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CONTAMINANT TRANSPORT AND FATE AT POINT-SOURCE  
RELEASES OF GASOLINE CONTAINING MTBE**

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**ABSTRACT**

Methyl *tert*-butyl ether (MTBE) is commonly found at concentrations above the current U.S. Environmental Protection Agency draft lifetime health advisory for drinking water (20 to 200 micrograms per liter) at many point-source gasoline release sites. MTBE is significantly more persistent than benzene, toluene, ethyl-benzene and xylenes (BTEX) in the subsurface. Therefore, evaluation of the implications of its presence in gasoline to monitored natural attenuation and engineered bioremediation alternatives is warranted. An interdisciplinary, field-based investigation of the subsurface transport and fate of MTBE and petroleum hydrocarbons is being conducted by the U.S. Geological Survey (USGS) Toxic Substances Hydrology Program at the site of an underground gasoline storage-tank release near Beaufort, South Carolina. The objective of the investigation is to provide a systematic evaluation of natural attenuation of MTBE compared to BTEX. Results of the field and laboratory studies at this site will be generalized to a broader range of hydrogeochemical conditions through experiments at other sites. Furthermore, newly developed methods of analysis can be applied to sites across the Nation. This investigation of MTBE at point-source release sites is coordinated with investigations of the occurrence of MTBE in shallow ground water, surface water, precipitation, and the atmosphere being conducted by the USGS National Water-Quality Assessment Program.

**INTRODUCTION**

Monitored natural attenuation and engineered bioremediation are acknowledged in many cases as cost-efficient alternatives for remediation of subsurface gasoline plumes. In a recent summary of trends in remediation technologies, a U.S. Environmental Protection Agency (USEPA) report states:

"...State UST (Underground Storage Tank) program managers report significant increases in the use of in situ processes, especially bioremediation, which is effective because of inherent biodegradability of

petroleum hydrocarbons. New technology development programs... help meet this demand by emphasizing in situ technologies, in particular bioremediation and enhancements to soil vapor extraction." (USEPA, 1997, p.13).

The use of oxygenated gasoline has increased in response to the 1990 Clean Air Act Amendments. Currently, methyl *tert*-butyl ether (MTBE) is the most widely used fuel oxygenate. The USGS National Water-Quality Assessment (NAWQA) Program found that MTBE was the second most frequently detected chemical of 60 volatile organic chemicals analyzed in samples of shallow, ambient ground water collected in urban areas during 1993-1994 (Squillace and others, 1996). While the effectiveness of MTBE-amended fuels is debated (National Research Council, 1996), the widespread use and environmental behavior of MTBE warrant close attention to its persistence in subsurface gasoline plumes and the attendant implications for natural attenuation and engineered bioremediation alternatives. MTBE is a significantly greater component of reformulated or oxygenated gasoline (as much as 14.8 percent by volume to achieve the Clean Air Act Amendment requirement of 2.7 percent by weight oxygen content) than the aromatic hydrocarbon constituents (BTEX). Also compared to BTEX, MTBE has a much higher water solubility, has a greater tendency for air-water partitioning to water, has less tendency to partition to subsurface solids, and has been found to be significantly less likely to degrade under field conditions (Squillace and others, 1997). All of these characteristics indicate that the environmental behavior of MTBE can result in significantly less natural attenuation than BTEX. Furthermore, it is likely that MTBE concentrations in ground water affected by point-source releases of oxygenated gasoline will exceed the current USEPA draft lifetime health advisory for drinking water (20 to 200  $\mu$ g/l (micrograms per liter)). For instance, the maximum aqueous MTBE concentration measured at the site discussed below is 251,000  $\mu$ g/l.

The USGS Toxic Substances Hydrology Program has conducted intensive field investigations of the transport and fate of petroleum hydrocarbons in the subsurface at the site of a petroleum pipeline rupture near Bemidji, Minnesota, and at a gasoline storage-tank leak in Galloway Township, New Jersey. Research at these sites has contributed to development of methods to characterize and evaluate the potential for natural attenuation of petroleum hydrocarbons at similar sites elsewhere (Baedecker and others, 1993; Cozzarelli and others, 1994; and Lahvis and Baehr, 1996). In 1996, the Program initiated research at a gasoline storage-tank release near Beaufort, South Carolina (SC). The purpose of this paper is to describe the design of that research, to provide a summary of preliminary results, and to describe the relation of this research to other ongoing USGS research activities on the environmental occurrence, transport, and fate of MTBE.

## **RESEARCH APPROACH**

The investigation focuses on a contaminated field site typical of many subsurface gasoline-contamination sites across the Nation. Extensive field characterization conducted by an

interdisciplinary team of scientists provides an understanding of field conditions conducive to evaluating the physical, chemical, and biological processes that affect natural attenuation at the site, including dilution by mixing, adsorption, biotic and abiotic transformation, and volatilization. Field and laboratory experiments at the site are generalized to a broader range of hydrogeochemical conditions through experiments at other sites. Furthermore, newly developed methods of analysis can be applied to sites across the Nation.

