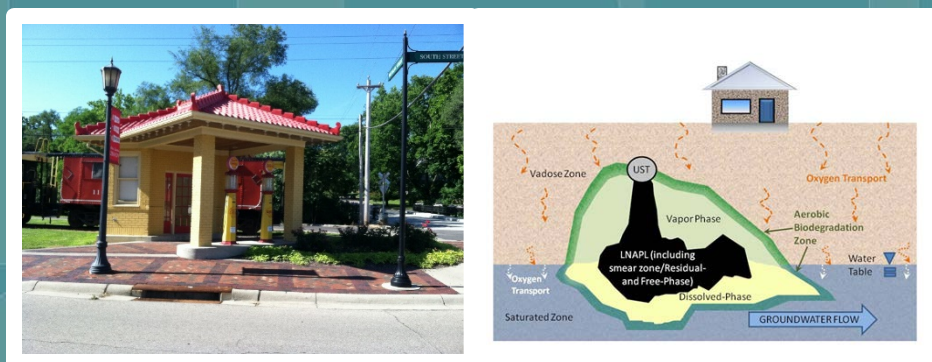


Gasoline Composition in the U.S. from Three Datasets 1976-2017



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by

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Notice/Disclaimer

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Foreword

The U.S. Environmental Protection Agency (US EPA) is charged by Congress with protecting the Nation's land, air, and water resources. Under a mandate of national environmental laws, the Agency strives to formulate and implement actions leading to a compatible balance between human activities and the ability of natural systems to support and nurture life. To meet this mandate, US EPA's research program is providing data and technical support for solving environmental problems today and building a science knowledge base necessary to manage our ecological resources wisely, understand how pollutants affect our health, and prevent or reduce environmental risks in the future.

The National Risk Management Research Laboratory (NRMRL) within the Office of Research and Development (ORD) is the Agency's center for investigation of technological and management approaches for preventing and reducing risks from pollution that threaten human health and the environment. The focus of the Laboratory's research program is on methods and their cost-effectiveness for prevention and control of pollution to air, land, water, and subsurface resources; protection of water quality in public water systems; remediation of contaminated sites, sediments and ground water; prevention and control of indoor air pollution; and restoration of ecosystems. NRMRL collaborates with both public and private sector partners to foster technologies that reduce the cost of compliance and to anticipate emerging problems. NRMRL's research provides solutions to environmental problems by: developing and promoting technologies that protect and improve the environment; advancing scientific and engineering information to support regulatory and policy decisions; and providing the technical support and information transfer to ensure implementation of environmental regulations and strategies at the national, state, and community levels.

This report discusses and summarizes regulations that impact gasoline composition, as an introduction to a review of major components in historical gasoline samples from major cities and regions in the United States.

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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 of ether 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.

Acknowledgements

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Table of Contents

| | |
|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----|
| Notice/Disclaimer | ii |
| Foreword..... | iii |
| Abstract..... | iv |
| Acknowledgements..... | v |
| List of Figures | vii |
| List of Tables | ix |
| Acronyms and Abbreviations..... | x |
| 1.0 Introduction | 11 |
| 2.0 Outline of Gasoline Requirements..... | 11 |
| 3.0 Leaded Gasoline | 13 |
| 4.0 Reformulated and Conventional Gasoline | 14 |
| 4.1 Reformulated Gasoline | 14 |
| 4.2 Conventional Gasoline | 17 |
| 5.0 Oxygenated Gasoline | 19 |
| 6.0 Ethanol Mandates..... | 23 |
| 7.0 MTBE Bans | 24 |
| 8.0 Data and Methods..... | 26 |
| 9.0 Data Combination | 27 |
| 10.0 Results..... | 27 |
| 10.1 Reformulated Gasoline | 28 |
| 10.2 California Reformulated Gasoline..... | 32 |
| 10.3 Conventional Gasoline | 34 |
| 10.4 Oxygenated Gasoline | 38 |
| 11.0 Conclusions | 42 |
| References | 43 |
| Appendices..... | 48 |
| Appendix A..... | 48 |
| Conventional Gasoline Cities not appearing in the main text: Denver, Colorado | 48 |
| Appendix B | 50 |
| Reformulated gasoline areas not appearing in the main text: Baltimore, MD; Boston-Worcester, MA, Chicago-Lake Co, IL –Gary, IN; Dallas-Fort Worth, TX; Philadelphia, PA—Wilmington, DE—Trenton, NJ; Portsmouth-Dover, NH; St. Louis, MO; Washington DC area. | 50 |

List of Figures

| | |
|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| 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. | 14 |
| Figure 2: 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. CFR, 2007, 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. | 18 |
| Figure 3: Amounts of total, conventional and reformulated gasoline produced per week in the U.S. (EIA, 2018). | 19 |
| Figure 4: 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. | 22 |
| Figure 5: Total weekly gasoline production and the total containing ethanol. (data from EIA, 2018). | 23 |
| Figure 6: Advertisements at a gasoline station in Oklahoma City, July, 2018, advertising the availability of ethanol-free gasoline and gasoline with 10% ethanol. | 24 |
| 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 alcohol. | 25 |
| Figure 8: Benzene content in RFG from Houston and Galveston, Texas. | 29 |
| Figure 9: Benzene content in RFG from the New York City area, including New York, New Jersey, and Connecticut. | 30 |
| Figure 10: Oxygenates (MTBE and ethanol) in Houston-Galveston gasoline. | 31 |
| Figure 11: Oxygenates (MTBE and ethanol) in NY-NY-CT area reformulated gasoline. | 32 |
| Figure 12: Benzene content in California RFG (Alliance data: North American Auto Alliance, 2018). | 33 |
| Figure 13: Oxygenates (MTBE and ethanol) in Los Angeles gasoline (Alliance data: North American Auto Alliance, 2018). | 34 |
| Figure 14: Benzene content in Atlanta, GA conventional Gasoline (Alliance data: North American Auto Alliance, 2018). | 35 |
| Figure 15: Benzene content in Seattle, WA conventional Gasoline (Alliance data: North American Auto Alliance, 2018). | 36 |
| Figure 16: Oxygenates (MTBE and ethanol) in Atlanta, GA conventional gasoline (Alliance data: North American Auto Alliance, 2018). | 37 |
| Figure 17: Oxygenates (MTBE and ethanol) in Seattle, WA conventional gasoline (Alliance data: North American Auto Alliance, 2018). | 38 |
| Figure 18: Benzene content in Phoenix, AZ conventional/oxygenated gasoline (Alliance data: North American Auto Alliance, 2018). | 39 |
| Figure 19: Oxygenates (MTBE and ethanol) in Phoenix, AZ conventional/oxygenated gasoline (Alliance data: North American Auto Alliance, 2018). | 40 |
| Figure 20: Median values of oxygenate concentration (MTBE and ethanol) in Phoenix, AZ conventional/oxygenated gasoline (Alliance data: North American Auto Alliance, 2018). ... | 41 |
| Figure 21: Benzene content in conventional gasoline from Denver, Colorado. | 48 |
| Figure 22: Oxygenates (MTBE and ethanol) in Denver, Colorado conventional gasoline. | 49 |
| Figure 23: Benzene content in reformulated gasoline from the Baltimore, Maryland area. | 50 |
| Figure 24: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Baltimore, | |

| | |
|-------------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| Maryland area..... | 51 |
| Figure 25: Benzene content in reformulated gasoline from the Boston-Worcester, Massachusetts area..... | 52 |
| Figure 26: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Boston- Worcester, Massachusetts..... | 53 |
| Figure 27: Benzene content in reformulated gasoline from the Chicago-Lake Co, Gary Indiana area..... | 54 |
| Figure 28: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Chicago Lake Co—Gary Indiana area..... | 55 |
| Figure 29: Benzene content in reformulated gasoline from the Dallas-Ft Worth, Texas area..... | 56 |
| Figure 30: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Dallas—Ft Worth, Texas area..... | 57 |
| Figure 31: Benzene content in reformulated gasoline from the Philadelphia, Pennsylvania— Wilmington, Delaware—Trenton, New Jersey area..... | 58 |
| Figure 32: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Philadelphia, Pennsylvania—Wilmington, Delaware—Trenton, New Jersey area..... | 59 |
| Figure 33: Benzene content in reformulated gasoline from the Portsmouth—Dover, New Hampshire area. Only data from OTAQ were available..... | 60 |
| Figure 34: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Portsmouth Dover, New Hampshire area. Only data from OTAQ were available..... | 61 |
| Figure 35: Benzene content in reformulated gasoline from the St Louis, Missouri area..... | 62 |
| Figure 36: Oxygenates (MTBE and ethanol) in reformulated gasoline from the St. Louis Missouri area..... | 63 |
| Figure 37: Benzene content in reformulated gasoline from the Washington DC area..... | 64 |
| Figure 38: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Washington DC area..... | 65 |

List of Tables

Table 1 Oxygen and benzene requirements for reformulated gasoline.....15

Table 2 Oxygen and required amounts of oxygenates to meet requirements of reformulated and winter oxygenated gasoline. Values given as weight percent.16

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).16

Table 4 Selected parameters of the statutory baseline for conventional gasoline (40 CFR 80.91 and 40 CFR 80.45).17

Table 5 Cities currently implementing the winter oxygenates program (U.S EPA, 2008c).20

Table 6 State ethanol mandates.....21

Acronyms and Abbreviations

| | |
|---------|-----------------------------------------------------|
| API | American Petroleum Institute |
| Cal-RFG | California Reformulate Gasoline |
| CAAA | Clean Air Act Amendments |
| CG | Conventional Gasoline |
| EPA | Environmental Protection Agency |
| EPAct | Energy Policy Act of 2005 |
| EISA | Energy Independence and Security Act of 2007 |
| MTBE | Methyl tert-butyl ether |
| NIPER | National Institute of Petroleum and Energy Research |
| OG | Oxygenated Gasoline |
| OTAQ | Office of Transportation and Air Quality |
| RFG | Reformulated Gasoline |

1.0 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 BTEX¹ fractions, organic lead, methyl tert-butyl ether (MTBE) and other ethers, and alcohols. The impact of a given type of gasoline depends on several factors, not the least 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.

2.0 Outline of Gasoline Requirements

Gasoline is designed to meet performance specifications and regulations based on the Clean Air Act. The 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. To date, 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 (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. 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 Leaking Underground Storage Tanks (LUST) sites were the total ban on lead in gasoline, and new requirements for

¹ BTEX is benzene, toluene, ethylbenzene and xylenes

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. 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%.

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 (EPA 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., 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 the results below.

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 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.

3.0 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. 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.

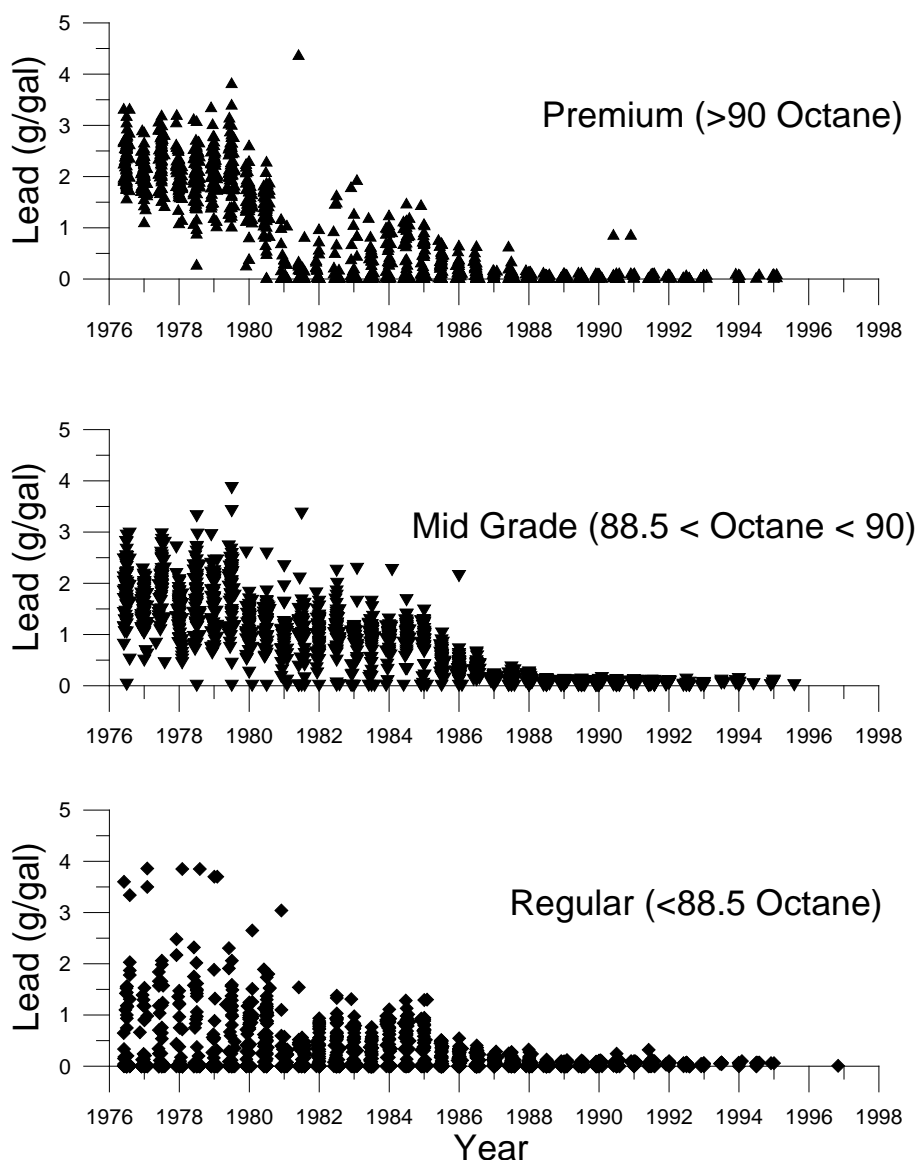


Figure 1: Data illustrating the phasing out of lead from gasoline in the United States in premium (top), mid-grade (middle), and regular (bottom). After declining through the 1980s, lead was completely eliminated by January 1, 1996 by the Clean Air Act Amendments of 1990.

4.0 Reformulated and Conventional Gasoline

4.1 Reformulated Gasoline

EPA defines reformulated gasoline as gasoline that is certified 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 was 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 | Effective Dates | RFG Content Requirements | | |
|---------------------------|------------------------|--------------------------|-------------------------|--------|
| | | Per-gallon Basis | Averaged 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 through 2010 | ≤ 1.00 | ≤ 0.95 | ≤ 1.30 |

*After Jan 1, 201 all gasoline must meet an annual average benzene content standard of 0.65% (Vol) and after Jan 1, 2012 a maximum average benzene standard of 1.3 vol % must be met.

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.

| Weight percent of oxygen | Per-gallon Basis | Required Oxygenate Concentration | | | |
|---------------------------------------------------------------------------|------------------|----------------------------------|---------|------------------|-------------|
| | | RFG | | Winter Oxygenate | |
| | | Averaged Basis | | 2.7% | 3.1% oxygen |
| | | Standard | Minimum | oxygen required | required |
| | 2.0 % | 2.1% | 1.5% | | |
| Common Oxygenates and Required Content to Meet Oxygen Requirements | | | | | |
| Methyl <i>tert</i> -butyl ether | 11.0% | 11.6% | 8.25% | 14.9% | 17.1% |
| Ethyl <i>tert</i> -butyl ether | 12.75% | 13.4% | 9.6% | 17.2% | 19.8% |
| <i>Tert</i> -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).

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

4.2 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 40CFR 80.91(c)(5)(i) 40CFR 80.45(b)(2) | Summer 40CFR 80.91(c)(5)(ii) 40CFR 80.45(b)(2) | Average 40CFR 80.91(c)(5)(ii) |
|------------------------------------|-----------------------------------------------------|------------------------------------------------------|----------------------------------|
| Benzene (vol %) | 1.64 | 1.53 | 1.60 |
| Aromatics (vol %) | 26.4 | 32 | 28.6 |
| Olefins ^(*) (vol %) | 11.9 | 9.2 | 10.8 |
| Reid Vapor Pressure (psi) | 8.7 | 8.7 | 8.7 |
| API Gravity ^(**) (°API) | 60.2 | 57.4 | 59.1 |

(*) Olefins are also known as alkenes

(**) API Gravity (°API) is defined by:

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

A map of locations required to use federal reformulated and conventional gasoline is shown in Figure 2. 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. Detailed exceptions to these dates are given on the figure.

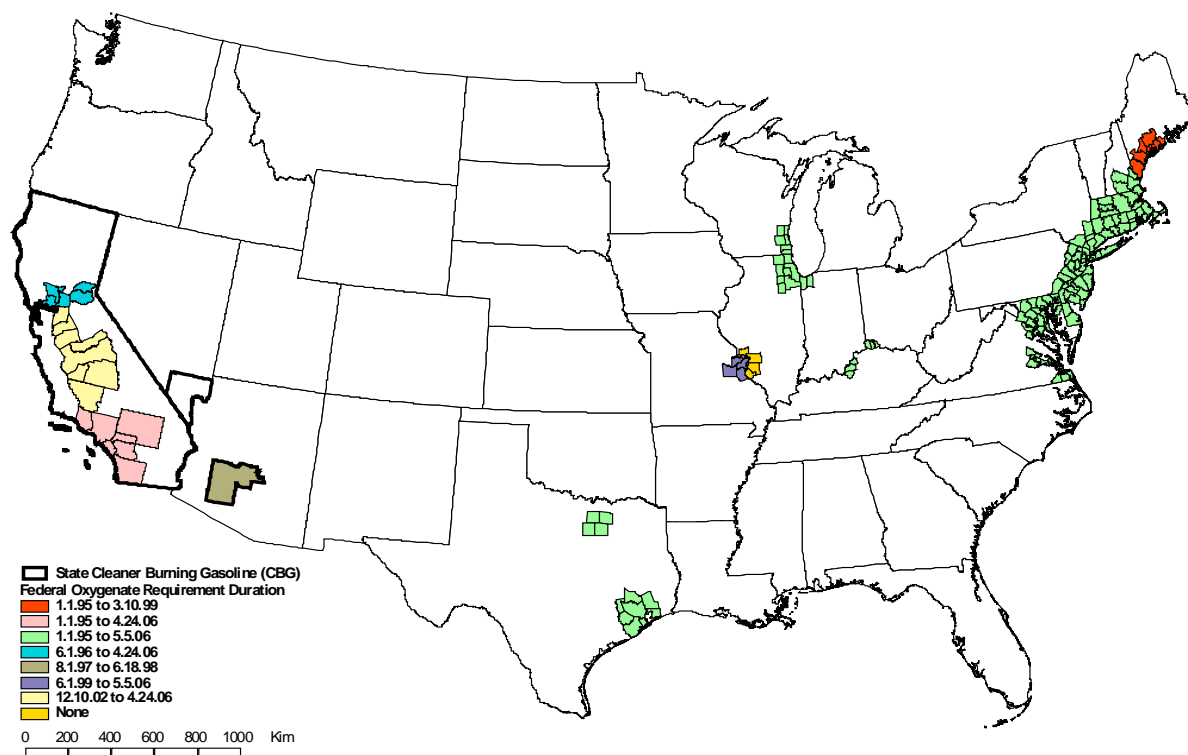


Figure 2: Federal reformulated gasoline and state cleaner-burning gasoline programs in the United States (California, 2003a; Clark County, Nevada, 2003; U.S. CFR, 2005, Title 40, Part 80, Section 40 U.S. CFR, 2007, Title 40, Part 80, Section 40, U.S Federal Register, 2004). Durations of oxygenate mandates 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.

The total gasoline production in the U.S. is thus split between conventional and reformulated (Figure 3). The trend in gasoline production has been upward through the period from 1990 to 2018, with a notable leveling off associated with the economic crisis of 2008. Production follows an annual cycle with higher production associated with higher demand in the summertime. The first reformulated gasoline was produced in late 1994 and quickly rose to approximately one-third of U.S. gasoline production. This ratio has remained approximately the same over time, although reformulated gasoline plateaued in 2008.

