

# **Polychlorinated Biphenyl Emissions to Urban Atmospheres: Enhanced Concentrations, Atmospheric Dynamics and Controlling Processes**

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**October, 2000**

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## **Funding Agencies:**

**Hudson River Foundation**

**D. Suszkowski**

**New Jersey Sea Grant (NOAA)**

**M. Weinstein**

**New Jersey Department of Environmental Protection**

**L. McGeorge, S. Nagourney, M. Aucott**

**US Environmental Protection Agency**

**A. Hoffman, D. Pahl, G. Foley, G. Evans**

## **Polychlorinated Biphenyl Emissions in Urban Atmospheres: Enhanced Concentrations, Atmospheric Dynamics and Controlling Processes**

**By Steven J. Eisenreich**

### **Abstract**

Regions of major urban-industrial activity emit hazardous air pollutants (HAPs) into the atmosphere at sufficient rates to yield significantly elevated concentrations relative to regional background. Elevated urban atmospheric concentrations lead to transport at the local and regional scale and to deposition to regional lakes, estuaries and landscapes. The PCB concentrations in the urban atmosphere range from perhaps 100-300 pg m<sup>-3</sup> in winter to 5000 to 16000 pg m<sup>-3</sup> on hot summer days. Examples from the cities of Chicago, Baltimore, Jersey City and Camden-Philadelphia are presented. The atmospheric PCB concentrations are controlled by air (probably surface) temperature and the size of the mobile environmental reservoir (MER). The size of the MER and the emission dynamics must be established for areas such as Chicago using ambient temperature as a forcing function. The 'footprint' of elevated gaseous PCB concentrations down gradient of source areas may be on the order of 5- 60 km, although much larger 'footprints' are expected under sustained winds blowing from the urban-industrial region across wide expanses of water. Atmospheric PCBs are lost in the urban atmosphere in the summer time due to OH radical attack. Evidence exists that this process is important at local scale in contaminated areas and at the continental scale and global scale. Recognizing that old urban-industrial areas exhibit high atmospheric concentrations especially in the warm summer time, that this reflects strong local emissions, and that there is ample evidence that urban-industrial emissions of PCBs into the local atmosphere results in enhanced atmospheric signals and deposition, the PCB sources must be identified and limited, controlled and/or eliminated. Given that the influence of temperature on atmospheric PCB concentrations is more pronounced in urban complexes, then the size of the MER must be substantial.

**Recommendation 1.** *Stockpiles of PCB (Arochlor oils) must be identified, collected and disposed properly before they become accessible to environmental exchange processes. This may require 'buy-back' of PCB stockpiles without concern for liability or penalties.*

**Recommendation 2.** *PCB source and/or emission inventories must be constructed recognizing that normal regulatory protocols do not function for banned and widely-distributed chemicals that cycle constantly in the environment.*

**Recommendation 3.** *Renovation or destruction of existing old buildings in urban-industrial areas should be performed in such a way as to greatly limit PCB emissions.*

**Recommendation 4.** *Quantify the rate of gaseous PCB discharges to the atmosphere from sanitary landfills and control emissions if above agreed-upon rates.*

### **Introduction**

Wet deposition via rain and snow, dry deposition of fine/coarse particles, and gaseous air-water exchange are the major atmospheric pathways for persistent organic pollutant (POP) input to the Great Waters such as the Great Lakes, Chesapeake Bay, the Hudson-

River Harbor Estuary, and coastal waters. The Integrated Atmospheric Deposition Network (IADN) operating in the Great Lakes and the Chesapeake Bay Atmospheric Deposition Study (CBADS) formerly operating in Chesapeake Bay were designed to capture the *regional* atmospheric signal, and thus sites were located in background areas away from local sources. However, many urban/industrial centers are located on or near coastal estuaries (e.g., Hudson River Estuary and NY Bight; Chesapeake Bay) and the Great Lakes (e.g., southern Lake Michigan proximate to Chicago). Emissions of pollutants into the urban atmosphere are reflected in elevated local and regional pollutant concentrations and localized intense atmospheric deposition that is *not* observed in the regional signal. The southern basin of Lake Michigan and northern Chesapeake Bay, as two such locations, are subject to contamination by air pollutants such as polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs), Hg and trace metals because of its proximity to industrialized and urbanized Chicago, IL and Baltimore, MD, respectively. Concentrations of PCBs and PAHs are significantly elevated in Chicago and coastal Lake Michigan and in the air of Baltimore and nearby Chesapeake Bay as compared to the regional signal (Offenberg and Baker, 1997, 1999; Simcik et al., 1997; Franz et al., 1998; Zhang et al., 1998; Dachs and Eisenreich, 2000; Brunciak et al., 2000).