## **Field Site Description**

In 1991, a leaking underground gasoline storage tank was detected at Laurel Bay Exchange, Marine Corps Air Station, Beaufort, SC (fig. 1). The leak is believed to have occurred shortly before detection, and the amount of gasoline leaked was estimated at 5,678 liters. The UST and surrounding sediments were excavated in October 1993. In 1994, the USGS and the Southern Division, Naval Facilities Engineering Command initiated an assessment of contaminant transport of benzene and toluene and the potential for human or wildlife exposure. Results of field and laboratory studies and interpretation with a numerical solute-transport model indicated that these contaminants would not discharge to nearby surface waters (Landmeyer and others, 1996). These results were used to assess the potential for natural attenuation at the site. Although the fate of MTBE in the gasoline was not considered at that time, MTBE was monitored during the assessment.

The sandy water-table aquifer is approximately 14 meters thick; and is underlain by an extensive clay-rich confining unit. The uncontaminated part of the aquifer has dissolved oxygen of about 4 mg/l and ferric oxyhydroxide grain coatings. The hydraulic conductivity of the aquifer is estimated to be 3.4 meters/day. Ground-water flow directions and rates (about 18.9 meters/year) have been consistent in measurements from 1993 to 1997 (fig. 1).

## **Saturated Zone**

The phase-partitioning properties of MTBE indicate that dissolved MTBE can advance farther and faster than dissolved BTEX. The rate and extent of that advance can have a significant effect on the decision to employ natural attenuation to remediate a site.

Periodic monitoring of ground-water quality at the site provides a means to evaluate the evolution of the dissolved plume. Specifically, MTBE and BTEX compounds are monitored to observe the rate of separation of these compounds within the plume. Monitoring for the occurrence of potential degradation products of MTBE can indicate whether transformation of MTBE is occurring at the site. Additionally, monitoring for terminal electron acceptors enables description of the redox and general geochemical conditions within the plume, as well as how those conditions change with time.

Laboratory microcosm experiments using  $^{14}\text{C}$ -labeled MTBE enable evaluation of biotic or abiotic transformation of MTBE under both aerobic and anaerobic conditions. Aquifer sediments from both near the source and at the periphery of the plume, corresponding to in situ anaerobic and aerobic conditions, respectively, are being studied. Samples from areas with both high and low dissolved concentrations of MTBE and BTEX compounds also are being tested. Additional laboratory column experiments on sediments from the site, based on detecting the occurrence of degradation products, can provide an independent means for evaluating degradation of MTBE under controlled conditions that nearly duplicate those in the field.

