



Bioslurry treatment of a soil contaminated with low concentrations of total petroleum hydrocarbons

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Abstract

The objective of this study was to investigate the degradation patterns of petroleum hydrocarbons during bioremediation of soils containing low levels of contaminants. The study was conducted in pilot-scale bioslurry reactors (70 l) under aerobic conditions. The reactors were equipped with a process-gas-recirculation system to ensure complete containment and eventually complete degradation of all contaminants. The concentrations of benzene, toluene, ethyl benzene, and xylenes (BTEX-compounds) and of naphthalene, anthracene, and phenanthrene were found to decrease rapidly. But, polyaromatic hydrocarbons (PAHs) containing >3 aromatic rings did not show significant biodegradation. Addition of rapidly metabolizing substrates such as sodium acetate and/or phenanthrene did not enhance the degradation of PAHs containing >3 aromatic rings. However, the augmented phenanthrene was rapidly metabolized.

Keywords: Bioremediation; PAH; Slurry bioreactors; BTEX; Aerobic process

1. Introduction

Petroleum hydrocarbon-based fuels are one of the most prevalent soil pollutants in the United States of America. The source of this contamination is usually an accidental spill or improper storage. Contamination from leaking underground storage tanks is a particularly large problem. Use of tar and creosote in wood-treatment has also contributed to considerable contamination of surface and subsurface environments with aromatic as well as chlorinated hydrocarbons [1]. Benzene, toluene, ethyl

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benzene, xylenes (BTEX chemicals) and polyaromatic hydrocarbons (PAHs) are the key chemical-constituents of petroleum fuels. These chemicals are also present in flue-gas condensates and in refinery and coal-gasification wastes. Several members of this family of chemicals are considered toxic [2], teratogenic [3], mutagenic and carcinogenic [4]. Therefore, these compounds are categorized as priority pollutants by the US Environmental Protection Agency. Due to their adverse impact on human health and regulatory concern, several methods have been explored for restoration of sites contaminated with BTEX and PAHs.

Until recently, treatment of soils containing organic contaminants has essentially been limited to thermal-destruction technologies. Although incineration destroys most organic contaminants, it is expensive and often difficult to implement due to political and regulatory issues. Recent developments in innovative treatment technologies based on physical, chemical, and biological processes, offer remediation alternatives that are technically, economically, and politically attractive. Among these, biological treatment systems (biotreatment) are considered to be very promising [5, 6].

Soil biotreatment systems can be divided into two main implementation categories; in situ and above-ground. In situ systems use natural environments in which additives are strategically injected in an attempt to create the necessary treatment conditions. Successful introduction of the additives into potential in situ treatment zones can be very challenging. The aquifers are open systems with only a partial access for determining the extent and location of contamination and for introduction of the required additives.

On the other hand, the above-ground biotreatment systems use engineered bioreactors which can be designed and equipped with various process-control instruments to create ideal conditions for biodegradation. Use of above-ground systems requires excavation of the contaminated soils followed by treatment in bioreactors. Examples of such systems include land farming [7, 8], composting [9, 10] and bioslurry treatment [11].

Among the above-ground biotreatment systems for contaminated soils [12], slurry reactors offer maximum control, most flexibility, and usually the highest rates of biodegradation. Bioslurry treatment of soils and sludges is a relatively new treatment technology for destruction of contaminants sorbed to solids and/or in solution in the interstitial pore water. It is essentially an engineering reconfiguration of other more widely used biotreatment technologies, such as land treatment and composting, that have been successfully used for decontamination of soils and sludges [7]. Bioslurry treatment is similar to other soil and sludge biotreatment technologies in terms of microbiological interactions and contaminant degradation pathways [6]. However, it differs from the other technologies because the availability of contaminants, electron acceptors, nutrients, and other additives to the microbial populations is enhanced in a slurry, thus substantially increasing the rates of biodegradation [6].

