Application and Performance of Technologies for Treatment of MTBE and Other Oxygenates

Abstract

Introduction

As a result of fuel spills, thousands of sites across the United States have contaminated groundwater and soil. Most of these sites are service stations that have stored gasoline in leaking underground storage tanks. Recent data from the U.S. Environmental Protection Agency's (EPA's) Office of Underground Storage Tanks (OUST) indicate that out of 436,500 confirmed releases of gasoline into the environment, 139,500 still require cleanup.

Many of these sites are contaminated with fuel hydrocarbons, most often benzene, toluene, ethylbenzene, and xylene (BTEX) compounds, as well as fuel oxygenates such as methyl tert-butyl ether (MtBE) and tert-butyl alcohol (TBA). MtBE has been detected nationwide in soil, groundwater, and surface water. Federal and state studies have found that MtBE contamination has reached drinking water sources in many locations, including areas where the use of oxygenated fuel has not been mandated (GAO, 2002). Thirty-one states have established standards for MtBE in drinking water, and 42 states have established MtBE cleanup levels for soil and groundwater.

EPA Evaluation of Treatment Technologies Used to Remediate Oxygenate Sites

EPA is in the process of publishing a new report that provides a summary and evaluation of available data about seven technologies used to treat MtBE and other oxygenates. The report evaluates data from EPA's on-line database of MtBE Treatment Profiles (*http://cluin.org/products/mtbe*), and other sources, that include information about the design, operation, performance, and cost of treatment at sites contaminated with MtBE. The report also provides information about the anticipated performance of these technologies for treating other oxygenates, such as ethyl tert-butyl ether (ETBE), tert-amyl methyl ether (TAME), diisopropyl ether (DIPE), TBA, ethanol, and methanol, although information about treatment and occurrence of these other oxygenates is limited. The sources of these project data are journal articles, conference proceedings, interviews with practitioners, and direct data input from site managers and technology vendors. The technologies consist of *in situ* bioremediation, *in situ* chemical oxidation, air sparging, soil vapor extraction (SVE), multi-phase extraction, phytoremediation, and pump-and-treat. Often these technologies are used in combination to cost-effectively address soil and groundwater or high and low contaminant concentrations.

Purposes for Paper

The purposes for this paper are to (1) provide a summary of EPA's new report on MtBE treatment technologies; (2) provide an update on EPA's MtBE database/web application; and (3) present an updated, detailed evaluation of the performance data for treatment technologies used for MtBE and other fuel oxygenates. This evaluation will consist of a review of the available performance data for MtBE and the anticipated performance of these technologies for treating non-MtBE oxygenates. The paper will discuss factors that potentially impact the performance of the technologies and provide observations about the use of technologies for treating MtBE and other oxygenates.

Notice and Disclaimer

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Application and Performance of Technologies for Treatment of MtBE and Other Fuel Oxygenates

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As a result of fuel leaks and spills, thousands of sites across the United States are contaminated with fuel hydrocarbons and oxygenates. Most of these sites are service stations that have stored gasoline in leaking underground storage tanks. Recent data from the U.S. Environmental Protection Agency's Office of Underground Storage Tanks indicate that out of 436,500 confirmed releases of gasoline into the environment, 139,500 still require cleanup (Blue Ribbon Panel, 1999).

Many of these sites are contaminated with fuel hydrocarbons, most often benzene, toluene, ethylbenzene, and xylene (BTEX) compounds, as well as fuel oxygenates such as methyl tert-butyl ether (MtBE) and tert-butyl alcohol (TBA). MtBE has been detected nationwide in soil, groundwater, and surface water. Federal and state studies have found that MtBE contamination has reached drinking water sources in many locations, including areas where the use of oxygenated fuel has not been mandated (GAO, 2002). Thirty-one states have established standards for MtBE in drinking water, and 42 states have established MtBE cleanup levels for soil and groundwater (Delta Environmental, 2004).

There are many challenges associated with the assessment and remediation of sites contaminated with MTBE and other oxygenates. For example, fuel oxygenates are generally more soluble, less likely to partition to organic matter in soil, and slower to biodegrade than other contaminants in fuel, such as BTEX. This can result in larger and more widespread groundwater plumes and challenges with employing certain types of treatment technologies. In addition, characterization at sites contaminated with fuel oxygenates must consider these factors to assure that the nature and extent of contamination is accurately assessed.

