

# Gasoline Composition Regulations Affecting LUST Sites

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by

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#### **Notice**

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#### **Abstract**

Passage of the Clean Air Act Amendments in 1990 imposed requirements on gasoline composition in the United States. Impacts to ground water are affected by the provisions that required oxygenated additives and limited benzene concentration. Reformulated and oxygenated gasoline were required to contain an oxygenated additive at 2.0 wt % and 2.7 wt %, respectively. In most cases, the additive initially was methyl tert-butyl ether (MTBE). The amount of benzene in both reformulated and conventional gasoline was limited: reformulated gasoline contains less than 1 % benzene by volume, while benzene levels in conventional gasoline were set by producer baselines. The allowable benzene levels vary among these producers and, unlike reformulated gasoline, are not tied to use at specific locations in the U.S. In 2000, states began to pass bans on MTBE, other ethers, and/or alcohols; consequently, production and use of MTBE in reformulated gasoline declined. The Energy Policy Act of 2005 removed the oxygenate requirement from reformulated gasoline and industry responded by removing ethers from U.S. gasoline, with some limited exceptions.

#### **Foreword**

The National Exposure Research Laboratory's Ecosystems Research Division (ERD) in Athens, GA conducts research on organic and inorganic chemicals, greenhouse gas biogeochemical cycles, and land use perturbations that create direct and indirect chemical and non-chemical stresses, exposures, and potential risks to humans and ecosystems. ERD scientists develop, test, apply and provide technical support for exposure and ecosystem response models that assess and manage risks to humans and ecosystems within a watershed /regional context. The Regulatory Support Branch (RSB) at ERD conducts problem-driven and applied research, develops technology tools, and provides technical support to customers in program and regional offices, states, municipalities, and tribes. Models are distributed and supported by EPA's Center for Exposure Assessment Modeling (CEAM) and its Internet tools (www.epa.gov/athens/onsite).

Research on gasoline composition is a fundamental part of assessing exposures to contamination from leaking underground storage tanks and surface water releases of petroleum fuels. Statistical analysis of gasoline samples, however, is not a sufficient basis for understanding variations in gasoline composition across the United States. Familiarity with the United States' unique regulatory requirements is a necessary precursor to studying gasoline composition. This report gives an overview of the regulations affecting major contaminants at leaking underground storage tank sites.

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### Leaking Underground Storage Tank Assessment Report Series

A series of research reports is planned that will present data and models for leaking underground storage tank risk assessments. To date, these include:

#### 1. Gasoline Composition

- Weaver, James W., Linda R. Exum, Lourdes M. Prieto, 2008, Gasoline Composition Regulations Affecting LUST Sites, United States Environmental Protection Agency, Washington, D.C., EPA/600/R-10/001.
- Weaver, James W., Sheldon A. Skaggs, David L. Spidle, and Guthrie C. Stone, 2009, Composition and Behavior of Fuel Ethanol, United States Environmental Protection Agency, Washington, D.C., EPA/600/R-09/037.
- Weaver, James W., Lewis Jordan and Daniel B. Hall, 2005, Predicted Ground Water, Soil and Soil Gas Impacts from US Gasolines, 2004: First Analysis of the Autumnal Data, United States Environmental Protection Agency, Washington, D.C., EPA/600/R-05/032.

#### 2. Simulation Models

- Gorokhovski, Vikenti M. and James W. Weaver, 2007, A Catalog of Ground Water Flow Solutions for Plume Diving Calculations, United States Environmental Protection Agency, Washington, D.C.
- Weaver, James W., 2004, On-line Tools for Assessing Petroleum Releases, United States Environmental Protection Agency, Washington, D.C., EPA 600/R-04/101.

#### 3. Model Background and Evaluation

Weaver, James W. and C. S. Sosik, 2007, Assessment of Modeling Reports for Petroleum Release and Brownfields Sites, United States Environmental Protection Agency, Washington, D.C., EPA 600/R-07/101.

As reports are added to this series, they can be found on EPA's web site: http://www.epa.gov/athens/publications.

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#### Introduction

The magnitude of environmental contamination from gasoline releases depends, among other factors, on composition of the gasoline. The amount of certain components in gasoline varies for a number of reasons, including fuel performance characteristics, refinery equipment and capabilities, crude oil sources, industry standards and regulation. Of the many causes of variability, regulatory requirements may be dominant for some components. Limits on certain component concentrations and properties have been set by regulation. Benzene, ethers and alcohols are the best examples of components with limitations, and vapor pressure is the best example of a limited property. In addition to being regulated for their impact on air pollution, benzene and methyl *tert*-butyl ether (MTBE) and other oxygenates are ground water contaminants. This report gives an overview of the air quality regulations that affect major contaminants at leaking underground storage tank sites.<sup>1</sup>

#### **History**

The Clean Air Act of 1970 established the framework for setting nation-wide air quality goals (NAAQs) for pollutants Subsequently, states developed implementation plans (SIPs) to meet those goals. To date, the U.S. Environmental Protection Agency (EPA) and states have set goals for six pollutants: sulfur dioxide, particulate matter, nitrogen oxides, carbon monoxide (CO), ozone, and lead (Ayres and Kornreich, 2004). EPA was authorized to require registration and testing of specified fuels and fuel additives (US Federal Register, 1975, 1976). Because of these requirements, we know MTBE was registered for use in 1979, *tert*-amyl methyl ether (TAME) in 1981, and ethyl *tert*-butyl ether (ETBE) in 1981 (Stikkers, 2002). Under authority granted by the Clean

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<sup>&</sup>lt;sup>1</sup> The information provided in this paper is not intended for determining compliance with state or federal regulations for air quality, gasoline composition or other similar purposes. For these purposes, information should be obtained from the appropriate local, state and federal regulatory agencies, the Clean Air Act, the United States Code, the Code of Federal Regulations, the Federal Register and other sources.

