Evaluating HAP Trends: A Look at Emissions, Concentrations, and Regulation Analyses for Selected Metropolitan Statistical Areas

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ABSTRACT

EPA's annual *Trends Report*[1] is tasked with characterizing the state of air quality for the nation. Most of the reports in the past focused on criteria pollutant trends for emissions and concentrations; few analyses were conducted for hazardous air pollutants (HAPs) due to the limited amount of data available. Since the passage of the Clean Air Act Amendments in 1990, much time and resources have been directed at quantifying HAP emissions and concentrations and regulating HAP emission sources.

Historical concentration and emissions data were retrieved from EPA for select metropolitan statistical areas (MSAs) spanning from 1990-2003, for the purpose of evaluating trends for specific HAPs. Within the last 10 years, EPA has implemented several air regulations to reduce stationary and mobile source HAP emissions, and these reductions should correspond to reductions in ambient monitoring concentrations. Annual and seasonal trend graphs were generated to evaluate possible correlations between the imposition of HAP regulations and reductions in HAP emissions and ambient concentrations. Targeted HAPs for this study include: benzene, ethylbenzene, toluene, xylenes (total), acetaldehyde, formaldehyde, and some metal compounds.

INTRODUCTION

Under earlier versions of the Clean Air Act (1960, 1963, 1977), the primary legislative focus from the United States Environmental Protection Agency (EPA) was on criteria pollutants (lead, NO_x, SO_2, CO, PM, and VOCs). Extensive work on source characterization, control device options, and monitoring took place under these Acts. The vast monitoring network of ambient air instruments across the nation is used to assess the nation's air quality. These findings are reported by EPA in its annual *Trends* report. However, due to limited availability of data, hazardous air pollutant (HAP) trends were not calculated.

Since the passage of the 1990 Clean Air Act Amendments (CAAA),[2] EPA has spent considerable time and resources in establishing and enabling federal regulations to reduce emissions for HAPs. The goal of this study is to review HAP ambient monitoring, emissions, and regulation data from the last 14 years across ten metropolitan statistical areas (MSAs) with the purpose of characterizing HAP trends at each of these MSAs, and across the nation.

POLICY-RELEVANT QUESTIONS

In this type of study, it was important to have definitive conclusions based on the available data. To guide our analyses, we asked three policy-relevant questions:

1. What are the HAP concentration trends?

2. Have HAP-specific regulations been effective on reducing ambient concentrations?

3. Do HAP emissions show a decline due to HAP-specific federal regulations?

To answer these three questions, we focused our study to a select number of HAPs, specific MSAs, and a study period from 1990-2003. Detailed information is described below.

Pollutants of Interest

The 1990 CAAA shaped early monitoring programs. The amendments emphasized collecting, reporting, and regulating HAPs (also called air toxics). Air toxics are those pollutants known or suspected to cause cancer or other serious health effects. Currently, 188 HAP categories (with over 700 subspecies) are regulated under the Clean Air Act.[3] These HAPs may cause a wide variety of adverse ecosystem and health problems, including cancer, neurological effects, reproductive effects and developmental effects. Emissions from multiple sources, including major stationary, area, and mobile sources, result in population exposure to these air toxics compounds. In some cases the public may be exposed to an individual air toxic. More typically, however, people experience exposures to multiple air toxics from many emission sources. Exposures result not only from the direct inhalation of air toxics, but also from multi-pathway exposures such as drinking water contaminated from airborne deposition of HAP-laden particles, deposition on skin, and various routes to ingestion in contaminated food.

Several EPA programs have been built around subsets of the total HAPs, such as the 112(c)(6) program,[4] the 112(k) program,[5] and the core HAPs designated by EPA under the National Air Toxics Assessment (NATA).[6] Some of these programs examine only carcinogenic and/or non-carcinogenic HAPs, while others may focus on HAPs from mobile sources. For this study, we targeted the following HAPs to represent stationary and mobile sources:

HAP	Type	Cancer Compound	Noncancer Compound
Acetaldehyde	Carbonyl	X	X
Benzene	VOC	X	X
Cadmium	Metal	X	X
Ethylbenzene	VOC	X	X
Formaldehyde	Carbonyl	X	X
Lead	Metal		X
Mercury	Metal		X
Toluene	VOC		X
Xylenes (total)	VOC		X

Benzene, ethylbenzene, toluene, acetaldehyde, and formaldehyde do not have multiple isomers or species, and are also considered their own pollutant group. For pollutants that have multiple isomers or species, such as the metallic HAP compounds, lead compound, cadmium compound, and mercury compound averages were computed. For the individual xylene species, the isomer concentrations were summed to compute a total xylene value. Each of the targeted HAPs has corresponding cancer and/or noncancer toxicity factors.

MSAs of Interest

The initial list of MSAs targeted comes from ERG's work operating EPA's Urban Air Toxics Monitoring Program (UATMP). For year 2004, several state agencies from large MSAs participated in the UATMP. Previous year-end reports limited the geographic area to the monitor, thus limiting the number of years for constructing a trend. For the 2004 report, the trends analyses portion increases from the participating monitor to the monitors within the MSA, thereby potentially increasing the amount of study years available.

Of the ten MSAs ERG chose, six have monitors participating in the 2004 UATMP. Each of the six monitors also represents one of EPA's ten regions. The remaining four MSAs represent the four EPA regions not chosen from the UATMP report. The MSAs for this study are, as follows (2004 UATMP MSAs are denoted with an *):

- Region 1: (*) Boston-Cambridge-Quincy, MA-NH MSA
- Region 2: (*) New York-Northern New Jersey-Long Island, NY-NJ-PA MSA
- Region 3: (*) Philadelphia-Camden-Wilmington, PA-NJ-DE-MD MSA
- Region 4: (*) Tampa-St. Petersburg-Clearwater, FL MSA
- Region 5: (*) Detroit-Warren-Livonia, MI MSA
- Region 6: Dallas-Fort Worth-Arlington, TX MSA
- Region 7: (*) St. Louis, MO-IL MSA
- Region 8: Denver-Aurora, CO MSA
- Region 9: Los Angeles-Long Beach-Santa Ana, CA MSA
- Region 10: Seattle-Tacoma-Bellevue, WA MSA

It should be noted that although the Dallas, TX MSA and Denver, CO MSA are not participating in the 2004 UATMP, they were participants of previous UATMP years. Each MSA in this study showed moderate to substantial increases in population and vehicle miles traveled (VMT) during the time period considered (Table 1).

An MSA is defined by the counties associated with the MSA from the U.S. Census Bureau designations.[7] For example, Camden County, NJ (FIPS = 34007), in which CANJ (AQS site ID = 34-007-0003) is a UATMP monitor in that county, is part of the Philadelphia-Camden-Wilmington, PA-NJ-DE-MD MSA. According to the 2003 U.S. Census Bureau, ten other counties are part of this MSA:

- New Castle County, DE (FIPS = 10003);
- Cecil County, MD (24015);
- Burlington County, NJ (34005);
- Gloucester County, NJ (34015);
- Salem County, NJ (34033);
- Bucks County, PA (42017);
- Chester County, PA (42029);
- Delaware County, PA (42045);
- Montgomery County, PA (42091);
- Philadelphia County, PA (42101)

Time Period of Interest

Our time period for this study spanned from 1990-2003. The first HAP emission inventory developed by EPA was for the 1990 base year to coincide the passage of the 1990 CAAA. HAP emission inventories were also developed for the 1996, 1999, and 2002 base years, thus providing emissions data before and after several regulations from the CAAA were implemented. Specifically over the last 10 years, EPA has implemented several air regulations to target stationary and mobile source HAP emissions, and these reductions should correspond to reductions in ambient monitoring concentrations and emissions.

This time period also captures the period where a number of federal, state, and local agency HAP monitors and networks were placed or expanded across the nation, including the UATMP, PAMS, IMPROVE, and Pilot City. The time period also does not conflict with EPA's plans for calculating HAP trends. Beginning in 2004, EPA established a HAP monitoring network of 22 sites called the National Air Toxics Trends Subsystem (NATTS), which is to serve a similar function as the well-established criteria pollutant monitoring network.

METHODOLOGY

In calculating trends for this study, ERG retrieved three types of information from EPA: 1) HAP ambient monitoring data; 2) HAP emissions data; and 3) implemented federal stationary and mobile source rules.

Ambient Monitoring Data

The primary data sources for the HAP ambient monitoring data were from the EPA historical archive (HA),[8] the Air Quality Subsystem (AQS),[9] and from the IMPROVE[10] network. The HA contained nationwide HAP data from 1990-2000, the AQS data contained state/local/tribal-submitted data for 2001-2003, and the IMPROVE data covered specific metal HAPs from 2001-2003. Under contract to EPA, ERG compiled, supplemented, and quality-assured the three data sources into a single comprehensive database. The concentrations were standardized to $\mu g/m^3$. Additional quality assurance/quality control (QA/QC) checks were performed on a subset of the entire data set for approximately 30 HAPs.

To evaluate trends, ERG chose to calculate annual and seasonal MSA averages using a modified approach to an EPA-approved procedure.[11] The following steps were performed:

1. *Calculate pollutant group averages*. As described earlier, the metal compounds were averaged, while the xylene species were summed together.

2. *Calculate valid daily site averages from the pollutant group averages*. Most of the data in the merged database were daily samples, and no adjustments were needed. For sub-daily data (hourly, 3-hour, 6-hour, etc.), a minimum of 18 hours of sampling data within a day was needed to establish a valid daily average. Thus, if a site had seventeen 1-hour concentrations in a particular day, the average of those concentrations would not be considered a valid daily average.

3. *Calculate valid seasonal site averages from the valid daily site averages*. Each season is assumed to have 91 days. If samples were collected 1-in-6 days, then up to15 samples can be collected per season. Some sites sampled with less frequency, such as 1-in-12 days, which is less than 8 samples per season. For a site to have a valid seasonal average, a minimum of seven valid daily averages is needed.

4. *Calculate seasonal MSA averages from the valid seasonal site averages*. The valid seasonal averages for each site within the MSA were averaged together.

5. *Calculate valid annual site averages from the valid seasonal site averages*. An annual average is the average of the valid seasonal averages. A minimum of two seasons is needed to compute a valid annual average.

6. *Calculate annual MSA averages from the valid annual site averages*. The valid annual averages for each site within the MSA were averaged together.

Emissions Data

Data from the National Emissions Inventory (NEI)[12] for base years 1990, 1996, 1999, and 2002 were retrieved from EPA for the targeted HAPs. Using an approach similar to that used for the concentration data, total MSA emissions were calculated using the following steps:

1. *Retrieve emissions from NEI for base years 1990, 1996, 1999, and 2002*. Emissions data for 1990 were at the county-level, but still delineated between stationary and mobile sources. Emissions data for 1996, 1999, and 2002 contained stationary source data at the facility- and county-level.

2. *Calculate county-level emissions by HAP*. Emissions for each base year were summed to the county-level by targeted HAP. Stationary and mobile source emission types were retained.

3. *Calculate MSA emissions by HAP*. Using the MSA-county designations, the MSAs of interest were summed by HAP and emission type.

Implemented HAP Regulations

The final component for the trends analyses involves identifying implemented HAP regulations. Federal air regulations for stationary and mobile source HAPs were researched for applicability and for implementation dates (i.e., when sources need to comply). These dates were compiled for over sixty regulations which affect our time period. All legislation corresponded to the passing of the 1990 CAAA. Specifically, Titles I, II and III of the 1990 CAAA contain legislation to reduce HAP emissions from stationary and mobile sources:

- Under Title I, NSPS and NAAQS Programs, Solid Waste Combustion MACT Regulations and National Volatile Organic Compound Emission Standards (NVOCES) were two of several sub-programs to be enacted;

- Under Title II, Mobile Sources Program, the following sub-programs were to be enacted:

    1. Motor Vehicle Emission Standards (also called Tier I and Tier II);
    2. Fuel and Fuel Additives;
    3. Aircraft Emission Standards; and
    4. Clean-Fuel Vehicles.

- Under Title III, NESHAP Program, the following sub-programs were to be enacted:
    1. National Urban Air Toxics Strategy;
    2. NESHAP Standards (post-1990);
        A. 112(c) – Specific regulations for 8 HAPs;
        B. 112(d) – Specific regulations for 170+ MACT source categories;
        C. 112(f) – Residual Risk Program; and
        D. 112(k) – Specific regulation for 30+ HAPs
    1. State Programs; and
    2. Accidental Release Prevention Program

Of the specific sub-programs listed above, only the Solid Waste Combustion MACT (Section 129), the National VOC Emission Standards (NVOCES, Section 183), the Motor Vehicle Emission Standards (Section 202), Fuel and Fuel Additives (Section 211), and over forty Section 112(d) NESHAPs have federal regulations implemented from 1990-2003.

To identify federal stationary source regulations for the above sub-programs, ERG used the 2002 NEI for point and area nonpoint sources for each MSA. In these inventories, MACT codes are identified for each facility (in the point inventory) or source category (in the area nonpoint inventory). Since each MSA is unique in its emission source makeup, each MSA is subject to different stationary

source regulations. Since 1990, seventy-three of 143 MACT source categories promulgated have been implemented.

Mobile source rules are generally for the entire country (e.g. – National Low Emissions Vehicles) or for specific MSAs (e.g. – Reformulated Gasoline for some ozone nonattainment areas). NVOCES categories, such as architectural surface coating, consumer products, and autobody refinishing, typically are found in all areas.

Table 2 is a summary of the CAAA federal regulations that were implemented during our study period. Sixty-four HAP-specific regulations were implemented during our study period; forty-four were implemented between 1995-2001. While it's difficult to definitively conclude that concentrations decreased due to the implementation of certain regulations, it's useful to analyze concentrations pre- and post-implementation to conclude an apparent affect.

