

Air Toxics

<http://www.epa.gov/oar/aqtrnd99/chapter5.pdf>

Worth Noting:

- For all 188 HAPs, there is a 23-percent reduction in emissions between the 1990–1993 baseline and 1996. For the 33 urban HAPs, there is a 30-percent reduction in air toxics emissions between baseline and 1996. The majority of these reductions are attributable to two source types with existing regulatory programs: major sources and onroad mobile sources.
- Ambient monitoring results generally reveal downward trends for most monitored HAPs. The most consistent improvements are apparent for benzene which is predominantly emitted by mobile sources; and for total suspended lead. From 1994–1999, annual average concentrations for these two HAPs declined 40 and 47 percent respectively.

Background

Hazardous air pollutants (HAPs), commonly referred to as air toxics or toxic air pollutants are pollutants known to cause or suspected of causing cancer or other serious human health effects or ecosystem damage. The Clean Air Act (CAA) lists 188 HAPs and directs EPA to regulate sources emitting major amounts of these identified pollutants.¹ Examples of HAPs include heavy metals (e.g., mercury and chromium), volatile chemicals (e.g., benzene and perchlo-

roethylene), combustion byproducts (e.g., dioxins) and solvents (e.g., carbon tetrachloride and methylene chloride). In addition, EPA has recently listed diesel particulate matter plus diesel exhaust organic gases as a mobile source air toxic and has addressed diesel exhaust in several regulatory actions. EPA's list of mobile source air toxics also includes 20 other pollutants which are included among the list of 188 HAPs.

Hazardous air pollutants are emitted from literally thousands of sources including large stationary industrial facilities (such as electric power plants), smaller area sources (such as neighborhood dry cleaners), mobile sources (such as automobiles), indoor sources (such as some building materials and cleaning solvents), and other sources (such as wildfires).

Factors such as weather, the terrain (i.e., mountains, plains, valleys), and the chemical and physical properties of a pollutant determine how far it is transported, its concentration at various distances from the source, what kind of physical and chemical changes it undergoes, and whether it will degrade, remain airborne, or deposit to land or water. Some HAPs (such as chromium) remain airborne and contribute to air pollution problems far from the pollution source. Other HAPs (such as mercury) are released into the air and can be deposited to land and water bodies through

precipitation, or by settling directly out of the air onto land or water.

Potential Effects of Air Toxics

Human Health

- Cancer
- Birth defects
- Developmental delays
- Reduced immunity
- Difficulty in breathing and respiratory damage
- Headache, dizziness, and nausea

Environmental

- Reproductive effects and developmental delays in wildlife
- Toxicity to aquatic plants and animals
- Accumulation of pollutants in the food chain

Health and Environmental Effects

The degree to which a toxic air pollutant affects a person's health depends on many factors, including the quantity of pollutant the person is exposed to, the duration and frequency of exposures, the toxicity of the pollutant, and the person's state of health and susceptibility. The different health effects that may be caused by HAPs include cancer; neurological, cardiovascular, and respiratory

effects; effects on the liver, kidney, immune system, and reproductive system; and effects on fetal and child development. The timing and severity of the effect (e.g., minor or reversible vs. serious, irreversible, and life-threatening) may vary among HAPs and with the exposure circumstances. In some cases effects can be seen immediately; in other cases the resulting effects (e.g., liver damage or cancer) are associated with long-term exposures and may not appear until years after exposure. Roughly half of the 188 HAPs have been classified by EPA as “known,” “probable,” or “possible” human carcinogens. Known human carcinogens are those that have been demonstrated to cause cancer in humans. Examples include benzene, which has caused leukemia in workers exposed over several years in their workplace air, and arsenic, which has been associated with elevated lung cancer rates in workers at metal smelters. Probable and possible human carcinogens include chemicals that we are less certain cause cancer in people, yet for which laboratory animal testing or limited human data indicate carcinogenic effects. For example, EPA concluded that diesel exhaust is likely to be carcinogenic to humans at environmental levels that the public faces (classifying it as a “probable human carcinogen”).²

Some HAPs pose particular hazards to people of a certain stage in life (e.g., young children, adolescents, adults, or elderly people). Some HAPs are developmental or reproductive toxicants in humans. This means that exposure before birth or during childhood may interfere with normal development into a healthy adult. Other such exposures may affect the ability to conceive or give

birth to a healthy child. Ethylene oxide, for example, has been associated with increased miscarriages in exposed workers and has affected reproductive ability in both male and female laboratory animals.

Some HAPs are of particular concern because they degrade very slowly or not at all, as in the case of metals such as mercury or lead. These persistent HAPs can remain in the environment for a long time and can be transported great distances. Persistent and bioaccumulative HAPs are of particular concern in aquatic ecosystems because the pollutants accumulate in sediments and may biomagnify in tissues of animals at the top of the food chain through consumption or uptake to concentrations many times higher than in the water or air. In this case, exposure to people occurs by eating contaminated food from waters polluted from the deposition of these HAPs. As of July 2000, for example, 40 states and the American Samoa have issued fish consumption advisories for mercury. Thirteen of those states have issued advisories for all water bodies in their state and the other 27 states have issued advisories for more than 1900 specific water bodies.³

Hazardous air pollutants can have a variety of environmental impacts in addition to the threats they pose to human health. Like humans, animals can experience health problems if they are exposed to sufficient concentrations of HAPs over time. For example, exposures to PCBs, dioxins, and dibenzo-furans are suspected of causing death and deformities to various bird chicks.⁴ These pollutants are also thought to have had adverse impacts on reproduction of lake trout.⁵ Mercury is also thought to pose a significant risk to wildlife. Meth-

ylmercury levels in fish in numerous waterbodies have been shown to exceed levels associated with adverse effects on birds.⁶ These and other observations have led some scientists to conclude that fish-eating birds and mammals occupying a variety of habitats are at risk due to high levels of methylmercury in aquatic food webs.

National Air Toxics Control Program

Since 1990, EPA has made considerable progress in reducing emissions of air toxics through regulatory, voluntary, and other programs. To date, the overall air toxics program has focused on reducing emissions of the 188 air toxics from major stationary sources through the implementation of technology-based emissions standards as specified by Congress in the 1990 CAA Amendments. These actions have resulted in, or are projected to result in, substantial reductions in air toxics emissions. Additionally, actions to address mobile sources under other CAA programs have achieved significant reductions in air toxics emissions (e.g., the phase-out of lead from gasoline). Many motor vehicle and fuel emission control programs of the past have reduced air toxics. Several current EPA programs further reduce air toxics emissions from a wide variety of mobile sources. These include the reformulated gasoline (RFG) program, the national low emission vehicle (NLEV) program, and Tier 2 motor vehicle emission standards and gasoline sulfur control requirements. In addition, EPA has recently issued regulations to address emissions of toxic air pollutants from motor vehicles and their fuels as well as stringent standards for heavy-duty trucks and buses and diesel fuel that will lead to a reduction in emis-

Table 5-1. List of 33 Urban Air Toxics Strategy HAPs

VOCs	Metals (Inorganic Compounds)	Aldehydes (Carbonyl Compounds)	SVOCs & Other HAPs
acrylonitrile	arsenic compounds	acetaldehyde	2,3,7,8-tetrachlorodi benzo-p-dioxin (& congeners & TCDF congeners)
benzene	beryllium and compounds	formaldehyde	coke oven emissions
1,3-butadiene	cadmium compounds	acrolein	hexachlorobenzene
carbon tetrachloride	chromium compounds		hydrazine
chloroform	lead compounds		polycyclic organic matter (POM)
1,2-dibromoethane (ethylene dibromide)	manganese compounds		polychlorinated biphenyls (PCBs)
1,3-dichloropropene	mercury compounds		quinoline
1,2-dichloropropane (propylene dichloride)	nickel compounds		
ethylene dichloride, EDC (1,2-dichloroethane)			
ethylene oxide			
methylene chloride (dichloromethane)			
1,1,1,2,-tetrachloroethane			
tetrachloroethylene (perchloroethylene, PCE)			
trichloroethylene, TCE			
vinyl chloride			

EPA’s Integrated Urban Air Toxics strategy identified 33 HAPs which are judged to pose the greatest threat to public health in urban areas.⁷ These 33 “urban HAPs” are a subset of EPA’s list of 188. Under EPA’s urban strategy, the Agency is developing area source regulations that will control those sources responsible for 90 percent of the total emissions of the 33 HAPs in urban areas. The list of the 33 urban HAPs is presented in Table 5-1 and is grouped according to chemical properties (volatile organic compounds (VOCs), metals, aldehydes, and semi-volatile organic compounds [SVOCs]). This grouping is the same breakdown EPA uses for ambient monitoring which is discussed in a subsequent section of this chapter.

In addition to national regulatory efforts, EPA provides leadership and technical and financial assistance for the development of cooperative federal, state, local, and tribal programs to prevent and control air pollution. EPA’s risk initiatives include comprehensive local-scale assessments, as well as federal and regional activities associated with air toxics deposition (e.g., the Great Waters program (includes the Great Lakes, Lake Champlain, Chesapeake Bay, and many U.S. coastal estuaries) and Agency initiatives concerning mercury and other persistent and bio-accumulative toxics [PBTs]).

EPA also has an ongoing comprehensive evaluation of air toxics in the United States which is called the National Air Toxics Assessment (NATA). These NATA activities help EPA identify areas of concern, characterize risks, and track progress toward meeting the air toxics program goals to reduce risk to human health and the environment. They include expansion of air toxics monitoring,

sions of diesel particulate matter by over 90 percent between 1996 and 2020. From 1996 (the year of the most up-to-date emissions inventory estimates) to 2020, the existing proposed mobile source programs are also expected to lower onroad emissions of benzene by 61 percent, formaldehyde by 78 percent, 1,3-butadiene by 60 percent, and acetaldehyde by 73 percent from the 1996 levels. There will also be substantial reductions from other gaseous onroad HAPs and from nonroad mobile sources.

EPA expects, however, that the emission reductions that will result from these actions may only be part of what is necessary to protect public health and the environment from air toxics. In accordance with the 1990 CAA Amendments, EPA has begun to assess the risk remaining (i.e., the

residual risk) after implementation of technology-based standards in order to evaluate the need for additional stationary source standards to protect public health and the environment. During 2001, EPA will also begin the process for assessing new standards for nonroad engines such as construction and farm equipment. In addition, after extensive study, EPA determined mercury emissions from power plants pose significant hazards to public health and must be reduced. EPA will propose regulations by 2003 and issue final rules by 2004. By July 2003, EPA will reassess the need for and feasibility of controls for onroad and nonroad sources of air toxics, and propose any additional vehicle and fuel controls that the Agency determines are appropriate. This rulemaking will be finalized by July 2004.

improving and periodically updating emissions inventories, developing better air toxics emission factors for nonroad sources, improving national- and local-scale modeling, continued research on health effects and exposures to both ambient and indoor air, and improvement of assessment tools.

