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Review

Bioventing soils contaminated with petroleum hydrocarbons

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SUMMARY

Bioventing combines the capabilities of soil venting and enhanced bioremediation to cost-effectively remove light and middle distillate hydrocarbons from vadose zone soils and the groundwater table. Soil venting removes the more volatile fuel components from unsaturated soil and promotes aerobic biodegradation by driving large volumes of air into the subsurface. In theory, air is several thousand times more effective than water in penetrating and aerating fuel-saturated and low permeability soil horizons. Aerobic microbial degradation can mitigate both residual and vapor phase hydrocarbon concentrations. Soil venting is being evaluated at a number of U.S. military sites contaminated with middle distillate fuels to determine its potential to stimulate in situ aerobic biodegradation and to develop techniques to promote in situ vapor phase degradation. In situ respirometric evaluations and field pilot studies at sites with varying soil conditions indicate that bioventing is a cost-effective method to treat soils contaminated with jet fuels and diesel.

INTRODUCTION

Recent investigations indicate that several hundred-thousand underground tanks containing petroleum fuels may be leaking [5,6]. While the greatest number of leaking tanks contain gasoline, the largest tanks frequently contain less volatile petroleum distillate fuels, such as diesel and jet fuels. Because these middle distillate fuels are considerably different in composition than gasoline, unique remediation methods for their destruction in soils may be required. Unlike gasoline fuels, which may contain up to 50% alkylbenzenes [30,35], middle distillate fuels (e.g., diesel, kerosene, JP-4, JP-5, and JP-8) generally contain only 5 to 12% alkylbenzenes and consist mainly of C_5 to C_{16} normal and branched chain alkanes [10,11]. While most alkylbenzenes are slightly to moderately soluble in water, the higher molecular mass alkanes have extremely low solubilities in water [48]. Lower molecular mass alkanes and alkylbenzenes generally display greater volatilities and water solubilities than their higher molecular mass counterparts. Thus the volatilities and water solubilities of the hundreds of hydrocarbon components of gasoline are significantly higher than those of middle distillate fuels, which favors both their evaporation into the gaseous phase and dissolution into the water phase. Middle distillate fuels have liquid densities only slightly higher than gasoline and float on the groundwater table [9].

IN SITU REMEDIATION TECHNOLOGIES

Potentially cost-effective methods for destroying or detoxifying wastes include treatment in situ. In situ technologies that have been used to remediate fuel-contaminated sites include soil washing, low-temperature thermal treatment, soil venting, and enhanced bioreclamation.

Soil washing involves injecting a synthetic surfactant or solvent into the contaminated zone to promote greater release of hydrophobic contaminants to the aqueous phase. Unfortunately, this technology has met with limited success in both laboratory [1] and field evaluations [29]. Surfactants may also serve to increase bioavailability; however, when 53 synthetic surfactants were screened and tested for their ability to enhance natural biodegradation rates in jet fuel-contaminated soils, actual degradation rates were either unaffected or inhibited [1].

Low-temperature thermal methods for treating fuel contamination in situ include heated gas or steam injection [7] and radio frequency heating of inplace soils [34]. Heated air or steam injection technologies can be

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energy and equipment intensive, resulting in costs of about \$300/cu yd [7]. Thus these methods are not presently being routinely used to remediate hydrocarbon spillage.

Soil venting has been widely used in the field for removing volatile hydrocarbons from the subsurface, especially from the unsaturated vadose zone above the groundwater table [3,6,38]. It involves pulling a vacuum in the vadose, usually through vapor extraction wells or dewatering points emplaced in the contaminated zone. This stimulates in situ volatilization, and contaminant vapors are drawn to the extraction point. Typically, much of the contamination resulting from a hydrocarbon release in the subsurface resides in the vadose zone and capillary fringe immediately above the water table, where it serves as a reservoir for prolonged contamination of groundwater. Since the diffusion of gases averages about 10 000 times those of solutes, soil venting has the potential to remove volatile contaminants from less permeable soils by diffusion. The transport of volatile organics through the soil gas phase will be affected by sorption on soil particles, partitioning into soil water, temperature, and biodegradation [32]. Soil venting is a potentially cost-effective technology for removing volatile components from soils, with a cost as low as \$15/ton (\$20/cu yd) [22]. Another estimate is about \$70/gallon of gasoline fuel without off-gas treatment; if activated carbon sorption is used to treat the vapors, the cost approximately doubles and the hydrocarbons have not been destroyed [14].

