Great Lakes Binational Toxics Strategy

2009 Biennial Report

EPA

Canada

Cover Photo Credits:

1: Chicago Lighthouse, Photograph courtesy of U.S. EPA

2. Niagara Falls, Photograph courtesy of The Center for Great Lakes Aquatic Sciences

3. Sandhill Crane at Nest, Photograph by Thomas A. Schneider

EN 161-1/2-2009E-PDF

978-1-100-17486-0

TABLE OF CONTENTS

ABBREVIATIONS

AFFF	Aqueous Fire-Fighting Foam
AHTN	Acetyl Hexamethyl-Tetrahydronaphthalene
ANOVA	Analysis of Variance
ANPR	Advance Notice of Proposed Rulemaking
AOC	Area of Concern
B(a)P	Benzo(a)pyrene
BCF	Bioconcentration Factor
BDE	Brominated Diphenyl Ethers
BFR	Brominated Flame Retardants
BGSU	Bowling Green State University
BTBPE	1,2-bis(2,4,6-tribromophenoxy)ethane
CAA	Clean Air Act
CAC	Criteria Air Contaminant
CAD	Confined Aquatic Disposal
CADAMP	California Ambient Dioxin Air Monitoring Program
CAMR	Clean Air Mercury Rule
CAMU	Corrective Action Management Unit
CanMETOP	Canadian Model for Environmental Transport of Organochlorine Pesticides
CCME	Canadian Council of Ministers of the Environment
CDF	Confined Disposal Facility
CEC	Commission for Environmental Cooperation
CEPA	Canadian Environmental Protection Act
CGLI	Council of Great Lakes Industries
ChAMP	Chemical Assessment and Management Program
CI	Confidence Interval
CMP	Chemical Management Plan
CMTI	Clean Manufacturing Technology Institute (Indiana)
COA	Canada-Ontario Agreement
COP4	Fourth Meeting of the Conference of the Parties to the Stockholm Convention
CTS	Coal Tar Sealants
CWS	Canada-wide Standard(s)
DBDPE	Decabromodiphenyl ethane
DDD	Dichlorodiphenyldichloroethane
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichlorodiphenyltrichloroethane
DEHP	Bis(2-ethylhexyl) phthalate
D/F	Dioxin/Furan
DLPCB	Dioxin-Like PCBs
DP	Dechlorane Plus
DSL	Domestic Substances List
EAF	Electric Arc Furnace
EC	Environment Canada
EC-NWSB	Environment Canada's National Wildlife Specimen Bank
EMS	Environmental Management System
ENEV	Estimated No-Effect Value
ENGO	Environmental Non-Governmental Organization
EPA	Environmental Protection Agency
EPI	Estimation Program Interface
EROD	Ethoxyresorufin-O-deethylase
ESCO	Environmental Services and Consulting
EU	European Union
FE	Fort Erie
FSU	Former Soviet Union
GIS	Geographic Information System
GLBTS	Great Lakes Binational Toxics Strategy
GLLA	Great Lakes Legacy Act

GLNPO	Great Lakes National Program Office
GLRC	Great Lakes Regional Collaboration
GLRI	Great Lakes Restoration Initiative
GLWQA	Great Lakes Water Quality Agreement
HARP	Hayton Area Remediation Project
HBB	Hexabromobenzene
HBCD	Hexabromocyclododecane
HCB	Hexachlorobenzene
HCH	Hexachlorocyclohexane
HE	Heptachlor epoxide
Hg	Mercury
HHCB	Hexahydrohexamethylcyclopentabenzopyran
HIT	Harbin Institute of Technology
HPBA	Hearth, Patio and Barbeque Association
HPV	High Production Volume
HRAI	Heating, Refrigeration and Air Conditioning Institute of Canada
HWC	Hazardous Waste Combustors
H2E	Hospitals for a Healthy Environment
IADN	Integrated Atmospheric Deposition Network
IDEM	Indiana Department of Environmental Management
IJC	International Joint Commission
IJRC-PTS	International Joint Research Center for Persistent Toxic Substances
I-TEQ	International Toxic Equivalents
IUR	Inventory Update Reporting [Rule]
LaMPs	Lakewide Management Plans
LDR	Land Disposal Restrictions
LRAT	Long-Range Atmospheric Transport
MACT	Maximum Achievable Control Technology
MCDI	Midwest Clean Diesel Initiative
MDEQ	Michigan Department of Environmental Quality
MHSW	Municipal Hazardous or Special Waste
MLE	Maximum Likelihood Estimation
MOE	Ministry of the Environment (Ontario)
MPCA	Minnesota Pollution Control Agency
MUCC	Michigan United Conservation Clubs
MWC	Municipal Waste Combustor
MWI	Medical Waste Incinerator
NA	North America
NAPS	National Air Pollution Surveillance
NARAP	North American Regional Action Plan
NCEP	National Centers for Environmental Prediction
NDAMN	National Dioxin Air Monitoring Network
NEWMOA	Northeast Waste Management Officials' Association
NOAA	National Oceanic and Atmospheric Administration
NOTL	Niagara-on-the-Lake
NP	Nonyl Phenol
NPE	Nonylphenol [and its] Ethoxylate[s]
NPL	National Priority List
NPRI	National Pollutant Release Inventory (Canada)
NRDA	Natural Resource Damage Assessment
NRTMP	Niagara River Toxics Management Plan
NWF	National Wildlife Federation
NYSDEC	New York State Department of Environmental Conservation
O&M	Operation and Maintenance
OCP	Organochlorine Pesticide
OCS	Octachlorostyrene
OMOE	Ontario Ministry of the Environment
OPPTS	Office of Prevention, Pesticides and Toxic Substances (US EPA)
OTS	Ontario Tire Stewardship
OU	Operable Unit
P&B	Persistent and Bioaccumulative
P2	Pollution Prevention
PAH	Polycyclic Aromatic Hydrocarbon
PBDE	Polybrominated Diphenyl Ether

PBEB	Pentabromoethylbenzene
PBT [PB&T]	Persistent, Bioaccumulative, and Toxic
PCBs	Polychlorinated Biphenyls
PCDD	Polychlorinated Dibenzo-Para-Dioxin
PCDF	Polychlorinated Dibenzofuran
PCP	Pentachlorophenol
PFAS	Perfluoroalkyl Sulfonates
PFC	Perfluorinated Compound
PFCA	Perfluorocarboxylic Acid
PFDS	Perfluorodecasulfonate
PFHxS	Perfluorohexane Sulfonate
PFOA	Perfluorooctanoic Acid
PFOS	Perfluorooctanesulfonate
PFOSA	Perfluoro-1-octanesulfonamide
PFSA	Perfluorosulfonates
PM	Particulate Matter
PMRA	Pest Management Regulatory Agency
PNEC	Predicted No-Effect Concentration
POPs	Persistent Organic Pollutants
PPCPs	Pharmaceuticals and Personal Care Products
PPM	Parts per Million
PTS	Persistent Toxic Substances
PUF	Polyurethane Foam
PVOC	Polar Volatile Organic Compounds
RAP	Remedial Action Plans
RCO	Recycling Council of Ontario
RCRA	Resource Conservation and Recovery Act
RMA	Rubber Manufacturers Association
RMS	Risk Management Strategy
ROPS	Remedial Options Pilot Study
R/V	Research Vessel
SAB	Science Advisory Board
SAR	Structure-Activity Relationship
SLRIDT	St. Louis River/Interlake/Duluth Tar
Sn	Tin
SOLEC	State of the Lakes Ecosystem Conference
SOP	Strategic Options Process / Standard Operating Procedure
SPMD	Semi-permeable Membrane Device
SPP	Security and Prosperity Partnership
SWAC	Surface Weighted Average Concentration
SWARU	Solid Waste Area Reduction Unit
SVOC	Semivolatile Organic Compound
TBBPA	Tetrabromo Bisphenol A
TBE	1,2-bis(2,4,6-tribromophenoxy)ethane
TBT	Tributyltin
TCDD	Tetrachlorodibenzodioxin
TCRA	Time Critical Removal Action
TDSB	Toronto District School Board
TEQ	Toxic Equivalent
TRC	Thermostat Recycling Corporation
TRI	Toxics Release Inventory (U.S.)
TSCA	Toxic Substances Control Act
TSMP	Toxic Substances Management Policy
UNEP	United Nations Environment Programme
US ACE	United States Army Corps of Engineers
US EPA	United States Environmental Protection Agency
USFWS	United States Fish and Wildlife Service
USGS	United States Geological Survey
UVCB	Unknown or Variable Composition, Complex Reaction Products and Biological Materials
VOC	Volatile Organic Compound
WDNR	Wisconsin Department of Natural Resources
WRDA	Water Resources Development Act
WG	Workgroup
WHO	World Health Organization
WLSSD	Western Lake Superior Sanitary District
WQS	Water Quality Standard
WWTP	Wastewater Treatment Plant

Executive Summary

Sleeping Bear Dunes, Photograph by Robert DeJonge

Introduction

During 2008 and 2009, the Great Lakes Binational Toxics Strategy (GLBTS, or Strategy) forum continued to pursue a new path forward in addressing emerging chemical threats to the Great Lakes Basin. Many of the challenge goals established by Environment Canada (EC) and the United States Environmental Protection Agency (US EPA) with the signing of the Strategy in 1997 have been met. Thirteen of the Strategy's original 17 challenge goals for Level 1 substances have been achieved, and significant progress has been made toward the remaining four. The GLBTS is hoping to build upon the successes of the past to identify and address new chemicals of concern to the Basin.

This report documents the progress achieved and actions taken to reduce the use and release of GLBTS Level 1 substances. This report also highlights the activities of a new group focused on emerging substances of concern and presents environmental monitoring data collected by Great Lakes monitoring and surveillance programs.

About This Report

This report contains a compilation of activities and progress achieved under the GLBTS for the years 2008 and 2009. Chapters 1 through 4 present highlights for the Level 1 substance workgroups for mercury, polychlorinated biphenyls (PCBs), dioxins and furans, and hexachlorobenzene (HCB) and benzo(a)pyrene (B(a)P), respectively. These highlights include a summary of progress toward the GLBTS challenge goals, a review of workgroup meetings, and descriptions of activities undertaken to reduce the use or emissions of the Level 1 substances. Chapter 5 documents the progress of the Substance/Sector Workgroup. Chapter 6 presents a summary of Integration Workgroup activities, including four workgroup meetings, and three Stakeholder Forums held in 2008 and 2009. Chapter 7 reports progress in remediating contaminated sediments in the Great Lakes Basin, including descriptions of Great Lakes sediment remediation projects, estimated sediment volumes remediated or capped, and estimated volumes of contaminated sediment remaining in specific Areas of Concern (AOCs). Chapter 8 presents examples of efforts to evaluate the contribution and significance of the long-range transport of Strategy substances. Chapter 9 presents the State of the Great Lakes with regards to contaminant trends in ambient air, fish, herring gull eggs, mussels, and sediments and surface waters. Appendix A includes a compendium of activities related to the GLBTS that have been undertaken from 1997 to 2009.

Highlights of the report are presented below.

- The Mercury Workgroup is being phased-out, as both Canada and the United States have met their challenge goals. Canada has reduced mercury releases by greater than 90%, and the U.S. has reduced uses and releases of mercury by more than 50%.
- The PCB Workgroup is active and continues to make progress toward reaching the PCB challenge goals outlined in the Strategy.
- The Dioxin/Furan Workgroup has suspended further active work, as the challenge goals have been met for both countries. However, both countries will continue to monitor dioxin in the environment, investigate dioxin data as available, and look for reductions in uncontrolled combustion sources such as burn barrels. The 2007 inventory of dioxin/furan releases in Ontario totals 25.6 g I-TEQ

(international toxic equivalents)/year. The U.S. has not updated the dioxin inventory since 2000. Burn barrels and household garbage burning are the largest quantified sources of dioxin emissions in both countries.

» The work of the HCB/B(a)P Workgroup has continued. For example, EC conducted testing of certified wood stoves to evaluate emission factors and completed a polycyclic aromatic hydrocarbon (PAH) Source Apportionment Modeling project. US EPA continued its Midwest Clean Diesel Initiative and launched a national Burn Wise educational campaign to help reduce wood smoke pollution.

» The Substance/Sector Workgroup met several times in person or by teleconference during 2008 and 2009 and gathered information on emerging contaminant monitoring and surveillance efforts in the Great Lakes.

» In 2008, approximately 740,000 yd^3 of contaminated sediment were remediated from U.S. and Canadian sites in the Great Lakes Basin.

» Research continues into the contribution and significance of long-range transport of toxic substances to the Great Lakes. For example, present modeling investigations indicate that U.S. and Canadian emission sources made the largest contribution to the loading of penta-brominated diphenyl ether (penta-BDE) to North American terrestrial surfaces, followed by China, India, and Western Europe.

» Canadian monitoring data indicate declining ambient air concentrations of dioxins, furans, coplanar PCBs, B(a)P, and HCB at Ontario sites.

» Similarly, data from US EPA's Great Lakes Fish Monitoring Program and EC's Great Lakes Fish Contaminant Surveillance Program show declining concentrations of several Strategy substances in Great Lakes fish. Brominated flame retardants have been reported in fish tissues for several years throughout the Great Lakes Basin, and retrospective analyses have been conducted on archived tissue samples.

» Contaminant levels in herring gull eggs collected from Great Lakes colonies by the Canadian Wildlife Service indicate that concentrations of several flame retardants have accumulated in herring gull eggs. PBDE contamination increased rapidly from 1981 to 2000, primarily associated with the penta-BDE formulation. Congeners derived mainly from penta-BDE and octa-BDE mixtures showed no increasing trend post-2000. From 1982 to 2006, concentrations of BDE-209 and the octa- and nona-BDE congeners, which result from the debromination of BDE-209, continued to increase, with BDE-209 doubling times ranging from 2.1 to 3.0 years.

» Recent data of legacy contaminant concentrations in herring gull eggs from 1997 to 2007 suggest that there has been virtually no significant decline in concentrations of most legacy contaminants in gull eggs over the last 10 years.

» Data from NOAA's Mussel Watch Program (National Oceanic and Atmospheric Administration) indicate varying concentration trends in mussel tissues and sediment for several Strategy substances from 1993 to 2008. Many substances show decreasing trends or no trend at all. However, sediment concentrations of several substances in the Great Lakes remain high compared to national levels. Beginning in 2009, NOAA is making several enhancements to the Mussel Watch Program with the primary goal of improving data and information sharing, and coordinating with the monitoring efforts of other federal and state agencies.

1.0 MERCURY

Workgroup Status: Less active information-sharing group
Canadian Workgroup co-chair: Robert Krauel
U.S. Workgroup co-chair: Alexis Cain

Cascade River Falls into Lake Superior, Photograph by Robert F. Beltran

Progress Toward Challenge Goals

U.S. Challenge: Seek by 2006, a 50% reduction nationally in the deliberate use of mercury and a 50% reduction in the release of mercury from sources resulting from human activity.

Canadian Challenge: Seek by 2000, a 90% reduction in the release of mercury, or where warranted the use of mercury, from polluting sources resulting from human activity in the Great Lakes Basin.

Ontario:
Progress Toward the GLBTS Challenge

In Ontario, releases of mercury have been reduced by slightly more than 90% between the 1988 baseline and 2006, thus achieving the Canadian 90% reduction target. Figure 1-1 illustrates the progress made toward the Canadian reduction target.[1] This figure shows that releases in Ontario have been cut by more than 12,600 kg since 1988, based on EC's 2006 mercury inventory. Note that some of the sources listed in the legend of Figure 1-1 (e.g., paint, pesticides) refer to the baseline year of emissions and are no longer current sources. Figure 1-2 illustrates the 2006 sources of mercury releases in Ontario. This figure shows that the primary sources of releases are municipal (primarily land application of biosolids), electric power generation, iron and steel, cement and lime, and incineration. However, all of these sectors have reduced releases when compared to the 2003 inventory reported in the previous progress report.[2] Most notable is the reduction in the electric power generation sector, which contributed 19% of total releases in 2006 compared to 29% of total releases in 2003.

United States:
Progress Toward the GLBTS Challenge

The U.S. has met its challenge goals of a 50% reduction in the deliberate use of mercury and a 50% reduction in releases of mercury nationwide. According to the National Emissions Inventory,

[1] This target is considered as an interim reduction target and, in consultation with stakeholders in the Great Lakes Basin, will be revised if warranted, in accordance with periodic Canada-Ontario Agreement (COA) reviews of mercury use, generation, and release from Ontario sources.

[2] US EPA and EC. (2006). *Great Lakes Binational Toxics Strategy 2006 Annual Progress Report*, Tenth Anniversary Edition. Prepared by US EPA and Environment Canada. Report No. En161-1/2006E; 978-0-662-45249-2. Available at http://binational.net/bns/2006/2006GLBTS_en.pdf.

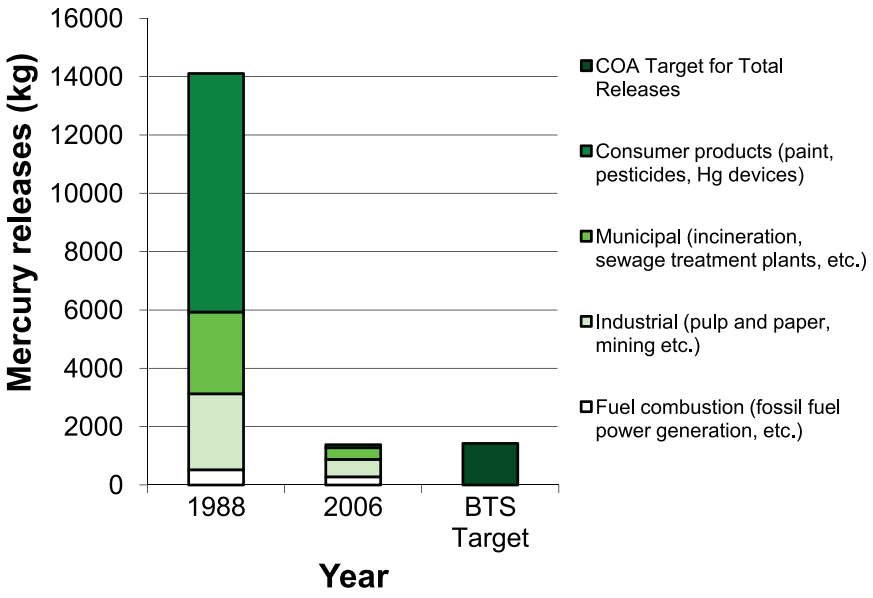


Figure 1-1. Reductions in Mercury Releases in Ontario from 1988 to 2006, by Sector. Source: Environment Canada, Ontario Region/Ontario Ministry of the Environment (2007)

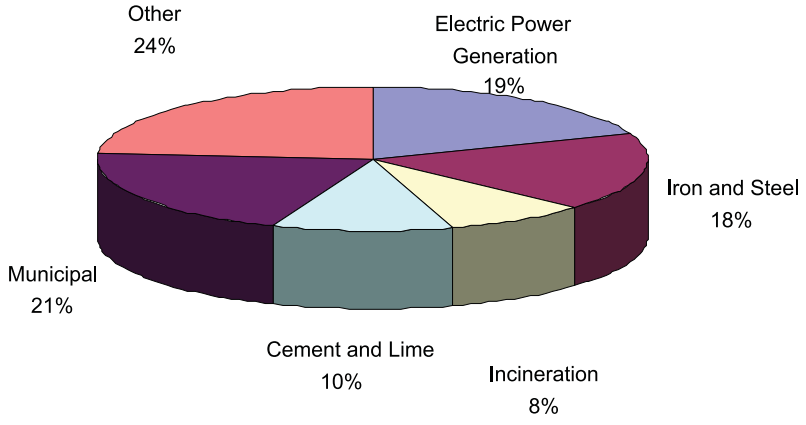


Figure 1-2. Sources of Mercury Releases in Ontario (2006). Source: Environment Canada, Ontario Region/ Ontario Ministry of the Environment (2007)

U.S. mercury emissions decreased approximately 58% between 1990 and 2005.[3] Figure 1-3 presents the reduction in U.S. mercury emissions from 1990 to 2005 compared to the challenge goal.

Total mercury use in the U.S. is estimated to have declined by more than 50% between 1995 and 2003, based on data reported by the chlor-alkali, lamp, and dental industries. Mercury use has continued to decline since 2003. Figure 1-4 depicts the reductions in mercury uses since 1995, compared to the challenge goal.

Workgroup Activities

On November 17-18, 2009, a Mercury Science & Policy Conference with a Special Focus on the Great Lakes and Northeast Regions was held in Chicago. The GLBTS co-sponsored the conference with the Northeast Waste Management Officials' Association and the Council of Great Lakes Industries (CGLI). Some points of significance recorded during the conference include:

- The general trend regarding atmospheric mercury levels was consistently shown to be downward.
- The historic large sources of mercury to the atmosphere were incinerators, which had considerable local impact. They no longer exist or have now been controlled.
- The latest modeling results show that little, if any, reduction in fish tissue mercury levels are predicted to result from significant reduction, or even elimination, of remaining local sources. It will take a substantial reduction in long range transport contributions to provide significant fish tissue mercury level reductions.
- The importance of the form of mercury relative to the impact it has in the environment was highlighted by many researchers. Oxidized mercury is of primary concern. Focus should not be placed on total mercury releases.
- Understanding of the health implications of mercury exposure for both humans and the ecosystem is increasing substantially. Better communication of the risks of human exposure is needed without overstating the risks.
- While mercury is a factor in Great Lakes fish consumption advisories, other contaminants are the predominant controlling factor at most locations. For example, the Ontario Ministry of the Environment (MOE) suggested that, for the general population in Ontario, mercury was found to be responsible for between just 1% and 17% of advisories.[4] For most areas, PCBs or other substances are the basis for which advisories will remain in place, even if mercury could be completely eliminated from the system.
- Model results are useful for suggesting policy needs, and the models continue to improve. However, comparisons of model predictions with actual test data are extremely important and must be utilized for good decision-making. For example, measurements have shown that mercury emissions declined by 50% between 1996 and 2008. At the same time, measured deposition rates have declined by only 10%.[5] The models have typically shown higher rates of decline in deposition.
- Reductions of contaminants in environmental media significantly lag reductions in emissions. As a result of emission reductions already made, additional reductions in fish and wildlife levels are predicted to follow. This encouraging news must be highlighted.

[3] NEI (2007). National Emissions Inventories for the U.S. Web site prepared by US EPA. Available at http://www.epa.gov/ttnchie1/trends/.

[4] USEPA & Northeast Waste Management Officials' Association. 2009. Mercury in Canadian Great Lakes Fish: A Concern for Human Consumption? Presentation at 2009 Mercury Science & Policy Conference with a Special Focus on the Great Lakes & Northeast Regions, November 2009. Available at http://www.newmoa.org/prevention/mercury/conferences/sciandpolicy/agenda.cfm.

[5] USEPA & Northeast Waste Management Officials' Association. 2009. Comparison of Fish Tissue, Deposition, & Emission Trends. Presentation at 2009 Mercury Science & Policy Conference with a Special Focus on the Great Lakes & Northeast Regions, November 2009. Available at http://www.newmoa.org/prevention/mercury/conferences/sciandpolicy/agenda.cfm.

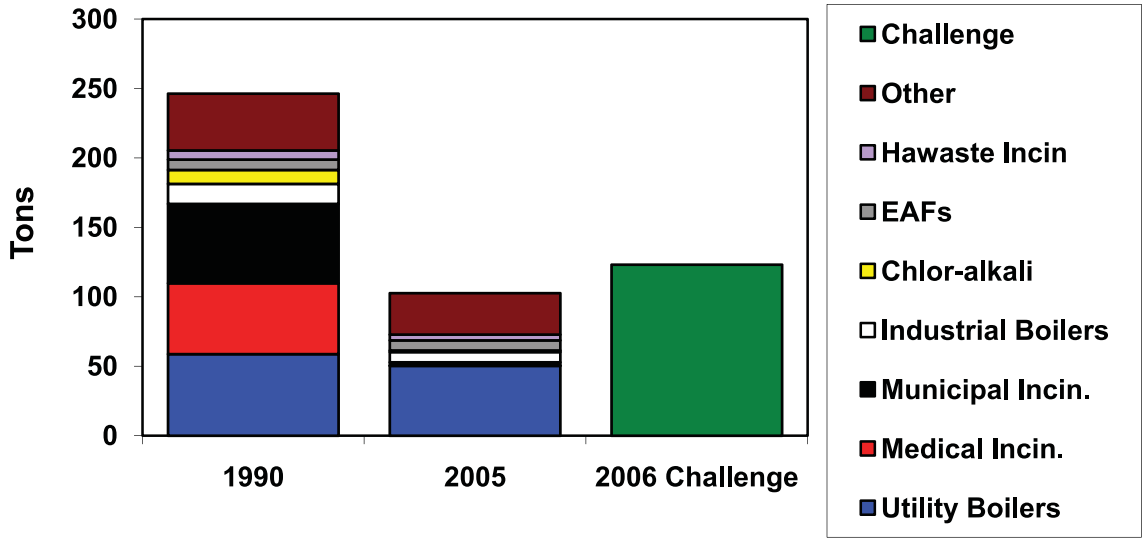


Figure 1-3. U.S. Mercury Emissions: 1990 Baseline and 2005 Estimates, Versus 2006 Challenge [6]

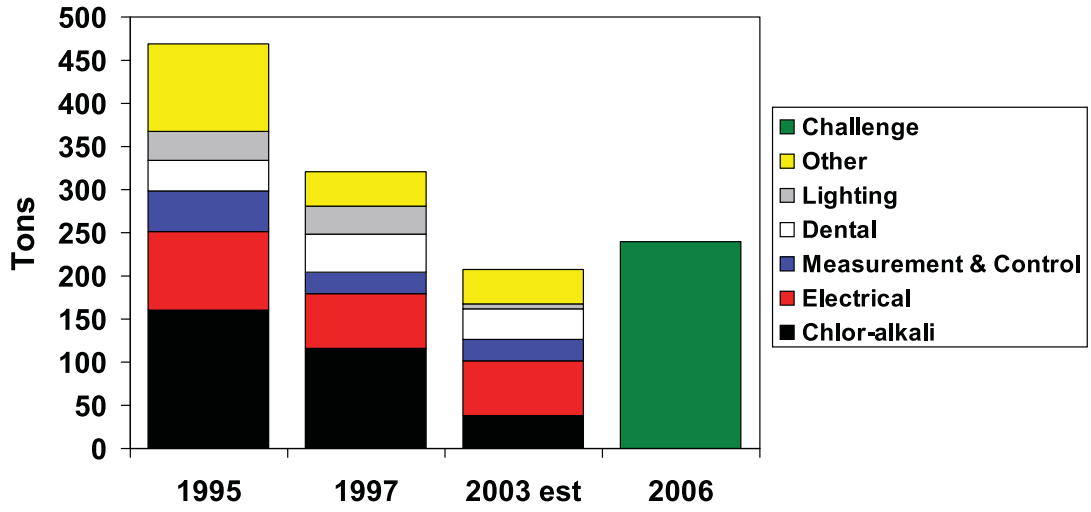


Figure 1-4. U.S. Mercury Use: 2006 Challenge, 2003 and 1997 Estimates, and 1995 Baseline.[7,8,9]

[6] US EPA 1990 NEI for HAPs, revised November 14, 2005; 2005 NATA NEI for HAPs, completed July 1, 2009.

[7] USGS. (1995, 1997). *Minerals Yearbook*. Mercury 1995, by Josef Plachy; Mercury 1997, by Robert G. Reese, Jr. United States Geological Survey. Available at http://minerals.usgs.gov/minerals/pubs/myb.htm.

[8] Chlorine Institute. (2004). *Seventh Annual Report to EPA*. Prepared by The Chlorine Institute, Arlington, Virginia.

[9] NEMA. (2004). National Electrical Manufacturers Association, direct communication.

- The nature of the ecosystem (chemical, biological, and physical characteristics) appears to be the controlling factor regarding the rate of mercury methylation in the environment.
- Given the current advanced state of mercury science, a review of the research agenda is needed to focus attention on the factors most likely to lead to additional progress on mercury issues. Topics for which conference participants suggested research priority included: improved inventories, improved understanding of the role of "new" atmospheric mercury oxidizers (halogens) compared to traditional ones (ozone, hydrogen oxide, etc.), indications that dry mercury deposition is a larger portion of total mercury deposition (wet and dry) than previously thought, and emerging evidence of the reduction of divalent mercury to its elemental form in coal-fired power plant plumes.

U.S. Reduction Activities

Elemental Mercury Collection and Reclamation Program

An Elemental Mercury Collection and Reclamation Program formally began at Bowling Green State University (BGSU) in Ohio in January 1998. The program involves the collection and recycling of uncontaminated elemental mercury that is present in a variety of devices. These sources include thermometers, manometers, barometers, sphygmomanometers (blood pressure measurement devices), mercury-containing heating thermostats, and mercury switches, as well as individual containers of elemental mercury. The program is available and free to individuals, academic institutions, small businesses, industries, medical and dental facilities, emergency response and other governmental agencies, spill response companies, and any additional entity having unwanted, uncontaminated elemental mercury.

Collaborative partners in the program include BGSU, Ohio EPA (Division of Emergency and Remedial Response), Rader Environmental Services, Toledo Environmental Services, and ESCO (Environmental Services and Consulting). The Wood County Emergency Management Agency and the Wood County Health Department have also assisted in this effort. Since the program began, mercury has been removed from numerous sources throughout Ohio as well as from locations in Illinois, Indiana, Michigan, Minnesota, Pennsylvania, Wisconsin, West Virginia, Kentucky, Tennessee, Nebraska, Texas, Georgia, and California. To date, nearly 24,500 lbs of elemental mercury have been collected and recycled.

A more detailed explanation of BGSU's collection and reclamation program is available at: http://www.bgsu.edu/offices/envhs/page18364.html.

Thermostat Recycling Corporation

The Thermostat Recycling Corporation (TRC) reported that it collected over 135,000 mercury-switch thermostats in its national U.S. program in 2008, a 19% increase over 2007. This effort diverted almost 1300 pounds of mercury from solid waste in one year. "TRC collections have now exceeded 100,000 thermostats per year for three years running," said Executive Director Mark Tibbets.

National Vehicle Mercury Switch Recovery Program

The National Vehicle Mercury Switch Recovery Program was initiated in August 2006 through an agreement among vehicle manufacturers, steelmakers, vehicle dismantlers, auto shredders, brokers, the environmental community, state representatives, and US EPA. The program was designed to recover an estimated 40 million mercury-containing light switches from scrap vehicles by promoting a voluntary program and providing incentives for removal of mercury switches from automobiles at the end of life. In February 2008, the program collected its millionth mercury-containing automotive switch, which represents more than 1 ton of mercury that has been removed from the environment. In July 2009, the program's voluntary incentive fund was depleted. Incentive payments continue in states where they are required by law (AR, IL, IA, MA, NJ, RI, UT, MD) or have a state funded program (NC, SC,WA) but ceased in voluntary states. All other aspects of the switch collection program continue, and participants are encouraged to continue

removing switches. The program is scheduled to continue until 2017, based upon an estimate that all available mercury vehicle switches will have been collected by that year, and continues to accept switches at no cost to participants.[10]

Canadian Reduction Activities

Canada-wide Standards for Mercury

Since 2000, Canada-wide Standards (CWS) have been developed by the Canadian Council of Ministers of the Environment (CCME) for specific mercury-containing products and sources of mercury emissions. Currently, standards exist for mercury-containing lamps, dental amalgam waste, emissions from base metal smelting, incinerators, and the coal-fired electric power generation sector. In Canada, progress in reductions related to these standards includes:

- Under the CWS for lamps, the mercury content of fluorescent tubes has decreased by more than 74%.
- As a result of implementation of the Ontario Amalgam Waste Disposal Regulation,[11] 100% of dentists in Ontario installed amalgam separators, which capture waste mercury, before October 2008.
- As a result of CWS on Mercury for Dental Amalgam Waste, 70% of dentists across Canada installed amalgam separators in 2007. In 2002, only 27% of dentists across Canada had installed separators.[12]
- Mercury emissions from coal plants have decreased by approximately 55%, or more than 300 kg, relative to the 1988 baseline, as shown in Figure 1-1. The closure of coal-fired power plants, installation of control technologies, reduced use of coal and the increased use of alternative energy sources for power generation (e.g., hydroelectric, nuclear, wind) have contributed significantly to the reduction in mercury emissions from power plants. Ontario is phasing out coal burning in power plants by 2014. Ontario Power Generation currently has four coal-fired power stations in operation. Ontario provincial regulation ON 496/07 requires that these four plants cease using coal for electricity generation after December 31, 2014.[13]
- Mercury emissions from incineration have decreased by over 70%, or more than 300 kg, relative to the 1998 baseline, as shown in Figure 1-1.

Final Pollution Prevention Notice on Mercury Switches in End-of-Life Vehicles

A Final Notice requiring the preparation and implementation of Pollution Prevention (P2) Plans with respect to mercury releases from mercury switches in end-of-life vehicles processed by steel mills was published in the *Canada Gazette* Part I in December 2007. The P2 Notice requires the targeted vehicle manufacturers and steel mills to prepare and implement P2 plans to reduce mercury releases from the mercury switches in end-of-life vehicles. The targeted companies must consider the participation of each vehicle manufacturer for 15 years after the last model year in which mercury switches were installed, and it requires the participation of targeted steel mills until December 31, 2017. The P2 Notice also requires that a P2 Plan be prepared by June 2008 and implemented by December 2011.

[10] US EPA. (October 07, 2009). National Vehicle Mercury Switch Recovery Program. Available at http://www.epa.gov/mercury/switch.htm.

[11] Ontario (2003). Dentistry Act, 1991; Ontario Regulation 205/94; Part III, Amalgam Waste Disposal Regulation 196/03. Citing Standard Practice of the Profession for Amalgam Waste Disposal, published by the Royal College of Dental Surgeons of Ontario. Also citing Best Management Practices for the Disposal of Dental Amalgam and Mercury Wastes in Ontario, Environment Canada, October 2003. Available at http://www.search.e-laws.gov.on.ca/navigation?file=home&lang=en/.

[12] CCME 2007. Canada-Wide Standards for Mercury. A Report on Compliance and Evaluation-Mercury from Dental Amalgam Waste. A Report on Progress-Mercury Emissions and Mercury-Containing Lamps. 2007.

[13] Government of Ontario, Canada. 2007. e-Laws: Ontario Regulation 496/07. Available at http://www.e-laws.gov.on.ca/html/regs/english/elaws_regs_070496_e.htm.

Pollution Prevention Notice on Dental Amalgam Waste

A Final Notice regarding P2 planning with respect to mercury releases from dental amalgam waste was published in the Canada Gazette Part I on May 8th 2010. The P2 Notice requires targeted dental facilities to prepare and implement pollution prevention plans. They must consider implementing Best Management Practices to reduce mercury releases to the environment in order to contribute to a 95% national reduction in mercury releases from dental amalgam waste relative to a base year of 2000. The P2 Notice also requires that a P2 Plan be prepared by August 2010 and implemented by November 2010.

Risk Management Strategy for Mercury-Containing Products

EC developed a Risk Management Strategy (RMS) to manage mercury-containing products. Mercury can be found in everyday products such as thermometers, compact fluorescent lights, switches and relays, and some measuring devices and batteries. The RMS provides a framework for the development of control instruments to manage the environmental effects of mercury used in products. The objective is to reduce mercury releases to the environment from consumer products to the lowest possible level by prohibiting or limiting the mercury content in new consumer products and by preventing releases from the end-of-life mercury-containing products. EC held public consultations on the proposed RMS in 2007. A consultation document proposing a regulation to implement the objective of the RMS was published in December 2007. In 2008, consultation sessions were delivered to stakeholders from industries, associations, governments, environmental organizations, and health organizations. EC expects to publish a proposed regulation in the Canada Gazette Part I by fall 2010.

For more information on EC's mercury-related initiatives, please visit the "What's New?" section on the Mercury and the Environment website at: http://www.ec.gc.ca/MERCURY/EN/wn.cfm.

Summerhill Impact Builds on Successful "Switch Out" Program

Summerhill Impact (formerly Clean Air Foundation), a Canadian environmental not-for-profit organization, manages two mercury recovery programs in Canada. Switch Out (www.switchout.ca) is Canada's national automotive mercury switch recovery program that operates in partnership with automotive recyclers across Canada. Switch the 'Stat (www.switchthestat.ca) is a residential and commercial thermostat exchange program delivered in partnership with the Heating Refrigeration and Air Conditioning Institute of Canada (HRAI) and their member contractors. Both initiatives aim to reduce the amount of mercury released to the environment from disposal of the end-of-life consumer products, vehicles and thermostats.

Switch Out Program Results

Since the Switch Out program began in 2001, through the voluntary participation of auto recyclers across Canada in British Columbia, Alberta, Ontario, Quebec, and Nova Scotia, more than 352,403 mercury-containing switches have been safely removed from end-of-life vehicles prior to recycling in Canada. This is equivalent to the recovery of approximately 300 kg of mercury. Specifically, since national program funding began in September 2007, approximately 188,699 mercury switches have been recovered, resulting in the safe capture and storage of approximately 160 kg of mercury. More information about the Switch Out program can be found at http://www.switchout.ca.

Switch the 'Stat Program Results

Switch the 'Stat was officially launched by Summerhill Impact (formerly Clean Air Foundation) in partnership with 1,330 heating and cooling contractors in the Province of Ontario. Contractors encourage the installation of energy-efficient programmable thermostats, while simultaneously recovering older mercury-containing thermostats. This diverts the older mercury-containing thermostats from landfills to

safe storage facilities. An old thermostat can contain 2.5 to 10 grams of mercury. Since the launch of the pilot project in April 2006, 20,000 thermostats (containing approximately 78 kg of mercury) have been collected in Ontario. Program partners and funders include Enbridge Gas Distribution, Union Gas, HRAI, Aveitas Inc. (formerly Fluorescent Lamp Recyclers), and Purolator. More information about the Switch the 'Stat program can be found at http://www.switchthestat.ca.

Take Back the Light Program Managed by Recycling Council of Ontario (RCO)

In 2005, the RCO studied and undertook a pilot study with the Grand Erie District School Board, which explored the feasibility of changing the end-of-life management of fluorescent lamps. Building upon this experience, the RCO worked with the larger Toronto District School Board (TDSB) in 2007. In the TDSB pilot, Osram-Sylvania and Wolf Electric and Lighting worked with the RCO to develop a reverse distribution system for spent lamps. The RCO rolled out a Fluorescent Lamp Stewardship program (called Take Back the Light) to the institutional, commercial, and industrial sectors in 2008. Its goal is to work with both sellers and buyers of fluorescent lamps to recover and recycle 10 million fluorescent lamps by 2012 in Ontario. A total of 623,071 fluorescent lamps have been recycled to date. The program managed by RCO will continue to work with industrial, commercial, and institutional sectors to recycle additional fluorescent lamps.

Municipal Hazardous or Special Waste Program in Ontario

On September 22, 2009, the Ontario Minister of the Environment approved the consolidated Municipal Hazardous or Special Waste (MHSW) Program Plan. It expands on the current MHSW program (phase 1), which started July 1, 2008. The MHSW program includes wastes discarded in the residential stream and small quantities in the business stream. The consolidated MHSW Program is scheduled to commence in July 2010 and will accept additional wastes including mercury-containing wastes such as thermostats, mercury switches, mercury-containing measuring devices (e.g., thermometers and barometers), and fluorescent bulbs. The program is a producer-responsibility diversion program that will make industry responsible for full program costs, including the collection and management of wastes.

Next Steps

The Mercury Workgroup has provided input to the development of a draft Great Lakes Mercury Emission Reduction Strategy sponsored by the Great Lakes Regional Collaboration (GLRC). The workgroup is being phased-out, as both Canada and the United States have met their challenge goals. In place of regular workgroup meetings, the GLBTS plans to periodically organize and/or sponsor larger science and policy conferences. The first of these was held in Chicago on November 17-18, 2009. GLBTS progress reports will continue to report on biennial activities related to mercury.

2.0 POLYCHLORINATED BIPHENYLS (PCBs)

Workgroup Status: Active
Canadian Workgroup co-chair: Ken De
U.S. Workgroup co-chairs: Tony Martig and Brad Grams

Lake Huron Beach, Photograph courtesy of The Michigan Travel Bureau

Progress Toward Challenge Goals

U.S. Challenge: Seek by 2006, a 90% reduction nationally of high-level PCBs (>500 ppm) used in electrical equipment. Ensure that all PCBs retired from use are properly managed and disposed of to prevent accidental releases within or to the Great Lakes Basin.

Canadian Challenge: Seek by 2000, a 90% reduction of high-level PCBs (>10,000 ppm) that were once, or are currently, in service and accelerate destruction of stored high-level PCB wastes which have the potential to enter the Great Lakes Basin, consistent with the 1994 COA.

The U.S. and Canada both continue to make progress toward reaching the PCB challenge goals outlined in the Strategy. However, as described below, some data gaps still exist regarding the amount of PCBs in remaining equipment and storage. Information continues to be gathered and assessed by US EPA and EC to determine whether the U.S. and Canadian PCB challenge goals have been met in their entirety. While the U.S. has made progress in reducing the amount of equipment in service containing >500 ppm PCBs, the U.S. is still unable to determine, with accuracy, the status of progress toward the goal due to a lack of information. Based on preliminary data received from EC on the Canadian National Inventory system for Ontario, it appears that Ontario has achieved a 90.2% reduction of high-level PCBs (>10,000 ppm PCB) in storage. For PCBs that are still in service or in use in PCB equipment in Ontario, preliminary analyses indicate that approximately 68 to 70% have been eliminated or destroyed. Further reductions are expected due to the accelerated mandatory phase-out of PCBs in service and in storage as required by Canada's PCB regulations.[14]

The PCB Workgroup is active and continues to pursue reduction opportunities and outreach activities, and plans to prioritize recommendations developed in the 2006 Management Assessment for PCBs, which are outlined below:

» Continue existing Level 1 programs that:

[14] Canada Gazette. (November 4, 2006). PCB Regulations. Proposed under Subsection 93(1) of CEPA, 1999. Canada Gazette Part I, Vol. 140, no. 44. Available at http://www.ec.gc.ca/ceparegistry/documents/regs/g1-14044_r1.pdf.

- Promote decommissioning of PCBs in use/service (PCB equipment and small and large capacitors containing > 50 ppm PCBs).
- Identify and control releases from storage and disposal facilities.

» Promote compliance activities for mandatory phase-out of PCBs in service as required by new Canadian PCB regulations.

» Continue data gathering and assessment to determine additional PCB sources and to plan for future resource commitments.

» Prioritize PCB inventory update and source emission studies.

» These recommendations have been reviewed and accepted by the PCB Workgroup. The workgroup plans to address the following recommendations:

» Review the literature annually for new information on PCB sources and new or updated data on PCB levels and trends in the Great Lakes.

» Prepare annual summary reports on the literature reviews but consider that, even though more information may be published, specific information on PCB releases from some sources are still poorly documented (e.g., contaminated sites, dispersive PCB sources).

Both Canada and the U.S. are evaluating opportunities to comply with the Stockholm Convention (Canada has both signed and ratified the convention; the US is also a signatory, but has not ratified it), which includes international goals to phase-out PCBs.[15] The PCB Workgroup will continue to work with the COA program in order to achieve COA goals in Ontario. [16]

Ontario: Progress Toward the GLBTS Challenge

EC continues to update its inventory information annually. The information below summarizes previously compiled and evaluated inventory information through 2006.

According to EC's 2006 PCB Inventory reports, about 90.2% of previously stored high-level PCB waste had been destroyed (compared to 1993 baseline; see Figure 2-1), and the number of PCB storage sites had been reduced from 1,529 in 1993 to less than 400 (see Figure 2-2). As of 2006, 90% of high-level PCBs in storage were reduced, which exceeded the GLBTS target goal. Less than 400 PCB storage sites remain in Ontario, down from 1,529 in 1993. A new Canadian PCB regulation is accelerating mandatory phase-outs of PCBs in storage and in use.

As noted above, Canada continues to update its inventory of PCBs remaining in in-service equipment. New mandatory reporting requirements will help improve Canada's inventory information. In Ontario at the end of 2006, there were still approximately 2,771 tonnes (in net tonnes) (5.5 million lbs) of high-level PCBs in use/service that needed to be targeted for phase-out (see Figure 2-3). Canada hopes to meet its challenge goal of 90% reduction of high-level PCBs in service (approximately 70% was achieved as of the end of 2006).

United States: Progress Toward the GLBTS Challenge

US EPA uses two sources of information to evaluate the estimated inventory of PCB transformers remaining in use: (1) annual reports submitted by PCB disposers and (2) the PCB Transformer Registration Database. The annual report data has been compiled up to and including 2007. It shows that PCB transformers and PCB capacitors are still being disposed of each year, at an average annual rate of 7500 and 2700 units, respectively,

[15] Stockholm Convention. (May 22, 2001). Stockholm [Sweden] Convention on Persistent Organic Pollutants. Available at http://www.pops.int/.

[16] EC. (2002-2007). Canada-Ontario Agreement Respecting the Great Lakes Basin Ecosystem. Prepared by Environment Canada. Available at http://www.ec.gc.ca/CEPARegistry/documents/agree/Fin-COA07/toc.cfm.

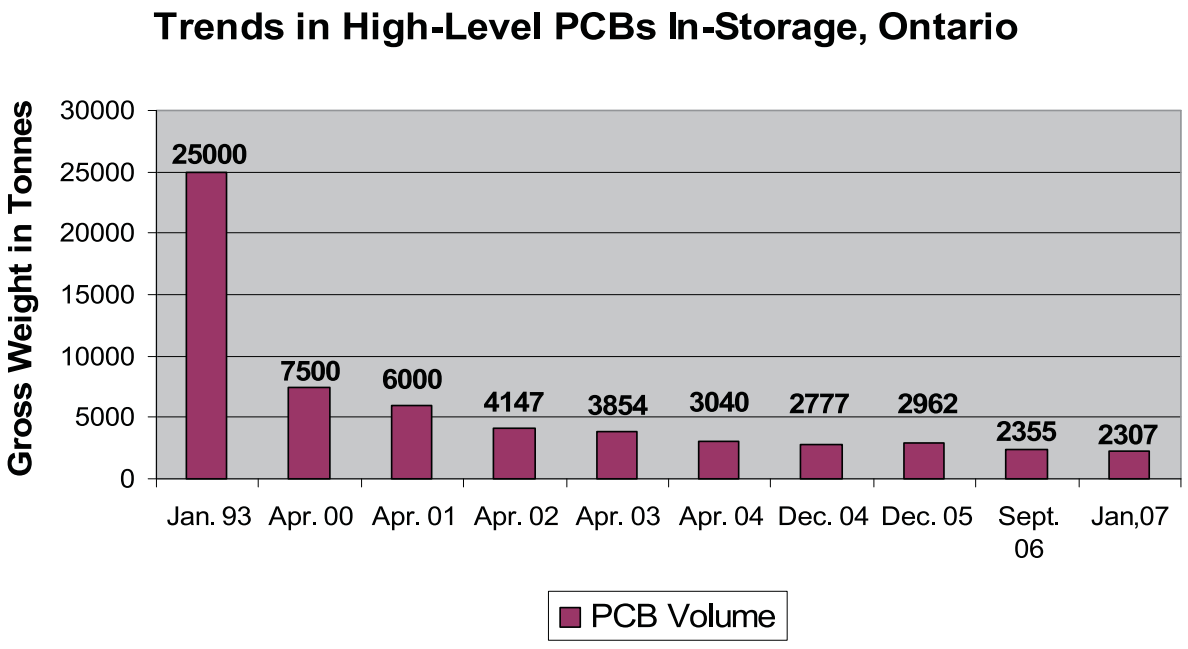


Figure 2-1. **High-Level PCBs (Gross Tonnes) in Storage in Ontario. Source: Environment Canada and Ontario Ministry of Environment PCB Database**

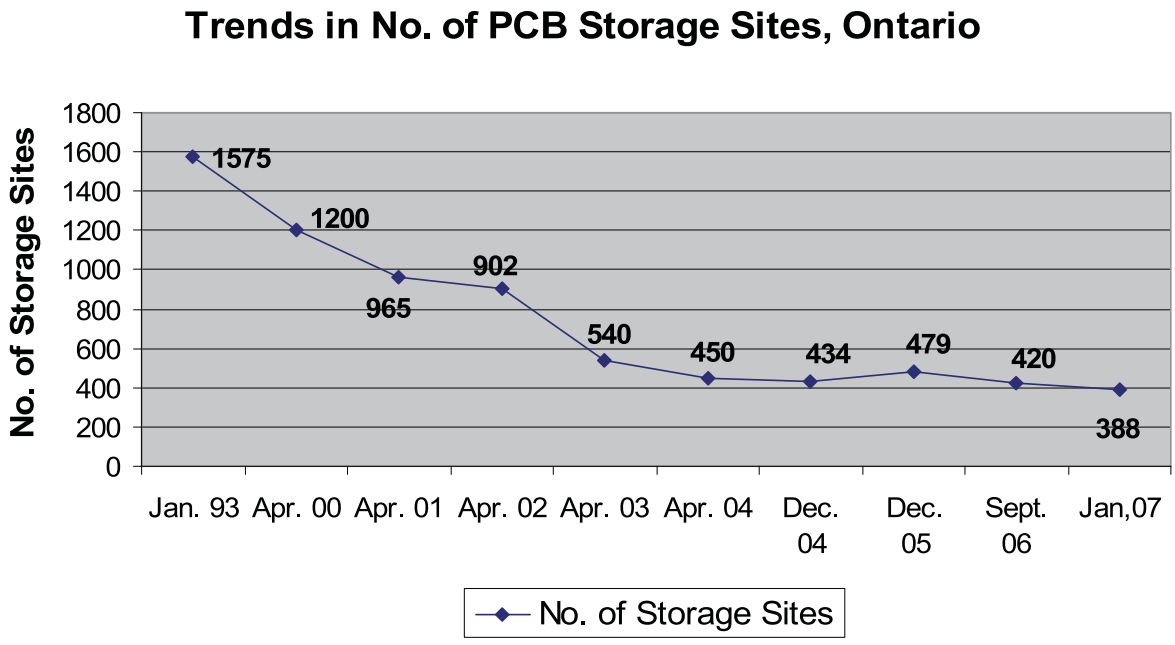


Figure 2-2. **Trends in Number of PCB Storage Sites in Ontario. Source: Environment Canada**

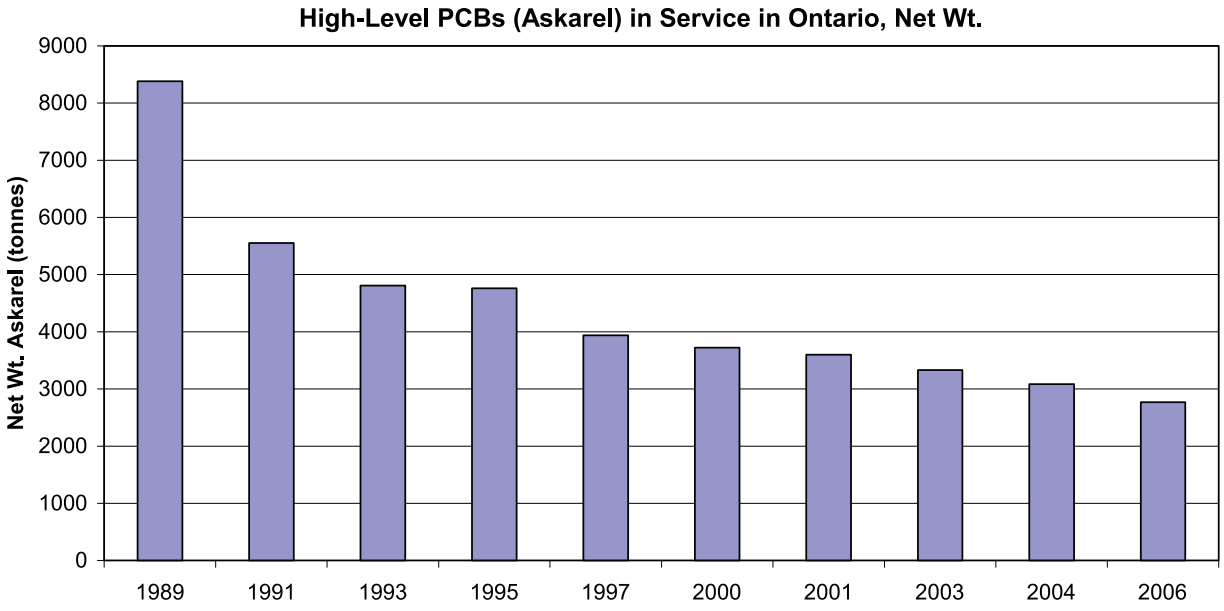


Figure 2-3. Trends in High-Level (Askarel) PCBs (Net Tonnes) in Service in Ontario. Source: Environment Canada

for the past five years. Based on the annual report data through 2007, an estimated 64,312 PCB transformers and 1,293,000 large PCB capacitors remained in use at the end of 2007. The estimates for the amount of equipment remaining in use in 2007 were obtained by subtracting the annual disposal data from the 1994 estimated baseline.[17] However, according to the PCB Transformer Registration Database (updated in January 2008), only about 14,150 PCB transformers were registered with US EPA. Although the data from the annual reports is important for compliance purposes and can be used to compare trends for and between facilities and years, it is not particularly useful for determining the amount of PCB equipment that is remaining in service. In the absence of more specific or detailed data, US EPA will continue to use this data to provide some insight into the amount of PCB equipment that may remain in service.

Workgroup Activities

Workgroup Meetings

The PCB Workgroup met on December 3, 2008. This meeting focused on four topics: (1) current PCB data trends for the Great Lakes; (2) EC, the Minnesota Pollution Control Agency (MPCA), and US EPA collected data trends and challenge goals; (3) regulatory framework agendas, and (4) the PCB Management Assessment. Much of the discussions centered around the issue of providing better accessibility for acquired or developed data and programs.

The PCB Workgroup also met on December 1, 2009. This meeting focused on several topics: (1) an update of the PCB equipment inventory; (2) the anticipated US EPA Advance Notice of Proposed Rulemaking on the current remaining authorized uses of PCBs; (3) PCBs in caulk; (4) PCBs in used oil; and (5) an initiative to track potential remaining sources of PCBs based on a PCB sales list provided by Monsanto.

The main topic areas discussed at the meetings which have follow-up activities are identified later in this chapter.

[17] The baseline was determined by the US EPA Office of Prevention, Pesticides and Toxic Substances based on PCB registration data, industry- and association-provided estimates, and other government-acquired data.

PCB Management Framework

The PCB Workgroup distributed the final Management Assessment for PCBs, dated January 2007, at its December 2008 workgroup meeting and discussed the final management outcome from the assessment. As identified in the Management Assessment, the PCB Workgroup will retain an active Level 1 status and as such, continue to pursue the decommissioning of PCBs in use and/or service. The PCB Workgroup will also pursue the following activities identified in the Management Assessment:

» Further data gathering and assessment to determine additional PCB sources and to consider where and how to focus resources;

- Collect better information on PCB sources, including updating the PCB inventory;
- Review literature annually for new information on PCB sources and new or updated data on PCB levels and trends in the Great Lakes.

» Prepare annual summary reports on the literature reviews but consider that, even though more information may be published, specific information on PCB releases from some sources are still poorly documented (e.g., contaminated sites, dispersive PCB sources).

U.S. Reduction Activities

US EPA Advance Notice of Proposed Rulemaking on PCBs

US EPA is reevaluating the current remaining authorized uses of PCBs and is planning to issue an Advance Notice of Proposed Rulemaking (ANPR) on PCBs. For background on the ANPR, Section 6(e)(2) of the Toxic Substances Control Act (TSCA) prohibits, among other activities, the distribution in commerce and use of PCBs in a manner other than in a totally enclosed manner, unless the US EPA Administrator authorizes such activity by rule. To make such an authorization, the US EPA Administrator must find that the activity will not present an unreasonable risk of injury to health or the environment. US EPA is reevaluating its TSCA PCB use and distribution in commerce regulations at 40 CFR Part 761 subparts B and C, to address: (1) the use, distribution in commerce, marking and storage for reuse of liquid PCBs in equipment; (2) the use of air, gas, and liquid pipelines and transmission systems containing or contaminated with PCBs; (3) the use of non-liquid PCBs in carbonless copy paper; and (4) the use and distribution in commerce of PCBs in porous surfaces. US EPA is also reevaluating certain definitions in 40 CFR section 761.3. In the ANPR, US EPA will solicit written comments on these and other areas of the PCB use regulations. However, US EPA is not soliciting comments on the PCB disposal regulations in this notice. The ANPR is tentatively scheduled to be announced in early 2010, and US EPA is planning to have several public meetings on the ANPR, including one in Chicago.

U.S. PCBs-in-Building Materials Program

In September 2009, US EPA began outreach work for schools and childcare facilities related to PCBs-in-Building Materials (also known as "PCBs-in-Caulk"). While the program is relatively new, baseline educational materials are now available and guidance is currently being developed to assist any facility with building materials or debris having potential PCB contamination (caulk containing PCBs); however, the highest priority facilities would be those with children in day-to-day attendance.

For some states where TSCA PCB wastes are also listed as Resource Conservation and Recovery Act (RCRA) hazardous wastes, such as Minnesota, additional guidance and discussion over the following year will be crucial to assisting stakeholders. The PCB Workgroup will share information on this effort, as possible, as source and emission reductions are voluntarily made.

At the December 1, 2009, PCB Workgroup meeting, the US EPA workgroup co-leads provided an overview of the current issue and available information, which is also available at the following website:

http://www.epa.gov/epawaste/hazard/tsd/pcbs/pubs/caulk/index.htm.

U.S. PCB Data Collection Efforts

The U.S. is continuing work on the identification of potential abandoned and contaminated sites through novel applications of older datasets. Through a comparison and harmonization of several older datasets (e.g., Monsanto sales and distribution lists from 1970-1975) with other datasets (e.g., RCRA and TSCA generator notification datasets), the U.S. hopes to better locate and identify potential PCB sites that may be of concern.

The data are being used as a starting point in targeting potential sites of PCB concern. Since the data have not been evaluated completely to date (due to data, funding, and resource considerations), specific sources or proposed pathways have not been identified at this time. Prior to beginning workgroup efforts, US EPA PCB workgroup members will develop a more detailed plan for employing older datasets and will review this with stakeholders. For instance, in narrowing and focusing efforts by applying current work to high priority areas (e.g., environmental justice areas, Great Lakes Areas of Concern, etc.), it is expected that the data may be better evaluated and analyzed.

PCBs in Used Oil

Related to the aforementioned efforts to collect data and identify potential sources of PCBs, US EPA initiated an informal evaluation of occurrences of PCBs at regulated levels being found in the used oil recycling industry. Used oil can be any type of oil, but it is generally considered to be used motor/engine oil(s). US EPA Region 5 has found that, in the past several years, there have been at least 7 occurrences across the U.S. where PCB contaminated oil was found in the used oil recycling sector, shipped to used oil facilities as non-PCB oil for recycling or processing. The sources of these PCBs have varied; some sources come from criminally investigated facilities (who are trying to dilute PCBs), others come from facilities decommissioning manufacturing engines/processes, and others from unknown sources of used oil that contain PCBs.

Some of these occurrences resulted in hundreds of thousands of gallons, or a million gallons of oil becoming contaminated and therefore having to be managed and disposed of as a regulated PCB waste. US EPA will follow up on this issue with representatives of the used oil industry to better understand the extent and potential sources of this problem and to determine ways to better respond, utilizing "lessons learned" from these incidents and possibly by identifying protocols to address PCB-contaminated waste materials.

PCB Software – Financial Analysis of PCB Transformer Phase-Outs – A Study of the Costs and Benefits of PCB Phase-Out

Under a grant from US EPA, EMA Research & Information Center, subcontractor to the Tellus Institute, developed a spreadsheet tool to determine and compare the costs of phasing out PCB transformers against the costs of continued use. The tool was developed with the input of industry representatives and was based on actual case study information. During the December 6, 2006, PCB Workgroup meeting and GLBTS Stakeholder Forum, Dr. Deborah Savage of EMA Research & Information Center discussed and gave a demonstration on the PCB transformer phase-out tool. Some of the major cost drivers and considerations were: the transformer age, size, type and rating; the fluid volume and PCB concentration; the location and accessibility of the equipment; spill containment and fire prevention; equipment reliability and importance; and regulatory compliance. The software specifically enables a firm to conduct an itemized financial assessment for the scenarios of keeping, removing, and retrofilling a PCB transformer, including such factors as net present value and payback, depreciation, taxes, inflation, and discounting.

The tool is currently available by contacting the US EPA co-leads for the PCB Workgroup. The workgroup is discussing options for marketing the tool and making it available online. CGLI has offered to make the software tool available to its constituents and other interested parties.

Canadian Reduction Activities

Canadian PCB regulations[18] set deadlines for ending the use and storage of PCBs, consistent with Canada's obligations and international agreements. The regulations aim to achieve accelerated destruction and phase-outs of PCB, as well as mandatory reporting and labeling of PCB-containing equipment. The new regulations require that equipment containing high-level PCBs (over 500 ppm) and low-level PCBs (50 to 500 ppm) in sensitive locations must be phased-out by December 2009. They also limit the maximum duration of storage by generators to 1 year, to 1 year at authorized transfer stations, and to 2 years at disposal/destruction facilities. Mandatory annual reporting to a federal online reporting system will provide current PCB inventory data. Training videos and factsheets explaining the online reporting system are available on EC's website. More information concerning this regulation can be accessed at:

http://www.ec.gc.ca/CEPARegistry/regulations/detailReg.cfm?intReg=105.

The Canadian government conducted 10 information workshops and question and answer sessions across Ontario during 2009. There are plans to conduct a few more in 2010 in Northern Ontario or as requested.

Next Steps

The workgroup and government agencies plan to continue seeking PCB reduction commitments and evaluate PCB Management Assessment recommendations for implementation.

PCB Reduction Commitments

The PCB Workgroup will continue seeking commitments to reduce PCBs through PCB reduction commitment letters and other PCB phase-out efforts, and to publicize other significant voluntary achievements in PCB reductions as information on such achievements is available.

Both EC and US EPA will also pursue outreach and education on the regulations related to using PCBs, final PCB phase-out regulations in Canada, and the Advanced Notice of Proposed Rulemaking in the U.S.

PCB Management Assessment Recommendations

The Final Management Assessment for PCBs was discussed at the PCB Workgroup meeting of December 3, 2008. The workgroup has begun working on the recommendations presented in the report.

Because the workgroup has determined that several data issues exist (e.g., data quality and comparability issues as well as completeness) regarding PCB sources, levels, and trends in the environment, future workgroup activities will include further evaluation of the available data before final conclusions are made.

At this time, the workgroup recommends that PCBs should continue an active Level 1 status. As such, work targeting PCB-containing equipment in service should continue (such as outreach to industry), due to the potential for the equipment to be a source of future releases. This work should be coordinated with other efforts.

However, a priority will be placed on collecting and assessing a more complete set of data on PCB sources and environmental levels. The primary goals of this exercise will be to: (1) prioritize the remaining PCB sources (better defining relative source contributions), (2) clarify PCB trends and impacts on the environment, and (3) assess the ability of the GLBTS to effect further reductions.

The PCB Workgroup will continue to gather data to identify and determine relative contributions of PCBs to the environment from known and potential sources. Once sufficient progress on this work is made, a better determination can be made of the activities that can be undertaken, and by whom, to reduce releases from particular sources.

[18] Environment Canada CEPA Environmental Registry: http://www.ec.gc.ca/CEPARegistry/regulations/detailReg.cfm?intReg=105.

The workgroup will also consider future resource commitments by workgroup members for any future work.

Some of the specific activities regarding potential sources the PCB Workgroup will pursue include (as discussed above):

- Continuing work on the identification of potential abandoned and contaminated sites through novel applications of older datasets;
- Follow-up with representatives of the used oil industry to better understand the extent and potential sources of the finding of PCBs at regulated levels in used oils shipped for recycling.

In addition, the PCB Workgroup will update its website (or evaluate other/better ways) to share information on the above efforts.

Finally, although the PCB Workgroup will retain an active Level 1 status, it does not plan to continue having face-to-face meetings. Instead, the co-lead will arrange conference calls to discuss and follow-up on specific/focused activities during the course of the year.

3.0 DIOXINS/FURANS

Workgroup Status: Inactive
Canadian Workgroup co-chair: Anita Wong
U.S. Workgroup co-chair: Erin Newman

Prairie Phlox, Photograph courtesy of U.S. EPA

Progress Toward Challenge Goals

U.S. Challenge: Seek by 2006, a 75% reduction in total releases of dioxins and furans (2,3,7,8-TCDD toxicity equivalents) from sources resulting from human activity. This challenge will apply to the aggregate of releases to the air nationwide and of releases to the water within the Great Lakes Basin.

Canadian Challenge: Seek by 2000, a 90% reduction in releases of dioxins and furans from sources resulting from human activity in the Great Lakes Basin, consistent with the 1994 COA.

The U.S. has met its goal of a 75% reduction in dioxin/furan releases (at 89% as of 2000), and Canada has reached its 90% dioxin/furan reduction goal, by achieving a 90% reduction (230 grams) of total releases within the Great Lakes Basin in 2007, relative to the 1988 Canadian baseline. Now that the GLBTS challenge goals have been met for both countries, the Dioxin Workgroup is suspending further active work. However, both countries will continue to monitor dioxin in the environment, investigate dioxin data as available, and look for reductions in uncontrolled combustion sources such as burn barrels.

During the past year, US EPA and EC have worked to reduce burn barrels and household garbage burning, which is the largest quantified source of dioxin emissions in both countries. US EPA continued to distribute its toolkit for municipalities, which is available online (http://www.iisgcp.org/learnnot2burn/). EC also conducted outreach and widely distributed burn barrel information materials. Due to the change in status of the Dioxin Workgroup, the Burn Barrel Subgroup will continue to operate, but under HCB/B(a)P Workgroup leadership. Other sources of uncontrolled combustion such as outdoor wood-fired boilers, wood stoves, and agricultural burning remain a concern for dioxins, HCB, and B(a)P.

Ontario: Progress Toward the GLBTS Challenge

Canada has met the goal of a 90% reduction in releases of dioxins/furans. This reduction is based on the 2007 release inventory update for Ontario sources,[19] which estimates a total annual dioxin/furan release of 25.6 grams. Figure 3-1 illustrates

[19] Point sources are mostly based on: EC. (2005). National Pollutant Release Inventory Data (NPRI) data. Web site of Environment Canada. Available at http://www.ec.gc.ca/pdb/npri/npri_dat_rep_e.cfm#highlights.

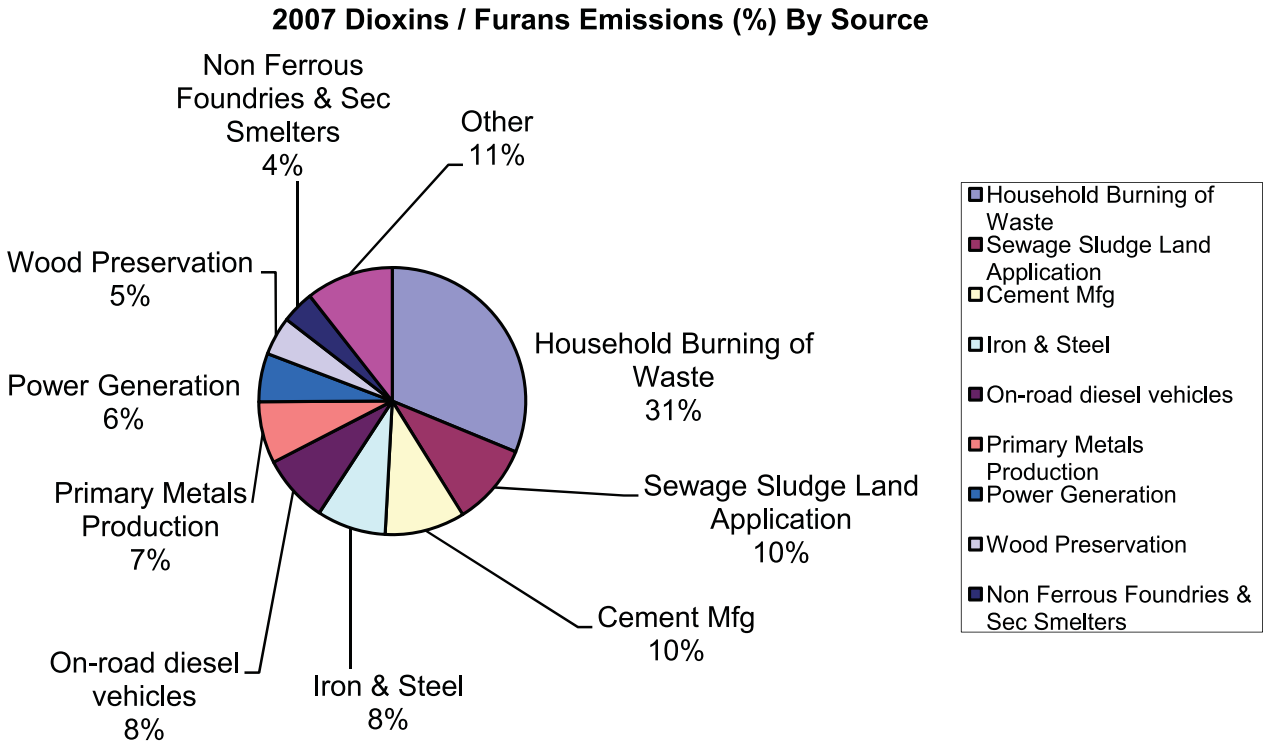


Figure 3-1. **Dioxin/Furan Emissions by Sources (%), 2007. Source: Environment Canada, Ontario Region**

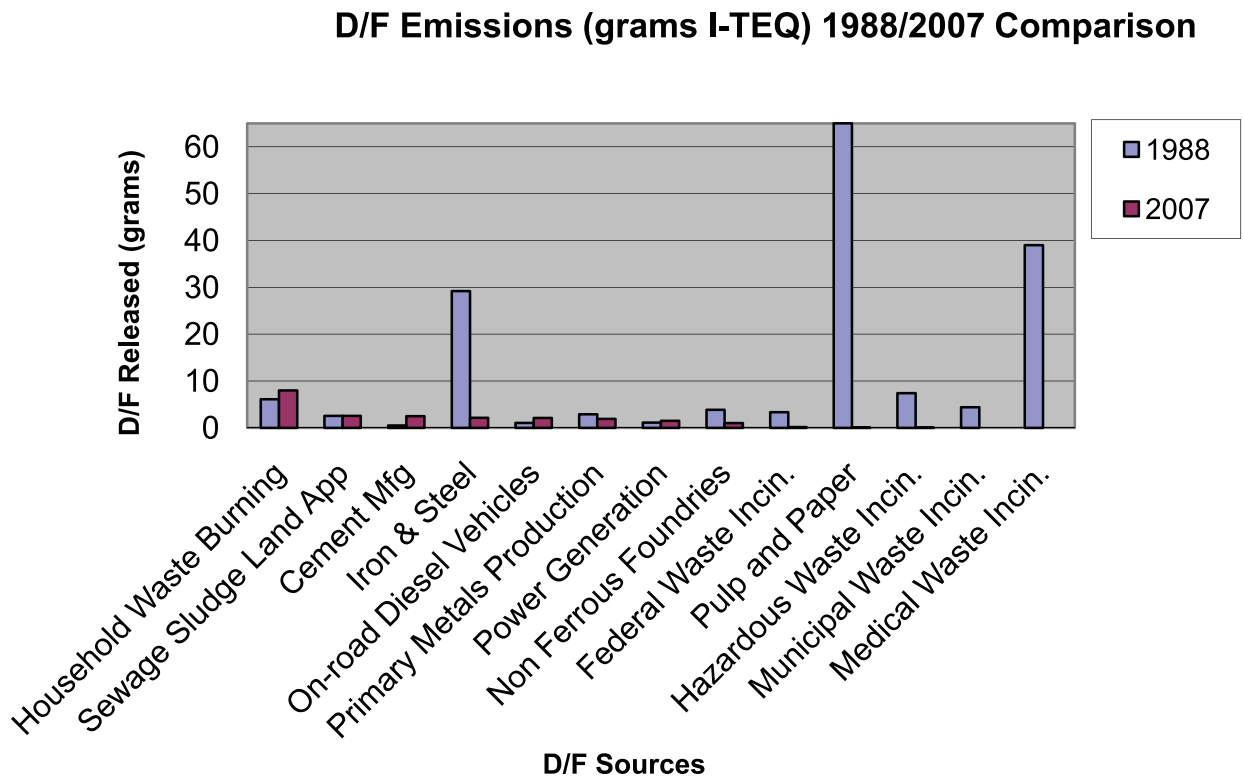


Figure 3-2. **Comparison of Dioxin/Furan Emissions (grams), 1988 and 2007. Source: Environment Canada, Ontario Region**

the remaining Ontario dioxin/furan release sources for 2007. Figure 3-2 illustrates reductions in the top Canadian (Ontario) dioxin/furan release sources since 1988.