RESULTS

The combination of the ambient monitoring data, emissions data, and implementation dates of stationary and mobile source rules were the basis for our trends analysis. The following sections describe the concentration and emissions trends for each MSA. Tables 3a-i provide an emissions and concentration trends summary by HAP and MSA. Due to availability of ambient monitoring data, we chose to average 1990-1994 concentrations and 2002-2003 concentrations. To evaluate a trend, we compared the average concentrations from two time periods. For each MSA and HAP, there were 57 combinations which had concentration values during both of these time periods; of those, over 85% of the HAPs measured across the ten MSAs presented a decrease in their HAP concentrations.

These sub-time periods overlap with the NEI baseyear (1990-1993) and latest year (2002) emissions inventory. When comparing emission estimates from the 1990 NEI and the most recent 2002 NEI, HAP emissions for each MSA decreased substantially; total emissions across the MSAs decreased from 578,493 tpy to 268,890 tpy (53% reduction).

Regulation impact analyses figures were constructed for each MSA and pollution group, and are posted at Eastern Research Group's FTP site[13] due to space limitations for this paper. However, for this paper, we have included two figures per MSA. Implemented regulations are coded in the figures using the graph key from Table 2.

<u>Boston MSA</u>

- The Boston MSA experienced a 7.4% increase in population and a 73.9% increase in vehicle miles traveled (VMT) from 1990-2003. This MSA participated in the winter-oxygenated program from 1992-1996 and the reformulated gasoline program during the study period.

- Although there was increase in population and VMT during the study period, the emissions for all the pollutants of interest decreased from 30% (acetaldehyde) to 84% (mercury). The VOC and metal HAPs decreased in their average concentrations, ranging from 47% (ethylbenzene) to 94% (total xylenes). Acetaldehyde and formaldehyde concentrations, in contrast, increased during our study period (+639% and +296%, respectively).

- The Boston MSA phased out of the winter-oxygenated fuel program in 1996, but still participates in the reformulated gasoline (RFG) program. According to Figures 2 and 3, acetaldehyde and formaldehyde concentrations appeared to increase after the implementation of RFG Phase II and the POTW MACT. Since emissions for these HAPs have decreased

substantially, this may suggest that they are forming as secondary pollutants. Research has shown increases in carbonyl concentrations due to implementations of the RFG Program (Phase I and Phase II).[14]

New York City MSA

- The New York MSA experienced a 10.5% increase in population and a 28.9% increase in VMT from 1990-2003. This MSA participated in the winter-oxygenated program and the reformulated gasoline program during the study period.

- Although there was increase in population and VMT during the study period, the emissions for all the pollutants of interest decreased from 29% (total xylenes) to 84% (mercury). Similarly, all the pollutants of interest decreased in their average concentrations, ranging from 24% (cadmium) to 95% (mercury).

- Benzene concentrations in the New York MSA decreased, as shown in Figure 4, after implementation of mobile source rules (winter-oxygenated fuel and reformulated gasoline). The federal rules targeting stationary sources (Degreasing, Gasoline Distribution Stage I, etc.) did not appear to reduce benzene concentrations.

- Mercury concentrations appeared to decrease after implementation of the Petroleum Refineries MACT, Reformulated Gasoline Phase II program, and the Large Municipal Waste Combustors MACT (Figure 5).

Philadelphia MSA

- The Philadelphia MSA experienced a 6.2% increase in population and a 56.6% increase in VMT from 1990-2003. This MSA participated in the winter-oxygenated program and the reformulated gasoline program during the study period.

- Although there was an increase in population and VMT during the study period, the emissions for all the pollutants of interest, except for cadmium, decreased from 49% (lead) to 71% (mercury). Cadmium emissions increased 114%. All the pollutants of interest, except cadmium, decreased in their average concentrations, ranging from 65% (benzene) to 99% (lead). Average cadmium concentrations increased from 0.45 ng/m^3 to 4.58 ng/m^3.

- Cadmium compound concentrations appeared to increase throughout the study period in the Philadelphia MSA (Figure 6), however, limited or no data are available from 1998-2000.

- As shown in Figure 7, lead compound concentrations appeared to decrease substantially after implementation of the Secondary Lead Smelter MACT and the Gasoline Distribution Stage I MACT.

Tampa Bay MSA

- The Tampa Bay MSA experienced a 22.4% increase in population and a 72.8% increase in VMT from 1990-2003. This MSA does not participate in either the winter-oxygenated program or the reformulated gasoline program.

- Although there was an increase in population and VMT during the study period, the emissions for all the pollutants of interest decreased, ranging from 22% (benzene) to 96% (cadmium). Lead was the only HAP to have a 1990-1994 average calculated, and that concentration decreased by 61%. Limited or no data were available for the other HAPs during the1990-1994 time period.

- According to Figure 8, lead compound concentrations appeared to decrease overall during the study period, most notably after the Secondary Lead Smelting MACT.

- Although toluene concentrations have decreased during the study period (Figure 9), there is limited toluene data in the Tampa MSA for the last five years, thereby making it difficult to establish a trend.

Detroit MSA

- The Detroit MSA experienced a 5.5% increase in population and a 28.9% increase in VMT from 1990-2003. This MSA does not participate in either the winter-oxygenated program or the reformulated gasoline program.

- Although there was an increase in population and VMT during the study period, the emissions for all the pollutants of interest, except cadmium and lead, decreased ranging from 32% (benzene) to 79% (mercury). Cadmium and lead emissions increased by 36% and 67%, respectively. Concentrations for the VOC HAPs and for lead decreased during the study period, ranging from 9% (ethylbenzene) to 39% (lead). Limited or no data were available for the acetaldehyde, cadmium, formaldehyde, or mercury during the1990-1994 time period.

- Lead concentrations in Detroit appeared to decrease steadily during the study period (Figure 10). The biggest effect appears to be related to the implementation of the Large Municipal Waste Combustors MACT.

- Although total xylene concentrations had little variance throughout the 1990s (Figure 11), it is unclear how stationary source rules affected xylene concentrations due to the limited data availability of xylene measurements from 1997-2000, when several regulations were implemented.

Dallas MSA

- The Dallas MSA experienced a 40.1% increase in population and a 36.1% increase in VMT from 1990-2003. This MSA participated in the reformulated gasoline program during the study period.

- Although there was an increase in population and VMT during the study period, the emissions for all the pollutants of interest decreased from 36% (total xylenes) to 83% (cadmium). All the pollutants of interest decreased in their average concentrations, ranging from 36% (benzene and lead) to nearly 100% (mercury). Limited or no data were available for acetaldehyde, cadmium, or formaldehyde during the1990-1994 time period.

- Benzene concentrations in Dallas show a downward trend (Figure 12), primarily in response to implementation of Tier 1 Mobile Standards.

- Mercury compound concentrations appeared to have decreased substantially with the implementation of the Reformulated Gasoline Phase 1 Program (Figure 13). However, according to the NEI, mercury emissions from Hazardous Waste Combustors decreased by 97% from 1996 to 2002 for this MSA, most likely as a result of impending regulations.

St. Louis MSA

- The St. Louis MSA experienced a 6.1% increase in population and a 37.9% increase in VMT from 1990-2003. This MSA participated in the reformulated gasoline program during the study period.

- Although there was an increase in population and VMT during the study period, the emissions for all the pollutants of interest decreased from 46% (acetaldehyde) to 89% (lead). All the pollutants of interest, except acetaldehyde, ethylbenzene, and formaldehyde decreased in their average concentrations, ranging from 21% (toluene) to nearly 89% (total xylenes). Average acetaldehyde, ethylbenzene, and formaldehyde concentrations increased 79%, 9%, and 222%, respectively.

- Overall, cadmium compound concentrations in the St. Louis MSA appeared to have decreased during the study period (Figure 14). The implementation of the Reformulated Gasoline Phase 2 Program and the Primary Lead Smelting MACT coincide with these reductions. However, no implemented regulations explain the increases and decreases in the 1996, 1997, and 1999 concentrations.

- Total xylene concentrations in the St. Louis MSA declined dramatically in apparent response to several implemented stationary and mobile source regulations targeting VOCs (Figure 15). However, little or no xylene data measeurements between 1995 and 2000 limit the certainty of these conclusions.

Denver MSA

- The Denver MSA experienced a 39.4% increase in population and a 75.2% increase in VMT from 1990-2003. This MSA participated in the winter-oxygenated program, but not the reformulated gasoline program during the study period.

- Although there was an increase in population and VMT during the study period, the emissions for all the pollutants of interest decreased ranging from 11% (lead) to 97% (mercury). Cadmium and lead were the only HAPs to have 1990-1994 averages calculated, and those concentrations decreased by 90% and 54%, respectively. Limited or no data were available for the other HAPs during the1990-1994 time period.

- As shown in Figure 16, over the last four years, acetaldehyde concentrations appeared to decrease after implementation of mobile source rules (winter-oxygenated fuel program and the National Low Emissions Vehicle Program Phase II).

- For the cadmium trend (Figure 17), no implemented stationary source regulations were identified in the Denver MSA which affected cadmium concentrations. To understand this decline, we reviewed the NEI and Toxic Release Inventory[15] from 1990-1994 for cadmium emissions. One facility, Asarco, Inc. Globe Plant, which is a cadmium refining and cadmium oxide production plant reported emissions in 1990 as 0.198 tpy. By 1994, the emissions decreased to 0.077 tpy, a 61% decrease in emissions. Emissions at this plant through 2002 remain constant. It's likely

that the decrease in cadmium emissions from this plant contributed to the decrease in ambient cadmium concentrations.

Los Angeles MSA

- The Los Angeles MSA experienced a 13.8% increase in population and a 15.7% increase in VMT from 1990-2003. This MSA also participated in the winter-oxygenated program and the reformulated gasoline program during the study period.

- Although there was an increase in population and VMT during the study period, the emissions for all the pollutants of interest decreased from 40% (acetaldehyde) to 81% (mercury). Similarly, all the pollutants of interest decreased in their average concentrations, except for cadmium and formaldehyde, ranging from 24% (acetaldehyde) to 85% (lead). Average cadmium concentrations increased from 0.78 ng/m^3 to 3.79 ng/m^3, while formaldehyde concentrations increased by 65%.

- Ethylbenzene concentrations in the Los Angeles MSA declined steadily in apparent response to several implemented stationary and mobile source regulations targeting VOCs (Figure 18).

- Formaldehyde concentrations have steadily increased during the study period (Figure 19). This is most likely due to the implementations of the RFG Program (Phase I and Phase II), which creates formaldehyde secondarily.

Seattle MSA

- The Seattle MSA experienced a 22.8% increase in population and a 27.1% increase in VMT from 1990-2003. This MSA participated in the winter-oxygenated program from 1992-1996, but not the reformulated gasoline program.

- Although there was an increase in population and VMT during the study period, the emissions for all the pollutants of interest decreased ranging from 25% (ethylbenzene and total xylenes) to 91% (cadmium and mercury). Lead was the only HAP to have a 1990-1994 average calculated, and that concentration decreased by 96%. Limited or no data were available for the other HAPs during the1990-1994 time period.

- Over the last three years, formaldehyde concentrations appeared to have increased, noticeably after the implementation of the Publicly Owned Treatment Works MACT (Figure 20). However, formaldehyde data prior to 2001 are limited or unavailable, thus making it difficult to characterize a trend.

- During the study period, lead concentrations appeared to have decreased (Figure 21). After implementation of the Petroleum Refineries and Aerospace Manufacturing MACTs, concentrations decreased substantially. Interestingly, lead concentrations increased after the prohibition of leaded gasoline.

CONCLUSIONS

Nine HAPs were analyzed for MSAs across the United States. Each MSA was chosen from one of EPA's ten regions. ERG used a database consolidated from EPA's Historical Archive, the IMPROVE network, and from the Air Quality Subsystem to calculate annual average concentrations. Emissions for each HAP were retrieved from EPA's National Emissions Inventory, 1990 to 2002.

Information on federal regulations were researched from the 1990 Clean Air Act Amendments (CAAA). Three policy-relevant questions were used to guide our study, and we answer these questions below:

- ***What are the hazardous air pollutant (HAP) concentration trends?*** When comparing concentrations between 1990-1994 and 2002-2003, over 85% of the MSA-HAP combinations measured across the ten MSAs realized a decrease in their HAP concentrations, while less than 15% realized an increase. This observation would suggest that most HAPs had a decreasing trend during the study period. For example, lead compound concentrations decreased in all ten MSAs (range 36% to 99%), while benzene decreased in seven (range 19% to 79%). However, acetaldehyde, cadmium compound, ethylbenzene, and formaldehyde each had at least one MSA that computed an increasing trend.

- ***Have HAP-specific regulations been effective on reducing ambient concentrations?*** Sixty-four HAP-specific regulations were implemented between 1992-2003. During that time period, most HAP concentrations decreased, suggesting a correlation between the two. The most effective regulations on pollutant types, based on visual inspection of the regulation impact analysis figures, were:

    1. VOCs: Reformulated Gasoline Phase I, VOC rules, Printing/Publishing MACT, Tier 1 Mobile Source Standards, Reformulated Gasoline Phase II

    2. Carbonyls: Reformulated Gasoline Phase I, National Low Emissions Vehicle Program Phase II, Pharmaceuticals Production

    3. Metals: Prohibition of Leaded Gasoline, Aerospace Manufacturing MACT, Petroleum Refineries MACT, Reformulated Gasoline Phase II, Large Municipal Waste Combustors MACT, Secondary Lead Smelter MACT, Stage I Gasoline Distribution MACT, Primary Lead Smelter MACT

- ***Do HAP emissions show a decline due to HAP-specific federal regulations?*** When comparing emission estimates from the 1990 NEI and the most recent 2002 NEI, HAP emissions for each MSA decreased substantially. Total emissions across the ten MSAs for each HAP decreased from 578,493 tpy to 268,890 tpy (53% reduction). Emissions in the Los Angeles MSA decreased the most among the MSAs (69% reduction, 86,447 tpy). For the HAPs, mercury emissions realized the highest percent reduction (80%), while toluene emissions realized the highest mass reduction (141,469 tpy). Between 1991 and 2001, over 40 HAP-specific regulations were implemented, suggesting that HAP emissions declined due to these regulations.

