EX SITU BIOLOGICAL PETROLEUM REMEDIATION: LANDFARM CASE HISTORY

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INTRODUCTION

In the late 1980s, the University of Idaho (UI) negotiated an option to purchase an 11-acre industrial parcel in Moscow, Idaho, for possible university expansion. Phase I and II environmental site investigations took place at three former petroleum bulk plants on this site, and at four underground storage tank sites (USTs) on the UI campus. The investigations identified ≈20,00 yd^3 of soil with weighted average total petroleum hydrocarbon (TPH) concentrations of 947 ppm oil, 465 ppm diesel, and 624 ppm gasoline. The Idaho Division of Environmental Quality (IDEQ) petroleum cleanup requirements mandated remediation levels of 40 ppm gasoline, 100 ppm diesel, and 40 ppm oil based on depth to groundwater (<25 ft) and proximity to surface water (<100 ft). The low permeability of the contaminated silt-clay-sand sediments and refractory long-chain hydrocarbons indicated that in situ remediation would be unlikely to achieve cleanup levels in an acceptable time period. Excavation was considered necessary, but the high cost of low-temperature thermal desorption ($1.3 million at $65/ton, plus trucking) and lack of a local commercial landfarm required that UI design and construct its own landfarm to bioremediate the petroleum-contaminated soil (PSC). An administrative decision was made to maintain ownership of the PCS over concerns about potential liabilities from the landfarm operation or residual contamination. Negotiations with the responsible parties were conducted on a sliding scale based on shared responsibility with a service fee of ≈$35/$yd^3$.

Idaho Division of Environmental Quality siting criteria for land application of petroleum-contaminated soils (IDEQ, 1994) specified six major landfarm siting criteria: 12% maximum slope, 100-ft minimum distance from surface water, 100-ft minimum distance to water wells, 931-ft setback from buildings (based on estimated contaminant concentrations and volumes), and >25 ft to groundwater. In addition, treated soils were not be used for food-chain agricultural purposes, regardless of the level of remediation. On the basis of these criteria, three critical questions were raised: (1) Could the landfarm be designed to prevent surface and groundwater contamination? (2) Would the contaminants be degraded in the acceptable 2-year time frame? (3) Would residual metal contamination perpetually classify the soil as hazardous waste, preventing the return of the land to agricultural use?

SITE DESCRIPTION AND EXPERIMENTAL METHODS

Hydrogeologic site investigations consisted of a literature review, consulting water well boring logs in a 2-mile radius, and drilling a total of 12 hollow-stem-auger boreholes to characterize the shallow stratigraphy. The chosen landfarm site is located on a northwesterly trending ridge ≈900 ft long, 200 to 450 ft wide, and covering ≈10 acres within the city limits of Moscow, Idaho. The average slope is about 9%, with 80 ft of elevation change from top to bottom. Previous land use at the site was pasture for the UI beef herd.

Since the landfarm was designed to be placed on an unlined parcel of agricultural land, careful consideration was given to the regional and site hydrogeology to assess probable impacts to groundwater. Detailed hydrogeologic investigations previously conducted at the UI Aquaculture Site, located ≈1600 ft to the northeast of the landfarm (Kopp, 1994), and the UI Groundwater Research Site, located 2500 ft northeast (Li, 1991), indicated that a basalt aquitard separates the shallow alluvial and upper fractured basalt sub-aquifer from the upper drinking water aquifer. This site-specific hydrological barrier, combined with a relatively thick vadose zone (135 ft) composed of silt, clay, and basalt, suggested that impacts to groundwater would be minimal at the landfarm site. Interpretation of the data indicated that the petroleum was at residual saturation in the soil, so free product migration was unlikely to be a problem. Dissolved organics were a potential problem but were considered degradable in the aerobic vadose zone beneath the landfarm, or in the shallow drainfield in the stormwater discharge zone. Absence of surface water runon, generally well-drained soils with a high soil moisture-holding capacity, and reinfiltration of the limited quantity of stormwater runoff prevented any direct surface water impacts from the landfarm. The primary concern for off-site contaminant migration was from soil erosion. The fine-grained, unconsolidated contaminated soils placed on an 8% slope were highly erodible, so considerable efforts were expended to prevent offsite soil migration.