For indoor air toxics, EPA's program has relied on education and outreach to achieve reductions. EPA's voluntary programs that focus on indoor air pollution have been very successful in reducing indoor air pollution. For example, through EPA's voluntary *Tools for Schools Pro-*

Examples of Source Types

- **Major sources:** large industrial sources such as chemical plants, oil refineries and steel mills.
- **Area and other sources:** smaller industrial sources such as drycleaners, gas stations and landfills, as well as natural sources like wildfires.
- **Onroad mobile sources:** cars, heavy-duty trucks, buses and other highway vehicles.
- **Nonroad mobile sources:** construction and farm equipment as well as recreational vehicles.

gram, there have been significant reductions in children's exposure to air toxics in 4,000 schools across the country. EPA is also developing a specific strategy for indoor air toxics that will present an approach to evaluate information, characterize potential indoor exposures and risks, and identify methods to reduce air toxics indoors. Additional information about indoor air toxics activities is available at: www.epa.gov/iaq/pubs/index.html.

Figure 5-1. National contribution of source types to 1996 NTI emissions for the 188 HAPs.

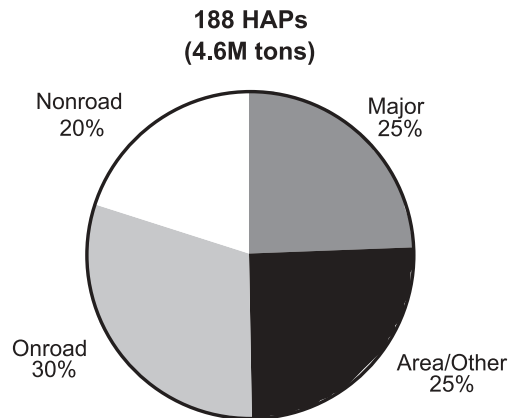
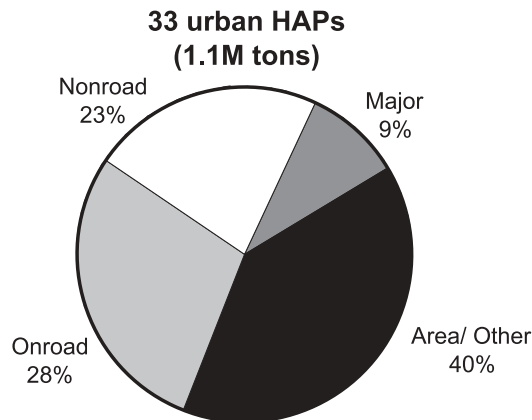


Figure 5-2. National contribution of source types to 1996 NTI emissions for the urban HAPs.

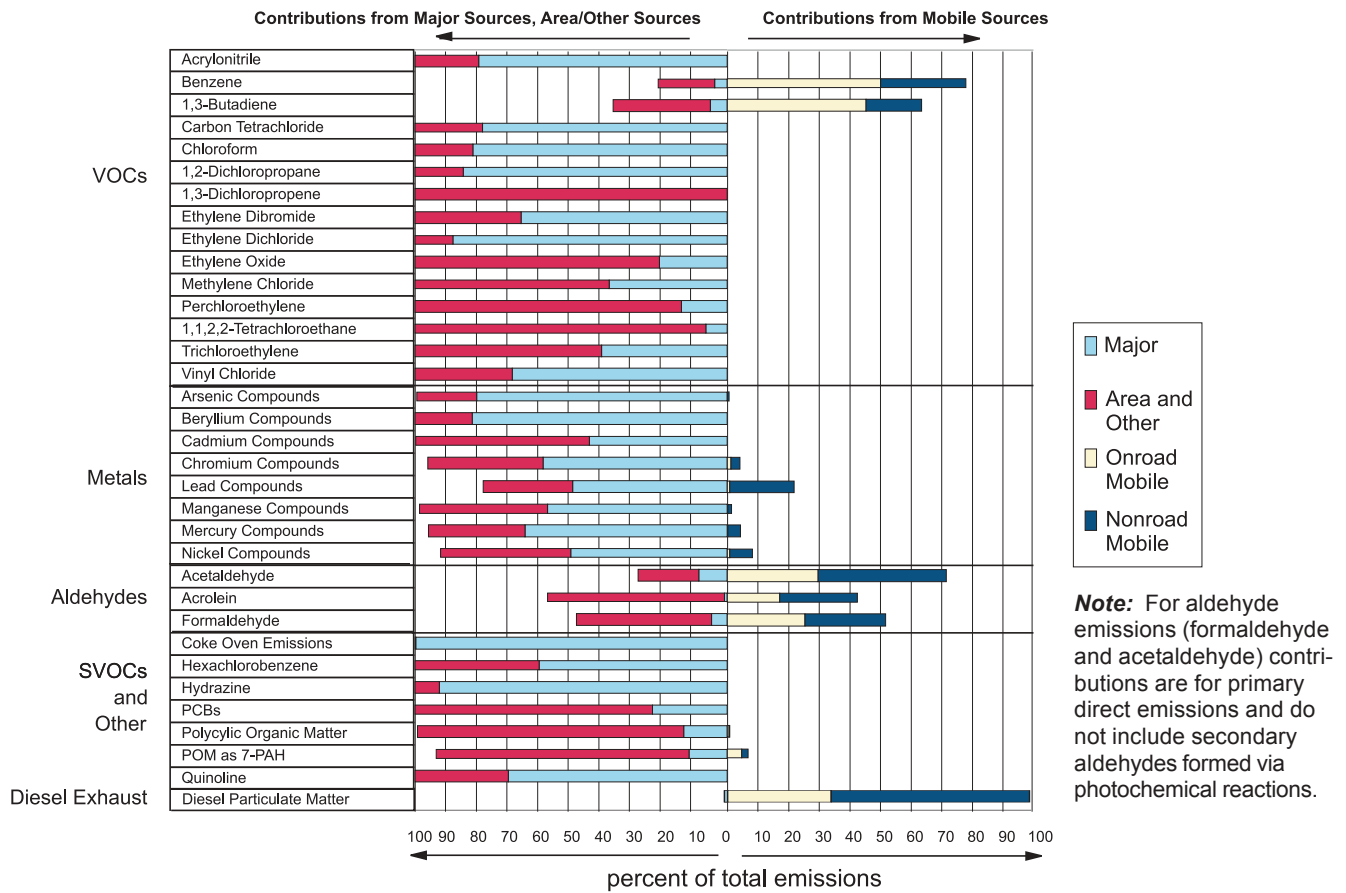


Air Toxics Emissions in 1996

The National Toxics Inventory (NTI) is EPA's compilation of quantitative information concerning the mass of emissions of HAPs emitted into the atmosphere (through smokestacks, tailpipes, vents, etc.) from stationary and mobile sources. The NTI is developed every 3 years. EPA has compiled both a baseline period (1990–1993) as well as 1996 emissions estimates for the 188 HAPs. As of the date of this publication, the 1996 NTI contains the most complete, up-to-date air toxics emissions estimates available. However, EPA has not yet included the 1996

dioxin emissions in the 1996 NTI since they are still under review. Since dioxin emissions are relatively small, its exclusion does not affect the summary information presented for the 188 or the 33 urban HAPs. In addition, these emission summaries do not include diesel particulate matter. For purposes of this report, the information in the NTI has been divided into four overarching source types: 1) large industrial or "major" sources; 2) "area and other sources," which include smaller industrial sources, such as small drycleaners and gasoline stations, as well as natural sources, such as wildfires; 3) "onroad"

Figure 5-3. National contribution by emission source type for individual urban HAPs and diesel particulate matter, 1996.



Note: For aldehyde emissions (formaldehyde and acetaldehyde) contributions are for primary direct emissions and do not include secondary aldehydes formed via photochemical reactions.

mobile, including highway vehicles; and 4) "nonroad" mobile sources, like aircraft, locomotives, and lawn mowers.

Figures 5-1 and 5-2 provide a summary of the national emissions in the 1996 NTI based on source types for the 188 HAPs as well as the 33 urban HAPs, respectively. Note that emissions of the 33 urban HAPs represent roughly a quarter (23 percent) of the 1996 emissions of the 188 HAPs. As shown in Figure 5-1, the national emissions of the 188 HAPs are relatively equally divided between the four types of sources. For the 33 urban HAPs, however, area and other sources are the largest overall contributor (40 percent), while major sources account for less than 10 percent of the nationwide emis-

sions and mobile sources make up the remaining 51 percent.

Figure 5-3 provides the percent of emissions by source type for each of the 33 urban HAPs that have available emissions information (i.e., excluding dioxin). It also contains information on diesel particulate matter. Note that for each bar, the individual contributions total to 100 percent. Also, the center vertical line in the chart is zero so that the mobile source contributions are shown on the right side of the chart for ease of display. The contributions from each source type vary by pollutant. For example, acetaldehyde and benzene have mobile sources as the dominant contributor, hydrazine and coke oven emissions are dominated by major

sources, and perchlorethylene is predominantly from area and other sources. Since the other 156 HAPs are not represented here, this graph provides a subset of information on what source types emit which HAPs. For example, nine of the 21 HAPs EPA has identified as mobile source air toxics are not included in Figure 5-3. Table A-21 shows the 21 mobile source air toxics, including diesel particulate matter, and their contributions from mobile sources.

Also, note that Figure 5-3 does not provide any information about the relative magnitude of emissions. For example, benzene and formaldehyde together represent about 64 percent

(roughly 32 percent each) of the total emissions of these 32 urban HAPs. Conversely, 23 of the urban HAPs, including lead, chromium, and PCBs each represent less than 1 percent of the total emissions of the 33 urban HAPs.

Figure 5-4 provides additional detail on the source sector emissions from the 1996 NTI to show the relative percentages of sources that are found in urban versus rural areas for all 188 HAPs. Figure 5-5 shows this same breakdown for the 33 urban HAPs subset. For the 188 HAPs, urban sources dominate the emissions for all source types. For the 33 urban HAPs, there is one source type, area and other sources, which has roughly the same percentage contribution of urban and rural sources.

Trends in Air Toxics Emissions

Trends in air toxics emissions are shown in Figure 5-6 based on comparison of a baseline period of NTI emissions data (1990–1993) to the 1996 NTI. The bar for each time period includes both the national total for the 188 HAPs as well as the fraction of the national emissions that are associated with the urban HAPs. For all 188 HAPs, there is a 23-percent reduction between the baseline and 1996. For the 33 urban HAPs, there is a 30-percent reduction between baseline and 1996. The majority of these reductions are attributable to two source types with existing regulatory programs: major sources and onroad mobile sources. For the 188 HAPs, major source emissions (which accounted for 25 percent of the total emissions in 1996) decreased by 58 percent and onroad mobile source emissions (which accounted for 30 percent of the total emission in 1996) decreased by 16 percent. Although differences in how EPA compiled the

Figure 5-4. Urban/rural splits by source type for the 1996 national emissions of 188 HAPs.

