

**IN-SITU BIOREMEDIATION TECHNOLOGICAL INNOVATION CORE RECOVERY
SYSTEM AND BENCH SCALE STUDY TESTING AT THE SANDIA NATIONAL
LABORATORY, LIVERMORE, CALIFORNIA**

James L. Craig
Los Alamos National Laboratory
Los Alamos, New Mexico

David Miller
Roy F. Weston, Inc.
Albuquerque, New Mexico

ABSTRACT

In-Situ bioremediation at depth is being developed as a remedial action technology for a diesel spill at Sandia National Laboratories, Livermore (SNL) site. A large diameter core recovery program was developed to provide core that most closely represents the diesel fuel contaminated soils at in-situ conditions for bench scale studies. This work was authorized by SNL in conjunction with the California Regional Water Quality Control Board (RWQCB). The Remedial Investigation/Feasibility Study (RI/FS) recommended several technologies to be investigated during bench scale study testing. These technologies included: 1) In-situ volatilization 2) In-situ bioremediation and 3) Soil washing/surfactant flooding. Preliminary results indicate in-situ bioremediation is the preferred remedial action alternative to remediate this site.

INTRODUCTION

The Los Alamos National Laboratory Environmental Restoration - Technical Support Office (ER TSO) is directly funded by the Department of Energy - Albuquerque Operations Office (DOE-AL). ER TSO provides personnel to monitor environmental and remediation activities, provide document review (RI/FS, etc) to assure quality and regulatory continuity, and provide program management for several of the sites directed by the DOE-AL office. This office has several facilities under its direction. Included in these sites is the Sandia National Laboratory at Livermore, California.

In February 1975 an above ground diesel storage tank, with a capacity of 179,000 gallons, developed a leak thru a puncture in a distribution pipe. Over the next several days approximately 59,000 gallons of diesel fuel leaked into the surrounding soil. Upon discovery of the leak, an initial investigation was conducted and a small volume of the fuel recovered. Subsequently, site investigations were initiated. Results from these investigations determined benzene, a component of diesel fuel and a known carcinogen, had reached the groundwater in levels that exceeded the State of California mandated MCL of 0.7 ppb. This information triggered a formal RI/FS. The results of this investigation determined the extent of the contamination and providing sufficient data to propose several remedial action alternatives, including a state-of-the-art cleanup alternative, in-situ bioremediation. The State of California RWQCB approved the technology selected and authorized an extension to the Cleanup Order to enable SNL to conduct the necessary bench scale studies to determine the feasibility of this technology.

**DEVELOPMENT OF CORE RECOVERY
TECHNIQUE**

Since in-situ bioremediation has never been implemented at a site with contamination extending to depths of 105 ft below land surface, a drilling and core recovery technique was developed to collect samples and generate the data necessary to support the bench scale testing and engineering design. Roy F. Weston, Inc. a subcontractor to the Los Alamos National Laboratory was responsible for developing and implementing the needed sampling technique.

In particular, a field program was designed to allow the collection of undisturbed soil samples that would accurately represent on-site field conditions required for the laboratory bench scale testing. In order to allow the most accurate laboratory testing to be performed, it was necessary to collect undisturbed soil samples that most accurately represented in-situ conditions. Samples collected needed to minimize wall effects and changes in the soil structure due to drilling and sampling.

Sample requirements for the bioremediation bench scale study were to collect large diameter soil columns to the depth of 110 ft. The soil horizons to be encountered included alluvial deposits of unconsolidated clays, silts, sands and gravels. The samples were to be collected in the area of greatest concentration of contaminated soils near the center of the spill site. Additional sampling requirements were: 1) columns had to be constructed of a material strong enough to withstand the pressures derived from auger drilling, 2) allow visual inspection of the containerized soil, necessary for geologic description, and the soil profile to ensure all soil horizons and contaminated zones were sampled, and 3) the containerized soil columns could be

drilled into and cut or modified by laboratory personnel during the bench scale study tests.

To accomplish these specific requirements, a CME-75 hollow stem auger rig with 12-inch diameter auger flight was used to collect the samples. The drill rig was equipped with an 8-inch outside diameter continuous core Laskey barrel that could be advanced inside the auger flights during borehole installation. The stainless steel core barrel was sealed and advanced in the borehole using A-rods.

Tenite butyrate was selected as the soil column container material. In addition to meeting the requirements specific to the sampling program, this material has been evaluated to be chemically inert to petroleum compounds. Four, 15-inch long polybutyrate sample tubes were inserted inside the 5-ft long core barrel for each sample interval.

During the installation of the first core barrel, sufficient heat was generated inside the auger to partially melt the polybutyrate tubing. This was the result of friction created by the core barrel spinning inside the auger flight during drilling. To prevent damage to additional sample tubes, vegetable oil was applied to the outside of the core barrel. The outside of the core barrel does not come into contact with the soil nor the sample tubes. This lubrication reduced the friction sufficiently so that no sample tube was damaged during subsequent drilling.

Eleven, five-ft long sample intervals were targeted for collection during borehole installation. The 11 sample intervals were collected at depths of 20-25, 25-30, 40-45, 45-50, 55-60, 60-65, 70-75, 75-80, 82-87, 95-100, 101-106 ft. These sample intervals contained the greatest levels of contamination and represented the various alluvial soil types identified during the RI/FS at the Fuel Oil Spill site.

