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

Regi Oommen

Eastern Research Group, Inc. (ERG, Inc.)
1600 Perimeter Park
Morrisville, NC

September 27, 2005

ACKNOWLEDGEMENTS

- Jaime Hauser
- Garry Brooks
- Dave Dayton
- Heather Perez
- Susan McClutchey
- Andy Blackard
- Randy Bower
- Jody Tisano

CLEAN AIR ACT AMENDMENTS - BACKGROUND

- The Clean Air Act – first promulgated in 1960
- Amended 1963 and 1977
- Primary focus was on Criteria Air Pollutants (CAPs)
  - Carbon monoxide
  - Lead
  - Oxides of nitrogen
  - Sulfur dioxide
  - Particulate matter
  - Volatile organic compounds*

CLEAN AIR ACT AMENDMENTS – HAPS FOCUS

- Major revisions in 1990
- Primary focus was on Hazardous Air Pollutants (HAPs)
- Focus included:
  - source characterization
  - control device options
  - monitoring

TRENDS WORK

- EPA has done a good job of characterizing the trends of the CAPs through its annual Trends report
- However, characterization of HAP trends have been few due to:
  - Limited monitoring data (monitors, coverage areas)
  - Limited emissions data (HAPs not required)
  - Limited resources (e.g., AQS submittals not required)
  - EPA staff/contractor expertise
- ERG was recommended by EPA staff to provide support due to our work on the UATMP reports and emission inventory support.

TRENDS WORK

- Summer 2004, ERG received a Work Assignment
- Primary work was to compile a database of HAP ambient monitoring concentrations from:
  - Historical Archive (compiled by previous contractors)
  - IMPROVE data
  - AQS data supplemental (2001-2003)
- SQL Server and Access were chosen to house the data
- Database contained over 14,000,000 records
- Data would be a great resource for trends analysis in the UATMP reports.

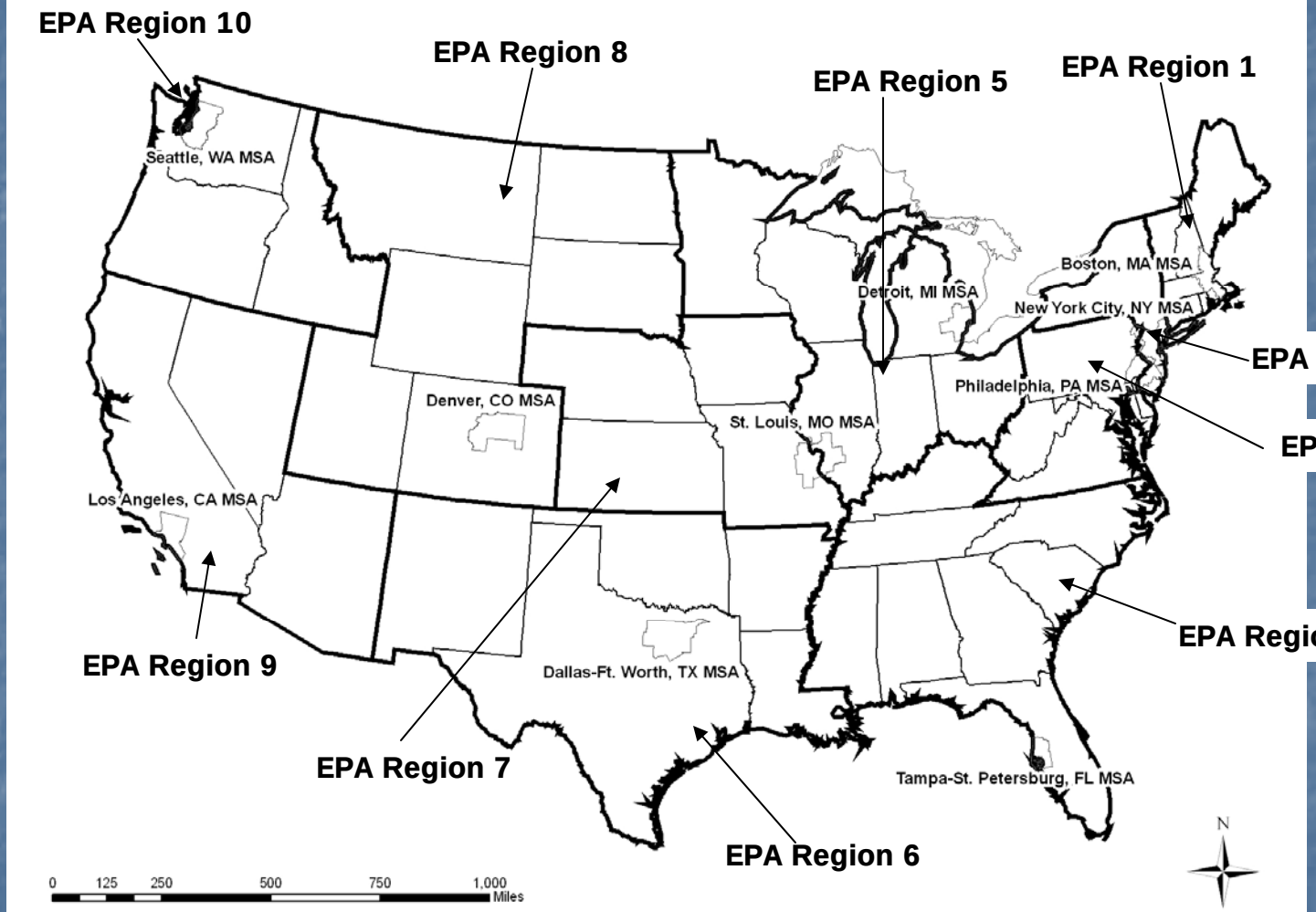
POLICY-RELEVANT QUESTIONS

- What are the HAP concentration trends?
- Have HAP-specific federal regulations been effective at reducing ambient concentrations?
- Do HAP emissions show a decline due to HAP-specific federal regulations?

APPROACH – MSAs OF INTEREST

- We reviewed the 2004 monitoring sites for the UATMP. Six MSAs from the UATMP were chosen:
  - Boston (BOS)
  - New York (NYC)
  - Philadelphia (PHL)
  - Tampa (TAM)
  - Detroit (DET)
  - St. Louis (STL)
- We decided to try and cover the U.S. by selecting one MSA per EPA region. Chose the following MSAs:
  - Dallas (DAL)
  - Denver (DEN)
  - Los Angeles (LOS)
  - Seattle (SEA)

APPROACH – MSAs OF INTEREST



APPROACH – HAPs OF INTEREST

- Nine HAPs were chosen:
  - Acetaldehyde
  - Benzene
  - Cadmium
  - Ethylbenzene
  - Formaldehyde
  - Lead
  - Mercury
  - Toluene
  - Xylenes (total)

- EPA Core HAPs: acetaldehyde, benzene, cadmium, formaldehyde, and lead

APPROACH – HAPs OF INTEREST

- Pollutant Types:
  - Carbonyls – acetaldehyde and formaldehyde
  - VOCs – benzene, ethylbenzene, toluene, and xylenes
  - Metals – cadmium, lead, mercury
- Cancer compounds: acetaldehyde, benzene, cadmium, ethylbenzene, and formaldehyde
- Noncancer compounds: all nine HAPs

