Cleaning Up Underground Contaminants

Dynamic underground stripping combines steam and electrical heating of underground soils with vacuum extraction of vapors and fluids, guiding these processes by real-time monitoring methods.

AT hundreds of industrial and government sites across the United States, environmental consulting firms are designing permanent containment systems for underground contaminants such as hydrocarbon fuels, cleaning solvents, and industrial chemicals. In quantities of thousands of liters or more, these chemicals threaten to contaminate drinking water supplies for hundreds of years. Typical containment systems (e.g., deep walls of cement or clay, or hydraulic pumping to control groundwater movement) can keep the chemicals from further contaminating groundwater if they are properly maintained for many years, but they do not remove the contaminants. Clearly, removing the contaminants from the soil is a much preferable solution than containing them and attempting to prevent their spread.

In a fairly typical example of this problem, between 1952 and 1979, tens of thousands of liters of gasoline leaked from an underground tank at the former LLNL filling station. The amount of gasoline leaked is not well known. Estimates made using data from borehole core analysis placed the amount at about 30,000 liters; discrepancies in inventory logs from the 1970s gave the upper limit at 70,000 liters. In the 1970s, agricultural pumping in the vicinity ceased and the water table rose, trapping gasoline below it and "smearing" the gasoline through clay-rich soils of low permeability. (See the box on p. 14 for a description of the water table.) Figure 1 shows a diagram of this contaminated region.

Hydrocarbons trapped below the water table are especially difficult to clean up by traditional methods. The pump-and-treat method—now in use at some 300 Superfund cleanup sites—requires the pumping of huge amounts of groundwater up through an extraction well, followed by removal of whatever captured contaminant comes up with the water. Unfortunately, the pumped water carries very little contaminant—in the case of gasoline, no more than 10 liters of contaminant per million liters of water. We estimated that removing the free-product gasoline from the LLNL spill site by the pump-and-treat method alone could take up to 200 years. (See the box on p. 15 for a description of free-product gasoline.)

Instead of using the pump-and-treat method, a team of researchers from LLNL and from the University of California (UC) at Berkeley demonstrated a new and unique combination of technologies—collectively called dynamic underground stripping—to rapidly remove some 29,000 liters of free-product gasoline from beneath the LLNL site. As shown in Figure 2, our experiment targeted a portion of the LLNL spill area, which contained the majority of the free-product gasoline. Pools of free-product gasoline are known to lie outside the targeted area, as indicated by the circles in Figure 2, and a small fraction (much less than 1%) of the gasoline is known to have dissolved into the groundwater outside this region. The purpose of our demonstration was not to perform an entire cleanup of the spill area but to prove that dynamic underground stripping is successful in removing the free-product gasoline. This first demonstration of the new method on an actual spill site accomplished in less than a year what the conventional method would have taken decades to do. (In 1991, we tested the dynamic underground stripping technique on an uncontaminated underground site.[1])

Dynamic Underground Stripping: A General Description

In dynamic underground stripping, a targeted volume of earth is heated to vaporize the trapped contaminants. Two methods of heating—steam injection and electrical resistance—are used to heat all layers in the soil. Permeable layers (e.g., gravels) are amenable to heating by steam injection, and impermeable layers (e.g., clays)

