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WSRC-MS-94-0324

A PETROLEUM CONTAMINATED SOIL BIOREMEDIATION FACILITY

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A document prepared for SPECTRUM '94 at Atlanta, GA, USA from 8/14/94 thru 8/18/94.

DOE Contract No. DE-AC09-89SR18035

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DISCLAIMER

A PETROLEUM CONTAMINATED SOIL BIOREMEDIATION FACILITY

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ABSTRACT

The amount of petroleum contaminated soil (PCS) at the Savannah River Site (SRS) that has been identified, excavated and is currently in storage has increased several fold during the last few years. Several factors have contributed to this problem: 1) South Carolina Department of Health and Environmental Control (SCDHEC) lowered the sanitary landfill maximum concentration for total petroleum hydrocarbons (TPH) in the soil from 500 to 100 parts per million (ppm), 2) removal and replacement of underground storage tanks at several sites, 3) most recently SCDHEC disallowed aeration for treatment of contaminated soil, and 4) discovery of several very large contaminated areas of soil associated with leaking underground storage tanks (LUST), leaking pipes, disposal areas, and spills. Thus, SRS has an urgent need to remediate large quantities of contaminated soil that are currently stockpiled and the anticipated contaminated soils to be generated from accidental spills. As long as we utilize petroleum based compounds at the site, we will continue to generate contaminated soil that will require remediation.

The facility has no precedence in South Carolina or Georgia and as such represents new technology for the area. However, since other states have demonstrated similar facilities, it represents low risk and has high public acceptance. The facility also provides South Carolina with the opportunity to demonstrate and evaluate an innovative, environmentally sound, on site and cost effective remediation technique that can be used to help clean-up the growing LUST problem. The basic concepts of this technology are expected to be applicable to other sites in the Department of Energy complex having PCS. The experience gained at the SRS facility will provide the basis for designs for other sites. The simplistic design contributes direct benefits associated with the ease of management, operations and safety.

I. INTRODUCTION

Biodegradation of petroleum hydrocarbons in soil (petroleum land farming) has been used by the oil industry for more than 30 years as an efficient way to destroy oil sludges (Bartha and Bossert, 1984). Indigenous microorganisms in the soil can degrade large quantities of petroleum hydrocarbons if they are provided sufficient amounts of water, oxygen, and other limiting nutrients, usually nitrogen (N) and phosphorus (P). By applying oil to the soil surface, adding fertilizer (N & P), water, and then tilling to aerate (oxygenate), the soil microbes have been shown to completely degrade large quantities of oil. A demonstration of this technology using waste oil was done at SRS near Central Shops in 1980 (Watts and Corey, 1982).

Until recently, the state-of-the-art approach to soil remediation was excavation and disposal at a secure landfill. Changes in liability concerns, costs, and regulatory constraints have decreased the popularity of excavation and disposal as a soil cleanup alternative. Landfill disposal of contaminated soil does not remove the future liability of its generator, who will be held jointly liable with the landfill operator for any future associated contamination. Thus, on-site permanent solutions must be sought whenever possible.

The basic treatment design is referred to by the U.S. Environmental Protection Agency (USEPA) as a "prepared bed" bioreactor (Sims et al., 1989). This is a proven technology, having been demonstrated in several states by several different concerns. Sims (1986) reported 50-100% reduction of fossil fuels in soil after only 22 days. St. John and Sikes (1988) reported that a prepared bed system, complete with fugitive air emissions control, at a Texas oil field was able to reduce volatile organic carbon by >99% after 94 days, with semivolatiles being reduced by more

than 89%. In California, Ross et al. (1988) reported that four acres of soil 15 inches deep, contaminated with diesel and waste motor oils were decreased from 2,800 ppm TPH to less than 380 ppm in only four weeks. He also reported that at another site owned by a heavy equipment manufacturer, 7,500 yd^3 (5,734.16 m^3) were reduced to <100 ppm TPH after nine weeks and an additional 9,000 yd^3 (6,880.99 m^3) with 180 ppm TPH were reduced to <10 ppm after only five weeks. Another site in California had 600 yd^3 (458.73 m^3) reduced from 1000 ppm TPH to <200 ppm in 35 days. Molnaa and Grubbs (1989) report other sites in California where similar results were obtained, e.g., 2000 yd^3 (1,529.11 m^3) with 2800 ppm TPH were reduced to less than 38 ppm in 74 days, a truck stop where 15,000 yd^3 (11,468.32 m^3) were reduced from 3000 ppm TPH to less than 30 ppm TPH in 62 days, and a site contaminated with lubricating oils where 25,000 yd^3 (19,113.38 m^3) were reduced from 4800 ppm down to 125 ppm in 58 days. Clearly, the higher rain fall and higher ambient temperatures of this climatic region would suggest that rates would be even greater in this area than in many of the previous sites where this technology has proven successful.