APPROACH – TIME PERIOD OF INTEREST

- CAAA promulgated 1990
- First EPA HAP emission inventory covered base years 1990-1993; Latest EPA HAP emission inventory covers 2002 base year
- Monitoring data: Several ambient monitoring sites began sampling post-1995. EPA's NATTS Network begins monitoring in 2004.
- Several stationary source and mobile source rules have been promulgated/implemented in the last 10 years
- Chose 1990-2003 as our time period of interest

METHODOLOGY

- Compare *emissions* at the MSA-level
  - 1990 to 2002
  - Grouped individual HAPs
    - Xylene species (*o*-, *m*-, *p*-) summed
    - Metal species averaged
  - MACT code assignments were retained
  - EPA is currently revising 1990 and 2002 NEI
- Compare *concentrations* at the MSA-level
  - 1990-94 to 2002-03
  - Limited data in early 90s
  - Grouped individual HAPs
    - Xylene species (*o*-, *m*-, *p*-) summed
    - Metal species averaged

RESULTS – POPULATION/VMT CHANGES

- **Look at Population/VMT changes across each MSA**

MSA	1990 MSA Population	2003 MSA Population	% Change in MSA Population	1990 MSA VMT (1000s)	2003 MSA VMT (1000s)	% Change in MSA VMT	Winter-Oxygenated Time Period Implemented	MSA Reformulated Gasoline Designation
Boston MSA	4,133,895	4,439,971	+ 7%	18,738,370	32,582,820	+ 74%	1992-1996	Opt-In
New York MSA	16,863,671	18,640,775	+ 11%	82,128,650	105,869,710	+ 29%	1992-2000	Required
Philadelphia MSA	5,435,550	5,772,947	+ 6%	24,002,035	37,576,750	+ 57%	1992-1996	Required
Tampa Bay MSA	2,067,959	2,531,908	+ 22%	12,304,150	21,258,330	+ 73%	NA	NA
Detroit MSA	4,248,699	4,483,853	+ 6%	28,551,395	36,788,715	+ 29%	NA	NA
Dallas MSA	3,989,294	5,589,670	+ 40%	29,273,000	39,848,145	+ 36%	NA	Opt-In
St. Louis MSA	2,599,893	2,759,440	+ 6%	16,530,120	22,794,250	+ 38%	NA	Opt-In
Denver MSA	1,650,489	2,301,116	+ 39%	9,909,750	17,358,670	+ 75%	1992-2003	NA
Los Angeles MSA	11,273,720	12,829,272	+ 14%	91,495,645	105,856,570	+ 16%	1992-2003	Required
Seattle MSA	2,559,136	3,141,777	+ 23%	19,203,015	24,413,025	+ 27%	1992-1996	NA

RESULTS – EMISSION CHANGES

- **Look at HAP emission changes across each MSA**

HAP	EMISSION CHANGES
Acetaldehyde	-30% (BOS) to -57% (NYC)
Benzene	-22% (TAM) to -79% (LOS)
Cadmium	+110% (PHL) to -97% (TAM)
Ethylbenzene	-25% (SEA) to -75% (LOS)
Formaldehyde	-38% (TAM) to -62% (NYC)
Lead	+67% (DET) to -89% (STL)
Mercury	-70% (DAL) to -97% (DEN)
Toluene	-35% (TAM) to -70% (PHL)
Xylenes (Total)	-25% (SEA) to -73% (LOS)

RESULTS – EMISSION CHANGES

- What's going on in PHL?

HAP	EMISSION CHANGES
Cadmium	+110% (PHL) to -97% (TAM)

- Emission Inventory Validation?
  - 1990 PHL cadmium emissions = 2.9 tpy
  - 2002 PHL cadmium emissions = 6.3 tpy

RESULTS – EMISSION CHANGES

- Emission Inventory Validation?
  - 1990 PHL cadmium emissions for:
    - Commercial Heating: residual oil = 0.012 tpy
    - Industrial Boilers: bitum. & lignite = 0.018 tpy
    - Industrial Boilers: residual oil = 0.008 tpy
  - 2002 PHL cadmium emissions for these categories:
    - Commercial Heating: residual oil = 0.70 tpy
    - Industrial Boilers: bitum. & lignite = 0.68 tpy
    - Industrial Boilers: residual oil = 4.27 tpy
  - These emissions increased from 0.038 tpy to 5.65 tpy (+14,770%)

RESULTS – EMISSION CHANGES

- What's going on in DET?

HAP	EMISSION CHANGES
Lead	+67% (DET) to -89% (STL)

- Emission Inventory Comparison
  - 1990 DET lead emissions = 19.0 tpy
  - 2002 DET lead emissions = 31.7 tpy

RESULTS – EMISSION CHANGES

- Emission Inventory Validation?
  - 1990 DET lead emissions for:
    - Utility oil-fired boilers = 0.21 tpy for entire MSA
  - 2002 DET lead emissions for these categories:
    - Detroit Edison Greenwood Energy Center = 13.7 tpy
    - St. Clair/Belle River Power Plant = 3.5
  - These emissions increased from 0.21 tpy to 17.2 (+8,100%)

RESULTS – CONCENTRATION CHANGES

- **Look at HAP concentration changes across each MSA**

HAP	Significant Increases	Significant Decreases
Acetaldehyde	BOS, STL	NYC, PHL, LOS
Benzene	None	BOS, NYC, PHL, DET, DAL, STL, LOS
Cadmium	PHL, LOS	NYC, STL, DEN
Ethylbenzene	None	BOS, NYC, PHL, DAL, LOS
Formaldehyde	BOS, STL, LOS	NYC, PHL
Lead	None	All MSAs
Mercury	None	NYC, DAL, STL, LOS
Toluene	None	BOS, NYC, PHL, DET, DAL, LOS
Xylenes (Total)	None	BOS, NYC, PHL, DET, DAL, STL, LOS

RESULTS – APPLICABLE IMPLEMENTED REGULATIONS

- Title I: NSPS and NAAQS Programs
  - Solid Waste Combustion MACT rules
  - National VOC Emission Standards
- Title II: Mobile Sources Program:
  - Motor Vehicle Emission Standards (Tier I and II)
  - Fuel and Fuel Additives
- Title III: NESHAP Program: NESHAP Standards

RESULTS – REGULATIONS NOT CONSIDERED FOR STUDY

- **Title IV: Acid Rain Program – PM HAPs indirectly reduced through use of PM control devices**
- **Title V: Permitting Program – reduction of individual HAP sources**
- **Title VI: Stratospheric Ozone Protection Program**
- **Title VII: Enforcement**