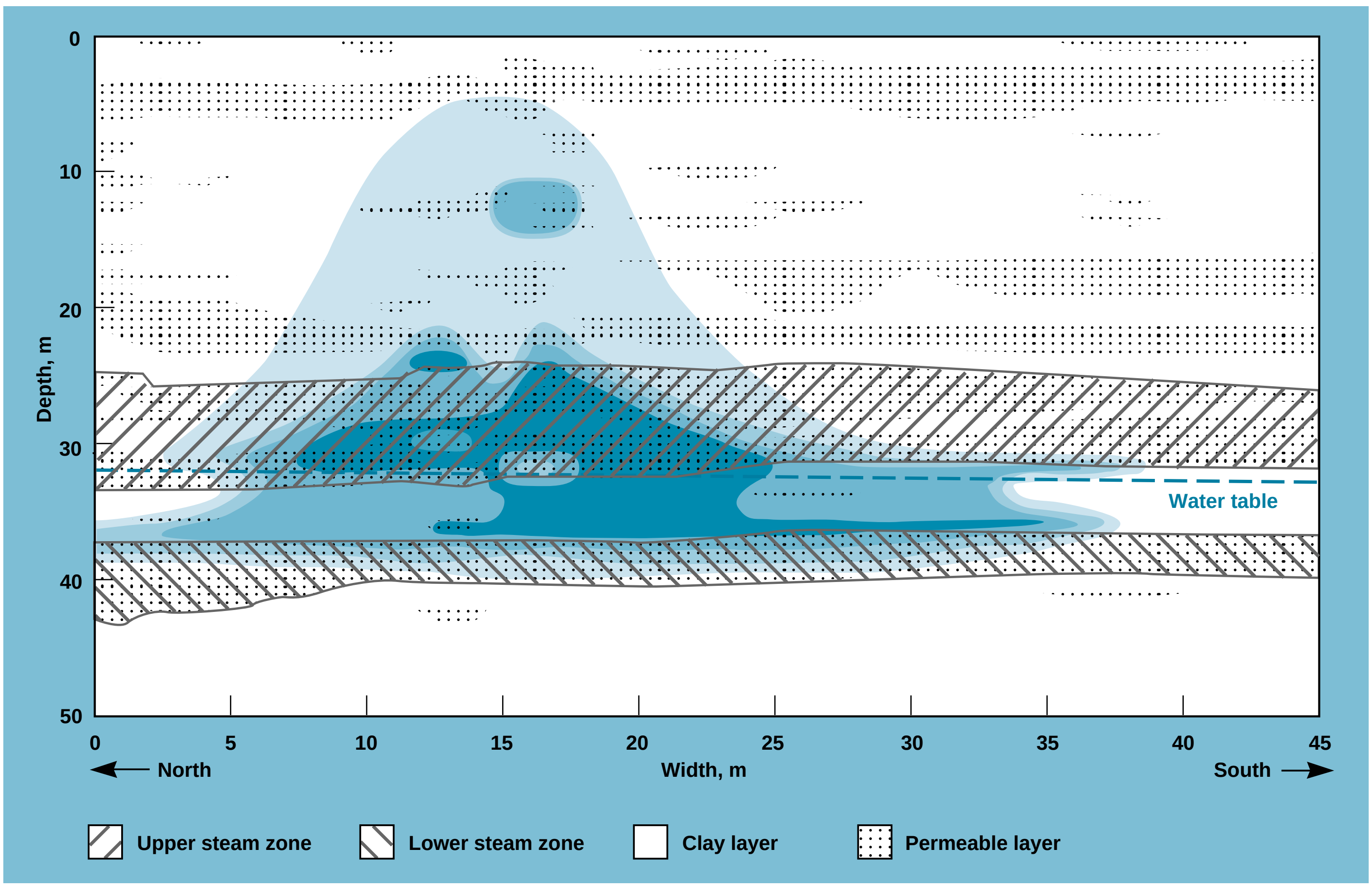


Figure 1. Cross section showing an approximation of the gasoline contamination at the treatment site before dynamic underground stripping began. The darker areas represent higher concentrations or free-product gasoline. The dashed line denotes the level of the water table.

can be heated by electric current. Because of these complementary heating techniques, dynamic underground stripping is the best technique available to treat heterogeneous soils. Once vaporized, the contaminants are removed by vacuum extraction. All these processes—from the heating of the soil to the removal of the contaminated vapor—are monitored and guided by underground imaging.

Researchers from UC Berkeley, led by Professor Kent Udell of the Mechanical Engineering Department, developed the powerful steam-injection technique by combining vacuum extraction with a steam-injection method that oil companies sometimes use in late stages of oil recovery. LLNL researchers developed the electrical resistance method of heating and an underground imaging technique—electrical resistance tomography.

Steam Injection and Vacuum Extraction

Injection wells drilled around an area of concentrated contamination are used to supply both steam and electric current. Extraction wells placed as close as possible to the center of the contamination are used to extract the contaminant. The steam is pumped in through the injection wells and advances in a wall, or front, toward the extraction wells. Concurrently, groundwater is pumped and vapor is extracted from the extraction wells. As the steam front advances, the permeable soils are heated to the boiling point of water (100°C), and volatile organic contaminants are vaporized from the hot soil. After the steam front reaches the extraction wells, steam injection is stopped; vacuum continues to be applied at the extraction wells. The lowered vapor pressure (resulting from the applied vacuum) forces the contaminants to boil, and the concentrated, contaminant-carrying vapor is then pumped to the surface and treated. When the steam zone collapses, groundwater reenters the treatment zone. The steam injection/vacuum extraction cycle is repeated, and additional contaminants are vaporized and removed.

Electrical Resistance Heating

Electric current is used to heat the impermeable soils. It operates on the same principle that makes a heating coil work—heat builds up in a conductor that resists current flow. For this technique, the clay itself supplies the resistance. In the steam injection wells, electrodes are sunk into the ground. Each electrode supplies several hundred amperes of current at up to 600 V, heating the impermeable clays. Water and contaminants trapped in these (relatively) conductive regions are vaporized and forced into the steam zone for vacuum extraction.

These combined heating processes achieve a hot, dry zone surrounded by cool, damp, untreated areas. Electrical heating and steam injection are repeated as long as underground imaging shows that cool (and therefore untreated) regions remain.

Underground Imaging and Monitoring

Several geophysical techniques are used to monitor the underground movement of steam and the progress of heating, including temperature measurements, electrical resistance tomography, and tiltmeters.

Temperature measurements made in monitoring wells throughout the treatment area reveal details of the complex heating phenomena in the alternating gravel and clay layers.

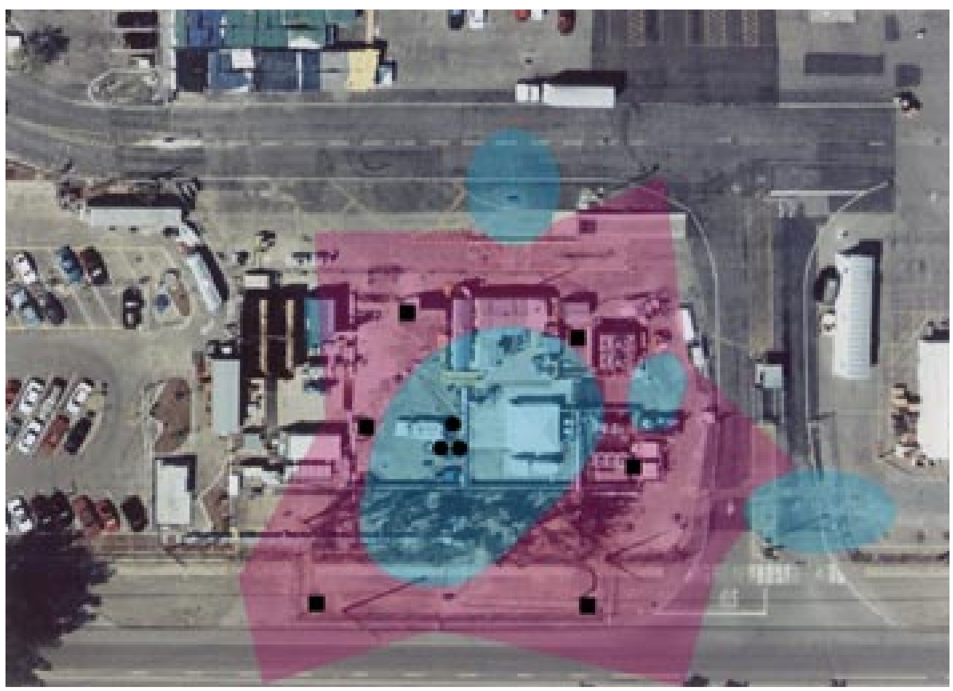


Figure 2. Top view of LLNL spill area, showing regions of known or suspected free-product gasoline contamination (circled in blue) and where we applied dynamic underground stripping (in pink). As noted by the circles outside the treatment area, additional free-product gasoline remains at the site.