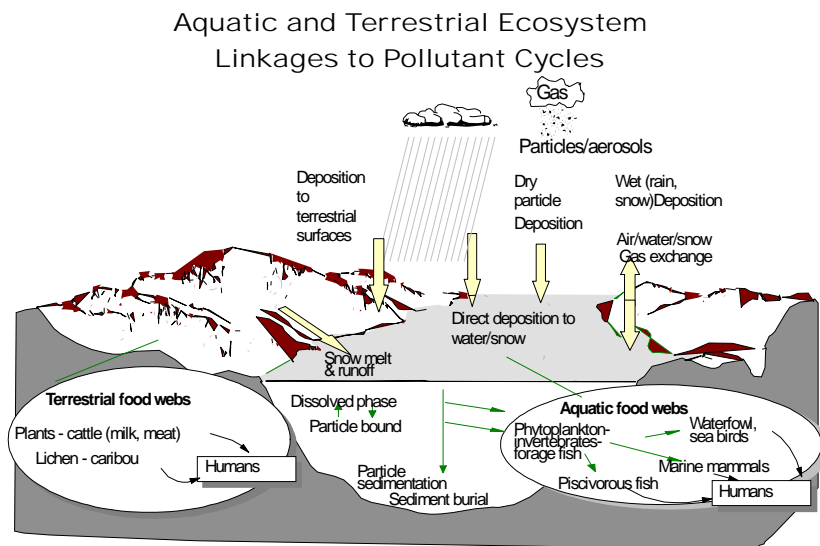


Figure 1. Pathways of Contaminant Emissions and Deposition to the Great Waters.

Higher atmospheric concentrations of pollutants are ultimately reflected in increased precipitation and dry particle fluxes of contaminants to the lake/estuarine waters as well as enhanced air-water exchange fluxes of organic compounds such as PCBs (Zhang et al., 1998; Miller et al., 2000) and PAHs. Of course, the contribution of mobilized in-place pollutants, discharges from wastewater treatment facilities and agricultural runoff, and upstream riverine and tributary flow to overall water concentrations and loads must be evaluated to determine the relative importance of the atmospheric pathway.

Processes of wet and dry deposition and air-water exchange of atmospheric pollutants reflect loading directly to the water surface (Figure 1). This is especially important for aquatic systems that have large surface areas relative to watershed areas (e.g., Great Lakes; coastal seas), as well as to estuaries near major sources. Also, water bodies may be sources of contaminants to the local and regional atmosphere representing losses to the water column and inputs to the local atmosphere of the Great Lakes and Chesapeake Bay (Hornbuckle et al., 1994, 1995; Zhang et al., 1998; Nelson et al., 1998; Bamford et al., 1999). This has also been demonstrated in the NY/NJ Harbor Estuary for PCBs, PAHs and nonylphenols. However many aquatic systems have large watershed to lake/estuary areas emphasizing the importance of atmospheric deposition to the watershed (forest, grasslands, crops, paved areas, and wetlands) and subsequent leakage of deposited contaminants to the downstream water body (Figure 1). Most lakes and estuaries in the Mid-Atlantic States have large watershed/water area ratios emphasizing the potential importance of atmospheric pollutant loading to the watershed and subsequent release to rivers, lakes and estuaries. Direct deposition to Lake Michigan from the proximate urban-industrial area of Chicago, IL – Gary, IN is believed to be a dominant contributor to the load of many contaminants.