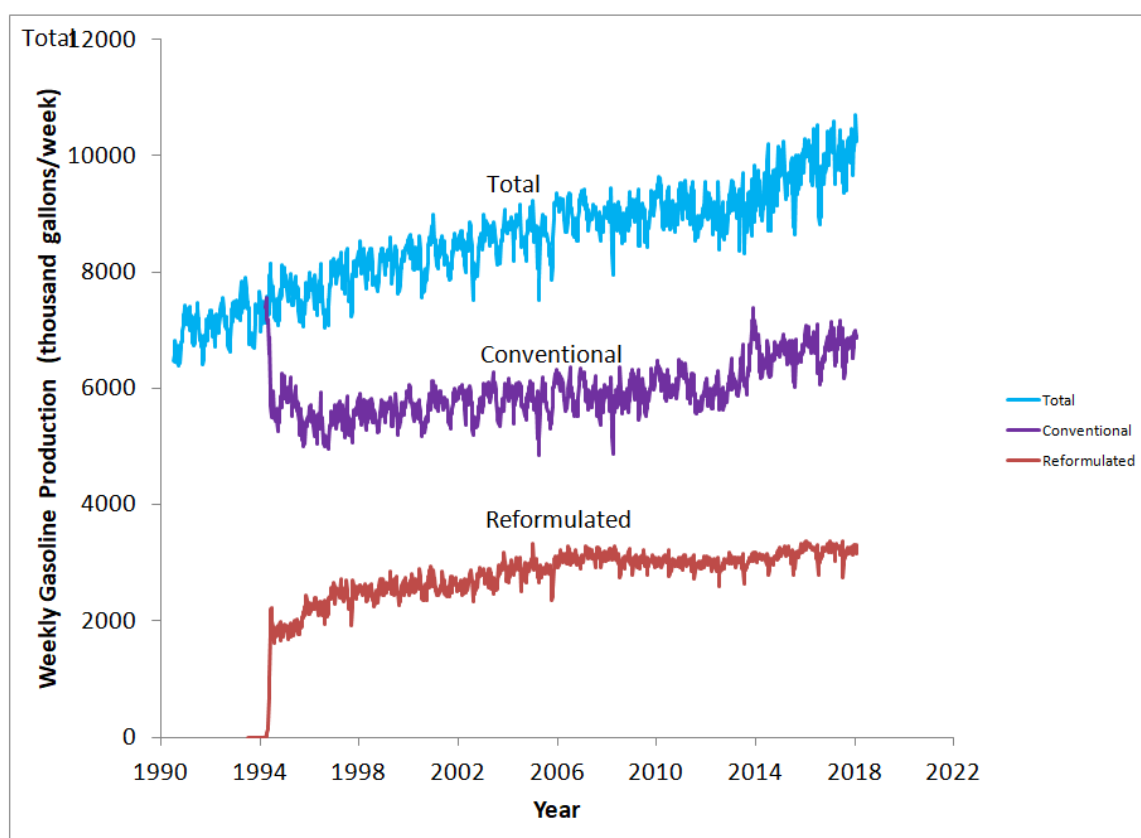


Figure 3: Amounts of total, conventional and reformulated gasoline produced per week in the U.S. (EIA, 2018).

5.0 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% oxygen by weight was required.

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 (Stickers,

2001). Table 5 lists the nine areas still in the program to the present, along with their required oxygen content and effective months. Detailed lists of the exit dates for cities that have left the program are given in Weaver et al. (2010).

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

| Control Period | 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 *State ethanol mandates.*

| State | Ethanol Requirement | Effective Date | Citation |
|------------|---------------------------|----------------|---------------------------------|
| Vol % | | | |
| Florida | 9 to 10; Repealed 2013 | Dec 21, 2010 | Florida, 2009; Florida, 2013 |
| Hawaii | 10 | 1994 | Hawaii, 2004 |
| Minnesota | 2.7 ^(a) | Oct 1, 1997 | Minnesota, 1997 |
| | 9.2 to 10 | 2003 | Minnesota, 2003 |
| Missouri | 10 | Jan 1, 2008 | Missouri, 2008 |
| Oregon | 10 | Nov 1, 2009 | Oregon, 2009 |
| Washington | “2% of gasoline” | Dec 1, 2008 | Washington, 2009 |

^(a)Oxygenated fuel requirement, Minnesota ethanol producer credit caused ethanol usage.

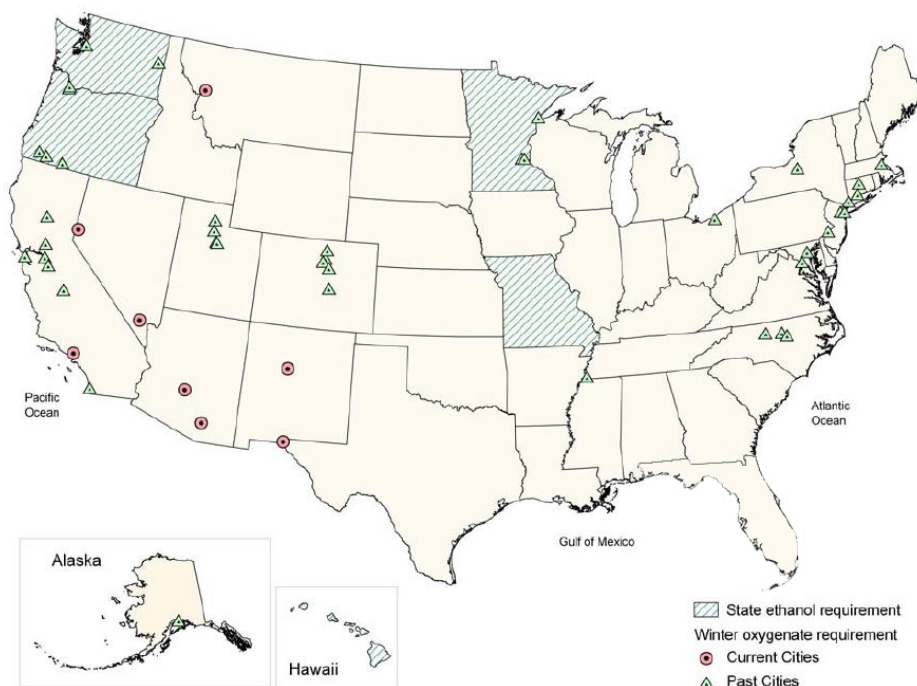


Figure 4: Oxygenated gasoline programs in the United States. Cities where an oxygenated additive was, or is, required for winter time gasoline are indicated by triangles and circles, respectively. Six states have imposed year-round oxygenate mandates.

6.0 Ethanol Mandates

Five states have continuing state-wide oxygenate mandates (Table 6). 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 ban on MTBE (Montana, 2005). The ethanol mandate was repealed in 2017 (Montana, 2018). The Florida requirement for ethanol was rescinded in 2009.

The federal Renewable Fuel Standard (RFS) was created by the Energy Policy Act of 2005 and extended by the Energy Independence and Security Act of 2007. It requires the replacement of petroleum fuel by renewable fuels which can be conventional, advanced or cellulosic. The program established renewable fuel targets for each year until 2020. The current target for 2020 is 36 billion gallons (EPA, 2018). 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.

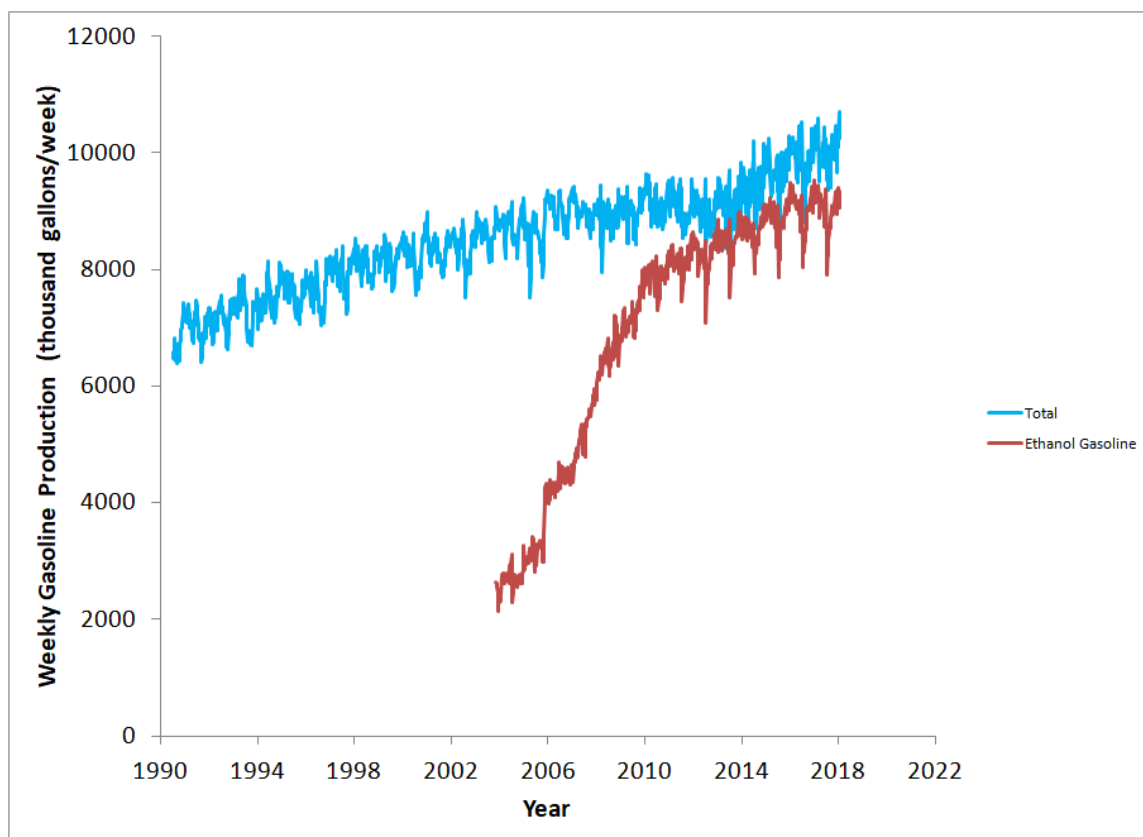


Figure 5: Total weekly gasoline production and the total containing ethanol. (data from EIA, 2018).



Figure 6: Advertisements at a gasoline station in Oklahoma City, July 2018, advertising the availability of ethanol-free gasoline and gasoline with 10% ethanol.

7.0 MTBE Bans

Beginning in 2000, Twenty-seven states, two counties, and one city banned MTBE or ether oxygenates in gasoline (Figure 7). Three states have imposed absolute bans, while most allow some MTBE to remain in gas (Weaver et al. 2010, Table 9). In 17 states, the level was 0.5%, while in others ranged from 0.05% in California to 1% in Nebraska. Six states banned MTBE and other oxygenates which included the other gasoline oxygenate ethers². The bans did not affect requirements to use reformulated or oxygenated-gasoline, but did cause a shift to another oxygenate. Data presented below show that the primary substitute was ethanol. Dates and specific requirements for each MTBE-banning state are given by Weaver et al. (2010).

² ethyl *tert*-butyl ether (ETBE); *tert*-amyl methyl ether (TAME); diisopropyl ether (DIPE); and alcohols, mostly *tert*-butyl alcohol (TBA)

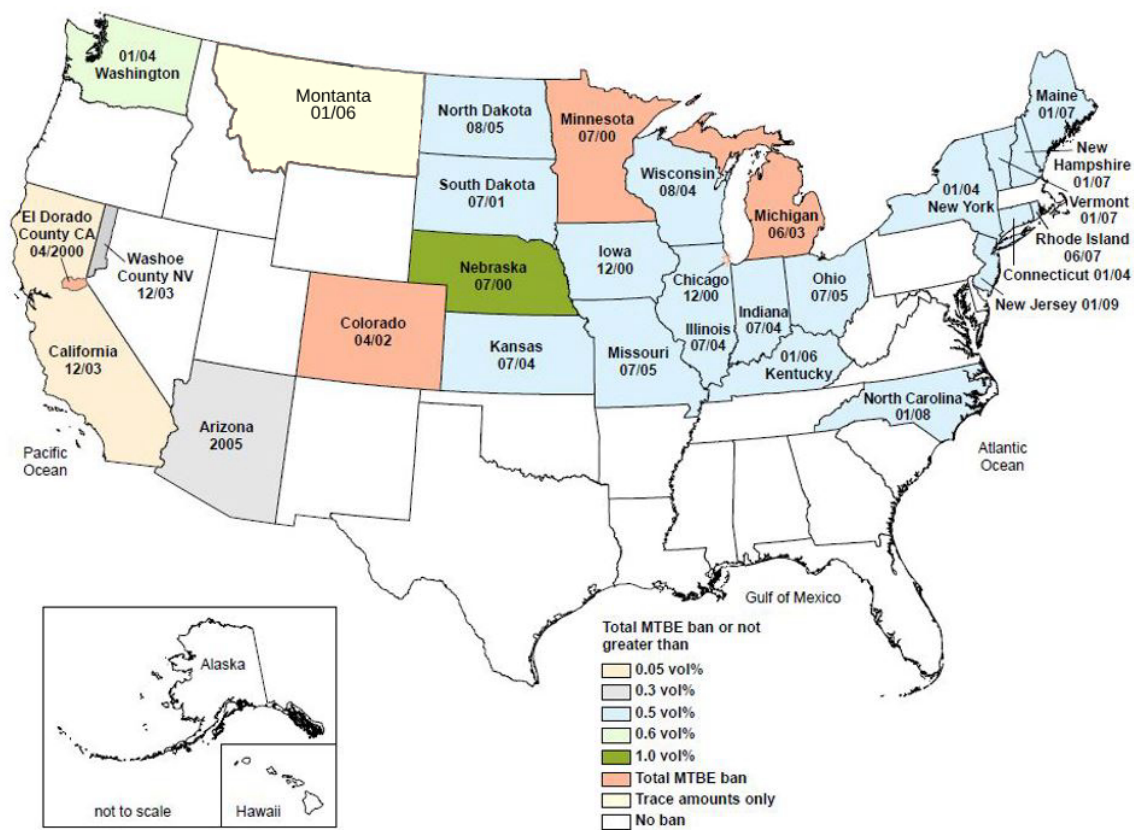


Figure 7: State MTBE, ether, and/or alcohol bans, showing effective dates and maximum levels allowed for MTBE. Please refer to Table 9 for full details on other ethers and alcohol.

8.0 Data and Methods

Data were obtained from three surveys: the National Institute of Petroleum and Energy Research (NIPER) and its successors (e.g., American Petroleum Institute (API), Northrop-Grumman), the Alliance of Automobile Manufacturers (Alliance), and the U.S. EPA Office of Transportation and Air Quality (OTAQ) in collaboration with industry (i.e., API, National Petroleum Refiners Association). In the next two sections, data from these three sources were compared to the regulatory time lines. First was the NIPER data set (e.g., Dickson, 2006), which published results from an industry consortium that has collected data since the 1930s. This company was the latest successor to the well-known NIPER. Its analyses were performed using ASTM standard methods for analysis of aromatics, ethers and alcohols. Northrup-Grumman data from 1976-2009 were obtained in an electronic format. From the late 1930s until 2009, NIPER and its successors collected and distributed gasoline survey data. The cities included and the parameters measured varied throughout time. Formerly the successors to NIPER distributed electronic data for the summer of 1976 through the winter of 2010. As many as 174 cities were included at various times. About 35 of the cities included have fairly complete records for the period of 1990 to 2010.

The second data source was the Alliance of Automobile Manufacturers (Alliance), and their data were used with permission. The data developed by the Alliance represents snapshots of retail fuel collected from retail service stations located in North American cities and do not represent a statistical or market based survey (North American Fuel Survey, 2018). The cities and specific service stations can vary from survey to survey. In accordance an agreement with the Alliance only data for conventional gasoline and California RFG were used from their sources, as no other data source covers these in later years. The primary presentation of all the data are as a minimum, median and maximum for each survey date.

The third source was data generated by the Reformulated Gasoline Survey Association (RFGSA). The RFGSA is a non-profit trade association that was formed through a collaboration of the American Petroleum Institute (API), the National Petroleum Refiners Association (NPRA) and EPA. Since 1995, RFGSA has conducted surveys of RFG required by the Clean Air Act and reports the results to EPA for evaluation of RFG compliance. EPA supplied a publicly-available version of RFGSA data which spanned the time period from 1995 to 2017.

The RFGSA survey designs utilize an aerial-based random sampling methodology with probability proportional to size to generate samples that represent the fuel sold without bias. Thus, the quantity of samples follow from the regulatory program—here reformulated gasoline and coordination between industry and EPA (RFGSA, 2018). California is not included in these surveys because of its exemptions under the Clean Air Act. So the California data below was selected from NIPER and Alliance data.

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

- Conventional gasoline cities of
 - a. Atlanta, GA
 - b. Denver, CO
 - c. Phoenix, AZ
 - d. Seattle, WA
- Federal RFG areas of
 - a. NY-NJ-Long Is.-CT
 - b. Baltimore, MD

- c. Boston-Worcester, MA
- d. Chicago-Lake Co., IL; Gary, IN
- e. Dallas-Fort Worth, TX
- f. Houston-Galveston, TX
- g. Philadelphia, PA-Wilmington, DE-Trenton, NJ
- h. Portsmouth-Dover, NH
- i. St. Louis, MO
- j. Washington, D.C.-area
- Cal-RFG
 - a. Los Angeles

The 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. 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.

9.0 Data Combination

The data were combined using a custom-written java program. The program reads each available instance of each survey and combines the three data sets based on location, which is done in two ways: by city name for CG and Cal RFG cities, and by RFG compliance region. This process is straightforward for NIPER and Alliance data from CG and Cal RFG cities, as the city names were recorded in a similar way (i.e., New York City versus New York). Combining OTAQ data NIPER data, however, requires that the RFG regions be assigned to cities within the NIPER data set. In both cases, a list of equivalent location (city and RFG compliance region) names were developed for combining the data.

The surveys also differed in parameters reported and their nomenclature. For example, NIPER reported the month and year of the survey, while OTAQ reported a date, and Alliance reported either summer or winter. Because the main purpose was to differentiate summer and winter survey results, NIPER data were assigned to the 15th day of the collection month, and Alliance data were either assigned to January 15 or July 15. Some parameters (MTBE, ethanol, and others) were reported either as percent by weight or percent by volume. As the data were processed all of the percent by weight values were converted to percent by volume, either by using the actual or an averaged fuel density.

10.0 Results

Cities were selected for discussion to highlight characteristics of reformulated, California-reformulated, conventional, and oxygenated gasoline. These were selected from the list given in Section 3. Data from the cities not specifically discussed below are presented in Appendix A.

10.1 Reformulated Gasoline

Data from NIPER and OTAQ patterns in reformulated gasoline. The Houston-Galveston and NY-NJ-CT areas are used as examples, while data from the other RFG areas are presented in Appendix A. The pre-1995 data were drawn from the NIPER data (dashed lines), and OTAQ data (solid lines) extend the time range through 2017. The data are presented as minimum, median, and maximum values in each survey. The median values establish the general trends. The minimum and maximum 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 the 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. After the imposition of the MSAT benzene requirement of <0.62% in 2011, the median benzene in Houston-Galveston was less than 0.535% and that of NY-NJ-CT less than <0.73%.