A typical implementation scenario for bioslurry systems has been illustrated in Fig. 1. Soils are excavated and prescreened to remove soil particles that are too large for treatment in the bioslurry reactors. After screening, the soil is dispersed in water (typically 20–50% solids by weight) and introduced into an agitated bioreactor for

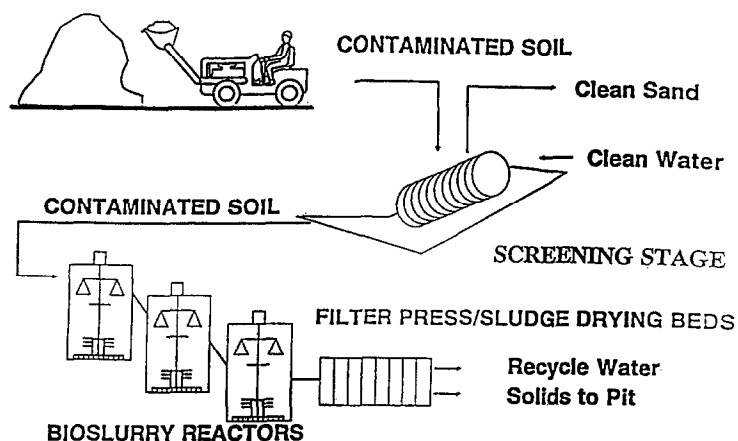


Fig. 1. Slurry process for soil bioremediation.

treatment. Bioslurry reactors can be operated under aerobic or anaerobic conditions. For aerobic systems, oxygen is delivered either by sparging air or oxygen into the soil/water slurry or by addition of hydrogen peroxide. Once treatment levels are met, the treated soils are removed, dewatered, and disposed. Cost estimates for bioslurry treatment of petroleum hydrocarbons at various sites range from \$90 to \$130 per cubic yard [13].

Biodegradation of BTEX and PAHs has recently been summarized by Gunnison et al. [14]. These chemicals are commonly found in our environment due to their large production volumes and widespread usage. Microorganisms capable of degrading these chemicals are quite ubiquitous. Biodegradation occurs under aerobic [15, 16] as well as anaerobic conditions [17, 18]. In general, the rates under aerobic conditions are considerably higher than those under anaerobic conditions. BTEX and low molecular weight aromatic hydrocarbons are generally easily biodegraded [19]. On the other hand, PAHs containing four or more benzene rings are highly recalcitrant [15]. This group encompasses the most carcinogenic compounds also [15]. In natural systems, the higher molecular weight PAHs are cometabolized in presence of phenanthrene, anthracene, or other faster-degrading chemicals that act as primary carbon and energy sources [20]. Several publications have reported successful bioremediation of soils containing relatively high concentrations of PAHs [21–23]. When present at high concentration levels, the faster-degrading contaminants can provide energy for significant cometabolization of high molecular weight PAHs. When the contaminant concentrations are low, the faster-degrading primary chemicals may be exhausted quickly before any significant degradation of high molecular weight PAHs occurs. This is a source of major concern for soils with low-level contamination. However, reports describing soils with low-level PAH contamination are conspicuous by their absence in published literature. The objective of this study was to investigate the biodegradation of low levels of BTEX and PAH contaminants in a soil using a slurry reactor with process-gas-recirculation.

2. Materials and methods

The contaminated soil sample used in this study was collected from a US Army installation where storage of petroleum fuels (gasoline) in leaking underground storage tanks resulted in the localized contamination of sub-surface soils. The contaminant concentrations in soil are presented in Table 1. The soil contained high levels of clays and silts indicating poor hydraulic conductivity. This soil, therefore,