This document focuses on the emission of PCBs into the urban-industrial atmosphere yielding enhanced concentrations, transport and deposition. Three research programs will provide the data to demonstrate these phenomena: The AEOLUS Study conducted in 1994-98, its daughter, the New Jersey Atmospheric Deposition Network in the Mid-Atlantic States, and the IADN. The hypothesis of AEOLUS (Atmospheric Exchange Over Lakes and Oceans Study) is that emissions of hazardous air pollutants into the urban atmosphere increase atmospheric depositional fluxes to proximate Great Waters, focusing on southern Lake Michigan near Chicago, IL during the Lake Michigan Mass Balance Study (LMMBS) and northern Chesapeake Bay near Baltimore. The research effort is derived from Section 112m of the 1990 Clean Air Act Amendments that mandated that the US EPA conduct an investigation of atmospheric depositional fluxes of contaminants from urban areas to the Great Waters and to identify their source(s) and strengths. The objectives of this research were to determine the magnitude of wet, dry and air-water exchange fluxes of PCBs to Lake Michigan as impacted by the Chicago and Baltimore air plumes, to quantify the urban influence on annual and spatial inputs, and to assess resulting concentrations and accumulations on an impacted Lake Michigan and Chesapeake Bay. The strategy of this research (e.g., Simcik et al., 1997; Offenberg and Baker 1997;) was to make simultaneous air and water measurements over the lake and at shore source areas during seasonal intensive campaigns within the southern basin. The measured air and rain concentrations over the lake and land are combined with meteorological conditions to estimate the annual cycle of atmospheric PCBs over the basin. Finally, the data on emissions, atmospheric concentrations, transformations and deposition are synthesized into a quantitative evaluation of the urban plume's impact on Lake Michigan. The principal mode to accomplish this is to quantify the urban influence on the PCB mass budget in Lake Michigan.

The New Jersey Atmospheric Deposition Network (NJADN) was established in October 1997 (1) to support the atmospheric deposition component of the NY/NJ Harbor Estuary

Program; (2) to support the Statewide Watershed Management Framework and the National Environmental Performance Partnership System (NEPPS) for New Jersey; (3) to assess the magnitude of toxic chemical deposition throughout the State; and (4) to assess in-state versus out-of-state sources of air toxic deposition. The NJADN design is based on the well-developed experience in the Great Lakes and Chesapeake Bay, and reflects a combination of systematic measurements in 9 site well-developed network, and intensive field campaigns to address key research questions such as air-water exchange and air-terrestrial exchange, and source identification. The NJADN is a *research and monitoring* network designed to provide scientific input to the management of the various affected aquatic and terrestrial resources. This report will also rely on the excellent data set coming from the IADN courtesy of Drs. I. Basu and R. Hites (Indiana University).

The objectives of this report are to demonstrate (1) the elevated atmospheric PCB concentrations in the urban atmosphere compared to sites over water or land nearby; (2) the dramatic influence of air and surface temperatures in mobilizing environmental reservoirs of PCBs; (3) the spatial 'footprint' of PCBs emitted in the urban areas of Chicago and Camden-Philadelphia; (4) key processes controlling downwind transport and deposition such as air-water exchange and OH-radical attack on gas-phase PCBs; and (5) to make recommendations on major questions and actions.

### **Enhanced PCB Concentrations in Urban Atmospheres**

PCB concentrations in the urban-industrial atmospheres of Chicago, Baltimore, Camden/Philadelphia and the NY-NJ corridor are elevated over sites 10 to 50 km distant by factors of 5 to 20 (Table 1). These polluted atmospheres exhibit PCB gas-phase concentrations in cold months that are only 2-3x background concentrations (100 to 300 pg/m<sup>3</sup>) but increase in the summer months to as high as 5000 to 16000 pg/m<sup>3</sup>. The highest PCB concentrations occur in the urban atmosphere on the hottest days under low wind speeds reflecting the role of temperature in driving air-surface exchange in accessing the large mobile environmental reservoir (MER) in the urban areas. The concentrations of gas-phase PCBs decrease with distance from the urban-industrial source area although the functional form of that decrease is unknown.

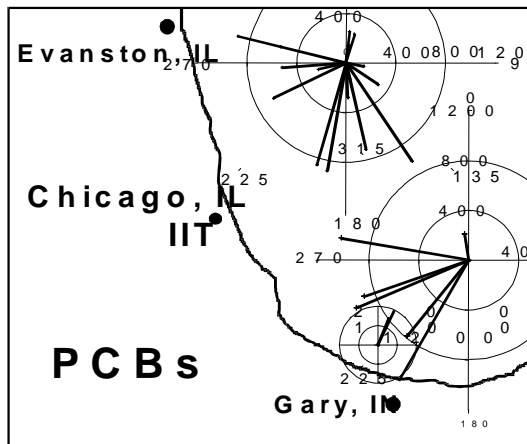
Figure 6 shows that  $\Sigma$ PCB concentrations over southern Lake Michigan are highly dependent on wind direction (Simcik et al., 1997). The highest air concentrations occurred when the wind was from a direction of the urban-industrial sector inclusive of the shoreline from Evanston, IL to Gary, IN, and they were low when the winds came from any other direction. On average, over-water concentrations were enhanced 4x over regional background values. This pattern is observed in the Baltimore-Chesapeake Bay

