents, and these bacteria were naturally found in soil and aquifer material. Thus the test consisted of injection of methane mixed with air into the contaminated aquifer via a horizontal well and extraction from the vadose zone via a parallel horizontal well. This configuration has the advantage of simultaneously stimulating methanotrophic activity in both the groundwater and vadose zone, and inhibiting spread of the plume. Groundwater was monitored biweekly from 13 wells for a variety of chemical and microbiological parameters. Groundwater from wells in affected areas showed increases in methanotrophs of more than one order of magnitude every two weeks for several weeks after 1% methane in air injection was started. Simultaneous with the increase in methanotrophs was a decrease in water and soil gas concentrations of trichloroethylene (TCE) and tetrachloroethylene (PCE). Two wells declined in TCE/PCE concentration in the water by more than 90% to below 2 ppb. All of the wells in the affected zone showed significant decreases in contaminants in less than 1 month. Four of five vadose zone piezometers (each with three sampling depths) declined from concentrations as high as 10,000 ppm (vol/vol) to less than 5 ppm in less than six weeks. The fifth cluster also declined by more than 95%. A variety of other microbial parameters increased with methane injection indicating the extent and type of stimulation that had occurred.

Introduction

This project is designed to demonstrate **in situ** bioremediation of groundwater and sediment contaminated with chlorinated solvents. Indigenous microorganisms were stimulated to degrade TCE, PCE and their daughter products **in situ** by addition of nutrients to the contaminated zone. **In situ** biodegradation is a highly attractive technology for remediation because contaminants are destroyed, not simply moved to another location or immobilized, thus decreasing costs, risks, and time, while increasing efficiency and public and regulatory acceptability. Bioremediation has been found to be among the least costly technologies in applications where it will work (Radian 1989).

Subsurface soils and water adjacent to an abandoned process sewer line at the SRS have been found to have elevated levels of TCE (Marine and Bledsoe 1984). This area of subsurface and groundwater contamination is the focus of a current integrated demonstration of new remediation technologies utilizing horizontal wells. Bioremediation has the potential to enhance the performance of **in situ** air stripping as well as offering stand-alone remediation of this and other contaminated sites (Looney et al. 1991). Horizontal wells could also be used to enhance the recovery of groundwater contaminants for bioreactor conversions from deep or inaccessible areas (e.g., under buildings) and to enhance the distribution of nutrient or microbe additions in an **in situ** bioremediation.

Materials and Methods

The principal nutrient to be supplied via the horizontal wells in this test was methane, at a low concentration in air (<4%). The lower horizontal well provides a very efficient delivery of gas throughout the contaminated region (Figure 1). A vacuum was applied to the upper well (vadose zone) to encourage air/methane movement through the upper saturated zone and lower vadose zone and inhibit spreading of the plume. Air/methane mixtures have been demonstrated to stimulate selected members of the indigenous microbial community that have the capability to degrade TCE (Wackett et al. 1989). An extensive characterization and monitoring program using existing monitoring wells and periodic borings for sediment was used to measure the response of the soil and water following injection of air/methane (Eddy et al. 1991). In addition, offgas from the upper horizontal well was assayed for methane, total VOC, TCE, PCE, and potential break down products of TCE/PCE (e.g., DCE, VC, and carbon dioxide). For a complete listing of all analytical assays, protocols, permits, collaborators, expert panel, etc. see Hazen (1991).

Initially 1% methane/air was injected continuously into the lower well; however, to ensure process optimization (i.e., to further stimulate the indigenous microorganisms to peak biodegradation rates and efficiencies), the injection protocol was altered for subsequent campaigns. At three-month intervals during the 14-month demonstration, the data from the current operating campaign and process support activities were examined by an expert panel and a decision was made as to how to alter the injection protocol for the subsequent campaign. Thus, the final test consisted of the following operating campaigns:

1. air extraction alone from the upper well at 240 scfm (2/26/92–3/18/92)
2. air only injection was added at 200 scfm (3/18/92–4/20/92)
3. injection with 1% methane/air (4/20/92–8/5/92)
4. injection with 4% methane/air (8/5/92–10/23/92)
5. pulsing 4% methane/air (10/23/92–1/25/93)
6. pulsing 4% methane and continuous injection of nitrous oxide at 0.07% in air and tri-ethyl phosphate at 0.007% in air (1/25/93–4/30/93)

