METHYL *TERT*-BUTYL ETHER IN SURFACE DRINKING WATER SUPPLIES

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1.0 INTRODUCTION AND BACKGROUND

This contribution to the overall UC report is intended to provide background information and the supporting data base for our discussion of the (1) current levels of MTBE in surface drinking water supplies (i.e. lakes, reservoirs, channelized flow) and (2) fate and transport of MTBE in these waterbodies as it appears in the Summary Report. In addition, a separate evaluation of MTBE at Lake Tahoe is included as required by SB 521.

We begin by presenting a summary of the state-wide data used in our analysis of current levels of MTBE in California's surface drinking water supplies (section 5 of Summary Report). This is followed by a copy of a peer-reviewed journal article scheduled to appear in the December issue of Environmental Science and Technology. This article discusses concentrations, sources and fate of the gasoline oxygenate MTBE in Donner Lake, CA, a multiple use lake located in the Sierra Nevada. On the basis of our 1998 investigations at Lake Tahoe and on the basis of the state-wide data base, a number of the observations and conclusions reached as part of the Donner Lake study have application to many other waterbodies. Finally, we present a summary of the major findings at Lake Tahoe. In that discussion we present MTBE concentration data for Lake Tahoe from 1997 and 1998, assess groundwater contamination by MTBE, and report on the limited data available from testing by water purveyors which use the lake as a drinking water source.

Simultaneous to this SB 521 study is an investigation designed to assess the impacts of motorized watercraft at Lake Tahoe. This is a multi-investigator effort done by the Lake Tahoe Motorized Watercraft Committee under the auspices of the Tahoe Regional Planning Agency. Direct contributors to that report, which has an expected release date of December 1998, include, but are not limited to, Tahoe Regional Planning Agency, U.S. Geological Survey, University of California - Davis, University of Nevada - Reno, California Air Resources Board, Lahontan Regional Water Quality Control Board - California, Nevada Division of Wildlife, and Miami University of Ohio. Aspects of that effort which are relevant to MTBE at Lake Tahoe are summarized herein and in the Summary Report.

It is important to note, and as stated in the Summary Report, the state-wide data on MTBE in surface drinking water supplies includes mostly 1996 and 1997. It does not include waterbodies which are not used as drinking water sources, however, we assume that these waterbodies are unlikely to be sampled for MTBE. To our knowledge, this is perhaps the most complete compilation of this data to date. While the specific statistics which we derived from this data base will change with time as monitoring continues, we believe that the conclusions are sound. This data is currently not available on the CAL-DHS web site but was developed through an extensive effort to directly contact the managers of each waterbody.

2.0 STATE-WIDE DATABASE FOR MTBE IN SURFACE DRINKING WATER SUPPLIES

Ms. Andrea Buxton significantly contributed to this section and the following tables and figures.

- Table 1. Summary of statewide MTBE database for surface drinking waterbodies by CAL-DHS region.
- Figure 1. Statewide database for MTBE. Frequency of waterbodies with sample numbers within specified ranges.
- Figure 2. Statewide database for MTBE. Summary of concentration data.
- Waterbodies sampled for MTBE by district.

3.0 CONCENTRATIONS, SOURCES AND FATE OF THE GASOLINE OXYGENATE MTBE IN A MULTIPLE USE LAKE†

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Discovery of the fuel additive MTBE (methyl *tert*-butyl ether) in drinking water supplies is of concern to public health officials, water suppliers and the public. Despite recent policy decisions, few published studies exist on concentrations, sources and fate of MTBE in surface waters. The purpose of this study was to determine the (1) relative contribution of motorized watercraft as a source of MTBE, (2) its seasonal distribution, (3) loss from the water column, (4) extent of vertical transport, and (5) persistence between years; this work was done in Donner Lake, California, a multiple-use lake in the Sierra Nevada Mountains. MTBE measurements were made at 9 individual depths from surface to bottom on 16 dates. Recreational boating was the most important source of MTBE. Statistically, 86% of the change in MTBE was explained by variation in motorized watercraft use. Neither highway runoff nor precipitation contributed significantly. MTBE concentration ranged from $<0.1\ \mu g \cdot L^{-1}$ to a high of $12\ \mu g \cdot L^{-1}$. Between July 1-7, 1997, MTBE content rose dramatically from 115 kg to 365 kg. By January, levels declined to a minimum of 15 kg suggesting little inter-annual persistence. The major loss of MTBE appeared to be volatilization at the air-water interface characterized by two distinct periods. During the boating season, MTBE decline was $1.2\ kg \cdot d^{-1}$ (193 day half-life). At the end of the boating season, MTBE loss increased to $8.1\ kg \cdot d^{-1}$ (14 day half-life). Thermal stratification acted to retard MTBE transport to deeper depths.

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3.1 INTRODUCTION

Addition of fuel oxygenates to gasoline to enhance octane level, increase burning efficiency, and reduce the emission of atmospheric pollutants has become more common in recent years. The most frequently used fuel oxygenate is methyl *tert*-butyl ether (MTBE) *(1)*. Other ether-based

additives that are far less frequently used include ethyl *tertiary*-butyl ether (ETBE) and *tertiary*-amyl methyl ether (TAME). As of 1992, urban areas classified as serious or moderate nonattainment for carbon monoxide (CO) under the 1990 Clean Air Act Amendments were required to sell oxygenated fuel containing a minimum of 2.7% oxygen by weight. For MTBE this would correspond to 14.8 percent by volume *(2)*. Since October 1992 the State of California required fuel to be oxygenated during the wintertime in areas of CO nonattainment. In 1995 California's South Coast Region was using reformulated gasoline year round, and by June 1996 gasoline containing MTBE as the primary fuel oxygenate was at service stations state-wide.