Table 1. Enhanced Urban Concentrations of Atmospheric PCBs

		$\Sigma$ PCBs (pg/m <sup>3</sup> )	
Site	Urban	Over-Water or Background	References
	Range	Range	
Chicago-AEOLOS '94-95	270 - 14000	130 – 1200 70 - 800	Simcik et al., 1997 Zhang et al., 1998
Chicago-LMMB '94-95	500 - 6800	100 – 500 (Sleeping Bear Dunes)	Miller et al. 2000 (In Review)
Chicago-LMMB '94-95	450 – 4600 (modeled)	350 – 3200 (modeled)	Green et al. 2000
Chicago-IADN 1998 1999	460 – 6800 335 - 7000		Hites and Basu, Unpubl. data
Baltimore-AEOLOS-06-96	380 - 3360	210 - 740	Offenberg and Baker, 1999
Baltimore-AEOLOS-06-97	760 - 2280	290 - 990	Brunciak et al., 2000
NY-NJ Area '97-00 (NJADN)	100 – 3300 (Jersey City)	60 – 2340 (Suburban) 90 – 1600 (Coastal)	Brunciak et al., 2000
Camden/Philadelphia '99-00 (NJADN)	1020 - 16000	45 – 550 (Pinelands) 200 – 540 (Washington's Crossing)	VanRy and Eisenreich Unpubl. data

(Offenberg and Baker, 1999; Brunciak et al., 2000), and the NY-NJ Harbor Estuary (Brunciak et al., 2000). PCB emissions in the urban atmosphere result in elevated concentrations down wind.

Let us assume that the bulk of the PCB emissions from the urban landscapes of Chicago, Baltimore and Camden-Philadelphia into the local atmosphere are focused in a 5 x 5 km area with an atmospheric mixing height of 1 km. Then the inventory of atmospheric  $\Sigma$ PCBs at 1 ng/m<sup>3</sup> concentration is 1 ug/m<sup>2</sup> or 25 g in the 25-km<sup>3</sup> atmosphere. If the concentration is 10 ng/m<sup>3</sup>, then the inventory is 10 ug/m<sup>2</sup> or 250 g. Daily emission rates of perhaps 1 to 10 ug/m<sup>2</sup> d are necessary to achieve the observed atmospheric concentrations assuming transport into the grid is negligible. These emission rates may

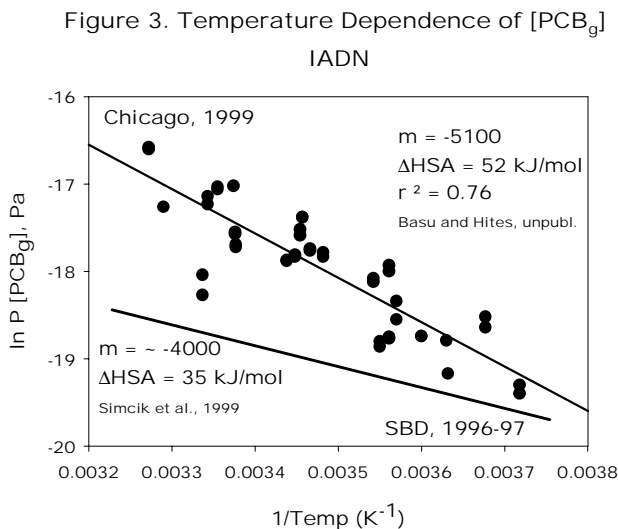


**Figure 2.** Concentrations of  $\Sigma$ PCBs ( $\text{pg}/\text{m}^3$ ) over southern Lake Michigan generally downwind of Chicago during AEOLUS in 1994 (Simcik et al., 1997).

be compared to atmospheric deposition rates to Lake Michigan of perhaps  $0.05$  to  $0.15 \text{ ug}/\text{m}^2 \text{ d}$  and PCB accumulation rates in lake sediments of  $0.05$  to  $0.1 \text{ ug}/\text{m}^2 \text{ d}$ . Thus PCB mass in the MER of the urban-industrial center of tonnes can support emission fluxes for many decades into the future.