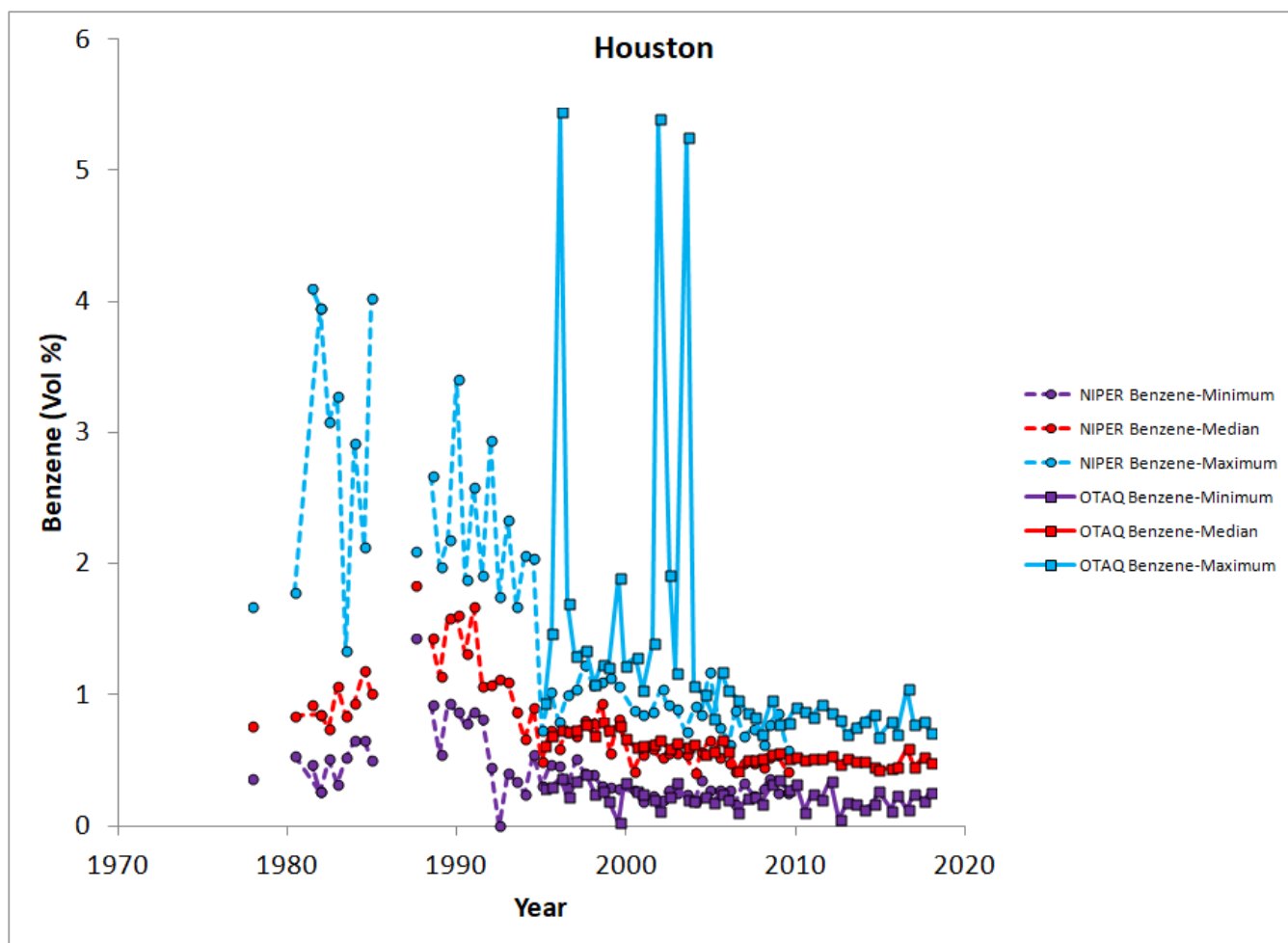


Figure 8: Benzene content in RFG from Houston and Galveston, Texas.

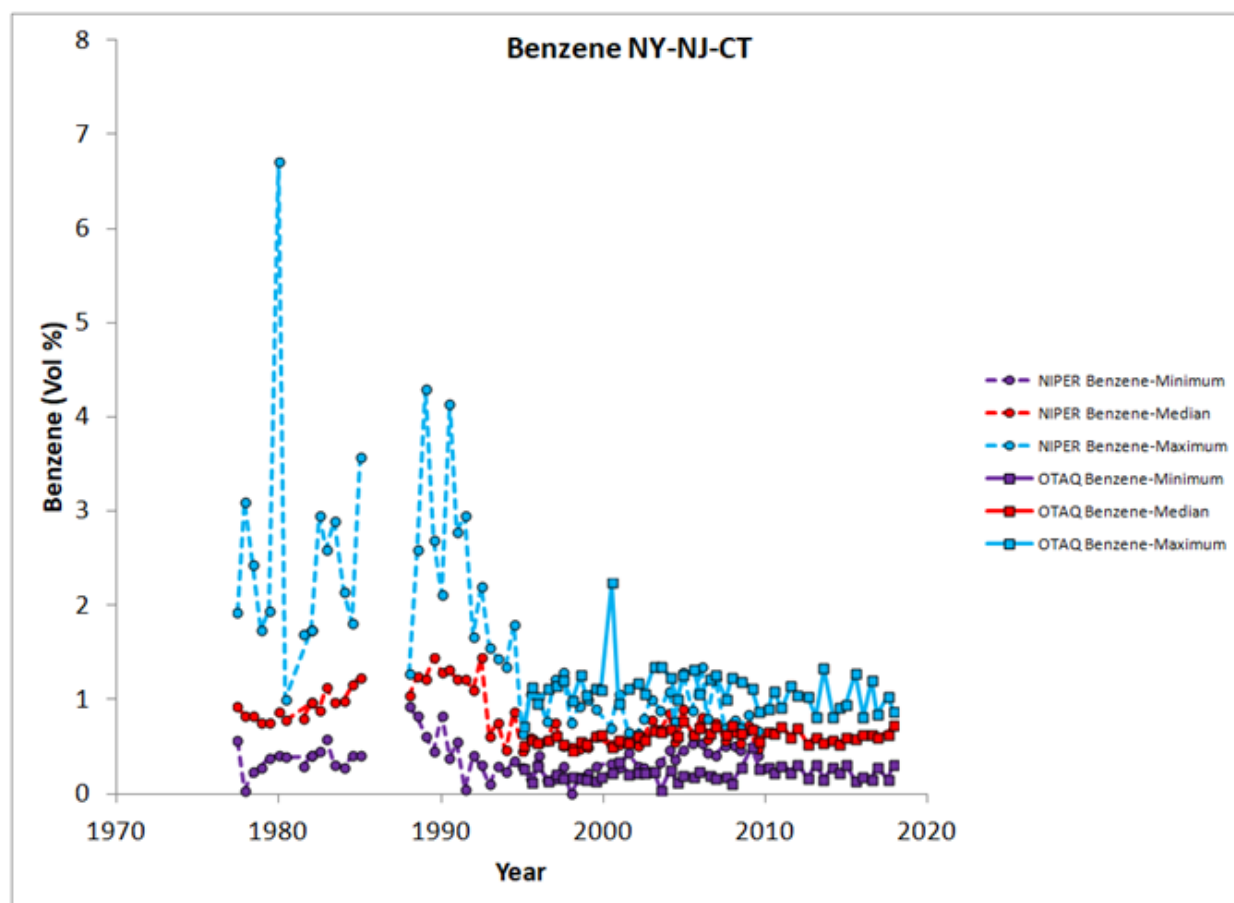


Figure 9: Benzene content in RFG from the New York City area, including New York, New Jersey, and Connecticut.

Prior to the EPAct removal of the oxygenate mandate from RFG, MTBE was the dominant oxygenate in Houston-Galveston area gasoline (Figure 10). The median concentration of MTBE was around 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, MTBE 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 EPAct did, however, require use of biofuels at levels up to 10%.

NY-NJ-CT area gasoline shows a more complex picture in two ways (Figure 11). First, this area participated in the oxygenated gasoline program from its inception in 1992 until it 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. From 1995 to 2000, the median MTBE is around 11%. Some oxygenate was supplied by other ethers during this time (not shown). The second feature that New York and Connecticut banned MTBE in 2004 while New Jersey did not until 2009 (Figure 7). 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 showing MTBE usage, while New York and Connecticut samples showed ethanol use. 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).

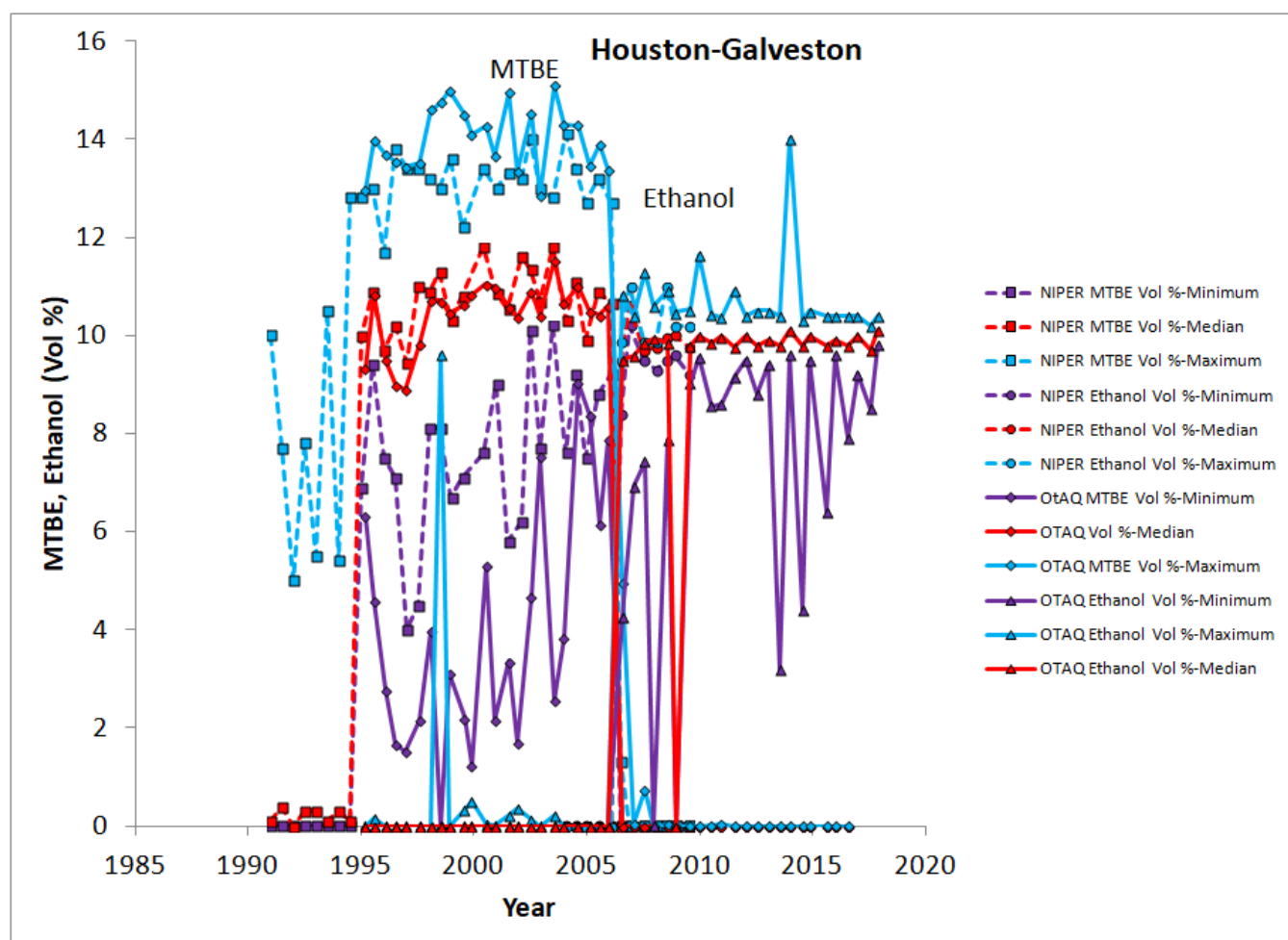


Figure 10: Oxygenates (MTBE and ethanol) in Houston-Galveston gasoline.

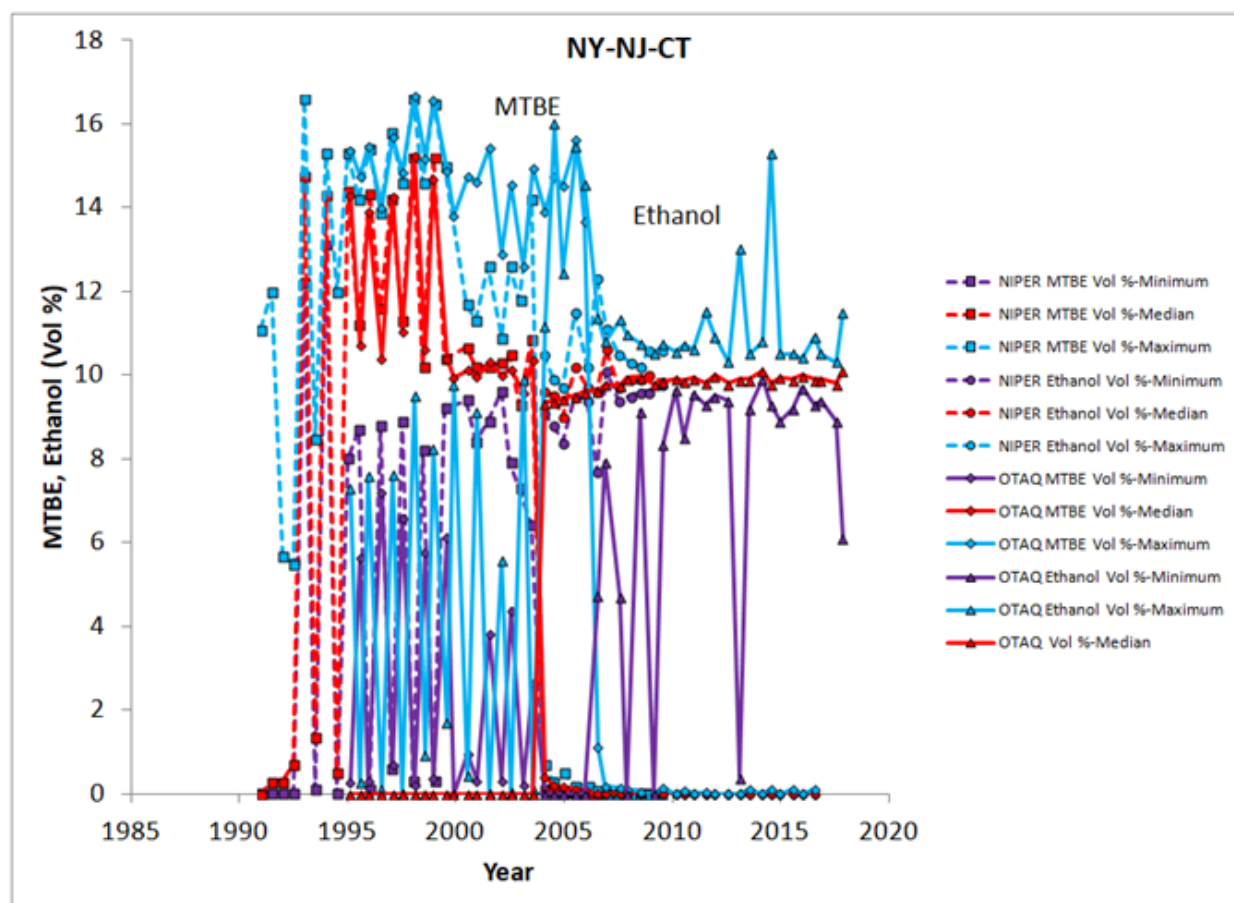


Figure 11: Oxygenates (MTBE and ethanol) in NY-NY-CT area reformulated gasoline.

10.2 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. As for New York, Los Angeles was in the oxygenated gasoline program from 1992 to until the 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.

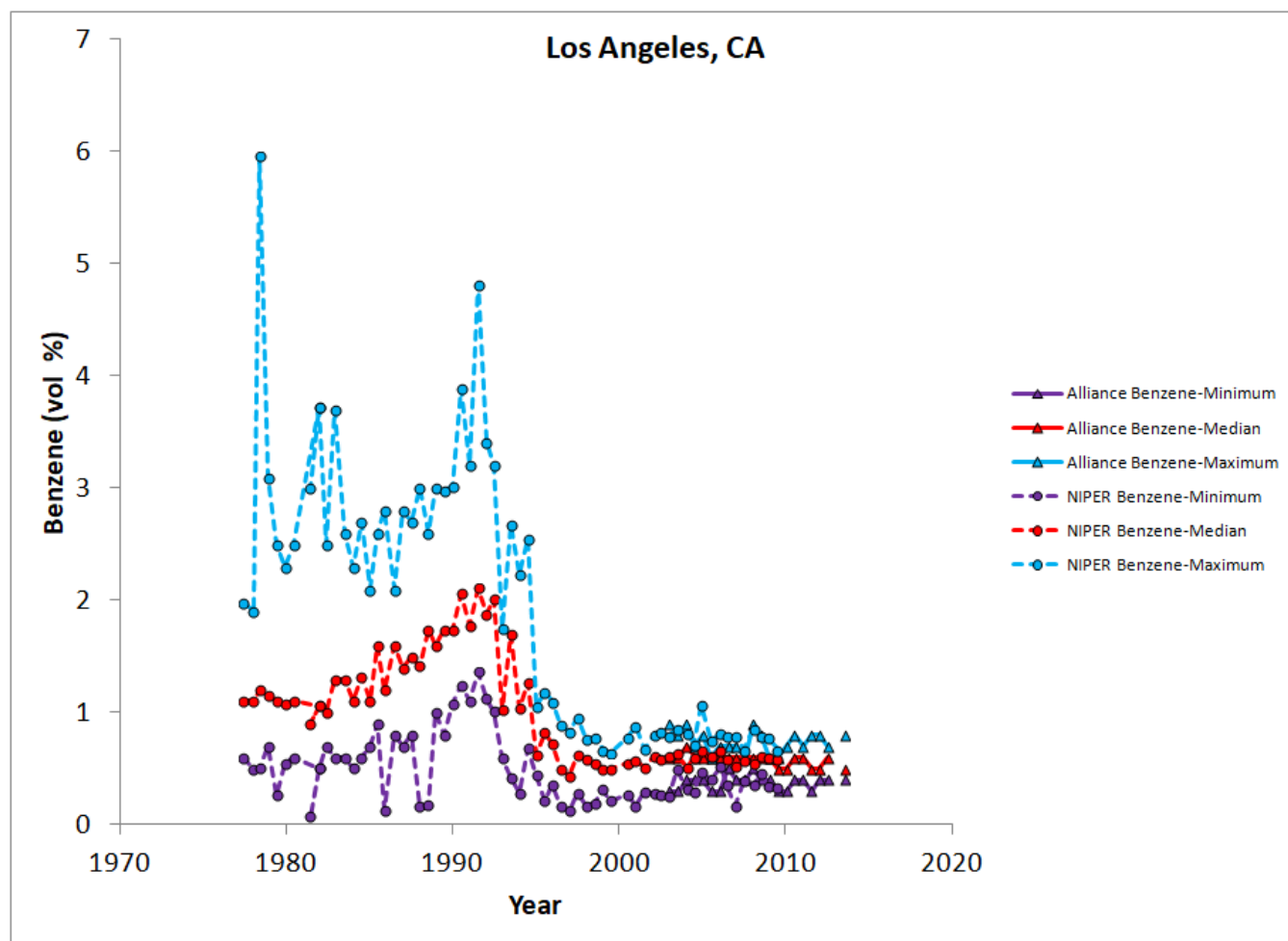


Figure 12: Benzene content in California RFG (Alliance data: North American Auto Alliance, 2018).