RESULTS – IMPLEMENTED REGULATIONS

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

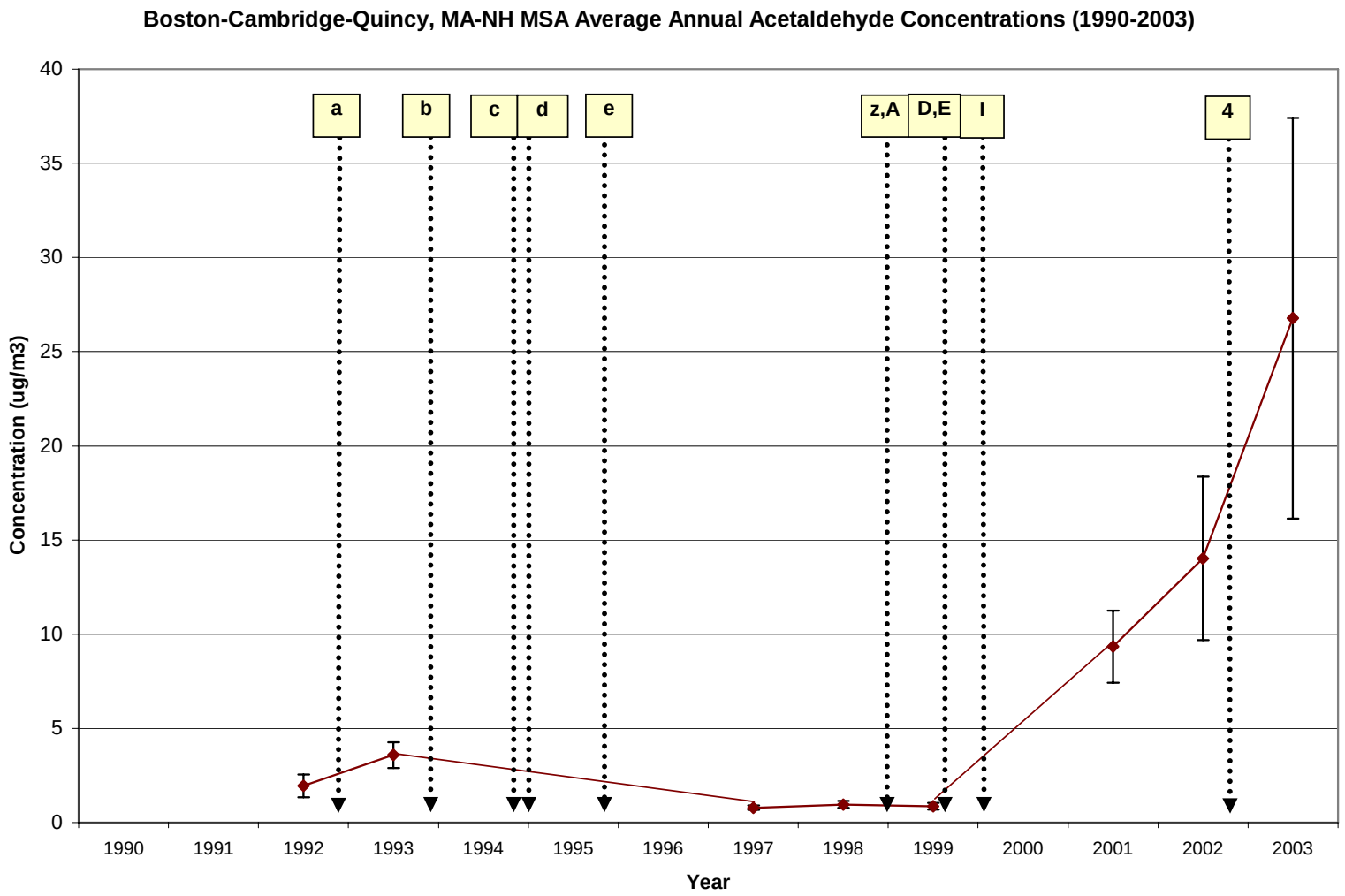
RESULTS – IMPLEMENTED REGULATIONS

Graph Key	Regulation	Implementation Date	Targeted HAPs[1]
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

RESULTS – IMPLEMENTED REGULATIONS

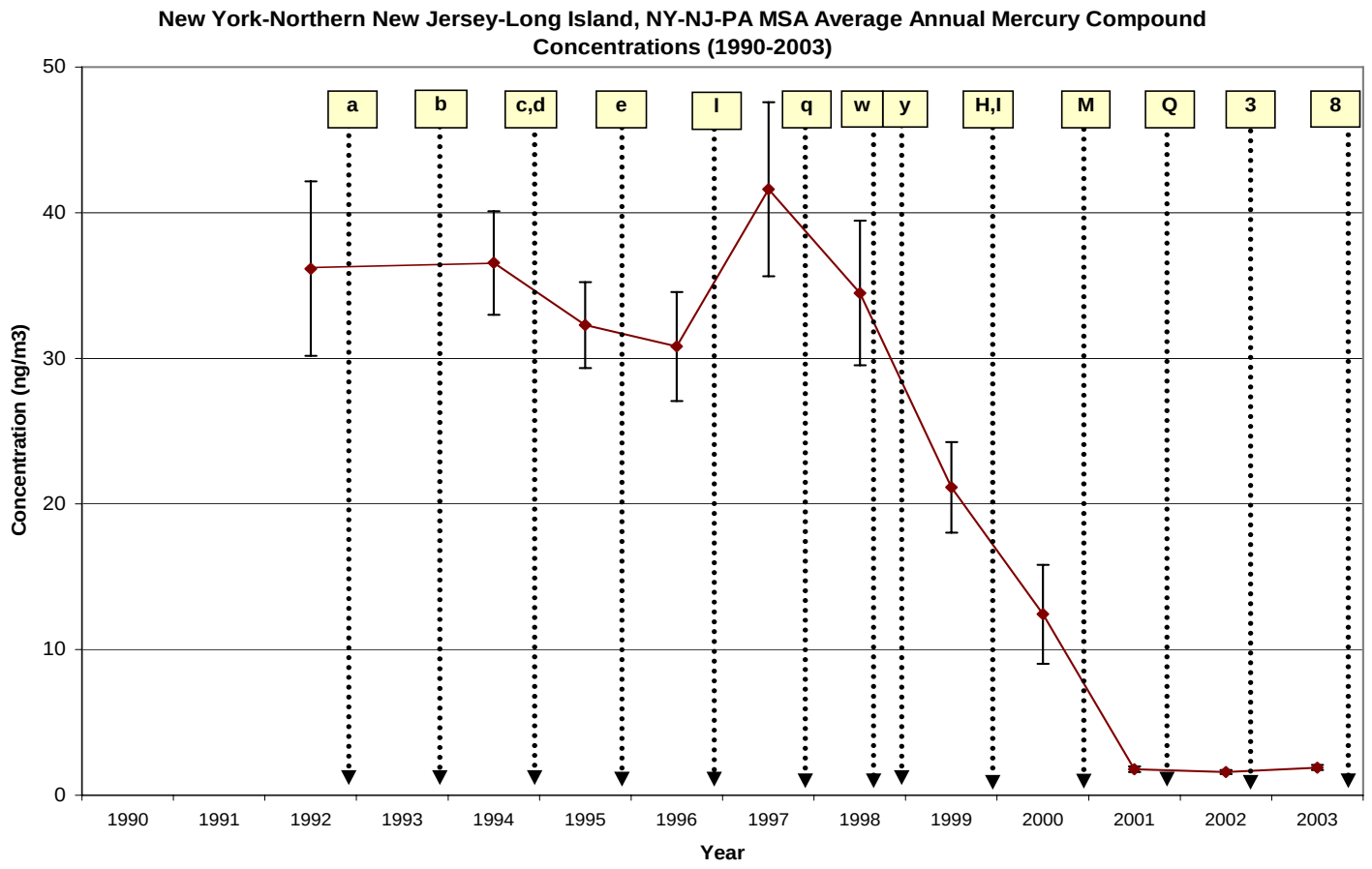
Graph Key	Regulation	Implementation Date	Targeted HAPs[1]
P1	Steel Pickling – HCl Process	6/22/2001	NA
Q	Pharmaceuticals Production	9/21/2001	V, C, M
R	Flexible Polyurethane Foam Production	10/8/2001	V
S	Winter-Oxygenated, Season 10	11/1/2001	V, C, M
T	Ferroalloys Production	11/21/2001	NA
U	Mineral Wool 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
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	12/23/2003	V

