

## Widespread Potential for Microbial MTBE Degradation in Surface-Water Sediments

PAUL M. BRADLEY,\*  
JAMES E. LANDMEYER, AND  
FRANCIS H. CHAPELLE

U.S. Geological Survey, 720 Gracern Road, Suite 129,  
Columbia, South Carolina 29210-7651

Microorganisms indigenous to stream and lake bed sediments, collected from 11 sites throughout the United States, demonstrated significant mineralization of the fuel oxygenate, methyl-*tert*-butyl ether (MTBE). Mineralization of [U-<sup>14</sup>C]MTBE to <sup>14</sup>CO<sub>2</sub> ranged from 15 to 66% over 50 days and did not differ significantly between sediments collected from MTBE contaminated sites and from sites with no history of MTBE exposure. This result suggests that even the microbial communities indigenous to newly contaminated surface water systems will exhibit some innate ability to attenuate MTBE under aerobic conditions. The magnitude of MTBE mineralization was related to the sediment grain size distribution. A pronounced, inverse correlation ( $p < 0.001$ ;  $r^2 = 0.73$ ) was observed between the final recovery of <sup>14</sup>CO<sub>2</sub> and the percentage content of silt and clay sized grains (grain diameter < 0.125 mm). The results of this study indicate that the microorganisms that inhabit the bed sediments of streams and lakes can degrade MTBE efficiently and that this capability is widespread in the environment. Thus aerobic bed sediment microbial processes may provide a significant environmental sink for MTBE in surface water systems throughout the United States and may contribute to the reported transience of MTBE in some surface waters.

### Introduction

Identification of indigenous mechanisms for in situ biodegradation of the fuel oxygenate methyl *tert*-butyl ether (MTBE) has become an environmental priority due to the ongoing, pervasive contamination of ground- and surface-water systems by MTBE (1-4). In light of the USEPA's recently announced intention to reduce or eliminate the use of MTBE in domestic fuels over the next 3 years, identification of MTBE attenuation mechanisms is crucial to understanding the long-term behavior and final fate of MTBE in the environment. MTBE has a high aqueous solubility, has a low taste and odor threshold (5), is tentatively classified by the USEPA as a possible human carcinogen (2, 5), and has a USEPA drinking water advisory of 20-40 µg/L. MTBE contamination in surface water systems is a particular concern, because these systems are notoriously vulnerable to numerous point- and nonpoint-sources of contamination and because approximately 60% of the drinking water consumed in the continental U.S. comes from surface water sources. MTBE contamination of surface water can result from atmospheric deposition (5-7), storm-

water runoff (5, 8-10), direct releases by industrial (5, 8, 10, 11) and recreational activities (5, 12-14), and discharge of MTBE contaminated groundwater (5, 15-17). The growing number of reports documenting MTBE contamination in streams and lakes (5, 8, 11-14) emphasizes the need to understand natural attenuation mechanisms for MTBE in these surface water systems.

A recent study documenting microbial MTBE mineralization in aerobic microcosms containing stream bed sediments indicated that indigenous microbial activity can provide a sink for MTBE contamination in surface water systems (17). Because that study was limited to two petroleum contaminated sites in South Carolina, however, the importance of aerobic microbial degradation as a mechanism for natural attenuation of MTBE in surface water systems throughout the U.S. remains unclear. Moreover, because MTBE can contaminate remote locations as the result of nonpoint source processes such as atmospheric deposition (5-7), the response of previously uncontaminated surface water systems to MTBE also requires clarification. The purpose of this study was to examine microbial MTBE mineralization in lake- and stream-bed sediments collected throughout the U.S. in order to evaluate the potential environmental relevance of MTBE biodegradation as a natural attenuation mechanism.

### Methods

**Chemicals.** The potential for microbial degradation of MTBE was investigated in lake- and stream-bed-sediment microcosms using uniformly labeled [U-<sup>14</sup>C]MTBE (10.1 mCi/mmol; 10% MTBE in ethanol) obtained from New England Nuclear Research Products, Du Pont (Boston, Massachusetts). The radiochemical purity of the [U-<sup>14</sup>C]MTBE was determined by the manufacturer and independently confirmed in our lab using radiometric detection gas chromatography (GC/GRD) to be greater than 97%.

