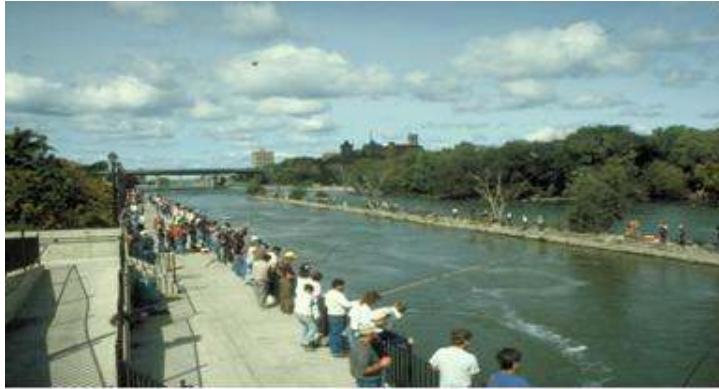


# Survey of New Findings in Scientific Literature Related to Atmospheric Deposition to the Great Waters:

## Polychlorinated Biphenyls (PCB)



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Survey of New Findings in Scientific Literature  
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Polychlorinated Biphenyls

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

## 1.0 Introduction and Highlights

Atmospheric deposition of pollutants, including polychlorinated biphenyls (PCB), has been recognized as a significant contributor in many locations to water quality problems, including toxic contamination of fish and bioaccumulation in wildlife and humans. 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<sup>1</sup> of the coastal United States. PCB are included in the group of pollutants of concern for the Great Waters.<sup>2</sup> 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 “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

### HIGHLIGHTS Polychlorinated Biphenyls

- **Environmental Progress.** Decreases in PCB bioconcentration in fish and fish-eating birds, and increased populations of these birds in the Great Lakes and southern New Jersey indicate improvements in ecosystem health. Nationally, PCB concentrations in mollusks are also showing a decreasing trend, although many individual locations show no trends. PCB contamination continues to be a concern to human health in the Great Lakes, Lake Champlain, northeast coast of the U.S., Chesapeake Bay, and some areas of the Pacific coast, including San Francisco Bay and Puget Sound, as evidenced by fish consumption advisories in effect in 2004.
- **Temporal and Spatial Trends.** In the Great Lakes, PCB concentrations in sediment, surface water and ambient air have been declining since the 1970s, although the rate of decline appears to be leveling off. In general, urban and industrial sites exhibit higher PCB concentrations than suburban sites, which show higher concentrations than rural sites. Across many of the Great Waters, net fluxes of PCB are in the direction of volatilization from the surface water to the surrounding atmosphere, making these waterbodies sources of PCB to the atmosphere.
- **Sources.** Volatilization from, and combustion of, materials containing PCB result in emissions. The 2002 National Emission Inventory cites open burning of residential household waste as a major contributor nationally. In Chicago, sources include transformer storage yards, municipal sludge drying beds, and landfills, among others. Several geographically based studies looked at a variety of inputs to the waterbodies, including emission sources, rivers and suspended sediment, discharges, and global sources. Areas included the mid-Atlantic area, Gulf Coast, and San Francisco Bay, as well as the Great Lakes.

<sup>1</sup> The estuaries that are part of the Great Waters are those that are 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).

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

information related to environmental concentrations, deposition trends and sources of PCB 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. References are provided at the end of this summary.

The recent scientific research highlights the declining trend in PCB concentrations and the associated ecosystem health improvement in the Great Waters. At the same time, the research also points to continuing concerns due to PCB contamination.

## 2.0 Background

PCB are mixtures of similar synthetic organic compounds, known as congeners, that were used widely in electrical, heat transfer, and hydraulic equipment; as plasticizers in paints, plastics and rubber products; in pigments, dyes and carbonless copy paper and many other applications. Manufacture of PCB in the U.S. ceased in 1977; however, PCB are still being emitted into the atmosphere via vaporization from, and open burning of, products containing PCB.

Additionally, PCB have persisted and bioaccumulated in the environment as they are transferred continuously between air, water, and soil by natural

chemical and physical processes such as weathering, runoff, precipitation, atmospheric deposition, and advection (Sun et al. 2006). Figure 1 represents a general schematic of the release, transport, and deposition process for pollutants. While PCB are not emitted from natural or mobile sources or transformed in the environment, the transport from the atmosphere to deposition is consistent with Figure 1.

### Definition of Common Terms

**Direct deposition:** The 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.

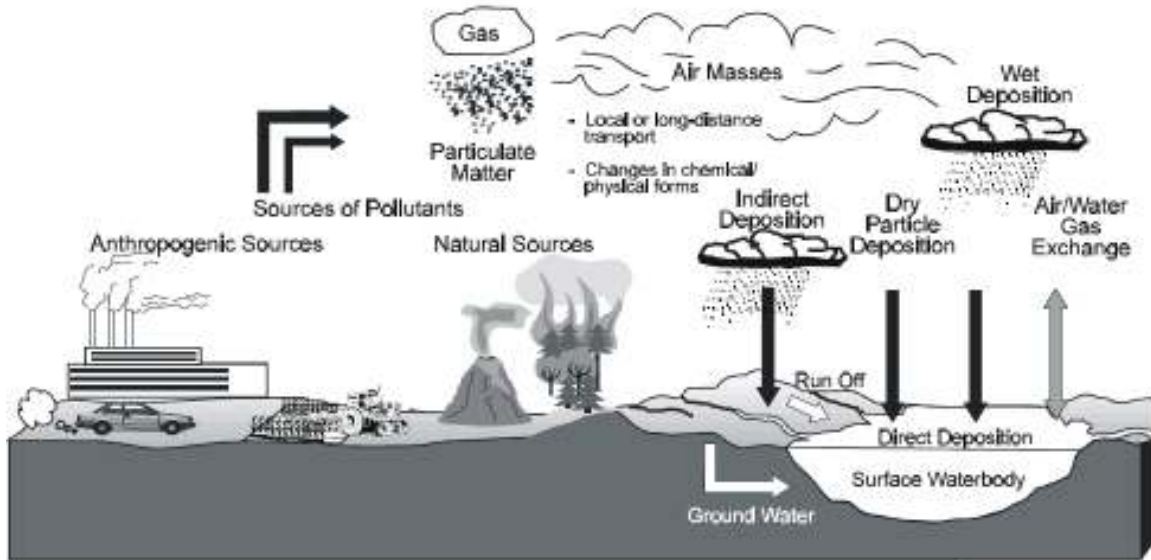
**Indirect deposition:** The deposition of air pollution to the rest of the watershed. Once pollutants are deposited in the watershed, some portion is transported through runoff, rivers, streams, and groundwater to the waterbody of concern. The portion that reaches the waterbody by passing through the 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

**Figure 1. Atmospheric Release, Transport, and Deposition Processes**



Source: U.S. EPA 2000

PCB have been shown to cause cancer and a number of serious non-cancer health effects in animals, including effects on the immune system, reproductive system, nervous system, and endocrine system. Studies in humans provide supportive evidence for the potential carcinogenicity and non-carcinogenic effects of PCB (U.S. EPA 1998). Of the various PCB congeners, several are considered to be similar to dioxin because of their structure and toxicity. The research results summarized in this report did not distinguish between these dioxin-like PCB congeners and other congeners. The scientific results described in this survey report typically are the sum of a subset of PCB congeners; the congeners included in the subset varied among researchers and can be found in the original references.

### **3.0 Improvements in Ecosystem Health**

Several examples of improved ecosystem health – as reflected in biomarkers – are reported in the Third Report to Congress. Newly published information is consistent with those findings.

