INNOVATIVE TECHNOLOGY
Summary Report

In Situ Bioremediation for the Hanford Carbon Tetrachloride Plume

Subsurface Contaminants
Focus Area

Prepared for
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Office of Environmental Management
Office of Science and Technology

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INNOVATIVE TECHNOLOGY

Summary Report

In Situ Bioremediation for the Hanford Carbon Tetrachloride Plume

OST Reference #1742

Subsurface Contaminants
Focus Area

Demonstrated at
Hanford Site
Richland, Washington

INNOVATIVE TECHNOLOGY
Summary Report

Purpose of this document

Innovative Technology Summary Reports are designed to provide potential users with the information they need to quickly determine if a technology would apply to a particular environmental management problem. They are also designed for readers who may recommend that a technology be considered by prospective users.

Each report describes a technology, system, or process that has been developed and tested with funding from DOE's Office of Science and Technology (OST). A report presents the full range of problems that a technology, system, or process will address and its advantages to the DOE cleanup in terms of system performance, cost, and cleanup effectiveness. Most reports include comparisons to baseline technologies as well as other competing technologies. Information about commercial availability and technology readiness for implementation is also included. Innovative Technology Summary Reports are intended to provide summary information. References for more detailed information are provided in an appendix.

Efforts have been made to provide key data describing the performance, cost, and regulatory acceptance of the technology. If this information was not available at the time of publication, the omission is noted.

All published Innovative Technology Summary Reports are available on the OST Web site at http://OST.em.doe.gov under "Publications."

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SECTION 1

SUMMARY

Technology Summary

Problem

Nuclear production operations have contributed various types of contamination to the subsurface at a number of DOE sites. In many cases, organic contaminants have migrated to the water table thus affecting the shallow aquifer at the site. Organic contaminants in the groundwater are often laterally dispersed over large areas and vertically dispersed to depths to hundreds of feet below the ground surface. These groundwater contaminants are difficult to treat; the baseline techniques of excavation and/or pump and treat are very expensive over the life cycle of the project; often projected to be in excess of 30-200 years.

The 200 Area at Hanford (also called the Central Plateau) contains approximately 817 waste sites, 44 facilities to be demolished, and billions of gallons of contaminated groundwater resulting from chemical processing plants and associated waste facilities (e.g., waste tanks). From 1955 to 1973, carbon tetrachloride, nitrate, and other materials were discharged to subsurface liquid waste disposal facilities in the 200 Area. As much as 600,000 kilograms of carbon tetrachloride may have entered the soil column and a portion of this has contaminated the underlying aquifer.

How It Works

In Situ Bioremediation for the Hanford Carbon Tetrachloride Plume (ISB), which is the term used in this report for an in situ treatment process using indigenous micro-organisms with a computer based Accelerated Bioremediation Design Tool (ABDT), remediates groundwater contaminated with volatile organic compounds (VOCs) and nitrates under anaerobic conditions. ISB involves the injection of nutrients into the groundwater with subsequent extraction and re-injection of the groundwater to provide nutrient distribution in the aquifer (Figure 1). The technology was developed by Battelle's Pacific

