Status of PCBs in the Great Lakes

Keri C Hornbuckle[1], Daniel L Carlson[2], Deborah Swackhamer[2], Joel E Baker[3], Steven J Eisenreich[4]

[1]University of Iowa
[2]University of Minnesota
[3]University of Maryland
[4]EU Joint Research Centre Ispra

Results and Discussion

Properties of PCBs. The 209 different PCB congeners vary widely in their properties because of differing degrees and patterns of chlorination (Figure 1). This is especially evident in the saturation vapor pressure and water solubility of the congeners. Because both water solubility and vapor pressure have similar trends, the air-water partitioning coefficient (Henry's Constant) does not show a clear trend with chlorination number.

Many of the same properties that are described above, such as degree of chlorination, low water solubility, and hydrophobicity, are the same properties that make PCB compounds toxic. They are resistant to breakdown or metabolism, are bioaccumulative, and possess the ability to pass through cell membranes and bind to a variety of receptors that elicit their toxicity. Exposure to PCBs results in the risk of adverse impacts on wildlife populations as well as human populations. These impacts can vary by species, and the degree and level to which they are exposed, thus resulting in a wide variety of toxicological endpoints (e.g. reproductive impairment, cancer, chloracne). Much of our current understanding of these impacts arose from research on wildlife and human populations within the Great Lakes Basin. Despite being banned in North America more than 25 years ago, the toxicological risks from PCB exposure is still a present-day concern. For example, current concentrations in fishes in the Great Lakes are still sufficiently great enough as to drive the need for fish consumption advisories for sport fishes and to restrict commercial fisheries.

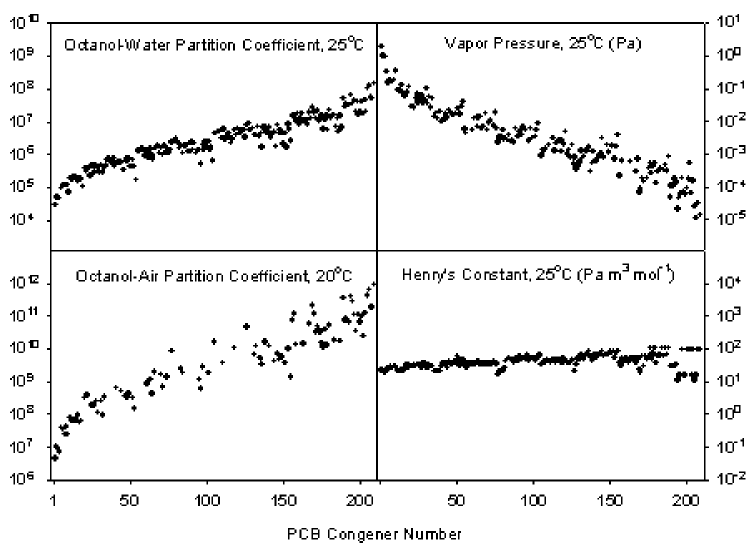

Figure 1. Properties of PCB congeners.

Sources of PCBs. It has been long assumed that eating fish is the major exposure route for humans. Eating Great Lakes fish has been shown to cause adverse neurotoxicological effects in humans. Surprisingly, the source of PCBs in those fish is not clear, and little is known about the relative magnitudes of PCB sources to natural waters.

The major sources of PCBs to the Great Lakes now include tributary flow, resuspension of contaminated sediments and atmospheric deposition. Over the last decade, there have been several major research studies to quantify all the PCB sources to Lake Michigan and Green Bay. Although there have been studies of PCB sources in Saginaw Bay (Lake Huron), Lake Erie, and Lake Ontario, only Lake Michigan has benefited from a coordinated field study to assess of all the possible sources. From studies completed in Lake Michigan, it is now clear that atmospheric processes dominate current PCB inputs to the Great Lakes.

Atmospheric inputs are the largest source of PCBs to Lake Michigan (Figure 2). At least 2000 kg of gas-phase PCBs enter Lake Michigan each year *(7, 8)*. The majority of that input is through gas exchange. Deposition of PCBs associated with aerosol particles accounts for an additional 100 kg each year. Deposition of PCBs in rain contributes a similar amount. Atmospheric deposition (gas, dry particle, and wet deposition) is larger than inputs from resuspension of contaminated sediments and larger than inputs from direct discharge and contaminated tributaries. Resuspension of contaminated sediments is an important source of PCBs to the water, but still not as large as gross atmospheric deposition. For example, a field study of resuspension after major storms concluded that annual wintertime storms contribute as much as 400 kg of PCBs per event *(7)*. Although there may be more than one major event each year, the total amount of PCBs resuspended is still only about 1200 kg, and a large fraction of that mass may immediately return to the lake floor. The tributary load is only about 380 kg and that includes the Fox River discharge to Green Bay. The importance of the atmospheric pathway is widely misjudged. In fact, sediment export from contaminated tributaries as a source of PCBs to Lake Michigan is often cited as a justification for dredging. But if a goal is to reduce concentrations of PCBs in the open lake, more effort should focus on reducing or eliminated atmospheric sources.

Fate and Transport. Once in the waters of the Great Lakes, PCBs partition to particles and plankton and can accumulate in the aquatic food chain. In the Great Lakes, hydrolysis, photolysis, and microbial decay are considered negligible loss mechanisms for PCBs. Loss though volatilization and sediment burial are the most efficient loss mechanisms. The importance of atmospheric exchange processes is one of the most striking findings of the mass budget analysis. This implies that the atmosphere is also a major source of PCBs ultimately accumulated in fish.