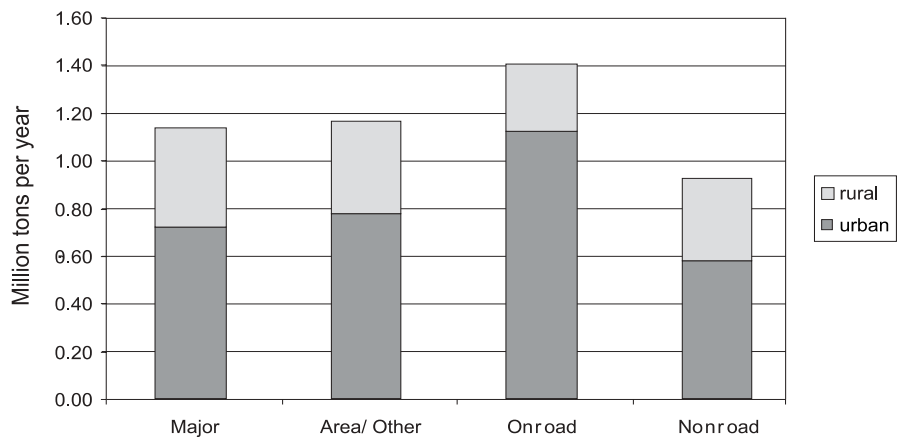
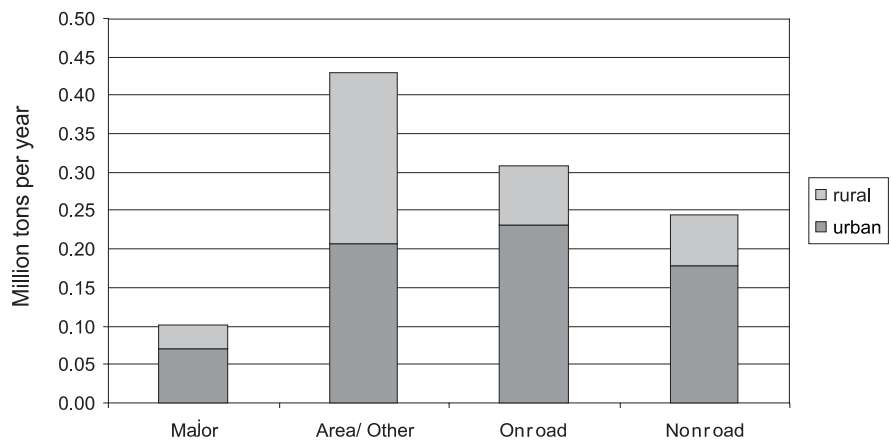


Figure 5-5. Urban/rural splits by source type for the 1996 national emissions of 33 urban HAPs.



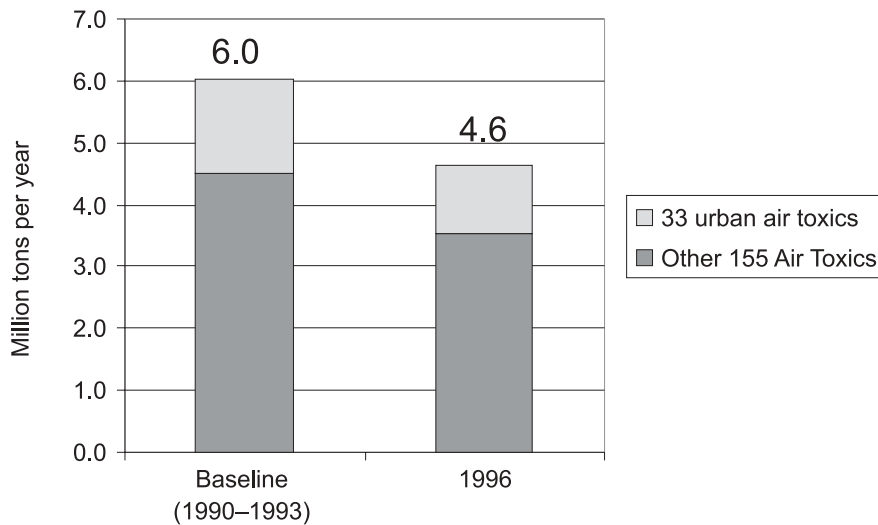
inventory over time could account for some of the current estimates of changes in emissions, EPA and state regulations, as well as voluntary reductions by industry, have clearly achieved large reductions in overall air toxic emissions.

Ambient Monitoring

Ambient measurements, which provide the concentration of a HAP at a particular monitored location at a point in time, are useful to characterize air

quality. These measurements are used to derive trends in HAP concentrations to help evaluate the effectiveness of HAP reduction strategies. They also can provide data to support and evaluate dispersion and deposition models.

Unlike criteria air pollutants, such as carbon monoxide and sulfur dioxide (which have been monitored since the 1970s), there is no national air toxics monitoring system. However, there are approximately 300 monitoring sites currently producing ambient data on

Figure 5-6. Change in national air toxics emissions – baseline (1990–1993) to 1996.

HAPs. These include sites within several states that have long-standing air toxics monitoring programs as well as sites of the Interagency Monitoring of Protected Visual Environments (IMPROVE) visibility network which provides historical information about HAP trace metals in rural areas. The current monitoring sites also include those participating in the Urban Air Toxics Monitoring Program which provides a year's worth of measurements of 39 HAP VOCs and 13 carbonyl compounds.⁸ In addition, the Agency's Photochemical Assessment Monitoring Stations (PAMS) program requires routine year-round measurement of VOCs which include nine HAPs: acetaldehyde, benzene, ethylbenzene, formaldehyde, n-hexane, styrene, toluene, xylenes (m/p-xylene, o-xylene) and 2,2,4-trimethylpentane. For a more detailed discussion of the PAMS program, see the ozone section in Chapter 2 of this report. At the present time, the collection of current state and local air toxics monitoring data and PAMS data is limited in its geographic scope and it

does not cover many HAPs for most states. In addition the sites are not necessarily at locations which represent the highest area-wide concentrations. Nevertheless, they can still be used to provide useful information on the trends in ambient air toxics at this time.

EPA is working together with state and local air monitoring agencies to build upon these sites to develop a monitoring network with the following objectives: to characterize air toxics problems on a national scale; to provide a means to obtain data on a more localized basis as appropriate and necessary (e.g., to evaluate potential "hot spots" near sources), and to help evaluate air quality models. However, there are a significant number of the 188 HAPs for which EPA does not yet have a monitoring method developed. For this reason, EPA is devoting its resources on building up the air toxics monitoring network by first focusing on the 33 urban HAPs. The states currently have the capability to monitor for 28 of the 33 urban HAPs. As the moni-

toring network is enhanced, EPA will assist the states to continue to add to both the geographic scope of the monitoring as well as the number of HAPs included. The network will represent an integration of information from many monitoring programs, including existing state and local air toxic monitoring sites; PAMS, and the new urban PM_{2.5} chemical speciation and rural IMPROVE program networks. This new national network will be developed over the next several years.⁹

Trends In Ambient Concentrations

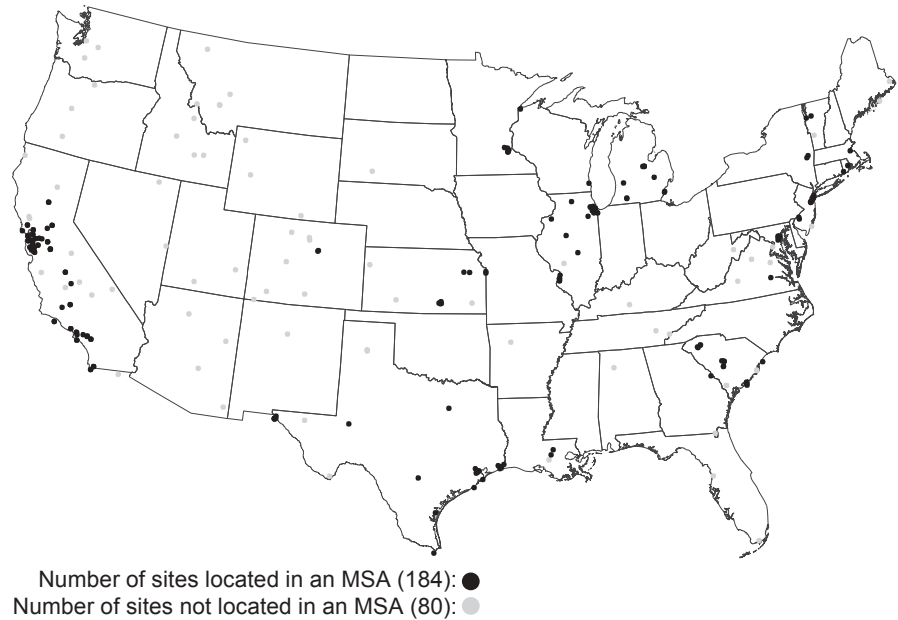
The most widely measured HAP has been lead, which is also a criteria pollutant. Until recently, it has been monitored in most states, both in metropolitan and non-metropolitan areas. Nineteen states have monitored other urban HAPs in their metropolitan areas since 1994. In addition, several VOCs, aldehydes and metals have good data history in metropolitan areas. Most of these monitors, however, are concentrated in a few states, with 36 percent of them in California alone. Nevertheless, these data can be used to provide a preliminary picture of nationwide trends in air toxics. A good history of several trace metal concentrations in rural areas is derived from the IMPROVE program. However, long-term monitoring in rural areas for VOCs and aldehydes has generally been more limited. The locations for the urban and rural monitors with long-term data are shown in Figure 5-7.

Trends derived from these data are separately presented for metropolitan (urban) and non-metropolitan (rural) sites. Table 5-2 presents a national summary of these 6-year trends in

ambient air toxics concentrations in metropolitan statistical areas. Among the 33 HAPs on the urban strategy list, 25 pollutants have sufficient historical data for this 6-year trends assessment. These air contaminants include 13 of the 15 urban VOCs, all eight urban HAP trace metals, the three aldehydes and several specific polycyclic aromatic hydrocarbons (PAHs). Also included are styrene and toluene, which are two additional pervasive air toxics whose monitoring sites have good nationwide coverage. The table presents the number of sites with increases and decreases in measured ambient concentrations from 1994–1999. For trace metals, the table includes results representing more than one particulate size fraction. Similarly, trends are shown separately for several individual PAHs which are constituents of polycyclic organic matter (POM). For each of these HAPs with sufficient historical data, the number of sites with statistically significant changes are highlighted. When most individual locations reveal a consistent change (and when many are statistically significant), this is more characteristic of a national trend.

Although these ambient air toxics data are only available for a limited number of metropolitan areas, the results generally reveal downward trends for most monitored HAPs. The most consistent improvements are apparent for benzene which is predominantly emitted by mobile sources; and for total suspended lead. From 1994–1999, annual average concentrations for these two HAPs declined 40 and 47 percent respectively. The majority of ambient concentrations of lead once came from the tail pipe of cars. Since the mid-90s, however, lead has been largely

Figure 5-7. Locations for urban and rural air toxics monitors with long-term data.



removed from gasoline and almost all of these trace elements now typically emanate from major point sources and aircrafts with piston engines (e.g., small commuter aircraft). The criteria pollutant section in Chapter 2 of this report contains more information about particulate lead. The change in national benzene emissions is attributed to a combination of new car emission standards, use of cleaner fuels in many states as well as stationary source emission reductions. Ambient concentrations of toluene (emitted primarily from mobile sources) also show a consistent decrease over most reporting locations. Similar to benzene, annual average toluene concentrations dropped 48 percent. Other HAPs (including styrene) also reveal air quality improvement, but the downward trends are not significant across large numbers of monitoring locations.