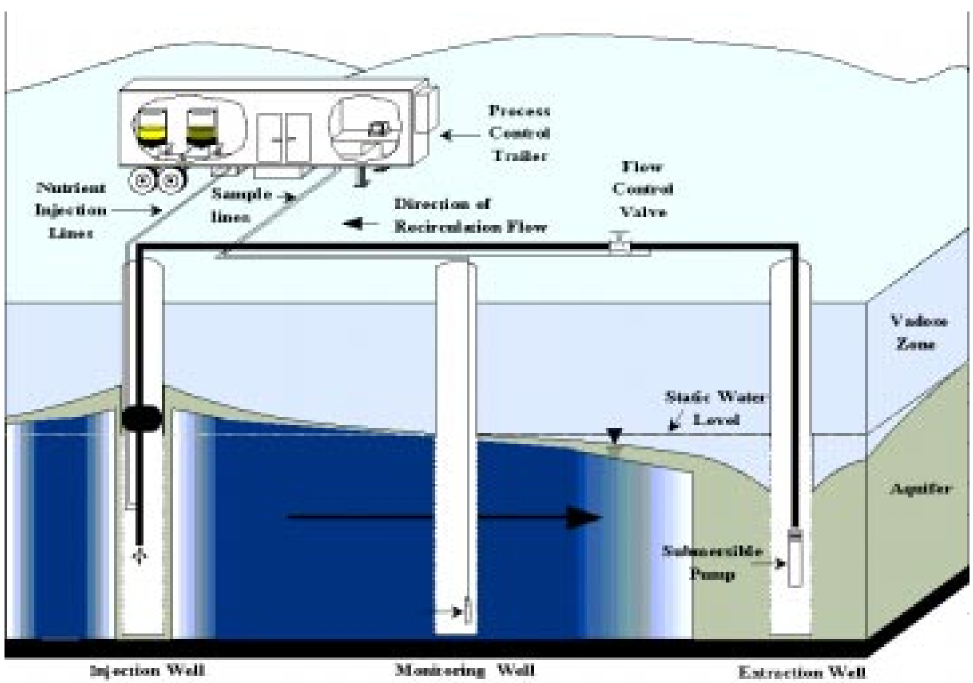

Figure 1. In Situ Bioremediation Concept

Northwest National Laboratory (PNNL) with support from the U. S. Department of Energy (DOE) Office of Science and Technology.

ISB involves injection of nutrients into the groundwater through injection wells, and extraction of groundwater through extraction wells. The extracted groundwater is filtered, nutrients are added, and then the groundwater is re-injected. Nutrients are added in pulses. Operations are conducted from a process-control trailer using a personal-computer (PC)-based process-control system, the Accelerated Bioremediation Design Tool (ABDT), which automated the nutrient injection, the required sampling operation, and the data collection. Using a flow and transport model, the ABDT quickly extrapolates data collected at the monitoring locations to induce good conditions for biological contaminant destruction, to limit formation of undesirable by-products, and to encourage microbial growth. The major innovation of ISB is in the use of the computer-based engineering design tool, the ABDT, to aid in selecting appropriate system designs and to determine optimal operating strategies.

Potential Markets

The potential markets for this technology include DOE, DOD, and commercial sites, which have moderately permeable, relatively homogeneous sediments contaminated with VOCs and/or nitrates within an anaerobic aquifer system.

Advantages Over Baseline

The advantages of ISB over baseline methods include :

- ISB has the potential to be a more efficient and cost-effective treatment of contaminated groundwater than the baseline pump-and-treat technology.
- The ISB may be effective in treating plumes caused by dissolution of non-aqueous phase liquids (NAPLs).

Demonstration Summary

This report covers demonstrations that took place between January 1995 and March 1996 at the DOE Hanford Site in Washington State. The field-scale demonstration included two separate tests, which were conducted in distinct, unconnected water-bearing units at the same test site.

The demonstration site was located in the 200 West Area, approximately 250 feet north of the sanitary tile field and 750 feet west of the the 221-T plant. The aquifer at this location contains elevated concentrations of both carbon tetrachloride and nitrate. Carbon tetrachloride concentrations are approximately 2 mg/l and nitrate concentrations are about 250 mg/l. The source is upgradient, approximately 1 mile away. The unsaturated zone at the demonstration site is 75 meters thick and is uncontaminated. Two highly permeable zones occur at depths of 75 to 78 meters and at 87 to 92 meters with an intervening low permeability unit. The two permeable zones do not interact significantly. The Site demonstration characteristics are described in more detail in Section 3, Performance.

Key Results

- Almost 2 kg of carbon tetrachloride were destroyed during the upper and lower zone tests.
- The upper zone test produced more than 20 kg (dry weight) of bacteria while the lower zone tests produced more than 10 kg (dry weight).
- Contaminant destruction was effective without plugging of the injection well.

- The ABDT developed for this project proved effective in the design and operation of the ISB demonstration.
- ISB could reduce the cost and time for remediation by as much as 50% compared to the baseline pump-and-treat technology.

Commercial Availability/Status

- Battelle has signed nonexclusive agreements with three leading environmental companies to work as teams in applying and commercializing ISB at DOD and industrial sites.
- At the DOE Idaho National Engineering and Environmental Laboratory, ISB is scheduled to be used in remediation of TCE-contaminated groundwater.
- At Point Mugu Naval Facility in California, ISB is being used for remediation of PCE- and TCE-contaminated groundwater.

Contacts

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Other

All published Innovative Technology Summary Reports are available online at HTTP ://em-50.em.doe.gov. The Technology Management System, also available through the EM50 web site, provides information about OST programs, technologies, and problems. The OST reference number for ISB is 1742.

TECHNOLOGY DESCRIPTION

Overall Process Definition

The field test system consisted of an injection/extraction well pair (dual multi-screened wells using pneumatic packers to isolate well screen intervals) approximately 17 meters apart, two monitoring wells, a nutrient injection system, and a groundwater sampling system (Figure 2).

