Development of the Integrated In Situ Lasagna Process

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4.3   Development of The Integrated *In Situ* Lasagna Process

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Introduction

Contamination in deep, low permeability soils poses a significant technical challenge to in-situ remediation efforts. Poor accessibility to the contaminants and difficulty in uniform delivery of treatment reagents have rendered existing in-situ methods such as bioremediation, vapor extraction, and pump and treat rather ineffective when applied to low permeability soils present at many contaminated sites.

Recently the use of electrokinetics as an *in situ* method for soil remediation has received increasing attention due to its unique applicability to low-permeability soils (2-11). Electrokinetics includes the transport of water (electroosmosis) as well as ions (electromigration) as a result of an applied electric field. Electroosmosis in particular has been used since the 1930s for dewatering clays, silts, and fine sands (1). For remedial applications, water is typically introduced into the soil at the anode to replenish the water flowing towards the cathode due to electroosmosis. The water flow is utilized to flush organic contaminants from the subsurface soil to the ground surface at the cathode region for further treatment or disposal.

Advantages with electroosmosis include uniform water flow through heterogeneous soil, high degree of control of the flow direction, and very low power consumption. As currently

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practiced, however, the technology suffers several limitations on large-scale operations. These include low speed (liquid flow induced by electroosmosis typically moves about 1 inch per day for clay soils), additional aboveground treatment and unstable long-term operation resulting from soil drying and cracking, steep pH gradient in the soil bed, and precipitation of metals and minerals near the cathode. Recent efforts to mitigate the pH problem primarily involve conditioning the anode and cathode solutions through external recirculating loops (9,10).

There is a need for a low-cost, in-situ technology that can effectively treat a wide range of contaminants in low-permeability soils or soils containing low-permeability zones. In this paper, we describe an integrated approach that minimizes the drawbacks associated with the use of electrokinetics alone, and present the experimental results of the initial field test.

Technical Approach

Monsanto's new approach is an integrated in-situ treatment in which established geotechnical methods are used to install degradation zones directly in the contaminated soil and electrokinetics is utilized to move the contaminants through those zones until the treatment is completed.

One possible configuration of the process has the following components:

a) create permeable zones in close proximity sectioned through the contaminated soil region, and turn them into treatment zones by introducing appropriate materials (sorbents, catalytic agents, microbes, oxidants, buffers, etc.). Hydraulic fracturing and related technologies may provide an effective and low-cost means for creating such zones horizontally in the subsurface soil. The treatment zones can also be vertical, which can be constructed using sheet piling, trench, slurry wall, etc. Instead of being discrete, the treatment zones can be continuous using *in situ* soil mixing devices.

b) utilize electrokinetics for transporting contaminants from the soil into the treatment zones. Since these zones are deliberately located close to one another, the time taken for the contaminants to move from one zone to the adjacent one can be short. In the horizontal configuration, the zones above and below the contaminated soil area can be injected with graphite particles during the hydrofracturing process to form in-place electrodes.

c) liquid flow can be periodically reversed, if needed, simply by switching the electrical polarity. This mode would enable multiple passes of the contaminants through the treatment zones for complete sorption/destruction. The polarity reversal also serves to minimize complications associated with long-term operation of uni-directional electrokinetic processes as discussed above. Optionally, the cathode effluent (high pH) can be recycled directly back to the anode side (low pH), which provides a convenient means for pH neutralization as well as simplifies water management.

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Electrodes and treatment zones can be of any orientation depending upon the emplacement technology used and the site/contaminant characteristics. Schematic diagrams of two typical configurations, horizontal and vertical, are shown in Figure 1. The process has been called Lasagna™ (12) due to the layered configuration of electrodes and treatment zones. Conceptually, the technology could treat organic and inorganic contamination as well as mixed wastes.

Project Description

A consortium was formed in 1994 consisting of Monsanto, E. I. du Pont de Nemours & Co., Inc. (DuPont) and General Electric (GE). With participation from the Environmental Protection Agency (EPA) Office of Research and Development and the Department of Energy (DOE), the consortium combined resources to accelerate the development of the Lasagna technology. The consortium's activities have been facilitated by Clean Sites, Inc., under a Cooperative Agreement with EPA's Technology Innovation Office.

The initial Lasagna technology development is referred to as Phase I in which trichloroethylene (TCE), a major contaminant at many DOE and industrial sites, was selected as the target contaminant . Phase I consisted of three major efforts: 1) developing key technical components of the Lasagna process, 2) conducting field experiments to evaluate the coupling of individual components, and 3) evaluating the overall performance including cost of remediation. For the field test, the Paducah Gaseous Diffusion Plant (PGDP) site was chosen from a list of Department of Energy sites based on the two major criteria of low-permeability soils and TCE as the single contaminant. DOE contributed by providing the contaminated site in Paducah for the test, soil sampling and analysis support (through Martin Marietta), and funds through a Research and Opportunity Announcement (ROA) grant. CDM-Federal, Inc., was hired by the consortium to construct and manage the field experiment.

The purpose of the field test was to experiment with the coupling of electroosmotic removal of TCE from the contaminated soil with in-situ adsorption by activated carbon in treatment zones in the vertical configuration. This was the initial phase of the Lasagna project, to be followed by a subsequent one incorporating in-situ degradation of TCE, either catalytically or biologically.

The major technical objectives for conducting the field test included studying the scaled-up characteristics of key operating parameters and the effectiveness of TCE removal. Important operating issues involved
- design of treatment zones and electrodes, emplacement methods and cost of installation
- electrical effects: voltage, current, power, soil conductivity, heating, etc.
- electrokinetic effects: electroosmotic flow, pH profile, solution conductivity
- responses to polarity & flow reversal

Issues related to TCE removal included extent of soil cleanup, effectiveness of carbon adsorption in an electroosmotic environment, and overall mass balance. Field data were also needed for the development of a mathematical model and for refining an economic model estimating the cost of treatment using the Lasagna process.

Results of the Field Experiment

The field test covered a soil section 15 feet long by 10 feet wide and approximately 15 feet deep. Each electrode zone consisted of 8 steel panels. There were a total of 4 treatment zones installed, each consisting of 11 or 12 individual wickdrains filled with activated carbon. The treatment zones were installed with 21 inches of soil between each zone. Figure 2 shows the layout of the field unit. A 4 ft by 4 ft by 15 ft deep control zone was installed at the west end of the unit and isolated hydraulically from the test site with interlocking sheetpiles. The control zone contained one carbon wickdrain.

The field unit was constructed during November and December 1995. The experiment was started (power on) on January 3, 1995 and lasted for four months. The voltage was then reversed for about one week, primarily to collect voltage and current data, before the power was turned off. Key operating characteristics of the test are reported below.

Electrokinetic Effects

Voltage and Current: The initial voltage was set at 138 volts and the corresponding current was about 41 amps. With the current held constant at 40 amps, the voltage slowly decreased with time and stabilized at about 105 volts after one month of operation. The lower voltage was due to the soil heating up (discussed below), which increased the electrical conductivity of the soil. The voltage gradient ranged from 0.45 to 0.35 volts/cm. The voltage drop across the unit was fairly linear and did not change over the course of the experiment. Total voltage and current trends are shown in Figure 3.

