

## Implementation of natural attenuation at a JP-4 jet fuel release after active remediation

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### Abstract

After eighteen months of active remediation at a JP-4 jet-fuel spill, a residual of unremediated hydrocarbon remained. Further site characterization was conducted to evaluate the contribution of natural attenuation to control exposure to hazards associated with the residual contamination in the subsurface. Activities included the detailed characterization of ground-water flow through the spill; the distribution of fuel contaminants in groundwater; and the analysis of soluble electron acceptors moving into the spill from upgradient. These activities allowed a rigorous evaluation of the transport of contaminants from the spill to the receptor of groundwater, the Pasquotank River. The transport of dissolved contaminants of concern, that is benzene, toluene, ethyl benzene, xylene isomers (BTEX) and methyl-tertiary-butyl ether (MTBE), into the river from the source area was controlled by equilibrium dissolution from the fuel spill to the adjacent groundwater, diffusion in groundwater from the spill to permeable layers in the aquifer, and advective transport in the permeable layers. The estimated yearly loading of BTEX compounds and MTBE into the receptor was trivial even without considering biological degradation. The biodegradation of hydrocarbon dissolved in groundwater through aerobic respiration, denitrification, sulfate reduction, and iron reduction was estimated from changes in ground-water chemistry along the flow path. The concentrations of target components in permanent monitoring wells continue to decline over time. Long term monitoring will ensure that the plume is under control, and no further active remediation is required.

### Introduction

Implementation of natural attenuation is a contaminant management strategy that uses intrinsic attenuation processes to control the hazard associated with the release of the contaminant. It has long been recognized that the terrestrial subsurface environment has the ability to recover from damage caused by certain chemical release such as spills and leaks of petroleum hydrocarbons from storage and transport facilities. When release of these chemicals to the subsurface environment has harmed ground-water quality, natural processes such as biodegradation, dilution, dispersion, and sorption often limit the migration of contaminants in groundwater. Results from a large number of field

sites, laboratory studies, and computer models of field sites have demonstrated that natural attenuation can significantly limit the migration of contaminants and reduce the potential impact of hydrocarbon release by preventing the transfer of contaminants to sensitive receptors [1]. Natural attenuation is not a passive 'let-it-go' process, but an active management strategy with planning, site characterization, data analysis, and monitoring throughout the entire period of implementation. A protocol developed for the U.S. Air Force [2] includes various steps for the implementation of natural attenuation including: review of site data, developing a preliminary conceptual model, performing additional site characterization to fill gaps in the data required for a complete assessment, documenting the geochem-

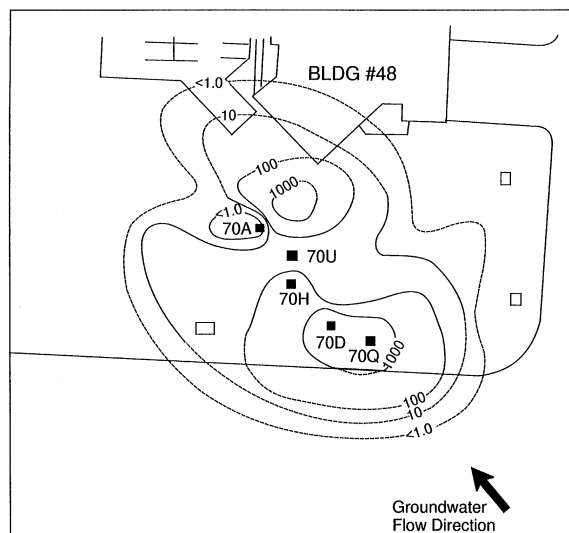


Figure 1. Total BTEX isoconcentration map for groundwater, JP-4 pipeline release site, US Coast guard support center, Elizabeth City, North Carolina. — 100 —, Total BTEX Isoconcentration line ( $\mu\text{g/L}$ ). Samples collected on 12/6/94

ical indicators of intrinsic remediation, modeling the behavior of the plume as part of an exposure assessment, preparing long-term monitoring plans and alternative remediation plans, and finally presenting the findings to regulatory agencies. Under ideal situations, the appropriate regulatory agencies are involved in the process from the planning stages forward to the final report.

