

## ABSTRACT

Gasoline composition in the U.S. varies due to market, technical and regulatory factors. Technical factors include seasonal and elevation adjustments for drivability, and needed anti-knock properties. Regulatory factors derive from the Clean Air Act and its amendments. The Clean Air Act Amendments (CAAA) of 1990 created two major types of gasoline in the U.S.: reformulated and conventional gasoline. Since the implementation of this Act, conventional gasoline has tended to be approximately two-thirds of U.S. gasoline and is used in rural areas and smaller cities. Major urban areas are required to use reformulated gasoline (RFG) to meet ozone and carbon monoxide standards and reduce toxic air pollutants. In addition, the CAAA specified that oxygenated gasoline could be used in certain cities to meet carbon monoxide standards in winter. Given the combination of technical and regulatory requirements, and that they have varied over time, the best way to understand the composition of gasoline at any specific location is to evaluate historical data. In this report, three historical datasets were used to trace the historical composition of benzene, oxygenates and alcohols in 15 cities or RFG compliance areas. For the RFG cities, the benzene content dropped when the CAAA mandates went into effect. The median benzene content was consistently less than 1% after 1995. Typically, methyl tert-butyl ether (MTBE) was used to meet the requirement for an oxygenated additive until, either a state ban or other use was imposed or until MTBE was removed from the fuel supply in 2006. Ethanol use typically replaced MTBE to meet continuing requirements of RFG. In conventional gasoline, benzene was not uniformly limited, but producer baselines were developed. Benzene levels typically decreased over time. When the Mobile Sources Air Toxics Act requirements were imposed in 2011, all U.S. gasoline was seen to have reduced benzene levels. The ether and alcohol content of conventional gasoline varies widely, and is best assessed through historical data. This follows because both ethers and alcohols can be used as octane boosters and they could be used for a variety of market reasons. Oxygenated gasoline, which can be conventional or reformulated, shows the benzene characteristics of its type. The ether and alcohol levels vary seasonally prior to the imposition of national requirements for biofuel usage in 2006 and later years.

## OUTLINE OF GASOLINE REQUIREMENTS

Gasoline is designed to meet performance specifications and regulations based on the Clean Air Act.

- *Clean Air Act, established the framework for setting nation-wide air quality goals (NAAQS) for six pollutants.*
- *Subsequently, states developed implementation plans (SIPs) to meet those goals.*
- *MTBE was registered for use in gasoline in 1979, tert-amyl methyl ether (TAME) in 1981, and ethyl tert-butyl ether (ETBE) in 1981 (Stickers, 2002).*
- *Under authority granted by the Clean Air Act, EPA phased out leaded gasoline over a period of time that ended on January 1, 1996.*

Clean Air Act Amendments (1990) 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 Leaking Underground Storage Tanks (LUST) sites:*
  - *Total ban on lead in gasoline*
  - *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.*

Parts of the country not using RFG were also affected by CAAA under an anti-dumping provision to prevent air quality deterioration in areas using conventional gasoline (CG).

- *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: CG baseline limitations are applied to producers/importers, while RFG requirements apply to where the fuel is used.*
- *Historically, at a given location the benzene concentration in CG was usually variable and not very predictable.*
- *In 2011 the Mobile Sources Air Toxics rule (U.S. Federal Register, 2007) reduced benzene levels in all gasoline to an average of 0.62%.*

## INTRODUCTION

Gasoline composition has changed throughout the period of extensive automobile use to balance the needs of ever more powerful engines with environmental concerns. Thus, there has been a progression of additives and gasoline components ("blending components") that have contributed to mechanical needs but may have generated environmental concerns. The most well-known of these are the aromatic and BTEX<sup>1</sup> fractions, organic lead, methyl tert-butyl ether (MTBE) and other ethers, and alcohols. The impact of a given type of gasoline depends upon several factors, not the least of which is the composition. Is a certain component present above a threshold or present at all? If gasoline composition didn't vary due to technical factors—season of the year, elevation—refinery capacity, and oil supply, and the varying regulations by the U.S. Environmental Protection Agency (EPA) and the states, the answer to this question could be answered in a straightforward way. Under the framework of the Clean Air Act of 1970 and its amendments, the states have also imposed requirements on gasoline composition. These have varied by location and time. So, to answer the question of the composition of leaked gasoline, historical data provide the best source of information. The purpose of this report is to better understand gasoline composition through reviewing requirements for gasoline composition by various regulatory programs and through presenting historical data illustrating the variation in benzene, MTBE and ethanol for cities across the U.S.