Electrokinetic Effects: During the first month, an electroosmotic flow rate of about 2 L/hr was measured, which was less than one half of the predicted value based on the bench-scale's measurements. This turned out to be due to pluggage in the cathode siphon tubes, causing water to "overflow" the cathode wicks. Once this problem was fixed, the flow rate averaged about 4 to 5 L/hr, corresponding to an electroosmotic permeability of 1.2×10^{-5} cm^2/V-s. This value, as well as the water flow rates, agreed very well with the lab data. With the first month's flow rate corrected to a value of 4 L/hr, the unit moved three pore volumes of water (between adjacent treatment zones) over the 4 month operating period. Figure 4 shows the flow rate and the corresponding pore volumes during the field test.

Conductivity and pH: The pH of the anode fluid dropped from near neutral at the beginning to between 5 and 6 for most of the test. This mild pH behavior for an electroosmosis experiment, which had also been observed in laboratory units, was a direct consequence of iron corrosion rather than water electrolysis as and the predominant anodic reaction. During the last month of the test, however, the anode fluid pH dipped rather unexplainably down to 2-3. The pH of the cathode fluid rose rapidly to about 12 and stayed there for the whole experiment. This is normal for electroosmosis due to the electrolysis of water generating OH^- at the cathode.

The conductivity of the anode solution ranged between 1000 to 3000 μS/cm for most of the test, except near the end when it fluctuated widely with occasional spikes up to 25,000 μS/cm with the prime ion species being iron and chloride. This behavior was probably related to

the low pH noted above during this period. The cathode solution conductivity was fairly constant at about 10,000 μS/cm with the primary ion species being sodium. These values were consistent with the laboratory data. A plot of pH and conductivity values over time are shown in Figure 5.

Temperature Effects: The initial temperature of the soil at the 10-foot depth was 15°C. The temperature rise at various locations due to the electrical power is shown in Figure 6. The core temperature (hottest spot) reached a maximum of 45.2°C at the end of the test. Notice the control zone temperature raises also. A possible reason for the increase in temperature in the control zone is that stray current could flow through it, causing heating. It could also be due to thermal conduction since the control zone was less than one foot from the anode.

To predict temperature rises in field experiment, a mathematical model was developed by General Electric using FIDAP®, a commercial computational fluid dynamics program. The governing equations included energy transport by conduction, electroosmotic convection, and heat generation from Joule heating. Temperature dependencies of electrical conductivity and electroosmotic permeability were accounted for. Specific examples simulating possible scenarios for the Lasagna pilot test in Paducah, KY were studied. With the appropriate values for thermal conductivity and heat capacity of the soil , the model predictions correspond very well with the field data as shown in Figure 7.

Polarity Reversal: Polarity of the DC power to the unit was reversed for 1 week at the end of the test to study the system response. The unit ran stably at a reverse voltage of 90 volts and a current of 39 amps. There was an increase in electrical conductivity of the soil when the polarity was reversed and the pH at the electrodes was shifting from high to low at the new anode and from low to high at the new cathode. These results were expected and similar to laboratory data. Thus, polarity reversal can be readily utilized in the field, if needed for reversing flow or neutralizing pH and/or osmotic gradients.

Overall, the field unit behaved very much like the laboratory and pilot experiments. The unit scale-up was excellent. The electroosmotic conductivity, pH and conductivity trends, power requirements, temperature trends and operational stability all were consistent with laboratory data.

TCE Removal and Mass Balance

Pre-Test Soil Samples: Prior to the installation of the Lasagna field unit, soil samples were taken and analyzed by Oak Ridge National Laboratories (ORNL) personnel. A total of 12 bore holes were made with samples taken every 1 foot to a depth of 15 feet below ground surface. Nine bore holes were completed within the boundary of the Lasagna unit, two in the control area and one to the outside west of the unit. The locations of the bore holes with respect to the wicks are shown in Figure 2. The results show TCE concentrations in the soil ranged from 1 mg/kg to over 500 mg/kg with an overall average of 83.3 mg/kg for the 12 bore holes from 4 to 15 foot depths (Table 2). The average concentration of TCE within the test site (15' x 15' x 10') is almost the same, 83.2 mg/kg, resulting in a total amount of TCE of about 9.25 kg.

Intermediate Carbon Samples: In order to assess the progress of the test, the carbon cassette from wick C4-7 was removed for analysis after 2 pore volumes of water exchange were obtained. Carbon samples were taken every foot from 3 to 15 feet. TCE levels on the carbon were found to be quite high, ranging from several thousand to over 10,000 mg TCE/kg carbon (Figure 8). This indicates that TCE was being effectively flushed from the soil and trapped on the carbon. Also shown in the figure are the additional amounts of TCE trapped on the fresh carbon inserted into cassette of wick C4-7 for the remainder of the test (1 additional pore volume). The data show that, except at the very low depths, not much additional TCE was trapped by the carbon, an indication that the soil was probably clean of TCE.

Post-Test Soil and Carbon Samples: At the completion of the test (3 pore volumes total), all the carbon sampling cassettes were removed and analyzed in the same manner as the intermediate samples. Soil samples were also taken. Twelve bore holes were dug near the original pre-test bore holes to a depth of 15 feet and slightly lower. Results in Table 2 show very high as well as uniform removal of TCE from the treated soil. The final soil concentrations were mostly below 1 mg/kg, with an average of 1.1 mg/kg for the 9 bore holes within the test boundary. TCE removal ranged from 92.4 to 99.8%, with an overall average of 98.4%. The soil samples taken either outside or deeper than the test zone (below 15 feet) still showed substantial amounts of TCE present. TCE removals outside the test boundary were 26% and 51% from the two spots in the control zone, and 68% from the west of the Lasagna unit. The removal in the control zone was probably due to diffusion into

the carbon wick (see modeling below). The removal in the area to the west was probably due to both migration and diffusion from the anode area and/or sampling inaccuracies. Despite significant degrees of removal from the control areas, the definite contrast between TCE levels in the soils within the treatment area and without (outside or deeper) is quite remarkable and shows conclusively the effectiveness of the process for cleanup of the contaminated soil.

Several of the pre-test soil samples showed TCE concentrations greater than 225 mg/kg, corresponding to an equilibrium pore water concentration of 1100 mg/l, thus indicated a residual DNAPL situation (12 through 15 foot samples of borehole L-08). In these likely DNAPLs locations, TCE levels were reduced to less than 1 mg/kg, except for the 15 foot deep sample which was reduced to 17.4 mg/kg. These results show that the process could be effective for removing residual DNAPL TCE from the soil.

TCE Mass Balance: Attempts were made to determine the mass balance for TCE taking into account the amounts in the soil before and after the test and the amount trapped on the carbon. This is a complex task due to the following reasons:

a) TCE levels in soil varied in all directions, so an averaging process was needed.
b) Not all the soil samples were taken directly in front of the corresponding carbon cassettes, so another estimating procedure was required.
c) The carbon cassettes were installed at the site about a month before the experiment actually started. So some TCE from the soil was lost due to passive diffusion to

the carbon cassette in the back row opposite the direction of electroosmotic flow. Based on diffusion modeling (Table 3 below), the loss could be about 5%.

d) Volatilization of TCE from the soil into the atmosphere did occur to some extent. Estimates calculated from measurements of the air samples, however, indicated that the volatilization loss was less than 5% of the total.

e) possible TCE degradation

Because of the above complications, the mass balance results ought not to be viewed as more than a gross indication of the fate of TCE.

Contour maps of TCE levels in the treated area were obtained using the kriging method. Table 4 shows the mass balances obtained for various locations, each from 4 to 15 foot depth. As can be seen, the mass balance ranges from about 20 to 78%, with an overall average of about 50%. These numbers are judged to be very good considering the uncertainties mentioned above.