**Temperature Control on Atmospheric PCB Concentrations and Emissions**

Atmospheric concentrations of PCBs at any site are dependent upon concentrations in air masses advecting into the area, the magnitude of surface contamination, and the temperature driving the magnitude of air-surface exchange. This phenomenon is frequently expressed in a modified Clausius-Clapyron equation of the form:  $\ln P = m/T + b$ , where  $P$  is the partial pressure of the gas in the atmosphere (Pa),  $T$  is temperature (K), and  $b$  is the intercept. Thus a plot of  $\ln [\text{PCB}]$  vs.  $1/T$  yields a straight line with slope equal to  $m$  ( $= \Delta H_{SA}/R$ , where  $\Delta H_{SA}$  is the pseudo-enthalpy of air-surface exchange, and  $R$  is the universal gas constant). Figure 3 shows a typical plot of gaseous  $\Sigma$ PCB



concentrations at the urban Chicago IADN site in 1999 (Basu and Hites, unpubl.) and similar behavior at the remote Sleeping Bear Dunes site in the northern basin of Lake Michigan in 1996-97 (Simcik et al., 1999). The observations here and elsewhere state that higher temperatures generate higher atmospheric PCB concentrations, and the higher concentrations are observed in areas with higher surface contamination. As supported by Wania et al. (1998), the slope and therefore the apparent energy of air-surface exchange is driven by the local exchange processes, advection of PCBs in incoming air, and the size of the local mobile environmental reservoir. Although Hoff et al. (1998), Wania et al. (1998), and Simcik et al. (1999) differ in the details of the mechanistic explanation, it is clear that high temperatures in contaminated areas yield high atmospheric concentrations, whose inventory is then transported away from the urban area to proximate water bodies and terrestrial landscapes where deposition processes load clean environments. It should be possible to derive a relationship of temperature and atmospheric concentrations in urban areas to derive the magnitude of the mobile environmental reservoir. We are actively pursuing this issue in the NJ-NY and

Table 2. Temperature-Dependence of PCB Partial Pressure in the Atmosphere Derived from the slope (m) of the  $\ln Pa$  versus  $1/T(K)$  expressions. The slope (m) =  $\Delta H_{SA} / R$  Other values cited in Wania et al., 1998.

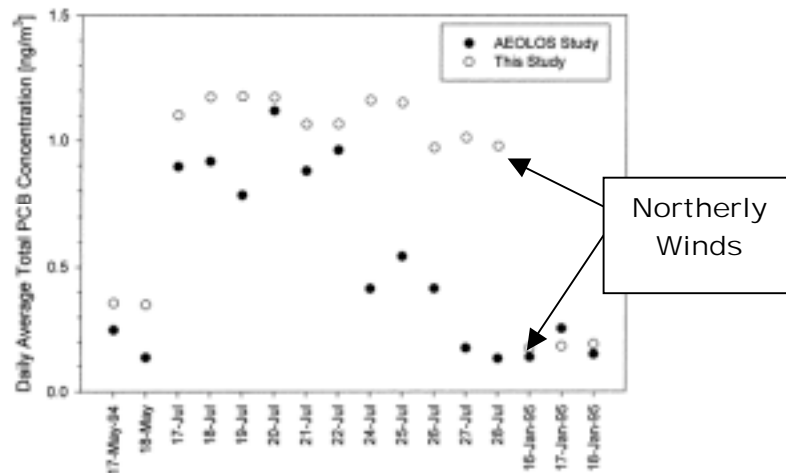
Location	Slope, m	Reference
<b>URBAN/INDUSTRIAL</b>		
Bloomington, IN	-6700 to -7800	Hermanson and Hites, 1989 Panshin and Hites, 1994 Wallace and Hites, 1996
Chicago, IL	~ -6000 (1996-97)	Simcik et al., 1999 IADN
Chicago, IL	~ 5100 (1999)	Basu and Hites, unpubl. IADN
Jersey City, NJ	~ -6200	Brunciak et al., 2000 NJADN
Lista, Norway	-6700	Haugan et al., 1999
Camden/Philadelphia	~ -6500	VanRy and Eisenreich, unpub.; NJADN
<b>BACKGROUND/REMOTE</b>		
Great Lakes Superior, Michigan, Erie	~ -4500 ~ -3800 to -4600	Hillery et al., 1997 Simcik et al., 1999
Suburban, NJ	~ - 4400	Brunciak et al., 2000 NJADN
Marcel, MN	~ - 2000	Hornbuckle and Eisenreich, 1996
United Kingdom	~ -4300	Lee and Jones, 1999
Pinelands, NJ	~ -4500	VanRy and Eisenreich, unpubl.; NJADN

Philadelphia/Camden areas where atmospheric PCB concentrations are among the highest in the US. Table 2 below summarizes some relevant estimates of the slope of the  $\ln P$  vs.  $1/T$  observed in urban and background areas.