Several source sectors offer opportunities for potential reductions. For example, efforts by the GLBTS Burn Barrel Subgroup, such as education and outreach, can help reduce emissions from household garbage burning, the largest source of dioxin emissions in Ontario. In addition, Ontario has drafted a regulation to phase-out coal-fired power units by 2014, and the last iron sinter plant was shut down in 2007. Canada will continue to track increases in emissions and emerging sources of dioxins/furans.

The top source of dioxins/furans continues to be household burning of waste. The Burn Barrel Subgroup remains active in addressing this source. The contribution of dioxin/furan releases from the remaining sources ranges from less than 1% to 10%. Most of these sources are being addressed directly or indirectly through existing initiatives, as indicated in Table 3-1.

Table 3-2 includes some of the sources in the "other" category that currently release less than 1 g I-TEQ/year. The waste incineration and pulp and paper sectors have been dominant sources in the past but have since made significant reductions in releases of dioxins/furans through a combination of control instruments and facility shutdowns.

Table 3-1. 2007 Total Dioxin/Furan Releases in Ontario (g I-TEQ/year)

Sector	1988 D/F Total	2007 D/F Total	2007 Percent	Initiatives
Household Burning of Waste	6.10	8.00	31.2%	Burn Barrel Subgroup
Sewage Sludge Land Application	2.55	2.55	9.9%	MOE/EC 2004 study showed insignificant impact to environment
Cement Mfg	0.51	2.48	9.7%	Current fed/prov developing stds for CAC, may look into toxics
Iron & Steel	29.20	2.14	8.4%	CWS for EAF and Iron sinter, the last sinter plant shut down by 2007
On-road diesel vehicles	1.06	2.11	8.2%	Cobenefits from regs. on vehicle emissions (CAC) and fuel quality
Primary Metals Production	2.90	1.92	7.5%	CEPA Code of Practice and P2 Plan
Power Generation	1.13	1.50	5.9%	Ontario to phase-out coal-fired power plants by 2014
Wood Preservation	5.40	1.20	4.7%	PMRA – levels of D/F dropped significantly from late 90s in PCP mfg
Non Ferrous Foundries & Sec Smelters	3.86	1.01	3.9%	Current EC studies examining sectors
Other	203.19	2.71	10.6%	See below
Ontario Total	**255.90**	**25.63**	**100.0%**	

Source: Environment Canada

List of Acronyms Used:

CAC: Criteria Air Contaminants
CEPA: Canadian Environmental Protection Act
CWS: Canada-Wide Standard(s)
EAF: Electric Arc Furnace
EC: Environment Canada
MOE: Ministry of the Environment (Ontario)
PMRA: Pest Management Regulatory Agency

Table 3-2. Other Sources of Dioxins/Furans in Ontario (g I-TEQ/year)

Sector	1988 D/F Total	2007 D/F Total
Residential Wood Combustion	0.84	0.82
Crematorium	NA	0.79
On-road Gasoline vehicles	0.14	0.20
Federal Waste Incineration	3.34	0.16
Pulp and Paper	147	0.04
Hazardous Waste Incineration	7.40	0.00
Municipal Waste Incineration	4.40	0.00
Medical Waste Incineration	39	0.00

Source: Environment Canada

United States: Progress Toward the GLBTS Challenge

According to *An Inventory of Sources and Environmental Releases of Dioxin-Like Compounds in the United States for the Years 1987, 1995, and 2000*, the U.S. has achieved an 89% reduction in dioxin releases nationally. A significant portion of those reductions are a direct result of the maximum achievable control technology (MACT) standards enacted under the Clean Air Act (CAA). For example, MACT standards reduced municipal waste combustion emissions from 8,905 grams TEQ in 1987 to 83 grams in 2000. Other source categories with significant reductions resulting from the enactment of MACT standards include Medical Waste Incinerators (MWIs), hazardous waste-burning cement kilns, and secondary copper smelting. These reductions result from a combination of changes in processes and equipment to comply with standards, pre-existing actions in the design and retrofitting of facilities, and facility closures. The total U.S. inventory for dioxin releases has dropped from 13,965 to 1,422 g TEQ_{DF}-WHO_{98}/year. These figures, however, do not reflect full implementation of the MACT standards for medical waste incinerators. So while that source is shown as the second largest source of dioxin releases, US EPA has found substantial reductions while monitoring MACT implementation in subsequent years. It is now clear from these inventory figures that the largest source of quantified dioxin releases is household garbage burning.

The U.S. has not conducted a dioxin inventory since 2000. However, revisions to the 2000 inventory are underway. Additionally, US EPA Administrator Lisa Jackson has publicly committed to completion of US EPA's "*Exposure and Human Health Reassessment of 2,3,7,8-Tetrachlorodibenzo-p-Dioxin (TCDD) and Related Compounds*", more commonly referred to as the Dioxin Reassessment by the end of 2010.[20]

Reduction Activities

Burn Barrels and Household Garbage Burning

The use of burn barrels and other household garbage burning methods remains a high reduction priority for the workgroup. Household garbage burning is the largest quantified source of dioxin emissions in both countries. The practice of household garbage burning typically is carried out in old barrels, open pits, wood stoves, or outdoor boilers. The Burn Barrel Subgroup is working to address this issue through continued outreach and education. However, the subgroup now reports to the HCB/B(a)P Workgroup due to the inactive status of the Dioxin Workgroup.

Over the past two years, US EPA developed a web-based burn barrel toolkit entitled *Learn Not to Burn*, which provides resources for local officials to reduce trash burning in their communities. The toolkit includes individual fact sheets for each state and case studies of efforts to reduce household garbage burning in various communities. The toolkit is available free of charge online at http://www.iisgcp.org/learnnot2burn/.

In Ontario, open burning outreach material is being developed for Canadian citizens and for the building industry. Representatives attended the Spring 2009 Toronto Cottage Life show to share information on open burning. The show attracted about 27,000 visitors. EC's dioxin brochure entitled, *What Goes*

[20] Administrator Lisa Jackson testimony, Hearing on Scientific Integrity, before the U.S. Senate Environment and Public Works Committee, June 9, 2009, available at http://www.epa.gov/ocir/hearings/testimony/111_2009_2010/2009_0608_lpj.pdf.

Up Must Come Down, was distributed at the show. EC plans to include open burning and burn barrel materials on the EC website in the near future.

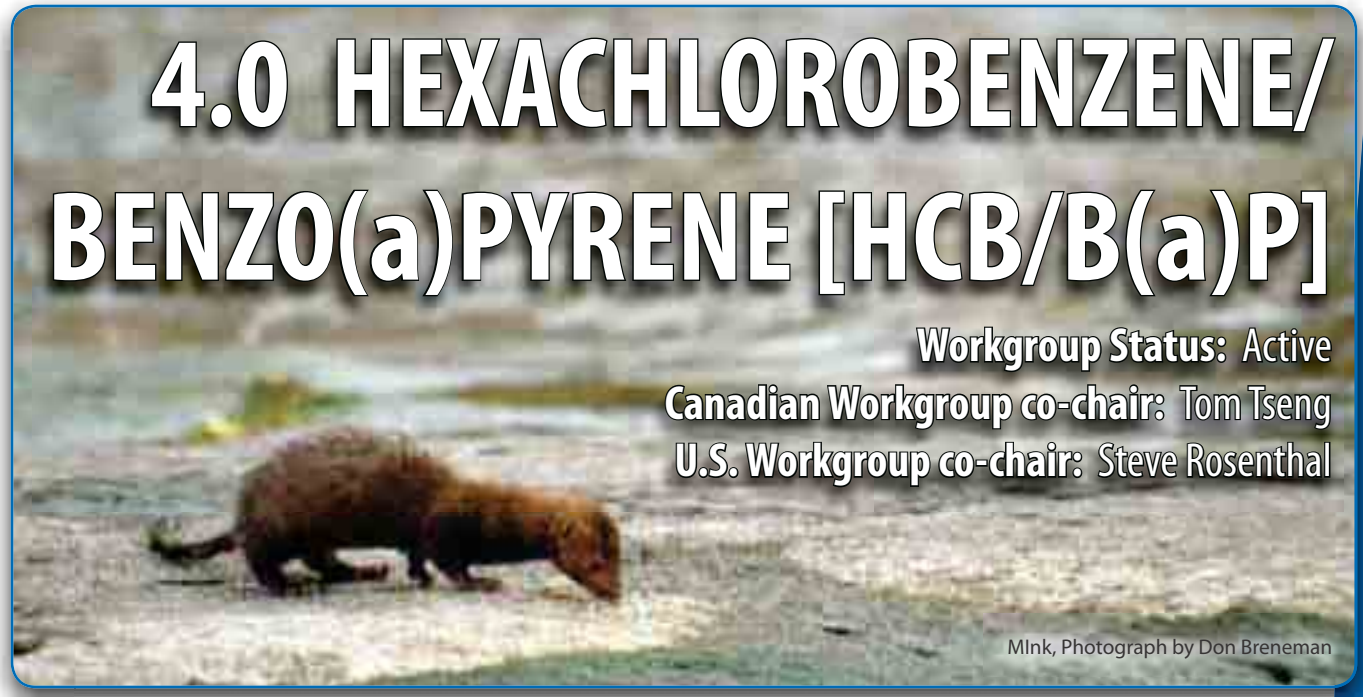
Great Lakes states and tribes are continuing activities, consistent with the Burn Barrel Subgroup's Household Garbage Burning Reduction Strategy, to educate residents and influence behavioral change, supported by infrastructure and the institution of local by-laws. Of particular note, the New York State Department of Environmental Conservation (NYSDEC) passed a statewide ban on open burning. This new rule went into effect October 19, 2009, and prohibits burn barrels, as well as leaf burning and agricultural plastic burning.

Next Steps

The GLBTS challenge goals have been met for both countries. The Dioxin Workgroup considered its ability to affect remaining sources of dioxin to the Great Lakes Basin and decided to suspend further work but to continue Burn Barrel Subgroup activities (including the Burn Barrel subgroup website). The Burn Barrel Subgroup now reports to the HCB/B(a)P Workgroup. The Dioxin Workgroup co-chairs will continue to track sources of dioxin through release inventories and environmental monitoring data. Canada is undertaking a modeling project to assess the global transport of dioxins/furans and its impact to Canada and the North American region. The co-chairs may reactivate the workgroup if warranted as new issues arise. The co-chairs will also investigate potential opportunities to reduce agricultural waste burning, through the Burn Barrel Subgroup, and other poorly characterized sources of dioxins/furans.

4.0 HEXACHLOROBENZENE/ BENZO(a)PYRENE [HCB/B(a)P]

Workgroup Status: Active
Canadian Workgroup co-chair: Tom Tseng
U.S. Workgroup co-chair: Steve Rosenthal



Mink, Photograph by Don Breneman

Progress Toward Challenge Goals

U.S. Challenge: Seek by 2006, reductions in releases that are within, or have the potential to enter, the Great Lakes Basin, of HCB and B(a)P from sources resulting from human activity.

Canadian Challenge: Seek by 2000, a 90% reduction in releases of HCB and B(a)P from sources resulting from human activity in the Great Lakes Basin, consistent with the 1994 COA.

The U.S. and Canada have both made significant reductions in HCB/B(a)P emissions to the Great Lakes Basin.

Ontario: Progress Toward the GLBTS Challenge

HCB Reduction

From a 1988 baseline, Canada has reduced HCB emissions to the Great Lakes Basin by approximately 71% as of 2007 (the latest year for which data are available).[21] Figure 4-1 shows the release estimates and progress achieved toward meeting the 90% reduction target.[22] Over 80% of the reductions achieved to date are due to:

- Lower residual HCB levels in pesticides and reduced usage of certain pesticides known to contain HCB;
- Implementation of a CWS for waste incinerators and the closure of solid waste incinerators, such as Hamilton's Solid Waste Area Reduction Unit (SWARU);
- Reductions reported by the iron and steel sector and the closure of Algoma's Wawa sintering facility; and
- Process changes within Ontario's chlorinated chemical manufacturing sector.

Canada's 2007 HCB releases in the basin are estimated at 32 pounds (14.7 kilograms). Major sources are pesticide application, household waste burning, and ferric/ferrous chloride use.

[21] Emission estimates for 2007 were slightly higher than those for 2006 due to higher reported industrial release emissions.

[22] Based on "Hexachlorobenzene Sources, Regulations and Programs for the Ontario Great Lakes Basin 1988, 1998 and 2000 Draft Report (No. 1), July 13, 2000" prepared for Environment Canada by Benazon Environmental Inc., with releases updated by Environment Canada - Ontario Region, based on NPRI facility release data, recent sector release assessments, and pesticide application release information received from Health Canada's Pest Management Regulatory Agency on August 29, 2005.

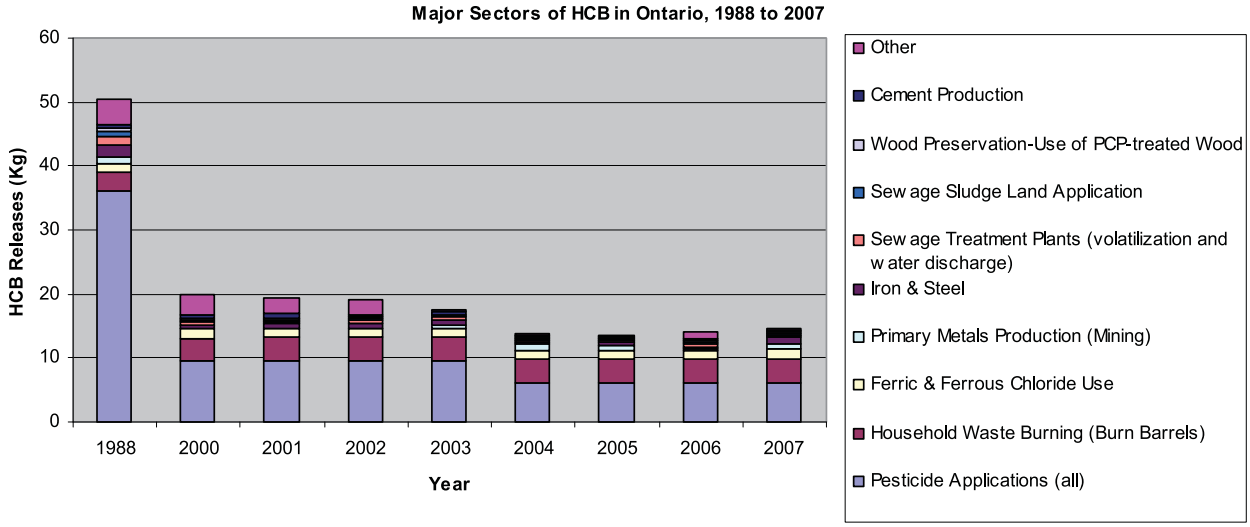


Figure 4-1. Estimated HCB Releases (to Air and Water) in Ontario by Sector, 1988-2007. Source: Environment Canada (Environmental Protection Operations Division – Ontario Region) Inventory as of November 2009

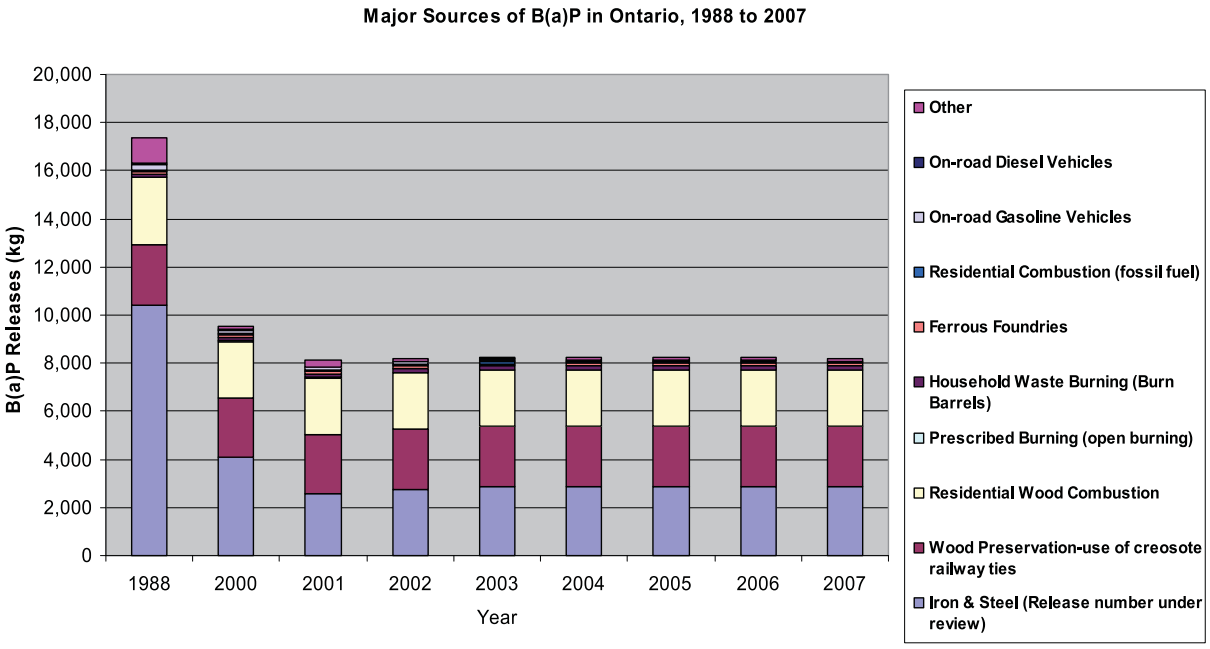


Figure 4-2. Estimated B(a)P Releases in Ontario by Sector, 1988-2007. Source: Environment Canada (Environmental Protection Operations Division – Ontario Region) Inventory as of November 2009

B(a)P Reduction

From a 1988 baseline, Canada has reduced B(a)P emissions to the Great Lakes Basin by approximately 53% as of 2007 (the latest year for which data are available). Figure 4-2 shows the release estimates and progress achieved toward meeting the 90% reduction target.[23] Most of the B(a)P reductions achieved to date have resulted from the following activities:

- The iron and steel sector's implementation of a best practices manual entitled "Environmental Best Practice Manual for Coke Producers – Controlling and Reducing Emissions of Polycyclic Aromatic Hydrocarbons (PAH) from Metallurgical Coke Production in the Province of Ontario," which is consistent with EC's "Environmental Code of Practice for Integrated Steel Mills";[24]
- Decreases in estimated wood consumption; however, reliance on wood heat is expected to increase due to rising oil and gas costs;
- Implementation of control technologies by the petroleum refining sector; and
- Decreases in creosote-treating activities and shutdown of the Northern Wood Preservers Inc. facility in Thunder Bay.

Canada's 2007 B(a)P releases in the basin from anthropogenic sources are estimated at 17,969 pounds (8,168 kilograms). Major sources are residential wood combustion and the use of creosote-treated railway ties. The release number for the steel manufacturing sector is under review due to changes in the sector's release estimation methods.

United States: Progress Toward the GLBTS Challenge

From a 1990 baseline, the U.S. has reduced releases of HCB from approximately 8,519 pounds in 1990 to 2,911 pounds in 1999. From 1999 to 2002, HCB emissions were reduced by an additional 28%. Figure 4-3 shows national HCB release estimates and progress achieved between 1990 and 1999.[25] This reduction is mainly attributed to lower residual HCB levels in pesticides, along with reduced HCB emissions from chlorinated solvent production and pesticide manufacturing. These three categories combined account for roughly 5,000 pounds per year of HCB reductions.

Differences in the 1990 and the 1999 emission inventories and source categories complicate the determination of the exact emission reductions that have occurred. The inventories represent the best emission estimates that are available and provide a useful snapshot of HCB emissions from several source categories in 1990 and 1999. However, due to inconsistencies in the sources included in the two inventories, they cannot be used to establish a specific reduction in HCB emissions between 1990 and 1999. During 2006, US EPA commissioned work on an HCB Inventory, similar to the EPA's 2000 Dioxin Inventory. The HCB Inventory will employ emission factors and activity estimates for source category emissions rather than facility- or state-reported data. The expected date for completion of the inventory is uncertain.

Figure 4-4 shows B(a)P release estimates and reduction progress within the U.S. Great Lakes Basin from 1996 to 2001.[26] B(a)P emissions from the eight Great Lake states have been reduced by approximately 77% during that time, with annual

[23] Based on "B(a)P/PAH Emissions Inventory for the Province of Ontario 1988, 1998 and 2000 Draft Report (No. 1), May 16, 2000" prepared for Environment Canada by Benazon Environmental Inc., with releases updated by Environment Canada - Ontario Region, based on NPRI facility release data and recent sector release assessments.

[24] Available at http://www.ec.gc.ca/nopp/docs/cp/1mm7/en/toc.cfm

[25] Based on EPA's 1990 National Toxics Inventory (with 1999 open burning estimates added) and 1999 National Emissions Inventory (updated with 1999 pesticide application emissions data).

[26] Based on the Great Lakes Regional Air Toxic Emissions Inventory for 1996 through 2001, with Ontario emissions removed and petroleum refining emissions reduced to approximately 5 lbs beginning in 1997, per revised estimates provided by the American Petroleum Institute (API, 2001).

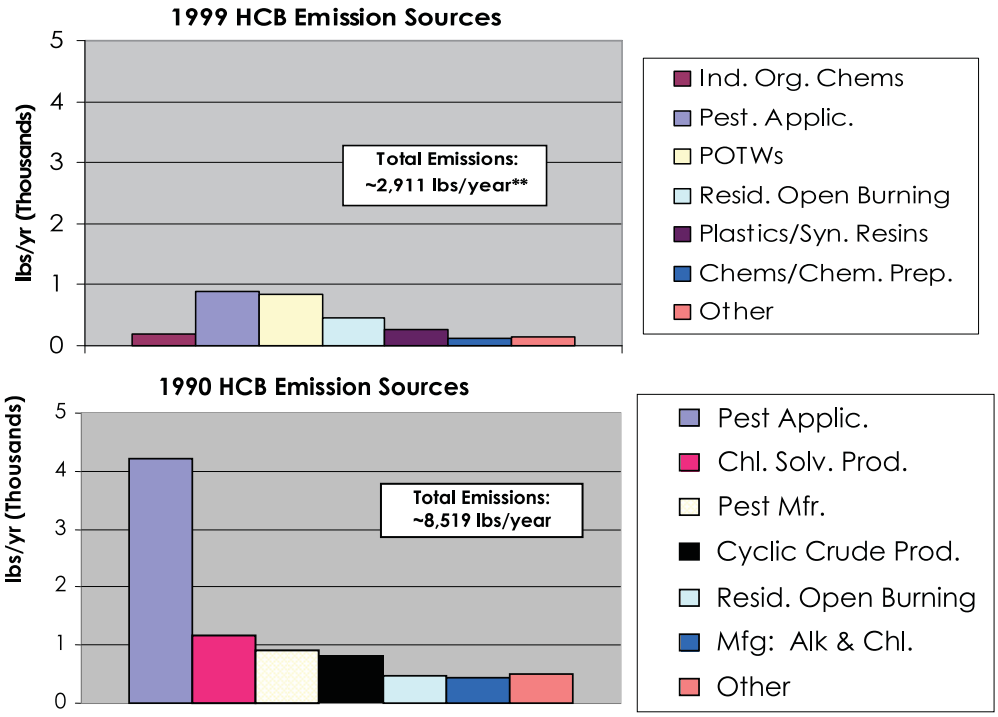


Figure 4-3. Estimated U.S. HCB Releases for 1990 and 1999 (lbs/year) Source: US EPA 1990 National Toxics Inventory, adjusted to reflect residential open burning emissions, and 1999 National Emissions Inventory data updated with 1999 pesticide application emissions data[27,28,29]

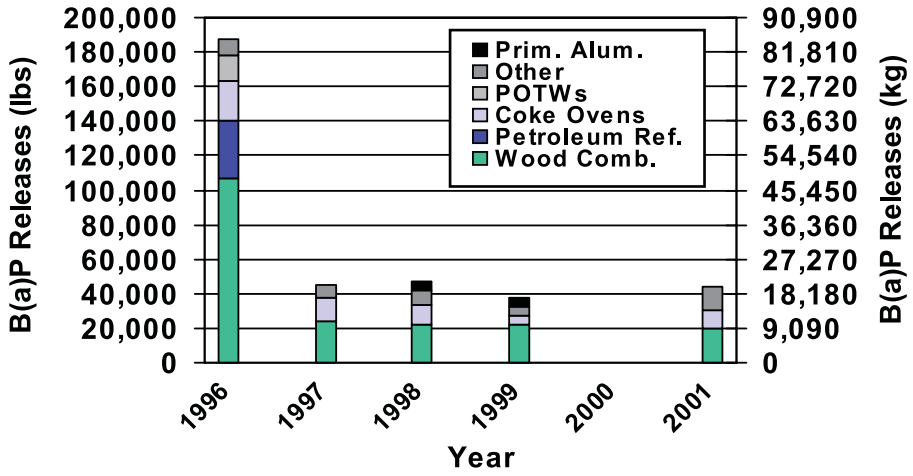


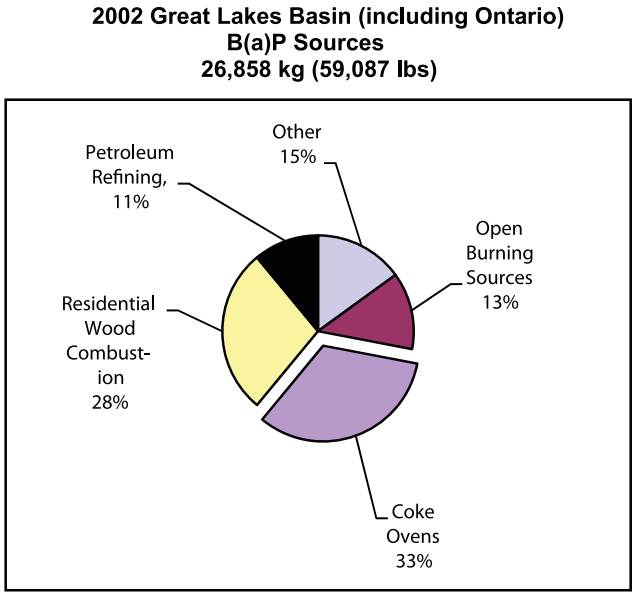
Figure 4-4. B(a)P Releases from the U.S. Great Lakes States, 1996-2001.[30]

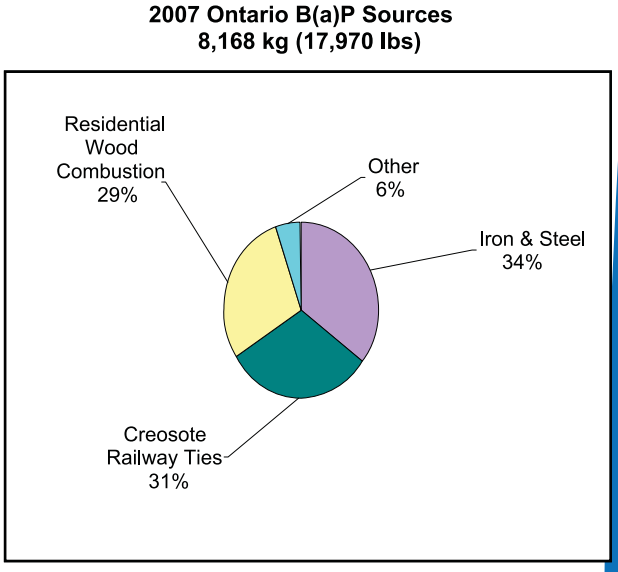
[27] **1999 NEI data excludes ~8,500 lbs of HCB emissions which could not be verified by the reporting facility.

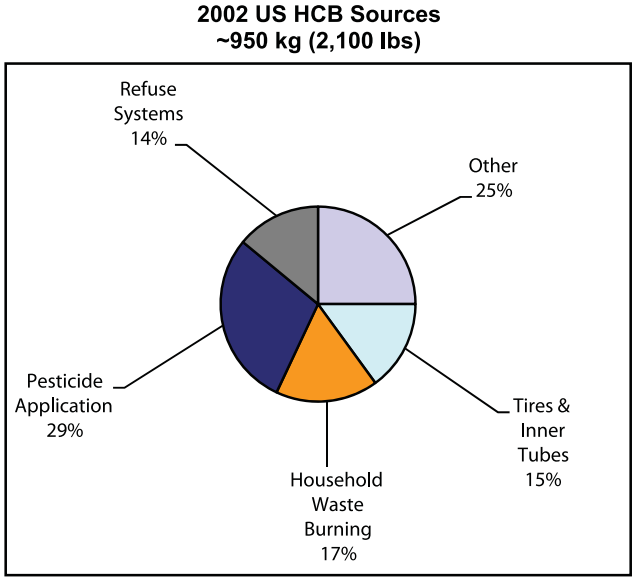
[28] Pesticide application data assumes 100% volatilization of the HCB contaminant in pesticides.

[29] 1999 emissions from POTWs could not be verified.

[30] Based on the Great Lakes Regional Air Toxic Emissions Inventory for 1996 through 2001, with Ontario emissions removed and petroleum refining emissions reduced to approximately 5 lbs beginning in 1997, per revised estimates provided by the American Petroleum Institute (API, 2001).







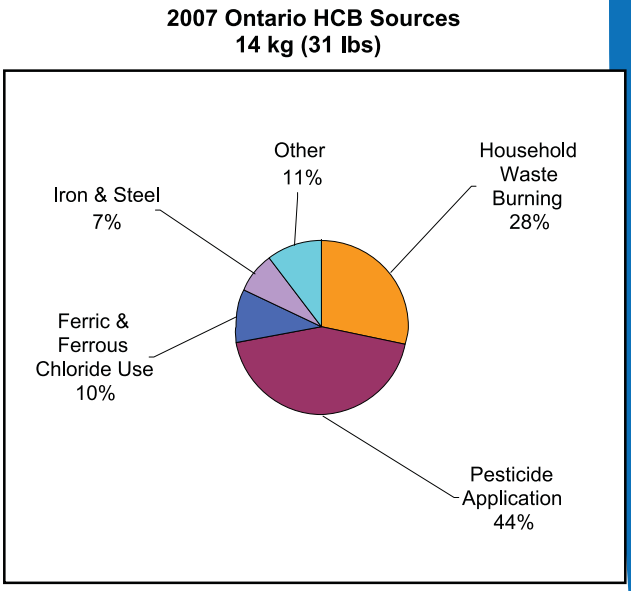


Figure 4-5. **HCB and B(a)P Sources in the Great Lakes Sources: Great Lakes Commission, 2002 Inventory of Toxic Air Emissions for the Great Lakes Region and Environment Canada (Environmental Protection Operations Division – Ontario Region) Inventory as of November 2009.**

emissions in 2001 estimated at 43,700 pounds. Since the 2001 inventory was prepared, B(a)P emissions from the petroleum refinery sector have been essentially eliminated and emissions from primary aluminum manufacture and coke ovens substantially reduced. In 2001, residential wood combustion was the largest B(a)P emission source in the Great Lakes.

Data from a reassessment of the 2002 Great Lakes Regional Air Toxic Emissions Inventory became available in 2007. Total B(a)P emissions from the eight Great Lake States and Ontario were estimated at 59,087 (see Figure 4-5) in this reassessment. Estimated annual B(a)P emissions were higher in the 2002 inventory than in the 2001 inventory primarily due to improvements in the inventory. The 2002 Inventory of Toxic Air Emissions is available at www.glc.org/air/inventory/2002/.

U.S. Reduction Activities

Midwest Clean Diesel Initiative

» The Midwest Clean Diesel Initiative (MCDI) is a collaboration of federal, state, and local agencies, along with communities and private companies, working together to reduce emissions from diesel engines in the Midwest (U.S. EPA Region 5). (See the MCDI website at http://www.epa.gov/midwestcleandiesel.) MCDI reduces diesel emissions by fostering projects that use one or more of the "5 R's" of clean diesel actions: retrofitting, reducing idling, refueling, repowering, and replacing diesel engines in the Midwest. In the past year, the initiative has provided over $44 million in grants for retrofits and other clean diesel technologies. Diesel retrofits have been performed on school buses, construction equipment, marine vessels, and municipal and private fleets. The installation of Advanced Truck Stop Electrification systems provides diesel trucks the opportunity to "plug in" rather than keep their diesel engines idling for auxiliary power, and US EPA's SmartWay Transport Partnership promotes voluntary measures that will reduce fuel use and emissions. As of November 2009, MCDI had impacted over 761,000 diesel engines (in a way that would reduce their emissions). The MCDI goal is to reduce emissions from 1 million diesel-powered engines by 2010.

Burn Wise Campaign

On October 22, 2009, US EPA launched its Burn Wise educational campaign (*http://www.epa.gov/burnwise*) to help reduce wood smoke pollution. The program encourages people to burn the right wood the right way, in the right wood-burning appliance. This campaign follows a recent US EPA announcement of particulate matter (PM) designations. In many areas across the nation, wood smoke is a significant contributor to particle pollution. The message is fairly simple: If people burn wood, they can save money and have a safer and healthier home by following these tips:

1. Burn only dry, seasoned wood. It's better for the air – and your wallet. Look for wood that is darker, has cracks in the end grain, and sounds hollow when hit against another piece of wood. Dry seasoned wood is more efficient at heating your home and therefore can add up to significant savings over the winter. Never burn painted or treated wood or trash.

2. Maintain your wood stove or fireplace and have a certified technician inspect them yearly. A certified technician can clean dangerous soot from your chimney, and keep your wood stove or fireplace working properly – reducing your risk of a home fire.

3. Change to a US EPA-certified wood stove or fireplace insert. These models are more efficient than older models, keeping your air cleaner, your home safer and your fuel bill lower, while keeping you warm in the winter. An estimated 12 million Americans heat their homes with wood stoves each winter, and nearly three-quarters of these stoves are not EPA-certified. An EPA-certified wood stove can emit nearly 70% less smoke than older uncertified models.

Wood Stove/Fireplace Initiatives

- US EPA has combined several websites into one comprehensive website containing information on wood smoke (www.epa.gov/burnwise). The Burn Wise website provides consumers with information on the health effects of wood smoke, the benefits of using US EPA-certified stoves, and guidance on burning wood efficiently and safely. This website also provides a guide for implementing a wood stove change-out campaign. A wood stove change-out campaign provides information and incentives (e.g., rebates or discounts) to encourage people to replace their old, conventional wood stove with a US EPA-certified wood-burning appliance that burns more cleanly and efficiently or with alternative heating devices, including pellet, gas, and propane appliances.

- A wood stove change-out fact sheet has been developed that explains the problems with using older, higher polluting wood stoves and discusses the quantity and type of emissions from residential wood combustion, the adverse health effects from wood smoke, and a way to address the problem by facilitating the replacement of old and inefficient wood stoves with newer, more efficient and cleaner burning technologies through education, outreach, and incentives (e.g., cash rebates).

- The *Great Michigan Wood Stove Change-out Campaign* allowed Michigan residents to take advantage of a more efficient, clean, and safe way to heat their homes at a discounted cost through an initiative by the Michigan United Conservation Clubs (MUCC), who partnered with the Michigan Department of Environmental Quality (MDEQ), and the Hearth, Patio and Barbeque Association (HPBA). This initiative provided up to $500 in rebate and discount incentives toward the replacement of a conventional wood-burning stove with a more efficient, cleaner wood, pellet, corn, gas, or electric stove or fireplace insert certified by the EPA. Made possible by a MDEQ/US EPA grant, "The Great Michigan Woodstove Change-out Campaign" aimed to educate Michigan residents about the economic, health, safety, and environmental benefits of switching to modern home heating stoves, improving air quality, and lowering wood smoke emissions. MUCC performed extensive outreach on this campaign through its magazine, television shows, trade shows and public service announcements. A $290 mail-in rebate from MUCC was issued to qualified consumers who performed the change-out through a participating retailer. Retailers also offered significant discounts and/or rebates as a part of the campaign. In order to receive the $290 rebate from MUCC, Michigan residents must have agreed to have their old stove rendered inoperable. This extremely successful program ran from May through July 2008 and resulted in the replacement of 500 old, highly polluting wood stoves.

- In addition to promoting Burn Wise tips, US EPA has developed a guidance document for state, local, and tribal agencies. "Strategies for Reducing Residential Wood Smoke" provides a comprehensive list of strategies to help communities reduce wood smoke from residential heating. The document includes education and outreach tools, information on regulatory approaches to reduce wood smoke, as well as voluntary programs to change out old, inefficient wood stoves and fireplaces. To download a copy of "Strategies for Reducing Residential Wood Smoke" visit, *http://www.epa.gov/ttn/oarpg/t1/memoranda/strategies-doc-8-11-09.pdf*

Outdoor Wood-Fired Boilers

- Outdoor wood boilers have combustion chambers in small sheds outside of the home. Burning occurs in the shed with no emission control devices, and emissions are vented through a small stack (generally less than 12 feet). The cyclic nature of the boiler operation does not allow for complete combustion, which results in much higher emissions than from wood stoves. The use of outdoor wood boilers is increasing, with about 500,000 expected to be in place nationwide by 2010, primarily in the Northeast and Midwest, including the Great Lakes area. Although US EPA has not yet adopted regulations to address outdoor wood boilers, it has taken

the following steps: (1) development of a test method specific to outdoor wood boilers is complete; and (2) a voluntary incentive program has resulted in an agreement with the major outdoor wood boiler manufacturers (see *http://www.epa.gov/burnwise/testmethods.html*). As a result of this agreement, wood boilers that emit 70% less emissions have been available since 2007, and wood boilers emitting over 90% less emissions are now available. In addition, a model rule has been developed for states and local agencies that includes limits requiring a reduction of approximately 80% in particulate matter emissions, zoning and stack height restrictions, information on proper operation and maintenance, and labels for new boilers that verify that the model in question meets applicable emission level. The status of all aspects of this program is available at *www.epa.gov/burnwise*.

Scrap Tires

- The Rubber Manufacturers Association (RMA) reported that scrap tire reuse now approaches 90% nationwide in the U.S. In 2007, 89.3% of the scrap tires generated in the U.S. by weight were consumed in end-use markets, which include tire-derived fuel, civil engineering and ground rubber applications. The total volume of scrap tires consumed in end-use markets in the U.S. reached approximately 4.1 million tons of tires. "Old piles of scrap tires are shrinking," said RMA Vice President Michael Blumenthal. Managing scrap tires to prevent tire fires that release B(a)P and other pollutants is a priority of the HCB/B(a)P Workgroup.
- Under a Scrap Tire Pile Mitigation Support Project, US EPA finished developing a scrap tire pile inventory for the Great Lakes States, along with Geographic Information System (GIS) mapping of large tire piles (>500 tires). For more information about this project, see http://www.epa.gov/reg5rcra/wptdiv/solidwaste/tires/index.htm.
- Between 2005 and 2007, there has been a reduction of about 24 million stockpiled tires in the Great Lakes States of New York and Pennsylvania, which now report less than 2 million tires. Michigan will continue to abate stockpiles and should have nearly all pre-1991 piles abated within the next year. Ohio has cleaned up all known major abatement sites and Minnesota, Wisconsin, Illinois, and Indiana all report less than 1 million tires.
- In January 2006, US EPA completed a best practices *Scrap Tire Cleanup Guidebook* on how to manage scrap tire piles.
- Scrap tire market development, and the protection of existing markets, must be a top priority of states and industry.

Coke Ovens

- Amendments to the 1993 MACT standards for coke ovens, which contain more stringent emission limits for coke oven doors, charge port lids, and off-take piping on 17% of U.S. coke batteries, were promulgated in April 2005. This action, which addressed "residual risk," was the first of its kind by US EPA. In April 2006, new MACT rules went into effect for coke plant emission points, not included in the 1993 rules, for pushing, combustion stacks, and quench towers. These MACT rules apply to all U.S. coke plants.
- According to the American Coke and Coal Chemicals Institute, coke production did not change from 2006 to 2007 in either Canada or the United States. In Canada, 100% of coke is produced in the Great Lakes Basin. There are some increases in heat recovery capacity in the U.S., but nothing in Canada. Some of the planned upgrades for the upcoming year have been postponed or cancelled due to the current state of the economy. No upgrades are planned for the Great Lakes region. Worldwide, China is the largest producer of steel and has the largest demand for coke. However, it too is affected by the poor economy and has seen a reduction in demand.

Industry Reduces HCB Releases Reported to Toxics Release Inventory (TRI)

- The number of facilities reporting HCB releases to TRI remained around 40 from 2004 to 2007, a decline from ~50 facilities that reported annually from 2000 to 2003.
- Reductions in HCB emissions reported to TRI have leveled off in recent years (2006-2007, the latest data available).
- Several companies reported declines in HCB releases from 2006 to 2007, primarily due to lower production levels.
  - Dow Chemical Co. (Plaquemine, LA) reported a reduction in stack HCB emissions by 98%, from 53 lbs in 2006 to 1 lb in 2007.
  - Dow's Freeport facility, in Freeport, TX, reported a reduction in stack HCB emissions from 28 lbs in 2006 to 10 lbs in 2007.
  - Occidental Chemical Corp. Geismar Plant (Geismar, LA) reported reductions in fugitive HCB air emissions from 17 lbs in 2006 to 1 lb in 2007.
  - Clean Harbors Deer Park LP (La Porte, TX) reported a decline in HCB water releases from 6 lbs in 2006 to 0.1 lb in 2007. (The facility had reported ~ 6 lbs since 2004.)

Coal Tar Sealants

There are two main kinds of driveway and parking lot sealants: refined coal tar-based and asphalt-based. The variation in the PAH content of each can be significant. One study reported that refined Coal Tar-Based Sealants (CTS) contain 3.4% to 20% PAH dry weight basis, compared to 0.03% to 0.66% in asphalt-based sealants, up to 670 times less than CTS. The net difference in a lifecycle analysis, however, may not be as significant; the experience of users suggests that CTS lasts significantly longer and is replaced less often than asphalt-based alternatives. The reason for concern about the use of CTS is the potential additional PAH contamination of nearby watersheds from runoff close to driveways and parking areas treated with CTS. Gravel and concrete are other available alternatives to CTS which could be considered. Like CTS and asphalt-based sealants, each alternative has advantages and disadvantages. In response to concerns raised about CTS, some retail stores have stopped selling products with CTS, and some local municipalities have instituted laws prohibiting their use.

A study by the Stormwater Center of the University of New Hampshire was conducted on a parking lot test facility at the university and provided some evidence of increased PAH levels on newly applied CTS after the first rain, compared to a similar application of asphalt material. However, adverse weather conditions inhibited curing of the newly applied CTS, which may account for some or all of the observed relative increase in PAH levels. The findings underscore the importance of following recommended application procedures to ensure that CTS have sufficient time to cure before the first rain event. An expansion of the University of New Hampshire project was funded by US EPA to determine total PAH loads transported offsite from coal tar and asphalt sealed pavements by means of wind and tire tracking.

A study supported by the Pavement Coatings Technology Counci[31] was performed in Austin, Texas, which was the first city to ban the use of CTS within its jurisdiction. The study looked at pre- and post-ban levels of contaminants. The study showed no significant differences in the levels or sources of PAHs in runoff after the ban compared to before the ban. The industry continues to work with the HCB/B(a)P Workgroup to provide additional information regarding the contribution of PAHs from CTS.

Canadian Reduction Activities

Residential Wood Combustion

- EC has restructured the Residential Wood Combustion focus to develop regulations and

[31] DeMott, R.P., Gauthier, T.D., Wiersema, J.M. and Crenson, G. 2009. PAHs in Austin Sediments after a Ban on Pavement Sealers. Environmental Forensics, In press.

has reduced the outreach aspects of the work since 2008.

» A DVD, developed by EC, containing three videos (*Advanced Technology Woodstoves - EPA, Firewood Preparation*, and *Woodstove Operation*) has become very popular among retailers and other interest groups. This DVD continues to be distributed to participants of woodstove change-out programs in the United States and Canada.

In early 2009, EC completed an EPA-certified wood stove testing study on two appliances. The purpose was to verify the emission factors from these types of stoves under real-world conditions. The results of the study indicate that the real-world emission factors were either comparable to or less than the average literature value. This is explained by the fact that the two stoves were modern stoves.

Ontario Tire Stewardship (OTS) Program (from www.ontariots.ca)

» On September 1, 2009, the Ontario Tire Stewardship program was launched. The program will eliminate the "disposal fee" that consumers have paid to dispose of their old tires – whether or not they are buying new ones – making it easy and free for Ontarians to get their old tires recycled by dropping them off at registered collectors across Ontario.

» OTS will provide financial incentives for registered organizations that collect, transport, and process used tires or manufacture recycled products in accordance with the program plan. These incentives will promote sustainable development and new markets for recycled materials and innovative uses for recycled rubber products. In the first year of the program, this will represent a $23 million investment in the Ontario tire recycling industry in the first year alone, stimulating economic growth and helping to increase capacity.

» Within five years, the Program is expected to divert 90% of scrap on-road tires and collect and recycle 50% of all scrap off-road tires.

» OTS is also working with the MOE to develop a tire stockpile abatement schedule and is looking forward to starting clean-up projects in municipalities in the spring as part of its 3-year plan to eliminate the millions of stockpiled tires in sites across Ontario.[32]

PAH Source Apportionment Modeling

» Research has been completed on identifying and quantifying sources contributing to ambient PAH levels in both urban and rural sites in Ontario using receptor modeling techniques. Results are currently being reviewed.

Next Steps

The workgroup will continue ongoing efforts to improve the accuracy of the U.S. and Canadian HCB and B(a)P emission inventories to ensure that all significant emission sources have been identified and included. The workgroup will also continue to pursue emission reduction activities from significant B(a)P source sectors, namely:

» **Residential Wood Combustion** – Research activity will be pursued to learn more about the extent of wood burning and emissions from certified EPA woodstoves. In addition, voluntary wood stove and outdoor wood boiler reduction activities, e.g., wood stove change-out programs and "Burn it Smart" and "Burn Wise" outreach programs, remain a top priority.

» **Scrap Tires** – *U.S. EPA Best Practices Guidebook* and additional training materials are available. Also, scrap tire pile mapping and inventory initiatives should continue; tracking progress made by the OTS program should also continue.

» **Coal Tar Sealants** – EC is performing an additional study to better establish the environmental impact of coal tar driveway sealers. Also, field measurements are anticipated

[32] OTS News. Used Tires Program Benefitting Bottom Line. October 2009. Available at http://www.ontariots.ca/. Accessed January 2010..

as a follow-up to an inventory that was developed to identify the extent of CTS use in Ontario municipalities. This work could be developed alongside a similar investigation into CTS on the U.S. side.

The workgroup will also support other actions and ideas that impact HCB releases to the Great Lakes Basin. Specifically, the workgroup will

- Continue to implement the Household Waste Burning Strategy (Burn Barrel Subgroup of Dioxin/Furan Workgroup).
- Examine potential opportunities for reductions for major sources (pesticide application, ferric and ferrous chloride use).
- Continue solicitation of voluntary HCB reductions by chemical companies.

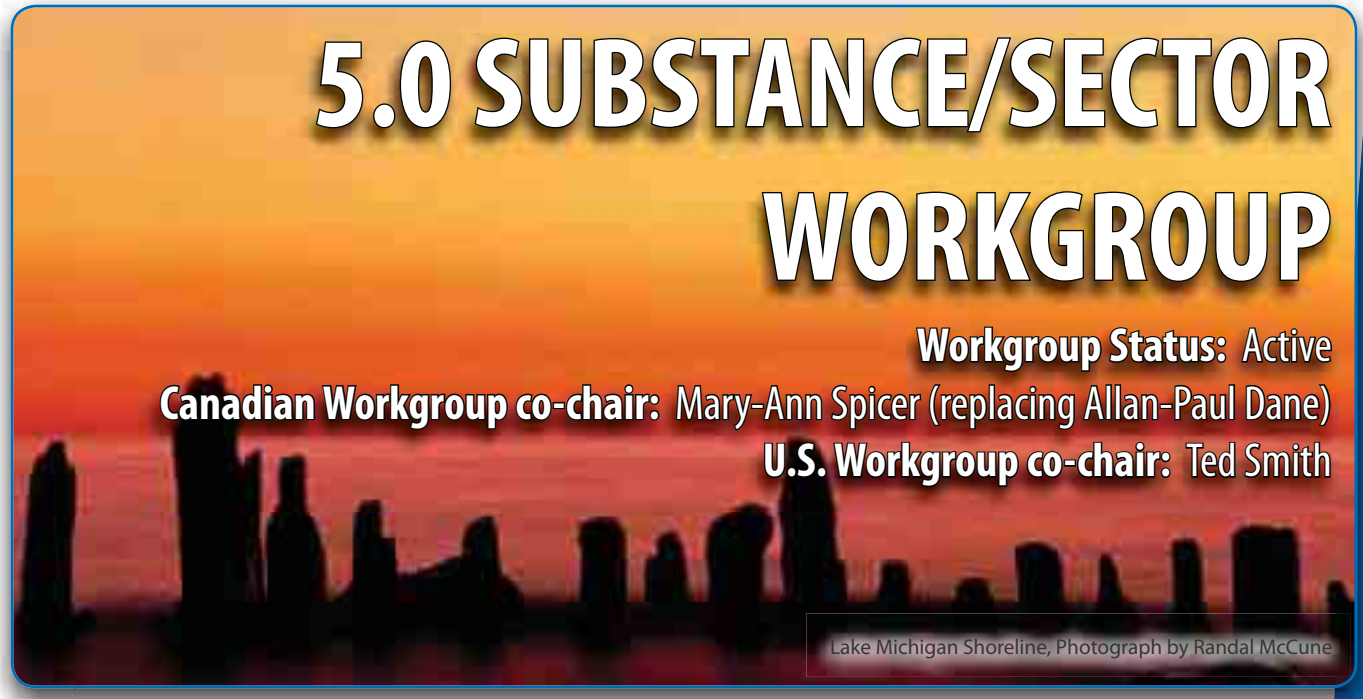
The workgroup will consider expanding its scope to track other GLBTS substances closely associated with HCB and B(a)P, namely, chlorobenzenes and other PAHs.

5.0 SUBSTANCE/SECTOR WORKGROUP

Workgroup Status: Active

Canadian Workgroup co-chair: Mary-Ann Spicer (replacing Allan-Paul Dane)

U.S. Workgroup co-chair: Ted Smith



Lake Michigan Shoreline, Photograph by Randal McCune

Under the Strategy, EC and US EPA agreed to consider new substances that may pose threats to the Great Lakes ecosystem, for potential reduction activities. The Strategy challenges the Parties (EC and US EPA) to consider:

"… whether new substances which present threats to the Great Lakes ecosystem should be considered for inclusion on the Level I or II lists."

The following efforts were undertaken in support of the above challenge.

Substance/Sector Workgroup Activities

During 2008 and 2009, the Substance/Sector Workgroup met, either in person or by teleconference, as follows:

- April 8, 2008 meeting in Chicago
- June 2-3, 2008 meeting in Burlington
- August 7, 2008 teleconference
- September 24, 2008 meeting in Chicago
- December 2-3, 2008 meeting in Chicago
- March 31, 2009 meeting in Toronto
- December 2, 2009 meeting in Chicago

In addition, the Substance/Sector Workgroup reported progress and discussed future directions at GLBTS Integration Workgroup meetings.

At these meetings, the Substance/Sector Workgroup explored a new path forward under the GLBTS by considering potential chemical threats to the Great Lakes Basin. A draft *General Framework for Identifying Substances to be Considered in the Great Lakes Basin* was developed. The framework illustrates a process by which substances may be identified for consideration under the GLBTS. The workgroup prepared examples of using the framework to consider potential threats to the basin. Based on the amount of data available, the following three candidate substances were chosen to illustrate examples of implementing the general framework:

- Nonylphenol and its Ethoxylates (NPEs)
- Polybrominated Diphenyl Ethers (PBDEs)
- Perfluorooctane Sulfonate (PFOS)

To determine substances that may be national priorities for both Canada and the U.S. in the Great Lakes, the Substance/Sector Workgroup

conducted an analysis of substances that are common across Canada's Domestic Substances List (DSL), US EPA's Inventory Update Reporting (IUR), and the International Joint Commission's (IJC's) List of Substances of Emerging Concern. The analysis identified approximately 30 common substances (or groups of substances). The analysis demonstrated one approach to a GLBTS substance selection process. The workgroup illustrated a similar approach to identifying sectors for GLBTS discussion. The analysis identified four common sectors based on the DSL/IUR/IJC substance analysis described above. Further discussions with EC, US EPA, and stakeholders are needed to refine the substance and sector selection processes.

The Substance/Sector Workgroup gathered information on emerging contaminant monitoring and surveillance efforts in the Great Lakes. The workgroup learned of monitoring and surveillance activities being conducted under Canada's Chemical Management Plan (CMP), EC's Great Lakes Fish Contaminant Surveillance Program, EC's Herring Gull Egg Monitoring Program, EC's Great Lakes Sediment Assessment Program, Integrated Atmospheric Deposition Network (IADN), US EPA's Great Lakes Fish Monitoring Program, Muir/Howard North American Chemical Inventory Screening Project, NOAA's Mussel Watch Program, U.S. Geological Survey (USGS) tributary monitoring in the Great Lakes, USGS monitoring of contaminant effects on Great Lakes indicator species, and other projects. Information gathered from these monitoring programs will help inform the workgroup's considerations of potential threats to the basin.

In an effort to maintain consistency with the efforts of various groups that may influence the future direction of the GLBTS, the Substance/Sector Workgroup kept up to date on a number of current issues, including: renegotiation of the Great Lakes Water Quality Agreement, IJC Chemicals of Emerging Concern Workgroup, US EPA's Chemical Assessment and Management Program (ChAMP)[33], and MOE's activities to address Level 1 substances and chemicals of emerging concern. A few of these efforts are described in further detail below.

Related New Substance Work

Various efforts related to identifying and prioritizing new chemicals serve to inform the Substance/Sector Workgroup of the GLBTS. A few of these efforts are summarized below. In addition, environmental monitoring results for a limited number of emerging substances of concern are presented in Chapter 9 of this report.

Canada's Chemical Management Plan

The Canadian Great Lakes Substance Priorities Working Group has been charged with providing direction and recommendations regarding Canada's priorities for substances in the Great Lakes Basin for federal, joint-jurisdictional, and binational programs. This will ensure that actions taken are complementary to Canada's CMP, through a coordinated Great Lakes approach to chemicals management.

While the working group is charged with providing recommendations concerning Canadian Great Lakes Basin substance priorities, it will not conduct assessments, nor determine specific management actions. Substances identified by the working group will be recommended for management, assessment, review, and/or monitoring as necessary, within best-placed programs. Specific actions and further subsets of priorities can then be established within these programs.

In order to achieve its objectives, the working group is currently developing a chemical selection and prioritization process. This process contains four key elements, which are presented below.

[33] ChAMP has been superseded by a comprehensive approach to enhancing EPA's current chemicals management program. Under the new approach, EPA released action plans in December 2009 that describe steps EPA will take to manage concerns for phthalates, long-chain perfluorinated chemicals (PFCs), PBDEs in products, and short-chain chlorinated paraffins.

1) Triggers for Considering Substances for Action through a Great Lakes Approach

There are two triggers which identify substances to be considered by the process, based on indication of potential risk to the environment and/or human health in the Great Lakes Basin:

a) Prioritization within national chemical programs; and

b) Early warnings from monitoring and research initiatives.

National program priority is considered a primary trigger, in order to be responsive to the national CMP while implementing a coordinated Great Lakes approach. Categories of substances in various stages of assessment and/or management that may be considered national priorities include:

- Non-challenge substances (previously assessed and/or managed substances)
- High priorities (challenge substances, as defined under Canada's CMP)
- CMP II substances

Ideally, as each national priority substance undergoes assessment and risk management, the relevance of action through a Great Lakes approach is routinely evaluated. In the immediate future, it will be necessary to perform this evaluation for a backlog of current national program priorities.

A Great Lakes approach can also provide utility by recognizing the Great Lakes Basin as a sentinel ecosystem for early warning and feedback to the national programs. A substance may not currently be a national priority because it has not been recognized as a concern by national programs, it is already under management in Canada, or it is not used or released within Canada. However, the substance may be in commerce in the U.S., deposited in the basin from international sources through long-range atmospheric transport, or be the subject of new research that indicates potential concerns not previously considered (e.g., endocrine disrupting properties). Therefore, the substance is not a national priority but is emerging or re-emerging as a concern and should be considered under the chemical selection and prioritization process.

2) Relevance to the Great Lakes

The primary reason that national priorities or substances of emerging concern would be addressed under a coordinated Great Lakes approach is that they are present in the Great Lakes Basin ecosystem. One means of accomplishing an evaluation of presence is through the use of overlay analyses of national program priorities with substances currently detected in the Great Lakes Basin through monitoring and surveillance initiatives. Steady or rising trends, multiple detections, and/or the presence of sectors as a potential source may help in establishing presence. In order to prevent the bias of finding only what is looked for, close links with research and monitoring are necessary.

3) Present Management Considerations

If a substance is present in the Great Lakes Basin ecosystem and carries an environmental and/or human health concern, it is a strong candidate for action under a coordinated Great Lakes approach. The present management status of the substance in national programs is evaluated to determine whether actions are necessary to complement any existing efforts. Management of a substance within the U.S. and through engagement in international fora is also considered. Consultation with risk managers and/or substance coordinators is suggested as an effective way to evaluate the present management status and to perform triage, in order to help determine whether a substance should be recommended for action (monitoring, assessment, management, and/or review) within best-placed programs.

4) Stakeholder Input

Stakeholder consultation constitutes an important component of the substance selection process, as it provides valuable insight from an "on the ground" capacity and also facilitates engagement at the subsequent risk management stage. Unless circumstances demand otherwise,

stakeholder consultation will occur once chemicals have been recommended for action, under best-placed programs. Consultation at this stage will allow for the refinement of priorities and for the development of specific actions within these programs. Additionally, most best-placed programs will already have an active and engaged stakeholder base, which will facilitate the external consultation process.

Delisting

By using the national programs as triggers for consideration, formal delisting may not be necessary. Substances should move forward as appropriate within the monitoring, assessment, management, and review processes of the best-placed programs. Substances not recommended for action may simply be categorized by the working group as "no recommendation at this time," with the second trigger of "emerging and re-emerging concerns" available to elevate the status of a substance, should it become necessary.

US EPA Chemical Management Activities

EPA's Enhanced Chemical Management Program

In late 2009, US EPA Administrator Lisa P. Jackson announced that US EPA would develop a comprehensive approach to enhance the current TSCA chemicals management program. The most important components involve:

- Identifying chemicals that pose a concern to the public;
- Moving quickly to evaluate them and determine what actions need to be taken to address the risks they may pose; and
- Initiating appropriate action.

US EPA's chemical actions may include initiating regulatory action to label, restrict, or ban a chemical, or to require the submission of additional data needed to determine a chemical's risk. If US EPA determines that a chemical does not present a need for action, US EPA will make respective information available.

US EPA has posted four action plans to date.[34] These plans: (1) summarize available hazard, exposure, and use information; (2) outline the risks that each chemical may present; and (3) identify the specific steps US EPA is taking to address those concerns. As these actions continue, US EPA will make opportunities available for public and stakeholder comment and involvement. Chemicals were chosen on the basis of multiple factors, including:

- Chemicals identified as persistent, bioaccumulative, and toxic;
- High production volume chemicals;
- Chemicals in consumer products;
- Chemicals of particular potential concern for children's health because of reproductive or developmental toxicity;
- Chemicals subject to review and potential action in international fora;
- Chemicals found in human blood in biomonitoring programs; and
- Chemicals in categories generally identified as being of potential concern in the new chemicals program.

Region 5 Leadership in US EPA's Enhanced Chemical Management Program

Region 5 is currently the US EPA Office of Prevention, Pesticides and Toxic Substances (OPPTS) National Coordinator for "Reviewing and Reducing TSCA Chemical Risk." As part of that role, Region 5 works closely with OPPTS, other regional offices, state and local governments, tribes, stakeholders, and the public to assist with chemical, chemical risk, or other related projects in which there is potential concern to human health and the environment. Region 5 projects include:

[34] Plans posted to date include: phthalates, perfluorinated compounds (PFCs), PBDEs in products, and short-chain chlorinated paraffins.

- Communicating national and regional chemical program goals to US EPA offices, labs, and regions.
- Strengthening coordination between OPPTS, other US EPA offices, labs, and regions in multi-media chemical and chemical risk projects.
- Coordinating with OPPTS and other US EPA offices, labs, and regions and states regarding: chemical or chemical risk assessment, communication, identification, and management methods and strategies; program activities and respective tool, strategy, and network development; enhanced resource leveraging, and better communication strategies.
- Facilitating enhanced stakeholder dialogs on chemical and chemical risk issues.

New Chemical Screening Work by Howard/ Muir

US EPA funded a project conducted by Philip Howard of Syracuse Research Corporation and Derek Muir of EC to identify emerging contaminants and persistent, bioaccumulative, and toxic (PBT) chemicals that were not being sought out or analyzed in current Great Lakes contaminant monitoring and surveillance programs and to determine how they could be chemically analyzed. The Canadian DSL list totaling 11,317 compounds was combined with the US EPA high production volume (HPV) list, a list of 3,059 substances of "Unknown or Variable composition, Complex reaction products and Biological materials" (UVCBs), the US EPA TSCA IUR database for years 1986, 1990, 1994, 1998, 2002, and 2006, and 500 chemicals from US EPA's HPV program, which covered substances that were not in the HPV program but were produced in amounts over 1 million pounds during 2002. The CAS numbers were cross-compared to remove duplicates, yielding a total of 22,263. From that list, 610 chemicals were identified by Structure Activity Relationships (SARs US EPA EPI Suite) and using expert judgment. Toxicity was also assessed using SARs for aquatic toxicity and cancer potential, but was not used to prioritize the chemicals. This study has yielded some interesting potential persistent and bioaccumulative (P&B) substances that could be considered for further study and monitoring and surveillance in the Great Lakes region.

Spectacle Reef Lighthouse, Straits of Mackinac
Photograph courtesy of US Coast Guard

The major chemical groups in this analysis include brominated, chlorinated, fluorinated, silicone, and non-halogenated substances. Top 10 priorities were selected from each of the five chemical groups, in order to identify a first round of priority substances for further investigation. The major criteria used to select the top 10 were production volume, bioconcentration factor (BCF), and persistence (atmospheric oxidation half life: AO t½). Representatives of important classes of compounds such as tetrabromo bisphenol A (TBBPA) derivatives, cyclic siloxanes, chlorinated pyridines, and cyclopentane/enes were also identified. Chemicals for which there were already measurements, for example, PBDEs, synthetic musks, triaryl phosphates, and haloalkyl phosphates, were omitted.

Most of the 50 top priorities identified are not currently analyzed, yet most are in commerce

based on the 2002 and 2006 TSCA IUR information. All of the top 50 and most of the larger list of 610 could likely be analyzed in environmental media, although suitable analytical standards would need to be available and method testing/refinements would need to be conducted. The next phase of this work includes the development of analytical methods for the top priority substances.

The full report can be found at *http://epa.gov/greatlakes/p2.html* under Identification of New, Possible PB&T Substances Important in the Great Lakes Region by Screening of Chemicals in Commerce.

International Joint Commission Review of Chemicals of Emerging Concern and Analysis of Environmental Exposures in the Great Lakes Basin

The U.S. co-chair of the Substance/Sector Workgroup participated in a review of chemicals of emerging concern and analysis of environmental exposures in the Great Lakes Basin. The review and analysis were conducted by an advisory workgroup to the IJC, and the results were compiled into a report that was published by the IJC.[35] The objectives of this report were to review and compile all peer reviewed scientific studies and reports since 1997 in relation to chemicals of emerging concern that may pose threats to water quality in the Great Lakes watershed. Emphasis was placed on chemicals discharged to the Great Lakes nearshore waters from wastewater treatment plants as well as from other point and non-point sources of rural and urban pollution. The concentrations of chemicals in various environmental media were assembled into a database, which was statistically analyzed to develop a quantitative understanding of current environmental exposures. To develop an initial assessment of their potential ecological significance, the concentrations were compared with currently available regulatory standards, guidelines, or criteria. The abstract of the report is presented below. A full summary of the study is provided in Appendix B.

Over the past 10 years, 80 investigations have reported the concentrations of a variety of chemicals of emerging concern in the Great Lakes Basin and watershed. This study was conducted to develop a statistical understanding of environmental exposures in the basin to a variety of environmental contaminants, including current use pesticides, pharmaceuticals, organic wastewater contaminants, alkylphenol ethoxylates, perfluorinated surfactants, flame retardants, and chlorinated paraffins. The available literature was critically reviewed and used to develop a database containing 19611 values for 326 substances. Many of the papers characterized the sampling locations as being downstream from municipal wastewater discharges, receiving waters for industrial facilities, areas susceptible to agricultural or urban contamination, or harbors and ports. Concentrations in surface waters (n = 14841) and biota (n = 3742) represented the majority of the available data, with fewer values reported for sediments (n = 1028). The analysis showed that many chemicals of emerging concern are present in the Great Lakes watershed. Concentrations were generally the highest in the vicinity of sources such as wastewater treatment discharges, agricultural operations, or manufacturing sites; declined with increasing distance from the source; and were generally low or non-detectable in the open waters of the Great Lakes. To develop an initial assessment of their potential ecological significance, the concentrations were compared with currently available regulatory standards, guidelines, or criteria.

Ontario Ministry of the Environment Activities to Address Level 1 Substances and Chemicals of Emerging Concern

The Ontario government continues to address harmful pollutants in the Great Lakes Basin through a

[35] Great Lakes Chemicals of Emerging Concern Advisory Work Group to the International Joint Commission (IJC), 2009. Great Lakes Water Quality Agreement Priorities 2007-2009 Series. Work Group Report on Great Lakes Chemicals of Emerging Concern, 2009 IJC, Special Publication 2009-01, Windsor, Ontario, Canada. Available at http://www.ijc.org/en/priorities/2009/chemicals.

[36] Canada-Ontario Agreement Respecting the Great Lakes Basin Ecosystem. 2007. Available at http://www.ec.gc.ca/CEPARegistry/documents/agree/Fin-COA07/toc.cfm. Accessed: January 2010.

number of regulatory and non-regulatory programs and in cooperation with federal partners under the *Canada-Ontario Agreement Respecting the Great Lakes Basin Ecosystem*.[36]

In 2008, the Ontario government announced its commitment to remove and destroy PCB-contaminated soils stored at the Pottersburg PCB Storage Site in London, Ontario. This PCB site was established in the 1980s to securely contain PCB-contaminated soil, sediment, and debris from the remediation of Pottersburg Creek and some adjacent industrial properties. The ministry acquired the facility and operated it as a storage site until it became possible to destroy the PCBs in an economical manner at an approved PCB destruction facility. Also in 2008, the MOE continued its support to the Summerhill Impact (formerly Clean Air Foundation) "Switch the Stat" program to divert nearly 7,000 old thermostat switches containing mercury from the waste stream. On September 22, 2009, the Minister approved Waste Diversion Ontario's revised program plan for MHSW. The MHSW program diverts common household hazardous or special wastes, such as paints and solvents, from being disposed in landfills or sewers. The program places the management and funding responsibility on producers of these products, and as of July 1, 2010, the revised program will divert additional types of wastes, including mercury-containing products such as thermostats, thermometers, fluorescent bulbs, and switches.