Table 1
Concentrations of PAHs and volatile chemicals

	Initial (mg/kg)	After			Percentage reduction ^a in 22 days (mg/kg)
		2 days	8 days (mg/kg)	22 days	
<i>Soil phase</i>					
Naphthalene	1.1	0.23	0.14	0.04	96
Phenanthrene	0.44	0.31	0.18	0.11	75
Anthracene	0.11	0.09	0.06	0.03	73
Fluoranthene	0.67	0.70	0.40	0.29	57
Benzo(a)anthracene	0.32	0.33	0.23	0.16	50
Chrysene	0.40	0.40	0.33	0.27	33
Pyrene	0.56	0.60	0.61	0.44	22
Benzo(b)fluoranthene	0.19	0.18	0.28	0.27	
Benzo(a)pyrene	0.21	0.18	0.26	0.22	
Benzo(k)fluoranthene	0.28	0.24	0.29	0.21	
Fluorene	0.07	0.05			
Indeno[1,2,3-cd-]pyrene	0.06	0.06	0.11	0.12	
Benzo(g,h,i)perylene	0.06	0.06	0.11	0.04	
					(In 2 days)
Xylene (total)	19.00	0.30			98
4-Methyl, 2-pentanone	14.00	0.28			98
Toluene	1.30	0.06			95
Ethylbenzene	0.35	0.02			94
Benzene	0.16	0.01			94
Chlorobenzene	0.15	0.008			95
TRPH	70.00	40.0	<25		82 ^b
	Initial sample (mg/l)	After		Percentage reduction in 8 days	
		3 days (mg/l)	8 days (mg/l)		
<i>Aqueous phase</i>					
Naphthalene	0.25	–			100
2-Methyl naphthalene	0.15	–			100
Benzene	0.16	–			100
Xylene (total)	5.65	0.0023			100
TRPH	25.00		<0.5		99 ^b

^a Percentage reduction = $(1 - \text{final concentration}/\text{initial concentration}) * 100$.

^b On the basis of half of detection limit.

was not considered amenable to in situ-based technologies such as bioventing and bioflooding.

The biodegradation studies were carried out in three 70 l (working volume) Biolift™ slurry reactors manufactured by Eimco Inc. (Salt Lake City, Utah). The reactors were fitted with shaft-mounted raker arms, located close to the reactor bottom, that forced the settled soil particles to the center [12]. An air-lift system lifted the settled particles from the center of the reactor bottom and discharged them at the slurry surface. Oxygen was introduced into the soil slurry through three air-sparging membrane-canisters mounted on the raker arms. The reactors were equipped with a gas-recirculation system in order to prevent any loss of volatile chemicals from the reactor due to air sparging. Typical bioslurry reactors in the past have had to utilize activated carbon for capturing volatilized contaminants from process off-gases. With the use of gas-recirculation system, there were no process-off gases and this permitted a reliable estimate of the extent of bioremediation.

The organic content in the gas phase was continuously monitored using a photoionizing detector (PID) equipped with data logging capability. Gas-phase carbon dioxide and oxygen concentrations were monitored using in-line gas analyzers (Rosemount brand, Houston, Texas). Carbon dioxide which is produced as a result of microbial activity, was removed using an in-line packed-tower caustic-scrubber that was activated when the CO₂ levels in the gas phase exceeded 1%. Pure oxygen was injected into the process gas line on an as-needed basis to maintain oxygen levels of at least 19% in the gas phase. Fig. 2 illustrates the gas-recirculation system used in this study.

A 26.7% (w/w dry weight basis) slurry of contaminated soil in tap water was loaded into the reactors. The slurry was supplemented with nutrients (ammonium