Using the above observations, the implementation of HAP-specific federal regulations coincides favorably with reductions in emissions and ambient concentrations for benzene, ethylbenzene, lead compounds, mercury compounds, toluene, and xylenes.

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KEYWORDS

Hazardous Air Pollutants (HAPs)
HAP Concentration Trends
Regulation Analysis
HAP Emission Trends
Metropolitan Statistical Areas
Ambient Air Monitoring
Ambient Concentration Trends

Table 1. Population and Vehicle Miles Traveled (VMT) Profiles for Each MSA

MSA	1990 MSA Population	2003 MSA Population	% Change in MSA Population	1990 MSA VMT	2003 MSA VMT	% Change in MSA VMT	Winter-Oxygenated Time Period Implemented	MSA Reformulated Gasoline Designation
Boston MSA	4,133,895	4,439,971	+ 7.4%	18,738,370	32,582,820	+ 73.9%	1992-1996	Opt-In
New York MSA	16,863,671	18,640,775	+ 10.5%	82,128,650	105,869,710	+ 28.9%	1992-2000	Required
Philadelphia MSA	5,435,550	5,772,947	+ 6.2%	24,002,035	37,576,750	+ 56.6%	1992-1996	Required
Tampa Bay MSA	2,067,959	2,531,908	+ 22.4%	12,304,150	21,258,330	+ 72.8%	NA	NA
Detroit MSA	4,248,699	4,483,853	+ 5.5%	28,551,395	36,788,715	+ 28.9%	NA	NA
Dallas MSA	3,989,294	5,589,670	+ 40.1%	29,273,000	39,848,145	+ 36.1%	NA	Opt-In
St. Louis MSA	2,599,893	2,759,440	+ 6.1%	16,530,120	22,794,250	+ 37.9%	NA	Opt-In
Denver MSA	1,650,489	2,301,116	+ 39.4%	9,909,750	17,358,670	+ 75.2%	1992-2003	NA
Los Angeles MSA	11,273,720	12,829,272	+ 13.8%	91,495,645	105,856,570	+ 15.7%	1992-2003	Required
Seattle MSA	2,559,136	3,141,777	+ 22.8%	19,203,015	24,413,025	+ 27.1%	1992-1996	NA

Table 2. Implemented Federal Regulations from 1990 Clean Air Act Amendments, 1992-2003

Graph Key	Regulation	Implementation Date	Targeted HAPs[1]
a	Winter-Oxygenated, Season 1	11/1/1992	V, C, M
b	Winter-Oxygenated, Season 2	11/1/1993	V, C, M
c	Winter-Oxygenated, Season 3	11/1/1994	V, C, M
d	Reformulated Gasoline (RFG) – Stage I	1/1/1995	V, C, M
e	Winter-Oxygenated, Season 4	11/1/1995	V, C, M
f	Coke Ovens	12/31/1995	V
g	Prohibition of Leaded Gasoline for Motor Vehicles	1/1/1996	M
h	Chromium Electroplating	1/25/1996	V
i	Industrial Cooling Towers	3/8/1996	NA
j	Final phase-in of Tier 1 Standards	8/1/1996	V, C
k	Dry Cleaners	9/23/1996	NA
l	Winter-Oxygenated, Season 5	11/1/1996	V, C, M
m	Magnetic Tape (surface coating)	12/15/1996	NA
n	Shipbuilding and Ship Repair (surface coating)	12/16/1996	V
o	Polymers and Resins Manufacturing I	7/31/1997	NA
p	Polymers and Resins Manufacturing IV	7/31/1997	V, M
q	Winter-Oxygenated, Season 6	11/1/1997	V, C, M
r	Wood Furniture (surface coating)	11/21/1997	V, C
s	Degreasing Organic Cleaners	12/2/1997	V
t	Gasoline Distribution Stage I	12/15/1997	V, C
u	Secondary Lead Smelting	12/23/1997	M
v	National Low Emissions Vehicle Program – Stage I	8/1/1998	V, C
w	Petroleum Refineries	8/18/1998	V, C, M
x	Aerospace Manufacturing (surface coating)	9/1/1998	V, C, M
y	Winter-Oxygenated, Season 7	11/1/1998	V, C, M
z	National VOC Emission Standard for Consumer Products	12/10/1998	V, C
A	National VOC Emission Standard for Autobody Refinishing	1/1/1999	V, C
B	Hazardous Organic NESHAP	5/12/1999	V, C
C	Printing and Publishing (surface coating)	5/30/1999	V, C, M
D	California Low Emissions Vehicle Program – Stage I	8/1/1999	C
E	National VOC Rule for Architectural Surface Coating	9/13/1999	V, C
F	Marine Vessel Loading	9/19/1999	V
G	Primary Aluminum Manufacturing	10/7/1999	V
H	Winter-Oxygenated, Season 8	11/1/1999	V, C, M
I	Reformulated Gasoline (RFG) – Stage II	1/1/2000	V, C, M
J	Off-Site Waste Recovery Operations	2/1/2000	NA
K	National Low Emissions Vehicle Program – Stage II	8/1/2000	V, C
L	Winter-Oxygenated, Season 9	11/1/2000	V, C, M
M	Municipal Waste Combustors – Large Units	12/19/2000	C, M
N	Pulp and Paper I	4/15/2001	V, M
O	Pulp and Paper II	4/16/2001	V, M
P	Primary Lead Smelting	6/4/2001	M
P1	Steel Pickling – HCl Process	6/22/2001	NA
Q	Pharmaceuticals Production	9/21/2001	V, C, M
R	Flexible Polyurethane Foam Production	10/8/2001	V
S	Winter-Oxygenated, Season 10	11/1/2001	V, C, M
T	Ferroalloys Production	11/21/2001	NA
U	Mineral Wool Production	6/1/2002	NA
V	Polyether Polyols Production	6/1/2002	NA
W	Phosphate Fertilizer Production	6/10/2002	NA
X	Phosphoric Acid Manufacturing	6/10/2002	M
Y	Portland Cement Manufacturing	6/14/2002	V, C, M
Z	Wool Fiberglass Manufacturing	6/14/2002	V, C

Table 2. Implemented Federal Regulations from 1990 Clean Air Act Amendments, 1992-2003 (Continued)

Graph Key	Regulation	Implementation Date	Targeted HAPs[1]
0	Natural Gas Transmission and Storage	6/17/2002	V, C
1	Oil and Natural Gas Production	6/17/2002	V, C, M
2	Generic MACT	6/29/2002	M
3	Hospital, Medical, Infectious Waste Incinerators	9/15/2002	M
4	Publicly Owned Treatment Works	10/26/2002	V, C, M
5	Winter-Oxygenated, Season 11	11/1/2002	V, C, M
6	Polymers and Resins Production III	1/20/2003	V, C, M
7	Secondary Aluminum Production	3/24/2003	V, C, M
8	Hazardous Waste Combustion	9/30/2003	M
9	Winter-Oxygenated, Season 12	11/1/2003	V, C, M
10	Pesticide Active Ingredients Manufacture[2]	12/23/2003	V

[1] = NA: Not Applicable; V = VOC HAPs; C = carbonyl HAPs; M = metal HAPs

[2] = The implementation date for this MACT, although occurring in 2003, would be considered part of the winter 2004 time period, and was not be considered for this study.

Table 3a. Acetaldehyde Emission (tpy) and Concentration ($\mu g/m^3$) Comparison

MSA	1990 Emissions	2002 Emissions	% Change in Emissions	1990-1994 Average Concentration	2002-2003 Average Concentration	% Change in Concentration
Boston MSA	1,130	791	-30.0%	2.76 ± 0.53	20.40 ± 5.27	+639.1%
New York MSA	3,077	1,320	-57.1%	4.21 ± 1.28	1.89 ± 0.12	-55.0%
Philadelphia MSA	1,236	617	-50.1%	3.37 ± 0.45	0.95 ± 0.17	-71.7%
Tampa Bay MSA	543	360	-33.6%	NA	2.11 ± 0.09	NA
Detroit MSA	1,179	629	-46.6%	NA	1.79 ± 0.10	NA
Dallas MSA	1,229	568	-53.8%	NA	1.75 ± 0.12	NA
St. Louis MSA	819	445	-45.6%	2.57 ± 1.21	4.60 ± 0.35	+79.4
Denver MSA	657	439	-33.2%	NA	2.47 ± 0.23	NA
Los Angeles MSA	2,418	1,448	-40.1%	4.74 ± 0.30	3.58 ± 0.25	-24.6
Seattle MSA	1,125	625	-44.4%	NA	1.70 ± 0.14	NA

Table 3b. Benzene Emission (tpy) and Concentration ($\mu g/m^3$) Comparison

MSA	1990 Emissions	2002 Emissions	% Change in Emissions	1990-1994 Average Concentration	2002-2003 Average Concentration	% Change in Concentration
Boston MSA	6,262	2,229	-64.4%	3.93 ± 0.64	0.81 ± 0.05	-79.5%
New York MSA	16,653	7,512	-54.9%	3.24 ± 0.15	1.35 ± 0.04	-58.5%
Philadelphia MSA	5,961	2,577	-56.8%	3.60 ± 0.27	1.26 ± 0.06	-64.9%
Tampa Bay MSA	3,103	2,408	-22.4%	NA	NA	NA
Detroit MSA	6,480	4,388	-32.3%	4.19 ± 0.46	3.40 ± 1.15	-18.7%
Dallas MSA	7,933	2,832	-64.3%	1.21 ± 0.13	0.78 ± 0.04	-35.8%
St. Louis MSA	4,358	2,304	-47.1%	5.16 ± 2.20	1.43 ± 0.10	-72.3%
Denver MSA	2,800	1,913	-31.7%	NA	2.75 ± 0.22	NA
Los Angeles MSA	19,762	4,168	-78.9%	8.97 ± 0.48	2.34 ± 0.08	-73.9%
Seattle MSA	5,844	4,315	-26.2%	NA	1.39 ± 0.17	NA

Table 3c. Cadmium Compound Emission (tpy) and Concentration (ng/m^3) Comparison

MSA	1990 Emissions	2002 Emissions	% Change in Emissions	1990-1994 Average Concentration	2002-2003 Average Concentration	% Change in Concentration
Boston MSA	1.0	0.4	-62.1%	NA	2.07 ± 0.23	NA
New York MSA	3.7	0.9	-75.1%	5.07 ± 0.40	3.85 ± 0.30	-24.0%
Philadelphia MSA	2.9	6.3	114.0%	0.45 ± 0.11	4.58 ± 0.51	906.6%
Tampa Bay MSA	9.0	0.3	-96.4%	NA	5.00 ± 0.79	NA
Detroit MSA	1.7	2.3	36.4%	NA	1.27 ± 0.16	NA
Dallas MSA	4.7	0.8	-83.1%	NA	3.03 ± 0.30	NA
St. Louis MSA	8.7	2.7	-69.3%	16.26 ± 3.15	3.42 ± 0.47	-79.0%
Denver MSA	0.4	0.2	-54.1%	31.73 ± 8.36	3.21 ± 0.62	-89.9%
Los Angeles MSA	2.9	1.4	-51.4%	0.78 ± 0.07	3.79 ± 1.06	385.0%
Seattle MSA	1.0	0.1	-91.1%	NA	2.00 ± 0.25	NA

Table 3d. Ethylbenzene Emission (tpy) and Concentration (μg/m^3) Comparison

MSA	1990 Emissions	2002 Emissions	% Change in Emissions	1990-1994 Average Concentration	2002-2003 Average Concentration	% Change in Concentration
Boston MSA	2,576	1,074	-58.3%	0.96 ± 0.09	0.51 ± 0.05	-47.1%
New York MSA	8,439	4,205	-50.2%	2.08 ± 0.12	0.74 ± 0.02	-64.7%
Philadelphia MSA	3,387	1,347	-60.2%	1.56 ± 0.15	0.53 ± 0.04	-66.2%
Tampa Bay MSA	1,565	1,146	-26.8%	NA	NA	NA
Detroit MSA	3,266	1,827	-44.0%	1.38 ± 0.20	1.26 ± 0.33	-8.8%
Dallas MSA	3,469	1,496	-56.9%	0.72 ± 0.11	0.26 ± 0.02	-64.4%
St. Louis MSA	2,066	1,092	-47.2%	0.76 ± 0.41	0.83 ± 0.08	9.3%
Denver MSA	1,259	785	-37.6%	NA	1.57 ± 0.18	NA
Los Angeles MSA	7,645	1,904	-75.1%	3.86 ± 0.31	1.51 ± 0.13	-60.8%
Seattle MSA	2,280	1,714	-24.8%	NA	NA	NA