Role of Diffusion: Due to the significant removal of TCE in the control area, the effect of diffusion on the removal of TCE from the contaminated soil was modeled. The results obtained are shown in Table 3. The model assumes

- slab configuration of 22 in. thick bracketed with infinite sinks (zero TCE concentration in the carbon wick drain), which is essentially correct since the activated carbon used has very high capacity for TCE (5000 mg/g carbon) compared to the amount of TCE in soil (equivalent to less than 15 mg TCE/g carbon).

- no retardation from soil interaction, a good assumption due to the very low adsorption of TCE (maximum amount adsorbed = 4.5 mg TCE/kg soil)

As can be seen, TCE removal by diffusion after four months is 22.4% at 15°C, 26.5% at 25°C, and 33% at 40°C. These levels are way below the removal levels obtained in the field test. It is interesting that even after 10 years passive diffusion only removes about 70% at 15°C. Larger treatment zone spacings will drastically reduce the amount of TCE removal from passive diffusion. As shown in the table, after 10 years the TCE removal by diffusion alone for a 9 foot spacing is less than 20%.

In summary, the field experiment was conducted for 4 months, covering a region of 15 ft by 10 ft by 15 ft deep. Both electrical and electrokinetic parameters were found to scale up very well. Soil analyses showed very high and uniform removal of TCE, averaging about 98% with most samples exhibiting 99+% removal from the soil. In addition, good TCE material balance was achieved.

Applications/Benefits

Lasagna has the potential of a very broad and effective in-situ technology for soil remediation. It treats the difficult case of contamination in low-permeability or heterogeneous soils. It requires very little maintenance, has built-in flexibility for handling diverse site situations, is practically non-intrusive to the environment and cost competitive. Based on the initial field results, the treatment cost for a typical 1-2 acre site with contamination to a depth of 40-50 feet would be about $50 / yd³. This compares well with other technologies such

as soil mixing (an ex-situ method), soil vapor extraction/bioventing (in-situ method but not effective for clay soils), and excavation/incineration (ex-situ and very high cost). Once developed, the Lasagna technology will have great benefits over existing ones in many aspects including environmental impacts, cost effectiveness, waste generation, treatment flexibility, and breadth of applications.

Future Activities

Phase II development activities are being planned to provide the technical and performance data necessary to advance the technology to the point where it can be commercially utilized as a full scale remediation technique. Phase II will consist of IIa, a commercial-scale development demonstration, and IIb, a full-scale first application demonstration at the approximately 1/2 acre contaminated site at the DOE Gaseous Diffusion Plant in Paducah. Phase II will utilize iron filings as the reagent in the treatment zones for degrading TCE to innocuous products.

Various treatment processes are also being investigated to handle other types of contaminants, DNAPLs situation, heavy metals and mixed wastes. For highly non-polar contaminants, surfactants could be introduced into the circulating water or incorporated into the treatment zones to solubilize the organics. For mixture of organics and metals, the treatment zones can contain sorbents for binding/immobilizing the metals, and microbes or catalysts for degrading the organics.

Acknowledgements

This research addresses the Plumes Focus

Area. METC's COR is Mr. Kelly Pearce and the period of contract performance is September 26, 1994 - November 25, 1995. DOE's Office of Technology Development, Office of Environmental Restoration at PGDP, Battelle Pacific Northwest Laboratories, Oak Ridge National Laboratory/Grand Junction, EPA Remediation Technology Development Forum, DuPont, General Electric, CDM Federal Programs, API Contractors, Nilex Corporation, Stanford University, and Clean Sites Inc. provided critical resources and inputs for the successful completion of these studies.

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12. P.H. Brodsky and S.V. Ho, U.S. Patent No. 5,398,756 entitled *In Situ Remediation of Contaminated Soils*, March 1995. The term Lasagna^{TM} has also been trademarked by Monsanto.
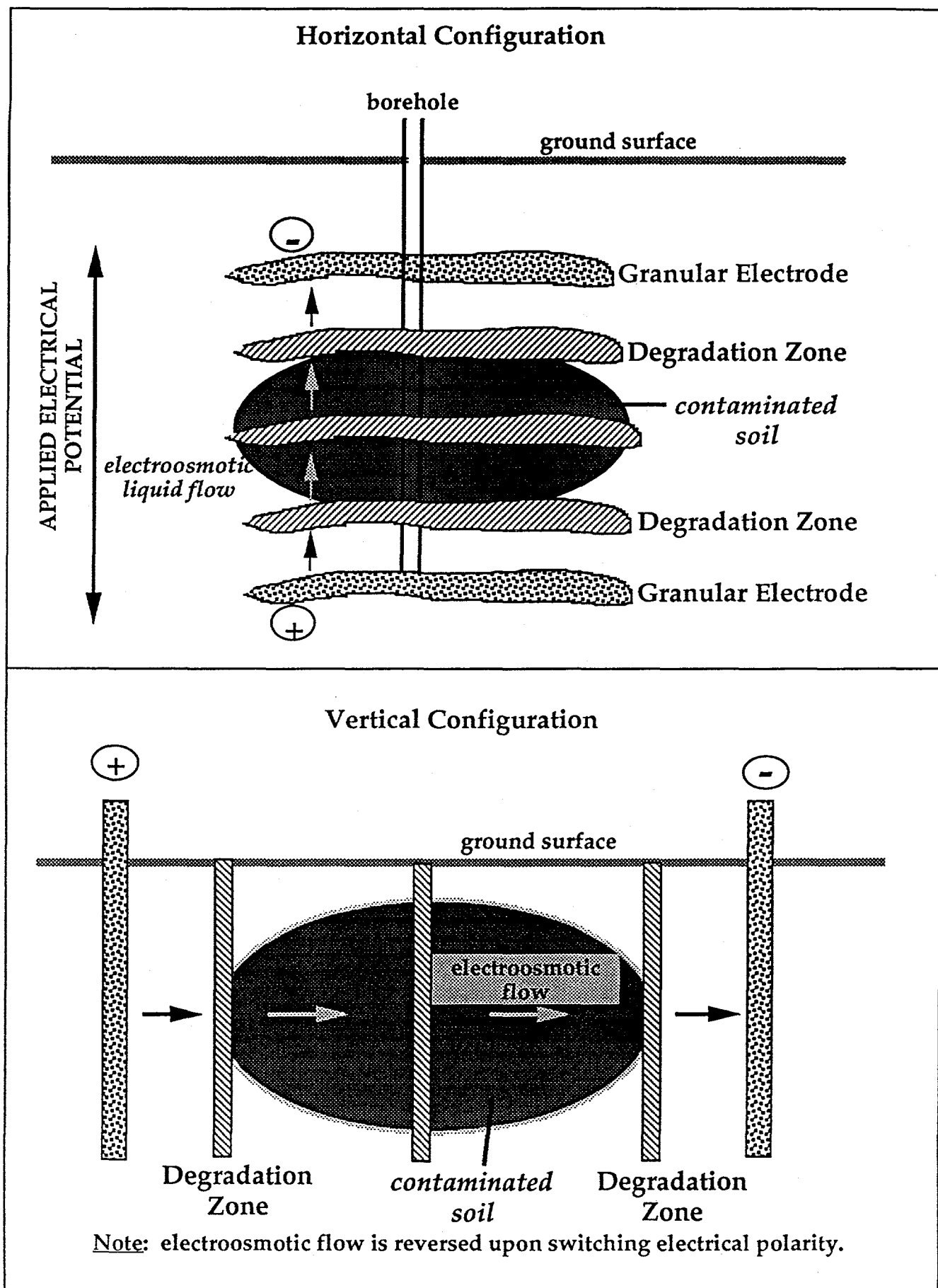
Figure 1. Integrated In-Situ Remediation Process

Horizontal Configuration

borehole

ground surface

APPLIED ELECTRICAL POTENTIAL

Granular Electrode

Degradation Zone

contaminated soil

electroosmotic liquid flow

Degradation Zone

Granular Electrode

Vertical Configuration

ground surface

electroosmotic flow

Degradation Zone

contaminated soil

Degradation Zone

Note: electroosmotic flow is reversed upon switching electrical polarity.

