

Fate, Transport and Remediation of MTBE

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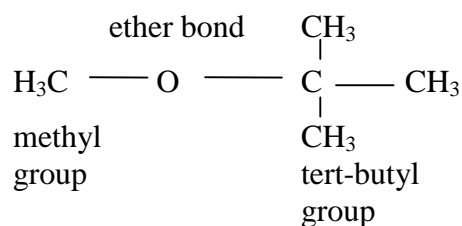
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I. Fate and Transport of MTBE in the Environment

The fate of methyl tertiary butyl ether (MTBE) in the environment and remediation of ecosystems once they are contaminated with it are directly related to the compound's chemical and physical properties. MTBE is comprised of a methyl (CH₃ group) bonded by an oxygen atom to a butyl (C₄H₉) group.



The C-O-C linkage makes this compound an ether. Ethers are generally hydrophilic (water-loving) and are therefore very soluble in water. For example, the solubility of pure MTBE in water is 50,000 mg/L. This differs greatly from the other natural constituents of gasoline (i.e., petroleum hydrocarbons) that consist of carbon and hydrogen alone and are relatively insoluble in water. In contrast, pure benzene, one of the gasoline hydrocarbons that most readily dissolves in water, has a maximum solubility of 1780 mg/L. MTBE is generally 30 times more soluble in water than the hydrocarbon constituents in gasoline (Table 1). If MTBE is released into air, as a result of incomplete combustion of fuel, it will readily dissolve into precipitation because it is 42 times more soluble in water than air. Hence, during precipitation events MTBE is transferred from the atmosphere to the aqueous phase.

Once MTBE is dissolved in water it prefers to stay there. Most gasoline hydrocarbons, such as benzene, are readily removed from water to air. Henry's Constant expresses the ratio of a compound's concentration in air relative to its concentration in water at a given temperature. The larger the Henry's Constant, the more volatile a compound is (i.e., the more readily it moves from water to air). The Henry's Constant for MTBE is 0.022 at 25°C. This contrasts to benzene with a Henry's Constant of 0.22 at 25°C. Compounds with values ≥ 0.05 are considered very volatile. This does not mean, however, that MTBE is not volatile. For example, if a container

Table 1
Water Solubilities of Hydrocarbon Compounds

Compound	Solubility	
<i>Solubility (mg/L) at 20°C from conventional gasoline^a</i>		
<i>Aromatic gasoline hydrocarbons</i>		
Benzene	18	
Toluene	25	
Ethylbenzene	3	
Xylenes (total)	20	
<i>Solubility (mg/L) at 20°C</i>		
<i>Chlorinated solvent compounds from the pure compound</i>		
trichloroethylene (TCE)	1440	
perchloroethylene (PCE)	240	
<i>Solubility (mg/L) at 20°C</i>		
<i>Alkyl ether compounds</i>		
	<i>from RFG^b</i>	<i>from oxyfuel^c</i>
methyl <i>tert</i> -butyl ether (MTBE)	4700	6300
ethyl- <i>tert</i> -butyl ether (ETBE)	1300	1750
<i>tert</i> -amyl methyl ether (TAME)	1400	1850
di-isopropyl ether (DIPE)	1200	1600

^a Assumes release of a conventional gasoline containing 1% benzene, 5% toluene, 1.5% ethylbenzene, and 10% total xylenes.

^b Assumes release of reformulated (RFG) gasoline containing 2.0% by weight oxygen, which would correspond to 11.1% MTBE, 12.9% ETBE, 12.4% TAME, or 12.9% DIPE (all by volume).

^c Assumes release of oxygenated gasoline containing 2.7% by weight oxygen, which would correspond to 15.0% MTBE, 17.5% ETBE, 16.8% TAME, or 17.4% DIPE (all by volume).

Source: Johnson et al., 2000

of **pure** MTBE was open to the atmosphere, it would be 2.6 times more likely to volatilize into the air than would **pure** benzene under the same conditions. The important distinction is that MTBE becomes relatively non-volatile once it is dissolved in water.