II. TECHNICAL NEED

As sources of clean surface water steadily decline, our reliance on ground water will undoubtedly continue to increase far into the next century. Thus, with increasing urgency, ways have been sought to clean-up, i.e. remediate, PCS. The sOILS Facility, named to highlight "OILS", will provide SRS and the state of South Carolina with the opportunity to demonstrate and evaluate an innovative, environmentally sound, on site and cost effective remediation technique that can be used to handle fuel spill clean-ups and the growing LUST problem.

The basic concepts of this technology are expected to be applicable to other sites in the Department of Energy complex having PCS. However, the particular process designs will be site specific. The experience gained at the SRS facility will provide the basis for designs for other sites. Regulatory drivers for this activity are the Resource Conservation and Recovery Act (RCRA) (40 CFR Parts 280, 280.20, 280.21 and 280 22. and Parts 280.70-280.74), the State Underground Petroleum Environmental Response Bank Act of 1988 (SUPERB), S.C. Code Ann. §§ 44-2-10 et seq., State Safe Drinking Water Act of 1976, S.C. Code Ann. § 44-55-10, et seq., Pollution Control Act of 1970 (PCA) and Federal Safe Drinking Water Act (40 CFR 141).

III. ALTERNATIVES

A variety of alternative technologies to land disposal of untreated PCS exist today. They include in situ/ex situ bioremediation, soil stabilization and solidification, vapor extraction, bioventing, soil washing and/or chemical treatment and incineration. Several of these technologies have disadvantages that far out weigh the advantage of being used in lieu of ex situ bioremediation. For example, soil stabilization and solidification, although relatively low cost, generate a volume increase of disposable material and can create possible limitations on future use of sites where this method is used as it does not destroy the contaminants it only immobilizes them. Vapor extraction and bioventing are viable methods but require extensive site characterization and they are very site specific applications, as is in situ bioremediation. Soil washing (flushing) and chemical treatment can be used as a permanent treatment method but additional waste streams are generated requiring further treatment and expense. High and low temperature incineration is an effective method to destroy PCS. The performance of these systems is measured by the destruction and removal efficiency (DRE). Meeting the mandates of a high temperature DRE of 99.9% can be costly, requiring the use of large amounts of supplemental fuel to meet minimum operating parameters. Low temperature incineration is more applicable to PCS remediation but with both applications, permit conditions, contaminant concentrations, soil volume, incinerator efficiency and heating values of the soil all control what the final cost of operations will be, not withstanding the NIMBY factor (Not In My Backyard).

IV. BENEFITS

There are distinct advantages associated with the on site remediation of PCS. By excavating the contaminated material to an above ground treatment cell, the engineer/scientist has better control over the critical factors which dictate the rate at which the degradation takes place. Nutrient concentration, moisture content, oxygen availability and in many cases temperature can be controlled to maximize the efficiency of the process (Lombard, 1990).

Sampling and analysis of the PCS and the excavated area become simplified. It is far easier to demonstrate that the area is clean when the contaminated material has been removed from the site. In order to optimize the sampling and analysis many agencies and/or customers have required the material be excavated to insure complete clean up.

The simplistic design contributes direct benefits associated with the ease of management, operations and safety. A minimal staff is required to operate the facility adding to the low risk factor by limiting exposure to operations personnel. This action supports the efforts of SRS to remain cost effective while providing Site personnel with a safe working environment. The results of many studies, by independent researchers, indicates that bioremediation is the most cost effective way to treat PCS (Lombard, 1990).