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Figure 2. Electrodes and Treatment Zones Installation Scheme
Phase I Field Test
15' x 10' x 15' deep
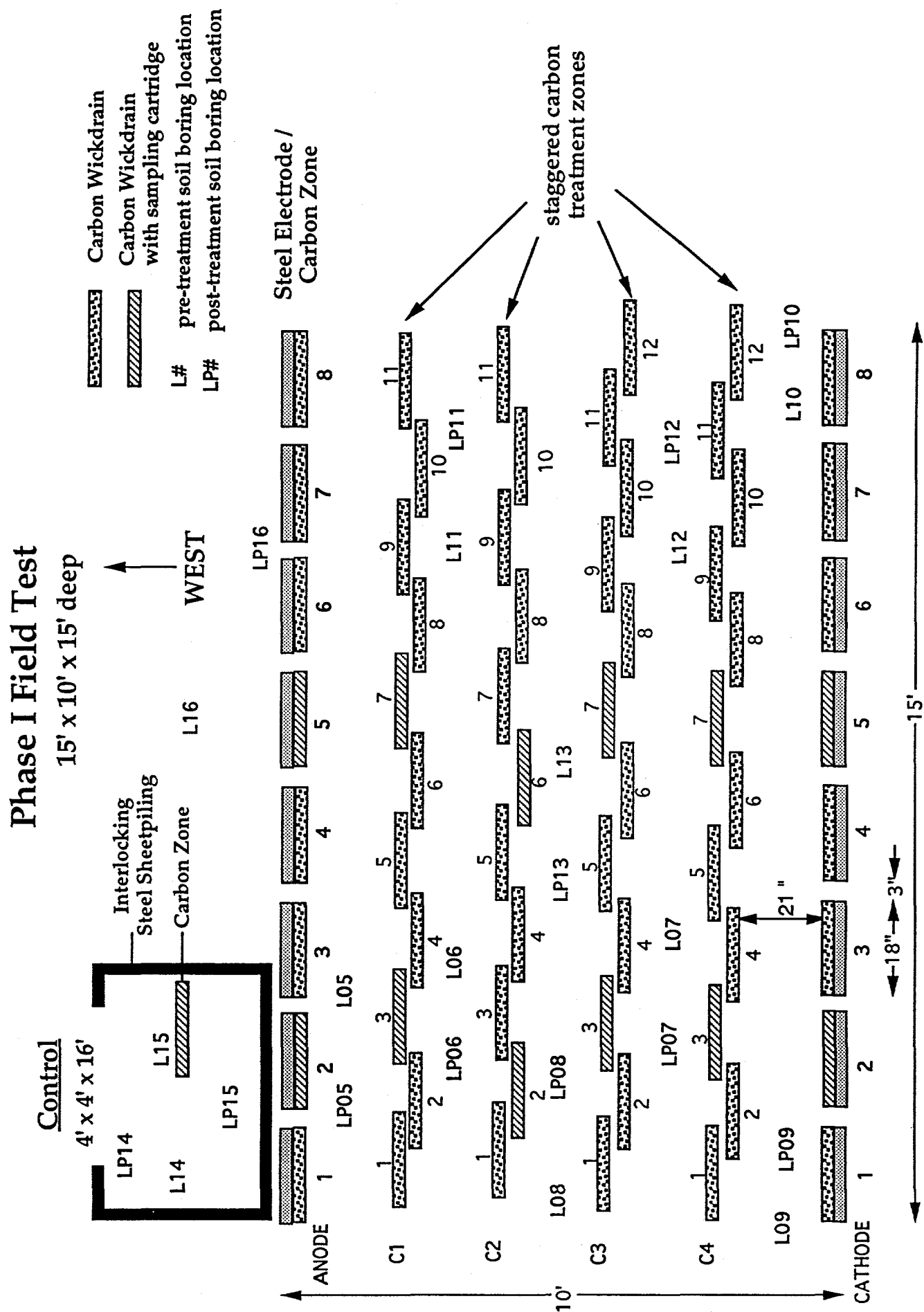
Figure 3. Voltage and Current Trends for Lasagna Field Experiment

