

Survey of New Findings in Scientific Literature Related to Atmospheric Deposition to the Great Waters:

Polycyclic Aromatic Hydrocarbons (PAH)



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Survey of New Findings in Scientific Literature Related to Atmospheric Deposition to the Great Waters: Polycyclic Aromatic Hydrocarbons

1.0 Introduction and Highlights

Atmospheric deposition of pollutants, including polycyclic aromatic hydrocarbons (PAH), is recognized as a significant contributor in many locations to water quality problems, including toxic contamination of the fish and shellfish living in the waters. PAH are a subset of a set of compounds known as polycyclic organic matter (POM), which are organic compounds primarily formed from the incomplete combustion of organic materials, such as coal and wood. Several PAH compounds have been classified as probable human carcinogens (ATSDR 1995).

The U.S. Environmental Protection Agency (U.S. EPA) has been directed by the Clean Air Act to consider the contribution of atmospheric deposition to pollution in the “Great Waters,” which comprise the Great Lakes, Lake Champlain, Chesapeake Bay, and many of the estuaries of the coastal United States.¹ POM are included in the group of pollutants of concern for the Great Waters.² Background information on the sources, deposition, and environmental concentrations of the pollutants of concern is summarized in detail in a series of reports, the most recent of which is

HIGHLIGHTS Polycyclic Aromatic Hydrocarbons

- **Long-term Temporal Trends.** No consistent long-term trends in PAH deposition are reported for the Great Lakes region. In Chicago, however, PAH concentrations in precipitation and ambient air are decreasing. In mollusks, a decreasing trend in the national median concentration is becoming apparent, although at most of the individual sites, no trends are evident. In lake sediments in several urban areas, PAH concentrations have been increasing over the past 20 to 40 years.
- **Seasonal Trends.** Studies in multiple areas found higher PAH air concentrations and deposition during winter. This trend was attributed in part to domestic heating. No seasonal trends were found in PAH concentrations in surface water and zooplankton in southern Lake Michigan, near Chicago, indicating relatively constant PAH deposition.
- **Spatial Trends.** The new studies echo the findings in the Third Report to Congress that PAH air concentrations and deposition are greater in urban and industrial areas than in rural areas. There are several newly-studied geographic areas, including Tampa Bay, San Francisco Bay, and the New Jersey coast. A variety of statistical modeling techniques were used to examine the source regions that influence the Great Lakes. Two studies demonstrated passive sampling techniques to look at spatial trends.
- **Emissions and Source Profiles.** The dominant sources of emissions of 15 PAH compounds nationally are related to residential, wood-fired boilers, and open burning. The distribution of sources varies by region, with significant sources including vehicular traffic, coal combustion, coke ovens, jet exhaust, and natural gas combustion.

¹ The estuaries that are part of the Great Waters are those that part of the National Estuary Program (NEP) administered by EPA or the National Estuarine Research Reserves (NERR) Program administered by the National Oceanic and Atmospheric Administration (NOAA).

² The Great Waters pollutants of concern include POM, mercury, cadmium and lead (and their compounds), several banned or restricted pesticides, polychlorinated biphenyls (PCB), nitrogen compounds, tetrachlorodibenzo-p-dioxin, and tetrachlorodibenzofuran. More specific information is at <http://www.epa.gov/oar/oaqps/gr8water>.

“Deposition of Air Pollutants to the Great Waters Third Report to Congress” (U.S. EPA 2000), hereafter referred to as “the Third Report to Congress.”

U.S. EPA is no longer required to submit reports to Congress on deposition of air pollutants to the Great Waters. However, much new information related to environmental concentrations, deposition trends, and sources of PAH in the Great Waters has been published since the Third Report to Congress, and is compiled here. The recent research also is compared to findings described in the Third Report to Congress. Much of the information in this survey report relates to recent findings in temporal and spatial trends in both the Great Lakes and Chesapeake Bay regions. Other studies address the creation of new source profiles in different areas. For example, studies for Galveston Bay and the city of Miami have recently been completed, which detail sources of PAH to these regions. Highlights of long-term temporal and seasonal trends, findings and spatial trends in various locations, and sources and emissions from the newly published scientific literature are

presented in the textbox in this section. References are provided at the end of this summary.

DEFINITIONS OF COMMON TERMS

Direct deposition: The process of deposition of air pollution directly into a body of water (e.g., a large body of water like an estuary or large lake). The amount of pollution reaching the water in this way is called the direct load from atmospheric deposition.

Indirect deposition: The process of deposition of air pollution to the rest of the watershed (both the land and the water). Once pollutants are deposited in the watershed, some portion is transported through runoff, rivers, streams, and ground water to the waterbody of concern. The portion that reaches the waterbody by passing through the watershed is called the indirect load from atmospheric deposition.

Wet deposition: Pollutants deposited in rain, snow, clouds, or fog. Acid rain, which has been recognized as a problem in Europe, eastern Canada, Asia, and areas of the United States, is an example of wet deposition of sulfur and nitrogen compounds.

Dry deposition: Pollutants deposited during periods of no precipitation. This is a complicated process that happens in different ways depending on the size and chemical nature of the particle or gas being deposited and the “stickiness” of the surface. Dry deposition of particles can be thought of as similar to dust collecting on a table.

Source: U.S. EPA 2001

2.0 Temporal Trends

One area of new research since the Third Report to Congress concerns temporal trends of PAH associated with deposition and concentrations in environmental media. Findings relating to long-term temporal and seasonal PAH trends are discussed below.

2.1 Long-term Trends

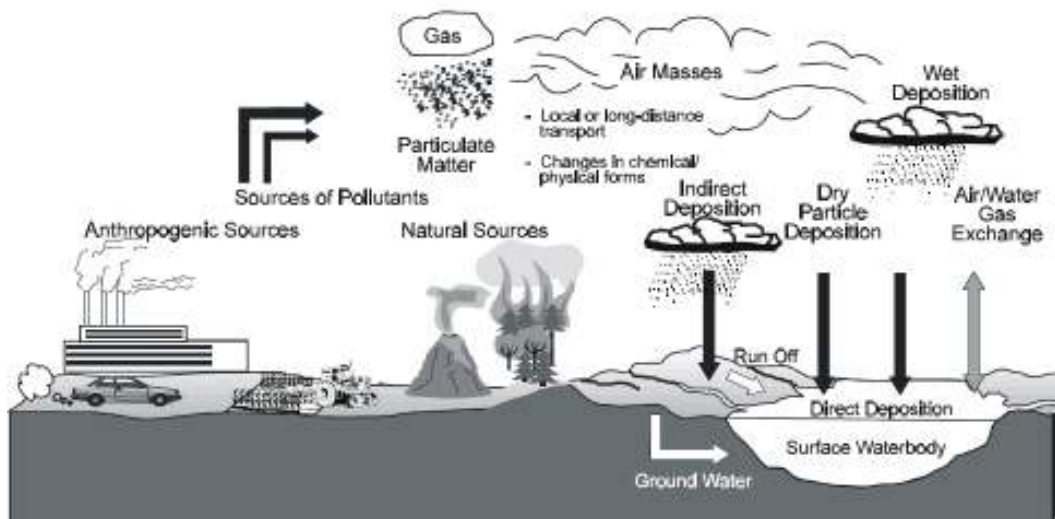
2.1.1 Ambient Air and Deposition

All recent literature regarding long-term temporal trends in ambient air and deposition for PAH pertains to the Great Lakes region. The trends analyses are based on data measured at sites in the Integrated Atmospheric Deposition Network (IADN). The IADN consists of monitoring stations located on each of the Great Lakes since 1990 and at several satellite locations in the surrounding areas in both the United States and Canada. Gas and particle-phase air samples and precipitation data are collected throughout the year for several pollutants, including 13 PAH compounds (Buehler and Hites 2002). This new data demonstrates that

heavier PAH compounds (e.g., benzo(a)pyrene) enter the lakes from the atmosphere through wet deposition and dry deposition of particles, whereas the lighter PAH compounds (e.g., phenanthrene) enter through gas absorption (EC and U.S. EPA 2004).