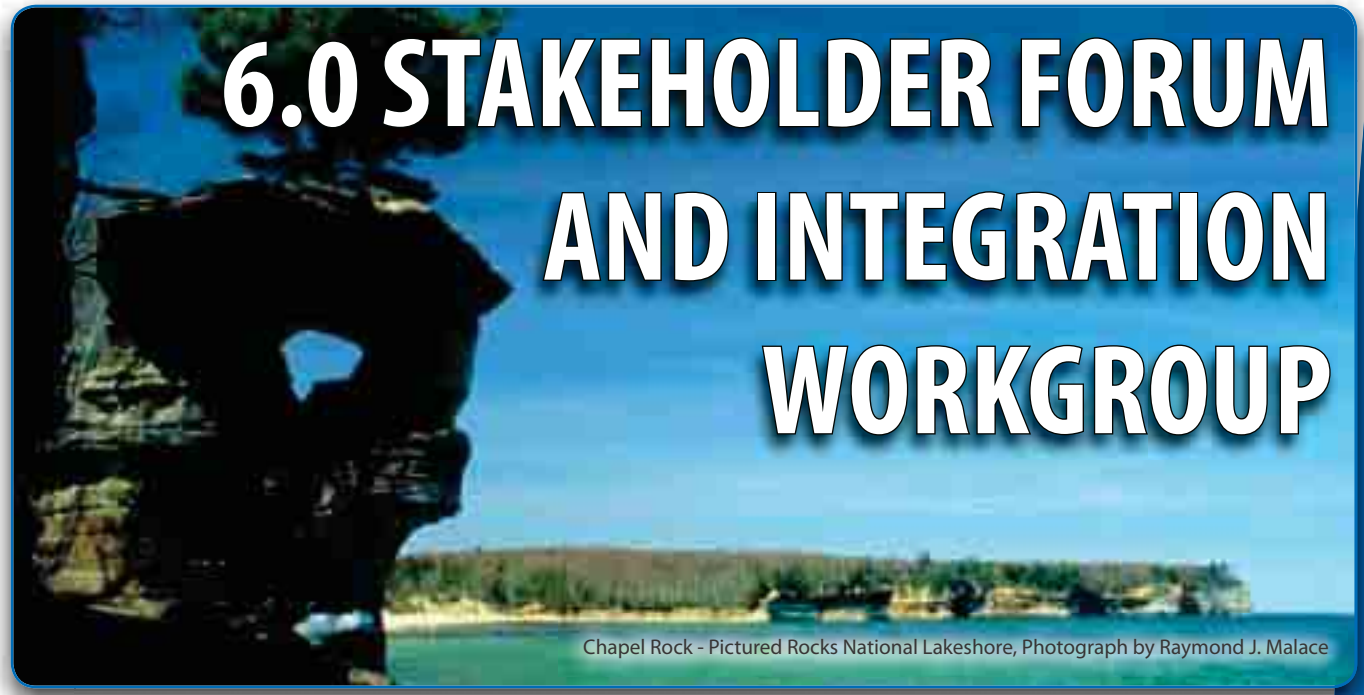
In June 2009, the Ontario government passed the *Ontario Toxics Reduction Act*, 2009. The Act requires owners and operators of regulated facilities to develop plans to reduce their use and creation of toxic substances, to track and quantify the toxics that they use, create, and release, and to report to the ministry and the public. Several GLBTS Level 1 and Level 2 substances have been identified for the first phase of the Act's implementation, including mercury, dioxins and furans, HCB, cadmium, 4,4-methylenebis (2-chloroaniline), and PAHs including B(a)P. Subsequent phases of the Act's implementation would include over 300 substances on Canada's National Pollutant Release Inventory, as well as information-gathering on selected substances of concern, for which use and emissions are not yet tracked in Ontario.

The MOE's science and monitoring programs continue to track harmful pollutants, including chemicals of emerging concern, in the Great Lakes. Recently, the MOE conducted a screening survey of chlorinated flame retardants in Great Lakes sediment and fish. Ongoing collaborative projects include: (1) examining sediments in nearshore areas of the Canadian Great Lakes for presence and trends of perfluorinated compounds (PFCs), halogenated flame retardants, and dioxin-like chemicals; (2) assessing atmospheric contributions of persistent chemicals of emerging concern to the Great Lakes by examining remote sediment cores in proximity to the lakes; (3) developing new analytical methods for the analysis of halogenated flame retardants and chlorinated flame retardants; (4) carrying out passive sampling for pharmaceuticals and personal care products in nearshore areas of Lake Erie and Lake Ontario; and (5) assessing nearshore inputs of current and past-use chemicals from an urban area.

Next Steps

Monitoring and surveillance activities report a great diversity of substances in the Great Lakes environment. The Substance/Sector Workgroup will continue to work with the Canadian and U.S. federal and provincial/regional governments, and others conducting monitoring in the Great Lakes, to identify potential threats to the Great Lakes Basin from emerging chemicals of concern. This work will also involve input from stakeholders.

6.0 STAKEHOLDER FORUM AND INTEGRATION WORKGROUP



Chapel Rock - Pictured Rocks National Lakeshore, Photograph by Raymond J. Malace

Stakeholder Forum and Integration Workgroup meetings have long been a tradition of the GLBTS. The meetings provide an opportunity for stakeholders and the governments to come together, get to know one another, share information, and try to resolve issues of toxic substances affecting the Great Lakes. Beginning in 2009, the Parties decreased the frequency of Stakeholder Forum/ Integration Workgroup meetings to one face-to-face meeting per year. This change reflected several factors affecting the GLBTS, including a declining role of the Level 1 workgroups and transition to new chemicals of concern, and limited travel budgets for many GLBTS stakeholders.

Brief summaries of Stakeholder Forum and Integration Workgroup meetings held over the past two years are presented below.

Stakeholder Forum – December 12, 2007, Chicago

The December 12, 2007, Stakeholder Forum featured a keynote address by Jim Willis of US EPA's Office of Pollution Prevention and Toxics. Mr. Willis presented an overview of the U.S./Canada/Mexico Trilateral Security and Prosperity Partnership (SPP) Agreement on Chemical Management Activities.

With the signing of the SPP agreement in August 2007, the leaders of the U.S., Canada, and Mexico committed to specific goals for enhancing regulatory cooperation among the three countries, accelerating and improving the effectiveness of actions to safeguard health and the environment, providing cost-effectiveness for business and government, and retaining national regulatory authority. U.S. commitments under the SPP include, by the end of 2012, assessing and initiating needed action on the over 9,000 existing chemicals produced above 10 tons/yr in the U.S. Canadian commitments under the SPP include, by the end of 2012, completing assessments and taking regulatory action on the highest priority substances resulting from the DSL categorization, and initiating assessments of medium-priority substances, and by 2020, updating the DSL.

The substance workgroup leaders also reported on progress toward the Strategy challenges for mercury, dioxins/furans, PCBs, and HCB/B(a)P. The forum was followed by substance workgroup break-out sessions for mercury, PCBs, dioxins/ furans, and HCB/B(a)P.

Integration Workgroup Meeting – December 13, 2007, Chicago

The December 13, 2007, Integration Workgroup meeting included updates from the co-chairs of the active substance workgroups (mercury, PCBs, dioxins/furans, and HCB/B(a)P) on the previous day's workgroup meetings. The Dioxin/Furan Workgroup decided to move to inactive status and have the Burn Barrel Subgroup report to the HCB/B(a)P Workgroup (backyard burning is also a source of HCB and B(a)P). The Integration Workgroup also discussed several programs related to the new GLBTS Substance Group.

Presentations at this meeting included:

» *North American Commission for Environmental Cooperation Tri-National Chemicals Management*—Vic Shantora, Commission for Environmental Cooperation Sound Management of Chemicals Program

» *Substance Workgroup Reports*

- *Mercury*—Alexis Cain, US EPA
- *PCBs*—Ken De, EC
- *Dioxins/Furans*—Erin Newman, US EPA
- *HCB/B(a)P*—Tom Tseng, EC

» *Overview of Canada's Chemicals Management Plan (CMP)* —Suzanne Easton, EC

» *Great Lakes Chemicals Screening Project*—Ted Smith, US EPA

» *Terms of Reference for the Substance and Sector Groups*—Ted Smith, US EPA

Stakeholder Forum – June 4, 2008, Burlington

The first Stakeholder Forum of 2008 featured a keynote address by Ms. Susan Boehme, director of the New York/New Jersey Harbor Project from 2000 to 2005. Ms. Boehme presented the findings of the Harbor Project and implications for the GLBTS. Dr. Jianmin Ma of EC presented the results of a modeling study conducted by EC to investigate the impact of intercontinental atmospheric transport of lindane on the North American environment. The meeting also included presentations on the status and achievements of the mercury, PCB, and HCB/B(a)P Workgroups. Having met the GLBTS challenge goals, the Mercury Workgroup discussed decreasing the frequency of face-to-face meetings and examined alternative means of sharing information, such as web-based meetings or focused two-day meetings that are held periodically (e.g., every 2 years). The PCB Workgroup announced the discontinuation of the PCB Recognition and Award program for Canadian companies that voluntarily decommission 90% or more of their in-service PCB equipment. New Canadian PCB regulations have mandated the phase-out of PCB equipment. The HCB/B(a)P Workgroup continued to investigate sources of release to the Great Lakes Basin and identified coal tar sealants as a source of PAHs in storm water runoff. The Stakeholder Forum was followed by an afternoon Integration Workgroup meeting.

Integration Workgroup Meeting – June 4, 2008, Burlington

The June 4, 2008, Integration Workgroup meeting was a shortened half-day meeting that followed a morning GLBTS Stakeholder Forum. The Integration Workgroup meeting focused on the path forward for the Substance and Sector Workgroups. The workgroup discussed the mission and scope of the Substance and Sector Workgroups, a decision framework as an approach for identifying substances of potential concern to the Great Lakes Basin, and stakeholder participation and public engagement in the substance identification process. To reach their goals, it was decided that the two groups would be joined with one name: Substance/Sector Workgroup.

Integration Workgroup Meeting – September 25, 2008, Chicago

At its September 25, 2008, meeting, the Integration Workgroup continued discussions of the path forward for the Substance/Sector Workgroup. The

Stakeholder Highlights: National Wildlife Federation

Progress under the Binational Toxics Strategy, Fall 2008 - Fall 2009

The National Wildlife Federation (NWF) has been involved in several activities involving the GLBTS and chemicals policy more broadly in the Great Lakes region over the past year, including the following:

- NWF made progress on a project assessing the impact of environmental management system (EMS) programs on releases of persistent, bioaccumulative and toxic chemicals at firms in the region. This included working with the Indiana Clean Manufacturing Technology Institute (CMTI) at Purdue University in identifying (through an analysis of TRI data) several dozen candidate firms in the basin to contact concerning serving as potential case studies, contacting a subset of firms, and receiving three completed questionnaires on EMS programs and chemical releases. In addition, NWF received a questionnaire from a publicly owned treatment works facility, and is in the process of finalizing a report which will summarize case study findings and include general recommendations.
- NWF made progress in assessing broader environmental non-governmental organization (ENGO) awareness of the GLBTS process, potential involvement, and interest in broader chemicals policy work through revision to a survey and identification of over 150 Canadian and U.S. groups to survey, with goals of having results and finalizing the report in early 2010.
- NWF also maintained significant involvement in related chemicals policy work, including Michael Murray's involvement in the IJC Chemicals of Emerging Concern project (including providing comments on the draft policy framework document, taking part in the 1½ day Expert Consultation in March 2009, and researching and providing a bibliography of additional papers for the project to consider).
- In addition to involvement in GLBTS Substance/Sector and Integration Workgroup meetings, NWF has maintained involvement in other chemicals policy work involving the GLBTS, including providing written comments on the draft 2008 GLBTS Newsletter and the draft Mercury Phasedown Strategy under the Great Lakes Regional Collaboration. NWF has also continued correspondence with other ENGOs on the future of chemical policy in the region (including discussions involving the Great Lakes Water Quality Agreement), and taken part in webinars by (and shared information with) the Great Lakes Green Chemistry Network.

co-chairs of the Substance/Sector Workgroup proposed to identify candidate substances, in consultation with national programs, to analyze using the group's decision framework provisionally by June 2009. A coalition of ENGOs advocated that the governments pursue the zero discharge and virtual elimination goals of the Great Lakes Water Quality Agreement (GLWQA) and the GLBTS Strategy for toxic chemicals into the Great Lakes (as understood in those agreements, and neither amended nor limited by later Canadian or U.S. legislation). The co-chairs of the active substance workgroups provided updates on the status of the workgroups for mercury, HCB/B(a)P, and PCBs. The Integration Workgroup discussed the frequency of future GLBTS meetings and the format of reporting through the GLBTS annual report and periodic update brochures. The Mercury Workgroup decided that biannual meetings were no longer needed and instead supported larger gatherings to be held occasionally and in collaboration with other regions of the country. The PCB and HCB/B(a)P Workgroups supported reducing the frequency of face-to-face meetings to once a year, with other means of communication utilized between meetings (e.g., teleconferences). The Integration Workgroup also decided to reduce the frequency of preparing a GLBTS progress report

from annually to biennially, or once every two years. Less formal reporting mechanisms, such as newsletters, could be prepared in the interim.

Stakeholder Forum/Integration Workgroup Meeting – December 4, 2008, Chicago

On December 4, 2008, the GLBTS Stakeholder Forum and Integration Workgroup meeting were combined in a full, one-day meeting. This meeting was a milestone in that it was Danny Epstein's last meeting as Canadian co-chair of the GLBTS. Margaret Kenny of EC and Jim Willis of US EPA discussed the status of new chemical management programs in their respective governments, including similarities and differences between the two. The co-chairs of the substance workgroups provided updates on the status of the workgroups for mercury, HCB/B(a)P, dioxin, and PCBs. Of note was the announcement of Canada's PCB regulation, which will significantly improve Canada's progress in achieving the GLBTS goals for PCBs. The meeting included an update of progress in developing a Mercury Emissions Phase-Down Strategy under the GLRC. Progress of the Substance/Sector Workgroup and a timeline for the workgroup from December 2008 to December 2009 were also presented.

GLBTS Update Teleconference – June 23, 2009

In lieu of face-to-face meetings in Windsor, as previously scheduled, a teleconference was held on June 23, 2009, to provide updates on several issues affecting the GLBTS:

- Linda Klaamas of EC and Mark Elster of US EPA, Office of International Affairs, provided an update on the status of the GLWQA Revision.
- Karrisa Kovner of US EPA, Office of Pollution Prevention and Toxics, presented an update, from a U.S. perspective, of a United Nations Environmental Program (UNEP) Persistent Organic Pollutants (POPs) fourth meeting of the Conference of the Parties (COP4) to the Stockholm Convention. Nav Khera of EC, Chemicals Management Division, presented a Canadian perspective of the UNEP POPs Conference and explained other key issues that were discussed at COP4.
- Keith Houck of US EPA, National Center for Computational Toxicology, presented an overview and update on a US EPA Strategic Plan for Evaluating the Toxicity of Chemicals—ToxCast Chemical Prioritization Project.
- Alan Waffle of EC presented a brief overview of the IJC Contaminants of Emerging Concern Nearshore Workgroup Recommendations.
- Ted Smith of US EPA provided a Status Update on Toxics under the Great Lakes Restoration Initiative (GLRI).

GLBTS Update Teleconference – September 23, 2009

On September 23, 2009, a teleconference was held to update GLBTS stakeholders on several initiatives:

- Allan-Paul Dane of EC provided an overview of a new Canadian Great Lakes Chemical Priorities Working Group, which brings together key director-level representatives from Canadian federal government agencies to recommend Canadian chemical priorities in the Great Lakes Basin and to ensure that these are communicated to all programs that address chemicals in the Great Lakes.
- Julie Schroeder of Ontario MOE reported on Ontario's Toxics Reduction Strategy, which includes the Toxics Reduction Act, passed by MOE in June 2009, and subsequent regulations in support of the legislation.
- Linda Klaamas of EC provided information on progress made by the U.S. and Canada to renegotiate the GLWQA.
- Ted Smith of US EPA described an upcoming IJC GLWQA Biennial Meeting scheduled for October 7-8, 2009, in Windsor, Ontario.

Stakeholder Highlights: Industry Continues to Value GLBTS Process

Industry has continued to work extensively with GLBTS program managers and other stakeholders on the evolution of the GLBTS chemical substance management effort throughout a time of transition. Since inception of the GLBTS program, industry has appreciated the opportunity to work with governments, environmental advocacy organizations, and others on chemical issues in this voluntary stakeholder-based forum. Through these interactive activities, industry has joined other stakeholders to successfully achieve chemical release reductions, provide chemical inventory and characterization information, and draft chemical management policies. The process is taking on new dimensions to shift focus from legacy substances to those now being discussed as materials of emerging concern. Industry representatives continue to believe that the unique GLBTS multi-stakeholder voluntary process provides the best opportunity to gain understanding of the significance of the presence of these materials in the ecosystem and to seek the most appropriate action for long term sustainability.

In 2009, highlights of industry participation facilitated by CGLI include:

- CGLI recruited several experts on toxicology and chemical effects to the GLBTS process as the discussion shifted to new substances. The experts evaluated and provided substantive comments on framework proposals for the evaluation of chemicals of emerging concern. Information provided demonstrated the importance of considering both hazard and risk when the significance of presence for these substances is evaluated. Risk factors such as potential for exposure and the specifics regarding exposures are critical elements in an assessment of the significance of a substance's presence in the environment.
- CGLI provided information and experience regarding models used to predict chemical toxicity characteristics based on molecular structure.
- CGLI contributed observations and experience related to ecosystem monitoring and surveillance methodologies that can help differentiate ecosystem impacts related to differing stressors. Industry supports a robust ecosystem-based monitoring and surveillance program in the Great Lakes. This will enable monitoring and surveillance efforts to be guided, reviewed, and the results that are obtained interpreted through a GLBTS workgroup charged to do this work.
- CGLI continues to develop and maintain a robust network of industry personnel that meet regularly via teleconference and actively participate in GLBTS workgroups. They bring important expertise and perspectives to the process.

The GLBTS process can efficiently bring government, academic, ENGO, and industry scientists together to best review and draw conclusions from Great Lakes ecosystem characterization work on a continuing basis.

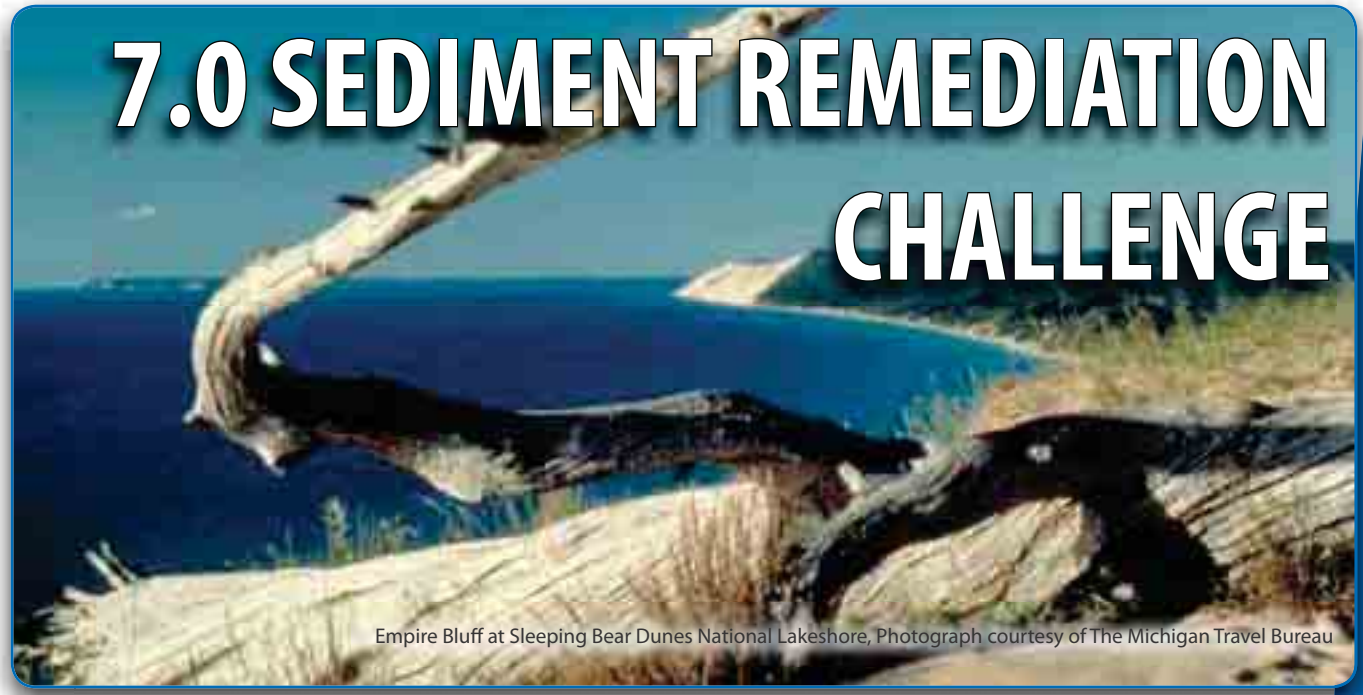
- Alexis Cain of US EPA provided an overview of an upcoming conference scheduled for November 17-18, 2009, in Chicago: 2009 Mercury Science and Policy Conference with a Special Focus on the Northeast and Great Lakes Regions, led by the Northeast Waste Management Officials' Association (NEWMOA). Alexis also described the status of the GLRC Great Lakes Mercury Emissions Reduction Strategy.
- Melanie Neilson and Sean Backus of EC presented an overview of Great Lakes monitoring and surveillance activities under Canada's CMP.
- Ted Smith of US EPA moderated a GLRI Toxics Monitoring and Surveillance panel presentation, which featured the following speakers:

- Kimani Kimbrough of NOAA discussed NOAA's Enhanced Mussel Watch Program in the Great Lakes.
- Dave DeVault of the U.S. Fish and Wildlife Service (USFWS) described an early warning system to identify effects of new contaminants.
- Charlie Peters of the USGS described a proposed effort to monitor contaminants in Great Lakes tributaries.
- Tom Custer of USGS described monitoring effects of contaminants on Great Lakes indicator species.
- Todd Nettesheim of US EPA Great Lakes National Program Office (GLNPO) described US EPA's efforts to develop critical information through monitoring and surveillance.

Stakeholder Forum/Integration Workgroup Meeting – December 3, 2009, Chicago

A combined Stakeholder Forum and Integration Workgroup meeting was held in Chicago on December 3, 2009. The meeting included updates on the status and progress of the PCB, HCB/B(a)P, Dioxin/Furan, Mercury, and Substance/Sector Workgroups. The Dioxin/Furan and Mercury Workgroups are inactive, but the workgroup co-chairs provided updates such as the 2007 inventory of dioxin emissions in Ontario. The formation of a new GLBTS Monitoring and Surveillance Workgroup was proposed for the purpose of evaluating ongoing monitoring and surveillance efforts and identifying potential new chemical threats to the Great Lakes ecosystem. The meeting featured a Green Chemistry Panel discussion with Rui Resendes of Green Centre Canada and Lin Kaatz Chary of the Great Lakes Green Chemistry Network.

7.0 SEDIMENT REMEDIATION CHALLENGE



Empire Bluff at Sleeping Bear Dunes National Lakeshore, Photograph courtesy of The Michigan Travel Bureau

Under the Great Lakes Binational Toxics Strategy, EC and US EPA committed to:

"Complete or be well-advanced in remediation of priority sites with contaminated bottom sediments in the Great Lakes Basin by 2006."

Highlights of sediment assessment and remediation activities undertaken in the U.S. and Canada are described below.

2009 Sediment Assessments with US EPA's Research Vessel *Mudpuppy*

Contaminated sediments are a significant concern in the Great Lakes Basin. Although toxic discharges have been reduced over the past 30 years, high concentrations of contaminants still remain in the sediments of many rivers and harbors. These sediments are of potential risk to the health of aquatic organisms, wildlife, and humans.

To assist in determining the nature and extent of sediment contamination at these polluted sites, US EPA GLNPO operates the Research Vessel (R/V) *Mudpuppy*. The R/V *Mudpuppy* is a 32-foot-long, flat-bottom boat that is specifically designed for sampling sediment deposits in shallow rivers and harbors. The boat is able to sample at water depths between 2 feet and 50 feet. Using a vibrocoring unit, the R/V *Mudpuppy* can take sediment core samples of up to 20 feet in depth.

To adequately characterize a site, GLNPO uses an integrated sediment assessment approach. This involves collecting data for sediment chemistry, toxicity, and the benthic community at a specific site, and then using the results to determine the extent of contamination that could be impacting the aquatic ecosystem.

Since 1993, the R/V *Mudpuppy* has conducted surveys at 41 locations, including 28 of the 31 original U.S. Great Lakes Areas of Concern (AOCs). In 2009, the following surveys were conducted with the assistance of the R/V *Mudpuppy*:

- **Lake Calumet, Chicago, IL** – Assisted the University of Illinois at Chicago with sampling to investigate in-situ PBDE debromination in sediments.
- **Indiana Harbor, East Chicago, IL** – Assisted the University of Iowa with sampling to determine the potential for PCB flux from the sediments.
- **Rouge River, Detroit, MI** – Conducted sampling to determine the nature (chemistry,

toxicity, geotechnical properties) and extent of sediment contamination.

- **River Raisin, Monroe, MI** – GLNPO collected sediment samples to define chemical and physical properties of sediment and to delineate horizontal and vertical extent of contamination.
- **Ashtabula River, Ashtabula, OH** – GLNPO sampled surface sediment to evaluate post-remediation sediment concentrations at the Great Lakes Legacy Act (GLLA) site.
- **Cuyahoga River, Cleveland, OH** – GLNPO collected sediment samples to determine the nature and extent of contamination in the sediments.
- **Trenton Channel, Trenton, MI** – Assisted the US EPA RCRA program with oversight of field sampling activities.

Great Lakes Sediment Remediation Projects - 2008[37]

In 2008, approximately 740,000 yd^3 of contaminated sediment were remediated from seven U.S. sites and one Canadian site in the Great Lakes Basin. Remedial action was initiated for the first time in 2008 at one U.S. site and one Canadian site; that same Canadian site and two U.S. sites completed their remedial actions in 2008. Four U.S. sites, each under a different cleanup authority, continued to make progress in their remedial actions. The following is a list of specific details about each site.

U.S. Sites

St. Louis River/Interlake/Duluth Tar, Duluth, Minnesota – The St. Louis River/Interlake/Duluth Tar (SLRIDT) Superfund site is a state-led National Priority List (NPL) site. In 2008, remedial activities consisted of the completion of placement of cover sand and armor material in Stryker Bay; completion of the 54th Avenue south wetland excavation of approximately 4,000 yd^3; placement of covers on both the south and north wetlands; dredging of approximately 26,000 yd^3 of contaminated sediments located in the Federal Navigation Channel and waters of the State of Wisconsin (south of the confined aquatic disposal end dike in Slip 6 and Minnesota Channel dredging); and completion of Slip 7 capping, cover, and armoring. Additionally, the Stryker Bay cap/surcharge continued to settle.

Hayton Area Remediation Project, Calumet County, Wisconsin – The 2008 removal was the first phase of removing what is likely the largest PCB deposit in the project area. PCB-contaminated sediment has accumulated in a series of wetlands formed by glacial esker constrictions of the Pine Creek valley about three miles downstream from the release point. PCB concentrations in the first wetland are as high as 2,600 ppm with much of the deposit having concentrations of more than 50 ppm. Removal activities will continue in 2009. Removal is being conducted by isolating and pumping the work area followed by mechanical removal. Contaminated sediment with concentrations of less than 50 ppm is being disposed of at a local landfill. A landfill in Michigan is the disposal location for sediment with concentrations of 50 ppm or more.

Lower Fox River, Operable Unit (OU) 1, Green Bay, Wisconsin – In June 2008, the dredging portion of the remedial work in OU1 (Little Lake Butte des Morts) was completed by two responsible parties under a court-approved consent decree with Superfund and the Natural Resource Damage Assessment (NRDA) Trustees. Approximately 41,000 yd^3 were hydraulically dredged in 2008, bringing the total volume of contaminated sediment removed up to 370,000 yd^3. PCB-contaminated sediment was placed into geotubes for dewatering; the water was treated on-site and returned to the river. Contaminated sediment was taken to a nearby landfill for proper disposal. Remaining areas with lower levels of PCBs were capped with approximately 245,000 yd^3 of sand and gravel. The OU 1 project has a 1 ppm action level for PCBs and a surface weighted average concentration (SWAC) standard of 0.25 ppm.

Allied Paper, Inc./Portage Creek/Kalamazoo River, Kalamazoo, Michigan – The second phase of a Time Critical Removal Action (TCRA) was

[37] Sediment remediation data for 2008 are presented because data lag a year behind in reporting (i.e., 2009 data will become available in 2010).

initiated by Georgia-Pacific and Millennium Holdings contractors as a result of agreements negotiated by the two companies, US EPA Superfund, MDEQ, and the Natural Resource Trustees. In March 2008, approximately 83,000 yd^3 of PCB-contaminated sediment were dredged from the Kalamazoo River near the Plainwell Impoundment. Sediment with more than 50 ppm PCB content was sent to MDEQ's Wayne Disposal facility in Belleville, MI. Less contaminated material below 50 ppm was sent to Allied Waste's C & C Landfill near Marshall, MI, and its Ottawa Farms Landfill near Coopersville, MI.

Tittabawassee River, Reach D, Midland, Michigan – In April 2008, approximately 130 yd^3 of dioxin-contaminated sediment were hydraulically dredged from Reach D of the Tittabawassee River, completing the two-year removal project required by a consent order between US EPA and the Dow Chemical Company. Sediment was pumped via pipeline to a containment facility for dewatering, and then disposed of at Dow's Salzburg Landfill.

Ashtabula River, Ashtabula, Ohio – In 2008, the U.S. Army Corps of Engineers (US ACE) Buffalo District hydraulically dredged 132,904 yd^3 from the Ashtabula River as authorized by Operations and Maintenance (O&M) under Section 1 of the Rivers and Harbors Act of 1937 and Water Resources Development Act (WRDA) of 1986, and by Section 312(a) and (f)(3) of WRDA 1990, Public Law 101-640, as amended (33 U.S.C. 1272). To determine the final sediment disposition, the sediment was sampled, analyzed, and evaluated in accordance with guidance contained in the Great Lakes Dredged Material Testing and Evaluation Manual. Based on this approach, the dredged material was determined to be unsuitable for open lake placement, and was therefore pumped via pipeline to the TSCA-permitted disposal facility specifically constructed for Ashtabula River sediments as part of the Great Lakes Legacy Act project.

Buffalo River, Buffalo, New York – In 2008, the US ACE Buffalo District mechanically dredged 78,460 yd^3 from the Buffalo River as part of the US ACE's O&M dredging mission. To determine the disposal location, the sediment was sampled, analyzed, and evaluated in accordance with

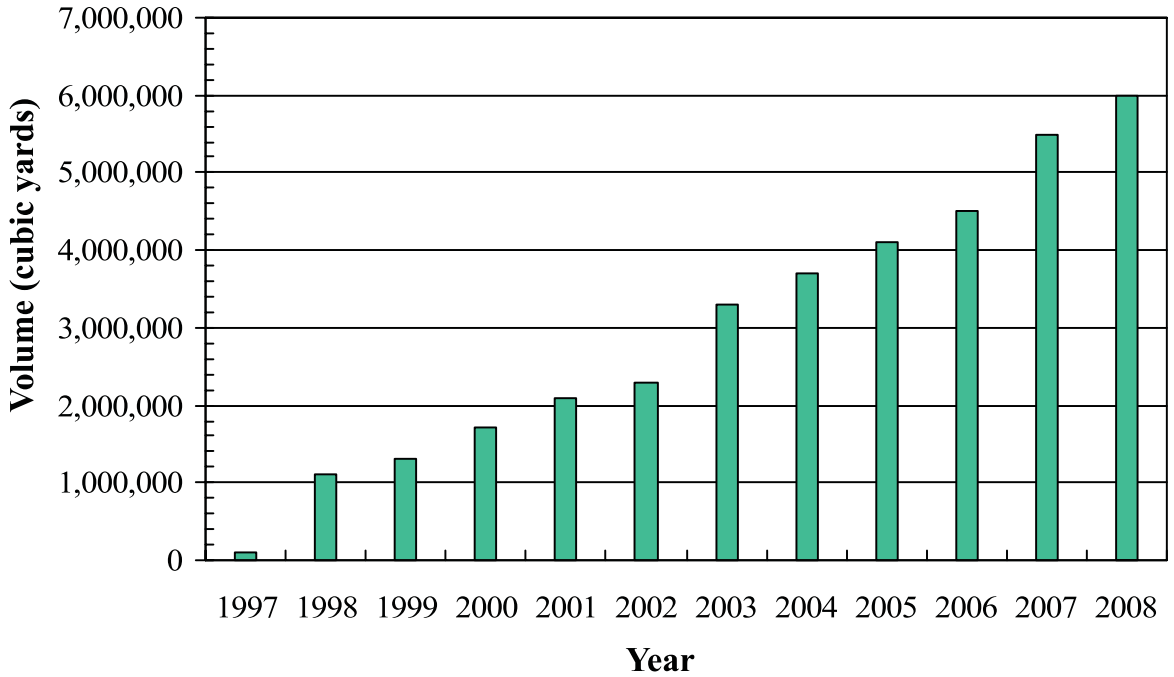


Figure 7-1. Cumulative Volume of Sediment Remediated in the U.S. Since 1997. Source: US EPA Great Lakes National Program Office[38]

[38] Volumes in bar graph are quantitative estimates as reported by project managers, summed, and then rounded to the nearest one hundred thousand cubic yards. Data collection and reporting efforts are described in the "Great Lakes Sediment Remediation Project Summary Support" Quality Assurance Project Plan (GLNPO, June 2008). Detailed project information is available upon request from project managers.

guidance contained in the Great Lakes Dredged Material Testing and Evaluation Manual. Based on this approach, the dredged material was determined to be unsuitable for open lake placement, and was therefore barged to and placed in the Buffalo confined disposal facility (CDF).

Figure 7-1 presents the cumulative volume of sediment remediated in the U.S. since 1997. Information included in the bar graph represents quantitative estimates as reported by project managers. Data collection and reporting efforts are described in the *Great Lakes Sediment Remediation Project Summary Support, Quality Assurance Project Plan*.[39] Detailed project information is available upon request from project managers.

Canadian Sites

Sediment Remediation Guidance

Canada-Ontario Decision-Making Framework for Assessment of Great Lakes Contaminated Sediments – A risk-based decision-making framework for contaminated sediments was completed under the 2002-2007 *Canada-Ontario Agreement Respecting the Great Lakes Basin Ecosystem* (COA). The MOE has integrated the document with existing guidance to produce *Guidelines for Identifying, Assessing and Managing Contaminated Sediments in Ontario: An Integrated Approach*. The guidance document is currently applied throughout the province. The Canada-Ontario Decision Making Framework is being applied to evaluate the need for sediment management actions in a number of project sites in the AOCs.

Remediation Update

The following information provides a status report on all sites in the Canadian AOCs that involve sediment investigations and known or potential sediment remediation projects.

Bay of Quinte (Trent River) – As part of the ongoing monitoring work to assess sediment quality, elevated levels of dioxins and furans were found in sediment at the mouth of the Trent River in 2001. An Ecological Risk Assessment completed in 2007 predicted that there is negligible risk to piscivorous wildlife and fish exposed to the contaminated sediment. As such, monitored natural recovery was chosen as the preferred management option for this site. Studies to control the off-site migration of contamination into the river are continuing in the area.

Detroit River (Turkey Creek) – Turkey Creek upstream of Walker Road has elevated PCB and metal concentrations. Local stakeholders were consulted, and negotiations with MOE, EC and municipal and industry representatives regarding cleanup of this site were completed. The creek and its banks were successfully remediated between August and November in 2008, which resulted in the removal of 975 m^3 of contaminated sediment (including 8 kg of PCBs).

Hamilton Harbour (Randle Reef) – An engineering design study for the Randle Reef remedial option is nearing completion. An engineered containment facility about 7.5 hectares in size is being designed to contain in-situ 130,000 m^3 and another 500,000 m^3 of hydraulically dredged PAH-contaminated sediments. An Environmental Comprehensive Study Report is being completed for agency and public review. Federal and provincial funding commitment of $60 million has been made for the remediation itself, and further municipal and industry stakeholder participation is being sought. Construction could begin in 2011 and extend to 2019.

Niagara River (Lyon's Creek, East & West) – The Lyon's Creek watercourse is bisected by the Welland Canal.

Arsenic-contaminated sediment from Lyon's Creek West was excavated (300 m^3) in the summer of 2007 and placed in a secure landfill facility. The geographic extent of the PCB contamination in the sediment and soil of Lyon's Creek West is currently being investigated by Transport Canada, the major landowner of the contaminated site. It is anticipated

[39] US EPA. (2008). *Quality Assurance Project Plan for Great Lakes Sediment Remediation Project Summary Support*. Revision 1.0. Unpublished GLNPO document available from Mary Beth G. Ross (ross.marybeth@epa.gov).

that removal and offsite disposal will take place in 2010/2011.

In August 2008, Monitored Natural Recovery was selected as the preferred management option to address PCB-contaminated sediments in Lyons Creek East in order to protect the Provincially Significant Wetland.

Peninsula Harbour – Results of assessments of mercury and PCB bioaccumulation and ecological risk have indicated the need for sediment management. Sediment management options were assessed in consultation with local stakeholders. In 2008, thin layer capping was chosen as the preferred remedial option.

Port Hope Harbour – Remedial investigations on harbour sediments are focusing on the uranium and thorium series radionuclides, and secondarily on heavy metal contamination (particularly arsenic, copper, lead and nickel) and PCBs. Remediation of harbour sediment is planned as part of the *Port Hope Area* Initiative to clean up historic low-level radioactive waste in the Port Hope area, pursuant to a March 2001 agreement between the federal government and local municipalities.

Preliminary design descriptions indicate that hydraulic dredging has been identified as the most appropriate means of remediating the approximately 110,000 m^3 of contaminated sediments.

The sediment slurry will be conveyed through a floating pipeline to a sediment dewatering area, where it will be injected into sediment containment tubes. De-watered sediment will be deposited in a long-term waste facility designed for the safe disposal of low-level radioactive waste. Detailed engineering design is planned for 2010. Remediation is planned for 2013 and 2014.

St. Clair River (Zones 2 & 3) – Zones 2 and 3 are downstream from the "Chemical Valley" area of Sarnia. Various sediment investigations have been undertaken. The Canada-Ontario Decision-Making Framework is being applied to data from 2004 to the present (post Zone 1 remediation). It is anticipated that priority areas will be identified and an evaluation of the need for management interventions conducted in 2009.

St. Marys River (Bellevue Marine Park) – Assessments of contaminated sediment at the Bellevue Marine Park (BMP) location were undertaken in the fall of 2006. In addition, an assessment of contaminated sediment at two areas downstream of BMP was completed in 2008. Reports from both assessments will be completed in 2009.

Thunder Bay (North Harbour) – Results of assessments of mercury bioaccumulation and ecological risk have indicated the need for sediment management. Sediment management options are currently being assessed in consultation with local stakeholders.

Wheatley Harbour – An Ecological Risk Assessment undertaken in 2007 concluded that there is negligible risk of PCB effects to piscivorous wildlife in the Muddy Creek wetland. Therefore, the Wheatley Harbour Implementation Team recommended that no further action is required in this AOC prior to delisting.

Supporting Table and Graphics

Table 7-1 reports progress on sediment remediation projects at both AOCs and non-AOCs in the U.S. and Canada, from 1997 through 2008. Figure 7-2 illustrates the progress and achievements made in sediment remediation activities in the Great Lakes in 2008. Information included in the tables and map represents quantitative estimates as reported by project managers. Data collection and reporting efforts are described in a US EPA Quality Assurance Project Plan.[40] Detailed project information is available upon request from project managers. On occasion, project managers may submit to GLNPO updated sediment remediation estimates on projects previously reported. Readers should always refer to the most current version of the *GLBTS Progress Report* or GLNPO's Contaminated Sediments website at *www.epa.gov/glnpo/sediment/remed/index.html* for the most up-to-date sediment remediation estimates.

[40] US EPA. (2008). Op. cit.

Table 7-1. Progress on Sediment Remediation in the Great Lakes since 1997*

Site/AOC/non-AOC (*)	Cumulative Mass of Contaminant Remediated (kg)												Cumulative Volume Sediments Remediated 1997 thru 2008 (yd^3)	Volume Sediments Remediated 2008 (yd^3)	Ultimate Disposition
	aldrin/ dieldrin	benzo(a) pyrene	chlordane	DDT (+DDE/DDD)	hexachloro-benzene	alkyl-lead	mercury & compounds	mirex	octachloro-styrene	PCBs	dioxins/ furans	toxaphene			
U.S. Sites															
Alma Iron and Metal/Smith Farms Property*													15,904		Encapsulated on-site
Ashtabula River, OH - Great Lakes Legacy Act - Navigation Dredging										 6,000			**629,490** 496,586 132,904	 132,904	On-site TSCA landfill
Black River, OH															
Black River, MI* - CR 681													 25,000		 Landfilled
Buffalo River, NY - Buffalo Color - Area D - Navigation Dredging													**206,421** 45,000 161,421	 78,460	 Encapsulated on-site CDF
Clinton River, MI															
Cuyahoga River, OH															
Deer Lake - Carp River, MI															
Detroit River, MI - Monguagon Creek - Black Lagoon - BASF Riverview													**166,500** 25,000 115,000 26,500		 Landfilled CDF Encapsulated on-site
Eighteenmile Creek, NY															
Fields Brook Superfund, OH*													53,094		Landfilled
Fox River, Green Bay, WI - Deposit 56/57 - Deposit N - Deposit O - OU 1 - Phase 1										**1,829** 950 51 828			**917,809** 81,662 7,149 1,026 695,972 132,000	 366,485	 Landfilled Landfilled Landfilled Landfilled/capped Landfilled
Grand Calumet, IN - U.S. Steel/Gary Works - U.S.S. Lead							382			7,897	0.03		**865,570** 840,200 25,370		 On-site CAMU CAMU & TSCA facility
Kalamazoo River, MI - Bryant Mill Pond - Allied Paper/Portage Creek										10,000			**274,000** 150,000 124,000	 87,000	 Landfilled Off-site TSCA/landfill

Site/AOC/non-AOC (*)	Cumulative Mass of Contaminant Remediated (kg)												Cumulative Volume Sediments Remediated 1997 thru 2008 (yd^3)	Volume Sediments Remediated 2008 (yd^3)	Ultimate Disposition
	aldrin/ dieldrin	benzo(a) pyrene	chlordane	DDT (+DDE/DDD)	hexachloro-benzene	alkyl-lead	mercury & compounds	mirex	octachloro-styrene	PCBs	dioxins/ furans	toxaphene			
Manistee Lake, MI*															
Manistique River, MI										4,771			161,162		Landfilled
Manitowoc River, WI* - HARP										1,570			27,150	9,750	Off-site TSCA facility and landfilled
Maumee River, OH - Fraleigh Creek (Unnamed Tributary)										25,400			8,000		Landfilled
Menominee River, MI/WI - Ansul Eighth Street Slip													13,000		Landfilled/ awaiting further management
Milwaukee Harbor, WI - North Ave. Dam - Moss American													**29,960** 8,000 21,960		 Landfilled Landfilled
Muskegon Lake, MI - Ruddiman Creek													90,000		Landfilled
National Gypsum** - Alpena, MI															
Niagara River, NY - Scajaquada Creek - Gill Creek - Cherry Farm/River Road - Niagara Transformer													**77,850** 17,500 6,850 42,000 11,500		Landfilled
Paw Paw River, MI* - Aircraft Components													349		Landfilled
Pine River, MI* - Velsicol Chemical SF Site - TPI Petroleum, Inc.				351,080									**718,076** 669,975 48,101		 Landfilled Landfilled
Presque Isle Bay, PA															
River Raisin, MI - Ford Monroe Outfall - Consolidated Packaging Corp.										16,795			**57,000** 27,000 30,000		 On-site TSCA facility TSCA landfill/landfilled
Rochester Embayment, NY															
Rouge River, MI - Evan's Product Ditch - Newburgh Lake										**250,000** 4,000 246,000			**406,900** 6,900 400,000		Off-site TSCA facility and landfilled
Saginaw River/Bay, MI - NRDA										4,500			**360,213** 342,433		Off-shore CDF

Site/AOC/non-AOC (*)	Cumulative Mass of Contaminant Remediated (kg)												Cumulative Volume Sediments Remediated 1997 thru 2008 (yd^3)	Volume Sediments Remediated 2008 (yd^3)	Ultimate Disposition
	aldrin/ dieldrin	benzo(a) pyrene	chlordane	DDT (+DDE/DDD)	hexachloro-benzene	alkyl-lead	mercury & compounds	mirex	octachloro-styrene	PCBs	dioxins/ furans	toxaphene			
- Lake Linton - Wickes Park													17,000 780		Landfilled Landfilled
Sheboygan Harbor, WI										250			20,727		Off-site TSCA facility & landfilled
Shiawassee River, MI*													63		Landfilled
St. Clair River, MI															
St. Lawrence River, NY - Reynolds Metals/Alcoa E. - Alcoa Grasse River ROPS										10,000			**112,000** 86,000 26,000		Landfilled/capped Landfilled
St. Louis River/Bay, MN/WI - Newton Creek/Hog Island Inlet - Interlake/Duluth Tar													**359,643** 52,143 307,500	68,000	Landfilled Capped/on-site CAD
St. Marys River, MI - Cannelton - Tannery Bay							33						**42,912** 3,000 39,912		Landfilled Landfilled
Ten Mile Storm Drain* - St. Clair Shores, MI													18,500		Landfilled
Tittabawasee River, MI* - Reach D - Reach O													**28,528** 12,130 16,398	130	Landfilled
Torch Lake, MI															
Torch Lake, MI USX Vessel Slip*													3,200		On/off-site landfilled
Waukegan Harbor, IL															
White Lake, MI - Tannery Bay - Occidental Chemical Corp.					495†					495†			**105,500** 95,000 10,500		Landfilled Landfilled
Willow Run Creek, MI*										200,000			450,000		On-site TSCA facility
Wolf Creek - Unnamed Tributary, MI*													1,948		Landfilled
TOTALS				**351,080**	**495†**		**415**			**539,507**	**0.03**		**6,246,469**	**742,729**	

*Denotes non-area-of-concern sites.

† Mass displayed is the combined total of PCBs and HCB.

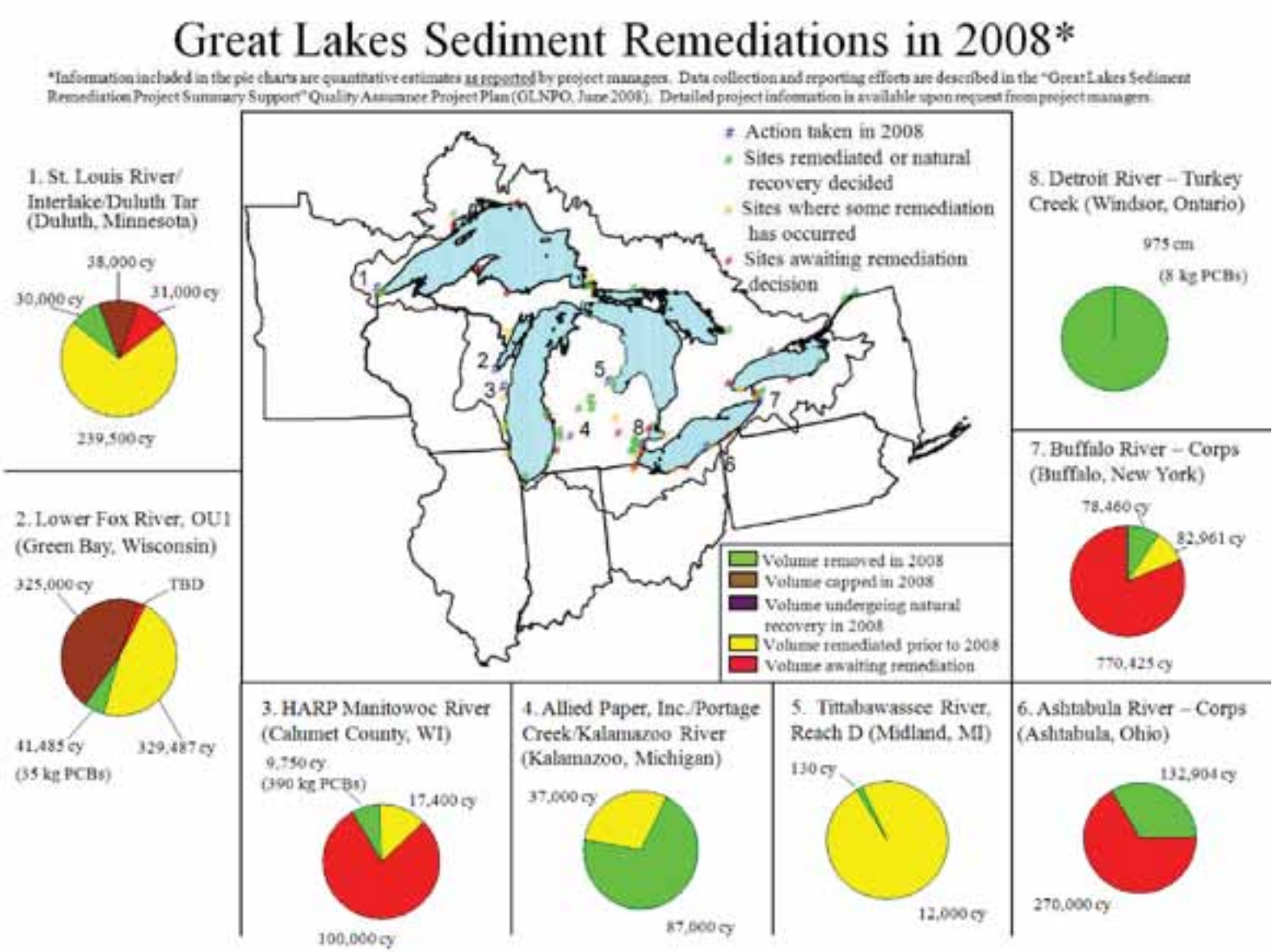
Abbreviations: CAD = confined aquatic disposal; CAMU = corrective action management unit; CDF = confined disposal facility; TSCA = Toxic Substances Control Act

Footnote: *Information included in the matrix are quantitative estimates as reported by project managers. Data collection and reporting efforts are described in the "Great Lakes Sediment Remediation Project Summary Support" Quality Assurance Project Plan (GLNPO, June 2008). Detailed project information is available upon request from project managers.*

Site/AOC/non-AOC	Cumulative Mass of Contaminant Remediated (kg)												Cumulative Volume Sediments Remediated 1997 thru 2008 (cm)	Volume Sediments Remediated 2008 (cm)	Ultimate Disposition
	aldrin/ dieldrin	benzo(a) pyrene	chlordane	DDT (+DDE/DDD)	hexachloro-benzene	alkyl-lead	mercury & compounds	mirex	octachloro-styrene	PCBs	dioxins/ furans	toxaphene			
Canadian Sites															
Bay of Quinte - Trent River															
Detroit River - Turkey Creek (Windsor)										8			975	975	Landfilled
Hamilton Harbour - Randle Reef - Windermere Basin - Dofasco Boatslip															
Jackfish Bay															
Niagara River (Ontario) - Lyons Creek East - Lyons Creek West													300		Landfilled
Nipigon Bay															
Peninsula Harbor															
Port Hope															
St. Clair River - Dow Chemical - Zones 2 & 3							19.3						13,690		Landfilled
St. Lawrence River - Cornwall															
St. Marys River - Algoma Boatslip - Bellevue Marine Park													2,630		Landfilled
Severn Sound															
Spanish River															
St. Lawrence River - Cornwall															
Thunder Bay - Northern Wood Preservers - North Harbour		2,700											11,000 21,000		Thermal treatment Berm enclosure & capped
TOTALS		**2,700**					**19.3**			**8**			**49,595**	**975**	

Footnote: Information included in the matrix are quantitative estimates as reported by project managers. Data collection and reporting efforts are described in the *Great Lakes Sediment Remediation Project Summary Support Quality Assurance Project Plan* (GLNPO, June 2008). Detailed project information is available upon request from project managers.

Figure 7-2. Great Lakes Sediment Remediations in 2008. Source: US EPA Great Lakes National Program Office



8.0 LONG-RANGE TRANSPORT CHALLENGE

Canadian Workgroup co-chair: Jianmin Ma (replacing S. Venkatesh)
U.S. Workgroup co-chair: Todd Nettesheim

Storm Front, Photograph courtesy of U.S. EPA

Under the Great Lakes Binational Toxics Strategy, EC and US EPA committed to:

"Assess atmospheric inputs of Strategy substances to the Great Lakes. The aim of this effort is to evaluate and report jointly on the contribution and significance of long-range transport of Strategy substances from worldwide sources. If ongoing long-range sources are confirmed, work within international frameworks to reduce releases of such substances."

The following efforts are presented as examples of projects undertaken in support of the above challenge.

Numerical Assessment of the Impact of Regional and Global Emissions, Intra- and Inter-continental Atmospheric Pathways of Polybrominated Diphenyl Ethers (PBDEs) and Dioxins/Furans on the North American and the Great Lakes Environments - Current Research Program and Progress

Prepared by: Jianmin Ma, Yifan Li, and Anita Wong, Environment Canada

EC's global atmospheric transport model for persistent toxic chemicals, Canadian Model for Environmental Transport of Organochlorine Pesticides (CanMETOP), has been applied to simulate the atmospheric transport and multi-compartment fate of PBDEs and dioxins/furans. A gridded global emissions inventory of PBDEs has been established subject to the usage, human development index and population intensity index. Based on currently available information of PBDE usage, the U.S. is the largest source of penta-BDE, followed by Western Europe, Canada, and China. Multiple model scenario runs have been conducted using this emissions inventory. The contribution of emissions from those major source regions to the total deposition (dry deposition + wet deposition) of PBDEs to the North American environment was assessed numerically. The present modeling investigations indicate that U.S. and Canadian emission sources made the largest contribution to the loading of penta-BDE to North American terrestrial surfaces, followed by China, India, and Western Europe. The modeling results also suggest that episodic trans-Pacific atmospheric transport is a primary atmospheric pathway that delivers PBDEs from East and South Asia to North America. While sources of dioxins/furans in the U.S., Canada and Western Europe

have been well-identified, China has been regarded as a major source of dioxins/furans globally in recent years. Identifying dioxin/furan emissions in China is a major gap in compiling a global dioxin/furan emissions inventory. Progress has been made in the establishment of an emissions inventory of dioxins/furans in China, especially southern and eastern China, which are the most industrialized regions. Ongoing modeling studies of global atmospheric transport and source-receptor relationships of dioxins/furans will provide detailed information on air and soil concentrations, atmospheric transport, and depositions to Canada and the Great Lakes environment.

Quantifying the Contributions to γ-HCH Deposited to North America and Great Lakes from Major Source Regions

Prepared by: Yi-Fan Li[41, 42], Chong-Guo Tian[42], Nan-Qi Ren[42], Jianmin Ma[41], S. Venkatesh[41]

Abstract

A joint project "China – North America Joint Project on Reduction of Lindane Usage in China and its Impact Globally and on North America" between the North American (NA) Commission for Environmental Cooperation (CEC) and the International Joint Research Center for Persistent Toxic Substances (IJRC-PTS), Harbin Institute of Technology (HIT) began in 2005. Funded by EC, CEC, US EPA, and HIT, the project explored the impact of airborne HCH from China and other major sources to the environment of North America and the Great Lakes. The project aimed to quantify the contributions of γ-HCH deposited to North America and the Great Lakes from major source regions worldwide. Hexachlorocyclohexane (HCH) is a Level 2 substance under the GLBTS.

Using a recently constructed global γ-HCH emission inventory as input, the CanMETOP was employed to simulate the atmospheric transport and deposition of γ-HCH in this study. Modeled air concentrations of γ-HCH matched well with measured data both spatially and temporally, indicating the reasonable accuracy of both the inventories and modeled results. Total depositions of γ-HCH due to global sources in 2005 were 30 tonnes (t) in Canada, 12 t in the USA, and 1 t in Mexico. In Canada, the percentage contribution from major global sources was 93%, of which 7% was from China, 8% from India, 2% from the Former Soviet Union (FSU), 3% from Europe (the FSU is excluded), and 72% from North America. In the USA, total deposition of γ-HCH due to global sources was 82%, of which 17% was from China, 31% from India, 6% from Europe, 4% from FSU, and 25% from North America. In Mexico, global sources contributed 71% of total deposition, of which 11% was from China, 39% from India, 9% from Europe, 2% from FSU, and 10% from North America. Total deposition of γ-HCH in the Great Lakes due to global sources in 2005 was 386 kg, and contributions from the five major sources were 3.2% from Europe, 68% from North America, 7.7% from China, 1.6% from FSU, and 12% from India. The remaining 7.5% was from other sources.

Introduction

1,2,3,4,5,6-HCH, is an organochlorine pesticide (OCP) and belongs to the group of POPs. HCH is available in two technical formulations. Technical HCH is the mixture of several isomers in the proportions α, 53–70%; β, 3–14%; γ, 11–18%; δ, 6–10%; ε, 3–4%. The other formulation, lindane, contains the only insecticidal isomer, γ-HCH. Due to its effectiveness and low price, technical HCH was one of the most widely used insecticides in the world (Willett, et al., 1998). Although production and use of technical HCH has been banned worldwide, lindane had still been produced and used in some countries until the middle of the 2000s. Since HCH is a toxic and persistent pollutant of concern, the CEC North American Regional Action Plan (NARAP) on lindane (γ-HCH) and other HCH isomers seeks to identify and quantify the sources of both HCH production and

[41] Science and Technology Branch, Environment Canada, 4905 Dufferin Street, Downsview, Ontario, Canada M3H 5T4
[42] International Joint Research Center for Persistent Toxic Substances (IJRC-PTS), State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, Harbin, China

atmospheric transport of various HCH isomers in order to quantify and assess its impact on the North American and the Arctic environment. The three North American countries, Canada, Mexico, and the U.S., are seeking to determine practical options for managing the risks associated with HCH isomers, especially γ-HCH.

Methods

Global Soil Residues of γ-HCH in 2005

Gridded soil residue inventories of γ-HCH are crucial to the modeling of atmospheric transport and deposition for this chemical. Global γ-HCH soil residues in 2005 were used as initial conditions for the model simulations (Figure 8-1). Original global γ-HCH soil residues were obtained based on data from historical usage of technical HCH and lindane in the world (Li et al., 2004). As shown in the figure, the major sources of γ-HCH across the world in 2005 were in India, China, central Europe, and the Canadian Prairie provinces. According to our calculation, total soil residues of γ-HCH in 2005 were 13600 t in the world and 1900 t in China, 3000 t in India, 1200 t in the FSU, 3700 t in Europe (excluding the FSU, used throughout this paper or specified), and 2200 t in North America. The total amount of γ-HCH in soils of the five regions consists of 88% of global total residues. Fresh use of substances containing γ-HCH, such as the current use of lindane in India and accidental emission due to agitation of external force on those sinks, such as cultivation of agricultural soil, may lead to a sharp rise in concentrations of the chemical in the atmosphere in the local region. However, detailed information on these causes is missing (Abhilasha et al., 2008). Thus, it was assumed in the research that neither technical HCH nor lindane was used in 2005, and only emission of γ-HCH from soil was considered.

Results and Discussion

Annual Air Concentrations

Figure 8-2 shows modeled global annual mean air concentrations of γ-HCH at 1.5 m above ground level in 2005. It is expected that the high air concentration happened in the major source regions, such as India, Europe, China, and Canadian Prairie provinces. It is interesting to note that the annual mean air concentration was the highest in India but the soil concentration was not (see Figure 8-1). This phenomenon is attributed to the effect of higher temperature in India, which leads to higher volatilization of γ-HCH (Wania et al., 1995; Wania et al., 1998). The figure also depicts that long-range atmospheric transport (LRAT) of the chemical is weak near ground due to influences of surface drag over land, turbulent diffusion and exchange between water/soil and air at the near ground, since relatively high air concentrations of γ-HCH are only found in and/or close to those mainly source regions (Zhang et al., 2008; Koziol et al., 2006). This distribution pattern is different at 3000 m height, as shown in Figure 8-2b, due to the LRAT occurring in the mid-troposphere (Zhang et al., 2008; Koziol et al., 2006). Relatively high air concentrations can be identified almost all over the northern hemisphere in the upper air, demonstrating the existence of LRAT in upper air, as reported by Zhang et al., 2008 and Koziol et al., 2006. This can be attributed to the pattern of atmospheric circulations (Zhang et al., 2008). A marked extending plume of γ-HCH from the major source regions toward the east is clearly found in Figure 8-2b, which is associated with prevailing westerly winds over mid-high latitude zone. Another extending air plume of the substance appears toward the west near the equatorial zone caused by trade winds over the belt. Therefore, North America seems to experience a converging attack from both sides (Zhang et al., 2008).

Figure 8-3 shows modeled gridded annual mean air concentrations of γ-HCH at 1.5 m above ground level in the Great Lakes in 2005. It is expected that annual mean air concentrations are the highest around Lake Erie and Lake Ontario. This phenomenon is attributed to soil residues of γ-HCH around the lakes due to historical use of technical HCH and lindane on agricultural lands, especially around Lake Erie and Lake Ontario (Figure 8-1).

Annual Deposition

Atmospheric γ-HCH dry and wet deposition was simulated by the model. Dry deposition was calculated from the effective deposition velocity of particles at 1.5 m multiplied by the concentration

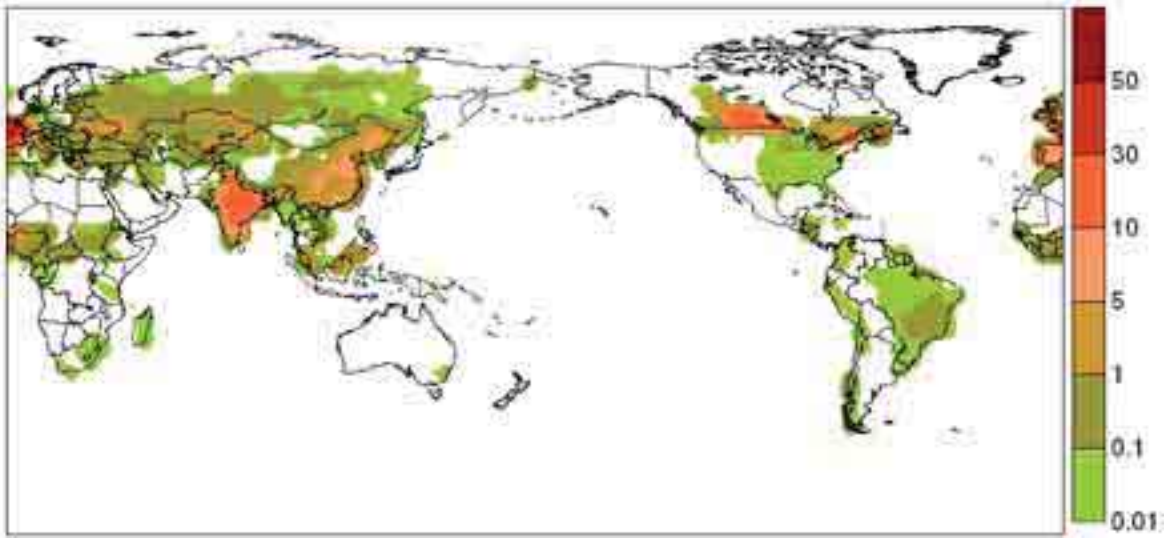


Figure 8-1. **Gridded Global γ-HCH Soil Residues (t cell^{-1}) in 2005 with 1°×1° Latitude/Longitude Resolution. Source: Environment Canada.**

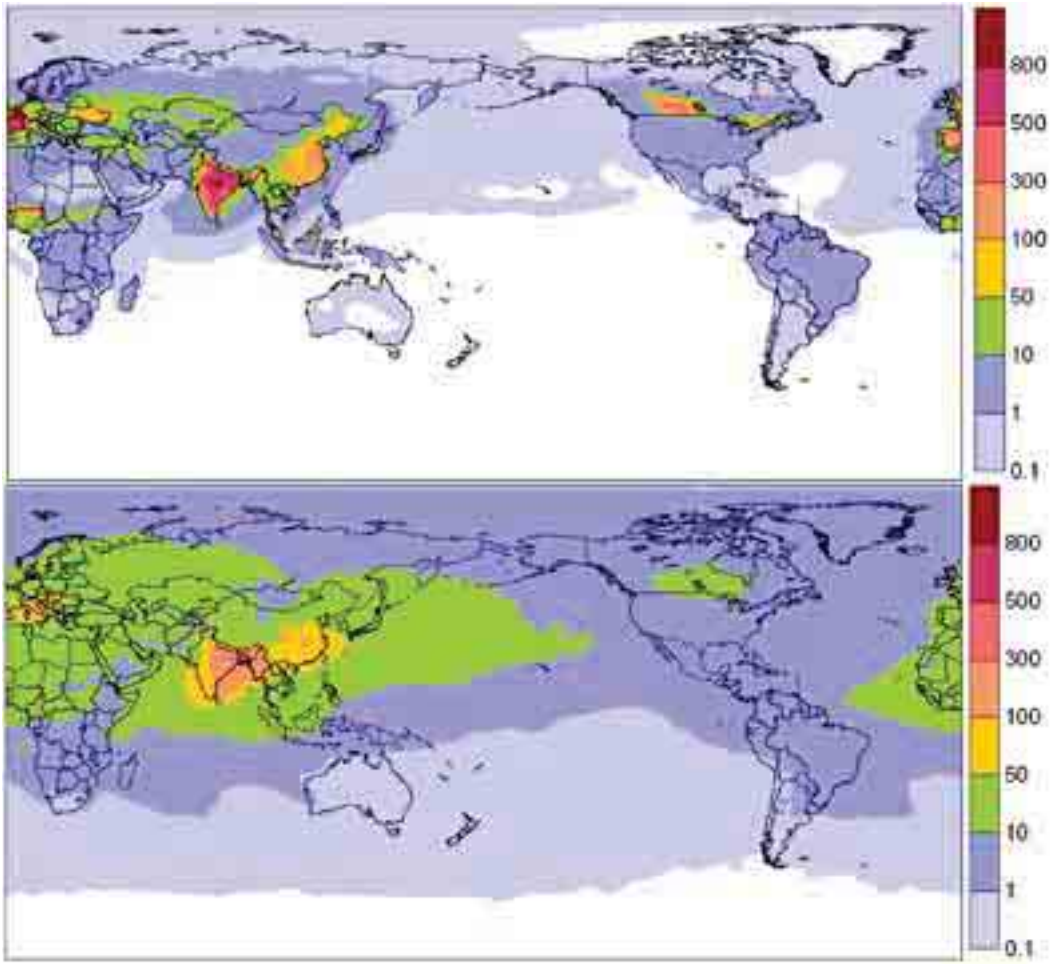


Figure 8-2. **Modeled Average Daily Air Concentrations (pg m^{-3}) of γ-HCH in 2005 at: (a) 1.5 m Height Above Ground Level, and (b) 3000 m Height Above Ground Level. Source: Environment Canada.**

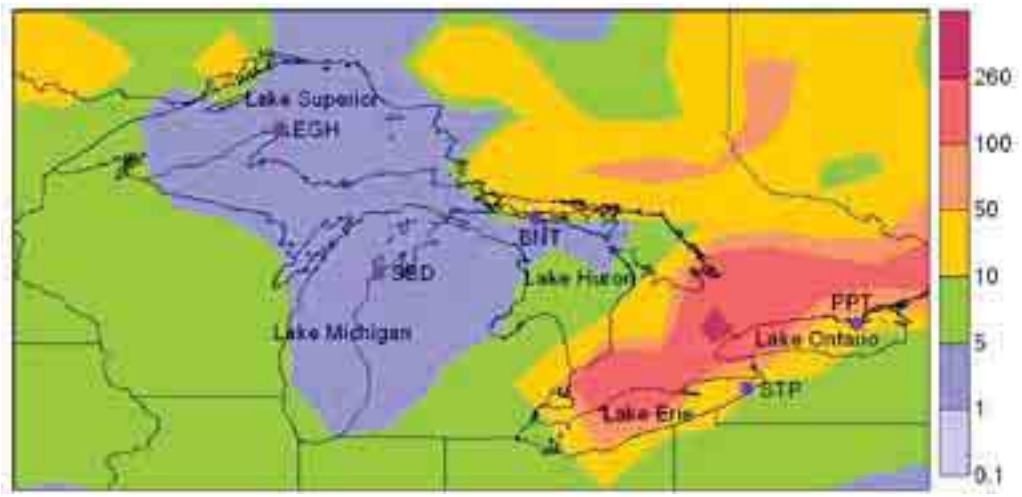


Figure 8-3. **Gridded Air Concentrations (pg/m³) of γ-HCH in the Great Lakes with 1°×1° Latitude/ Longitude Resolution. Five major monitoring sites under IADN (Integrated Atmospheric Deposition Network) are also shown. Source: Environment Canada.**

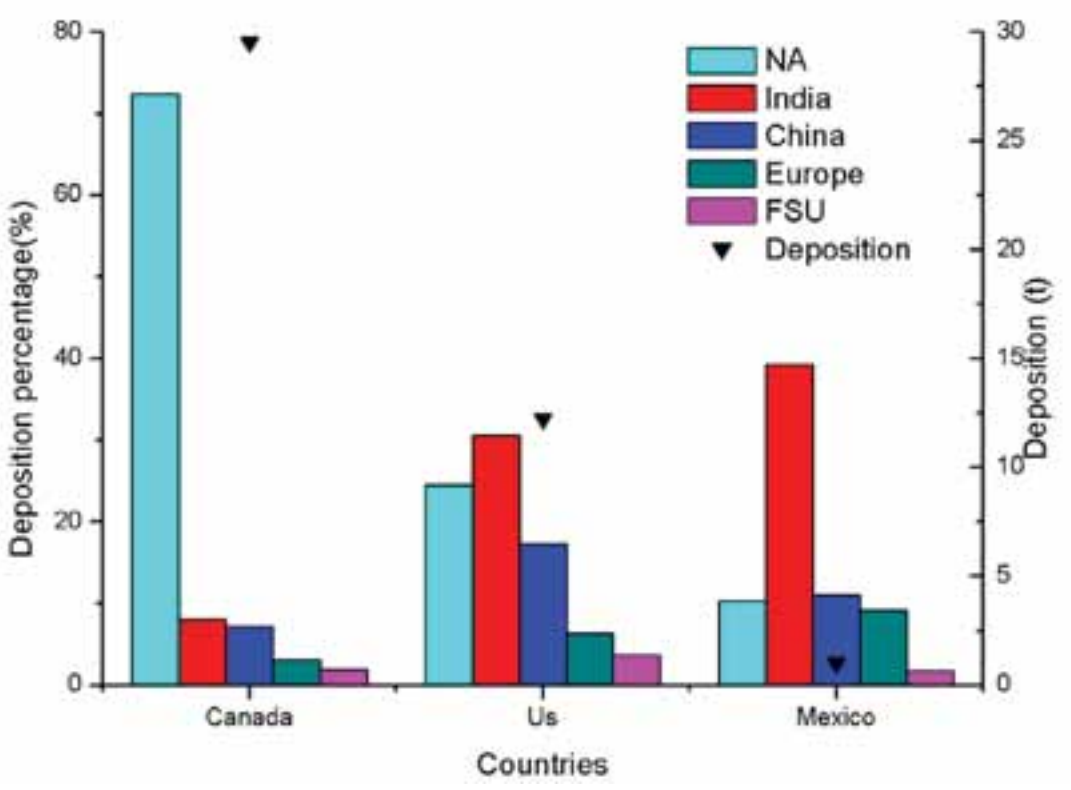


Figure 8-4. **Total Depositions of γ-HCH in Three Countries in North America (Canada, Mexico, and the USA) due to Global Sources and the Contributions of Five Major Sources (China, India, the FSU, Europe, and North America). Source: Environment Canada.**

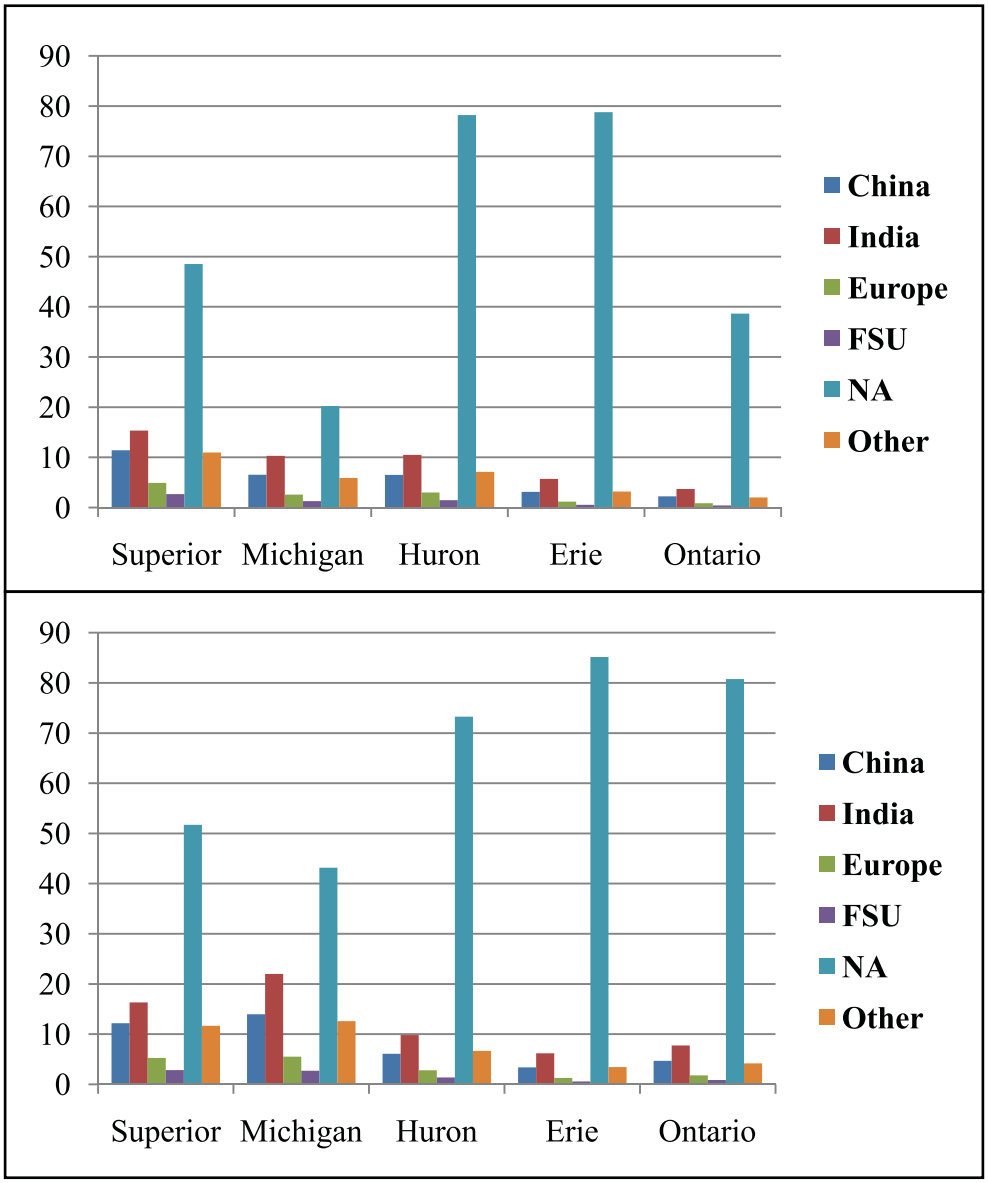


Figure 8-5. **Deposition of γ-HCH to Five Great Lakes from Different Sources. Top: in kg; bottom: in percentage. Source: Environment Canada.**

in air at the same height. The wet deposition flux was calculated from the product of the vertically integrated concentration and a scavenging ratio. A detailed description related to the computational methods can be found in the previous study (Ma et al., 2004).