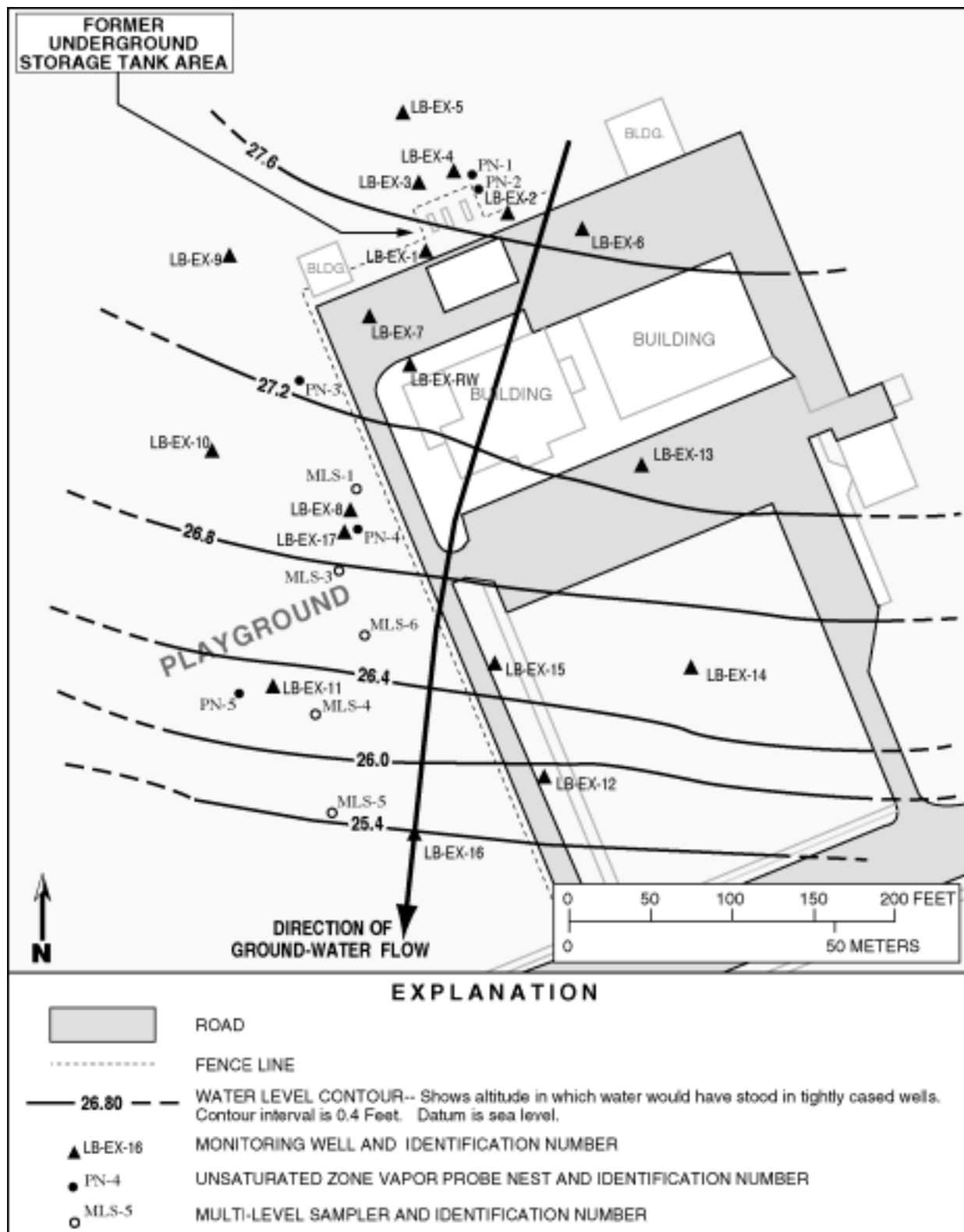


Figure 1. Study area showing location of monitoring wells, multi-level samplers, vapor probe nests, and representative water table (March 10, 1994).

## **Unsaturated Zone**

BTEX has been shown to volatilize from the water table and undergo aerobic biodegradation that contributes significantly to natural attenuation at the plume scale (Ostendorf and Kampbell, 1991; and Lahvis and Baehr, 1996). The predicted behavior of MTBE at the water table is fundamentally different from that for BTEX (Baehr and others, 1997a; and Pankow and others, 1997). This, together with its occurrence in oxygenated gasoline, has led researchers at the site to investigate the potential loss of MTBE through the unsaturated zone pathway under field conditions.

Nested probes installed in the unsaturated zone allow collection of unsaturated-zone gas samples at various intervals between the water table and land surface. Samples are analyzed for BTEX, MTBE, and other constituents. The flux of MTBE from the water table is estimated using these data and a transport model (Baehr and others, 1997a). Effective diffusion coefficients estimated from laboratory experiments on core material are used in the analysis. Analysis of the MTBE gas concentration profiles provides evidence for the loss of MTBE relative to BTEX in the unsaturated zone pathway. Laboratory experiments using unsaturated zone sediments from the site provide an independent means for evaluation of aerobic degradation of MTBE in the unsaturated zone.

## **Generalization to Other Sites**

Although intensive investigation of a field site provides unique research opportunities, steps are required to assure that results are transferable to other sites across the Nation. To generalize results from the intensive field site, selected field and laboratory studies are conducted at other sites with a broad range of hydrogeologic and geochemical conditions. Ground waters from other point-source and nonpoint-source sites where MTBE has been detected in ground water are sampled and analyzed for degradation products to identify field conditions where MTBE transformation may occur. In addition, laboratory experiments using sediments from other sites enable evaluation of transformation under widely varying field conditions, and provide the opportunity to modify geochemical conditions under controlled circumstances.

## **PRELIMINARY RESEARCH RESULTS**

This summary of research results is intended to provide an overview of the progress of the ongoing investigation, including recent results of field and laboratory studies. More detailed descriptions of these results can be found in the literature cited herein.

## Saturated Zone

The site has been instrumented with 15 monitoring wells and 5 multilevel sampling wells, each with 4 to 6 sampling ports at varying depths (fig. 1) (Landmeyer and others, 1996). Ground water was sampled periodically from 1993 to 1997. Samples were analyzed for petroleum compounds, BTEX and MTBE; potential products of MTBE degradation, *tert*-butyl alcohol (TBA) and *tert*-butyl formate (TBF); and other microbially-reactive solutes, including H<sub>2</sub>, Fe(II), sulfide, SO<sub>4</sub><sup>-2</sup>, CH<sub>4</sub>, dissolved inorganic carbon, and dissolved oxygen concentrations.

Maps of dissolved BTEX and MTBE concentrations in the evolving gasoline plume are shown in figure 2. Decreases in BTEX and MTBE concentrations near the source (wells LB-EX-1, -7, -8 and -RW) from 1993 to 1997 suggest that source removal and natural attenuation has been effective in decreasing contaminant concentrations. Continual increase in MTBE concentration in well (LB-EX-16), approximately 120 meters downgradient from the source, to over 40,000 µg/l in January 1997, indicates that MTBE still is migrating downgradient. The arrival of very low concentrations of BTEX (101 µg/l or less) at this well in January 1997 indicates that MTBE has advanced ahead of BTEX. BTEX and MTBE were not detected in samples from a well located downgradient from well LB-EX-16 and near Laurel Bay Boulevard.