Table 3e. Formaldehyde Emission (tpy) and Concentration (μg/m^3) Comparison

MSA	1990 Emissions	2002 Emissions	% Change in Emissions	1990-1994 Average Concentration	2002-2003 Average Concentration	% Change in Concentration
Boston MSA	3,687	1,795	-51.3%	3.90 ± 0.76	15.45 ± 4.82	295.7%
New York MSA	10,430	3,988	-61.8%	6.49 ± 2.41	3.15 ± 0.26	-51.5%
Philadelphia MSA	4,348	2,139	-50.8%	4.24 ± 0.31	1.68 ± 0.45	-60.5%
Tampa Bay MSA	1,745	1,081	-38.1%	NA	3.42 ± 0.44	NA
Detroit MSA	4,078	1,657	-59.4%	NA	3.98 ± 0.70	NA
Dallas MSA	4,299	1,755	-59.2%	NA	3.75 ± 0.34	NA
St. Louis MSA	2,658	1,263	-52.5%	4.05 ± 1.72	13.03 ± 1.31	221.7%
Denver MSA	2,237	1,107	-50.5%	NA	2.83 ± 0.22	NA
Los Angeles MSA	8,004	4,743	-40.7%	3.27 ± 0.19	5.38 ± 0.28	64.7%
Seattle MSA	3,180	1,879	-40.9%	NA	2.62 ± 0.56	NA

Table 3f. Lead Compound Emission (tpy) and Concentration (ng/m^3) Comparison

MSA	1990 Emissions	2002 Emissions	% Change in Emissions	1990-1994 Average Concentration	2002-2003 Average Concentration	% Change in Concentration
Boston MSA	15.1	7.0	-53.5%	47.63 ± 2.69	9.01 ± 0.65	-81.1%
New York MSA	86.2	22.3	-74.2%	109.60 ± 16.64	10.89 ± 1.45	-90.1%
Philadelphia MSA	69.8	35.6	-49.0%	848.02 ± 113.62	11.72 ± 0.87	-98.6%
Tampa Bay MSA	21.8	7.2	-66.8%	612.81 ± 83.85	236.99 ± 42.96	-61.3%
Detroit MSA	19.0	31.7	66.6%	24.93 ± 1.84	15.24 ± 0.90	-38.9%
Dallas MSA	38.1	22.9	-39.8%	124.20 ± 8.72	79.29 ± 8.46	-36.2%
St. Louis MSA	223.1	23.7	-89.4%	785.09 ± 77.69	293.77 ± 60.05	-62.6%
Denver MSA	7.7	6.9	-10.9%	71.31 ± 4.08	32.93 ± 5.04	-53.8%
Los Angeles MSA	63.5	28.4	-55.3%	125.37 ± 14.17	19.03 ± 1.88	-84.8%
Seattle MSA	16.3	3.7	-77.5%	116.45 ±19.68	4.40 ± 0.27	-96.2%

Table 3g. Mercury Compound Emission (tpy) and Concentration (ng/m^3) Comparison

MSA	1990 Emissions	2002 Emissions	% Change in Emissions	1990-1994 Average Concentration	2002-2003 Average Concentration	% Change in Concentration
Boston MSA	3.4	0.5	-84.0%	NA	0.93 ± 0.09	NA
New York MSA	7.6	1.3	-83.6%	36.36 ± 3.16	1.75 ± 0.11	-95.2%
Philadelphia MSA	4.4	1.3	-71.2%	NA	2.29 ± 0.16	NA
Tampa Bay MSA	1.3	0.2	-87.1%	NA	2.25 ± 0.29	NA
Detroit MSA	2.5	0.5	-79.2%	NA	2.22 ± 0.24	NA
Dallas MSA	1.8	0.5	-69.8%	842.85 ± 124.50	0.89 ± 0.08	-99.9%
St. Louis MSA	2.3	0.7	-70.4%	10.31 ± 0.92	2.35 ± 0.41	-77.2%
Denver MSA	1.2	0.0	-97.0%	NA	2.15 ± 0.27	NA
Los Angeles MSA	5.6	1.1	-80.5%	7.74 ± 0.47	2.34 ± 0.45	-69.8%
Seattle MSA	1.5	0.1	-91.1%	NA	0.83 ± 0.08	NA

Table 3h. Toluene Emission (tpy) and Concentration (µg/m^3) Comparison

MSA	1990 Emissions	2002 Emissions	% Change in Emissions	1990-1994 Average Concentration	2002-2003 Average Concentration	% Change in Concentration
Boston MSA	17,936	6,169	-65.6%	9.24 ± 1.56	3.70 ± 0.38	-59.9%
New York MSA	56,702	28,487	-49.8%	12.08 ± 0.70	4.75 ± 0.28	-60.7%
Philadelphia MSA	24,908	7,565	-69.6%	11.96 ± 1.58	3.37 ± 0.32	-71.8%
Tampa Bay MSA	9,708	6,268	-35.4%	NA	NA	NA
Detroit MSA	25,103	11,907	-52.6%	7.93 ± 0.94	5.32 ± 0.59	-32.9%
Dallas MSA	23,917	8,959	-62.5%	3.20 ± 0.47	1.71 ± 0.16	-46.5%
St. Louis MSA	13,682	6,501	-52.5%	4.49 ± 1.23	3.57 ± 0.33	-20.5%
Denver MSA	8,256	5,190	-37.1%	NA	8.44 ± 1.31	NA
Los Angeles MSA	53,919	17,489	-67.6%	24.27 ± 1.96	8.85 ± 0.34	-63.5%
Seattle MSA	16,465	10,592	-35.7%	NA	NA	NA

Table 3i. Total Xylenes Emission (tpy) and Concentration (µg/m^3) Comparison

MSA	1990 Emissions	2002 Emissions	% Change in Emissions	1990-1994 Average Concentration	2002-2003 Average Concentration	% Change in Concentration
Boston MSA	10,615	4,301	-59.5%	9.27 ±2.18	0.54 ± 0.07	-94.2%
New York MSA	35,141	25,055	-28.7%	2.13 ± 0.10	0.78 ± 0.03	-63.4%
Philadelphia MSA	15,071	5,174	-65.7%	6.77 ± 0.66	0.73 ± 0.07	-89.2%
Tampa Bay MSA	6,206	4,345	-30.0%	NA	NA	NA
Detroit MSA	15,393	7,751	-49.6%	1.78 ± 0.24	1.11 ± 0.17	-37.7%
Dallas MSA	14,862	9,535	-35.8%	0.85 ± 0.13	0.26 ± 0.02	-69.7%
St. Louis MSA	9,039	4,638	-48.7%	12.88 ± 6.43	1.48 ± 0.20	-88.5%
Denver MSA	5,376	3,170	-41.0%	NA	2.15 ± 0.29	NA
Los Angeles MSA	33,236	8,826	-73.4%	19.77 ± 2.06	5.15 ± 0.44	-73.9%
Seattle MSA	9,143	6,872	-24.8%	NA	NA	NA

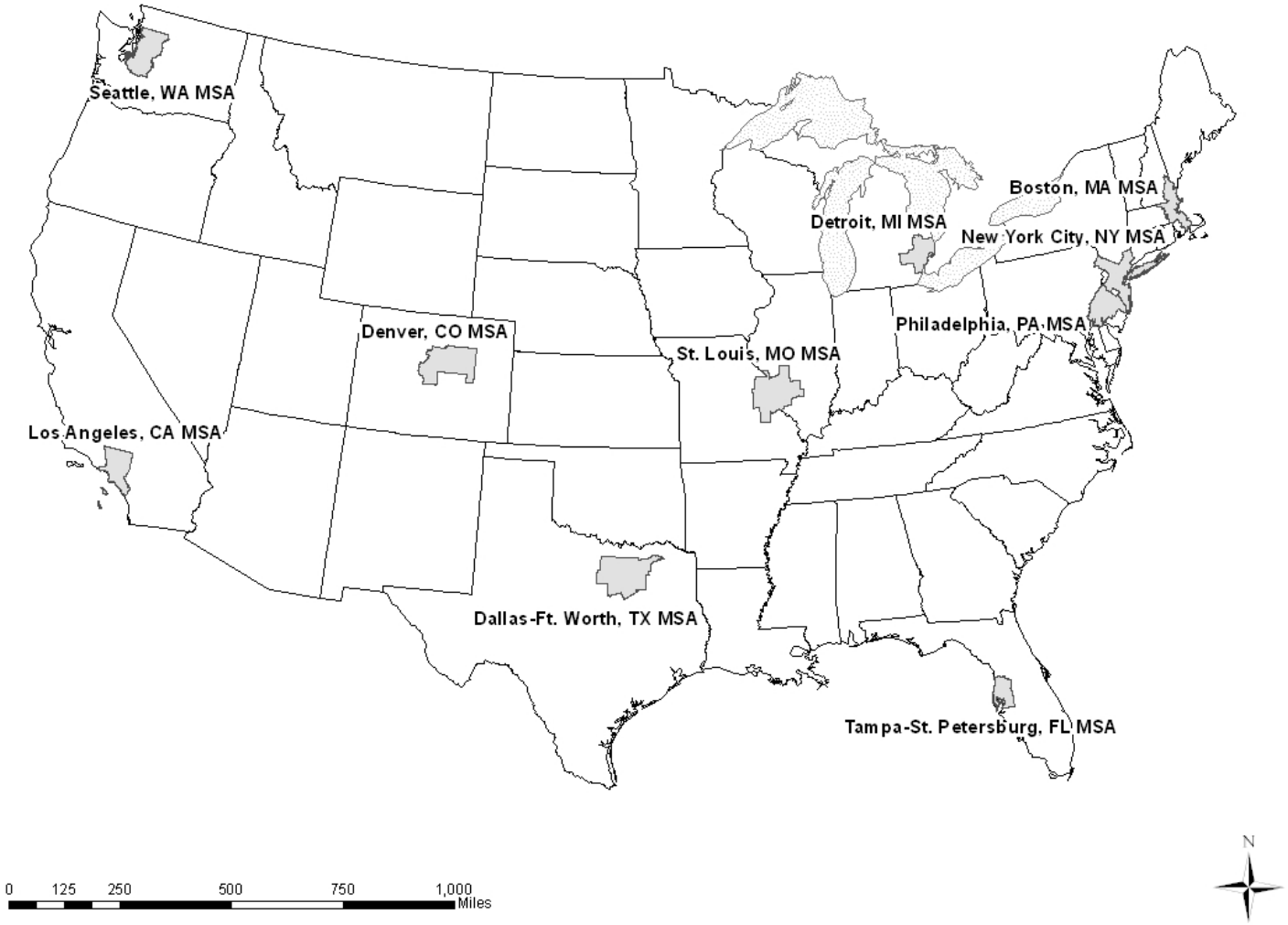

Figure 1. Metropolitan Statistical Areas In This Study

Each MSA represents an EPA Region. The Boston, New York City, Philadelphia, Tampa, Detroit, and St. Louis MSAs are all participants in EPA's 2004 Urban Air Toxics Monitoring Program.

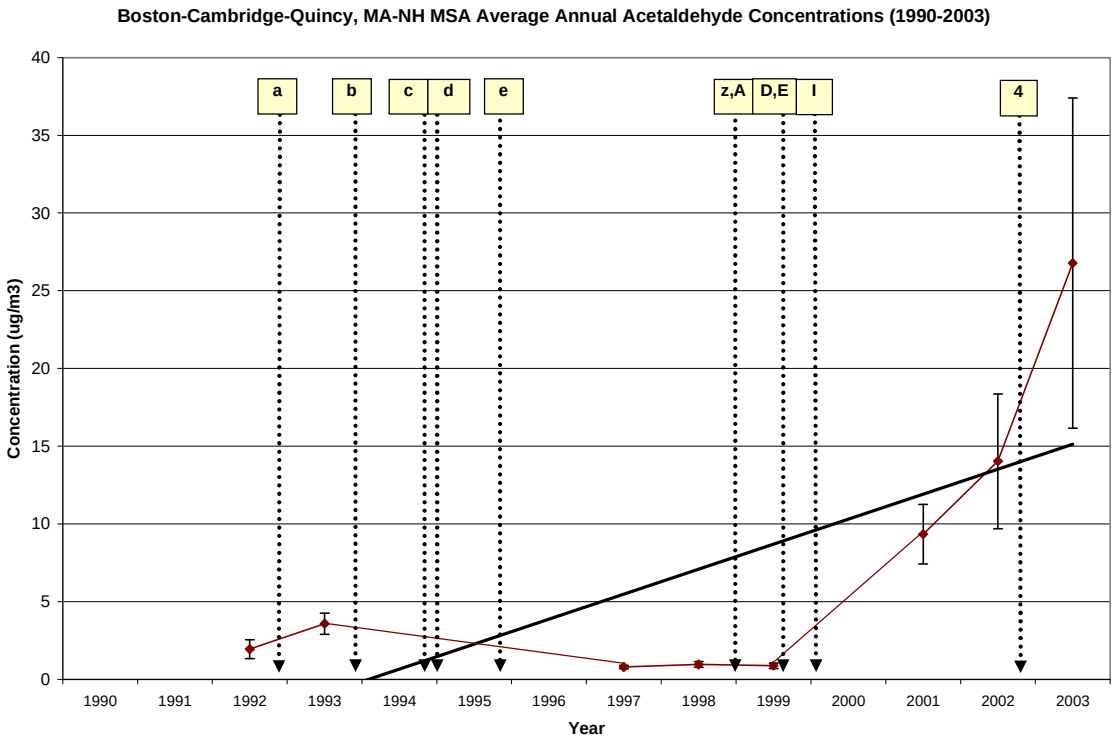

Figure 2. Boston MSA Acetaldehyde Regulation Impact Analysis
After the implementation of RFG Phase II and the POTW MACT, acetaldehyde concentrations appeared to have increased in the Boston MSA.