These measurements are difficult to make in areas of high thermal gradients, such as in a steam flood. UC Berkeley researchers developed an optical temperature-logging system that can provide detailed daily borehole-temperature logs. In addition, thermocouples permanently installed in boreholes throughout the treatment area record temperatures around the clock.

Electrical resistance tomography provides near-real-time images of the underground processes and permits the identification of areas that are affected by the dynamic stripping process. Because electrical conductivity varies with temperature, measuring the resistance of the soil can reveal the progress of the steam front and the heated zones. Electrical measurements thus provide good measurements of steam movement and reveal changes in formation properties over a broad zone. Because the electrical properties of the soil are controlled by soil type, fluid saturation, and chemistry, electrical resistance tomography is also useful for characterizing a given site and for predicting steam pathways.

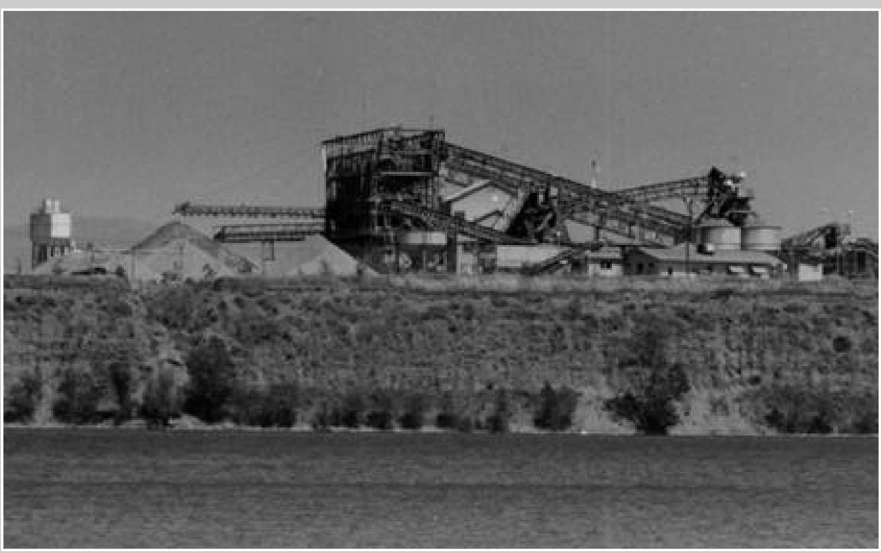
Tiltmeters are used to track the movement of the steam front. These devices are capable of detecting very small angular deformations in the ground surface that result from subsurface pressure changes, such as those that occur with the movement of the underground steam front. They are sensitive enough to detect pressure changes as small as a few hundred pascals (~0.05 psi) in the heated zone.

The Water Table

A child digging a hole at the beach will eventually reach a depth where the hole fills with water despite all efforts to bail it out. This is an early experience with the "water table"—the depth at which all the space between the medium (in this case, the sand grains) is water-filled. At any point on Earth, one can dig a hole deep enough to encounter such standing water.

The depth of the water table varies greatly from region to region. On the south side of the LLNL site (the location of the gasoline spill), the depth of the water table is about 30 meters; in more arid regions, the water table can be more than ten times as deep. During periods of rain and drought, the water table might rise and fall, respectively. Also, in areas where water is pumped for agricultural reasons, the level of the water table might change, as occurred at the LLNL site in the 1970s when agricultural pumping in the region ceased.

Not all holes make good wells, however. At the site of the LLNL gasoline spill, for instance, a 35-meter-deep hole could end in a fine-grained, clay-rich soil in which the water would not flow—but only seep. It could also end in a gravely soil; a hole in this medium would fill rapidly and would not be emptied even by pumping the water out at a rate of 100 liters/minute.



At Shadow Cliffs Regional Park—an old gravel quarry near LLNL—the local water table forms the surface of the lake.

Demonstration of Dynamic Underground Stripping at a Contaminated Site

In this first application of dynamic underground stripping at a contaminated site, our goals were to:

- Determine how well the process removes gasoline.
- Determine how well the monitoring methods can be used to control heat input and map heated zones.
- Determine whether there are any deleterious effects with this process (such as dispersal of contaminant).
- Determine how the several components of this technique can be varied in relation to one another to maximize extraction efficiency.
- Demonstrate the engineering and operational practices required for safe and effective operation of this cleanup technique.

Site Characterization

The soils at the gasoline spill site are alluvial, ranging from very fine silt and clay layers to coarse gravels, with permeabilities ranging over several orders of magnitude. There are two principal permeable zones: one above and one below the water table.

Our aim was to remove all the free product gasoline at the treatment area (see Figure 2). Approximately half of this gasoline was above the water table (at a depth of 30 m) and half was trapped below. The treatment zone was in the shape of a distorted cylinder about 40 m in diameter and extending from a depth of 20 to 45 m.

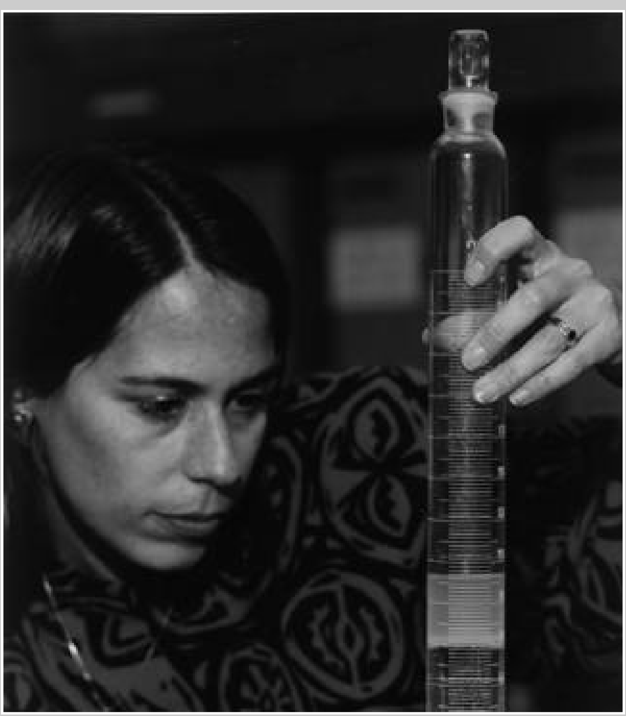
We drilled six injection wells around the spill perimeter to deliver both steam and electric current. Three extraction wells were drilled close to the center of the spill site.