When gasoline is released from a leaking underground storage tank or vehicular accident, the MTBE is dissolved in the gasoline. Typical, concentrations of MTBE in reformulated gasoline (RFG) or oxygenated fuel (oxy fuel) range from 10 to 15%. Because MTBE is soluble in gasoline as well as water, it will partition itself between both phases. For example, the concentration of MTBE in water at 25°C in contact with a gasoline containing 10% MTBE will be 5000 mg/L (5,000,000 µg/L). While this concentration is an order of magnitude lower than if pure MTBE were present, the massive environmental implications of this (5,000,000 µg/L) solubility are clear if one contrasts it to the existing USEPA health advisory concentration of 70 µg/L (a 10,000 fold difference). The concentration of MTBE must be <20-40 µg/L to avoid taste and odor problems. Some states, such as New Hampshire and California, have set strict primary drinking water standards for MTBE. In New Hampshire, the standard is 13 µg/l. Compared to other organic contaminants that commonly trigger remediation actions in water such as TCE and benzene, MTBE has a very high solubility in water. For example, the solubility of the other gasoline hydrocarbons in water is only 120,000 µg/L. As a result of this very high aqueous phase solubility, one gallon of gasoline containing 10-15% MTBE can potentially contaminate 4 million gallons of drinking water.

MTBE that volatilizes into the air can be degraded to CO₂ and water when it absorbs ultraviolet light from the sun. However, if the MTBE comes in contact with precipitation in the atmosphere first, it will dissolve into the water and be stripped from the atmosphere. It will move with the precipitation as it flows over impervious surfaces such as roadways or as it infiltrates into the ground. The runoff from the impervious surfaces will either flow into streams

and rivers or be collected in a storm sewer and ultimately discharged into a local water body, contaminating it with MTBE.

Uncombusted fuel from outboard engines and jet skis is released directly in surface waters and serves as a point source of MTBE. If the water infiltrates into the ground, it will move down through the unsaturated zone of soil until it reaches and mixes with the groundwater, contaminating the aquifer with MTBE.

MTBE from fuel spills will move with the gasoline until it reaches a water body (via runoff) or it will percolate down through the soil until it reaches the groundwater. Because gasoline is relatively insoluble and less dense than water, it will pool on top of the water. The MTBE will then begin to move from the gasoline into the water. This is the mechanism by which MTBE in gasoline contaminates surface water bodies and groundwater.

Unlike many other contaminants in groundwater, MTBE does not readily sorb to soil or rock surfaces, because it is highly soluble in water. A contaminant's mobility in groundwater is expressed as a Retardation Factor (R) where:

$$R = \frac{\text{groundwater velocity}}{\text{compound's velocity}}$$

For MTBE, R is approximately 1, indicating that MTBE will move through the subsurface at the same velocity as the groundwater in which it is dissolved. This contrasts with other gasoline constituents such as the BTEX compounds (benzene, toluene, ethylbenzene, xylenes) that are retarded because they sorb to soil/rock surfaces. The high solubility of MTBE in water, in concert with its low sorption characteristics, mean the MTBE plumes can move on the scale of kilometers away from the source of contamination. BTEX plumes typically move ≤ 100 meters from the source. Groundwater velocity, and hence the velocity of MTBE, is dependent upon the permeability and porosity of the formation (which is a function of the type of soil or rock) and the hydraulic gradient. In addition, the pumping of drinking water wells can induce a stress on

the groundwater affecting its velocity. Natural groundwater velocities range from millimeters per year to as much as one meter per day.

Three processes control the fate of MTBE in groundwater: dispersion, dilution and degradation. Dispersion causes the plume of contamination to expand both perpendicular to the direction of groundwater flow and in the direction of flow. It is caused on the microscopic scale by the irregular surface of the soil or rock through which the water is flowing. Microscopic dispersion is particularly important in fractured rock. Macroscopic dispersion is caused by inhomogeneities such as gravel lenses or clay pockets (i.e., layering) in soils.

Dilution occurs when the MTBE plume water is mixed with uncontaminated groundwater. This phenomenon can occur, for example, when a water supply well draws groundwater from a radius around itself. In this case, the plume water constitutes only a part of the water within the well's radius of influence, with clean water comprising the remaining portion of the water drawn into the well.

II. Remediation of MTBE

Dispersion and dilution reduce the concentration of MTBE by mixing the contaminated water with cleaner water, but the total mass of MTBE present *in situ* does not change. Degradation reduces the concentration of MTBE in water by converting it to other compounds. More importantly, degradation reduces the mass of MTBE *in situ*. MTBE in surface and ground water can be treated by pumping the water into an engineered system (*ex situ* treatment) or it can be treated directly in the environment (*in situ* treatment).

A. Ex Situ Remediation

When MTBE is removed from groundwater or surface waters using engineered reactors, the water must be pumped out of the surface water body or aquifer. In the latter case, it is essential to place wells strategically within or near the plume to collect the contamination.