Ground water from the site was analyzed for TBA and TBF using a direct aqueous injection technique (Church and others, 1997). TBA was detected in 6 of 14 wells sampled in June 1996 and January 1997 (detection limit 0.1 µg/l). This occurrence does not necessarily indicate that transformation of MTBE is occurring, as TBA may also have been a component of the source gasoline. TBA decreased between June 1996 and January 1997 by at least an order of magnitude in 4 of the 6 wells. TBA concentrations in well LB-EX-1 (at the source) remained constant and concentrations in well LB-EX-16 (at the downgradient extent of the plume) increased from 20.4 to 32.4 µg/l. TBA was detected only in wells where MTBE also was detected. TBF was not detected in samples at the site at a detection limit of 5 µg/l (Landmeyer and others, 1997).

Aerobic conditions in uncontaminated wells and wells in the periphery of the plume had low ferrous iron concentrations, relatively high sulfate, low sulfide, and no detectable methane. Anaerobic conditions near the center of the plume are characterized by higher ferrous iron and sulfide concentrations, and hydrogen concentrations indicating that iron-reducing, sulfate-reducing, and possibly methanogenic conditions exist or have existed.

Laboratory microcosm experiments using <sup>14</sup>C-MTBE show that, after 50 days of incubation under both aerobic and Fe(III)-reducing conditions, mineralization to <sup>14</sup>CO<sub>2</sub> was not significant. These results do not rule out the possibility that MTBE mineralization can occur in saturated or unsaturated sediments. It is possible, for example, that the lack of observed mineralization reflects preferential microbial mineralization of more labile BTEX compounds (which were



present in the contaminated sediments) or lack of microbial acclimation (in the uncontaminated sediments). These possibilities can be addressed only by using longer-term incubations, which are currently ongoing.

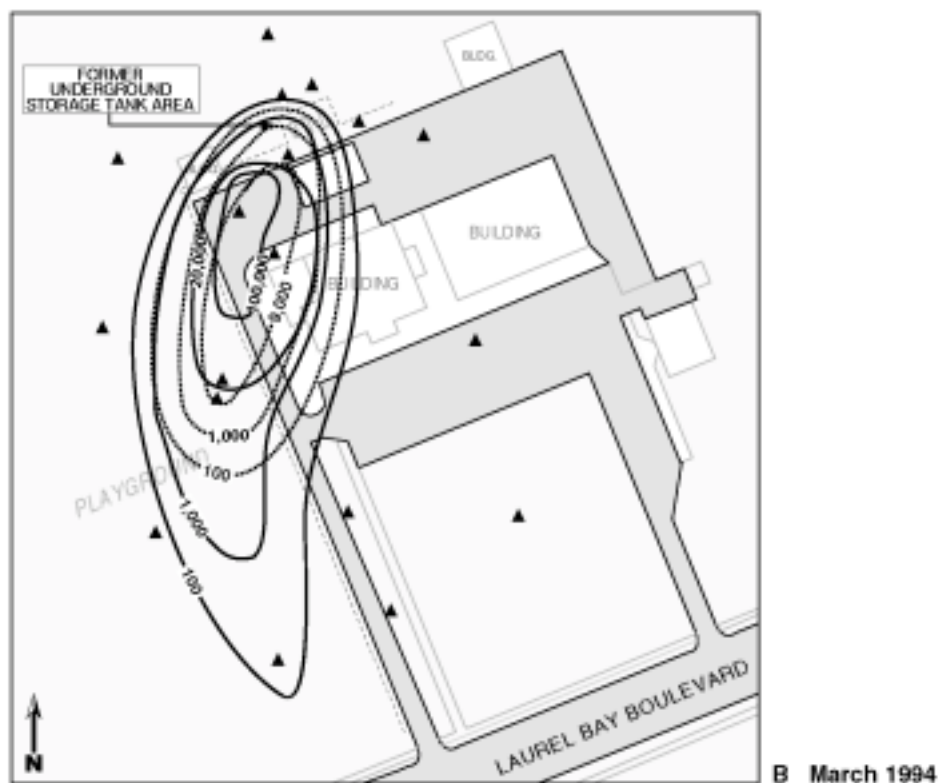
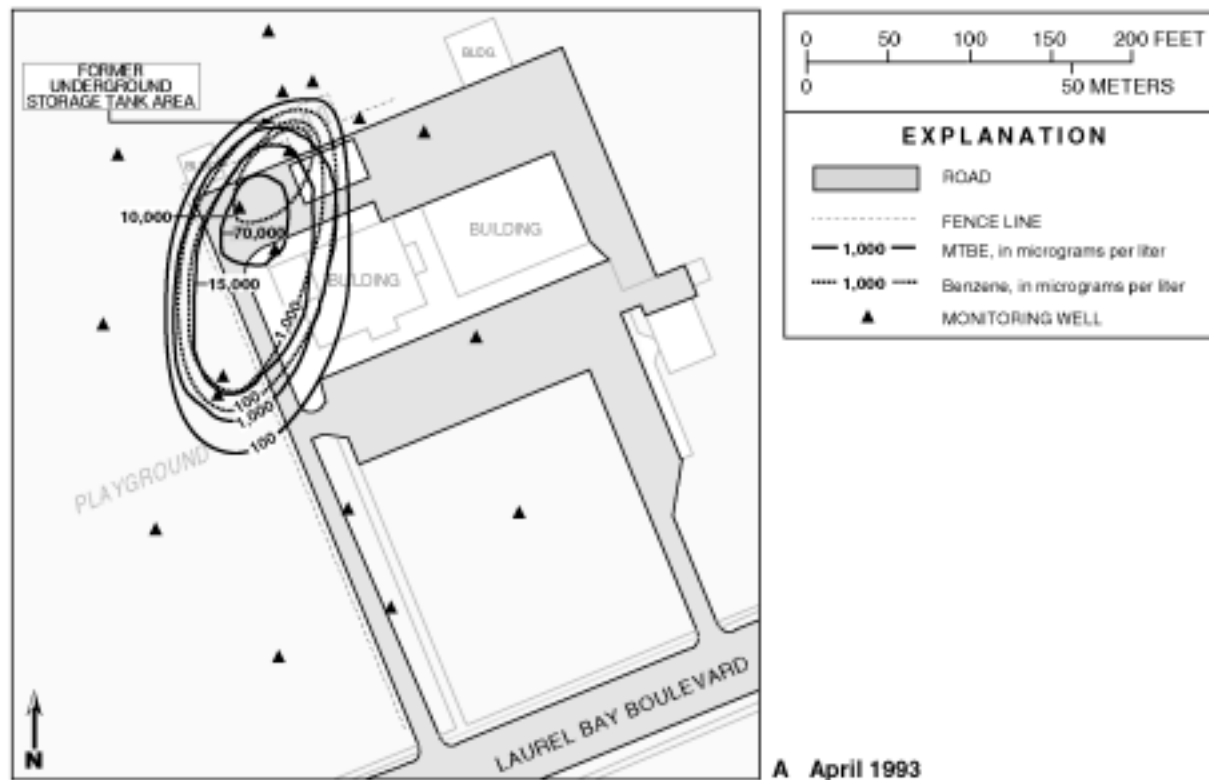