Air Act, EPA began to phase out leaded gasoline over a period of time that ended on January 1, 1996. The original focus of the Clean Air Act was to limit emissions through improved vehicle technology. As time went on this approach became less viable for solving persistent pollution problems, and Congress believed that lead was being replaced by toxic organic chemicals (Martels, 2004). Consequently, the Clean Air Act Amendments (CAAA) of 1990 (42 U.S. Code 4701) expanded regulation of fuels to help meet NAAQs for ozone and carbon monoxide and reduce toxic air pollutants (e.g., benzene).

The CAAA introduced several requirements that have had a major impact on gasoline composition throughout the United States, beginning with implementation in 1992 and 1995, and continuing to the present. The most important requirements for LUST sites were the total ban on lead in gasoline, and new requirements for three types of gasoline: conventional, reformulated, and oxygenated. Both reformulated gasoline (RFG) and oxygenated gasoline (OG) required oxygen-containing additives, because the fuel would burn cleaner. Initially, the most common oxygenate was MTBE. The RFG program limited the amount of benzene and total aromatics in reformulated gasoline. Since RFG areas were specified at county or partial-county level or, in a few cases, at the city level, there are different requirements in adjoining counties. This spatial distinction between RFG and CG might not be absolute, however, because market forces guide gasoline sales, and there are no restrictions on selling RFG in CG counties.

Parts of the country not using RFG were also affected by the CAAA because the Act contained an anti-dumping provision to prevent air quality deterioration in areas using conventional gasoline (CG). This requirement prevented benzene from being moved out of the RFG and into the CG supply by establishing benzene concentration limitations from producer/importer baseline conditions that existed in 1990. An important distinction between CG and RFG is that CG baseline limitations are applied to producers/importers, while RFG requirements apply to where the fuel is used.

Consequently, at a given location the benzene concentration in CG is usually variable and not very predictable<sup>2</sup>.

In response to dissatisfaction with ground water contamination, a number of state legislatures banned MTBE and, in some cases, other ethers and alcohols, beginning in 2000. These state bans did not affect federal oxygen requirements for RFG and OG, however, so MTBE typically was replaced by ethanol. In 2005, Congress passed the Energy Policy Act (EPAct 2005) which removed the oxygenate mandate from the RFG program. Gasoline suppliers responded by reducing the use of MTBE and other ethers.

A composition-related aspect of gasoline is octane number. Some gasoline -called straight run gasoline -- is a direct output from distilling crude oil, but its octane
number is too low to prevent engine knock in modern engines. Therefore, the octane
number is boosted in a number of ways. These commonly include the use of alkyl leads,
aromatic hydrocarbons, ethers, alkylate and alcohols (Owen and Coley, 1995). Shifts
among these have occurred, partly due to laws and regulations that address different goals
(see e.g., Stikkers, 2002). Only small amounts of certain additives, 1 g/L or less of alkyl
leads or 0.017 g/L methylcyclopentadienyl manganese tricarbonyl (MMT), are needed to
increase octane levels from 5 to 25 octane numbers, depending on the blendstock (Owen
and Coley, 1995). Blending higher amounts of some organic compounds also increases
resistance to engine knock, but their levels typically must be one percent or higher. In
1979, MTBE was registered as an octane enhancer, so it may have appeared in gasoline
for octane purposes even when there was no regulatory mandate for oxygenated
additives. This has been borne out by gasoline composition studies in which MTBE
appeared in CG when it was not required (Weaver et al., 2005).

Timing of the mandates varied according to federal requirements or state implementation plans. Nationally, the RFG program had an implementation date of January 1, 1995, so RFG began appearing on the market in late 1994. Opt-out and opt-in

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 $<sup>^2</sup>$  In 2011 the Mobile Sources Air Toxics rule (U. S. Federal Register, 2007) will reduce benzene levels in all gasoline.

provisions allowed areas to enter or leave the programs at different times. The oxygenated gasoline program began in the fall of 1992. Since it specified oxygenated fuel to be used only during a few months in winter, there was a characteristic pattern of oxygenate usage in these locations. Where NAAQs for CO were met later, implementation plans were revised to remove oxygenated gasoline requirements; for example, see U. S. Federal Register (1999, 2000) for an example from New York State.

#### **Data Sources**

In the next two sections, data from three sources were compared to the regulatory time lines. First was the Northrup-Grumman data set (e.g., Dickson, 2006), which published results from an industry consortium that has collected data since the 1930s. This company is the latest successor to the well-known National Institute for Petroleum and Energy Research (NIPER). Its analyses were performed using ASTM standard methods for analysis of aromatics, ethers and alcohols. Northrup-Grumman data from 1976-2007 were obtained in an electronic format, then sorted by city and octane number (ON). Three octane number classes were used: regular (less than 88.5 ON), mid-grade (88.5 ON to 90 ON), and premium (above 90 ON).

The second source was data from the Reformulated Gasoline Survey Association. This group conducts surveys of RFG required by the Clean Air Act and reports the results to EPA, which then evaluates the data for compliance. California is not included in these surveys.

Lastly, the U.S. Department of Energy's Energy Information Agency (EIA) collects weekly data from producers on their production of gasoline and a variety of fuel components.