Electrical Preheating

In November 1992, we began the electric preheating of the site. Electricity preferentially flows in areas of high conductivity; the hotter the soil, the higher the conductivity. Initially, the clays are much more conductive than the gravels. Our preheating ensured that the conductivity in the clay-rich zones would remain higher than in the gravel zones even after they were elevated to steam temperatures. Had we not taken this step and simply started with steam heating, the gravel would have been more conductive than the clay, electricity would have flowed into the gravel, and the clay would not have been heated. In November and December 1992, the electrical heating system operated at a maximum power of 800 kW, heating the clay layers in some areas to temperatures exceeding 70°C.

First Steam Pass

We began injecting steam into the lower of two permeable layers in early February 1993. For 37 days, a gas-fired boiler of ~8 MW put out 11,000 kg/h (190 liters/min) of steam. The spreading steam rapidly heated the permeable layers to the boiling point of water, and, within just 12 days, the steam front reached the extraction wells. A small fraction (about 15%) of the free-product gasoline was pushed ahead of the steam front and recovered as liquid; most of the gasoline was removed as vapor after the steam zone was fully established. Prior to the experiment, we did not know that the fraction of vapor would be so high. The amount of water and gasoline vapor removed during this phase (~6400 liters) was limited by the capacity of the vapor treatment system (~95 liters/day); subsequently, the vapor treatment system was redesigned to increase its capacity. Figure 3 shows the daily and cumulative volumes of gasoline removed during this and the other two extraction phases of the project.



Free-Product Gasoline

When large amounts of gasoline contact water, most of the gasoline remains as a separate liquid (see photo to the left). Often called “free product” because it is independent of the water, this gasoline can exist in soils as droplets, coatings on soil particles, or pools of underground gasoline. Because gasoline does not dissolve readily in water, it is very difficult to remove free product by pumping water. It can be pumped out as a separate liquid when there are large underground pools of gasoline, but some gasoline remains trapped in small spaces between soil particles. Held there by the strong forces of surface tension, this trapped gasoline can only be removed by dissolving it in thousands of times its volume of water or by boiling it away, as is done with dynamic underground stripping.

Robin Newmark, a key researcher on the dynamic underground stripping project, holds a sample of free-product gasoline and water recovered from the LLNL treatment area. The yellow gasoline floats on the heavier water, with a salad-oil-like emulsion—a mixture of droplets of both—between. The water below is tainted by dissolved gasoline (about 1 part gasoline per 1000 parts water).

Second Steam Pass

After a three-month shutdown, during which we upgraded the effluent treatment facility and improved the in-process sampling and analyses, we began the second steam pass, which ran from May into July 1993. During this pass, we intended to establish better control of the effluent stream by applying the knowledge gained during the first steam pass. We also wanted to explore ways of increasing the cost effectiveness of the dynamic stripping process.

Extraction rates were high at the beginning of the second pass because the residual heat in the soil had vaporized the gasoline during the shutdown period. To maximize the extraction rate during this second pass, we increased the amount of time the treatment zone was kept under vacuum. We also used a pulsed mode of operation, alternating steam injection and vacuum-only phases on a five- to six-day cycle. We found that the extraction rate varied considerably depending on the amount of steam injected and the total vacuum applied; more gasoline was extracted when steam was not being injected and thus the vacuum effect was greater. During this pass, the average extraction rate was more than 380 liters/day of gasoline (compared to 3 liters/day for pump-and-treat).

The effluent from the extraction wells was first directed to heat exchangers. Most of the gasoline was removed as vapor, and some of the gasoline vapor was condensed along with a large amount of water in the heat exchanger. A gasoline–water separator allowed us to measure the volume of the condensed gasoline. The stream of gasoline vapor out of the heat exchanger was used to help power two internal combustion engines that created the vacuum for extraction. Figure 4 shows the vapor extraction and treatment system.

At the end of this second steam-injection phase, we drilled six boreholes across the treated site from which we made temperature measurements and took core samples for analysis. We found that most of the soil within the treatment volume was heated to the boiling point of water. Only a thick clay layer at 30 to 34 m was not, having reached only 80°C in places. This “cold spot” was where the largest concentration of free-product gasoline remained, an estimated 3000 liters. Recovered soil samples revealed that free-product gasoline had been removed from the edges of the spill and from the zone above the water table. They also revealed that gasoline concentrations had not increased in the soil outside the treatment volume, a very important finding because we wanted to demonstrate that our method did not spread the contaminant. Figure 5 shows a geologist’s interpretation of the data from the six boreholes in the treatment area.