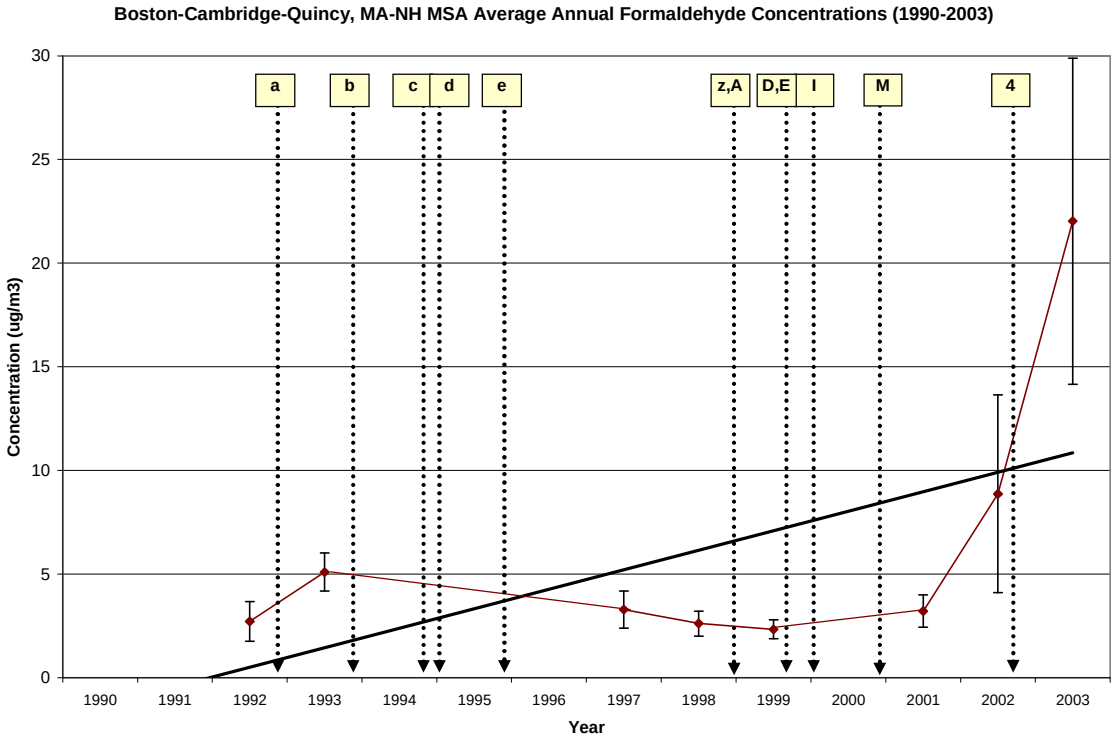

Figure 3. Boston MSA Formaldehyde Regulation Impact Analysis
Formaldehyde concentrations in the Boston MSA were steady throughout the 1990s. After the implementation of the Large Municipal Waste Combustors MACT and POTW MACT, concentrations appeared to have increased.

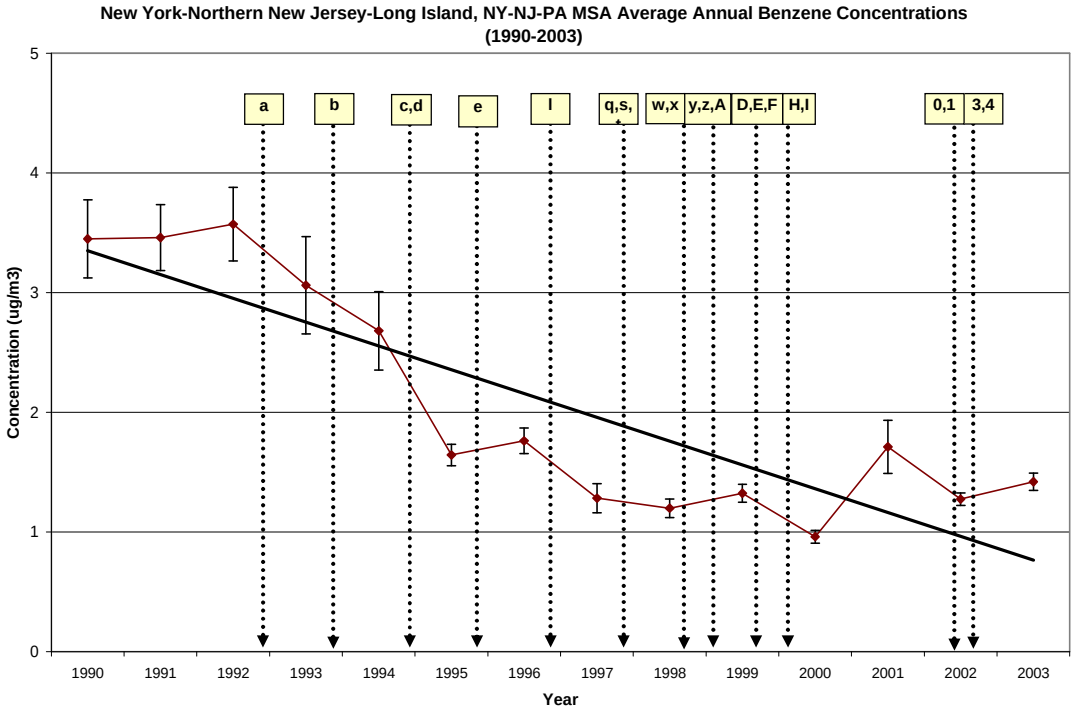

Figure 4. New York City MSA Benzene Regulation Impact Analysis

Benzene concentrations in the New York MSA appeared to have decreased after implementation of mobile source rules (winter-oxygenated fuel and reformulated gasoline). Rules targeting stationary sources did not appear to reduce benzene concentrations.

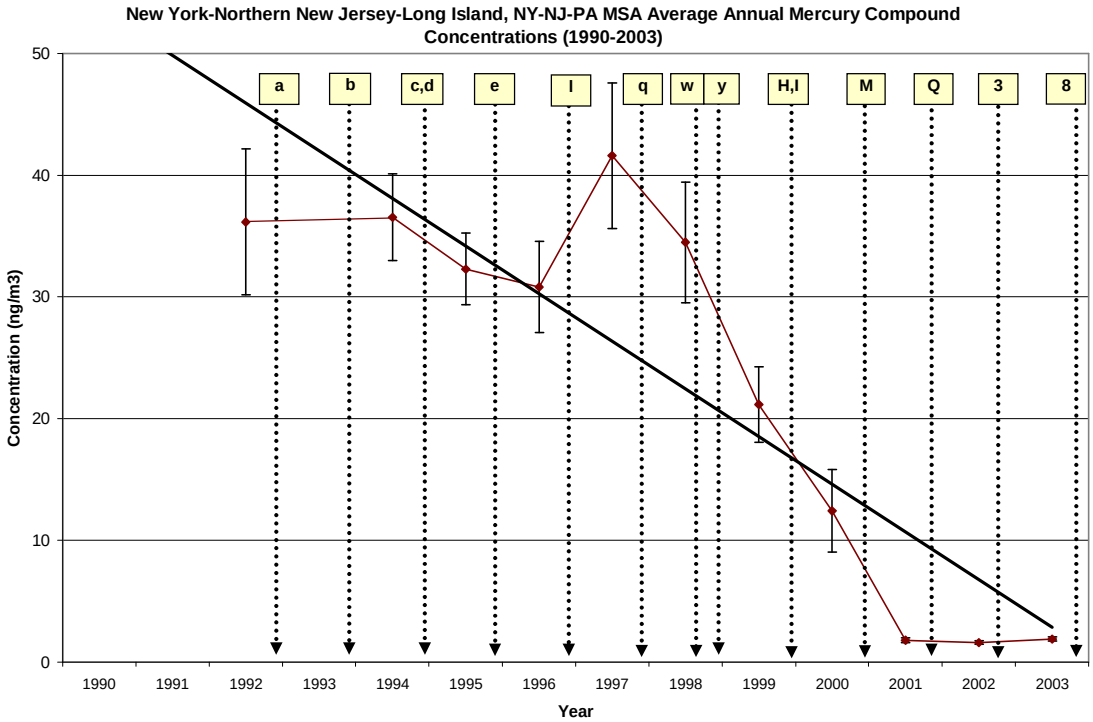

Figure 5. New York City MSA Mercury Regulation Impact Analysis

Mercury concentrations appeared to have decreased after implementation of the Petroleum Refineries MACT, Reformulated Gasoline Phase II program, and the Large Municipal Waste Combustors MACT.

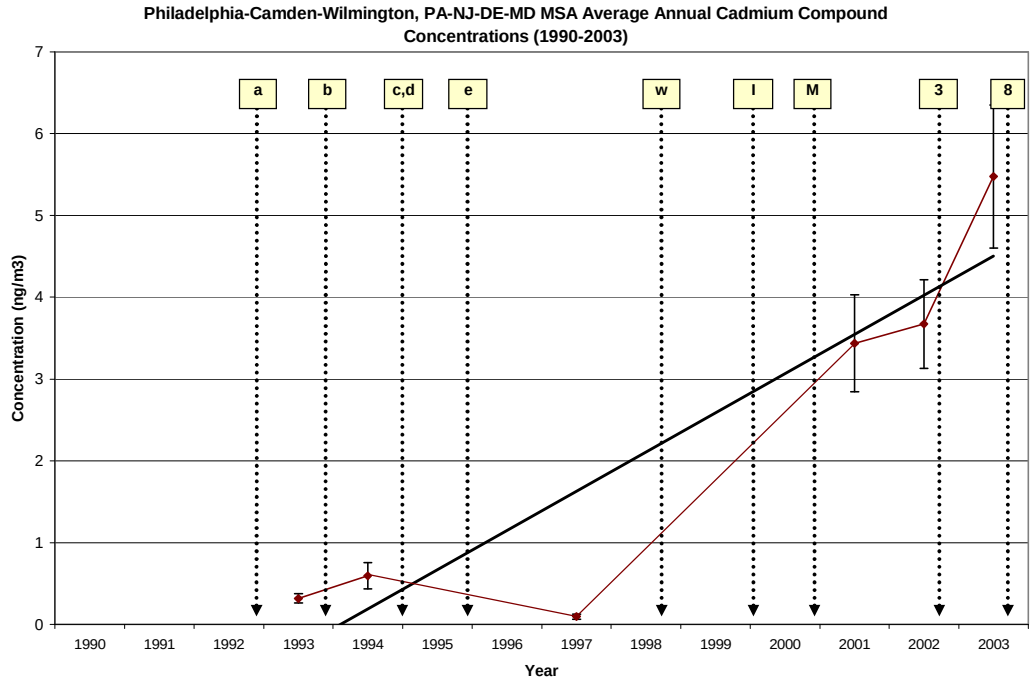

Figure 6. Philadelphia MSA Cadmium Compounds Regulation Impact Analysis

Cadmium compound concentrations appeared to have increased throughout the study period in the Philadelphia MSA, however, limited or no data is available from 1998-2000.

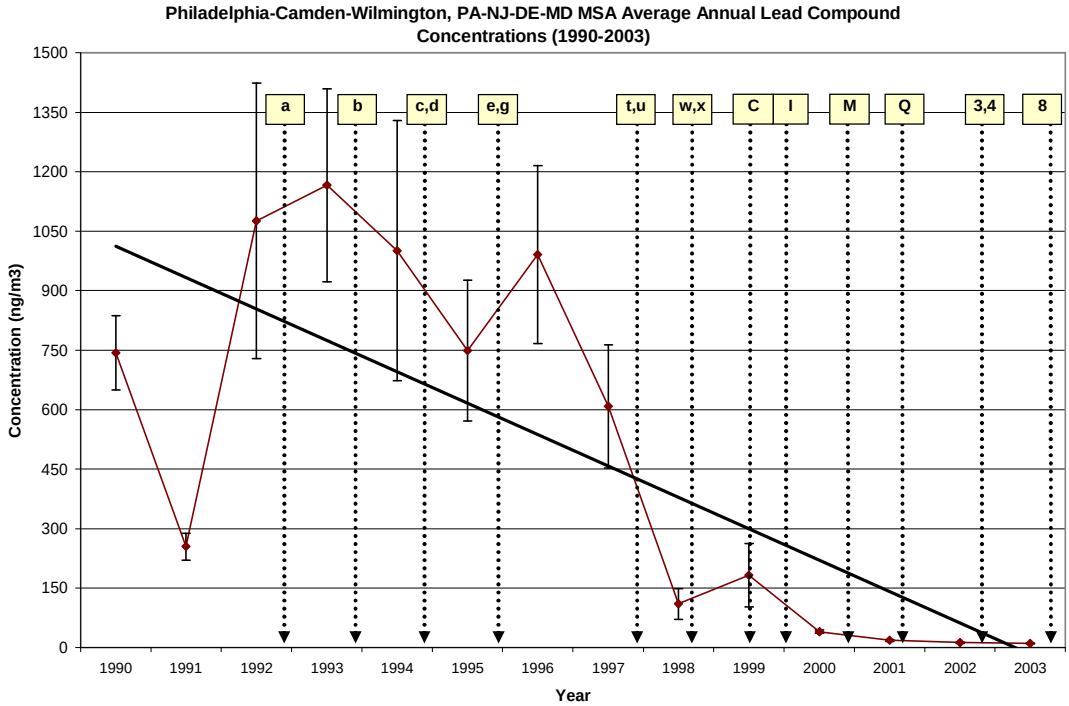

Figure 7. Philadelphia MSA Lead Compounds Regulation Impact Analysis

Lead compound concentrations appeared to have decreased substantially after implementation of the Secondary Lead Smelter MACT and the Gasoline Distribution Stage I MACT.