Figure 2. Benzene and methyl *tert*-butyl ether in ground water, (A) April 1993 and (B) March 1994.

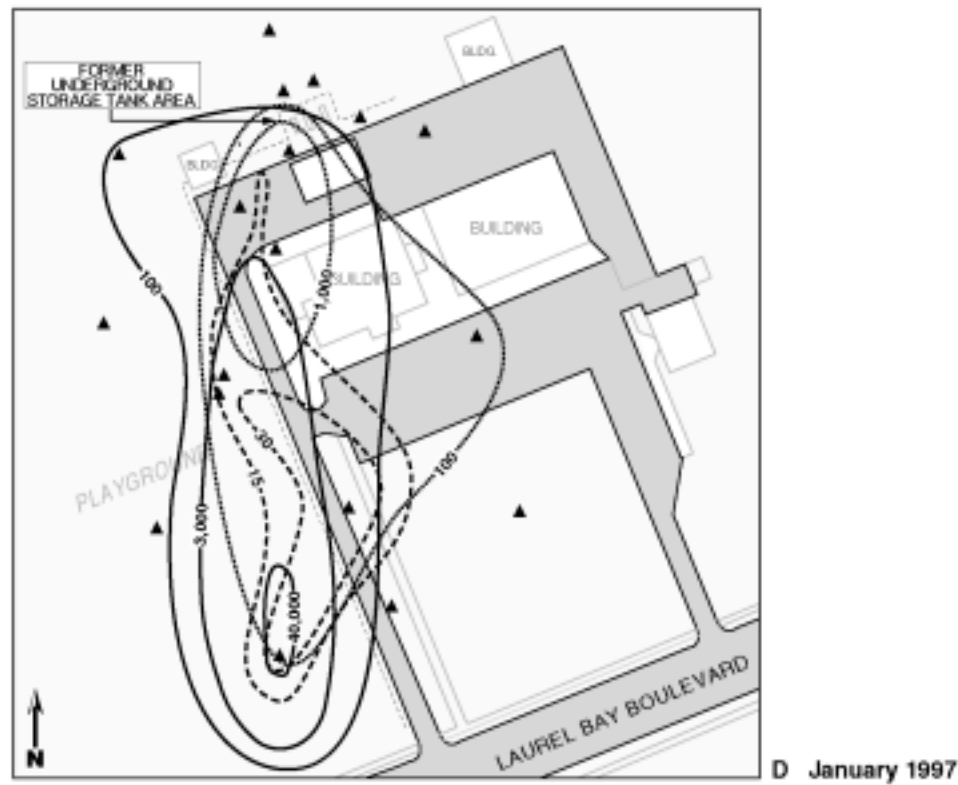
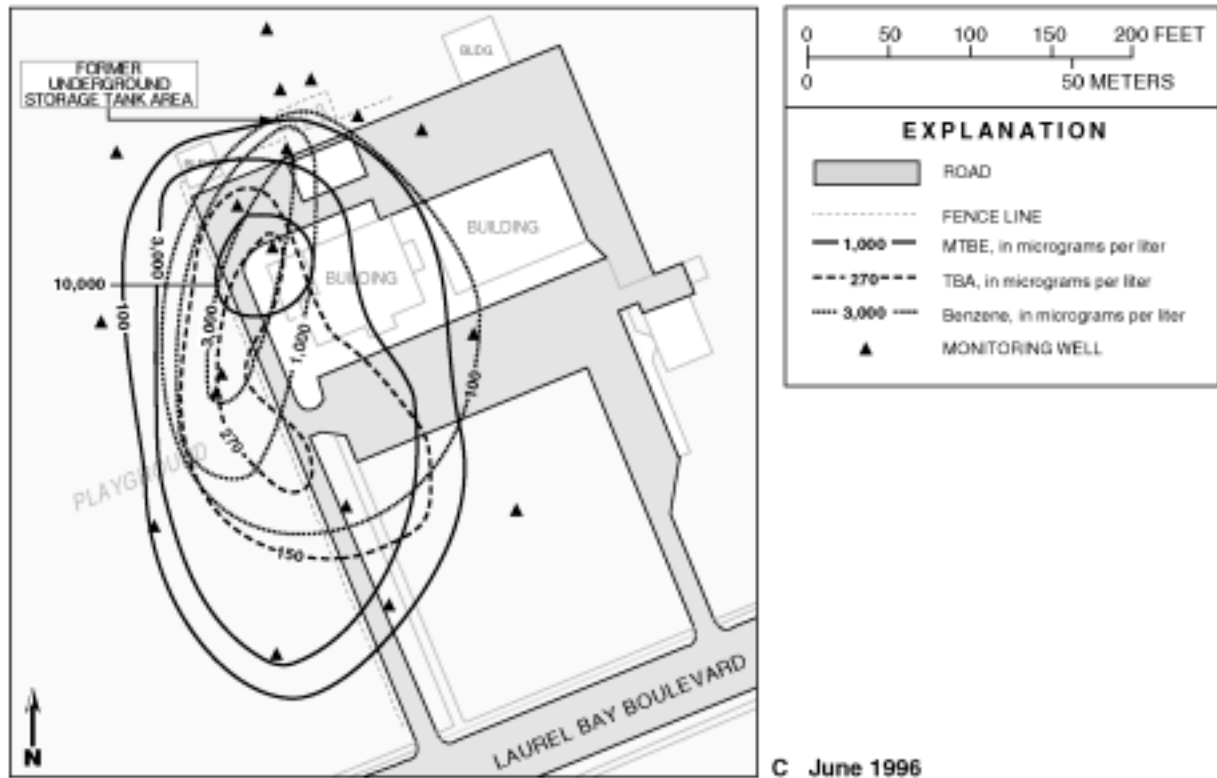