Figure 5-8 presents boxplots of the composite urban trends for six HAPs: benzene, 1,3-butadiene, lead, perchlorethylene, styrene and tolu-

ene. These figures depict the concentration distributions among annual averages in metropolitan areas from 1994–1999. The accompanying map displays the number and location of the monitoring “trend” sites. For comparison, the maps also show the number of sites that produced any measurement data during the 6-year period. The average trend lines for benzene, lead and toluene show more improvement in the first few years. The trend for toluene continues through 1999. The benzene trend reveals a small increase between 1998 and 1999.

For the other HAPs in Table 5-2, most urban locations do not reveal predominant or consistent trends among all monitoring areas. In addition, most observed trends for these HAPs are not statistically significant. This is attributed in part to few states with long-term HAP monitoring, to the large year-to-year variability in computed annual average concentrations for some HAPs and the large variety of contributing emission

Table 5-2. National Summary of Ambient HAP Concentration Trends in Metropolitan Areas, 1994–1999

Pollutant Name	Number of Urban Sites by HAP					
	Total	Significant* UP Trend	Non-Significant UP Trend	No Trend	Non-Significant DOWN Trend	Significant* DOWN Trend
Acrylonitrile	4		4			
Benzene	84	2	8		52	22
1,3-Butadiene	62	3	23	5	22	9
Carbon tetrachloride	57	1	10	6	26	14
Chloroform	76	5	24	13	34	
1,2-Dibromoethane	26		3	17	3	3
1,2-Dichloropropane	30		2	11	16	1
Ethylene dichloride	58		5	26	21	6
Methylene chloride	74		19	2	39	14
1,1,2,2-Tetrachloroethane	11		4	3	4	
Perchloroethylene	76		7	5	50	14
Trichloroethylene	66	2	17	8	37	2
Vinyl chloride	55		2	32	18	3
Arsenic (coarse)	9			9		
Arsenic (fine)	8			1	7	
Arsenic (PM ₁₀)	13		1	2	8	2
Arsenic (TSP)	64		8	37	12	7
Beryllium (PM ₁₀)	6			6		
Beryllium (TSP)	25		3	20	2	
Cadmium (PM ₁₀)	6		3		3	
Cadmium (TSP)	58	2	12	10	30	4
Chromium (coarse)	9		1		8	
Chromium (fine)	8		1	1	5	1
Chromium (PM ₁₀)	12	1	7		4	
Chromium (TSP)	70	4	27	9	27	3
Chromium VI	19				9	10
Lead (coarse)	9				7	2
Lead (fine)	8	1			6	1
Lead (PM ₁₀)	26	2	3	14	5	2
Lead (TSP)	241	8	52	2	124	55
Manganese (coarse)	9		1		7	1
Manganese (fine)	8		4		4	
Manganese (PM ₁₀)	12		1		11	
Manganese (TSP)	63		20	1	34	8
Mercury (fine)	8		1	7		
Mercury (PM ₁₀)	6		3		3	
Mercury (TSP)	22	1	16	2	3	
Mercury compounds	2		1		1	
Nickel (coarse)	9		2		5	2
Nickel (fine)	8			1	6	1
Nickel (PM ₁₀)	12		3		9	
Nickel (TSP)	69		12	3	39	15
Acetaldehyde	18	1	9		7	1
Formaldehyde	18	1	12		4	1
Acrolein	6	1	2	3		
Benzo(a)pyrene (total PM ₁₀ & vapor)	18	1	13		4	
Dibenz(a,h)anthracene (total PM ₁₀ & vapor)	18	3	11		4	
Indeno(1,2,3-cd)pyrene (total PM ₁₀ & vapor)	18	1	13		4	
Benzo(b)fluoranthene (total PM ₁₀ & vapor)	18	3	13		2	
Benzo(k)fluoranthene (total PM ₁₀ & vapor)	18	3	11		4	
Styrene	61		13	5	38	5
Toluene	80	1	4		42	33

*Statistically significant at the 10-percent level (See Appendix B: Methodology, Air Toxics Methodology section).

Figure 5-8a. National trend in annual average benzene concentrations in metropolitan areas, 1994–1999.

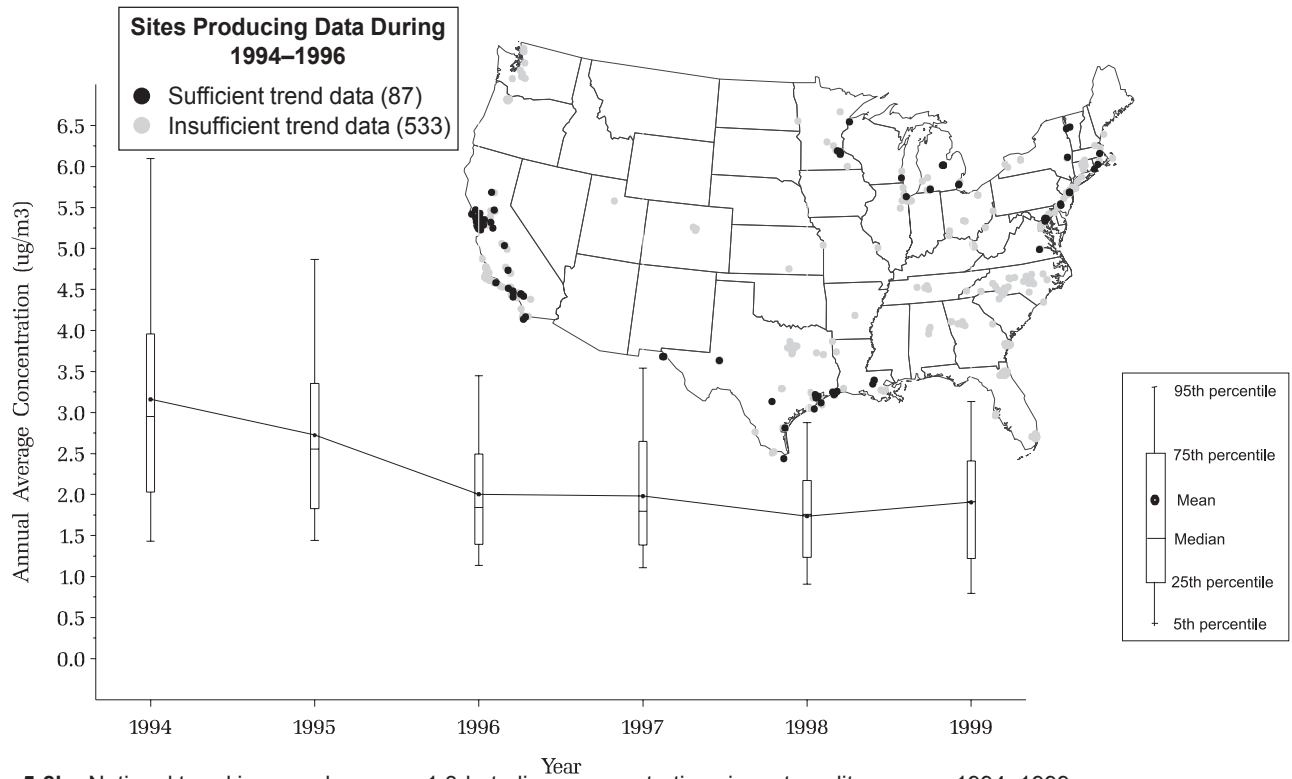


Figure 5-8b. National trend in annual average 1,3-butadiene concentrations in metropolitan areas, 1994–1999.

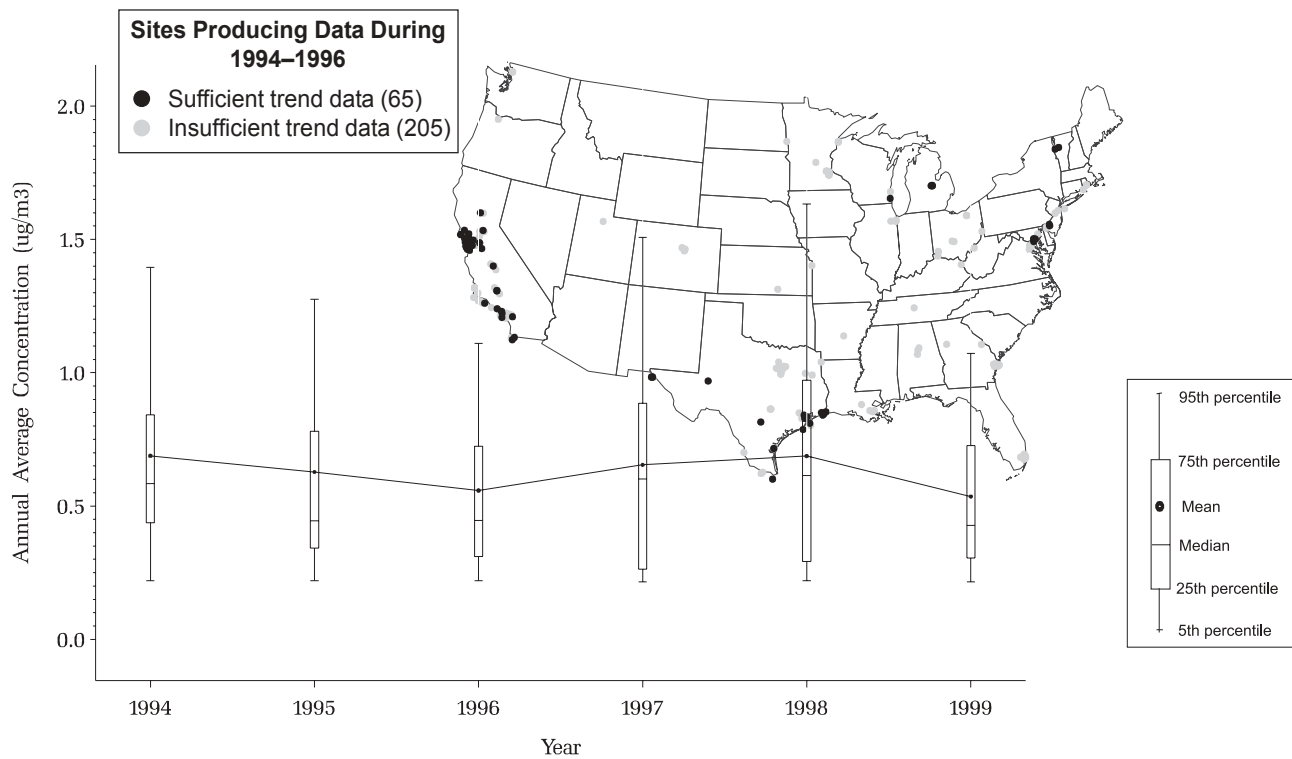


Figure 5-8c. National trend in annual average total suspended lead concentrations in metropolitan areas, 1990–1999.