There are consistent long-term trends neither in PAH measurements in ambient air nor in PAH deposition loading throughout the Great Lakes region. This lack of consistent trends echoes the Third Report to Congress, and is congruous with the fact that PAH compounds continue to be emitted to the atmosphere, largely as the byproduct of incomplete combustion (Buehler and Hites 2002, EC and U.S. EPA 2004). Once in the atmosphere, PAH are transferred continuously between air, water, and soil by natural chemical and physical processes such as weathering, runoff, precipitation, dry deposition of dust, and stream/river flow (see Figure 1).

Figure 1. Atmospheric Release, Transport, and Deposition Processes



Source: U.S. EPA 2000

Several studies reported trends analyses of IADN data for total PAH concentrations in precipitation, the particle phase in ambient air, and the gas phase in ambient air as shown on Table 1. Results are reported for three U.S. sites: Eagle Harbor site near Lake Superior, the Sleeping Bear Dunes site near Lake Michigan, and the Sturgeon Point site near Lake Erie. These sites are situated to measure regional background levels of pollutants.

Table 1. Long-Term Atmospheric Trends in Total PAH Concentrations in the Great Lakes Region between 1991 and 2003

PAH Measurement	Years	Lake Superior	Lake Michigan	Lake Erie	Source
Precipitation	1991-1997	↔	↔	↔	Simcik et al. 2000
	1997-2003	↔	↔	↔	Sun et al. 2006a
Ambient particle phase	1991-1997	↔	↓	↔	Cortes et al. 2000 ^a
	1996-2003	↔	↔	↓	Sun et al. 2006b
Ambient gas phase	1991-1997	↓	↓	↓	Cortes et al. 2000 ^a
	1996-2003	↓	↓	↓	Sun et al. 2006b

↓ = decreasing trend

↔ = no trend

^aTrends for Lake Superior (Eagle Harbor) and Lake Erie (Sturgeon Point) are based on the majority of reported compounds, not total PAH.

The studies of the gas-phase PAH estimated half-lives³ for these compounds. Based on the data through 1997, the half lives for gas-phase PAH compounds ranged from two to nine years. The more recent data showed a slower rate of decline. Based on the data through 2000, the half-lives ranged from four to 18 years, and over the most recent period (1996-2003), half lives were greater than 15 years. Sun et al. (2006b) note that a slower rate of decline in recent years is reasonable if atmospheric PAH concentrations are approaching a nonzero steady state.

At the IADN site in Chicago, concentrations of all the PAH compounds measured in precipitation decreased significantly between 1997 and 2003. Similarly, PAH compounds in the gas- and particle-phases in the ambient air also decreased over this period; significant trends were found for total PAH, as well as for most of the individual PAH compounds. Efforts to improve the air quality in Chicago, such as reductions in pollution from motor vehicles, may explain the decreasing trends (Sun et al. 2006a, Sun et al. 2006b).

The deposition information measured by IADN can be supplemented by measurements of PAH compounds in the ambient air in the Ontario Great Lakes Basin by Environment Canada. These sites include rural areas and three classes of urban areas: residential, commercial, and industrial. For the period from 1996 to 2002, total PAH concentrations in air decreased at urban commercial sites near Lake Ontario in Toronto and Hamilton, and at an urban residential site in Windsor near Lake Erie. The authors did not hypothesize as to why these trends exist. At rural sites, there were no significant trends (Environment Canada 2004).

2.1.2 Biota and Sediments

The National Oceanic and Atmospheric Administration (NOAA) National Status and Trends (NS&T) Mussel Watch Program has monitored concentrations of trace chemicals in mussels and

³ The half-life is the number of years for the concentration to decrease by a factor of two, and therefore is an indication of the rate of decline.

oysters in the coastal U.S. since 1986. The sites were selected to be representative of large areas, rather than smaller-scale areas that would be influenced directly by particular local sources of contaminants. Since mollusks concentrate chemicals from their surrounding waters in their tissues, they provide an integrated measurement of contamination over time. For PAH compounds, the Mussel Watch Program groups low-molecular-weight (LMW) PAH (two- and three-ring compounds) separately from high-molecular-weight (HMW) PAH (four- and five-ring compounds), because the LMW compounds are relatively more concentrated in oil than in combustion products (O'Connor 2002).

BIOAVAILABILITY OF PAH IN BENTHIC ORGANISMS

What is bioavailability? Bioavailability is a measure of the extent and rate that a substance is absorbed into an organism and how much of it becomes available within various physiological systems. Bioavailability is an important component that should be taken into account when chemical exposures are assessed.

Are PAH compounds readily bioavailable to benthic organisms? The total concentration of PAH in sediment is not the only factor involved in how much of the PAH can enter a benthic organism. In sediment, several factors influence bioavailability. For example, the organic carbon content is important because carbon soot content reduces the bioavailability, amount, and quality of organic material present in sediment (Burgess et al. 2003). Rust et al. (2004) provides some examples of particle-derived PAH and their level of bioavailability to benthic organisms; low-bioavailable PAH compounds include those in coal dust, those with medium bioavailability are associated with diesel soot and tire rubber powder, and PAH with high bioavailability include those associated with fuel oil, creosote, and crude oil.

Are all benthic organisms equally susceptible? PAH bioavailability depends not only on environmental conditions and the type of PAH but also on organism properties. In organisms, some of the properties influencing tissue PAH concentrations include the lipid concentration of an organism (PAH are attracted to fat), their lifestyle and feeding habits, and gut chemistry. For example, deposit feeders have greater ingestion rates than suspension feeders and therefore tend to accumulate more PAH. And the conditions in the gut of some benthic organisms make it more likely that PAH compounds will desorb from the sediments, increasing bioavailability (Burgess et al. 2003). All of these factors should be considered when determining potential effects.

Data from the Mussel Watch Program from 1986-1996 were discussed in the Third Report to Congress, and showed no trends for PAH. By contrast, a new national-scale analysis of Mussel Watch Program data showed that the median concentration in mollusks decreased between 1986 and 2002 for both total LMW and total HMW PAH (U.S. EPA 2004).

At 206 individual Mussel Watch sites, trends in total PAH in mollusk tissue vary. Between 1986 and 1999, 162 sites showed no trends, 26 sites showed decreasing trends, and 18 sites showed increasing trends. This analysis also examined specifically the trends at National Estuarine Research Reserves (NERR) designated by NOAA. Three of the NERR had a Mussel Watch site showing an increasing trend for total PAH: North Inlet – Winyah Bay, SC; Rookery Bay, FL; and Tijuana River, CA. One NERR site (Wells, ME) had a Mussel Watch site showing a decreasing trend. Some of the increasing trends may be explained by changes in the analytical methods for PAH over this period. Early in the program, the laboratory analytical method was less sensitive, so values now recorded as low PAH concentrations were originally recorded as zero (NOAA 2002). Higher values of total HMW PAH were correlated with sites in close proximity to urban areas,

indicating that levels are likely due to human activity. Levels of PAH in urban areas are in a range that may have detrimental effects to the mollusks by causing alterations in lysosomes of digestive cells (O'Connor 2002).

Long-term trends of PAH concentrations in lake sediments show recent increases related to urban pollution. While the 38 lakes studied are not Great Waters, they do indicate possible effects of urbanization on a watershed. The lakes represent a diverse group of geographic regions and ecoregions, and were categorized by land use: densely urban, light urban, and reference (less than 1.5 percent urban land use). Between 1970 and 2001, concentrations of total PAH in sediment increased at 42 percent, decreased at five percent, and showed no trend at 53 percent of all the lakes. None of the reference lakes showed a trend in total PAH concentrations in this period. To evaluate the potential impact to the aquatic biota, the researchers compared the mean concentrations in the sediment in the decades from 1965 to 1975 and the 1990s to a consensus-derived probable effect concentration (PEC). For PAH, the frequency of exceedances of the PEC approximately doubled in the 1990s compared to the decade from 1965 to 1975. The highest frequency of exceedances occurred in densely urban lakes (Van Metre and Mahler 2005).