Voltage and Current vs Time
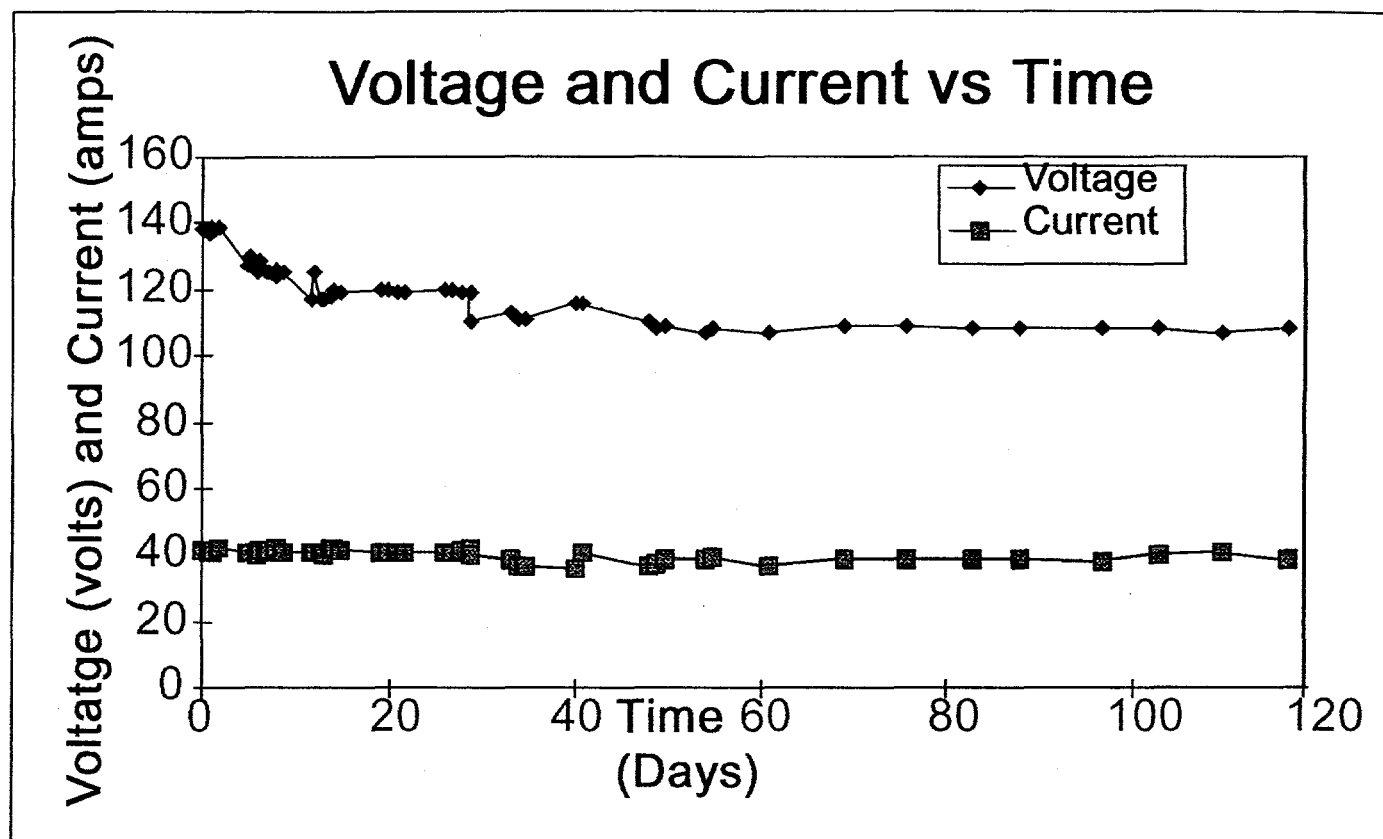
Figure 4. Flow Rate Measurements andAccumulated Pore Volumes
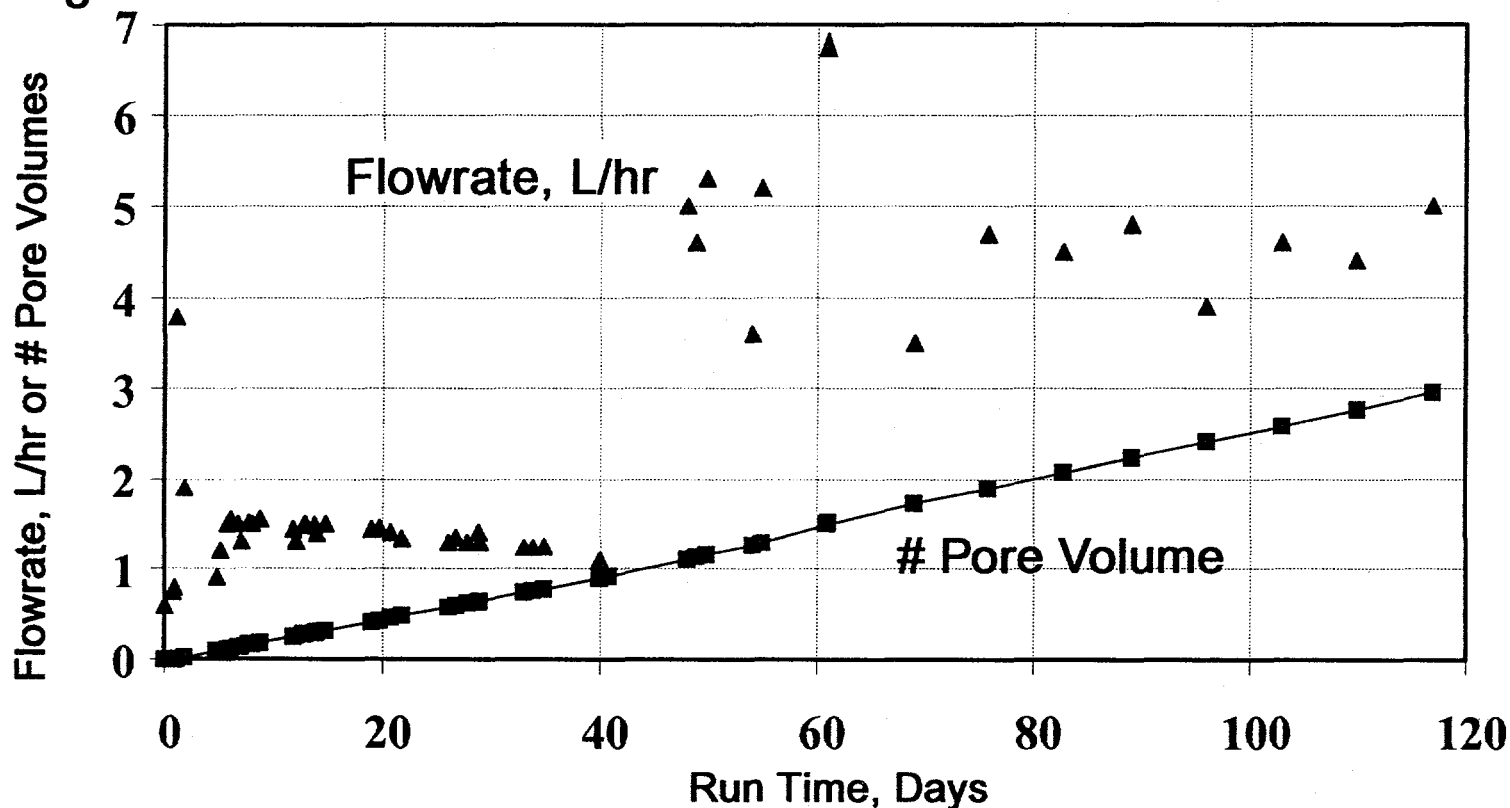
from Intact Core: (ke = 1.2 x 10-5 cm2/v-s)   Est. F = 3.6 L/hr @ 20˚C, 0.40 V/cm
                                                 F = 5.0 L/hr @ 40˚C, 0.37 V/cm