Total depositions of γ-HCH in the countries in North America (Canada, the USA, and Mexico) due to global sources and contribution ratios from the five major regions are illustrated in Figure 8-4. Total depositions of γ-HCH due to global sources in 2005 were 30 t in Canada, 12 t in the USA, and 1 t in Mexico. In Canada, the percentage contribution from major global sources was 93%, of which 7% was from China, 8% from India, 3% from Europe, 2% from FSU, and 72% from North America. In the USA, total deposition of γ-HCH due to global sources was 82%, of which 17% was from China, 31% from India, 6% from Europe, 4% from FSU, and 25% from North America. In Mexico, global sources contributed 71% of total deposition, of which 11% was from China, 39% from India, 9% from Europe, 2% from FSU, and 10% from North America.

Total depositions of γ-HCH to the Great Lakes due to global sources and contribution ratios from the five major regions are illustrated in Figure 8-5. The total deposition of γ-HCH in the Great Lakes due to global sources in 2005 was 386 kg, and contributions from the five major sources were 3.2% from Europe, 68% from North America, 7.7% from China, 1.6% from FSU, and 12% from India. The remaining 7.5% was from other sources.

References

Abhilasha PC and Singh N (2008) Distribution of hexachlorocyclohexane isomers in soil samples from a small scale industrial area of Lucknow, North India, associated with lindane production *Chemosphere* 73(6):1011-1015.

Koziol A and Pudykiewicz J (2001) Global-scale environmental transport of persistent organic pollutants. *Chemosphere* 45(8):1181-1200.

Li Y, Struger J, Watie D, & Ma JM (2004) Gridded Canadian lindane usage inventories with 1/6×1/4 Latitude and Longitude resolution. *Atmospheric Environment* 38:1117-1121.

Li Y, Venkatesh S, and Li DC (2004) Modeling global emissions and residues of pesticides. *Environmental Modeling and Assessment* 9(4):237-243.

Li Y, Zhulidov A, Robarts R, and Korotova LG (2004) Hexachlorocyclohexane use in the former Soviet Union. *Archives of Environmental Contamination and Toxicology* 48:10-15.

Ma J, Daggupaty S, Harner T, Blanchard P, and Waite D (2004) Impacts of lindane usage in the Canadian Prairies on the Great Lakes ecosystem. 2. Modeled fluxes and loadings to the Great Lakes. *Environmental Science & Technology* 38(4):984-990.

Wania F, Haugen J, Lei Y, and Mackay D (1998) Temperature dependence of atmospheric concentrations of semivolatile organic compounds (SVOC). *Environmental Science & Technology* 32(8):1013-1021.

Wania F and Mackay D (1995) A global distribution model for persistent organic chemicals. *Science of the Total Environment* 160/161:211-232.

Willett K, Ulrich E, and Hites R (1998) Differential toxicity and environmental fates of hexachlorocyclohexane isomers. *Environmental Science & Technology* 32:2197-2207.

Zhang L, Ma J, Venkatesh S, Li Y-F, and Cheung P (2008) Modeling Evidence of Episodic Intercontinental Long-Range Transport of Lindane. *Environmental Science & Technology* 42(23):8791-8797.

9.0 STATE OF THE GREAT LAKES

Satellite Image of the Great Lakes, Photograph courtesy of The National Oceanic and Atmospheric Administration (NOAA)

Environmental monitoring programs maintained by government agencies and other organizations collect data with which to assess the state of the Great Lakes with respect to toxic substances.

This chapter presents monitoring data for environmental indicators in the air over the Great Lakes and in Great Lakes fish, herring gull eggs, bivalves (mussels), sediment, and surface waters. Trends in atmospheric concentrations are described by ambient air monitoring data collected by the IADN and the National Air Pollution Surveillance (NAPS) network. Levels in fish tissue are illustrated by data collected from Canada's Great Lakes Fish Contaminant Surveillance Program and US EPA's Great Lakes Fish Monitoring Program. The status of toxic substances in Great Lakes herring gull eggs is described by data collected and analyzed by the Canadian Wildlife Service. The NOAA Mussel Watch Program provides monitoring data with which to track trends of legacy substances and emerging contaminants of concern. Spatial and temporal trends in Great Lakes sediment are described by data collected from various water and sediment contaminant monitoring programs operating in the Great Lakes. The State of the Lakes Ecosystem Conference (SOLEC), hosted every 2 years by US EPA and EC, provides another opportunity for Great Lakes researchers to report the results of monitoring and surveillance efforts in the Great Lakes. For more information about SOLEC, see http://www.epa.gov/solec/ and http://binational.net/solec/pub_e.html.

Trends in Ambient Air

Photo: Lake Michigan beach, Petoskey, Michigan Michigan Travel Bureau. Courtesy of US EPA GLNPO.

Ambient Air Monitoring of Great Lakes Toxics

Submitted by Tom Dann and Liisa Jantunen, Environment Canada, and Todd Nettesheim, US EPA

This section presents data from two air monitoring networks in the Great Lakes Basin. The National Air Pollution Surveillance (NAPS) network was established in 1969 as a joint program of the federal and provincial governments to monitor and assess the quality of ambient air in Canadian towns and cities. The NAPS network collects data on ambient air levels of a variety of toxics at rural, suburban, city-centre, and industrial sites in Canada. The Integrated Atmospheric Deposition Network (IADN) is a joint United States/Canada atmospheric monitoring network that has been in operation since 1990.

National Air Pollution Surveillance (NAPS) Network

The NAPS program includes measurement of volatile organic compounds (VOCs), including toxics and ground-level ozone precursors; polar volatile organic compounds (PVOCs) such as aldehydes and ethers; components of fine particulate matter (PM), including metals and inorganic and organic ions; and persistent, toxic semi-volatile organic compounds (SVOCs)[43], such as B(a)P and polychlorinated dibenzo-p-dioxins (PCDDs) and furans (PCDFs), coplanar PCBs, HCB, pentachlorophenol (PCP) and octachlorostyrene (OCS). NAPS began sampling for PBDEs in 2009 at 10 sites across Canada, including 5 in Ontario, but data are not yet available.

Examples of trends in GLBTS Level 1 and Level 2 substances are shown in Figures 9-1 to 9-7. The box plots show median, 25th and 75th percentiles, and non-outlier minimum and maximum. Note that the vertical axes in the figures have different scales, and in some cases the plots are logarithmic rather than linear.

Ambient concentrations of dioxins, furans, and coplanar PCBs, represented as TEQ, have decreased over time (Figures 9-1 and 9-2), with the largest declines at urban sites, where concentrations were the highest. Ambient air concentrations are well below the Ontario Ambient Air Quality Criteria for dioxins/furans. Similarly, the NAPS data show B(a)P concentrations in urban areas decreasing slightly over time (Figure 9-3). B(a)P concentrations in rural areas are significantly lower than concentrations in urban areas and are near the method detection limit. HCB (Figure 9-4) and PCP (Figure 9-5) concentrations at Ontario sites appear to have slowly declined over the past decade. Concentrations of the Level 2 compounds cadmium (Figure 9-6) and lead (an indicator for alkyl lead, Figure 9-7) have decreased in the past few years at Ontario sites.

Integrated Atmospheric Deposition Network (IADN)

IADN was initiated in 1990 to measure atmospheric concentrations of persistent toxic pollutants in the Great Lakes Basin. These measurements have been conducted at five master stations, one on each of the Great Lakes, as well as 10 satellites stations (see Figure 9-8). Concentrations of PCBs, PBDEs, organochlorine pesticides, PAHs, and trace metals are measured in air (gas phase) and precipitation and on suspended particles at each station. These data are used to examine spatial and temporal trends of toxic contaminants and to calculate atmospheric loadings to the Great Lakes.

In the figures of IADN data that follow, the master stations are represented by lake names: "Lake Superior" represents data collected at Eagle Harbor, "Lake Michigan" represents data collected at Sleeping Bear Dunes, "Lake Huron" represents data collected at Burnt Island, "Lake Erie" represents data collected at Sturgeon Point, and "Lake Ontario" represents data collected at Point Petre.

PCBs. Figure 9-9 illustrates that there has generally been a decline in total PCB concentrations in Great Lakes air over the past 30 years. Figure 9-10 depicts the changes since the year 1992. Half-lives for temperature-corrected IADN data (data since 1992) for gas-phase PCBs are 7 to 27 years; the longer half-lives are for the more remote sites on Lakes Superior and Huron (Sun et al., 2007). Using data through

[43] SVOC measurements are made with a high-volume filter/Poly Urethane Foam (PUF) sampling system. The filter and PUFs are extracted together to represent a total (particle + vapor phase) measurement.

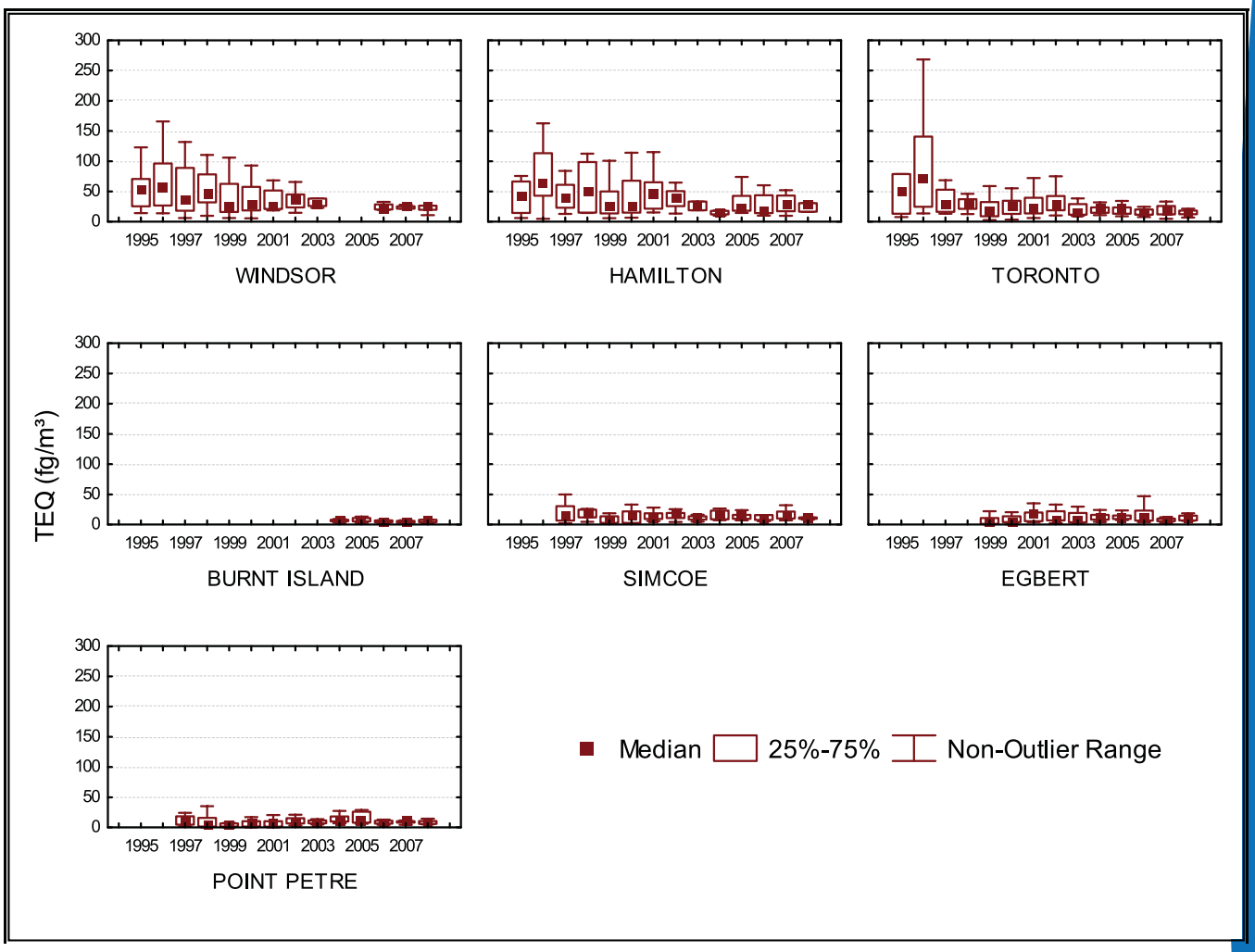


Figure 9-1. **Trends in 2,3,7,8-TCDD Toxic Equivalents (TEQ) (fg/m³) (1995-2008) at Urban (Windsor, Hamilton, Toronto) and Rural (Burnt Island, Simcoe, Egbert, Point Petre) Ontario Sites[44]**

[44] Unpublished data, Tom Dann, Environment Canada.

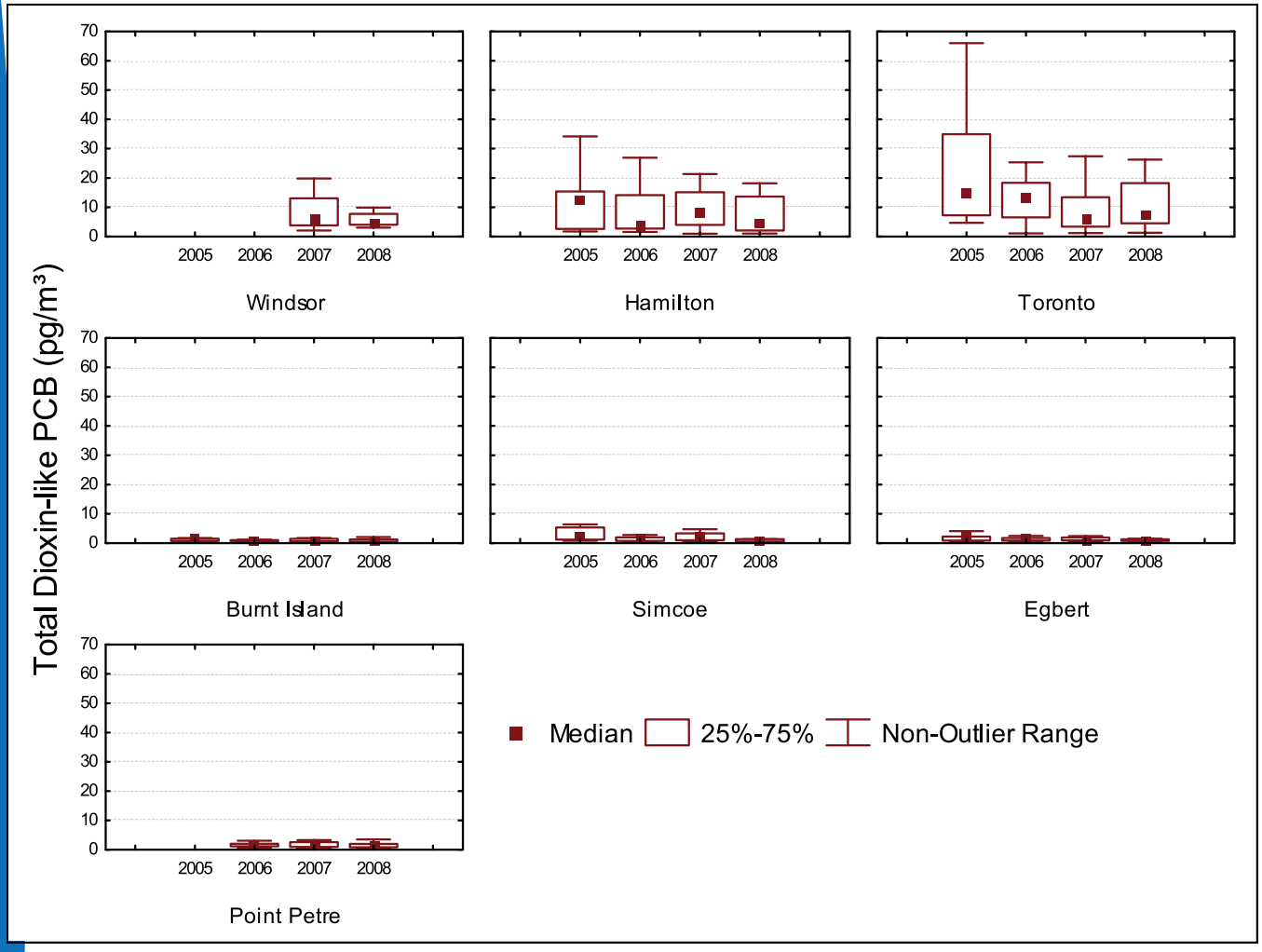


Figure 9-2. Trends in Dioxin-Like PCB Concentrations (pg/m³) (2005-2008) at Urban (Windsor, Hamilton, Toronto) and Rural (Burnt Island, Simcoe, Egbert, Point Petre) Ontario Sites[45]

[45] Unpublished data, Tom Dann, Environment Canada.

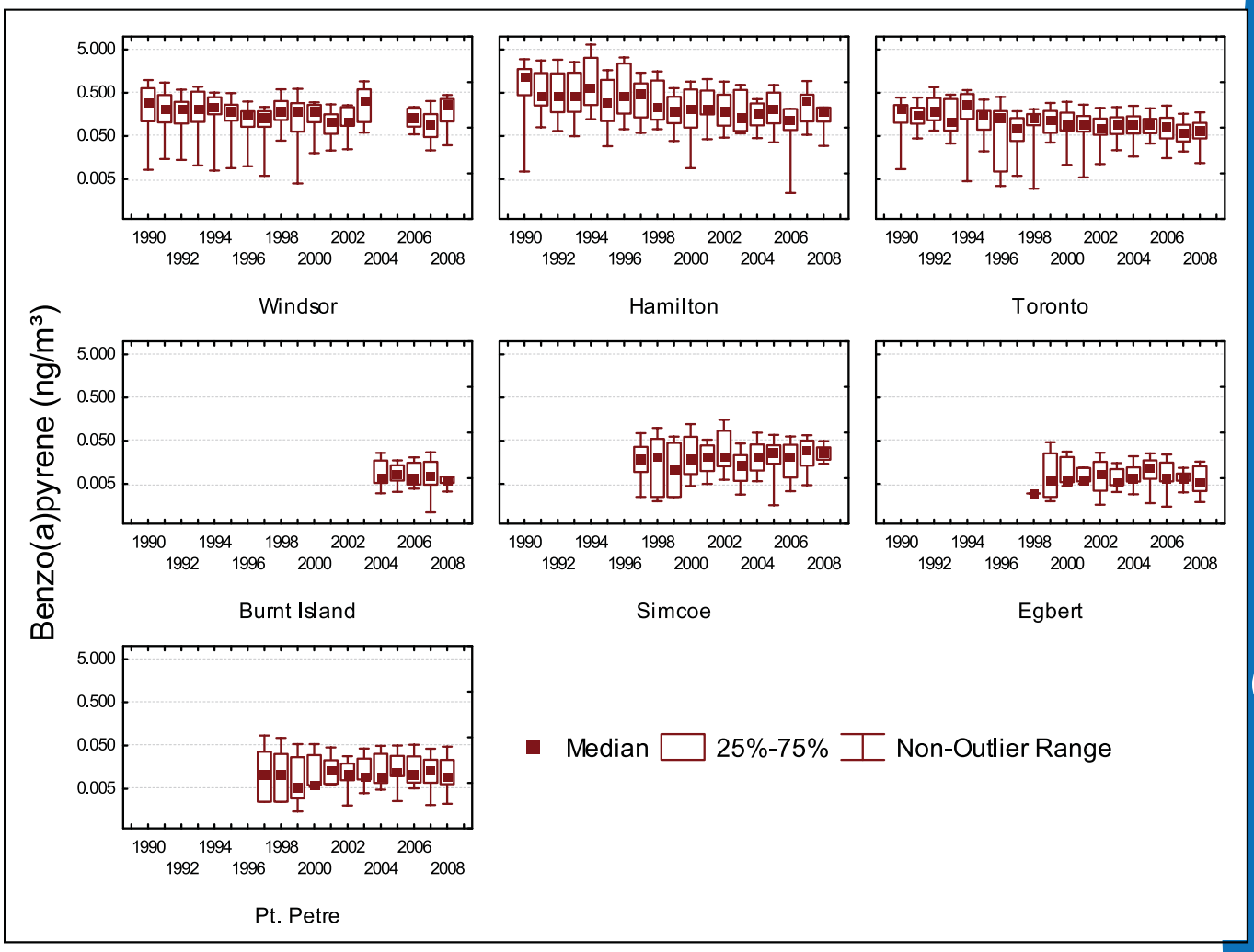


Figure 9-3. **Trends in Benzo(a)pyrene Concentrations (ng/m³) (1990-2008) at Urban (Windsor, Hamilton, Toronto) and Rural (Burnt Island, Simcoe, Egbert, Point Petre) Ontario Sites[46]**

[46] Ibid.

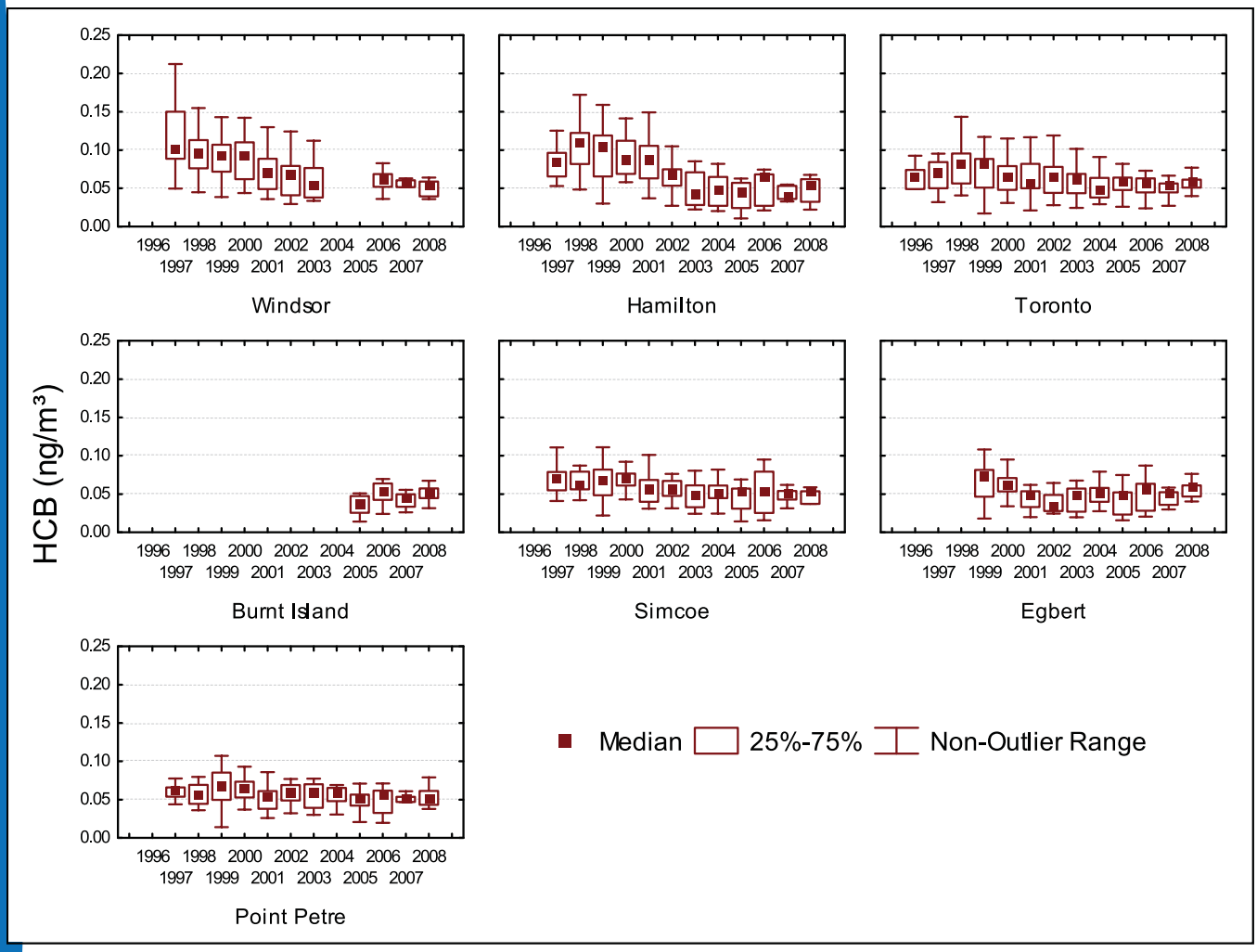


Figure 9-4. Trends in HCB Concentrations (ng/m³) at Urban (Windsor, Hamilton, Toronto) and Rural (Burnt Island, Simcoe, Egbert, Point Petre) Ontario Sites (1996-2008) [47]

[47] Ibid.

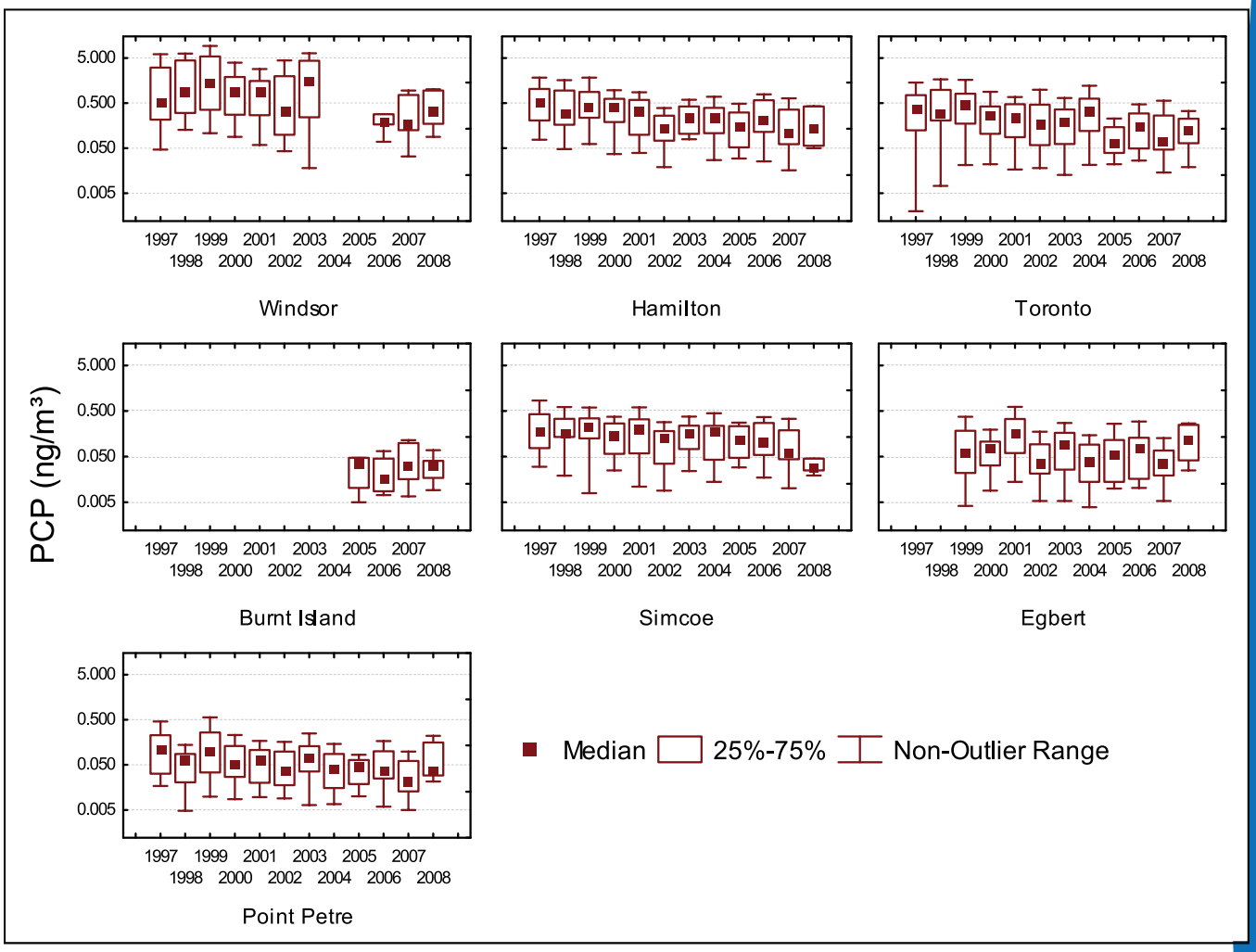


Figure 9-5. **Trends in PCP Concentrations (ng/m³) at Urban (Windsor, Hamilton, Toronto) and Rural (Burnt Island, Simcoe, Egbert, Point Petre) Ontario Sites (1997-2008)[48]**

[48] Ibid.

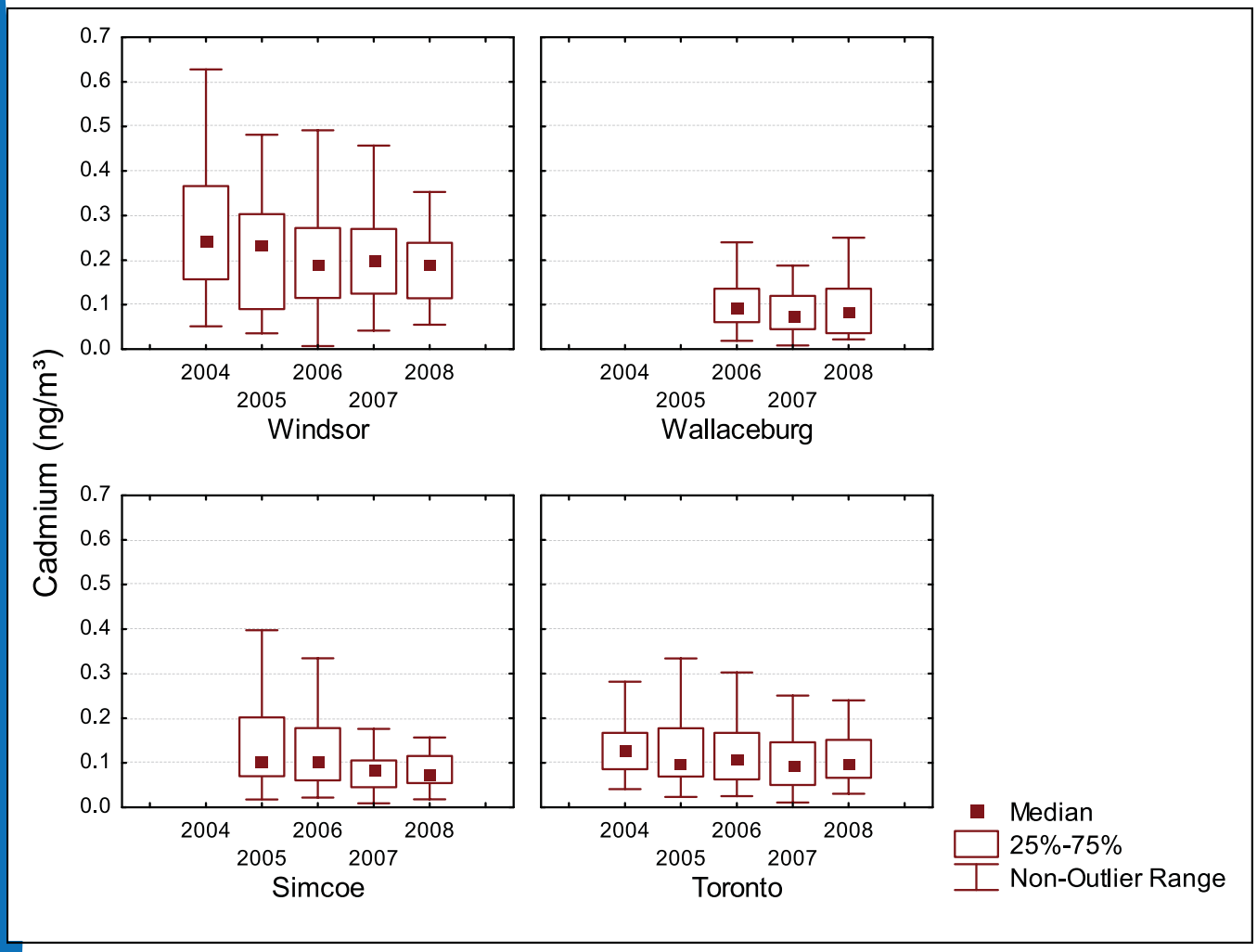


Figure 9-6. Trends in Cadmium Concentrations (ng/m³) (2004-2008) at Ontario Sites[49]

[49] Unpublished data, Tom Dann, Environment Canada.

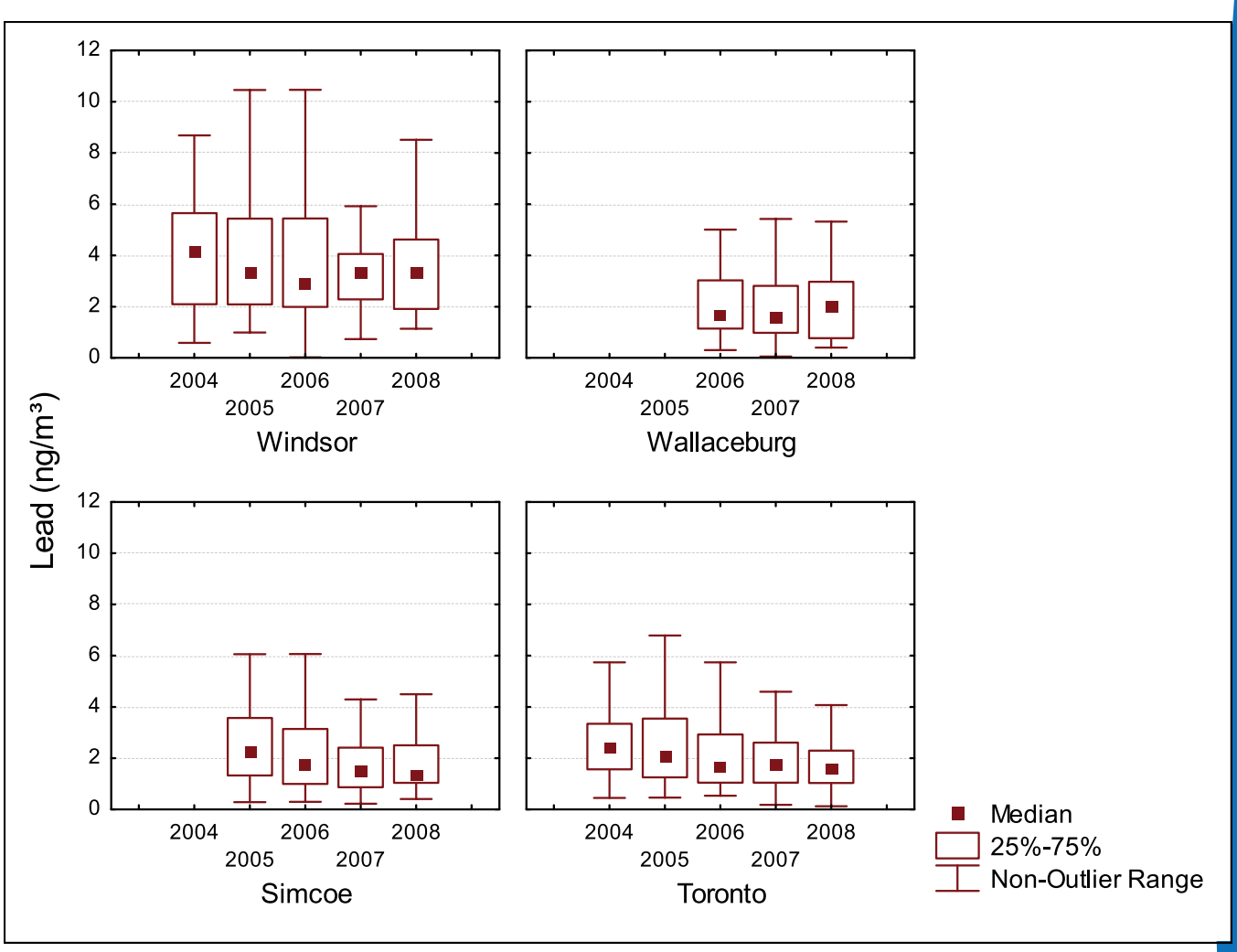


Figure 9-7. Trends in Lead Concentrations (ng/m³) (2004-2008) at Ontario Sites [50]

[50] Unpublished data, Tom Dann, Environment Canada.

Figure 9-8. Map of IADN Monitoring Stations

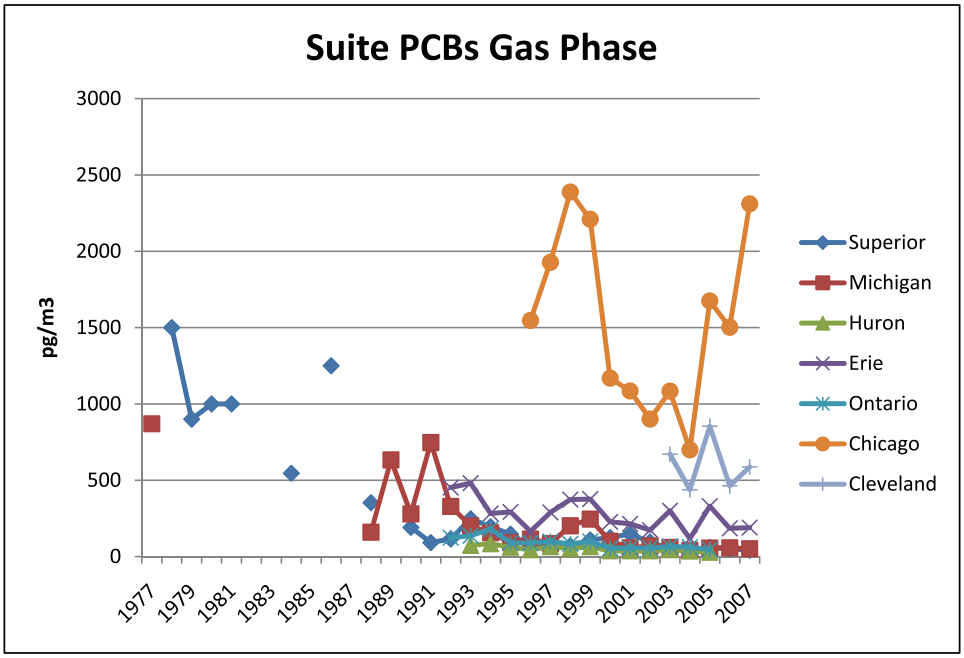


Figure 9-9. Long-term Gas-Phase Annual Average Total PCB Concentrations (pg/m³) [51]

[51] IADN Steering Committee, unpublished data, 2009. Sources for pre-1992 PCB data: Achman et al., 1993; Baker and Eisenreich, 1990; Cotham and Bidleman, 1995; Doskey and Andren, 1981; Eisenreich et al., 1981; Eisenreich, 1987; Hornbuckle et al., 1993; Hornbuckle et al., 1994; Manchester-Neesvig and Andren, 1989; Monosmith and Hermanson, 1996.

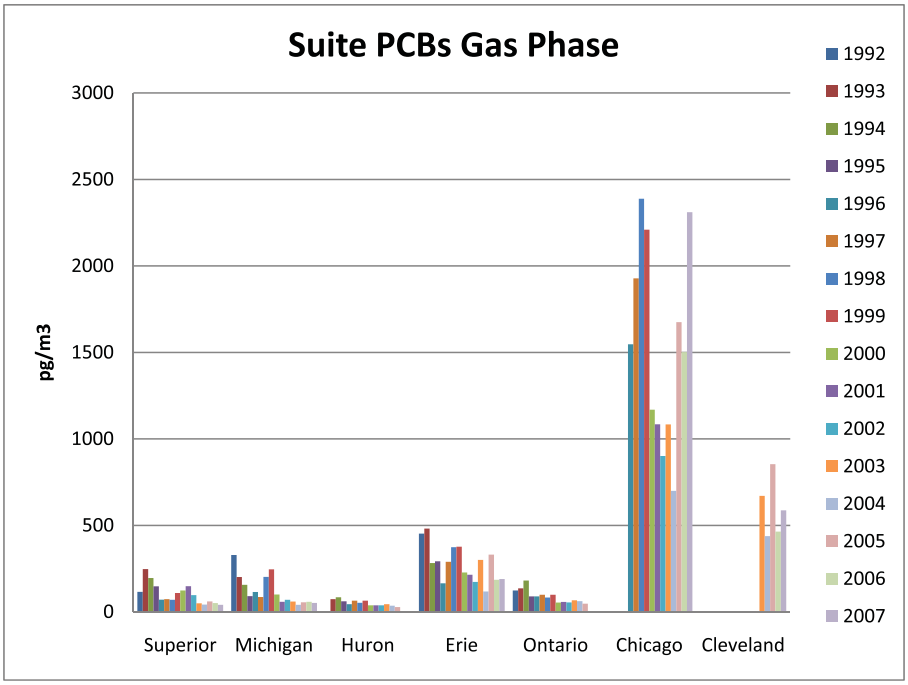


Figure 9-10. Gas-Phase Annual Average Total PCB Concentrations (pg/m³) [52]

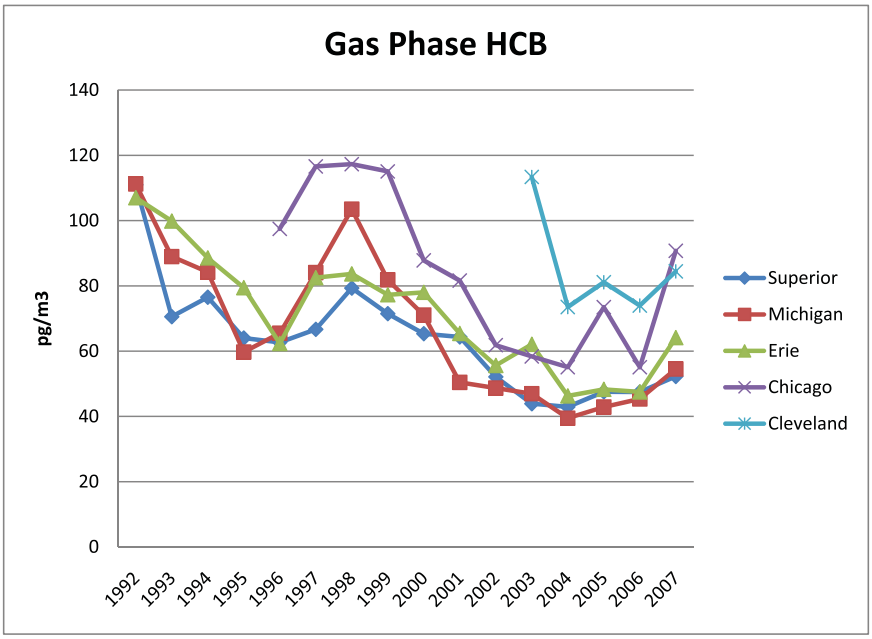


Figure 9-11. Annual Average Gas-Phase HCB Concentrations (U.S. sites only) (pg/m³)[53]

[52] IADN Steering Committee, unpublished data, 2009.

[53] IADN Steering Committee, unpublished data, 2009. HCB data not available for Canadian stations due to breakthrough on polyurethane foam (PUF) sampling media.

2007 and correcting for other factors including temperature, population, time, wind speed and wind direction, the half-life of PCBs in the Great Lakes atmosphere is estimated to be 13 +/- 0.7 years (Venier and Hites 2010).

It is assumed that PCB concentrations will continue to decrease slowly. However, as concentrations decrease, the absolute size of subsequent decreases will diminish, as shown by the fairly consistent values from the mid-1990s to the present. Further data will confirm whether concentrations continue to decline and whether remaining sources of PCBs, including legacy sources in the U.S. and long-range transport from other countries, may be contributing to the relative stability of PCB levels in the Great Lakes region.

Sturgeon Point, the Lake Erie master station, consistently shows higher PCB concentrations compared to the other master stations. Back-trajectory analyses have shown that this is due to possible influences from upstate New York (the site is 20 km southwest of Buffalo) and the East Coast (Hafner and Hites, 2003). PCB concentrations at the satellite stations in the urban areas of Chicago and Cleveland are about 10 times higher than those at the more remote master stations. It is expected that PCB concentrations should be elevated in the urban areas because of the widespread use of PCBs in industrial applications in the mid-20th century. Back-trajectory analyses have revealed that the influence of the Chicago urban area as a source of PCBs may reach as far away as Lake Superior (Hafner and Hites, 2003). Data from the Cleveland station, where monitoring began in 2003, indicate that PCB levels in that city are lower than those in Chicago, but higher than at the master stations. A multiple linear regression model for IADN air concentrations demonstrates that local human population is the most important factor in explaining the variability of PCB concentrations, accounting for about 55% of the total variability. Seasonality also plays a significant role in explaining PCB concentrations, accounting for about 23% of the total variability (Venier and Hites 2010).

HCB. IADN data for HCB from the three U.S. master stations on Lakes Superior, Michigan, and Erie show decreasing trends with half-lives of 12 to 18 years (Sun et al., 2006e). However, like PCBs, HCB concentrations increased somewhat during the late 1990s (see Figure 9-11), perhaps due to atmospheric circulation phenomena such as the North Atlantic Oscillation and El Niño-Southern Oscillation (Ma et al., 2004). A similar increase has been observed in recent years. The longer half-lives may be due to continued releases of HCB into the environment as a byproduct of manufacturing processes and contaminant pesticides. HCB also has an atmospheric lifetime of about 2 ½ half years (Brubaker and Hites, 1998), making it capable of global transport and therefore making the Great Lakes susceptible to inputs from global emissions.

Organochlorine Pesticides. In general, gas-phase concentrations of banned or restricted pesticides measured by the IADN are decreasing over time in the air, with half-lives generally between 4 and 9 years (Sun et al., 2006e). The half-life of α-HCH of 3.3 +/- 0.04 years recently reported with data through 2007 using a multiple linear regression model (Venier and Hites 2010) is similar to the half-life of 3.8 years reported with data through 2003 (Sun et al., 2006e). It is worthwhile to note that α-HCH continues to be eliminated rapidly from the environment after its ban about 40 years ago.

Figures 9-12 and 9-13 present data for α-HCH and total p,p'-DDT. These declining trends correlate well with declining global use of these pesticides. Some pesticides, including chlordane and DDT, are found at higher levels in urban areas. This is demonstrated for DDT in Figure 9-13, but other compounds such as HCHs do not show an urban source (Figure 9-12 and 9-14). A multiple linear regression model for IADN air concentrations demonstrates that local human population is a significant factor in explaining the variability of concentrations of total-DDT and total-chlordane, accounting for about 22% and 28% of the total variability. Chlordane was used as a termiticide in buildings, and DDT was sprayed in urban areas in the U.S. to control mosquitoes (and is still used in some other countries for malaria control). However, seasonality plays the most important role in explaining the concentrations of total-DDT and total-chlordane, accounting for about 43% and 47%

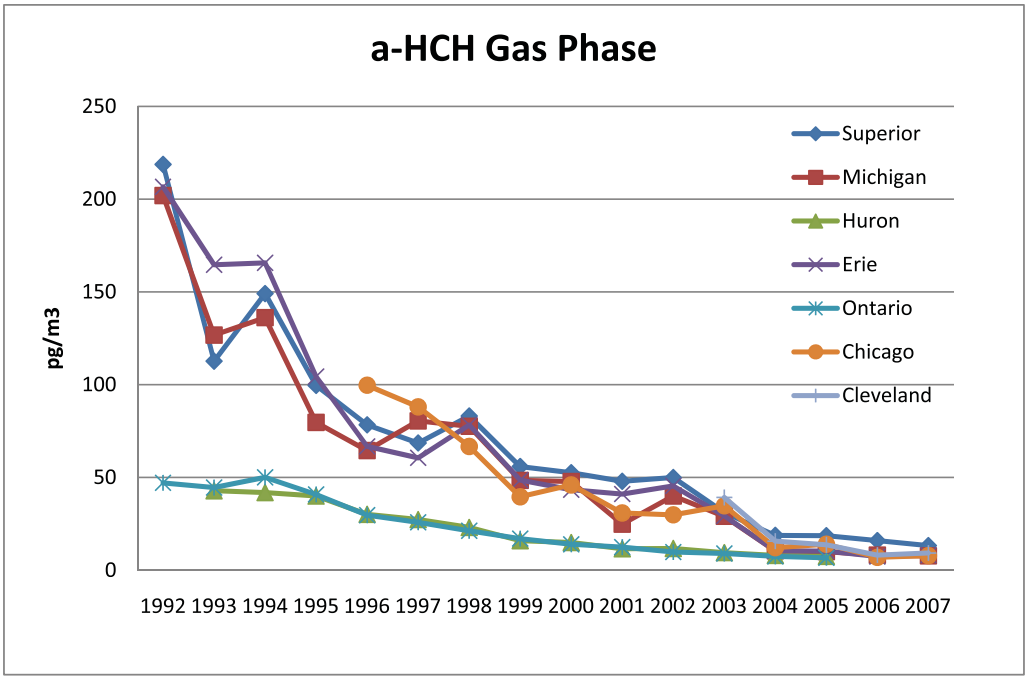


Figure 9-12. Annual Average Gas-phase Concentrations of α-HCH (pg/m³)[54]

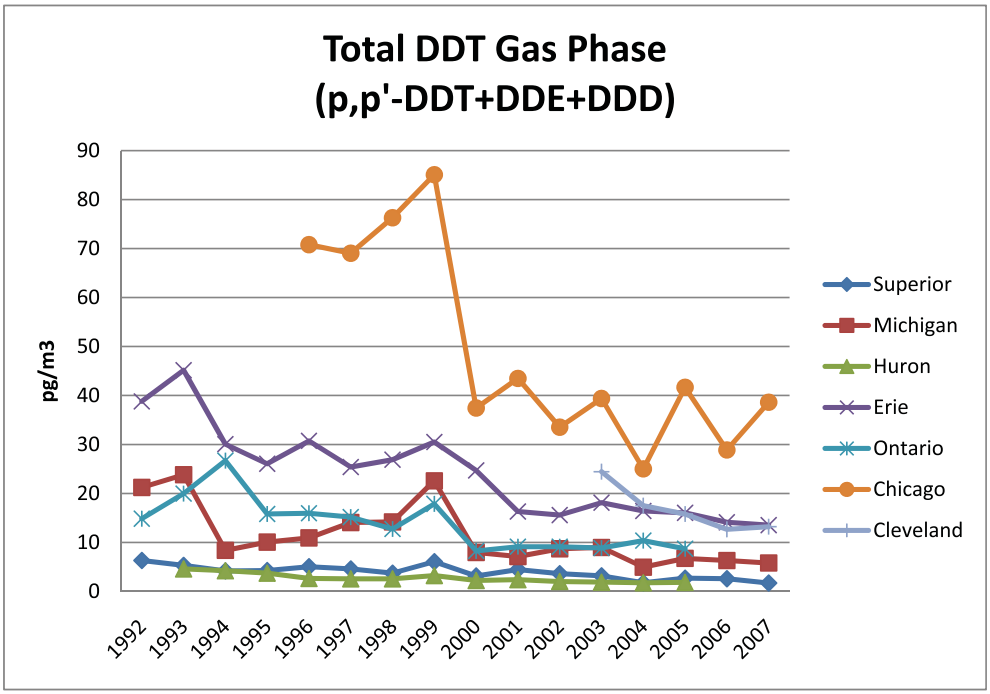


Figure 9-13. Annual Average Gas-phase Total DDT (p,p'-DDT+DDE+DDD) Concentrations (pg/m³)[55]

[54] IADN Steering Committee, unpublished data, 2009.
[55] Ibid.

of the total variability (Venier and Hites 2010). This suggests that soil reservoirs continue to release these compounds in the atmosphere through temperature-dependent exchanges.

Vapor-phase concentrations of lindane (γ-HCH) at Chicago, Sleeping Bear Dunes, and Sturgeon Point were similar but significantly higher than concentrations at Eagle Harbor, Point Petre, and Burnt Island (Sun et al., 2006e). Lindane was used in Canada through 2004. On January 1, 2005, Canada withdrew registration of lindane for agricultural pest control. U.S. registrants agreed to a voluntary cancellation of the registrations for lindane in December 2006, which was ratified by the U.S. Office of Pesticide Programs in January 2007. Use of existing stocks for seed protection in the U.S. was allowed to continue until October 2009. Levels of lindane at IADN sites have decreased in recent years (Figure 9-14), and it is expected that this trend will continue as use ends in North America. The half-life of lindane reported with data through 2007 using the multiple linear regression model (3.8 +/- 0.08 years) is considerably lower than the half-life estimated with data through 2003 (6.1 +/- 2.1 years). The more rapid rate of decline may be the result of Canada's ban on lindane's use in 2004. An even more rapid rate of decline might be expected in future years as the programs implemented in the U.S. take full effect. Lindane concentrations have generally peaked in the summer in concordance with agricultural usage.

Endosulfan. Endosulfan concentrations show significant decreases at some sites in some phases, but no decrease in the vapor phase at Eagle Harbor, Sleeping Bear Dunes, or Sturgeon Point (Sun et al., 2006e). However, a multiple linear regression model using data through 2007 estimates that the half-life of total endosulfans (α-endosulfan, β-endosulfan, and endosulfan sulfate) increased from 5.9 +/- 3.6 years (Sun et al., 2006e) to 12 +/- 1.4 years (Venier and Hites 2010). Higher endosulfan concentrations were observed at Point Petre, Sturgeon Point, and Sleeping Bear Dunes in all phases, which could be explained by agricultural usage in surrounding areas (Hoh and Hites, 2004). Similar to lindane, concentrations of endosulfan are also generally higher in the summer following use. Seasonality (i.e., temperature) explains about 68% of the variability in total endosulfan concentrations based on the multiple linear regression model.

PAHs. In general, concentrations of PAHs can be roughly correlated with population, with the highest levels observed in Chicago and Cleveland and lower concentrations at the remote master stations (Sun et al., 2006a,d). In general, PAH concentrations in Chicago and Cleveland are about 10 to 100 times higher than at the master stations. A multiple linear regression model for IADN air concentrations demonstrates that local human population is the most important factor in explaining the variability of PAH concentrations, accounting for about 74% of the total variability. Other factors including temperature, time, wind speed, and wind direction only accounted for 7% of the variability in explaining concentrations of PAHs (Venier and Hites 2010).

Concentrations of PAHs in the particle and gas phase are decreasing at Chicago, with half-lives of 3-10 years in the gas phase and 5-15 years in the particle phase. At other sites, most gas-phase PAH concentrations showed significant, but slow, long-term decreasing trends (half-lives >15 years). Using data through 2007 and correcting for other factors in the multiple linear regression model, the half-life of PAHs in the Great Lakes atmosphere is estimated to be 14 +/- 3.6 years (Venier and Hites 2010). For most PAHs, decreases in PAHs measured on particles and in precipitation were only found at Chicago (Sun et al., 2006d; Sun et al., 2006a).

Figure 9-15 shows the annual average particle-phase concentrations of B(a)P as an example of PAH concentrations.

Octachlorostyrene. Gas-phase octachlorostyrene (OCS) data, available for the U.S. stations only, is shown in Figure 9-16. OCS concentrations are low, in the single pg/m^3 range, and appear to be decreasing. Initial data from Cleveland indicate that concentrations of OCS are higher there than at the remaining stations, including Chicago, suggesting nearby sources in that metropolitan area.

Dioxins/Furans. From 2004 to 2007, dioxins and furans were measured at four U.S. IADN sites. The average concentrations of dioxins and furans are

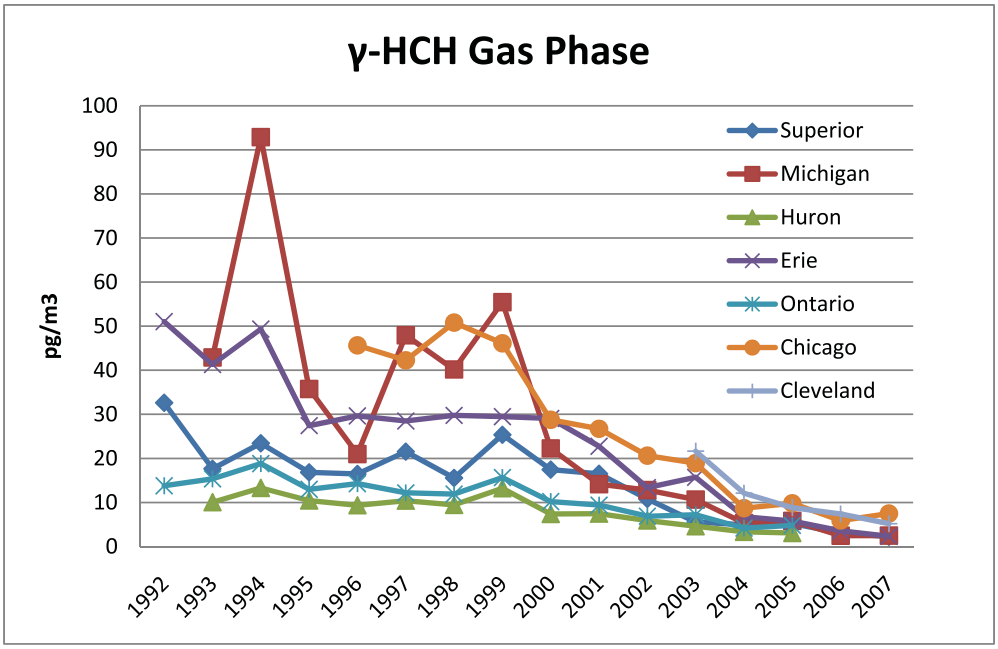


Figure 9-14. Annual Average Gas-Phase Concentrations (pg/m³) of Lindane (γ-HCH) [56]

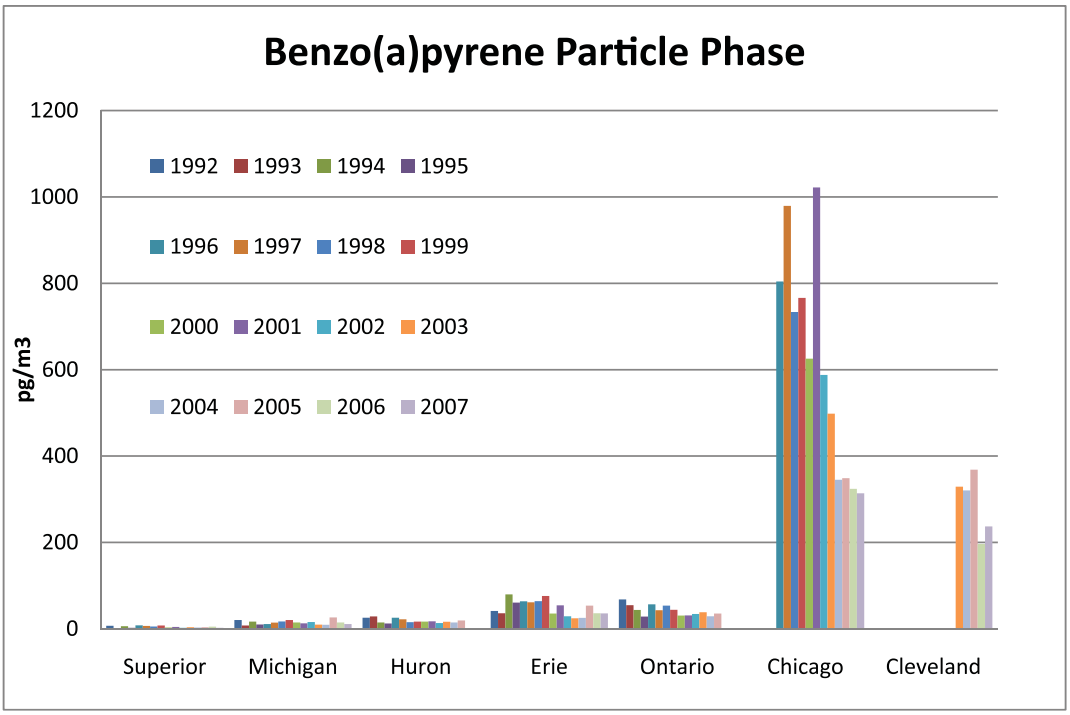


Figure 9-15. Annual Average Particle-phase B(a)P Concentrations (pg/m³) [57]

[56] Ibid.
[57] Ibid. [Note: B(a)P data for 2007 are preliminary.]

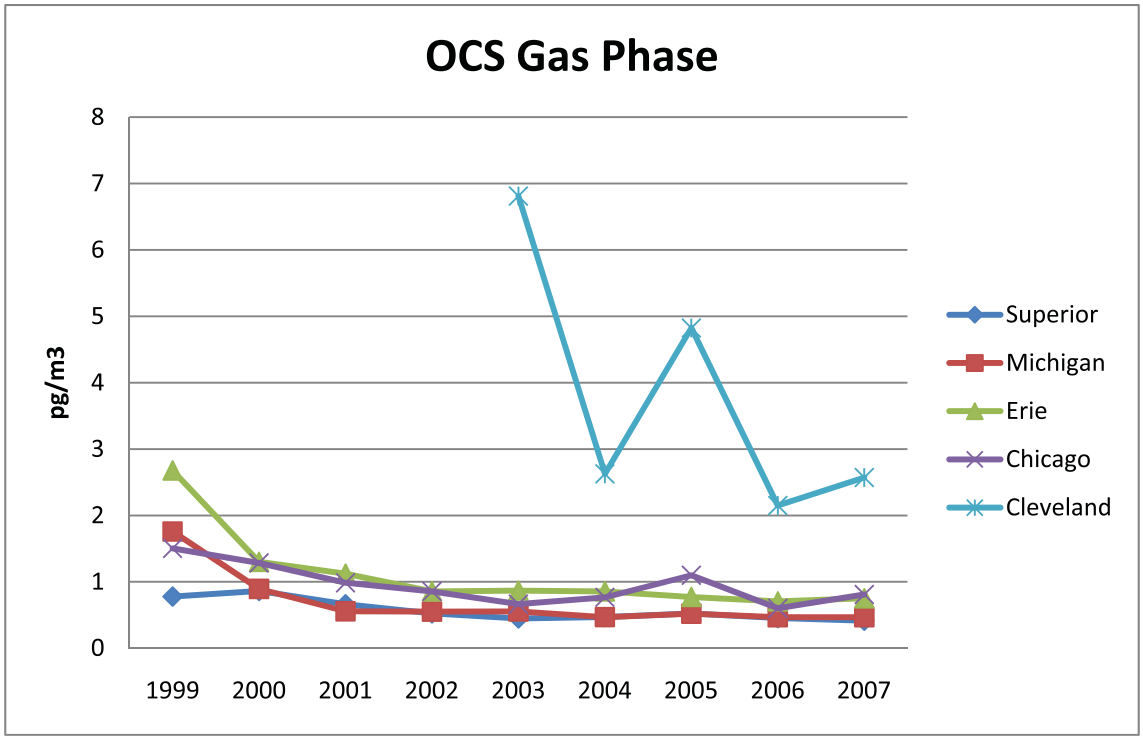


Figure 9-16. Annual Average Gas-phase OCS Concentrations (pg/m^3) [58]

displayed in Figure 9-17 as total PCDD/F and total TEQ_{DF}. While there was no significant temporal trend, concentrations of dioxins and furans showed a significant seasonal trend at all sites, except Chicago. The date of maximum concentration averaged December 6, which is consistent with residential heating being an important source of PCDD/Fs to the atmosphere (Venier et al., 2009). Using data from IADN and other monitoring networks in North America, concentrations of dioxins and furans were shown to be significantly related to local population, as evidenced by Figure 9-18.

PBDEs. PBDEs are a group of brominated flame retardant chemicals used in a variety of commercial products, including furniture and electronics, and the penta- and octa-formulations have been banned by the Stockholm Convention due to their toxicity, persistence, bioaccumulation and potential for long range transport. PBDEs have been found in the Great Lakes environment, including in air at the IADN stations. The highest mean concentrations of total PBDEs have been found at the urban sites in Chicago and Cleveland, while the lowest concentrations were found at the remote site in Eagle Harbor, as shown in Figure 9-19. This figure also illustrates that BDE-47, BDE-99, BDE-100, and BDE-209 comprise about 70-80% of the total PBDEs, with BDE-47 being the most abundant congener. The different congener distribution pattern seen in Cleveland is probably the result of a few samples containing very high concentrations of BDE-209 (Venier and Hites, 2008).

Figure 9-20 shows the concentration of total PBDEs and BDE-209 with temperature, indicating no significant seasonality. Insignificant temperature dependence was found of the lighter congeners (BDE-47 and 99), suggesting the importance of advective inputs.

PBDEs were measured separately in gas and particle-bound phases at Point Petre (Figure 9-21). BDE-209 was found only on particles, where BDE-47 and 100 were in the gas and particle phases. In the

[58] Ibid.

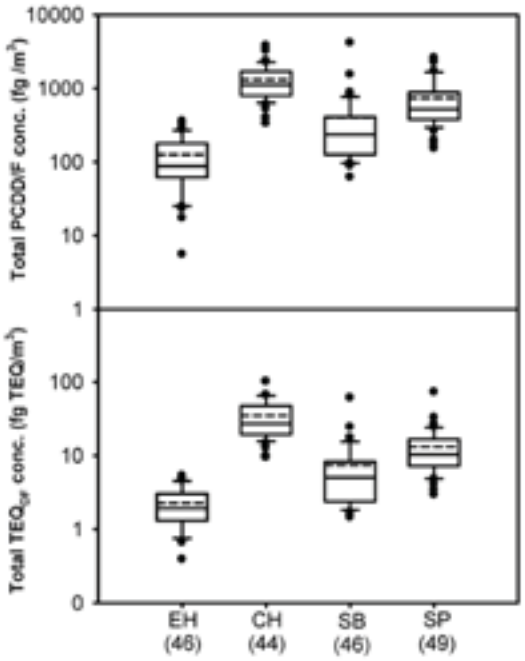


Figure 9-17. **Total PCDD/PCDF Concentrations (fg/m³) and Total TEQ_{DF} Concentrations (in fg TEQ per m³) at Four Great Lakes Sites Ordered from West to East. The horizontal lines represent the median, and the dotted lines represent the mean. The boxes represent the 25th and 75th percentiles, and the whiskers represent the 5th and 95th percentiles. The numbers in parentheses represent the number of samples reported at each site.[59]**

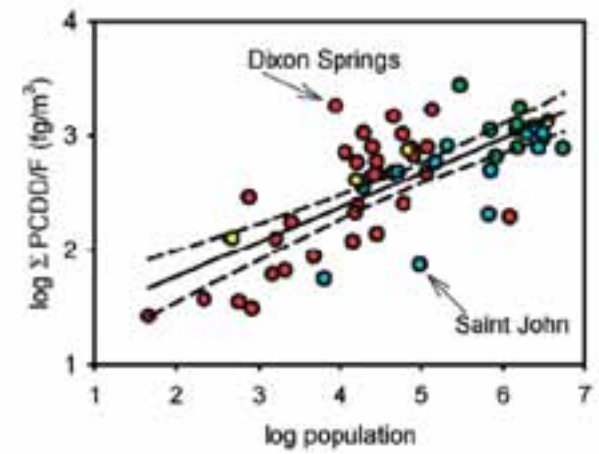


Figure 9-18. **Atmospheric Total PCDD/F Concentrations (fg/m³) as a Function of Human Population within a 25-km Radius of the Sampling Site in North America (n = 60). The black line represents a linear regression of the data. The symbols are color-coded as follows: red) NDAMN sites, cyan) NAPS sites, green) CADAMP sites, yellow) sites reported in Venier et al., 2009. The black dotted lines represent the 95% confidence limits.[60]**

[59] Venier et al., 2009..