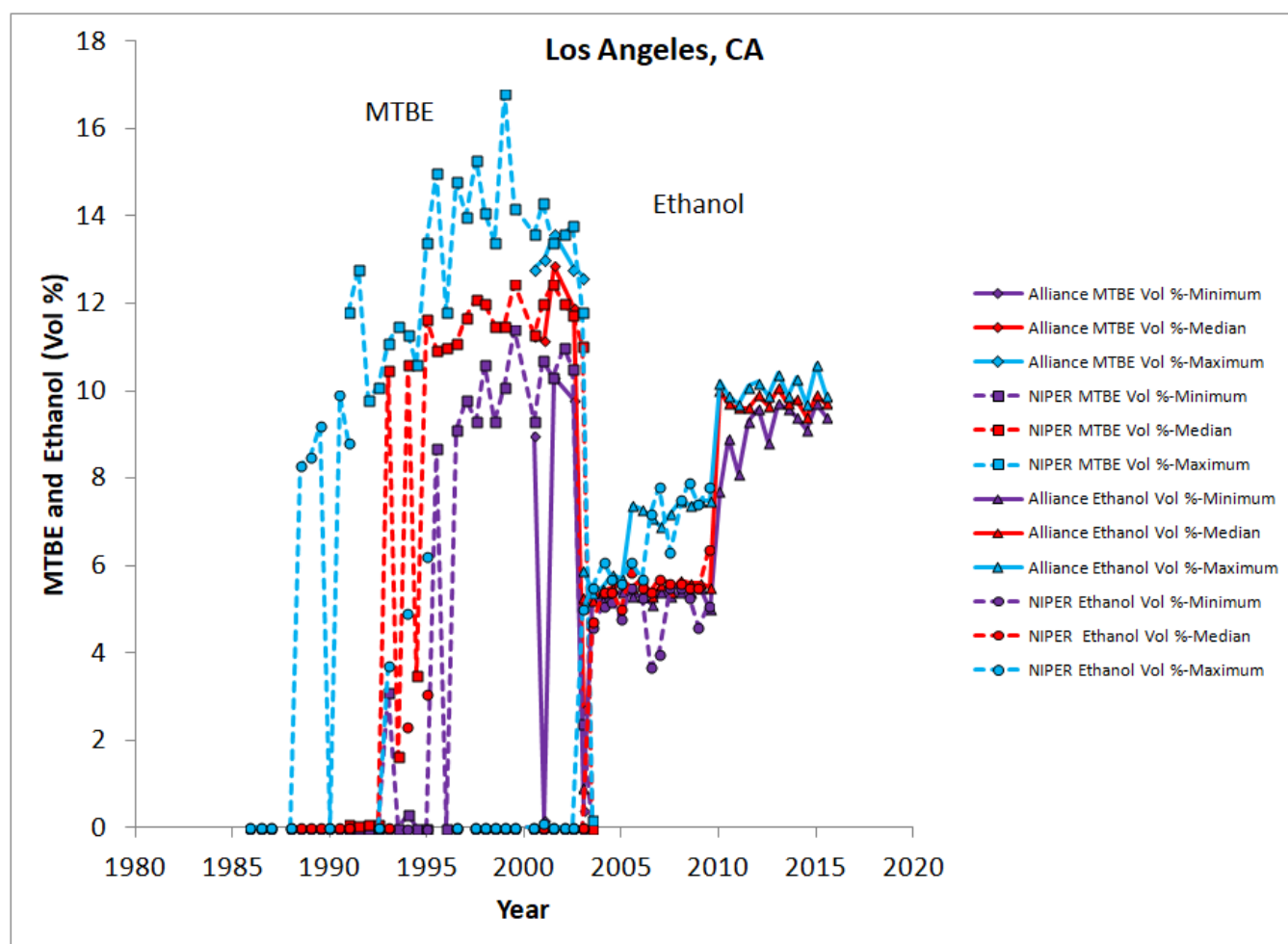


Figure 13: Oxygenates (MTBE and ethanol) in Los Angeles gasoline (Alliance data: North American Auto Alliance, 2018).

10.3 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 (Table 4). Thus, the benzene in use at a specified city could include products from several refineries, each with a different baseline. Cities like Atlanta and Seattle (Figure 14 and Figure 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 usage of MTBE (and other oxygenates) varied greatly, but typically at lower levels than mandated for RFG or OG (Figure 16 and Figure 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 EPA mandated biofuel use on a nationwide basis.

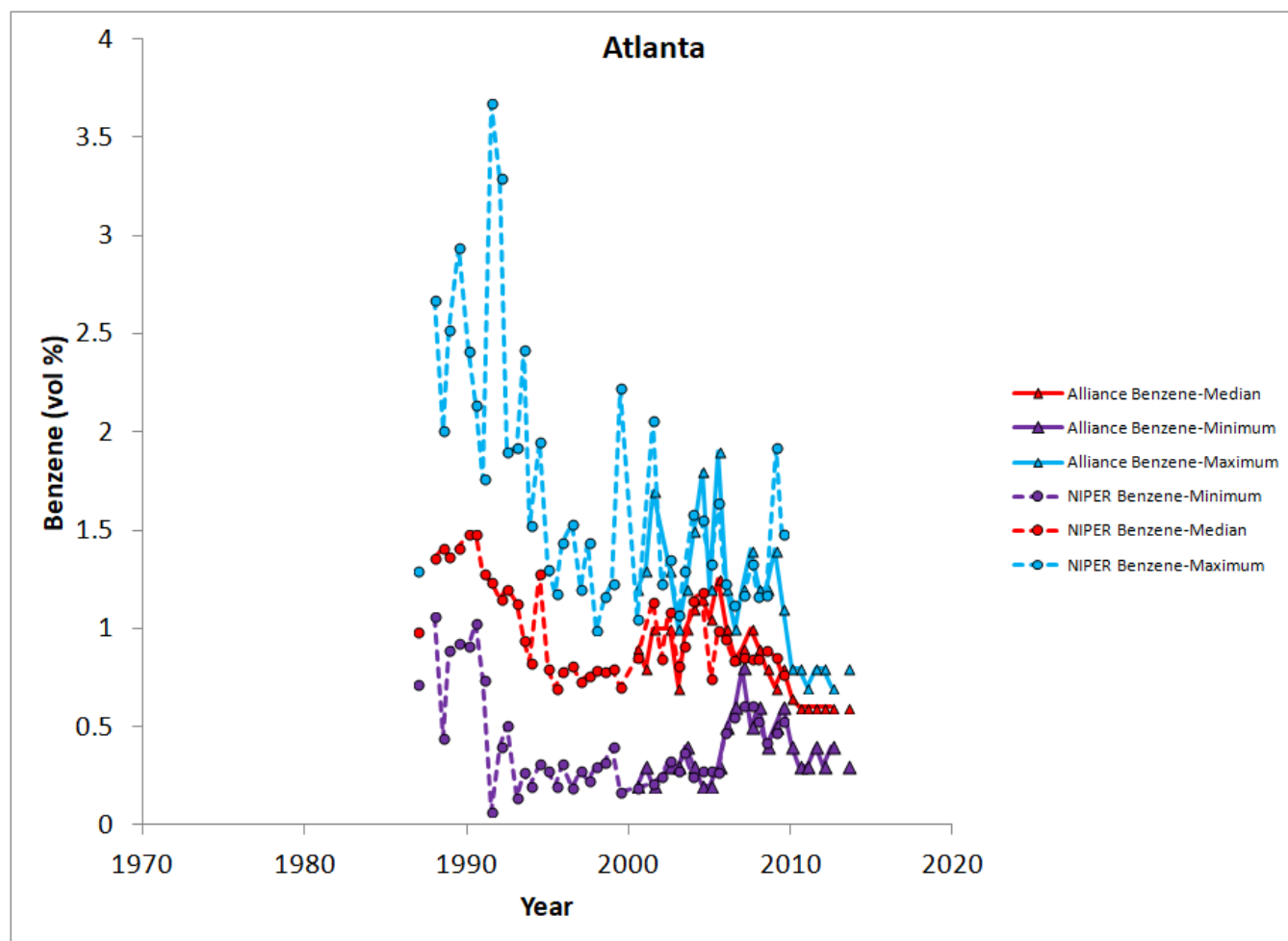


Figure 14: Benzene content in Atlanta, GA conventional Gasoline (Alliance data: North American Auto Alliance, 2018).

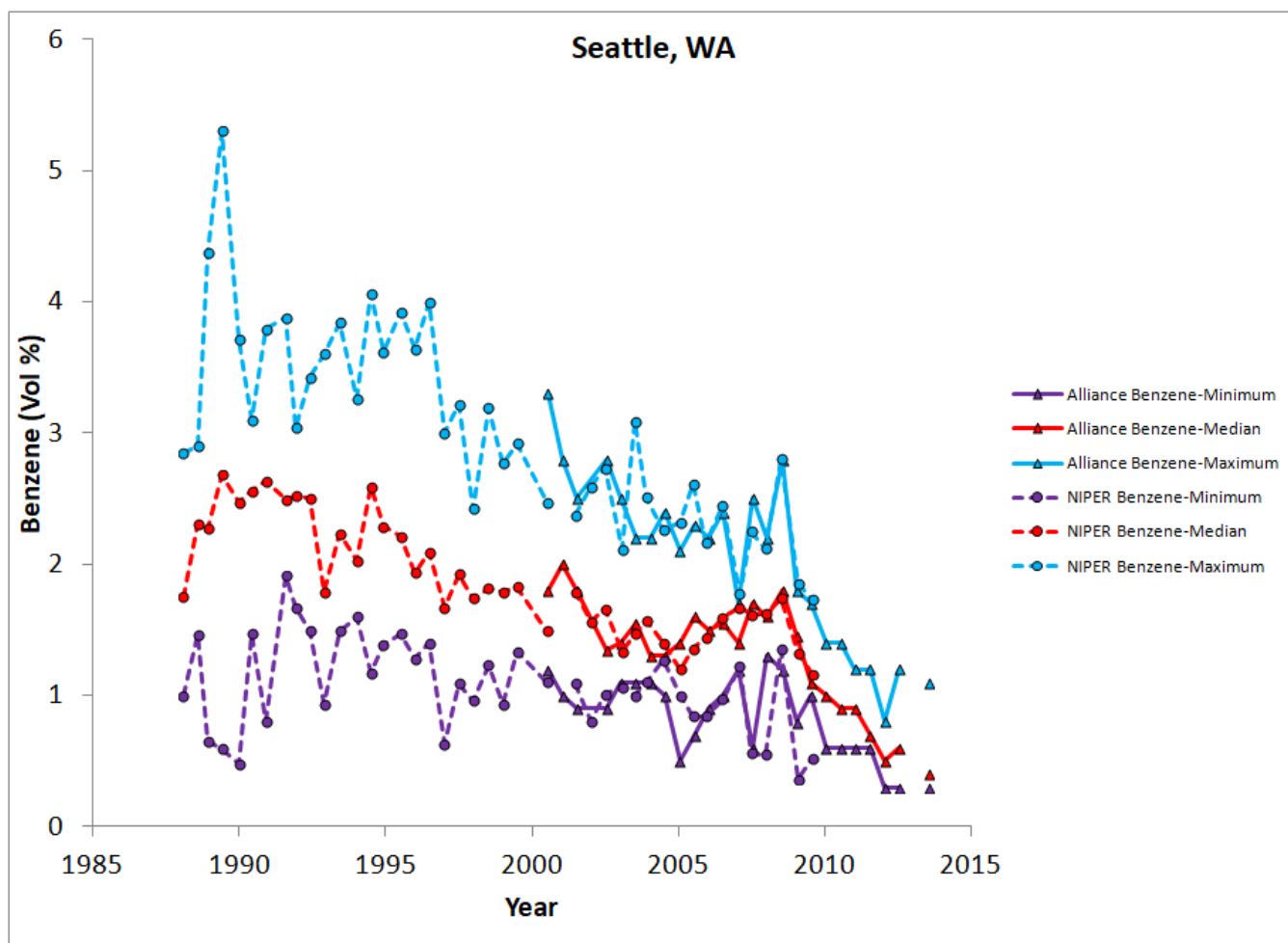


Figure 15: Benzene content in Seattle, WA conventional Gasoline (Alliance data: North American Auto Alliance, 2018).

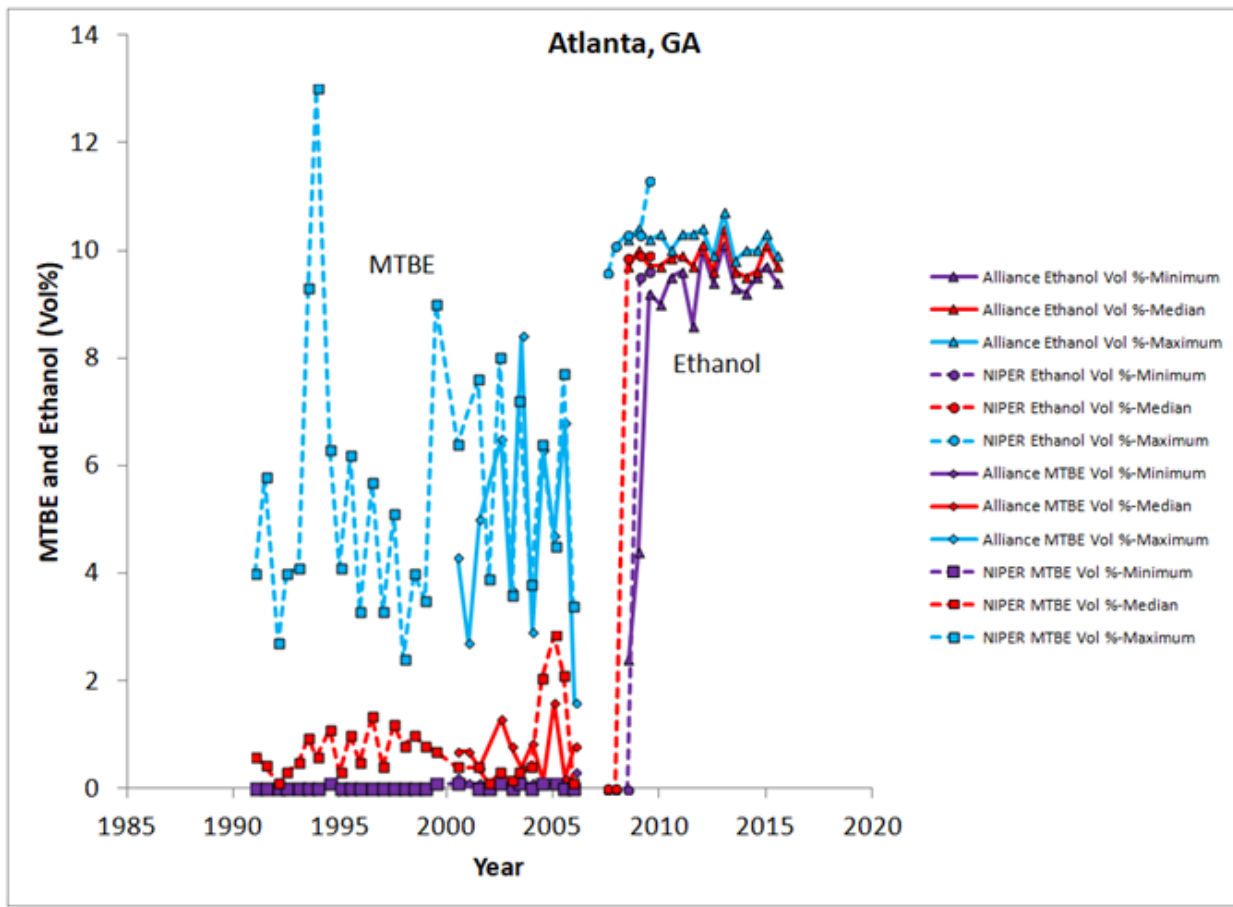


Figure 16: Oxygenates (MTBE and ethanol) in Atlanta, GA conventional gasoline (Alliance data: North American Auto Alliance, 2018).

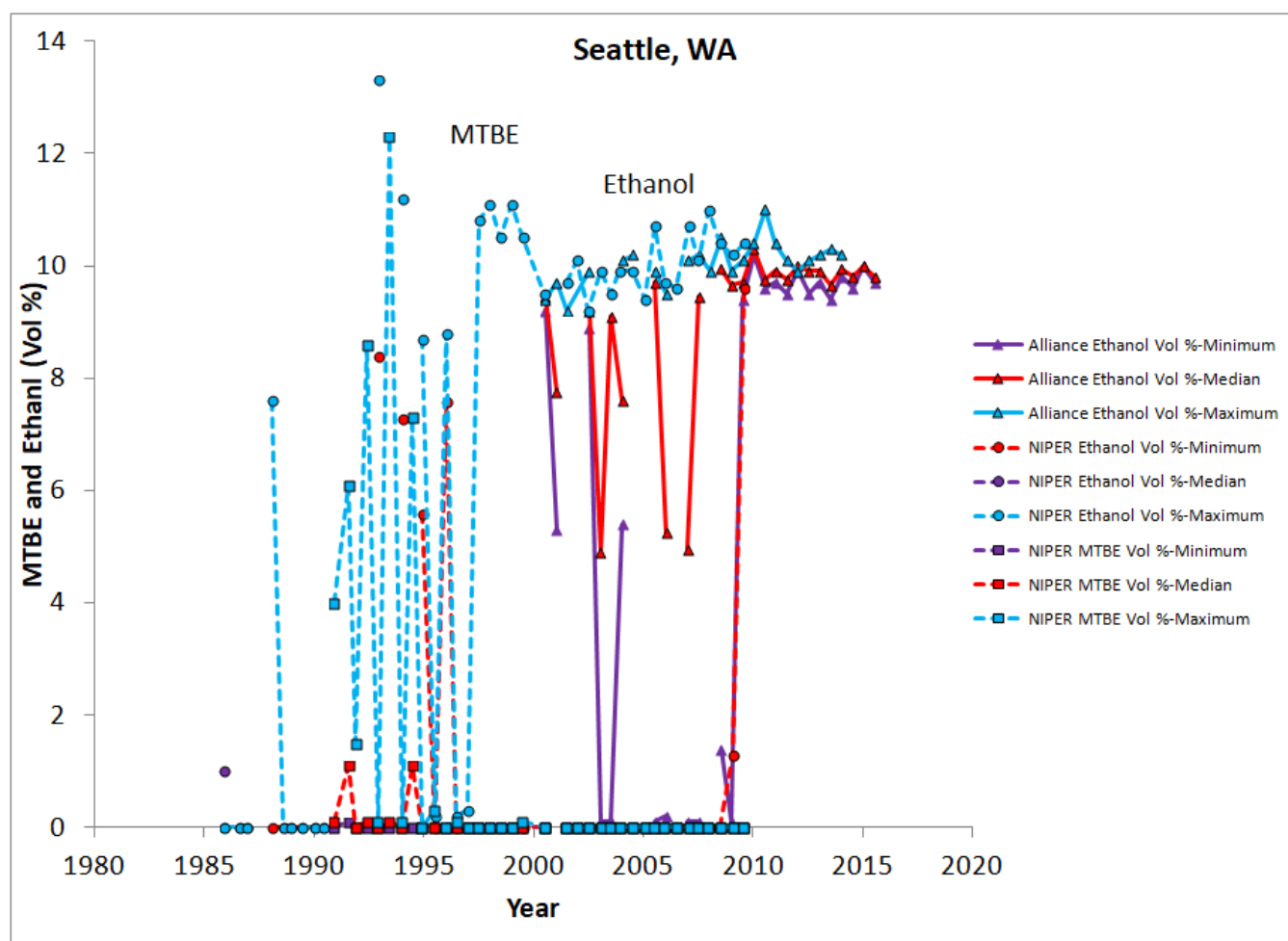


Figure 17: Oxygenates (MTBE and ethanol) in Seattle, WA conventional gasoline (Alliance data: North American Auto Alliance, 2018).

10.4 Oxygenated Gasoline

Phoenix has participated in the oxygenated gasoline program and requires oxygenated gasoline from November through the end of March (Table 5). As a conventional gasoline city, the 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 the 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%.