Treated water can be discharged back into the water body or aquifer. There are three major types of engineered remediation processes that are used for MTBE: adsorption systems, air stripping systems, and advanced oxidation processes (AOPs).

The most common adsorbent is Granular Activated Carbon (GAC). This charcoal-like material has a very high surface area and is made from processing wood, bone, or similar products at high temperatures and pressures. GAC has a large capacity for adsorbing organic compounds. As a result, it is commonly used in adsorption systems that treat water containing gasoline compounds. Unfortunately, GAC has a relatively low sorptive capacity for MTBE, in part because the latter is so soluble in water. For example, in the Malley et al. (1993) study, a GAC unit used to treat gasoline-contaminated groundwater worked for only two weeks before it was saturated with MTBE and no more treatment occurred. Once saturated, the GAC must be replaced or regenerated by thermal oxidation or steam cleaning. While GAC can achieve 99% removal of MTBE, the short operational time is problematic in that it results in higher costs because of frequent replacement/regeneration. It is possible to use GAC as a polishing unit after another treatment process (e.g., air stripping). Synthetic sorbents, such as the commercially-fabricated resin Ambersorb[®], have been used instead of GAC because they have higher sorptive capacities for MTBE (Malley et al., 1993). However, they have a high capital cost and cannot be easily regenerated, further escalating the total cost.

Air stripping systems exploit the principle that MTBE can be transferred into the gas phase by bubbling air through the water (diffused bubble aeration) or passing it by a thin film of water (packed tower aeration). Air stripping works well for many gasoline compounds that are much more volatile than MTBE when dissolved in water (i.e., they have a higher Henry's Constant). Several types of air stripping devices are used including packed towers and diffused bubble aeration systems. Research conducted in New Hampshire by Malley et al. (1993) indicated that even when very large amounts of air were used (air: water ratio of 150:1), MTBE

removals were only 60%. Some air stripping systems have been reported to have removals of 99%, but lower removals are more typical because of MTBE's high solubility in water. In addition, the MTBE-laden air exiting the air stripping unit must be treated before discharge. This adds expense and complexity to the system and must be accomplished using sorption of the gas phase MTBE onto GAC or by thermal oxidation (combustion of the MTBE). If GAC is used, it must be regenerated after saturation, typically by thermal processing.

AOPs exploit the principle that MTBE can be chemically or physically oxidized to CO_2 and H_2O . The types of oxidants that can be used are: hydrogen peroxide (H_2O_2), ultraviolet (UV) light, and ozone (O_3). Use of AOPs to treat MTBE-contaminated water is promising, with some research indicating 99% removals are possible. As advances are made in AOP technology, its use in MTBE degradation may grow. There is much research ongoing (e.g., the American Water Works Research Foundation is funding an MTBE research project on AOPs in California) that could improve our understanding of AOP application in MTBE removal from water and reduce the high cost of operation.

Perhaps the greatest disadvantage of *ex situ* treatment processes for aquifers is that in order to insure the remediation has met the treatment goal, one must be satisfied that all of the contaminated water is completely captured by the well network. This is often very difficult to accomplish because it is hard to know the location of all of the contaminated water underground, especially because MTBE moves so readily in water. In addition, in the subsurface there is usually significant heterogeneity in the soils/aquifer materials, thereby making it difficult to completely predict where and how the groundwater and MTBE move. In bedrock systems, knowledge of contaminant plumes and movement are further complicated by the very complex pattern of fractures in the rock.

B. In Situ Remediation

Treating MTBE in the environment where it occurs (i.e., *in situ*) eliminates the need to be able to completely remove it from the aquifer. *In situ* remediation is accomplished using microorganisms that live in the environment and can biodegrade MTBE.

In surface and ground waters and soils, most MTBE degradation is a result of microbiological activity. The only other naturally-occurring degradation process – photolysis – is limited because UV light from the sun does not penetrate far into these media. The degradation rate of a contaminant in the environment is often expressed in terms of half-life ($t_{1/2}$). $t_{1/2}$ is the time it takes for $\frac{1}{2}$ of the original compound present to degrade. The $t_{1/2}$ of MTBE in surface waters is greater than 1 day. For example, it will take approximately 6.7 half-lives (7 days) to achieve the two orders of magnitude of degradation needed to reduce a concentration of MTBE of 1000 $\mu\text{g/L}$ to the New Hampshire primary drinking water standard of 13 $\mu\text{g/L}$. In a river flowing at 1 foot/second, the MTBE plume would have traveled 108 miles in this time. In groundwater, the $t_{1/2}$ is not well known, but is at least 2 years. In this case, assuming a 2-year $t_{1/2}$, it will take approximately 13 years to achieve the two order of magnitude reduction in MTBE. This contrasts with a 2-3 month $t_{1/2}$ for BTEX in groundwater. MTBE is resistant to biodegradation because the tertiary butyl carbons are associated with the ether linkage and the branching off the base molecule is only one carbon long making it hard for microbes to attack the molecule.