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Figure 5. Conductivity and pH of Anode and Cathode Liquid
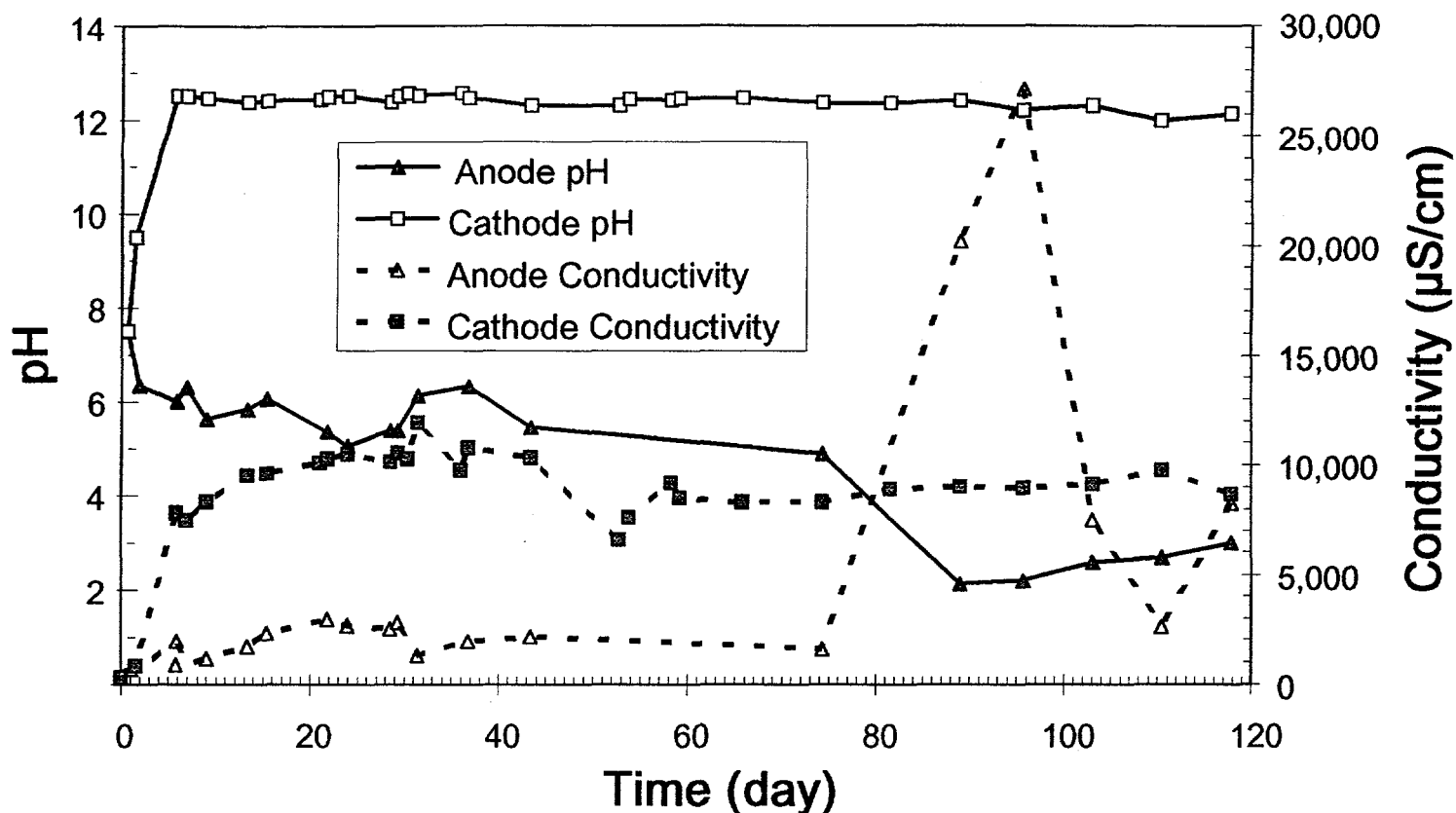
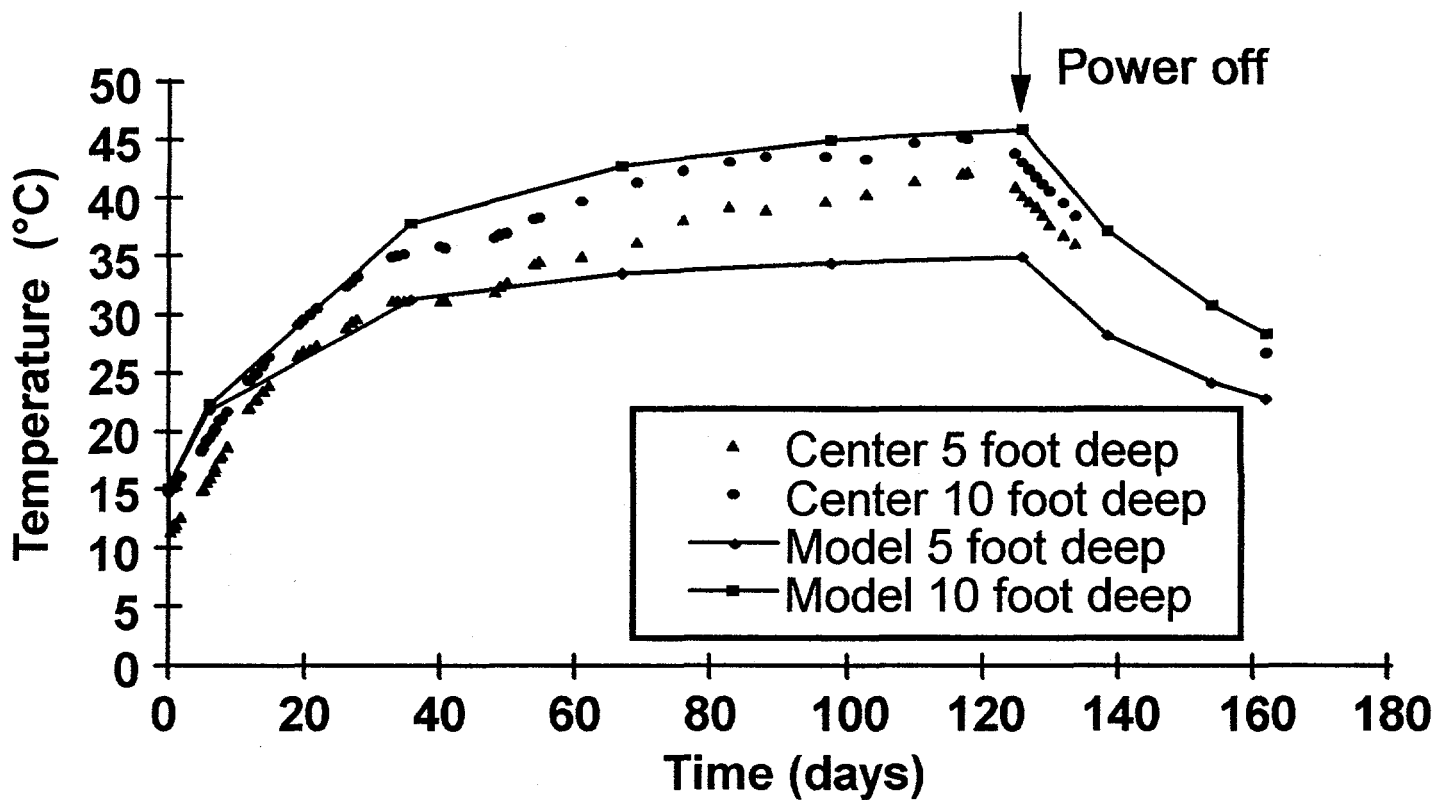# Figure 6. Temperature Trends for Various Locations