V. FACILITY DESIGN and OPERATIONS

The facility design consists of a reinforced concrete floor 400 ft (121.92 m) long and 40 ft (12.19 m) wide. This base is divided into four cells 200 ft (60.96 m) long and 20 ft (6.096 m) wide. The bases slope to the center where a leachate control system collects any water in a reinforced concrete holding tank which can reapply any leachate or rainwater via a pumping system powered by a portable electric generator with a Burks model 5WT5, 1/2 horsepower self priming centrifugal type pump connected to a sprinkler system with distribution heads mounted on the center wall. The sprinkler heads are designed to provide water to each cell or to the entire unit as needed. The leachate collection system is designed to hold water from a catastrophic 25 year rainfall event. Each cell is equipped with a full size dark brown, ultraviolet light resistant tarp to act as a watershed and heat sink during cold weather operation. The tarps are light weight, high strength, tear resistant material which can be easily removed for roto-tilling or quickly installed by one person in the case of heavy rain. A base of clean soil will be applied to a depth of 6-9 inches (15- 23 cm) to provide good soil drainage. The contaminated soil will be applied to the top surface of the drainage bed to a depth of 6-12 inches (15-30 cm). The design loading requirements take into consideration the utilization of existing site equipment, e.g., trucks and graders, for transporting, applying, and distributing the soil in the facility. Each cell is open on the end to provide easy access by large vehicles. The facility is able to treat approximately 900 yd^3 (688 m^3) of contaminated soils every 6-12 weeks, depending on the concentration level of the PCS, ambient temperature and weather conditions. Thus, 3,000- 8,000 yd^3 (2,294-6,116 m^3) of soil could be processed every year. Although relatively simplistic, the design provides excellent environmental control and operating conditions that minimize fugitive air emissions and maximize biodegradation rates.

To operate the sOILS Facility requires a simple four step approach; 1) screening the soil and loading the bioremediation cells with PCS; 2) treatment of the material, 3) interim sampling and analysis and 4) removal and disposal of the remediated soil. Upon receipt, the PCS is checked for appropriate documentation, accepted, classified and stockpiled according to contaminant type (i.e. diesel fuel, gasoline, waste oil etc.). As space is avaiable for additional PCS at the sOILS Facility, the maintenance department will transport commonly classified PCS to the facility and place it according to the direction of the sOILS Facility operator. A material tracking database and inventory system has been set-up to track PCS for its complete life cycle, from the time it arrives to its final disposition or disposal location.

The treatment process begins with an initial screening of the PCS for inorganic nutrients, pH, contaminant concentrations, moisture, and microbes. Adjustments to the pH, nutrients, and moisture are made, as needed, to stimulate and optimize he biodegradation rate. The soil is kept moist but not damp and aerated via roto-tilling once a week. Monthly analysis continue for inorganic nutrients, contaminants, and microbes. Weekly measurements for air emissions, pH, and moisture are taken by random samples to assure that volatilization and particulate emissions are below annual air emission limits. The bioremediation treatment process continues until the soil analysis has demonstrated by gas chromatography (GC) to be below 100 ppm for TPH and 10 ppm for BTEX.

The final concentration of contaminants in the soil dictates the final disposition of the material or its disposal location. Soils with TPH and/or BTEX below detectable levels (< 1 ppm) may be used as erosion control material or road base material; soils (i.e.. above detection but below clean-up criteria) are removed to a sanitary landfill per SCDHEC mandate.

VI. AIR EMISSIONS

Stockpiled or containerized contaminated soils are transported to the sOILS Facility and placed directly into cells. Fugitive dust, Total Particulate Matter (TPM) is emitted from unloading (placing into cells) and loading (removal of treated soil to the landfill) operations. Another emission source is from volatilization of organic constituents (i.e., TPH and BTEX) from PCS during soil turn-over or tilling operations. TPM and volatile organic (VO) emissions have been calculated using the U.S. Environmental Protection Agency (USEPA), recommended AP-42 formula (USEPA, 1985), and have been determined to be 317.2 lb/mo (143.88 kg/mo) TPM and 288.1 lb/mo (130.68 kg/mo) VO using the highest median value (i.e.; 6.34K ppm TPH) presented in current test data. In order to remain in compliance with SCDHEC air emission standards and develop a baseline inventory, weekly air emission samples are being taken for the first six months of operations. When the baseline is established, the air sampling frequency will be reduced to a monthly time interval to monitor long term emissions trends. A standard surface emissions sampling protocol using a modified isolation flux chamber procedure (Dupont, 1997) is used in field air emissions sampling activities.

The operations of the sOILS Facility will result in airborne emissions of particulates, TPH, and the air toxics benzene, toluene, ethyl benzene and xylenes. The toxics are regulated emissions under SCDHEC, Air Pollution Regulation 62.5, Standard 8, Toxic Air Pollutants (1991). Both median and worst case data for emissions of particulates and toxics were modeled using initial values from the Bureau of Air Quality Control Modeling Toxics Questionnaire.