#### **Leaded Gasoline**

Use of lead in gasoline declined throughout the 1980s (Figure 1) and this phasing out contributed to the rise in use of ether in gasoline (Stikkers, 2002). Averaged data

from NIPER/Northrup-Grumman show that lead usage was highest in premium gasoline and, on average, lower in mid-grade and regular gasoline. For example, Figure 1 shows that in 1978, premium gasoline contained at least 1 g/gal, mid-grade at least 0.5 g/gal, and regular may have contained no lead at all. After reaching levels as high as 4 g/gallon, by 1986 most lead was reduced to concentrations of 1 g/gallon. Complete removal of lead was mandated by the Clean Air Act Amendments of 1990, and occurred on January 1, 1996. Lead can still be used, however, in aviation gasoline and racing fuel.

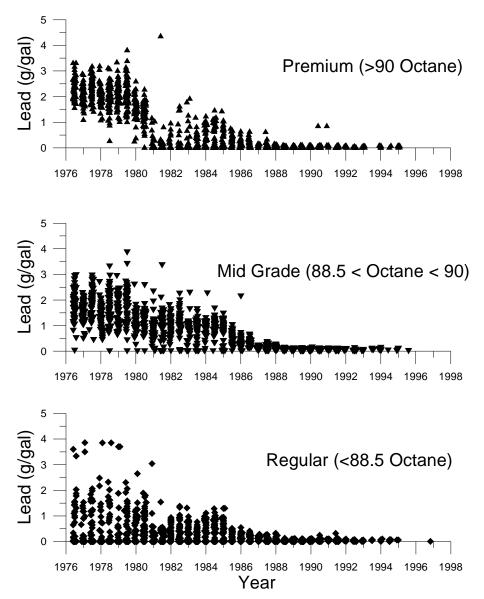


Figure 1 Data illustrating the phasing out of lead from gasoline in the United States in premium (top), mid-grade (middle), and regular (bottom). After declining through the 1980s, lead was completely eliminated by January 1, 1996 by the Clean Air Act Amendments of 1990.

#### **Gasoline Production in the United States**

Gasoline production data compiled by EIA document the production of conventional and reformulated gasoline. The data show that production of RFG began in September 1994, shortly before the RFG program was implemented when RFG replaced part of total gasoline production in the U.S. (Figure 2 A). The remainder of production became conventional gasoline (see U.S. CFR, 2007, Title 40, Part 80, Section 2). Production of both types of gasoline tended to increase over time. In 2004, EIA began differentiating ether-, alcohol-, and non-oxygenated RFG (Figure 2 B). The data show declining use of ether in RFG from 2004 until mid-2006, when ethers finally were replaced by alcohol. The slow decline of ether use corresponds to when MTBE was being banned by the states. When the oxygenate mandate was removed by the Energy Policy Act of 2005, ethers were removed from RFG. While ether use was declining, alcohol use was increasing until almost all RFG produced after the EPAct 2005 took effect contained alcohol. This legislation also required increased use of ethanol, as shown by the increased amount of all gasoline containing ethanol. By December 2009, 77% of all U.S. gasoline contained alcohol.

RFG Survey Association data indicate that MTBE in RFG outside California decreased, on average, from 7.9% to 0.01%; ethanol increased from 2.2% to 9.2%; and benzene remained nearly the same -- 0.61% in 2002 versus 0.69% in 2006. These data also document removal of ethers from the U.S gasoline supply (Figure 4). Ether levels in gasoline declined throughout the summer months of 2006, so that the date when the oxygenate requirement was removed did not correspond to immediate removal of ethers. Some U.S. gasoline is imported, but no RFG with ether has been imported since June 2006<sup>3</sup> (Figure 3 A). Production of RFG with ether has continued sporadically at very low levels from 2004 through 2009 (Figure 3 B)<sup>4</sup>.

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<sup>&</sup>lt;sup>3</sup> Conventional gasoline is also imported, but EIA data show only very small amounts of alcohol-containing gasoline has been imported, and none since Mar, 2008.

<sup>&</sup>lt;sup>4</sup> EIA export data show only the total of petroleum products exported.

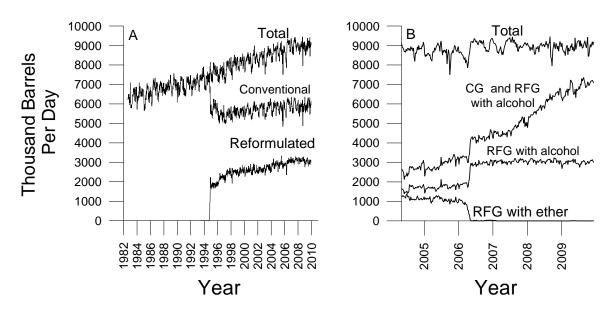


Figure 2 DOE gasoline production data. A. Prior to the Clean Air Act Amendments, there was no distinction between conventional and reformulated gasoline. Since 1995, the proportion of conventional gasoline has remained roughly constant. B. Since removal of the oxygenate mandate in 2006, the amount of reformulated gasoline with ether has declined dramatically, although there has been a commensurate increase in RFG with alcohol. Mandates for increased use of ethanol have increased the amount of CG with alcohol, while the amount of RFG with alcohol has remained roughly constant. The total amount of gasoline with alcohol has increased to be approximately 77% of the total in December 2009.

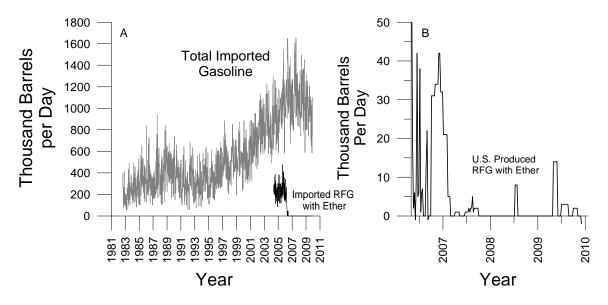


Figure 3 A. Gasoline imported into the U.S. from 1982 to 2009. Some RFG with ether was imported prior to 2006, but none since. B. From 2006 to 2009, small amounts of RFG with ether have been produced in the U.S.