In response to concerns 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.*
- *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., Stickers, 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), and as seen in the results at right.*

## 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 (Stickers, 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.

After declining through the 1980s, complete removal of lead, as mandated by the Clean Air Act Amendments of 1990, occurred on January 1, 1996. Lead can still be used, however, in aviation gasoline and racing fuel.

## REFORMULATED AND CONVENTIONAL GASOLINE

EPA definition: Gasoline that is certified to meet requirements and standards specified in U.S. CFR, Title 40, Part 80, Section 42.

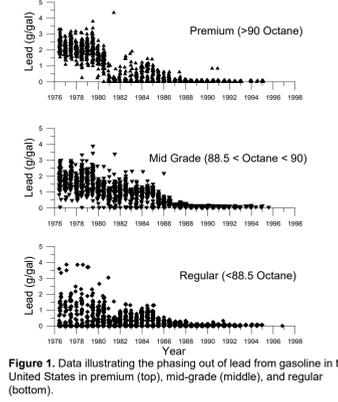
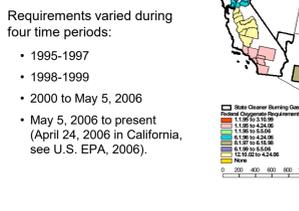


Figure 1. Data illustrating the phasing out of lead from gasoline in the United States in premium (top), mid-grade (middle), and regular (bottom).

Requirements varied during four time periods:

- 1995-1997
- 1998-1999
- 2000 to May 5, 2006
- May 5, 2006 to present (April 24, 2006 in California, see U.S. EPA, 2006).

Although other requirements of RFG changed over these times, required oxygen content and benzene limitation did not change until the Energy Policy Act passed in 2005.

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 was reduced to 0.62 vol % to comply with the Mobile Sources Air Toxics Rule (U.S. Federal Register, 2007). Three states, Arizona, California and Nevada, have implemented cleaner burning gasoline (CBG) programs. Figure 2 shows the areas of the U.S. that at some time have used RFG and, in the legend, the duration of oxygenate use.

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 %. 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)(1)).

## Conventional Gasoline

Conventional gasoline: gasoline that has not been certified as RFG (i.e., meeting the U.S. CFR, 2007, Title 40, Part 80, Section 91(c)(5) and Section 45, (b)(2)). 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).

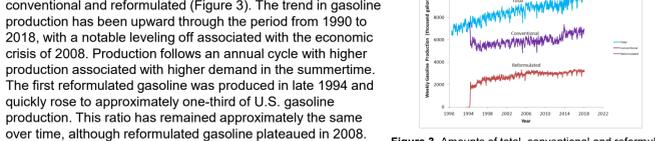


Figure 3. Amounts of total, conventional and reformulated gasoline produced per week in the U.S. (EIA, 2018).

## OXYGENATED GASOLINE

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.
- 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% oxygen by weight was required.
- Program began in late fall 1992 in 39 areas, with one addition in 1993.
- Six cities began winter oxygenate programs earlier (1989/1990 winter season)
  - Denver, Colorado
  - Reno and Las Vegas, Nevada
  - Tucson and Phoenix, Arizona
  - Albuquerque, New Mexico (Stickers, 2001)
- Detailed lists of the exit dates for the 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.

## ETHANOL MANDATES

- Five states have continuing state-wide oxygenate mandates (Weaver, 2018).
- Federal Renewable Fuel Standard (RFS).
  - Created by the Energy Policy Act of 2005.
  - Extended by the Energy Independence and Security Act of 2007.
  - Requires the replacement of petroleum fuel by renewable fuels which can be conventional, advanced or cellulosic.
- Program established renewable fuel targets for each year until 2020.
- Current target for 2020 is 36 billion gallons (EPA, 2018b).
- For conventional vehicles, ethanol is approved at concentration of up to 10%. Because the refining of gasoline produces a petroleum product that is intended to be used with 10% ethanol, ethanol is typically seen in gasoline at 10% by volume.
- By mid-2018, 90% of gasoline in the U.S. was formulated with ethanol (Figure 5).
- Although the majority of U.S. gasoline is formulated for use with 10% ethanol, there are places where non-ethanol gasoline is available (Figure 6).
  - These are located in states without ethanol mandates, such as Oklahoma.