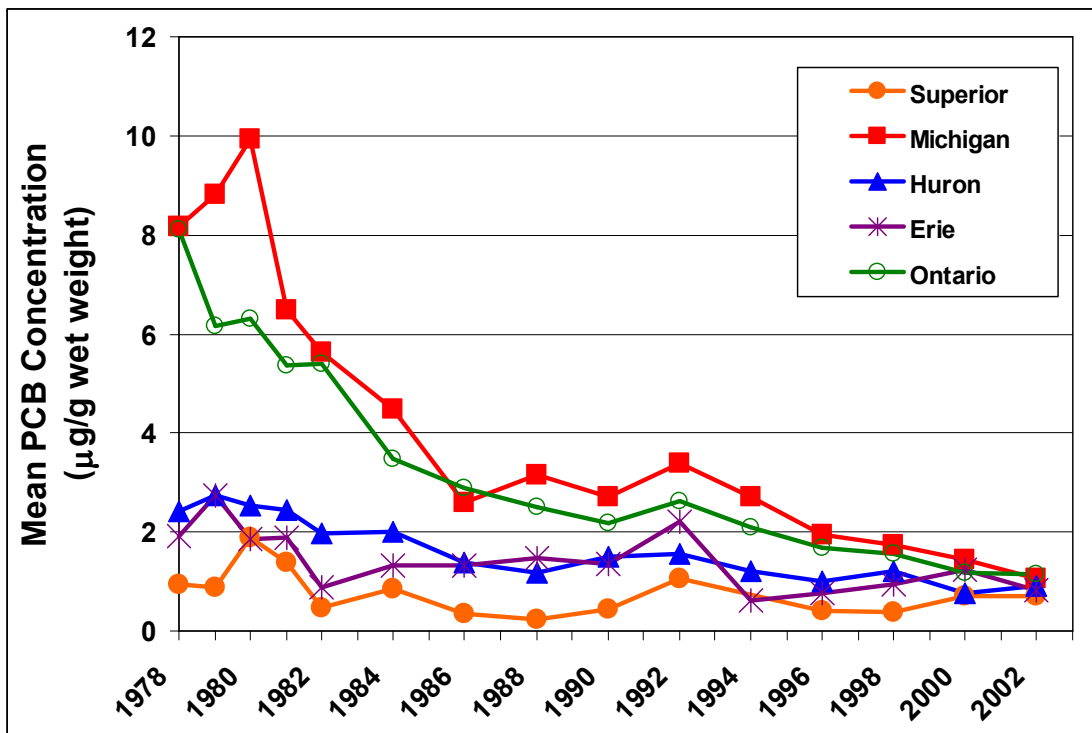
#### **3.1 Great Lakes**

##### **3.1.1 Fish**

Concentrations of PCB in fish generally have been declining in the Great Lakes since monitoring began in the 1970s and 1980s. Recent analytical results indicate that on average, total PCB concentrations in whole Great Lakes top predator fish have declined five percent annually between 1990 and 2003. This decline has been due largely to various remedial, mitigative, and pollution prevention efforts, such as the remediation of contaminated sediments and the reduction of PCB loadings to the Great Lakes (U.S. EPA 2007a, EC and U.S. EPA 2005).

Figure 2 shows these trends for each of the lakes. In Lake Michigan, PCB concentrations in lake trout have declined consistently over the last two decades. PCB concentrations in lake trout of Lake Superior fluctuated through the early 1980s, with greater stabilization after that period. A slight increase was observed in 2000; however, this increase may have resulted from a change in collection sites, or the fact that the sample population was consuming more contaminated prey than the previous sample population collected from that site in 1998. In Lake Huron, an overall decline in PCB concentrations was observed with some periodic increases seen through 2000. As noted in Figure 2, walleye are collected in Lake Erie as the top predator fish instead of lake trout because they are more representative of conditions in that lake. PCB concentrations in Lake Erie increased in the late 1980s through the early 1990s, after which PCB concentrations suddenly declined. The period of increase corresponds with the introduction of zebra mussels into Lake Erie. Zebra mussels remove PCB contamination from open water and deposit it in the sediment making it available to bottom feeding fish. Consequently, higher concentrations are observed in walleye since they prey on some bottom feeding fish. Finally, in Lake Ontario, PCB concentrations have declined through 2000 with little observed fluctuation since the late 1990s (EC and U.S. EPA 2005).

**Figure 2. Decline of PCB Fish Contamination in Great Lakes Top Predator Whole Fish<sup>a</sup>**



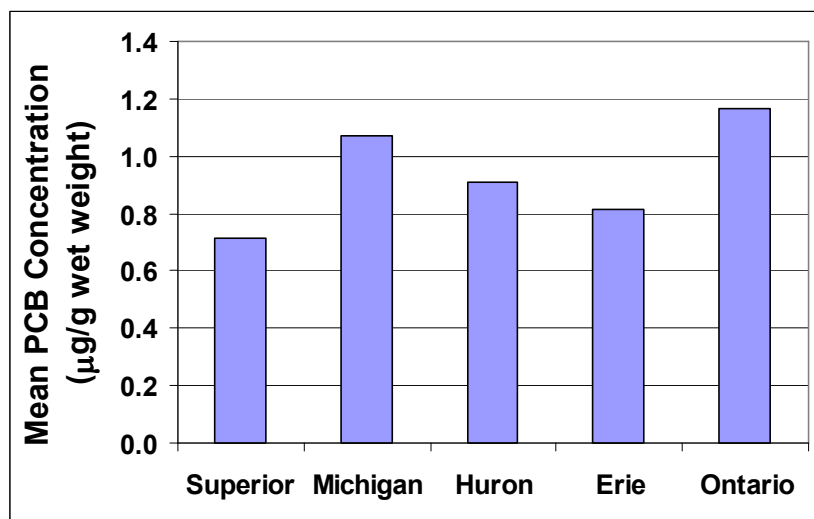
Source: U.S. EPA 2006

<sup>a</sup> Data collected for lake trout and reported for sites sampled in even years (the collection site locations are in different parts of each lake during odd and even years). Note that annual sampling was done at these sites from 1978 – 1982. Walleye collected in Lake Erie instead of lake trout due to the limited number of lake trout and the difficulty in collecting them.



Despite the observed trends discussed above, concentrations in Great Lakes fish are still high enough to be of concern for consumption by wildlife and humans. The Great Lakes Water Quality Agreement includes a threshold concentration of total PCB in fish tissues (whole fish, calculated on a wet weight basis) of no more than 0.1 micrograms per gram ( $\mu\text{g/g}$ ) or parts per million (ppm) for the protection of birds and animals which consume fish (U.S. and Canada, 1987). The U.S. EPA has established a protection value for fish-consuming wildlife and birds at 0.16 ppm in fish tissue (U.S. EPA 2005a). Figures 2 and 3 demonstrate that PCB concentrations in all of the Great Lakes for top predator fish exceed these values. Furthermore, PCB levels in Great Lakes fish are high enough to warrant fish consumption advisories for humans for all five of the Great Lakes; these are discussed later in this section.

**Figure 3. PCB Fish Contamination in Great Lakes Top Predator Whole Fish in 2002**



Source: U.S. EPA 2006

### 3.1.2 Fish-eating Birds

Reductions in PCB levels in herring gull eggs at Lakes Ontario and Erie were discussed in the Third Report to Congress. Current literature also reported reductions in PCB concentrations in gull eggs. For example, at sites on Lake Ontario, concentrations in herring gull eggs have decreased from approximately  $70 \mu\text{g/g}$  (wet weight) in 1970 - 1974 to less than  $10 \mu\text{g/g}$  in 1995 - 1999 (EC and U.S. EPA 2003). Levels of contamination vary spatially around the Great Lakes. Mean values of contaminants in herring gull eggs were calculated for the five-year period 1998-2002 around the Great Lakes. Across the 15 colonies examined, mean values ranged between two and  $22 \mu\text{g/g}$  (wet weight) for total PCB; the highest concentration was in Saginaw Bay in Lake Huron (Weseloh et al. 2006). Herring gull eggs provide a bio-marker for colonial waterbirds, including other species such as terns and cormorants. Populations of most waterbird species in the Great Lakes have increased and become healthier than they were 30 years ago, as the majority of contaminant levels have decreased. However, more subtle health effects may still be found in these bird populations (EC and U.S. EPA 2005).