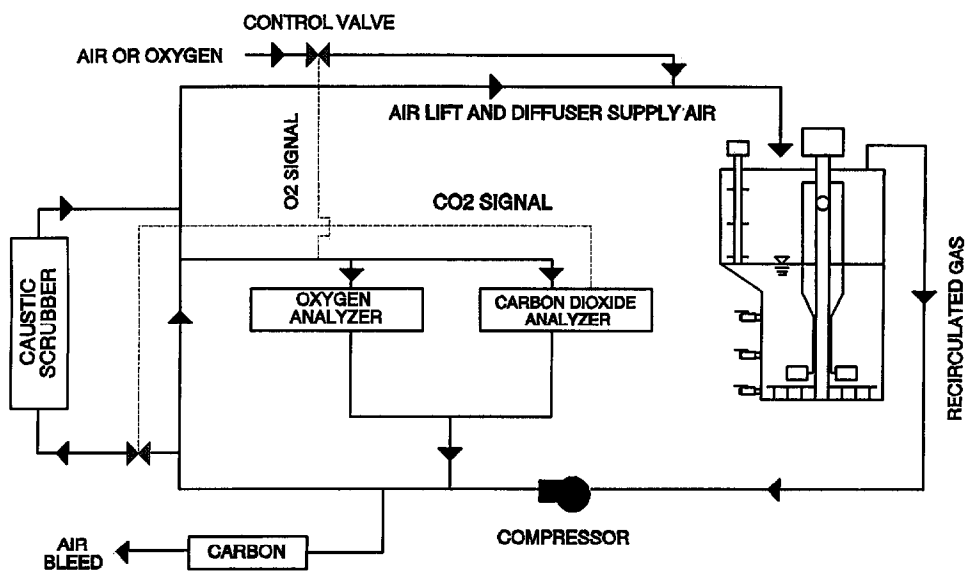


Fig. 2. Schematics of process gas recirculation system.

sulfate as a nitrogen source and KH_2PO_4 as a phosphate source). The study was conducted without any bioaugmentation and all the biodegradation activity was attributed to the indigenous microbial population in the contaminated soil. The liquid samples were analyzed every other day for ammonia nitrogen and phosphate (PO_4^{3-}) concentrations, and the nutrients were added in sufficient quantities in order to maintain 50 mg/l ammonia nitrogen and 10 mg/l phosphate in the slurry. Dissolved oxygen, pH, temperature, suspended solids, volatile suspended solids, total recoverable petroleum hydrocarbons (TRPH), and oxygen uptake rates were also monitored regularly. The concentration of dissolved oxygen remained >5.0 mg/l throughout the course of this study. The slurry pH was maintained at 7.2 ± 0.4 and the temperature at 24 ± 3 °C. Measurements of oxygen uptake rate were conducted by withdrawing a sample of slurry in a biochemical oxygen demand (BOD) bottle to zero head space and measuring the rate of change of dissolved oxygen concentration with a dissolved oxygen probe for a period of 10 min. The total volume of samples was insignificant compared to the volume of slurry in the reactors. On the 42nd day of the experiment and every eighth day for the next three weeks, the first reactor was supplemented with sodium acetate (66.73 g), the second reactor with nonionic surfactant Tween 80 (66.73 g), and the third reactor with a mixture of sodium acetate (6.73 g) and phenanthrene (6.73 g). These supplementations were made to evaluate the effect of different supplements on the residual concentrations of high molecular weight PAHs.

Ammonia nitrogen in the samples was measured with a 901 Ion Analyzer and an Orion 95-121 ion-specific probe. Phosphate content was analyzed using Hach kit PO-24 which uses a digestion/colorimetric technique. The concentrations of total recoverable petroleum hydrocarbons (TRPH) and of PAHs and BTEX (benzene, toluene, ethyl benzene, and total xylene) in soil and liquid samples were determined either by USEPA-approved methods [24, 25] or by standard methods [26].

3. Results and discussions

The biological activity, as indicated by oxygen uptake rate measurements, in all the three reactors was evident from the beginning and it was persistent throughout the experiment. The average oxygen uptake rate in the first six weeks was ~ 3 mg/l/h. Addition of supplements (sodium acetate, phenanthrene, or Tween 80) to the slurries resulted in short term increases in the oxygen uptake rates which quickly settled around the average values. The concentrations of PAHs and volatile chemicals in the soil and aqueous phases, initially and at selected times, are presented in Table 1. Percent reduction in the concentrations were calculated from these data and these too are presented in Table 1.

3.1. BTEX and TRPH degradation

Xylenes and 4-methyl-2-pentanone were the predominant volatiles in the soil phase (Table 1) and these were rapidly degraded in the slurry reactors. The total soil-phase

concentration of BTEX chemicals dropped from 21 mg/kg to below detection limit within two days. The total BTEX level in aqueous phase also reduced from 5.81 to 0.0023 mg/l within three days, which is well below the EPA's off-site aquifer cleanup guideline of 0.150 mg/l BTEX [27]. By the seventh day, BTEX levels in the liquid phase were also undetectable.