Enhanced bioreclamation, when applied in situ, involves stimulating the development and metabolic capabilities of soil microorganisms that degrade or detoxify contaminants residing within the soil and groundwater. Although in situ bioreclamation can involve the addition of previously isolated specialized microorganisms, this is not generally recommended for the degradation of petroleum products because they consist of several hundred individual compounds and many microorganisms present in the natural environment possess fuel hydrocarbon-degradative abilities [2,24,31]. The most common methods presently used to stimulate soil microflora include the addition of oxygen (or other electron acceptors) and nutrients (usually soluble nitrogen and phosphorus compounds). In situ bioreclamation is considered to be effective only in more permeable soils such as sandy profiles (permeabilities greater than 10^{-4} cm/s). A detailed account of current requirements and techniques used for in situ bioreclamations is summarized by Thomas and Ward [41]. Bioreclamation is the only currently available in situ technology that, in itself, can totally destroy most organic contaminants, other than considerably more costly in situ vitrification [4]. The majority of compounds found in refined petroleum products are readily biodegradable, especially under aerobic conditions [2,24]. Yet most in situ bioreclamations of fuel-contaminated soils require extended time periods for adequate site cleanup or have met with questionable success. Enhanced bioreclamation has the potential of being an effective remediation method, but additional research is required before this technology may be considered beyond the experimental stage. Because of frequent field problems and lack of non-proprietary information associated with enhanced bioreclamation of fuel contamination, cost estimates vary widely from \$26 to \$200/ton (\$35–\$270/cu yd) [11,22,45]. Another cost estimate, for a non-research project, is \$230–\$300/gallon of residual fuel in the soil [22].

PROBLEMS WITH ENHANCED BIORECLAMATION

As typically applied in the U.S., enhanced bioreclamation characteristically uses water to carry oxygen and nutrients into the subsurface. This is a logical extension of previously developed wastewater treatment technology and pump-and-treat technology used to surface-treat groundwater containing soluble contaminants. However, using water to drive in situ bioremediations, especially of hydrophobic contaminants common to most fuels, has several major drawbacks. Two of the most important obstacles include difficulty in aerating heavily contaminated soils and groundwater [18,20,23,44], and inability of the hydrocarbon-degrading microflora to interact with hydrophobic fuel components [12,28].

The most common methods used today to deliver oxygen gas into and through the subsurface to promote aerobic biodegradation involve sparging groundwater with air or oxygen gas or adding hydrogen peroxide to pumped and re-injected groundwater. If air is sparged into groundwater, maximum dissolved oxygen concentrations of approximately 8–9 mg/l would be attained; sparging pure oxygen gas would increase this concentration about five-fold. Even when pure oxygen is used, approx. 75 000 kg of water must be delivered to the subsurface to mineralize 1 kg of fuel hydrocarbons. This compares with delivering 12 000 kg of water when hydrogen peroxide is added at 500 mg/l as the oxygen source [15]. However, hydrogen peroxide is rapidly decomposed in soils by free iron and microbial hydroperoxidase enzymes, especially catalase [1,37]. Hydrogen peroxide stability and toxicity to microorganisms seems to be dependent on specific site conditions, but rapid breakdown was noted during laboratory [20,36] and field studies [18]. Although molecular oxygen released from decomposed hydrogen peroxide in soil column studies was utilized within the columns, only 55% of it was transferred to the water phase and hydrogen peroxide concentrations at low as 100 mg/l may have caused microbial inhibition [20]. Hydrogen peroxide decomposition rates for an in situ field study at Eglin Air Force Base (AFB) in Florida were consistently much higher than oxygen utilization rates and the majority of the oxygen gas appeared to be released to the atmosphere within a meter of the injection well [18]. Degradation of high molecular mass alkanes under denitrifying or obligate anaerobic conditions has not been shown to be feasible [24]. Such recent findings suggest perhaps that water is a poor medium in which to transport sufficient oxygen to maintain aerobic metabolism for degrading large volumes of fuels sorbed to soils [18,20].

Hydrophobic compounds are often recalcitrant to biodegradation because membrane-bound enzymes are unable to bind with the target molecules [26]. Since microorganisms do not proliferate in hydrophobic fuel fractions, such as the alkane fraction, biodegradation rates are often limited by the surface area of the water-immiscible fraction in soil pores that is in contact with the soil pore water. Water and hydrophobic fuel components do not readily intersperse with one another and the hydrophobic component could also retard solubilization of the more soluble aromatic fraction. The inability of water to interact with fuels occluded in soil pores has been confirmed by both laboratory [46] and field [8] studies. Ways to help overcome these problems include using synthetic emulsifiers, promoting conditions that favor production of bioemulsifiers secreted by bacteria, and vaporizing the volatile fraction. As noted, problems have been encountered while attempting to increase biodegradation rates by using biodegradable synthetic surfactants [1,29]. Furthermore, laboratory studies using biosurfactants have indicated that the interactions are complex, with both stimulation [33] and inhibition [13] of alkane biodegradation being observed. Inhibition of alkane degradation by the bioemulsifier, emulsan, was thought to be caused by interference with passage of hydrocarbons through the bacterial membrane [13]. If the hydrocarbon surface area is increased by volatilization, then no interfering molecules will be involved. Also, volatilization favors the removal of lower molecular mass alkanes and aromatics, which may be more toxic to microorganisms [2].