In bald eagles, the Third Report to Congress included a study showing a significant reduction in PCB concentrations in added eggs between 1969 and 1993 near Lake Superior. In more recent studies of bald eagles nesting on the shores of the Great Lakes, PCB concentrations in eggs, blood and feathers are either decreasing or stable (EC and U.S. EPA 2005). At nests within eight kilometers of Lake Michigan, PCB concentrations in added eggs showed no temporal trend over the period 1986-2000. The overall mean PCB concentration in these eggs was 33.8  $\mu\text{g/g}$  wet weight, which is near a concentration associated with reproductive failure (33  $\mu\text{g/g}$  wet weight) in an earlier study of bald eagles. A caveat related to this threshold, however, is that there is a strong correlation between levels of dichlorodiphenyl-dichloroethene (DDE) and PCB in eagle eggs. As a result, it is difficult to separate the effects of PCB from DDE (Dykstra et al. 2005).

The number of active bald eagle territories in the Great Lakes basin has risen substantially since the 1970s, when there were practically no successful bald eagle nests along the shores of the Great Lakes. For example, over the time period between 1997 and 2001, there were on average about 80 occupied territories near Lake Michigan, and about 190 near Lake Erie. Furthermore, over this same period, the average number of young fledged per territory per year was close to one (EC and U.S. EPA 2005). This number of young fledged is indicative of a healthy expanding population (Dykstra et al. 2005). Yet there are several gaps in the pattern of reproductive recovery, including reaches of shoreline where the population has not recovered (EC and U.S. EPA 2005).

### *3.1.3 Great Lakes Food Web*

Morrison et al. (2002) found that differences in concentrations of PCB measured in biota in Lakes Erie and Ontario were chemical-, species-, and food web-specific. The effects of varying PCB concentrations in water and sediment on several aquatic biota were modeled and field-verified. This study suggests that PCB transfer between water and air occurs more quickly than between water and sediment. This, combined with reductions in deposition of PCB to surface waters over time in Lake Ontario and eastern Lake Erie, leads to lower PCB concentrations in the water column, while PCB still remain in sediment in these areas. Therefore, aquatic biota in Lake Ontario and eastern Lake Erie tend to accumulate less of their PCB body burden from surface water and more from bottom sediments. As bottom sediments become the predominant source of PCB to aquatic biota, the concentrations of PCB in aquatic biota are expected to approach chemical equilibrium with the sediment. This was observed in PCB concentrations in Lake Ontario lake trout, which are declining more slowly in recent years than previously (Huestis et al. 1996). However, in the western basin of Lake Erie, PCB continue to be input from the Detroit River and aquatic biota continue to derive a large proportion of their PCB body burden from suspended PCB in water. Therefore, remediation of contaminated bottom sediments may greatly reduce contaminant levels in aquatic biota in ecosystems similar to Lake Ontario and the eastern basin of Lake Erie. On the other hand, a two-pronged remediation approach focused on both reducing inputs of PCB and decontaminating bottom sediment would be needed in ecosystems similar to the western basin of Lake Erie (Morrison et al. 2002).

Stapleton et al. (2001) also studied the pathways for PCB inputs to the foodweb in Lake Michigan. With a slowing of the decline of PCB concentrations in Lake Michigan lake trout in the mid-1980s, the researchers wanted to understand whether contaminated sediment or atmospheric

inputs were the primary sources of PCB in the food web. This study was done by analyzing samples of sediment, settling organic matter, suspended particulate matter (seston) and biota in various levels of the food web for PCB and stable isotopes of nitrogen and carbon. Stapleton et al. (2001) concluded that sediments were not the source of PCB to the food chain and that PCB body burdens are affected by contaminant levels in both seston and settling particles in the water column. Algal material and seston derive PCB from atmospheric inputs. Furthermore, sediments are frequently resuspended in Lake Michigan due to storms and other events, which increase the residence time of particles in the water column. This study, therefore, suggests that response times for changes in PCB body burden in lake trout is governed by the air-water exchange, residence time of settling particles, mass flux of particles in the water column, and partitioning of PCB between the dissolved phase and seston (Stapleton et al. 2001).

### ***3.2 Other Geographic Areas***

In southern New Jersey, contamination in osprey eggs and prey fish was studied in 1989 and a decade later in 1998. Concentrations of PCB in eggs showed about a 60 percent decline from 1989 levels as compared to 1998 levels, while concentrations in prey fish showed about a 35 percent decrease from 1989 levels. The 1998 concentrations of PCB were found to be below levels considered toxic to egg development, which was exemplified in improved osprey nest success in the Delaware Bay study area and a 200 percent increase in osprey nesting populations in the mid-Atlantic coast and Maurice River study areas (Clark et al. 2001).

In coastal waters across the U.S., the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends Mussel Watch Program has monitored concentrations of trace chemicals in mussels and oysters since 1986. The sites were selected to be representative of large areas, rather than smaller-scale areas that are expected to be influenced directly by a particular source of contaminants. Because mollusks concentrate chemicals from the surrounding waters in their tissues, they provide an integrated measurement of contaminant trends, rather than a single point in time (O'Connor 2002). The Mussel Watch Project samples more than 220 sites regularly (U.S. EPA 2005a). A national-scale analysis of the median PCB concentrations in mollusks from 1986 to 2002 showed a statistically significant decreasing trend (U.S. EPA 2005a).

For 206 individual Mussel Watch sites, 171 sites showed no trend, 30 sites showed a decreasing trend, and five sites showed an increasing trend for total PCB in mollusk tissue between 1986 and 1999. Specific to sites located in the National Estuarine Research Reserves (NERR), PCB levels were increasing at one NERR; other sites showed no trend (Lauenstein and Cantillo 2002). A spatial analysis done by O'Connor (2002) found a correlation between high concentrations of total PCB and sites in close proximity of urban areas, which is not surprising since PCB are synthetic chemicals. Many of these high concentration areas are in the northeastern U.S. Concentrations at some of the urban areas are in a range that possibly may have detrimental effects to the mollusks by causing alterations in lysosomes of digestive cells (O'Connor 2002). In one instance, a site exceeded the levels of concern for human consumption of mussels and oysters set by the Food and Drug Administration (FDA); this was the Angelica Rock site in Buzzards Bay, Massachusetts in 1989 (O'Connor 2002). The U.S. EPA has more stringent human consumption guidelines for PCB than FDA. Based on the U.S. EPA's guidelines, there were 47 exceedances for the 222 sites sampled in 2001 and 2002 (U.S. EPA 2005a).

### **3.3 Fish Consumption Advisories**

The contamination of fish due to PCB continues to be of concern for human consumption in many of the Great Waters. Fish consumption advisories can vary in breadth of coverage by population, fish species, chemical contaminant, and waterbody type, among others. As of 2004, all of the Great Lakes and Lake Champlain had active fish consumption advisories for them due to PCB contamination. All the states in the northeastern U.S. had statewide coastal advisories related to PCB. The Chesapeake Bay and its tributaries also had active advisories. On the west coast of the U.S., there were advisories for fish and shellfish consumption in San Francisco Bay and Puget Sound. While advisories to limit fish consumption were rescinded in the Columbia River in Oregon, there continued to be guidelines to not consume fatty parts of the fish tissue due to PCB contamination (U.S. EPA 2005b).