Discovery of MTBE in groundwater, lakes and reservoirs used for drinking water has raised considerable concern among health officials and water suppliers. The U.S. Environmental Protection Agency has classified MTBE as a possible human carcinogen and has set a drinking water advisory at 20-40 $\mu g \cdot L^{-1}$ based on consumer acceptance. A California state drinking water interim action level of 35 $\mu g \cdot L^{-1}$ has been established. However, State legislation in California has required that primary and secondary drinking water standards be in place by 1999 and 1998, respectively. Currently under consideration are values of 5 $\mu g \cdot L^{-1}$ based on taste and odor concerns and 14 $\mu g \cdot L^{-1}$ as a public health goal. These concentrations are recommendations and not enforceable at this time. Because of MTBE's possible health affects and the fact that it is highly soluble in water and difficult to biodegrade, its potential persistence in water supplies has recently raised numerous research and management questions. In a nation-wide survey of the occurrence and possible sources of MTBE and other VOCs in groundwater, the U.S. Geological Survey's National Water-Quality Assessment program reported MTBE as the second most frequently detected chemical in shallow groundwater from urban areas *(2)*. Of the 210 shallow urban wells and springs sampled, 27% were found to contain MTBE at a concentration at or above a reporting limit of 0.2 $\mu g \cdot L^{-1}$. Three percent of the wells exceeded 20 $\mu g \cdot L^{-1}$ MTBE. In this study it was noted that since MTBE is mobile in groundwater, it may move from shallow to deeper aquifers, suggesting the possibility of its transport into subsurface drinking water supplies over time. Sources of MTBE in shallow groundwater include direct contamination from leaking storage tanks, and indirect contamination from stormwater flow and urban precipitation *(3, 4, 5)*.

Despite the fact that a body of literature on MTBE is now growing, very few published studies exist on sources, fate and transport in lakes and reservoirs. This is highlighted in a recent review which discusses the environmental behavior and fate of MTBE *(6)*. With some notable exceptions, most of the investigations on MTBE in lakes to date focus on describing point-in-time, regulatory monitoring designed to determine whether or not MTBE is present. The results of these surveys are primarily accessible from agency monitoring files. Concentrations of MTBE measured in the open-water portions of a variety of reservoirs in

California to date typically range from <1-20 μg·L^{-1} *(7)*. The Metropolitan Water District of Southern California has conducted an extensive MTBE monitoring program in six surface water reservoirs of varying recreational activity. The occurrence of MTBE correlated with the general pattern of recreational use by motorized watercraft *(8)*. These investigators determined seasonal trends at Lake Perris and found that epilimnetic concentrations of MTBE reached as high as 25 μg·L^{-1}, the thermocline acted to retard transport of MTBE to deeper waters and, MTBE was diluted to nondetect-4 μg·L^{-1} in the late fall as lake mixing commenced.

At the same time, concentrations of MTBE in and around marinas, or in other areas expressly used for boating, levels can be much higher. In Shasta Lake, a large hydropower and recreational-use reservoir in northern California, concentrations ranged from 9-88 μg·L^{-1} over the Labor Day 1996 weekend. Maximum values were associated directly with large boats entering a docking area or with engine exhaust from these same vessels *(9)*. MTBE was also measured in a temporary lake constructed in southern California for a jetski event in the summer of 1996. After the 3-day event, concentrations ranged from 50-60 μg·L^{-1} and were well-mixed in this shallow water body *(10)*. Concentrations of MTBE at Lake Tahoe in the vicinity of boating activity were often within the range of 20-25 μg·L^{-1}, with a single maximum measured value of 47 μg·L^{-1} *(11)*. Between July-September 1997, the U.S. Geological Survey conducted a preliminary survey of MTBE and other VOC's in Lake Tahoe. Concentrations ranged from 0.18-4.2 μg·L^{-1} with larger concentrations nearer sites with substantial boating activity *(12)*. Despite the apparent link between the use of motorized watercraft and levels of MTBE, this source has not been widely acknowledged in the literature reviews on MTBE to date.

The work of Pankow et al. *(13)* provides a theoretical basis for predicting volatilization of MTBE from flowing surface waters. Rates were predicted as a function of mean flow velocity, mean flow depth, ambient temperature and wind speed. Half-life values for MTBE were calculated to range from hours in shallow (0.3 m), high velocity (3 m·s^{-1}) waterbodies to tens of days in deep (10 m), slow moving (0.1 m·s^{-1}) waters. These calculations apply to streams and river which are assumed to be characterized by relatively defined flow velocities which roughly correspond to much of the channel. However, the direct application of these calculations in predicting MTBE volatilization in lakes and reservoirs is complicated by the fact that there is less likely to be a well-defined cross section of flow which applies to the entire waterbody *(13)*. Models to simulate the transport and fate of MTBE in lakes and reservoirs are in various stages of development *(15, 16, 17)*. However, comprehensive field studies which provide sufficient data to identify important physical-chemical processes and for use in calibrating and verifying these models is essential.

From March 1997 to January 1998 we extensively studied the concentrations, sources, transport, and fate of MTBE in Donner Lake which is located at the summit of Interstate 80 as it passes through the Sierra Nevada in California (Figure D-1). Donner Lake lies at an elevation of 1,809 m above sea level with a surface area of 3.9 km^2. It's volume is approximately 1.26 x 10^8 m^3 (102,000 acre-feet) with a maximum depth of 70 m and an average depth of 33 m. Among its designated beneficial uses Donner Lake is a source of drinking water for lakeside residents and contributes to the water supply for downstream communities including Reno, Nevada.

This study was designed to address a number of important issues including: (a) the relative contribution of motorized watercraft as a source of MTBE, (b) the seasonal distribution of MTBE, (c) the extent of MTBE transport from surface waters into deeper portions of the lake, (d) the loss rate of MTBE from the water column, and (e) carry-over of MTBE between years.