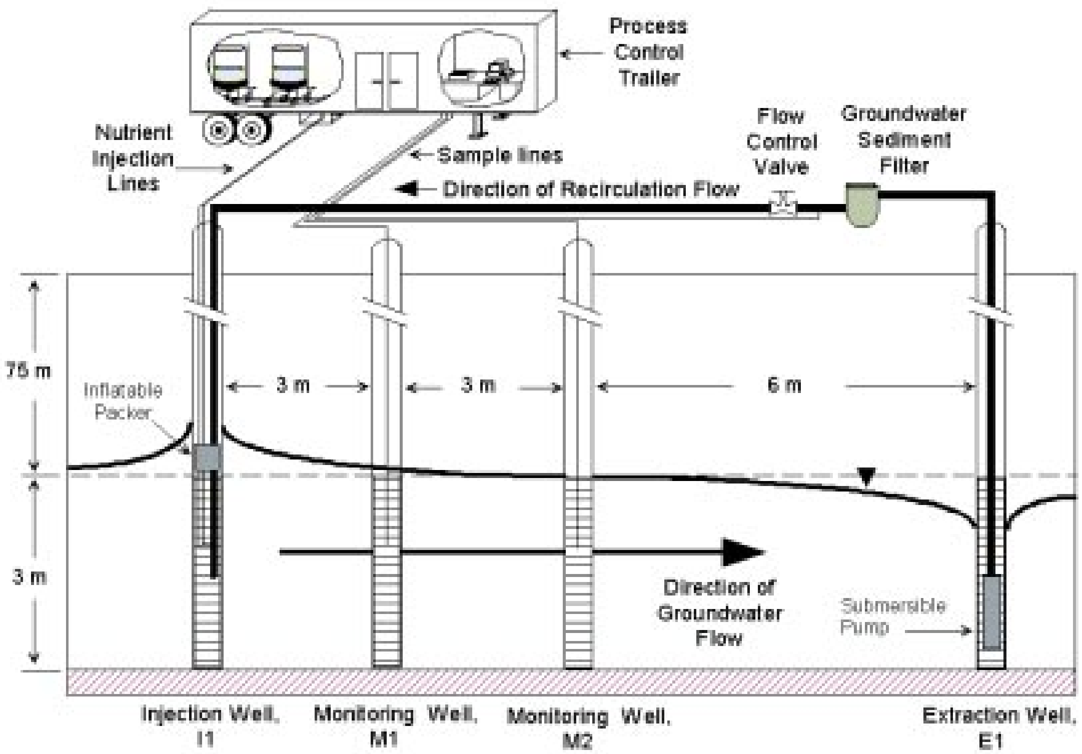

Figure 2. Recirculation/Monitoring Well System Used for Biostimulation Operations

- The two monitoring wells were located on the centerline between the two recirculation wells.
- Only two monitoring wells were used due to the costs associated with the installation of 320 foot-deep wells.
- Only partial hydraulic control was possible during the test, because of the use of only two wells.
- Two separate tests were performed in distinct, non-interacting aquifer layers at the same test site, an upper aquifer zone biostimulation and a lower aquifer zone biostimulation.

System Operation

- Nutrient injection campaigns
  - — Acetate pulses were added at about 24 hour intervals to promote dechlorination of carbon tetrachloride by indigenous microorganisms.

- Nitrate was added in pulses at about 24 hour intervals, skewed in time by approximately 10 hours from the acetate pulses.

- System controls
  - The ABDT was used to determine the concentration, pulse duration, pulse period, and skew time required between acetate and nitrate pulses.
  - Acetate nutrient solution was injected into the upper and lower zones of the aquifer in separate operation phases. Nitrate was also added in pulses as required to maintain a specified concentration.
  - Ground water was extracted, passed through a filter and flow control valve, and reinjected with nutrients added.
- Concentrations of carbon tetrachloride and the by-product chloroform were measured every two weeks, while pH, temperature, and oxidation/reduction potential were monitored continuously.
  - Indigenous anaerobic microorganisms reduced nitrate to nitrogen gas, and carbon tetrachloride was dechlorinated, resulting in the formation of CO_2 and chloride ions, In addition, some chloroform was produced as a byproduct.
- Operations were conducted from a process-control trailer.
- A personal-computer (PC) based process control system (ABDT) automated the nutrient injection, the required sampling operation, and the data collection
- Using a flow and transport model, the ABDT quickly extrapolated data collected at the monitoring locations to induce optimum conditions for contaminant destruction rates, limit by-product formation, and encourage microbial growth.

SECTION 3

PERFORMANCE

Demonstration Plan

Performance of the technology is based upon a field-scale demonstration in the 200 Area at the Hanford Site.