Results and Discussion

The flow and vacuum conditions of the extraction system have remained constant with a flow rate of 240 scfm and 7.6 in. Hg. VOCs in the offgas were composed entirely of TCE and PCE. Dissolved oxygen content in the water did not changed during this period. Overall VOC concentrations increased slightly during the first 5 days and then steadily declined. During the previous extraction demonstration with this same well the VOC concentration started 10 times higher and declined rapidly over the next 5 days. Since the previous test extraction rate was double the current rate, the current stabilized VOC concentration is about what would be expected at the end of the previous demonstration, taking into consideration the lower flow rate. Because the previous demonstration finished nearly 15 months ago, we believe this result indicates that the effect of this type of extraction is long term and that a permanent reduction has occurred in the amount VOCs in the vadose zone at the site. Comparison of VOCs in pretest and post-test borings support this observation since sediment concentrations decreased by more than 30%. Interim borings at four holes done at the end of the 1% methane injection also reveal a further 50% decline in the amount of VOCs in the sediment. Indeed, few of these samples had detectable levels remaining.

Air injection (200 scfm) seemed to have little effect on the extraction efficiency. One percent and 4% methane injection had little effect on extraction efficiency or offgas quality though overall there was a small but significant decline in VOC concentration over time for both operating campaigns. In addition, the ratio of TCE/PCE significantly and consistently declined over time. This observation is consistent with our knowledge that methanotrophs will degrade TCE but not PCE and that PCE is degraded at a slower rate by syntrophic anaerobes. However, pulsing of methane injection has caused a significant decrease in VOC concentration in the extraction well. When the methane was injected again for five days after air-alone injection, the VOC concentration increased but declined again as soon as this pulse was stopped. These observations coincide with our understanding of competitive inhibition (i.e., when the methane is withdrawn once high biomass is achieved, more contaminants are degraded since there is more available enzyme active sites). In addition, it appears that the long interval pulsing decreased methanotroph density during the first six weeks of the pulsing campaign; during the subsequent six weeks, the short-interval pulsing increased methanotroph densities. Carbon dioxide concentrations from the extraction well suggest an upward trend beginning 2–3 weeks post air-injection startup; this may be indicative of increased microbial respiration in the subsurface caused by the air injection. There is also a striking positive correlation between VOC concentration in vadose zone soil gas and CO_2 concentrations. After VOCs disappeared, the CO_2 concentration subsequently declined. When new VOCs move into the area, the CO_2 concentra-

tions subsequently increase until after the VOCs have declined again. Since pulsing began vadose zone concentrations declined significantly and then increased in some wells. Since nitrogen and phosphorus (N&P) injection began, the concentration of VOC in all vadose zone wells has declined dramatically, more than 90%. This again supports the theory of competitive inhibition and nutrient limitations discussed above. More than 108,206,345 scf of air were injected during this test. As expected, even though more than 1,392,774 scf of methane were injected into the subsurface during 53 weeks, only trace quantities of methane were detected in the extraction wells or any of the vadose zone piezometers during the 1% methane injection campaign (i.e., most if not all of the methane injected was consumed by the subsurface TCE-degrading microflora). Simultaneous injection of helium as a conservative tracer has shown that more than 50% of the injected methane is being consumed.