Figure 2. Benzene and methyl *tert*-butyl ether (MTBE) in ground water, --continued (C) June 1996 (also showing *tert*-butyl alcohol (TBA)). (D) January 1997 (also showing *tert*-butyl alcohol (TBA)).

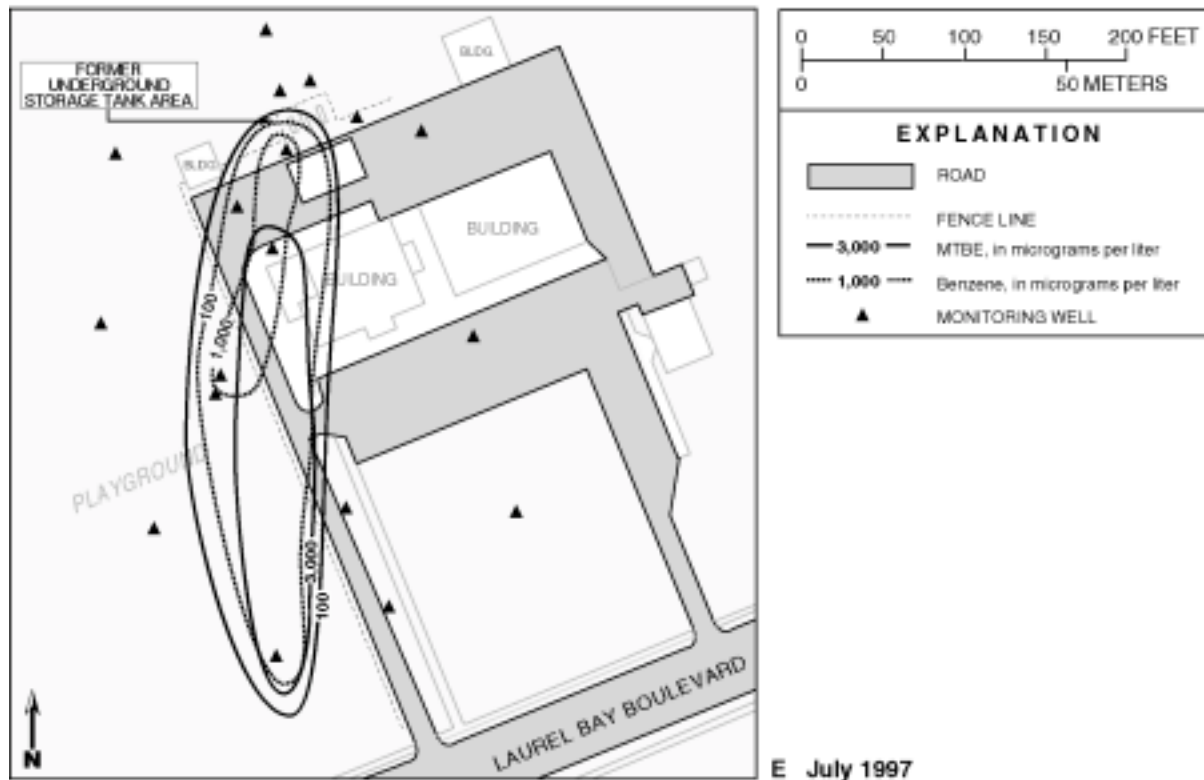


Figure 2. Benzene and methyl *tert*-butyl ether (MTBE) in ground water, --continued (E) July 1997.

## Unsaturated Zone

Five sets of nested vapor probes have been installed at the site (fig. 1). Samples were collected in June 1995, October 1996, and March 1997. The calculated flux of MTBE across the water table at probe nest PN-1 (located near the source) is given in Table 1.

Table 1. Flux of MTBE across the water table to the unsaturated zone and depth to water table.

Sampling Period	MTBE Flux (grams/meter <sup>2</sup> /year)	Water Table Depth (meters)
June 1995	19.7	3.35
October 1996	3.6	3.23
March 1997	22.1	3.47

The calculated mass fluxes of MTBE are comparable to those of BTEX, which for March 1997 were 36.6, 22.4, 29.4, and 192.5 grams/meter<sup>2</sup>/year, respectively. Temporal variations in mass flux may be attributable to the variability of product in the unsaturated zone caused by water-table fluctuation.