13
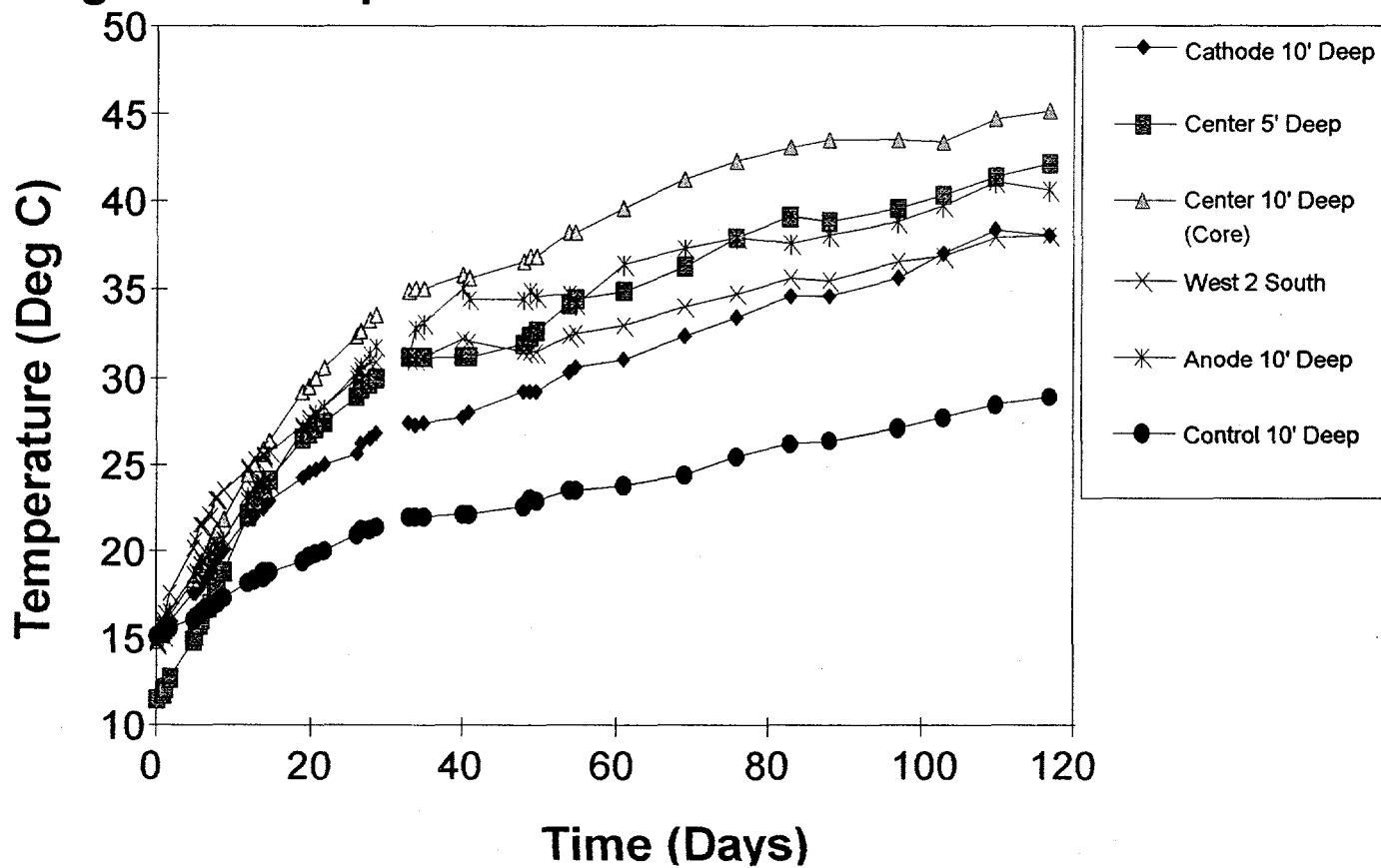
Figure 7. Comparison of Model and Actual Temperatures

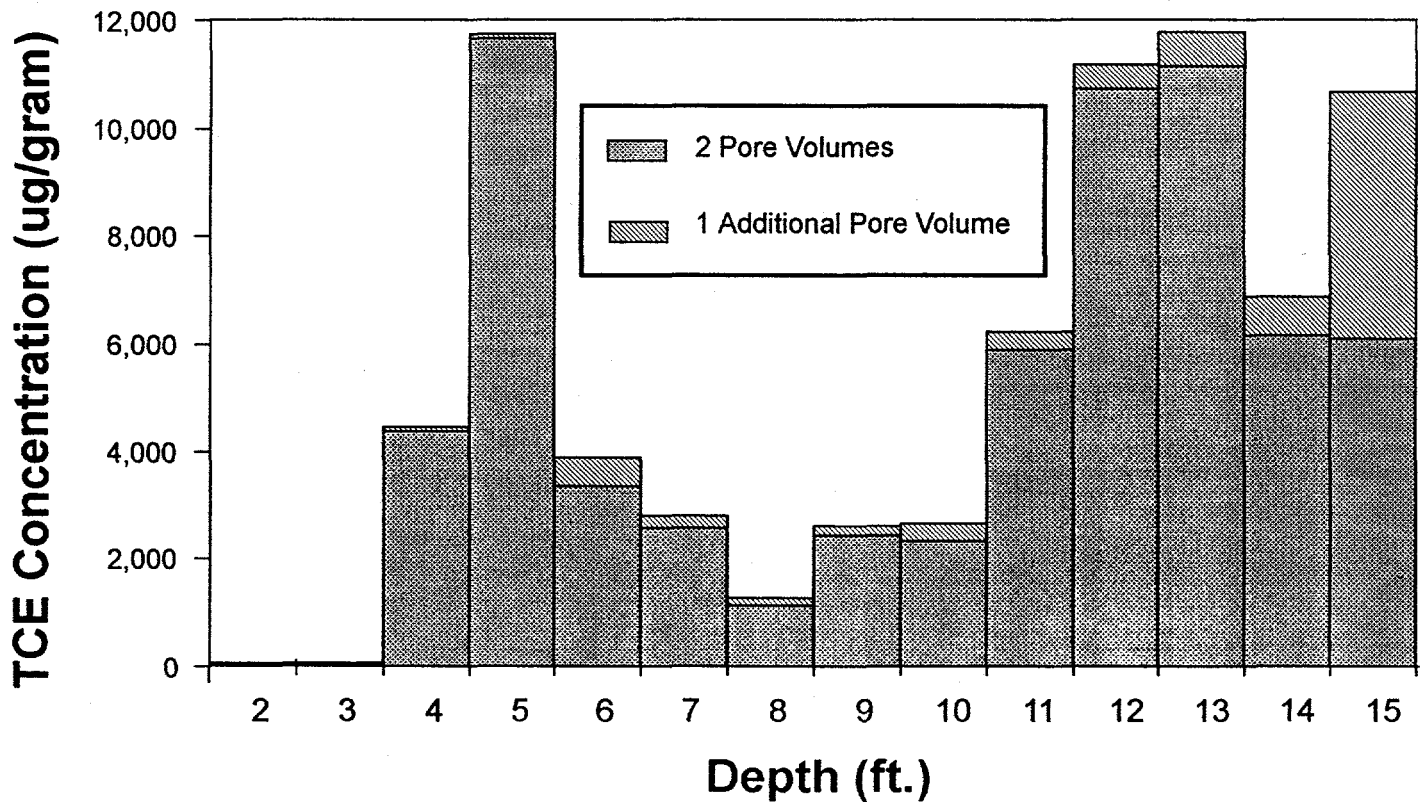# Figure 8. Intermediate Carbon Cassette Sampling
{C4-7} Carbon Cassette

Table 1. TCELevels in Soil Before Experiment

Paducah Drop Test Site (Samples from 12 locations)

	TCE, mg/Kg Soil			Estimated TCE in Water, mg/l
Depth	Minimum	Maximum	Mean	Maximum
4	8.60	61.6	22.8	308.0
5	1.00	63.1	34.5	315.5
6	1.00	79.4	38.3	397.0
7	2.60	114.5	45.1	572.5
8	2.30	79.4	44.4	397.0
9	5.90	129.0	60.3	645.0
10	7.80	138.9	79.5	694.5
11	6.90	165.1	109.7	825.5
12	2.60	292.0	140.6	1460.0
13	4.70	402.5	150.2	2012.5
14	2.10	507.3	151.4	2536.5
15	3.20	237.3	123.2	1186.5
Average	4.06	189.2	83.3	