Monitoring of the groundwater has shown that methanotrophs increased at the rate of one order of magnitude every two weeks since methane injection (1%) began (Figure 2). However, increases substantially slowed and began declining slightly. This change coincides with reduction in nitrates in the water of these same wells. Several other measures of microbial activity and abundance have also increased dramatically concomitantly with the start of methane injection and have shown a similar response to nitrates. After 4% methane injection was started (8/5/92), methanotroph densities continued to increase. The wells showing the greatest decrease in TCE/PCE concentrations have experienced as much as a five order-of-magnitude increase in methanotrophs. These same wells have also shown increased concentrations of chloride in the water, an aerobic biodegradation end product for TCE. Stimulation of biodegradation activity by the indigenous microflora appears to have been great during the initial phase of the 1% methane injection. After two months of the 4% methane/air campaign, it appeared that the methanotroph population was further stimulated but that nitrogen-fixing bacteria may have been inhibited causing severe nitrogen limitations. However, the outer wells started showing significant densities of methanotrophs and for the first time the concentrations of TCE/PCE either remained the same or declined slightly. Prior to this they had been slowly increasing. The 4% methane injection may have been inhibitory to nitrogen-transforming bacteria; therefore, we began the pulsing campaign, which initially consisted of air injection alone for 5–14 days, followed by injection of 1% methane for 4–5 days. It was believed this would reduce competitive inhibition of the methane and TCE for the same enzyme and reduce the inhibition of nitrogen fixers shown to be stimulated by air injection alone. Pulsing caused a significant increase in nitrogen-transforming bacteria, a decrease in TCE in the well water and vadose zone, and a decrease in methanotroph densities. On December 11, 1992, we started a short pulse interval of 8 h of 4% methane every other day. The final campaign (1/25/93) included pulsed injection of methane and continuous injection of nitrous oxide at 0.07% in air and tri-ethyl phosphate at 0.007% in air. This decision was based on enrichment and mineralization studies. It was felt that this last injection would overcome both N&P limitations and allow higher biomass and higher degradation rates of TCE to be achieved by the methane-stimulated subsurface bacteria. Since injection of N&P started, and with only limited analyses complete, we can report that densities in the water have gone up, and TCE concentration in the vadose zone and water has declined.