[60] Venier et al., 2009. The analysis was restricted to large sampling networks in the North American region: US EPA National Dioxin Monitoring Network (NDAMN), which deployed samplers mainly in rural and remote locations around the U.S.; the California Ambient Dioxin Air Monitoring Program (CADAMP), which collected samples predominantly in heavily populated areas of California; and the Canadian NAPS, which sampled air throughout Canada. For consistency among data sets, only data collected in 2002 were used, with the exception of data from Venier et al., 2009. Two outliers are highlighted in the figure: Saint John, New Brunswick, Canada, and Dixon Springs, Illinois.

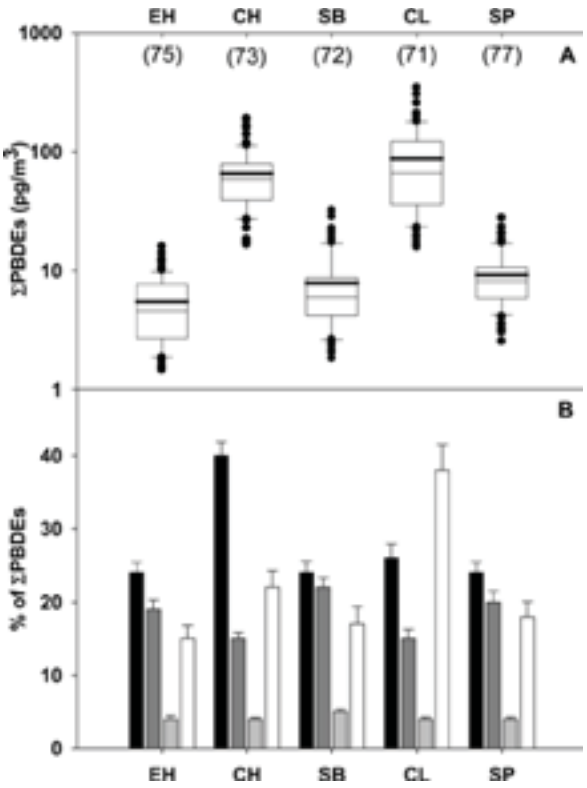


Figure 9-19. **(A) Total PBDEs Concentrations (pg/m3) at the Five IADN Sites, Ordered from West to East. The thin black horizontal line represents the median and the thick one the mean. The box represents the 25th and 75th percentiles, and the whiskers represent the 5th and the 95th percentiles. The numbers in parentheses are the number of detects. (B) Percentage of Total PBDEs of BDE-47 (black), BDE-99 (dark grey), BDE-100 (light grey), and BDE-209 (white). Standard errors are included.** [61]

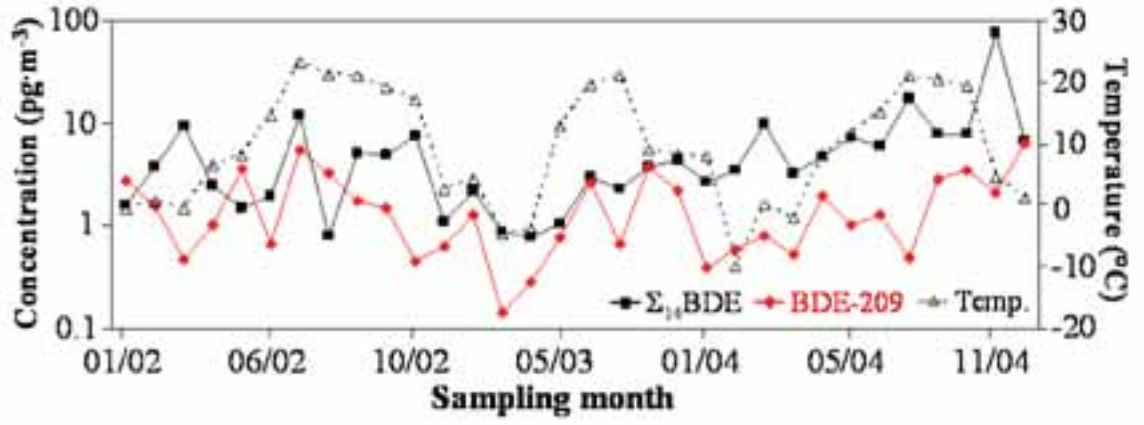


Figure 9-20. Concentration of ∑PBDEs and BDE-209 with Temperature at Point Petre, 2002-2004.[62]

[61] Venier and Hites, 2008.
[62] Su et al., 2009.

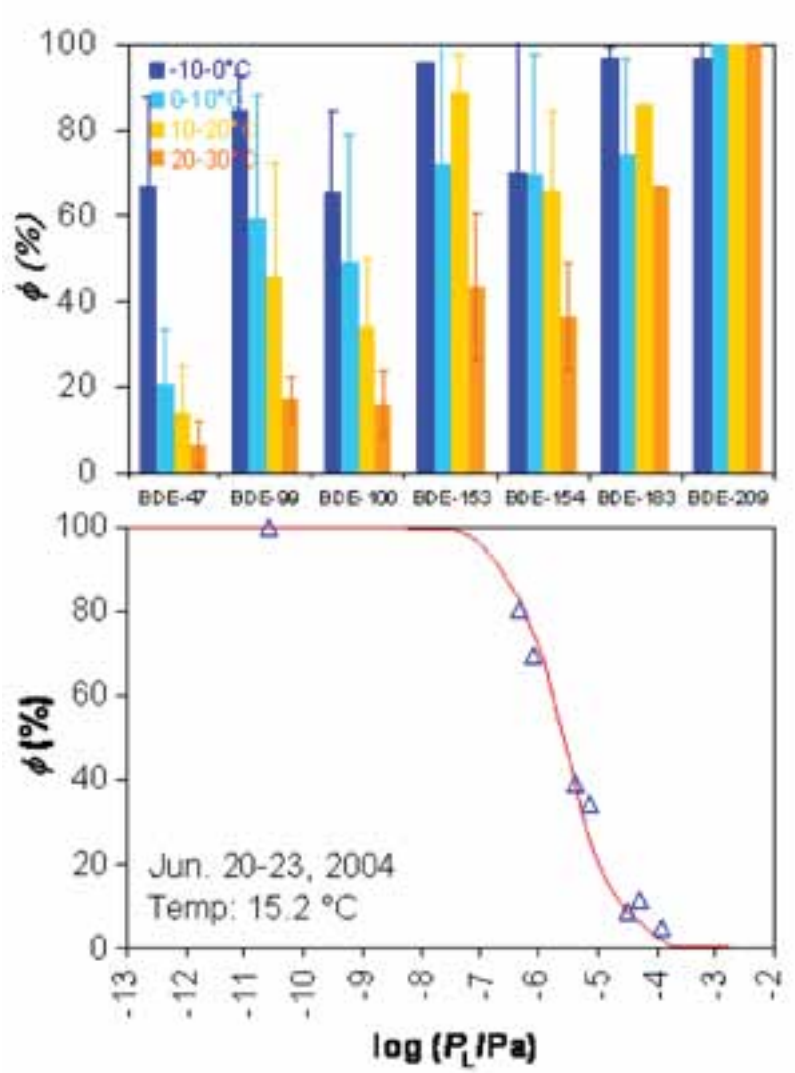


Figure 9-21. Gas-particle Partitioning at Point Petre. Top) Particle-bound percentage of seven PBDEs in four different temperature ranges. Bottom) Example of the gas-particle partition, fitted to the Junge-Pnakow model. [63]

same temperature ranges (the same color bars in Figure 9-21), heavier congeners were more particle-bound than lighter ones. For the less brominated congers, the particle-bound percentage increases with decreasing temperature. This is consistent with laboratory studies, and the gas-particle partitioning of PBDEs fits well to the Junge-Pankow model (Su et al., 2009).

Using a little over 3 years of data (2003-2006), temporal trends were assessed for total (gas + particle phase) PBDEs, BDE-47, BDE-99 and BDE-209 in Figure 9-22. For total PBDEs, statistically significant decreasing trends were found at the urban sites of Chicago and Cleveland, corresponding to half-lives of 4 +/- 1.5 and 3.4 +/- 1.6 years, respectively. For BDE-47 and BDE-99, the atmospheric concentrations are decreasing rapidly (half-lives of approximately 2 years) at all sites except for Chicago. This rapidly decreasing trend seems to indicate that the voluntary phase-out of the penta- and octa-BDE formulations by the sole U.S. manufacturer in 2004 has had immediate environmental benefits. Additional

[63] Ibid.

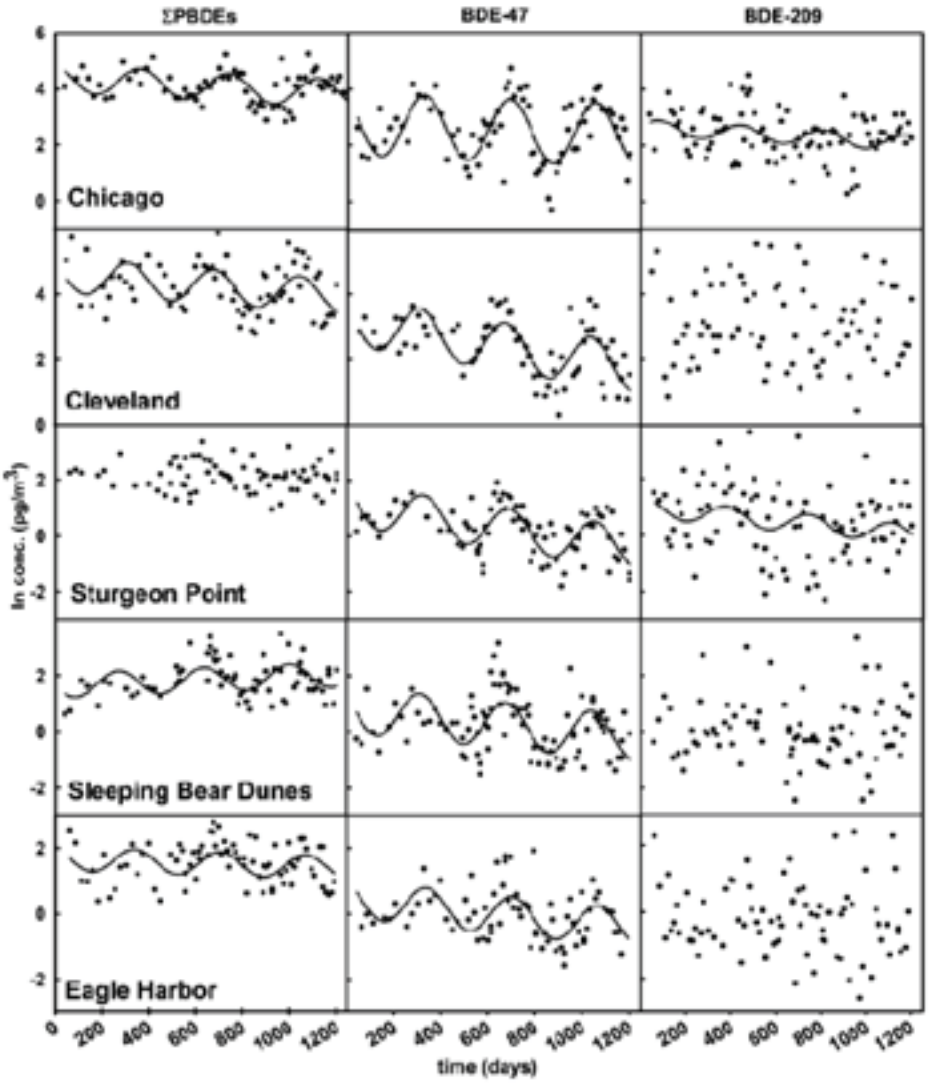


Figure 9-22. Temporal Trends of Total PBDEs, BDE-47, and BDE-209 (gas plus particle phase concentrations) in pg/m^3 at Five IADN Sites. The curves were fitted as described in Venier and Hites, 2008; if no curve is given, the regression was not significant. [64]

[64] Ibid.

Figure 9-23. Mercury Deposition Network Sites [65]

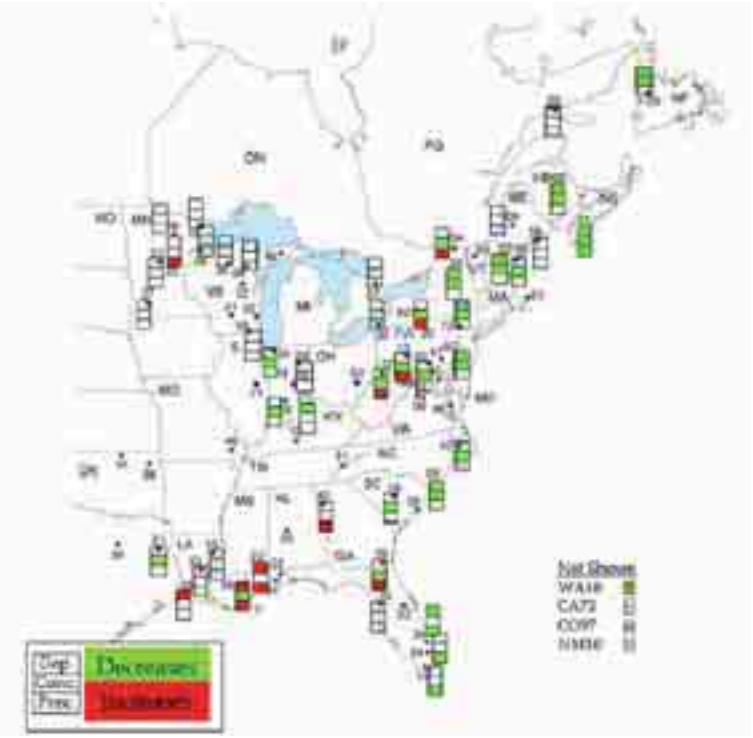


Figure 9-24. Trends in Wet Deposition of Total Mercury, Concentration in Precipitation, and Amount of Precipitation from the Mercury Deposition Network, 1996-2005[64]

[65] National Atmospheric Deposition Program, Mercury Deposition Network. 2009. http://nadp.sws.uiuc.edu/mdn/.
[66] Prestbo and Gay 2009.

data will help determine whether these trends will continue or level off. Concentrations of BDE-209 are not decreasing at any of the five U.S. sites (Venier and Hites 2008). This was expected given that the deca-BDE formulation was still being manufactured. On December 17, 2009, as the result of negotiations with US EPA, the two U.S. producers of decabromodiphenyl ether (deca-BDE), Albemarle Corporation and Chemtura Corporation, and the largest U.S. importer, ICL Industrial Products, Inc., announced commitments to voluntarily phase-out deca-BDE in the U.S.

Other Brominated Flame Retardants. 1,2-bis(2,4,6-tribromophenoxy)ethane (TBE) was heavily produced in the U.S. from 1986 to 1994, but this compound may have been recently reintroduced to the market as a substitute for octa-BDE. Decabromodiphenyl ethane (DBDPE) was introduced to the market as an alternative for BDE-209. Dechlorane plus (DP) was introduced to the market as a replacement for dechlorane (aka mirex) when its use was restricted in the 1970s. TBE, DBDPE and DP were detected at all sites, but there were insufficient data to determine temporal trends for these compounds (Venier and Hites 2008).

More information about IADN, including a report published by EC and US EPA on the atmospheric loadings of monitored PBTs to the Great Lakes using data through 2005, is available online at http://www.epa.gov/glnpo/monitoring/air2/iadn/resources.html.

Mercury Deposition Network (MDN)

Another important North American monitoring network is the Mercury Deposition Network (MDN), which is part of the National Atmospheric Deposition Program (NADP). This program began monitoring pH and major inorganic ions related to "acid rain" in the United States in 1978. In 1995, NADP began an experimental monitoring program for wet deposition of mercury, the MDN. This program has grown into an international network with sites in the U.S. and Canada (Figure 9-23). MDN collects weekly precipitation samples and analyzes them for total mercury and, at the option of the sponsoring agency, for methylmercury. MDN data show that concentrations of total mercury in precipitation are decreasing for much of the U.S., but there is no trend for the stations in the upper Midwest (Figure 9-24) (Prestbo and Gay, 2009).

References

Brubaker, Jr., W.W. and Hites, R.A. 1998. OH reaction kinetics of gas-phase a- and g-hexachlorocyclohexane and hexachlorobenzene. *Environmental Science and Technology* 32 (6):766-769.

Hafner, W. and Hites, R. 2003. Potential Sources of Pesticides, PCBs, and PAHs to the Atmosphere of the Great Lakes. *Environmental Science and Technology* 37(17):3764-3773.

Hoh, E. and Hites, R. A. 2004. Sources of toxaphene and other organochlorine pesticides in North America as determined by air measurements and potential source contribution function analyses. *Environmental Science and Technology* 38(15): 4187-4194.

Ma, J., Hung, H., Blanchard, P. 2004. How Do Climate Fluctuations Affect Persistent Organic Pollutant Distribution in North America? Evidence from a Decade of Air Monitoring. *Environmental Science and Technology* 38(9):2538–2543.

Prestbo, E.M. and Gay, D.A. 2009. Wet Deposition of Mercury in the U.S. and Canada, 1996-2005: Results and analysis of the NADP Mercury Deposition Network (MDN). *Atmospheric Environment* 43: 4223-4233.

Su, Y., Hung, H., Brice, K.A., Su, K., Alexandrou, N., Blanchard, P., Chan, E., Fellin, P., 2009. Air concentration of polybrominated diphenyl ethers (PBDEs) in 2002-2004 at a rural site in the Great Lakes: *Comparison to measurements in the Arctic, Atmosp. Pollut.*43, 6230-6237.

Sun, P., Backus, S., Blanchard, P., Hites, R.A. 2006a. Annual Variation of Polycyclic Aromatic Hydrocarbon Concentrations in Precipitation Collected near the Great Lakes. *Environmental Science and Technology*, 40(3): 696-701.

Sun, P., Basu, I., Hites, R.A. 2006b. Temporal Trends of Polychlorinated Biphenyls (PCBs) in Precipitation and

Air at Chicago. *Environmental Science & Technology*, 40(4): 1178-1183.

Sun, P., Backus, S., Blanchard, P., Hites, R.A. 2006c. Temporal and Spatial Trends of Organochlorine Pesticides in Great Lakes Precipitation. *Environmental Science and Technology*, 40(7): 2135-2141.

Sun, P., Blanchard, P., Brice, K.A. and Hites, R.A. 2006d. Trends in Polycyclic Aromatic Hydrocarbon Concentrations in the Great Lakes Atmosphere. *Environmental Science and Technology*, 40(20), 6221-6227.

Sun, P., Blanchard, P., Brice, K.A., Hites, R.A. 2006e. Atmospheric Organochlorine Pesticide Concentrations near the Great Lakes: Temporal and Spatial Trends. *Environmental Science and Technology*, 40(21): 6587-6593.

Sun, P., Basu, I., Blanchard, P., Brice, K.A. and Hites, R.A. 2007. Temporal and Spatial Trends of Atmospheric Polychlorinated Biphenyl Concentrations near the Great Lakes. *Environmental Science and Technology*, 41(4): 1131-1136.

Venier, M. and Hites, R.A. 2008. Flame Retardants in the Atmosphere near the Great Lakes. *Environmental Science and Technology*, 42 (13): 4745–4751.

Venier, M., Ferrario, J., Hites, R.A. 2009. Polychlorinated Dibenzo-p-dioxins and Dibenzofurans in the Atmosphere Around the Great Lakes. *Environmental Science and Technology*, 43 (4): 1036-1041.

Venier, M. and Hites, R.A. 2010. Regression Model of Partial Pressures of PCBs, PAHs, and Organochlorine Pesticides in the Great Lakes' Atmosphere. *Environmental Science and Technology*, 44 (2): 618–623.

Trends in Great Lakes Fish

Photo: Lake trout, Lake Superior Minnesota
Steve Geving, Minnesota Department of Natural Resources. Courtesy of US EPA GLNPO.

Open Lake Fish Contaminants Monitoring Program – Great Lakes:

Contaminants in Whole Fish

Submitted by
Elizabeth Murphy, US EPA Great Lakes National Program Office
Sean Backus, Environment Canada

The Great Lakes Fish Monitoring Program (operated by US EPA GLNPO) and the Great Lakes Fish Contaminant Surveillance Program (operated by EC)[67] monitor contaminant burdens in open water fish species from throughout the Great Lakes. These programs provide data to describe temporal and spatial trends of bioavailable contaminants as an indicator of ecosystem health. The two monitoring programs annually monitor the burden of a suite of toxic chemicals in fish and fish communities throughout the Great Lakes. They were developed in direct response to the needs of Annex 11 (Surveillance & Monitoring) of the GLWQA (1978), which states the need "To provide information for measuring local and whole lake response(s) to control measures using trend analysis and cause/effect relationships and to provide information which will assist in

[67] In the spring of 2006, Environment Canada assumed the responsibilities of the Department of Fisheries and Ocean (DFO) Fish Contaminant Surveillance Program. All data included in this report were produced by DFO.

the development and application of predictive techniques for assessing the impact of new developments and pollution sources." Annex 11 also contains a requirement for the identification of emerging problems and provides support for the development of Remedial Action Plans (RAPs) at AOCs and Lakewide Management Plans (LaMPs) for Critical Pollutants pursuant to Annex 2 of the GLWQA.

The programs also address requirements of GLWQA Annex 12, Persistent Toxic Substances. They provide the specific monitoring capabilities required in section 4 (a-d) of the Annex plus an early warning system capability (section 5a) and the development and maintenance of a biological tissue bank (section 5e) to permit retrospective analysis of recently identified compounds.

Since its inception in 1997, significant progress had been made towards the GLBTS challenge goals. In order to ensure that this pathway of progress continues into the future, Canada and the U.S., with help from the many partners involved in the GLBTS, continue to identify opportunities to reduce GLBTS substances on the road to virtual elimination. To further this effort, a number of actions have been undertaken, including, but not limited to, continued monitoring in air, water, sediment, and biota, and the consideration of impacts to the Great Lakes Basin ecosystem from Level 2 substances and other potential chemicals of concern.

Program Background Information

Long-term (>25 yrs), basin-wide monitoring programs that measure whole body concentrations of contaminants in top predator fish (lake trout and/or walleye) and in forage fish (smelt) are conducted by US EPA GLNPO through the Great Lakes Fish Monitoring Program and by EC through the Great Lakes Fish Contaminants Surveillance Program.

The U.S. program annually monitors contaminant burdens in similarly sized lake trout (600-700 mm total length) and walleye (Lake Erie, 400-500 mm total length) from alternating locations by year in each lake. Approximately 50 whole body fish are collected at each site annually. Samples are then composited by size and location into 10, 5 fish composites, for a total of 10 composites per site. The Canadian program annually monitors contaminant burdens in similarly aged lake trout (4+ to 6+ year range), walleye (Lake Erie), and in smelt. The program monitors approximately 10 Great Lakes sites annually. On Lake Ontario, four stations (Niagara, Port Credit, Cobourg, Eastern Basin) are monitored annually, while Lake Erie has sites in both the eastern and western basins. There are traditionally two sites per year monitored each on Lake Superior and Lake Huron. The two annual sites are rotated among four indicator stations on each of the Lakes (Lake Superior: Thunder Bay, Jackfish Bay, Marathon, Whitefish Bay–Sault Ste. Marie; Lake Huron: North Channel, French River, Meaford, Goderich) with the intent of collecting two consecutive years of data at any single site every three to four years. Lake trout (or walleye for western Lake Erie) are collected at each site, and elements of the food web (alewife/sculpin/smelt + invertebrate diet items) are collected at a subset of the 10 sites annually. Approximately 450 individual (top predator) and composite (forage species) fish samples are analyzed annually.

While both US EPA and EC fish monitoring programs collect and analyze contaminant burdens in Great Lakes fish on an annual basis, differences in the programs' collection and analytical methods do not allow for direct comparisons between the two programs. However, although the programs differ, they both show the same general declining trend for legacy contaminants. Recently, the two programs have begun sharing samples between analytical laboratories for comparison. Results are expected shortly.

Great Lakes Top Predator Fish Contaminant Concentrations

Since the late 1970s, concentrations of historically regulated contaminants such as PCBs, dichlorodiphenyl-trichloroethane (DDT), and mercury (Hg) have generally declined in most monitored fish species. Concentrations of other currently regulated and unregulated contaminants have generally demonstrated slowing declines. The changes are often lake-specific and relate to the characteristics and sources of the substances

involved and the biological composition of the fish community. For example:

- **Lake Superior** – Contaminants in Lake Superior are typically atmospherically derived. The dynamics of Lake Superior allow for the retention of contaminants much longer than any other lake.
- **Lake Michigan** – Food web changes are critical to Lake Michigan contaminant concentrations, as indicated by the failure of the alewife population in the 1980s and the presence of the round goby. Aquatic invasive species have had a significant impact on the food web; zebra and quagga mussels in particular have been associated with major declines in Diporeia, an important food source for many fish species. The threat of Asian carp entering Lake Michigan through the Chicago Sanitary and Ship Canal is also a major concern due to the danger the fish pose to the food web.
- **Lake Huron** – Contaminant loadings to Saginaw Bay in Lake Huron continue to be reflected in fish tissue contaminant concentrations. Significant changes to the Lake Huron food web in recent years, including significant declines in many zooplankton and forage fish, have had a critical impact on fish contaminant concentrations.
- **Lake Erie** – Aquatic invasive species are of major concern to Lake Erie because of the potential to alter the pathways and fate of persistent toxic substances. This results in differing accumulation patterns, particularly near the top of the food chain.
- **Lake Ontario** – The impact of historic point sources in the Lake Ontario Basin and along the Niagara River continue to be reflected in fish tissue contaminant concentrations. Food web changes are critical for fish contaminant concentrations. Dioxins and furans, contaminants of concern for Lake Ontario, were added to the monitoring program's analyte list in recent years, but trend data are not yet available.

Monitored Contaminants

Σ PCBs. In general, total PCB concentrations in Great Lakes top predator fish have declined since their phase-out in the 1970s (Figures 9-25 and 9-26). However, rapid declines are no longer observed, and concentrations in fish remain above the US EPA wildlife protection value of 0.16 ppm (US EPA, 1995) and the GLWQA criteria of 0.1 ppm for the protection of birds and animals that eat fish. Concentrations remain high in top predator fish due to the continued release of uncontrolled sources and their persistent and bioaccumulative nature.

Σ DDT. In general, total DDT concentrations in Great Lakes top predator fish have declined since the chemical was banned in 1972 (Figures 9-27 and 9-28). However, large declines are no longer observed; rather, very small annual percent declines predominate, indicating near steady-state conditions. The concentrations of this contaminant remain below the GLWQA criterion of 1.0 ppm. There is no US EPA wildlife protection value for total DDT because the PCB value is more protective. The CCME guideline for the protection of wildlife consumers of aquatic life is 14.0 ppm for total DDT.

Mercury. Concentrations of mercury are similar across all fish in all lakes (Figure 9-29). It is assumed that concentrations of mercury in top predator fish are atmospherically driven. Current concentrations in GLNPO top predator fish in all lakes remain above the GLWQA criterion of 0.5 ppm, and Canadian smelt have never been observed to be above the GLWQA criteria. Mercury was only recently added to the GLNPO routine analyte list, in 2001.

Σ Chlordane. Concentrations of total chlordane have consistently declined in whole top predator fish since its ban in the late 1980s (Figures 9-30 and 9-31). Total chlordane is composed of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane, with *trans*-nonachlor being the most prevalent of the compounds. While trans-nonachlor was the minor component of the total chlordane mixture, it is the least metabolized and

predominates within the Laurentian Great Lakes aquatic food web (Carlson and Swackhamer, 2006).

Mirex. Concentrations of mirex are highest in Lake Ontario top predator fish due to historical releases from sources near the Niagara River (Figures 9-32 and 9-33).

Dieldrin. Concentrations of dieldrin in lake trout appear to be declining in all lakes and are lowest in Lake Superior and highest in Lake Michigan (Figures 9-34 and 9-35). Concentrations in Lake Erie walleye were the lowest of all lakes. Aldrin is readily converted to dieldrin in the environment. For this reason, these two closely related compounds (aldrin and dieldrin) are considered together by regulatory bodies.

Toxaphene. Decreases in toxaphene concentrations have been observed throughout the Great Lakes in all media following its ban in the mid-1980s. However, concentrations have remained the highest in Lake Superior due to its longer retention time, cold temperatures, and slow sedimentation rate. It is assumed that concentrations of toxaphene in top predator fish are atmospherically driven (Hites, 2006).

Data used in this section are from whole body fish and are not intended for fish advisories or statements regarding human consumption. However, levels of mercury and PCBs in some sport-caught fish are sufficiently high, in some cases, to trigger fish consumption advisories issued by the states and the Province of Ontario.

Current Contaminants of Concern

There are a number of chemicals of current concern within the Great Lakes Basin. Several of these have been detected in Great Lakes fish. The foremost is the group of brominated flame retardants (BFRs), which include PBDEs and HBCD. These contaminants have been reported in fish tissues for several years throughout the Great Lakes Basin, and retrospective analyses have been conducted on archived tissue samples.

PBDEs. Both the U.S. and Canada analyze for PBDEs in whole top predator fish. PBDEs are used in everyday items, such as furniture upholstery and foam, to make them difficult to burn. Analyses of whole lake trout (walleye in Lake Erie) indicate a declining trend in total PBDE concentrations in the Great Lakes from 1999 to 2005. The declining trends seen in the Great Lakes are an example of the success that can be achieved through voluntary efforts—the sole U.S. manufacturer of PBDEs agreed to voluntarily phase-out production of these chemicals by the end of 2004. As illustrated in Figure 9-36, the highest concentrations are found in Lake Michigan. This is consistent with the large human population and intense industrial activity surrounding Lake Michigan (Zhu and Hites, 2004).

HBCD. One of the most widely used BFRs is HBCD. This chemical is mainly used as a flame retardant in polystyrene insulation boards and the back coating of upholstery fabric. Based on its use pattern, as an additive BFR, it has the potential to migrate into the environment from its application site. Recent studies in Lake Ontario (Tomy et al., 2004) have confirmed that HBCD isomers do bioaccumulate in aquatic ecosystems and do biomagnify as they move up the food chain. Table 9-1 presents total HBCD concentrations (α and γ isomers) for various species in the Lake Ontario food web.

Table 9-1. Lake Ontario Food Web Bioaccumulation of HBCD Isomers

SPECIES (ng/g wet wt ±S.E.)	ΣHBCD (α+γ isomers)
Lake Trout	1.68± 0.67
Sculpin	0.45± 0.10
Smelt	0.27± 0.03
Alewife	0.13± 0.02
Mysis	0.07± 0.02
Diporeia	0.08 ±0.01
Plankton	0.02± 0.01

Source: Tomy et al., 2004

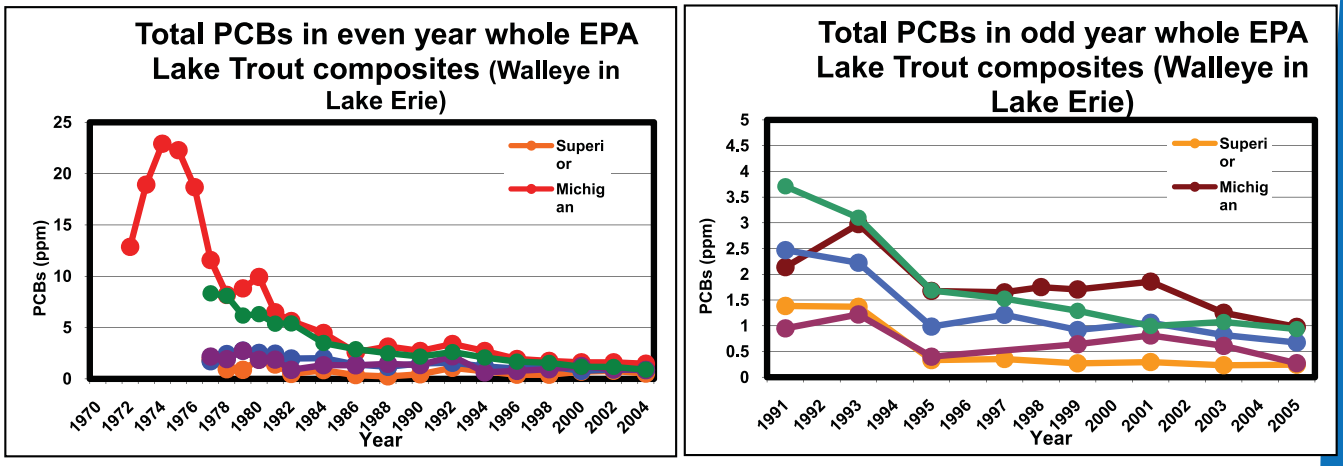


Figure 9-25. **Total PCBs in Great Lakes Top Predator Fish, Even Year (left) and Odd Year (right) Sites. Source: US EPA GLNPO Great Lakes Fish Monitoring Program**

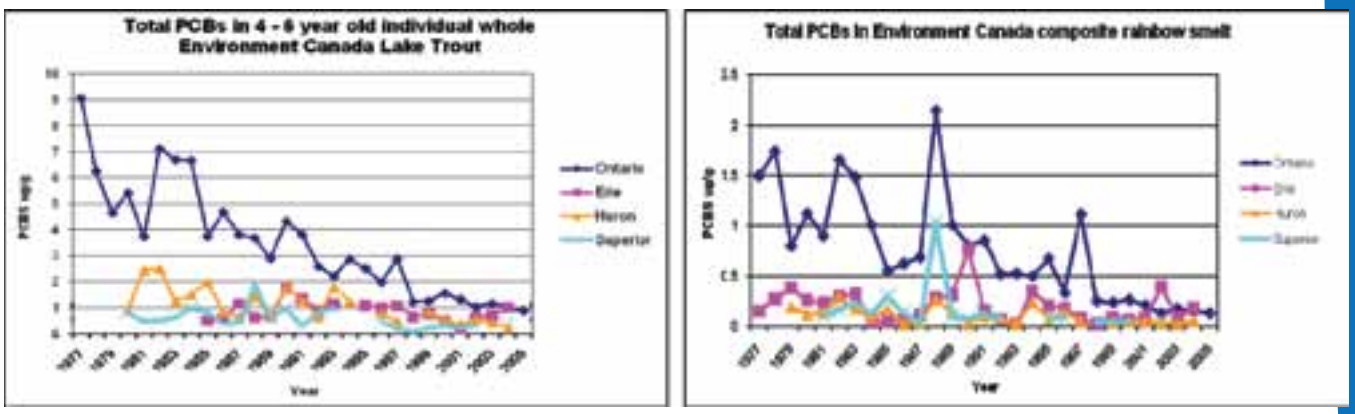


Figure 9-26. **Total PCBs in Great Lakes Lake Trout (left) and Great Lakes Smelt (right). Source: Environment Canada Great Lakes Fish Contaminant Surveillance Program**

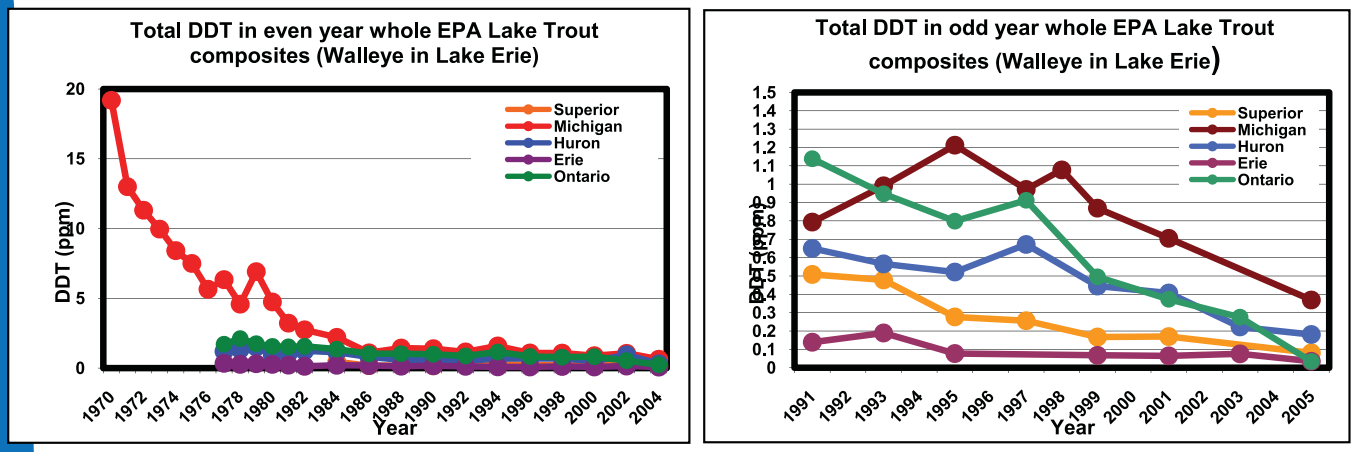


Figure 9-27. Total DDT in Great Lakes Top Predator Fish, Even Year (left) and Odd Year (right) Sites. Source: US EPA GLNPO Great Lakes Fish Monitoring Program

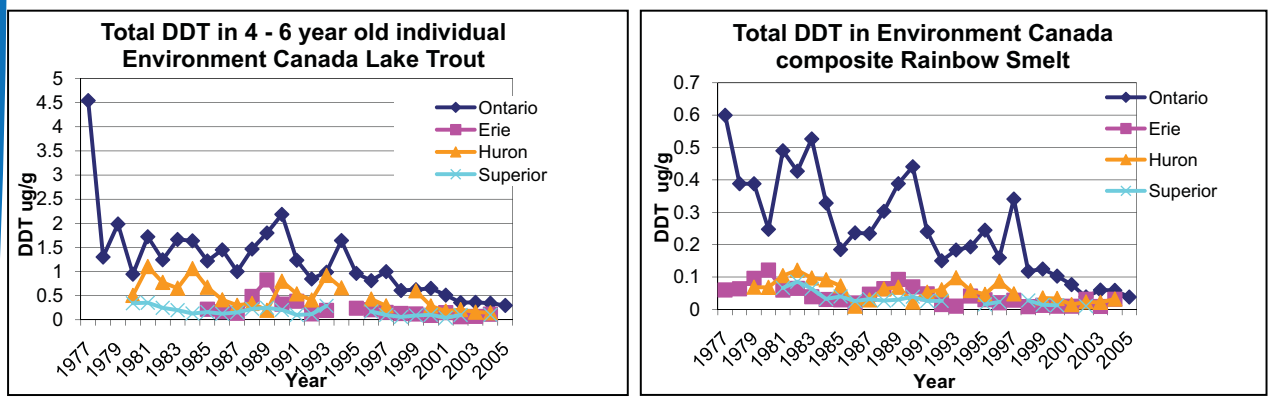


Figure 9-28. Total DDT in Great Lakes Lake Trout (left) and Great Lakes Smelt (right). Source: Environment Canada Great Lakes Fish Contaminant Surveillance Program

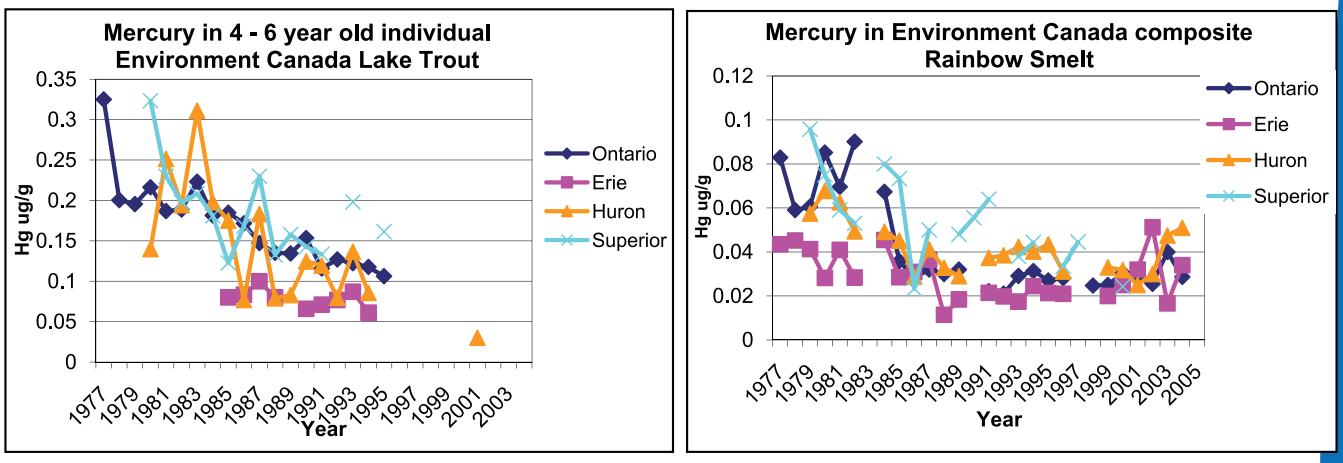


Figure 9-29. **Mercury in Great Lakes Lake Trout (left) and Great Lakes Smelt (right). Source: Environment Canada Great Lakes Fish Contaminant Surveillance Program**

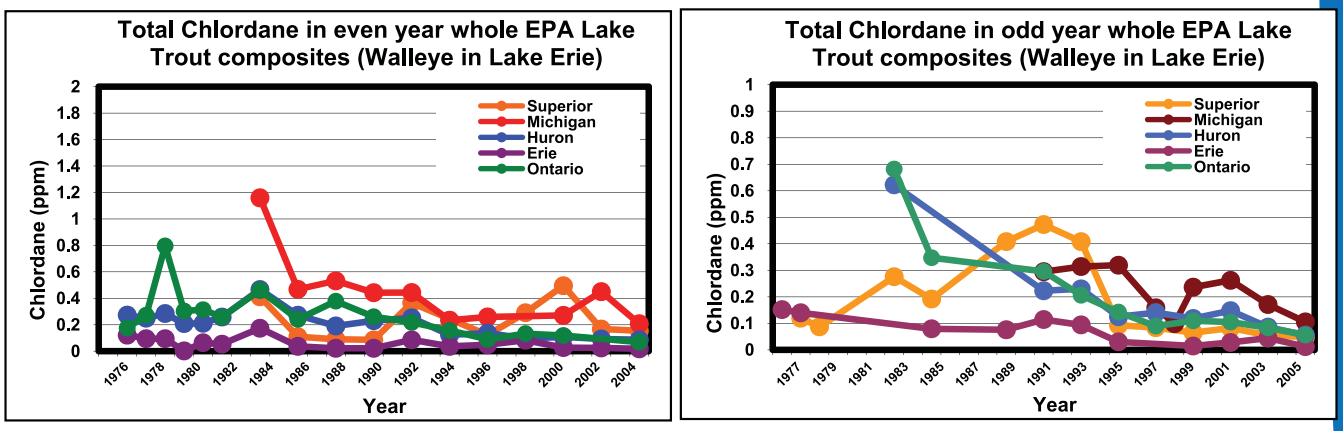


Figure 9-30. **Total Chlordane in Great Lakes Top Predator Fish, Even Year (left) and Odd Year (right) Sites. Source: US EPA GLNPO Great Lakes Fish Monitoring Program**

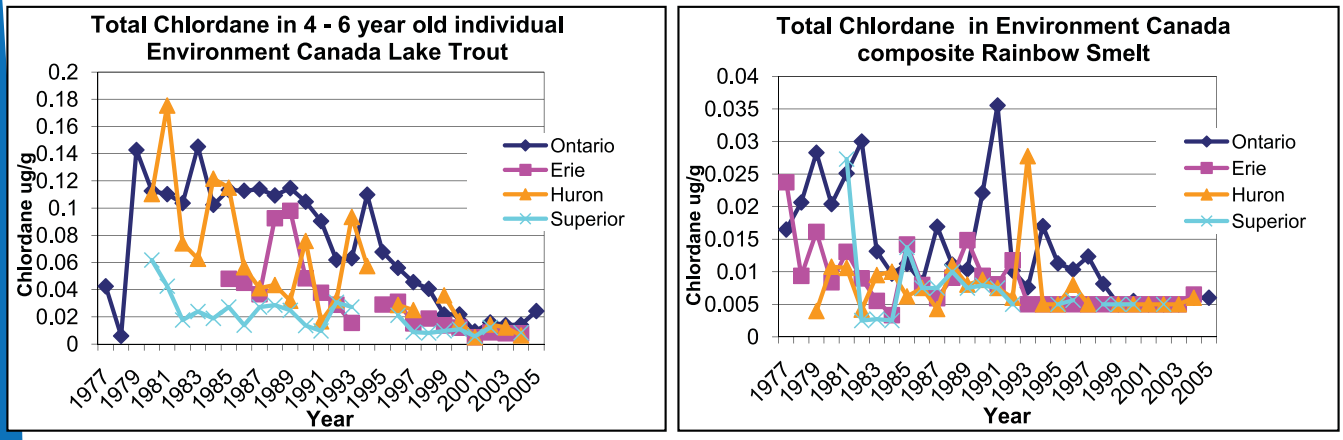


Figure 9-31. Total Chlordane in Great Lakes Lake Trout (left) and Great Lakes Smelt (right). Source: Environment Canada Great Lakes Fish Contaminant Surveillance Program

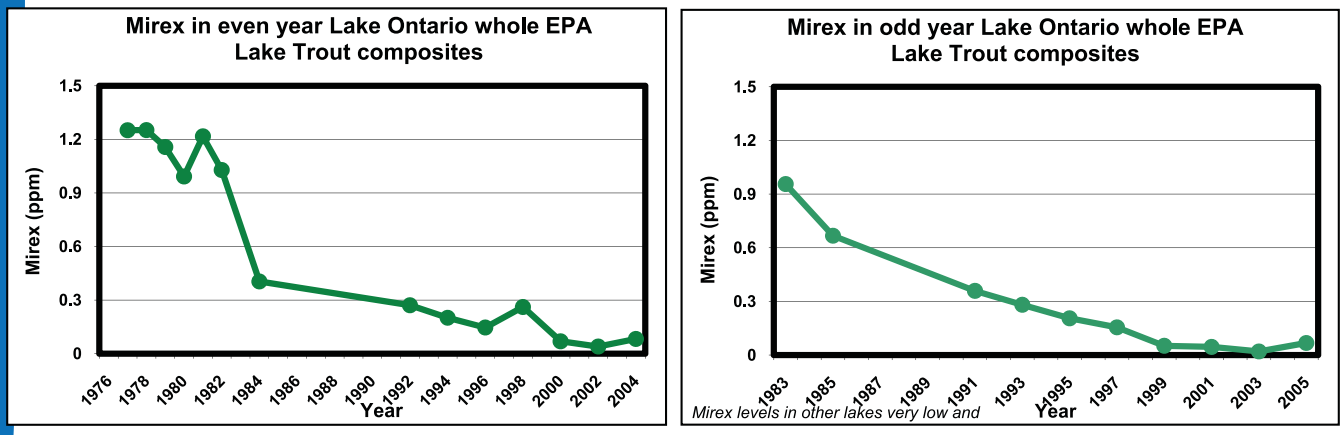


Figure 9-32 Mirex in Lake Ontario Lake Trout, Even Year (left) and Odd Year (right) Sites. Source: US EPA GLNPO Great Lakes Fish Monitoring Program

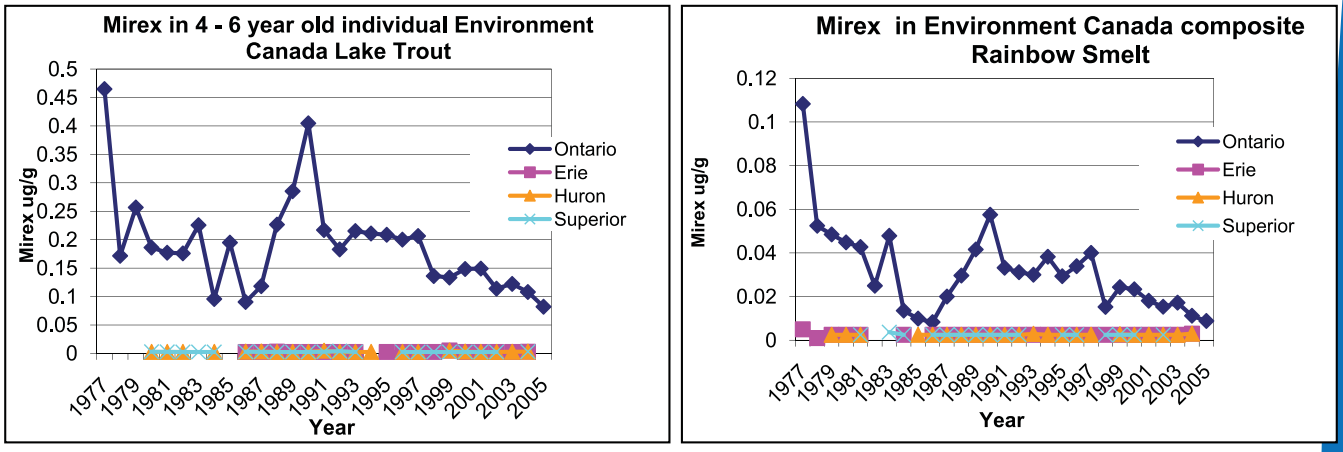


Figure 9-33. **Mirex in Great Lakes Lake Trout (left) and Great Lakes Smelt (right). Source: Environment Canada Great Lakes Fish Contaminant Surveillance Program**

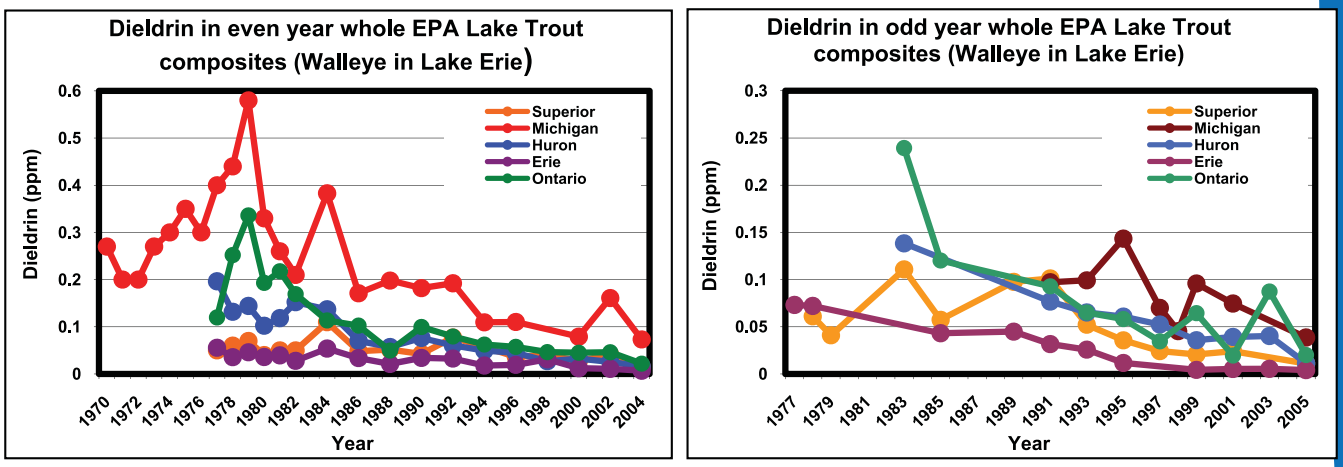


Figure 9-34. **Dieldrin in Great Lakes Top Predator Fish, Even Year (left) and Odd Year (right) Sites. Source: US EPA GLNPO Great Lakes Fish Monitoring Program**

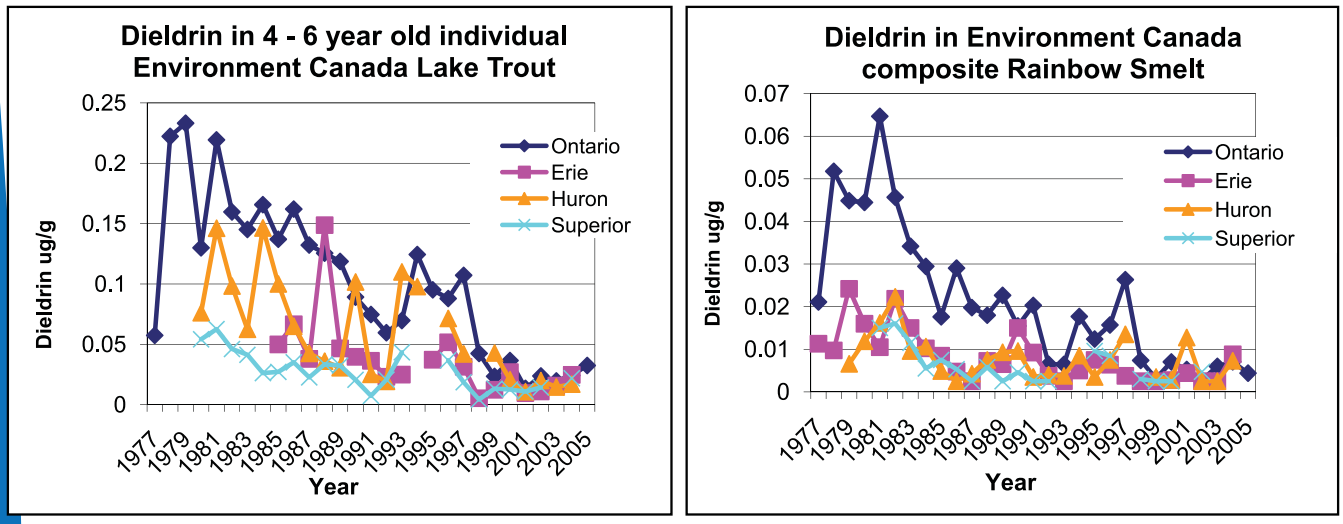


Figure 9-35. Dieldrin in Great Lakes Lake Trout (left) and Great Lakes Smelt (right). Source: Environment Canada Great Lakes Fish Contaminant Surveillance Program

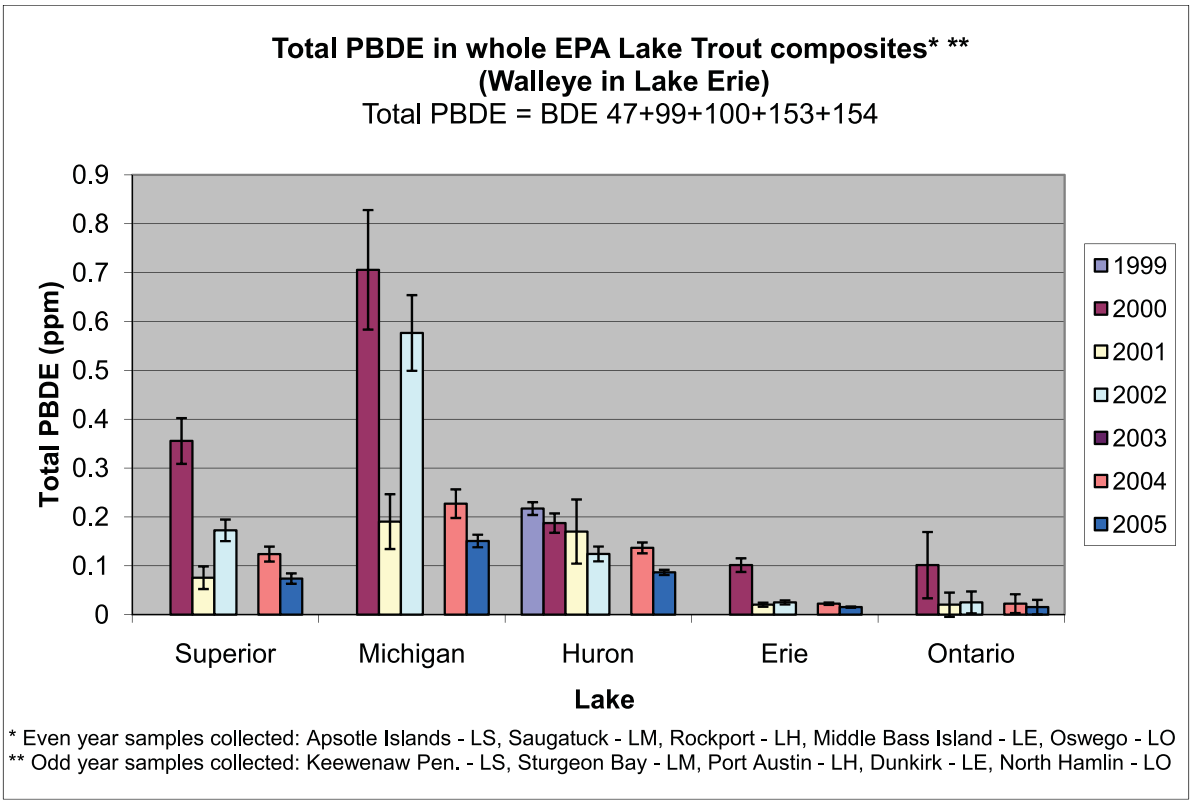


Figure 9-36. Temporal Trends in Total PBDE Concentrations in Whole Fish in the Great Lakes (1999-2005). Source: US EPA GLNPO

References

Carlson, D.L. and Swackhamer, D.L. 2006. Results from the U. S. Great Lakes fish monitoring program and effects of lake processes on bioaccumulative contaminant concentrations. *Journal of Great Lakes Research*, 32 (2), 370-385.

Cook, P.M., Robbins, J.A., Endicott, D.D., Lodge, K.B., Guiney, P.D., Walker, M.K, Zabel, E.W., and Peterson, R.E. 2003. Effects of Aryl Hydrocarbon Receptor-Mediated Early Life Stage Toxicity on Lake Trout Populations in Lake Ontario during the 20th Century. *Environmental Science and Technology*. 37(17) 3878-3884.

Great Lakes Water Quality Agreement of 1978. International Joint Commission, United States and Canada. September, 1989. http://www.ijc.org/rel/agree/quality.html.

Hites, R.A. 2006. Persistent Organic Pollutants in the Great Lakes. Hites, R. A. (editor), Persistent Organic Pollutants in the Great Lakes: An Overview (pp. 13-70). Berlin Heidelberg, Germany: Springer.

Martin, J.W., D.M. Whittle, D.C.G. Muir, and S.A. Mabury. 2004. Perfluoroalkyl Contaminants in the Lake Ontario Food Web. Environ. Sci. Technol. 38. 5379-5385.

Muir, D.C.G. and P.H. Howard. Are There Other Persistent Organic Pollutants? A Challenge for Environmental Chemists. Environ. Sci. Technol., ASAP Article 10.1021/es061677a S0013-936X(06)01677-4 Web Release Date: November 2, 2006.

Stock, N.L., J. Bonin, D.M. Whittle, D.C.G. Muir, and S.A. Mabury. 2003. Perfluoronated Acids in the Great Lakes. SETAC Europe 13th Annual Meeting, Hamburg, Germany, April 27 - May 1, 2003.

Tomy, G.T., W. Budakowski, T. Halldorson, D.M. Whittle, M. Keir, C. Marvin, G. MacInnis, and M. Alaee. 2004. Biomagnification of α- and γ-hexabomocyclododecane in a Lake Ontario Food Web. *Environ. Sci. Technol*. 38 (2298-2303).

US EPA Document # 820B95008. *Great Lakes Water Quality Initiative Criteria Documents for the Protection of Wildlife: DDT Mercury 2378-TCDD PCBS*. March 1995. http://tinyurl.com/3cruv9.

Whittle, D.M., D.C. MacEachen, D.B. Sergeant, M.J. Keir, and M.D. Moles. Food Web Biomagnification of Brominated Diphenyl Ethers in Two Great Lakes Fish Communities. 3rd International Workshop on Brominated Flame Retardants (pp. 183-186). Toronto, ON, June 6-9, 2004.

Zhu, L. Y. and R.A. Hites. 2004. Temporal Trends and Spatial Distributions of Brominated Flame Retardants in Archived Fishes from the Great Lakes. *Environ. Sci. Technol*, 38(10):2779-2784.

Trends in Great Lakes Herring Gull Eggs

Photo: Herring gull, unknown location National Park Service, Indiana Dunes National Lakeshore. Courtesy of US EPA GLNPO.

Canadian Wildlife Service Great Lakes Herring Gull Egg Monitoring Program: Trends in Emerging and Legacy Contaminant Levels in Herring Gull Eggs

Submitted by
D.V. Chip Weseloh, Dave Moore, and Robert Letcher
Canadian Wildlife Service
Environment Canada, Ontario Region

As part of EC's Great Lakes Herring Gull Egg Monitoring Program, the Canadian Wildlife Service and the Wildlife and Landscape Science Directorate have analyzed the temporal and spatial trends of contaminant concentrations (e.g., PCBs, organochlorine pesticides, and PCDD/PCDFs) in herring gulls via analysis of eggs collected from 15 colonial sites on the Great Lakes for over 35 years. Eggs have been collected since the early 1970s from the surroundings of up to eight water bodies within the Great Lakes Basin: the St. Lawrence, Niagara, and Detroit Rivers and Lakes Ontario, Erie, Huron, Michigan, and Superior. This section presents an analysis of trends in legacy contaminants in herring gull eggs from 1974 to 2007, an analysis of recent data only on trends in gull eggs (1997-2007), and an update on spatial trends to identify which sites are the most (and least) contaminated by legacy substances, based on herring gull egg data from 2003 to 2007. Changes in aquatic food webs and their impact on the contaminants being monitored in herring gulls are also discussed. Results are also presented for emerging compounds. In recent years, spatial and retrospective temporal trends of emerging contaminants have been carried out using egg homogenates that have been archived in EC's National Wildlife Specimen Bank (EC-NWSB). For example, emerging contaminants classified as brominated flame retardants (BFRs) and perfluorinated compounds (PFCs), and their degradation and precursor products, respectively, have been identified and trends assessed. In addition, there have been studies on Great Lakes herring gulls that investigated effects, for example, on the competitive binding to thyroid hormone transport proteins as an indicator of effects on circulating thyroid hormones. Recent results are available in the published literature (see References).

Study Areas and Methods

The methods and protocol for the Great Lakes Herring Gull Egg Monitoring Program have been described previously (Mineau, et al., 1984; Ewins, et al., 1992; DiMao, et al., 1999; Hebert et al., 1999, Weseloh et al., 2006). Briefly, 10 to 13 fresh herring gull eggs from 15 colonies spanning all five Great Lakes, as part of EC's Great Lakes Herring Gull Egg Monitoring Program, were collected (Figure 9-37). Collections were made in late April to early May ranging from 1974 to 2009 (depending on the study). Eggs were sent to the Canadian Wildlife Service National Wildlife Research Centre (Ottawa, Canada), where they were processed into individual and pooled egg homogenates and then refrigerated at -40°C, prepared, and analyzed for various legacy POPs within 8 weeks of collection (Won et al., 2001). For emerging POPs, and in very recent years, analysis has occurred within the year the eggs were collected (Gauthier et al., 2007; Gebbink et al. 2009). However, for retrospective temporal trends studies, egg samples have been obtained from the EC-NWSB archive (Gauthier et al., 2008, 2009; Gauthier and Letcher, 2009). Prior to 1986, all eggs were analyzed individually. Although eggs are still prepared individually, since 1986 a sub-sample from each egg has been taken to form a single site pool homogeny

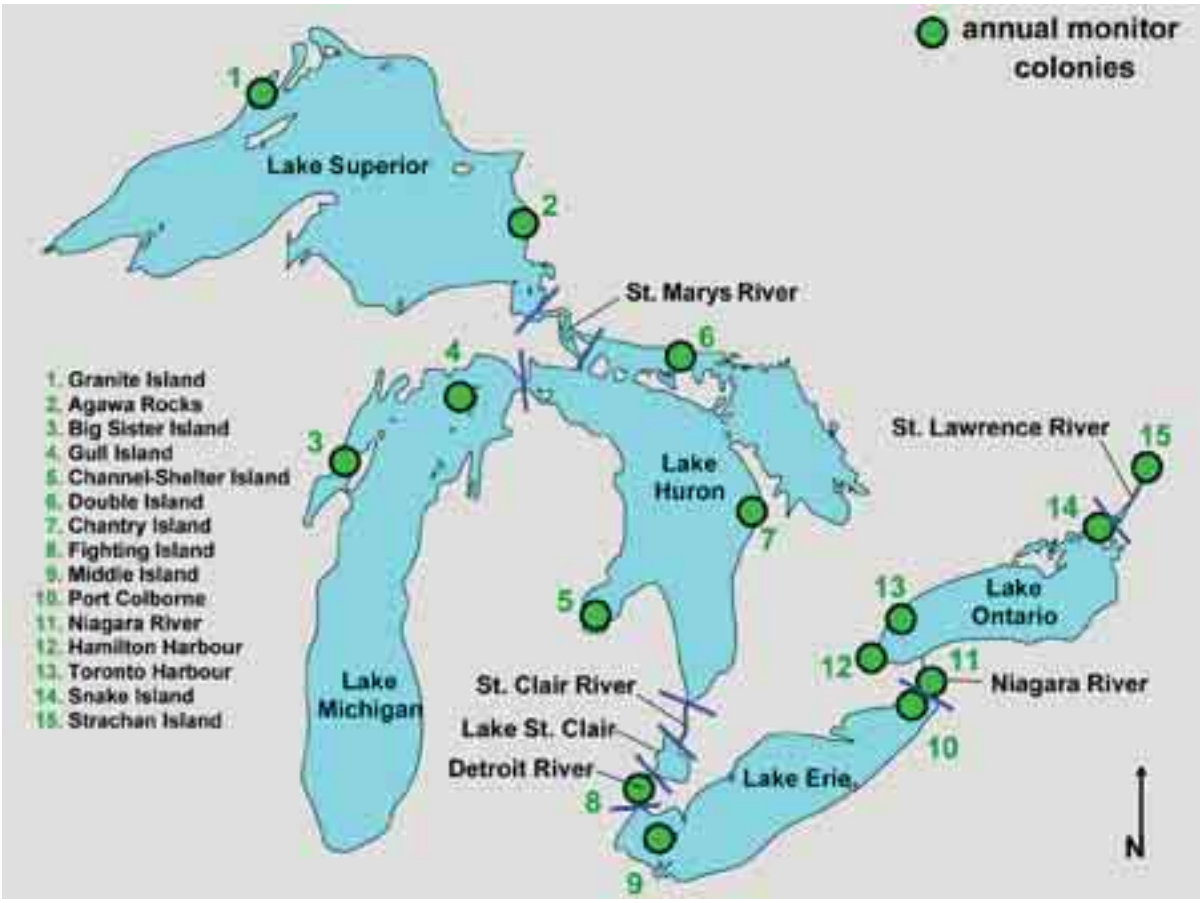


Figure 9-37. Locations of the 15 Herring Gull Colonies Sampled in This Study. Source: Canadian Wildlife Service.

on an equal wet weight basis (ng/g wet wt.), which is then analyzed.

Many of the compounds presented in this report include different kinds of flame retardants, such as total PCBs, PBDEs (penta-BDE, octa-BDE and deca-BDE derived isomers, including BDE-209), various non-PBDE brominated flame retardants (including hexabromobenzene (HBB), pentabromoethylbenzene (PBEB), pentabromotoluene, 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), (isomer-specific) hexabromocyclododecanes (HBCDs), and derivatives of tetrabromobisphenol A (TBBPA)), as well as isomers of the chlorinated flame retardant DP. In addition, this report includes various kinds of polyfluorinated precursor compounds including perfluorosulfonates (PFSAs; including PFOS and its isomers and precursors), perfluorocarboxylic acids (PFCAs), and the PFOS precursor to perfluoro-1-octanesulfonamide (PFOSA). Due to analytical difficulties, there are no new results for dioxins and furans beyond those presented in the 2006 GLBTS Progress Report. [68]

It is well-known that there have been dramatic declines in virtually all the legacy compounds in gull eggs since the program started; many compounds at most sites have declined more than 90% (Pekarik and Weseloh, 1998; Weseloh et al., 2003, 2005; Hebert et al., 2008a). Therefore, the new data presented in this report deal mainly with temporal and spatial trends over the last 10 years for which we have data, 1997-2007.

[68] USEPA and Environment Canada. 2007. Great Lakes Binational Toxics Strategy Progress Report, February 2007. Available at http://www.epa.gov/glnpo/bns/reports/2006glbtsprogressreport.pdf.

All temporal trends were determined by linear regression on log transformed data, using sequential Bonnferroni-adjusted p-values to assess significance (due to multiple independent tests, by site, for each compound; Rice, 1989). Individual annual data for all compounds and sites can be found in Bishop et al. (1992), Pettit et al. (1994), Pekarik et al. (1998), and Jermyn et al. (2002; Canadian Wildlife Service, unpublished report). To determine spatial trends, mean values of seven major contaminants in herring gull eggs were calculated for each site for the five-year period 2003-2007. The sites were ranked according to the concentrations of each compound relative to fish flesh criteria for the protection of piscivorous wildlife, and a single overall rank was calculated for each site (Weseloh et al., 2006; S. deSolla, unpubl. data).

Herring gull eggs were collected from the following sites (Figure 9-37):

- **St. Lawrence River (SLR)** – Strachan Island (near Cornwall)
- **Lake Ontario (LO)** – Snake Island (near Kingston), Tommy Thompson Park (Toronto Harbour) and Neare Island (Hamilton Harbour)
- **Niagara River (NR)** – an unnamed island 300 m above Niagara Falls
- **Lake Erie (LE)** – Port Colborne Lighthouse and Middle Island
- **Detroit River (DR)** – Fighting Island
- **Lake Huron (LH)** – Chantry Island, Double Island (North Channel), and Channel-Shelter Island (Saginaw Bay)
- **Lake Michigan (LM)** – Gull Island and Big Sister Island (Green Bay)
- **Lake Superior (LS)** – Agawa Rocks and Granite Island (Black Bay)

Current concentrations of eight contaminants and percentage change during the study period were calculated as the average value of the sites within each water body (Table 9-2). One site in Lake Ontario (Hamilton Harbour, site 12) and one in Lake Huron (Saginaw Bay, site 5) were not included for this calculation because their time series were not continuous with the two other sites from each of those lakes.