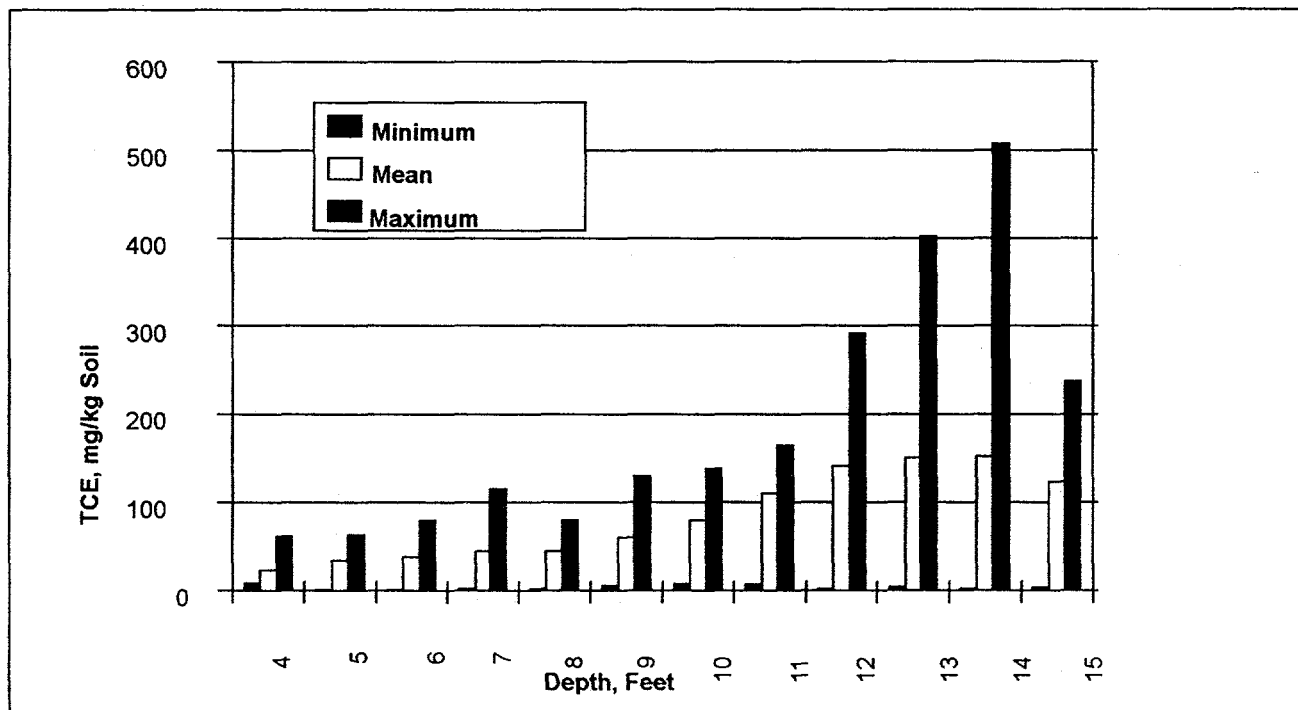TCE Solubility in Water @ 25°C =1100 mg/L

Table 2. TCE Removal Summary Based on Soil Cores

Borehole Number	Bore Holes Within Test Boundary									Avg. in unit	Bore Holes Outside unit		
	L-05	L-06	L-07	L-08	L-09	L-10	L-11	L-12	L-13		L-14	L-15	L-16
Pre-Test Concentration													
High mg/kg	169.4	167.3	176.7	368.6	113.1	7.6	144.3	76.9	140.2		178.2	196.8	114.2
Low mg/kg	20.3	4.9	37.2	3.0	13.7	1.0	4.1	12.0	38.3		10.4	43.7	10.6
Average mg/kg	73.0	82.0	101.6	110.4	66.6	4.0	72.2	44.8	99.1	72.6	108.2	118.6	63.0
Post-Test Concentration													
High mg/kg	0.29	1.22	8.62	17.40	15.20	0.47	1.84	5.23	2.29		101.7	109.5	50.8
Low mg/kg	0.004	0.21	0.055	0.018	0.002	0.042	0.40	0.002	0.038		24.6	6.7	2.6
Average mg/kg	0.14	0.50	1.06	2.07	3.73	0.30	0.89	0.89	0.64	1.14	80.0	58.0	20.1
TCE Removal %	99.8	99.4	99.0	98.1	94.4	92.4	98.8	98.0	99.4	98.4	26.1	51.1	68.1

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Table 3.   Model of TCE Removal by Diffusion in Paducah Soil

Model assumptions & parameters:
- diffusion in slab
- carbon treatment zones as infinite sinks
- no retardation of TCE due to interaction with the soil matrix
- $\varepsilon = 0.4$ (soil porosity); $\tau = 3$ (soil tortuosity)
- $D_{TCE} = 1.03 \times 10^{-5}$ cm^2/s at 25°C

Carbon Zone | Paducah Soil Ave. TCE = 85 mg/kg | Carbon Zone
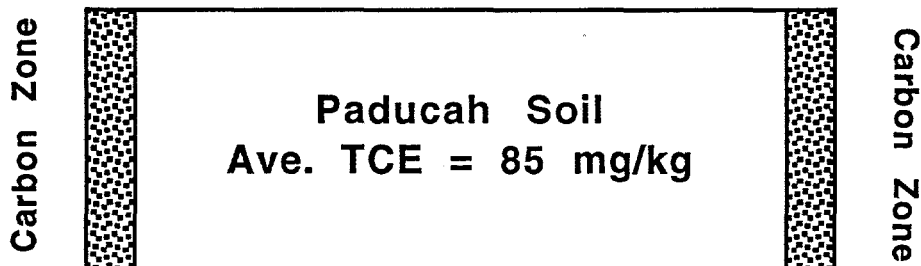
Carbon Zones Spacing = 22"

3. a	Percent TCE Removal at Indicated Soil Temperature			
Exposure Time	15°C	25°C	40°C	90°C
1 week	7.1	7.3	7.7	9.3
4 months	13.3	15.4	19.0	31.5
1 yr	22.4	26.5	33.0	54.8
5 yrs	49,9	59.0	71.7	95.7
10 yrs	69.2	79.3	90.1	99.8

3.b	%TCE Removal at 25°C	
Exposure Time	3-ft carbon zone spacing	9-ft carbon zone spacing
1 week	6.9	6.7
4 months	10.4	7.1
1 yr	16.4	8.0
5 yrs	36.3	12.6
10 yrs	51.2	17.2

Table 4. TCE Mass Balance

Carbon Wick Number	Wick TCE Concentration (mg/kg)	Wick TCE Mass (grams)	Pre-test Soil TCE Concentration (mg/kg)	Pre-test Soil TCE Mass (grams)	Post-test Soil TCE Concentration (mg/kg)	Post-test Soil TCE Mass (grams)	TCE Mass Removed From Soil (grams)	TCE Material Balance (%)
C1-3	1694	10.0	95.5	52.1	0.14	0.074	52.0	19.4%
C1-7	1861	11.0	90.3	49.2	0.14	0.074	49.1	22.5%
C2-2	4273	25.2	121.1	66.0	0.50	0.27	65.7	38.6%
C2-6	4239	25.0	91.2	49.7	0.89	0.49	49.2	51.3%
C3-3	4757	28.1	106.8	58.2	2.10	1.1	57.1	50.2%
C3-7	6788	40.1	95.8	52.2	0.64	0.35	51.9	77.5%
C4-3	2239	13.2	106.6	58.1	1.10	0.58	57.5	23.7%
C4-7	5862	34.6	87.8	47.9	0.89	0.48	47.4	73.3%
Cathode-2	5652	33.4	99.4	54.2	3.70	2.0	52.2	65.3%
Cathode-5	6005	35.4	84.3	46.0	0.30	0.16	45.8	77.4%
Overall Average	**4337**	**25.6**	**97.9**	**53.4**	**1.04**	**0.558**	**52.8**	**49.0%**

TCE soil mass (grams) is figured using 6 inch long by 21 inch wide by 11 feet deep zone of soil in front (up-stream) of the 6 inch wide sampling cassette and the average TCE concentration based on the pre-test soil core kriged data.