The purposes for this paper are to (1) provide a summary of EPA's new report on MtBE treatment technologies; (2) provide an update on EPA's MtBE database/web application; and (3) present an updated, detailed evaluation of the performance data for treatment technologies used for MtBE and other fuel oxygenates. This evaluation will consist of a review of the available performance data for MtBE and the anticipated performance of these technologies for treating non-MtBE oxygenates. The paper will discuss factors that potentially impact the performance of the technologies for treating MtBE and other oxygenates.

EPA Report on Treatment Technologies Used to Remediate Oxygenate Sites

EPA is in the process of publishing a new report, called *Technologies for Treating MtBE and Other Fuel Oxygenates*, that provides a summary and evaluation of available data about seven technologies used to treat MtBE and other oxygenates. The report evaluates data from EPA's on-line database of MtBE Treatment Profiles (http://www.cluin.org/products/mtbe), and other sources, and includes information about the design, operation, performance, and cost of treatment at sites contaminated with MtBE. The report also provides information about the anticipated performance of these technologies for treating other oxygenates, such as diisopropyl ether (DIPE), ethyl tert-butyl ether (ETBE), tert-amyl methyl ether (TAME), TBA, ethanol, and methanol, although information about treatment and occurrence of these other oxygenates is limited. The sources of these project data are journal articles, conference proceedings, interviews with practitioners, and direct data input from site managers and technology vendors. Technologies discussed in the report consist of in situ bioremediation, in situ chemical oxidation, air sparging, soil vapor extraction (SVE), multi-phase extraction, phytoremediation, and pump-and-treat. Often these technologies are used in combination to cost-effectively address soil and groundwater or high and low contaminant concentrations. Results from EPA's report are provided later in this paper.

Update of EPA's MtBE Database/Web Application

As recently as several years ago, there was limited information available about using remediation technologies to address sites contaminated with fuel oxygenates. To address this information need, EPA has worked to make available information about the characterization and treatment of sites contaminated with MtBE and other oxygenates, including the publication of fact sheets, technical reports, and other documents. In 2000, EPA's Office of Solid Waste and Emergency Response began an effort to compile information about actual cleanup sites where the treatment of MtBE and other oxygenates has taken place. In April 2002, EPA published an online database of this information as MtBE Treatment Profiles, located at the website http://cluin.org/products/mtbe/. This website is intended to be used by cleanup professionals and researchers; federal (Superfund and RCRA remediation site managers), state, and local regulators; remediation consultants; water treatment plant designers and operators; and other interested parties as a starting point for identifying technologies that have been used for the treatment of MtBE and other oxygenates, as well as for identifying other environmental professionals and technology providers or remediation consultants that may serve as resources.

As of April 2004, the database contained information about 390 projects, consisting of 263 ongoing and 126 completed projects. From April 2002 through April 2004 the website has had over 64,000 hits.

The MtBE Treatment Profiles website is a searchable database of projects at sites that performed treatment of MtBE and other oxygenates in groundwater, soil, or drinking water. The website contains project treatment profiles that include project information, cost and performance information, points of contact, and references, as shown in Exhibit 1.

Exhibit 1: Types of Information in Treatment Profiles

Project Information	Site name, location, type, lithology, depth to groundwater Contaminant(s) and media treated Area of contamination Technology design and operation, including the number of wells, scale, vendor, period of operation, and status
Cost and Performance Information	Cleanup goals Concentration data for MTBE, TBA, & BTEX (before and after treatment) Cost for remediation (Capital, O&M, Assessment, and Monitoring)
Point of Contact	Contact information (name, title, affiliation, mailing address, phone, fax, and e-mail address)
References	Sources of information used to prepare profile

The website allows users to search for treatment profiles and to submit new profiles or update existing profiles. New or updated profiles are submitted frequently. The site provides a search engine that allows a user to search the profiles by contaminant, media, technology, scale, status, state, site name, or by performing a keyword search. Alternately, a user may browse a list of all profiles in the database.