The measurements of total volatile organics in the gas phase have been presented in Fig. 3. Though the initial values started at ~ 7 ppm (v/v), these quickly dropped to < 0.2 ppm within two days. This suggests that the reductions in the concentrations of BTEX chemicals are not due to transfer to the gas phase. Since the gas-phase was contained within the system due to the gas recirculation, the reductions in BTEX concentration are attributable only to microbial degradation.

The aqueous-phase concentration of total xylenes dropped from 5.65 mg/l to 0.073 mg/l in two days and below the detection limits ($5 \mu\text{g/l}$) in three days; it remained below the detection limits during the rest of the experiment. Their concentration in soil phase also dropped from 19 to 0.3 mg/kg in two days. Similar trends were observed also for the concentrations of benzene, toluene, and ethyl benzene.

The concentration of TRPH in the soil phase dropped from an initial value of 70 to < 25 mg/kg (detection limit) by the end of eighth day. The aqueous-phase concentration of TRPH was 25 mg/l in the first sample taken immediately after mixing the slurry and it was reduced to < 0.5 mg/l (detection limit) within the same time period. Based upon these observations, it may be concluded that slurry-phase biotreatment of soils is very effective in reducing BTEX and TRPH contamination even at these low levels.

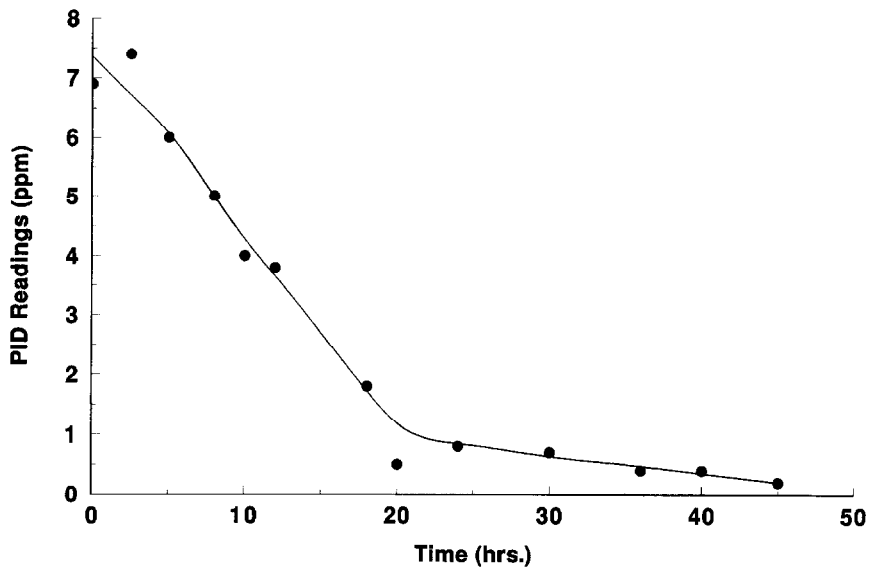


Fig. 3. Changes in head-space contamination in the slurry bioreactor, measured by a photo-ionizing detector.

3.2. PAH degradation

The concentrations of the different PAHs in the soil and aqueous phases at selected times during the experiment are presented in Table 1 while the detailed changes in soil-phase concentration of selected PAHs with time are presented in Figs. 4 and 5. Naphthalene was the contaminant with highest concentration in the soil; its levels dropped from 1.1 mg/kg soil to <0.05 mg/kg by the 21st day of the study (Fig. 4). The most rapid change was observed in the first few days. Similar degradation patterns were also observed for phenanthrene and anthracene. The concentration of phenanthrene decreased from 0.44 to 0.11 mg/kg (Fig. 5) and that of anthracene decreased from 0.11 to 0.03 mg/kg in 20 d.