One to two feet of topsoil was stripped from the landfarm , placed in a berm along the perimeter, and covered with straw and coconut fiber erosion control mats to stabilize the berm and to contain all contaminated soils within the landfarm area. To minimize erosion on the landfarm during the winter and spring, the soil was aerated with a subsoil ripper along contour and covered with composted manure and animal bedding, and silt fences were installed and spaced on 50-ft centers parallel to contour (Gill, et al., 1997).

A seasonal perched water table was identified on the lower third of the landfarm (Brackney, 1995). A stormwater runoff and leachate collection system was keyed into a low-permeability "B-soil" horizon. It was designed as an underdrain to prevent ponding by gathering water into two 1000-gallon sediment-settling and oil-water separator tanks. A 12-inch culvert drains the tanks and discharges to a trapezoidal flume where stormwater effluent is gauged with an ISCO 4230 bubbler flow meter and sampled with an ISCO 3700 stormwater sampler prior to discharging to a shallow drainfield. Six groundwater monitoring wells were installed, four upgradient and two downgradient from the landfarm, including three multi-point standpipe well clusters. These were sampled to characterize petroleum and inorganic impacts to the shallow groundwater.

A literature review indicated the following optimal conditions for petroleum biodegradation: Maintain soil O_2 levels at near atmospheric conditions by thin spreading the soil and tilling whenever levels are <5%; maintain soil moisture levels to 60-70% of field capacity; and balance the carbon:nitrogen:phosphorus levels to 10-20:1:0.1

The landfarm was irrigated with secondarily treated sewage effluent from the municipal wastewater treatment plant which was applied with a wheel line sprinkler irrigation system. Soil moisture was measured in the field by Soil Moisture Equipment tensiometers (model 2725AR) installed in three arrays from 6 to 36 inches deep. The contaminated soils were aerated monthly during the growing season by a subsoil ripper with 2.5 ft ripper teeth, spaced on 2.5 ft. centers, with an attached ripper shoe to increase soil fracturing. Soil O_2 levels were sampled with an AMS soil vapor probe and analyzed with a hand held combustible gas/O_2 meter.

Because of the heterogeneity of the petroleum concentrations from seven sites in various stages of weathering, three 30-ft-diameter grids were installed across the site. Although the contaminated soil had been placed on the site in fall 1994, change in TPH concentrations over time were measured in each grid in May and October 1995 to estimate the progress of degradation over the entire site. Three different treatments were designed for the grids: Grid 1 was planted in perennial ryegrass and irrigated, Grid 2 was tilled and irrigated, and Grid 3 was tilled without irrigation. Most probable number (MPN) bacterial counts, with hexadecane as a sole-carbon source, were measured from soil samples collected during the May and October 1995 sampling periods. A random number radial-coordinate system was generated to select sample locations within each grid. At the end of the first season, a crop of rapeseed and perennial ryegrass was planted for erosion control. At the beginning of the second season (June 1996) the entire site was sampled, establishing uniform gridlines across the site. On each gridline random numbers were generated to select sample locations along the line. Because the oil fraction was considered to be the slowest to degrade, EPA Method 418.1, TPH-Oil, was selected to monitor remediation progress. In June 1997 another round of soil samples will be collected to measure residual contamination.

RESULTS

Very little dissolved or miscible petroleum products were transported offsite from stormwater runoff (Figure 1). Ethylbenzene and xylene were detected only on November 1, 1994, with total concentrations of 0.036 mg/L. Maximum TPH-diesel was 1 mg/L in December 1994, while the maximum TPH-oil of 2.6 mg/L was measured in March 1995. The low levels of TPH-diesel and oil may reflect interference from the high organic concentrations leached from the composted animal manure. The erosion controls at the site worked remarkably well, and no contaminated soil left the landfarm, largely as a result of the compost covering the site.