In addition to serving as a tool for identifying existing and completed cleanup projects, the website provides a portal to other environmental professionals and technology providers. Each profile provides information on point(s) of contact, allowing more detailed information about the profile to be acquired directly from those individuals involved with the site. EPA strongly encourages communication between environmental professionals involved with treatment of fuel oxygenate sites and is actively working to expand and update the treatment profiles in the database.

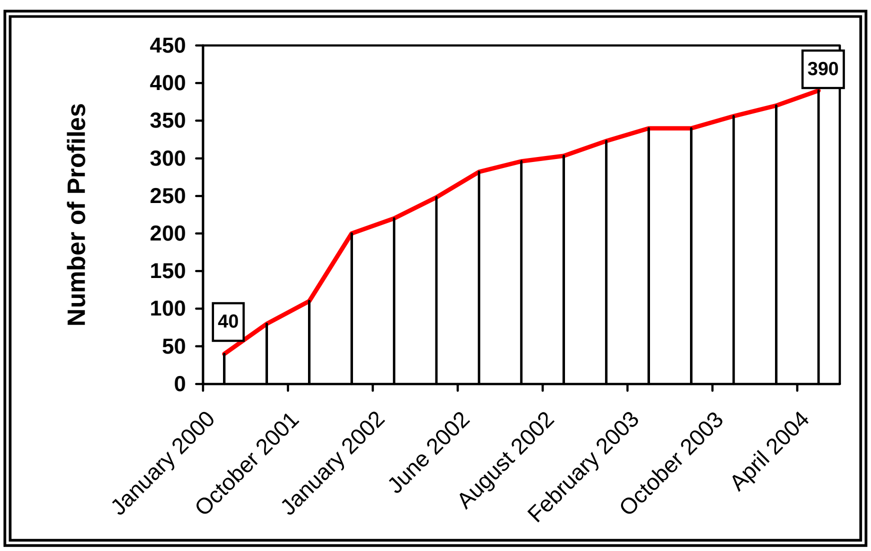
To prepare the profiles, EPA obtained data from site managers, regulatory officials, and technology providers, as well as from published reports, conference proceedings, and other available reference materials. Consequently, each profile has a varying level of detail, depending on the data and information that was available. In addition, some of the profiles include active links to more detailed case studies, which present in-depth information about the treatment sites. No additional testing of technologies was performed during the preparation of the treatment profiles and no independent review was performed for the data provided by project managers and technology vendors. The performance and cost data included for the projects are provided as general information and should not be used as a sole basis to select future MTBE remediation projects or to compare among technologies. Thus, EPA cannot guarantee the accuracy or completeness of these data.

EPA encourages project managers, site owners, and technology providers to add new treatment profiles to this site. To submit a new profile, a user only needs to select the 'Submit a New MTBE Treatment Profile' button and fill in the site information as prompted by selecting options from the drop down boxes where appropriate, and providing numerical or text data where drop down boxes are not provided. As an effort to keep the site profiles current and up-to-date, EPA also encourages users to update site profiles as appropriate. To update a profile, a user may select the 'Update an MTBE Treatment Profile' button and fill in the site information that has changed. For instance, an end user may submit a site profile while it is still an on-going project, but may later update the profile information once the treatment project is complete and the site has reached closure. This functionality allows users to track current remediation projects at all stages of development.

Analysis of Available Information

Since EPA began collecting information for the first 40 treatment profiles in 2000, the number of profiles has grown steadily as the number of MTBE treatment projects increased and the information about these projects became available. As shown in Exhibit 2, the database contained a total of 390 profiles as of April 2004.