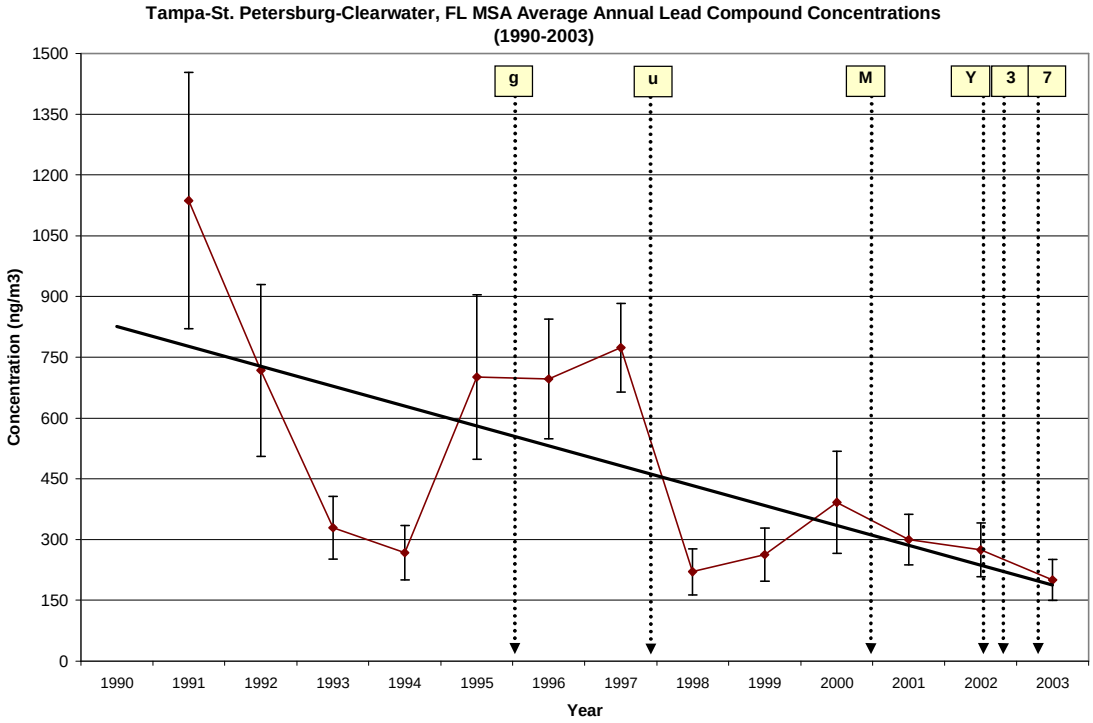

Figure 8. Tampa MSA Lead Regulation Impact Analysis

Lead compound concentrations appeared to have decreased overall during the study period, most notably after the Secondary Lead Smelting MACT.

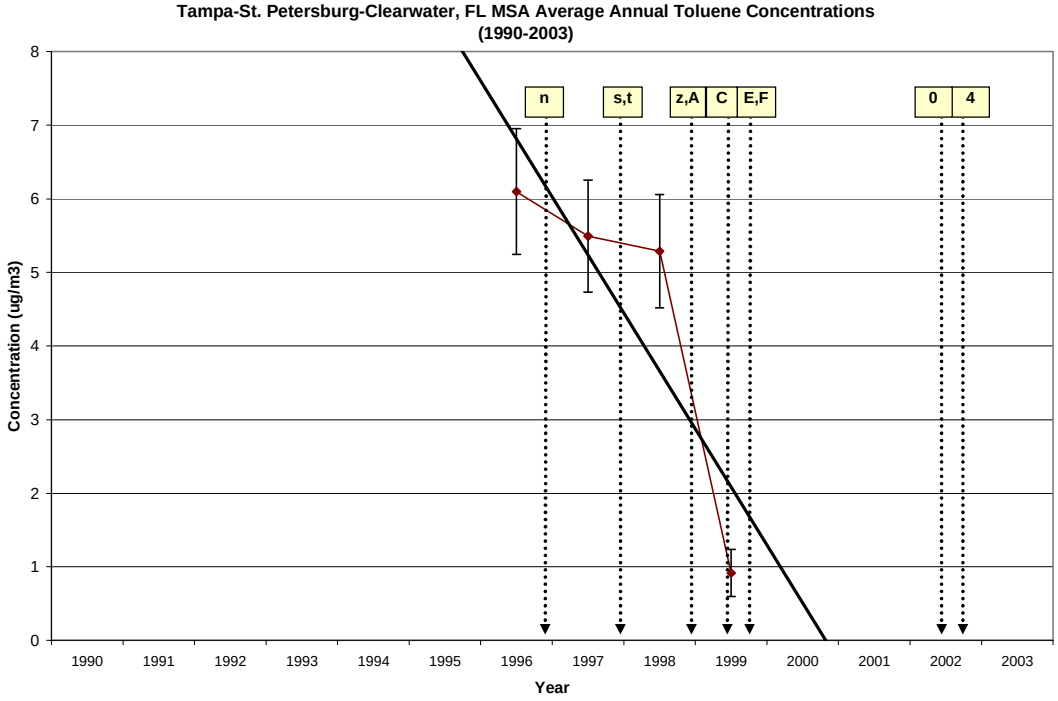

Figure 9. Tampa MSA Toluene Regulation Impact Analysis

Limited toluene data in the Tampa MSA for the last five years limits the conclusion that concentrations are continuing to decrease.

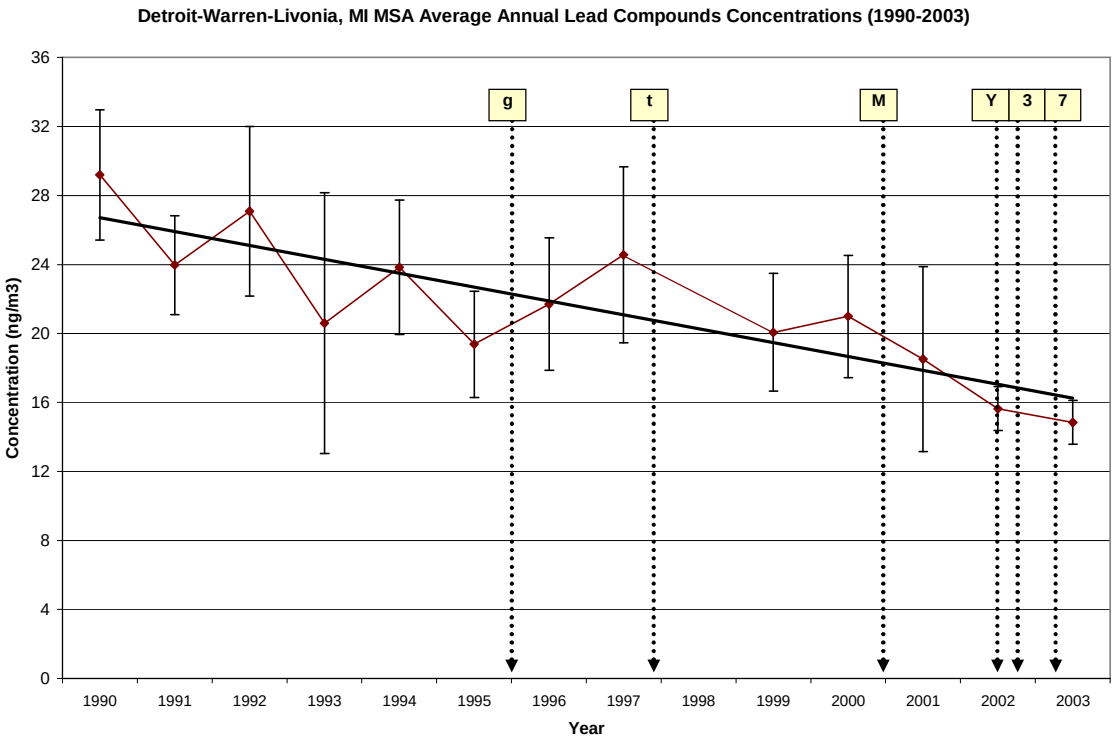

Figure 10. Detroit MSA Lead Regulation Impact Analysis

Lead concentrations in Detroit appeared to have decreased steadily during the study period. The biggest affect appears to be the implementation of the Large Municipal Waste Combustors MACT.

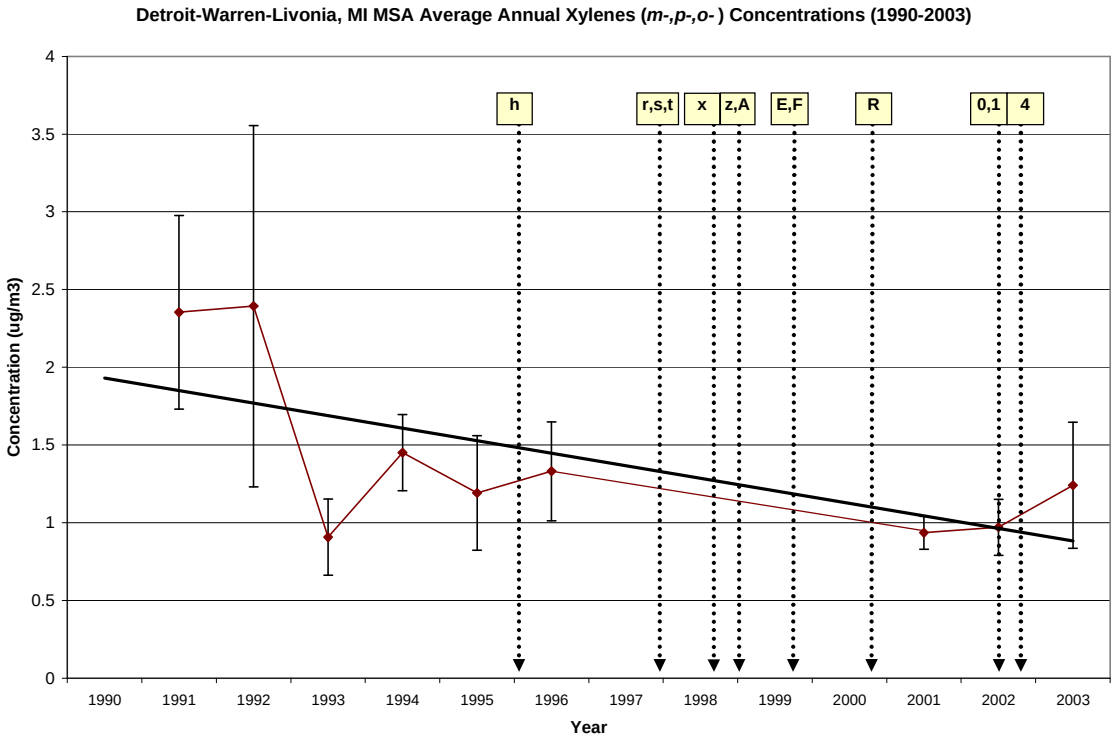

Figure 11. Detroit MSA Xylenes (total) Regulation Impact Analysis

Although total xylene concentrations had little variance throughout the 1990s, it's unsure how stationary source rules affected xylene concentrations due to the limited data availability of xylene measurements from 1997-2000, when several regulations were implemented.

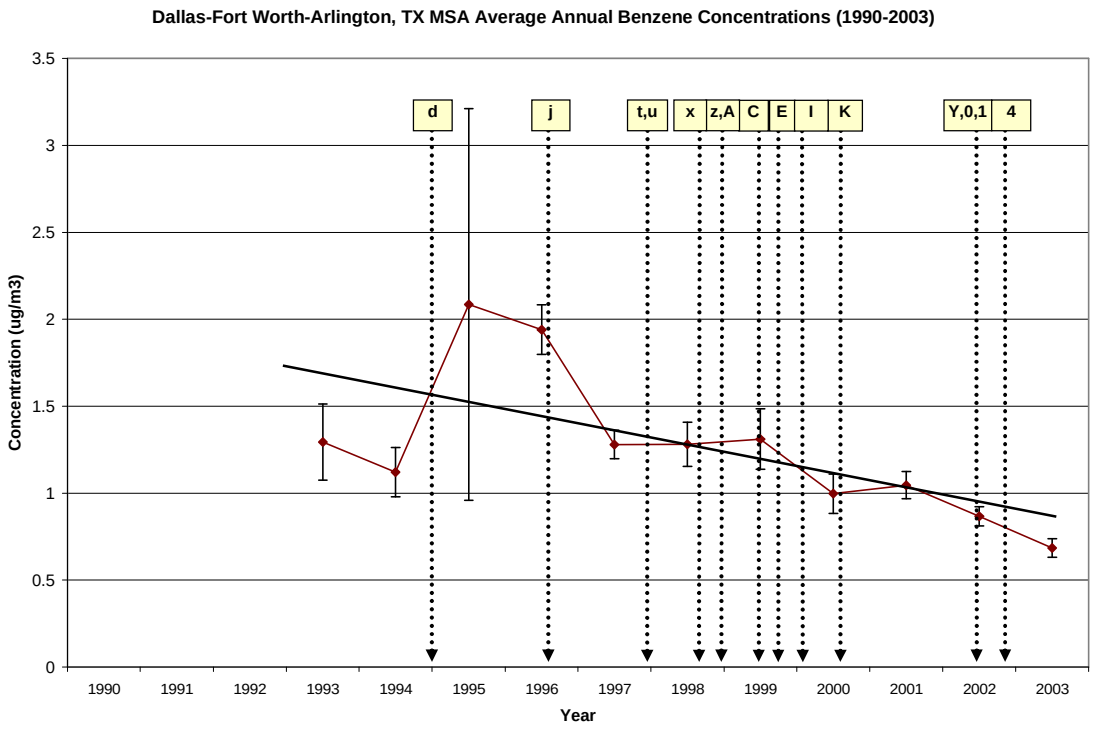

Figure 12. Dallas MSA Benzene Regulation Impact Analysis

Benzene concentrations in Dallas show a downward trend, apparently in response to implementation of Tier 1 Mobile Standards.