**Study Sites.** Microcosm studies were conducted using bed sediments from 11 sites located throughout the U.S. (Figure 1). Sites were selected to provide a range in sediment geochemistry, contaminant exposure, and geographical location. Select characteristics of the sediment samples are given in Table 1. Grain size analyses were performed by dry sieving. For the purposes of this study, sand sized grains are defined as those between 0.125 mm and 2 mm in diameter. The combined silt and clay fraction is defined as grains of less than 0.125 mm in diameter. The sediment organic content was estimated by the loss on ignition method. The following is a brief description of the collection sites and sample sediments.

The Laurens (Laurens, South Carolina) and Oasis 2 (Charleston, South Carolina) sites were described in detail previously (17). At both sites, groundwater containing soluble components of oxygenated gasoline (including BTEX and MTBE) discharges to shallow streams located 15-30 m downgradient. Oasis 1 sediments, poorly sorted fine to medium sands, were collected from a nearby stream at a point approximately 20 m from Oasis 2. No MTBE was detected (MDL = 2 µg/L) in the sediments collected from Laurens or Oasis.

Bed sediment samples were collected from a stream at the Naval Air Station (NAS) Cecil Field, Jacksonville, Florida (18). The Cecil Field bed sediment sample was a poorly sorted fine sand. The site contains an anaerobic, chlorinated solvent plume that discharges to the stream. Previous studies indicated that the microbial community indigenous to the stream bed sediments at this site actively degrade chlorinated

\* Corresponding author phone: (803)750-6125; fax: (803)750-6181; e-mail: pbradley@usgs.gov.

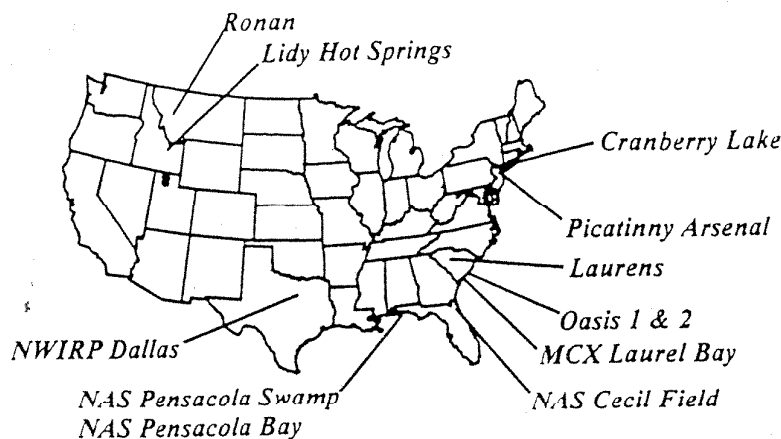


FIGURE 1. Location of sample collection sites.

TABLE 1. Characteristics of Bed Sediment Samples Collected from 11 Sites throughout the U.S.<sup>a</sup>

sediment	bulk density, <sup>a</sup> g/mL	water, <sup>b</sup> %	sand, <sup>c</sup> %	silt + clay, <sup>c</sup> %	organic, <sup>c</sup> %	biomass <sup>d</sup> × 10 <sup>3</sup>
Cecil Field	1.4	28 ± 1	83 ± 1	17 ± 1	0.1 ± 0.2	0.7 ± 0.1
Cranberry Lake	0.1	94 ± 1	19 ± 11	54 ± 11	27 ± 0	12 ± 3
Laurens	1.4	28 ± 1	98 ± 0	2 ± 0	0 ± 0	0.7 ± 0.0
Lidy Hot Spring	1.0	45 ± 0	20 ± 6	79 ± 8	1 ± 1	2.0 ± 0.2
NWIRP Dallas	0.3	73 ± 1	0 ± 0	95 ± 0	5 ± 0	0.6 ± 0.3
Oasis 1	1.5	27 ± 1	89 ± 0	11 ± 0	0 ± 0	5.4 ± 0.4
Oasis 2	0.2	66 ± 4	5 ± 2	78 ± 2	17 ± 3	1.4 ± 0.0
Pensacola Bay	1.7	24 ± 0	100 ± 0	0 ± 0	0 ± 0	0.7 ± 0.0
Pensacola Swamp	1.4	26 ± 1	100 ± 0	0 ± 0	0 ± 0	4.0 ± 0.3
Picatinny	1.0	47 ± 10	63 ± 2	36 ± 1	1 ± 1	6.6 ± 1.2
Ronan	0.3	79 ± 0	13 ± 1	75 ± 6	12 ± 6	3.5 ± 1.2