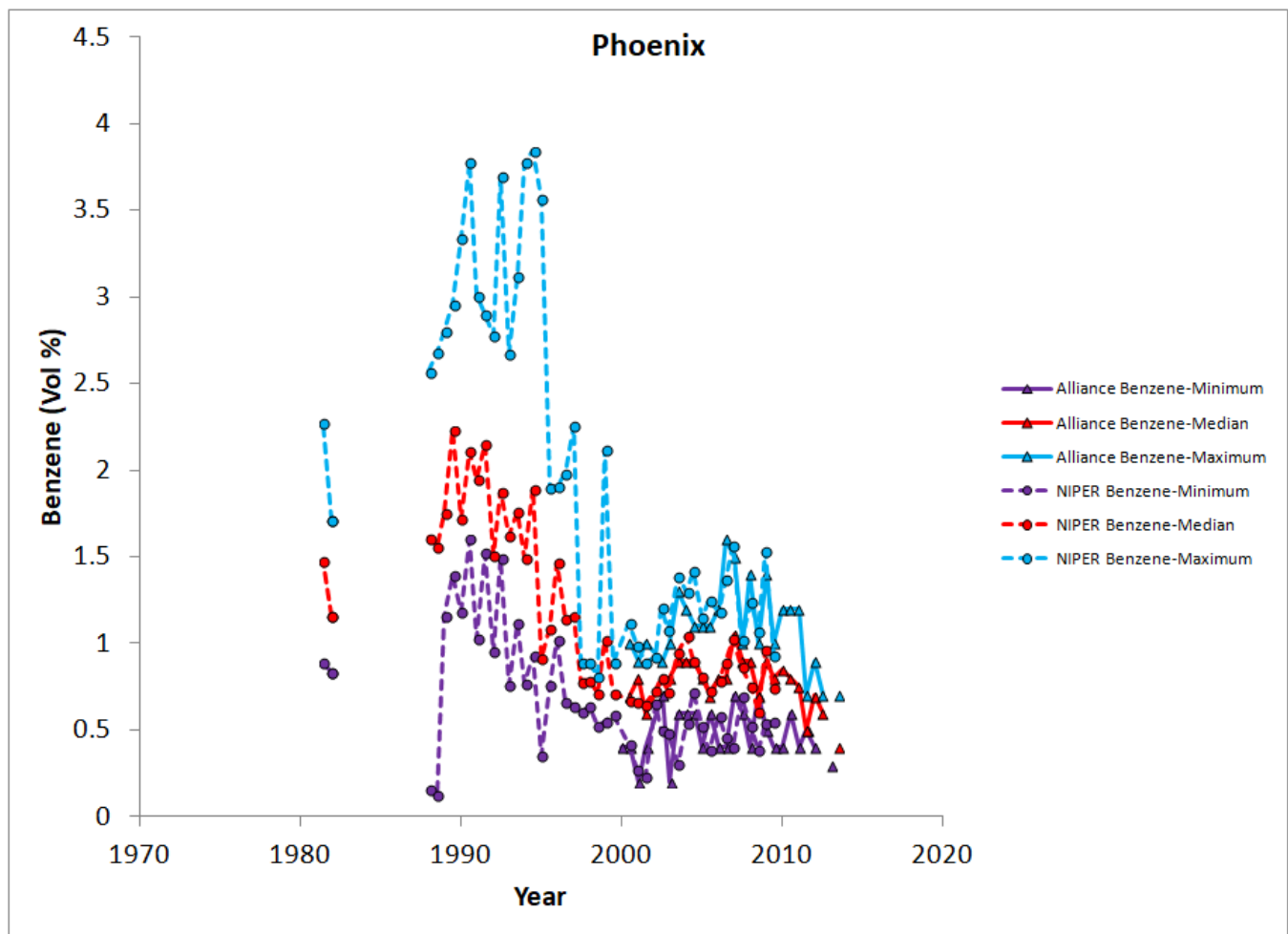


Figure 18: Benzene content in Phoenix, AZ conventional/oxygenated gasoline (Alliance data: North American Auto Alliance, 2018).

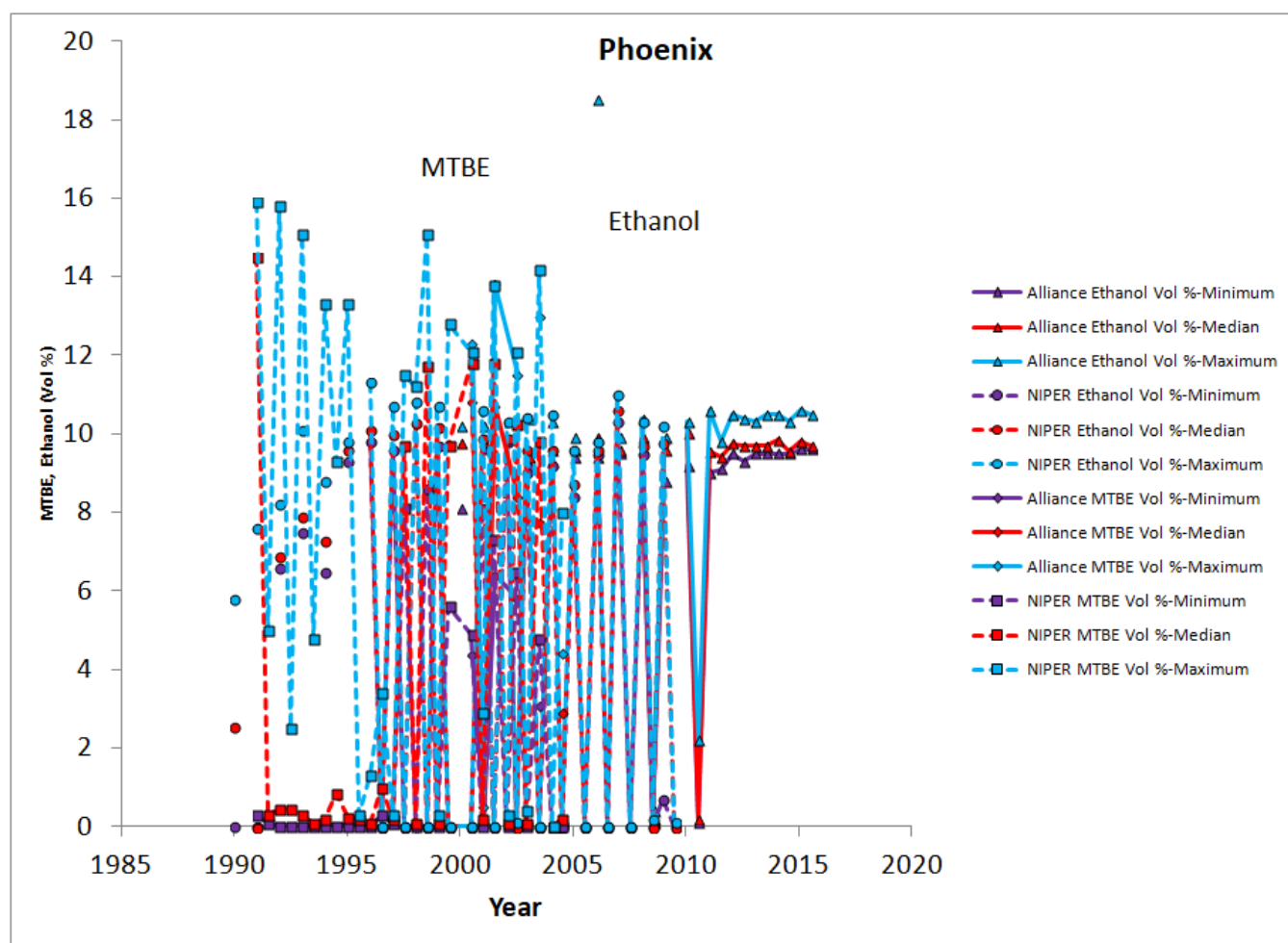


Figure 19: Oxygenates (MTBE and ethanol) in Phoenix, AZ conventional/oxygenated gasoline (Alliance data: North American Auto Alliance, 2018).

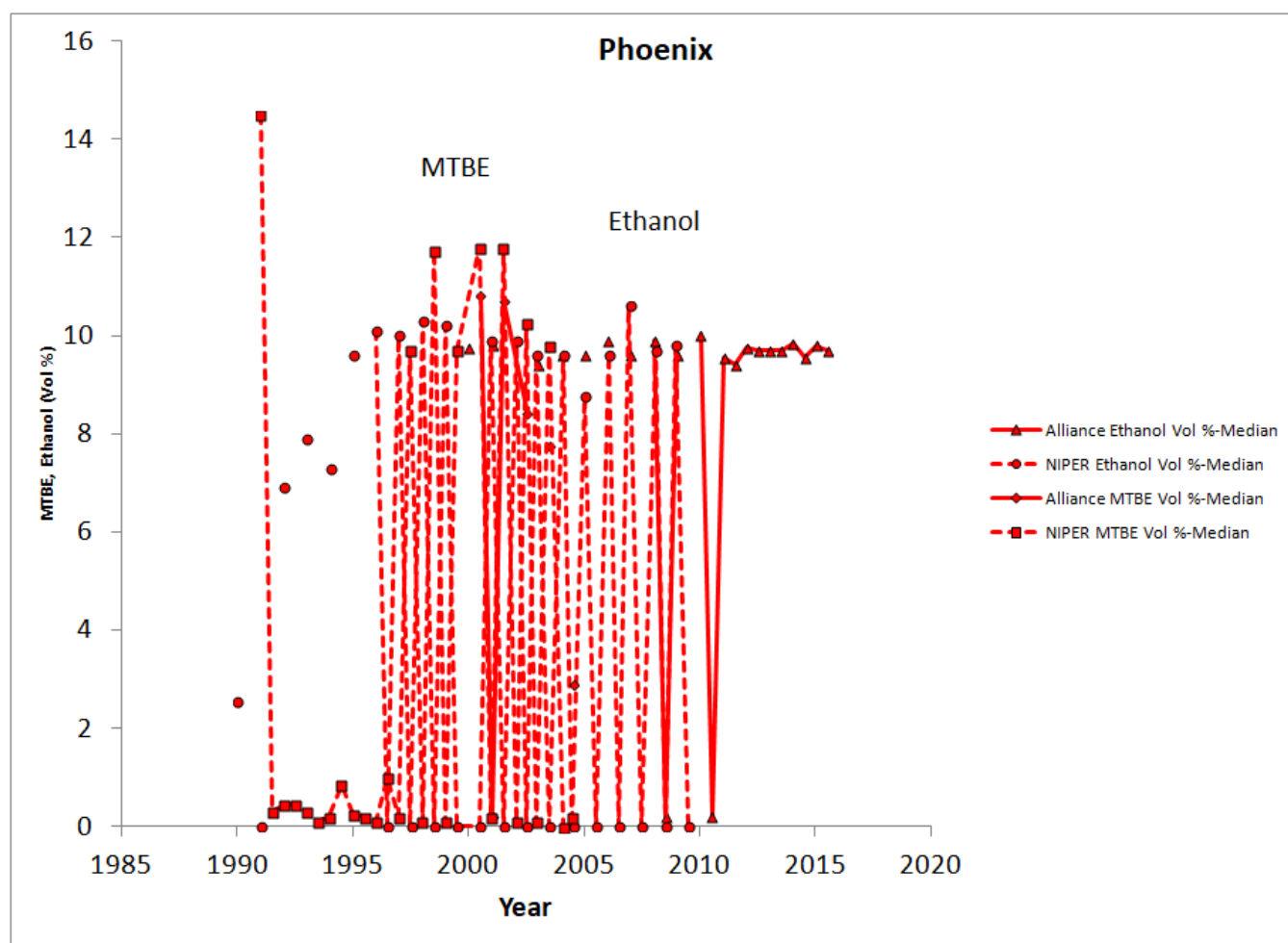


Figure 20: Median values of oxygenate concentration (MTBE and ethanol) in Phoenix, AZ conventional/oxygenated gasoline (Alliance data: North American Auto Alliance, 2018).

11.0 Conclusions

The 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 EPAct, 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.

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Appendices

Appendix A

Conventional Gasoline Cities not appearing in the main text: Denver, Colorado

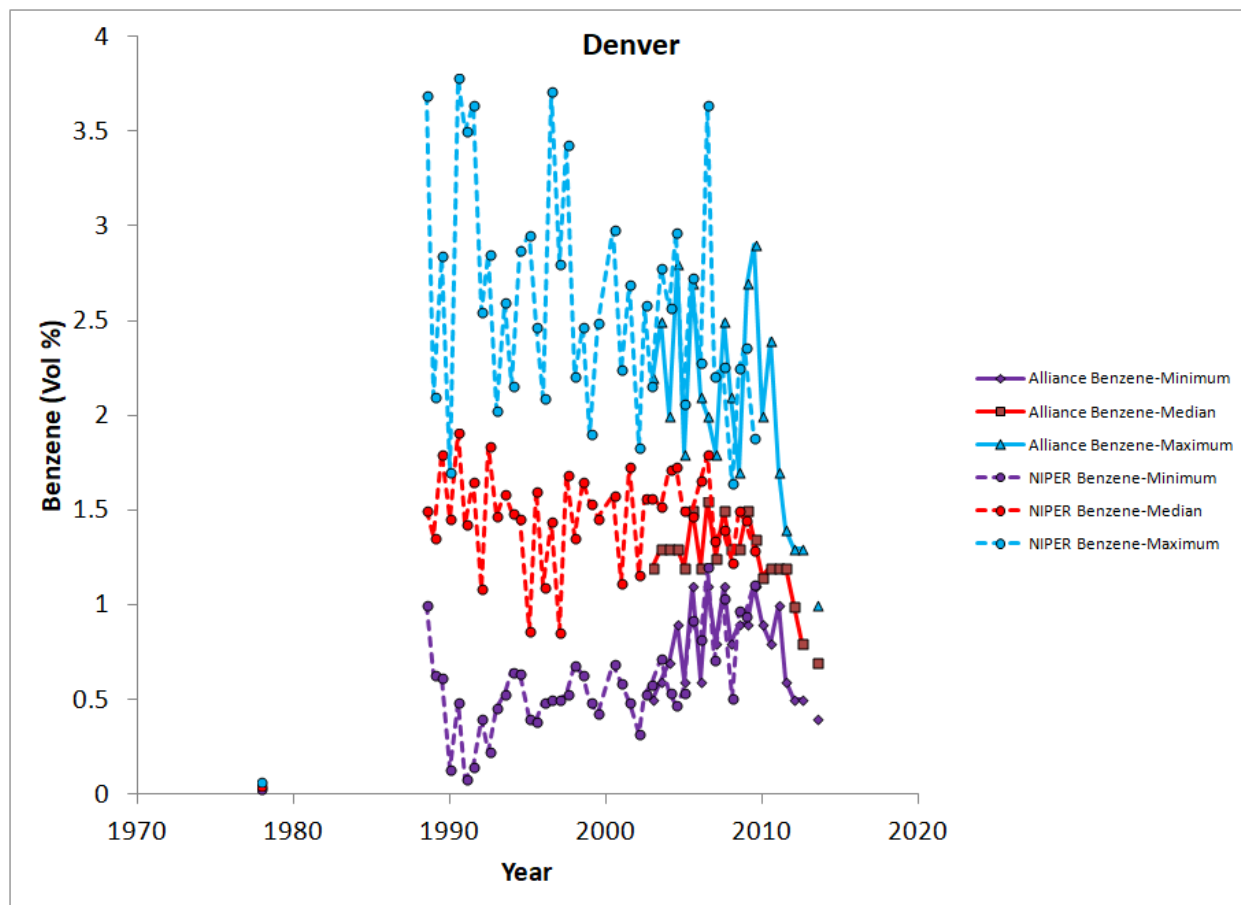


Figure 21: Benzene content in conventional gasoline from Denver, Colorado.

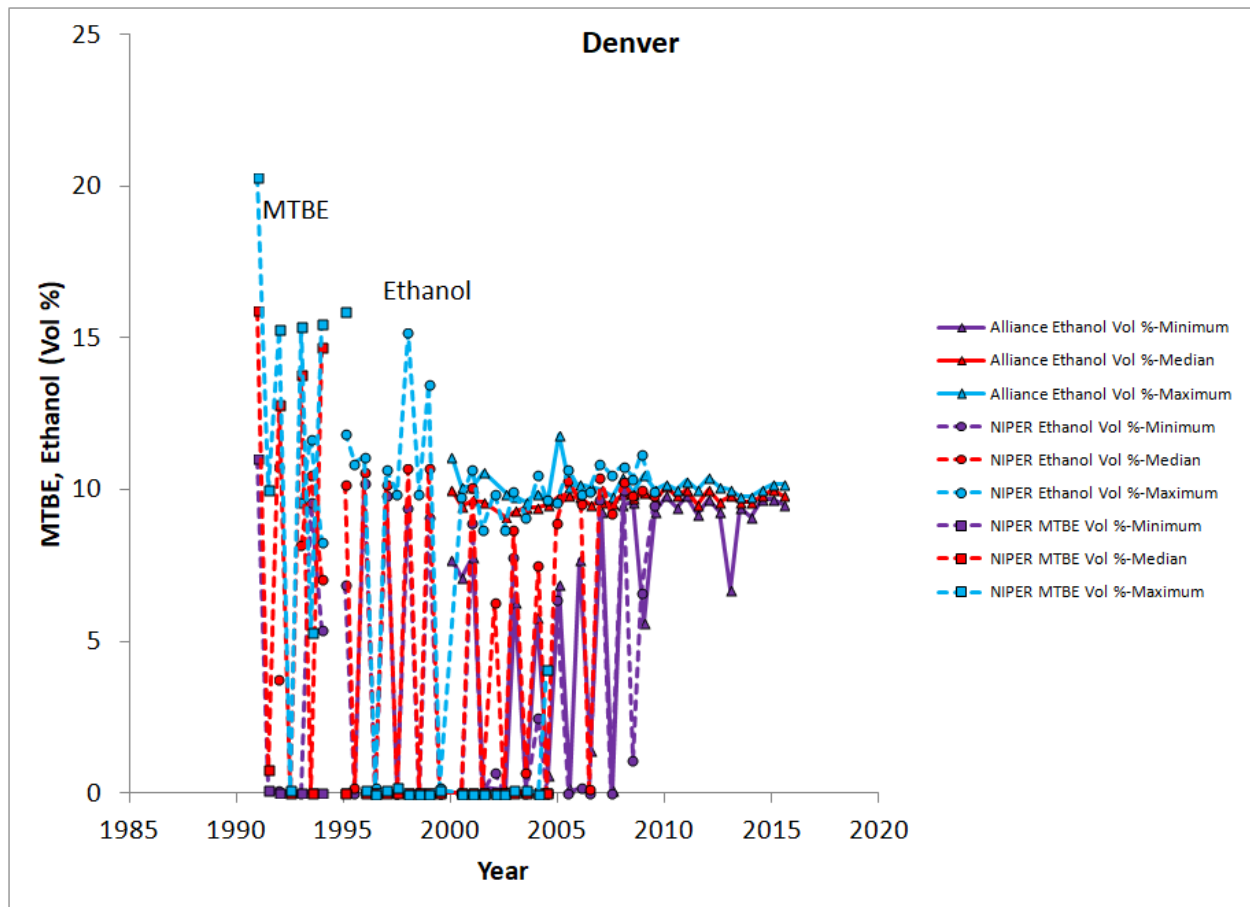


Figure 22: Oxygenates (MTBE and ethanol) in Denver, Colorado conventional gasoline.

Appendix B

Reformulated gasoline areas not appearing in the main text: Baltimore, MD; Boston-Worcester, MA, Chicago-Lake Co, IL –Gary, IN; Dallas-Fort Worth, TX; Philadelphia, PA—Wilmington, DE—Trenton, NJ; Portsmouth-Dover, NH; St. Louis, MO; Washington DC area.