#### **Fish Consumption Advisories**

U.S. states, territories and Native American tribes are responsible for protecting their residents from possible health effects of eating fish caught in their boundaries. Fish consumption advisories are their primary means of limiting human exposure when fish taken from a particular waterbody contain levels of pollutants that exceed recommended intake levels.

These advisories are issued in several forms - from a comprehensive consumption guide to a listing of state waterbodies and their associated consumption advice. They can be issued to either the general population or subpopulations potentially at greater risk (e.g., children, pregnant or nursing women) to restrict or avoid consumption of specific species of fish and other wildlife caught locally from specific waters or waterbody types (U.S.EPA 2003b, 2005b). All advisories are publicly available through the EPA "Fish Advisories" website (<http://www.epa.gov/waterscience/fish/>).

## **4.0 Temporal and Spatial Trends in PCB Concentrations in Environmental Media**

### **4.1 Temporal Trends in Environmental Media**

In general, PCB concentrations in the Great Lakes (Marvin et al. 2002, Marvin et al. 2003, Morrison et al. 2002, Offenberg and Baker 2000) and Great Lakes atmosphere (Buehler et al. 2002, Sun et al. 2006) are continuing on a downward trend, although the rate of decline in the Great Lakes may have leveled off in recent years. Marvin et al. (2002) found a three-fold decline for PCB sediment concentrations between 1971 and 1997 in Lake Erie and a six-fold decline for PCB sediment concentrations between 1981 and 1998 in Lake Ontario. Similarly, Offenberg and Baker (2000) reported that the concentration of total PCB in Lake Michigan surface waters has declined ten-fold between 1980 and 1994. However, while the 1994 average concentration was clearly lower than concentrations measured in 1991 and 1992, it was similar to the average concentration measured in spring 1993. In regards to the declining levels observed in the Great Lakes atmosphere, Buehler et al. (2002) found that atmospheric PCB concentrations decreased

with a half-life of 10 years at the Sleeping Bear Dunes and six years at the Eagle Harbor Integrated Atmospheric Deposition Network (IADN) stations. In Chicago, Sun et al. (2006) found significant decreasing trends in both precipitation and the atmosphere based on the period 1996 – 2003, with half-lives of about seven years.

The IADN was established in the Great Lakes area to regularly measure select pollutants in air and precipitation for atmospheric deposition (EC and U.S. EPA 2002). The IADN collects gas, particle, and precipitation phase samples at each of its master stations and some of its satellite sites. Master stations are located on Lakes Erie (Sturgeon Point), Michigan (Sleeping Bear Dunes), Superior (Eagle Harbor), Huron (Burnt Island), and Ontario (Point Petre), while satellite stations are located in Chicago and Cleveland and various locations throughout Canada (see Figure 4 for a map of the Great Lakes). The master stations are situated in rural locations to obtain representative regional values, while the Chicago and Cleveland stations provide useful information about levels of toxic substances in urban air and precipitation. Volatilization from the Great Lakes is estimated using water concentrations from other monitoring or research projects for some compounds (EC and U.S. EPA 2004). As a result of the various sources of data, there exists the potential for inconsistencies in the data between years and lakes.

**Figure 4. The Great Lakes**



The United States - Canadian IADN Scientific Steering Committee (EC and U.S. EPA 2002) combined IADN gas-phase PCB air monitoring data with other available historic data back to 1977 to analyze changes in atmospheric concentrations since the ban of PCB. The results indicate an overall decreasing trend (by a factor of seven to 10) in gas-phase PCB air concentrations over Lakes Superior and Michigan from the late 1970s to the mid-1990s, after which gas-phase PCB concentrations start to level off.

The precipitation data from IADN stations were analyzed by Simcik et al. (2000) to determine if wet deposition of semivolatile organic compounds, including PCB, decreased at the same rate as concentration reductions observed in fish, water and air for data available from 1991 to 1997. The data indicated similar concentration half-lives in Great Lakes precipitation, air, water, and biota, suggesting that most of these contaminants are at long-term equilibrium among all environmental compartments (Simcik et al. 2000). Recent IADN data show PCB concentrations in precipitation are very low and approaching blank concentrations, most notably at the master stations of Eagle Harbor and Sleeping Bear Dunes on Lake Superior and Lake Michigan (EC and U.S. EPA 2004).

## KEY TERMS

An **environmental concentration** is the amount (i.e., mass) of a substance within a certain volume or mass of a given medium. Concentrations are reported in different units, depending on the type of medium. For example, ambient air concentrations may be reported in units of nanograms per cubic meter ( $\text{ng}/\text{m}^3$ ), while concentrations in fish are expressed as parts per million (ppm) – or the milligrams of PCB per kilogram of fish tissue or muscle.

**Mass Flux** is the rate of mass flow or transfer of a chemical across a unit area. In this report, for example, flux is reported between surface water and air. It is the rate of diffusion or transport of PCB across the surface or interface (e.g., nanograms per square meter per day or  $\text{ng}/\text{m}^2/\text{day}$ ). Several fluxes discussed in this report are:

- **Wet deposition flux**, which is the rate of transfer of a chemical from the atmosphere to the surface of the earth based on the intensity and frequency of precipitation.
- **Dry deposition flux**, which is the rate of transfer of a chemical from the atmosphere to the surface of the earth by diffusion and the gravitational settling of contaminated atmospheric particles.
- **Net gas exchange flux**, which is dependent on gas absorption (from the air) and volatilization (from water to the air).
- **Net flux**, which is the sum of wet deposition, dry deposition and net gas exchange.

### *4.2 Spatial Trends in Environmental Media*

Several new studies evaluated the spatial trends in PCB concentrations for various environmental media, often with a focus on comparisons between urban, suburban, and rural locations. Spatial trends consistent with those reported in the Third Report to Congress also have been reported in the new studies. That is, waterbodies and/or the air in urban and industrial settings show higher concentrations than those in suburban settings, which show higher concentrations than those in remote locations (Brunciak et al. 2001, Chiarenzelli et al. 2000, Totten et al. 2003, Van Ry et al. 2002, Buehler and Hites 2002, Liu 2003, Reinfelder et al. 2004, Totten et al. 2004). Table 1 summarizes the relative concentrations of PCB in different media at various geographic locations based on comparisons made within the studies.

A number of researchers demonstrated how passive air samplers could be used to examine the urban-to-rural changes in PCB concentration. Passive air samplers were deployed for three 4-month integration periods from June 2000 to July 2001 from Toronto urban sites to a rural site about 75 kilometers to the north. Urban concentrations were about five to 10 times rural concentrations. The researchers also found that the PCB mixture was different between urban and rural sites. The higher-chlorinated PCB, typically associated with the particle phase, were found in greater concentration at the urban sites. At the rural sites, the PCB mixture had more PCB congeners with lower chlorination, typically associated with the gas phase. The researchers believe that this is due to higher particle concentration and deposition fluxes in urban areas (Harner et al. 2004, Motelay-Massei et al. 2005).