Exhibit 2: Growth of Treatment Profile Database

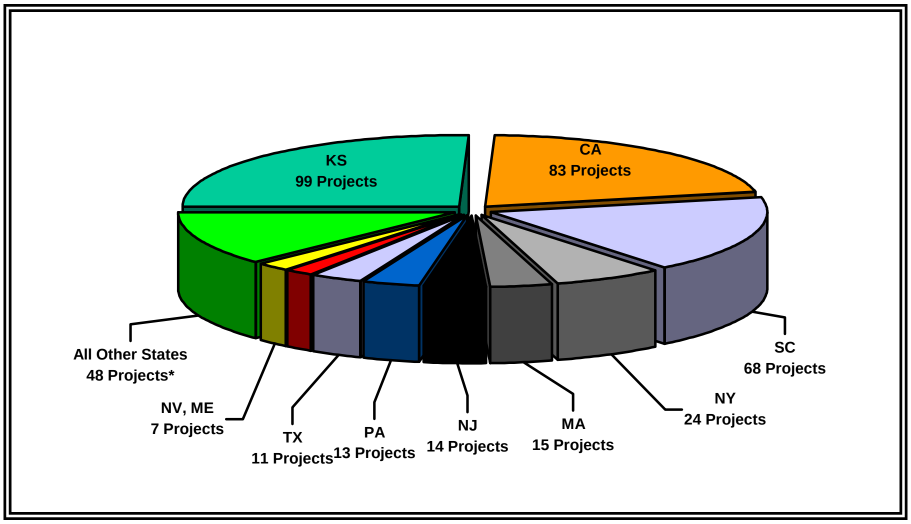


EPA has analyzed the dataset from these 390 treatment profiles as it relates to the locations of the treatment projects, types of technologies employed, types of contaminants treated, treatment technology performance and cost. The analyses were based solely on data from the 390 treatment projects that was current as of April 2004. Their results are not intended to represent all projects that performed treatment for MTBE or other fuel oxygenates, but should provide environmental professionals and other interested parties at sites contaminated with MTBE and other oxygenates with information that can assist them in evaluating treatment technologies.

Geographical Location

Exhibit 3 shows the distribution of projects by location for the 390 profiles. The majority of the projects in the database are in Kansas, California, and South Carolina.

Exhibit 3: Geographical Distribution of 390 Projects in the Profile Database



*****The following states had fewer than 7 projects: FL, CO, WI, NM, IN, MI, OR, NC, CT, UT, IL, AZ, MD, DE, LA, MO, NE, WY, NH, RI, TN, VT, WA, MT

Treatment Technologies Employed

The 390 projects employed all of the technologies commonly used to treat MTBE and other oxygenates, including air sparging, bioremediation (*in situ* and *ex situ*), *in situ* chemical oxidation, pump-and-treat, MPE, SVE, and phytoremediation to remediate MTBE in groundwater and soil. The projects included in the database focus on those using active treatment technologies to treat MTBE and other oxygenates. Therefore, it does not include projects that primarily employed non-active treatment remedies such as natural attenuation or institutional controls. In addition, the database generally does not include projects where the only remedial technology employed was a removal technology, such as excavation or product recovery. Exhibit 4 summarizes the types of technologies employed at the 390 projects. For the projects in the database, SVE, air sparging, pump-and-treat, and in situ bioremediation were used more frequently to remediate groundwater and soil contaminated with MTBE, and in situ chemical oxidation, MPE, and phytoremediation were used less frequently. Many of the projects used more than one technology, such as air sparging plus SVE. Most (84%) of the 390 projects are full-scale; 13% are pilot-scale and 3% are bench-scale.

Exhibit 4: Breakdown of Technologies for 390 Projects

Treatment Technologies	Number of Profiles*
Soil Vapor Extraction	175
Air Sparging	155
Pump and Treat	101
In Situ Bioremediation	92
In Situ Chemical Oxidation	26
Multi-Phase Extraction	17
Drinking Water Treatment	15
Ex Situ Bioremediation	12
Phytoremediation	9
Thermal Desorption	4

*Note: May be more than one technology per profile.

Contaminants Treated

While the treatment profiles primarily focus on projects where MTBE was the contaminant treated, a number of these projects also provided information about other contaminants that were treated along with MTBE. Where this information about other contaminants was provided, it was included in the treatment profiles. As shown on Exhibit 5, contaminant data was available for several contaminants including MTBE, TBA, TAME, DIPE, ethanol, and BTEX. Of these contaminants, all projects had MTBE as a contaminant with nearly three-quarters also having BTEX as a contaminant. TBA (11%), TAME (2%), DIPE (0.5%), and ethanol (0.1%) were reported as being present for only a small percentage of the projects. Information related to oxygenates other than MTBE was only included in the treatment profiles if such information was provided. For the 390 projects, 328 (84%) provided MTBE concentrations (either initial or final concentrations, or both) and 116 (30%) provided MTBE cleanup goals. Reported MTBE treatment goals ranged from 0.5 µg/L to more than 8,600 µg/L.