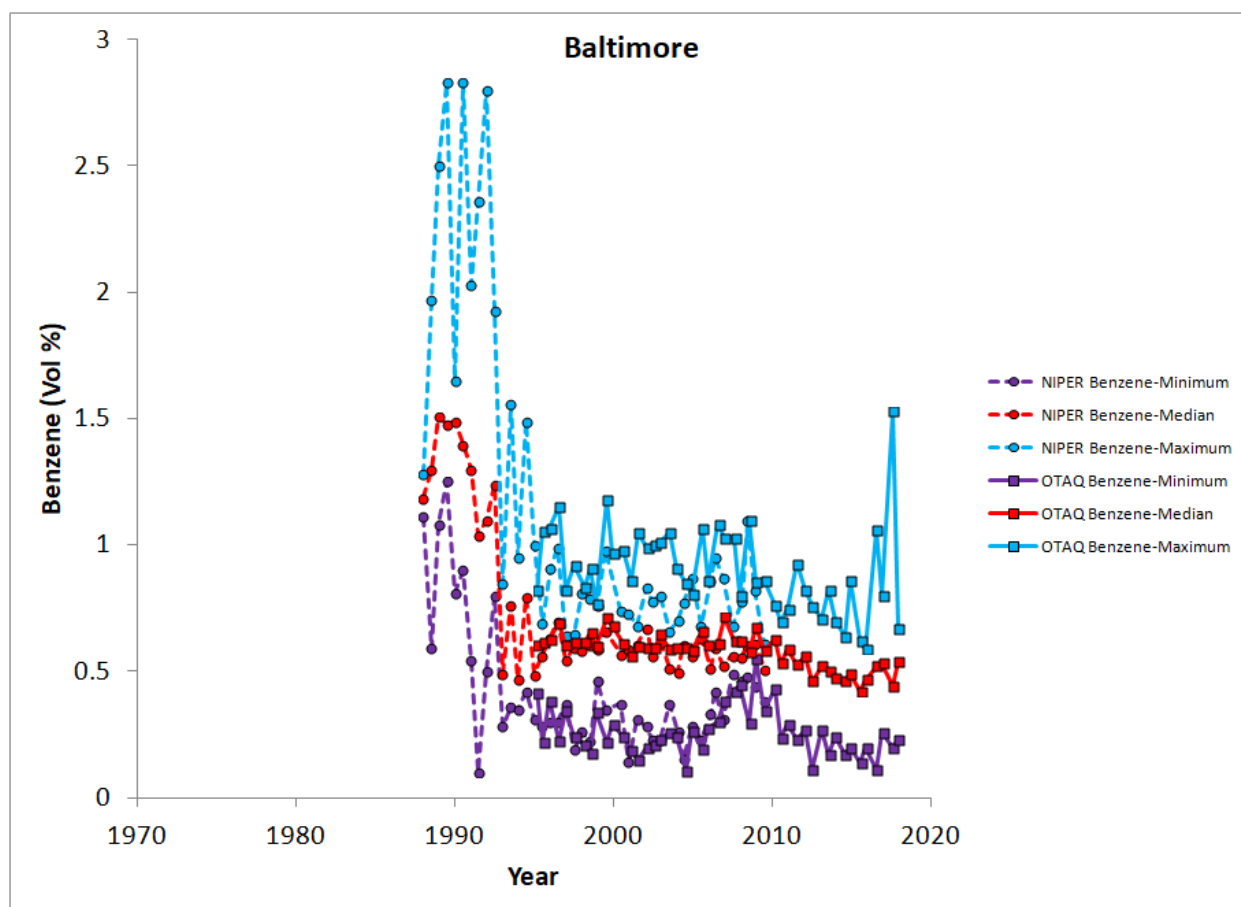


Figure 23: Benzene content in reformulated gasoline from the Baltimore, Maryland area.

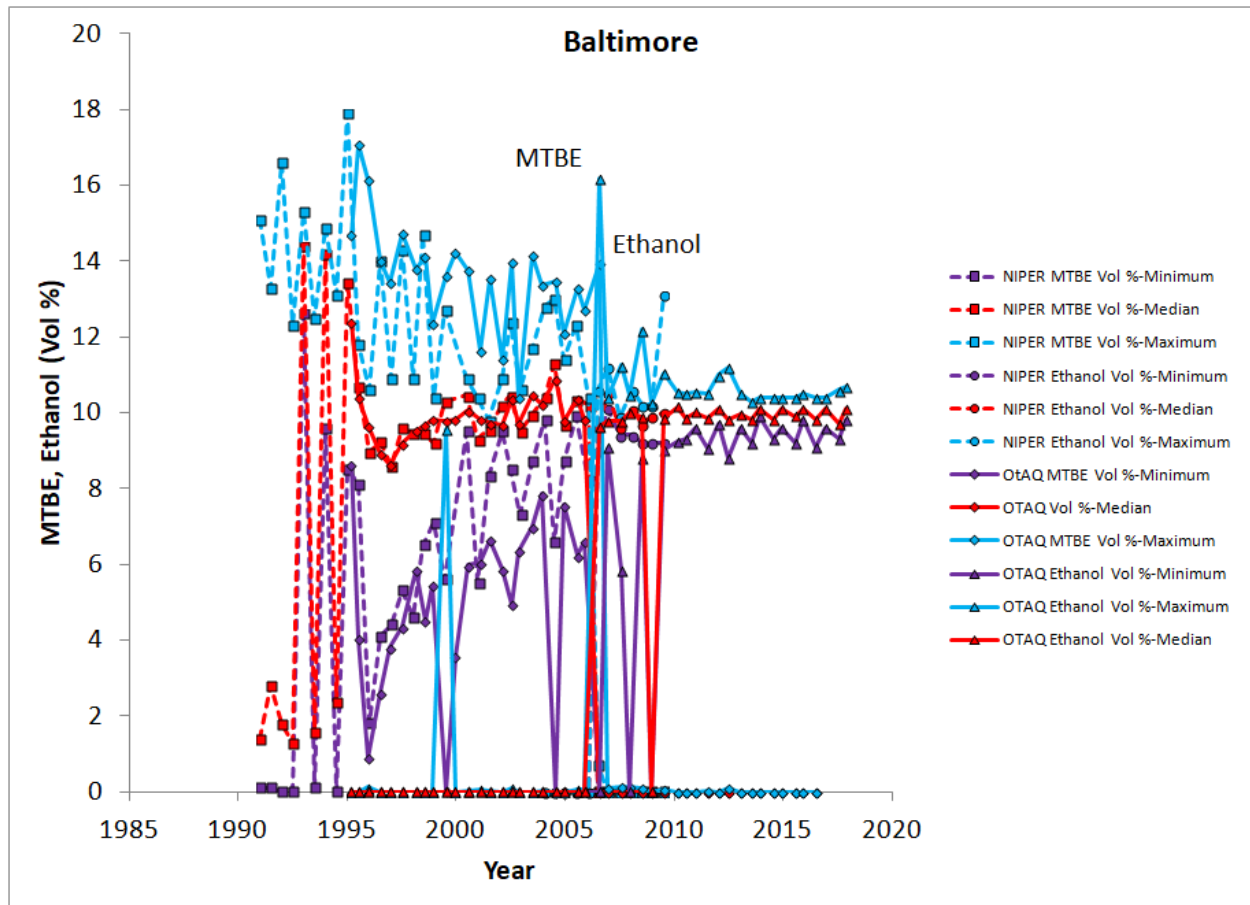


Figure 24: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Baltimore, Maryland area.

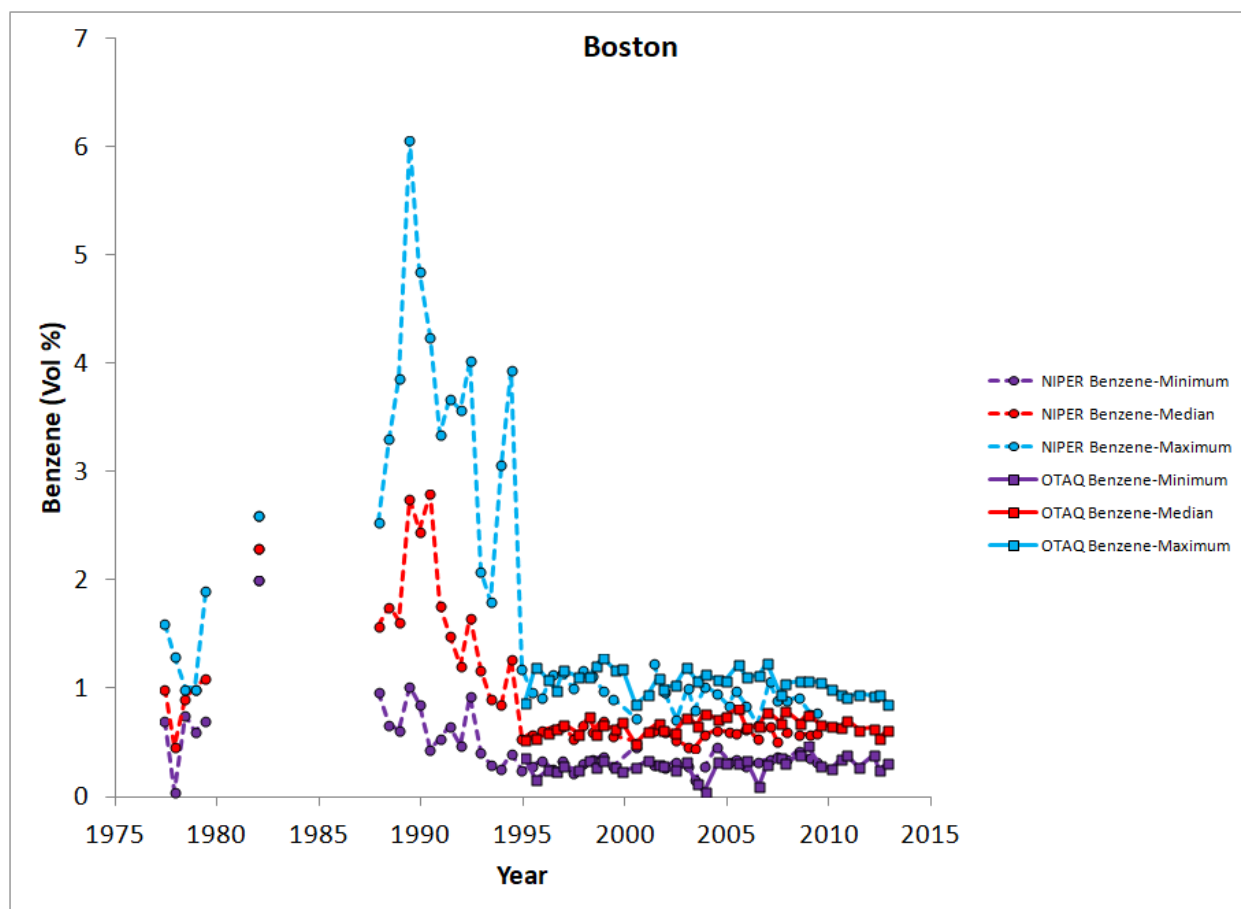


Figure 25: Benzene content in reformulated gasoline from the Boston-Worcester, Massachusetts area.

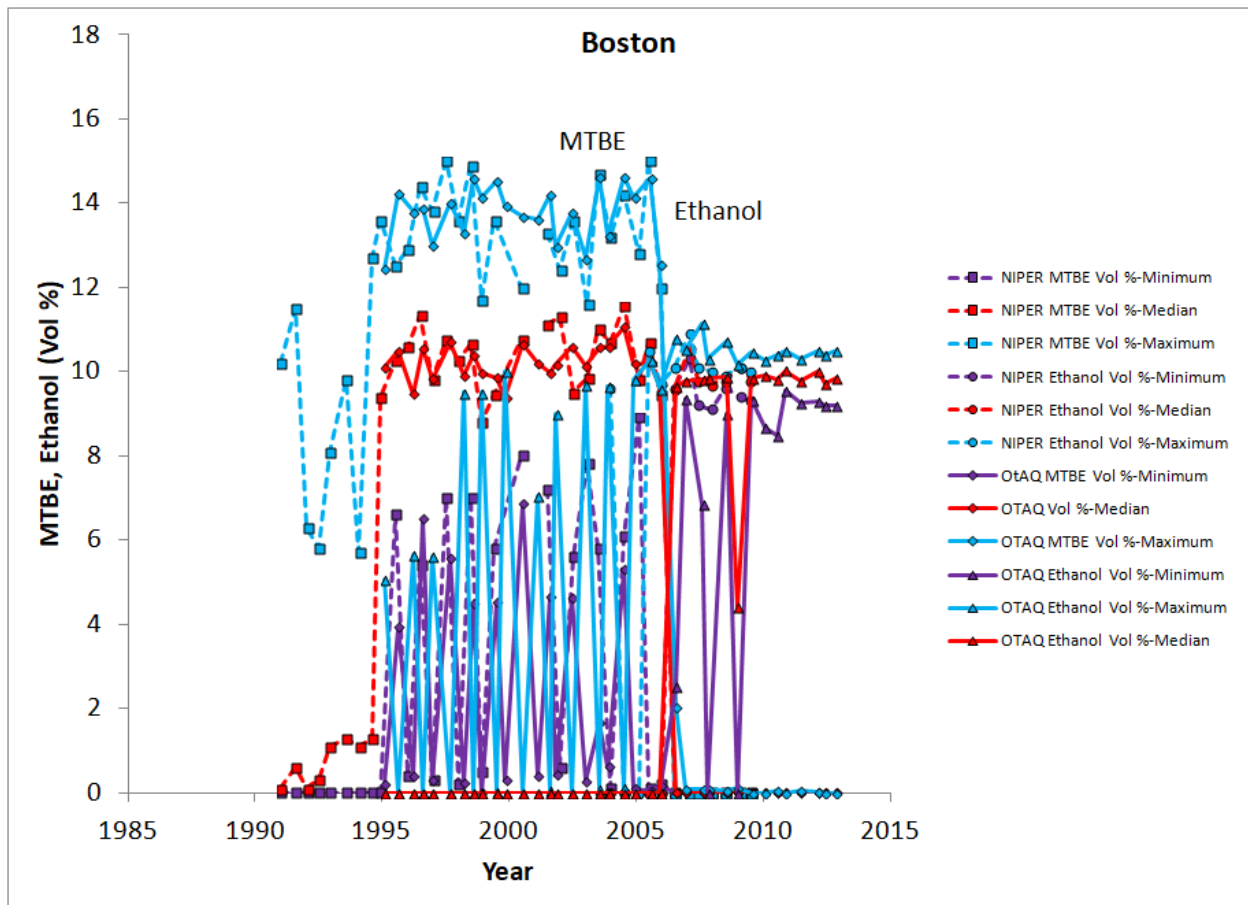


Figure 26: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Boston-Worcester, Massachusetts area.

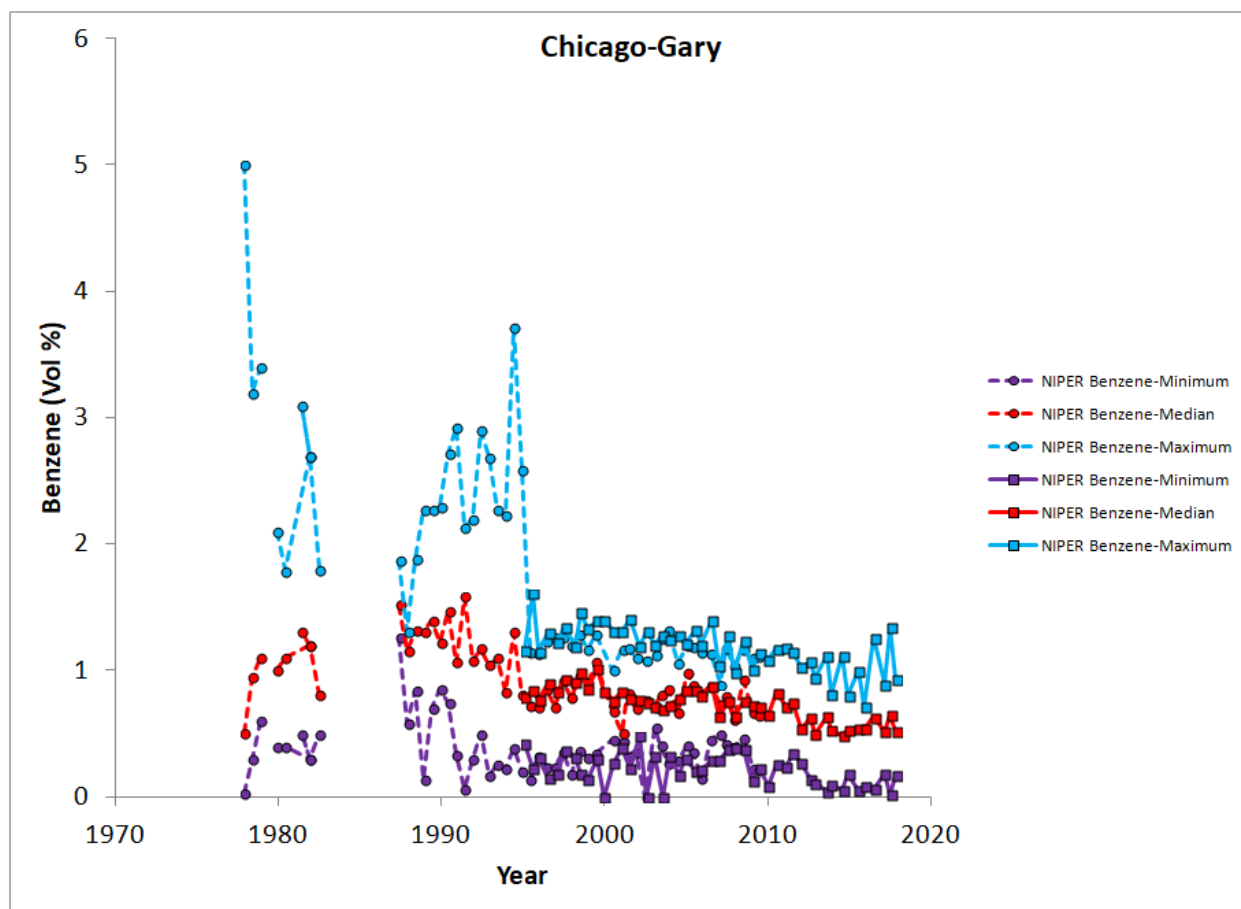


Figure 27: Benzene content in reformulated gasoline from the Chicago-Lake Co, Gary Indiana area.