A. Calculations

1. TPM associated with unloading/loading of contaminated soil and tilling of soil once weekly during soil treatment.

a. Unloading and loading operation:

$$E = K(0.0018)\frac{(\frac{s}{5})(\frac{U}{5})(\frac{H}{5})}{(\frac{M}{2})^2(\frac{Y}{6})^{0.33}}$$

where:

E = total suspended particulates
K = particle size, use 0.73 for < 30 μm
U = mean wind speed, use 6 mph
H = drop height, use 5 feet
M = moisture content, use 0.25%
Y = dumping device capacity, use 1 yd^3
s = soil silt content, use 18%

$$E = 0.73(0.0018)\frac{(\frac{18}{5})(\frac{6}{5})(\frac{5}{5})}{(\frac{0.25}{2})^2(\frac{1}{6})^{0.33}}$$

= (0.66 lb/ton) (900 yd^3/mo) (1.4 ton / yd^3)

E = 277.22 lb/mo = 0.385 lb/hr

b. Soil roto-tilling:

$E = K(4.8)(s)^{0.6}$ lbs/acre
where:

E = emission factor
s = silt content of the soil, use 18%
K = 1

$E = 1(4.8)(.18)^{0.6}$

= (27.19 lbs/acre) (2.2957 x 10^{-5} acre/ft^2)
(20 ft x 200 ft)
= (2.5 lbs/cell/tilling) (4 cells x 4 tilling/mo)
= 40 lbs/mo = 0.055 lbs/hr

= 277.22 + 40
TPM = 317.22 lbs/mo = 0.44 lbs/hr

2. VO emissions are estimated assuming 25% of the original constituent is emitted to the air. applying this estimate to a contaminated soil having a TPH = 6,340 ppm; Benzene = 15 ppm, Toluene = 11 ppm, Ethyl Benzene = 11 ppm, and Xylene(s) = 34 ppm. Estimating that 900 yd^3. of contaminated soil can be treated to acceptable levels every 4 - 12 weeks, the VO emissions to the air are approximately 0.40 lbs/hr as estimated below.

a. VO emissions: TPH =

0.25 (6340 ppm)
x (7.48 x 10^{-6} lbs/ft^3/ppm)
x 27 ft^3/yd^3 x 900 yd^3/mo
288.1 lbs/mo = 0.40 lbs/hr

b. Air toxics emissions: BTEX =

Benzene, Toluene, Ethyl benzene, and Xylene(s)

Benzene =
0.25 (15 ppm) (7.48 x 10^{-6} lbs/ft^3/ppm)
x 27 ft^3/yd^3 x 900 yd^3/mo
0.68 lbs/mo = 0.001 lbs/hr

Similarly,

Toluene = 0.50 lbs/mo = 0.001 lbs/hr

Ethyl benzene = 0.50 lbs/mo = 0.001 lbs/hr

Xylene(s) = 1.54 lbs/mo = 0.002 lbs/hr

VII. SAMPLING AND ANALYSIS

Initial sampling and analysis of the PCS put into the facility consists of 3 random samples per cell for inorganic nutrients (nitrogen/nitrates, sulfates, phosphorus/phosphate), pH, contaminants (TPH and BTEX), heavy metals, polycyclic aromatic hydrocarbons (PAHs), moisture, and microbes (direct counts and viable counts and enrichments). Fertilizer is applied, if necessary, depending on the results of the inorganic nutrient analysis. Monthly (weekly if necessary) random samples are taken from each of the four cells to determine if cleanup criteria has been met and/or if additional nutrients are required.

A. Sampling Protocol

The primary goal of the sampling activity is to obtain an unbiased statistical estimate of the mean TPH and/or BTEX concentration within the treatment cell(s). As discussed above, simple random samples are taken for analysis for the parameters shown in Table 1, after the contaminated soil is applied into a cell. Monthly, random samples are taken to monitor the soil biodegradation rate. Sampling and analysis are performed using EPA protocols (i.e., SW 846, Third Edition, 1986). Required holding times for soil samples can be seen in Table 2.