Detailed site characterization was conducted at a jet-fuel contaminated site to implement the natural attenuation as the plume management strategy after the termination of active remediation. Contamination of soil and groundwater was discovered at a site on the U.S. Coast Guard Support Center, Elizabeth City, North Carolina. The contamination was caused by a continuous release of JP-4 jet fuel from small corrosion holes in an underground pipeline. The U.S. Coast Guard and the U.S. EPA National Risk Management Research Laboratory initiated a joint cleanup demonstration using air injection, soil vacuum extraction (SVE), and enhanced biodegradation. Based on extensive core sampling, the initial spill was estimated to contain approximately 3600 kg of JP-4 jet-fuel. During the eighteen months of operation, about 1700 kg of hydrocarbon was collected through the SVE system. Soil core sampling after the termination of active remediation revealed that 55% of fuel hydrocarbon and more than 98% of benzene had been removed by the remedi-

ation effort. The majority of volatile and biodegradable components such as BTEX were removed by SVE and biodegradation, but still a large portion of fuel hydrocarbon remained in the subsurface material.

Ground-water sampling after remediation showed that the size of the plume was reduced substantially, but a few wells within the region that contained residual light non-aqueous phase liquids (LNAPL) continued to produce groundwater with concentrations of benzene, toluene, ethylbenzene, and xylenes (BTEX), and methyl-tertiary-butyl-ether (MTBE) that were far above the drinking water standards. The maximum concentration of BTEX encountered after remediation was  $2584 \mu\text{g/L}$  at the monitoring well 70D in Figure 1 [3].

Monitoring wells downgradient from the release produced groundwater with concentrations of MTBE and BTEX that were below the State of North Carolina's requirements. Although the ultimate receptor, the Pasquotank river, was only 100 m distant from the spill, natural attenuation apparently reduced contaminant concentrations below standards before groundwater reached the river boundary. To confirm and validate this empirical observation, intensive site characterization and mathematical modeling of ground-water flow and contaminant biotransformation were used to demonstrate that the expected behavior of the plume after active remediation was consistent with the monitoring data [4]. Because groundwater is not used for drinking water, there is no exposure to the human population. Based on the absence of human exposure, and the projection of the mathematical models that contaminants would not discharge to a surface water ecosystem, the State of North Carolina and the U.S. Coast Guard determined that further active remedy was unnecessary, and they implemented a program of long term monitoring. This paper presents the site characterization activities and results which provided the bases for the implementation of natural attenuation as the plume management strategy at the site. The purpose of site characterization activities and data analysis was to develop the conceptual model of the fate and transport processes from the plume source to the sensitive acceptor and to provide data for the natural attenuation implementation as the alternative plume management strategy. The conceptual model allowed the estimation of the contaminant loading and electron acceptors under given field condition. Similar approach can be applicable to the hydrocarbon contamination sites on flood plain landscapes.

Table 1. Amount of electron acceptors in groundwater equivalent to BTEX concentration

Degradation Processes	Stoichiometric Mass Ratio to BTEX	Maximum Acceptor or Product Concentration	Equivalent BTEX Concentration ( $\mu\text{g/L}$ )
Aerobic Respiration	3.14 g $\text{O}_2/\text{g}$ BTEX	2.5 mg/L of DO	800
Fe(III) Reduction	21.8 g Fe(II)/g BTEX	16.2 mg/L of Fe(II)	810
Denitrification	4.86 g $\text{NO}_3/\text{g}$ BTEX	2.2 mg/L of $\text{NO}_3$	450
Sulfate Reduction	4.7 g $\text{SO}_4/\text{g}$ BTEX	20 mg/L of $\text{SO}_4$	4200
Methanogenesis	0.78 g $\text{CH}_4/\text{g}$ BTEX	less than 30 $\mu\text{g/L}$ of $\text{CH}_4$	less than 38
Total Amount			6300
Highest BTEX Concentration			2584

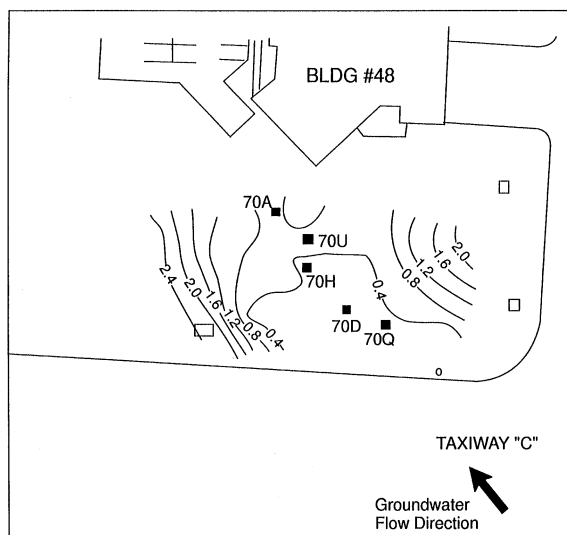


Figure 2. Dissolved oxygen isoconcentration map for groundwater. — 0.8 — Dissolved oxygen isoconcentration line (mg/L). Samples collected on 12/6/94