RESULTS – BOSTON MSA



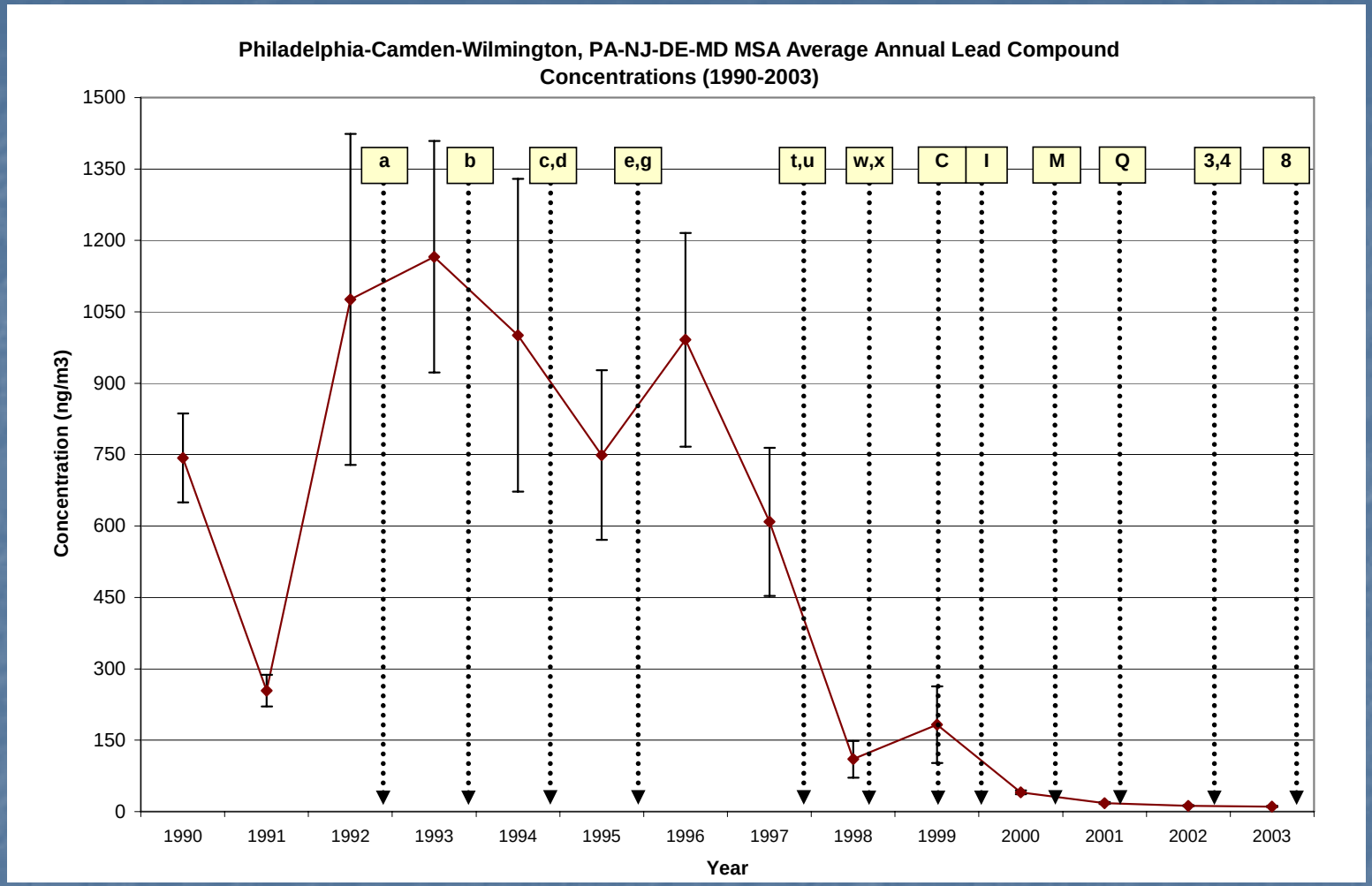
After the implementation of RFG Phase II (graph key = I) and the POTW MACT (graph key = 4), acetaldehyde concentrations appeared to have increased in the Boston MSA