3.2 METHODS

3.2.1 STUDY SITE AND FIELD SAMPLING

Water for MTBE measurements was collected on 16 dates during the period March 26, 1997 to January 15, 1998. On each date, samples were typically taken at three stations and from multiple depths along the long axis of Donner Lake, California (Figure D-1). Station 2 was located approximately 0.1 km off the mouth of Billy Mack Creek which is directly feed by Summit Creek and the major tributary inflow; the bottom depth at Station 2 was 39 m. Station 4 was over the deepest portion of the lake with a depth of 66 m. Station 5, at a depth of 33 m, was positioned 1.4 km from the eastern outlet of Donner Lake. While the exact location of the individual sampling depths varied somewhat over the 10 month sampling period, the sampling design typically included 0, 3, 6, 9, 12, 15, 20, 25 and 30 meters at Station 2 and Station 5, and 0, 3, 6, 9, 12, 22, 32.5, 45 and 57.5 meters at Station 4. In all cases, these depths extended from the surface to the bottom with extensive coverage of the upper, epilimnetic waters. For whole-lake calculations it was taken that Stations 2, 4 and 5 accounted for 17, 47 and 36 percent of the lake surface, respectively. The contribution of each depth layer to the whole-lake content was calculated as the product of mean lake concentration and the volume of each layer as determined from the morphometric contours (Figure D-1).

In situ temperature profiles were obtained at each station with a YSI temperature meter which was calibrated on each date with a hand-held thermometer.

Sampling for MTBE in Donner Lake was conducted from a 14 foot aluminum fishing boat powered by a 12 horsepower outboard motor. To ensure no contamination from the vessel, an auxiliary electric trolling motor was used to travel the final distance to each collection site. All sites were approached from down wind, and the boat anchored on location.

Water samples were collected from discrete depths using a Kemmerer well sampler. The two inch diameter stainless tube with polyurethane end seals was lowered to depth in the open position, and tripped closed with a messenger. The 0.6 L volume was hauled to the surface and used to completely fill pre-acidified 40 mL amber glass vials with septa sealed tops. These vials are used for volatile organic analysis (VOA) sampling and were acidified prior to use with two drops of 1:1 hydrochloric acid. Lake water was poured directly into these vials using the spring loaded purge at the bottom of the Kemmerer sampler. They were completely filled to ensure that no head space existed. Collected samples were kept in a cooler on ice and directly transported to the laboratory for analysis. Analysis of field blanks (laboratory deionzed water passed through the Kemmerer sampler and other phases of the field collection process) were always below the 0.1 $\mu g \cdot L^{-1}$ estimated limit of detection (LOD).

3.2.2 LABORATORY ANALYSIS

MTBE was analyzed at Alpha Analytical, Inc. (Sparks, NV) in 25 mL aliquots first using purge and trap procedures for concentration followed by gas chromatography/mass spectrometer detection (MSD) (EPA Method 524.2). Laboratory instrumentation consisted of a Tekmar auto sampler; Tekmar LSC 2000 Liquid Sample Concentrator with a VOCARB 3000 trap; a Hewlett Packard 5890 gas chromatograph with a 75 meter, 0.45 mm (I.D.) J&W DB-VRX column and glass-jet separator interface to a Hewlett Packard MSD. The standard LOD for this method is 0.5 $\mu g \cdot L^{-1}$; however in order to achieve the estimated LOD of 0.1 $\mu g \cdot L^{-1}$ used for this study, several modifications were necessary. Donner Lake samples were analyzed as independent batches, i.e. without other samples which could contain high levels of MTBE and possible result in even the slightest of sample-to-sample carry over. The MSD was tuned for maximum sensitivity and a 5-point calibration curve from 0.25 $\mu g \cdot L^{-1}$ to 5.0 $\mu g \cdot L^{-1}$ was generated. This curve was consistently linear through the origin, an in essence a 6-point calibration curve. The ability to quantify MTBE at concentrations between 0.1 and 0.5 $\mu g \cdot L^{-1}$ was verified for each batch run by analyzing MTBE Laboratory Control Spikes at 0.1 and/or 0.5 $\mu g \cdot L^{-1}$. QA/QC protocol required that for the 0.1 $\mu g \cdot L^{-1}$ spike, if the results were not in the range of 0.07-0.13 $\mu g \cdot L^{-1}$, those sample out of compliance were re-analyzed. To verify that reported sample concentrations above 0.1 $\mu g \cdot L^{-1}$ were not due to carry over or contamination of the analytical system, the normal 524.2 Method Blank was analyzed at the beginning of each batch with the inclusion of identical method blanks at several points during the analytical sequence. All the method blank analyses during the entire time that low-level MTBE analyses were made for this study shown no indication of MTBE in the laboratory or analytical system, or between samples.

3.3 RESULTS AND DISCUSSION

3.3.1 SEASONAL SURFACE HYDROLOGY

Sampling began prior to summer and during the period of annual spring snowmelt. Throughout March and early April, cumulative inflow to Donner Lake via its major tributary, Billy Mack and Summit Creeks, was moderate at 50-100 $m^3 \cdot min^{-1}$ despite a secondary peak in flow the last week in March (Figure D-2). During that period, lake level and volume were relatively uniform at approximately 68 m and 1.19 x 10^8 m^3, respectively. From April 16-28 a major snowmelt event occurred with creek discharge reaching as high as 305 $m^3 \cdot min^{-1}$. In the two month period from mid-April to mid-June the cumulative snowmelt discharge resulted in 2 m increase in lake level and a 6 percent increase in lake volume. Both inflow and lake outflow were negligible by late June. Lake level declined steadily during the summer months of June-September at a rate of approximately 0.5 $cm \cdot d^{-1}$ as a result of evaporation in concert with no significant precipitation. Summers are extremely dry in the Sierra Nevada mountains with the exception of occasional and brief thunderstorms which do not contribute significantly to the water budget of the medium to large subalpine lakes. Consequently, evaporative processes dominated the summer hydrology of Donner Lake until discharge began on September 12 for management purposes. This outflow is a regular feature of the lake's hydrology and is part of a release schedule which provides increased water supply for downstream use at the end of the dry summer and allows additional holding capacity in Donner Lake to accept fall rainstorms should they occur. Discharge on September 13 was 118

