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The Integrated Atmospheric Deposition Network (IADN)

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Summary

For almost two decades, a joint U.S.-Canadian project, operating at Indiana University, has measured the atmospheric contributions of chemical pollution to the Great Lakes. Over time, this project has produced significant information for scientists and policy makers alike. Key findings from the Hites' Laboratory at the School of Public and Environmental Affairs show that

1. most pollutants studied tend to peak in concentration during warm months,
2. concentrations of polychlorinated biphenyls, in particular, are declining more slowly than other pollutants, and
3. atmospheric concentrations of these pollutants are highly correlated with population density.

All three findings suggest opportunities for targeted policy interventions.

What is IADN?

The Great Lakes have become contaminated with many toxic organic contaminants over the past few decades. These contaminants include industrial chemicals such as polychlorinated biphenyls (PCBs), combustion-related pollutants such as polycyclic aromatic hydrocarbons, and chlorinated pesticides such as lindane and DDT. Large amounts of these chemicals enter the lakes by way of the atmosphere; thus, atmospheric measurements around the Great Lakes are needed to quantitate this atmospheric deposition process (as opposed to direct or tributary inputs) and to follow its temporal trends. To implement and interpret these measurements, the "Integrated Atmospheric Deposition Network" (IADN) was created by an agreement between the United States and Canadian governments. The United States' part of this network was initiated in 1990 in Illinois and moved to SPEA in 1994; the Canadian part has been operated by Environment Canada in Toronto.

The specific intent of IADN is to measure and evaluate more than 150 pollutant concentrations in the atmosphere (airborne vapor, airborne particles, and precipitation) at a lake-wide level of detail. Organic pollutants of interest include several chlorinated pesticides (some of which have been banned and some of which are still in use), PCBs, several polycyclic aromatic hydrocarbons, and more recently, brominated and chlorinated flame retardants. The Hites' Laboratory at SPEA uses these measurements to calculate flows of selected toxic organic contaminants into the Great Lakes and to explore spatiotemporal trends in atmospheric and precipitation concentrations. The Hites' Laboratory is also exploring the importance of short- vs. long-range atmospheric transport and the significance of in-use pesticide inputs.

Air samples have been collected continuously at the five sites shown in Figure 1. Air samples are taken every 12 days for 24 hours in such a way as to separately collect the particle and vapor phase contaminants. Precipitation is sampled using automated wet-only samplers and integrated for one month. Because one of the goals of this project is to compare concentrations over a 10-25 year time scale, it is important to have excellent long-term continuity of the measurements. For this reason, the Hites' Laboratory has made great efforts to ensure the longitudinal quality of this project's data stream. The simple fact that this program has been operated at one institution (Indiana University) for the last 16 years has been tremendously helpful. Perhaps the most important factor in maintaining continuity of data quality has been the presence of Dr. Ilora Basu since the inception of this project. Dr. Basu worked on this project when it was first initiated in Illinois in 1990, and when the project moved to Indiana University in 1994, Dr. Basu moved with it.

From 1994 until today, the Hites' Laboratory has taken and analyzed over 6,600 samples. These data have been quality assured and submitted to the U.S. Environmental Protection Agency for inclusion in the IADN master database. In addition, dozens of papers and reports have been prepared from these data and published in high-quality journals, such as *Environmental Science and Technology*, the highest ranked journal in environmental chemistry. IADN projects have also formed the base for numerous doctoral and master's theses, which have been submitted to Indiana University for academic credit. The faculty, staff, and students of the Hites' Laboratory have presented lectures about the IADN project at over 70 regional, national, and international meetings. Thus, IADN is contributing to the training of the next generation of environmental chemists.

Figure 1. *The locations of the five United States Integrated Deposition Network air sampling sites; the population density over this region is also shown.*

Three Important Findings

One of our earliest observations was that the atmospheric concentrations of almost all of these pollutants maximized in the warm summer months. This is true for the PCBs (see Figure 2, top) and for pesticides such as lindane (see Figure 2, bottom). The concentrations of these compounds maximize in the summer as their environmental reservoirs (soil and water) heat up and release increasing amounts of these semi-volatile pollutants. The only pollutant group for which the levels do not maximize in the summer is the polycyclic aromatic hydrocarbons, and this is because these compounds are produced and released into the atmosphere by combustion systems (for example, those used for space heating in the winter). The difference between the (usually) peak summer and the minimum winter concentrations is about a factor of eight.

Another important finding is the result of having data over a 17-year period (as shown in Figure 2), which is essential for determining temporal trends in atmospheric concentrations. For example, PCB concentrations in the atmosphere are decreasing very slowly, if at all, even though their production was banned in 1976. Figure 2, top, shows that the PCB levels in 2000-2002 were about the same as the levels in 1992-1996. Careful statistical analysis of these data indicates that the PCB concentrations at this location are decreasing by a factor of two in about 30 years. The levels of some other pollutants are going down a bit more quickly; for example, the atmospheric concentrations of lindane (see Figure 2, bottom) are decreasing by a factor of two in about 10 years, showing the positive effects of bans on the agricultural uses of this insecticide in both the U.S. and in Canada. The atmospheric levels of some of the other banned pesticides (DDT and chlordanes) are also declining with halving times of 8-10 years. With the exception of PCBs, this is generally good news.

The surprisingly slow rate of decrease for PCBs may be due to large amounts of PCBs that have not been permanently removed from the environment. Tons of PCBs are still out there, especially in electrical gear, such as transformers and capacitors. In addition, "decommissioned" PCBs have not really been removed from the environment either; rather, they have been placed in landfills and in other disposal facilities that may be leaking into the atmosphere. Of course, there may also be large reservoirs of some insecticides in agricultural and urban soils that are only slowly being depleted.