Gas-phase concentrations of both BTEX and MTBE within the unsaturated zone in March 1997 (fig. 3) decline more sharply than predicted, assuming diffusive transport with no transformation (Baehr and others, 1997a), suggesting aerobic degradation in the unsaturated zone. The mass flux for MTBE across the water table is significant enough to warrant further investigation of the unsaturated zone pathway. Laboratory column experiments to address the possibility of aerobic degradation are planned.

## Generalization to Other Sites

Ground water from five sites of MTBE occurrence has been analyzed for TBA and TBF. Three are point-source sites, 2 leaking underground storage-tank sites in California and a field test at Canada Forces Base Borden (Schirmer and others, 1997). Two are urban nonpoint-source sites in New Jersey and Michigan, identified during USGS NAWQA Program activities. To date, TBA

and TBF have not been detected at these sites at detection levels of 0.1 and 5  $\mu\text{g/l}$ , respectively (Church and others, 1997).

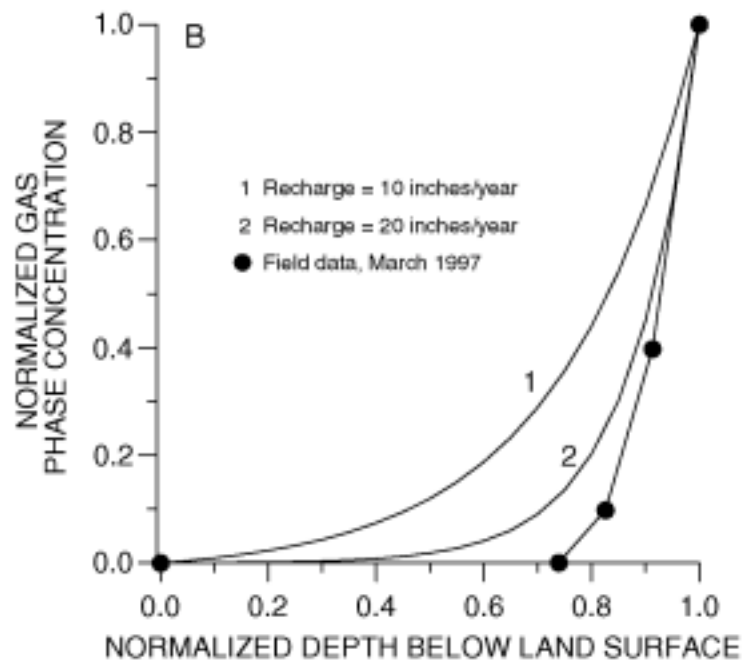
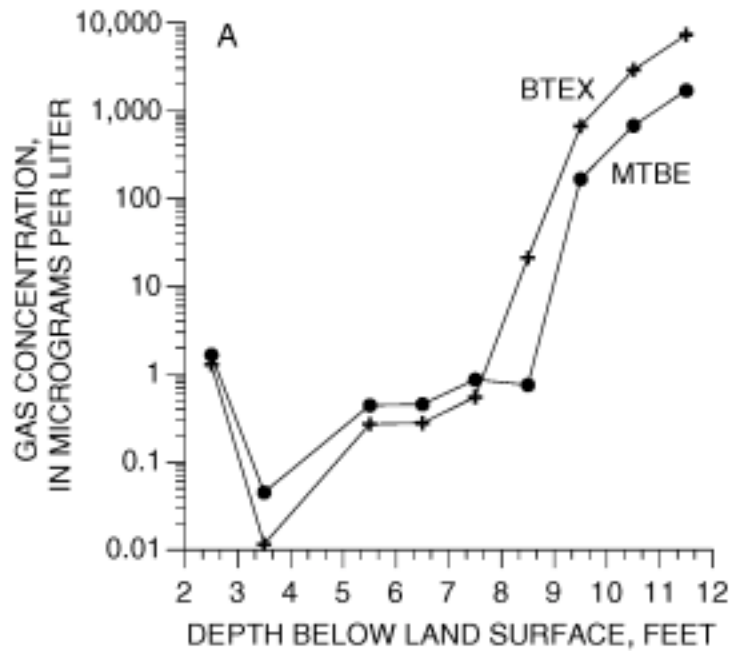