RESULTS – NEW YORK MSA



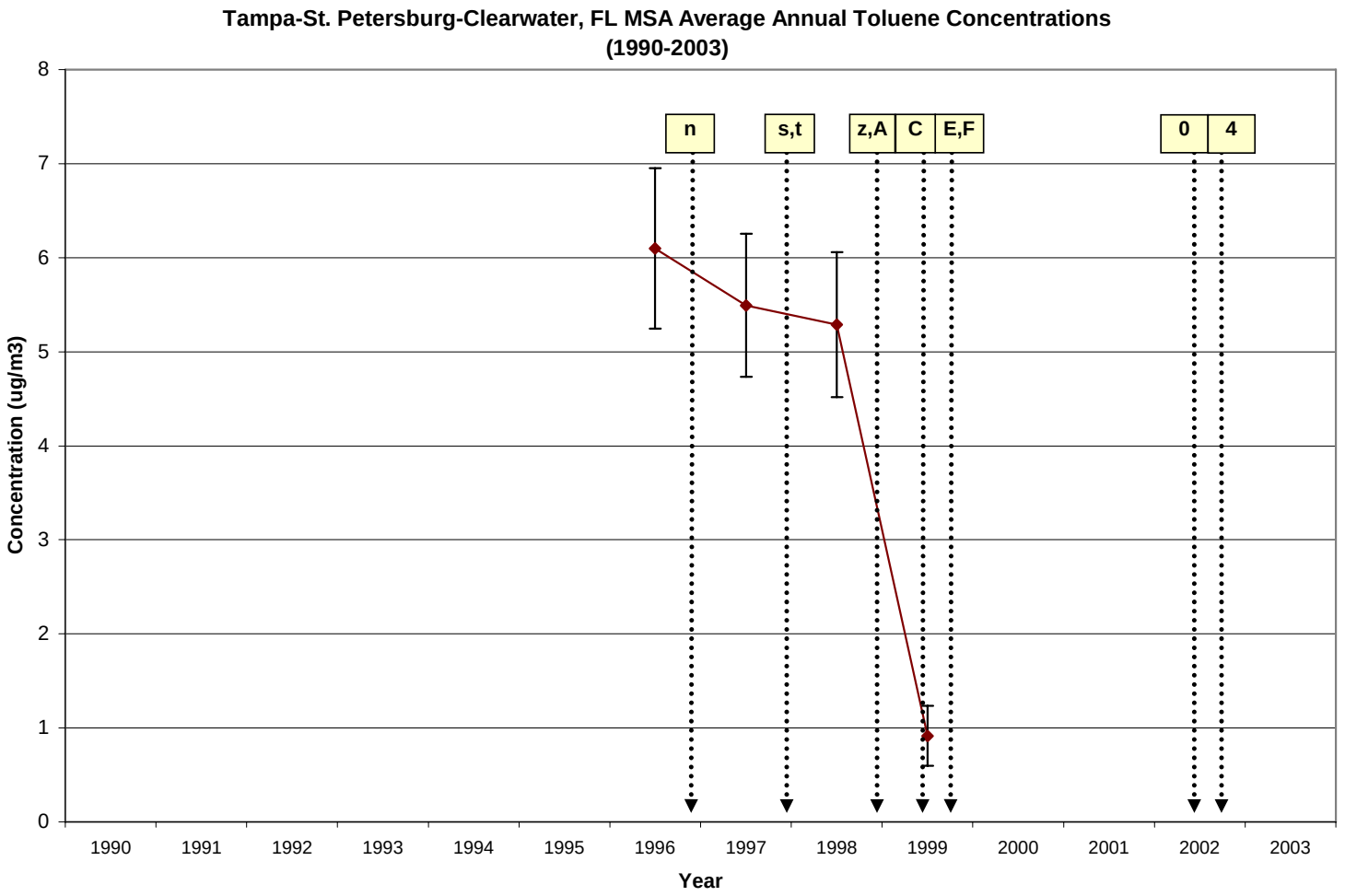
Mercury concentrations appeared to have decreased after implementation of the Petroleum Refineries MACT (graph key = w), Reformulated Gasoline Phase II program (graph key = I), and the Large Municipal Waste Combustors MACT (graph key = M)

RESULTS – PHILADELPHIA MSA



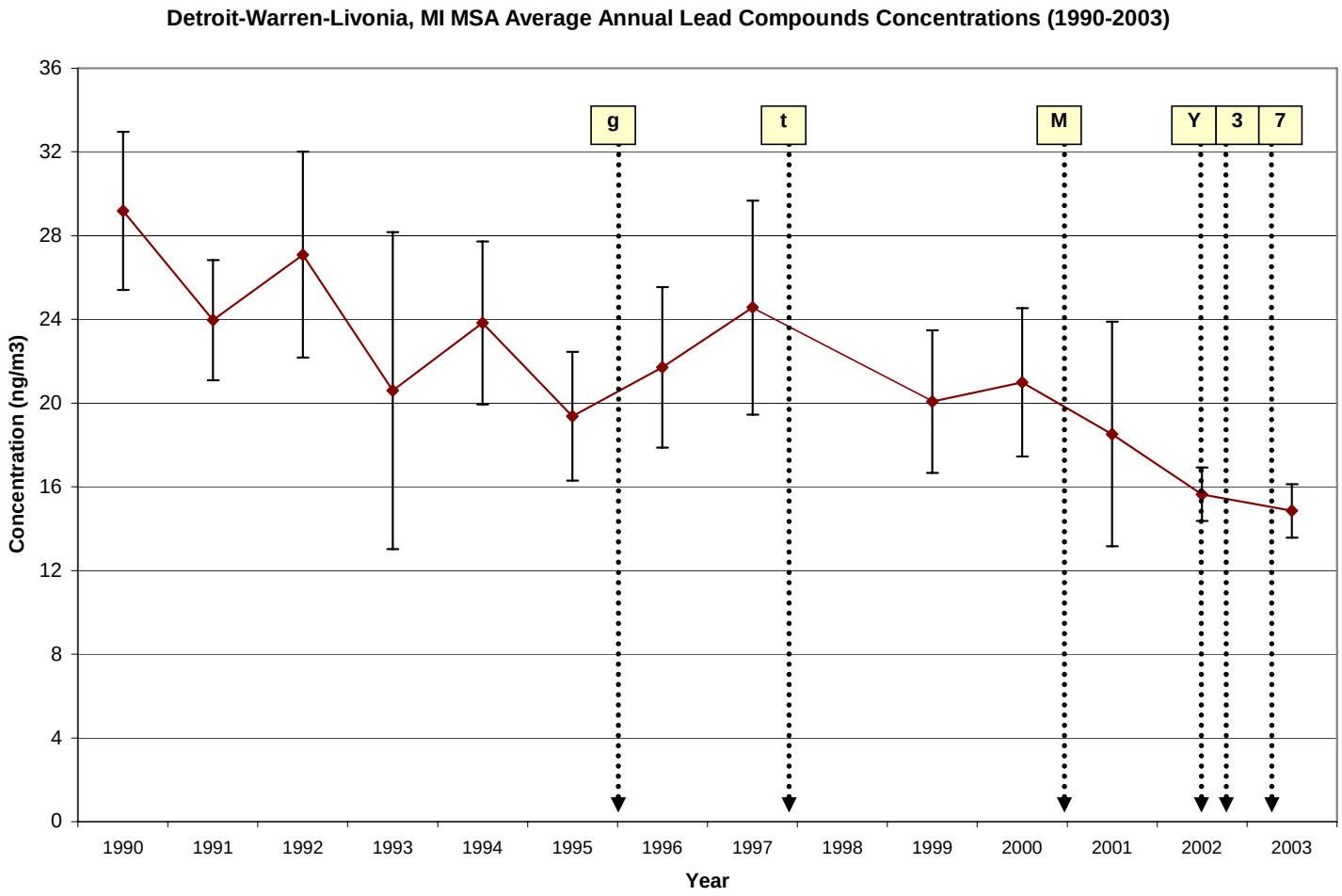
Lead compound concentrations appeared to have decreased substantially after implementation of the Secondary Lead Smelter MACT (graph key = u) and the Gasoline Distribution Stage I MACT (graph key = t)