<sup>a</sup> Grams dry weight per mL wet sediment. The coefficient of variation was 4%. <sup>b</sup> Content expressed as a percentage of sediment wet weight. <sup>c</sup> Content expressed as a percentage of sediment dry weight. <sup>d</sup> Total number of microorganisms per microcosm estimated by AODC. \* Data are means ± SD of duplicate analyses.

ethene contaminants. No previous exposure to MTBE has been reported for this site.

Cranberry Lake is a suburban lake located in a densely populated area of Sussex County New Jersey (14). Use of recreational watercraft fueled by reformulated gasoline is prevalent at Cranberry Lake. The sample from Cranberry Lake was a loosely suspended, organic-rich sediment of silt and clay. Dissolved MTBE concentrations as high as 29 µg/L have been observed in the lake (14).

Lidy Hot Springs is located approximately 50 km west of Dubois, Idaho. Bed sediments were collected from a small wetland that borders a geothermal spring. The sediment sample was a carbonate mud consisting primarily of silt and clay sized grains. At the time of sample collection, the groundwater temperature at the mouth of the spring was 58 °C, while the water temperature at the point of sample collection was approximately 35 °C. This was an uncontaminated site with no apparent MTBE exposure.

Lake bed sediments were collected from a site adjacent to the Naval Weapons Industrial Reserve Plant (NWIRP) Dallas, Texas. The NWIRP Dallas bed sediments were collected from a shallow, freshwater lake into which chlorinated ethene contaminated groundwater continuously discharges. The collected sediment was an organic-rich pluff mud consisting of silt and clay. Although no MTBE was detected in the collected sediments, this lake is accessible by gasoline-powered water craft. Consequently, periodic exposure of the bed sediment microbial community to low concentrations of MTBE is likely.

Both Pensacola sediment samples were collected from NAS Pensacola, Florida. The Pensacola swamp sediment was a well sorted, medium sand collected from an area of active

groundwater discharge. This sample site was located along the groundwater flow path of a chlorinated solvent plume which contained primarily chlorobenzene compounds. No MTBE was detected in this sample, and no previous exposure to MTBE has been reported for this sample location. The Pensacola Bay sample was collected approximately 40 m down-gradient of the swamp sample location at a point 5 m from the shore of Pensacola Bay. The bay sediment was a well-sorted coarse sand, and the interstitial salinity of the collected sample was 23.5 ppt. No MTBE or chlorinated solvents were detected in the Pensacola Bay sample. However, due to boating activity in Pensacola Bay, periodic exposure to MTBE cannot be ruled out.

Bed sediments were collected from a small stream at Picatinny Arsenal, New Jersey. The sample was a poorly sorted sediment with grain sizes ranging from medium sand to clay. The sediment sample was collected from a location where anaerobic groundwater contaminated with chlorinated ethene compounds discharges to the stream. No MTBE was detected in the sediment samples.

At the Ronan (Montana) site an anaerobic plume from a reformulated gasoline spill extends 600 m down-gradient from a leaking underground storage tank to Spring Creek. The contaminant plume discharges into a poorly drained oxbow that connects to the main channel during high streamflow events. Maximum dissolved MTBE concentrations of 44 mg/L have been reported in the source area of the plume. The sediment sample was collected from the plume side of the oxbow where the discharging groundwater contains up to 30 µg/L MTBE. The collected sediment was an organic-rich, silt and clay.