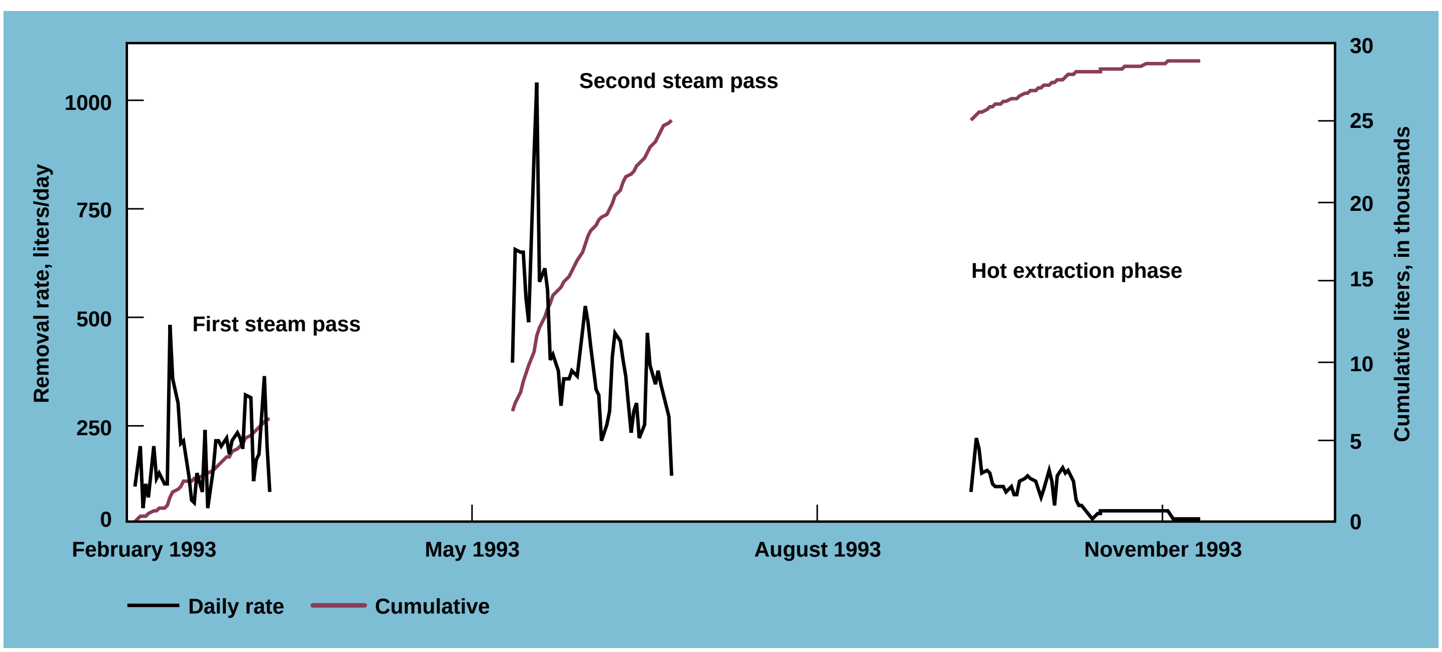


Figure 3. Daily extraction rates and cumulative volumes of extracted free-product gasoline show the three extraction phases. Extraction rates were highest during the second phase, when extraction systems were optimized and we used a pulsed mode of operation, alternating steam injection and vacuum-only phases.

Continued Hot Extraction Phase

After completing the initial experimental phase in July, we resumed extracting groundwater and vapor in October 1993 as part of the ongoing LLNL site cleanup. The initial spike in extraction rates at this time was smaller than after the first pass. In November 1993, we applied electric heating to the area. The overall temperature of the treated zone rose only slightly because the extraction systems were removing much of the deposited electrical energy. For this heating phase, we added four new, long electrodes; we are in the

Figure 4. View of the vapor-extraction treatment system. The internal combustion engines inside the trailer create the underground vacuum; the engines run directly off gasoline vapors pulled from the extraction wells.