### Geochemical analysis for intrinsic biodegradation

Microorganisms obtain energy for cell production and maintenance by catalyzing the transfer of electrons from electron donors to electron acceptors. Fuel hydrocarbons are completely degraded and detoxified if they are utilized as the primary electron donor for microbial metabolism [5,6,7]. Important electron acceptors in aquifers include dissolved oxygen, nitrate, and sulfate, and Iron III minerals in the aquifer matrix material. After the soluble electron acceptors are depleted, microorganisms may ferment hydrocarbon compounds through reactions that produce methane and carbon dioxide. The depletion of oxygen, nitrate, and sulfate within a plume compared to concentrations outside the hydrocarbon plume, and the elevated concentration of

ferrous iron and methane produced within the plume can be used to estimate the total amount of electron acceptors available in groundwater. According to Borden et al. [7], the average mass of BTEX compounds that can be degraded using the specific electron acceptor can be estimated from stoichiometric relationships and is listed in Table 1.

At the jet-fuel site on the USCG Supply Center, dissolved oxygen concentrations were measured with a DO meter at existing monitoring wells on December 6, 1994 (Figure 2). Based on the comparison of background concentrations of DO with DO concentrations within the plume, it is likely that oxygen is an important electron acceptor at the site. With a background DO concentration of approximately 2.5 mg/L, the shallow groundwater at the site has the capacity to provide oxygen which can microbially oxidize 0.8 mg/L of BTEX compounds.

Samples were collected and analyzed to measure concentrations of nitrate + nitrite (as N) in December 1994. EPA method 353.1 was used. Concentrations ranged from <0.05 mg/L to 0.27 mg/L. Additional ground-water samples collected using the Geoprobe<sup>TM</sup> on December 6, 1994 indicated a maximum total nitrogen concentration of 2.23 mg/L at a depth of approximately 4.5 m below land surface. Nitrate in groundwater can be used to assimilate 0.45 mg/L of BTEX compounds. Although several investigators have reported that benzene is recalcitrant to nitrate reduction [8, 9], other studies indicate that benzene may be degraded under nitrate reducing conditions [10, 11].

Once the available oxygen and nitrate are depleted, subsurface microorganisms may use oxidized iron minerals [Fe(III)] as an electron acceptor [12, 13]. The forms that are most easily reduced are amorphous and poorly crystalline Fe(III) hydroxides and Fe(III) oxyhydroxides [14]. The reduction of Fe(III) results in

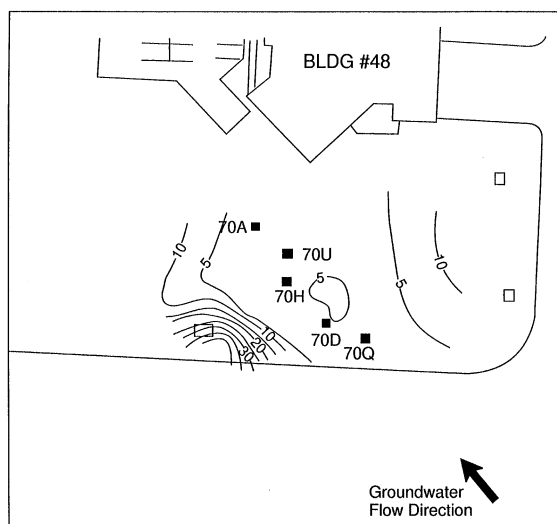


Figure 3. Sulfate isoconcentration map for groundwater. — 10 — Sulfate isoconcentration line (mg/L). Samples collected on 12/6/94

elevated concentrations of dissolved Fe(II) in contaminated aquifers. Lovely et al. [12] found that, in an aquifer contaminated by a crude oil spill, the selective removal of BTEX from the plume was accompanied by an accumulation of dissolved Fe(II) and depletion of Fe(III) oxides in the contaminated sediments. The highest concentration of ferrous iron was 16.2 mg/L at existing monitoring wells in July 1991. Groundwater had assimilated at least 0.81 mg/L of BTEX during iron reduction.

Sulfate concentrations were measured at existing monitoring wells in December 1994 (Figure 3). Water's capillary electrophoresis method N-601 was used for sulfate in ground-water samples. Areas with elevated BTEX concentrations showed depleted sulfate concentrations. This is a strong indication that anaerobic biodegradation of the BTEX compounds is occurring at the site through microbial sulfate reduction. With the background sulfate concentration of 20 mg/L, the shallow groundwater at this site has assimilated 4.2 mg/L of BTEX during sulfate reduction.