COMBINING SOIL VENTING AND BIOREMEDIATION: BIOVENTING

Bioventing is an innovative in situ technology that combines the physical processes of soil venting with the degradation potential of enhanced biodegradation. With soil venting, a vacuum is pulled on vapor extraction wells emplaced in the vadose zone within or adjacent to the zone of contamination. The negative pressure that develops in the soil pores greatly accelerates volatilization of hydrocarbon compounds sorbed to soils in the vadose zone. By lowering the groundwater table simultaneously through emplacement of dewatering points at and just beneath the groundwater table, additional vapor phase removal can occur in soils beneath the normal water table. In addition, using dewatering points allows for simultaneous removal of groundwater, free hydrocarbon product, and the vapor phase.

Besides fuel vapor removal, soil venting can greatly increase the rate of air diffusion into the subsurface to help satisfy the vacuum. The air likewise has the potential to diffuse through soils having low water permeability because of the greater diffusivities of gases compared with liquids. Since air contains over 200 000 parts per million of oxygen, soil venting can quickly overcome the oxygen deficits that usually occur in soils heavily contaminated with fuels. This oxygen can then serve as the terminal electron acceptor for aerobic biodegradation of fuel hydrocarbons, as discussed by Connor [6], Wilson and Ward [47] and Hinchee et al. [15,19]. Since middle distillate fuels are comprised mainly of low to medium volatility compounds, the primary purpose for soil venting in the contaminant zone is to stimulate aerobic biodegradation. Volatilization of the fraction with higher vapor pressures should also function to increase fuel surface areas and dilute the main hydrocarbon plume.

The value of using soil venting to stimulate in situ biodegradation of fuel hydrocarbons was first realized about a decade ago at the Texas Research Institute. In laboratory studies more than a third of the gasoline removed from vented soils may have been removed by biodegradation [39,40]. Dutch researchers first publicized field observations of venting-induced biodegradation in 1986 [36,49]. Bennedsen et al. [3] showed that soil venting promoted the movement of large quantities of atmospheric oxygen into the unsaturated soil profile, while Connor [6] suggested that biodegradation was stimulated by soil venting by noting temperature increases in vented soil gas. Based on limited published data, Ely and Heffner of Chevron Research patented a process that involves fuel vapor extraction from soils to enhance in situ biodegradation [10].

A typical approach to soil venting for gasoline removal is to pump large volumes of soil gas, usually from permeable soils, and to sorb the vapors on activated carbon, to flare or catalytically incinerate them, or to emit the hydrocarbons to the atmosphere. The release of volatile hydrocarbons from the soil during soil venting of fuel contamination may be greatly reduced if the rate of venting is equivalent to the rate of vapor phase biodegradation in the soil profile [27]. This should be accom-

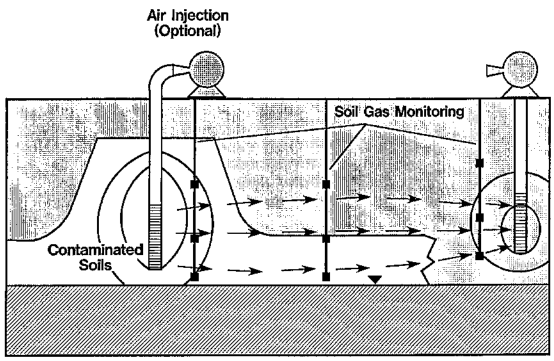

Fig. 1. Potential configuration for enhanced bioreclamation through soil venting (Air withdrawn from clean soil).

plished more easily when venting middle distillate fuel-contaminated soils than when venting gasoline-contaminated soils. Greater venting rates could be achieved if the vapor flow path is increased. This could be accomplished by emplacing vapor extraction wells outside of and lateral to the contamination zone but still within the zone of influence. Alternatively, air can be forced into the contamination zone and vapor can be extracted from uncontaminated soil. This should also promote complete mineralization of most of the vapor within the soil profile. These configurations are shown in Fig. 1.