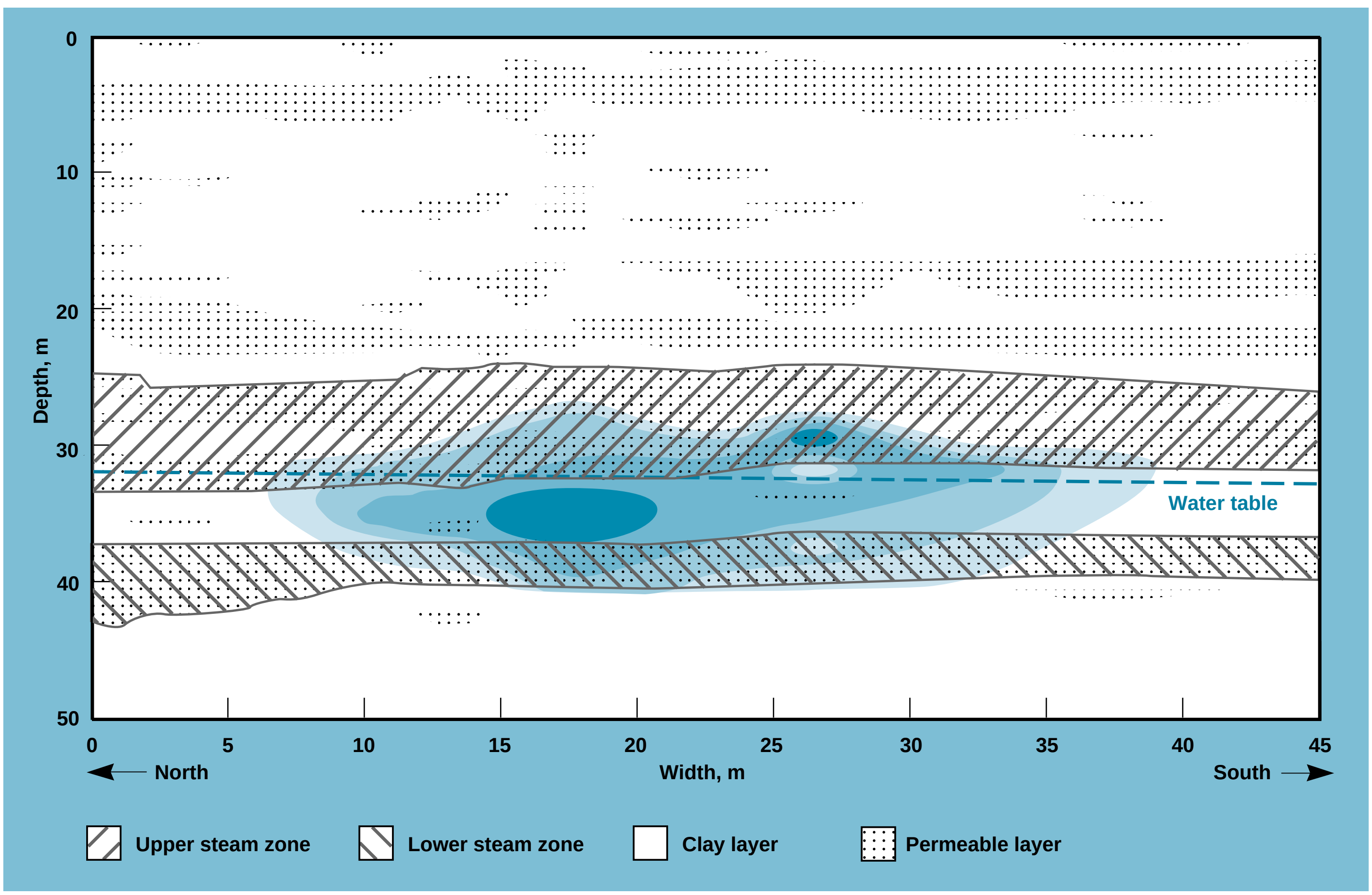


Figure 5. Approximate cross section of the treatment site after the two steam passes (compare with Figure 1). The area of gasoline contamination has contracted greatly.

process of modeling the effect these electrodes had on the process. We turned off all heating and extraction systems in mid-December 1993.

When we resumed groundwater pumping and vapor extraction in January 1994, gasoline concentrations in the recovered groundwater had decreased and the gasoline vapor concentrations increased only slightly, suggesting that no significant amount of free-product gasoline remained to be volatilized (unlike after the previous shutdown periods). Benzene concentrations in the extraction wells were less than 200 ppb, down from their peak of 7000 ppb before the first steam pass. (Benzene is the component in gasoline that is most closely regulated and thus is the chemical that we monitor to determine whether or not the cleaned-up site is within regulatory limits.) Similarly dramatic decreases in benzene concentrations were measured in the monitoring wells, from several thousand parts per billion before stripping to less than 300 ppb in January 1994. Although the site is not legally cleaned up—the regulatory limit for benzene is about 1 ppb—we have reduced the concentrations of free-product gasoline such that over a period of years microorganisms may degrade the remaining gasoline at the treatment area.

This last extraction phase removed about 3800 liters of gasoline, for a total of at least 29,000 liters. We believe that no significant free-product gasoline remains in the treatment zone (although this can only be confirmed by future drilling). We estimate that this cleanup procedure decreased the amount of gasoline in the treatment area by roughly 100 times. Sampling and analysis of cores from new boreholes must still be done to assess the cleanup/contamination status of this area.

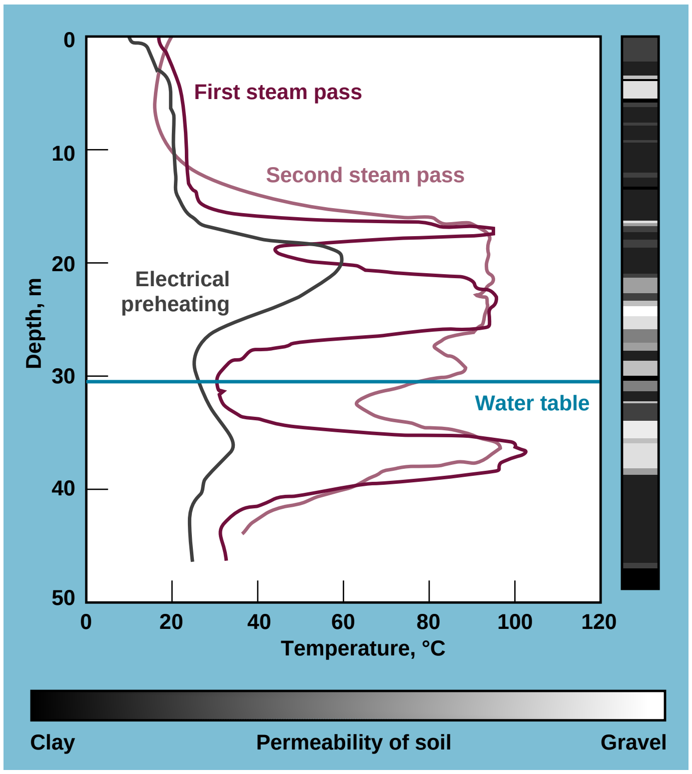


Figure 6. Temperature logs from a monitoring well inside the ring of injection wells. These logs show electrical heating of the clay-rich layers during the preheat, steam passing through the most permeable layers during the first steam pass, and conductive heating, and later penetration, by steam into less permeable layers during the second steam pass.

Monitoring and Controlling the Cleanup Process

Day-to-day monitoring of the dynamic stripping process assured that we were injecting enough steam to drive contaminant to the center of the treatment zone without driving too much steam (and, perhaps, contaminant) outside the zone. For example, we had agreed not to drive steam under the Sandia–California site, adjacent to LLNL. Such a need for rigorous containment would likely occur in other applications.

Because of variations in local geology within the treatment zone, each of the 12 injection ports (two each in six wells) injected a different amount of steam at a given pressure, ranging from 300 kg/h to the entire output of the boiler. Although such a range in steam injection rates is expected in such a geologically heterogeneous site, it requires that the location and size of the underground steam zones be measured *in situ*.

The 11 monitoring/imaging wells housed fixed thermocouples and infrared sensor systems from which were generated continuous temperature logs. Figure 6 shows the temperature logs at one monitoring well for the electrical preheating phase and the two steam passes. These temperature logs provided the most detailed local measurements of the vertical distribution of the steam; for example, they revealed temperature gradients during the first steam pass of up to 80°C over just a meter or so in depth.