- The demonstration site in the 200 Area at Hanford (also called the Central Plateau) overlies a portion of a ground water contaminant plume, which contains approximately 2 mg/l carbon tetrachloride and 250 mg/l nitrate in the vicinity. Contaminant concentrations are relatively constant over a large area of the plume surrounding the test site.
- The unsaturated zone is 75 meters thick and is uncontaminated at the demonstration site.
- A permeability profile at the demonstration site shows highly variability with depth due to cementation of certain layers by carbonate minerals.
- Hydrogeologic measurements indicate two highly permeable zones (approximately 10^{-3} cm/s at depths of 75 to 78 meters (245 to 255 feet) and 87 to 92 meters (286 to 300 feet) with an intervening low permeability unit (approximately 10^{-6} cm/s). The two permeable zones do not interact significantly even under pumping stresses.
- Predictions of microbial distsribution, nutrient usage, and carbon tetrachloride destruction agreed with field data, validating the ABDT tool to apply to full-scale ISB.

Results

- Biostimulation was responsible for the simultaneous destruction of carbon tetrachloride (dechlorination) and nitrate (denitrification) with less than 2% conversion to chloroform in both the upper and lower aquifer zones (Figure 3).

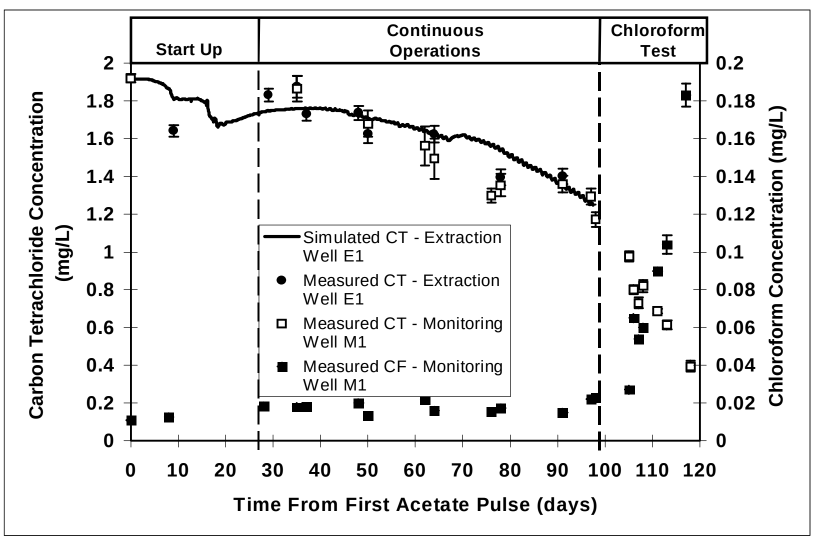

Figure 3. Dechlorination Results During Upper Zone Biostimulation

— Laboratory tests using Hanford Site cores demonstrated simultaneous carbon tetrachloride dechlorination by indigenous microorganisms under denitrification conditions.
— Decreases in carbon tetrachloride and nitrate concentrations in ground water coincided with the addition and subsequent depletion of the acetate nutrient.
— Increases in the numbers of indigenous microorganisms coincided with addition of acetate nutrient, depletion of nitrate, and destruction of carbon tetrachloride.
— Evidence of biostimulation : in both the upper zone and lower zone the numbers of microorganisms increased 1 to 2 orders of magnitude (biomass was increased by one order of magnitude up to 8m away from the lower injection well). The addition of acetate and nitrate stimulated the growth of indigenous denitrifying bacteria that can co-metabolically dechlorinate carbon tetrachloride.

- The rates of carbon tetrachloride destruction were 0.8mg/g-biomass/day in the upper zone, 0.9mg/g-biomass/day in the lower zone.
- Nitrate was destroyed at a rate of 100 mg nitrate/g biomass/day during initial denitrification.
- Growth of indigenous microorganisms was controlled to provide good contaminant destruction without plugging the reinjection well.
- Nitrite accumulation was controlled by introducing a large pulse of acetate and stopping recirculation, thus stimulating microbial nitrite reduction (Figure 4).