Leachate from the compost contributed high nitrogen concentrations (NH_3, NO_3, and organic N), chemical oxygen demand (COD), and total dissolved solids (TDS) to stormwater runoff (Figure 2). Maximum concentrations of total nitrogen (400 mg/L), COD (1,800 mg/L), and TDS (2,800 mg/L) occurred in December 1994. Dissolved lead concentrations were also elevated during this period, peaking at 22 μg/L (data not shown). These dissolved inorganic constituents declined during the first year and stabilized by March 1996, with lead concentrations dropping to below detection limit (1 μg/L).

The downgradient monitor well network showed elevated nitrate during spring 1995, with maximum concentrations of 61 mg/L NO_3-N in the perched aquifer, as compared to a back-

ground of 1-2 mg/L NO_3-N in the upgradient wells (Figure 3). COD increases in shallow groundwater is more difficult to assess, since several of the of the upgradient wells apparently have COD concentrations similar to those of the downgradient compliance wells.

During the first season, changes in TPH-oil concentrations were most significant in Grid 1, seeded with ryegrass (Figure 4). Mean TPH changed from 203 ±452 to 32 ±79 mg/kg (error band = 1 SD). Grid 2, tilled and irrigated, changed from 230 ±268 to 165 ±295 mg/kg. Grid 3, tilled but not irrigated, was essentially unchanged from 55±47 to 54±28 mg/kg. MPN bacterial counts measured from the samples ranged from 3.0×10^5 to 2.7×10^6 colony forming units per gram of soil (Table 1). When the entire landfarm was sampled at the onset of the second season, mean TPH-oil was estimated at 131±38 mg/kg ($p = 0.95$); 45% of the samples were below the remediation criteria of 100 mg/kg TPH. The maximum concentration from the June 1996 sampling was 620 mg/kg, compared to the May 1995 maximum of 2,004 mg/kg.

Spot checks of soil O_2 analysis indicate that at both 1- and 2-ft depths, O_2 concentrations were >5% (data not shown). Soil moisture was maintained at 60 to 70% of field capacity on all areas of the landfarm except for the upper 6 inches of Grid 1 which were seeded to ryegrass (data not shown). Though the ryegrass was green throughout the season, evapotranspiration from this plot exceeded our ability to maintain the desired soil moisture levels.

Metal concentrations were determined to be the greatest potential impediment to returning the landfarm to pasture once degradation was completed. IDEQ conservatively interprets all TPH-oil concentrations as used engine oil with potentially toxic levels of heavy metals, particularly lead. Lead analyses, made routinely during the environmental site investigation, showed 149 soil samples to have a mean lead concentration of 57±57 mg/kg (error band = 1 SD) with maximum concentrations of 640 mg/kg. TCLP (toxicity leaching control procedure) analysis, conducted on two of the most contaminated samples, showed <5 mg/L lead in the resulting leachate, indicating that these soils did not test positive for the toxicity characteristic of hazardous waste. It was determined that 74% of samples with lead concentrations greater than the mean came from depths <2.5 ft below ground surface. Of these, 40% had petroleum concentrations less than detection limit. Since background soil lead concentrations for native/undisturbed Palouse soils range from 10-20 mg/kg, mean lead concentrations for the contaminated soil are 3 to 6 times background.

DISCUSSION

The highly erodible soil at the landfarm was stabilized primarily by the high residue content of the composted manure, available at no charge and in unlimited quantities adjacent to the landfarm site. The unconsolidated and saturated contaminated soils were largely inaccessible to heavy equipment until they froze during the winter. Lack of adequate manure-spreading capability and a relatively short window of opportunity required that manure be spread with a dump truck and bulldozer, resulting in overapplication of compost. The unintended consequences were excessive nitrogen, COD, and TDS concentrations. The high organic content of the leachate apparently may have caused chelation of lead and other ions from the soil. These relatively short-lived problems had limited environmental consequences due to the minimal volumes of stormwater runoff from the site (data not shown). The high organic content of the compost may have helped limit the mobility of the petroleum by sorbing it onto the compost. Heavy fall rains brought earthworm populations to the surface through the contaminated soil, causing near-total earthworm mortality. However, by the next spring earthworms began to recolonize the site. The growth of healthy ryegrass and rapeseed crops indicates that the present soil toxicity is minimal.