Between the wells, electrical resistance tomography, supported by temperature logs, mapped the progress of the steam and heating fronts rapidly and accurately at resolutions of 1 to 2 m. Electrical resistance tomography provided images of the heated zones by comparing the electrical resistance distribution before and during heating. These images revealed several areas where steam was moving vertically in the treatment zone, which had not been detected by the borehole temperature logs or inferred from geological interpretations.

The speed of electrical resistance tomography made it the principal method for monitoring the dynamic stripping process. Data were obtained and analyzed in less than a day and were used to set the steam-injection rates for the next day. Figure 7 shows images that reveal the movement of the steam front. The placement of the electrical resistance tomography/temperature wells allowed good monitoring of the interior of the treatment zone (extending about 9 m outside the ring of steam injection wells) and lower-resolution monitoring of the surrounding area.

Surface-implanted tiltmeters—arranged in a larger array—monitored the full extent of the steam zone outside the treated area (Figure 8). These devices provided maps of the areal extent of the steam zone emanating from each well, particularly for the zone below the water table. They were extremely effective in mapping the lateral spread of the steam and the development of any preferential steam pathways.

Results of the Demonstration

By late 1993, dynamic underground stripping had removed about 29,000 liters of gasoline from the treatment site. We treated a volume of earth of about 80,000 m^3 between 20 and 45 m in depth. The maximum gasoline extraction rate was 950 liters/day. At the surface, about one-third of the fuel was condensed for recycling.

We believe that we removed virtually all the free-product gasoline from the treated area. After dynamic stripping has removed the bulk of the contamination, other methods of soil and groundwater cleanup (including pump-and-treat and bioremediation) can be used to remove the remainder.

This demonstration clearly showed that dynamic underground stripping quickly removes the concentrated, free-product contaminants, preventing them from continuously leaching into the soil. In larger-scale operations, such as removal of million-liter spills from refineries, the ability to recover usable fuel or solvents will be quite valuable.

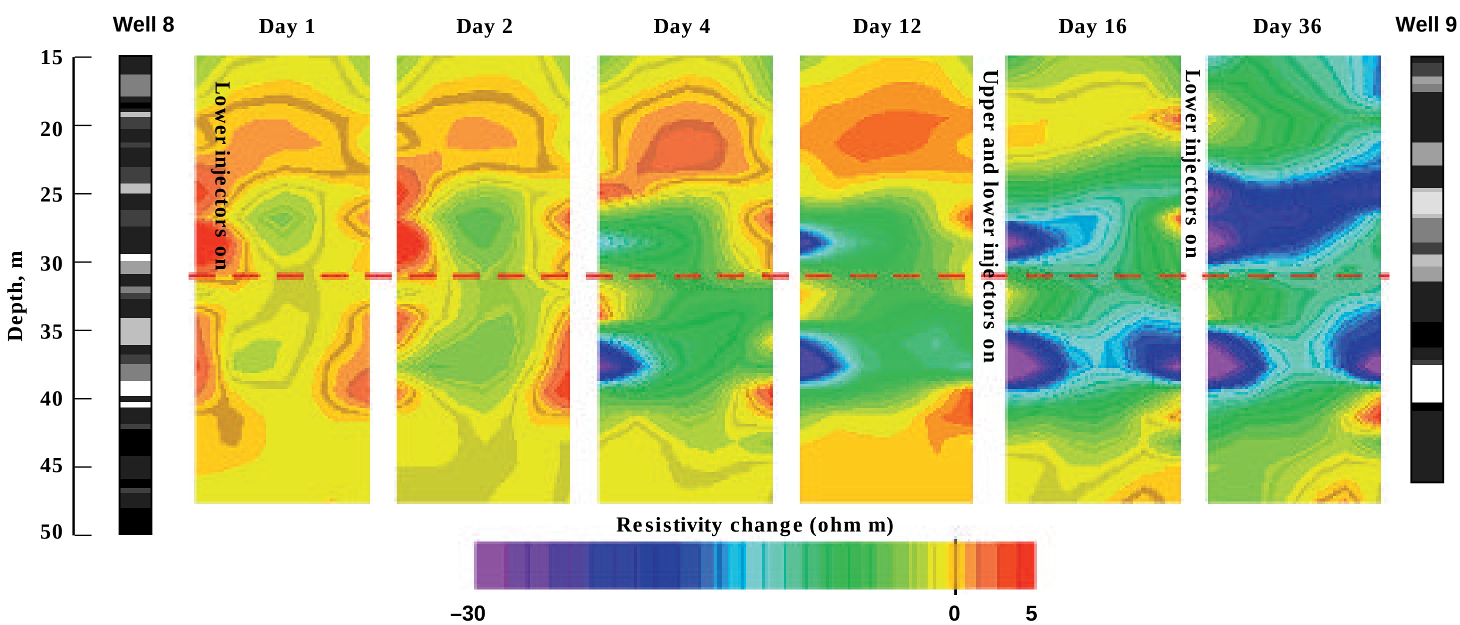


Figure 7. Electrical resistance tomography images show the passage of steam between two monitoring wells, starting from the first day of steam injection. The images compare initial baseline data with data taken during steaming. The steam zone appears as a zone of lower electrical resistivity (green, blue, and violet) passing across the image plane. This image plane is located about 6 m from the nearest injection well and is nearly perpendicular to a line linking it and the extraction wells. Small decreases in electrical resistivity are observed within hours of the start of steam injection. By the end of the first steam pass (Day 36), both the upper and lower steam zones were at or near steam temperature.

Effect of Dynamic Underground Stripping on Microorganisms

In areas where petroleum hydrocarbons exist naturally in soils, microorganisms have evolved to use these chemicals as food. Studies performed before we began stripping operations showed that on the edge of the spill, where gasoline was sufficiently dilute that it was not toxic to such organisms, bacteria were degrading the gasoline to some degree.

We expected that this flourishing ecosystem would be temporarily extinguished by the heating process. However, when we sampled the six post-test boreholes across the treatment site (drilled after the second steam pass, as mentioned above), we found a new and flourishing microbial ecosystem. We identified some species of bacteria and yeast that had been present before and others that had not. All were growing and were degrading gasoline at temperatures above 70°C. This unexpected assistance from nature is aiding in the continued cleanup of this treatment area.