Temporal and Spatial Trends of Emerging Contaminants

Results

Flame Retardants

For the Laurentian Great Lakes, reports on DP have been limited to sediment and fish, and temporal trends in Great Lakes wildlife are unknown. Both *syn*- and *anti*-DP isomers were detected in herring gull egg pools spanning collection years from 1982 to 2006, and from seven colonies in the five Laurentian Great Lakes (Gauthier et al., 2007; Gauthier and Letcher, 2009) (Figure 9-38). The sum (Σ) of *syn*- and *anti*-DP concentrations were generally <15 ng/g wet wt. and variable depending on the colonial site and year, although Σ-DP concentrations were generally higher post mid-1990s for all sites (Gauthier and Letcher, 2009). *Syn*- and *anti*-DP concentrations ranged from 310 to 1400 ng/g wet wt. and 130 to 4400 pg/g wet wt., respectively. There was a weak but significant, negative relationship (as determined by Spearman's rank correlation coefficient $r_S = -0.31$, $p < 0.001$) between the Σ-DP concentration and distance from the only DP production facility in North America at Niagara Falls, New York. However, the fraction of the *anti*-DP to the Σ-DP concentration (*fanti*) was 0.69 ± 0.08 (for all seven colonies and years, n = 101 pools). There was no significant, negative relationship ($r_S = -0.18$, $p = 0.07$) of *fanti* with increasing distance from the production facility at Niagara Falls, New York, which indicated that there was no temporal or spatial enrichment of either isomer relative to the commercial DP mixture. Over the past 25 years, it is clear that DP isomers have accumulated in the food web of female herring gulls, with subsequent transfer during ovogenesis.

Norstrom et al. (2002) reported on the geographical distribution of 25 di- to hepta-bromo-BDE congeners, derived from penta-BDE and octa-BDE mixtures, in Great Lakes herring gull eggs (13 egg pools) from the 15 monitoring colonies. PBDEs were found at concentrations ranging from 192 to

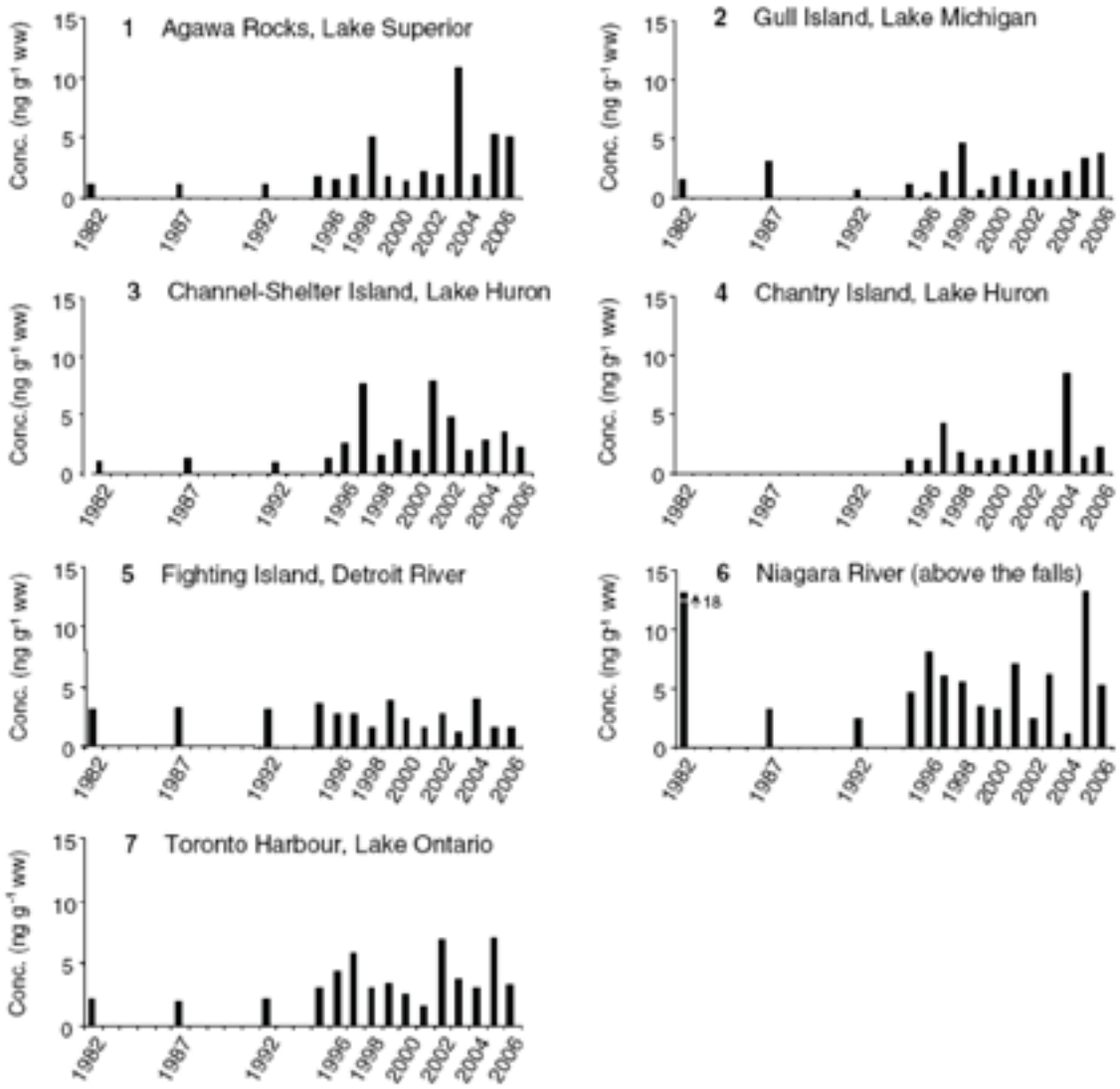


Figure 9-38. Total Concentrations (ng/g wet wt.) of Dechlorane Plus Isomers (*anti* and *syn*) and Temporal Distribution (1982–2006) in the Eggs of Herring Gulls at Seven Representative Colonies in the Laurentian Great Lakes. [69]

1400 ng/g, mean of 662 ± 368 ng/g wet wt. (of egg contents). The highest concentrations were found in northern Lake Michigan and Toronto Harbour (1000-1400 ng/g), and the lowest were found in Lake Huron and Lake Erie (192-340 ng/g). The distribution suggested that input from large urban/industrial areas through air or water emissions contributes local contamination to the herring gull food web in addition to background levels from regional/global transport. The congener composition was similar among sampling sites. Major congeners were BDE-47 (43%), BDE-99 (26%), BDE-100 (13%) BDE-153 (11%), BDE-154 (4%), BDE-183 (2%), and BDE-28 (1%).

Temporal trends of PBDE contamination from 1981 to 2000 were established by analysis of archived herring gull eggs (10 egg pools) from colonies in northern Lake Michigan, Saginaw Bay, Lake Huron, and eastern Lake Ontario (Norstrom et al., 2002). BDE-47, -99 and -100, and BDE-153, -154 and -183 concentrations were grouped

[69] Gauthier and Letcher, 2009.

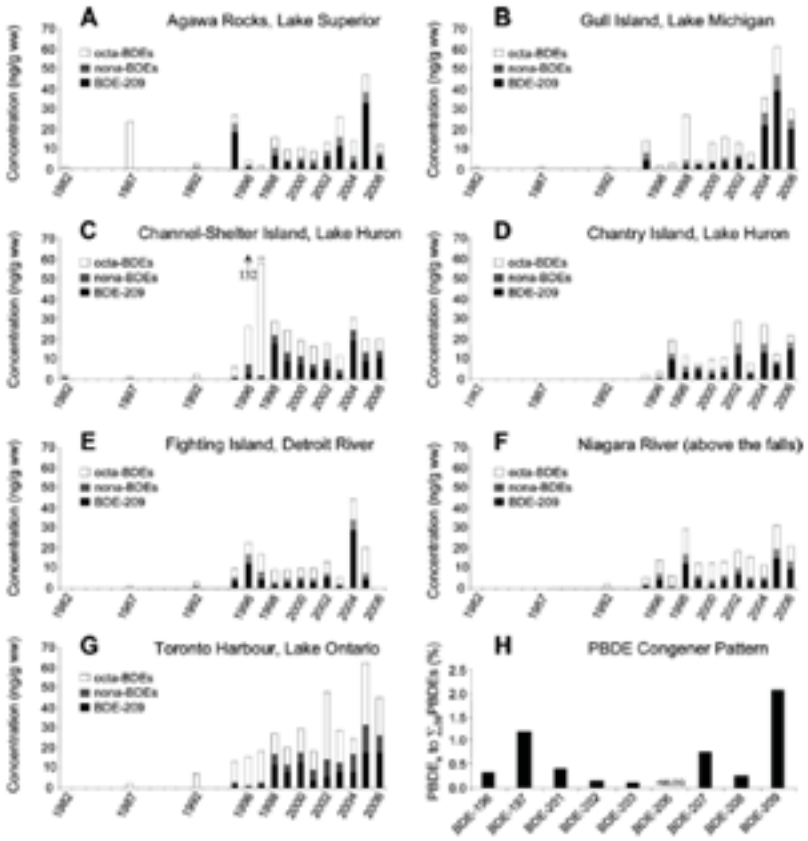


Figure 9-39. (A–G) Temporal Trends between 1982 and 2006 for Concentrations of BDE-209, ΣOcta-BDEs (BDE-194, -195, -196, -197, -201, -202, -203); ΣNona-BDEs (BDE-206, -207, -208) in Herring Gull Egg Pools from Seven Representative Great Lakes Colonies; (H) The Congener Pattern of Individual Octa- and Nona-BDE Congeners in Eggs from Toronto Harbour, Lake Ontario and Channel-Shelter Island Herring Gull Colonies (collected in 2006). [70]

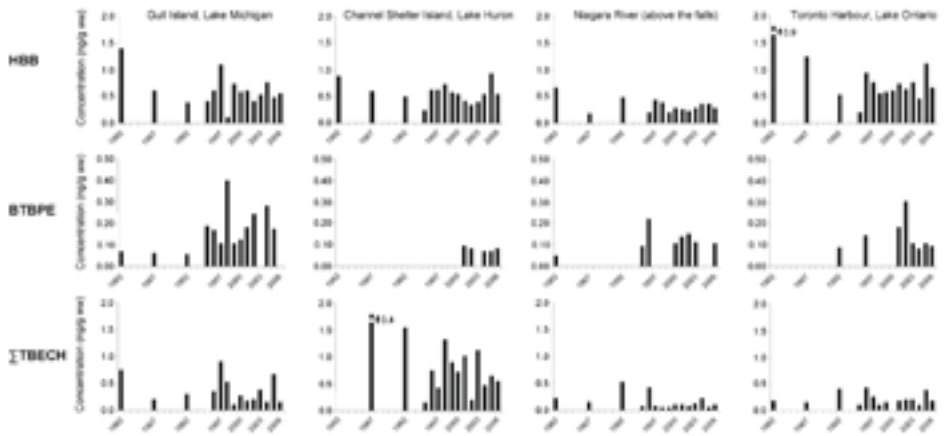


Figure 9-40. Temporal Distribution for Hexabromobenzene (HBB), 1,2-Bis(2,4,6-tribromophenoxy)ethane (BTBPE), and 1,2-Dibromo-4-(1,2-dibromoethyl)cyclohexane (sum of α- and β-isomers) at Four Representative Herring Gull Colonies on the Laurentian Great Lakes.[71]

[70] Gauthier et al., 2008.
[71] Gauthier et al., 2008.

separately for analysis because these two groups had different trends and are primarily associated with the penta-BDE and octa-BDE flame retardant formulations, respectively. BDE-47, BDE-99, and BDE-100 concentrations were 5-12 ng/g wet wt. in 1981-1983 and then increased exponentially ($p < 0.00001$) at all three sites to 400-1100 ng/g over the next 17 years. Doubling times were 2.6 years in Lake Michigan, 3.1 years in Lake Huron, and 2.8 years in Lake Ontario. BDE-154, BDE-153, and BDE-183 concentrations generally increased but varied in an erratic fashion among sites and decreased as a fraction of PBDE over time. Concentrations of BDE-154, BDE-153, and BDE-183 were 100-200 ng/g in eggs from all three colonies in 2000. Therefore, most of the dramatic increases in PBDE concentrations observed over the past 20 years in the Great Lakes aquatic ecosystem seem to be connected with the penta-BDE formulation, which is mainly used as a flame retardant in polyurethane foam in North America. It was concluded that, if these rates of change continued, concentrations of PBDEs would equal or surpass those of PCBs in Great Lakes herring gull eggs in 10-15 years.

However, in a very recent study, 43 PBDE congeners were monitored, and the temporal (1982–2006) and spatial trends were reported for quantifiable PBDEs, and in particular BDE-209, in pooled samples of herring gull eggs from seven colonies spanning the Great Lakes (Gauthier et al., 2007, 2008). BDE-209 concentrations in 2006 egg pools ranged from 4.5 to 20 ng/g wet wt. and constituted 0.6-4.5% of Σ_{39}PBDE concentrations among colonies, whereas Σocta-BDE and Σnona-BDE concentrations constituted from 0.5 to 2.2% and 0.3 to 1.1%, respectively. From 1982 to 2006, the BDE-209 doubling times ranged from 2.1 to 3.0 years, whereas for Σocta-BDEs and Σnona-BDEs, the mean doubling times ranged from 3.0 to 11 years and from 2.4 to 5.3 years, respectively (Figure 9-39). The source of the octa- and nona-BDE congeners (e.g., BDE-207 and BDE-197) is the result of BDE-209 debromination, and they are either formed metabolically in Great Lakes herring gulls and/or bioaccumulated from the diet and subsequently transferred to their eggs. In contrast to BDE-209 and the octa- and nona-BDEs, congeners derived mainly from penta-BDE and octa-BDE mixtures (e.g., BDE-47, BDE-99, and BDE-100) showed rapid increases up until 2000; however, there was no increasing trend post-2000. The data illustrated that PBDE concentrations and congener pattern trends in Great Lakes herring gull eggs had dramatically changed between 1995 and 2006. Regardless of BDE-209 not fitting the pervasive criteria as a persistent and bioaccumulative substance, it clearly has been of increasing concern in Great Lakes herring gulls, and provides evidence that regulation of deca-BDE formulations may be warranted.

The production and use of non-PBDE (BFR) alternatives have been on the rise, although their assessment in environmental samples is largely understudied. Several non-PBDE BFRs were found in the egg pools of herring gulls from seven colonies in the five Laurentian Great Lakes (collected from 1982 to 2006) (Gauthier et al., 2007, 2009). Of the 19 non-PBDE BFRs monitored, hexabromobenzene (HBB), 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE), decabromodiphenyl ethane (DBDPE), and α- and β-isomers of 1,2-dibromo-4-(1,2-dibromoethyl) cyclohexane (TBECH) were present in eggs from all the colonies (Figure 9-40). In 2005 and 2006 eggs, the concentrations of DBDPE were highest at three of the seven colonies (1.3 to 288 ng/g wet wt.) and surpassed concentrations of BDE-209. HBB (0.10 to 3.92 ng/g wet wt.), BTBPE (1.82 to 0.06 ng/g wet wt.), and ∑-TBECH (0.04 to 3.44 ng/g wet wt.; mainly the β-isomer 52 to 100% of ∑-TBECH) were detected at lower concentrations (and generally, ∑PBDE concentrations). Spatial trends were observed, although temporal trends were not obvious in most cases. Regardless, over the past 25 years, non-PBDE BFRs have accumulated variably in female herring gulls and have been transferred during ovogenesis to their eggs, indicating that there has been continual exposure and bioaccumulation of several BFRs in the Great Lakes.

Per- and Poly-Fluorinated Contaminants

Environmentally important PFCAs and PFSAs, as well as per- and polyfluorinated precursor compounds (PFCs) including several sulfonamides, telomer acids, and alcohols were determined in individual herring gull eggs collected (in 2007) from 15 colonies located at Canadian and some American sites across the Laurentian Great Lakes

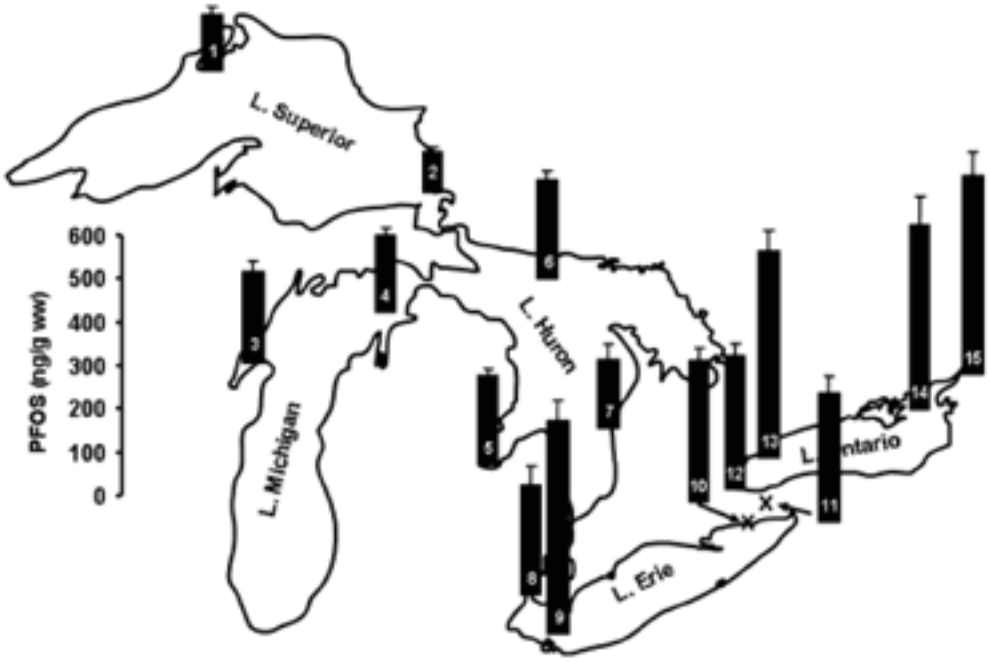


Figure 9-41. Arithmetic Mean PFOS Concentrations (ng/g wet wt. ± SE) in Individual Herring Gull Eggs (n = 13) Collected in 2007 from 15 Colonies in the Laurentian Great Lakes: (1) Granite Island, (2) Agawa Rocks, (3) Big Sister Island, (4) Gull Island, (5) Channel-Shelter Island, (6) Double Island, (7) Chantry Island, (8) Fighting Island, (9) Middle Island, (10) Port Colborne, (11) Niagara River, (12) Hamilton Harbour, (13) Toronto Harbour, (14) Snake Island, and (15) Strachan Island. [72]

(Chu and Letcher, 2009; Gebbink et al., 2009). The pattern of PFSAs (C6, C8, and C10 chain lengths) was dominated by PFOS (>90% of ΣPFSA concentration) regardless of collection location (Figure 9-41). Concentrations of ΣPFSA were significantly higher ($p < 0.03$) in eggs from Middle Island (western Lake Erie; 507 ± 47 ng/g wet wt.), Toronto Harbour (484 ± 49 ng/g wet wt.), and Strachan Island (486 ± 59 ng/g wet wt.) (Lake Ontario) compared to eggs from colonies on Lakes Superior, Michigan, and Huron. PFCAs ranging in chain length from C8 to C15 were detected in the eggs, with perfluoroundecanoic acid (PFUnA) and perfluorotridecanoic acid (PFTrA) being the dominant compounds. PFOA and perfluorononanoic acid (PFNA) were more abundant in the ΣPFCA in eggs from Lake Superior and Michigan colonies, and PFUnA and longer chain PFCAs were more abundant in the ΣPFCA in eggs from Lakes Erie and Ontario colonies. In contrast to ΣPFSA, the highest concentrations of ΣPFCA were found in eggs from Double Island, Lake Huron (113 ± 12 ng/g wet wt.), followed by eggs from colonies on Lakes Erie and Ontario. Among the PFOS or PFCA precursor compounds assessed (6:2, 8:2, and 10:2 fluorotelomer alcohols and acids and PFOSA), none were detectable in eggs from any sampling location. The exception was PFOSA (average concentration <1 ng/g wet wt.), which suggests that PFOS in the gulls and subsequently in their eggs may be due, in part, to biotransformation of PFOSA to PFOS in the gull and/or in their diet and food web. The accumulation of PFSA and PFCA from mainly aquatic dietary sources was suggested, and was highly lake- and/or colony-dependent, especially showing a northwest and southeast spatial trend, with higher concentrations in eggs from colonies in close proximity to highly urbanized and industrialized sites in Lakes Erie and Ontario.

Linear and branched [six mono(trifluoromethyl) and four di(trifluoromethyl)] isomers of PFOS were

[72] Gebbink et al., 2009.

analyzed for and the spatial patterns examined in individual herring gull eggs (n = 13 per site) collected (in 2007) from 15 colonies across the Great Lakes (Gebbink and Letcher, 2010). Linear PFOS (L-PFOS) consistently dominated the isomer pattern in all eggs, comprising between 95.0% and 98.3% of the ∑PFOS concentration. L-PFOS was highly enriched in the gull eggs, as the ∑branched-PFOS to L-PFOS isomer concentration ratios were very constant (overall average 0.038 ± 0.001) and much lower compared to technical PFOS (range 0.27 – 0.54). The highest proportions of L-PFOS were generally observed in the eggs from the lower lakes' colonies (Erie and Ontario). All six mono(trifluoromethyl) branched isomers were detected in the eggs from all the colonies. Comparable to technical PFOS (T-PFOS), the percentage of the mono(trifluoromethyl) isomer to ∑PFOS concentration was much lower than L-PFOS, and decreased as the branch substitution was located in the alkyl chain backbone closer to the sulfonate group (i.e., perfluoro-6-methyl-heptanesulfonate (P6MHpS), 0% - 2.5%; perfluoro-5-methyl-heptanesulfonate (P5MHpS), 0.43% - 1.18%; perfluoro-4-methyl-heptanesulfonate (P4MHpS), 0.25% - 0.69%; and perfluoro-3-methyl-heptanesulfonate (P3MHpS), 0.32% - 0.74%). This suggests that the apparent dilution/degradation of the mono(fluoromethyl) isomers from environmental processes that occur prior to final accumulation in herring gull eggs is independent of the mono(fluoromethyl) isomer structure. Although at even lower fractional composition than the mono(trifluoromethyl) isomers, of the di(trifluoromethyl) isomers, detected in >60% of the individual eggs per site was perfluoro-3,5-dimethyl-hexanesulfonate (P35DMHxS) and perfluoro-4,5-dimethyl-hexanesulfonate (P45DMHxS) for Toronto Harbour (Lake Ontario), P35DMHxS for Chantry (Lake Huron) and Fighting Island (Detroit River), and P45DMHxS for Gull Island (Lake Michigan). Relative to T-PFOS, and independent of colonial location, the high and consistent enrichment of L-PFOS in gull eggs is likely a function of several processes, including PFOS or precursor sources, and isomer-specific PFOS or precursor exposure, accumulation, biotransformation, retention and/or elimination.

Discussion

The spatial distribution of flame retardants and subsequent trends are affected by a variety of factors relating to bioaccumulation. Many concentrations are variable regardless of the year of collection and source site. This reflects the spatially different and temporarily variable diet of the gulls (Gauthier et al., 2007, 2008, 2009; Gauthier and Letcher, 2009; Gebbink et al., 2009; Hebert and Weseloh, 2006; Hebert et al., 2008a, 2008b, 2009a, 2009b). It is confirmed that changes in the food web and thus the diets of herring gulls are manifested in their eggs, including contaminant levels. Proximity to areas of concentrated human habitation and industrial activity also affect contamination levels spatially and temporally. For example, concentrations of PBDEs were highest from Gull Island, perhaps because gulls from the northern Great Lakes are known to migrate and over-winter close to urban centers like Milwaukee and Chicago (Gauthier et al., 2008; Norstrom et al., 2002).

Future studies should focus on a few different aspects in the study of these concentration levels. Gauthier et al. (2008) suggest that there is a need to reassess the need to restrict production and commercial usage of many formulations, including deca-BDE. Further studies are already underway regarding spatial and temporal trends assessments of many flame retardants including PBDEs and DP isomers; however, it is important that the scientific community continue to monitor new and existing flame retardants as well as other anthropogenic chemicals in the Great Lakes environment. Finally, Hebert et al. (2009a) suggest that incorporating an integrated application of ecological tracers will ultimately help lead to new insights in food web ecology, which will aid in understanding the health of herring gulls with respect to contaminants in this environment.

Table 9-2. Current, Recent and Historical Concentrations of Eight Contaminants in Herring Gull Eggs from 1974 (or year of first analysis*) to 2007[a]. Also shown are the direction and amount of change between 2005 and 2007, and the percentage decline from 1974 to 2007.

Water Body	Year	PCBs	DDE	HCB	OCS[b]	Mercury[c]	Dieldrin	Mirex	HE
Lake Superior (n=2)	1974*	62.75	16.72	0.253	0.0052	0.36	0.516	1.0528	0.1569
	2005	3.71	0.20	0.003	0.0005	0.14	0.008	0.0029	0.0072
	2007	3.29	0.70	0.009	0.0008	0.14	0.021	0.0146	0.0176
	[2007 – '05][1]	**-0.42**	**0.5**	**0.006**	**0.0003**	**0**	**0.013**	**0.0117**	**0.0104**
	[1974 – '07][2]	**94.70%**	**95.80%**	**96.40%**	**84.60%**	**61.10%**	**95.88%**	**99.72%**	**88.80%**
Lake Michigan (n=2)	1977*	107.99	29.17	0.128	0.0047	0.42	0.820	0.3570	0.4010
	2005	7.33	0.60	0.003	0.0005	0.12	0.010	0.0053	0.0072
	2007	4.86	1.05	0.007	0.0005	0.12	0.025	0.0099	0.0144
	[2007 – '05][1]	**-2.47**	**0.45**	**0.004**	**0**	**0**	**0.016**	**0.0046**	**0.0072**
	[1977 – '07][2]	**95.50%**	**96.40%**	**94.50%**	**89.40%**	**71.40%**	**96.90%**	**98.52%**	**96.41%**
Lake Huron (n=2)	1974*	71.01	17.40	0.383	0.0052	0.22	0.503	1.3370	0.1570
	2005	2.31	0.16	0.002	0.0005	0.09	0.005	0.0080	0.0027
	2007	2.76	0.38	0.006	0.0011	0.10	0.014	0.0203	0.0086
	[2007 – '05][1]	**0.45**	**0.22**	**0.004**	**0.0006**	**0.01**	**0.009**	**0.0124**	**0.0059**
	[1974 – '07][2]	**96.10%**	**98.70%**	**98.40%**	**78.80%**	**93.90%**	**97.28%**	**99.41%**	**94.52%**
Detroit River (n=1)	1978*	115.09	9.44	0.281	0.055	0.21	0.182	0.1273	0.0955
	2005	16.48	0.28	0.003	0.003	0.16	0.004	0.0001	0.0022
	2007	18.94	1.37	0.015	0.012	0.17	0.037	0.0108	0.0168
	[2007 – '05][1]	**2.46**	**1.09**	**0.012**	**0.009**	**0.01**	**0.033**	**0.0108**	**0.0146**
	[1978 – '07][2]	**83.50%**	**85.50%**	**94.50%**	**78.20%**	**19.00%**	**79.46%**	**99.96%**	**82.40%**
Lake Erie (n=2)	1974*	72.46	7.13	0.291	0.017	0.22	0.355	0.6400	0.1610
	2005	9.38	0.15	0.003	0.001	0.09	0.005	0.0050	0.0030
	2007	18.27	0.91	0.015	0.005	0.15	0.484	0.0219	0.0215
	[2007 – '05][1]	**8.89**	**0.76**	**0.012**	**0.004**	**0.055**	**0.478**	**0.0169**	**0.0185**
	[1974 – '07][2]	**87.70%**	**87.20%**	**94.80%**	**76.50%**	**33.18%**	**-36.39%**	**99.23%**	**86.68%**
Niagara River (n=1)	1979*	50.47	4.01	0.173	0.0052	0.24	0.465	7.0150	0.1549
	2005	3.98	0.16	0.003	0.0005	0.09	0.010	0.0844	0.0052
	2007	3.88	0.50	0.011	0.0017	0.11	0.028	0.2811	0.0137
	[2007 – '05][1]	**-0.1**	**0.34**	**0.008**	**0.0012**	**0.02**	**0.018**	**0.1967**	**0.0085**
	[1979 – '07][2]	**92.30%**	**87.50%**	**93.60%**	**67.30%**	**54.20%**	**94.06%**	**98.80%**	**91.16%**
Lake Ontario (n=2)	1974*	153.03	22.35	0.580	0.017	0.48	0.201	0.4930	0.0900
	2005	8.33	0.45	0.009	0.002	0.13	0.002	0.0192	0.0017
	2007	9.22	1.46	0.015	0.004	0.20	0.009	0.0572	0.0076
	[2007 – '05][1]	**0.89**	**1.01**	**0.006**	**0.002**	**0.07**	**0.007**	**0.0380**	**0.0059**
	[1974 – '07][2]	**94.00%**	**93.50%**	**97.40%**	**76.50%**	**58.30%**	**95.57%**	**96.11%**	**91.56%**
St. Lawrence River (n=1)	1986*	28.90	3.59	0.052	0.026	0.30	0.158	0.9454	0.0649
	2005	6.29	0.35	0.002	0.001	0.17	0.007	0.0560	0.0028
	2007	8.60	0.90	0.007	0.003	0.22	0.014	0.1866	0.0073
	[2007 – '05][1]	**2.31**	**0.55**	**0.004**	**0.002**	**0.05**	**0.007**	**0.1306**	**0.0045**
	[1986 – '07][2]	**70.2**	**74.90%**	**86.50%**	**88.50%**	**26.70%**	**91.39%**	**94.08%**	**88.75%**

* First year of analysis.

[a] All concentrations reported in µg/g wet weight. The average contaminant levels were calculated from the sites for each water body as listed under Study Areas and Methods, except for Lake Ontario, where only samples from Snake Island and Tommy Thompson Park (Toronto Harbour) were used, and Lake Huron, where only samples from Chantry and Double Islands were used.

[b] OCS first analyzed in 1987, at all sites except at Strachan Island, St. Lawrence River (1st yr = 1988).

[c] First year of mercury analysis on Lake Michigan was 1982; on the Detroit River was 1981; and on Niagara River was 1981.

[1] The change between 2005 and 2007.

[2] Percentage decline from the year of first analysis to 2007.

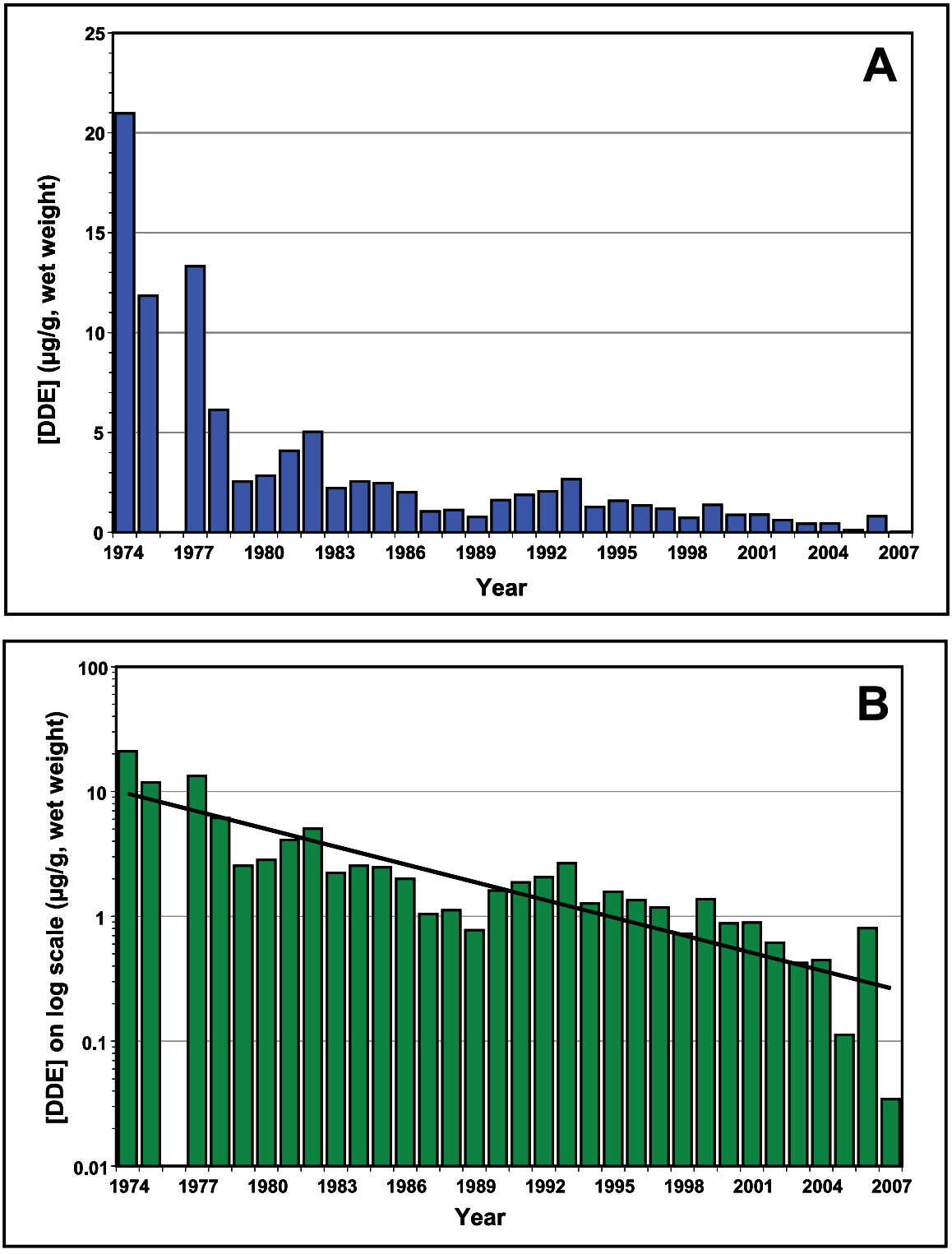


Figure 9-42. **DDE Concentrations in Herring Gull Eggs from Chantry Island, Lake Huron, 1974-2007. A) Linear scale; B) Log scale - Showing the significant regression line and an overall constant rate of decline. Source: Canadian Wildlife Service (see Study Areas and Methods).**

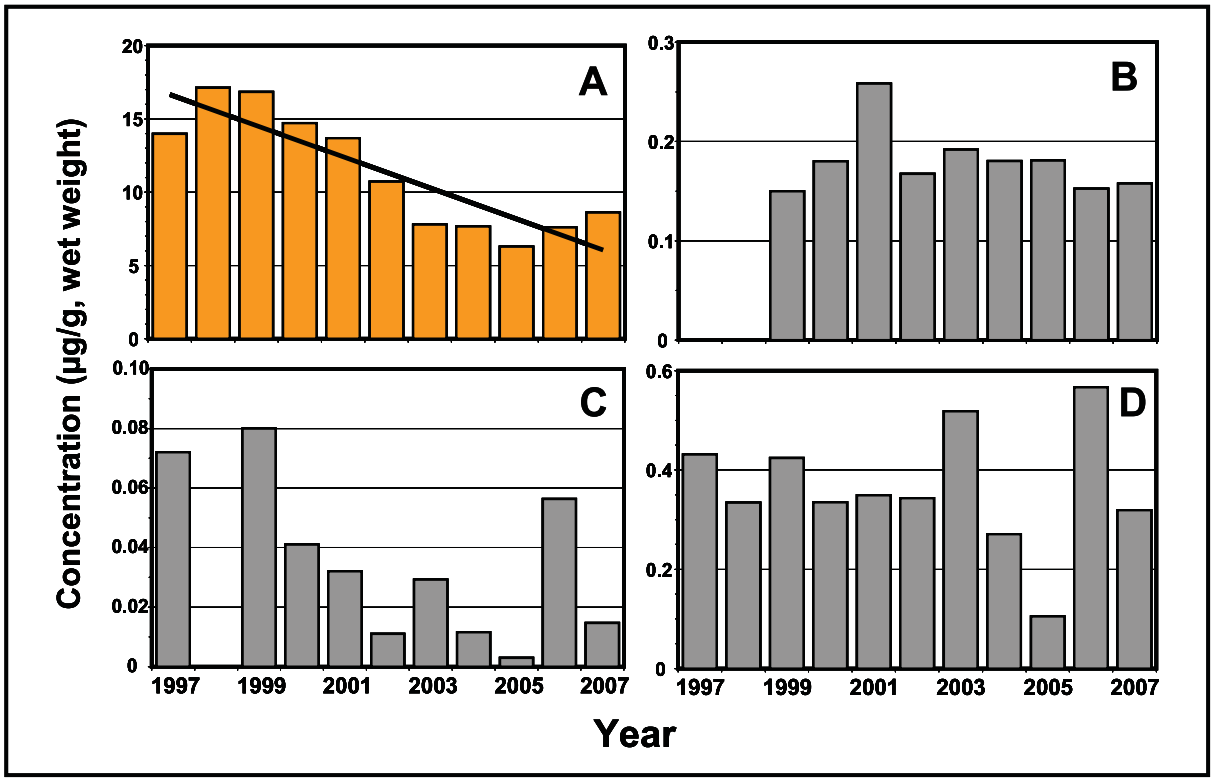


Figure 9-43. Contaminant Concentrations in Herring Gull Eggs, 1997-2007: A) Significant decline, PCBs, Strachan Island, St. Lawrence River (regression line included); B-D) Non-significant patterns: Mercury at Agawa Rocks, Lake Superior, dieldrin at Chantry Island, Lake Huron and mirex in Toronto Harbour, Lake Ontario, respectively. Source: Canadian Wildlife Service (see Study Areas and Methods).

Legacy Compounds

Results

Temporal Trends

To better establish the context of the recent 1997-2007 analysis, results of the regression analysis of the eight contaminants from 1974 (or from when first analyzed) to 2007 are presented first. In that analysis, there were significant ($P<0.001$) declining regressions for 95% (114 of 120) of the contaminant-site comparisons. All 15 regressions, for each of PCBs (1:1), DDE. mirex, dieldrin, HCB, and heptachlor epoxide (HE), showed significant declines over the 33-year period (see Figure 9-42 for a typical example). Fourteen of 15 regressions for OCS and eight of 15 for mercury also showed significant declines. These results are very similar to those reported in 2007 and in that sense they represent very little "new" information.

If one compares contaminant concentrations in herring gull eggs from 2007 with those from 2005 (Table 9-2), the most recent data in the 2006 GLBTS Progress Report,[73] there are an overwhelming number (61/64, 95.3%) of small increases or no changes in concentration; only three comparisons declined: PCBs in Lake Superior, Lake Michigan and the Niagara River. When comparing 2005 with 2003 (data not shown), most changes were in the other direction; they increased slightly. Therefore, to better assess recent data (and their variability) on contaminant trends in herring gull eggs, we also conducted a regression analysis on data from the last 10 years available, 1997 to 2007. When these data were examined, only 5.8% (7 of 120) of the

[73] Ibid.

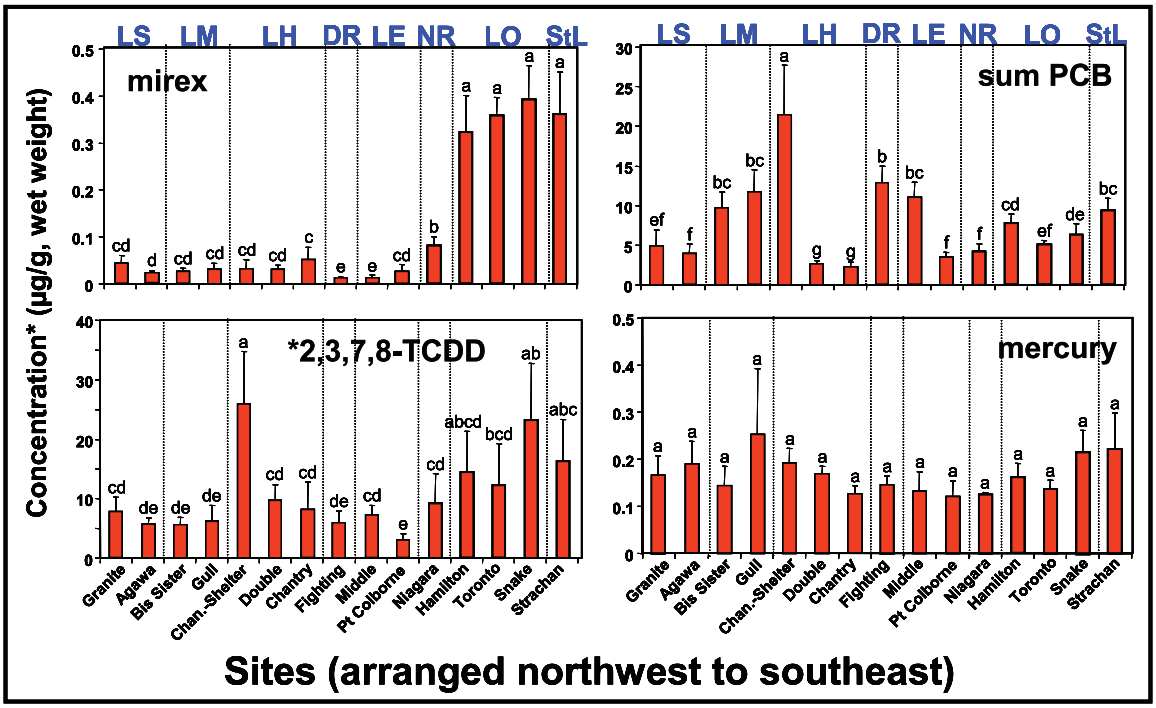


Figure 9-44. **Spatial Distribution of Contaminant Concentrations of Four Compounds among the 15 Herring Gull Egg Monitoring Sites, 2003-2007. *Measured in pg/g wet weight, 2003-2005. Source: Canadian Wildlife Service (see Study Areas and Methods).**

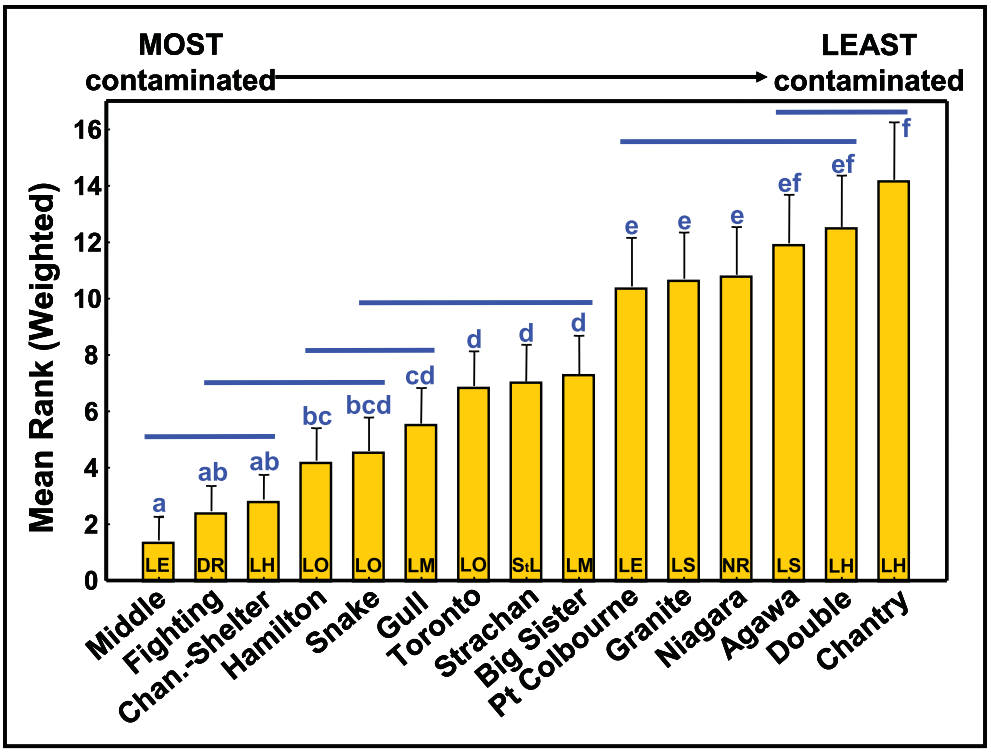


Figure 9-45. **Spatial Rankings of the 15 Herring Gull Egg Monitoring Sites From Most to Least Contaminated, 2003-2007. Source: Canadian Wildlife Service (see Study Areas and Methods).**

contaminant-site comparisons showed significant regressions ($P < 0.001$). The significant regressions were for PCBs at six locations and DDE at one (see Figure 9-43 for typical examples). This analysis suggests that, with the seven exceptions, mainly PCBs, there has been virtually no significant decline in concentrations of most legacy contaminants in gull eggs in the last 10 years.

A relatively recently recognized group of contaminants in the Great Lakes whose trend does not fit that portrayed by the legacy contaminants above are the polybrominated diphenyl ethers (PBDEs). Though their appearance in the Great Lakes ecosystem is relatively recent, Canadian Wildlife Service researchers have been able to analyze for them in archival herring gull eggs which have been maintained in the National Wildlife Specimen Bank. In the early 2000s, Norstrom et al. (2002) showed a continuously increasing trend for PBDEs in gull eggs starting in the early 1980s. More recently, Gauthier et al. (2008) have shown that lower brominated congeners have stablilized or declined, while highly brominated congeners, especially decabromodiphenyl ether (BDE-209) have increased. The Canadian Wildlife Service will continue to track these contaminants.

Spatial Trends

With 15 sample sites distributed among all five Great Lakes and three connecting channels, the Herring Gull Egg Monitoring Program also lends itself very well to a spatial analysis of contaminant concentrations. In the 2006 GLBTS Progress Report,[74] results from 1998 to 2002 were presented in a spatial context. For this report, we have conducted the spatial analysis on data from 2003 to 2007, i.e. updating the previous report by 5 years. The spatial distribution for mirex, sum PCBs, 2,3,7,8-TCDD, and mercury among the 15 herring gull sites is shown in Figure 9-44.

For an overall assessment of all 15 sites together, we used a weighted ranking scheme (see Study Areas and Methods), where concentrations of PCBs, sum DDT, and 2,3,7,8-TCDD contributed the most (83.5%, 9.5%, 5.0%, respectively) to this process. Gull eggs from Middle Island (western Lake Erie), Fighting Island (Detroit River) and Channel-Shelter Island (Saginaw Bay, Lake Huron) ranked as the three most contaminated sites. Eggs from Agawa Rocks (eastern Lake Superior), Double Island (North Channel, Lake Huron) and Chantry Island (southern Lake Huron) ranked as the three least contaminated sites (Figure 9-45, Table 9-3) (Weseloh et al., 2006; S. deSolla, unpubl. data).

Discussion

The major finding represented in this analysis is the near universal lack of a significant decline in concentrations of nearly all (legacy) contaminants measured in gull eggs at these 15 sites during the last 10 years, 1997-2007. The only exception to this was PCBs at several sites and DDE at one site; they continued to decline significantly. A slow-down in the rate of decline of contaminant concentrations has been noticed in fish since the late 1980s (Stow et al., 1995; DeVault et al., 1996; and Hickey et al., 2006) and has been addressed more recently (Bhavsar et al., 2007). Pekarik and Weseloh (1998) analyzed herring gull eggs from 1974 to 1995 by change-point regression and showed that only 19% of 143 contaminant-site comparisons showed a slower rate of decline in recent years. Also, in a short-term regression analysis, just over half of the contaminant comparisons that were significantly declining in the early 1980s had slowed in their rate of decline in the early 1990s.

The contaminant concentrations and their spatial and temporal trends shown in herring gull eggs presented in this section follow, to some extent, the data from fish monitoring programs in the

Great Lakes (Carlson and Swackhamer, 2006; Murphy et al., 2006). For example, DDE values were greatest in samples from Lake Michigan, mirex and OCS were greatest in Lake Ontario, and mercury values did not vary significantly at sites from across the Great Lakes water bodies. The gull data differ from the fish data in that most of the sites that had the greatest concentrations of various contaminants were located in the western Lake Erie to Saginaw

[74] USEPA and Environment Canada. 2007. Great Lakes Binational Toxics Strategy Progress Report, February 2007. Available at http://www.epa.gov/glnpo/bns/reports/2006glbtsprogressreport.pdf.

Table 9-3. **Mean Weighted Rank of Each Site, 2003-2007 (arranged from most to least contaminated) and Range in Rank (1 = most, 15 = least contaminated site).** [75]

Colony	Mean weighted rank	Least contaminated rank for this site	Most contaminated rank for this site	Homologous groups
Middle Island	1.4	12	1	A
Fighting Island	2.5	15	2	AB
Channel Shelter Island	2.9	11	1	AB
Hamilton Harbour	4.3	12	3	BC
Snake Island	4.6	8	1	BCD
Gull Island	5.6	13	1	CD
Leslie Street Spit	6.9	8	1	D
Strachan Island	7.1	14	2	D
Big Sister Island	7.4	15	1	D
Port Colborne	10.4	15	10	E
Granite Island	10.7	12	3	E
Niagara River	10.9	13	2	E
Agawa Rock	12.0	13	1	EF
Double Island	12.6	14	6	EF
Chantry Island	14.2	15	8	F

The ranks were weighted with a measure of contaminant toxicity using the ratio between mean egg concentrations of each compound and the corresponding fish flesh criteria for the protection of piscivorous wildlife (Newell et al., 1987).

* In or within herring gull feeding range of an Area of Concern.

1 Colonies with the same letter are not significantly different (SNK test, $\alpha = 0.05$).

[75] Data updated from Weseloh et al., 2006.

Bay (southwest Lake Huron) corridor and most of the sites with the least contaminated gull eggs were in eastern and northern Lakes Huron and Lake Superior. For the fish data, the areas of greatest and least contamination were Lake Michigan and Lake Superior, respectively. Part of the difference for the least contaminated lake may be because many of the herring gulls that breed on Lake Superior spend the winter on the lower lakes or the south end of Lake Michigan (Hebert 1998), where they are exposed, temporarily, to more elevated contaminant concentrations.

Fish and gull research both show that the food webs in the Great Lakes are changing due to a number of factors, including the presence of non-native species (Hebert et al., 2006, 2008b, 2009b; Hebert and Weseloh, 2006; Murphy et al., 2006). The net result of this is that, in some cases, predators may now be consuming prey from different food webs than in the past. This can have important impacts on contaminant trend interpretation (Hebert and Weseloh, 2006, Ismail et al., 2009). For example, feeding lower in the food web will reduce exposure of predators to biomagnifying contaminants, possibly leading to over-estimates of declines in contaminant availability in the environment. Conversely, changes in diet leading to increased contaminant exposure may give a false impression of increases in environmental contaminant availability. Clearly, altered food webs and concomitant impacts on the diets of biomonitoring species need to be considered when interpreting contaminant temporal trends.

Future studies of the Herring Gull Egg Monitoring Program include continuation of the annual monitoring and tracking of spatial and temporal contaminant trends and further research into the use of stable isotopes, fatty acids, and other ecological tracers to give more detailed meaning to the trends. Recent research is also highlighting the value of the program in identifying trends (Gauthier et al., 2007, 2008, 2009; Gebbink et al., 2009) and sources of emerging contaminants (Hebert et al., 2009a).

Acknowledgements

We are appreciative of Abde Idrissi and his Lab Services staff at the National Wildlife Research Centre (NWRC) for preparation and analysis of the gull eggs for legacy POPs and early analysis of emerging PBDE contaminants. For emerging POPs, later PBDE studies, non-PBDE BFRs and PFCs, we thank the staff and students in the Organics Research Group at NWRC. Clive Hodder provided field assistance; Craig Hebert provided comments on a previous version of the manuscript.

References

Bhavsar, S.P., D.A. Jackson, A. Hayton, E.J. Reiner, T. Chen and J. Bodnar. 2007. Are PCB levels in fish from the Canadian Great Lakes still declining? J. Great Lakes Res. 33:592-605.

Bishop, C.A., Weseloh, D.V., Burgess, N.B., Norstrom, R.J., Turle, R. and Logan, K.A. 1992. An atlas of contaminants in eggs of colonial fish-eating birds of the Great Lakes (1970-1988): Vols. I and II. Canadian Wildlife Service, Technical Report Series, No.153.

Carlson, D. L. and D.L. Swackhamer. 2006. Results from the U.S. Great Lakes fish monitoring program and effects of lake processes on bioaccumulative contaminant concentrations. J. Great Lakes Res. 32:370-385.

Chu, S. and Letcher, R.J. 2009. Linear and Branched Perfluorooctane Sulfonate Isomers in Technical Product and Environmental Samples by In-Port Derivatization-Gas Chromatography-Mass Spectrometry. *Analytical Chemistry*. 81: 4256-4262.

deSolla, S. Canadian Wildlife Service. Unpublished data.

DeVault, D.S., Hesselberg, R., Rodgers, P.W., and Feist, T.J, 1996. Contaminant trends in lake trout and walleye from the Laurentian Great Lakes. *J. Great Lakes Res.* 22:884-895.

DiMao, J., Pekarik, C. and D.V. Weseloh. 1999. Contaminant levels in Herring Gull eggs from Lake Erie and the Detroit and Niagara Rivers: change-point regression analysis, 1974 to 1996. In State of Lake Erie - Past, Present and Future. M. Munawar, T. Erdsall and I.F. Munawar, eds., pp. 399-416.

Ewins, P.J., Weseloh, D.V. and P. Mineau. 1992. Geographical distribution of contaminants and productivity measures of herring gulls in the Great Lakes: Lake Huron 1980. *J. Great Lakes Res.* 18:316-330.

Gauthier, L.T., Hebert, CE., Weseloh, D.V. and Letcher, RJ. 2007. Current-Use Flame Retardants in the Eggs of Herring Gulls (Larus argentatus) from the Laurentian Great Lakes. 2007. *Environmental Science and Technology*. 41:4561-4567.

Gauthier, L.T., Hebert, CE., Weseloh, D.V. and Letcher, RJ. 2008. Dramatic Changes in the Temporal Trends of Polybrominated Diphenyl Ethers (PBDEs) in Herring Gull Eggs from the Laurentian Great Lakes: 1982-2006. *Environmental Science and Technology*. 42: 1524-1530.

Gauthier, L.T., Potter, D, Hebert, CE. and Letcher, RJ. 2009. Temporal Trends and Spatial Distribution of Non-polybrominated Diphenyl Flame Retardants in the Eggs of Colonial Populations of Great Lakes Herring Gulls. *Environmental Science and Technology*. 43: 312-317.

Gauthier, L.T. and Letcher, RJ. 2009. Isomers of Dechlorane Plus flame retardant in the eggs of herring gulls (Larus argentatus) from the Laurentian Great Lakes of North America: Temporal changes and spatial distribution. *Chemosphere*. 75: 115-120.

Gebbink, W.A., Hebert, CE. and Letcher, RJ. 2009. Perfluorinated Carboxylates and Sulfonates and Precursor Compounds in Herring Gull Eggs from Colonies Spanning the Laurentian Great Lakes of North America. *Environmental Science and Technology*. 43: 7443-7449.

Gebbink, W.A. and Letcher, R.J. 2010. Linear and Branched Perfluorooctane Sulfonate Isomers and Patterns in Herring Gull Eggs from Colonial Sites across the Laurentian Great Lakes. *Environmental Science and Technology*. Submitted Feb. 2010.

Hebert, C.E. 1998. Winter severity affects migration and contaminant accumulation in northern Great Lakes herring gulls. *Ecological Applications*. 8:669-679.

Hebert, C.E., Norstrom, R.J. and Weseloh, D.V. 1999. A quarter century of environmental surveillance: The Canadian Wildlife Service's Great Lakes Herring Gull monitoring program. *Environmental Review* 7:147-166.

Hebert, C.E. and D.V. Weseloh. 2006. Adjusting for temporal change in trophic position results in reduced rates of contaminant decline. *Environmental Science and Technology*. 40: 5624-5628.

Hebert, C.E., M.T. Arts, and D.V. Weseloh. 2006. Ecological tracers can quantify food web structure and change. *Environmental Science and Technology*. 40: 5618-5623.

Hebert, C.E., D.V. Weseloh, T. Havelka, C. Pekarik, J.L. Shutt, M. Shieldcastle and F. Cuthbert. 2008a. Lake Erie colonial waterbirds, 1974-2002: Trends in populations, contaminant levels, and stable isotope indicators of diet. Pp. 497-528 In: Checking the Pulse of Lake Erie eds. M. Munawar and R. Heath. Ecovision World Monograph Series. Aquatic Ecosystem Health and Management Society, Burlington, ON, Canada.

Hebert, C.E., D.V.C. Weseloh, A. Idrissi, M.T. Arts, R. O'Gorman, O.T. Gorman, B. Locke, C.P. Madenjian, and E.F. Roseman. 2008b. Restoring piscivorous fish populations in the Laurentian Great Lakes causes seabird dietary change. *Ecology*. 89: 891-897.

Hebert, C.E., D.V. Weseloh., L.T. Gauthier, M.T. Arts, R.J. Letcher. 2009a. Biochemical tracers reveal intra-specific differences in the food webs utilized by individual seabirds. *Oecologia*.160: 15-23.

Hebert, C.E., D.V.C. Weseloh, A. Idrissi, M.T. Arts, and E.F. Roseman. 2009b. Diets of aquatic birds reflect changes in the Lake Huron ecosystem. *Aquatic Ecosystem Health and Management*. 12: 37-44.

Hickey, J.P., Batterman, S.A., and Chernyak, S.M. 2006. Trends in chlorinated organiccontaminants in Great Lakes trout and walleyes from 1970 to 1998. Arch. *Environ. Contam.Toxicol*. 50:97-110

Ismail, N., S.B. Gewurtz, K. Pleskach, D.M. Whittle, P.A. Helm, C.H. Marvin and G.T. Tomy. 2009. Brominated and chlorinated flame retardants

in Lake Ontario, Canada, lake trout (Salvelinus namaycush) between 1979 and 2004 and possible influences of food-web changes. *Environ. Toxicol. Chem*. 28:910-920.

Jermyn-Gee, K., C. Pekarik, T. Havelka, G. Barrett, and D.V. Weseloh. 2005. An atlas of contaminants in the eggs of fish-eating colonial birds of the great lakes (1998-2001). Volumes I. & 2. Accounts by location and chemical. Canadian Wildlife Service, Technical Report Nos. 417 & 418. 341 & 142 pp

Letcher, R.J. and Chu, S. 2010. Determination of tetrabromobisphenol-S and tetrabromobisphenol-A derivatives by liquid chromatography-atmospheric pressure photoionization-tandem mass spectrometry. *Environmental Science and Technology*. Submitted March 2010.

Mineau, P., Fox, G.A., Norstrom, R.J., Weseloh, D.V., Hallett, D., Ellenton, J.A. 1984. Using the Herring Gull to monitor levels and effects of organochlorine contamination in the Canadian Great Lakes. In Toxic Contaminants in the Great Lakes. Edited by J.O. Nriagu and M.S. Simmons. John Wiley and Sons, Inc. pp. 425-52.

Murphy, E., C. MacEachen, D.M. Whittle, M.J. Keir and J.F. Gorrie. 2006. Contaminants in whole fish. SOLEC Indicator # 121. Available at: http://www.solecregistration.ca/documents/0121%20Contaminants%20in%20Whole%20Fish%20(SOLEC%202006).pdf

Norstrom, R.J., Simon, M., Moisey, J., Wakeford, B., Weseloh, D.V. 2002. Geographic distribution (2000) and temporal trends (1981-2000) of polybrominated diphenyl ethers in Great Lakes Herring Gull eggs. *Environmental Science and Technology* 36: 4783-4789.

Pekarik, C. and D.V. Weseloh. 1998. Organochlorine contaminants in Herring Gull eggs from the Great Lakes, 1974-1995: Change point regression analysis and short-term regression. *Environmental Monitoring Assessment*. 53: 77-115.

Pekarik, C., Weseloh, D.V., Barrett, G.C., Simon, M., Bishop, C.A., Pettit, K.E. 1998. An atlas of contaminants in the eggs of fish-eating colonial birds of the Great Lakes (1993-1997): Vols I and II. Canadian Wildlife Service, Technical Report Series, No.321.

Pettit, K.E., Bishop, C.A., Weseloh, D.V., Norstrom, R.J. 1994. An atlas of contaminants in eggs of colonial fish-eating birds of the Great Lakes (1989-1992): Vols I and II. Canadian Wildlife Service, Technical Report Series, No.193.

Rice, W. R. 1989. Analyzing tables of statistical tests. *Evolution* 43: 223-225.

Stow, C.A., S.R. Carpenter, L.A. Eby, J.F. Amrhein and R.J. Hesselberg. 1995. Evidence that PCBs are approaching stable concentrations in Lake Michigan fishes. *Ecol. Appl*. 5:248-260.

Ucán-Marín, F., Arukwe, A., Mortenson, A., Gabrielsen, G.W., Fox, G.A. and Letcher, R.J. 2009. Rcombinant Transthyretin Purification and Competitive Binding with Organoalogen Compounds in Two Gull Species (Larus argentatus and Larus hyperboreus). 2009. *Toxicological Sciences*. 107 (2): 440-450.

Ucán-Marín, F., Arukwe, A., Mortenson, A., Gabrielsen, G.W., Letcher, R.J. 2010. Recombinant albumin transport protein from gull species (Larus argentatus and hyperboreus) and human: Chlorinated and brominated contaminant binding and thyroid hormones. *Environmental Science and Technology*. 44: 497-504.

Weseloh, D.V., C. Pekarik, R. Joos, J.F. Farquhar, J.L. Shutt, T. Havelka, I. Mazzocchi, G. Barrett, R. McCollough, R.L. Miller and A. Mathers. 2003. Monitoring Lake Ontario's waterbirds: contaminants in Herring Gull eggs and population changes in the Lake's nearly 1,000,000 colonial waterbirds. In: Munawar, M. (ed.). State of Lake Ontario (SOLO) – Past, Present and Future. pp 597-631. *Aquatic Ecosystem Health & Management Society*.

Weseloh, D.V.C., R.A. Faber and C. Pekarik. 2005. Temporal and spatial trends of organochlorine contaminants in herring gull eggs from Lake Michigan. Pp: 393-437. In: T. Edsall and M. Munawar [eds.] 2005. State of Lake Michigan: Ecology, Health, and Management. Ecovision World Monograph Series. Aquatic Ecosystem Health and Management Society. Goodword Books Pvt.Ltd, New Delhi, India.

Weseloh D.V., Pekarik C. and S.R. de Solla. 2006. Spatial patterns and rankings of contaminant concentrations in herring gull eggs from 15 sites in the Great Lakes and connecting channels, 1988-2002. *Environ Monitor Assess*. 113: 265-284.

Won, H.T., M.J. Mulvihil and B.J. Wakeford. 2001. Multiresidue methods for the determination of chlorinated pesticides and polychlorinated biphenyls (PCBs) in wildlife tissues by gas chromatography/ mass spectrometry. Technical Report Series Number 335E. Canadian Wildlife Service, Headquarters, Hull, Quebec, Canada.

Contaminant Trends in Mussels

Photo: Mussels, unknown location
National Oceanic and Atmospheric Administration, Center for Coastal Monitoring and Assessment.
Courtesy of US EPA GLNPO.

Mussel Watch Program

Submitted by
Kimani Kimbrough, Ed Johnson, Dennis Apeti and Gunnar Lauenstein
National Oceanic and Atmospheric Administration

Background

Founded in 1986, the Mussel Watch Program is one of the longest-running national monitoring programs for estuarine and coastal pollutants in the U.S. Mussel Watch was designed to monitor the status and trends of local chemical contamination of U.S. coastal waters, including the Great Lakes, and is based on yearly and decadal collection and analysis of bivalves (oysters and mussels) and sediment, respectively. Today the program monitors over 150 analytes. Initially, 145 test sites were established along the coasts, with additional sites in the Great Lakes added in 1992. The program has expanded over time to include nearly 300 monitoring sites (Figure 9-46).[76] Mussel Watch also stores samples in a specimen bank for future use, such as tracking trends of new and emerging contaminants of concern.

The information presented here details the status and trends of chemical concentrations in the Great Lakes between the years 1993 and 2008, and compares them to national concentrations. It was not until the 2009 summer sampling in the eastern Great Lakes that US EPA AOCs were first sampled; those data will become available in the near future. Our results showed few trends for trace metals. Many organic contaminants showed decreasing concentrations, probably resulting from state and federal regulation.

Bivalves are sessile organisms that filter particles and accumulate contaminants from water; making them good integrators of contaminants in a given area (Berner et al., 1976; Farrington et al., 1980; Farrington, 1983; and Tripp and Farrington, 1984), and surrogates for environmental quality (Roesijadi et al., 1984; Sericano, 1993). Using bivalves for monitoring adds another dimension beyond abiotic environmental monitoring because the presence of contaminants in bivalves is evidence of bioaccumulation. Additionally, contaminants found in bivalves may also be found in fish at higher concentrations as a result of consumption by organisms higher on the food chain.

Because one single species of mussel or oyster is not common to all coastal regions, a variety of species are collected to gain a national perspective. A target species is identified for each site based on abundance and ease of collection.

[76] Lake Superior was not included in the initial sites sampled in the Great Lakes but has been added to the monitoring program. However, current sampling in Lake Superior is not as extensive as in the other Great Lakes because zebra mussel densities are lower in Lake Superior.

Figure 9-46. NOAA Mussel Watch Program Monitoring Sites. Source: NOAA Mussel Match Program.

Mussels (*Mytilus* species) are collected from the North Atlantic and Pacific coasts, oysters (*Crassostrea virginica*) from the mid-Atlantic (Delaware Bay) southward and along the Gulf Coast, and zebra and quagga mussels (*Dreissena* species), both invasive species, are collected from sites in the Great Lakes (Figure 9-46). Mussel Watch began monitoring the Great Lakes in 1992, within a few years of the introduction of the invasive zebra mussels which first appeared in 1988 in Lake St. Clair (Hebert et al., 1989).

Where possible, sites were selected to coincide with historical mussel and oyster monitoring locations from other programs, such as the US EPA's Mussel Watch sites that were sampled from 1976 to 1978 (Goldberg et al., 1983), and to complement sites sampled through state programs, such as the California Mussel Watch Program (Martin, 1985). Hot spots were initially avoided; however, as a result of increased coordination with stakeholders, monitoring at polluted areas, such as US EPA AOCs, has been initiated by the program.

Sediments described in this report are used to compare Great Lakes contaminant measurements to the entire nation. Sediment samples are collected from Mussel Watch sites approximately once every 10 years, when new sites are established, or following extreme events such as oil spills. Bivalve and sediment sites are taken from areas in close proximity to one another. The top 3 cm of sediments, representing recent deposition, are used in this analysis. Three sediment grabs are collected from three stations and composited. Sediment collection sites are located as near as possible to, but generally not more than, 2 km from the bivalve site, and in low energy depositional areas.

Chemical concentration trends were assessed by correlating contaminant concentrations with time. Spearman's rank correlation was used to evaluate whether concentrations co-varied predictably as a function of time (Zar, 1999). That is, as time progressed from the beginning of our monitoring records (1992, Great Lakes) to our most current data (2008), did the concentration of contaminants also progress in an increasing or decreasing manner? The Spearman's rank correlation procedure is a nonparametric technique that is free of assumptions about concentrations being normally distributed with a common variance about sites. The variables used for the Spearman's test were year and site

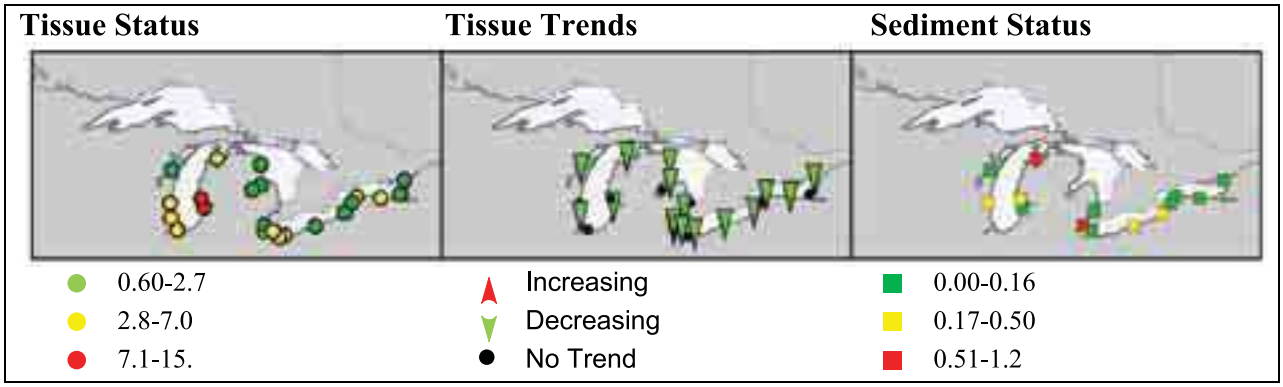


Figure 9-47. **Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Trends (Tissue Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for Aldrin/Dieldrin. All concentrations reported in ng/g dry wt. Source: NOAA Mussel Watch Program.**

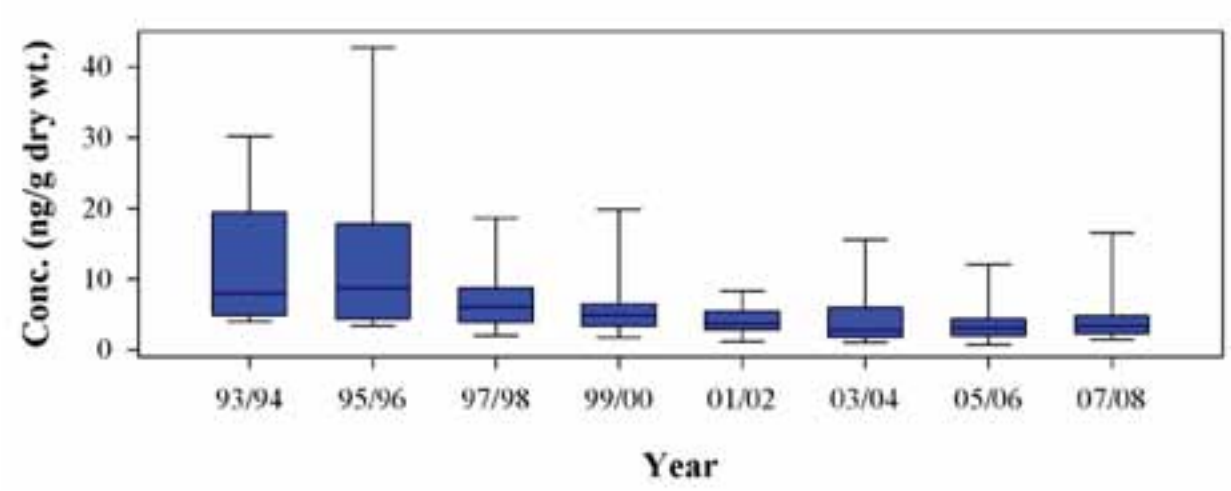


Figure 9-48. **Aldrin/Dieldrin Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.**

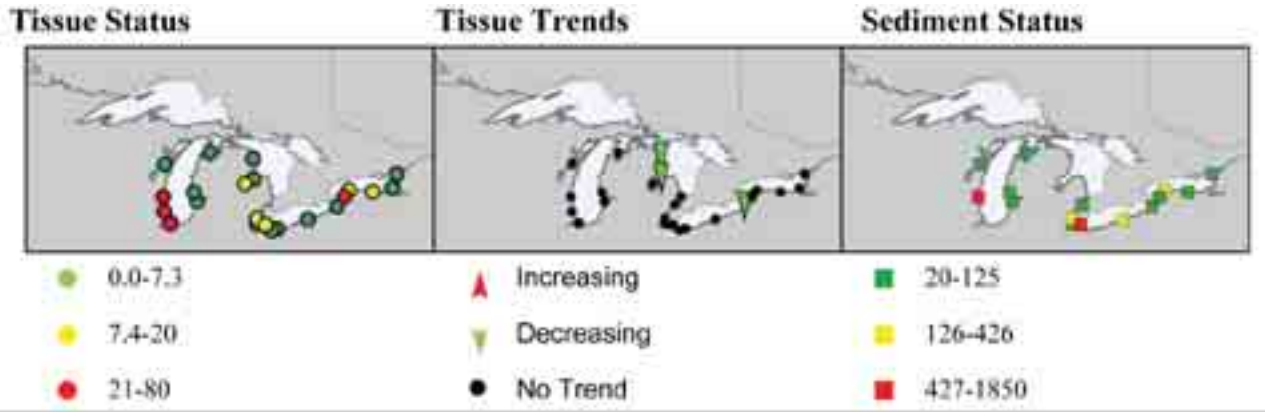


Figure 9-49. Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Tissue Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for B(a)P. All concentrations reported in ng/g dry wt. Source: NOAA Mussel Watch Program.