Landfarm management was capable of maintaining O_2, soil moisture, and nutrient levels at near optimum conditions for petroleum degradation. Precipitation during the first season was sufficient to prevent Grid 3 from receiving enough moisture stress to differentiate its treatment from that of Grid 2. Since the initial TPH concentrations of Grid 3 were lower than the remediation criteria of 100 mg/kg, the lack of significant change in TPH concentration in Grid 3 is inconclusive. Experience at the site showed that tillage was required less frequently than originally considered necessary. Tilling only when soil moisture levels were at near-optimum conditions prevented compaction of too-wet soils, and avoided pulverizing soils that were too dry. The subsoil tillage was highly effective at fracturing the soils, and O_2 stress was never detected. However, this was true even for the soil planted in ryegrass. This area, Grid 1, had the greatest total change in TPH concentration, possibly due to an enhanced microbial environment in the rooting zone of the ryegrass. Additional controlled research is needed to confirm this possibility.

Residual metal concentration in landfarm soils represents a potential long-term detriment to the site. The bioavailability of the lead is unknown. Even if the lead is not absorbed by forage crops,

some may be ingested in soil by grazing animals. The absorption characteristics of lead in the gut of an animal is beyond the scope of this research. However, it is planned to cover the treated soil with the stockpiled topsoil, reducing the potential for ingestion of lead-contaminated soil by cattle. Additional metal analysis planned for June 1997 will better assess the actual metal concentrations of the landfarm soils. Though the soils exceed the background concentrations for lead, preliminary data indicates that the values are still relatively low, and we are optimistic that the landfarm can be returned to productive agricultural use.

ACKNOWLEDGMENTS

We would like to thank Ken Gill for his assistance in managing the field operations at the landfarm and Jennifer Meehan for microbiological work. We are grateful to the University of Idaho College of Agriculture for lending us some highly productive grazing land for use in landfarming petroleum-contaminated soil. We are optimistic that it will soon be returned, no worse for the wear it has taken during the course of our research.

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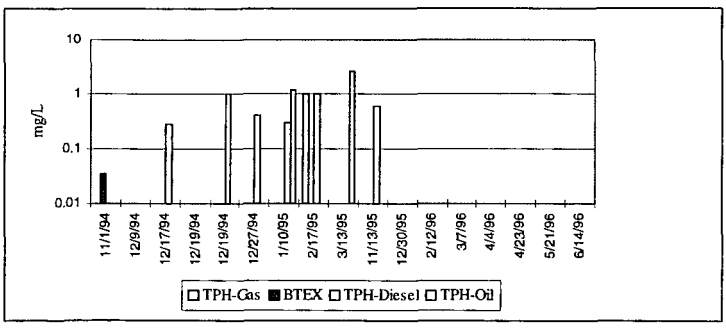

Figure 1. Petroleum concentrations of landfarm stormwater.

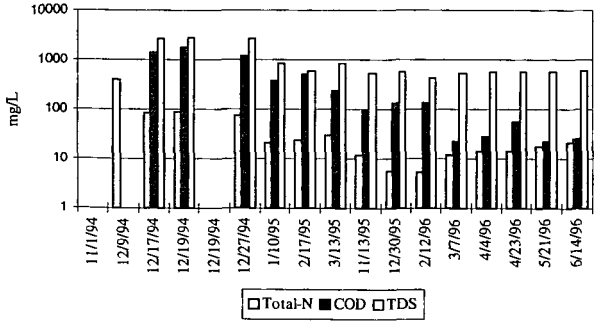

Figure 2. Stormwater analysis. Total N = NH_3-N, NO_3-N, and organic nitrogen; COD = chemical oxygen demand, TDS = total dissolved solids.