An analysis of trends for individual PAH compounds in these 38 lakes found that more of the lakes had increasing trends of compounds with higher molecular weight than those with lower molecular weight. As noted above, the higher molecular weight compounds are more typical of combustion by-products (Van Metre and Mahler 2005). In an earlier analysis for 10 urban lakes (a subset of the 38), the researchers found that increases in PAH concentration in the sediment cores outpaced the increase in urbanization since the mid-1970s at nine of the sites. They also found that the increases in PAH concentration followed closely with increases in automobile use, even in urban areas where there was a relatively minor increase in the degree of urbanization over the same time period. The authors noted that there are several sources of vehicle-related PAH in addition to exhaust, including asphalt wear, tire wear, and leaks and spills of engine oil (Van Metre et al. 2000). Other possible sources of the PAH are being considered as well, including parking lot sealants, many of which contain coal tar (Van Metre and Mahler 2005).

2.2 Seasonal Trends

Several studies found seasonal differences in PAH concentrations in air and deposition. Higher ambient air concentrations or dry deposition during late fall or winter have been observed in coastal New Jersey (Gigliotti et al. 2000), San Francisco Bay (Tsai et al. 2002), the Great Lakes (EC and U.S. EPA 2004, Sun et al. 2006b) and the New England coast (Golomb et al. 2001). Similar trends were found in wet deposition in the Great Lakes (Sun et al. 2006a) and New England (Golomb et al. 2001). The researchers believe these observations are likely due to fuel combustion for domestic heating, based on which PAH compounds are most prevalent. In addition, semi-volatile PAH compounds have a tendency to be in the particulate phase in colder temperatures.

In New Jersey, Gigliotti et al. (2000) found that during the winter months, the contribution to total PAH by methylated-phenanthrenes was greater than the contributions of phenanthrenes. During the summer months, the opposite was true, indicating that dominant sources differ between the two seasons. These results are based on the first year of sampling (October 1997 through October 1998) at the New Jersey Atmospheric Deposition Network (NJDAN) suburban site near New Brunswick and a coastal site in Sandy Hook. Based on an analysis of all the NJDAN sites across the state over

the approximately four years of sampling, there were no clear seasonal trends in gas-phase concentrations of PAH. However, increased particle-phase concentrations in winter were found to be driven by higher emissions from fossil fuel combustion at most of the NJDAN sites (Reinfelder et al. 2004).

In studies of the ambient air near Galveston Bay, Texas, higher concentrations were found in the winter and summer compared to the spring and fall, although these were not strong seasonal trends. Furthermore, no seasonal trends were found in a similar study at Corpus Christi Bay in Texas. The researchers speculated that the lack of seasonal trends in coastal Texas may be due to the temperate climate, and smaller seasonal variations in emissions sources than in more northern areas where seasonal trends are apparent (Park et al. 2001, Park et al. 2002).

Another study examined PAH concentrations in near-shore surface water and in zooplankton in southern Lake Michigan, near the greater Chicago area. The study was conducted in 1994 and 1995. Concentrations of PAH in surface water measured during the months of January, May, and July did not vary significantly according to season or wind direction. Based on these data, the study authors concluded that PAH levels in surface water remained relatively constant. The researchers also found that lipid-normalized PAH concentrations in zooplankton did not vary with season; concentrations in January and July were nearly identical. These findings suggest relatively constant deposition of PAH from urban sources throughout the year (Offenberg and Baker 2000).

3.0 Spatial Trends and Deposition Studies in Various Geographic Areas

A second major area of newly published PAH research addresses ambient air and deposition studies and analyses in several geographic areas, including the Great Lakes and multiple coastal locations. In the Third Report to Congress, the spatial trends in PAH concentrations in air and deposition between urban and rural locations were discussed at length. The newly published research details the urban influence even further. In general, studies show that PAH are largely urban pollutants that can travel tens of kilometers (km) through the atmosphere.

One study examined monitoring studies throughout the world, and statistically compared PAH air concentrations to the human population density within 25 km of the PAH measurements. As expected, the authors found a strong positive correlation between PAH concentrations and population density. In addition, spatial differences were seen. Sites located within 25 km of a coast had lower PAH concentrations than predicted by the regression analysis, due to dilution from cleaner ocean air, whereas sites near industrial areas had higher PAH concentrations than predicted (Hafner et al. 2005). The studies below are specific to Great Waters.

3.1 Great Lakes

3.1.1 IADN Master Stations

In a study of IADN data, dry deposition levels of PAH increased from west to east based on spatially comparable data for Lakes Superior, Michigan, and Erie (EC and U.S. EPA 2000). This trend also held true for Lakes Huron and Ontario. Wet deposition fluxes also tended to increase from west to east when looking at spatially comparable data for the Great Lakes Basin. It is not

clear whether this spatial trend is due to increasing urbanization in the east or to other aspects of the monitoring site locations (EC and U.S. EPA 2002a).

Multiple analyses have been performed to examine how well the master stations of IADN on each of the Great Lakes represent background concentrations and to determine what source regions may be influencing them. Cortes et al. (2000) considered how well the sites at Eagle Harbor, Sleeping Bear Dunes, and Sturgeon Point represent background concentrations. The authors concluded that for Eagle Harbor and Sleeping Bear Dunes, the measured PAH gas-phase concentrations represent regional background. However, at the Sturgeon Point site near Lake Erie, PAH gas-phase concentrations decreased when wind blew cleaner air from over Lake Erie. Conversely, concentrations increased when winds blew from the Buffalo, New York area, which is about 20 km to the northeast. Buehler and Hites (2002) had similar findings for particle-phase PAH concentrations, which also were elevated at Sturgeon Point when winds were blowing from Buffalo. Therefore, both studies concluded that Sturgeon Point does not represent PAH concentrations for the entire Lake Erie region, as PAH concentrations are strongly influenced by northeasterly winds from the Buffalo region. Another study compared PAH concentrations in air at Eagle Harbor and Brule River (a rural site on Lake Superior). Although concentration ranges were within global background concentrations at both sites, PAH concentrations were higher at Brule River, indicating that pollution from Duluth may be influencing this site (Buehler et al. 2001).

Further analysis of the IADN data was done by Hafner and Hites (2003) using the Potential Source Contribution Function (PSCF) model, a probabilistic back-trajectory modeling technique. The researchers found similar urban influences as found in the studies described above for the influence of Buffalo on Sturgeon Point and Duluth on Brule River. Across the four PAH compounds studied, the authors found that the source regions became less distinct as the molecular weight of the compound increased. They hypothesized that all four PAH compounds originated from the same source regions, and then the higher molecular weight PAH degraded over distance, due to greater gas-phase atmospheric reactivity with increased PAH molecular weight.

Another analysis of IADN data from sites in the U.S. compared a variety of methods for including direction into classic regression equations used to model the atmospheric concentration of a semi-volatile organic pollutant, like PAH, at a given sampling site. The typical regression model includes a factor for air temperature at the site, and a factor representing the change in emission rate as a function of time. This analysis examined three ways to incorporate a factor to represent direction from which the pollutant came: local wind direction; average backward trajectory of air coming to the site; and a nonparametric air trajectory based on hypothesized source regions determined by the PSCF model. For PAH, the source direction factors in the regression analyses were very important in helping to explain the variability of the atmospheric concentrations at all of these IADN sites. For example, the local wind direction explained much of the variability at each of these sites, ranging from about 10 percent of the variability at Eagle Harbor and Chicago to about 46 percent at Brule River on Lake Superior. The analysis also complemented the findings of Cortes et al. (2000) that PAH sources are located to the east of the Sturgeon Point site on Lake Erie, probably in the greater Buffalo, NY area (Hafner and Hites 2005).

3.1.2 Urban Areas near the Great Lakes

Another group of studies specifically examined gradients of concentrations in and around urban areas in the Great Lakes region. These studies consistently found that PAH concentrations were higher around urban areas than in the suburban and rural areas.