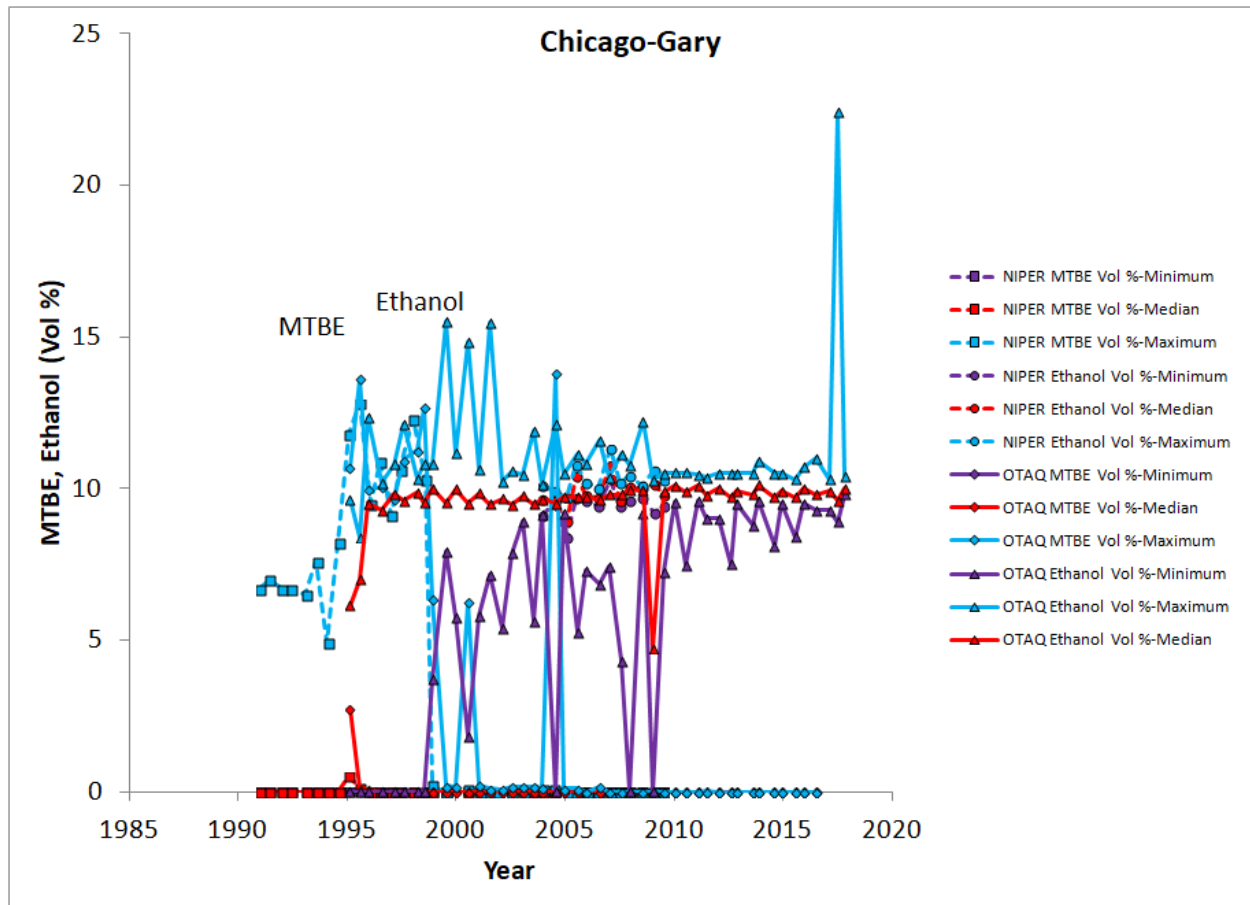


Figure 28: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Chicago Lake Co—Gary Indiana area.

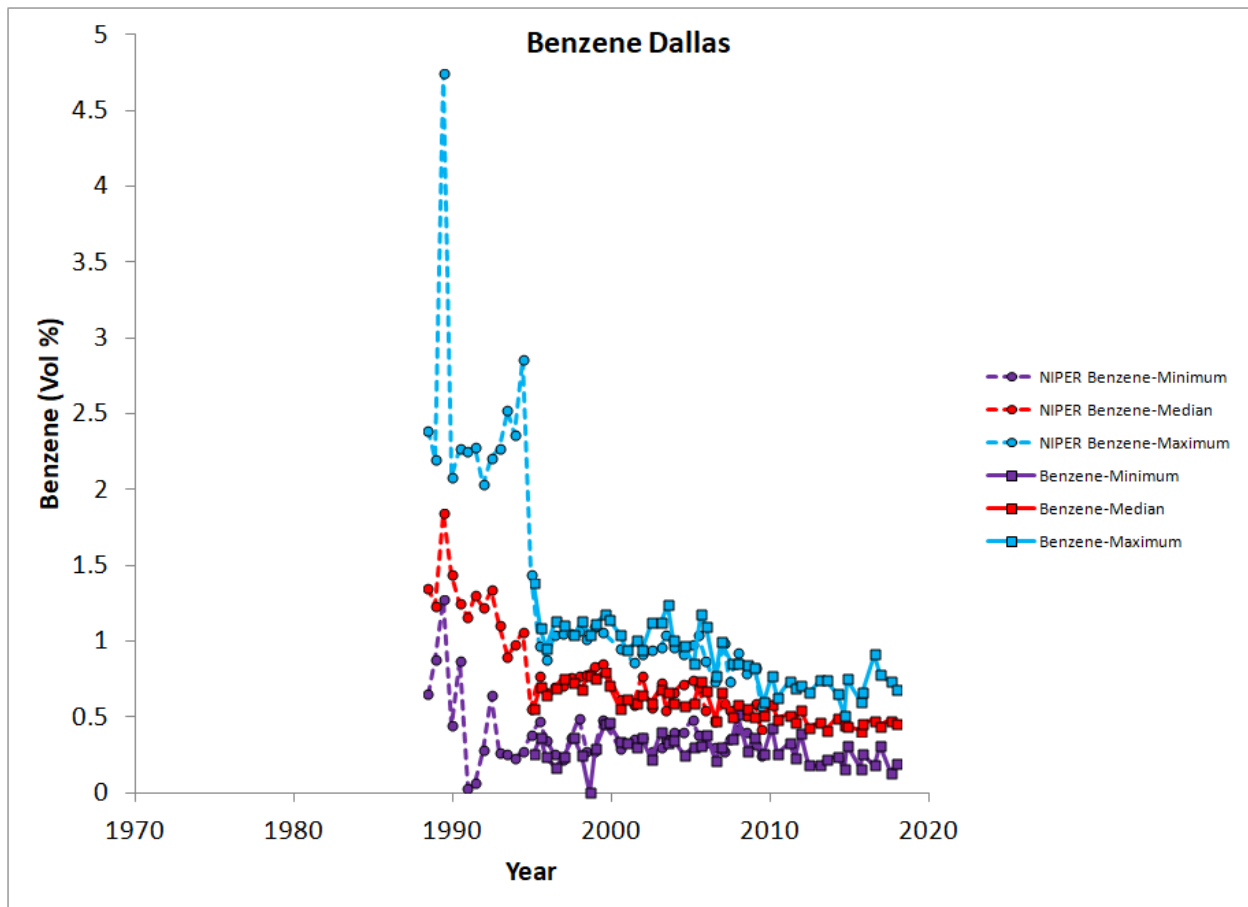


Figure 29: Benzene content in reformulated gasoline from the Dallas-Ft Worth, Texas area.

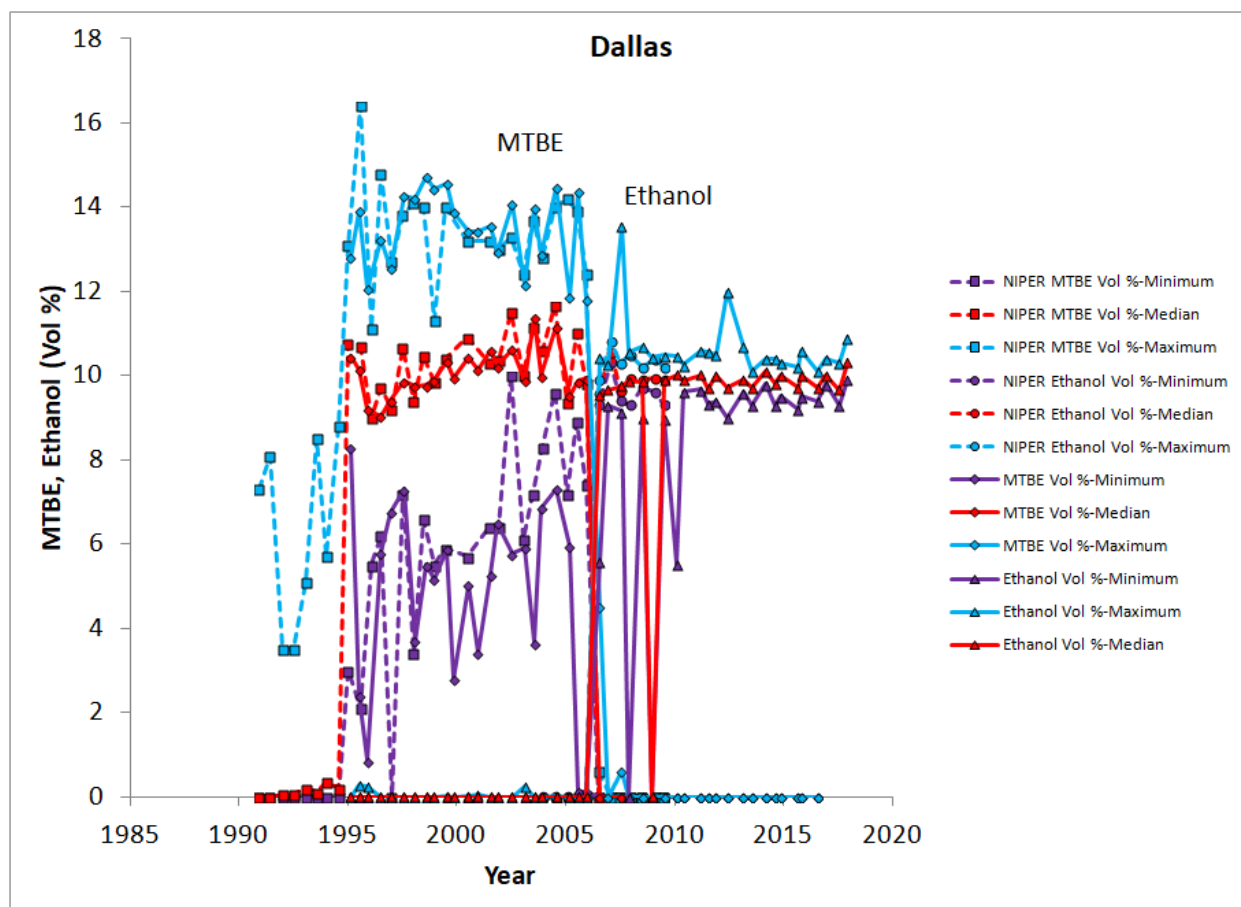


Figure 30: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Dallas—Ft Worth, Texas area.

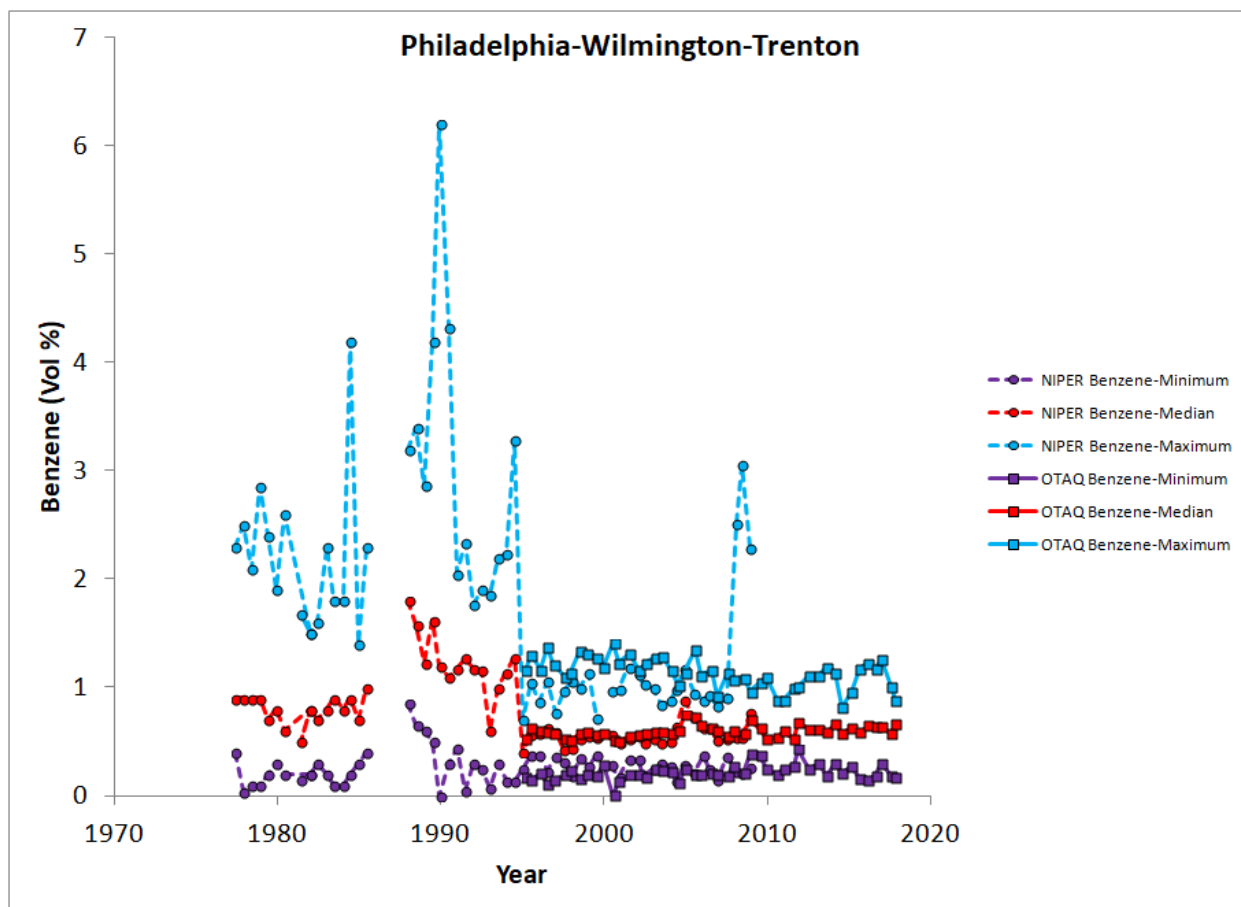


Figure 31: Benzene content in reformulated gasoline from the Philadelphia, Pennsylvania—Wilmington, Delaware—Trenton, New Jersey area.

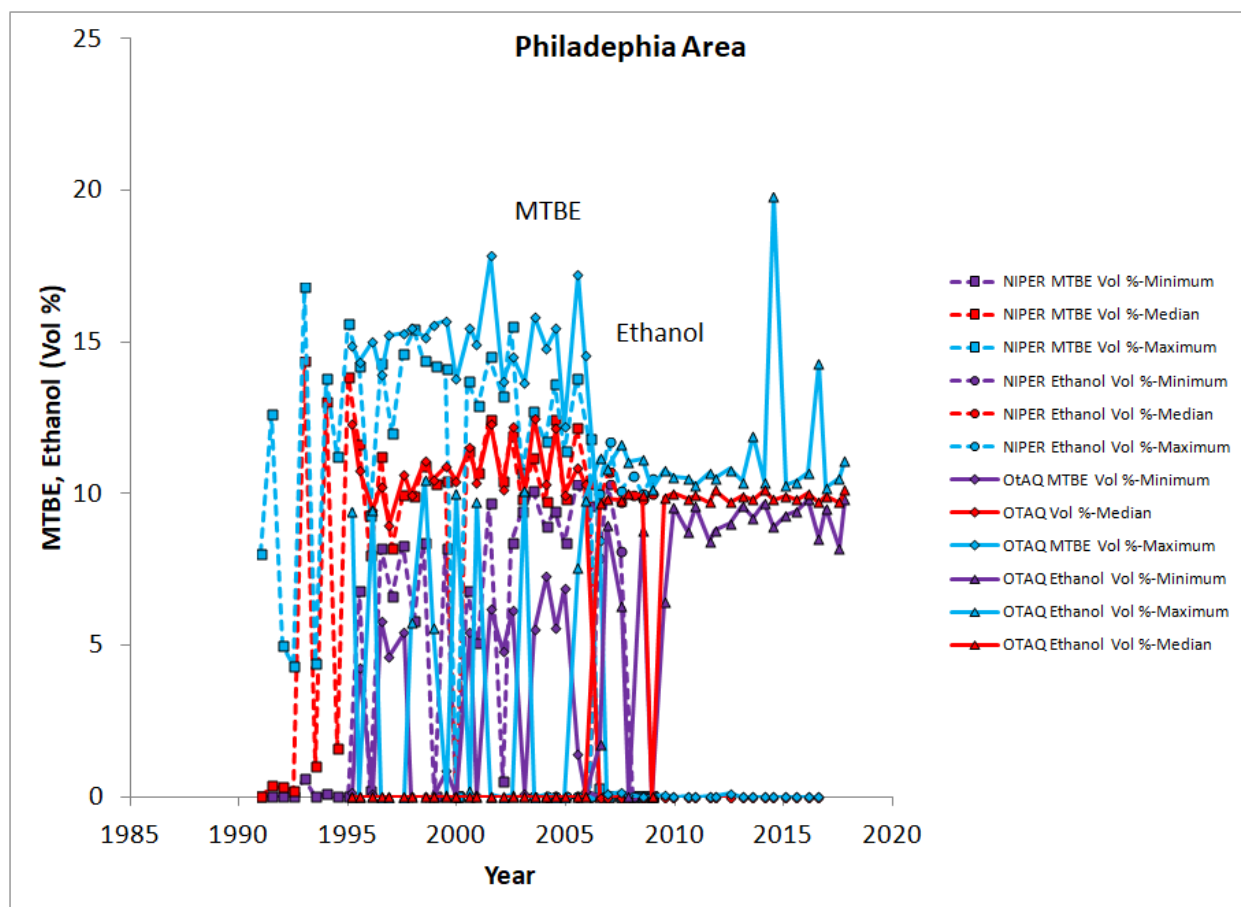


Figure 32: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Philadelphia, Pennsylvania—Wilmington, Delaware—Trenton, New Jersey area.

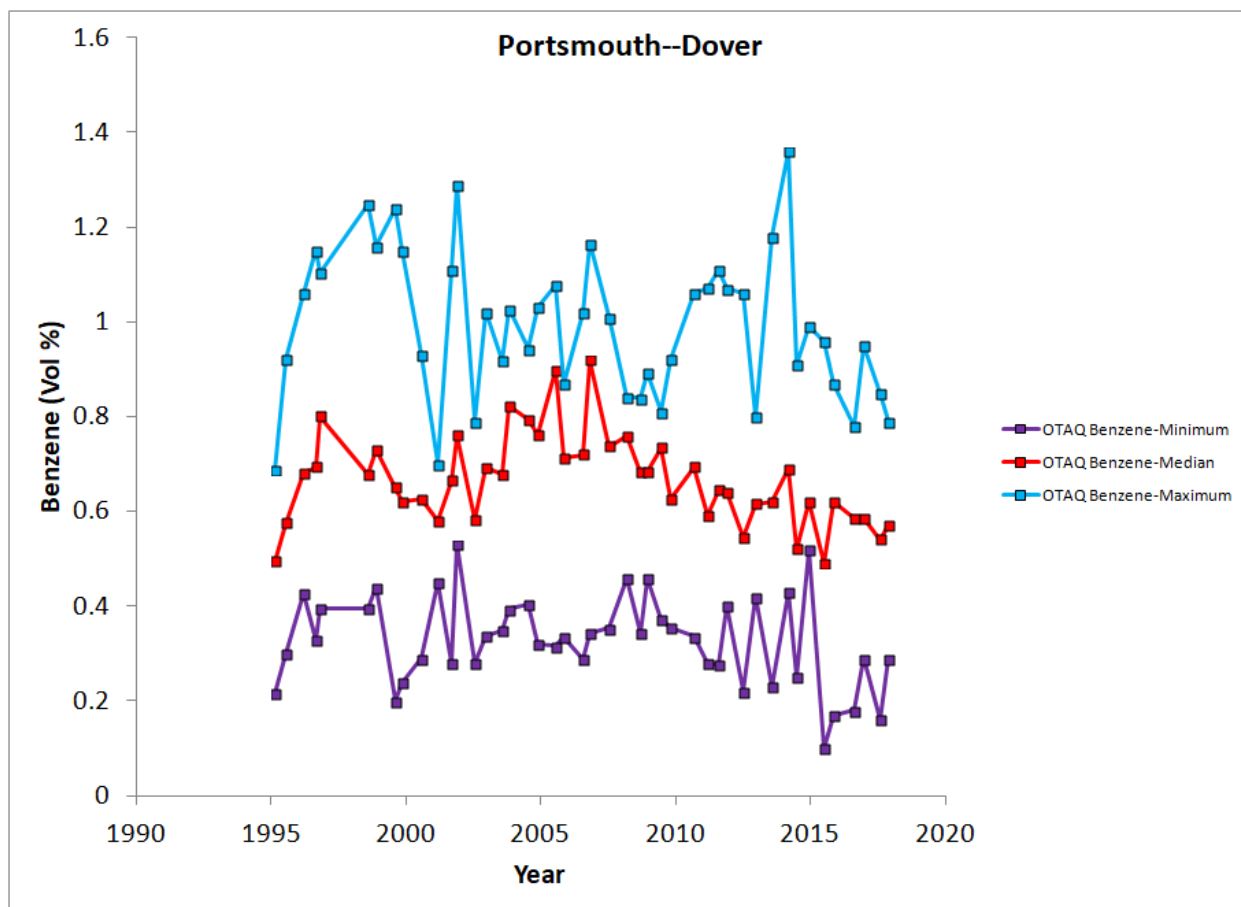


Figure 33: Benzene content in reformulated gasoline from the Portsmouth—Dover, New Hampshire area. Only data from OTAQ were available.