**Table 1. Spatial Comparisons of PCB Concentrations in Different Mediums**

Medium	Concentration Comparison <sup>a</sup>	Source
Atmosphere	<ul style="list-style-type: none"> <li>• Chicago &gt; coastal New Jersey sites &gt; Great Lakes<sup>b</sup></li> <li>• Chicago &gt; Galveston Bay &gt; Corpus Christi Bay<sup>c</sup> &gt; Chesapeake Bay<sup>d</sup></li> <li>• Akwesasne Mohawk Nation lands (New York, along the St. Lawrence River) &gt; Great Lakes</li> <li>• Downwind of Lake Ontario &gt; Great Lakes</li> <li>• Northern San Francisco Bay &gt; rural Chesapeake Bay &gt; Great Lakes</li> <li>• Camden &gt; Jersey City &gt; rest of New Jersey</li> </ul>	<ul style="list-style-type: none"> <li>• Brunciak et al. 2001</li> <li>• Park et al. 2001, Park et al. 2002</li> <li>• Chiarenzelli et al. 2000</li> <li>• Chiarenzelli et al. 2001</li> <li>• Tsai et al. 2002</li> <li>• Reinfelder et al. 2004, Totten et al. 2004</li> </ul>
Precipitation	<ul style="list-style-type: none"> <li>• Chicago ≈ urban-industrial New Jersey sites</li> <li>• Great Lakes rain concentrations ≈ snowpack concentrations in Minnesota within 50 km of Lake Superior</li> <li>• Chesapeake Bay ≈ urban-impacted suburban New Jersey sites</li> </ul>	<ul style="list-style-type: none"> <li>• Van Ry et al. 2002, Sun et al. 2006</li> <li>• Franz and Eisenreich 2000</li> <li>• Van Ry et al. 2002</li> </ul>
Surface Water	<ul style="list-style-type: none"> <li>• Lake Ontario &gt; Lake Erie<sup>e</sup></li> <li>• Green Bay (in northwestern Lake Michigan) &gt; southern Lake Michigan</li> </ul>	<ul style="list-style-type: none"> <li>• Morrison et al. 2002</li> <li>• Totten et al. 2003</li> </ul>
Sediment	<ul style="list-style-type: none"> <li>• Lake Ontario &gt; Lake Erie</li> <li>• Lake Ontario &gt; Lake Winnipeg ≈ Lake Superior ≈ Lake Michigan</li> </ul>	<ul style="list-style-type: none"> <li>• Morrison et al. 2002, Marvin et al 2002</li> <li>• Rawn et al. 2000</li> </ul>
Biota	<ul style="list-style-type: none"> <li>• Lake Ontario &gt; western Lake Erie &gt; eastern Lake Erie</li> </ul>	<ul style="list-style-type: none"> <li>• Morrison et al. 2002</li> </ul>

<sup>a</sup> Comparisons are designated based on subjective author determination in the source cited, and are denoted using “>” to indicate greater than and “≈” as approximately equivalent.

<sup>b</sup> Great Lakes refers to the IADN master stations located on Lakes Erie (Sturgeon Point), Michigan (Sleeping Bear Dunes), Superior (Eagle Harbor), Huron (Burnt Island), and Ontario (Point Petre).

<sup>c</sup> Galveston Bay atmospheric PCB concentrations are approximately four times greater than atmospheric concentrations at Corpus Christi Bay.

<sup>d</sup> Corpus Christi Bay atmospheric PCB concentrations are slightly greater than atmospheric concentrations at Chesapeake Bay.

<sup>e</sup> Water concentrations in Lake Ontario are 1.3 to 2.3 times greater than water concentrations in the western basin of Lake Erie, which is influenced by Detroit River industrial inputs, and approximately 1,000 to 6,000 times greater than water concentrations in Lake Erie’s eastern basin, which is more isolated from industrial inputs.

Relative to other IADN sites in the Great Lakes region, PCB continue to be found in the largest amounts at the Chicago IADN sampling site (Buehler and Hites 2002, Sun et al. 2006). Data from this site have been used to represent the whole Chicago urban area. Basu et al. (2004) conducted a short-term ambient air monitoring study at a second site closer to downtown Chicago and compared the results with data from the IADN site, to get an idea of possible differences among locations. Monitoring of vapor and particle-phase PCB in the ambient air at this second site was conducted from February to October 2002. The PCB congener patterns were virtually identical between the two sites. The average PCB concentrations at this second site ranged from 0.7 to 4.6 nanograms per cubic meter ( $\text{ng}/\text{m}^3$ ). These were about two times higher than the IADN site over the same period (0.3 to three  $\text{ng}/\text{m}^3$ ). If PCB loadings to southern Lake Michigan due to the Chicago area were recalculated using information from this second site, they would increase by about 13 kilograms/ year ( $\text{kg}/\text{yr}$ ). This comparison shows that there can be variations of PCB concentrations within urban areas. Further research is necessary to better understand these variations.

Similarly, several new studies found that Chicago continues to be a dominant source of PCB to southern Lake Michigan (Zhang et al. 1999, Green et al. 2000, Offenbergs and Baker 2000, Offenbergs and Baker 2002). Direct deposition of atmospheric PCB from the Chicago area to Lake Michigan occurs quickly relative to the horizontal mixing of the lake's surface waters. This rapid deposition is due to air-water gas exchange and wet and dry deposition that occur when winds carry the urban plume from Chicago over southern Lake Michigan. Green et al. (2000) noted the variability of the influence of Chicago, depending on temperature and wind direction. Offenbergs and Baker (2000) found that southwesterly winds, and associated storms, that cross over Chicago towards southern Lake Michigan coincide with a nearly two-fold increase in PCB concentrations in southern Lake Michigan surface waters. Zhang et al. (1999) estimated that the urban plume from Chicago affects as much as the entire southern quarter of Lake Michigan, while other researchers estimated the affected area to be only five percent or less (Offenbergs and Baker 2002). However, even with the inputs of PCB from Chicago, the PCB concentrations in surface waters of southern Lake Michigan are significantly less than those found in Green Bay, located in northwestern Lake Michigan at the mouth of the Fox River. Green Bay has historically received inputs from 13 paper mills and five major municipal wastewater treatment plants (Totten et al. 2003).

Several studies have examined Lake Ontario and the surrounding environment and have compared the results with Lake Erie and its surrounding environment. An ambient air monitoring study by Chiarenzelli et al. (2001) found PCB concentrations downwind of Lake Ontario, on its southeastern shore, to have higher PCB concentrations (with averages roughly around one  $\text{ng}/\text{m}^3$ ) than a site near Lake Erie, which had concentrations (averaging 0.22  $\text{ng}/\text{m}^3$ ) similar to regional backgrounds as measured by IADN. The samples near the Lake Ontario shore also showed higher chlorination, which ruled out volatilization from the lake as a primary source of the PCB. The researchers hypothesized that volatilization from local sources is a major contributor to the air concentrations. These could be reservoir sources, such as soil, sediment and vegetation from previous deposition, although further research would be needed to determine the exact source (Chiarenzelli et al. 2001).