Exhibit 5: Contaminant Distribution for 390 Projects

Contaminant Type	Profiles Reporting Contamination	Profiles Providing Concentration Data	Profiles Providing Cleanup Goals
MTBE	390 (100%)	328 (84.1%)	116 (29.7%)
TBA	43 (11%)	40 (10.3%)	19 (4.9%)
TAME	8 (2.1%)	0	0
DIPE	2 (0.5%)	0	0
Ethanol	3 (0.08%)	0	0
Other Contaminants			
BTEX	287 (73.6%)	210 (53.8%)	84 (21.5%)

Treatment Technology Performance

Treatment technology performance data in the report include information about maximum concentrations treated, comparisons with reported cleanup goals, and progress towards achieving the goals. Most performance data are for treatment of MtBE; the report also includes limited data about treatment of other oxygenates. Actual performance data for treatment of non-MtBE oxygenates is supplemented with a general evaluation of anticipated performance, based on their physical and chemical properties.

As discussed above, these performance data are based on the data provided by project managers and others in the source materials used to prepare the treatment profiles website. The profiles varied in the level of detail provided for performance data. Many of the treatment profiles contained only limited information about treatment performance, generally including a maximum concentration of MtBE for before-treatment and a maximum concentration after-treatment. These concentrations often were limited to discussing how the treatment technology performed relative to the cleanup goals for the site. Most treatment profiles did not provide detailed performance data, such as MtBE concentration over the duration of the project, or statistical evaluations of performance data with confidence limits.

As mentioned before, treatment performance is site-specific, and depends on many factors; thus it is difficult to extrapolate from one site to another. The data are provided to give a general indication of the technologies' performance.

Of the 390 projects, 126 (32%) were reported as being complete. Of those completed projects, 95 projects reported either before- or after-treatment concentration data. The available technology performance information for these projects is included in the treatment profiles primarily in terms of changes in concentration of MTBE in the groundwater (as opposed to concentrations of MTBE in the soil). In general, the highest concentration reported prior to beginning treatment and the highest concentration after treatment was completed (shown as "final concentration") is provided. A summary of the MTBE concentrations before- and after-treatment for the 95 completed projects is shown in Exhibit 6 (shown both graphically and in tabular format). Performance data, typically in the form of initial and the most current available concentration data, are also available for 232 of the 264 ongoing treatment projects in the database.