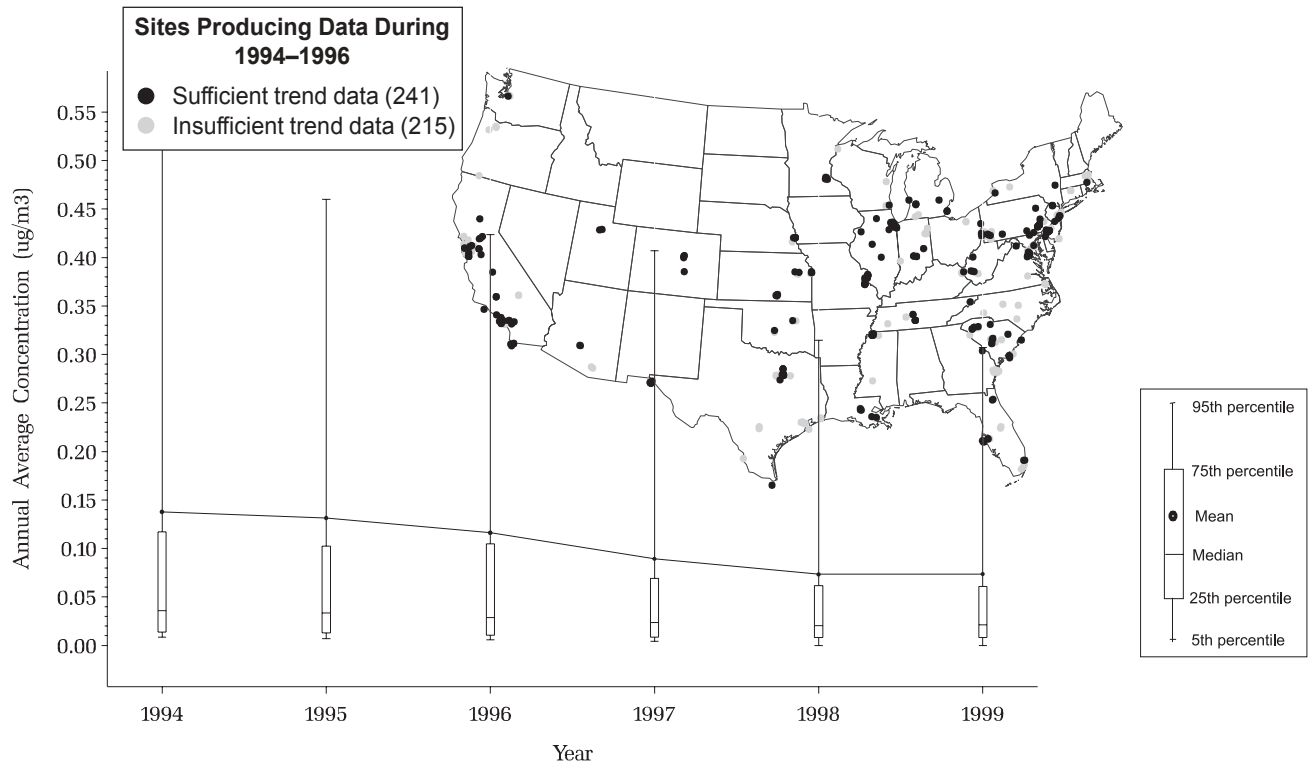


Figure 5-8d. National trend in annual average perchloroethylene concentrations in metropolitan areas, 1990–1999.

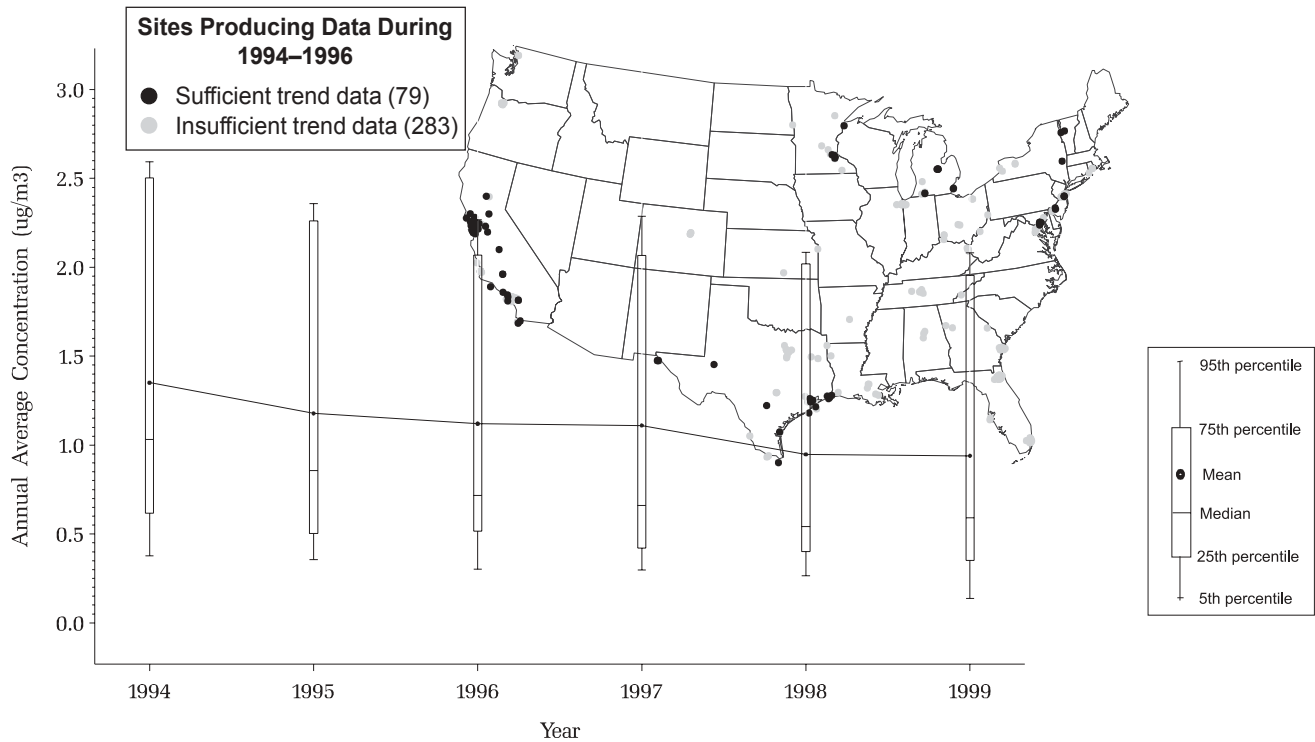


Figure 5-8e. National trend in annual average styrene concentrations in metropolitan areas, 1994–1999.

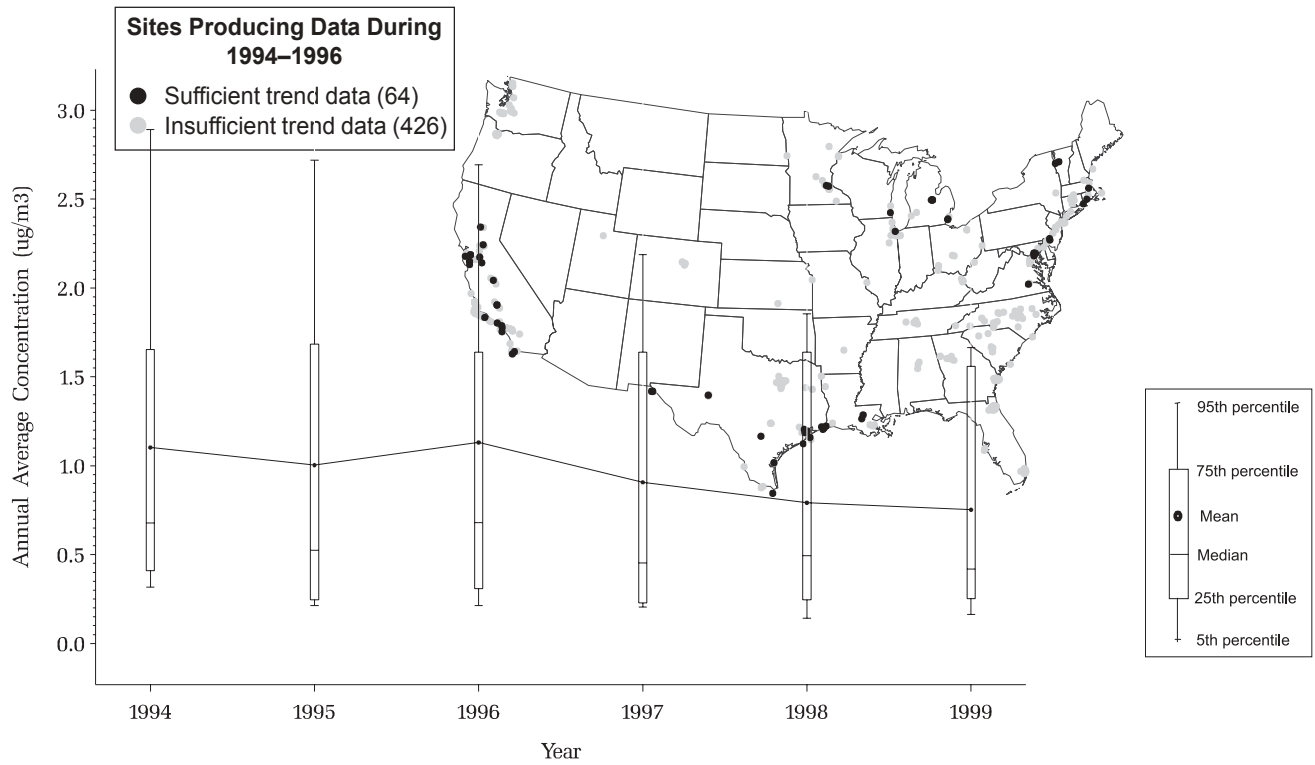
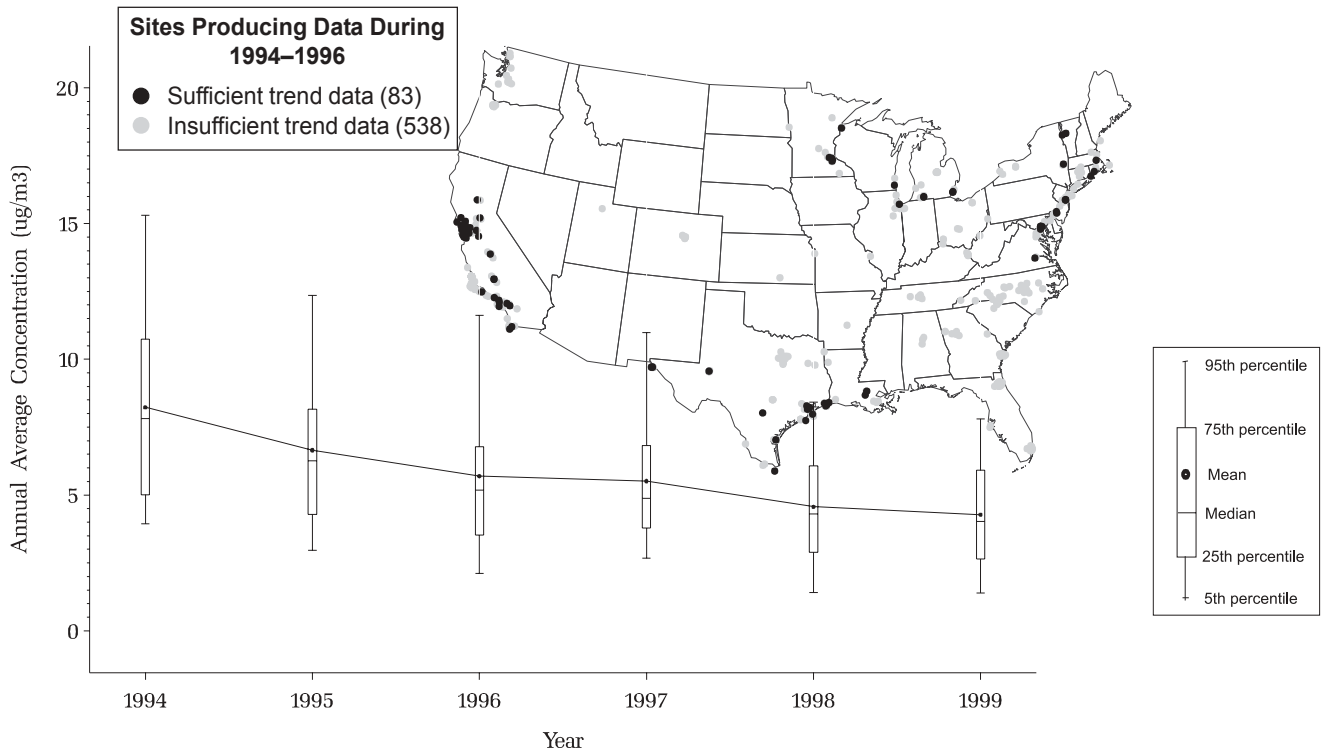