RESULTS – TAMPA MSA



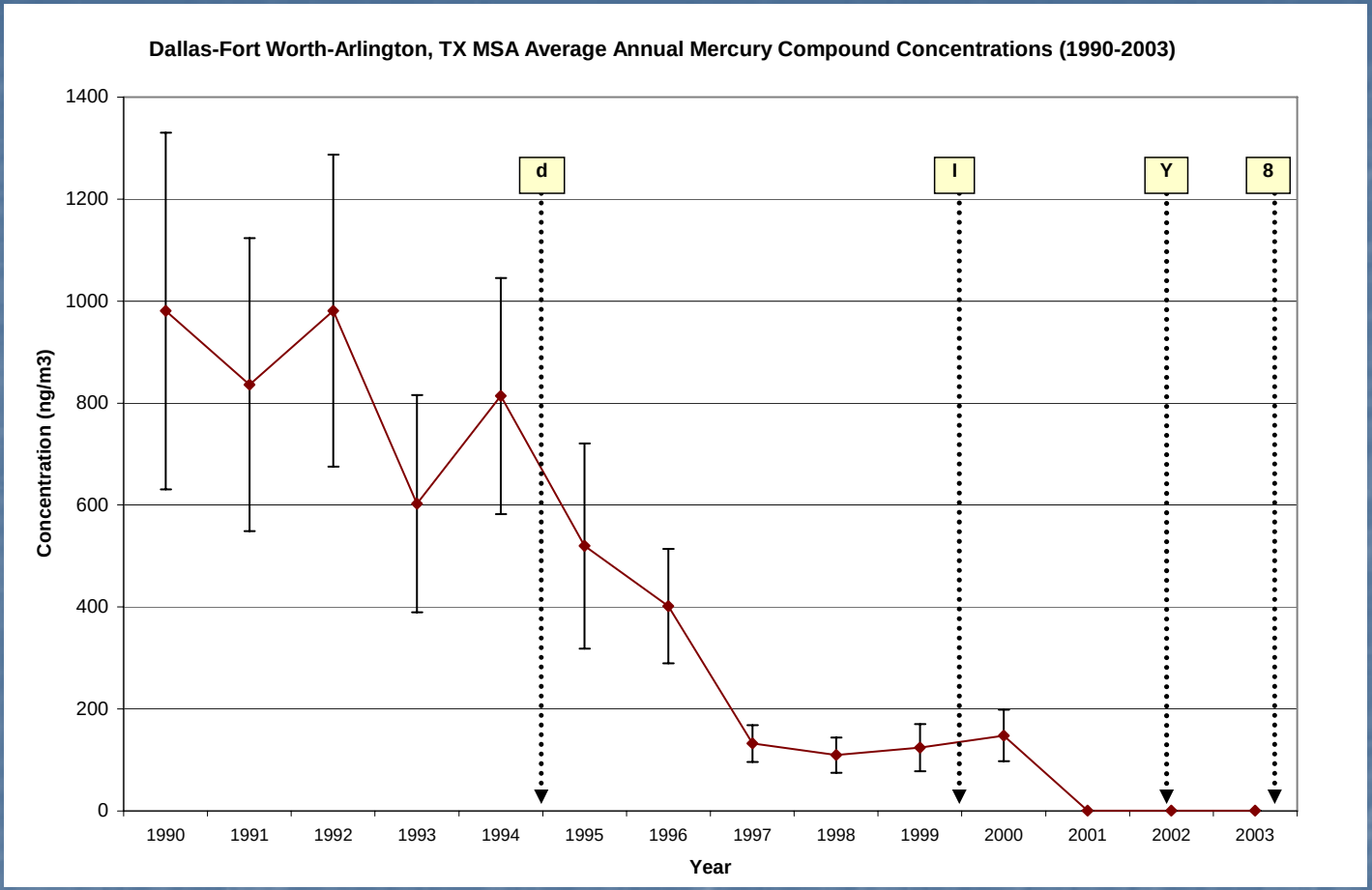
Limited toluene data in the Tampa MSA for the last five years limits the conclusion that concentrations are continuing to decrease.

RESULTS – DETROIT MSA



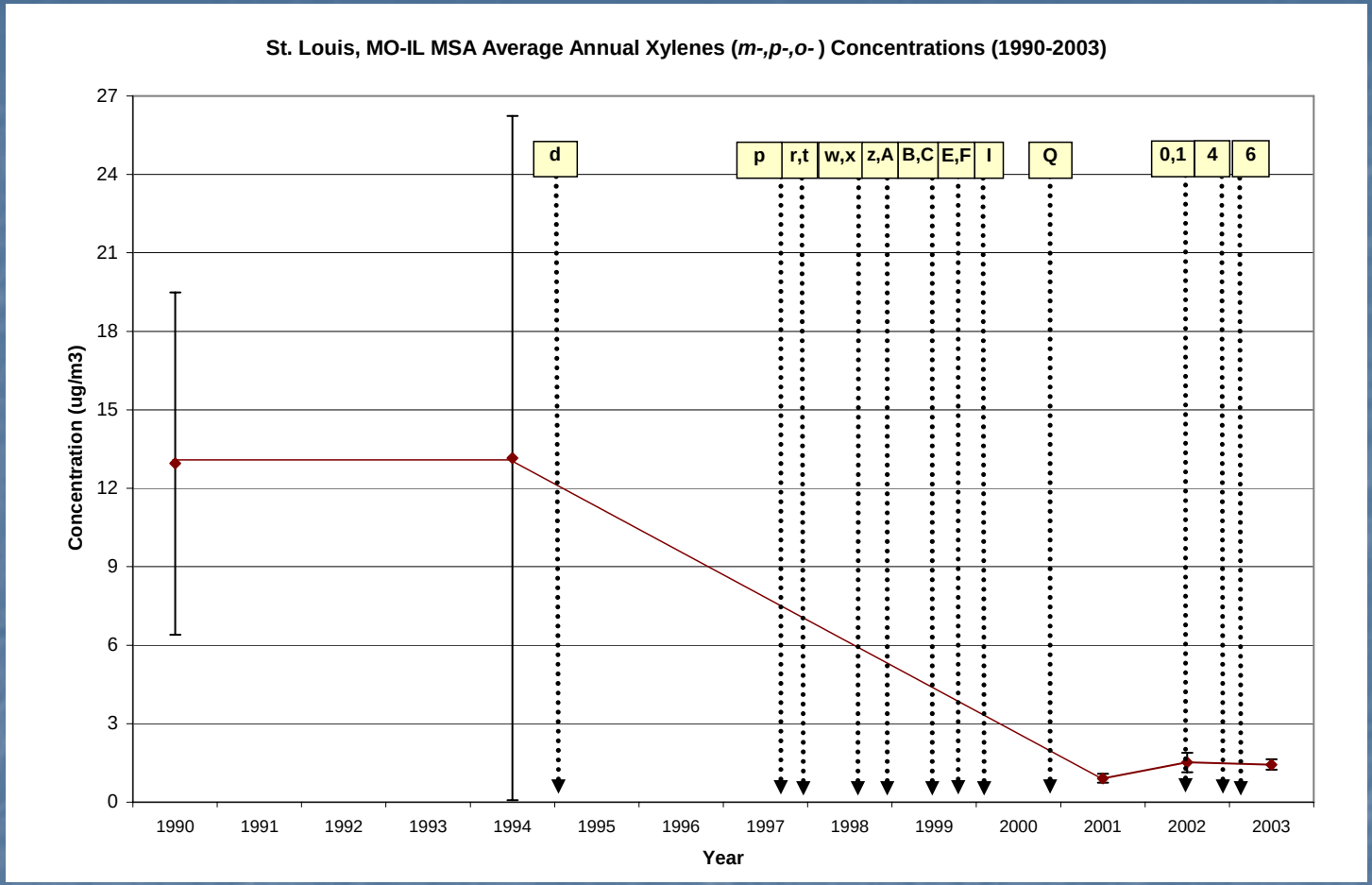
Lead concentrations in Detroit appeared to have decreased steadily during the study period. The biggest affect appears to be the implementation of the Large Municipal Waste Combustors MACT (graph key = M).