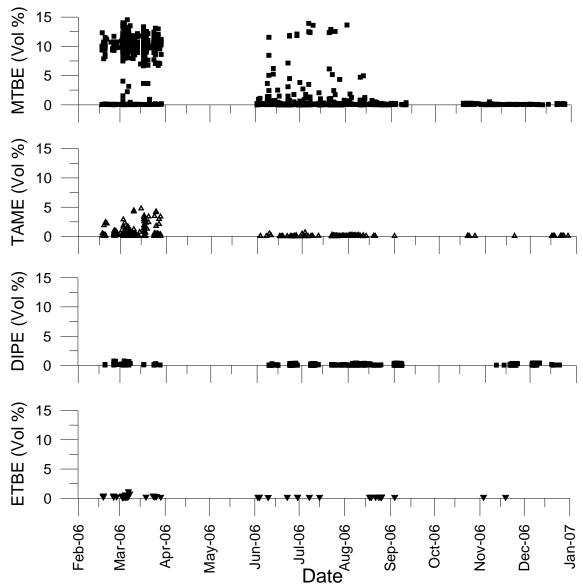


Figure 4 RFG Survey Association data, as reported to EPA, show that the removal of ethers from reformulated gasoline (except in California) occurred over the summer months of 2006.

#### **Reformulated and Conventional Gasoline**

#### Reformulated Gasoline

EPA defines reformulated gasoline as gasoline that is certified (U.S. CFR, 2007, Title 40, Part 80, Section 41) to meet requirements and standards specified in U.S CFR, 2007, Title 40, Part 80, Section 42. The requirements varied during four time periods: 1995-1997; 1998-1999; 2000 to May 5, 2006; and May 5, 2006 to the present (April 24, 2006 in California, see U.S. EPA, 2006). Although other requirements of RFG changed over these times, the required oxygen content and benzene limitation did not change until the Energy Policy Act passed in 2005 (Table 1). The oxygen requirement was removed in California effective April 24, 2006, and in the rest of the U.S. effective May 5, 2006. Beginning in 2011, benzene content in all U.S. gasoline will be reduced to 0.62 vol % to comply with the Mobile Sources Air Toxics Rule (U.S. Federal Register, 2007).

As shown in Table 1, standards were met on either an averaged or per-gallon basis. Using average basis, oxygen concentration in a gallon may have been as low as 1.5 wt %, but had to average 2.1 wt %. The total oxygen content could have been limited to 3.2 wt % when gasoline contained ethanol (U.S. CFR, 2007, Title 40, Part 80, Section 41 (g)(i)).

Table 1 Oxygen and benzene requirements for reformulated gasoline.

Component	<b>Effective Dates</b>	RFG Content Requirements			
		Per-gallon	gallon Averaged Basis		
		Basis	Standard	Limit	
Oxygen (weight percent)	1995 to May 5, 2005	≥ 2.0	≥ 2.1	≥ 1.5	
	May 5, 2005 to present	none	none	none	
Benzene (volume percent)	1995 to present	≤ 1.00	≤ 0.95	≤ 1.30	

The amounts of various ethers and ethanol that were needed to meet the oxygen requirement are shown in Table 2; for example, a gallon of gasoline containing 11.0 vol % MTBE met the per gallon oxygen requirement of 2.0 wt %. If the producers chose to

meet the standard on an averaged basis, there could have been compliant gasoline with MTBE content as low as 8.25 vol %. Thus, while a gallon of gasoline containing 11.0 vol % MTBE clearly complied with the standard, gasoline containing less than 11.0 vol % could comply in two ways. First, if the producer chose to meet the standard on an average basis and the batch met the 2.1 wt % oxygen requirement, then the fuel was compliant. The second was when more than one oxygenate was present: any single oxygenate could be present in a relatively low concentration, but the total oxygen supplied by all compounds had to meet the requirement. Similarly, RFG containing 1.0 vol% benzene complied with the per-gallon basis. One specific gallon of gasoline containing 1.30 vol % benzene could be compliant if the producer met the standard using the average basis.

Table 2 Oxygen and required amounts of oxygenates to meet requirements of reformulated and winter oxygenated gasoline. Values given as weight percent.

	Required Oxygenate Concentration					
		RFG		Winter	Oxygenate	
	Per-gallon	Averag	ed Basis	2.7%	3.1% oxygen	
	Basis	Standard	Minimum	oxygen	required	
Weight percent of oxygen	2.0 %	2.1%	1.5%	required		
Common (	Oxygenates and Re	quired Content to	Meet Oxygen Req	uirements		
Methyl tert-butyl ether	11.0%	11.6%	8.25%	14.9%	17.1%	
Ethyl tert-butyl ether	12.75%	13.4%	9.6%	17.2%	19.8%	
Tert-amyl methyl ether	12.75%	13.4%	9.6%	17.2%	19.8%	
Diisopropyl ether	12.75%	13.4%	9.6%	17.2%	19.8%	
Ethanol	5.8%	6.0%	4.3%	5.4%	6.2%	

#### State Cleaner Burning Gasoline Programs

Three states, Arizona, California and Nevada, have implemented cleaner burning gasoline (CBG) programs. A summary of requirements for California's program are given in Table 3. Arizona requires cleaner burning gasoline in the Phoenix area (U.S. Federal Register, 2004), and Nevada requires the same for the Las Vegas area (Clark County, Nevada, 2003). Arizona's cleaner burning gasoline matches the characteristics of either federal RFG or California CBG. Nevada imposes limits on sulfur and aromatic composition.