**TABLE 2. Final Percentage Recovery of  $^{14}\text{C}$ -Activity as  $^{14}\text{CO}_2$  and  $[1,2-^{14}\text{C}]$ MTBE in Microcosms Containing Bed Sediment from 11 Sites and Aquifer Sediment from Laurel Bay, SC**

sediment	$^{14}\text{CO}_2$		$[1,2-^{14}\text{C}]$ MTBE	
	exptl %	control %	exptl %	control %
Cecil Field	41 ± 6	0 ± 0	55 ± 7	96 ± 5
Cranberry Lake	21 ± 3	0 ± 0	67 ± 9	95 ± 8
Laurel Bay <sup>a</sup>	5 ± 2	0 ± 0	87 ± 7	95 ± 7
Laurens	31 ± 8	0 ± 0	51 ± 19	97 ± 4
Lidy Hot Spring	15 ± 2	0 ± 0	72 ± 9	95 ± 6
NWIRP Dallas	15 ± 6	0 ± 0	80 ± 22	94 ± 8
Oasis 1	50 ± 8	0 ± 0	51 ± 6	96 ± 6
Oasis 2	16 ± 9	0 ± 0	NA <sup>b</sup>	NA <sup>b</sup>
Pensacola Bay	66 ± 21	0 ± 0	21 ± 20	87 ± 18
Pensacola Swamp	62 ± 16	0 ± 0	44 ± 15	88 ± 17
Picatinny	32 ± 12	0 ± 0	69 ± 22	98 ± 3
Ronan	28 ± 13	0 ± 0	65 ± 10	98 ± 3

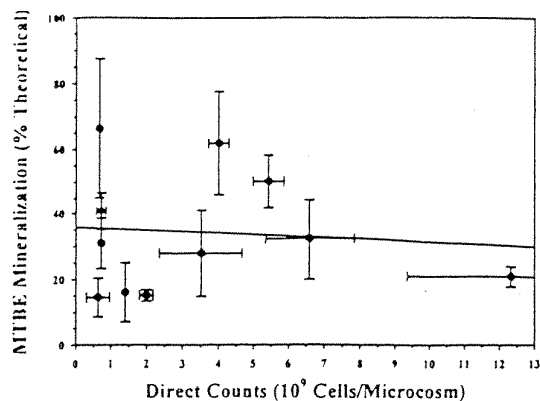
<sup>a</sup> Aquifer sediment from MCX Laurel Bay. <sup>b</sup> Not analyzed. Data are means ± SD for triplicate experimental and duplicate sterile control microcosms. The minimum detection limit for radiometric detection was equivalent to 1% final recovery.

In addition to the bed sediment samples described above, shallow aquifer material was collected from a gasoline spill site (MCX Laurel Bay) in Beaufort, South Carolina where contaminated groundwater containing 10,000  $\mu\text{g/L}$  dissolved MTBE is presently discharging to a nearby, concrete-lined stream (15). The hydrology, geochemistry, and contaminant distribution of the site have been described in detail previously (15).

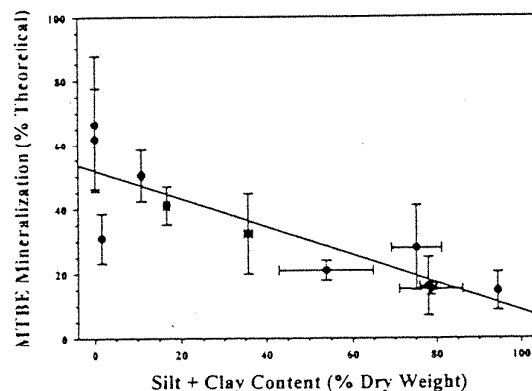
**Microcosm Studies.** Bed sediment microcosms were prepared as described previously (17, 18). In brief, 20 mL serum vials were amended with 10 mL of saturated bed sediment, sealed with Teflon-lined butyl rubber stoppers, and flushed with 1000 mL of zero air. Triplicate experimental treatments were prepared for each sediment. Duplicate killed control microcosms were prepared for each sediment and autoclaved twice for 1 h at 15 PSI and 121 C. Sediment microcosms were amended with approximately 0.3  $\mu\text{Ci}$  of  $[U-^{14}\text{C}]$ MTBE. Headspace concentrations of  $\text{CH}_4$ ,  $^{14}\text{CH}_4$ ,  $\text{CO}_2$ ,  $^{14}\text{CO}_2$ , MTBE, and  $[U-^{14}\text{C}]$ MTBE were monitored by analyzing 0.5 mL of headspace sample using GC/GRD combined with thermal conductivity detection. The headspace sample volumes were replaced with oxygen (99.9% Scotty II, Scott Specialty Gases, Plumsteadville, PA). Dissolved phase concentrations were estimated based on experimentally determined partition coefficients. The GC/GRD output was calibrated by liquid scintillation counting using  $\text{H}^{14}\text{CO}_3^-$  and  $[U-^{14}\text{C}]$ MTBE. The results of the  $[U-^{14}\text{C}]$ MTBE mineralization studies presented in Table 2 and Figures 2–4 were corrected for the loss of constituents due to headspace sample collection. The total number of microorganisms present in the sediment microcosms was estimated from acridine orange direct counts.