More recently, we have made a simple but previously overlooked observation: The atmospheric levels of most of the pollutants we

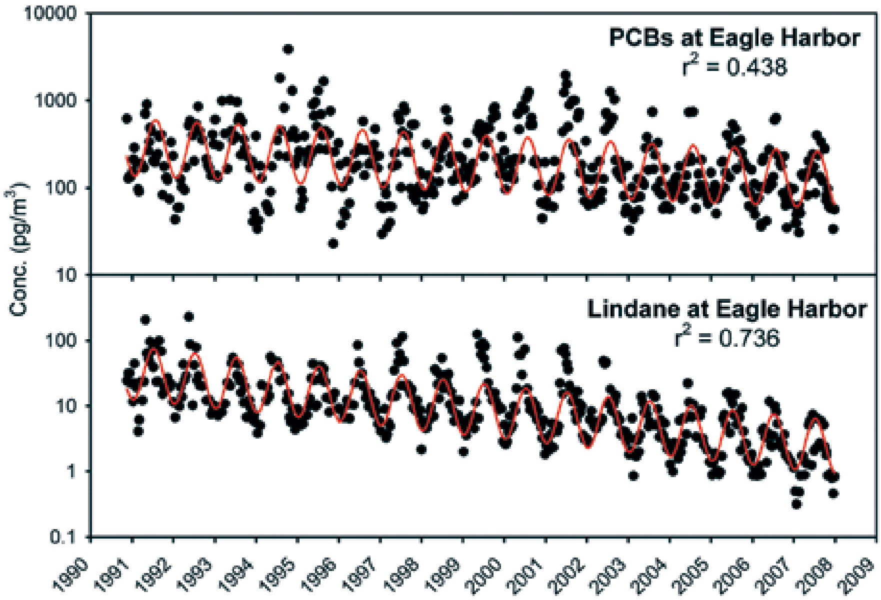

Figure 2. *Concentrations of PCBs (top) and lindane (bottom) measured every 12 days in the atmosphere at Eagle Harbor, Michigan, since November 1990. The concentration units are picograms per cubic meter of air. This sampling site is located at the northern-most tip of the Keweenaw Peninsula in Lake Superior. The red line is a fit of the following equation:* $ln(C) = a_0 + a_1t + a_2 sin(zt) + a_3 cos(zt)$*, where* t *is Julian Day relative to January 1, 1990 and* $z = 2\pi/365.25$*.*

measure are highly dependent on the human population living and working within a 25 km radius of the sampling site. The strongest dependence was for polycyclic aromatic hydrocarbons (PAH), and the weakest dependence was for the insecticide lindane (note the different slopes shown in Figure 3.) This makes sense given that polycyclic aromatic hydrocarbons are produced where people live and where they burn various fuels to keep warm, move about, and generate energy. On the other hand, lindane was primarily used on farms in the U.S. and Canadian prairies – far away from the crush of people. The presence of brominated and chlorinated flame retardants is also highly related to where people live; many household and commercial items (polyurethane foam and most hard plastics) are treated with these compounds to mitigate the danger of fires in the home and workplace. This general observation has important implications for pollution prevention and control: Focus pollution remediation in cities, and give the farmers a pass for now.

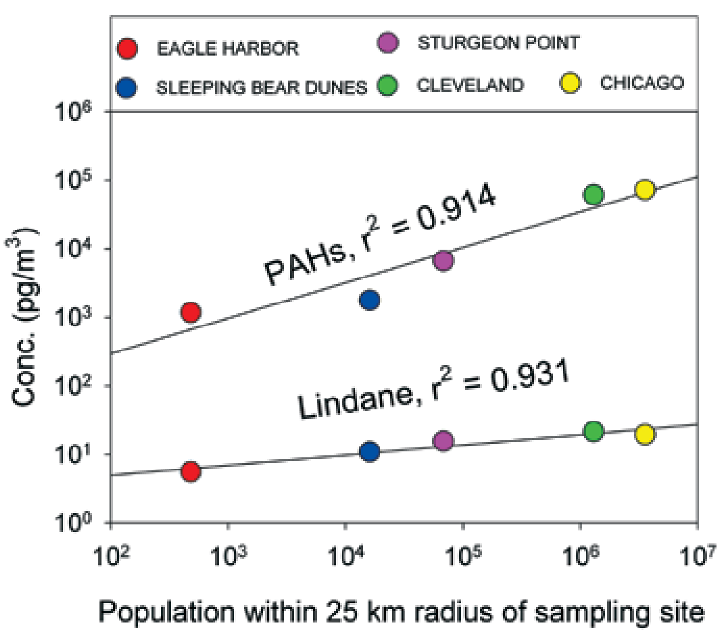

Figure 3. *Average concentrations (in picograms per cubic meter of air) of PAHs and lindane in the atmosphere at the five United States IADN sites as a function of the population living and working within a 25 km radius of the sampling site.*

The Future of IADN at SPEA

In 2008, an independent scientific peer review panel found that the implementation of IADN was technically sound and called for maintaining the continuity in network operations and analytical protocols. Given these recommendations, SPEA asked the United States Environmental Protection Agency to continue the cooperative agreement it had with Indiana University for the operation of IADN for another five years. They agreed and awarded SPEA a grant for $5,000,000 to support IADN until September of 2014.

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