Table 3 MTBE and benzene requirements for California cleaner burning gasoline (CBG), given in units of volume %. California allows refiners to meet standards for each batch of finished gasoline—the "flat" limit. Alternatively, a refiner can meet standards on an average over 180 days. In the latter case, the averaging limit must be met for all batches and no batch can exceed an upper/lower limit ("cap"). The lower cap limit of 1.8% in Phase 2 and Phase 3 apply to certain counties in the winter (California, 2003a).

	Phase 1	Phase 2			Phase 3			
	January, 1992	March, 1996			January, 2004			
		flat avg cap			flat	avg	cap	
Oxygen content	1.8%-2.3%	1.8% - 2.2%	n/a	1.8%-3.5%	1.8% - 2.2%	n/a	1.8%-3.5%	
				0% - 3.5%			0 %-3.5%	
Benzene	1.7%	1.0%	0.80%	1.20%	0.80%	0.70%	1.10%	

#### Conventional Gasoline

Conventional gasoline is gasoline that has not been certified as RFG (i.e., meeting the U.S. CFR, 2007, Title 40, Part 80, Section 41 requirements), but it must meet requirements of anti-dumping provisions of the CAAA (U.S. CFR, 2007, Title 40, Part 80, Section 90 and following sections). These were designed to prevent increased average per-gallon emissions of volatile organic compounds, nitrogen oxides, carbon monoxide and toxic air pollutants, in addition to requirements imposed on reformulated gasoline (CAAA Sec 211(k)(8)). Baselines for each refiner and producer were set, using their production/importation for 1990, or by statutory baseline with a benzene content of 1.53 vol % for winter and 1.64 vol % for summer (U.S. CFR, 2007, Title 40, Part 80, Section 91(c)(5) and Section 45, (b)(2)). Other features of the statutory baseline are

given in Table 4. Because of different producer baselines, benzene content of gasoline in conventional gasoline areas can vary. Recent EPA studies have shown benzene content in conventional gasoline ranged from 0.5 vol % to 3.0 vol % or in very limited instances to 5% (Weaver et al., 2005)...

Table 4 Selected parameters of the statutory baseline for conventional gasoline (40 CFR 80.91 and 40 CFR 80.45)

Parameter	Winter	Summer	Average		
	40CFR 80.91(c)(5)(i)	40CFR 80.91(c)(5)(ii)	40CFR 80.91(c)(5)(ii)		
	40CFR 80.45(b)(2)	40CFR 80.45(b)(2)			
Benzene (vol %)	1.64	1.53	1.60		
Aromatics (vol %)	26.4	32	28.6		
Olefins <sup>(*)</sup> (vol %)	11.9	9.2	10.8		
Reid Vapor Pressure	8.7	8.7	8.7		
(psi)					
API Gravity <sup>(**)</sup> (°API)	60.2	57.4	59.1		

<sup>(\*)</sup>Olefins are also known as alkenes

$$API Gravity = \frac{141.5}{Specific Gravity} - 131.5$$

#### Geographic Extent

A map of locations required to use federal reformulated and conventional gasoline is shown in Figure 5. The map shows that RFG has been used in the northeast corridor, large Midwestern cities, Arizona and California. It also shows that geographically, most of the U.S. uses conventional gasoline. For the majority of counties using RFG, the RFG oxygenate mandate was in force from January 1, 1995 to May 5, 2006. Counties in the Los Angeles area were included at the beginning of the program, but had an earlier date (April 24, 2006) for removing the oxygen mandate. Counties in the Sacramento metropolitan area were added in 1996, and those in the San Joaquin Valley in 2002. These latter two groups also share the April 2006 federal oxygen mandate removal date.

<sup>(\*\*)</sup> API Gravity (°API) is defined by:

Counties in the St. Louis, Missouri metropolitan area opted into the program on June 1, 1999. Four counties in Illinois opted into the RFG program in 2007 and, because of the late date, never were required to use oxygenates. In 1997, the Phoenix metropolitan area opted into the program, then opted out in 1998 because of Arizona's cleaner-burning gasoline program (see Figure 5 and U.S Federal Register, 2004). Seven counties in Maine opted out of the program in 1999.

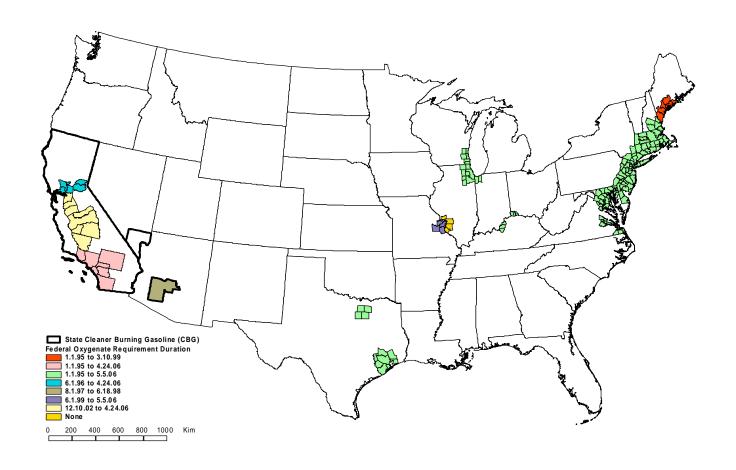


Figure 5 Federal reformulated gasoline and state cleaner-burning gasoline programs in the United States (California, 2003a; Clark County, Nevada, 2003; U.S. CFR, 2005, Title 40, Part 80, Section 40, U.S Federal Register, 2004). Durations of oxygenate mandates are shown on the legend. All areas not highlighted on the map use conventional gasoline. Reformulated gasoline is required above 4,500 ft elevation on Whiteface Mountain in northern New York.