Figure 3. Gas concentration profiles in the unsaturated zone at probe nest PN-1, March 1997, (A) Benzene, toluene, ethyl-benzene, xylenes (BTEX) and methyl *tert*-butyl ether (MTBE) and (B) theoretical MTBE gas concentration profiles assuming no degradation compared to field data.

A column experiment performed with aquifer material from the nonpoint-source site in Glassboro, New Jersey yielded TBA after 35 days. At about 44 days, loss of MTBE became apparent and TBA concentration of the column effluent was 18  $\mu\text{g/l}$ . Similar experiments are planned at a number of well-characterized field sites.

## **RELATION TO OTHER USGS RESEARCH ACTIVITIES**

MTBE has been detected in many parts of the hydrologic cycle in urban areas, including precipitation, stormwater runoff, surface water, and shallow ground water (Squillace and others, 1996; Zogorski and others, 1997). MTBE can enter shallow ground water through release from point sources or through an atmospheric pathway to the water table with recharge (Squillace and others, 1997). MTBE concentrations in shallow ground water from the atmospheric pathway are expected to be on the order of a few micrograms per liter (Squillace and others, 1997), whereas maximum concentrations of MTBE at point-source release sites can be very high. A maximum concentration of 251,000  $\mu\text{g/l}$  was measured near the source of the Laurel Bay Exchange site. Therefore, management decisions related to the occurrence of low levels of MTBE in shallow ground water should consider whether an atmospheric or land-surface pathway could account for its occurrence, or whether the occurrence should be attributed to point sources with the attendant potential for significantly higher concentrations.

The comprehensive approach to the investigation of MTBE by the USGS includes activities of the NAWQA Program, as well as the Toxic Substance Hydrology Program activities discussed herein. The NAWQA Program has developed laboratory analytical methods to detect MTBE and suspected transformation products in water at low detection levels (Raese and others, 1995; and Church and others, 1997). Through its ongoing study-unit investigations, the NAWQA Program is assessing the occurrence of MTBE in shallow ground water, streams, and the atmosphere in urban areas (Squillace and others, 1996; and Delzer and others, 1996). An investigation of the processes that affect the transport of MTBE and other VOCs from the atmosphere to the water table and through the hydrologic cycle is being conducted in an urban area near Glassboro, New Jersey (Baehr and others, 1997b).

## **SUMMARY**

An investigation of the fate and transport of MTBE from point-source gasoline releases is being conducted by intensive characterization of a field site in Beaufort, SC. The objective of the investigation is to evaluate the potential for natural attenuation of MTBE compared to BTEX by dilution from mixing, adsorption, biotic and abiotic transformation, and volatilization. Preliminary results indicate a decrease in aqueous BTEX and MTBE concentration near the source area from 1993 to 1997. This suggests that source removal and natural attenuation have been effective in decreasing contaminant concentrations in that area. Continuous increase in



MTBE concentration at a well located about 120 m downgradient from the source (to a high of 41,400  $\mu$ g/l in January 1997) indicates that MTBE still is migrating downgradient. Comparison with BTEX concentrations at the well indicates that BTEX is retarded with respect to MTBE. The presence of TBA in the plume at this time cannot be used to infer degradation of MTBE because of the possibility that TBA existed in the original gasoline. Aerobic conditions exist outside and along the periphery of the plume, and anaerobic conditions, with evidence of a range of redox conditions, are present within the plume, where BTEX and MTBE concentrations are highest. Analysis of gas-phase concentrations of BTEX and MTBE in the unsaturated zone indicates a mass flux of MTBE from the water table on the same order of magnitude as BTEX. Gas concentrations of both BTEX and MTBE within the unsaturated zone decline more sharply than predicted assuming transport with no transformation. To date, analysis of ground water from five other sites of both point-source and nonpoint-source release of MTBE have not detected TBA or TBF. This investigation of MTBE at point-source gasoline releases, sponsored by the USGS Toxic Substances Hydrology Program, is coordinated with USGS NAWQA Program activities to (1) assess the occurrence of MTBE in urban areas in shallow ground water, streams, and the atmosphere, and (2) investigate the transport of MTBE and other VOCs via the atmosphere and land surface to shallow ground water.

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