Methane concentrations were measured at groundwater monitoring points and at existing monitoring wells in July 1991. Methane concentrations (less than 0.03 mg/L) were not high enough to indicate substantial methanogenic bacterial activity.

Depletion of dissolved oxygen, nitrate, and sulfate and elevated concentrations of ferrous iron and methane suggest that mineralization of BTEX compounds was occurring through the microbially medi-

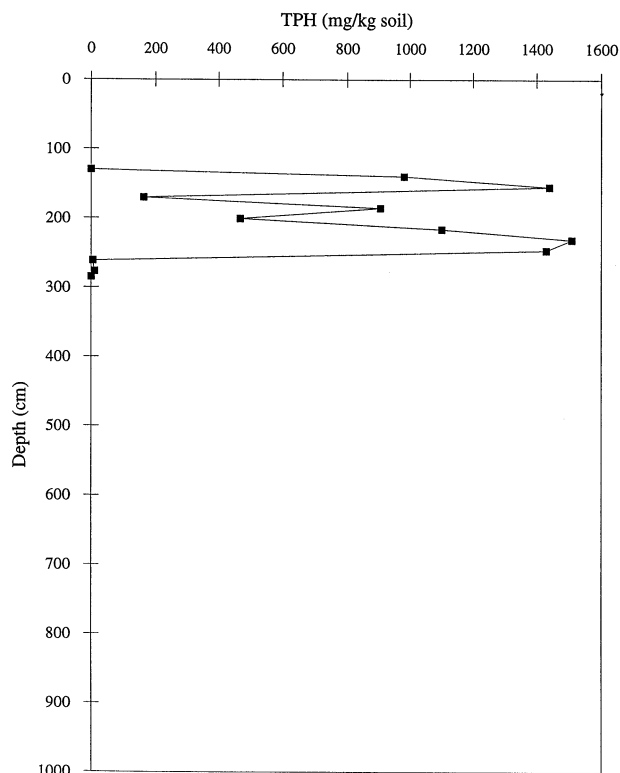


Figure 4. Vertical distribution of total petroleum hydrocarbon (TPH) in soil (mg/kg soil) near monitoring Well 70Q. Samples collected in September 1994

ated processes of aerobic respiration, iron reduction, nitrate reduction, and sulfate reduction. Based on stoichiometric relationships, electron acceptors in groundwater can degrade 6300  $\mu\text{g/L}$  in BTEX mass. The highest BTEX concentration in ground-water monitoring wells during the December 1994 sampling event was 2584  $\mu\text{g/L}$ . This is considerably less than the total amount of electron acceptors available in the groundwater. This provides a preliminary comparison of available electron acceptors and BTEX compounds based upon stoichiometric relations of BTEX oxidation. This ratio may be changed depending on the chemical and microbial conditions in groundwater. Vroblesky and Chapelle [15] showed that the dominant terminal electron accepting process can vary both temporally and spatially in an aquifer with fuel hydrocarbon contamination. Environmental conditions and microbial competition ultimately determine which processes will dominate [16].

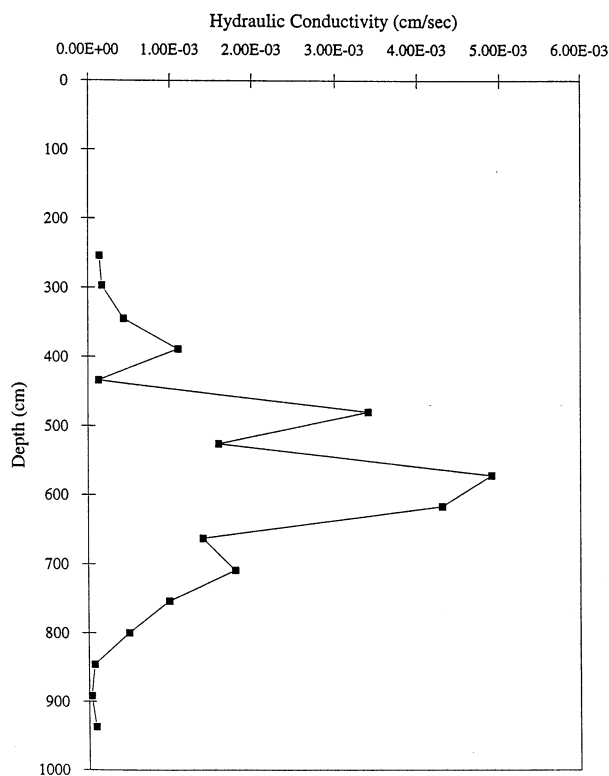


Figure 5. Vertical distribution of hydraulic conductivity (cm/sec) near monitoring Well 70Q, measurement in September 1994