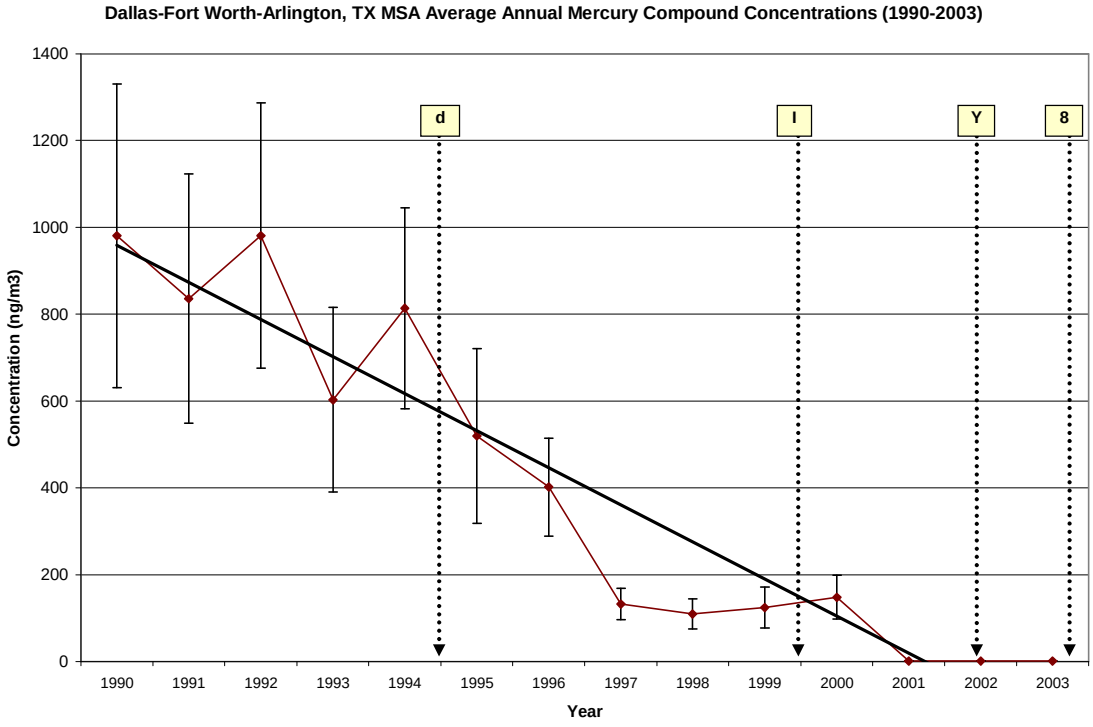

Figure 13. Dallas MSA Mercury Regulation Impact Analysis

Mercury compound concentrations appear to have decrease substantially with the implementation of the Reformulated Gasoline Phase 1 Program. However, according to the NEI, mercury emissions from Hazardous Waste Combustors decreased by 97% from 1996 to 2002 for this MSA, most likely as a result of impending regulations.

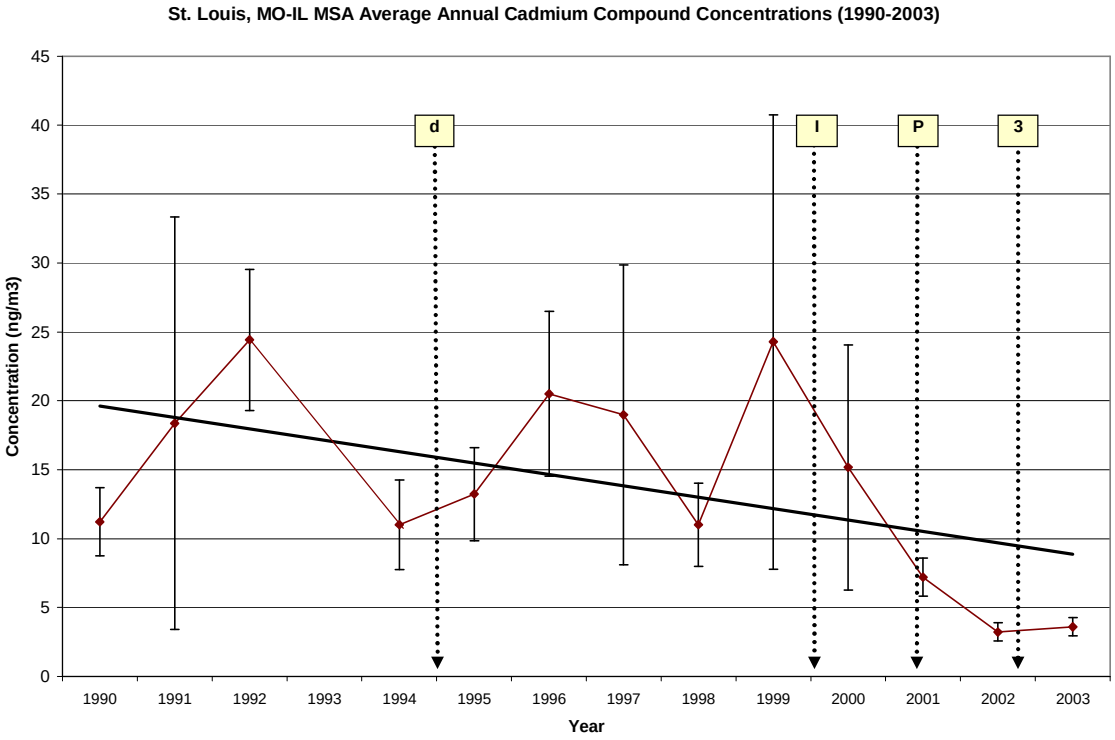

Figure 14. St. Louis MSA Cadmium Regulation Impact Analysis

Overall, cadmium compound concentrations in the St. Louis MSA appeared to have decreased during the study period. The implementation of the Reformulated Gasoline Phase 2 Program and the Primary Lead Smelting MACT coincide with these reductions. However, no implemented regulations explain the increases in the 1996, 1997, and 1999 concentrations.

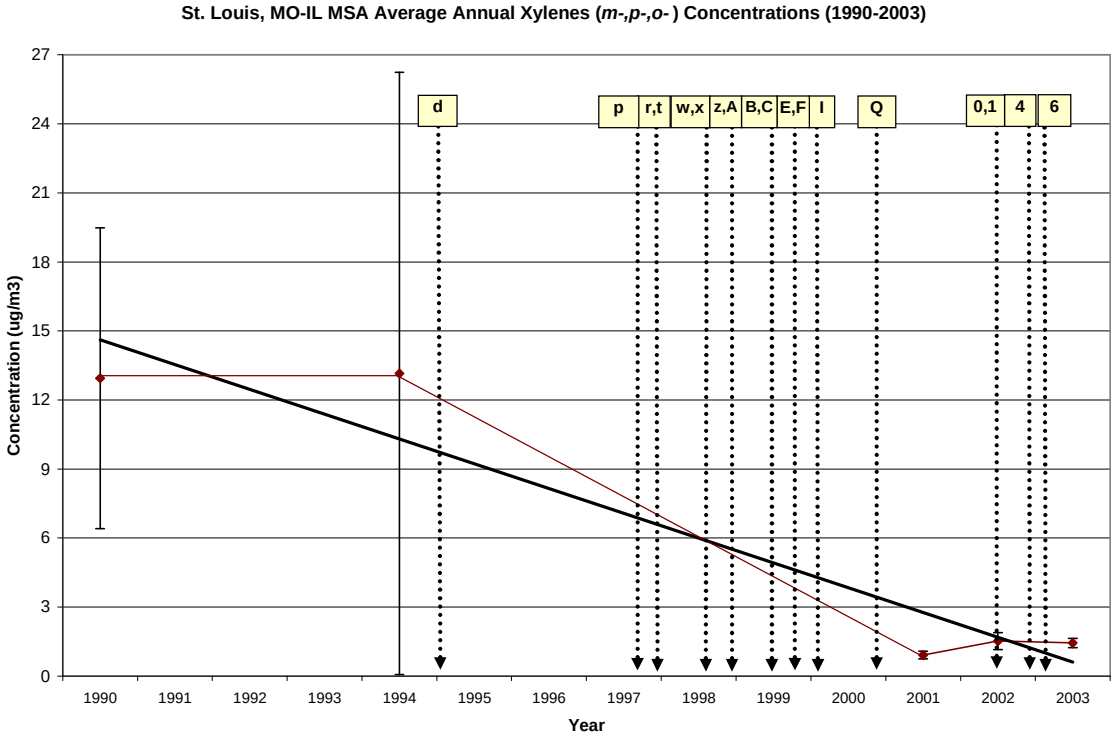

Figure 15. St. Louis MSA Xylenes (total) Regulation Impact Analysis

Total xylene concentrations in the St. Louis MSA declined dramatically from 1990 to 2003, apparently in response to several implemented stationary and mobile source regulations targeting VOCs. However, limited data availability of xylene measeurements between 1995 and 2000 limit the certainty of these conclusions.

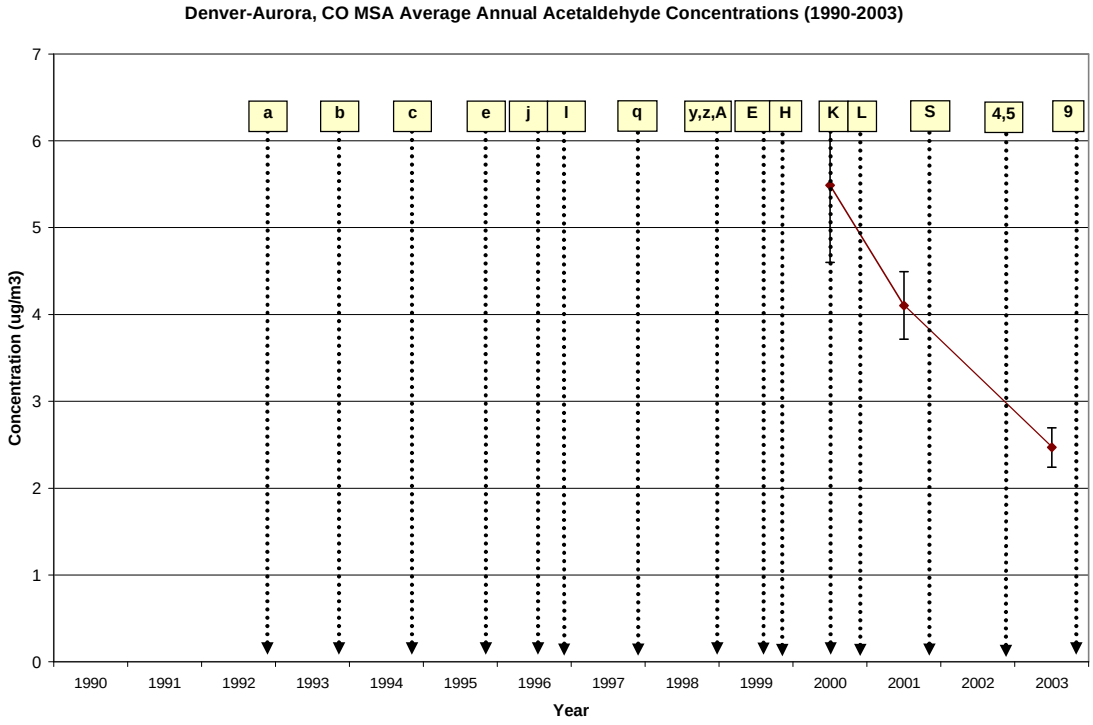

Figure 16. Denver MSA Acetaldehyde Regulation Impact Analysis

Over the last four years, acetaldehyde concentrations appeared to have decreased after implementation of mobile source rules (winter-oxygenated fuel program and the National Low Emissions Vehicle Program Phase II).

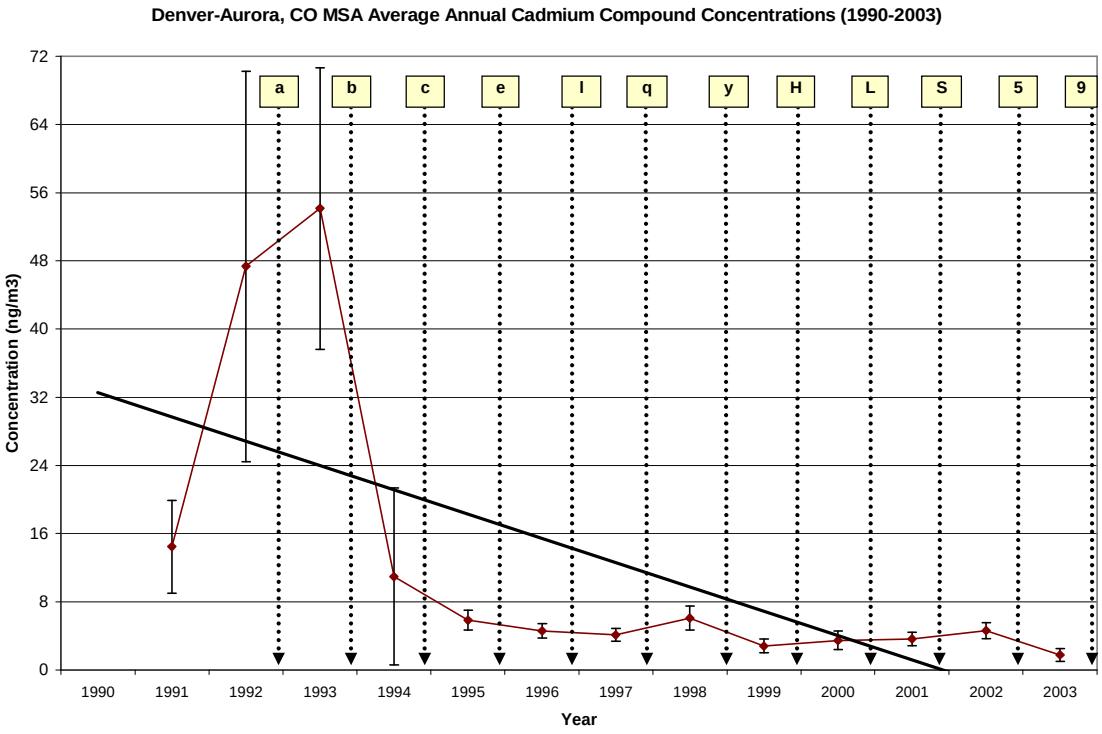

Figure 17. Denver MSA Cadmium Regulation Impact Analysis

No implemented stationary source regulations were identified in the Denver MSA which apparently affected cadmium concentrations. According to TRI, cadmium emissions from Asarco, Inc. Globe Plant decreased from 0.198 tpy in 1990 to 0.077 tpy in 1994, a 61% decrease. Cadmium emissions remained steady for this plant through the 2002 NEI.