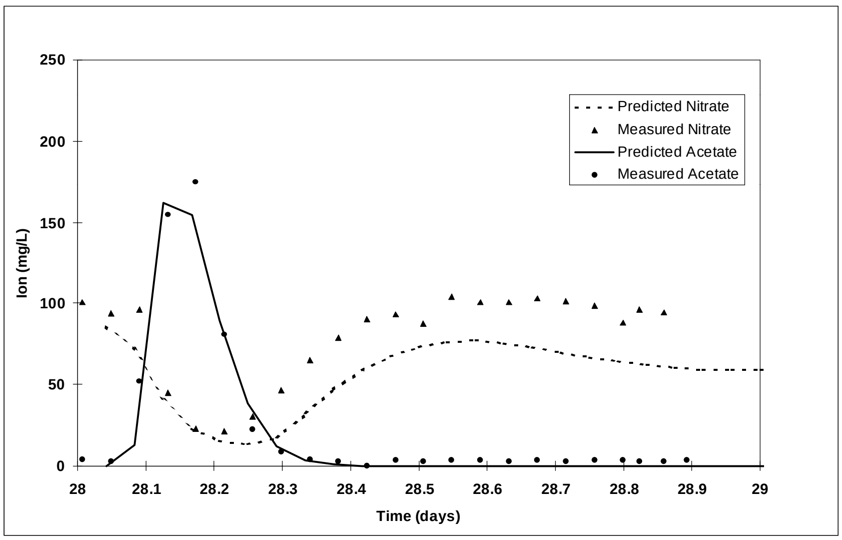

Figure 4. Measured and Predicted Acetate and Nitrate Concentrations at Well M2 on Day 30 of Upper Zone Operations

SECTION 4

TECHNOLOGY APPLICABILITY AND ALTERNATIVES

Competing Technologies

- ISB with aqueous nutrient injection is competitive with conventional baseline technologies of pump-and-treat and pump-and-treat combined with soil vapor extraction.
- **The** effectiveness of ISB was compared with performance data from ex situ air stripping and granular activated carbon (AS/GAC) for removal/destruction of carbon tetrachloride. This comparison was used as the basis of the cost analysis discussed in Section 5.
- Air sparging in vertical wells and in-well recirculation technologies have been implemented at similar sites across the U.S. and in Europe. In-well vapor stripping was demonstrated at Edwards AFB, supported by DOE funds.
- Aerobic ISB has been successfully demonstrated and is selected for use in remediation at the DOE Savannah River Site. It is also commercially available through several licensees.
- Anaerobic ISB, using similar processes, is being demonstrated at Dover AFB (DOE supported) and commercial sites.

Technology Applicability

- ISB is cost-effective for plumes or portions of plumes small enough for volumetric treatment (100-m diameter range).
- ISB is cost-effective where plumes exhibit non-equilibrium contaminant partitioning (i.e., caused by preferential flow paths, layering, sorption in soil particles/micropores).
- ISB is applicable for a source area plumes where significant contaminant sorption has occurred.
- ISB is effective for remediation of ground water contaminated with VOCs (carbon tetrachloride in this demonstration) and nitrate.
- Because ISB requires recirculation of ground water to distribute nutrients within the treatment zone, low permeability zones may not be treated or may require additional technology advancements for this technology to be effective.
- Aquifer conditions must be suitable to support microbial growth. Aquifer limiting conditions include toxicity due to high contaminant concentrations or other toxic substances, and pH. Aquifer-specific treatability tests are necessary.
- Currently the innovative Treatment and Remediation Demonstration Program is assisting Hanford in re-evaluating the use of ISB for this plume.
- Stimulation of indigenous microorganisms by injection of nutrients in water was demonstrated at Moffett Field, California, forming the technical basis for the design of this demonstration. However, the Moffett Field carbon tetrachloride bioremediation did not involve scale-up issues or mechanisms to control chloroform production, as did this demonstration.
- The ABDT is currently being demonstrated at other sites for anaerobic in situ bioremediation through agreements with industrial partners.

- Aerobic ISB has been successfully demonstrated and is selected for use in remediation at the DOE Savannah River Site. Anaerobic ISB is also commercially available through several commercial licensees and has been applied at a number of commercial sites.

Patents/Commercialization/Sponsor

- No patents have been sought nor issued to Battelle Pacific Northwest National Laboratory for the ISB technology.
- Battelle Pacific Northwest National Laboratory has signed nonexclusive agreements with OHM Remediation Services, Parsons Engineering, and Montgomery Watson to work as teams in applying and commercializing ISB at DOD and industrial sites.

SECTION 5

COST

Methodology

A preliminary cost analysis of ISB was developed by PNNL using data from this demonstration.

- The mass of contaminant removed or degraded by biological processes is difficult to quantify.
- The technology of pump and treat with ex situ air stripping/activated carbon was used as the baseline against which ISB was compared. To compare the two remediation systems several assumptions were made.