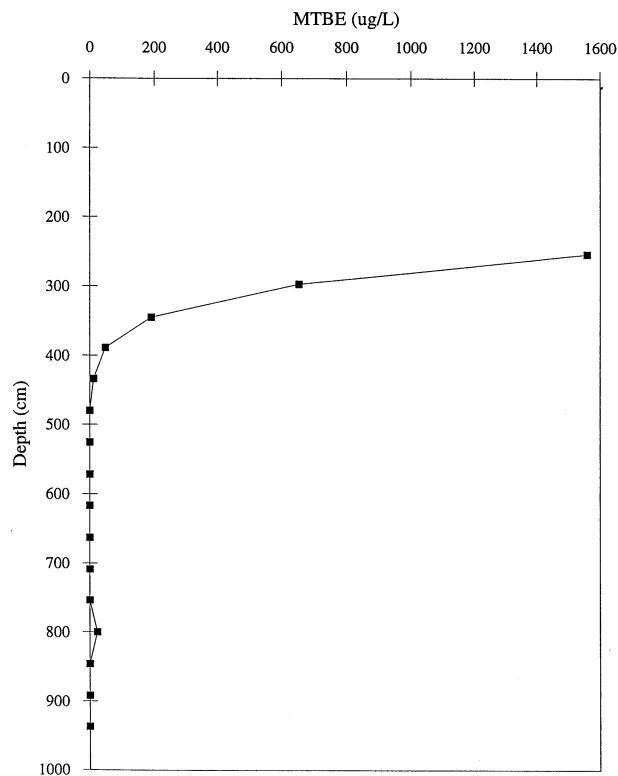


Figure 6. Vertical distribution of dissolved MTBE concentrations (µg/L) near monitoring Well 70Q in groundwater. Samples collected in September 1994

### Site investigation for fate and transport of BTEX and MTBE

Extensive soil and ground-water sampling and hydraulic conductivity measurement were conducted after the termination of active remediation [3, 4]. Soil samples were collected with a core barrel driven by the Geoprobe<sup>TM</sup>. A total of 71 samples from eight locations were collected and analyzed with GC/FID for total petroleum hydrocarbon (TPH) and GC/MSD for individual compound. Figure 4 shows the vertical distribution of TPH in soil at the location near monitoring well 70Q after the termination of the active remediation. This was the most heavily contaminated location prior to active remediation. The amount of TPH had been significantly reduced (about 55% was removed) by the active remedial effort. However, a substantial amount of hydrocarbon remained. Most of the residual fuel in the soil was detected within a narrow zone from 100 cm to 300 cm below the surface. This shallow, thin zone remains as a long term source for contamination in groundwater.

Hydraulic conductivity was measured with the Geoprobe<sup>TM</sup> ground-water sampling system [17]. The Geoprobe<sup>TM</sup> sampling tool drove hollow steel rods into the earth to serve as a temporary ground-water monitoring well. The rods were threaded to allow them to be joined together, and the leading rod was slotted to admit groundwater being sampled. The leading rod was slotted over a vertical interval of 45 cm with 0.51 mm slots. A polyethylene tube was inserted to the bottom of the rods and water was pumped from the tube until the flow rate came to equilibrium; then the flow rate was measured. The hydraulic conductivity was estimated following Wilson et al [17]. A vertical profile of the hydraulic conductivity measured near monitoring well 70Q is shown in Figure 5. A highly conductive zone existed at a depth of 400 to 700 cm. The conductivity was two orders of magnitude higher than the measurements at the shallower depth intervals. The majority of groundwater flows in this zone of higher conductivity.