#### Oxygenated Gasoline

The Clean Air Act Amendments (42 U.S.C. 7545, m, 1) required states to mandate at least 2.7 wt % oxygen in gasoline sold in areas where carbon monoxide standards were not attained. This requirement was imposed for at least four months of the year when ambient CO concentrations were highest. EPA could reduce the duration if a state demonstrated there were no exceedances of the carbon monoxide standard. If a carbon monoxide standard was not attained by a required date, gasoline containing 3.1% by weight was required.

The areas were defined as the larger of Consolidated Metropolitan Statistical Area or Metropolitan Statistical Area. The program began in late fall 1992 in 39 areas, with one more added in 1993. Six cities began winter oxygenate programs earlier -- in the 1989/1990 winter season: Denver, Colorado; Reno and Las Vegas, Nevada; Tucson and Phoenix, Arizona; and Albuquerque, New Mexico (Stikkers, 2001). Table 5 lists the nine areas still in the program to the present, along with their required oxygen content and effective months. Table 6 lists areas that no longer implement the program, and their effective ending dates. The geographic concentration of remaining cities in the southwest is shown in Figure 6. Table 7 lists cities that have exited the program, but retain the option of using oxygenated gasoline as a contingency measure.

Six states have state-wide oxygenate mandates (Table 8). Each of the state requirements differ, but the required ethanol contents do not exceed the federal limit of 10%. Montana imposed a conditional requirement for using ethanol in gasoline, in conjunction with a conditional ban on MTBE (Montana, 2005) that has so far not been imposed (Kuhn, 2010).

Table 5 Cities currently implementing the winter oxygenates program (U.S EPA, 2008c).

<b>Control Period</b>	Area (Consolidated Metropolitan Statistical Area)	Oxygen Content (wt %)
10/1 to 1/31	Reno, NV	3.5
10/1 to 3/31	El Paso, TX	2.7
	Las Vegas, NV	3.5
	Reno, NV	3.5
	Tucson, AZ	1.8
11/1 to 2/29	Albuquerque, NM	2.7
	Missoula, MT	2.7
	Los Angeles, CA	1.8 to 2.2
11/2 to 3/31	Phoenix, AZ	3.5

Table 6 Cities exiting the winter oxygenates program (U.S. EPA, 2008c).

Area (Consolidated Metropolitan Statistical Area)	<b>Effective Exit Date</b>
Syracuse, NY	Sep 29, 1993
Cleveland, OH	Mar 7, 1994
Greensboro, NC	Nov 7, 1994
Duluth, MN	Jun 13, 1994
Memphis, TN	Sep 26, 1994
Raleigh-Durham, NC	Sep 18, 1995
Baltimore, MD	Dec 15, 1995
Hartford, CT	Jan 2, 1996
Philadelphia, PA	Mar 15, 1996
Washington, DC	Mar 15, 1996
Boston, MA	Apr 1, 1996
Seattle, WA	Oct 11, 1996
Vancouver, WA	Oct 21, 1996
San Diego, Chico, Modesto, Sacramento, Fresno, San Francisco, and Stockton, CA	Jun 1, 1998
New Haven, CT	Dec 4, 1998
Salt Lake City, UT	Mar 22, 1999
Northern NJ/ Southwest CT/ New York, NY	Nov 22, 1999 Jan 31, 2000 May 19, 2000
Minneapolis/St. Paul, MN	Nov 29, 1999
Colorado Springs, CO	Feb 20, 2000
Ogden, UT	May 8, 2001
Ft. Collins, CO	Jan 1, 2004
Provo/Orem UT	Jan 3, 2006
Portland, OR	Feb 23, 2006
Denver/Boulder, CO	Oct 16, 2007
Longmont, CO	Oct 16, 2007

 $Table \ 7 \ Cities \ exiting \ the \ winter \ oxygenates \ program \ (U.S.\ EPA, 2008c), \ with \ oxygenated \ gasoline \ as \ a \ contingency \ measure \ in \ their \ states' \ maintenance \ plans.$ 

Area (Consolidated Metropolitan Statistical Area)	Redesignation/SIP Change Published in the		
	Federal Register		
Grants Pass, OR	Oct 30, 2000		
Klamath Co, OR	Nov 19, 2001		
Medford, OR	Sep 23, 2002		
Anchorage, AK	Jul 23, 2004		
Spokane, WA	Aug 29, 2005		

Table 8 State ethanol mandates.

State	Ethanol	<b>Effective Date</b>	Citation
	Requirement		
	Vol %		
Florida	9 to 10	Dec 21, 2010	Florida, 2009
Hawaii	10	1994	Hawaii, 2004
Minnesota	2.7 <sup>(a)</sup>	Oct 1, 1997	Minnesota, 1997
	9.2 to 10	2003	Minnesota, 2003
	20 or max allowed	Aug 20, 2013	Minnesota, 2009
	by EPA		
Missouri	10	Jan 1, 2008	Missouri, 2008
Oregon	10	Nov 1, 2009	Oregon, 2009
Washington	"2% of gasoline"	Dec 1, 2008	Washington, 2009

<sup>(</sup>a) Oxygenated fuel requirement, Minnesota ethanol producer credit caused ethanol usage.