— The size of the ground water plume is based upon a conservative estimate of the volume of ground water that can be treated by ISB using only 2 wells (30,000 cubic meters can be treated).
— Costs for each technology were based on removing/destroying the initial aqueous (1 mg/L) and sorbed contamination to reach a final aqueous concentration of 0.005 mg/L. The initial mass of contaminant is estimated as 9 kg aqueous and 9.9 kg sorbed.
— Cost comparisons are not based on life-cycle present value costs.
— Costs for ISB were based on process knowledge from the Hanford ISB demonstration and on the use of process simulations.

Cost Analysis

Since the completion of the ISB demonstration at the Hanford Site, further cost/benefit analysis for the use of ISB have been made with the RT3D software code developed by PNNL.

- RT3D is a Fortran 90-based software package for simulating 3-dimensional, multi-contaminant, reactive transport in ground water. RT3D can accomodate multiple sorbed and aqueous-phase contaminants with any reaction framework that the user wishes to define.
- Cost/benefit analyses for remediation of the carbon tetrachloride plume were made for the 100 microgram/l contour and the 1,000 microgram/l contour. Assumptions included equilibrium partitioning and a homogeneous aquifer.
- The remedial options evaluated were:

— ground water extraction with air stripping and the size granulated activated carbon adsorption and
— anaerobic bioremediation.

- The resulting analysis showed that the range of life-cycle costs for ISB were $8.8 million to $9.9 million, compared to $6.6 million to $7.0 million for the baseline for the duration of treatment in both cases. This cost difference is due primarily to the greater number of wells required for ISB.
- The Hanford demonstration and the bench-scaleanalyses performed for Point Mugu and INEEL demonstrate that ISB is cost-effective where plumes or portions of plumes are small enough for volumetric treatment (100-meter diameter range), in aquifers where contaminant plumes exhibit non-equilibrium contaminant partitioning (i.e., caused by preferential flow paths, layering, and sorption in soil particles/micropores), in source area plumes with significant contaminant sorption.

Capital Costs

- Capital costs for ISB are estimated to be substantially less than ex situ air stripping/GAC.
- Fixed equipment costs for ISB include nutrient mixing and injection equipment for providing nutrients required for stimulation of the bioremediation.
- The costs of installing wells is a major capital cost factor, since ISB may require twice the number of wells as would be required by AS/GAC. For this cost comparison, a 25 meter well with a 15 meter screen interval was used, and costs were estimated using $100/foot for well installation (typical for the Hanford case, but much higher than other sites).

Operating Costs

- The estimated annual operating costs are comparable between the baseline and the innovative treatment technology.
- The treatment time is estimated to be 4.5 years to remediate the 30,000 cubic meter ground water plume using the baseline air sparging/GAC technology, but only 1.9 years with ISB. Thus, ISB would require only about 42% of the treatment time with a comparably lower total operating cost.

Cost Conclusions

	ISB	AS/GAC
Capital cost	$88.0 k	$172 k
Operating cost	$87.5 k	$225 k
Treatment cost	$175.4 k	$397 k
Total	$5.80/cu m	$13.30cu m

- The estimated treatment time required for AS/GAC was directly related to the time required for contaminant extraction.
- Time for contaminant extraction was based upon very conservative assumptions that effective porosity equals actual porosity and that the aquifer is homogenous.

SECTION 6

REGULATORY AND POLICY ISSUES

Regulatory Considerations

- No specific permits were required for the field test at Hanford. A categorical exclusion was granted for NEPA requirements.
- Future application of ISB may require underground injection permits and NEPA review at other sites.

Safety, Risks, Benefits, and Community Reaction

Worker Safety

- There are no unusual health and safety issues related to the operation of in situ bioremediation.
- Reagents used in the process (acetate and nitrate) are easily managed using standard chemical handling procedures.
- Level D personnel protective clothing was used during installation and operation of the system.

Community Safety

- In situ bioremediation does not produce any significant routine release of contaminants.
- No unusual or significant safety concerns are associated with the transport of equipment, samples, waste, or other materials associated with in situ bioremediation.
- No harmful microbes were detected at the demonstration site after biostimulation

Environmental Impact

- Surface disturbance at the site is minimal.
- Nutritional enrichment does not promote the growth of harmful microbes at the demonstration site.

Socioeconomic Impacts and Community Perception

- In situ bioremediation has minimal economic or labor force impact.
- In situ bioremediation is viewed by the public as a “green” technology and is perceived as a preferred and acceptable technology based upon Hanford stakeholder input.