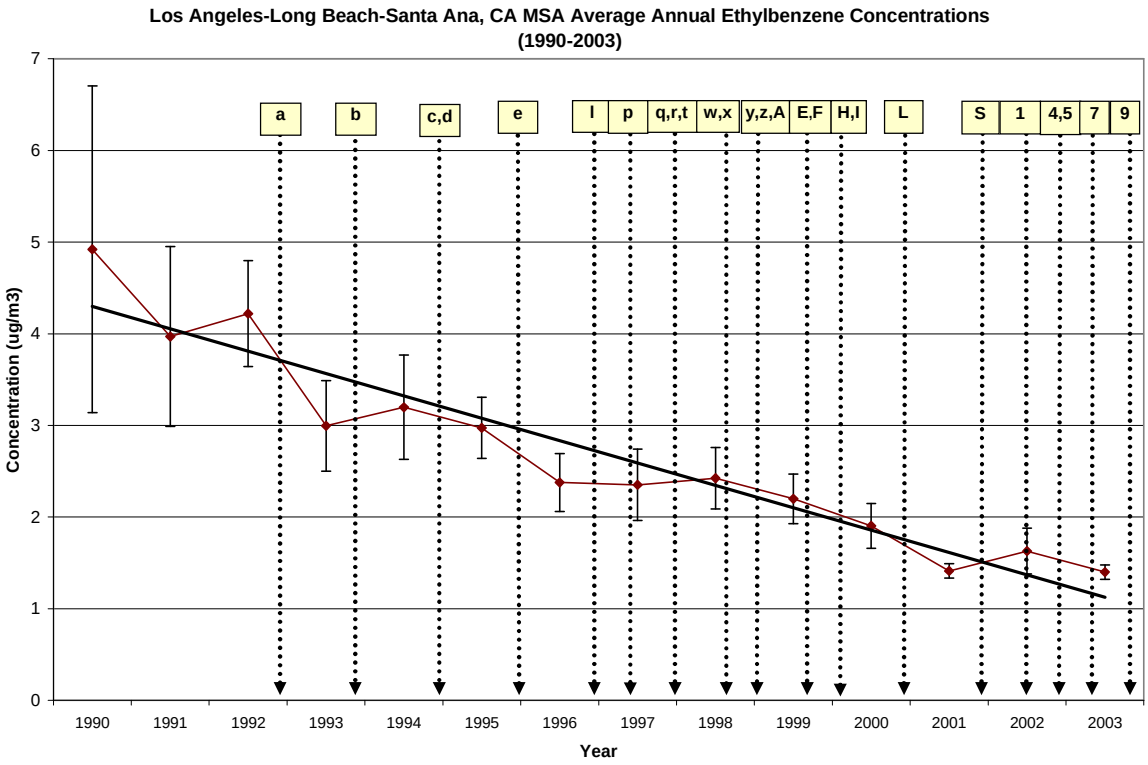

Figure 18. Los Angeles MSA Ethylbenzene Regulation Impact Analysis
Ethylbenzene concentrations in the Los Angeles MSA appeared to have declined steadily in response to several implemented stationary and mobile source regulations targeting VOCs.

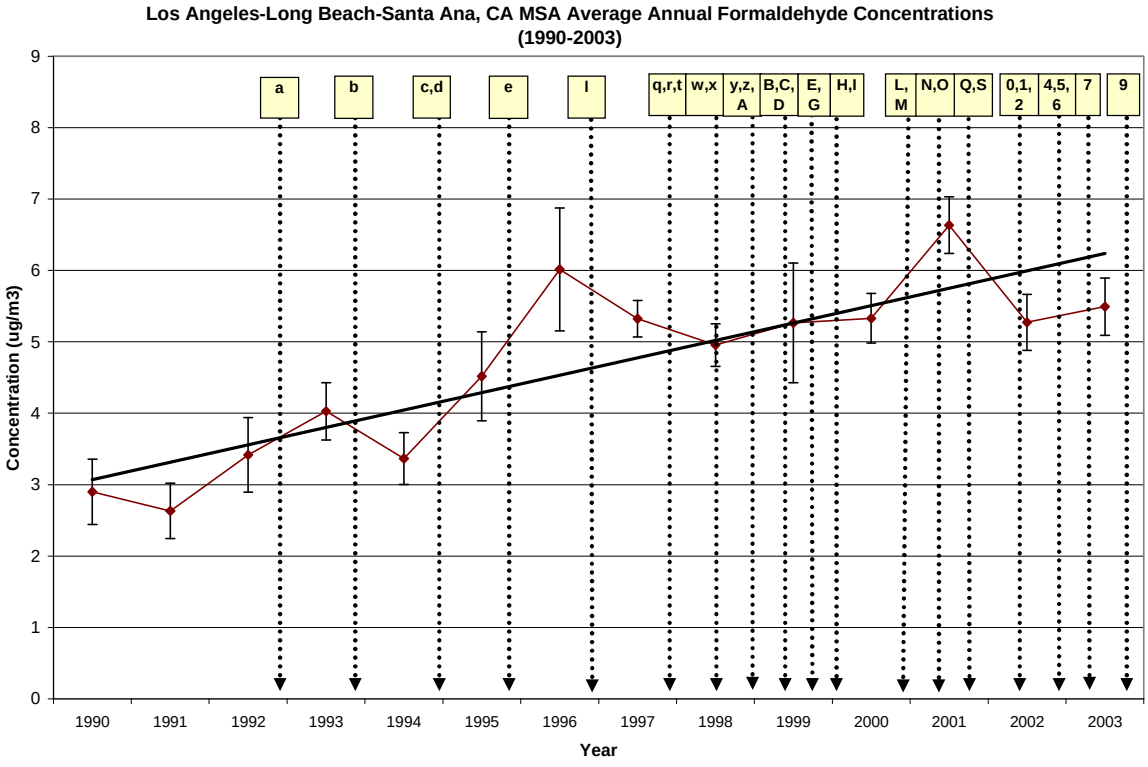

Figure 19. Los Angeles MSA Formaldehyde Regulation Impact Analysis
Formaldehyde concentrations have steadily increased during the study period. This is most likely due to the implementation of the Reformulated Gasoline Phase I Program.

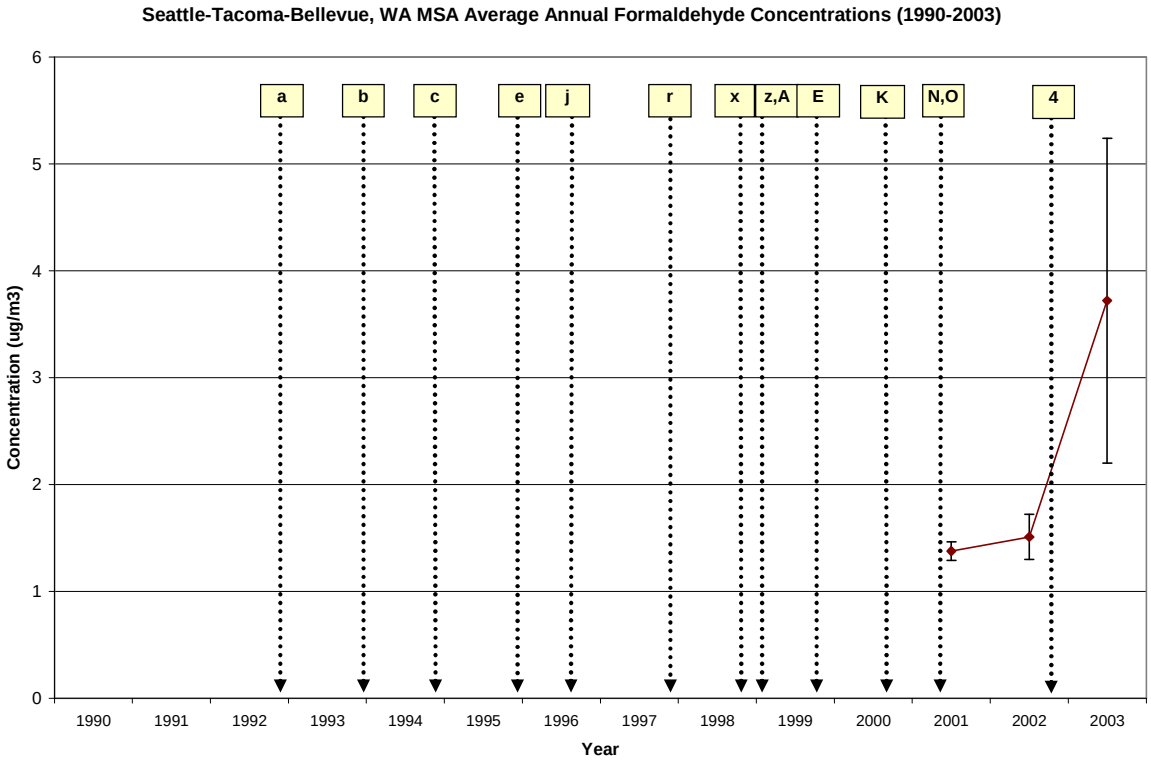

Figure 20. Seattle MSA Formaldehyde Regulation Impact Analysis

Over the last three years, formaldehyde concentrations have appeared to have increased, noticeably after the implementation of the Publicly Owned Treatment Works MACT. However, formaldehyde data prior to 2001 is limited or unavailable, thus making it difficult to characterize a trend.

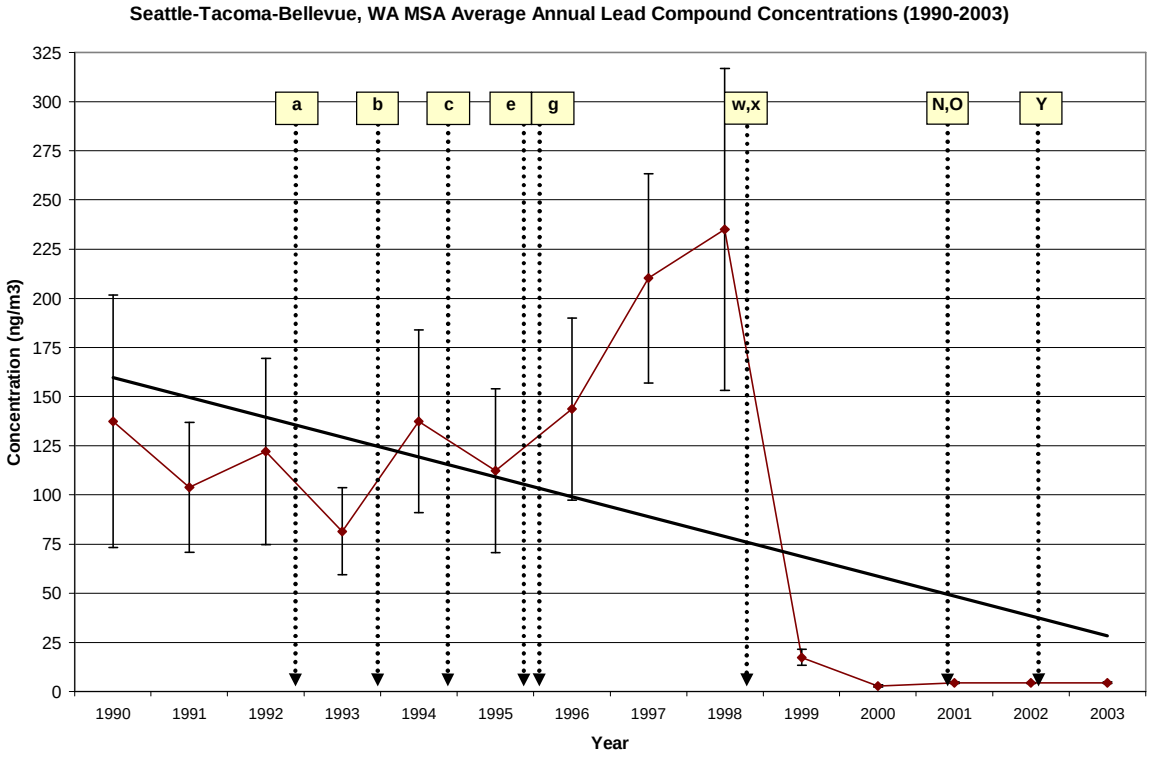

Figure 21. Seattle MSA Lead Regulation Impact Analysis

During the study period, lead concentrations appeared to have decreased. After implementation of the Petroleum Refineries and Aerospace Manufacturing MACTs, concentrations decreased substantially. Interestingly, lead concentrations apparently increased after the prohibition of leaded gasoline.