## Results and Discussion

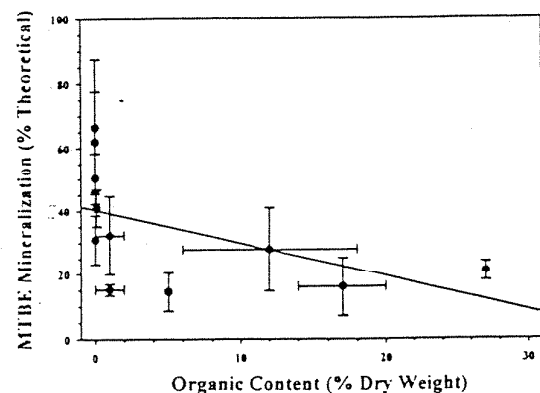
Every bed sediment examined in this study demonstrated significant aerobic mineralization of  $[U-^{14}\text{C}]$ MTBE to  $^{14}\text{CO}_2$  within 50 days (Table 2). Final mean recoveries as  $^{14}\text{CO}_2$  ranged from a minimum of 15% for sediments collected from the Lidy Hot Spring and NWIRP Dallas sites to a maximum of 66 ± 21% for the Pensacola Bay sediments. In all cases, the mineralization of  $[U-^{14}\text{C}]$ MTBE observed in bed sediment microcosms was greater than that observed in microcosms containing Laurel Bay aquifer material (Table 2). The loss of  $[U-^{14}\text{C}]$ MTBE observed in experimental microcosms was attributable to biological activity, because the loss of  $[U-^{14}\text{C}]$ MTBE in sterile controls was not statistically significant (Table 2). Similarly, mineralization of  $[U-^{14}\text{C}]$ MTBE to  $^{14}\text{CO}_2$  was



**FIGURE 2. Lack of correlation ( $p = 0.81$ ;  $r^2 = 0.007$ ) between the final recovery of  $[U-^{14}\text{C}]$ MTBE radiolabel as  $^{14}\text{CO}_2$  after 50 days and the number of microorganisms ( $\times 10^9$ ) present in bed sediment microcosms. Data are means ± SD. Solid line indicates the least squares linear regression of  $^{14}\text{CO}_2$  recovery and microbial numbers.**



**FIGURE 3. Strong correlation ( $p < 0.001$ ;  $r^2 = 0.73$ ) between the final recovery of  $[U-^{14}\text{C}]$ MTBE radiolabel as  $^{14}\text{CO}_2$  after 50 days and the percentage of bed sediment dry weight consisting of silt and clay sized grains (diameter < 0.0125 mm). Data are means ± SD. Solid line indicates the least squares linear regression of  $^{14}\text{CO}_2$  recovery and sediment silt/clay content.**



**FIGURE 4. Weak correlation ( $p = 0.10$ ;  $r^2 = 0.27$ ) between the final recovery of  $[U-^{14}\text{C}]$ MTBE radiolabel as  $^{14}\text{CO}_2$  after 50 days and bed sediment organic content expressed as a percentage of sediment dry weight. Data are means ± SD. Solid line indicates the least squares linear regression of  $^{14}\text{CO}_2$  recovery and sediment organic content.**

entirely attributable to biological activity, because no  $^{14}\text{CO}_2$  was recovered in autoclaved control microcosms (Table 2). In most cases, the degradation of  $[U-^{14}\text{C}]$ MTBE to  $^{14}\text{CO}_2$  was approximately stoichiometric. For the Cranberry Lake, Lau-