Figure 5-8f. National trend in annual average toluene concentrations in metropolitan areas, 1994–1999.



sources for many of the air toxics. For these pollutants, a national composite trend may not be meaningful at this time. Although the general direction of change is down for most HAPs on the urban list, several states reveal significant 6-year increases at a few locations. The HAPs and some of their influencing sources are: 1,3-butadiene (mobile sources); chromium (power plants, electroplating); lead (smelters and aircraft); and semi-volatile particulates (various combustion sources). This list also includes carbon tetrachloride, chloroform, and trichloroethylene whose ambient concentrations are estimated to have relatively high background contributions. Background concentrations are contributions to ambient air toxics concentrations resulting from natural sources, persistence in the environment of past years' emissions and long-range transport from distant source. To illustrate a few of the HAPs without consistent trends among the current set of trend sites, boxplots for 1994–1999 are presented for 1,3-butadiene, styrene, and perchloroethylene. The national trend lines for these HAPs show more year-to-year variability, but still appear to show 6-year air quality improvements.

To illustrate the behavior of selected HAPs in a particular region of the country, trends of monitoring sites in California are presented Figure 5-9. The state of California has the largest and longest running air toxics monitoring network. They have over 30 sites with a 10-year history for several VOCs and almost as many for several trace metals. These data allow us to take a look at air toxics trends over a longer period of time. Among the HAPs discussed in this section, notable improvements are

seen for benzene, 1,3-butadiene, lead, perchloroethylene, styrene and toluene. The impressive air quality improvement for urban benzene in California is shown in Figure 5-9a. This figure illustrates the large decrease in ambient concentrations which occurred during the early 1990s. Annual average concentrations declined 64 and 35 percent over the 1990–1999 and 1990–1999 periods. Ambient concentrations of perchloroethylene associated with dry cleaners is down 60 and 39 percent respectively (Figure 5-9d). Toluene associated with mobile sources also showed consistent 10-year declines which averaged 53 percent across the state (Figure 5-9f). Besides benzene, another HAP which predominantly comes from mobile sources is 1,3-butadiene. Although site-specific trends for this pollutant were mixed, the composite trend in Figure 5-9b shows an overall 40 percent and 28 percent decline in ambient concentrations for the 10- and 6-year periods.

As was the case nationally, the reductions in ambient concentrations of perchloroethylene are due to better controls on the use of solvents. The California improvements in benzene, 1,3-butadiene and toluene are primarily attributed to the reformulation of gasoline and new-car improvements in terms of emission controls. (For more information about trends in these emissions, see the ozone section in Chapter 2.) For lead in TSP, annual average concentrations in California declined 46 percent over the 10 years, but appear to have leveled off over the most recent years. For additional detail on the derivation of Figures 5-8a to 5-9f, see Appendix B: Methodology.

Ambient air toxics data in rural areas are much more limited, but the

results in Table 5-3 also indicate widespread air quality improvement for many monitored HAPs. Significant downward trends are noted among the few rural sites for benzene and several other VOCs. Lead concentrations in rural areas are also down.

While these data are useful to describe general trends and geographic variations in annual average concentrations, they only represent a selected group of monitoring sites. They do not necessarily highlight the range of concentrations or locate air toxics problem areas that exist nationwide. For example, a recent air toxics study conducted in the Los Angeles area has shown that higher concentrations of air toxics generally occur near their emission sources. In particular, concentrations of compounds that are emitted primarily from stationary and area sources tended to be highest within a few kilometers from the source location. More ubiquitous mobile source related compounds such as benzene and 1,3-butadiene were shown to be generally high throughout the South Coast Air Basin. However, the highest concentrations were estimated by air quality models to occur along freeway corridors and junctions. In addition, high levels of mobile source related compounds were estimated near major mobile source activities such as airports and other areas with major industrial activities. Also, annual averages may tend to average out peaks in the monitoring data. The study showed that there were strong seasonal variations to the levels of toxic air contaminants, primarily with those pollutants associated with mobile sources. For example, benzene and butadiene both had seasonal peaks in the late fall and winter

Figure 5-9a. Trend in annual average benzene concentrations for metropolitan sites in California, 1994–1999.

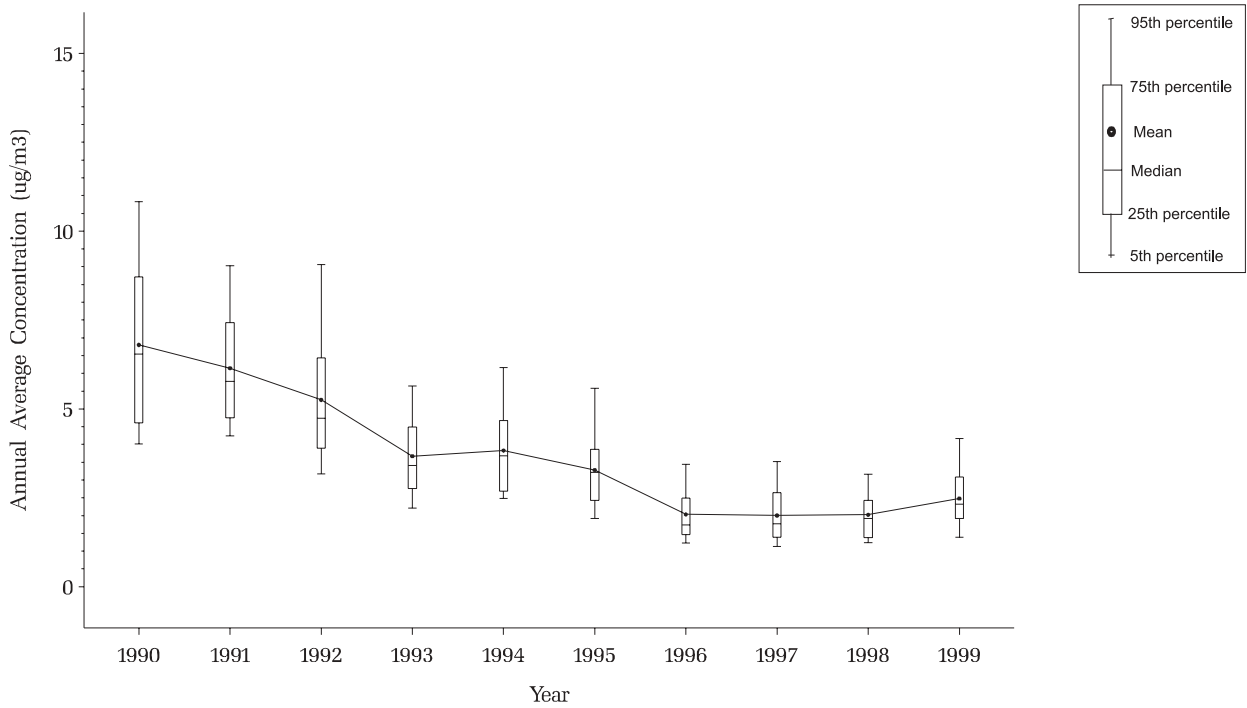


Figure 5-9b. Trend in annual average 1,3-butadiene concentrations for metropolitan sites in California, 1994–1999.

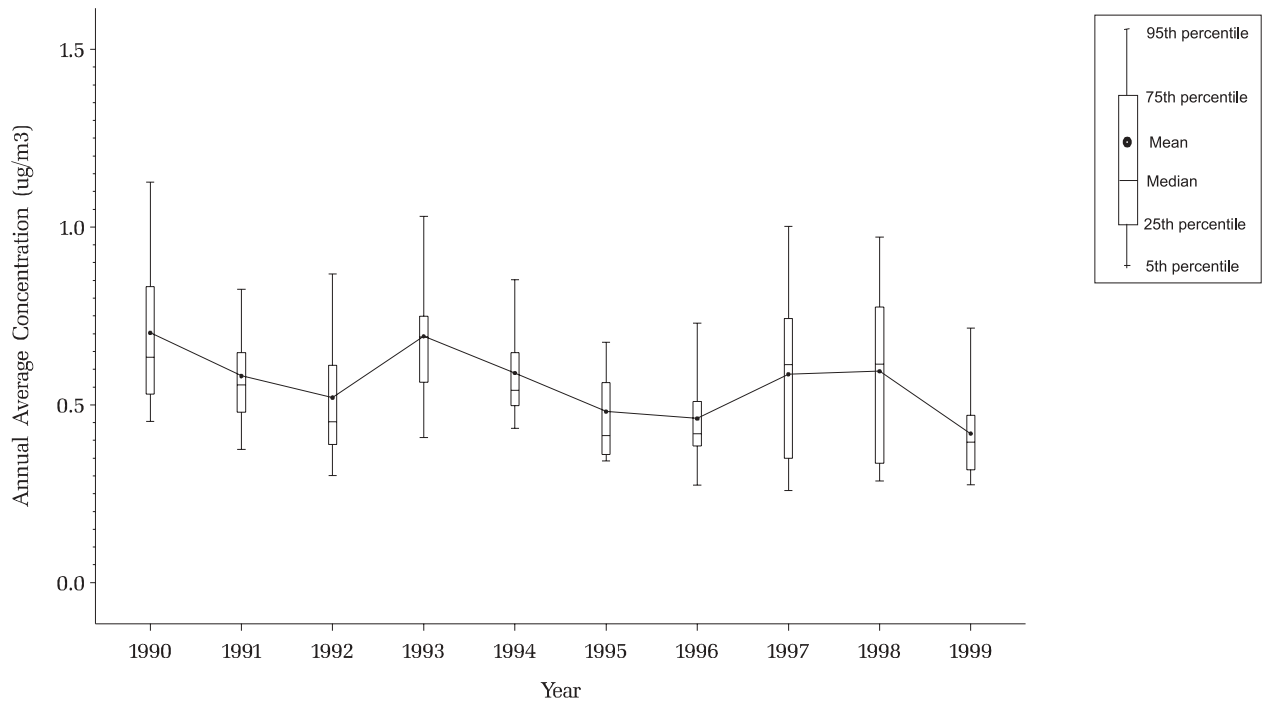


Figure 5-9c. Trend in annual average total suspended lead concentrations for metropolitan sites in California, 1990–1999.