Exhibit 6: Completed Project Performance Summary (95 Projects) – Graphical View*

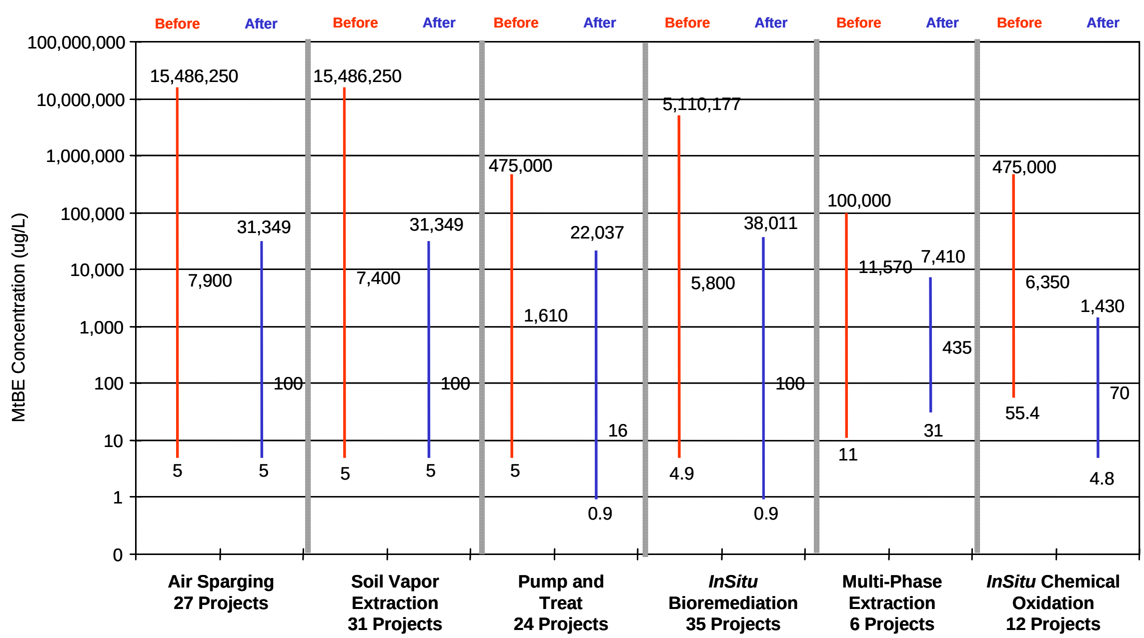


Exhibit 6: Completed Project Performance Summary (95 Projects) – Tabular View*

*Note: May be more than one technology per profile.

	Treatment	Max	Median	Min
Air Sparging	Before	15,486,250	7,900	5.0
	After	31,349	100	4.8
Soil Vapor Extraction	Before	15,486,250	7,400	5.0
	After	31,349	100	4.8
Pump and Treat	Before	475,000	1,610	5.0
	After	22,037	16	0.9
***InSitu* Bioremediation**	Before	5,110,177	4,300	4.9
	After	38,011	100	0.9
Mulit-Phase Extraction	Before	100,000	11,570	11.0
	After	7,410	435	31.0
***InSitu* Chemical Oxidation**	Before	475,000	6,350	55.4
	After	1,430	70	4.8

Factors Affecting Remedial Technology Performance and Cost

Many factors can affect the performance and cost for implementing a remedial technology at a given site, including the nature and extent of contamination, depth of contamination, physical and chemical characteristics of a site (such as dimensions and hydrogeology), design and operation of a treatment system, regulatory requirements, and logistical issues. Typically, these factors are quantified before an engineering level design is made for use of a technology at a given site. For example, sites with spilled or leaked gasoline often contain 200-300 distinct chemicals which can contaminate soil and groundwater; approximately 6-10% of the spilled/leaked gasoline typically consist of fuel oxygenates such as MtBE (McGarry, 2002).

Additional factors include duration of the release (such as a gasoline leak), presence of down-gradient water supply wells, and distribution of contaminants in soil and groundwater.

Observations About Remedial Treatment Technologies

The following observations are based on the data in the 390 treatment profiles discussed above. Cost data for the following treatment technologies can be found in the "Cost of *In situ* Treatment of Fuel Oxygenates" paper that was presented at the NGWA Conference on Remediation: Site Closure and Total Cost of Cleanup, in New Orleans, LA, on November 13 – 14, 2003 (Fiedler & Berman, 2003).

Air Sparging

- Widely used to remediate groundwater contaminated with MtBE
- Most often used alone or in conjunction with SVE; to a lesser extent has been employed with pump-and-treat or other technologies
- Has achieved MtBE concentration reductions greater than 99 percent
- Increased airflow (compared to a system employed to treat only BTEX) is required to physically strip common oxygenates using air sparging; based on their Henry's Law Constants, alcohol-based oxygenates would require greater airflow than ether-based oxygenates

Soil Vapor Extraction

- Widely used to remediate soil and groundwater contaminated with MtBE
- Has achieved MtBE concentration reductions greater than 99 percent
- All of the common oxygenates have vapor pressures that are greater than 10 mm Hg and are amenable to treatment using SVE, with ether-based oxygenates generally having greater vapor pressures than alcohol-based oxygenates
- GAC adsorption may be less effective in removing alcohol-based oxygenates from SVE off gas

Multi-Phase Extraction

- Has been used to remediate sites contaminated with MtBE, but less frequently than some other technologies, such as air sparging and *in situ* bioremediation
- Has achieved MtBE concentration reductions greater than 99 percent
- Most often used without other *in situ* technologies; to a lesser extent has been employed in conjunction with air sparging or ISCO
- While the contaminant properties are important considerations in the selection and design of an MPE system for a given site, the applicability of MPE is more dependent on media properties, primarily hydraulic conductivity and transmissivity