Chicago has significantly higher concentrations of PAH compounds in the atmosphere and deposition than at the IADN master stations. The overall mean concentration of total PAH in Chicago ambient air between 1996 and 2003 was 70 nanograms per cubic meter (ng/m^3) and $12 \text{ ng}/\text{m}^3$ for the gas and particle phases, respectively. This is over 10 times higher than those measured at the other IADN sites (Sun et al. 2006b). Over the same period, total PAH in precipitation in Chicago was 2,300 nanograms per liter (ng/L), compared to a range of 26 to 90 ng/L over the other IADN sites (Sun et al. 2006a).

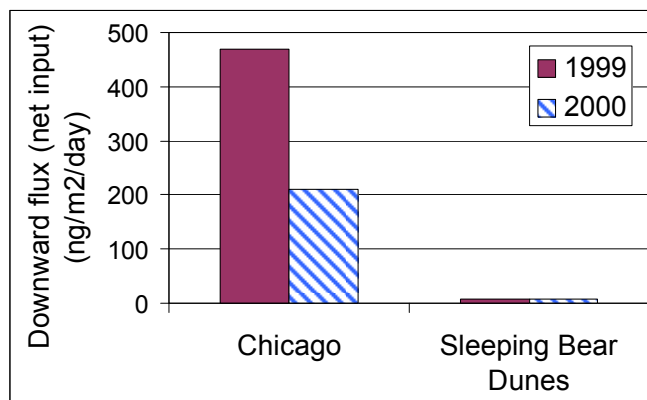
The net deposition downward flux of PAH to Lake Michigan is generally about 20-75 times higher at Chicago than at Sleeping Bear Dunes (Figure 2). This is based on data from IADN stations at both locations for 1999 and 2000 (EC and U.S. EPA 2004).

A study of winter snow packs in Minnesota and around Lake Superior discovered significantly higher concentrations of PAH at sites nearer to urban areas (Franz and Eisenreich 2000). The Cedar Creek Natural History Area and the Gray Freshwater Biological Institute are both sampling locations within approximately 50 km of the urban Minneapolis/St. Paul region. The study authors found that, between 1989 and 1992, these urban sampling locations had higher PAH concentrations than the more rural sampling locations at the Lake Itasca State Forest, the Marcell State Forest, and at the IADN Eagle Harbor site.

Ambient air monitoring in the Ontario Great Lakes Basin also found that urban sites had much higher concentrations than rural sites, indicating significant local releases. Over the years 1999 to 2003, the median concentrations of benzo-a-pyrene at most urban sites were generally an order of magnitude higher than at rural sites. The median concentrations of total PAH over this time period ranged from $0.59 \text{ ng}/\text{m}^3$ at the rural site of Burnt Island on Lake Huron to $28.6 \text{ ng}/\text{m}^3$ at an urban location in Toronto (Environment Canada 2004).

Several researchers demonstrated how passive air samplers could be used to examine the urban-to-rural changes in PAH concentration, as well as the vertical distribution of PAH in the urban atmosphere. Motelay-Massei et al. (2005) deployed passive air samplers for three 4-month integration periods from June 2000 to July 2001 along a transect from urban sites in Toronto to a rural site to the north, in Egbert. These samplers were polyurethane foam disks in a stainless steel

Figure 2. Net Downward Flux of Benzo(a)pyrene at IADN Stations on Lake Michigan in 1999-2000 (EC and U.S. EPA 2004)



domed chamber. Urban concentrations were roughly five times rural concentrations. Concentrations were highest in summer, which the researchers attributed to increases in the evaporative emission from petroleum products such as asphalt. As expected, the relative proportions of the various PAH compounds were different at the urban and rural sites. The compounds with lower molecular weight tended to travel farther, whereas those with higher molecular weight are associated with particles that tended to be deposited closer to the source. The significant finding was that this effect occurred over a relatively short distance (about 75 km).

The vertical distribution of PAH in the Toronto urban atmosphere was studied by Farrar et al. (2005) by deploying passive air samplers at various heights on a tower. They found that PAH concentrations declined sharply with height, indicating that ground-level emissions in urban areas are sources of these compounds. The study also demonstrated how a new design of passive air samplers could be used. These samplers consist of a thin film of polymer coating on a glass cylinder and are designed to equilibrate rapidly under ambient conditions.

3.2 New England Region

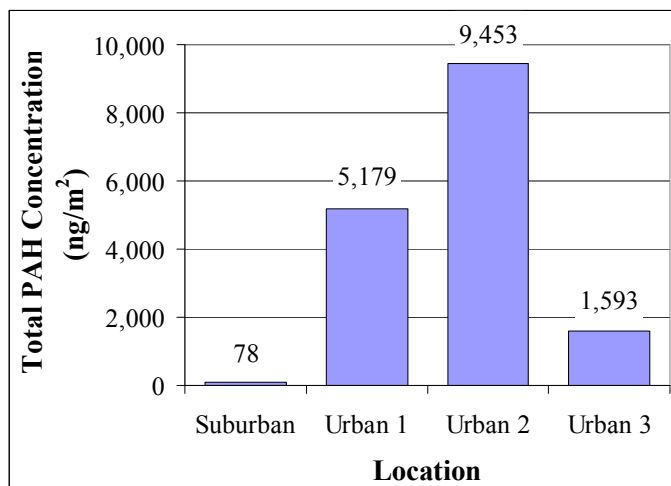
In New England, results from deposition monitoring of PAH to coastal waters from March 1998 through May 2000 (Golomb et al. 2001) indicated that the sampling location closer to the Boston metropolitan area and Logan International Airport (Nahant, Massachusetts) had a higher dry deposition rate of PAH than the more rural location (Wolf Neck, Maine). The urban sampling location averaged 95 nanograms per square meter per hour ($\text{ng}/\text{m}^2/\text{hr}$), while the rural site averaged $9.3 \text{ ng}/\text{m}^2/\text{hr}$. Conversely, the rural location experienced greater wet deposition of PAH, because the air masses that brought precipitation to this site carried more PAH from regional sources.

3.3 Mid-Atlantic Region

In Baltimore, Maryland, higher concentrations of PAH were found in films on the exterior window surfaces located in downtown areas than on those located in suburban areas. PAH samples were collected from one suburban and three urban sites in the summer of 1998 (Liu et al. 2003). Figure 3 shows the relative concentrations found in the suburban, as well as the urban areas. PAH concentrations were much higher in the urban areas, suggesting greater deposition of PAH in the urban areas, apparently from vehicle emissions. This is significant because the PAH that accumulate on window films are likely to be washed off into nearby surface waters during precipitation events.

Also in the Baltimore region, Dachs et al. (2002) determined that atmospheric gas- and particulate-phase PAH concentrations

Figure 3. Total PAH Concentrations Accumulated on Baltimore Windows (Liu et al. 2003)



were approximately two- to three-fold higher in the Baltimore urban area than over the Chesapeake Bay. Additionally, the study authors found that total suspended particulate concentrations in the Baltimore area (23 to 95 micrograms per cubic meter [$\mu\text{g}/\text{m}^3$]) were similar to concentrations observed over the Chesapeake Bay (40 to 84 $\mu\text{g}/\text{m}^3$) when the wind blew from Baltimore. Concentrations were lower over the Chesapeake Bay (4.5 to 40 $\mu\text{g}/\text{m}^3$) when the wind blew from other areas. This study, conducted in July 1997, demonstrates the influence of urban areas as a source of pollution to adjacent waterbodies.

In New Jersey, the concentrations of total PAH in ambient air and precipitation vary across the state. In the first year of sampling (October 1997 to October 1998) at NJADN, researchers found that the PAH concentrations in the atmosphere were about two times higher at the suburban New Brunswick site than at the coastal Sandy Hook site, which is consistent with the fact that New Brunswick is in closer proximity to urban and industrial areas. The Sandy Hook site results were two to 10 times higher than those at the remote Eagle Harbor on Lake Superior, indicating that Sandy Hook should not be classified as a remote site (Gigliotti et al. 2000). Over the full NJADN study period (about 1997 to 2001), PAH concentrations in the atmosphere and precipitation at the most urban/industrial sites (Jersey City and Camden) were four to six times greater than at the NJADN sites representing regional background. However, there was little spatial variation in the PAH profiles (i.e., the relative amounts of individual compounds to the total PAH), indicating that the mix of sources is similar throughout the state. Annual average deposition fluxes ranged from 540 to 7,300 $\mu\text{g}/\text{m}^2/\text{yr}$. The most prevalent component of the deposition flux was gas absorption (55 to 92 percent), followed by dry particle deposition (four to 31 percent), and wet deposition (three to 16 percent) (Reinfelder et al. 2004).