rens, and Lidy Hot Spring samples, however, the final recovery of radiolabel as  $^{14}\text{CO}_2$  and  $[\text{U-}^{14}\text{C}]\text{MTBE}$  was 82–88% in experimental microcosms, while the final recovery of  $[\text{U-}^{14}\text{C}]\text{MTBE}$  in sterile control microcosms was greater than 95%. For these three sediments, the radiolabel not accounted for by  $^{14}\text{CO}_2$  and  $[\text{U-}^{14}\text{C}]\text{MTBE}$  may have been incorporated into biomass or degraded to unidentified products. The results of this study demonstrate that bed sediment microorganisms are capable of extensive aerobic degradation of MTBE to nontoxic products and that this capability is widespread in the environment.

MTBE mineralization was not limited to sediments with a history of MTBE contamination. In this study, five sites (Cranberry Lake, Laurens, Oasis 1 and 2, Ronan) had significant MTBE contamination at the time of sample collection, and one site (NWIRP Dallas) was presumed to have a history of periodic exposure to low concentrations of MTBE. For these six sites, the final recovery of  $[\text{U-}^{14}\text{C}]\text{MTBE}$  radiolabel as  $^{14}\text{CO}_2$  ranged from 15 to 50%. Three other sites had no known history of MTBE contamination but were contaminated with chlorinated solvents. The Pensacola Bay sediment had no detectable contamination, but, considering its location in an urban harbor, periodic exposure to diverse contaminants is to be expected. For these four sites, the final  $^{14}\text{CO}_2$  recovery was similar to that observed for the MTBE sites and ranged from 32 to 66%. It is worth noting that the rapid mineralization of MTBE observed in the Pensacola Bay sediment microcosms suggests that microbial MTBE mineralization can be significant in estuarine and coastal environments as well as in freshwater systems. The Lidy Hot Spring sediment was collected in rural Idaho from a small wetland fed by a geothermal spring. The sediments were uncontaminated and prior exposure to MTBE was unlikely. Nevertheless, the sediment from Lidy Hot Spring demonstrated significant mineralization ( $15 \pm 2\%$ ) of  $[\text{U-}^{14}\text{C}]\text{MTBE}$ . Thus, not only is the potential for microbial MTBE mineralization widespread in the environment, the responsible microorganisms apparently do not require previous exposure in order to effectively degrade MTBE. This result suggests that even the microbial communities indigenous to newly contaminated surface water systems will exhibit some innate ability to attenuate MTBE under aerobic conditions.

In this study, the magnitude of MTBE mineralization was not related to the number of microorganisms present in the sediment samples (Figure 2). The total number of microorganisms per microcosm (i.e. per 10 mL sediment) was estimated by acridine orange direct counts and was found to vary by 2 orders of magnitude from  $0.6 \pm 0.3 \times 10^9$  cells in the NWIRP Dallas sediment to  $12 \pm 3 \times 10^9$  cells in the Cranberry Lake sample (Table 1). The final recovery of  $^{14}\text{CO}_2$  after 50 days, however, was not significantly correlated ( $p = 0.81$ ;  $r^2 = 0.007$ ) to microbial numbers (Figure 2).

The magnitude of MTBE mineralization was related to the grain size distribution of the sediment samples. A strong, inverse relationship ( $p < 0.001$ ;  $r^2 = 0.73$ ; Figure 3) was observed between the final recovery of  $^{14}\text{CO}_2$  and the percentage content of silt and clay sized grains (grain diameter  $< 0.125$  mm). Conversely, a strong, positive relationship ( $p < 0.001$ ;  $r^2 = 0.74$ ; data not shown) was observed between final  $^{14}\text{CO}_2$  recovery and the percentage content of sand sized grains (grain diameter between 0.125 and 2.0 mm). The probable explanation for the relationship between MTBE mineralization and grain size distribution is the reported dependence of microbial MTBE mineralization on aerobic conditions (15, 17, 19–24). A previous study, demonstrated that the mineralization of  $[\text{U-}^{14}\text{C}]\text{MTBE}$  observed in aerobic, stream-bed sediment microcosms was completely inhibited when the same sediment was incubated anaerobically (17). In the present study, all sediments were incubated statically under an air atmosphere. However, the diffusion of oxygen