Table 1. Frequency Parameters for Soil Samples

	Initial	Monthly	Weekly	Final
Organics				
TPH	Y	Y	as needed	Y
BTEX	Y	Y	as needed	Y
PAH	Y	as needed	as needed	as needed
Inorganics				
N	Y	Y	-	-
P	Y	Y	-	-
Moisture	Y	Y	Y	-
pH	Y	Y	Y	-
Metals	Y	-	-	-
Microbes	Y	Y	-	-

Table 2. Soil Sample Holding Times

CONTAMINANT	HOLDING TIME
BTEX (Benzene, Toluene, Ethylbenzene, Xylenes)	Analyze as soon as possible (max. 14 days)
Total Petroleum Hydrocarbon (TPH) (low to medium bp fuels)	Analyze as soon as possible (max. 14 days)
Total Petroleum Hydrocarbon (TPH) (high bp fuels)	Extract within 14 days Analyze within 40 days
Polycyclic Aromatic Hydrocarbons (PAH) (including naphthalene)	Extract within 14 days Analyze within 40 days
Mercury	28 days[1]
Metals (except mercury)	6 months[1]

Source: SCDHEC, 1992

1.) Soil samples must be at least 200 g and usually require no preservation other than storing at 4°C until analyzed.

Tables of uniformly distributed random x, y and depth coordinates are used to establish sampling locations and depths. These tables are generated using a computer software random number generator. The coordinates represent the distance in feet (meters) from the origin of each axis. Depth values represent the distance below the surface in inches centimeters). The range of values are based on the size of the cell and the extent of the distribution of contaminated material within the cell. Using the SRS coordinate grid system, the orientation of the x and y axis are North/South and East/West respectively, with the northerly and easterly directions from the origins being positive. Each cell has been assigned an alpha-numeric designator. The Northeast cell being "A", Southeast "B", Southwest "C", and Northwest "D". The center wall that forms the cell is numbered 1 through 35 in 5 ft intervals and in an easterly and westerly direction from the point of origin. The end of each cell is numbered 1 through 4 in 5 ft intervals forming 25 ft^2 sample plots in each cell.

B. Analytical Procedures

The EPA 8000 series analytical procedures are used in the analysis of PCS samples. The use of these methods is now nearly universal in public and private sector laboratories. Each of these methods has an associated list of target compounds for which it was specifically developed and evaluated. These methods use GC and mass spectrometers (MS) or a combination of both GC/MS techniques to detect organic compounds. These instruments are well known for their excellent sensitivity and selectivity for specific target compounds.

Detection of complex hydrocarbon mixtures is best achieved using a GC with a flame ionization detector (GC-FID). GC-FID analysis provides a more adequate representation of the degree of hydrocarbon contamination. EPA Method 418.1 does not provide information on the type of hydrocarbon contamination and the low boiling point components are easily lost. This method (418.1) is only being used for screening purposes and final disposal is governed by GC analysis (SCDHEC, 1992).

The procedures list different methodologies for the low to medium boiling point hydrocarbons (gasoline) and the high boiling point hydrocarbons (diesel motor fuels and light heating fuels). A purge and trap or head space is preferred for the more volatile contaminants whereas the high boiling point contaminants are to be analyzed using a GC-FID. The gas chromatographic analysis is equivalent to the well known "California Method" for testing TPHs for the Underground Storage Tank Program. For highly contaminated samples the waste dilution technique may be used but this must be documented with the analytical results. Any sample dilutions performed must also be documented with the analytical results (SCDHEC, 1992)

As part of the criterion for success of the sOILS Facility, the use of screening for the absence of soil contamination by immunoassay is a high priority. We hope to demonstrate, with a high level of confidence, that immunoassays have unique properties that should be utilized for environmental testing. Field soil vapor analyzers have been used for many years for measurement of indicator parameters. However, these instruments are not quantitative, and the values have been shown to correlate poorly with laboratory-derived results (Denehan et al., 1990; Preslo et al., 1990). Factors affecting variability in results are instrument response time, sensitivity, calibration procedures and environmental conditions. Immunoassays offer both sensitivity and specificity in a quick and relatively inexpensive format. In addition they can be performed using crude sample preparations and are easily adaptable to on-site testing (Hammock et al., 1990; Vanderlaan et al., 1990).

Recently, preliminary studies by the Savannah River Technology Center, Environmental Sciences Section have shown sensitivity and specificity levels by immunoassays to be within a 95% confidence interval for the detection of TPH and BTEX. Concentration ranges as low as 2.5 ppm for BTEX and 4 ppm for TPH have been consistently detected. Additional information on the laboratory techniques used and the results obtained in the analysis and evaluation of the various immunoassay test kits will be made available in a future publication.