3.4 Southeast and Gulf Region

A study conducted in Miami, Florida, between June 1994 and March 1995 found that air mass movement (i.e., wind direction and frequency) is the dominant factor in determining PAH levels in dry deposition and total suspended particulates (Lang et al. 2002). Winds from the north-northwest and east-northeast come from inland directions and carry more PAH into the city. PAH are most likely being carried from the downtown area by these winds, and are transporting PAH from Miami International airport, automobile exhaust, waste incineration, and natural gas usage areas. These winds increase the PAH concentration in the air over the most densely populated areas of Miami. Conversely, winds from the east and south are coming from the Atlantic Ocean and carry cleaner air. The study authors showed that PAH concentrations were lower when oceanic air was coming from the east and south, as it flushed anthropogenic aerosols out of the metropolitan area. Note that this analysis was based on monthly-averaged data, and therefore should be viewed as general trends, not as quantitative results.

Deposition of PAH compounds directly to Tampa Bay was studied by Poor (2002) and Poor et al. (2004). In the earlier study, measurements were made from March to October 2001. The average concentration for the total PAH in the ambient air was 14 ng/m^3 . Dry deposition of gas and particles was estimated to be about two $\mu\text{g}/\text{m}^2/\text{day}$, and wet deposition of gas and particles was estimated to be about 0.1 $\mu\text{g}/\text{m}^2/\text{day}$, assuming no flux of PAH from the water to the air. A comparison of these rates with others reported in the literature indicated that the rates in Tampa Bay are in the range of deposition rates at both rural and urban sites in the eastern coastal U.S.

(Poor 2002). The latter study used a different sampling method, which improved capture of gas- and particle-phase PAH compounds with lower molecular weights. Based on sampling between May and August 2002, the concentrations of total PAH were between 80 and 190 ng/m³, and dry deposition flux of gas and particles was estimated to be 11.5 µg/m²/day, assuming no flux of PAH from the water to the air. Differences in sampling methodologies are discussed further in Poor et al. (2004).

Deposition of PAH to Galveston and Corpus Christi Bays in Texas was studied by Park et al. (2001, 2002). In Galveston Bay, PAH in ambient air and precipitation was measured between February 1995 and August 1996. Total PAH concentrations ranged from 4 to 161 ng/m³ in air samples, and from 50 to 312 ng/L in rain samples. The annual wet deposition flux was estimated to be 130 µg/m²/yr, and dry particle deposition flux to be 99 µg/m²/yr. The net gas exchange from the air to the surface water was estimated to be 1,211 µg/m²/yr, although the authors noted that this estimate should be considered preliminary (Park et al. 2001).

In Corpus Christi Bay, PAH in ambient air and precipitation were measured between August 1998 and September 1999. Concurrently, water samples were taken to calculate air-water gas exchange. Total PAH concentrations ranged from about two to 57 ng/m³ in air samples. The annual wet deposition flux was estimated to be 182 µg/m²/yr, the dry particle deposition flux to be 68 µg/m²/yr, and the net gas exchange was -38.4 µg/m²/yr (the negative number indicating net exchange from the water to the air) (Park et al. 2002).

3.5 West Coast Region

For the northern San Francisco Estuary, PAH concentrations in the ambient air were studied, and fluxes between air and water were estimated by Tsai et al. (2002). Measurements were made at a single sampling site from June to November 2000. Average monthly concentrations ranged from eight to 37 ng/m³, and were predominantly in the vapor phase. The researchers noted that these concentrations were 10 times lower than in Chicago, and comparable to those found around urbanized areas in Baltimore and New Jersey. Dry deposition of particles ranged from 0.04 to 0.96 µg/m²/day. Gaseous PAH fluxes ranged from about 0.1 µg/m²/day efflux from the estuary to the air in August to about one µg/m²/day influx to the estuary from the air in November. Wet deposition was not measured.

4.0 Emissions and Source Profiles

The third major area of new information on PAH addresses source profiles in various regions as well as trends in PAH emissions from these sources. Many of the recent studies on sources of PAH are consistent with, and in some cases expand upon, the findings in the Third Report to Congress for the Great Lakes region. Furthermore, new information regarding sources of PAH nationally, as well as in Galveston Bay and Corpus Christi Bay, Texas, and the city of Miami has been reported.

4.1 National Emissions Inventory

U.S. EPA's 2002 national emissions inventory (NEI) lists aggregated emissions for 15 individual PAH compounds (15-PAH).⁴ Table 2 shows that the dominant 15-PAH emission sources are residential wood heating and open burning due to forest and wildfires. Comparisons with the National Toxics Inventory for 1990-1993, shown in the Third Report to Congress, reflect changes due to regulations, changes in industry, and changes in knowledge and information on the part of the organizations submitting data that are included in these national emission inventories. In the Third Report to Congress, consumer products usage was listed as the major source for 16-PAH; however, this category is almost completely attributed to naphthalene, which is no longer included in the aggregated list of 15 PAH for the NEI.

⁴ In the Third Report to Congress, naphthalene was included in the list of aggregated compounds (16-PAH); however, EPA is not using 16-PAH in the 1999 NEI for hazardous air pollutants (HAP), because naphthalene is one of the 188 HAP compounds. The chemicals that comprise the 15-PAH group are the ones that can be measured using EPA test method 610 and consist of: benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, chrysene, dibenzo(a,h)anthracene, indeno(1,2,3-c,d)pyrene, fluorene, acenaphthylene, acenaphthene, anthracene, benzo(g,h,i)perylene, fluoranthene, phenanthrene, and pyrene.

**Table 2. National Anthropogenic 15-PAH Air Emissions
(Based on U.S. EPA's 2002 National Emissions Inventory)**

Source Category	Anthropogenic Air Emissions (tons/year)	Percent Contribution to Total U.S. Anthropogenic Air Emissions
Residential Boilers – Wood/Wood Residue Combustion	5,899	52
Open Burning – Forest and Wildfires	2,300	20
Mobile Sources – Onroad	439	4
Open Burning – Residential, Household Waste	426	4
Open Burning – Prescribed Burning	359	3
Coke Ovens – Pushing, Quenching, & Battery Stacks ^a	319	3
Mobile Sources – Nonroad	253	2
Commercial Cooking – Charbroiling	221	2
Catastrophic/Accidental Releases	146	1
Mobile Sources – Aircraft Locomotive Marine	133	1
Industrial/Commercial/ Institutional Boilers & Process Heaters ^a	111	1
Others (<1 percent each) ^b	793	7
Total U.S. Anthropogenic 15-PAH Air Emissions (290 Categories)	11,400	100

^aThese source categories are associated with maximum achievable control technology (MACT) rules.

^bA list of the source categories that contribute less than one percent to total U.S. air emissions is provided in Appendix A.

Source: U.S. EPA 2007

Although it is not within the scope of this report to describe in detail the efforts to reduce current emissions of PAH, it is important to note that many are ongoing. U.S. EPA, other federal agencies, and state/local/tribal governments are involved in reducing emissions from burning through smoke management programs, regulations, and education/outreach activities. For residential wood combustion, U.S. EPA, state/local/tribal governments, and non-governmental partners are working to reduce emissions through voluntary programs, ordinances and education/outreach activities. U.S. EPA also has finalized several rules that will reduce air emissions of POM (of which PAH are a subset) from stationary sources; these rules are based on maximum achievable control technology (MACT). The Third Report to Congress noted the MACT standard for the primary aluminum production industry. Several other industries for which recent MACT standards will reduce POM emissions include asphalt processing and roofing, iron and steel foundries, and refractory products. U.S. EPA has also issued regulations to reduce the residual risk due to coke oven emissions remaining after implementation of the

MACT rule⁵ for coke ovens; constituents of coke oven emissions include POM compounds. In addition, in 2007, U.S. EPA published final rules to control hazardous air pollutants from mobile sources (72 FR 8427, February 27, 2007). The vehicle controls included in the rule may help to reduce deposition of POM.