After measurement of the hydraulic conductivity, water samples were collected. These samples were ana-

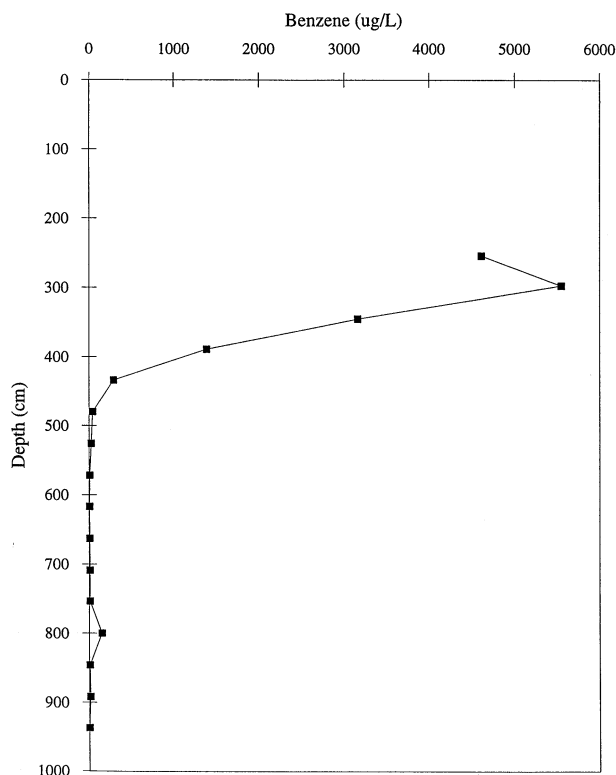


Figure 7. Vertical distribution of dissolved benzene concentrations ( $\mu\text{g/L}$ ) near monitoring Well 70Q in groundwater. Samples collected in September 1994

lyzed for MTBE, BTEX, and TMB with a headspace GC/MS in the laboratory. Vertical profiles of MTBE and benzene in the aqueous phase are shown in Figures 6 and 7. Both benzene and MTBE were detected at depths below the bottom of the interval with residual jet fuel at 300 cm. Benzene and MTBE had their highest concentrations in groundwater at a depth of 300 cm, at the deepest extent of the LNAPL. Their concentrations decreased with depth into the subsurface down to a depth of 500 cm, where increasing hydraulic conductivity allowed lateral transport of the contaminants.

The distribution of benzene and MTBE between the bottom of residual LNAPL layer and the top of the highly conductive zone suggests that the diffusion along the concentration gradient controls the transport of these contaminants from their source in the unremediated residual to the conductive part of the aquifer.

### Transport of MTBE and BTEX from the LNAPL source into groundwater

Three processes of chemical transport from the LNAPL source were considered: (1) equilibrium dissolution from LNAPL into water in soil pores; (2) downward diffusion due to concentration gradient; (3) and lateral advective transport with ground-water flow in the conductive depth intervals. The flux of MTBE diffusing from the concentration maximum in groundwater near the bottom of the layer of LNAPL down to the area with higher hydraulic conductivity was estimated by multiplying the diffusion coefficient by the concentration gradient and then dividing by the tortuosity.

The vertical concentration gradient of MTBE in the Geoprobe<sup>TM</sup> water samples was linear down to the first layer that could transport water at a depth of 3.6 to 4.0 m (compare Figure 5 and 6). The diffusion path was from depths of 3.0 to 3.9 m and the change in MTBE concentration along the diffusion path was from 1560 to 191  $\mu\text{g/L}$ , resulting in a concentration gradient of 15  $\mu\text{g/L cm}$ . The diffusion coefficient of MTBE in water was estimated to be  $0.85 \times 10^{-5} \text{ cm}^2/\text{sec}$ . The tortuosity was estimated to be 1.3. Multiplying diffusion coefficient and concentration gradient and dividing the tortuosity gave the diffusive flux of 9.6  $\text{mg/m}^2\text{sec}$ , equal to 30  $\text{mg/m}^2\text{year}$ . Integrating the vertical MTBE concentration in soil core extracts revealed that 600 mg of MTBE per  $\text{m}^2$  horizontal area remained in the subsurface around 70Q. With the given flux, five percent of the MTBE in the residual hydrocarbon would be transferred to the aquifer each year.

The concentration of benzene at 70Q changed from 5550  $\mu\text{g/L}$  at a depth interval of 2.9 m to 288  $\mu\text{g/L}$  at 4.3 m, a change of 5262  $\mu\text{g/L}$  over 137 cm. The concentration gradient for benzene was 38  $\mu\text{g/L per cm}$ . The diffusion coefficient of benzene was estimated as  $0.83 \times 10^{-5} \text{ cm}^2/\text{sec}$ . Multiplying the effective diffusion coefficient by the concentration gradient gave a diffusive flux of  $2.4 \times 10^{-6} \text{ mg/m}^2 \text{ sec}$  for benzene, equal to 76  $\text{mg/m}^2 \text{ year}$ .