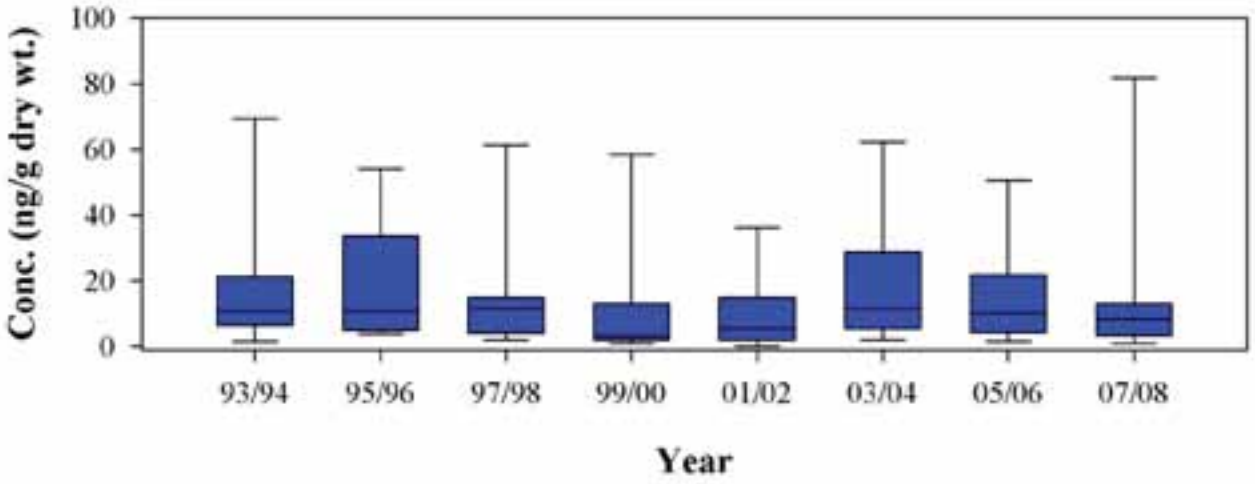


Figure 9-50. B(a)P Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.

concentration rank median (n = 8). Concentration was standardized by ranking to allow for inter-species comparison. All comparisons of sediment and tissue concentrations presented in this document are derived from the Mussel Watch data set.

Aldrin/Dieldrin

- The highest levels of aldrin/dieldrin were found in Lake Michigan tissue samples (Figure 9-47).
- Decreasing trends were observed throughout the lakes in more than half the sites, while no increasing trends were found. This is similar to national trend results for aldrin/dieldrin (Kimbrough et al., 2008). Overall, trends have decreased to an asymptotic level in the Great Lakes (Figure 9-48).
- High and medium sediment concentrations in the Great Lakes are elevated relative to the national Mussel Watch median and mean (0 and 0.15 ng/g dry wt.). However, they are lower than the maximum national Mussel Watch sediment value (8.5 ng/g dry wt.).

Benzo(a)pyrene [B(a)P]

- For both tissue and sediment, the highest B(a)P values occurred near urban areas (Figure 9-49).
- Most sites showed no trend; however, there were three decreasing trends and no increasing trends (Figure 9-49). Year-to-year variability for B(a)P supports the fact that sources of B(a)P and other PAHs still exist in the Great Lakes (Figure 9-50).
- The lowest concentrations found at Great Lakes sediment sites are above the national sediment median (14.7 ng/g dry wt.) but an order of magnitude lower than the highest national concentration (19,700 ng/g dry wt.). The national mean of 209 ng/g dry wt. is in the range of Great Lakes values.

Chlordane

- Elevated levels of chlordane were observed in sediment and tissue samples from urban and agricultural sites (Figure 9-51).
- There were only four decreasing trends, at tissue sites associated primarily with agricultural areas (Figures 9-51 and 9-52). Most sites showed no trend.
- All Great Lakes sediment concentrations were higher than the national median (0.04 ng/g dry wt.). The highest Great Lakes sediment concentrations were all higher than the national mean but several times lower than the highest national concentration (0.36 and 11.81 ng/g dry wt., respectively).

DDT (+DDD+DDE)

- Elevated levels of DDT (+DDD+DDE, or dichlorodiphenyldichloroethane and dichlorodiphenyldichloroethylene) are distributed throughout the Great Lakes in both mussels and sediment (Figure 9-53).
- Nine sites showed decreasing tissue concentrations, and an overall decreasing trend is exhibited for the Great Lakes (Figures 9-53 and 9-54).
- Elevated sediment concentrations in Great Lakes Mussel Watch samples are above the national mean but orders of magnitude lower than the national Mussel Watch maximum (2.8 and 107 ng/g dry wt., respectively).

Hexachlorobenzene (HCB)

- Elevated levels of HCB in tissue samples are associated primarily with urban/industrial areas (Figure 9-55).
- At most sites, tissue measurements showed no trend (Figure 9-55).
- Stable concentrations across all Great Lakes sites (Figure 9-56) are consistent with stable levels in HCB air and water releases reported to US EPA's TRI from 1990 to 2005 (US EPA, 2007).
- Sediment levels of HCB in the Great Lakes are high when compared to national median and mean concentrations of 0.03 and 0.53 ng/g dry wt., respectively. The highest Mussel Watch

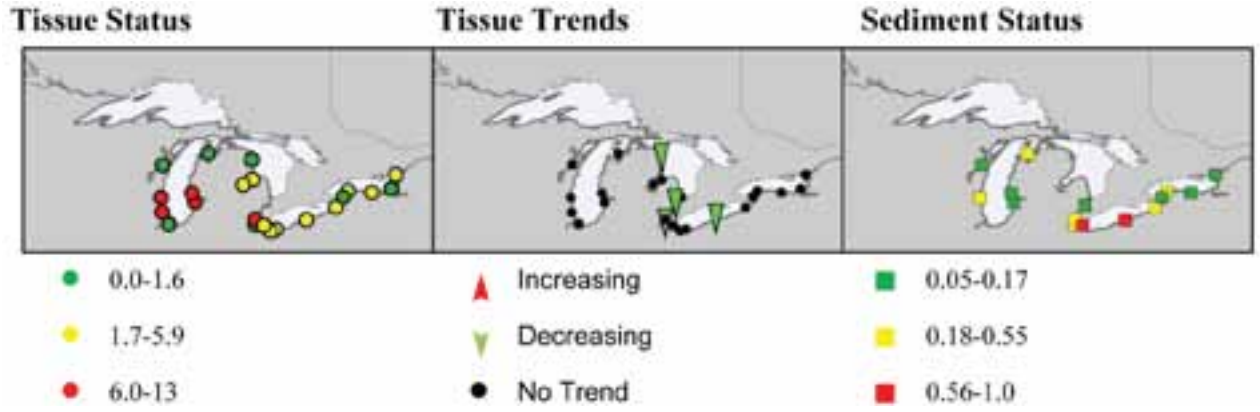


Figure 9-51. Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Tissue Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for Chlordane. All concentrations reported in ng/g dry wt. Source: NOAA Mussel Watch Program.

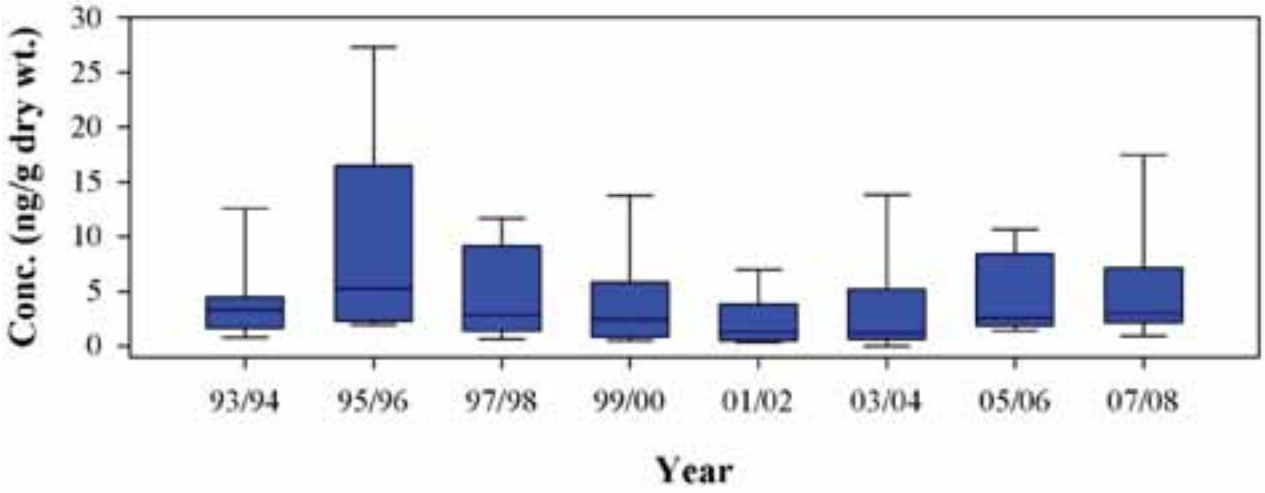


Figure 9-52. Chlordane Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.

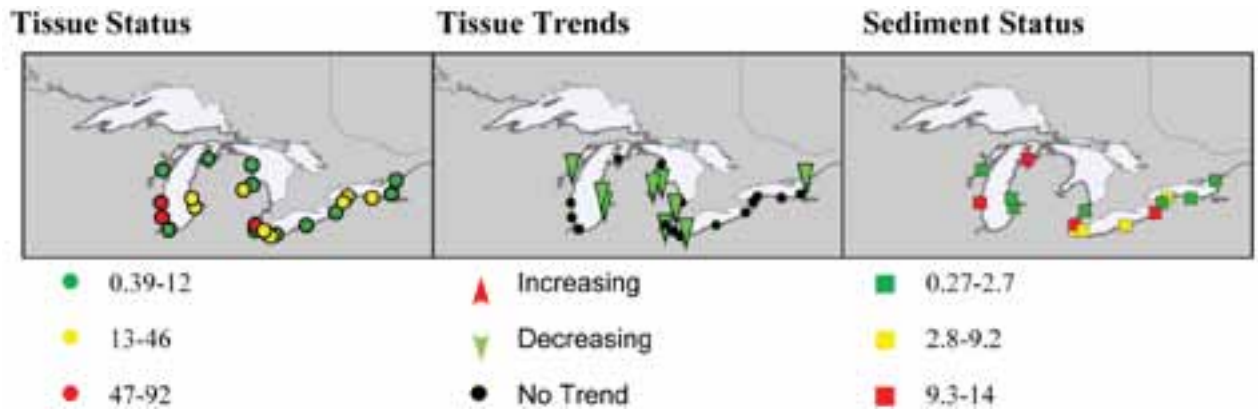


Figure 9-53. **Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Tissue Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for DDT (+DDD + DDE). All concentrations reported in ng/g dry wt. Source: NOAA Mussel Watch Program.**

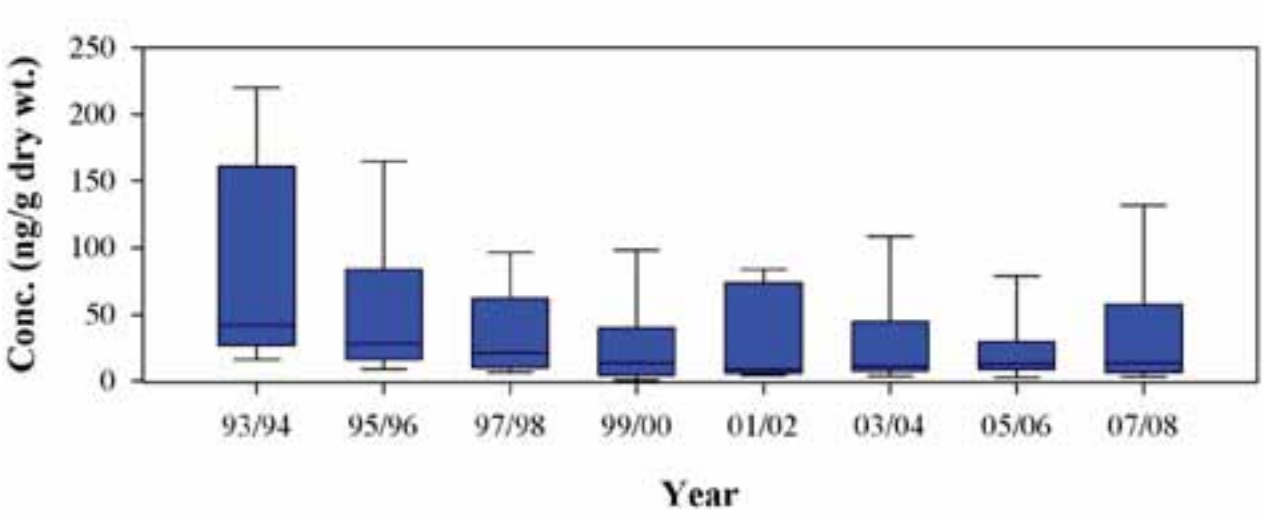


Figure 9-54. **DDT (+DDD + DDE) Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.**

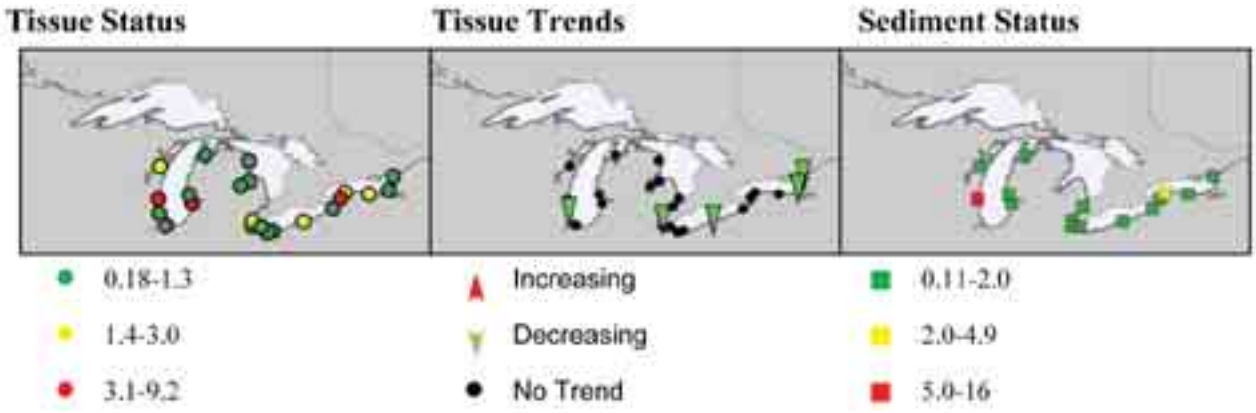


Figure 9-55. Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for HCB. All concentrations reported in ng/g dry wt. Source: NOAA Mussel Watch Program.

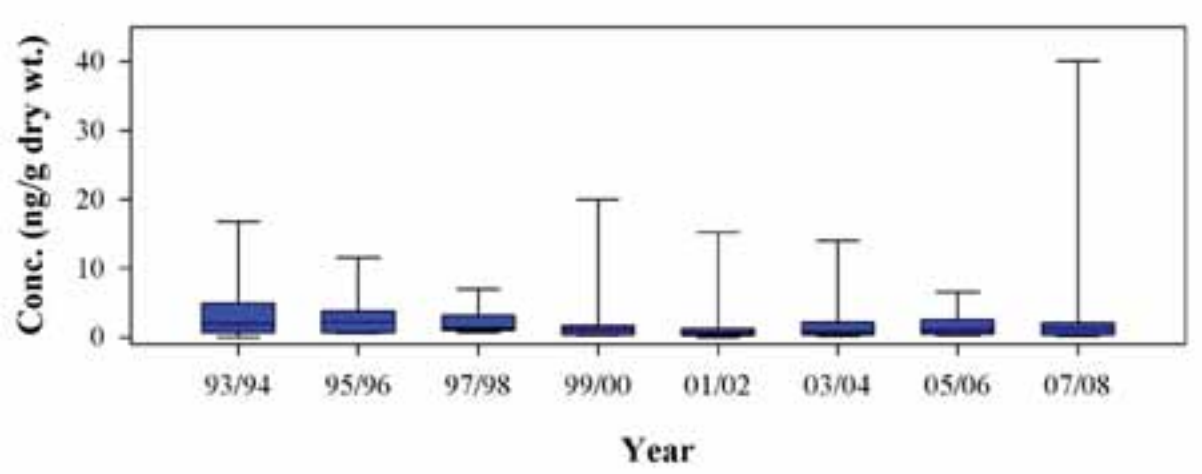


Figure 9-56. HCB Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.

sediment concentration (16 ng/g dry wt.) occurs in the Great Lakes (Figure 9-55).

Mirex

- High levels of mirex in both sediment and tissue occur in Lake Ontario (Figure 9-57). The distribution of mirex in Lake Ontario is consistent with its history of manufacture in the region.
- Three of the four Lake Ontario sites, where concentrations were the highest, showed decreasing trends; all other sites throughout the lakes showed no trend (Figure 9-57).
- Elevated sediment mirex concentrations found in Lake Ontario are the highest Mussel Watch sediment measurements in the nation (3.5 ng/g dry wt.; Apeti and Lauenstein 2006). Low sediment concentrations in the Great Lakes are below the national mean (0.06 ng/g dry wt.).
- Across all Great Lakes Mussel Watch sites, mirex tissue levels have declined since 1993 (Figure 9-58).

PCBs

- Great Lakes PCB tissue concentrations range several orders of magnitude (Figure 9-59).
- Most sites showed no trend; however, overall, PCB concentrations in mussels appear to be higher in the early years than in more recent years (Figure 9-60).
- All Great Lakes PCB sediment concentrations are higher than the national median (1.0 ng/g dry wt.), with elevated concentrations all being higher than the national mean (8.14 ng/g dry wt.). The highest national Mussel Watch sediment concentration is 124 ng/g dry wt.

Cadmium

- Elevated levels of cadmium in tissue samples are distributed throughout the Great Lakes (Figure 9-61).
- Decreasing trends in tissue concentrations were observed uniformly throughout the Great Lakes (Figure 9-62).
- All Great Lakes sediment measurements are higher than the national median and mean of 0.16 and 0.27 µg/g dry wt., respectively. The highest cadmium sediment concentration in the nation occurs in the Great Lakes (2.24 µg/g dry wt.; Apeti et al., 2009).

Heptachlor (+Heptachlor Epoxide)

- Elevated concentrations of heptachlor in tissue samples occur in all of the Great Lakes (Figure 9-63).
- About one-third of the sites showed decreasing trends; no trends were observed in Lake Erie and southern Lake Huron (Figure 9-64); however, across all sites, more recent concentrations are lower than historic tissue concentrations (Figure 9-64).
- The highest Mussel Watch heptachlor sediment concentration occurs in the Great Lakes (2.0 ng/g dry wt.). Most of the Great Lakes measurements are above the national Mussel Watch mean (0.065 ng/g dry wt.).

Pentachlorobenzene

- Most tissue levels were below detection limits; only four sites were elevated (Figure 9-65).
- No trends were observed in Lake Michigan and Lake Huron; in contrast, most sites in Lake Erie and Lake Ontario showed a decreasing trend (Figure 9-65).
- Across Great Lakes Mussel Watch sites, pentachlorobenzene concentrations in mussel tissue have declined since 1997 (Figure 9-66).

Tributyltin (TBT)

- Elevated levels of TBT in mussels were highest in western Lake Erie and southern Lake Michigan (Figure 9-67).
- Most sites showed no trend, but decreasing trends were observed at three sites in southern Lake Michigan. Increasing trends in TBT concentrations were observed at two sites in

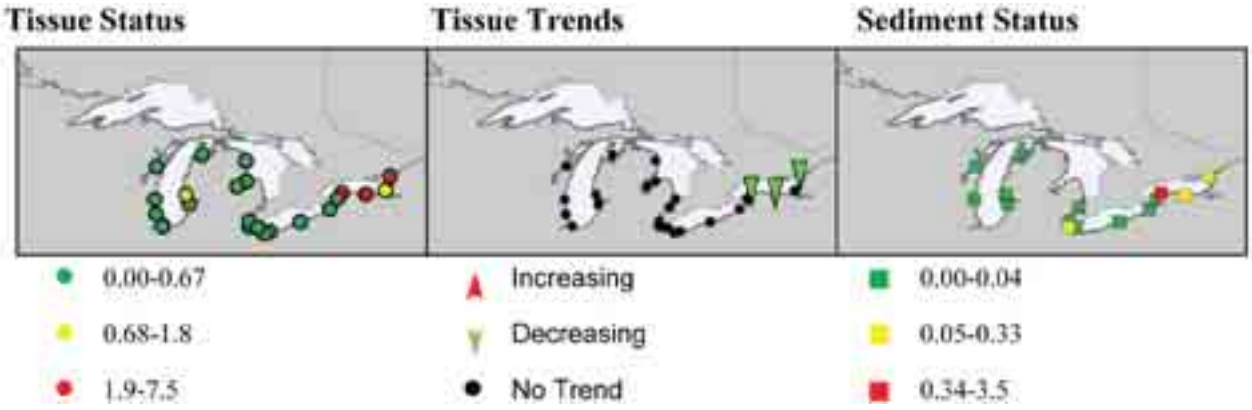


Figure 9-57. Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for Mirex. All concentrations reported in ng/g dry wt. Source: NOAA Mussel Watch Program.

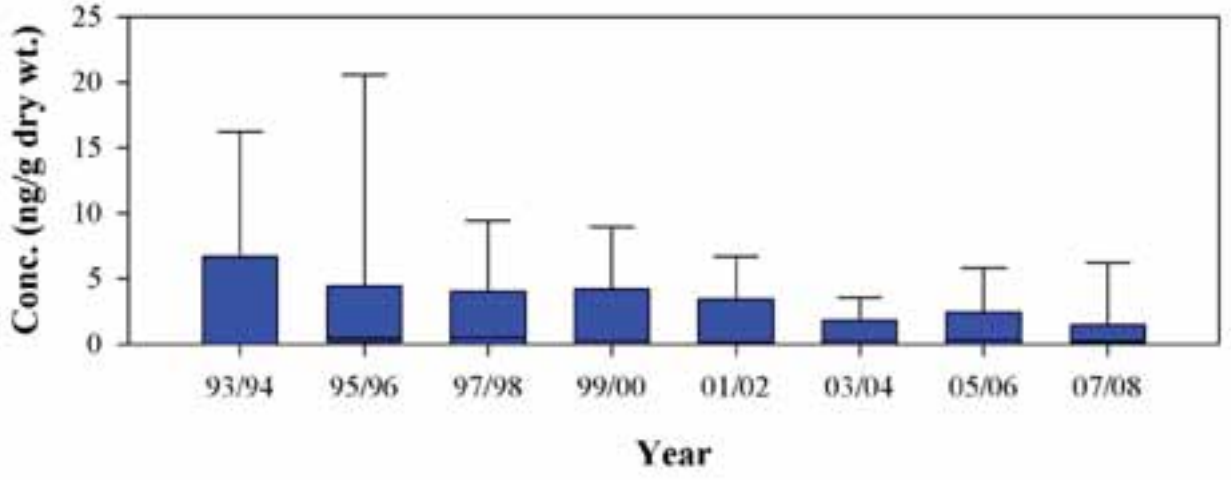


Figure 9-58. Mirex Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.

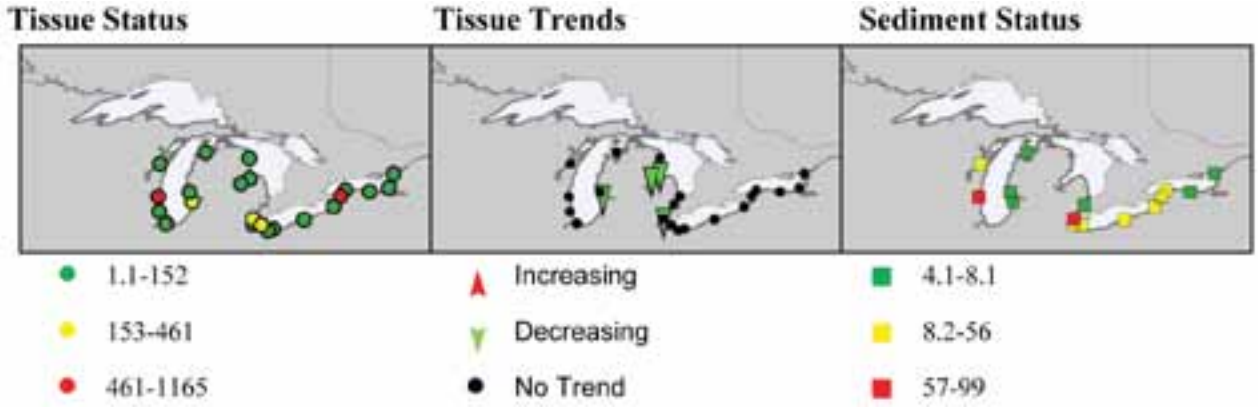


Figure 9-59. **Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for PCBs. All concentrations reported in ng/g dry wt. Source: NOAA Mussel Watch Program.**

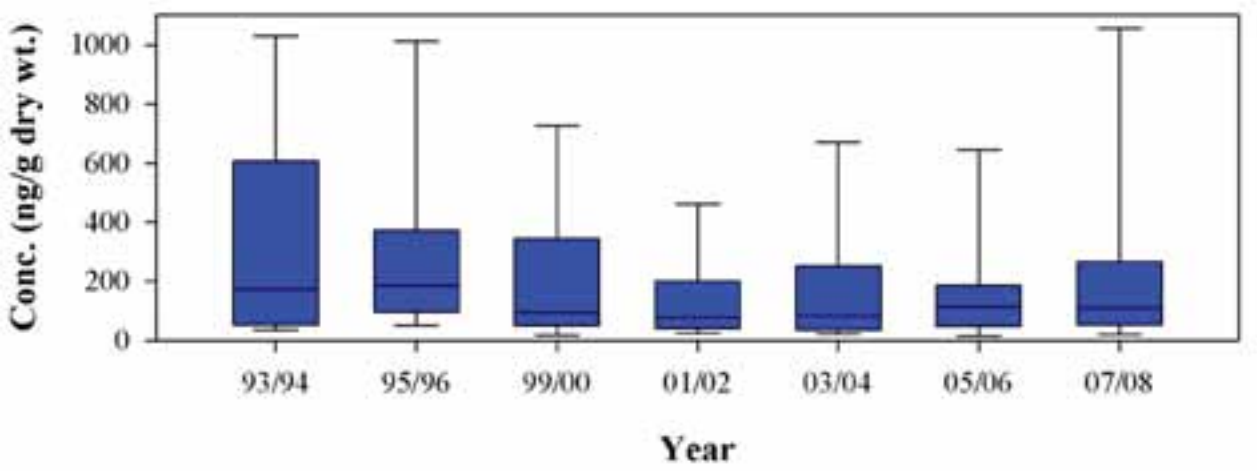


Figure 9-60. **PCB Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.**

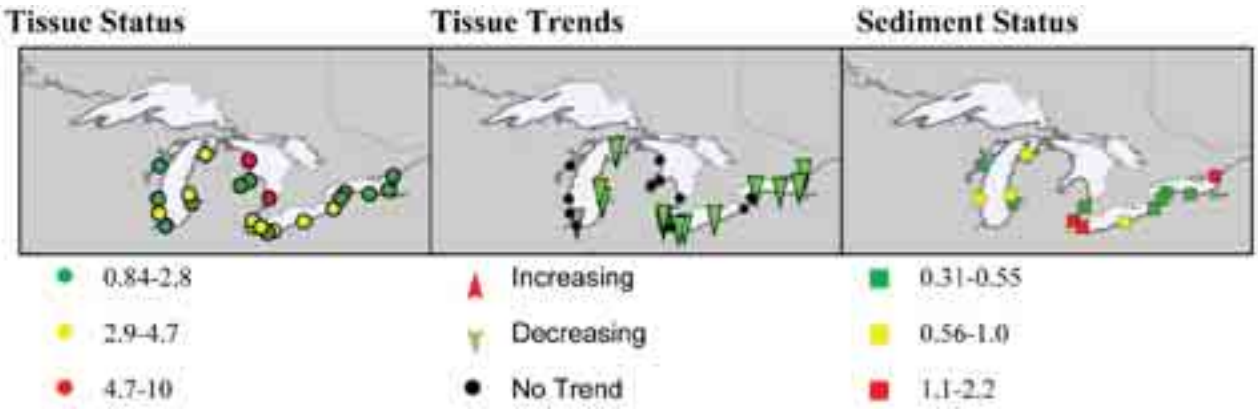


Figure 9-61. Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for Cadmium. All concentrations reported in µg/g dry wt. Source: NOAA Mussel Watch Program.

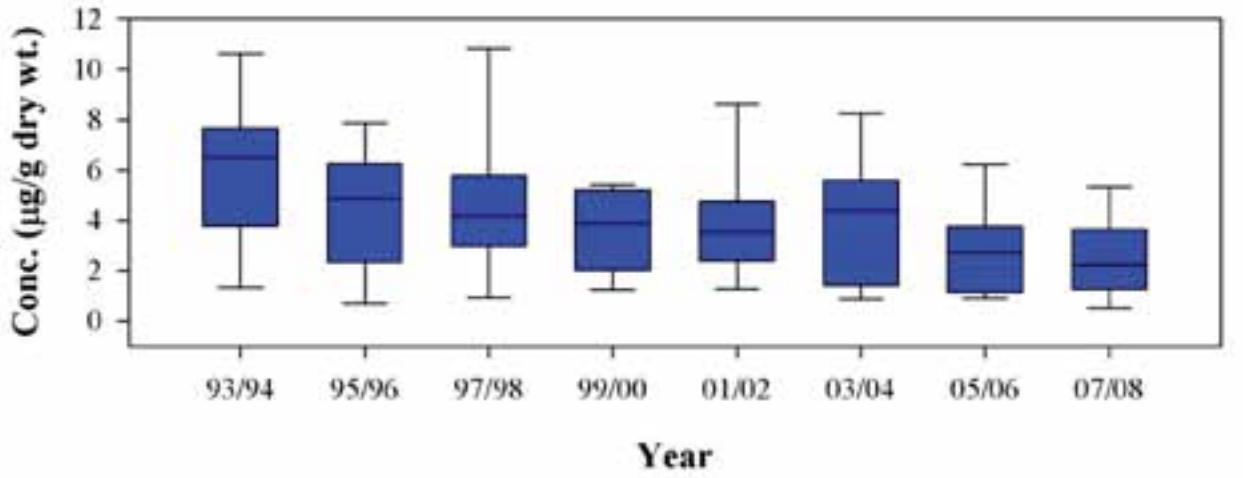


Figure 9-62. Cadmium Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.

western Lake Erie and may be associated with recreational boat use (Figure 9-67).

» Across all Great Lakes sites, Mussel Watch TBT tissue concentrations have declined considerably since 1993, largely the result of the phase-out of TBT compounds as an anti-fouling agent (Figure 9-68).

Mercury

» The highest levels of mercury were found in Green Bay, Thunder Bay, Saginaw Bay, Traverse Bay, and Black River Canal (Figure 9-69).

» Decreasing trends were observed at only three sites (Figure 9-69).

» High and medium mercury sediment levels were found in western Lake Erie at Stony Point and Reno Beach, Green Bay in Lake Michigan, and Lake Ontario near Olcott and Cape Vincent. Stony Point had the highest reported sediment mercury level in the country (0.68 µg/g dry wt).

» Across Great Lakes Mussel Watch sites, mercury tissue levels showed no increasing or decreasing trend (Figure 9-70).

Tetrachlorobenzene

» The highest tissue concentrations were observed in Lake Huron at Saginaw Bay and Lake Michigan at Milwaukee Bay but were below the national high of 18.8 ng/g dry wt.

» Most sites showed no trend; however, there were nine decreasing trends, primarily in Lakes Erie and Ontario, and no increasing trends (Figure 9-71).

» The highest sediment concentration measured in the Great Lakes was also the highest in the country (11.0 ng/g dry wt.).

» Across the Great Lakes, recent tetrachlorobenzene levels at Mussel Watch sites are lower than historic values (Figure 9-72).

Overall Findings

» Like national bivalve concentrations, tissue concentrations are higher than sediment concentrations.

» Nationally, sites are distributed in areas representative of ambient levels of contamination; therefore, hotspots are often avoided, though sites like Milwaukee were established in an AOC and therefore may indicate elevated contaminant levels, which may also be found once data for additional AOCs become available. This may result in Great Lakes concentrations that are higher than those found at the national level because of the high density of industry and urbanization in the Great Lakes or the slow water turnover rate in the Great Lakes.

» As with many of the compounds, concentrations are decreasing for those with relevant legislation. However, for others, concentrations may not appear to be decreasing because they have reached an asymptotic background level and may still be receiving input from atmospheric deposition, ground water, or rivers and streams.

» Our results showed few trends for trace metals. Most organic contaminants show decreasing concentrations, probably resulting from state and federal regulation.

Enhancements to the Mussel Watch Program

Beginning in 2009, NOAA is making several enhancements to the Mussel Watch Program. The primary goal of these enhancements is improved data and information sharing, and coordination with the monitoring efforts of other federal and state agencies. Specific to the Great Lakes, the Mussel Watch Program has expanded the number of monitoring sites and environmental measurements used to characterize Mussel Watch sites. Some of the benefits of these enhancements will be:

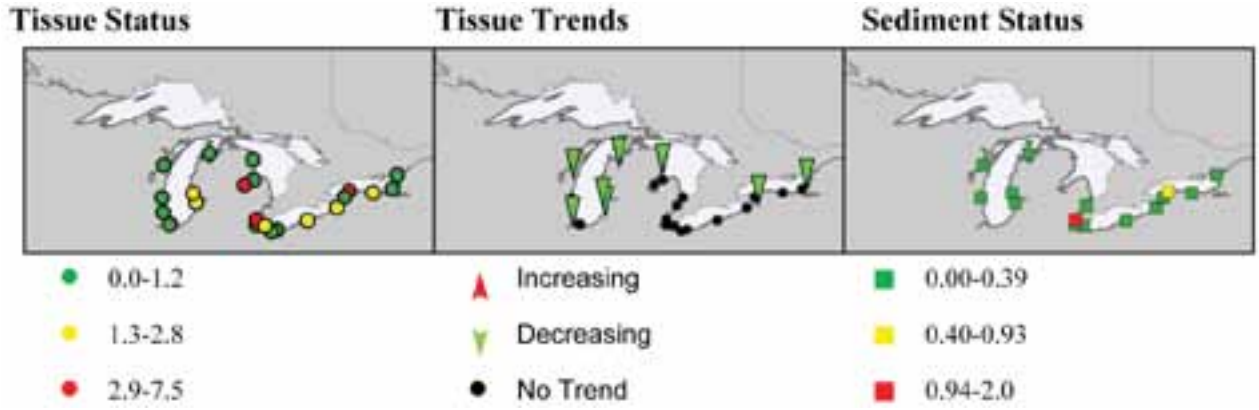


Figure 9-63. Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for Heptachlor (+ Heptachlor Epoxide). All concentrations reported in ng/g dry wt. Source: NOAA Mussel Watch Program.

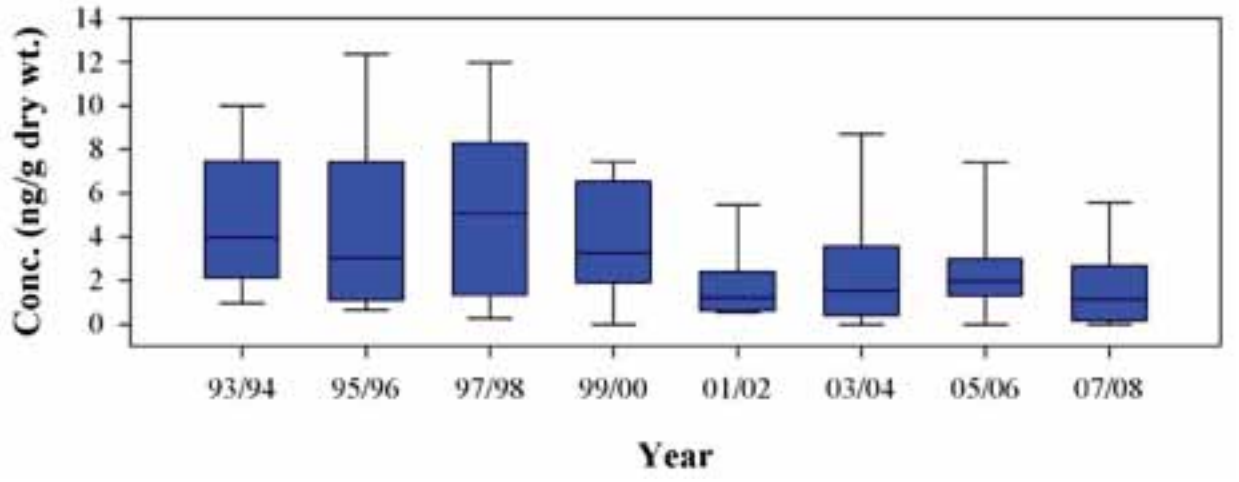


Figure 9-64. Heptachlor (+ Heptachlor Epoxide) Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.

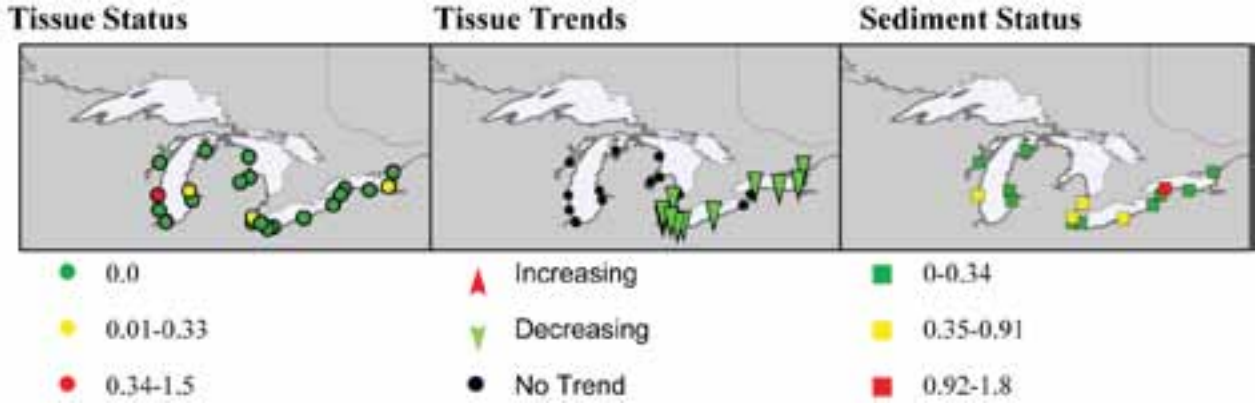


Figure 9-65. **Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for Pentachlorobenzene. All concentrations reported in ng/g dry wt. Source: NOAA Mussel Watch Program.**

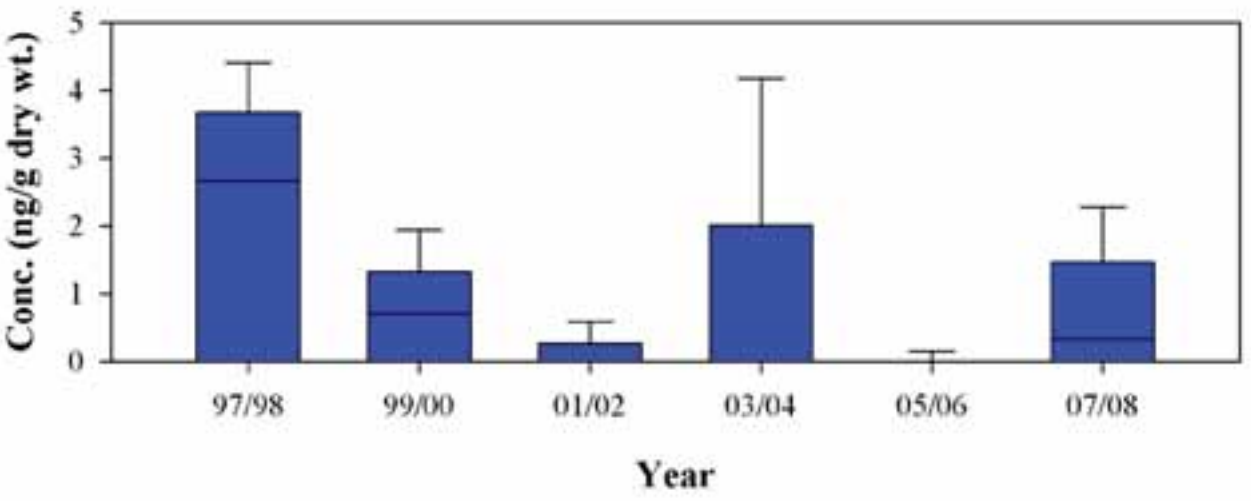


Figure 9-66. **Pentachlorobenzene Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1997-2008. Source: NOAA Mussel Watch Program.**

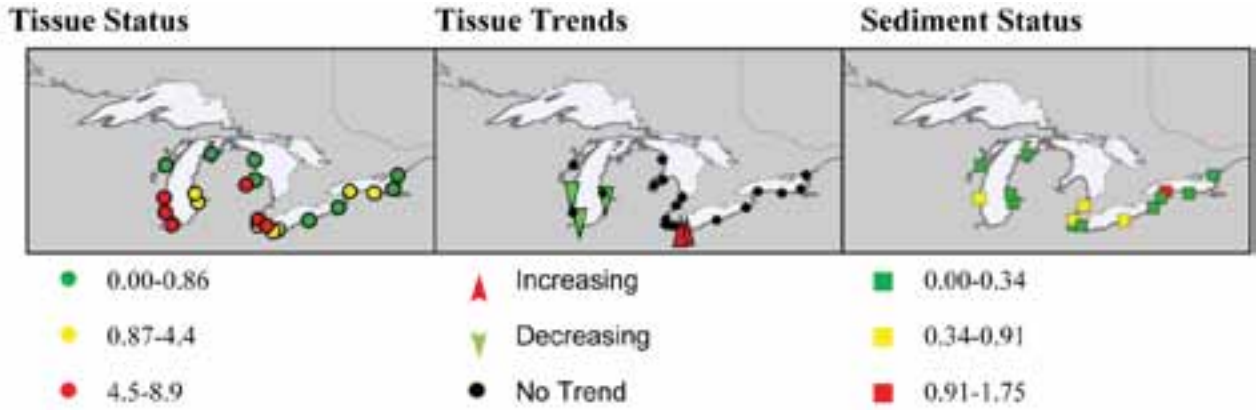


Figure 9-67. Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for TBT. All concentrations reported in ng Sn/g dry wt. Source: NOAA Mussel Watch Program.

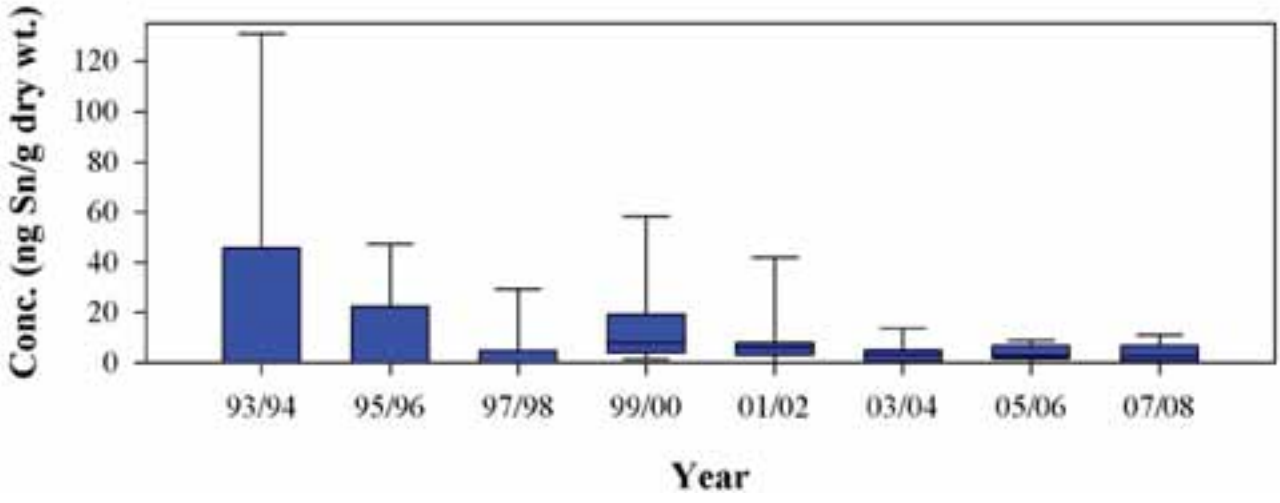


Figure 9-68. TBT Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.

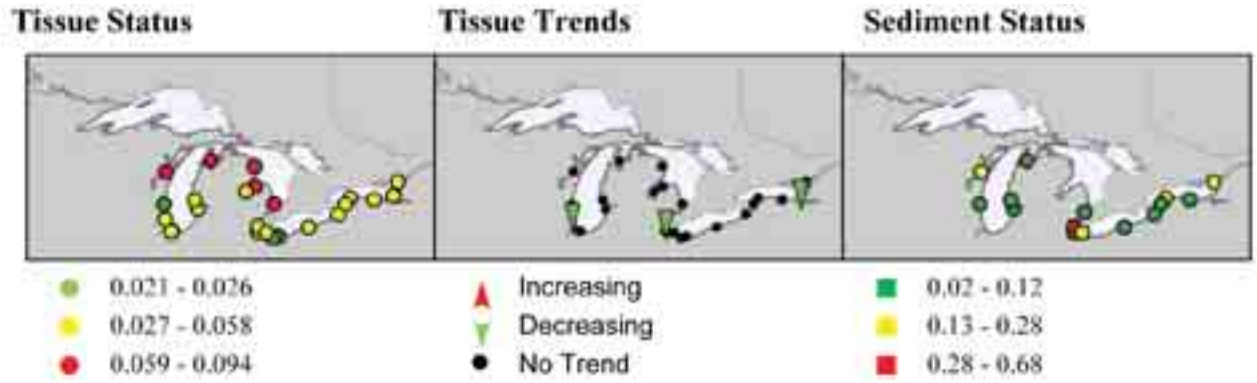


Figure 9-69. **Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for Mercury. All concentrations reported in µg/g dry wt. Source: NOAA Mussel Watch Program.**

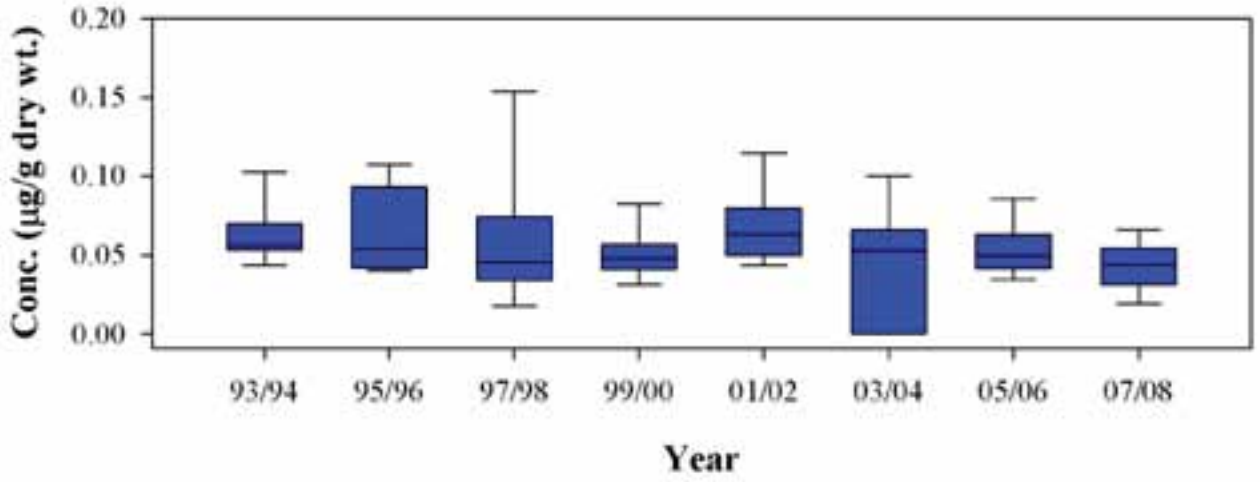


Figure 9-70. **Mercury Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1993-2008. Source: NOAA Mussel Watch Program.**

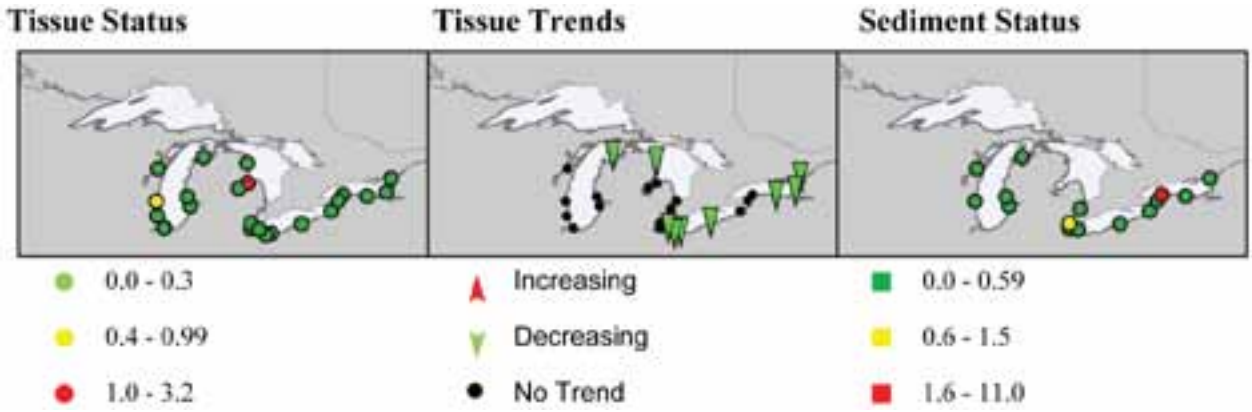


Figure 9-71. Maps with 2006/2007 Bivalve Concentrations (Tissue Status); 1992-2007 Bivalve Concentrations (Trends); and 2006/2007 Sediment Concentrations (Sediment Status) for Tetrachlorobenzene. All concentrations reported in ng/g dry wt. Source: NOAA Mussel Watch Program.

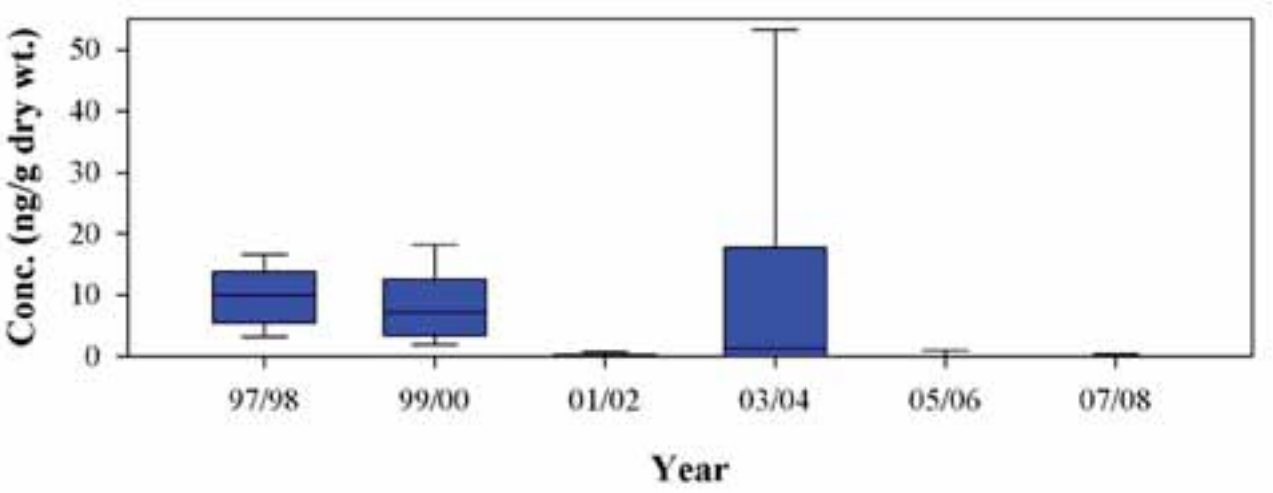


Figure 9-72. Tetrachlorobenzene Whisker Plots for Mussel Watch Tissue from Great Lakes Sites, 1997-2008. Source: NOAA Mussel Watch Program.

- Use of Mussel Watch data to assess the effectiveness of remediation efforts in the Great Lakes.
- Use of contaminant monitoring data for an AOC redesignation into Recovery Stage and for the formal delisting of an AOC.
- Increasing spatial coverage of contaminant monitoring.
- Creation of a warning network for detecting contaminants of emerging concern.
- Expand coordination of monitoring efforts with other agencies.

References

Apeti, D. A., Lauenstein, G. G. and Gerhardt, R. F. 2009. Cadmium distribution in coastal sediments and mollusks of the U.S. Marine Pollution Bulletin 58: 1016-1024.

Apeti, D. D. and Lauenstein, G. G. 2006. An assessment of mirex concentrations along the Great Lakes. *American Journal of Environmental Science* 2: 95-103.

Berner, L.H., J. McGowan, J.H. Martin, and J. Teal. 1976. Sampling marine organisms. In: Strategies for Marine Pollution Monitoring, E. D. Goldberg, (ed.). John Wiley & Sons, NY. pp. 269-73.

Farrington, J. W. 1983. Bivalves as sentinels of coastal chemical pollution: the Mussel (and oyster) Watch. Oceanus 26(2):18-29.

Farrington, J. W., J. Albaiges, K. A. Burns, B. P. Dunn, P. Eaton, J. L. Laseter, P. L. Parker, and S. Wise. 1980. Fossil fuels. In: The International Mussel Watch. National Research Council. National Academy of Sciences - Office of Publications, Washington, D.C. pp. 7-77.

Goldberg, E.D., M. Koide, V. Hodge, A.R. Flegal, and J. Martin. 1983. U.S. Mussel Watch: 1977-1978 results on trace metals and radionuclides. Estuarine Coastal Shelf Science 16:69-93.

Hebert, P. D. N., B. W. Muscaster, and G. L. Mackie. 1989. Ecological and genetic studies on Dreissena polymorpha (Palla): a new mollusk in the Great Lakes. Can. J. Fish. Aquat. Sci. 46: 1587-1591.

Kimbrough, K. L., W. E. Johnson, G. G. Lauenstein, J. D. Christensen and D. A. Apeti. 2008. An Assessment of Two Decades of Contaminant Monitoring in the Nation's Coastal Zone. Silver Spring, MD. NOAA Technical Memorandum NOS NCCOS 74. 105 pp.

Martin, M. 1985. State Mussel Watch: Toxics surveillance in California. Marine Pollution Bulletin 16(4):140-146.

Roesijadi, G., J.S. Young, A.S. Drum, and J.M. Gurtisen. 1984. Behavior of trace metals in Mytilus edulis during a reciprocal transplant field experiment. Marine Ecology Progress Series 18:155-70.

Sericano, J.L. 1993. The American Oyster (Crassostrea virginica) as a Bioindicator of Trace Organic Contamination. Doctoral dissertation, Texas A&M University, College Station, TX. 242 pp.

Tripp, B.W., and J.W. Farrington. 1984. Using sentinel organisms to monitor chemical changes in the coastal zone: progress or paralysis. Submitted to the Coastal Society, 9th Annual Conference, October 1984, Atlantic

City, NJ. Woods Hole Oceanographic Institution Contribution No. 5830.

Zar, J.H. 1999. Biostatistical Analysis. 4th Edition. Prentice-Hall: Upper Saddle River, NJ. 931 pp.

Trends in Great Lakes Sediments and Surface Waters

Photo: North Shore stream flows into Lake Superior
Lake Superior, Minnesota
Minnesota Extension Service, Dave Hansen.
Courtesy of US EPA GLNPO.

Spatial and Temporal Trends in Selected Pollutants in Great Lakes Waters and Sediments

Debbie Burniston, Brad Hill, Joanne Parrott, and Chris Marvin
Environment Canada
Burlington, ON

Water and sediment contaminant monitoring programs began in the late 1970s to the mid-1980s and are ongoing in the open waters and interconnecting channels of the Great Lakes (Figures 9-73a and b). Due to the comprehensive nature of these programs, spatial and temporal trends can be assessed over the breadth of the entire Great Lakes Basin and can illustrate the response in the ambient environment to toxic reduction initiatives at local and regional scales. Meanwhile, threat assessment studies can provide additional information on the occurrence of persistent toxic substances or emerging chemicals of concern. The following paragraphs summarize some of the recent results used to establish trends in Great Lakes sediments and surface waters.

A screening-level survey of recently deposited sediments was undertaken for Canadian tributaries to the Great Lakes over a five-year period ending in 2005. The geographical scope of the program was from the Quebec provincial border on Lake Ontario in the east to the Canadian/American border on Lake Superior in the northwest. A total of 431 tributaries were sampled and analyzed for 52 organic compounds.

PFCAs were detected in all of the tributary sediments analyzed. The distribution of concentrations is shown in Figure 9-74. Perfluorooctanoic acid (PFOA) had the highest mean concentration of PFCA in surficial sediment, which showed a west to east concentration gradient across the Great Lakes. There was no similar pattern for the other PFCAs.

Perfluorosulfonate compounds were not found in every tributary analyzed. While PFOSA was the most common, detected in all but two samples, the highest concentrations were found for both PFOS and perfluorodecasulfonate (PFDS). While high levels of PFOS accompanied with significant levels of PFDS and perfluorohexane sulfonate (PFHxS) often reflect an influence from released aqueous fire fighting foam (AFFF), the Lake Ontario tributaries often had much higher PFDS concentrations than PFOS. This difference likely indicates a significant source other than AFFF.

While the highest values for perfluoroalkyl sulfonates (PFAS) were found near large urban areas, not all large urban tributaries contained high concentrations of PFAS. The distribution of concentrations is shown in Figure 9-75. The highest values of total perfluorocarboxylates and total perfluorosulfonates were found in the tributaries of large centers such as Toronto, Hamilton, Burlington (Lake Ontario), Sarnia (Lake Huron), and Windsor. However, none of the six tributaries in Thunder Bay and Sault Ste Marie (Lake Superior) had elevated levels PFAS. It is also noteworthy that none of the sampled tributaries to Lake Erie had elevated concentrations, which may be a reflection of its

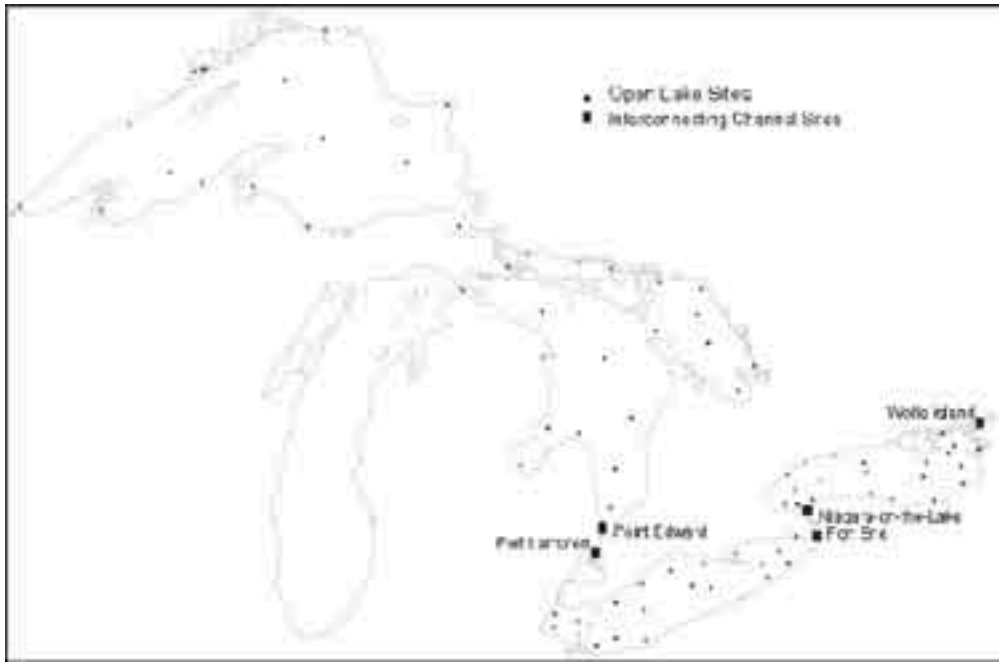


Figure 9-73a. **Open-lake and Interconnecting Channel Water Quality Sites Monitored for Persistent Toxic Substances. Source: Environment Canada**

Figure 9-73b. **Open-lake Bottom Sediment Sites Monitored for Persistent Toxic Substances. Source: Environment Canada**

rural character. Contrary to these observations, Marsh Creek, a tributary in the small community of Picton, Ontario, which drains into the Bay of Quinte (Lake Ontario), had the highest levels of perfluorocarboxylates and the second highest levels of perfluorosulfonate.

The results of this survey provide information about recently deposited sediment quality, and can be used to help determine whether Canadian watersheds are sources of pollutants to the Great Lakes.

Archived sediment samples taken from several Environment Canada monitoring programs established the occurrence and spatial distribution of PBDEs and perfluorinated compounds (PFCs) on sediment in the Detroit River. The Detroit River is one of the connecting rivers between Lake Huron and Lake Erie. Its watershed is highly urbanized and industrialized, and the resultant pollution contributes to its designation as a binational AOC. Levels of both new and emerging chemicals were relatively low compared to historic concentrations of PCBs. Of the PFCs, only PFOS was detected consistently. There appeared to be little influence on the concentrations from the tributaries. In contrast, PBDEs showed an opposite trend, with increasing levels down the river. While some PBDEs are still in production, several formulations have been prohibited, and there is no evidence that environmental levels have decreased in the Detroit River.

Figure 9-76 shows the occurrence and spatial distribution of PFOS in Detroit River suspended sediment in 2000. Levels decrease down the corridor leading to Lake Erie. Decreasing levels may be attributed to dilution by non-contaminated sediment and/or partitioning into the dissolved water phase. Further sources of PFOS down the corridor may include tributaries to the river. While Turkey Creek had the second highest levels of PFOS found in all of the Canadian tributaries to the Great Lakes, the level of 1.1 ng/g does not appear to influence sediment concentrations in the river, suggesting that loadings from the tributary are not great. Other tributaries along the corridor had only minimal concentrations of PFOS.

In contrast to PFOS, the concentration of total PBDEs did not decrease as it moved down the Detroit River (Figure 9-77). While this trend is less clear in 2000 due to the high value in the upper reaches of the river, it should be noted that concentrations between samples were highly variable. PBDE and PFOS show a significant increase in concentration at the top of the river; however, PBDE concentrations continue to increase as the sediment moves down the corridor in 2006, and after a decline in 2000. Differences in the levels of total PBDE at the bottom of the river in the two channels provide evidence that the majority of PBDE loadings are along the western shoreline. The distribution of PBDEs in the Detroit River is comparable to the distribution of HBCD, also a current-use flame retardant.

Environment Canada visited Lake Superior in 2001 and Lake Huron in 2002 to evaluate the current extent of sediment contamination, determine spatial trends of contaminants, and identify areas of associated sources. Tributary sediment surveys of Lakes Superior and Huron were conducted in 2006 and 2004, respectively. Nearshore sediment samples were collected in 2005 campaigns for Lake Superior, St. Marys River, North Channel, and in 2002 for Lake Huron. These samples were collected to determine the occurrence and spatial distribution of 2,3,7,8-substituted PCDDs and PCDFs, dioxin-like polychlorinated biphenyls (DLPCBs), and PBDEs; and to identify potential sources of these contaminants to the lakes. Results indicate PCDD/F and DLPCB levels at high-level sites are significantly different from the mean level across the Lakes Superior and Huron basins.

Figure 9-78 shows PCDD/F and DLPCB concentrations in sediments from the study regions. Generally, PCDD/Fs and DLPCBs at most sampling sites were found at low levels. Highest levels of these contaminants were observed at tributary sites and an off-shore site. PCDD/Fs across the Lakes Superior and Huron basins were generally lower than those observed in Lakes Ontario and Erie.

PBDE concentrations in Lake Huron sediment are shown in Figure 9-79. Generally, PBDEs in sediments were observed at low-ppb levels with a lakewide

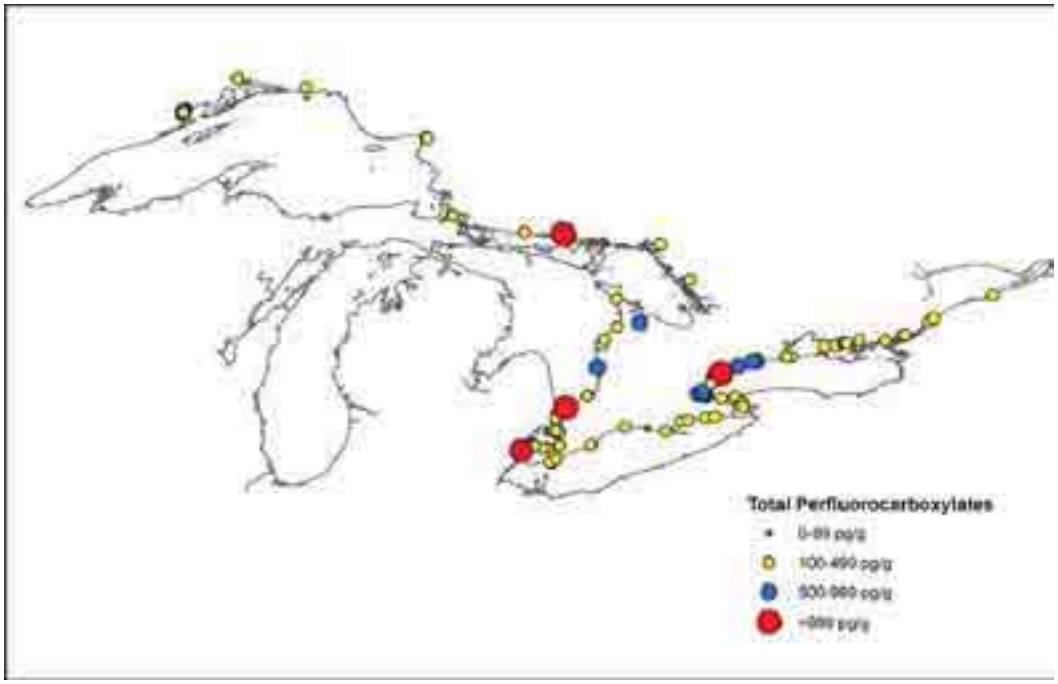


Figure 9-74. Levels of Perfluorocarboxylates in Canadian Tributaries to Great Lakes, 2000 – 2005.[77]

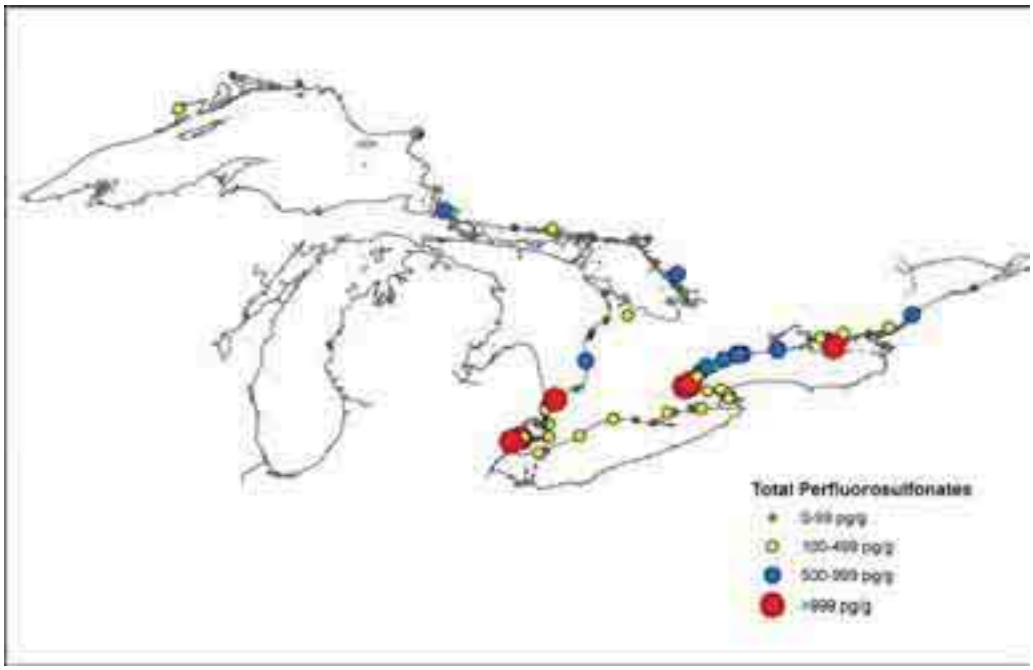


Figure 9-75. Levels of Perfluorosulfonates in Canadian Tributaries to Great Lakes, 2000 – 2005. [78]

[77] Burniston et al., 2006.
[78] Ibid.