The biodegradation pathways of MTBE are not yet fully understood. One generalized pathway is shown in Figure 1. Microorganisms can use the MTBE as a sole carbon and energy source or can degrade it along with another organic carbon source (e.g., pentane). The latter process is called co-metabolism. In the process of using MTBE as an energy source, the microbes remove electrons and hydrogen ions from the molecule and pass them down a chain of

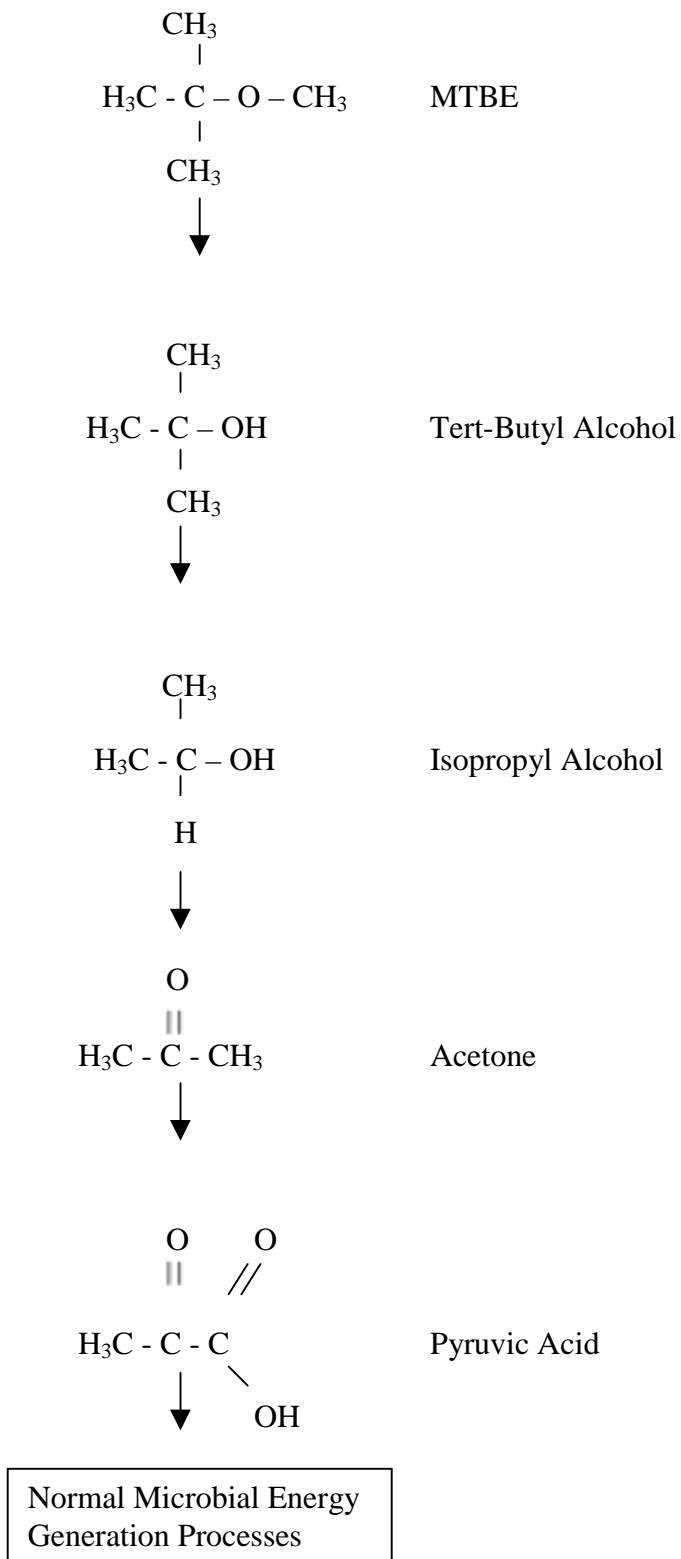


Figure 1. Generalized pathway of MTBE biodegradation based on Church et al. (2000) and Steffan et al. (1997).

oxidation-reduction reactions to a terminal electron acceptor (TEA). When oxygen is present, they will use it as a TEA because it produces the most energy to fuel cell maintenance and growth. Most lab studies on MTBE biodegradation have involved aerobic conditions. A variety of microorganisms, including pure cultures of bacteria and fungi and microbial consortia from waste treatment systems, and contaminated and uncontaminated environments have been found to degrade MTBE aerobically.