### Urban Footprint of PCB Transport

Recent studies clearly demonstrate that urban-industrial areas exhibit the highest concentrations of atmospheric PCBs, and distant areas exhibit the lowest. Recently Green et al. (2000) have interpolated the atmospheric PCB concentrations observed in IADN and during the LMMB over the whole of Lake Michigan on a monthly scale. Their data clearly show that highest concentrations over the lake occur when winds are from the southwest and south (out of the Chicago area) and when land surface temperatures are highest. They mean predicted zone of influence of Chicago described on a monthly basis was reported to be  $\sim 40$  km although the influence under sustained southerly flows is significantly larger. This is similar to the determination of Ondov et al. (2000) that southerly flows from the Chicago area over the lake of particles containing trace metals may be transported and the entire lake surface and subjected to depositional processes. During the period when winds blew from the Chicago area, modeled and measured PCB concentrations during the AEOLOS campaign agreed very well (Figure 4). However, when winds turned to the north and northeast, modeled PCB concentrations remained high but the measured values dropped by a factor of four to reflect regional concentrations.

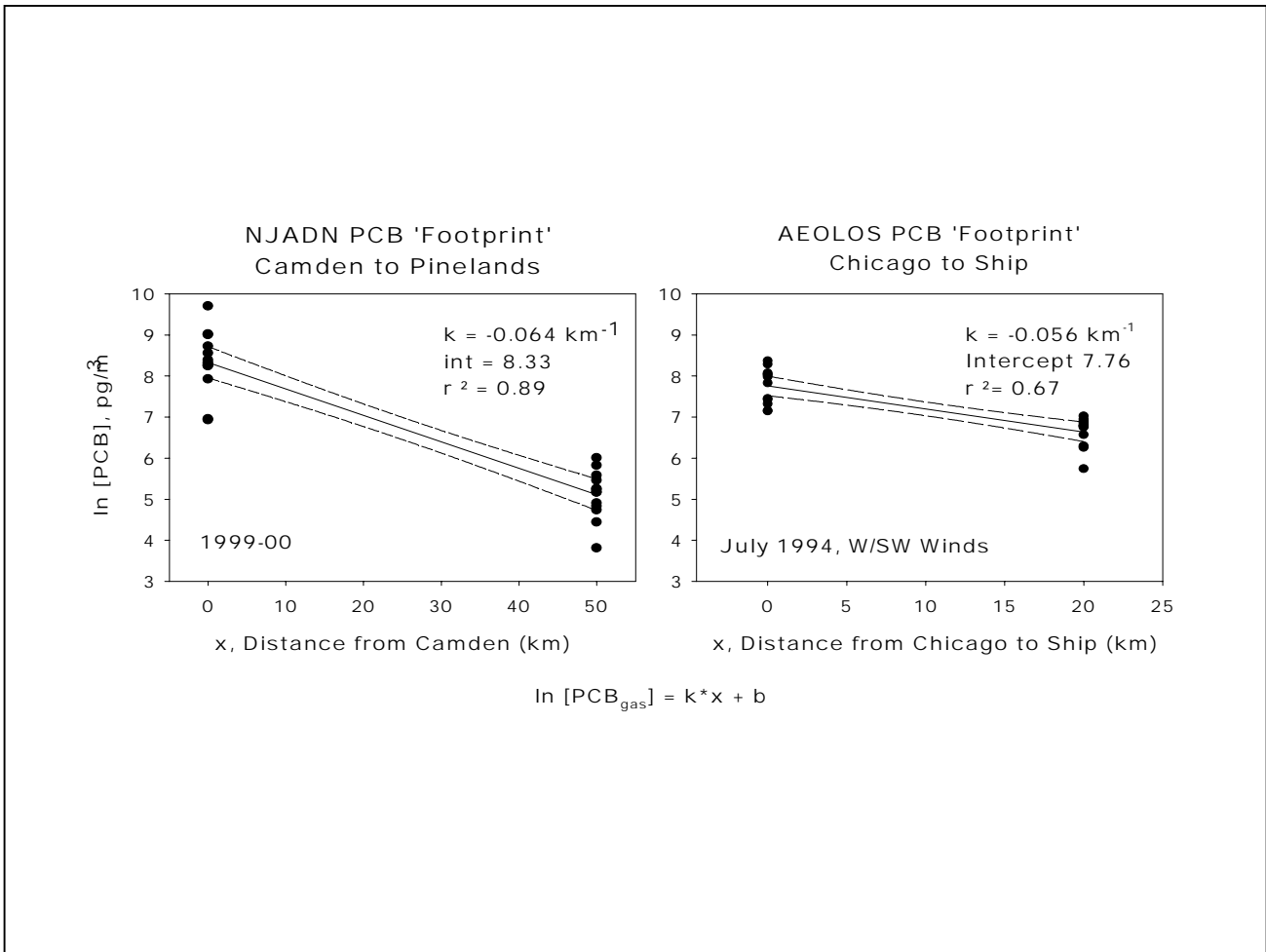
**Figure 4.** Comparison of model prediction for gas-phase PCB concentrations at LM Site 5 (Green et al., 2000) compared to AEOLOS measured values for 1994 (Simcik et al., 1997; Zhang et al., 1998).