$m^3 \cdot min^{-1}$ and between September 3-30 the lake lost 2-3 percent and 6-7 percent of the whole-lake and epilimnetic volumes, respectively. Winter flow to the lake began on January 12, 1998. This was not part of the annual snowmelt period, but rather was a short-lived precipitation event.

3.3.2 SOURCES OF MTBE

On each of the sixteen collection dates, MTBE was evenly distributed between the three sampling stations and presumably throughout the surface area of Donner Lake. The lake is characterized by an oval-shaped shoreline with no sheltered embayments or enclosed marinas. The long, east-west axis aligns with the prevailing wind direction contributing to the spatially well-mixed conditions. On those dates when MTBE exceeded a concentration of 1 $\mu g \cdot L^{-1}$, the coefficient of variation (standard deviation÷mean) for measurements taken at the three sampling stations was low and typically less than 10 percent. MTBE concentrations ranged from a minimum of <0.1 $\mu g \cdot L^{-1}$ to a maximum of 12.1 $\mu g \cdot L^{-1}$ for the entire study. Since the objective of this study was to examine MTBE on the spatial scale of the entire waterbody, no attempt was made to collect samples in the immediate vicinity of boat launching facilities, in the wake of passing watercraft engine exhaust, or at other locations which were not representative of whole-lake conditions.

Examination of the depth distribution of MTBE was facilitated by having measurements with a 0.1 $\mu g \cdot L^{-1}$ limit of detection. In March and April all measured concentrations ranged from 0.1-0.3 $\mu g \cdot L^{-1}$. Mean concentrations on these three sampling dates from surface to bottom were 0.3, 0.2 and 0.2 $\mu g \cdot L^{-1}$. The water column was isothermal on March 26 and April 9; on April 24 the difference between surface and bottom water temperature was only 1°C. This temperature structure allowed for wind-induced mixing to occur throughout the water column with no apparent vertical differences in MTBE distribution (Figure D-3).

These low concentrations during the spring months show that precipitation or highway runoff during this period did not significantly contribute to the MTBE content in Donner Lake. The March and April sampling dates were in the period when annual snowmelt was underway (Figure D-2). Maximum inflow occurred the week prior to the April 24 sampling. The partitioning of MTBE to precipitation in urban settings with MTBE use can result in concentrations as high as 2-4 $\mu g \cdot L^{-1}$ *(6)*. Since the ratio of annual inflow to total lake volume is on the order of 1:10, even if the entire snowmelt volume from the rural and subalpine drainage basin of Donner Lake contained these types of urban concentrations, the lake concentration would only increase by 0.2-0.4 $\mu g \cdot L^{-1}$. Preliminary data on MTBE content in snow was obtained on January 21, 1998 at three locations along Highway 80 at Donner Lake. Analysis of fresh snow fall indicated that in all samples MTBE was less than 0.1 $\mu g \cdot L^{-1}$ *(18)*. This is supported by data from fresh snow samples taken in the metropolitan Denver region which showed MTBE concentrations to be very low at 0.011-0.088 $\mu g \cdot L^{-1}$ *(19)*. Based on these considerations, precipitation as a source of MTBE to Donner Lake is of minor importance. Similarly, we suspect that because of the dry summer conditions in most of California, precipitation as a direct source of MTBE to reservoirs and lakes used as drinking water supplies should be of little consequence relative to advisories of 20-40 $\mu g \cdot L^{-1}$.

Interstate 80 is the major commercial trucking, bus and passenger car route through the Sierra Nevada mountains. It is a 4-6 lane highway which runs for 5 km along the northern edge of Donner Lake (see Figure D-1). At a minimum the roadway is 0.1 km from the lakeshore and at most it is 0.7 km away. Since Donner Lake is directly downslope of the highway, all highway runoff ultimately discharges to the lake. Nation-wide survey data on fuel oxygenates and BTEX compounds in urban runoff collected by the U.S. Geological Survey during the period 1991-1995 found MTBE in 41 of 592 stormwater samples *(3)*. While it was the seventh most frequently detected volatile organic compound found in these samples, the range of detected limits was 0.2-8.7 $\mu g \cdot L^{-1}$ with a median of only 1.5 $\mu g \cdot L^{-1}$. Given that the hydrologic contribution of highway runoff to Donner Lake is negligible relative to snowmelt flows, and that even the most elevated MTBE concentration in this runoff would not be expected to exceed 8-10 $\mu g \cdot L^{-1}$, the contribution of highway runoff to the MTBE content of this lake would be minimal.

It is noteworthy that despite the low March-April MTBE levels in 1997, they were nonetheless above detection and higher than the lowest measured concentrations during this study. For example, during the summer months it was not uncommon to observe MTBE concentrations below a depth of 30-40 m to be 0.1-0.2 $\mu g \cdot L^{-1}$, and in January 1998 MTBE throughout the water column was just above the 0.1 $\mu g \cdot L^{-1}$ limit of detection. This slight elevation in March-April might have been the result of a leak in a fuel pipeline which was first observed on March 1. Released fuel containing MTBE flowed under the snow cover and into the headwaters of Summit Creek. Sampling in Billy Mack / Summit Creek by state agencies, in cooperation with other responsible parties, revealed that just prior to entering Donner Lake, concentrations of MTBE in the inflow were only 1.0-1.5 $\mu g \cdot L^{-1}$ on March 26 when lake sampling began. During the period March 11-25, MTBE in Summit Creek declined uniformly from a concentration of 20-25 $\mu g \cdot L^{-1}$ to the lower level measured on March 26. While the affects of this spill may have caused the 0.1-0.2 $\mu g \cdot L^{-1}$ elevation in MTBE concentrations in March and April, the relative impact was negligible.