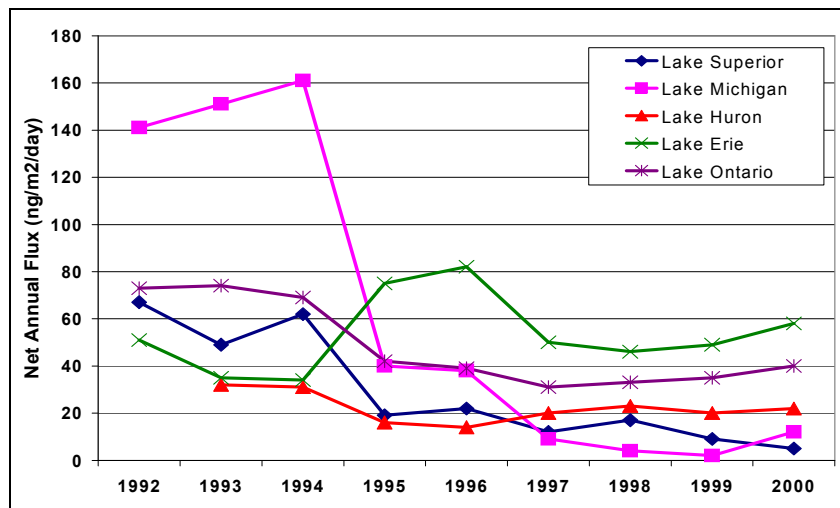
Marvin et al. (2002) and Morrison et al. (2002) found that sediment concentrations in Lake Ontario were greater than those in Lake Erie. Similarly, PCB concentrations in Lake Ontario surface waters were 1.3 to 2.3 times greater than those in the western basin of Lake Erie (which is heavily influenced by Detroit River industrial inputs), and 1,000 to 6,000 times greater than those in the more isolated eastern basin of Lake Erie (Morrison et al. 2002). This trend represents a shift from the 1970s, when concentrations were greater in Lake Erie's eastern basin relative to its western basin. This reversal is likely due to the remediation of several hazardous waste sites located along the eastern shore of Lake Erie, which suggests the success of the Superfund program (Marvin et al. 2002). Following the spatial trend of PCB found in the water column, Morrison et al. (2002) reported that PCB concentrations in biota are greatest in Lake Ontario, followed by western Lake Erie, and then eastern Lake Erie.

One study reported an exception to the land use spatial trend for PCB concentrations. Near Lake Superior, researchers found no clear spatial variation among PCB concentrations in snowpacks sampled in urban and suburban areas near Minneapolis/St. Paul, and those in more remote areas, including Eagle Harbor (on Lake Superior) and northern Minnesota. This observation suggests a well-mixed atmospheric source signal in the region (Franz and Eisenreich 2000).

## **5.0 Air/Water Fluxes**

Figure 5 shows the change in annual net gas exchange flux from 1992 through 2000 for the Great Lakes based on IADN monitoring data. Net gas exchange is the difference between gas absorption from the air to the water and volatilization from water to the air (*see key terms text box in Section 4*). These results indicate that the surface waters of the Great Lakes have been volatilizing PCB into the atmosphere for several years because the rate of volatilization generally decreases with time and approaches equilibrium between air and water (Buehler and Hites 2002, EC and U.S. EPA 2000, EC and U.S. EPA 2002, EC and U.S. EPA 2004). Lake Erie, which may be influenced by PCB sources in New York State, typically has the largest gas exchanges and shows an exceptional trend where the rate of volatilization may be increasing (EC and U.S. EPA 2000, EC and U.S. EPA 2004).

**Figure 5. Net Gas Exchange Flux for the Great Lakes  
(from Surface Water to Air)**



Source: EC and U.S. EPA 2004

The net gas exchange of PCB from the Great Lakes overwhelms the inputs via wet and dry deposition, making the Great Lakes a source of PCB to the atmosphere. Dry particle concentration measurements in IADN ceased after 1995 due to low reported levels. Wet deposition fluxes show no consistent change over time. These fluxes are similar among lakes for which there are recent data (Erie, Superior and Michigan); annual average wet deposition fluxes in these lakes have generally ranged between 1 and 4 ng/m<sup>2</sup>/day over the period from 1992 to 2000 (EC and U.S. EPA 2004). Lake Huron wet deposition fluxes decreased until 1994 when the last PCB precipitation measurements were taken there (EC and U.S. EPA 2000).

Researchers recently re-evaluated gaseous fluxes of PCBs between water and air from two locations on Lake Michigan: Green Bay (located in northwestern Lake Michigan), and southern Lake Michigan (near the urban/industrial areas of Chicago, Illinois and Gary, Indiana). They found that net volatilization fluxes are more important for the removal of PCB from these waters than previously recognized. Due to historically elevated concentrations in the waters of Green Bay, the net volatilization flux from Green Bay is greater than the flux reported for Lake Michigan and the other Great Lakes. Totten et al. (2003) updated previous calculations with improved Henry's law constant<sup>3</sup> estimates for PCB (Bamford et al. 2000) and new relationships for estimating mass-transfer rates across the air-water interface. They also suggested that the magnitude of the air-water gaseous exchange flux is a stronger function of daily meteorological conditions than previously recognized.

<sup>3</sup> Henry's law states that the amount of a gas that will be absorbed by water increases as the gas pressure increases. Henry's law constant is frequently used to define the distribution of a chemical across the air/water interface.

The resulting new calculations indicate that the net PCB volatilization flux from Green Bay is two to 20 times higher than had been estimated previously. In addition, the new evaluations indicate that net volatilization was the dominant loss process in both summer and winter from southern Lake Michigan during the summer of 1994 and January 1995. These results contrast with previous studies that calculated absorption of PCB by surface waters in the summer 1994, when air concentrations above southern Lake Michigan were elevated because of westerly winds blowing from the Chicago, Illinois, and Gary, Indiana, urban/industrial areas (Offenberg and Baker 2000, Zhang et al. 1999). However, due to the influence of local inputs from Chicago, wet deposition and dry particle deposition to southern Lake Michigan continues to outweigh volatilization. Volatilization and sediment burial are roughly equal in importance in removing PCB from southern Lake Michigan waters (Totten et al. 2003).

### **Scavenging as a Removal Mechanism**

Scavenging mechanisms for removing PCB from the atmosphere include gas exchange with surface waters, dry deposition to surface waters, and precipitation scavenging, also known as washout. Gaseous PCB can dissolve into water both within clouds and into falling precipitation. Similarly, PCB particulates can be melted into falling precipitation. In either form (gaseous or particle), PCB can be essentially washed into another medium, e.g., a lake, from the atmosphere. Offenberg and Baker (2002) studied this phenomenon around southern Lake Michigan.

The study was conducted to measure the effectiveness and relative importance of precipitation scavenging mechanisms during a variety of storm events, with measuring stations located at urban locations, over water, and following a downwind transect. Offenberg and Baker (2002) concluded that washout mechanisms are largely variable between storm events, and the relative importance of each removal mechanism generally does not vary greatly along a storm's path. Therefore, the storm type (i.e., meteorological and precipitation characteristics) controls precipitation scavenging, where particle scavenging, rather than gas scavenging, dominates washout mechanisms for PCB in a wide range of precipitation events. Van Ry et al. (2002) further confirm that, on average, 97 percent of total atmospheric washout is from particle scavenging at a variety of sites in the Mid-Atlantic States.

Table 2 shows the direction and magnitude of several reported net gas exchanges at the air-water interface for select Great Waters.

**Table 2. PCB Net Gas Exchange from Select Great Waters**

<b>Waterbody</b>	<b>Year and Period</b>	<b>Net Gas Exchange (ng/m<sup>2</sup>/day)</b>	<b>Source</b>
NY Harbor	1998 July	-2100	Totten et al. 2001
Green Bay, WI	1989 June through October average	-1200	Totten et al. 2003
Raritan Bay, NJ	1998 July	-400	Totten et al. 2001
Galveston Bay, TX	1995-96 annual	-195	Park et al. 2001
Corpus Christi Bay, TX	1998-99 annual	-184	Park et al. 2002
Chesapeake Bay	1992 annual	-96	Totten et al. 2003 <sup>a</sup>
Lake Erie (Sturgeon Point)	2000 annual	-58	EC and U.S. EPA 2004
Lake Ontario (Point Petre)	2000 annual	-40	EC and U.S. EPA 2004
Lake Huron (Burnt Island)	2000 annual	-22	EC and U.S. EPA 2004
Lake Michigan (Sleeping Bear Dunes)	2000 annual	-12	EC and U.S. EPA 2004
Lake Superior (Eagle Harbor)	2000 annual	-5	EC and U.S. EPA 2004
Northern San Francisco Estuary	2000 June through November	-2.2 to -24	Tsai et al. 2002

Note: Negative net gas exchanges indicate loss of concentration in surface water (i.e., volatilization).