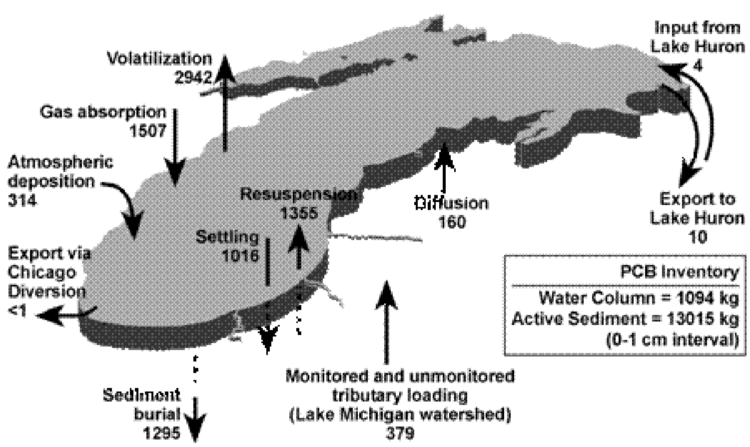

Figure 2. The magnitude and direction of various processes that move PCBs into and out of Lake Michigan each year, modified from *(8)*.

The partitioning and accumulation of PCBs from the major sources to humans is largely controlled by the physical-chemical properties of the congeners. As a result the PCB congener distribution patterns change as a function of the accumulation route. For example, PCBs in the air over the lakes are dominated by the lower chlorinated, more volatile congeners. In sediment samples, however, the more chlorinated congeners dominate. A similar trend is observed with length of the food chain. Organisms in the highest trophic levels, like lake trout or humans, show PCB congener patterns enriched in the more chlorinated congeners. This trend is illustrated in a plot of mean chlorine number for various sample categories (Figure 3). The mean chlorination level tends to increase from air to sediment and from lower to higher trophic levels. The figure illustrates data that are comparable over time and space and compared to the congener distributions of two technical mixtures of PCB Aroclors that were widely used in the Great Lakes Basin.

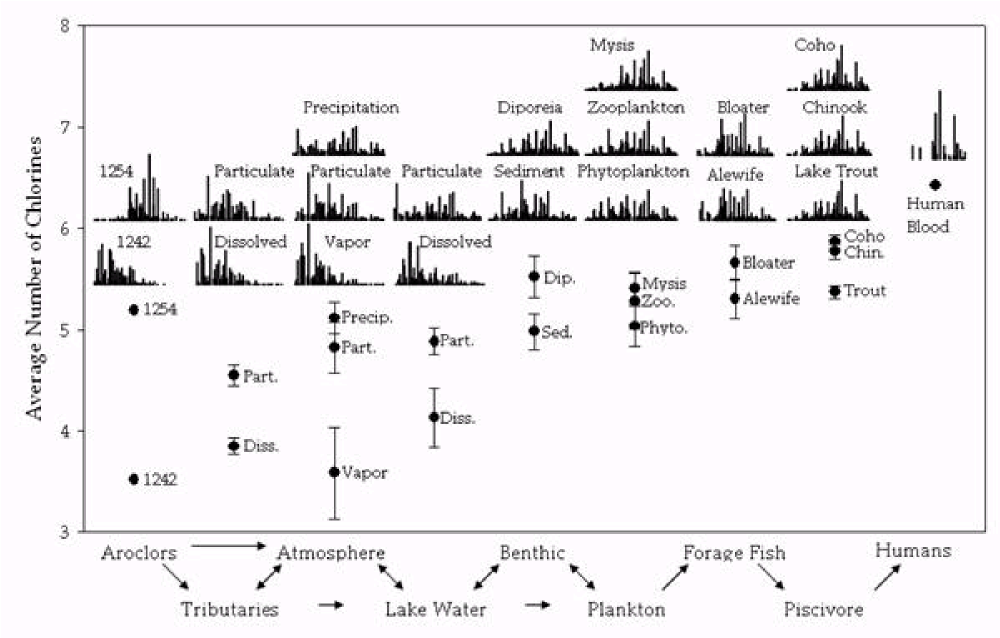

Figure 3. Mean congener distribution patterns and chlorine number for various environmental media in southern Lake Michigan. The samples illustrated were collected in the 1990s.

References

1. Veith, G.D., University of Wisconsin: 1968.

2. Jensen, S. Report of a new chemical hazard. *New Sci* 1966, *32*, 612.

3. Swackhamer, D.L. Studies of polychlorinated biphenyls in the Great Lakes. *Issues Environ. Sci. Technol.* 1996, *6*, 137-153.

4. Golden, K.A.; Wong, C.S.; Jeremiason, J.D.; Eisenreich, S.J.; Sanders, G.; Hallgren, J.; Swackhamer, D.L.; Engstrom, D.R.; Long, D.T. Accumulation and preliminary inventory of organochlorines in Great Lakes sediments. *Water Sci. Technol.* 1993, *28*, 19-31.

5. Brown, D.P. Mortality of workers exposed to polychlorinated biphenyls - an update. *Arch. Environ. Health* 1987, *42*, 333-339.

6. Hornbuckle, K.C.; Carlson, D.; Swackhamer, D.L.; Baker, J.E.; Eisenreich, S.J. in *Contaminants in the Great Lakes*, Hites, R.A., Ed. American Chemical Society: Washington, DC. 2005.

7. Hornbuckle, K.C.; Smith, G.L.; Miller, S.M.; Eadie, B.J.; Lansing, M.B. Magnitude and origin of PCB and DDT compounds resuspended in southern Lake Michigan. *J. Geophysical Res.-Oceans* 2004, *109, Art. CO5017 May 05018*.

8. USEPA *Lake Michigan Mass Balance Study Loadings Report*. Great Lakes National Program Office, US Environmental Protection Agency 2005. http://www.epa.gov/glnpo/lmmb/results/loadpcbs.html.