## MTBE BANS

- Beginning in 2000, twenty-seven states, two counties, and one city banned MTBE or other oxygenates in gasoline (Figure 7).
- Three states have imposed absolute bans, while most allow some MTBE to remain in gas (Weaver et al. 2010).
- In 17 states, the level was 0.5%.
- Others ranged from 0.05% in California to 1% in Nebraska.
- Six states banned MTBE and other oxygenates which included the other gasoline oxygenate ethers<sup>2</sup>.
- Bans did not affect requirements to use reformulated or oxygenated gasoline, but did cause a shift to another oxygenate.
- Data presented at right show that the primary substitute was ethanol.
- Dates and specific requirements for each MTBE-banning state are given by Weaver et al. (2010).

<sup>2</sup>ethyl tert-butyl ether (ETBE), tert-amyl methyl ether (TAME), diisopropyl ether (DIFE), and alcohols, mostly tert-butyl alcohol (TBA)

## DATA AND METHODS

Data were obtained from three surveys:

- National Institute of Petroleum and Energy Research (NIPER) and its successors (e.g., American Petroleum Institute [API], Northrup-Grumman)
- Alliance of Automobile Manufacturers (Alliance)
- U.S. EPA Office of Transportation and Air Quality (OTAQ) in collaboration with industry (i.e., API, National Petroleum Refiners Association).

Details on these sources and the methods for combining them are given in Weaver (2018).

## RESULTS

### Reformulated Gasoline

Data from NIPER and OTAQ patterns in reformulated gasoline. Houston-Galveston and NY-NJ-CT areas used as examples. Pre-1995 data drawn from NIPER data (dashed lines), OTAQ data (solid lines) extend the time range through 2017. Data are presented as minimum, median, and maximum values in each survey. Median values establish general trends. Minimum and maximum values define the limits of what was observed, and represent single values. They were chosen to represent the single-valued extremes of gasoline that could have been leaked.

Throughout the 1980s and 1990s, median concentration of benzene in Houston-Galveston gasoline ranged from 0.67 to 1.67% (Figure 8). Median concentrations above 1% do not appear after 1995, due to the Clean Air Act Amendments. Because the requirements can be met by average, rather than each individual sample, there were samples with higher concentration, including some with concentration of about 5.5%. NY-NJ-CT area gasoline followed a similar pattern with the median benzene concentration dropping and staying below 1% after the implementation of the RFG program in 1995 (Figure 9). After the imposition of the MSAT benzene requirement of <0.62% in 2011, median benzene in Houston-Galveston was less than 0.535% and that of NY-NJ-CT less than <0.73%.

Prior to the EPAAct removal of the oxygenate mandate from RFG, MTBE was the dominant oxygenate in Houston-Galveston area gasoline (Figure 10). Median concentration of MTBE was ~11% (as required) prior to 2006. Because other oxygenates were sometimes used, the total oxygenate content may have been provided by other compounds (not shown). NIPER data (dashed lines) show usage of MTBE prior to the 1995 start of RFG program, as MTBE, in addition to the later regulatory mandate, serves as an octane enhancer (Stickers, 2002). In 2006 MTBE was removed from the gasoline supply. After that time, ethanol was used to boost octane and help meet requirements of RFG, although a specific amount of oxygenate was not required. The EPAAct did, however, require use of biofuels at levels up to 10%.

NY-NJ-CT area gasoline shows a more complex picture in two ways.

- 1) This area participated in the oxygenated gasoline program from its inception in 1992 until 1999 (NJ) and 2000 (NY and CT). Under the oxygenated gasoline program, the oxygenate was required in the winter and not the summer, hence the data show fluctuating MTBE concentration from 1992 to 1995 (Figure 11). From 1995 to 2000, the median MTBE is around 11%. Some oxygenate was supplied by other ethers during this time (not shown).
- 2) New York and Connecticut banned MTBE in 2004 while New Jersey did not until 2009. Thus, in this data set MTBE use is seen to continue from the date of the New York and Connecticut bans in 2004 until it was removed from the fuel supply in 2006. New Jersey samples show MTBE levels similar to the New York and Connecticut samples until ethanol use (Figure 11). This is one of the few examples of a data set showing use of both MTBE and ethanol as an artifact of the sample area, rather than an indication of simultaneous use of MTBE and ethanol. Detailed characterization studies show that the two are not used together in individual gasolines (Weaver et al. 2005).