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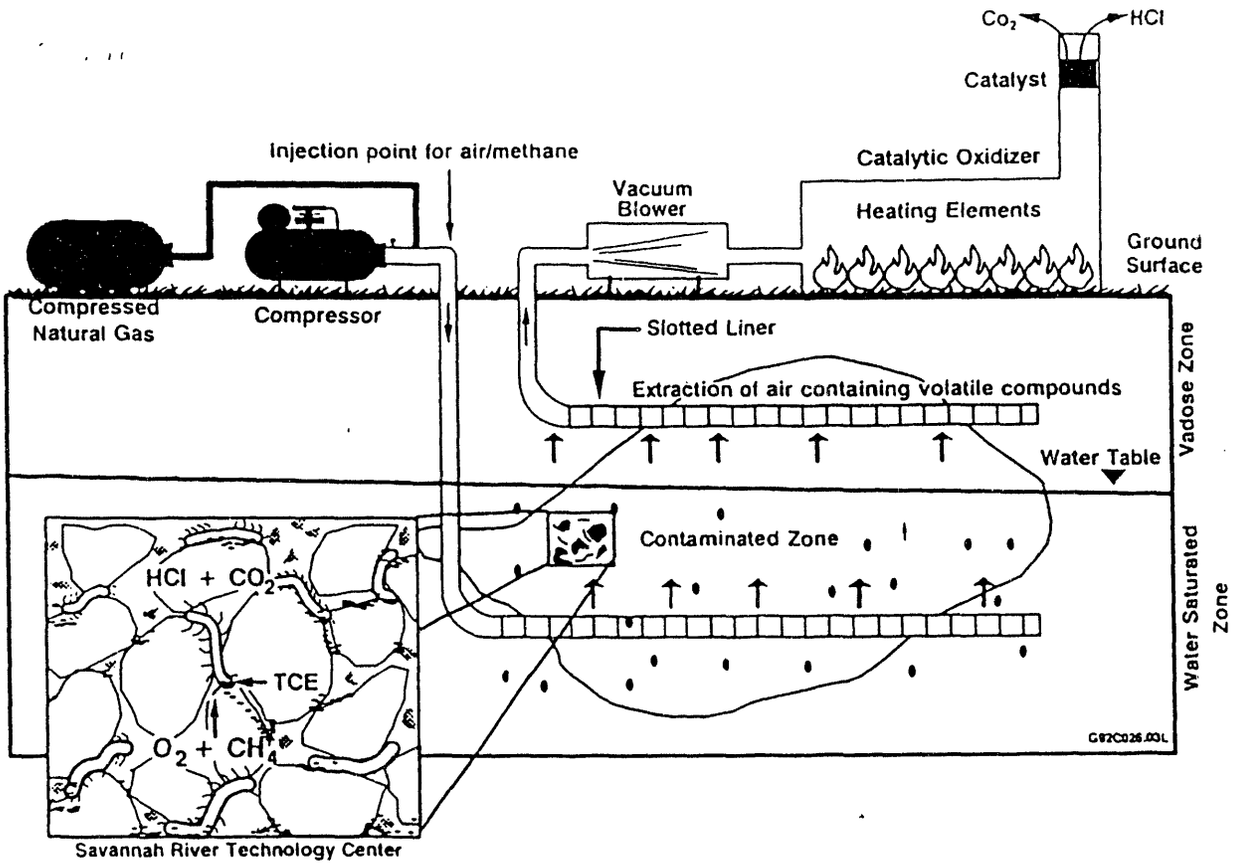
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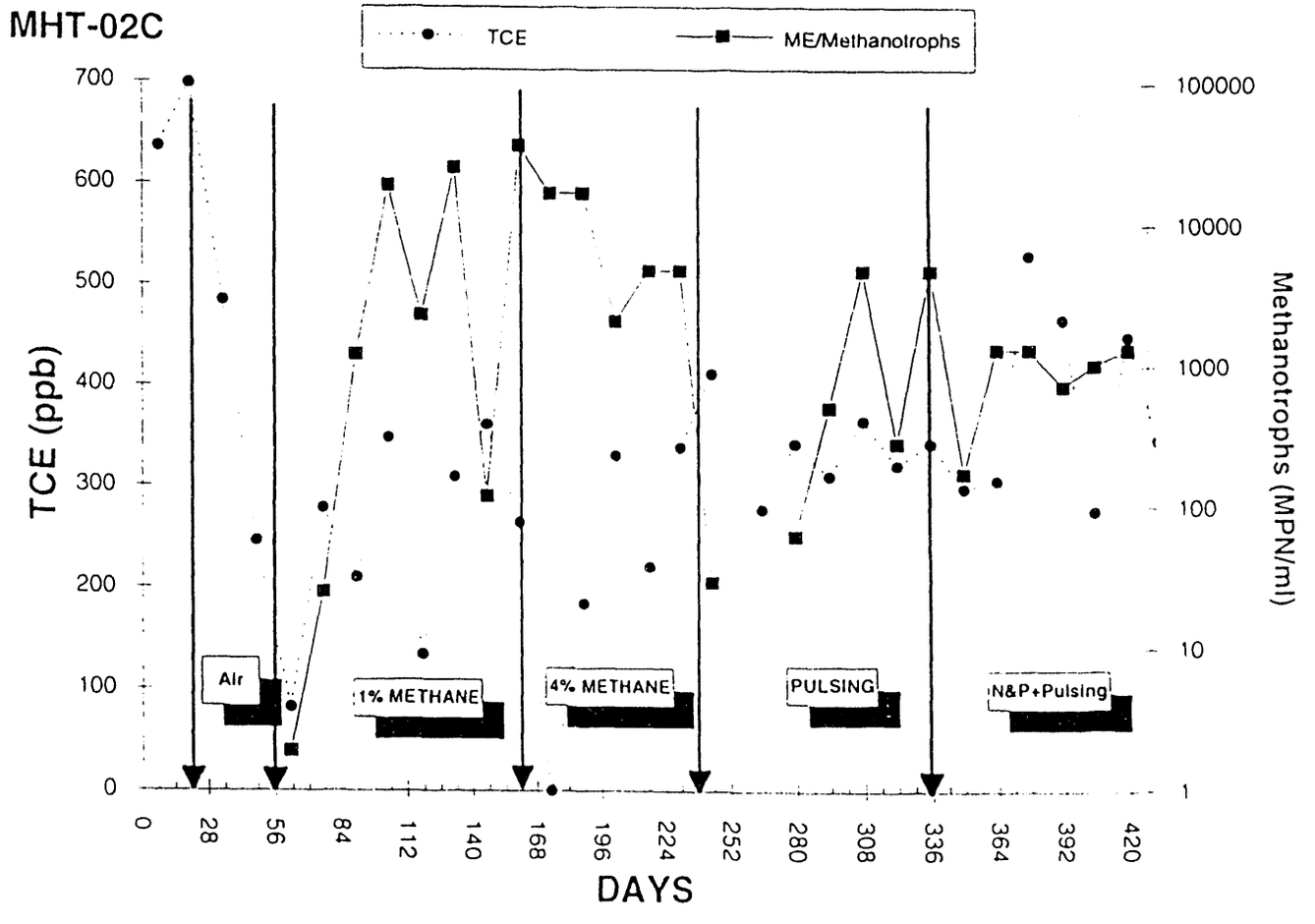
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