Conclusions

This first application of dynamic underground stripping to a spill confirmed its value as a cleanup technology. We demonstrated that:

- Steam injection and electric heating effectively heat permeable zones and clay zones, respectively.
- Establishing a complete steam zone in very permeable materials requires large amounts of steam.
- Most of the gasoline is removed through vapor recovery (rather than being extracted as liquid).
- The extraction rate varies greatly according to the amount of steam injection, the total vacuum applied, and the cycle times.
- Alternating steam injection and vacuum-only phases maximizes the extraction rate.
- The treatment systems (e.g., the heat exchanger and the gasoline–water separator) must be able to handle large peak extraction rates and rapid changes in rate.

Dynamic stripping is relatively inexpensive. From experience at other cleanup sites, we estimate that using the pump-and-treat method with vacuum extraction at this site would have taken up to 200 years to achieve the same level of cleanup. Rough cost estimates for pump-and-treat range from $20 million to $60 million.

A "low-tech, brute-force" alternative would have been to scoop out the contaminated material—leaving a hole 100 m across and 50 m deep—and haul the diggings to a broad expanse of disused flat earth. There the soil would be spread out and periodically plowed to expose fresh material so that aerobic bacteria could degrade the contaminants. Once cleaned, the soil would be hauled back to the site to fill up the hole. This treatment site, however, contained a number of underground

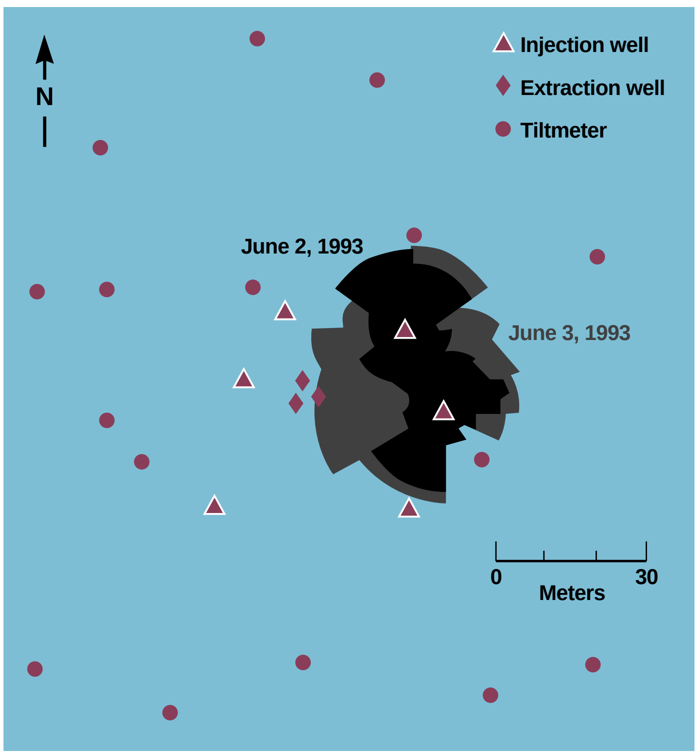


Figure 8. Tiltmeter maps show the growth of the steam fronts emanating from two injection wells on consecutive days. At this time, steam was being injected into only two wells. Steam broke through to the extraction wells the third day.

power, water, and sewer lines, which would have greatly complicated the excavation. This process would have taken a year and cost about $30 million.

In contrast, our dynamic stripping demonstration took 9 months of active time and cost $11 million for treatment and the supporting research. We are confident that if we applied what we learned from this first practical effort, we could perform the same cleanup in 6 months for about $6 million. In the future, improved commercial treatment systems optimized for high-effluent-volume, short-duration applications will probably yield further savings.

Future Work

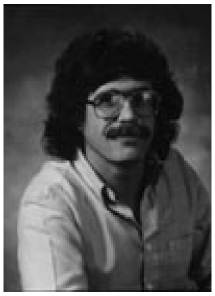
We are exploring the use of dynamic underground stripping to remove chlorinated solvents, which are used in common industrial processes (e.g., TCE or trichloroethylene, used in the manufacture of computer microchips) and are the most common contaminants at Superfund sites. These solvents have been difficult to remove using pump-and-treat methods because, unlike gasoline which is lighter than water, they are denser than water and tend to sink deeper and deeper into the earth. However, because they vaporize at 87°C (below the boiling point of water), they may well be amenable to extraction using dynamic underground stripping. We may also demonstrate dynamic underground stripping to remove solvents at a military base slated for closure.

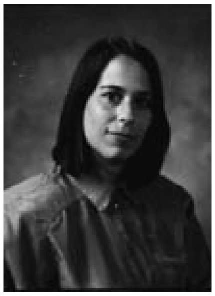
Work funded by the Department of Energy's Assistant Secretary for Environmental Restoration and Waste Management.

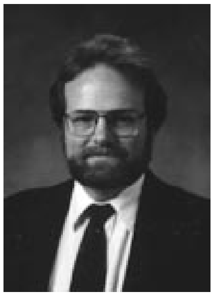
Key Words: dynamic underground stripping; electrical resistance heating; electrical resistance tomography; environmental cleanup; steam injection; underground gasoline contamination—cleanup.

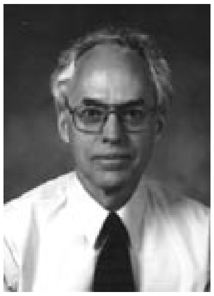
Reference

1. R. D. Aines and R. L. Newmark, "Rapid Removal of Underground Hydrocarbon Spills," *Energy and Technology Review* (UCRL-52000-92-7), July 1992, pp. 1–7.









For further information contact Roger Aines (510) 423-7184, Robin Newmark (510) 423-3644, John Ziagos (510) 422-5479, Alan Copeland (510) 422-8188, or Kent Udell (UC Berkeley) (510) 642-2928 (not pictured).