The concentrations of higher molecular weight PAHs are also presented in Table 1. Small changes in the concentrations of benzo(a)anthracene and chrysene were registered during the first 20 days, but then did not decrease any more. No significant reductions in the concentrations of pyrene and benzo(a)pyrene were observed during the course of this experiment (data not shown). Fluoranthene, benzo(b)fluoranthene, benzo(k)-fluoranthene, benzo(g,h,i)perylene, and indeno(1,2,3-cd)pyrene also behaved in a similar fashion. It was suspected that rapid disappearance of primary carbon and energy sources to support degradation of these more complex PAHs may be responsible for this lack of degradation. These PAHs also possess low solubilities and are strongly sorbed on the soil. Hence, bioavailability may also play an important role. The first possibility was explored by addition of sodium acetate (~3000 mg/kg soil) in one reactor and a mixture of sodium acetate and phenanthrene (~300 mg/kg soil each) to another reactor on the 42nd day of the study. In both of the reactors, microbial activity increased, as measured

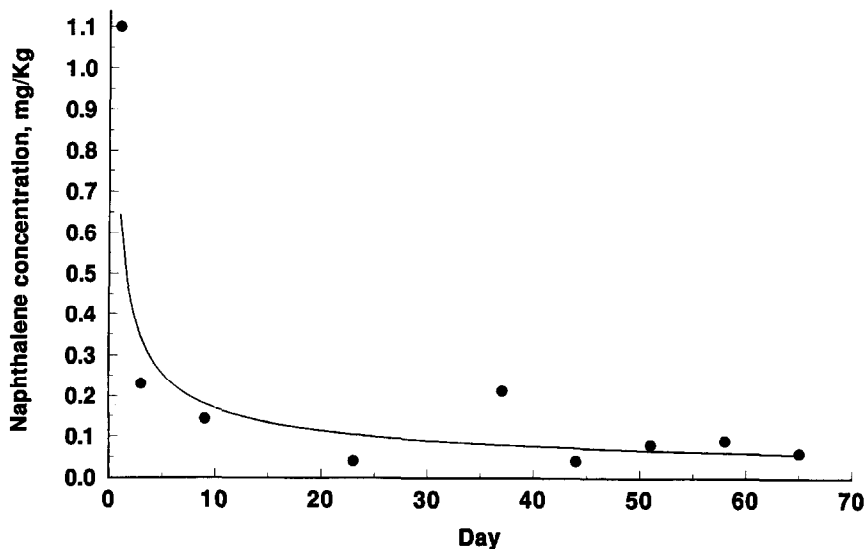


Fig. 4. Soil-phase naphthalene concentration in the slurry bioreactor.

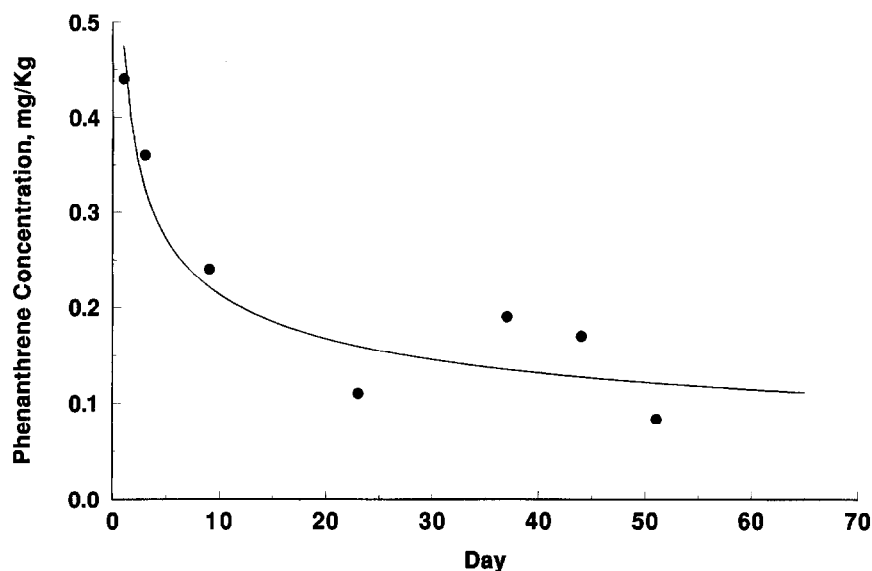


Fig. 5. Concentration of phenanthrene in the soil-phase of the slurry bioreactor.