### California Reformulated Gasoline

Los Angeles gasoline data are contained in the NIPER and Alliance data sets, and is used here as an example of California reformulated gasoline. Because California does not participate in the RFG surveys, OTAQ data does not cover Los Angeles or California. Los Angeles showed higher median benzene than New York or Houston (Figure 12), prior to the imposition of RFG requirements in 1995, which reduced the median benzene to below 1%. The Alliance stopped including benzene data after 2013, so later data are not available. Certain counties in California (including the Los Angeles area) are subject to Federal RFG requirements and the state cleaner burning gasoline program. Los Angeles, as in New York, has been in the oxygenated gasoline program from 1992 to present. Thus in the period from 1992 to 1995, the median MTBE content fluctuated seasonally (dashed red line on Figure 13). California banned MTBE in 2003 and MTBE disappeared from Los Angeles gasoline. Ethanol was in use in this gasoline at median concentration of around 5.5% from 2003 to 2010, and at median concentration of 10% thereafter.

### Conventional Gasoline

Conventional gasoline, which is by definition not reformulated, did not have an across-the-board benzene limitation prior to the implementation of the MSAT in 2011. Producer baselines determined the allowable benzene from each refinery. The benzene in use at a specified city could include products from several refineries, each with a different baseline. Cities like Atlanta and Seattle (Figures 14 and 15), had varying levels of benzene that have generally decreased over time. Seattle and the Pacific Northwest in general had higher levels of benzene than the rest of the U.S. In each of these cities the benzene levels were at or below 0.6% from 2011 to the end of the available data in 2013. Oxygenated additives were not required in conventional gasoline and the use of MTBE (and other oxygenates) varied greatly, but typically at lower levels than mandated for RFG or OG (Figures 16 and 17). Atlanta showed consistent usage through 2005, and Seattle's usage was more sporadic. Ethanol came into consistent use at about 10% by volume in these cities after the EPAAct mandated biofuel use on a nationwide basis.

### Oxygenated Gasoline

Phoenix has participated in the oxygenated gasoline program and requires oxygenated gasoline from November through the end of March. As a conventional gasoline city, benzene content followed a similar pattern as Atlanta and Seattle—showing a decrease over time and reduction to a median of 0.4% after the imposition of the MSAT requirements (Figure 18). Both ethanol and MTBE were in use prior to 2005, and these fluctuated seasonally (Figure 19). When only median values are shown (Figure 20), it can be seen that these were used separately: ethanol in the winter and MTBE in the summer. After 2010 ethanol use became constant throughout the year at a level around 10%.

To assess gasoline composition in locations around the U.S., the data were compiled for:

| Conventional Gasoline Cities                            | Federal RFG Areas  | Cal-RFG     |
|---|--|-------------|
| Atlanta, GA<br>Denver, CO<br>Phoenix, AZ<br>Seattle, WA | NY-NJ-Long Is.-CT<br>Baltimore, MD<br>Boston-Worcester, MA<br>Chicago-Lake Co.-IL, Gary, IN<br>Dallas-Fort Worth, TX<br>Houston-Galveston, TX<br>Philadelphia, PA-Wilmington, DE-Trenton, NJ<br>Portland-Dover, NH<br>St. Louis, MO<br>Washington, D.C.-area | Los Angeles |

- CG cities cover the east and west, high-elevation, and winter-oxygenate cities.
- Seattle is included as the Pacific Northwest was known to have historic elevated benzene levels.
- Each of the surveys use standard ASTM methods for gasoline analysis which facilitates the combination of data sets.
- NIPER and Alliance data were used because OTAQ does not supply CG compliance data to the public.
- Most of the major east-coast federal RFG cities (Baltimore, Boston, New York, Philadelphia, Washington) were included as were the midwestern and Texas cities included in the program (Chicago, St. Louis, Dallas, and Houston).
- NIPER and OTAQ data were combined for these cities.
- Los Angeles was the sole Cal-RFG city included in the study because it had the only complete data set (NIPER and Alliance).
- California areas are not included in the Federal RFG compliance surveys, so OTAQ data were not available.
- Either conventional or reformulated gasoline could be oxygenated.
  - NY-NJ-Long Is.-CT (RFG) and Phoenix (CG) are examples noted below.
- Full set of results is available in Weaver (2018).

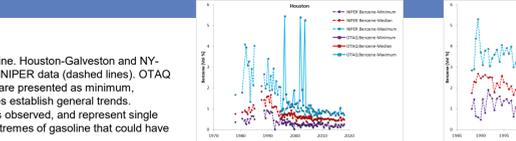


Figure 8. Benzene content in RFG from Houston and Galveston, Texas (Alliance data: North American Auto Alliance, 2018).