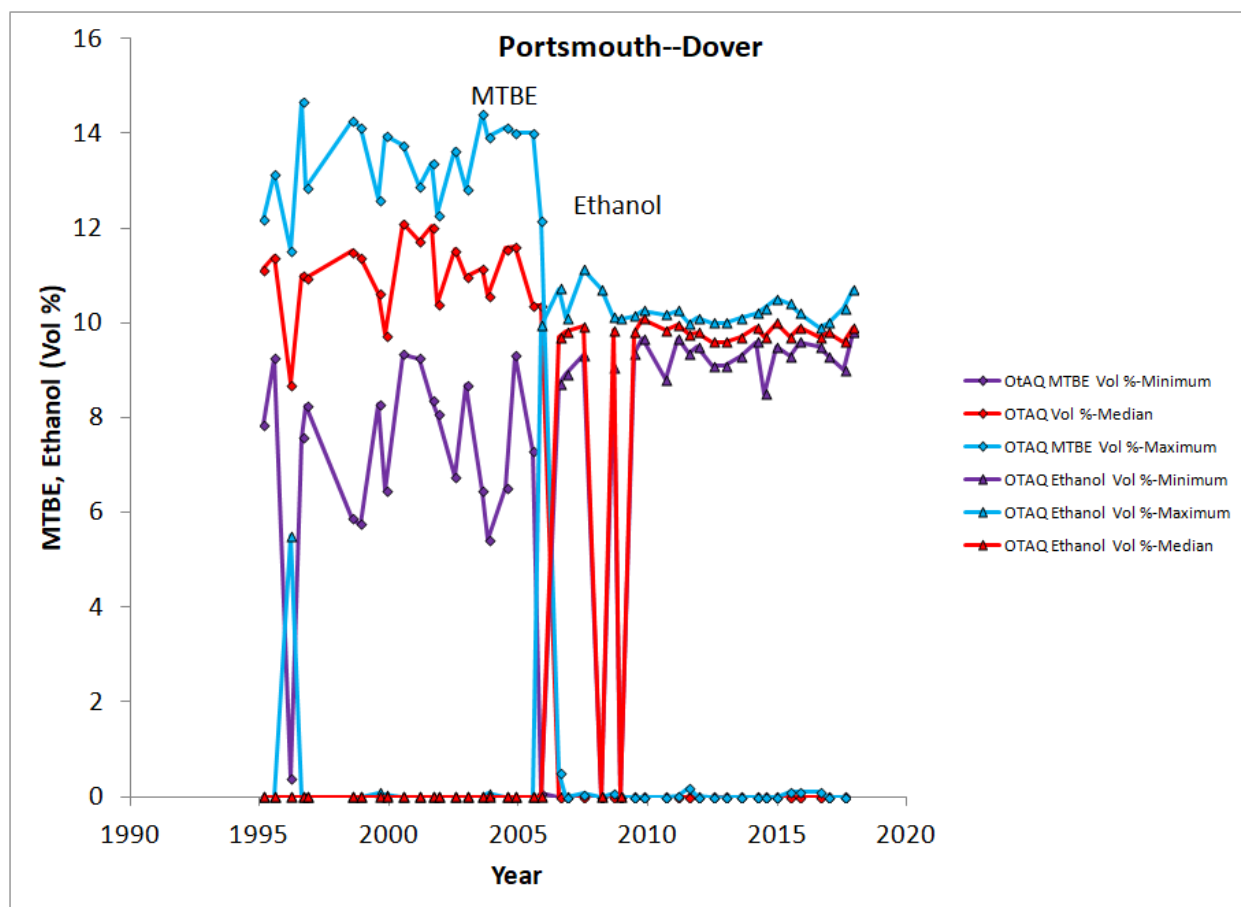


Figure 34: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Portsmouth Dover, New Hampshire area. Only data from OTAQ were available.

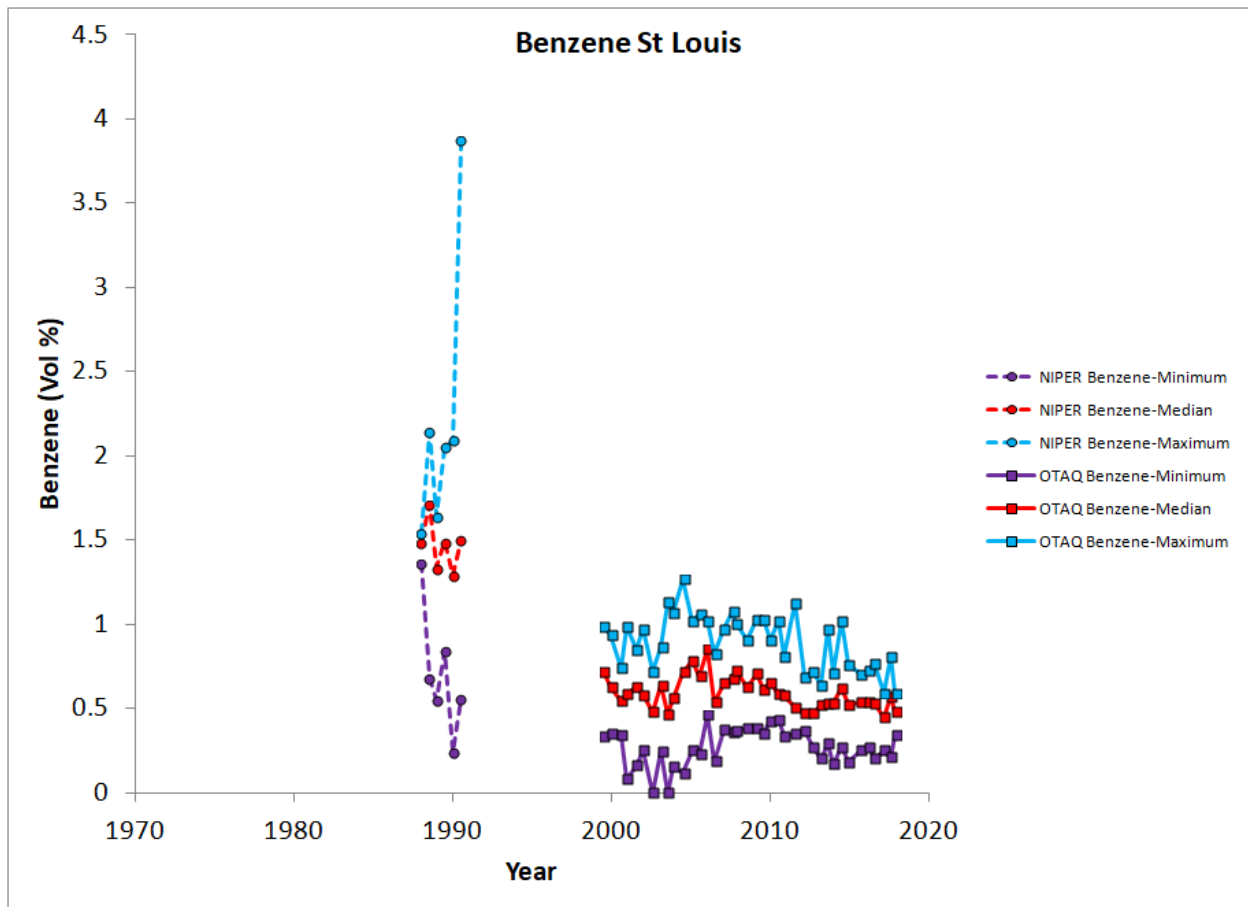


Figure 35: Benzene content in reformulated gasoline from the St Louis, Missouri area.

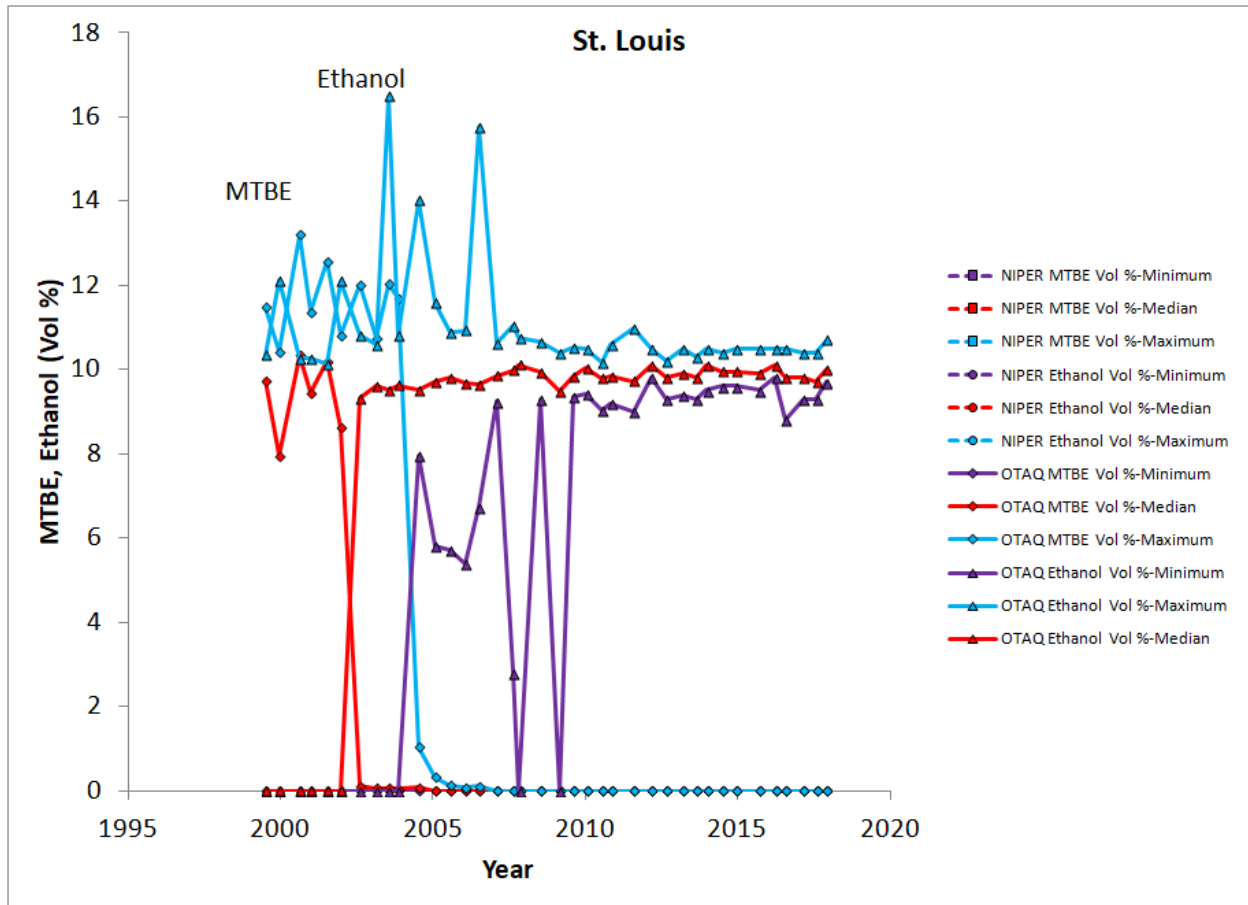


Figure 36: Oxygenates (MTBE and ethanol) in reformulated gasoline from the St. Louis Missouri area.

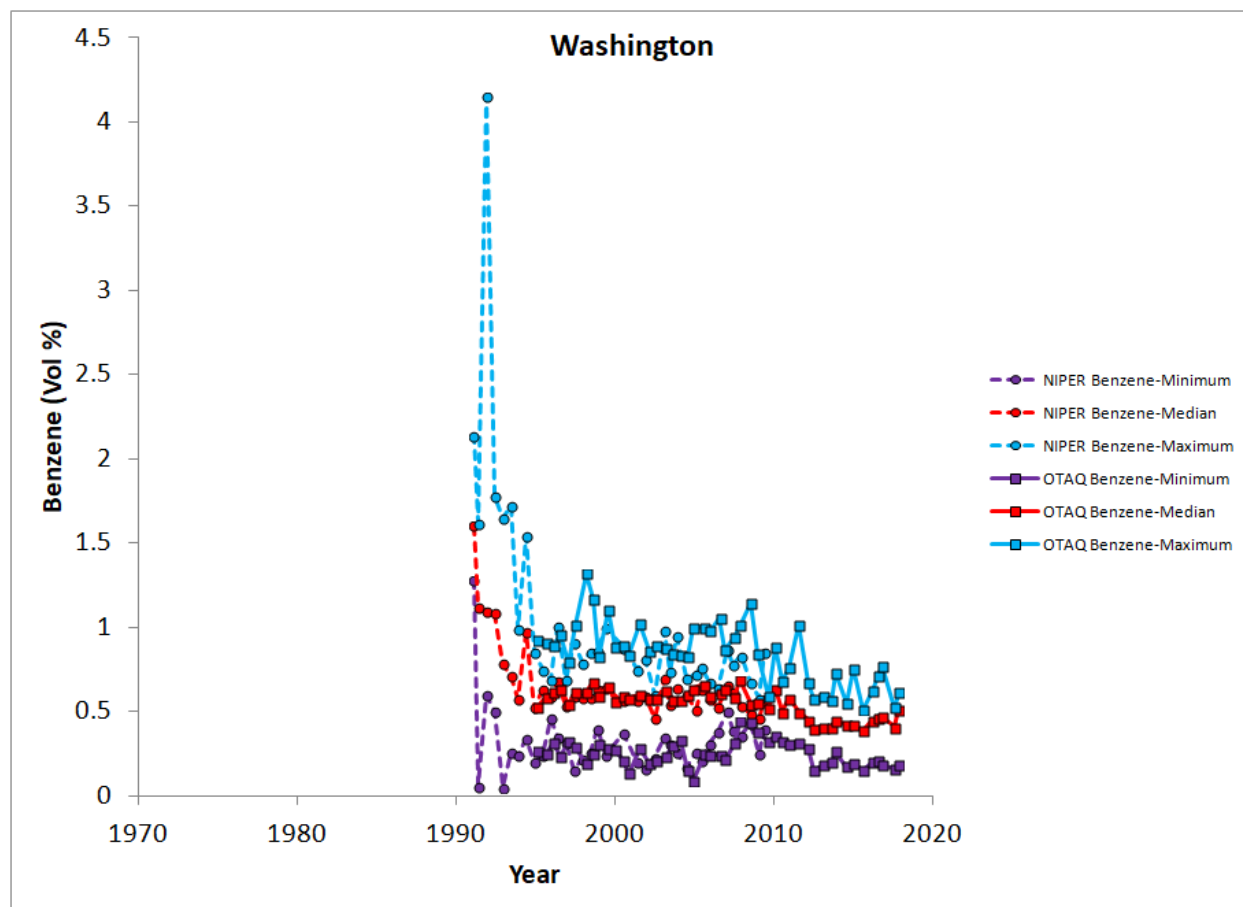


Figure 37: Benzene content in reformulated gasoline from the Washington DC area.

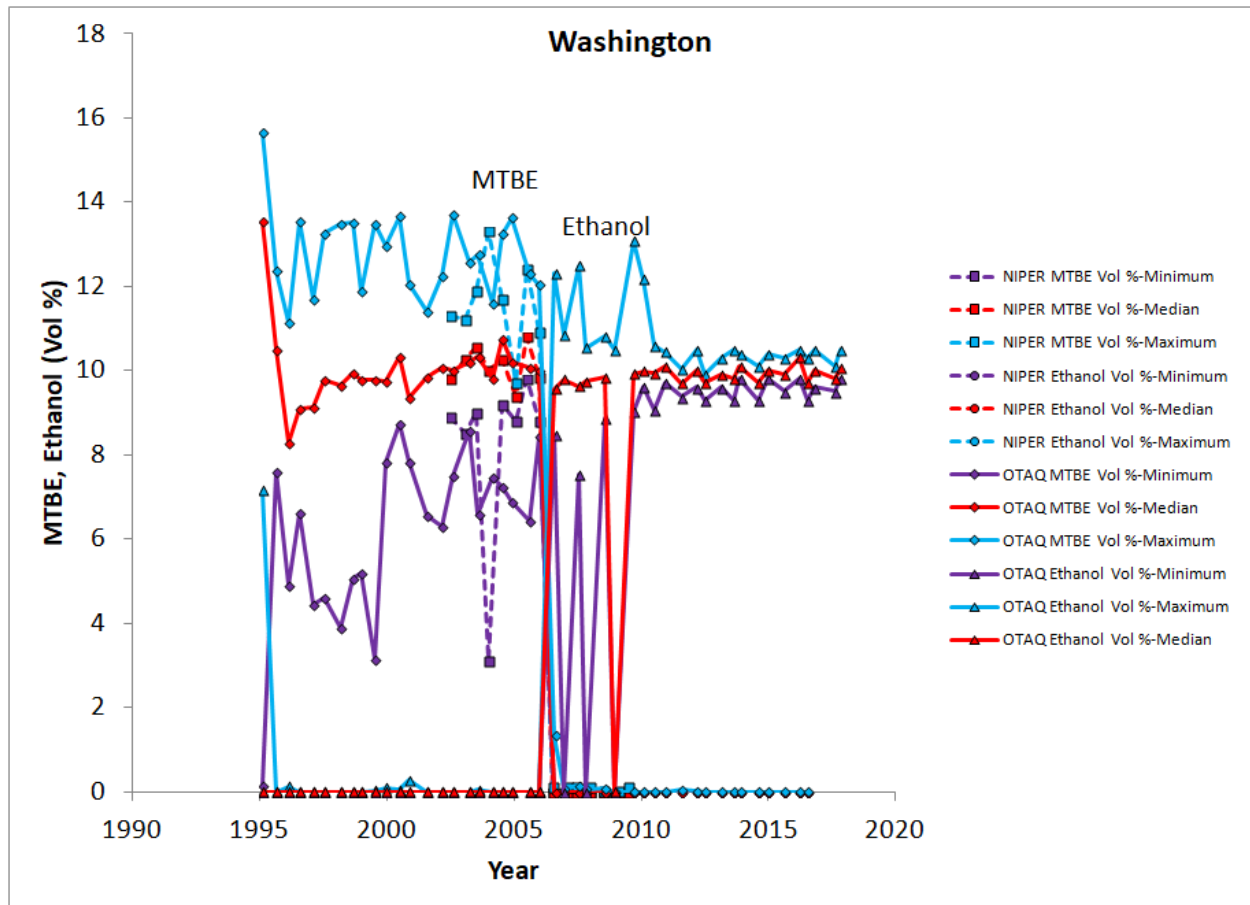


Figure 38: Oxygenates (MTBE and ethanol) in reformulated gasoline from the Washington DC area.

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