by an increase in oxygen uptake rates. Phenanthrene disappeared rapidly from the soil slurry (down from >300 mg/kg on the 42nd day to 88 mg/kg on the 44th day). But no significant change in the concentrations of higher molecular weight PAHs (pyrene, benzo(*a*)pyrene, fluoranthene, benzo(*b*)fluoranthene, benzo(*k*)fluoranthene, benzo(*g,h,i*)perylene, and indeno(1,2,3-*cd*)pyrene) was observed. A second and third doses of the same amounts of sodium acetate and phenanthrene were introduced again on the 49th and the 56th day of the study. Again phenanthrene disappeared rapidly (from >300 mg/kg on 49th day to 24 mg/kg on 51st day, and from >300 mg/kg on 56th day to 69 mg/kg on 58th day) without any observed reduction in the concentrations of the higher-ringed PAHs from the system. The metabolism of the amended compounds was also accompanied by an increase in the oxygen uptake rate in the reactor and a slight decrease in the dissolved oxygen concentration. The dissolved oxygen concentration, however, remained above 5 mg/l.

The low concentrations of higher molecular weight PAHs in the soil and their low solubility in aqueous phase suggest that their desorption from soil may be the critical step responsible for a lack of their biodegradation. This has been suggested to be true by the work of Edwards et al. [28]. A commercially available nonionic surfactant Tween 80 was added to the third reactor at a level of 950 mg/l slurry on the 42nd, 49th, and 56th days in order to evaluate this hypothesis. Again, however, no change in the concentrations of pyrene and other complex PAHs was observed. Their concentration in the aqueous phase remained below their detection limit (25 µg/l). Unfortunately, neither alternative concentrations of Tween 80 nor any other surfactant was explored in this study. As a result, it is difficult to speculate whether Tween 80 at these concentrations was ineffective or if bioavailability is not a

critical problem in biodegradation of the chemicals in this system. In a simultaneous study with another soil contaminated with higher concentrations of petroleum hydrocarbons [29], significant reductions in the concentrations of these higher ringed PAHs was observed. It implies that bioavailability was not an issue at least at higher concentrations.

It is also possible that an absence of suitable microorganisms was responsible for the observed lack of degradation of the complex PAHs. This is unlikely, as the soil was collected from a site that had been contaminated for a long time. Although microorganisms capable of growing on pyrene were not enumerated in this soil, a significant number of pyrene degraders were noted by Zappi et al. [29] in another heavily contaminated soil.

The biodegradation rates of the higher molecular weight PAHs observed in this study are in sharp contrast with those observed by Castaldi and Ford [22], Marks et al. [21], and of Zappi et al. [29]. All of these authors studied bioremediation of soils contaminated with high levels of PAHs and reported 90% or higher degradation in slurry reactors within 48 d. Rapid metabolization of BTEX and of aromatic hydrocarbons with up to three aromatic rings was observed in slurry reactors by all the investigators, including us. The discrepancy in the observations concerning higher ringed PAHs should be explored further.

4. Conclusions

In a slurry reactor containing soil with low-levels of fuel-hydrocarbon contamination, BTEX chemicals and TRPH were rapidly metabolized below their detection and regulatory limits. PAHs with fewer than four aromatic rings were also quickly metabolized. Higher ringed PAHs, however, were not degraded even when the system was supplemented with additional carbon and energy sources (sodium acetate and/or phenanthrene). The supplemented phenanthrene was rapidly metabolized, as evidenced by the decrease in its concentration, as well as by the dissolved oxygen and oxygen uptake rate data. Addition of surfactant Tween 80 also did not help with degradation of higher molecular weight PAHs. The results of this study suggest that while it is possible to biodegrade the BTEX-chemicals, TRPH, and PAHs with three or less aromatic rings in soils using a bioslurry system, low-level contaminations of PAHs containing four or more aromatic rings may remain even after long period of treatment. Supplying growth and energy source by itself did not improve their biodegradation rates.

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