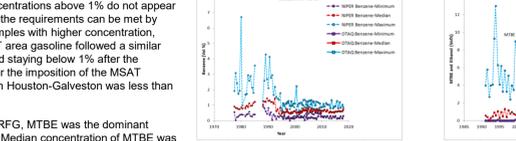


Figure 9. Benzene content in RFG from the New York City area, including New York, New Jersey, and Connecticut (Alliance data: North American Auto Alliance, 2018).

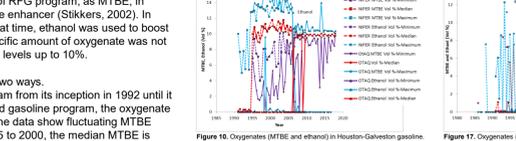


Figure 10. Oxygenates (MTBE and ethanol) in Houston-Galveston gasoline (Alliance data: North American Auto Alliance, 2018).

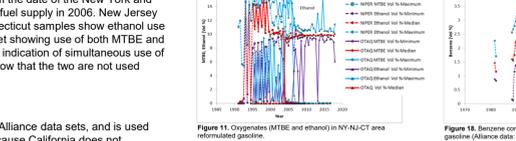


Figure 11. Oxygenates (MTBE and ethanol) in NY-NJ-CT area reformulated gasoline (Alliance data: North American Auto Alliance, 2018).

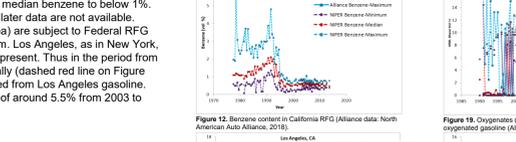


Figure 12. Benzene content in Phoenix, AZ conventional/oxygenated gasoline (Alliance data: North American Auto Alliance, 2018).

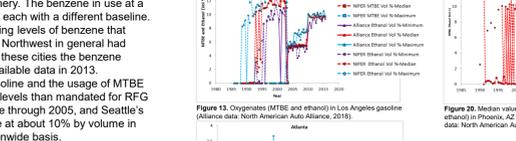


Figure 13. Oxygenates (MTBE and ethanol) in Phoenix, AZ conventional/oxygenated gasoline (Alliance data: North American Auto Alliance, 2018).

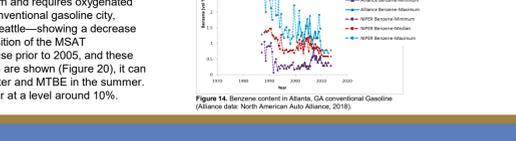


Figure 14. Benzene content in Atlanta, GA conventional Gasoline (Alliance data: North American Auto Alliance, 2018).

## CONCLUSIONS

Benzene, MTBE and ethanol content of gasoline across the U.S. is determined by a number of factors, including technical, market, and regulatory factors. For reformulated gasoline, the benzene level was explicitly capped at 1% in 1995. The median concentrations were then all maintained below 1%. Variability exists however, and both lower and higher values were found in the data. The benzene level in conventional gasoline has also been regulated at the refinery (or blender) since the Clean Air Act Amendments of 1990 went into effect. Because a city may receive gasoline from different refiners at various times, the producer baselines do not provide a prediction of the benzene content at a given retail location. In contrast the MSAT requirement for all benzene to be <0.62% is one of those time points where the benzene content is specified. As for reformulated gasoline, there is much variability in samples and historical data for individual locations is needed.

Oxygenated additives were required in oxygenated gasoline and reformulated gasoline. These requirements provide definitive specifications for MTBE or ethanol. RFG areas or cities (oxygenated gasoline) entered and left these programs at different times, so again historical data provide specific local knowledge. Even when seemingly straightforward, local complexities come into play. Phoenix is a good example, where MTBE was present in summertime gasoline and ethanol in wintertime gasoline. Another time point was created by the EPAAct, which effectively ended the use of MTBE in U.S. gasoline. As in the case of the NY-NJ-CT data, state MTBE or ether bans caused MTBE use to end sooner in some states than others. Imposition of the renewable fuel standard imposed the requirement to use ethanol at the time MTBE was discontinued. In conventional gasoline cities, however, there was no oxygenate mandated and use of MTBE or ethanol could have occurred prior to the beginning of the RFG program. This follows from the ability of MTBE and ethanol to boost octane ratings.

Regulatory requirements are important drivers of gasoline composition. Technical or market factors impact the use of various components and variability exists in the range of concentrations sampled. Historical data combined with regulatory knowledge provides the best way of understanding the composition of gasoline that might be leaked at a leaking underground storage tank site.

## ACKNOWLEDGEMENTS

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