In Situ Bioremediation

- Widely used to remediate soil and groundwater contaminated with MtBE
- Has achieved MtBE concentration reductions greater than 99 percent
- Most research has focused on bioremediation of MtBE and TBA; information on biodegradation of other ether-based oxygenates is limited, but similar degradation pathways are employed for many of these oxygenates
- Presence of ethanol may affect the aerobic biodegradation of MtBE and lead to longer MtBE plume; recent research has indicated that ethanol can deplete available electron acceptors and stimulate methanogenesis
- Effective biodegradation of MtBE and other oxygenates has been reported in a variety of applications of the aerobic pathway; results for anaerobic pathways have been confined to laboratory studies and have not been consistent
- Intermediate degradation products of MtBE and other oxygenates, such as TBA, may remain in the subsurface under natural conditions or when using anaerobic pathways
- Oxygenate-degrading microorganisms are typically slow growing; bench- and pilot-studies may be needed to confirm the applicability of bioremediation and to select an appropriate design

In Situ Chemical Oxidation

- Has been used to remediate sites contaminated with MtBE, but less frequently than some other technologies, such as air sparging and *in situ* bioremediation
- The most common oxidant applied was hydrogen peroxide with ferrous iron (Fenton’s chemistry)
- Has achieved MtBE concentration reductions greater than 99 percent
- All ether- and alcohol-based oxygenates susceptible to chemical oxidation
- Applicability to given site based primarily on factors not related to type of oxygenate (that is, all oxygenates can be treated using chemical oxidation, but certain other site conditions, like high concentrations of native organic matter or low permeability, may make other treatment technologies more attractive)

Pump-and-Treat

- Widely used to remediate soil and groundwater contaminated with MtBE
- Has achieved MtBE concentration reductions greater than 99 percent
- Properties of MtBE (high water solubility and low organic/water partition coefficient) make it amenable to groundwater extraction
- Air Stripping – Treatment of ether-based oxygenates may require greater air to water ratios than treating only BTEX; treatment of alcohol-based oxygenates may be impractical
- Adsorption – Ether-based oxygenates are less readily removed than BTEX using GAC and some alcohol-based oxygenates may not be adsorbable at all; synthetic resins that more selectively remove fuel oxygenates are available
- Chemical Oxidation – Fuel oxygenates can be destroyed using hydroxyl radical oxidation; oxidant dosage and contact time based more on overall oxidant demand of extracted groundwater than on type of oxygenate
- Biotreatment – Fuel oxygenates can be biodegraded given adequate retention time in a bioreactor with a sufficient mass of conditioned microbes

Next Steps

In an effort to ensure that the information in EPA's MtBE database/web site is correct and up-to-date, EPA periodically requests updates from the individuals listed as points of contact for the treatment profiles and any new information is included in the database. As a result of this effort, EPA has recently received responses and updated information for more than two-thirds of all the projects.

EPA also continues to increase the information available in the database, both by adding new profiles and by expanding the data fields for new and existing profiles.

With EPA's ongoing efforts and participation from environmental professionals to add new profiles and update existing profiles, EPA is making this website a more valuable tool for individuals remediating sites contaminated with MTBE and other oxygenates.

Other Resources

Some additional EPA resources about MTBE and other oxygenates are listed below.

EPA's MTBE Web Page – Provides a list of Frequently Asked Questions that provide basic background information on MTBE, as well as links to other websites. Available at http://www.epa.gov/mtbe
EPA's Office of Underground Storage Tanks MTBE Web Page – Provides general information about MTBE and USTs. Available at http://www.epa.gov/swerust1/mtbe/
Clu-In – A website that provides information about innovative treatment and site characterization technologies while acting as a forum for all waste remediation stakeholders. Available on line at http://www.cluin.org
TechDirect – Hosted by the U.S. EPA's Office of Superfund Remediation and Technology Innovation, TechDirect is an information service that highlights new publications and events of interest to site remediation and site assessment professionals. Sign up on line at http://www.cluin.org/newsletters/
Technology News and Trends – A newsletter about soil, sediment, and groundwater characterization and remediation technologies Available on line at http://www.clu-in.org/products/newsltrs/tnandt/.

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