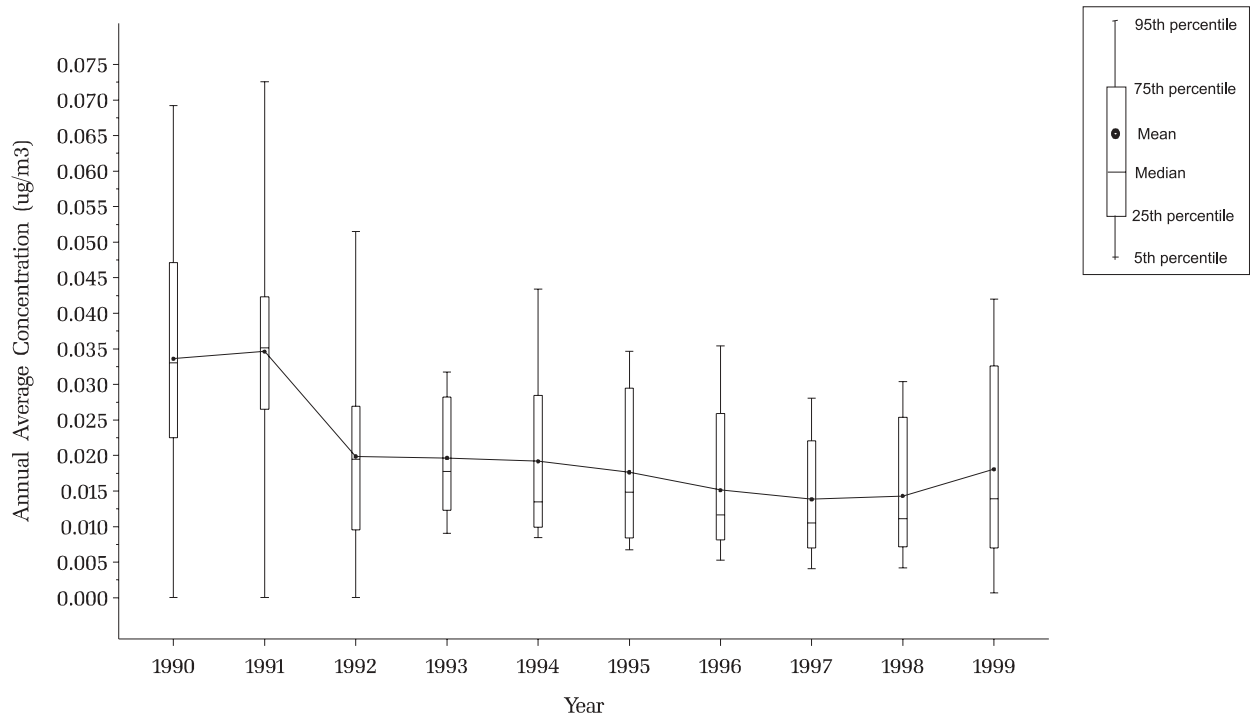


Figure 5-9d. Trend in annual average perchloroethylene concentrations for metropolitan sites in California, 1990–1999.

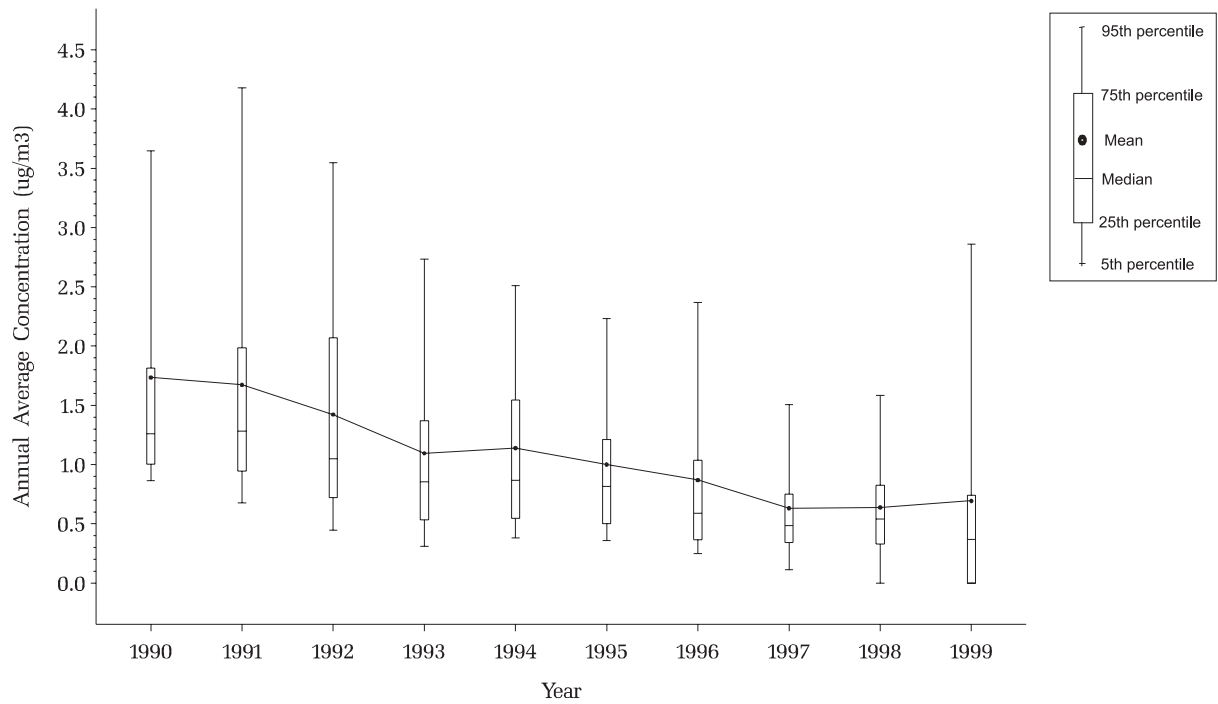


Figure 5-9e. Trend in annual average styrene concentrations for metropolitan sites in California, 1990–1999.

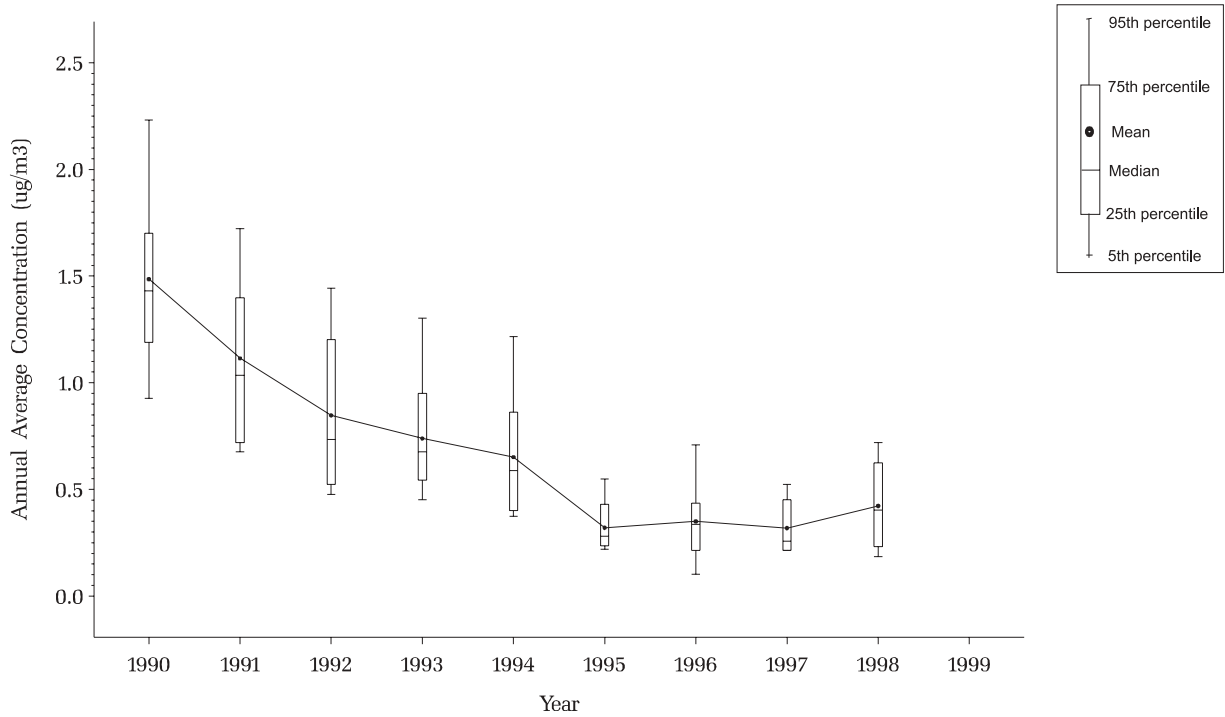


Figure 5-9f. Trend in annual average toluene concentrations for metropolitan sites in California, 1990–1999.