4.2 Great Lakes Region

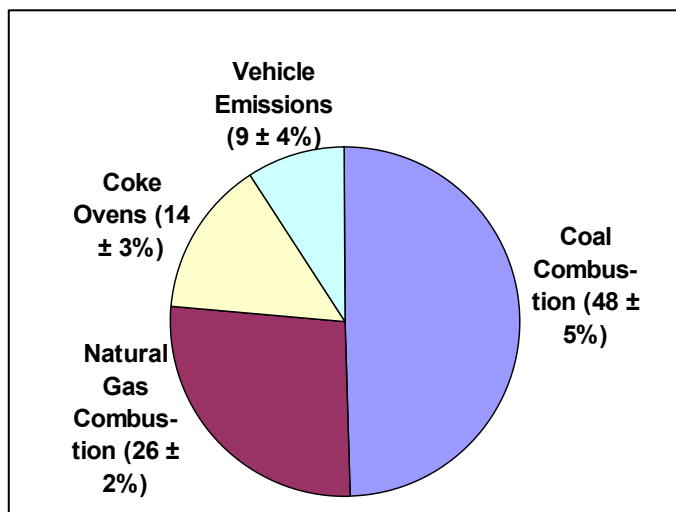
Numerous new studies examined the sources of PAH to the Great Lakes region. Simcik et al. (1999) applied multivariate statistical techniques to PAH concentrations to investigate source apportionment and source/sink relationships in the coastal atmosphere of Chicago and Lake Michigan over 1994 -1995. Figure 4 depicts the percentages of the various sources of PAH based on this analysis.

These sources are consistent with the sources for the region that were identified in the Third Report to Congress. Simcik et al. (1999) also determined that atmospheric deposition is the major source of PAH to the

sediments and water column particulate phase of Lake Michigan. Offenberg and Baker (2002) determined that particle scavenging (as opposed to gas scavenging) during storms is the dominant method for removal of PAH from the atmosphere sampled around southern Lake Michigan along the urban to over-water transect. A study of soot (e.g., partially combusted and/or pyrolyzed carbon) that had been deposited in the Great Lakes found a strong correlation between soot carbon and PAH sediment accumulation at a site in southern Lake Michigan, particularly for high molecular weight PAH. This finding provides additional evidence to the theory that soot is a major vector for deposition of PAH compounds. Furthermore, soot accumulation throughout the Great Lakes was found to be higher closer to large industrial areas, as might be expected due to the proximity of combustion sources (Buckley et al. 2004).

In another study, the chemical mass balance model CMB8.2 was used to determine the major sources of PAH to Lake Calumet and surrounding wetlands in southeastern Chicago (Li et al. 2003). The model, developed by U.S. EPA, has been used primarily to estimate source contributions to ambient air concentrations. In this study, the researchers applied the model to PAH found in aquatic sediment cores. They examined six source categories (coke oven, residential coal burning, coal combustion in power generation, gasoline engine exhaust, diesel engine exhaust, and traffic tunnel air) by establishing fingerprints based on the relative contribution of various PAH compounds for each category. The major sources of PAH to Lake Calumet were determined to be coke ovens and vehicular traffic.

Figure 4. Source Apportionment of Atmospheric PAH in the Coastal Atmosphere of Chicago and Lake Michigan in 1994-1995 (Simcik et al. 1999)



⁵ See <http://www.epa.gov/ttn/atw/> for more information on the MACT and residual risk standards.

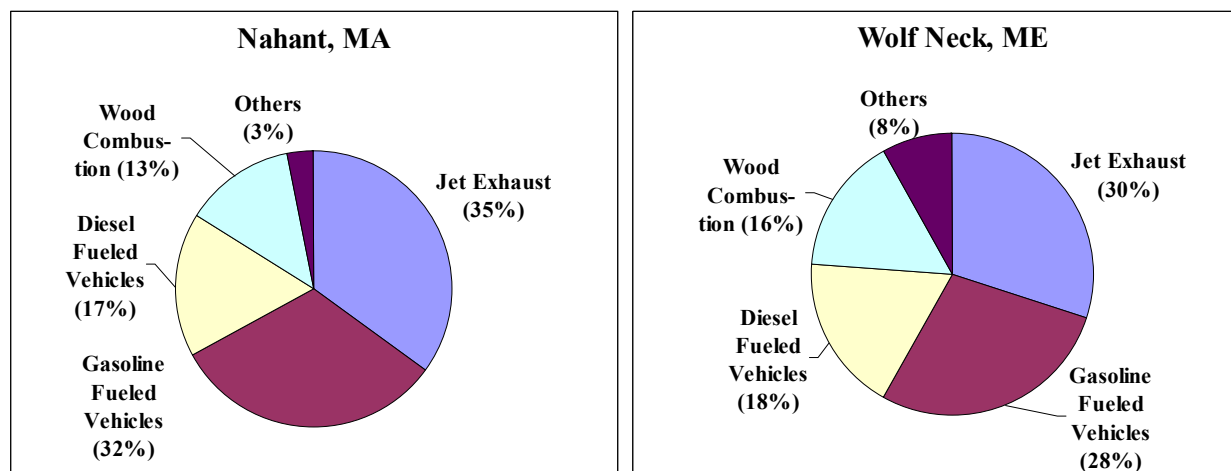
Su et al. (2000) examined sediment cores from the lower Fox River, near Green Bay, Wisconsin. The researchers used a chemical mass balance approach, along with an analysis of the carbon particles in the sediments, to evaluate source trends over time. They determined that the major PAH sources for this area were coke ovens, highway dust, coal gasification, and wood burning. Since 1896, the major source of PAH in the area has been coke ovens, contributing between 40 and 90 percent of the total PAH concentration. Highway dust contributes between 10 and 75 percent of the total PAH concentration. The contribution from coke ovens generally decreased between 1930 and 1990, while the contribution from highway dust has increased since 1930. The percentage of PAH from wood burning has been generally smaller than coke ovens and highway dust; it was estimated to be between three and 10 percent in 1995.

Wood burning is found to be more prevalent in northern and more rural areas around the Great Lakes. The contribution of retene, an indicator of wood combustion, to total PAH concentrations in ambient air was found to be higher in rural sites in Canada (Environment Canada 2004). Similarly, the ratios of retene concentrations to total PAH concentrations in precipitation and ambient air measured at rural IADN sites were significantly higher at the rural sites than in Chicago. The highest ratios in precipitation were found at the sites near Lake Superior (Brule River and Eagle Harbor) (Sun et al. 2006a, Sun et al. 2006b)

4.3 New England Region

The chemical mass balance model CMB8.0 was applied by Golomb et al. (2001) to examine source apportionment in two locations in New England: Nahant, Massachusetts, which is near the Boston metropolitan area and Logan International Airport, and Wolf Neck, Maine, which is more rural. These sites were chosen to represent deposition to Massachusetts Bay and Casco Bay, respectively. The researchers applied the model to samples from dry deposition monitoring of PAH in these locations from March 1998 through May 2000. Four source categories expected to be significant contributors in this region were chosen for the modeling analysis. Figure 5 depicts the source profiles of the two sites. It is interesting to note the differences in the source profiles at these locations, as compared to the Great Lakes site depicted in Figure 4. In the Great Lakes area, coal and natural gas combustion are the two largest sources of PAH, whereas in the New England area, jet exhaust and gasoline fueled vehicles are the two largest sources.