RESULTS – DALLAS MSA



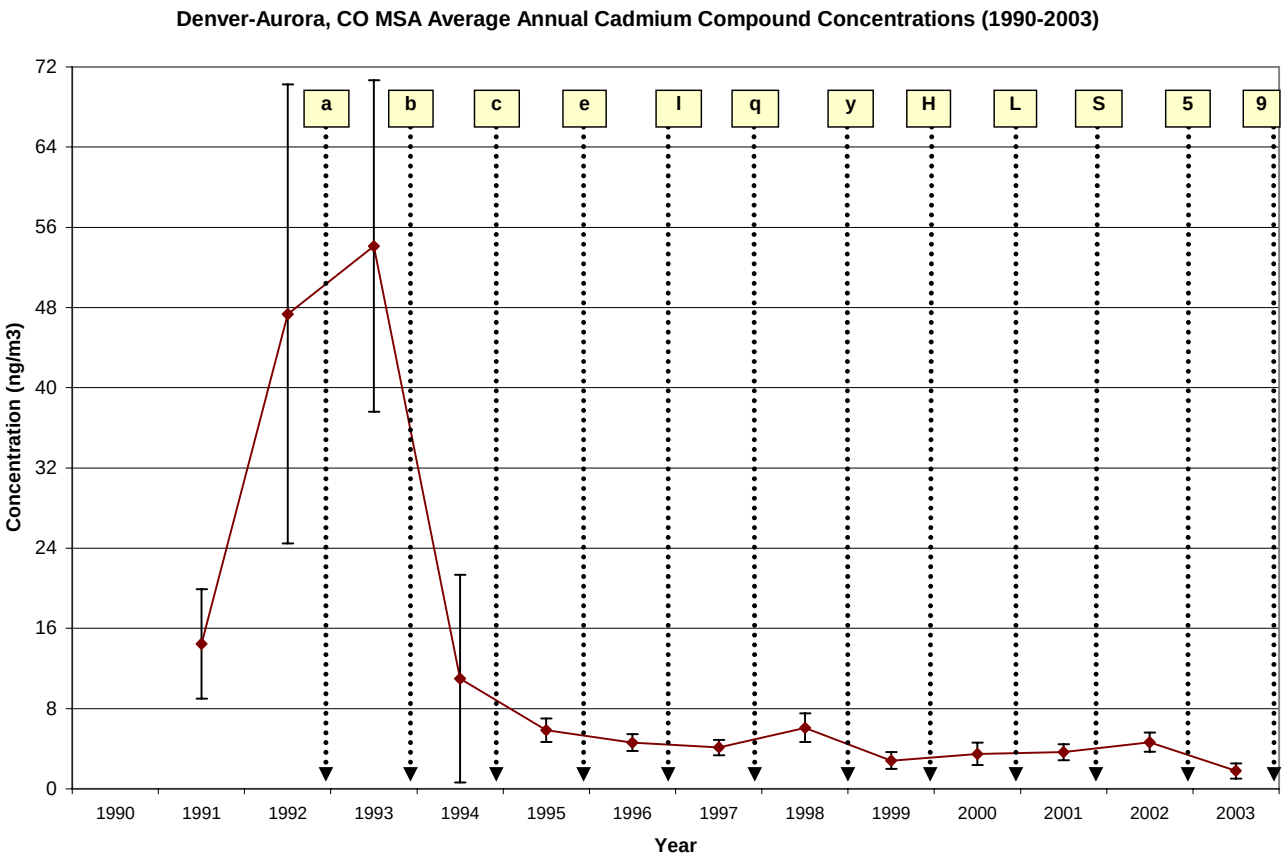
Mercury compound concentrations appear to have decrease substantially with the implementation of the Reformulated Gasoline Phase 1 Program (graph key = d). However, according to the NEI, mercury emissions from Hazardous Waste Combustors decreased by 97% from 1996 to 2002 for this MSA, most likely as a result of impending regulations (graph key = 8).

RESULTS – ST. LOUIS MSA



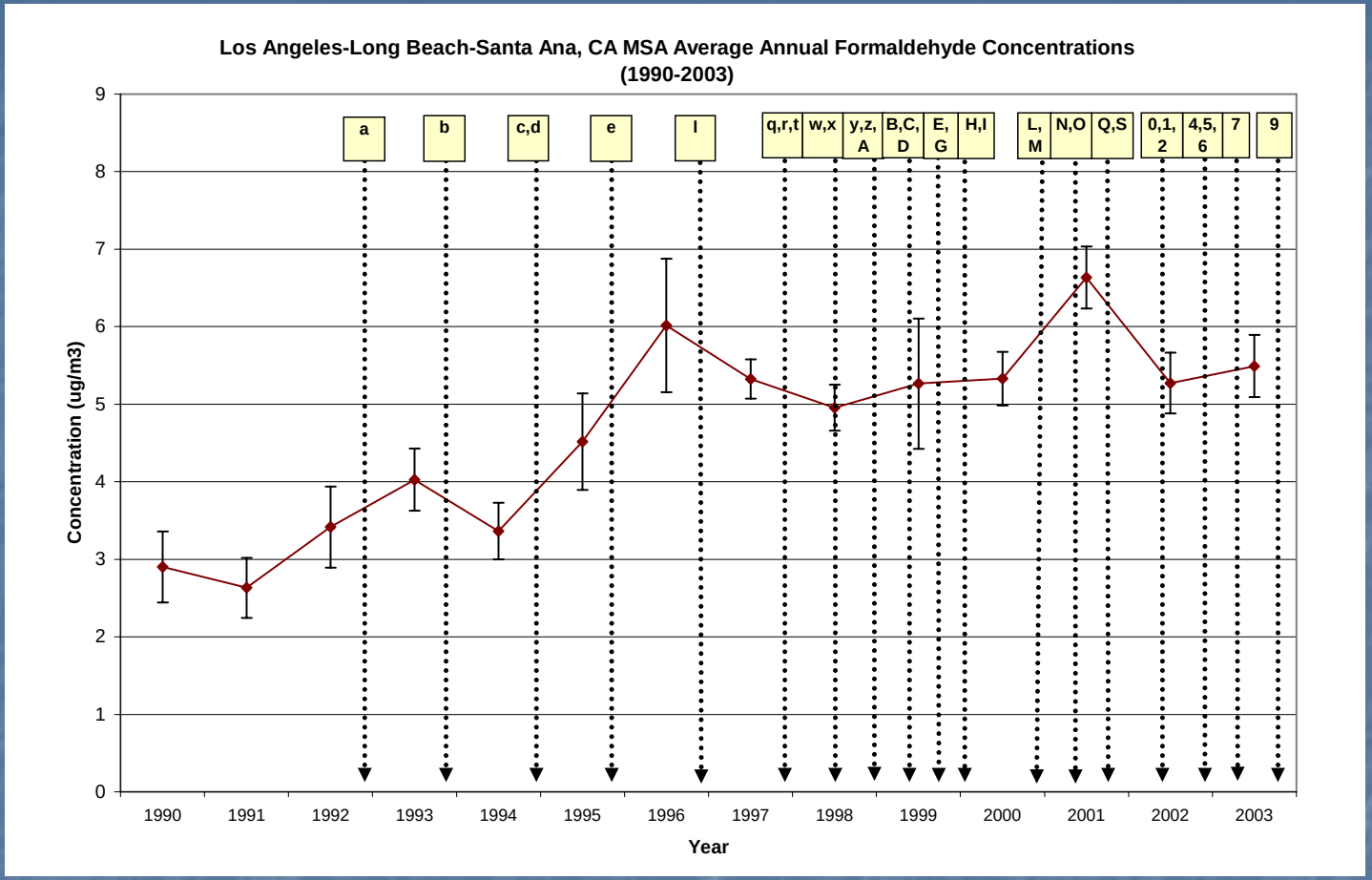
Total xylene concentrations in the St. Louis MSA declined dramatically from 1990 to 2003, apparently in response to several implemented stationary and mobile source regulations targeting VOCs (graph keys = d-Q). However, limited data availability of xylene measurements between 1995 and 2000 limit the certainty of these conclusions.