Concentrations of MTBE began to increase slightly in the surface waters by early-mid May when levels rose to 0.5-0.7 $\mu g \cdot L^{-1}$ in the upper 15 m. Even though there was not a well developed thermocline at that time, thermal stratification was becoming established as evidenced by a 5°C decrease in water temperature between the surface and a depth of 30 m. This was sufficient to prevent complete mixing and allow MTBE to accumulate in the surface water (Figure D-3). Coincident with the subsequent onset of warmer air and water temperatures, and the boating season, MTBE in the upper 10 m of the water column increased even further to 0.9-2.1 $\mu g \cdot L^{-1}$. This increased occurred between the end of May and the beginning of July. By July, a strong thermocline had developed at a depth of approximately 9-11 m, which hindered the transport of MTBE to the deeper, hypolimnetic waters. Some entrainment of MTBE into the most upper portion of the hypolimnion was observed when epilimnetic concentrations were the highest (Figure D-3).

Sampling on July 1 and July 7 allowed us to address the hypothesis that releases from motorized watercraft were a primary source of MTBE in Donner Lake. Maximum epilimnetic concentrations rose dramatically over this 7-day period as evidenced by the 6-fold increase in MTBE from 2.1 $\mu g \cdot L^{-1}$ on July 1 to 12.1 $\mu g \cdot L^{-1}$ on July 7. Since precipitation, snowmelt and highway runoff were virtually nonexistent at that time, and since stream inflow from Summit Creek was nearing its seasonal minimum, motorized watercraft associated with the July 4th holiday were the most likely source. During March and April, before the traditional seasonal boating activity began, Donner Lake contained between 20-30 kg of MTBE. The whole-lake

MTBE level provides a volume weighted value and is therefore more representative than depth-specific concentrations. By July 1 this had increased to 115 kg as concentrations rose. The sharp increase in concentration observed between July 1 and July 7 resulted in an additional increase of 250 kg MTBE to the lake.

The relationship between calculated values for whole-lake MTBE and boat use data obtained from day-use permit sales records taken at the sole public boat ramp on the lake shows the direct and strong relationship between MTBE and watercraft use throughout the study period (Figure D-4). Of particular interest is the observation that not only did the whole-lake MTBE level respond to large source events, i.e. the significant July 4th event, but also to less dramatic variations in boating patterns during the summer. A strong statistical relationship was found between these two variables based on regression analysis. The relationship was linear and with a high r-statistic of 0.93 indicating that 86 percent of the observed variation in whole-lake MTBE level was explained by boat use as measured by censusing at the boat launching facilities. This analysis does not distinguish between the relative contribution of the various motorized watercraft which operate on Donner Lake; however, outboard motors on fishing boats, personnel watercraft, and water-skiing are the most common motorized boat use. For reference, a total of 193 boats utilized the ramp facility during the week ending on July 6. There is a resident population of boats at Donner Lake which are active on the lake but which are not included in the boat ramp data. Visual observations and discussions with lakeshore residents indicate that the seasonal pattern of resident and non-resident boat use is similar. Consequently, while the relationship seen between boat use and MTBE level is accurate to describe the relative association, loading per boat based on lake MTBE content can not be obtained solely on this basis of the available boat ramp data.

While the data in Figure D-4 strongly suggest that the changes in MTBE level in Donner Lake were primarily related to boat use and most probably engine exhaust, there are a number of activities associated with boating which could also contribute MTBE to surface waters, and which must be considered. First, elevated MTBE in lakes and reservoirs has been found in the vicinity of protected marinas and gas-fueling stations. Since neither of these services are provided at Donner Lake, these were not sources of MTBE to the open lake. Second, gasoline can be spilled into the surface water upon engine startup, while switching between gas tanks, or during removal of bilge water. Over the period 1-7 July we know that 250 kg MTBE was loaded to the lake and 193 boats used the launching facility. Assuming that (a) the number of resident boats on the lake were equal to non-resident boats (i.e. a total of 386 boats, a high-side assumption), (b) MTBE was 11% by volume in the gasoline, and (c) the density of gasoline is approximately 0.75 $g \cdot cm^3$, we estimate that each boat on the lake would have had to spill approximately 2 gallons of raw gasoline directly to the lake over this period to account for the observed whole-lake increase. We consider this scenario highly unlikely. Direct exhaust of unburned fuel into surface waters during the operation of marine gasoline combustion engines appears to be a more likely source of increased MTBE. The greatest releases of VOCs come from two cycle engines which have exhaust ports at or below the water surface *(20)*. These engines are most commonly found in personal watercraft and outboard motors. Studies have shown that 4% to more than 50% of the fuel and oil mixture can pass uncombusted through a two cycle engine and into the water *(21, 22, 23)*. This wide range in values is due to different manufacturing considerations and engine operating conditions *(24)*. Data on the partitioning of MTBE in exhaust between air and water for the different watercraft motors is needed.