This sampling technique provided excellent sample recovery from the alluvial soils at the fuel oil spill site. The polybutyrate tubing material was extremely durable for sample collection and allowed the investigators to examine the undisturbed soils. The material could also be modified in the laboratory to provide easy access of analytical instrumentation. This process provided the necessary flexibility of the samples to enable the treatability and bench scale study tests to be conducted with sufficient confidence in testing actual in-situ samples.

BENCH SCALE TECHNOLOGIES ASSESSMENT

Argonne National Laboratory (ANL) in conjunction with The University of Notre Dame (UND) performed the bench scale studies and treatability studies with ANL providing management and contaminant modeling to design the pilot study system.

The collected cores were used to evaluate a number of remediation technologies including: 1) In-situ volatilization, 2) Soil Washing/Surfactant Flooding, and 3)

Bioremediation treatment studies. Results of these studies clearly demonstrate that the in-situ bioremediation is a viable technology for this particular contaminant at the SNL diesel fuel spill site.

In an effort to determine appropriate clean-up criteria, a series of leach tests were performed. Both the raw (contaminated) soil and the sample after treatment (other technologies as noted) were used in the leach tests. The leachate concentrations of total petroleum hydrocarbons (TPH) and target contaminants (benzene, toluene, ethylbenzene, xylenes, pyrene, and naphthalene) were determined by gas chromatography. The effect of aging on the treated samples was also determined by repeating the tests after 1 and 3 months.

In-situ Volitization

The in-situ Volatilization experiments addressed the following issues: 1) technical feasibility of using air-stripping and thermal air-stripping for soil decontamination, 2) the sensitivity of such parameters as air flow rate, temperature, air-to-water mass flow ratio, and pH to the removal efficiencies of the targeted organic contaminants, and 3) quantification and optimization for maximizing the contaminant removal.

Results of the air stripping experiments indicate the percentage of TPH which can be removed using this technology range from 0.21 to 1.52%. These results can be attributed to the relatively high boiling point of fuel oil and that the contaminants have been in place for over fifteen years with little migration from the original spill site.

Soil Washing/Surfactant Flooding

Soil washing/surfactant flooding studies indicate that after flushing the soil with over three pore volumes of water the TPH recovery was less than 3% with a typical value of about 1%. In an effort to improve this performance, 21 surfactants were evaluated at up to 2% concentrations. The typical enhancement of hydrocarbon removal over water alone was about 25% with a maximum improvement of 60% over water. These results indicate that soil washing/surfactant flooding is neither a technological nor economical option for decontamination at this site.

In-situ Bioremediation

The in-situ bioremediation engineering evaluation was divided into several efforts: 1) characterization of core soils for nutrients and their availability, and for geophysical and geochemical properties, 2) identification and qualification of soil micro-organisms with respect to growth potential, 3) development of in-situ model laboratory reactors from sections of field core samples containing undisturbed samples, and 4) utilization of slurry reactors to determine require-

ments for nutrients, surfactants, electron acceptors, and possible addition of specialized microbes.

Results of these laboratory studies conclusively demonstrate that the bioremediation of the fuel oil spill at SNL facility is scientifically feasible. In a period of approximately three months, the hydrocarbon concentration was reduced in the in-situ reactor from approximately 16,000 mg/kg to 6,000 mg/kg. Additionally, results of the soil slurry reactor studies verify that the hydrocarbon contamination at the SNL location can be bioremediated.

Two basic issues control the success of bioremediating the SNL fuel oil spill site. First, hydrologic control must be established in the subsurface environment. This is necessary to ensure that the contamination does not spread beyond the existing contaminated area. Second, a method of transporting the required nutrients and exogenous electron acceptors to the contaminated zones must be developed.

It appears the transport of nutrients and other essential compounds can be achieved by the selective addition of divalent cations into the soil. This addition of minerals can positively affect both the magnitude and maintenance of the observed hydraulic conductivity. Additionally, the macro-nutrient nitrogen, provided as ammonium, can also be more uniformly distributed to targeted soil domains. The biological degradation patterns of the more slowly degraded residues of diesel fuel, observed in the laboratory in-situ reactors, apparently mimic that observed for the closed slurry reactors in magnitude and chemical composition. Further, these residues were apparently minimally mobilized into the recycled waters.

Although the lowest level of TPH that can be obtained in the soil environment remains undetermined at this time,

the most significant problem to be anticipated apparently relates to the mass transfer of oxygen utilizing hydrogen peroxide.

The issue of maintaining hydrologic control over the environment is currently being addressed by an aggressive numeric modeling program. To support this effort, laboratory studies are being conducted to define the constitutive properties of the soil as they relate to the multi-phase transport of the contaminants at the fuel oil spill site. Specifically, the physics governing the transport of the fuel oil as it is displaced by other fluids is being examined. It is expected that a reliable geologic and numeric representation of the site will be developed which will facilitate the design of the pilot study project and address legitimate concerns regarding the uncontrolled movement of the subsurface contaminants.

CONCLUSIONS

A large diameter core can be recovered using conventional type drilling and available equipment to provide needed soil samples for laboratory testing. Preliminary results from several remediation alternatives performed on the large diameter core demonstrate that in-situ bioremediation will be the preferred alternative to remediate the diesel fuel contamination at the SNL facility. Additional testing on the soils can provide the geo-properties necessary to develop the pilot program will demonstrate bioremediation can be performed at depths to 105 ft in valley type fill above the saturated zone thereby avoiding the necessity and expense of an excavation and disposal remedial action alternative.