This suggests that the over lake PCB concentrations predicted by the model of Green et al. (2000) might be too high with predictable influence on over-lake wet and gaseous deposition. The PCB concentrations coincide again when winds have been from the north for a period of days. The zone of influence of Chicago, however, under southerly or westerly winds appears to be substantially greater than the reported mean of 40 km.

The decrease in atmospheric PCB concentrations away from Chicago and other source areas is due to dispersion and dilution under transport, and removal by deposition and atmospheric loss processes. If the decrease is considered as a 1<sup>st</sup>-order process, then we can estimate the apparent 1<sup>st</sup>-order loss coefficients ( $k$ ,  $\text{km}^{-1}$ ) due to these processes:  $\ln [\text{PCB}_g] = k \cdot x + b$  where  $x$  is distance (km). Figure 5 demonstrates this simple phenomenon for the apparent decrease of atmospheric PCB concentrations from Chicago to the over-water sampling site 20 km distance (AEOLOS, 1994), and from Camden-Philadelphia to the Pinelands NJ site in a forested reserve 50 km distant (NJADN, 1999-00). In both cases, the apparent rate of PCB decrease in concentrations is  $\sim -0.06 \text{ km}^{-1}$  or  $6\% \text{ km}^{-1}$ . If we define the zone of influence as the distance required to reduce concentrations to 5 half-distances ( $5x(\ln 2/k)$ ), then the zone of influence is on the order of 50-60 km in both cases. This is close to the mean value reported in the Green et al. (2000) study. Is it surprising that the apparent rate of decrease in gaseous PCB concentration is about the same when transport is over water or over forested land? Does this suggest something interesting about the forces controlling deposition out of urban areas?

**Figure 5.** The estimated “footprint” of the  $\Sigma\text{PCBs}$  signal in the Chicago area during AEOLOS and in the Camden-Philadelphia area in 1999-2000



### Loss of PCBs in Tropospheric Transport

Atmospheric PCB concentrations away from urban sources generally exhibit a diurnal pattern with highest concentrations during the day or on the warmest days, and lower concentrations at night or on the coolest days (see Figure 2). However, the diurnal fluctuation of atmospheric PCBs in urban-industrial areas frequently show higher concentrations at night, and lower concentrations during the day (Simcik et al., 1997; Totten et al., 2000; Brunciak et al., 2000). This pattern is not observable in 24-hour air samples as is typical in the IADN and the NJADN. Within intensive field campaigns of AEOLOS (Chicago and Baltimore) and the NJADN, 12-hour and 4-hour sampling protocols are common, thus permitting examination of diurnal patterns.

The most effective means of demonstrating the apparent anomalous behavior of higher PCB concentrations at night is to plot the daytime depletion of PCB homologue concentrations relative to nighttime values. Figure 6 shows the daytime depletion pattern for urban samples collected in Chicago AEOLOS in July 1994, and in Jersey City, NJ located across the Hudson river from Manhattan in July 1998 (NJADN). Daytime depletions of PCB homologues ranged from 60% for trichlorobiphenyls to <10% for hexachlorobiphenyls (Totten et al., 2000).