LESSONS LEARNED

Design Issues

- Effective ISB system designs and operational process control require an ABDT or similar process simulator.
- A flow and transport model predicted the contaminant destruction rate from inexpensive, rapid measurements of anions and a conservative bromide tracer.
- Factors that will control injection protocols, remediation system siting, and monitoring include site geology (especially permeability and heterogeneity), concentrations of native nutrients (such as total organic carbon), and natural oxidation potential of the subsurface (i.e. aerobic or anaerobic conditions).
- Stimulating microbial growth directly affects the permeability of the aquifer, which then may change flow patterns and
- the characteristics of the overall injection-extraction system.
- Use of the ABDT allows quick corrective action (changes in the amount/duration of nutrient pulse or the pulse period) to maintain rapid contaminant destruction during these changes.

Implementation Considerations

- A significant portion of ISB operations can be automated through a PC-based system.
- The ABDT controlled the nutrient pulses, collected electronic data for pressure, flow rate, pH, redox, and T, controlled a fraction collector and groundwater pumps, and provided signals if a measured parameter exceeded established control limits.
- Periodic sampling of ground water constituents is required to control the ISB process.

Technology Limitations and Needs for Future Development

- Better monitoring methods for determining mass balance and microbiological health of the subsurface population are required to facilitate implementation of ISB.
- Sustained operation of ISB requires managing the permeability changes caused by microbial growth. The ABDT developed as part of this demonstration proved to be an excellent platform for managing permeability changes, but more field experience with this system is required.
- Better mechanisms for effective distribution of nutrients into low permeability zones of an aquifer are required to broaden the applicability of ISB remediation.
- Treatability tests are required to facilitate better and more efficient ISB implementation.

Technology Selection Considerations

- This technology yields significant economic and efficiency gains over conventional baseline technologies for remediation of ground water contaminated with VOCs and nitrates.
- ISB may be effective for treating plumes caused by dissolution of non-aqueous phase liquids (NAPLs).

- The optimal geologic setting for successful ISB would be moderate to high soil permeability and a fairly homogenous saturated zone to allow for effective injection of nutrients and sufficient saturated thickness.

APPENDIX A

REFERENCES

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APPENDIX B

DEMONSTRATION SITE CHARACTERISTICS

Site History

- The Hanford Site's mission has been to support national defense efforts through the production of nuclear materials. From 1944 to 1989, as part of the plutonium recovery processes, tritium, carbon tetrachloride, chromium, nitrates, cobalt, strontium, cesium, technetium, iodine, plutonium, and uranium contamination were released to the soil and ground water, where it is presently found in many locations at the 560 square mile site.
- The 200 Area at Hanford (also called the Central Plateau) contains approximately 817 waste sites, 44 facilities to be demolished, and billions of gallons of contaminated ground water resulting from operations of the chemical processing plants and associated facilities (e.g., waste tanks).
- From 1955 to 1973 carbon tetrachloride, nitrate, and other materials were discharged to subsurface liquid waste disposal facilities in the 200 Area. As much as 600,000 kilograms of carbon tetrachloride may have entered the soil column and a portion of this has contaminated the underlying aquifer.

Contaminants of Concern

- An estimated 20,000 to 70,000 kg of carbon tetrachloride now contaminates a 10 square kilometer ground water plume at Hanford.
- The highest concentration of carbon tetrachloride in the 200 West Area ground water is approximately 7 mg/L.
- Carbon tetrachloride (2 mg/L) and nitrate (250 mg/L) are the only contaminants of concern at the ISB test site.

Contaminant Locations and Hydrogeologic Profiles

- The test area is a portion of the ground water contaminant plume that contains approximately 2 mg/L carbon tetrachloride and 250 mg/L nitrate. Contaminant concentrations are relatively constant over a large area of the plume surrounding the test site.
- The unsaturated zone is 75-m thick and is uncontaminated at the test area location.
- A permeability profile at the test site is highly variable with depth due to cementation by carbonate deposits.
- Hydrologic measurements indicate two highly permeable zones (approximately10-3 cm/s) in the ground water at depths of 75 to 78 m (245 to 255 ft) and 87 to 92 m (286 to 300 ft) and an intervening low permeability unit (approximately 10-6 cm/s). The two permeable zones do not interact significantly even under pumping stresses.
- Separate biostimulation operations were conducted in both permeable zones.
- The hydraulic and chemical characteristics of the test site upper zone are listed below. The lower zone hydraulic and chemical properties were similar except as noted in the text.