However, many contaminated aquifers do not contain significant amounts of oxygen. Some recent studies have shown that microbial consortia can use TEAs other than oxygen (e.g. nitrate, sulfate and carbonate/hydrogen), suggesting that oxygen may not be essential for MTBE biodegradation. However, the rates of some of these anaerobic reactions are slower than those with oxygen.

Some field studies have been conducted to monitor the natural rate of attenuation (degradation) of MTBE in gasoline-contaminated aquifers. Typically, BTEX degradation occurs first followed by MTBE. In some studies, MTBE is not degraded *in situ* at all. In a 1998 report by Hitzig et al., 13 of 70 sites contaminated by MTBE were relying on natural attenuation for remediation. One such site is the Borden Test Site in Ontario, Canada. In the first 16 months after a gasoline release, BTEX concentrations decreased significantly in the aerobic aquifer, but MTBE was hardly affected. Sampling that occurred 7 years later found MTBE concentrations reduced to <200 µg/L from several 1000 µg/L initially. Borden et al. (1997) reported MTBE degradation in a shallow coastal plain aquifer in North Carolina under a mix of aerobic and denitrifying conditions (using nitrate as a TEA). Wilson et al. (2000) conducted laboratory studies using sediment from an anaerobic aquifer contaminated with petroleum hydrocarbons. The rate of attenuation of MTBE under methanogenic conditions ($\text{CO}_3^{2-}/\text{H}_2$ as a TEA) was similar to that observed at the field scale. However, degradation rates under these conditions

were so slow that it will take 60 years for the MTBE concentration to be reduced from 1200 µg/L to 30 µg/L.

While naturally-occurring biodegradation offers an attractive alternative for *in situ* remediation of MTBE, it may be limited by the long lag times needed before degradation occurs consistently. In addition, because of the slow growth achieved and perhaps because MTBE is not similar to petroleum hydrocarbons or other organic contaminants, few microbes seem to degrade MTBE. In some cases, MTBE biodegradation has stopped at tert-butyl alcohol (Figure 1), which means it has not been completely degraded to CO₂ and H₂O. Some studies suggest that MTBE will not be degraded at all until other more readily degradable compounds (e.g., BTEX) are eliminated.

It may be possible to speed MTBE biodegradation *in situ* by adding substances to the subsurface. For example, Salanitro et al. (2000) injected oxygen, and oxygen and microorganisms to an MTBE plume in Port Hueneme, CA with favorable results (concentration decreased from 2000 to 2 µg/L). In addition, some bioremediation companies have reported accelerated MTBE degradation with the use of their proprietary products. These studies have been focused on soil-based aquifers and should have independent third party verification. Little to no research work has been performed on MTBE biodegradation in fractured rock.

The challenge of remediating MTBE in groundwater is one of the most difficult subsurface contamination problems facing the nation because of the compound's propensity to dissolve into water at high concentrations and to move with the water with little to no retardation. While *in situ* bioremediation offers the best hope for addressing the extent of MTBE contamination nationwide, natural processes will likely have to be accelerated using innovative engineering approaches, such as injecting TEAs, in order to meet regulatory standards in an acceptable timeframe.

The University of New Hampshire's USEPA-funded Bedrock Bioremediation Center (BBC) will be opening an MTBE/gasoline research site in Fall 2001 in New Hampshire. This site is currently being chosen in coordination with the New Hampshire Department of Environmental Services. The BBC develops and tests technologies to monitor and accelerate *in situ* bioremediation in bedrock. Currently, the BBC has a research site at the Pease International Tradeport (Portsmouth, NH) in a bedrock aquifer contaminated with chlorinated solvents. BBC researchers are developing drilling and monitoring technologies there and conducting evaluations of innovative and emerging bioremediation methods under controlled conditions, so that the true fate and degradation rates of the contaminants are known. It is essential to have such independent verification of monitoring and bioremediation strategies for regulators to accept bioremediation alternatives as viable options for *in situ* remediation. The BBC will be using the same approach at its new MTBE/gasoline site, which will have both bedrock and overburden MTBE contamination. In the future, the BBC may expand its focus to develop and test monitoring and *in situ* bioremediation technologies in the overburden at the test site, if there is interest among the regulatory agencies in this activity.

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