into the sediments is related to the sediment permeability which, in turn, is related to the grain size distribution. Thus, in this study, the diffusion of oxygen beneath the sediment surface is expected to be lowest in the low permeability, silt and clay sediments (ex. NWIRP Dallas) and greatest in the high permeability, well sorted, coarse sands (ex. Pensacola Bay). Qualitative evidence for the relationship between sediment type and the diffusion of oxygen beneath the sediment surface was provided by visual assessment of the depth of the surface oxidized zone in select sediment microcosms. In the NWIRP Dallas microcosms the oxidized zone was less than 2 mm in depth at the end of the study, while the oxidized zone in Pensacola Bay microcosms was greater than 1.3 cm in depth. The relationship between MTBE biodegradation efficiency and sediment type observed in this study under static conditions may be more pronounced in situ, where advection can transport oxygen deep into permeable sediments but oxygen supply in impermeable sediments is generally limited by diffusion to the upper few millimeters (25).

A weak inverse relationship was observed between MTBE mineralization and sediment organic content ( $p = 0.10$ ;  $r^2 = 0.27$ ; Figure 4). Only 27% of the variation in the final recovery of  $^{14}\text{CO}_2$  was associated with organic content, whereas 73% of the variation was associated with grain size distribution (Figure 3). This observation is consistent with previous studies indicating that competitive inhibition of MTBE biodegradation can occur in the presence of alternative carbon substrates (17, 21). The observed response may be due to preferential utilization of more favorable carbon substrates by the microbial community that is responsible for MTBE mineralization. Alternatively, the inverse relationship between MTBE mineralization and sediment organic content may be attributable to competitive consumption of oxygen and a resultant depletion in the oxygen available to support MTBE biodegradation.

The widespread potential for MTBE mineralization observed in this study is particularly important in light of recent evidence suggesting that MTBE contamination in surface water systems can vary seasonally with little apparent carry over from year to year (13). In that study (13), seasonal changes in MTBE concentrations were monitored in Lake Donner, a multiple-use lake located in northeastern California, and the use of motorized watercraft was identified as the primary source of MTBE contamination. Maximum MTBE concentrations of 12.1  $\mu\text{g/L}$  were associated with high recreational watercraft use during the summer months. During the off-season MTBE concentrations declined to the analytical detection limit of 0.1  $\mu\text{g/L}$ . Because MTBE losses to lake outflow could not account for the observed decline in MTBE and because MTBE biodegradation had not been demonstrated in surface water systems at that time, the sink for MTBE was assumed to be volatilization. The results of the current study, however, suggest that MTBE biodegradation in the bed sediments may contribute extensively to the loss of MTBE in Lake Donner (13). The results of these studies (13, 17, this study) suggest that efficient, natural sinks for MTBE exist in surface water environments and that the apparent persistence of MTBE in these systems is due to ongoing MTBE contamination rather than environmental recalcitrance. Moreover, because MTBE contamination in the environment is attributed to the use of MTBE as an oxygenate in domestic fuels, these findings (13, 17, this study) indicate that the EPA's proposed ban on MTBE use in domestic fuel supplies is an important step toward remediation of MTBE contaminated surface waters.

#### Acknowledgments

This research was supported by the South Carolina Department of Health and Environmental Control in cooperation

with the U.S. Geological Survey Toxic Substances Hydrology Program.