FIELD APPLICATIONS

The number of field demonstrations and pilot applications of bioventing reported in the literature is limited (Table 1). With the exception of the Hill AFB, UT, site, the kinetics (where they are reported) appear to be first order. Given the relatively high concentrations relative to the rates, it appears that hydrocarbons are not generally limiting. Miller and Hinchee [27] found that neither nutrients nor added soil moisture were limiting at the Tyndall AFB, FL, site. Results of a laboratory study suggest that inorganic nutrients are limiting at the Hill AFB site [16]; however, this has not been field-verified.

Hinchee et al. [17] developed a short-term in situ respirometry test to determine rates of biodegradation under field conditions. The test consists of injecting air into the vadose zone to provide aerobic conditions. The rate of oxygen utilization is then measured to determine biodegradation rates. Controls consist of similar respirometry measurements in an uncontaminated location, and injection of an inert gas into a contaminated location. This assures that the respiration measured is a result of hydrocarbon degradation verifies and the integrity of the samples.

TABLE 1

Summary of reported field applications of bioventing

Site	Scale of application	Contaminant	Biodegradation rates	Reference
Hill AFB, Utah	Full scale, 2 years	JP-4 jet fuel	Up to 10 mg/(kg day)[a,b]	[19]
Tyndall AFB, Florida	Field pilot, 1 year	JP-4 jet fuel	2–20 mg/(kg day)	[27]
The Netherlands	Undefined	Undefined	2–5 mg/(kg day)[b]	[41]
The Netherlands	Field pilot, 1 year	Diesel	8 mg/(kg day)	[42]
Undefined	Full scale	Gasoline and diesel	50 kg/(well day)[c]	[10]
Undefined	Full scale	Diesel	100 kg/(well day)[c]	[10]
Undefined	Full scale	Fuel oil	60 kg/(well day)[c]	[10]
Patuxent River NAS, Maryland	Short-term (1-week) pilot	JP-5 jet fuel	3 mg/(kg day)	[17]
Fallon NAS, Nevada	Short-term (1-week) pilot	JP-5 jet fuel	5 mg/(kg day)	[17]
Eielson AFB, Alaska	Short-term (1-week) pilot	JP-4 jet fuel	1–10 mg/(kg day)	[17]

[a] Rates reported by Hinchee et al. [19] were first order with respect to oxygen; for comparative purposes, these have been converted to zero order with respect to hydrocarbons at an assumed oxygen concentration of 10%.

[b] Rates were reported as oxygen consumption rates; these have been converted to hydrocarbon degradation rates assuming a 3 : 1 oxygen to hydrocarbon ratio.

[c] Units are in kg of hydrocarbon degraded per 30 standard cubic feet per minute (SCFM) extration vent well per day.

RESEARCH NEEDS

Bioventing has been performed and monitored at several field sites contaminated with middle distillate fuels, mainly JP-4 jet fuel. Yet the effects of environmental variables on bioventing treatment rates are poorly understood. In situ respirometry at additional sites with drastically different edaphic conditions has further defined environmental limitations and site-specific factors that are pertinent to successful bioventing. However, the relations between respirometric data and actual bioventing treatment rates have not been clearly determined. Additional field respirometry and closely monitored field pilot bioventing studies at the same sites are needed to determine what types of contaminants can be successfully treated in situ by bioventing and what the environmental limitations are. Studies to date clearly show that many notions regarding the factors that control bioventing rates can be wrong. For example, active respiration at a subarctic site at Eielson AFB near Fairbanks, Alaska, suggests that good hydrocarbon degradation can occur in situ at locations that are continually subjected to a cold environment. Failure to accelerate biodegradation rates by adding nitrogen fertilizer to biovented soils that contain low nitrogen levels indicates that nutrient addition at some sites may not be required. Also, fine-grained moist clayey soils in Nevada were readily aerated and showed excellent aerobic respiration, indicating that bioventing is feasible in soils having low permeabilities.

Vapor phase biodegradation occurs and it can take place in situ. The question of how soil sorption and partitioning of volatile organic compounds into soil air affects biodegradation rates was proposed earlier by McCarty [25]. This question remains mostly unanswered since the movement of the vapor phase in soils is complex and dependent on changing soil environmental conditions. Although degradation of chlorinated alkanes and aromatics occurs in situ, previous studies have involved treating these contaminants in groundwater. Since bioventing has not been investigated for volatile or semivolatile organics other than fuel hydrocarbons, additional research is required for determining its applicability in degrading or detoxifying non-petroleum hydrocarbons.

Bioventing rates need to be determined under varying vapor extraction rates since an important purpose for bioventing is to biodegrade the vapor within the soil profile. The minimal soil aeration levels that provide for high degradation rates must be determined under different soil conditions. Interaction of the vapor phase with soil particles and microorganisms in the uncontaminated soil profile needs further research in both the laboratory and in the field.

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