<sup>a</sup> From Nelson et al. 1998

NY Harbor, Green Bay and Raritan Bay, with the highest net volatilization rates, have all been impacted historically by industrial contamination (Totten et al. 2003). The Great Lakes net volatilization exchange is less than that for both Corpus Christi Bay and Galveston Bay, Texas, possibly due to the higher average surface water temperatures in the southern estuaries relative to the northern lakes (Park et al. 2002).

Wet deposition fluxes reported for Chesapeake Bay, Galveston Bay, Lake Superior, and Green Bay (Lake Michigan) were all similar (Park et al. 2001). Park et al. (2001) also found the dry deposition flux at Galveston Bay to be two-fold higher than the rate for Chesapeake Bay. PCB concentrations in air and rain samples collected at Corpus Christi Bay were four times lower than those found at Galveston Bay, but net volatilization fluxes were similar at both locations. For northern San Francisco Estuary, a combined flux of gaseous and particulate dry deposition PCB showed a net release of PCB from the water to the atmosphere, during a six-month study (June to

November) in 2000. The monthly fluxes ranged from 0.15 to 23 ng/m<sup>2</sup>/day; the highest volatilization rates occurred in the summer months. Wet deposition was not included in this study (Tsai et al. 2002). The studies referenced in Table 2 all note that net gas exchange is the dominant mode of transfer of PCB between the atmosphere and surface water. Therefore, the net flux is volatilization of PCB from the waterbody to the atmosphere.

Deposition fluxes were estimated for multiple sites across New Jersey through the New Jersey Atmospheric Deposition Network (NJADN). The network, which operated roughly between 1998 and 2001, included sites in urban, suburban, coastal and forested areas. The annual deposition fluxes (not accounting for volatilization) ranged from 20 to 931 ng/m<sup>2</sup>/day, with the highest values in the urban areas. For the sites near the Hudson River Estuary, which is adjacent to urban areas, these fluxes are at least two to 10 times those estimated for the Chesapeake Bay and Lake Michigan. For low molecular weight PCB (congeners containing two to five chlorines), volatilization from the Hudson River Estuary exceeds atmospheric deposition (Reinfelder et al. 2004, Totten et al. 2004).

## **6.0 Emissions and Sources**

### ***6.1 Emissions***

Numerous new studies reported on the sources of PCB to the environment. As noted earlier, PCB are no longer manufactured, and their use and handling are regulated under the Toxics Substances and Control Act (TSCA). Nevertheless, because PCB were widely used at one time, volatilization from or combustion of materials containing PCB can be sources to the atmosphere. Areas of past PCB contamination can also be sources. As noted in the previous section, many of the Great Waters themselves are sources of PCB to the atmosphere. Consequently, estimating PCB emissions to the atmosphere is challenging and can have a high degree of uncertainty.

The National Emissions Inventory (NEI) is a national database of air emissions information with input from numerous state and local air agencies, from tribes, and from industry. A list of the source categories that contribute to the 26 tons per year of PCB air emissions estimated for 2002 is provided in Appendix A. Open burning of residential household waste, is the largest contributor in the 2002 NEI (estimated to be 23 tons per year nationally) (U.S. EPA 2007b). However, this activity is not likely to represent the largest source of emissions in urban areas. The text box in this section highlights studies designed to better understand urban sources, specifically in Chicago.

### ***6.2 Source areas affecting the Great Lakes***

The Great Lakes continue to be impacted by atmospheric deposition and industrial point and nonpoint sources. There are no new PCB sources reported for Lake Erie. The Detroit River continues to be the dominant contributor of PCB to Lake Erie (approximately 75 percent), especially in the western basin and near shore areas along the Ohio shoreline (Morrison et al. 2002); the Clinton, Maumee, and Cuyahoga Rivers also contribute to PCB loadings in Lake Erie. Lake Erie also receives atmospheric inputs of PCB from the urban areas of Toledo, Cleveland, and Ashtabula, Ohio (Marvin et al. 2002). In contrast to Lakes Superior and Michigan, local

contaminant sources (primarily hazardous waste facilities on the Niagara River) play a greater role in Lake Ontario PCB loadings than does atmospheric deposition (Marvin et al. 2002). Franz and Eisenreich (2000) reported that falling snow, rather than dry deposition, is the dominant source of PCB to snowpacks; if gaseous PCB are sorbed to snow, it likely happens during snowfall. These researchers estimate that 30 to 40 percent of snowpack meltwater from the Lake Superior basin enters Lake Superior during a few weeks in the spring.

Hafner and Hites (2003) examined potential source regions for PCB measured at several of the Great Lakes IADN monitoring sites. Their analysis used the Potential Source Contribution Function (PSCF) Model, which is a probabilistic back-trajectory modeling technique. Sturgeon Point, on Lake Erie, is the most eastern of the sites, and appears to be strongly influenced by the heavily urban east coast. Sleeping Bear Dunes, on Lake Michigan, appears to be influenced by areas to the south and southwest; there are virtually no indications of influence from the north. The greater Chicago area appears to be the most influential region for Eagle Harbor, on Lake Superior. In later work, Hafner and Hites (2005) did further modeling analysis of the Great Lakes region. 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 PCB, the information derived from the models was not improved substantially by including a source direction factor.

### **Hybrid Receptor Models Indicate Chicago PCB Source Areas**

Two new studies using three types of hybrid receptor models and verified by field samples indicate a large potential atmospheric PCB source area southwest of Chicago between Kankakee and Joliet, Illinois, and two moderate PCB sources to the northwest in the direction of Madison, Wisconsin, and south of Chicago in the Lake Calumet neighborhood (Hsu et al. 2003a, Hsu et al. 2003b). Gary, Indiana, is also a potential PCB source to the air. The indication of sources to the south and southwest is consistent with higher temperatures associated with winds from this direction causing volatilization of previously deposited PCB (Hsu et al. 2003b). Specifically, a large transformer storage yard, municipal sludge drying beds, and a large landfill in Chicago were confirmed as contributors of atmospheric PCB. At the landfill, PCB may escape from either landfill gas or the wastewater sludge that is used as a cover. Other potential sources identified include the Chicago Confined Disposal Facility, which contains dredged sediment contaminated with PCB, and the Indiana Harbor and ship canal, which are in a highly industrial area with steel manufacturing and petroleum refineries and which contain some of the most highly contaminated sediments in Lake Michigan (Hsu et al. 2003b). Lake Michigan is not an atmospheric source to Chicago (Hsu et al. 2003a).

Modeling results were relatively consistent between the models; however, no single model provided information as complete as that obtained by using all three in conjunction (Hsu et al. 2003a). It should also be noted that many small sources not identified in these studies may collectively contribute a larger amount of PCB to the Chicago air (Hsu et al. 2003b). In turn, modeling results reported by the United States - Canada IADN Scientific Steering Committee (EC and U.S. EPA 2002) cite the urban area of Chicago and northwestern Indiana as a source of PCB to Lake Superior.

Although the majority of PCB in the Great Lakes atmosphere come from North American sources, analyses using the Lawrence Berkeley National Laboratory Berkeley-Trent (BETR) Global model estimates fractions of loadings from other areas of the world. A comparative analysis of cumulative historical emissions to emissions in 2002 indicates that relative contributions from global sources other than North America are increasing, as North American sources decrease. Specifically, Eastern Europe appears to be emerging as a relatively more important source of PCB deposition in the Great Lakes. However, there is a wide range of uncertainty associated with this assessment mainly due to uncertainty in emission estimates (U.S. EPA and EC 2005).