### Literature Cited

- (1) Squillace, P. J.; Pope, D. A.; Price, C. V. *U.S. Geological Survey Fact Sheet*, FS-114-95; 1995; 4 pp.
- (2) Squillace, P. J.; Zogorski, J. S.; Wilber, W. G.; Price, C. V. *Environ. Sci. Technol.* 1996, 30, 1721-1730.
- (3) Squillace, P. J.; Moran, M. J.; Lapham, W. W.; Price, C. V.; Clawges, R. M.; Zogorski, J. S. *Environ. Sci. Technol.* 1999, 33, 4176-4187.
- (4) Johnson, R.; Pankow, J.; Bender, D.; Price, C. V.; Zogorski, J. S. *Environ. Sci. Technol.* 2000, 34, 210A-217A.
- (5) Zogorski, J. S.; Delzer, G. C.; Bender, D. A.; Squillace, P. J.; Lopes, T. J.; Baehr, A. L.; Stackelberg, P. E.; Landmeyer, J. E.; Boughton, C. J.; Lico, M. S.; Pankow, J. F.; Johnson, R. L.; Thomson, N. R. *Proc. 1998 Ann. Conf. Am. Water Works Assoc.* In press.
- (6) Pankow, J. F.; Thomson, N. R.; Johnson, R. L.; Baehr, A. L.; Zogorski, J. S. *Environ. Sci. Technol.* 1997, 31, 2821-2828.
- (7) Baehr, A. L.; Stackelberg, P. E.; Baker, R. J. *Water Resources Res.* 1999, 35, 127-136.
- (8) O'Brien, A. K.; Reiser, R. G.; Gylling, H. *U.S. Geological Survey Fact Sheet*, FS-194-97; 1997; 6 pp.
- (9) Lopes, T. J.; Dionne, S. G. *U.S. Geological Survey Open File Report*, OFR-98-409; 1998; 67 pp.
- (10) Delzer, G. C.; Zogorski, J. S.; Lopes, T. J.; Bosshart, R. L. *U.S. Geological Survey Water Resources Investigation Report*, WRIR-96-4145; 1996; 6 pp.
- (11) Terracciano, S. A.; O'Brien, A. K. *U.S. Geological Survey Fact Sheet*, FS-063-97; 1997; 4 pp.
- (12) Boughton, C. J.; Lico, M. S. *U.S. Geological Survey Fact Sheet*, FS-055-98; 1998; 2 pp.
- (13) Reuter, J. E.; Allen, B. C.; Richards, R. C.; Pankow, J. F.; Goldman, C. R.; Scholl, R. L.; Seyfried, J. S. *Environ. Sci. Technol.* 1998, 32, 3666-3672.
- (14) Baehr, A. L.; Zapecza, O. S. *U.S. Geological Survey Water Resources Investigation Report*, WRIR-98-4264; 1998; 6 pp.
- (15) Landmeyer, J. E.; Chapelle, F. H.; Bradley, P. M.; Pankow, J. F.; Church, C. D.; Tratnyek, P. G. *Ground Water Monitor. Remed.* 1998, Fall issue, 93-102.
- (16) Happel, A. M.; Beckenbach, E. H.; Halden, R. U. *An evaluation of MTBE impacts to California groundwater resources*, UCRL-AR-130897; University of California: 1998; 68 pp.
- (17) Bradley, P. M.; Landmeyer, J. E.; Chapelle, F. H. *Environ. Sci. Technol.* 1999, 33, 1877-1879.
- (18) Bradley, P. M.; Chapelle, F. H.; Lovley, D. R. *Appl. Environ. Microbiol.* 1998, 64, 3102-3105.
- (19) Salanitro, J. P.; Diaz, L. A.; Williams, M. P.; Wisniewski, H. L. *Appl. Environ. Microbiol.* 1994, 60, 2593-2596.
- (20) Stefan, R. J.; McClay, K.; Vainberg, S.; Condee, C. W.; Zhang, D. *Appl. Environ. Microbiol.* 1997, 63, 4216-4222.
- (21) Mo, K.; Lora, C. O.; Wanken, A. E.; Javanmardian, M.; Yang, X.; Kulpa, C. F. *Appl. Microbiol. Biotechnol.* 1997, 47, 69-72.
- (22) Hanson, J. R.; Ackerman, C. E.; Scow, K. M. *Appl. Environ. Microbiol.* 1999, 4788-4792.
- (23) Mormile, M. R.; Liu, S.; Sufliata, J. M. *Environ. Sci. Technol.* 1994, 28, 1727-1732.
- (24) Sufliata, J. M.; Mormile, M. R. *Environ. Sci. Technol.* 1993, 27, 976-978.
- (25) Woessner, W. W. *Ground Water* 2000, 38, 423-429.

Received for review August 2, 2000. Revised manuscript received November 27, 2000. Accepted November 28, 2000.

ES0015489