3.3.3 FATE OF MTBE

From the maximum total lake content of 370 kg as measured on July 7, both MTBE content and concentration declined throughout the remainder of the year. The rate of MTBE loss from Donner Lake differed depending on whether motorized boating was still occurring, as in the very active summer season between July and August, or during the post-Labor Day holiday period in September and October when seasonal boating was significantly reduced. The mean concentration in the upper 10 m of the lake from July to September varied from 9.6 $\mu g \cdot L^{-1}$ to 5.4 $\mu g \cdot L^{-1}$ with the higher levels on July 7, July 22 and September 3. The September 3 date immediately followed the Labor Day weekend (August 30 to September 1). Overall the rate of decline over this period as determined by linear regression was 1.2 kg $MTBE \cdot d^{-1}$ for a half-life of 193 days (assuming first-order kinetics). This represents the loss rate following the large July 4th input, but still during the boating season when new sources of MTBE were present.

Following the Labor Day weekend, boat use decline dramatically (Figure D-4). This marks the end of the active summer recreation period and while some watercraft continued to use Donner Lake, the numbers are significantly reduced and indeed, boat use censusing by municipalities stops. In the 27-day period between 3 and 30 September, the whole-lake MTBE content dropped from 300 kg to 80 kg at a rate of 8.1 kg $MTBE \cdot d^{-1}$. The corresponding half-life without significant boat use was approximately 14 days. This is supported by the observation that between July 7 and July 22 when weekly boat use fell, the measured MTBE rate of loss was similar at 6.7 kg $MTBE \cdot d^{-1}$.

Calculations show that only a minor portion of the observed MTBE loss between September 3-30 could be accounted for by the increase in Donner Lake outflow during this period. Total discharge from the lake at that time was 0.034 x 10^8 m^3. Even with a high-side estimate of a 5.0 $\mu g \cdot L^{-1}$ MTBE concentration during that entire time (actual levels declined from a high of 6.0 to a low of 1.4 $\mu g \cdot L^{-1}$), the total MTBE loss would be approximately 15 kg or only 7 percent of the 225 kg loss. Volatilization appears to be the primary mechanism responsible for the major loss of MTBE from Donner Lake. MTBE volatilization also occurred during periods of watercraft activity but with at a reduced net loss from the lake because of the continued source from new emissions. It is also very noteworthy that this MTBE loss occurred prior to complete lake mixing. On September 30 a strong thermocline still existed at a depth of 13-14 m and indeed, even by November 18 the water column was still not isothermal.

Wind is an important meteorological forcing factor affecting the transfer velocity of MTBE at the air-water interface and consequently volatilization from the surface of lakes. For Donner Lake, mean daily wind speed data was available from nearby Truckee, CA during the entire period of our MTBE study *(25)*. While the mean daily wind speed varied from day-to-day, the magnitude of this variation was relatively low. From March to November 1997, the range of the monthly average for daily mean wind speed was only 1.7-2.1 $m \cdot sec^{-1}$. In comparison, these values declined to 1.1-1.5 $m \cdot sec^{-1}$ in the winter month (November-February). Visual comparison of the mean daily wind speed data to whole-lake MTBE content for Donner Lake clearly showed that (a) the MTBE increase during the late spring-summer period was not the result of a season decline in wind speed which could have allowed a build up of this compound, and (b) reductions in boat use and not changes in wind speed were most closely related to the observed pattern of MTBE content. Since MTBE data was collected on a bi-weekly or monthly schedule, it is not possible to ascertain the affect of wind on the daily change in MTBE during the period September 3-30.

Between September 30 and October 20, the mean MTBE concentration in surface waters (0-10 m) declined from approximately 1.4 $\mu g \cdot L^{-1}$ to 0.5 $\mu g \cdot L^{-1}$ (Figure D-3) with a fall in whole-lake MTBE content from 80 kg to 45 kg (Figure D-4). During September-November, MTBE concentration in the deeper waters (20 m) ranged from 0.2-0.4 $\mu g \cdot L^{-1}$. Complete mixing had not occurred by November 18 and surface water concentrations were still slightly above those found in the deeper waters. By the last sampling on January 15 (1998), the water column was isothermal at 3.9-4.0 °C, and MTBE was uniformly distributed from surface to bottom at concentrations slightly above the 0.1 $\mu g \cdot L^{-1}$ limit of detection (Figure D-3). Total lake MTBE in January was at a seasonal low of 15 kg. For reference, if all measured values in the water column were at the 0.1 $\mu g \cdot L^{-1}$ analytical detection, total lake MTBE content would be 12 kg. The minimal concentrations measured in January strongly indicate that in Donner Lake there was no inter-annual carry over of MTBE.

Motorized watercraft were clearly the most important source of MTBE in Donner Lake; unfortunately, the censusing data was not sufficient to separate the relative contribution of the various makes and models (e.g. personnel watercraft, outboard engines, etc.). The importance of these watercraft as a significant source of MTBE to lakes and reservoirs has not been emphasized in many of the early reviews on sources and environmental behavior of this compound *(4, 6)*. However, a number of data sets which are only now being collected from drinking water reservoirs, including this Donner Lake study, the work of the Metropolitan Water District in Southern California *(8)*, the preliminary survey at Lake Tahoe by the U.S. Geological Survey *(12)* and a recent survey by the Association of California Water Agencies *(26)* show the importance of this source. While many of the observations from Donner Lake may be applicable elsewhere, many lakes and reservoirs have unique features that must be accounted for in an analysis of MTBE transport and fate. When considering management options for individual lakes or reservoirs, these features include but are not limited to; thermocline stability, volume, lake hydrodynamics, water-use schedules, depth of water intake system, etc. Given the concern regarding MTBE in drinking water supplies, it is important that simulation models are developed for the purpose of lake management and environmental planning. Comprehensive monitoring is needed for a wide range of lake and reservoir types.