### ***6.3 Source areas affecting other Great Waters***

In the coastal New Jersey region, a study by Brunciak et al. (2001) concludes that atmospheric PCB derive from a dominant source type/area and process(es), possibly water treatment plants. In addition, volatilization from the Hudson River and surrounding rivers contributes PCB to the atmosphere. At individual sites, temperature, wind direction and speed, and distance from the source(s) determine the absolute concentration of PCB. In the Baltimore, Maryland area, a study conducted within the Chesapeake Bay watershed hypothesized that building materials used prior to 1980, paints and dielectric fluids used in electrical transformers and capacitors prior to the early 1970s, and a medical waste incinerator were potential sources of PCB found in films on exterior window surfaces (Liu et al. 2003). Precipitation readily removes the film, which may subsequently wash into nearby surface waters and eventually the Chesapeake Bay.

In the Corpus Christi Bay and Galveston Bay regions, as noted in section 5, new studies indicate that these surface waters are currently acting as sources of PCB to the atmosphere via net volatilization flux of vapor PCB from the water surface (Park et al. 2002, Park et al. 2001). Wind speed, wind direction, and air temperature affect local PCB concentrations in air and rain samples at both bays. Winds deriving from the Gulf of Mexico tend to exhibit a dilution effect on local atmospheric PCB concentrations, and winds from urban/industrial areas tend to increase local atmospheric PCB concentrations. Lower wind speeds were correlated with increased PCB concentrations in air, as were higher air temperatures. At Galveston Bay, concentrations of dissolved and particulate PCB also showed a weak dilution effect based on the volume of rain (Park et al. 2001). Regional airports may contribute PCB to Corpus Christi Bay (Park et al. 2002).

In the San Francisco Bay, Johnson et al. (2000) studied the parameters of PCB sorbed to particulates in the surface waters to better understand the sources of PCB in this area. Sediment resuspension is suspected to be the dominant source for three out of five PCB analytes studied because the highest levels were observed in the winter and spring when there was high freshwater inflow and high total suspended solids. The source of the fourth PCB analyte is suspected to be due to a restricted release or a spill and the fifth PCB analyte from either the atmosphere (because similar patterns have been observed in ambient samples of marine and urban air) or sewage inputs.



## 7.0 Summary and Some Future Directions in Research

Generally, PCB concentrations are declining or reaching steady-state in the environment (air, water, sediment, biota) surrounding the Great Waters, resulting in observed improvements to ecosystem health in most cases. Nevertheless, PCB concentrations in biota continue to be of concern for wildlife and humans, leading to fish consumption advisories in many of the Great Waters.

The PCB concentration in fish and fish-eating birds in both the Great Lakes and southern New Jersey are lower than in the 1970s, and populations of these birds in both areas have increased substantially. Yet, in the Great Lakes, PCB concentrations in top predator fish exceed thresholds established for fish-consuming birds and wildlife, and there are reaches of shoreline where the bald eagle population has not recovered.

State and tribal advisories for human consumption of fish were in effect in 2004 for all the Great Lakes, Lake Champlain, coastal waters of the northeastern states, Chesapeake Bay and its tributaries, San Francisco Bay and Puget Sound. Data from NOAA's Mussel Watch program indicated that the median concentration in mollusks nationally is decreasing. However, most individual Mussel Watch sites show no trend, including the majority of sites in NERR locations.

Several new studies evaluated spatial trends in PCB concentrations for various environmental media. Consistent with findings discussed in the Third Report to Congress, higher concentrations tended to be found in urban and industrial areas than in remote locations. Recent studies related to Corpus Christi, Galveston, and San Francisco Bays, New Jersey and the mid-Atlantic region provided information related to PCB air concentrations and air-water fluxes in Great Waters for which there previously were no data.

Similar to results identified in the Third Report to Congress, the new studies indicated that in many Great Waters, volatilization of PCB from the surface water to the air is taking place at a higher rate on average than PCB are entering the water from the air via wet and dry deposition and gas absorption, making these waterbodies sources of PCB to the atmosphere. However, deposition inputs from sources near Chicago outweigh volatilization in southern Lake Michigan.

Emission sources of PCB include volatilization from or combustion of material containing PCB. The National Emissions Inventory cites open burning of residential household waste as a major contributor nationally. A study in Chicago found that emission sources there included a transformer storage yard, municipal sludge drying beds, and a landfill, among others.

Recent studies considered source areas affecting waterbodies in various geographic areas. Inputs examined included rivers, suspended sediments, discharges, and global sources, in addition to domestic emission sources.

Several of the articles in the recent literature recommend continued research to improve knowledge related to PCB trends, and sources, fate, and transport. Investigators' specific recommendations for some future research are as follows:

- Research is needed to determine why there are higher airborne PCB concentrations and chlorination east of Lake Ontario compared to IADN sites throughout the Great Lakes Basin (Chiarenzelli et al. 2001).
- Volatilization from surface water is an important removal process of PCB from Corpus Christi Bay and Galveston 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; Park et al. 2001).
- Concentrations of PCB in water and sediment naturally change over time; therefore it is necessary to further research the fate and transport of PCB in the ecosystem so that effective measures can be taken to reduce the resulting contaminant burdens of aquatic biota (Morrison et al. 2002).
- Although the current IADN is extremely useful, it must continue to grow and adapt to emerging concerns in the region. Further research is still necessary to pinpoint potential sources of pollution to the Great Lakes using IADN meteorological and concentration data, and to develop new urban study sites (Buehler and Hites 2002). Long-term monitoring (i.e., decades of monitoring) is also necessary to fully understand how PCB compounds degrade in the environment (Buehler et al. 2002).
- Research is needed to address PCB sources in a global setting. In particular, further research is necessary in order to accurately characterize emissions in the global setting (U.S. EPA and EC 2005).

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## **Appendix A. Detailed Breakdown of Air Emissions Inventory Source Categories Emitting Polychlorinated Biphenyl**

Chemical Preparations <sup>a</sup>	Petroleum Lubricating Oil and Grease Manufacturing
Commercial and Industrial Solid Waste Incineration <sup>a</sup>	Plastic Materials and Resins Manufacturing <sup>a</sup>
Gasoline Distribution (Stage I) <sup>a</sup>	Polyethylene Terephthalate Production <sup>a</sup>
Incineration: On-site Commercial/Institutional and Industrial	Portland Cement Manufacturing <sup>a</sup>
Incineration: Commercial and Industrial Solid Waste	Printing, Coating, and Dyeing of Fabrics <sup>a</sup>
Industrial/Commercial/Institutional Boilers & Process Heaters <sup>a</sup>	Residential Heating: Wood/Wood Residue Combustion
Inorganic Pigments Manufacturing <sup>a</sup>	Secondary Aluminum Production <sup>a</sup>
Landfills – Commercial/Institutional and Industrial	Sewage Sludge Incineration <sup>a</sup>
Medical Waste Incinerators <sup>a</sup>	Solvent Use: Solvent Evaporation: Cold solvent Cleaning
Mineral Products: Abrasive Product Manufacturing	Solvent Use: Surface Coating: Auto Refinishing and Textile Products
Mining: Nonmetallic Mineral	Site Remediation <sup>a</sup>
Municipal Landfills <sup>a</sup>	Stationary Reciprocating Internal Combustion Engines <sup>a</sup>
Open Burning – Residential, Household Waste	Utility Boilers: Coal and Wood or Waste
Open Burning – Structure Fires	Waste Disposal, Treatment, and Recovery
Other Solid Waste Incineration <sup>a</sup>	Waste Disposal: Industrial

<sup>a</sup>These source categories are associated with regulations under the Clean Air Act for stationary sources known as maximum achievable control technology (MACT) rules.

Source: U.S. EPA 2007b

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