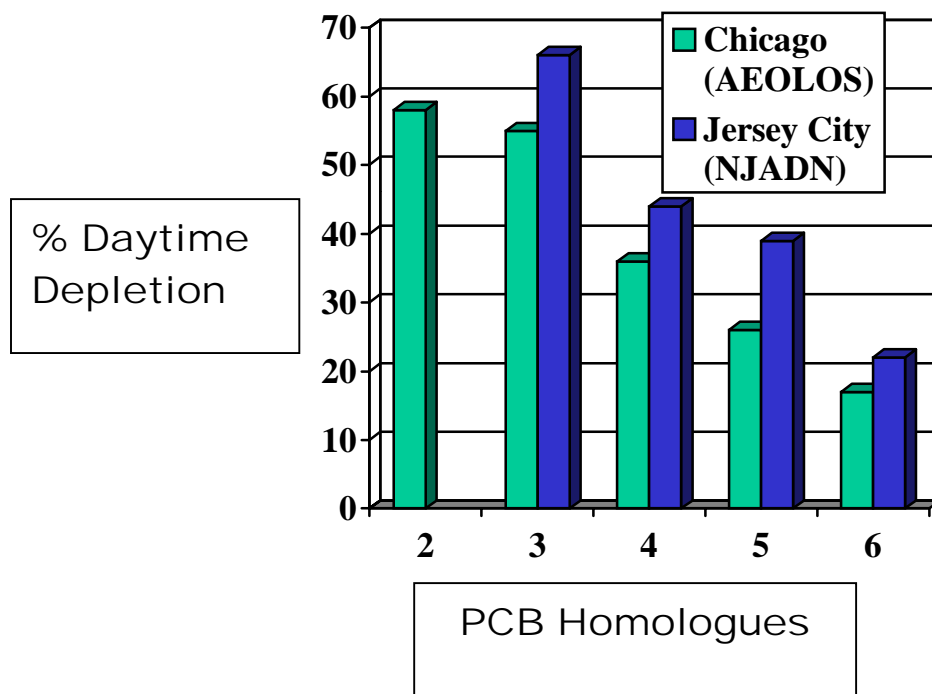


Figure 6. Percent daytime depletion of PCB concentrations by homologue For studies conducted in Chicago as part of AEOLOS and in New Jersey as part of the NJADN (Totten et al., 2000).

The data are consistent with the rate of OH radical attack on atmospheric PCB congeners, the effect of additional chlorines on the relative rate coefficient, and the absolute magnitude of the environmental rate constants for OH radical attack on PCB congeners  $\log k_e = k_{\text{obs}}/[\text{OH}] = -0.22 \text{ \#Cl} + \text{constant}$  (Anderson and Hites, 1996). Based on laboratory experiments of Anderson and Hites (1996), the half-lives for PCB homologues for range from 5-6 days for dichlorobiphenyls to 19-34 days for pentachlorobiphenyls. Totten et al. (2000) estimated values from 2-6 days to 10-20 days based on daytime depletion rates from urban-industrial atmospheres in the summer time. Brunciak et al. (2000) has estimated that the half-lives in the urban atmosphere of Jersey City, NJ over a year of changing environmental conditions are 145 days for trichlorobiphenyls to 460 days for pentachlorobiphenyls. Taken together, OH radical attack on atmospheric PCBs is an important loss process in urban atmospheres and on a global scale over long time periods, and acts preferentially on the low MW gas-phase species.

### **Recommendations**

Recognizing that old urban-industrial areas exhibit high atmospheric concentrations especially in the warm summer time, that this reflects strong local emissions, and that there is ample evidence that urban-industrial emissions of PCBs into the local atmosphere results in enhanced atmospheric signals and deposition, the PCB sources must be identified and limited, controlled and/or eliminated. Given that the influence of temperature on atmospheric PCB concentrations is more pronounced in urban complexes, then the size of the MER must be substantial.

**Recommendation 1. Stockpiles of PCB (Arochlor oils) must be identified, collected and disposed properly before they become accessible to environmental exchange processes. This may require 'buy-back' of PCB stockpiles without concern for liability or penalties.**

**Recommendation 2. PCB source and/or emission inventories must be constructed recognizing that normal regulatory protocols do not function for banned and widely-distributed chemicals that cycle constantly in the environment.**

One methodology now being examined by S. Eisenreich and K. Hornbuckle is to correlate measured (urban) PCB concentrations with temperature and surrogates of industrial/commercial/residential activities (county population in 1970; # Brownfields per county; measures of economic activity) to yield a calibrated function describing emission rates. A preliminary example will be presented.

**Recommendation 3. Renovation or destruction of existing old buildings in urban-industrial areas should be performed in such a way as to greatly limit PCB emissions.**

**Recommendation 4. Quantify the rate of gaseous PCB discharges to the atmosphere from sanitary landfills and control emissions if above agreed-upon rates.** In the case of the Fresh Kills' Landfill on Staten Island, PCB emissions to the local atmosphere may

exceed tonnes a year based on the emission of other chlorinated compounds. A preliminary example will be presented.

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