3.4 ACKNOWLEDGMENTS

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3.6 LIST OF FIGURES

4.0 LAKE TAHOE EVALUATION

4.1 SURFACE WATER

Four independent, although associated monitoring programs for MTBE have been conducted at Lake Tahoe since 1997. From July 29 to September 5, 1997 the U.S. Geological Survey (Carson City, NV) measured volatile organic compounds (VOCs) in Lake Tahoe and other nearby lakes within the Tahoe drainage area (Boughton and Lico 1998). While this was not an extensive survey (only 14 samples in Lake Tahoe and 6 samples in three additional lakes), it did serve as a preliminary data set which provided basin-wide information on the extent of MTBE and other fuel additives. The sampling design was not intended to look at the seasonal distribution of MTBE and other VOCs but rather to monitor (1) a series of nearshore areas where watercraft activity is common, (2) two open-water locations and (3) three background reference lakes which largely do not allow motorized boating. Findings in this initial survey suggested the following:

- MTBE concentrations in Lake Tahoe were higher in areas of boating activity (3 m depth). In early September, measured values from these areas ranged from 0.30 to 4.2 µg/L (ppb) with a mean of 1.3±1.3 µg/L (±SD).

- MTBE from the same depth but in the open-water was less at 0.51±0.12 µg/L.

- Each of the other three lakes which allow little or no motorized boating (Marlette Lake, Angora Lake, Cascade Lake) showed MTBE to be less than the 0.11 µg/L limit of detection (LOD).

- The maximum depth sampled was 30 m; therefore it was not possible to determine if MTBE mixed into the deeper waters of Lake Tahoe.

- The alternative oxygenates ETBE (ethyl *tert*-butyl ether) and TAME (*Tert*-amyl methyl ether) were also analyzed. ETBE was always below the 0.054 µg/LOD. TAME was measurable in three samples from the nearshore, boat-trafficked area in Lake Tahoe (0.20, 0.14 and 0.14 µg/L relative to the 0.11 µg/L LOD.

During the same year, Glenn Miller (University of Nevada, Reno) and Mary Fiore (Lahontan Regional Water Quality Control Board, CA) collected samples for MTBE analysis in areas of known boat use at Lake Tahoe (e.g. off shore from marina, adjacent to fuel pumps, end of piers). These samples were taken from June to November in order to observe the seasonal distribution. Concentrations varied depending on location. Offshore from Ski Run Marina (a jet ski storage area), concentrations were high exceeding 20 µg/L in both July and September. Next to a fuel pump in the same location a maximum of 47 µg/L was measured on 25 July 1997. Alternatively, other areas in the same general region showed values less or equal to 3 µg/L. In a prelude to a more extensive study in 1998, they also found that MTBE increased to 4.40 µg/L after a 2-stroke jet ski passed through relatively uncontaminated water (MTBE less than 0.1 µg/L LOD).

An important issue raised during the discussion of 2-stroke personal watercraft at Lake Tahoe was the possibility that MTBE might mix to the deeper depths of Lake Tahoe and accumulate in the bottom waters until the lake completely mixed. Since Lake Tahoe experiences full overturn on the average of only once every four years this was a concern. In February 1998 the Tahoe Research Group (University of California, Davis) began a monthly sampling for MTBE in Lake Tahoe which included up to 13 discrete depths from the surface to the bottom at 450 m. Eleven profiles were taken between February and October. Concurrently, surface water was taken at a nearshore area near Tahoe City where boating occurs. The highlights of this on-going monitoring program include:

- In the open waters, measurable concentrations of MTBE above the 0.1 µg/L LOD were first observed in early July. Because of colder than normal air temperatures boating activity had a later than usual start. MTBE at 0 and 3 m was typically very similar with levels of about 0.15-0.20 µg/L in July increasing to 0.25-0.35 µg/L in August and September. The maximum concentration at the mid-lake sampling station was 0.34 µg/L (Figure T-1).

- At no time has MTBE been found at concentrations above detection at depths of 20 m or greater, i.e. measurable levels have only been found at 0-10 m. As observed in other lakes, the presence of a stable thermocline retards the movement of MTBE into deeper waters prior to turnover.

- Lake Tahoe typically mixes to depth in the winter months of February and March. MTBE data from the October 7 profile were all below the 0.1 µg/L limit of detection showing that MTBE disappeared from the lake well in advance of mixing. This is supported by the February and March data which also showed no MTBE in the deep profile from surface to bottom (450 m). It is therefore highly unlikely that MTBE will persist and accumulate either at depth or indeed anywhere in the open water. In 1997, Miller and Fiore found that MTBE was at or near the limit of detection by early November even in areas where the concentration in late July was as high as 47 µg/L. The same conclusion was reach from research at Donner Lake (Reuter et al. in press).

- MTBE at the Tahoe City, nearshore location ranged from less than detectable to 3.77 µg/L, similar to the 1997 USGS findings. The mean concentration (±S.D.) at the combined depths of 0 and 3 m during the July-September boating season was 1.0±0.9 µg/L. There was no difference in the timing of MTBE appearance in the nearshore versus open waters, i.e. MTBE was first observed at both locations on July 2 and disappeared by October 7 (Figure T-1).

The fourth of the large studies of MTBE at Lake Tahoe is a continuation of the monitoring by the USGS. This work began in the summer of 1998 at Lake Tahoe and other regional waterbodies and will continue for one year. Preliminary data were not available for release but showed a great deal of similarity to USGS 1997 survey and the UC Davis-Tahoe Research Group's data.

Finally, a focused sampling for MTBE in the surface waters of nearby Fallen Leaf Lake was done by John Kleppe (University of Nevada, Reno). A 3-point transect was sampled twice in 1998; on September 4 and September 8. The range of concentrations found was 0.7-1.5 µg/L with most values in the 0.7-0.8 µg/L range.