Figure 5. Source Contributions to PAH Dry Deposition at the Nahant, MA and Wolf Neck, ME Sampling Locations (Golomb et al. 2001)



4.4 Baltimore/Chesapeake Bay Region

Larsen and Baker (2003) applied three statistical techniques for source apportionment to PAH concentrations in the Baltimore urban atmosphere, over the period from March 1997 through December 1998. Use of three methods allows for comparison and mitigates the weaknesses of each method. In general, the results from the methods agreed well with one another. Coal combustion contributed 28 to 36 percent of the PAH in ambient air. Vehicles, both gasoline and diesel, contributed 16 to 26 percent. Oil combustion contributed 15 to 23 percent. Wood combustion and other unidentified sources accounted for 23 to 35 percent of the total. During the summer, coal was the dominant source, while during the winter, oil dominated.

Dachs et al. (2002) conducted a study of atmospheric concentrations of PAH in Baltimore and the adjacent Chesapeake Bay in the summer of 1997 to examine short-term variability. Over the long term, the Bay is a receptor of PAH deposition due to regional urban and industrial influences (such as Baltimore). However, depending on certain meteorological conditions, such as wind speed and temperature, the Chesapeake Bay can act as a short-term source of PAH to the atmosphere when there is a relatively clean air mass above it.

Another study (Arzayus et al. 2001) indicated that the dominant mode of entry of PAH from the atmosphere to the southern part of the Chesapeake Bay is indirect atmospheric deposition (i.e., deposition to the surrounding watershed and subsequent runoff). Surface sediment samples were collected in various seasons from five sites in the southern Chesapeake Bay. For all sampling locations and times, increases in PAH concentrations were found to be similar, indicating that a single source or mode of entry controls the distribution of PAH to the Chesapeake Bay sediments. Due to the fact that most of the sampling locations were removed from point sources, the study authors determined that the atmosphere was the dominant source of PAH to the sediments in that region of the bay. Furthermore, the study authors concluded that PAH in the Chesapeake Bay sediments are from airborne soot particles, as the atmospheric soot signal is retained in the sediments.

4.5 Galveston/Corpus Christi Bay Region

In Galveston Bay, Texas, sediment and American oyster samples were analyzed to determine PAH sources and historical trends (Qian et al. 2001). Samples were collected at six sites in Galveston Bay between November and January, in selected years from 1986 to 1998. The primary source of PAH to the bay was determined to be from combustion products, followed by spilled or released petroleum products. There were no temporal trends, so these PAH sources were likely unchanged over the 13-year study. During some years, there was an increase in PAH that the study authors attributed to documented or suspected oil spills. Additionally, Qian et al. (2001) indicated that the bioavailability of the PAH sources determines the concentrations and composition of PAH in oysters. PAH concentrations in Galveston Bay oysters were high compared to national levels, although the authors did not explore the severity of the PAH contamination in this region.

In a study conducted at Galveston Bay between February 2005 and August 1996, PAH sampled in the air were found to be primarily from combustion and petroleum vaporization (Park et al. 2001). These sources are similar to those identified in Corpus Christi Bay, which acts as a sink for PAH. In a similar study conducted between August 1998 and September 1999 (Park et al. 2002), the majority of PAH found on particulates in air and rain were determined to be from combustion sources. This study also indicated that direct atmospheric inputs of PAH were not as large as inputs from land runoff or periodic oil spills. The researchers anticipate that as the population and industrial activity in the Bay increases, atmospheric input of PAH will also increase.

4.6 Miami Region

A study conducted in Miami between June 1994 and March 1995 determined that the main source of atmospheric PAH in that area was determined to be automobile exhaust, due to the elevated levels of benzo(ghi)perylene and coronene found in total suspended particulates analyzed (Lang et al. 2002). Other sources that contributed to the elevated PAH levels for dry deposition included waste incineration, power plants, and biomass accumulation. The authors also concluded that the air in the Miami area should be considered moderately polluted.

5.0 Summary and Some Future Directions for Research

A large amount of new research has been conducted since the publication of the Third Report to Congress on temporal and spatial trends in PAH concentrations and deposition in the Great Lakes region as well as in other Great Waters locations. In addition, several studies focused on PAH source profiling in new regions of interest (e.g., Miami, Galveston Bay). Most studies generally agree with or expand upon information that was provided in the Third Great Waters Report to Congress.

The Great Lakes region is the only Great Waters for which there are long-term data for PAH in the atmosphere and precipitation. There, trends are mixed; either no trends or decreasing trends have been found. In Chicago, decreases in concentrations in the atmosphere and deposition have been observed recently, perhaps due to reductions in sources in the city.

In coastal waters, Mussel Watch provides long-term information about PAH in the environment via concentrations in bivalves. At most of the over 200 sites, there is no trend in PAH concentration. However, the median PAH concentration across all sites nationally decreased between 1986 and 2002.

The greatest concentrations of PAH near the Great Waters are in urban and industrial areas. For example, in the coastal areas of New Jersey, PAH deposition was four to six times higher than at the most urban/industrial sites than at the more rural sites, representative of the regional background levels. Similarly, researchers in the Great Lakes have been determining ways to incorporate the effects of significant urban centers, like Chicago, into estimates of total loadings to the Lakes. Studies of lakes in growing urban areas are showing increasing trends in PAH concentrations in sediments. The urbanization brings increased vehicular traffic, which contributes to PAH from exhaust, asphalt wear, tire wear, and leaks and spills of oil. The findings from these areas may be transferable to urban areas near the Great Waters.

Seasonal trends in the PAH compounds in the ambient air and deposition were found in several areas, particularly those in more northern latitudes. Semi-volatile compounds tend to be more in a particulate state in cooler temperatures. In addition, more combustion-derived PAH emissions due to heating in the colder months add to the seasonality. No seasonal trends were found in PAH concentrations in surface water or in zooplankton in southern Lake Michigan; researchers' interpretation of this finding was that the deposition from the Chicago urban area was relatively constant throughout the year.

Since the Third Report to Congress, deposition studies have been done in several Great Waters areas not previously studied, including the Gulf coast areas of Tampa Bay, Galveston Bay and Corpus Christi Bay, and San Francisco Bay on the West coast. Furthermore, in New Jersey, where there are several Great Waters estuaries, the New Jersey Atmospheric Deposition Network provides a four-year monitoring study of multiple sites throughout the state.

The Third Report to Congress found that both local and regional sources can play a role in deposition of PAH to waterbodies. A new study in New England illustrates that finding. Dry deposition was higher near the urban area of Boston and Logan International Airport than in a rural area in Maine, likely due to local urban sources. However, wet deposition was higher at the rural Maine location because the air masses that brought precipitation to the site in Maine carried more PAH from regional sources than air masses that brought precipitation to the location near Boston.

On a national basis, emissions of PAH are dominated by residential wood burning and open burning. Regionally, however, other sources may dominate, depending on particular industrial or urban activities. Various statistical techniques have been applied to trace the sources in several geographic areas. Depending on the location, other dominant sources include coal combustion, coke ovens, jet exhaust, and vehicles.

Several of the articles in the recent literature recommend continued research to improve knowledge related to PAH trends and sources, and to assess the effects of recent programs to

reduce PAH emissions. Investigators' specific recommendations for future research are as follows.

- There is a need to better understand the effect of population and industrial activity on atmospheric PAH (Park et al. 2002).
- Volatilization from surface water is an important removal process of PAH from Corpus Christi Bay. However, despite technological advances over the past decade, further reliable estimates of physical-chemical parameters like Henry's Law constant and mass transfer coefficient, as well as deposition velocity and loadings from other sources are needed to improve the accuracy of atmospheric loading estimates for semi-volatile organic contaminants (Park et al. 2002).
- Because particle scavenging dominates washout mechanisms for PAH compounds in a wide range of precipitation events, more research is needed on the importance of in-cloud processing to overall scavenging. Direct measurement of in-cloud processing of these compounds would greatly improve the ability to interpret the scavenging and atmospheric processing of these compounds. Furthermore, such an investigation would allow for better explanation of the occurrence of the differing magnitudes of precipitation scavenging found over the course of more than 100 kilometers (Offenberg and Baker 2002).
- Future research and monitoring efforts should strive to include compounds that have been identified as key PAH source markers in models to more accurately identify the source origins of PAH (Larsen and Baker 2003).