If it is assumed that the average loading in the entire LNAPL zone follows the same diffusive flux as Geoprobe<sup>TM</sup> samples near monitoring well 70Q, then total loading from the entire LNAPL zone into the convective zone can be estimated by integrating the flux over the LNAPL boundary. The area was about 1600  $\text{m}^2$ ; hence, total loading of benzene into groundwater over the region would be 121 g/year. The total BTEX loading would be 146 g/year (TEX data not presented), and MTBE would be 50 g/year. It would

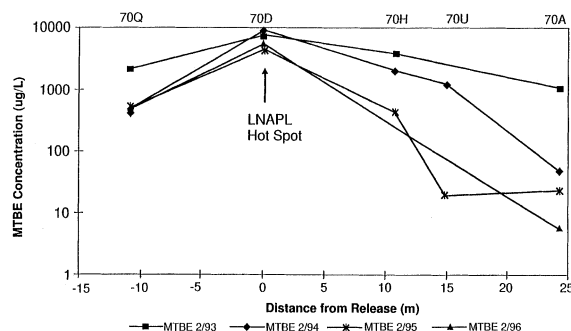


Figure 8. Temporal changes of MTBE concentrations along the center line of the plume. Samples from Well 70U in February 1993 and 70H and 70U in September 1996 were not available.

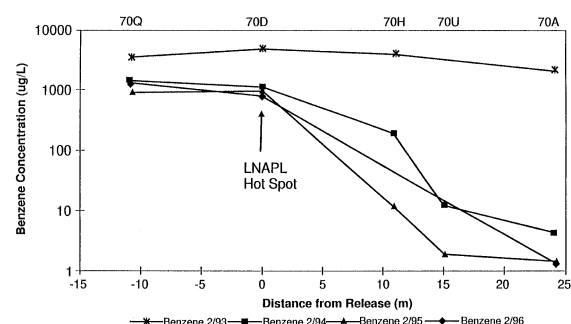


Figure 9. Temporal changes of benzene concentrations along the center line of the plume. Samples from Well 70U in February 1993 and 70H and 70U in September 1996 were not available.

take more than 20 years to completely deplete BTEX and MTBE from the LNAPL source at these rates. The LNAPL was 50 m wide. The conductive part of the aquifer was 7 m deep. The cross section affected by the LNAPL was 350 m<sup>2</sup>. The average hydraulic conductivity was 0.001 cm/s, and the average hydraulic gradient was 0.0015, resulting in a Darcy flow of 0.47 m/year. With a cross section of 350 m<sup>2</sup>, the discharge would be 170 m<sup>3</sup>/year. If the total flux of MTBE (50 g/year) is divided by the discharge of water receiving the MTBE, the dilution of MTBE in groundwater would result in a concentration of 300 µg/L when groundwater leaves the downgradient edge of the LNAPL boundary.

#### Comparison of electron acceptor influx and BTEX loading

Given the amount of electron acceptors available for biodegradation of hydrocarbon and groundwater discharge rate, the input rate of available electron

acceptors to the contaminated zone can be estimated. The amount of electron acceptors in groundwater (6300 µg/L) multiplied by the ground-water discharge rate (170 m<sup>3</sup>/year) yields the input rate of 1070 g/year electron acceptors. Comparing total loading of 145 g/year of BTEX from the LNAPL source, groundwater at the site can supply the sufficient amount of electron acceptors needed for biodegradation of hydrocarbons.

#### Temporal changes of MTBE and benzene concentrations

The concentrations of MTBE and benzene were monitored in wells along a flow path extending across the hydrocarbon release. Water samples were collected before, during, and after active remediation (Figures 8 and 9). Monitoring well 70Q was 10 m upgradient of the LNAPL hot spot, well 70D was at the hot spot, well 70H was 10 m downgradient of the hot spot but in the lens of LNAPL, well 70U was 15 m downgradient of the hot spot but at the edge of the lens of LNAPL, and well 70A was 24 m downgradient of the hot spot and just outside the margin of the LNAPL. Active remediation started March, 1993 and finished in December, 1994. By September, 1996 the concentrations of benzene and MTBE were below drinking water standards in the monitoring well just downgradient of the release at the monitoring well 70A. The concentrations of MTBE and benzene continued to decline after active remediation was terminated.

#### Summary and conclusions

Geochemical analysis of ground-water samples documented the contribution of sulfate reduction, Iron III reduction, aerobic respiration, and denitrification to removal of BTEX compounds from groundwater. Degradation of hydrocarbon by methanogenesis was minimal in groundwater at this site. The total dissolved electron acceptors available in groundwater exceeded the concentration of BTEX compounds in groundwater at any one time, indicating that the rate of natural biodegradation was controlled by the rate of supply of electron acceptors present in uncontaminated groundwater moving toward the LNAPL from upgradient.