4.2 SURFACE WATER DRINKING SUPPLIES

Lake Tahoe has been a source of drinking water since man first settled in the basin. Many of the old homes along the lakeshore still utilize Tahoe water for domestic use through private intakes. These are often located in shallow water (<20m) a short distance offshore. As the Tahoe basin resident and seasonal tourist populations increased, both public utility districts and private water companies plumbed the lake to meet the growing water demand. Today, Tahoe water is used exclusively by a few water companies, and in combination, or as a reserve for ground water sources by others. Those water companies utilizing the lake as a reserve supply generally start serving the surface water to customers during the peak demand, which occurs during the summer months.

There are currently eight water purveyors around Lake Tahoe serving lake water during at least some portion of the year to customers through upwards of 8,000 connections. The water companies and utility districts which use the lake as a reserve source of drinking water, generally tap into its supply during the summer months, coinciding with peak visitation to the basin. This is also the time when the greatest amount of boat traffic plies the lake's surface and fuel constituent concentrations reach an annual maximum.

The eight water purveyors draw water from 13 different intakes around the lake. These are concentrated in the northwest and southeast areas of the lake and are situated at the edge of the near shore zone as defined in this report (<10m depth and within 500 m of shore). The depth of the intakes range from 5 m to near 20 m depth, with nearly half being located in the top 10 m of water. The intakes cover a large range in lateral distance from shore (85 m to 670 m) with little correlation between offshore distance and the depth of the intake. Intakes that are situated on shelf areas tend to extend the furthest into the lake but do not reach the greatest depths.

Five of Tahoe's water purveyors have sampled the raw water from their intake lines for MTBE. While two of the purveyors had positive detections of MTBE, only one recorded MTBE levels consistently greater than the level of detection (>0.5 µg/L) throughout the boating season. These results were associated with the shallowest drinking water intake (approximately 5 m). Reported MTBE concentrations peaked just after the Labor Day weekend (2.4 µg/L, data provided by Round Hill General Improvement District).

While valuable information was provided by concerned and responsible water purveyors, it is suspected that if all of Tahoe's drinking water intakes were tested during the summer season, more positive results would have been reported. This conclusion is based on the fact that MTBE during the USGS, UCD and UNR nearshore surveys was often found to be above the limit of detection and in the range of 0.1 µg/L to a high in excess of 40 µg/L depending on boating activity. Testing by water purveyors should be more comprehensive.

4.3 GROUNDWATER

Graham Fogg, Jim Trask and Mary Meays, UC Davis, provided the groundwater data for this sub-section.

Within the Lake Tahoe Basin, 16 shallow wells (depth <30 feet) and a spring were sampled for MTBE analysis during summer 1998 by UC Davis scientists. Four of these wells had detectable MTBE (>0.1 µg/L), all at levels from 0.1 to 0.5 µg/L. For three of these four wells, sources of MTBE other than precipitation were strongly implicated. Two of these four wells were located within Pope Marsh, a wetlands areas. A snow pit at 8,600 feet elevation in Heavenly Valley was sampled for MTBE; no MTBE was detected (detection limit of 0.1 µg/L). Although further sampling of precipitation events would be needed to clarify the role (if any) played by precipitation in transporting atmospheric MTBE to surface and ground waters in the Lake Tahoe Basin, our data, together with that of others (Bruce and McMahon 1996), strongly indicate that the contribution of precipitation (snow and rain) to MTBE in groundwater is small (<0.5 µg/L) at most in the Lake Tahoe Basin. A similar finding was reported for snow at nearby Donner Lake (B.C. Allen and J.E. Reuter unpub. data). In addition, three measurements of MTBE levels entering water as a result of dry atmospheric deposition during the summer have been made at Lake Tahoe (B.C. Allen and J.E. Reuter unpub. data). Over the Labor Day weekend in 1997 the MTBE level in dry deposition near the lakeshore was less than the 0.1 µg/L limit of detection. During the 4th of July weekend in 1998, concentrations in dry deposition were above detection, albeit low, near the lakeshore (0.15 µg/L) but again below detection in the open water portion of the lake.

Gasoline LUFT sites that have contaminated groundwater with MTBE are ubiquitous in the Lake Tahoe Basin. 31 of 48 registered gasoline LUFT sites in the basin have reported MTBE in groundwater. MTBE levels have been over 1,000 µg/L at each of nine LUFT sites examined. MTBE plumes as long as 1500 feet have been detected, and the MTBE plumes are comparable in size to or larger than benzene plumes at nine of nine LUFT sites examined. MTBE has been detected as deep as 80 feet below the water table. Gasoline LUFT sites have had a large impact on public drinking water supply wells in the southern portion of the Lake Tahoe Basin. The South Lake Tahoe Public Utility District reports that 11 drinking water wells have been closed because of MTBE contamination. This represents 32 percent of the district's drinking water wells. As a consequence, water usage restrictions for its customers were required during the summer of 1998.

While there would otherwise be little immediate concern regarding LUFT sites as significant sources of MTBE to surface waters (i.e. via normal groundwater flow) on a state-wide basis because most of the drinking water lakes and reservoirs are not urbanized around the shoreline, Lake Tahoe could be a potential exception. Given that plumes of approximately 1500 feet have been identified, and that many of the existing and potential LUFT sites (gas stations) are within 500-1000 feet of the lake shore, the possibility exists that MTBE could enter Lake Tahoe or one of its tributaries from this source. Indeed, data supplied by the Lahontan Regional Water Quality Control Board (South Lake Tahoe) indicate that at least 5 LUFT sites may threaten surface waters. At these sites the leading edge of an MTBE plume is within 800-2000 feet of either Lake Tahoe or a stream. Using the assumption that regional horizontal groundwater velocities away from wells are on the order of 100 feet per year (Jim Trask, Department of Land, Air and Water Resources, UC Davis, pers. comm.), these sources may directly impact surface waters within 8-20 years. The potential magnitude of this impact has not yet been quantitatively assessed.

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