VII. MICROBIOLOGICAL PROCEDURES

Three microbiological analyses are done on a monthly basis. The soil samples from the facility are collected and processed on the same day the sampling is done. The first test gives total direct cell counts in the soil, utilizing an acridine orange stain for bacterial nucleic acids. This provides a total bacterial cell count, expressed in cells per gram dry weight. The second analysis being performed is viable counts, that gives the total number of organisms that can be cultured on an oligitrophic media. This number is expressed in colony forming units per gram dry weight. The third analysis is an enrichment for TPH. Bacteria are grown on a minimal salts media with trace metals and no available carbon source other than petroleum (diesel fuel) vapors.

A. Acridine Orange Direct Counts (AODC)

AODC provides a direct estimate of the total number of bacteria in the environment, regardless of the ability to grow on any media that might be used. Samples are preserved in phosphate buffered formalin. Samples (1 to 3 grams) are extracted three times with a non-ionic homogenizing detergent to remove bacteria from the sediment particles. Homogenates are cleared by low speed centrifugation and the supernatants pooled. Ten microliters of supernatant is spotted onto each well of a toxoplasmosis microscope slide, stained with 0.01% acridine orange, then rinsed with distilled water. The number of cells stained with acridine orange are counted by epifluorescence microscopy. The number of cells per sample is normalized by dividing by the dry weight of the sediment. Counts are reported as cells per gram (Sinclair and Ghiorse, 1989).

B. Aerobic Heterotrophic Plate Count.

This method provides an estimate of the total number of viable aerobic and facultatively anaerobic bacteria in the ground water. Low and high nutrient concentrations of a medium will be used to indicate differences in bacteria adapted to oligotrophic and eutrophic conditions. Samples (1 to 3 grams) are weighed directly into 15 ml conical centrifuge tubes containing 9 ml of pyrophosphate buffer. Subsequent serial dilutions are made in phosphate buffered saline. 0.1 ml of each appropriate dilution is inoculated onto a corresponding plate of appropriate medium. For this study, 1% peptone-trypticase-yeast extract-glucose (PTYG) is used (Balkwill, 1989). Plates are incubated at room temperature for at least two weeks prior to counting. Bacterial colonies are counted with the aid of low power magnification. Counts are normalized to sediment dry weights and reported as colony forming units (CFU) per gram dry weight.

C. TPH Enrichment.

This method provides an estimate of the total number of viable aerobic and facultatively anaerobic bacteria capable of living in a soil enriched with petroleum hydrocarbons. Successful bioremediation of TPH can also be in terms of increased microbial activity, increased biomass; particularly biomass which contains TPH

degrading machinery, increased biomass capable of consuming TPH as evidence of stimulation by treatments. Minimal salts media (Fogel et al. , 1986; AEM 51(4): 720-724) are used. The plates are incubated in an enclosed environment with petroleum (diesel fuel) vapors available to the bacteria as a source of carbon for metabolism. This count will also be in CFUs per gram dry weight. As petroleum will be the only carbon source available, this will be a count of TPH degraders only.

VIII. SUMMARY

The operation of the sOILS Facility shows the value added by using bioremediation as an effective and clean method to remediate PCS by: 1) demonstrating bioremediation as a permanent method for remediating soils contaminated with petroleum products; 2) establishing the best operating conditions for maximizing bioremediation and minimizing volatilization for SRS PCS during different seasons; 3) determining the minimum set of analyses and sampling frequency to allow efficient and cost effective operation; 4) determining how best to utilize existing Site equipment and personnel to optimize facility operations and conserve SRS resources, and 5) as an ancillary dividend, demonstrating and optimizing new and innovative analytical techniques that will lower cost, decrease time, and decrease secondary waste streams for required PCS assays.

ACKNOWLEDGMENTS

The information contained in this article was developed during the course of work for the Westinghouse Savannah River Company under Contact No. DE-AC09-89R180035 with the U.S. Department of Energy. In addition, the authors would like to thank Ms. Marilyn M. Franck and Mr. Sam L. Baxley for their contributions and assistance in the implementation of the sampling and analysis program for the sOILS Facility. Also, Bechtel Savannah River Inc. for providing the facility design and construction and Mr. Marc Waltz of the Central Services Works Engineering department, for providing operations support for the facility.

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