Simple field procedures using a direct push method were used to obtain vertical profiles of the hydraulic conductivity. Most of the advective flow of groundwater was confined to the zone of higher conductivity

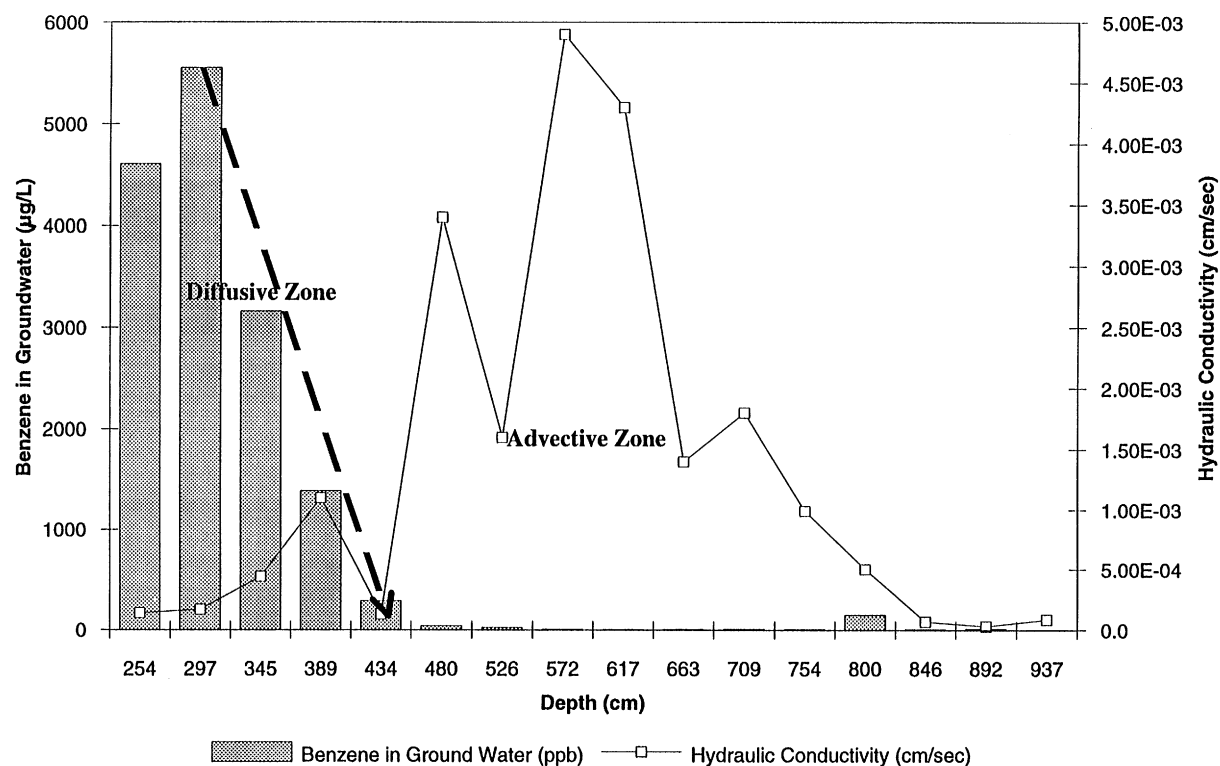


Figure 10. Vertical profiles of hydraulic conductivity (cm/sec) and dissolved benzene concentration ( $\mu\text{g/L}$ ) in groundwater near monitoring Well 70Q.  $\rightarrow$  Refers to the concentration gradient

underneath the interval containing the LNAPL. Water samples from the interval underneath the LNAPL revealed a concentration gradient of benzene and MTBE extending from the bottom of the LNAPL to the zone of higher hydraulic conductivity.

The processes of equilibrium dissolution into groundwater and diffusive transport due to the vertical concentration gradient were used to estimate the transfer of MTBE and benzene from the unremediated residual LNAPL to groundwater flowing underneath the LNAPL (Figure 10). Given loading rates, it would take more than 20 years to completely deplete BTEX and MTBE from the residual LNAPL source. Assuming no biodegradation of MTBE, dilution of MTBE in groundwater moving under the LNAPL toward the river would result in an equilibrium concentration of  $300 \mu\text{g/L}$ .

Benzene and MTBE analysis in the permanent monitoring wells along the ground-water flow showed marked reduction in the concentrations of MTBE and benzene since the termination of the active remediation. Groundwater at the site can supply a sufficient amount of electron acceptors to degrade dissolved

BTEX that partitions from the residual LNAPL in soil to groundwater.

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