Hydraulic properties

Horizontal hydraulic conductivity	5×10^{-3} cm/s
Longitudinal dispersivity	1.0 m
Effective porosity	0.15
Regional gradient	0.001 m/m

Sediment chemistry

carbon tetrachloride	-0.287 µg/gm
chloroform	-0.103 µg/gm
carbonate	-4%
leachable phosphorus	0.027-0.053%

Ground water chemistry

carbon tetrachloride	1918 µg/l
chloroform	12 µg/l
nitrate	240 mg/l
nitrite	nd
sulfate	55 mg/l
oxygen	5 mg/l
pH	7.4
temperature	19º C

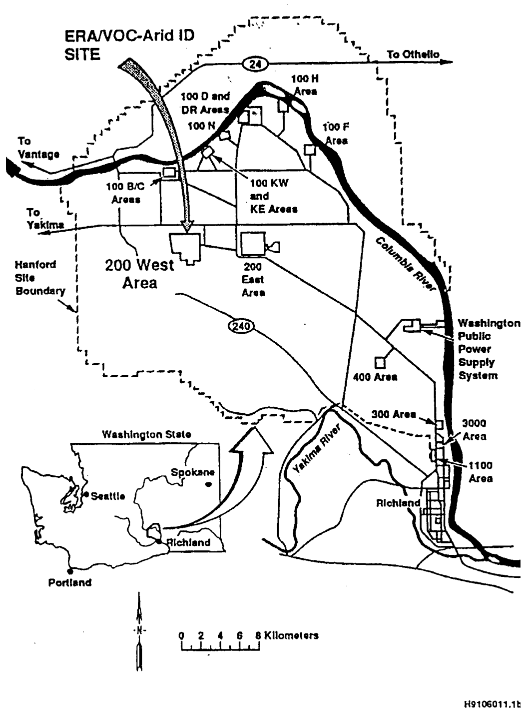

Figure B-1. Location of the ISB Demonstration Site

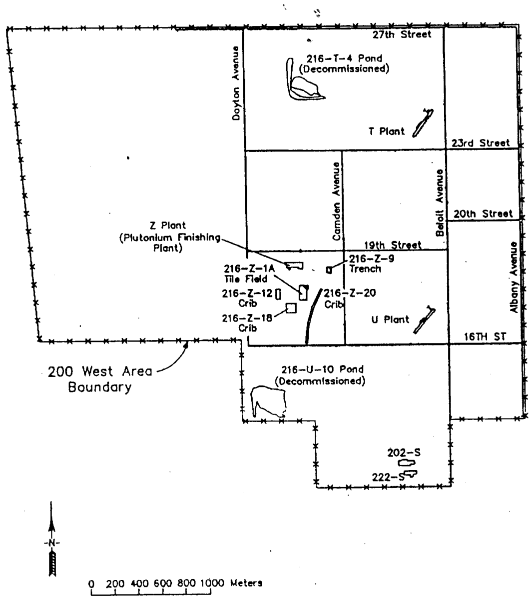

Figure B-2. Site Map of the 200 West Area

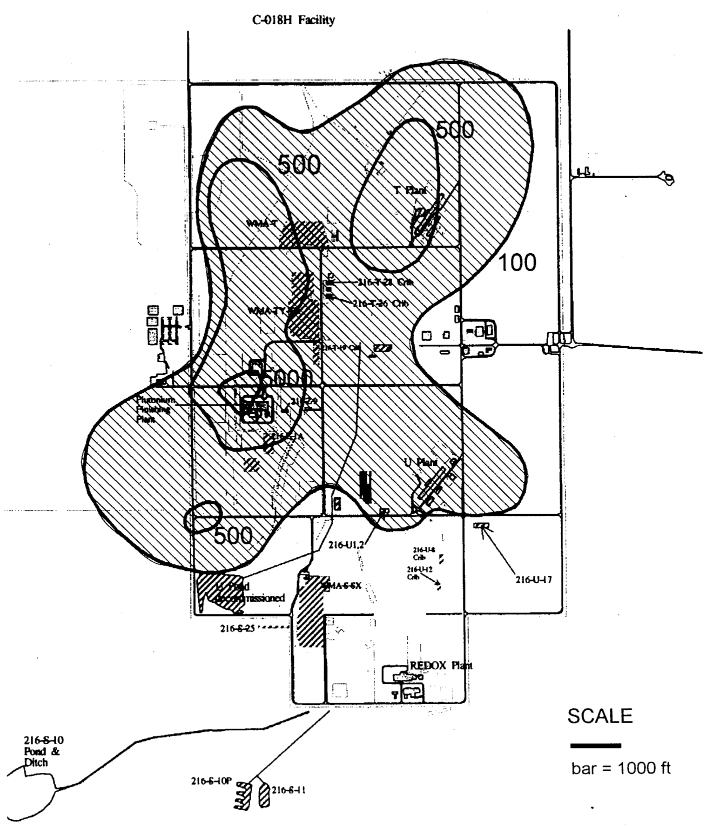

Figure B-3. Carbon Tetrachloride Plume in the Groundwater at Hanford