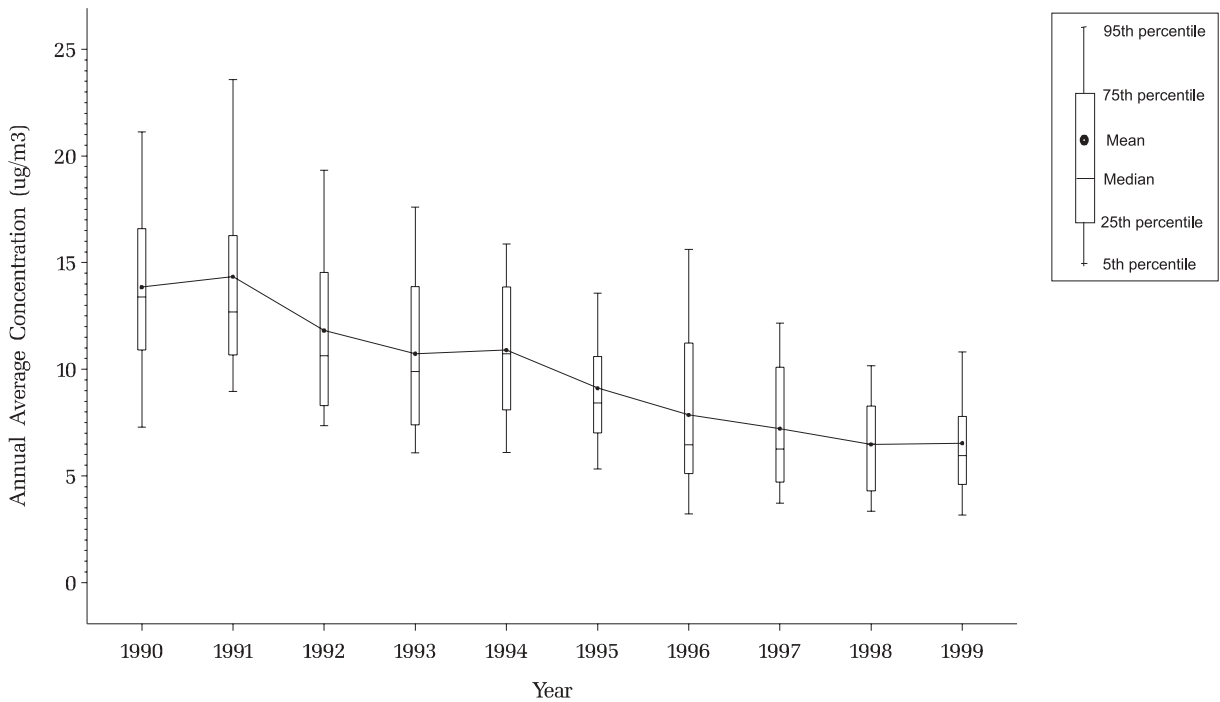


Table 5-3. National Summary of Ambient HAP Concentration Trends in Rural Areas, 1994–1999

Pollutant Name	Number of Rural Trend Sites by HAP					
	Total	Significant* UP Trend	Non-Significant UP Trend	No Trend	Non-Significant DOWN Trend	Significant* DOWN Trend
Benzene	6				6	
1,3-Butadiene	4		1		2	1
Carbon tetrachloride	2		2			
Chloroform	4		1		2	1
1,2-Dichloropropane	3			2	1	
Ethylene dichloride	3				2	1
Methylene chloride	4		1		3	
1,1,2,2-Tetrachloroethane	1				1	
Perchloroethylene	5		1		1	3
Trichloroethylene	5			1	3	1
Vinyl chloride	4		1	2	1	
Arsenic (coarse)	2		1	1		
Arsenic (fine)	59	2	18	1	36	2
Arsenic (PM ₁₀)	6		1	3	1	1
Arsenic (TSP)	5			1	2	2
Beryllium (PM ₁₀)	2		1	1		
Beryllium (TSP)	3			3		
Cadmium (PM ₁₀)	2			1	1	
Cadmium (TSP)	7			4	1	2
Chromium (coarse)	2		1		1	
Chromium (fine) *	59	32	22	1	4	
Chromium (PM ₁₀)	6	1	2		3	
Chromium (TSP)	8		3	1	4	
Chromium VI	1				1	
Lead (coarse)	2			1	1	
Lead (fine)	59	3	32		20	4
Lead (PM ₁₀)	8	1	2	2	2	1
Lead (TSP)	33		5		16	12
Manganese (coarse)	2		1		1	
Manganese (fine)	59	3	22		32	2
Manganese (PM ₁₀)	6		2		3	1
Manganese (TSP)	7		2		5	
Mercury (fine)	2			1	1	
Mercury (PM ₁₀)	4		2	1	1	
Mercury (TSP)	1		1			
Nickel (coarse)	2		1		1	
Nickel (fine)	59		12	1	32	14
Nickel (PM ₁₀)	6		1	1	3	1
Nickel (TSP)	8			1	6	1
Acetaldehyde	3		2		1	
Formaldehyde	4		1		3	
Acrolein	1				1	
Styrene	6		2		3	1
Toluene	7		3		3	1

*Statistically significant at the 10-percent level (See Appendix B: Methodology, Air Toxics Methodology section).

** The apparent up trends in fine chromium concentrations may be an artifact of the detection limits for these measurements.

months; their lowest levels were observed during the spring and summer months.

National Atmospheric Deposition Program/Mercury Deposition Network

The purpose of the National Atmospheric Deposition Program (NADP) is to address the problem of atmospheric deposition and its effects on agricultural crops, forests, rangelands, surface waters, and other natural resources. NADP began in 1978 as a cooperative program between federal and state agencies, universities, electrical utilities, and other industries to measure atmospheric deposition and determine geographical patterns and trends in wet deposition of sulfate, nitrate, hydrogen ion, ammonium, chloride, calcium, magnesium, and potassium. Wet deposition is atmospheric deposition that occurs when rain, snow, or fog carry gases and particles to the earth's surface.

The Mercury Deposition Network (MDN), which is a component of the NADP, measures mercury levels in wet deposition at over 40 NADP sites located in 16 states and two Canadian provinces. MDN is investigating the importance of atmospheric deposition as a source of mercury in lakes and streams. These MDN data enable researchers to compile a national database of weekly precipitation concentrations to determine seasonal and annual fluxes of mercury in precipitation falling on lakes, wetlands, streams, forested watersheds, and other sensitive ecosystems. As a result, state and federal air regulators can monitor progress in reducing mercury deposition and amend policy decisions accordingly. There are plans to expand the network in the near future, pending availability

of new funds. Additional information about the network is available on the Internet at <http://nadp.sws.uiuc.edu/mdn/>.

Data from 1998 indicate that the volume-weighted mean concentration of total mercury in precipitation from 30 sites ranged from 3.8–23.0 ng/L and annual deposition of mercury ranged from 4.0–20.3 µg/m². Most of the monitors are in the Great Lake states and eastern United States. While high concentrations in precipitation are found in many regions, the highest estimated deposition is in the southern states. In the eastern United States, average summer mercury concentrations are approximately twice the winter concentrations and average summer deposition values are three times winter values. This can be explained by higher concentrations of mercury in the rain and higher rainfall amounts during the summer.¹⁰

Integrated Atmospheric Deposition Network

The Integrated Atmospheric Deposition Network (IADN) was established in 1990 by the United States and Canada for conducting air and precipitation monitoring in the Great Lakes Basin. IADN collects data that can be useful in assessing the relative importance of atmospheric deposition to pollutant loadings in the Great Lakes. The first implementation plan, signed in 1990, committed the United States and Canada to work cooperatively towards the initiation of IADN. IADN measures concentrations of target chemicals in rain and snow (wet deposition), airborne particles (dry deposition), and airborne organic vapors.¹¹ PAHs, PCBs, and organochlorine compounds (which are all Semivolatile Organic Compounds, or SVOCs) are measured in air and pre-

cipitation samples in the United States and Canada. SVOCs are measured in both the gaseous and particulate phases in air. Canada also measures trace metals in air and precipitation, as well as PM_{2.5} (particles less than 2.5 microns in diameter) in air.

Under IADN, trends in pollutant concentrations in air and precipitation are assessed and loading estimates of atmospheric deposition and volatilization of pollutants are made every two years. The IADN network currently consists of one master station per Great Lake and 14 satellite stations. Stations are located in remote areas and do not assess urban sources of pollution.

General conclusions based on IADN data include the following:

- Levels in air and precipitation appear stable for current-use pesticides such as endosulphan, but levels for most other pesticides, PCBs, and lead are decreasing.
- Gas absorption appears to be the dominant deposition process for delivering SVOCs, including PCBs and PAHs, to lake surfaces, while wet and dry deposition dominate for the trace elements and higher molecular weight PAHs.
- For some IADN substances, like dieldrin and PCBs, the surface waters are behaving like a source since the amount that is volatilizing from the water is greater than the amount being deposited to the water.
- The lakes are sensitive to the atmospheric concentration of IADN chemicals, and this points out the fragility of these resources given that long-range transport from other regions may be a significant source of toxic pollutants.

- Air trajectory analyses indicate that many SVOCs are potentially originating from outside the Great Lakes basin, whereas trace metals and PAHs may be associated with local sources.

The Second Implementation Plan for IADN (IP2), signed in 1998, outlines goals and plans for IADN for the period 1998–2004. Under this Second Implementation Plan, the IADN will continue surveillance and monitoring activities, related research, and provision of information for intergovernmental commitments and agreements. Additional work to be completed under the Second Implementation Plan is the development of a database for all U.S. and Canadian data. Potential modifications will be discussed in relation to the placement of satellite stations to assess urban inputs and air-water gas exchange, criteria for changes to the IADN chemical list, coordination with other research activities, quality assurance and control of IADN operations, and communication of IADN results.¹²

References

1. This list originally included 189 chemicals. The CAA allows EPA to modify this list if new scientific information becomes available that indicates a change should be made. Using this authority, the Agency modified the list to remove caprolactam in 1996, reducing the list to 188 pollutants (*Hazardous Air Pollutant List; Modification*, 61 FR 30816, June 18, 1996).
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3. *Federal Register*, 65 FR 79827.
4. Giesy, J.P., Ludwig, J.P., and Tillitt, D.E. 1994. Deformities in birds of the Great Lakes region: assigning causality. *Environ. Sci. Technol.* 28,128A-136A.
5. Cook, P.M., Zabel, E.W., and Peterson, R.E. 1997. The TCDD toxicity equivalence approach for characterizing risks for early life stage mortality in trout. In: *Chemically Induced Alterations in the Functional Development and Reproduction of Fishes*, pp. 9–27. (Rolland, R., Gilbertson, M., and Peterson R., Eds.). SETAC Press, Pensacola, FL.
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7. "National Air Toxics Program: The Integrated Urban Strategy," *Federal Register*, 64 FR 38705, Washington, D.C., July 19, 1999. Available on the Internet at: <http://www.epa.gov/ttn/atw/urban/urbanpg.html>
8. "1997 Urban Air Toxics Monitoring Program (UTAMP)," EPA-454/R-99-036. RTP, NC 27711, January 1999. Available on the Internet at <http://www.epa.gov/ttn/amtic/airtxfil.html>.
9. "Air Toxics Monitoring Concept Paper," U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, RTP, NC, 27711. February 29, 2000. Peer Review Draft. Available on the Internet at: <http://www.epa.gov/ttn/amtic/airtxfil.html>.
10. Sweet, C.W., E. Prestbo, B. Brunette. 1999. Atmospheric wet deposition of mercury in North America. Proceedings of the 92nd Annual Meeting of the Air and Waste Management Association. June 21–23, 1999, St. Louis, MO.
11. The target chemicals include PCBs, pesticides, PAHs and metals. The compounds included as "target chemicals" were selected based on the following criteria: presence on List 1 of Annex 1 of the Great Lakes Water Quality Agreement (substances believed to be toxic and present in the Great Lakes); established or perceived water quality problem; presence on the International Joint Commission's Water Quality Board's list of criteria pollutants; evidence of presence in the atmosphere and an important deposition pathway; and feasibility of measurement in a routine monitoring network.
12. U.S./Canada IADN Scientific Steering Committee. 1998. Technical summary of progress under the integrated atmospheric depositions program 1990–1996.