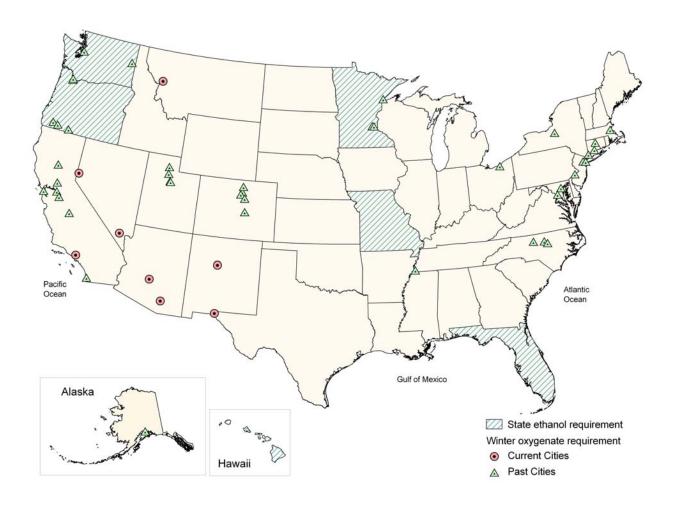


Figure 6 Oxygenated gasoline programs in the United States. Cities where an oxygenated additive was, or is, required for winter time gasoline are indicated by triangles and circles, respectively. Six states have imposed year-round oxygenate mandates. Montana has a future requirement that is conditional on in-state ethanol production.

#### **MTBE Bans**

Twenty-six states, two counties, and one city have banned MTBE in gasoline (Figure 7). Three states have imposed absolute bans, while most allow some MTBE to remain in gasoline (Table 9). In 17 states, the level was 0.5%, but ranged from 0.05% in California to 1% in Nebraska. Six states banned MTBE and other oxygenates which included the other gasoline oxygenate ethers: ethyl *tert*-butyl ether (ETBE); *tert*-amyl methyl ether (TAME); diisopropyl ether (DIPE); and alcohols, mostly *tert*-butyl alcohol (TBA). The bans did not affect requirements to use reformulated or oxygenated-gasoline, but did cause a shift to another oxygenate. One data set shows that the other oxygenate most commonly used was ethanol (Weaver et al., 2005). Oregon passed a conditional ban of various ethers and alcohols, but was only effective if these were banned by each of the California Air Resources Board, the California Environmental Policy Council and the US EPA (Oregon, 2009).

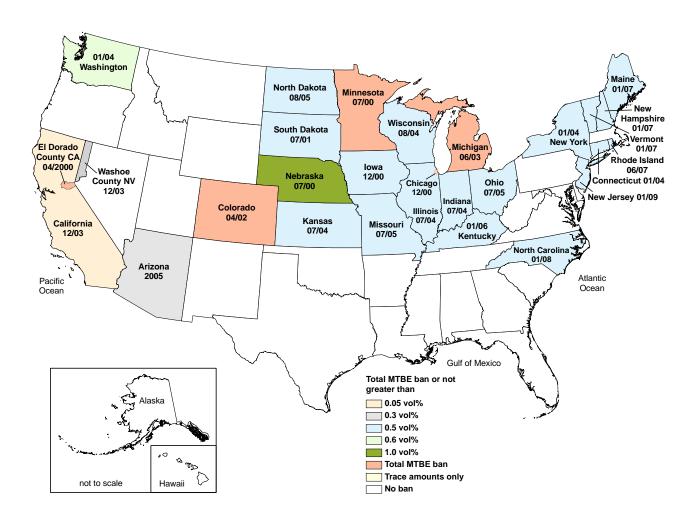


Figure 7 State MTBE, ether, and/or alcohol bans, showing effective dates and maximum levels allowed for MTBE. Please refer to Table 9 for full details on other ethers and alcohols.

Table 9 MTBE and other oxygenate ban dates, maximum allowable limits, and citation.

State/County/City	Ban Dates	mei oxygenau	de minin						Citations
	Enacted	Effective	MTBE	ETBE	TAME	DIPE	TBA	Others	
Arizona	05/11/04	01/01/05	0.3	*	*	*	*	*	Arizona (2004)
		12/31/03	0.60	**	**	**	**	**	California Air
C-1:C	02/14/02	07/01/04	0.30	**	**	**	**	**	Resources Board
California	03/14/03	12/31/05	0.15						(2003b)
		07/01/07	0.05						
Chicago	12/13/00	12/13/00	none						City of Chicago (2000)
Colorado	09/01/00	04/30/02	none						Colorado (2000)
Connecticut	06/18/03	01/01/04	0.5						Connecticut (2003)
El Dorado County, California	03/28/00	4/28/00	none						El Dorado County (2000)
Illinois	07/24/01	07/24/04	0.5						Illinois (2001)
Indiana	07/01/02	07/23/04	0.5				1		Indiana (2002)
Iowa	07/01/00	12/31/00	0.5						Iowa (2000)
Kansas	07/01/01	07/01/04	0.5						Kansas (2004)
Kentucky	07/15/02	01/01/06	0.5						Kentucky (2002)
Maine	07/30/05	01/01/07	0.5						Maine (2005)
Michigan	06/26/00	06/01/03	☆						Michigan (2000)
Minnesota	04/20/00	07/01/00	0.33	0.33	0.33				Minnesota (2000)
TVIIIIICSOtta	04/20/00	07/01/05	none	none	none				
Missouri	08/28/02	07/01/05	0.5						Missouri (2002)
Nebraska	04/11/00	07/13/00	1.0						Nebraska (2000)
New Hampshire	05/27/04	01/01/07•	0.5	<b>A</b>	<b>A</b>	<b>A</b>	0.5	0.5	New Hampshire (2004)
New Jersey	08/18/05	01/01/09	0.5						New Jersey (2005)
New York	05/24/00	01/01/04	0.5						New York (2000)
North Carolina	06/21/05	01/01/08	0.5						North Carolina (2005)
North Dakota	03/31/05	08/01/05	0.5						North Dakota (2005)
Ohio	08/28/02	07/01/05	0.5						Ohio (2002)
Rhode Island	07/06/05	06/01/07	0.5	0.5	0.5	0.5	•		Rhode Island (2005)
South Dakota	02/16/00	07/01/00	2.0						South Dakota (2000)
	02/28/01	07/01/01	0.5						
Vermont	05/23/05	01/01/07	0.5	0.5	0.5	0.5			Vermont (2005)
Washington	05/09/01	01/01/04	0.6						Washington (2001)
Washoe County,	10/25/00	12/31/03 10/1–1/31	0.30						Washoe County District Board of Health (2005)
Nevada	Revised 09/22/05	09/22/05 Yr round	0.30						(2000)
Wisconsin	08/11/03	08/01/04	0.5			1	1		Wisconsin (2003)