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Appendix A. Detailed Breakdown of Air Emissions Inventory by Pollutant for Source Categories Emitting Less than One Percent of Total U.S. Emissions

POLYCYCLIC AROMATIC HYDROCARBONS (MEASURED AS 15-PAH)

Acrylic/Modacrylic Fibers Production ^a	Glass Products Manufacturing
Aerospace Industries ^a	Hospital Sterilizers ^a
Agricultural Chemicals and Pesticides Manufacturing ^a	Incineration
Agricultural Field Burning	Industrial Cooling Towers ^a
Agriculture Production - Orchard Heaters	Industrial Inorganic Chemical Manufacturing ^a
Amino/Phenolic Resins Production ^a	Industrial Machinery and Equipment: Finishing Operations ^a
Asphalt Paving, Block and Roofing Manufacturing	Industrial Machinery Manufacturing
Asphalt Paving: Cutback Asphalt	Industrial Organic Chemical Manufacturing
Asphalt Paving: Emulsified Asphalt	Industrial Organic Chemical Manufacturing ^a
Asphalt Processing and Asphalt Roofing Manufacturing ^a	Industrial Processes
Auto & Light Duty Truck (Surface Coating) ^a	Inorganic Pigments Manufacturing ^a
Autobody Refinishing Paint Shops ^a	Instruments Manufacturing: Photographic and Photocopying Equipment
Boat Manufacturing ^a	Instruments Manufacturing: Surgical and Medical Instruments
Carbon Black Production ^a	Integrated Iron & Steel Manufacturing ^a
Chemical Manufacturing	Iron and Steel Forging ^a
Chemical Preparations ^a	Iron and Steel Foundries ^a
Chemicals and Allied Products Storage	Iron Foundries
Clay Ceramics Manufacturing ^a	Landfills - Commercial/Institutional
Coal Mining, Cleaning, and Material Handling	Landfills - Industrial
Coke Ovens: Charging, Top Side, and Door Leaks ^a	Large Appliance (Surface Coating) ^a
Commercial and Industrial Solid Waste Incineration ^a	Lead Acid Battery Manufacturing ^a
Commercial Cooking: Frying	Leather Tanning & Finishing Operations ^a
Commercial Sterilization Facilities ^a	Lime Manufacturing ^a
Cremation	Medical Waste Incinerators ^a
Crude Petroleum Pipelines	Metal Can (Surface Coating) ^a
Cyclic Crude and Intermediate Production ^a	Metal Furniture (Surface Coating) ^a
Electrical and Electronics Equipment Manufacturing	Mineral Products
Electrical and Electronics Equipment: Finishing Operations ^a	Mining
Engine Test Facilities ^a	Mining and Quarrying: Construction Sand and Gravel
Ethylene Processes ^a	Miscellaneous Coating Manufacturing ^a
Fabricated Metal Products Manufacturing	Miscellaneous Metal Parts & Products (Surface Coating) ^a
Fabricated Plate Work ^a	Miscellaneous Organic Chemical Manufacturing ^a
Fabricated Structural Metal Manufacturing ^a	Municipal Landfills ^a
Ferroalloys Production ^a	Municipal Waste Combustors: Large ^a
Food and Agriculture	Municipal Waste Combustors: Small ^a
Food Manufacturing	Natural Gas Distribution
Friction Materials Manufacturing ^a	Natural Gas Transmission
Gasoline Distribution (Stage I) ^a	Nonferrous Foundries, Not Elsewhere Classified ^a
General Laboratory Activities	Off-Site Waste and Recovery Operations ^a
General Laboratory Activities: Commercial Physical and Biological Research	Oil & Natural Gas Production: Crude Petroleum and Natural Gas Extraction
General Laboratory Activities: Noncommercial Physical and Biological Research	Oil & Natural Gas Production: Natural Gas Extraction
Glass Manufacturing: Flat Glass	

Oil & Natural Gas Production^a
 Open Burning - Managed Burning, Slash (Logging Debris)
 Open Burning - Commercial/Institutional
 Open Burning - Firefighting Training
 Open Burning - Industrial
 Open Burning - Municipal Yard Waste
 Open Burning - Yard Waste
 Organic Liquids Distribution (Non-Gasoline)^a
 Other Solid Waste Incineration^a
 Paper & Other Webs (Surface Coating)^a
 Petroleum and Coal Products Manufacturing
 Petroleum Lubricating Oil and Grease Manufacturing
 Petroleum Product Storage Tanks
 Petroleum Product Storage Tanks: Airports: Aviation Gasoline
 Petroleum Product Transportation
 Petroleum Product Transportation and Marketing: Bulk Stations and Terminals
 Petroleum Refineries
 Petroleum Refineries - Catalytic Cracking, Catalytic Reforming, & Sulfur Plant Units^a
 Petroleum Refineries - Other Sources Not Distinctly Listed^a
 Pharmaceutical Production^a
 Phosphate Fertilizers Production^a
 Phosphoric Acid Manufacturing^a
 Plastic Materials and Resins Manufacturing^a
 Plastic Parts & Products (Surface Coating)^a
 Plastics Products: Custom Compounding of Purchased Resins
 Plastics Products: Fiberglassing
 Plastics Products: Polystyrene Foam Product Manufacturing
 Plastics Products: Unlaminated Plastics Plate, Sheet, and Profile Shapes Manufacturing
 Plating and Polishing^a
 Plywood and Composite Wood Products
 Plywood and Composite Wood Products^a
 Polyether Polyols Production^a
 Polyethylene Terephthalate Production^a
 Polymers and Resins - NonMACT
 Portland Cement Manufacturing^a
 Prepared Feeds Manufacturing
 Prepared Feeds Manufacturing^a
 Pressed and Blown Glass and Glassware Manufacturing^a
 Primary Aluminum Production^a
 Primary Magnesium Refining^a
 Primary Metal Production

Primary Metal Products Manufacturing^a
 Printed Circuit Board Manufacturing
 Printing, Coating & Dyeing Of Fabrics^a
 Printing/Publishing (Surface Coating)^a
 Publicly Owned Treatment Works (POTWs)^a
 Pulp and Paper Production - NonMACT Facilities
 Pulp and Paper Production^a
 Refractory Products Manufacturing^a
 Reinforced Plastic Composites Production^a
 Residential Heating, NEC
 Rocket Engine Test Firing^a
 Rubber Tire Production^a
 Sawmills
 Secondary Aluminum Production^a
 Secondary Copper Smelting^a
 Secondary Lead Smelting^a
 Secondary Nonferrous Metals^a
 Semiconductor Manufacturing^a
 Sewage Sludge Incineration^a
 Shipbuilding & Ship Repair (Surface Coating)^a
 Site Remediation^a
 Solvent Extraction for Vegetable Oil Production
 Solvent Extraction for Vegetable Oil Production^a
 Solvent Use
 Stainless and Nonstainless Steel Manufacturing: Electric Arc Furnaces (EAF)^a
 Stationary Combustion Turbines - Gasified Coal
 Stationary Combustion Turbines - Natural Gas^a
 Stationary Combustion Turbines - Oil^a
 Stationary Reciprocating Internal Combustion Engines - Geothermal
 Stationary Reciprocating Internal Combustion Engines - Natural Gas^a
 Stationary Reciprocating Internal Combustion Engines - Oil^a
 Stationary Reciprocating Internal Combustion Engines^a
 Synthetic Organic Chemical Manufacturing (HON)^a
 Synthetic Rubber Manufacturing^a
 Utility Boilers
 Valves and Pipe Fittings Manufacturing^a
 Waste Disposal, Treatment, and Recovery: TSD
 Waste Disposal: Industrial
 Wastewater Treatment: Industrial
 Wood Building Products (Surface Coating)^a
 Wood Furniture (Surface Coating)^a
 Wood Preserving^a
 Wool Fiberglass Manufacturing^a

^aThese source categories are associated with MACT rules.
 Note: NEC indicates “not elsewhere classified.”

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