RESULTS – DENVER MSA



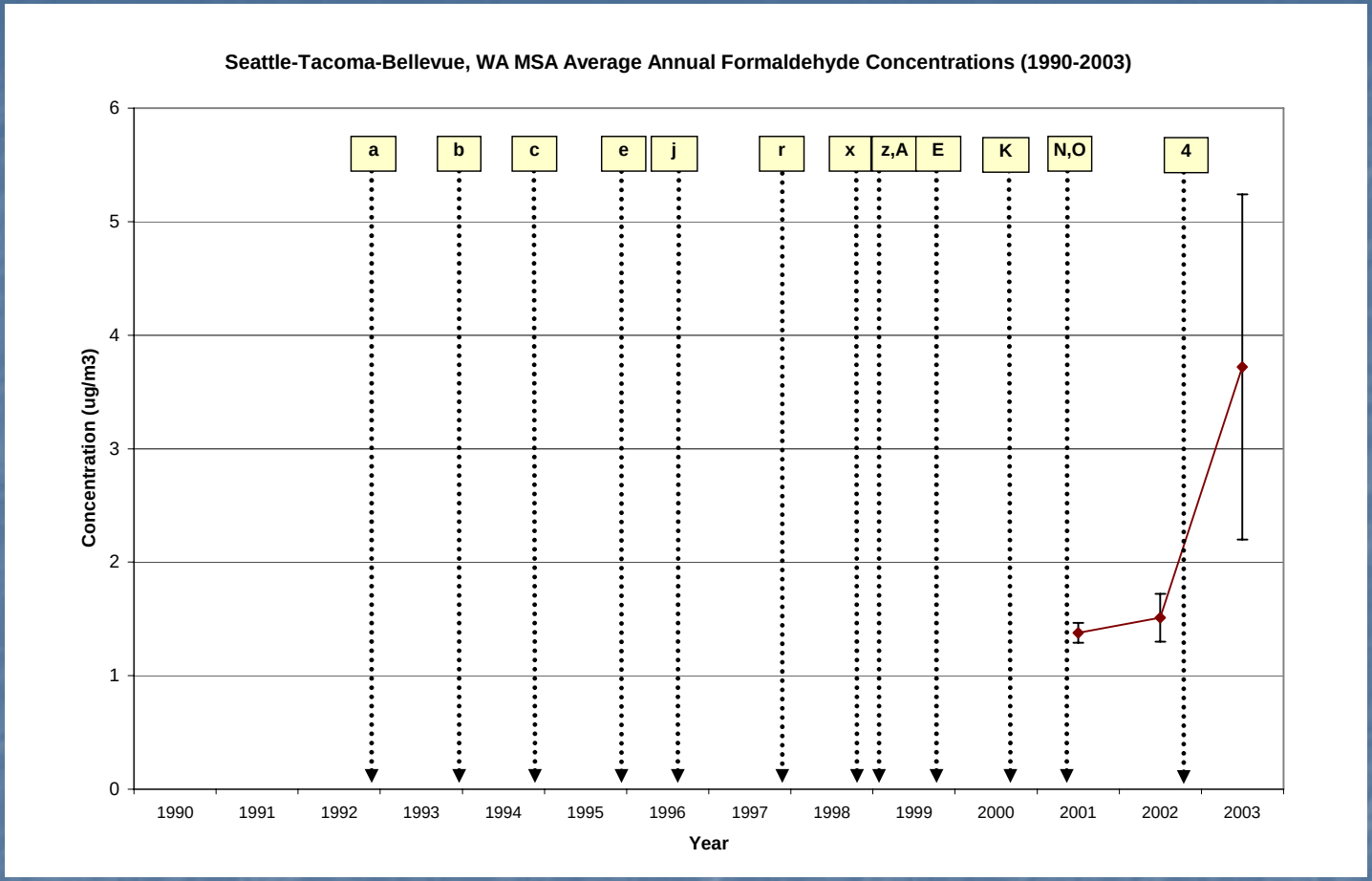
No implemented stationary source regulations were identified in the Denver MSA which apparently affected cadmium concentrations. According to TRI, cadmium emissions from Asarco, Inc. Globe Plant decreased from 0.198 tpy in 1990 to 0.077 tpy in 1994, a 61% decrease. Cadmium emissions remained steady for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

RESULTS – LOS ANGELES MSA



Formaldehyde concentrations have steadily increased during the study period. This is most likely due to the implementation of the Reformulated Gasoline Phase I Program (graph key =d).

RESULTS – SEATTLE MSA



Over the last three years, formaldehyde concentrations have appeared to have increased, noticeably after the implementation of the Publicly Owned Treatment Works MACT (graph key = 4). However, formaldehyde data prior to 2001 is limited or unavailable, thus making it difficult to characterize a trend.

CONCLUSIONS

What are the HAP concentration trends?

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.

Additionally, more than half of the percentage reduction comparisons for concentrations and emissions (30 of 57) were within 20% of each other, with one as close as 1% (e.g., Los Angeles total xylene emissions: 73% decrease in emissions, 74% decrease in average concentration).

CONCLUSIONS

Have HAP-specific federal regulations been effective at 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:

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

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

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

CONCLUSIONS

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

- HAP emissions for each MSA decreased substantially from 1990 to 2002.
- Total emissions across the ten MSAs for each HAP decreased from 580,000 tpy to 270,000 tpy (53% reduction).
- Emissions in the Los Angeles MSA decreased the most among the MSAs (69% reduction, 86,000 tpy).
- For the HAPs, mercury emissions realized the highest percent reduction (80%), while toluene emissions realized the highest mass reduction (140,000 tpy).

QUESTIONS?