<sup>\*</sup>Sum of diisopropylether (DIPE), ethyl *tert*-butylether (ETBE), isobutanol, isopropanol, methanol, n-butanol, n-propanol, *sec*-butanol, *tert*-amylmethylether (TAME), *tert*-butanol, *tert*-pentanol (*tert*-amylalcohol) not to exceed 0.1% by volume, starting 01/01/2006

<sup>\*\*</sup>Sum of methanol, isopropanol, n-propanol, n-butanol, isobutanol, sec-butanol, tert-butanol tert-pentanol (tert-amylalcohol), ethyl tert-butylether (ETBE), diisopropylether (DIPE), tert-amylmethylether (TAME) not to exceed 0.10 weight % oxygen, starting 12/31/2003, sum of same not to exceed 0.06 weight % oxygen, starting 07/01/2004

<sup>•&</sup>quot;"...within 12 months after the Department of Transportation has certified that the State of Montana has produced 40 million gallons of denatured ethanol and has maintained that level of production on an annualized basis for at least 3 months...ensure that all gasoline sold to consumers ....may not contain more than trace amounts of the additive methyl tertiary butyl ether." Montana Code Annotated 2005 82-15-121

<sup>•&</sup>quot;...shall take effect on the later of 01/01/07 or 6 months after federal approval" to opt out of the Federal Reformulated Gasoline Program (see paragraph II, Section 1 of Act 175-4)

<sup>▲ &</sup>quot;No person...shall sell...any neat gasoline ethers or gasoline containing MTBE, other gasoline ethers or tertiary butyl alcohol (TBA) in quantities greater than ½ of 1% by volume"

<sup>■</sup> Other ether oxygenates not greater than 0.5%: methanol, isopropanol, n-propanol, n-butanol, sec-butanol, tert-butanol, tert-pentanol (tert amylalcohol) isobutanol

#### **Discussion and Conclusions**

The Clean Air Act Amendments of 1990 imposed a set of requirements for gasoline in the United States. For LUST sites, the three most important are:

#### 1) Final Ban of Leaded Gasoline

Banning leaded gasoline had two major effects on LUST sites. First, removing lead resulted in new ways to boost the octane rating of gasoline. Most significant was the introduction of MTBE and other ethers, beginning in 1979; notably, this predates oxygenate requirements of the Clean Air Act Amendments. Second was the concurrent removal of lead scavengers. These compounds – ethylene dibromide and 1,2 dichlorethane – prevent precipitation of lead on engine parts. They recently were shown to persist in ground water at some sites (Falta, 2004, Falta et al., 2005), and continue to cause contamination problems.

#### 2) Conventional and Reformulated Gasoline

The Clean Air Act Amendments of 1990 created a framework for the two major types of gasoline sold in the U.S., reformulated and conventional. With a few exceptions, use of RFG has been on a county-by-county basis. Although most of these locations have used RFG for long periods, some changes have occurred as indicated on Figure 5.

The composition of RFG is specified by provisions of the Clean Air Act. Relevant for leaking underground storage tanks are provisions that limit benzene to a nominal level of 1.0% by volume and, until mid-spring of 2006, required an oxygenated additive at 2.0% by weight. Conventional gasoline is gasoline which is not reformulated -- the remaining gasoline in the U.S. The benzene content in conventional gasoline is limited by producer/importer baselines set in 1990. Because the baselines are not tied to point-of-use, they do not provide a guide for specific locations. Another complicating factor is the use of oxygenates as octane

enhancers. These could be used anywhere and evidence of their preferred use in premium gasoline appears in a companion study.

#### 3) Oxygenated Gasoline Program

The Clean Air Act Amendments of 1990 mandated states to require oxygenated gasoline in areas that did not attain carbon monoxide standards in the winter months. Beginning in 1992, these requirements were imposed in selected cities, but as carbon monoxide standards were met, cities gradually dropped out of the program.

The CAAA of 1990 indirectly led to a fourth impact on gasoline composition, which occurred because of the states' reaction to ground water contamination by MTBE:

#### 4) State MTBE and Other Oxygenate Bans

Beginning in 2000, state legislatures enacted bans or restrictions on oxygenate usage in gasoline. In some cases these included only MTBE, while in others a whole suite of ethers and alcohols were restricted. Most restrictions allowed MTBE content of up to 0.5% by volume. There continues to be sporadic production of small amounts of RFG with ether, which could be used in states without ether bans.

Despite changes caused by the states' oxygenate limitations and by removal of the oxygenate mandate for reformulated gasoline, the framework for reformulated and conventional gasoline is still in place in the United States. State oxygenated gasoline programs, mostly in the southwestern United States, continue to rely on ethanol to meet their oxygenate requirements. Ethanol usage has increased steadily so that 77% of U.S. gasoline now contains ethanol. Benzene concentrations in RFG were limited to 1 vol %, and will be further reduced to 0.62 vol % in all U.S. gasoline by 2011.

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