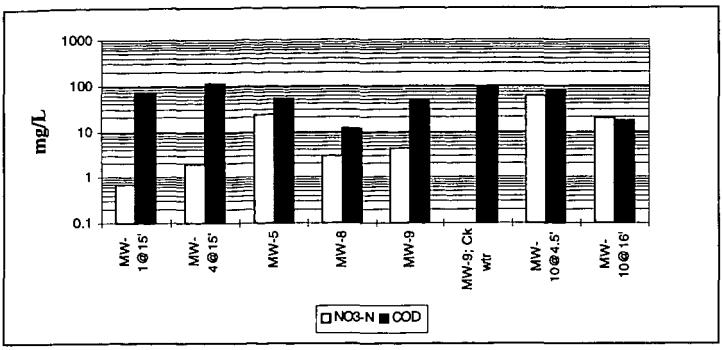

Figure 3. Groundwater concentrations of inorganic compounds, spring 1995. Monitor well cluster MW-10@4.5 ft and 16 ft is immediately downgradient from the landfarm. The 4.5 ft depth is located in the same perched water table that the stormwater collection facility is keyed into. Petroleum was not detected in any of the wells (data not shown). The 16-ft-deep well is located in a sand lens on top of the underlying basalt in a seasonal aquifer that is moderately protected by a clayey-silt aquitard. MW-9 is downgradient; the other wells are either upgradient or lateral to the landfarm. The nitrate and COD concentrations of wells, except for MW-5, probably indicate background levels on fertile pasture under range management. Elevated nitrate concentrations in MW-5 may result from lateral groundwater interflow from the landfarm, or the presence of cattle manure.

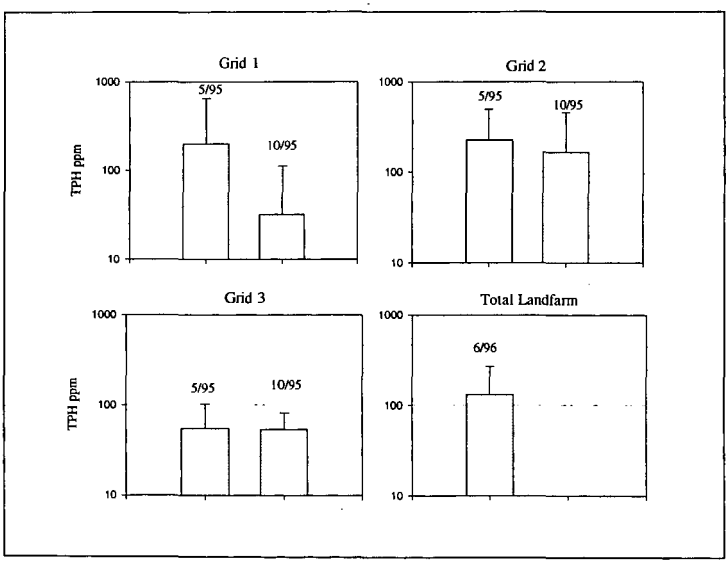

Figure 4. Total petroleum hydrocarbon (TPH) concentrations over time. Grid 1 (ryegrass and irrigation); Grid 2 (tillage and irrigation); Grid 3 (tillage only). Rapeseed and ryegrass were planted over the majority of the landfarm in October 1995. The total landfarm was sampled in June 1996, when a mature rapeseed crop covered the majority of the landfarm, except for Grid 1, which had a mature ryegrass covercrop. Error bars = 1 SD.

Date	Grid 1	Grid 2	Grid 3
May, 1995	$7.69 \pm 0.75 \times 10^5$	$1.61 \pm 0.12 \times 10^6$	$3.47 \pm 0.2 \times 10^6$
October, 1995	$2.67 \pm 0.08 \times 10^6$	$1.03 \pm 0.07 \times 10^6$	$3.01 \pm 0.0 \times 10^5$

Table 1. Most probable number bacterial counts in colony forming units per gram of soil.