Photo 9-1. Sediment Retrieval in Randle Reef. Courtesy of Joanne Parrott, Environment Canada.

Photo 9-2. Sprayer Suits on Randle Reef. Courtesy of Joanne Parrott, Environment Canada.

average of 4000 pg/g dry wt. The concentrations observed in this study are similar to those found in Lake Superior, and slightly lower than those previously reported in Lake Huron. Environmental releases of PBDEs to the Great Lakes are believed to be from the use of the penta- and/or deca-mixtures.

Environment Canada conducted sediment surveys to detect PBDEs in the open water of Lake Superior in 2001, Lake Huron including Georgian Bay and North Channel in 2002, and Lake Michigan in 2002 to evaluate the current extent of sediment contamination, determine spatial trends of contaminants, and identify areas of potentially associated sources. Environment Canada also conducted a tributary screening survey on Lake Superior in 2006 and Lake Huron in 2004 by sampling surficial sediments near the mouths of Canadian tributaries. The survey provides an indicator of water quality and contaminant loadings in Canadian watersheds around the lakes. Water quality in the nearshore areas of the Great Lakes is regularly monitored by the MOE through the Great Lakes Nearshore Monitoring and Assessment Program.

The sum of 17 PBDE concentrations is shown in Figure 9-80. PBDEs are widely dispersed and display a large variation across the watersheds of Lake Superior, Lake Huron, and Lake Michigan. In general, the open water areas of Lake Huron and Lake Michigan exhibit slightly higher levels of PBDEs than Lake Superior (Figure 9-81). Nearshore sediments had PBDE concentration ranges similar to offshore sediments in Lake Superior and Lake Huron (Figure 9-81, Shen et al., 2008).

Results from the Upstream/Downstream Program, part of the Niagara River Toxics Management Plan (NRTMP),[79] are intended to determine whether concentrations of specified chemicals at the mouth of the Niagara River at Niagara-on-the-Lake (NOTL) are statistically different from concentrations at the head of the Niagara River at Fort Erie (FE), and to assess trends over time. The Upstream/Downstream Program measures the concentrations of trace metals in whole water and trace organic contaminants in both water and suspended solids.

A comparison of recombined whole water and whole water sampling results (90% Confidence Interval) with the most stringent agency water quality criteria for the period 2001 through 2005 reveals:

[79] The NRTMP is approved by Four Parties: Environment Canada, United States Environmental Protection Agency (Region II), Ontario Ministry of the Environment, and New York State Department of Environmental Conservation.

Figure 9-76. PFOS Concentrations in Suspended Sediment in the Detroit River in 2000. [80]

Figure 9-77. PBDE Concentrations in Suspended Sediment in the Detroit River 2000 and 2006. [81]

[80] Burniston and Marvin, 2009.
[81] Ibid.

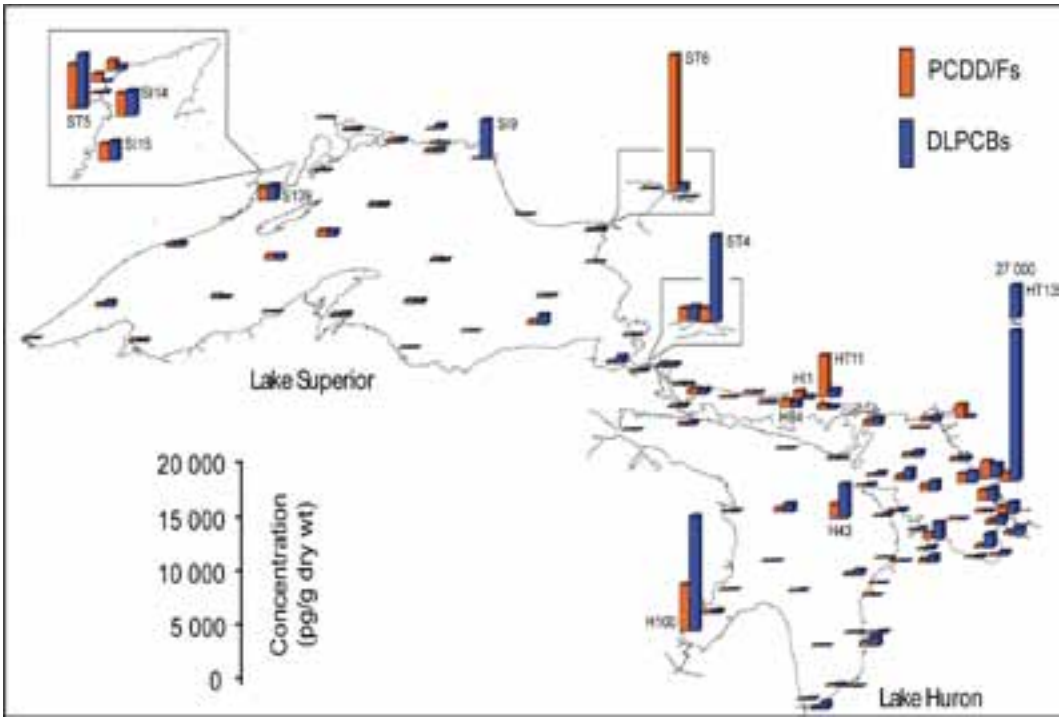


Figure 9-78. PCDD/F and DLPCB Concentrations (pg/g dry wt.) in Lakes Superior and Huron Sediment. [82]

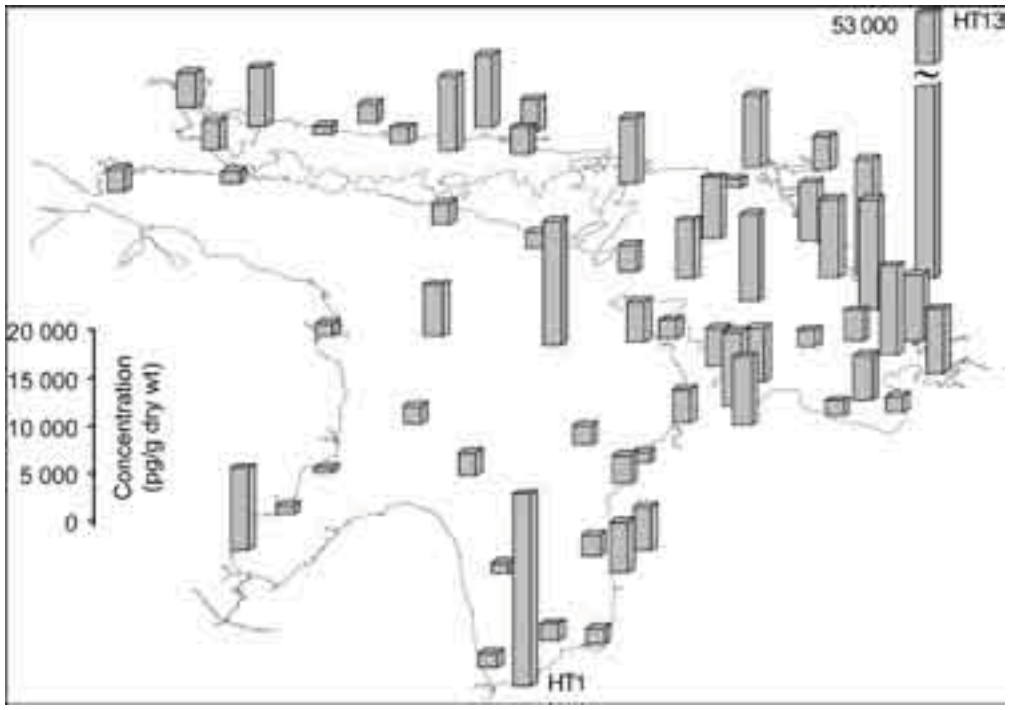


Figure 9-79. PBDE Concentrations (pg/g dry weight) in Lake Huron Sediment. [83]

[82] Shen et al., 2009.
[83] Shen et al., 2007.

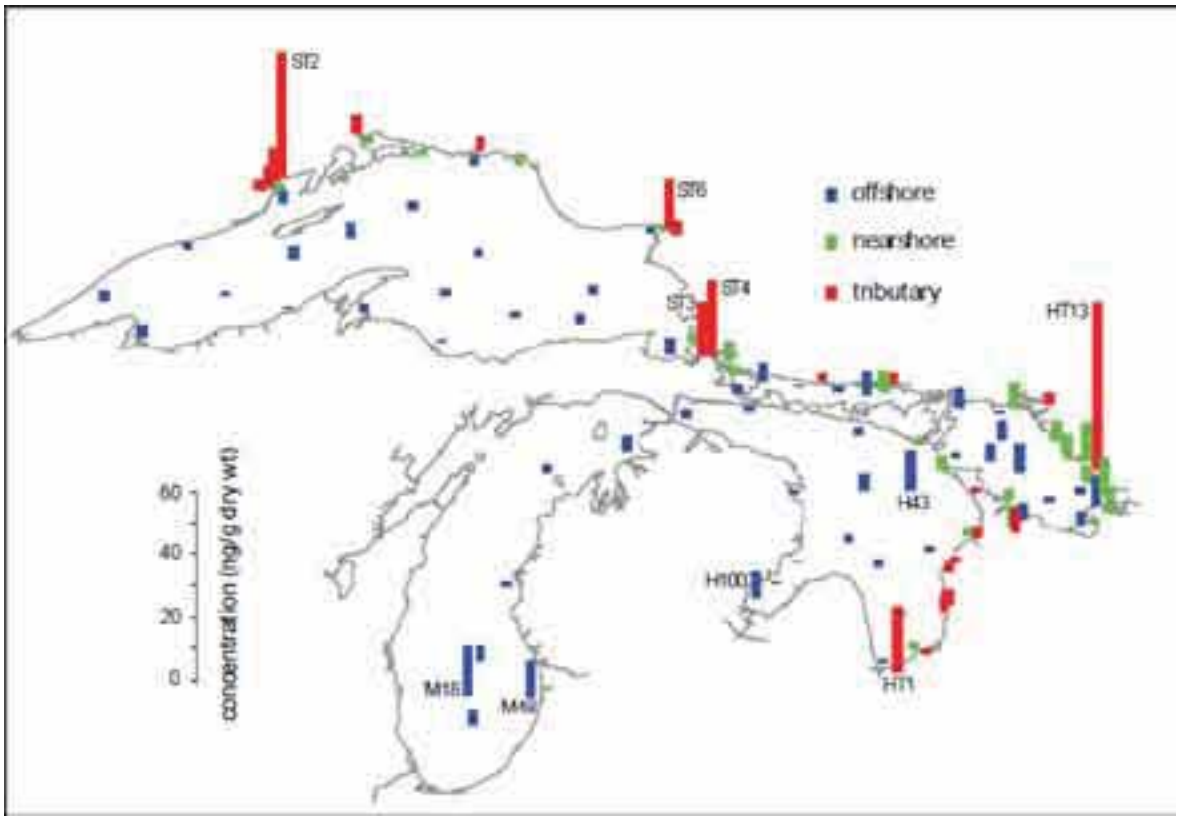


Figure 9-80. PBDEs in Surficial Sediments of Lakes Superior, Huron and Michigan. [84]

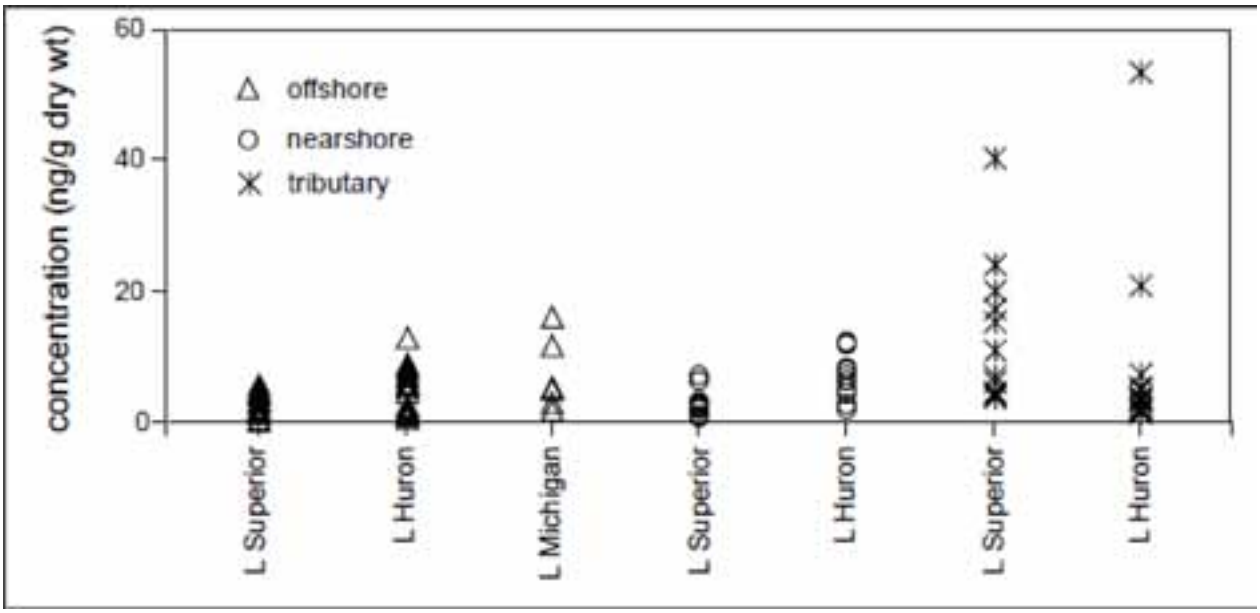


Figure 9-81. PBDE Concentrations in Surficial Sediments of Lakes Superior, Huron and Michigan. [85]

[84] Shen et al., 2008.
[85] Ibid.

- 17 of the 71 compounds sampled showed exceedences of the strictest agency guidelines between 2001-2002 and 2004-2005.
- 13 of the 17 compounds that show exceedences, including dieldrin, HCB, total chlordane, mirex, pp-DDT, pp-DDE, total DDT, total PCB, benz(a)anthracene, benzo(b/k) flouranthene, chrysene/triphenylene, benzo(a)pyrene, and mercury are part of the NRTMP's 18 "Priority Toxics".
- The remaining four compounds that exceed strictest agency guidelines include benzo(g,h,i)perylene, indeno(1,2,3-c,d) pyrene, aluminum, and iron.
- Mirex, HCB, chrysene/triphenylene, total chlordane, benzo(a)pyrene, benz(a) anthracene, indeno(1,2,3-c,d)pyrene, iron, and mercury exceeded their criteria only at NOTL.
- Dieldrin p,p-DDT, p,p-DDE, total DDT, total cogener PCBs (TCPCBs), benzo(b/k) fluoranthene, benzo(g,h,i)perylene, and aluminum exceeded strictest agency criteria at both FE and NOTL, suggesting Lake Erie/ upstream sources to the river.
- Based on the particulate phase only, mercury concentrations exceeded the strictest whole water criteria (1.3 ng/L) once in the four-year period (2001-2002) and only at the NOTL site.

In addition to identifying water quality criteria exceedences, the Niagara River Upstream/ Downstream Monitoring Program is used to examine trends in the concentrations and loadings of toxic compounds.

The trend of dieldrin concentration in the dissolved phase at NOTL and FE is shown in Figure 9-82. The concentrations and rate of decrease are similar at both stations. This suggests that the major input of dieldrin to the river is from Lake Erie/upstream, and that the changes occurring at both the FE and 16 NOTL stations are being dictated by changes in dieldrin concentrations upstream of the river.

Changes in HCB concentrations in suspended sediment using annual Maximum Likelihood Estimations (MLEs) at the NOTL and FE stations are shown in Figure 9-83. In contrast to dieldrin concentrations, HCB concentrations are vastly different at the two stations.

In some cases, compounds are not detected at the upstream FE site and trends can only be seen at NOTL. This is the case, for example, for OCS and mirex (Figure 9-84). This indicates that the chemical is originating from Niagara River sources, and the concentrations and changes in concentration reflect what is happening at those sources.

The PAHs benzo(b/k)fluorathene and benzo(a)pyrene are shown in Figures 9-85 and 9-86, respectively. The results suggest that there is an increasing trend for these contaminants in the suspended sediment at FE and NOTL. The reason for the increases is not known at present, but one theory is that the increases may be due to the change in the characteristics of the bottom sediments as a result of zebra and quagga mussel colonization of the eastern basin of Lake Erie. Evidence also seems to suggest that increasing PAH levels may be related to increased vehicular traffic at border crossings in the Niagara region (Van Metre, 2000).

The Aquatic Ecosystem Protection Research Division & Aquatic Ecosystem Management Research Division of Environment Canada[86] assessed the toxicity of Hamilton Harbour sediments and waters to laboratory fish prior to Randle Reef dredging and remediation activities. Sediments were collected (see Photo 9-1), and semi-permeable membrane devices (SPMDs) were deployed at several locations in Randle Reef (see Photo 9-2), Windermere Arm, Hamilton Harbour and Lake Ontario. Chemicals such as PAHs and PCBs taken up by fish can cause increases in detoxifying enzymes in the liver. Juvenile rainbow trout exposed for 4 days to sediments had increased liver enzyme activities (ethoxyresorufin-O-deethylase, EROD) 5- to 15-fold above controls. The most potent EROD-inducing sediments were

[86] Environment Canada, National Water Research Institute, Burlington, Ontario.

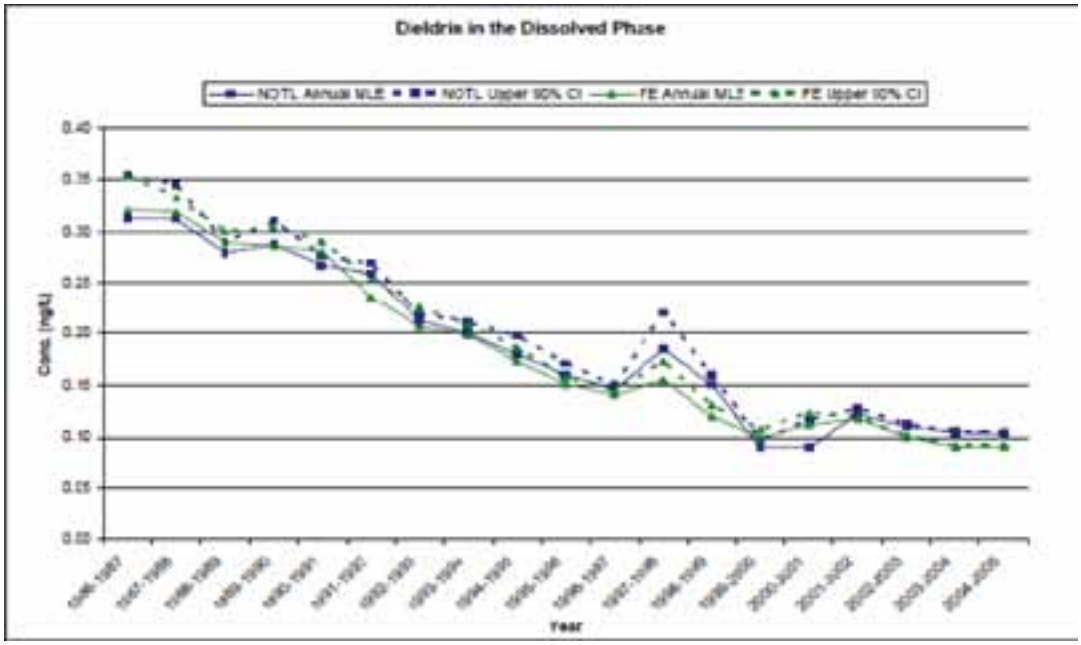


Figure 9-82. **Annual Dissolved Phase Maximum Likelihood Estimation (MLE) and Upper 90% Confidence Interval (CI) of Dieldrin from 1986-1987 to 2004-2005.** [87]

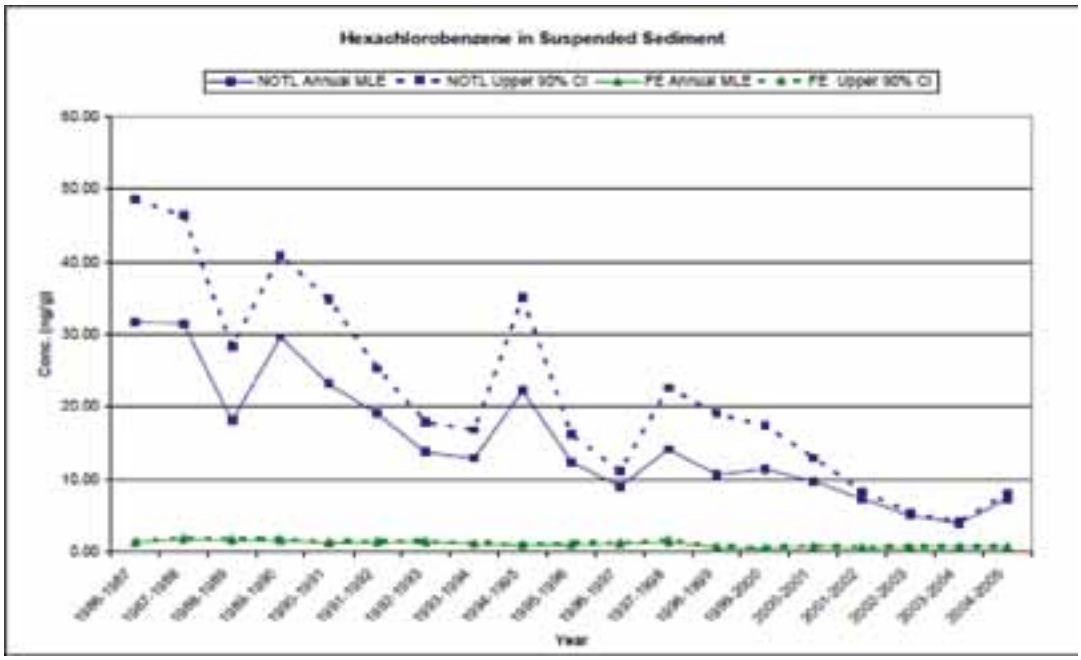


Figure 9-83. **Annual Suspended Sediment Maximum Likelihood Estimation (MLE) and Upper 90% Confidence Interval (CI) of HCB from 1986-1987 to 2004-2005.** [88]

[87] Niagara River Secretariat, 2007.
[88] Ibid.

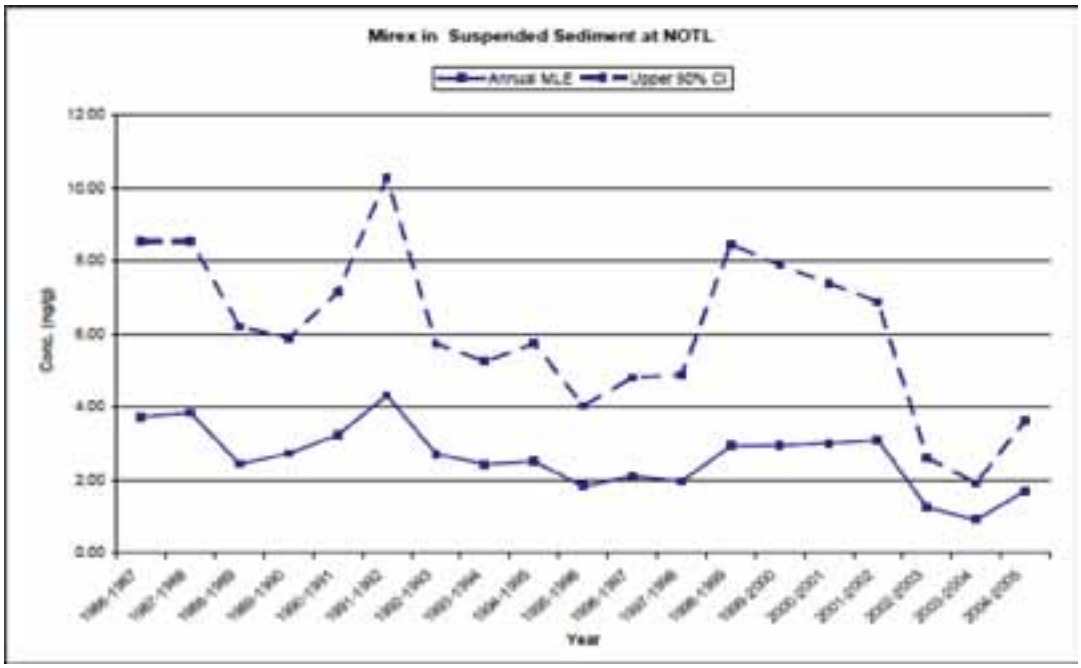


Figure 9-84. Annual Suspended Sediment Maximum Likelihood Estimation (MLE) and Upper 90% Confidence Interval (CI) of Mirex from 1986-1987 to 2004-2005. [89]

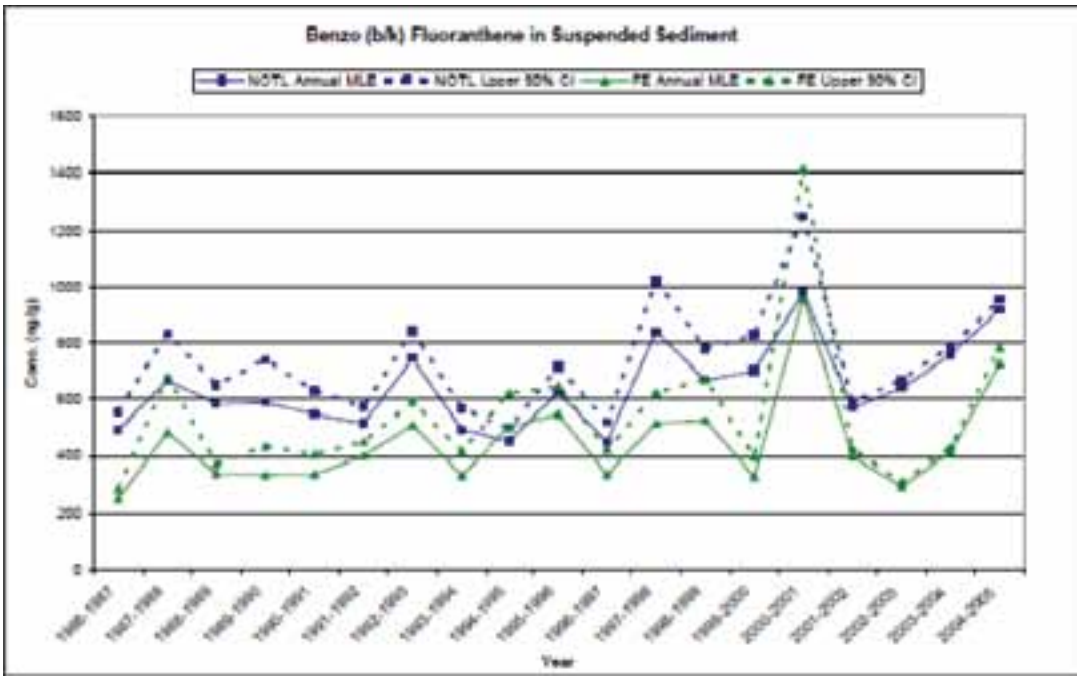


Figure 9-85. Annual Suspended Sediment Maximum Likelihood Estimation (MLE) and Upper 90% Confidence Interval (CI) of Benzo(b/k)fluoranthene from 1986-1987 to 2004-2005. [90]

[89] Ibid.
[90] Ibid.

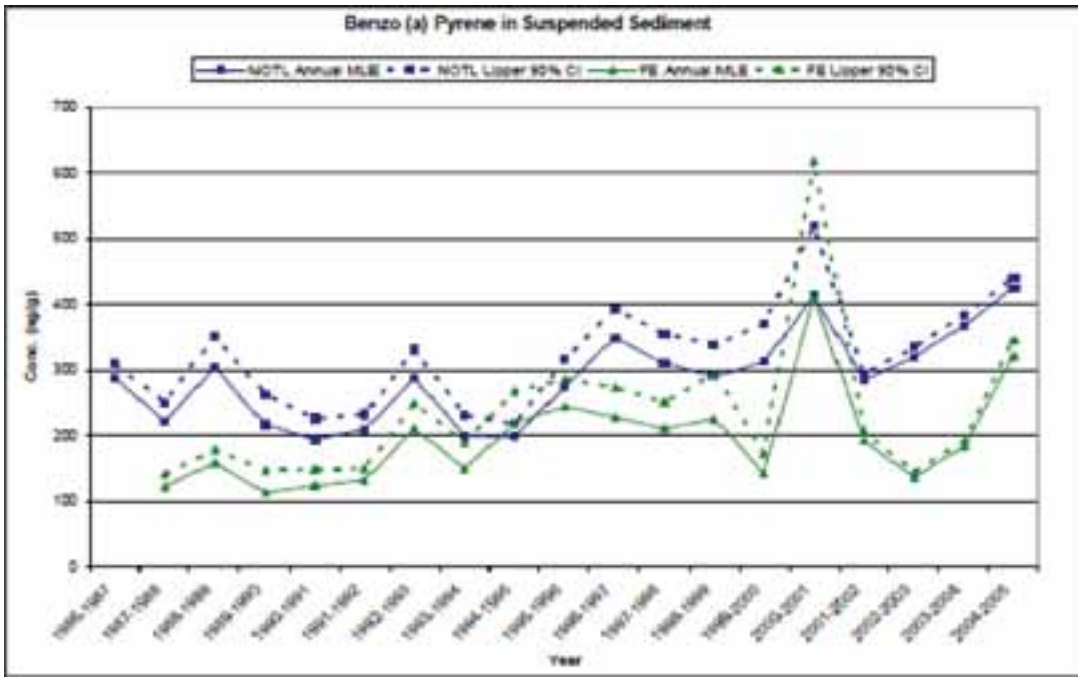


Figure 9-86. **Annual Suspended Sediment Maximum Likelihood Estimation (MLE) and Upper 90% Confidence Interval (CI) of Benzo(a)pyrene from 1986-1987 to 2004-2005.** [91]

[91] Ibid.

from Randle Reef, and these sediments contained the highest concentrations of PAHs, including benzo(a)pyrene, anthracene, benzo(a)anthracene, benzo(g,h,i)perylene, perylene, and phenanthrene.

Fish embryos exposed to Randle Reef sediments for 15 days showed changes in growth, development and survival of eggs and newly-hatched fish. Embryos exposed to 60 and 200 g of Randle Reef sediment/L had increased egg and larval mortality, as well as severe deformities (Figure 9-87). In addition, exposure to Randle Reef sediments reduced larval size compared to water controls and reference-exposed groups. Analysis is ongoing to determine if individual PAHs or groups of certain PAHs (in sediments or SPMD extracts) can account for most of the fish EROD and fish embryo toxicity (Figure 9-88). The results allow Environment Canada to assess and rank the potency of Hamilton Harbour sediments in terms of fish responses prior to clean-up. Fish responses will be compared to future post-remediation sediments to demonstrate changes in fish toxicity after remediation.

References

Burniston D, Dove A, Backus S, and Kraft J. 2006. Environment Canada, Sediment Quality in Canadian Lake Huron Tributaries: A Screening-Level Survey. Report No. ECB/EHD-OR/06-01/I.

Burniston D, Furdui V, Dove A, Backus S, Reiner E, Kraft, J. 2006. Perfluoroalkyl Compounds in Surficial Sediment from Canadian Great Lake Tributaries. 22nd International Symposium on Halogenated Environmental Organic Pollutants (2006), Organohalogen Compounds, Vol. 68.

Burniston, D and C Marvin. 2009. Polybrominated Diphenyl Ethers and Perfluorinated Compounds in Detroit River Sediment. 24th International Symposium on Halogenated Environmental Organic Pollutants (2009). *Organohalogen Compounds*, Volume 71.

Hill, B and P Klawuun, 2009. Niagara River Upstream/Downstream Monitoring Program Report 2001-2002 to 2004-2005. For Data Interpretation Group, River Monitoring Committee.

Niagara River Secretariat' 2007. Niagara River Toxics Management Plan (NRTMP) Progress Report and Work Plan.

Parrott J, Colavecchia M, and Marvin C, Environment Canada, Burlington, Ontario.

Shen L, Gewurtz S, Reiner E, Kolic T, MacPherson K, Helm P, Howell T, Burniston D, Brindle I, Marvin C, 2007. PCDD/Fs, Dioxin-like PCBs, PBDEs in Surficial Sediments of Lakes Superior and Huron. 23rd International Symposium on Halogenated Environmental Organic Pollutants (2007). *Organohalogen Compounds* Volume 69.

Shen L, Gewurtz S, Reiner E, Kolic T, MacPherson K, Helm P, Howell T, Burniston D, Brindle I, Marvin C, 2008. PBDEs In Surficial Sediments of the Great Lakes: Lakes Superior, Huron and Michigan. 23rd International Symposium on Halogenated Environmental Organic Pollutants (2008). *Organohalogen Compounds* Volume 70.

Shen, L., Gewurtz, S.B., Reiner, E.J., MacPherson, K.A., Kolic, T.M., Khurana, V., Helm, P.A., Howell, E. T., Burniston, D. A., Brindle, I. D., Marvin, C. H., 2009. Occurrence and sources of polychlorinated dibenzo-p-dioxins, dibenzofurans and dioxin-like polychlorinated biphenyls in surficial sediments of Lakes Superior and Huron. *Environmental Pollution*, 157 (4), p.1210-1218.

Van Metre PC, Mahler BJ, and ET Furlong, 2000. "Urban Sprawl Leaves its PAH Signature", *Environ. Sci. Technol.*, v34, p.4064-4070.

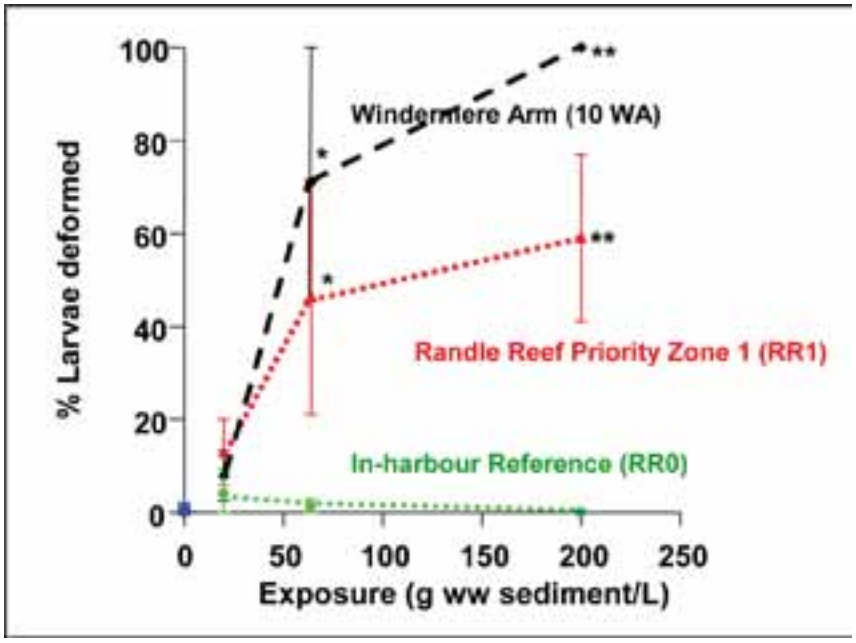


Figure 9-87. **Percentage of Larva Deformed after Exposure to Sediments from Reference Site (green line), Randle Reef (red line) or Windermere Arm (black line). Source: Environment Canada**

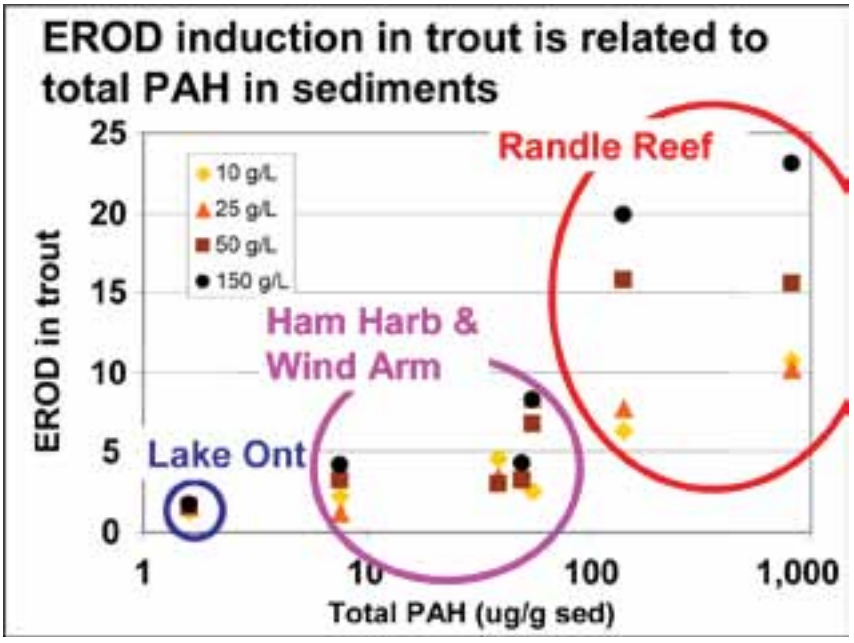


Figure 9-88. **EROD Induction (pmoles/mg protein/minute) in Trout Exposed to Different Concentrations (10 to 150 g/L) of Various Sediments from Hamilton Harbour. Randle Reef sediments contained the highest concentrations of PAHs and caused the highest EROD responses in fish. Source: Environment Canada**

APPENDIX A:
GREAT LAKES BINATIONAL TOXICS STRATEGY:
COMPENDIUM OF ACTIVITIES 1997 – 2009

Lighthouse Kewaunee, Wisconsin, Photograph by Carole Y. Swinehart

GREAT LAKES BINATIONAL TOXICS STRATEGY (GLBTS) PROGRESS OVERVIEW 1997 – 2009

GLBTS Development, Integration Workgroup, and Stakeholder Forum

1997

- 4/7/97 U.S. and Canada sign the GLBTS: *Canada-United States Strategy for the Virtual Elimination of Persistent Toxic Substances in the Great Lakes*
- 6/26/97 Stakeholders invited to workshop to develop a draft GLBTS Implementation Plan
- 12/97 GLBTS Implementation Plan distributed and Substance participation solicited
- 12/97 GLBTS Website is developed

1998

- 3/23/98 Kick-off implementation meeting in Chicago to form seven substance workgroups
- 6/19/98 The first GLBTS Integration Workgroup meeting is convened in Romulus, Michigan
- 6/98 GLBTS Website is redesigned; PCBs and Mercury Workgroup pages added
- 7/98 GLBTS Website is redesigned; Integration, Dioxins, Pesticides, HCB/B(a)P, Alkyl-lead, and OCS Workgroup pages added
- 10/21-23/98 GLBTS display and presentation (including GLBTS handouts, a brochure, Website cards, GLBTS progress timeline and activity sheets) at SOLEC in Buffalo, NY
- 11/16/98 The first GLBTS Stakeholder Forum is convened in Chicago, IL
- 11/16/98 The first GLBTS Progress Report is distributed

1999

- 1/26/99 GLBTS Integration Workgroup meets in Windsor, Ontario
- 4/27/99 GLBTS Stakeholder Forum is held in Toronto, Ontario
- 4/28/99 GLBTS Integration Workgroup meets in Toronto, Ontario
- EC and US EPA develop draft communications strategy, present it to Integration Workgroup, and revise strategy based on stakeholder comments
- 8/24/99 GLBTS Integration Workgroup meets in Detroit, Michigan
- 9/23-26/99 US EPA, EC and invited speakers give GLBTS session presentation at the IJC Great Lakes Water Quality Forum in Milwaukee, WI
- 9/24/99 A preliminary draft GLBTS Progress Report issued at IJC meeting in Milwaukee, WI
- 10/99 GLBTS main and Mercury Workgroup web pages are redesigned
- 10/7/99 A Canadian *GLBTS Report on Level II Substances* is posted on the GLBTS Website
- 11/18/99 GLBTS Stakeholder Forum is held in Chicago, IL
- 11/19/99 GLBTS Integration Workgroup meets in Chicago, IL
- 12/99 Preliminary planning initiated for a PCP Workshop (to include the GLBTS pesticides, HCB and Dioxin/Furan Workgroups)
- 12/3/99 a U.S. *GLBTS Report on Level II Substances* is posted on the GLBTS Website

- 12/15/99 Draft (Full) 1999 GLBTS Progress Report issued
- 1999 (various dates) Development of a Canadian GLBTS communications plan

2000

- 1/28/00 Municipal Solid Waste and Incineration Workgroup planning conference call
- 2/11/00 Municipal Solid Waste and Incineration Workgroup planning conference call

- 2/15/00 GLBTS Integration Workgroup meets in Windsor, Ontario
- 5/15/00 Protecting the Great Lakes, Sources of PBT Reductions Workshop on Municipal Solid Waste Management is held in Toronto, Ontario
- 5/16/00 GLBTS Stakeholder Forum is held, with the theme "Meeting the Challenge"
- 9/22/00 GLBTS Integration Workgroup meets in Chicago, IL
- 2000 (various dates) GLBTS communications plan is finalized by EC; "key messages" finalized; various communications products in development (brochure, business cards, display unit, letterhead, Website improvements, success stories)

2001

- 2/20/01 GLBTS Integration Workgroup meets in Windsor, Ontario
- 2/21/01 GLBTS 2000 Progress Report is posted to GLBTS Website
- 5/17/01 GLBTS Stakeholder Forum is held in Toronto, Ontario
- 5/18/01 GLBTS Integration Workgroup meets in Toronto, Ontario
- 6/18/01 GLBTS Sector Subgroup begins a series of conference calls to select a short list of sectors for a pilot effort
- 8/28/01 GLBTS Integration Workgroup meets in Chicago, IL
- 9/19/01 GLBTS Sector Subgroup begins information-gathering phase focusing on the short list of sectors
- 11/14/01 GLBTS Stakeholder Forum is held in Chicago, IL, with the theme "Implementation – Partners in Progress"
- 11/15/01 GLBTS Integration Workgroup meets in Chicago, IL
- 11/16/01 GLBTS/LaMP Workshop in Chicago, IL, with the theme of "Program Synergies – Partners in Progress, Exploring how we can mutually support the pollutant reduction needs and efforts of each program synergistically"

2002

- 1/25/02 GLBTS Sector Subgroup begins summarizing findings
- 2/26/02 GLBTS Sector Subgroup presents summary of findings to Integration Workgroup
- 2/26/02 GLBTS Integration Workgroup meets in Windsor, Ontario
- The GLBTS EC/US EPA Website "binational.net" is created
- 5/29/02 GLBTS Stakeholder Forum and Five-Year Anniversary event are held in Windsor, Ontario
- 5/29/02 GLBTS Five-Year Perspective report issued
- 5/30/02 GLBTS Integration Workgroup meets in Windsor, Ontario
- 9/16/02 GLBTS Sector Subgroup holds conference call to discuss a pilot sector project
- 9/18/02 GLBTS Integration Workgroup meets in Chicago, IL
- 12/3/02 GLBTS Stakeholder Forum is held in Chicago, IL
- 12/3/02 Draft GLBTS 2002 Progress Report issued
- 12/4/02 GLBTS Integration Workgroup meets in Chicago, IL

2003

- 2/25/03 GLBTS Integration Workgroup meets in Windsor, Ontario
- 3/01/03 GLBTS Binational.net bookmark created as a marketing tool
- 4/01/03 GLBTS CD ROM containing the Strategy, annual progress reports (1998, 1999, 2000, 2001, & 2002), Five-Year Perspective, and various Strategy Updaters (all in both French and English) is created and 5,000 copies are sent to basin stakeholders and Washington and Ottawa government officials
- 4/03/03 GLBTS presentation to the Lake Superior LaMP Forum in Duluth, Minnesota
- 5/05/03 GLBTS presentation to International Pulp and Paper Conference in Portland, Oregon
- 5/13/03 GLBTS presentation to Commission for Environmental Cooperation, Sound Management of Chemicals (SMOC) meeting in Windsor, Ontario
- 5/14/03 Final GLBTS 2002 Progress Report posted at www.epa.gov/glnpo/bns and binational.net
- 5/14/03 GLBTS Stakeholder Forum held in Windsor, Ontario, in conjunction with CEC SMOC public meeting
- 5/15/03 GLBTS Integration Workgroup meets in Windsor, Ontario
- 6/01/03 GLBTS Update prepared, as well as GLBTS displays in French, Spanish, and English
- 6/11/03 GLBTS presentation to Canadian P2 Roundtable in Calgary, Alberta
- 6/16/03 Conference call with Agricultural Subgroup of Integration Workgroup
- 6/23/03 GLBTS presentation to IAGLR in Chicago, Illinois
- 7/31/03 GLBTS Public outreach tent set up at Chicago Tall Ships event in Chicago, Illinois
- 8/11/03 GLBTS presentation at Emerging Chemicals Workshop in Chicago, Illinois
- 8/19/03 Conference call with LaMP leads to discuss GLBTS/LaMP Crosswalk of priorities
- 9/01/03 *GLBTS 2003 Activity Update* prepared
- 9/04/03 Conference call held with small number of Integration Workgroup members to discuss draft GLBTS Level I Substance Assessment Process
- 9/11/03 GLBTS Integration Workgroup meets in Toronto, Ontario
- 9/11/03 *GLBTS Fall 2003 Workgroup Activity Update* distributed
- 9/18/03 GLBTS attendance at the IJC Public Forum in Ann Arbor, Michigan
- 10/24/03 GLBTS presentation to European delegation at EU REACH Program in Chicago, Illinois
- 11/25/03 Conference call with LaMP and GLBTS Stakeholders to discuss GLBTS Level I Substance Assessment Process
- 12/02/03 GLBTS presentation to Lake Superior LaMP Task Force in Thunder Bay, Ontario
- 12/16/03 GLBTS Stakeholder Forum is held in Chicago, IL
- 12/16/03 Draft *GLBTS 2002 Progress Report* issued
- 12/17/03 GLBTS Integration Workgroup meets in Chicago, IL

2004

- 2/04 Final *GLBTS 2003 Progress Report* posted at www.epa.gov/glnpo/bns and binational.net
- 4/13/04 – 4/15/04 GLBTS Management Framework Workshop in Chicago, Illinois
- 6/17/04 GLBTS Stakeholder Forum is held in Toronto, Ontario
- 6/18/04 GLBTS Integration Workgroup meets in Toronto, Ontario
- 10/07/04 GLBTS Integration Workgroup meets in Toronto, Ontario: Draft *Management Assessment for OCS and Management Assessment for Dioxin and Furans* presented
- 10/07/04 GLBTS Fall 2004 Workgroup Activity Update distributed
- 11/16/04 – 11/18/04 Presentation at Workshop on Environmental Health Effects of Persistent Toxic Substances – Hong Kong: "The GLBTS as a Governance Model to reduce PTS"
- 11/30/04 GLBTS Stakeholder Forum is held in Chicago, IL
- 12/01/04 Draft *GLBTS 2004 Progress Report* issued
- 12/01/04 GLBTS Integration Workgroup meets in Chicago, IL

2005

- 2/10/05 GLBTS update presented to Lake Superior LaMP Chemical committee in Marquette, MI, given by Alan Waffle and E.Marie Wines
- 3/09/05 GLBTS update presented at GLRPPR in Chicago, IL, given by Alan Waffle
- 3/11/05 GLBTS attendance (Alan Waffle) at EC's Workshop on Pharmaceuticals and Personal Care products in Burlington, Ontario
- 3/23/05 GLBTS Integration Workgroup meets in Windsor, Ontario: Draft *Management Assessments for HCB, B(a)P, PCB, mercury, alkyl-lead, and pesticides* presented
- 3/29/05 GLBTS attendance at IJC Chemical Exposure Workshop in Chicago, IL
- 4/11/05 GLBTS display presented at US National Environmental Partnership Summit
- 5/05 Final GLBTS 2004 Progress Report posted at http://binational.net/bns/2004/index.html
- 5/17/05 GLBTS Stakeholder Forum is held in Toronto, Ontario
- 5/18/05 GLBTS Integration Workgroup meets in Toronto, Ontario
- 5/24/05 GLBTS presentation given by Ted Smith at IAGLR in Ann Arbor, MI
- 6/01/05 GLBTS presentation at Canadian Pollution Prevention Roundtable in Victoria, British Columbia, given by Tricia Mitchell and Alan Waffle
- 9/15/05 GLBTS Integration Workgroup meets in Chicago, IL
- 9/27/05 GLBTS update presented to Lake Superior LaMP Workgroup in Thunder Bay, Ontario, given by Alan Waffle
- 9/29/05 GLBTS attendance (Ted Smith and Alan Waffle) at SOLEC Chemical Integrity Workshop in Windsor, Ontario
- 11/02/05 GLBTS attendance (Alan Waffle) at IJC GLWQA Public Meeting in Windsor, Ontario
- 12/06/05 GLBTS Stakeholder Forum is held in Chicago, IL
- 12/07/05 Draft *GLBTS 2005 Progress Report* issued
- 12/07/05 GLBTS Integration Workgroup meets in Chicago, IL

2006

- 2/08/06 Presentation to Binational Executive Committee in Chicago on GLBTS successes and path forward by Gary Gulezian and Danny Epstein
- 2/16/06 GLBTS Integration Workgroup meets in Windsor, Ontario
- 3/07/06 to 3/08/06 GLBTS attendance (Ted Smith and Alan Waffle) at Environment Canada/Ontario Ministry of the Environment "Emerging Chemicals Workshop" in Toronto, Ontario
- 3/29/06 to 3/30/06 GLBTS attendance (Alan Waffle and Tricia Mitchell) at Environment Canada's "Workshop on Pharmaceuticals" in Burlington, Ontario
- 4/26/06 to 4/27/06 GLBTS attendance (Alan Waffle) at CEC SMOC meeting in Windsor, Ontario
- 4/28/06 GLBTS attendance (Ted Smith and Alan Waffle) at EC & US EPA GLWQA Review in Chicago
- 4/28/06 to 12/06 GLBTS participation as the US (Ted Smith) and Canadian (Alan Waffle) co-chairs of the Toxics Workgroup reviewing the GLWQA
- 5/17/06 GLBTS Stakeholder Forum is held in Toronto, Ontario
- 5/18/06 GLBTS Integration Workgroup meets in Toronto, Ontario
- 5/31/06 GLBTS presentation to Lake Superior LaMP Workgroup in Duluth, Minnesota, given by Alan Waffle
- 6/14/06 GLBTS presentation at Canadian Pollution Prevention Roundtable in Halifax, given by Alan Waffle
- 6/22/06 GLBTS attendance (Alan Waffle) at Great Lakes Cities Initiative meeting in Perry Sound, Ontario
- 7/31/06 Final *GLBTS 2005 Progress Report* posted at http://binational.net/bns/2005/2005-GLBTS-English-web.pdf
- 08/02/06 GLBTS and GLWQA presentations at DePaul University, Chicago, given by Danny Epstein and Susan Nameth
- 8/03/06 to 8/07/06 GLBTS promotion booth at Tall Ships event on the Chicago Waterfront, hosted by staff

from EC (Canadian lead Tricia Mitchell) and US EPA (US EPA Lead E.Marie Wines)
- 9/19/06 GLBTS Integration Workgroup meets in Chicago, IL
- 9/19/06 GLBTS presentations at Harbin Institute of Technology in Harbin, China, given by Alan Waffle
- 9/25/06 to 9/26/06 *International Workshop on Contaminated Site of Lindane and POPs in China*, Xian, China, given by Alan Waffle, S. Venkatesh, and Yi-Fan Li
- 10/11/06 to 10/12/06 GLBTS attendance (Alan Waffle) at State of Lake Huron Workshop in Honey Harbour, Ontario
- 11/01/06 GLBTS display booth at SOLEC

- 11/05/06 to 11/09/06 GLBTS attendance (Tricia Mitchell) at Society of Environmental Toxicology and Chemistry 27th Annual Meeting in Montreal
- 11/20/06 GLBTS Presentation at University of Toronto, given by Alan Waffle, S. Venkatesh, and Tricia Mitchell
- 12/06/06 GLBTS Stakeholder Forum is held in Chicago, IL
- 12/07/06 Draft *GLBTS 2006 Progress Report* issued
- 12/07/06 GLBTS Integration Workgroup meets in Chicago, IL
- 12/12/06 to 12/14/06 GLBTS attendance (Ted Smith and Alan Waffle) at first U.S. Conference Characterizing Chemicals in Commerce in Austin, Texas

2007

- 1/24/07 GLBTS presentation to Richview Collegiate physics students, Toronto, given by Alan Waffle and Tricia Mitchell
- 2/21/07 Integration WG meeting, held in Windsor
- 3/5/07 GLBTS attendance by Tricia Mitchell at Pharmaceuticals and Personal Care Products in the Canadian Environment: Research and Policy Directions, Niagara-on-the-Lake, Ontario
- 3/27/07 GLBTS attendance by Tricia Mitchell at Lake Ontario Contaminant Monitoring & Research Workshop - Planning for the 2008 Cooperative Monitoring Year, Grand Island, New York
- 3/28/07 GLBTS attendance by Tricia Mitchell at Lake Ontario LaMP Workgroup meeting, Grand Island, New York
- 4/16/07 GLBTS presentation by Danny Epstein at CEC Sound Management of Chemicals Meeting, Monterey, Mexico
- 5/23/07 BTS 10 Year Anniversary Evening Reception and Dinner, held in Chicago, along with Stakeholder Forum
- 5/24/07 to 5/25/07 BTS 10th Anniversary Workshop: Strategy's Future Focus and Challenges, held in Chicago
- 6/14/07 Presentation to Canadian Pollution Prevention Roundtable, Winnipeg, given by Alan Waffle
- 6/25/07 GLBTS attendance by Ted Smith at American Water Resources Association, Vail, Colorado
- 8/9/07 GLBTS attendance by Ted Smith at New England Interstate Water Pollution Control Commission on PPCPs, Portland, Maine
- 9/20/07 Integration WG meeting, held in Windsor
- 9/26/07 GLBTS presentation of proposal for new Substance and Sector Groups to Binational Executive Committee, by Danny Epstein
- 10/9/07 GLBTS presentation by Ted Smith at North American Hazardous Materials Management Association, San Diego, California
- 10/23/07 Attendance at Lake Ontario LaMP WG meeting, Grand Island, NY, by Alan Waffle
- 10/29/07 Attendance at "Making a Great Lake Superior," Duluth, Minnesota, by Alan Waffle
- 11/12/07 GLBTS presentations by Ted Smith at Society of Environmental Toxicology and Chemistry, Milwaukee, Wisconsin
- 11/15/07 GLBTS presentation at Univ. of Toronto, by Alan Waffle and Tricia Mitchell

- 12/12/07 Stakeholder Forum, held in Chicago, IL
- 12/13/07 Integration WG meeting, held in Chicago, IL

2008

- 01/25/08 to 01/26/08 GLBTS participation in Lake Superior Binational Forum in Two Harbors, MN, by Alan Waffle and Martin Nantel
- 05/09/08 Attendance at Lake Superior Binational Forum meeting, Nipigon, Ontario, by Martin Nantel
- 06/04/08 Stakeholder Forum, held in Burlington, Ontario
- 06/04/08 Integration WG meeting, held in Burlington, Ontario
- 06/11/08 to 06/12/08 GLBTS participation at Lake Superior LaMP WG meeting, Thunder Bay, Ontario, by Martin Nantel and Alan Waffle
- 09/02/08 to 09/04/08 GLBTS participation at Lake Superior LaMP WG meeting, Bayfield, Wisconsin, by Alan Waffle and Martin Nantel
- 09/25/08 Integration WG meeting, held in Chicago, IL
- 10/15/08 to 10/16/08 GLBTS presentation at Lake Erie LaMP meeting, Erie, PA, by Alan Waffle
- 10/22/08 to 10/23/08 GLBTS display at SOLEC, Niagara Falls, Ontario, by Alan Waffle and Martin Nantel
- 12/04/08 Stakeholder Forum and Integration WG meeting, held in Chicago, IL
- 12/09/08 to 12/11/08 GLBTS participation at *Meeting Current and Emerging Environmental Challenges within the Great Lakes - St Lawrence Basin: An Environment Canada Environmental Prediction Needs Assessment Workshop*, Cornwall, Ontario, by Alan Waffle and Martin Nantel

2009 and ongoing

- 01/30/09 to 01/31/09 Attendance at Lake Superior Forum Meeting, Superior, Wisconsin, by Martin Nantel
- 03/20/09 GLBTS presentation at Univ. of Toronto, by Alan Waffle and Martin Nantel
- 03/25/09 to 03/26/09 GLBTS attendance and presentation at the Lake Superior Think Thank (Restoring and Protecting the Lake Superior Basin: Actions Today and Ideas for Tomorrow), Thunder Bay, Ontario, by Alan Waffle and Martin Nantel
- 04/14/09 GLBTS attendance at BEC meeting, with presentation of proposed GLBTS reporting and operational changes, by Alan Waffle
- 04/21/09 to 04/22/09 GLBTS presentation at Lake Erie LaMP meeting, Erie, PA, by Alan Waffle
- 04/29/09 to 04/30/09 Attendance at Cooperative Science and Monitoring Workshop (by Alan Waffle and Martin Nantel) and Chemical Session lead (Martin Nantel), Duluth, MN
- 05/06/09 to 05/07/09 Attendance and GLBTS presentation at Lake Ontario Management Committee Meeting, Picton, Ontario, by Martin Nantel
- 06/02/09 to 06/04/09 Attendance at Lake Huron Binational Partnership Program, Les Chenaux, Michigan, by Alan Waffle
- 06/16/09 to 06/18/09 Attendance at Lake Superior LaMP WG meeting, Sault Ste. Marie, Ontario, by Alan Waffle and Martin Nantel
- 06/22/09 Cooperative Science and Monitoring Initiative (CSMI) Priority Setting Discussion for Lake Superior Chemicals, Burlington, Ontario, by Martin Nantel
- 06/23/09 GLBTS Status Update Teleconference with stakeholders
- 09/22/09 to 09/24/09 Attendance at Lake Superior LaMP WG meeting, Munising, Michigan, by Alan Waffle and Martin Nantel
- 09/23/09 GLBTS Status Update Teleconference with stakeholders
- 10/07/09 to 10/08/09 Attendance and workshop participation (Eutrophication and Chemicals of Emerging Concerns) at the IJC Biennial Meeting, Windsor, Ontario, by Alan Waffle, Martin Nantel, Tricia Mitchell, and Kelly Phillips
- 10/17/09 to 10/18/09 GLBTS green chemistry meeting with Executive Director of the Green Centre Canada,

Kingston, Ontario, by Alan Waffle
- 11/05/09 to 11/06/09 Attendance at Lake Superior Binational Forum Meeting, Thunder Bay, Ontario, by Martin Nantel
- 11/05/09 Release of the GLBTS 2008 Status Report (hard copy, web, CD)
- 11/20/09 to 11/20/09 GLBTS attendance at BEC meeting, by Alan Waffle and Tricia Mitchell
- 12/03/09 Stakeholder Forum and Integration WG meeting, held in Chicago, IL

Substance Activities: Mercury (Hg)

GLBTS Workgroup Activities and Reports

1998

- 3/23/98 Workgroup (WG) is formed at the first implementation meeting
- 5/5/98 WG conference call is held
- 8/24/98 *Background Information on Mercury Sources and Regulations* is posted on the GLBTS Website
- 9/10/98 Options Paper *Developing a Virtual Elimination Strategy for Mercury* is posted on the GLBTS Website
- 11/16/98 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- 11/17/98 GLBTS workshop on Potential Mercury Reductions at Electric Utilities is held in Chicago

1999

- 1/99 GLBTS web postings include: *Wisconsin Mercury Source Book* on community Hg reduction plans, findings of the Mercury Reduction at Electric Utilities workshop, and *Mercury Success Stories*
- 2/99 Information and FAQs on mercury fever thermometers posted on the GLBTS Website
- 3/99 GLBTS web postings include: The WDNR guide, *Mercury in your Community and Environment*, and a manual for hospitals, Reducing Mercury Use in Health Care
- 4/99 Workshop on community initiatives for reducing Hg
- 4/27/99 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 11/18/99 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- 11/99 Draft GLBTS Step 1&2 *Sources and Regulations* report for mercury is posted on the GLBTS Website

2000

- 5/16/00 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 6/00 GLBTS web page on Mercury Thermometers and FAQs is updated
- 8/00 Memo on progress in reducing mercury use posted on the GLBTS Website
- 9/1/00 A final draft GLBTS *Reduction Options* (Step 3) report for mercury is prepared and posted on the GLBTS Website on 9/29/00
- 10/17/00 Expansion of mercury web page links
- 11/18/00 WG meeting at the GLBTS Stakeholder Forum in Toronto

2001

- 5/17/01 WG meeting at the GLBTS Stakeholder Forum in Toronto
- 11/14/01 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2002

- 5/29/02 – 5/30/02 WG meeting at the GLBTS Stakeholder Forum in Windsor, Ontario
- 12/2/02 WG meeting in Chicago, IL on reducing impact of dental mercury
- 12/3/02 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2003

- 5/14/03 – 5/15/03 WG meeting at the GLBTS Stakeholder Forum in Windsor, Ontario
- 12/16/03 – 12/17/03 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2004

- 6/17/04 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 8/04/04 Workgroup report revised: *Options for Dental Mercury Reduction Programs: Information for State and Local Governments*
- 11/30/04 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2005

- 5/17/05 WG meeting in Toronto, Ontario
- 12/06/05 WG meeting in Chicago, IL

2006

- 02/06 WG finalizes Management Assessment for Mercury
- 5/17/06 WG meeting in Toronto, Ontario
- 12/06/06 WG meeting in Chicago, IL

2007

- 1/31/07 WG teleconference to discuss possible new challenge goals
- 12/12/07 WG meeting in Chicago, IL

2008 - 2009

- 6/03/08 WG meeting in Burlington, Ontario
- 11/17/09 to 11/18/09 2009 Mercury Science and Policy Conference with a Special Focus on the Northeast and Great Lakes Regions, held in Chicago, IL

Other Mercury Related Activities

1997 and Earlier

- Chlorine Institute voluntary mercury commitment to reduce mercury use by 50 percent by 2005
- 12/97 *Mercury Report to Congress* is released by US EPA

1998

- 5/8/98 Chlorine Institute releases progress report on voluntary mercury commitment
- 6/25/98 US EPA and AHA sign an MOU on reducing medical wastes

- 9/15/98 Three northwest Indiana steel mills commit to developing mercury inventories and reduction plans
- 10/98 IDEM household mercury collection efforts
- Dow Chemical Company commits to mercury reductions
- PBT Strategy grant to the Northeast Waste Management Officials' Association to encourage state mercury reduction efforts

1999

- 8/99 As part of 1998 agreement, mercury inventories at Indiana steel mills are completed
- 10/99 Mercury waste collection component of the Cook County (Illinois) Clean Sweep pilot begins
- Six Ontario hospitals sign MOU to voluntarily reduce Hg
- Pollution Probe investigates Hg reduction options for electrical products sector in Ontario
- Automotive Pollution Prevention Project efforts to phase-out Hg
- US EPA grant to Ecology Center of Ann Arbor: promoting mercury P2 in the health care industry
- Western Lake Superior Sanitary District (WLSSD) begins multimedia zero discharge pilot / focus on Hg
- Michigan Mercury Pollution Prevention Task Force
- 11/16/98 Draft *PBT National Action Plan* for Mercury is released by US EPA
- Total mercury used in lamps declines from an estimated 17 tons in 1994 to an estimated 13 tons in 1999, even though significantly more mercury-containing lamps are sold in 1999 than in 1994.

2000

- Chlorine Institute reports 42 percent reduction, production-adjusted, in mercury use
- US EPA, state agencies, and academic researchers conduct meetings with chlor-alkali industry representatives to coordinate mercury reduction projects
- Olin Corp. cooperates with US EPA, state, and academic researchers on mercury monitoring project at chlor-alkali plant
- Indiana steel mills complete mercury reduction plans; extend invitation to suppliers to commit to developing mercury inventories and reduction plans
- Auto Alliance commits to eliminate mercury switches in auto convenience lighting; New York DEC and Michigan DEQ implement mercury removal programs at auto scrap yards
- Hospitals for a Healthy Environment produces a Mercury Virtual Elimination Plan for hospitals under the AHA-US EPA MOU. State and local governments provide technical assistance to hospitals, and the National Wildlife Federation (NWF) continues its outreach and education efforts, signing up nearly 600 medical facilities to NWF's "Mercury Free Medicine Pledge."
- Wisconsin DNR and Department of Agriculture conduct a dairy mercury manometer replacement program; approximately 375 mercury manometers are recycled.
- University of Wisconsin extension creates a Website and list server to share information about mercury in schools.
- The Thermostat Recycling Corporation collects over 500 lbs of mercury from over 57,000 thermostats collected and processed from January 1, 1998 to June 30, 2000. The program is expanded to the Northeast and will gradually be expanded to include the entire U.S.
- The Great Lakes Dental Mercury Reduction Project funded by the Great Lakes Protection Fund produces a brochure template: *Amalgam Recycling and Other Best Management Practices*. Great Lakes Dental Associations reprint and distribute this document to their memberships. The University of Illinois-Chicago dental school and the Naval Dental Research Institute conduct research on controlling mercury in dental wastewater and help to educate dentists about best management practices.
- Coalitions including Health Care Without Harm and the National Wildlife Federation successfully encourage several national retailers to stop the sale of mercury-containing thermometers to the public. Duluth,

Minnesota, Ann Arbor Michigan, unincorporated areas of Dane County, Wisconsin, and several Dane Country municipalities, ban the sale of mercury thermometers.

2001

- 651 hospitals join the National Wildlife Federation's Mercury-Free Hospitals campaign
- Ispat-Inland Indiana Harbor Works, Bethlehem Steel-Burns Harbor Division, US Steel-Gary Works, the Delta Institute, and Lake Michigan Forum created the *Guide to Mercury Reduction in Industrial and Commercial Settings*
- Mercury Switch-out Pilot Program launched by Pollution Probe, Ontario Power Generation, Ontario Ministry of the Environment, and Environment Canada to collect mercury switches from old vehicles
- 2/21/01 A workshop entitled "Extended Producer Responsibility and the Automotive Industry" is sponsored by the Canadian Autoworkers Union'sWindsor Regional Environment Council and Great Lakes United

2002

- 2/27/02 Great Lakes United kicks off series of information-sharing sessions about auto mercury-switch removal programs for State agency staff
- 4/5/02 Chlorine Institute releases its *Fifth Annual Report to EPA*, showing a 75 percent reduction in mercury use by the U.S. chlor-alkali industry between 1995 and 2001, more than meeting this sector's commitment to reduce mercury use 50 percent by 2005
- 10/1/02 Thermostat Recycling Corporation announces that it collected 28,000 thermostats and 231 lbs of mercury in the first half of 2002, a 15 percent increase from mercury collections in the first half of 2001. The program began to serve the 48 continental U.S. States in the fall of 2001.
- 10/18/02 The Hospitals for a Healthy Environment (H2E) program has 335 partners representing 1,019 facilities: 347 hospitals, 618 clinics, 22 nursing homes and 32 other types of facilities. These partners are health care facilities that have pledged to eliminate mercury and reduce waste, consistent with the overall goals of H2E.

2006

- 6/06/06 US EPA reaffirms Clean Air Mercury Rule (CAMR)
- 8/06 National Vehicle Mercury Switch Recovery Program established by agreement among vehicle manufacturers, steelmakers, vehicle dismantlers, auto shredders, brokers, the environmental community, state representatives, and the US EPA.
- 12/9/06 EC published a Proposed Notice under Part 4 of the Canadian Environmental Protection Act of 1999 outlining proposed requirements to prepare and implement pollution prevention plans for mercury releases from mercury switches in end-of-life vehicles processed by steel mills. The Notice targets vehicle manufacturers and steel mills.
- 12/20/06 EC posted a Risk Management Strategy (RMS) for Mercury-containing products and is holding consultations to obtain the views of Canadians. The RMS provides a framework for the development of control instruments to manage the environmental effects of mercury used in products.

2007

- 2/07 NWF issues report, *Putting the Brakes on Quicksilver: Removing Mercury from Vehicles in Ohio*.
- 4/17/07 *Report to Congress: Mercury Contamination in the Great Lakes* released. Available at http://www.arl.noaa.gov/data/web/reports/cohen/NOAA_Great_Lakes_Mercury_Report.pdf
- 5/07 Chlorine Institute releases its *Tenth Annual Report to EPA*, showing an 89 percent capacity-adjusted reduction in mercury consumption by the U.S. chlor-alkali industry between 1995 and 2005.

- 8/07 GLRC released draft *Great Lakes Mercury in Products Phase-Down Strategy* for public comment.
- 9/07 Switch the 'Stat program launched by the Clean Air Foundation in partnership with 850 heating and cooling contractors in Ontario, to encourage programmable thermostats and collect mercury-containing thermostats.

2008 and Ongoing

- 8/07 Draft *Mercury Phase-Down Strategy* posted to GLRC website.
- 6/08 Final *Mercury Phase-Down Strategy* posted to GLRC website.
- 04/09 EC posts Proposed P2 Notice in *Canada Gazette Part I* requiring targeted dental facilities to prepare and implement BMPs regarding mercury releases from dental amalgam waste.
- 9/09 Draft *Mercury Emission Reduction Strategy* posted for comment to GLRC website.

Substance Activities: Polychlorinated Biphenyls (PCBs)

GLBTS Workgroup Activities and Reports

1998 and Earlier

- As of January 1993, approximately 25,000 tonnes of high-level PCBs are either in use or in storage in Ontario; 1529 active PCB storage sites in Ontario
- 3/23/98 WG is formed at the first implementation meeting
- 6/15/98 WG requests that the IG develop a strategy on sediments
- 11/10/98 Options Paper *Virtual Elimination of PCBs* is posted on GLBTS Website
- 11/16/98 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

1999

- 4/27/99 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 11/18/99 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- 11/99 Draft *GLBTS Step 1&2 Sources and Regulations* report for PCBs is posted on the GLBTS Website
- WG solicits and gains commitment of 3 U.S. auto manufacturers to reduce PCBs
- WG solicits commitment of steel producers to reduce PCBs

2000

- 5/16/00 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- Final draft GLBTS Step 3 *Reduction Options* report for PCBs is prepared (7/14/00) and posted (9/29/00) on the GLBTS Website
- WG continues to use PCB reduction commitment letters, through EC and US EPA, to seek commitments to reduce PCBs. Specific companies are targeted, primarily major owners of PCB transformers and capacitors, and associations, such as CGLI
- WG solicits and gains commitment to reduce PCBs from 2 Canadian auto manufacturers, 4 Canadian steel producers, and over 30 municipal electrical utilities in Ontario
- WG leaders and Council of Great Lakes Industries (CGLI) finalize outreach letters used to seek PCB reduction commitments from trade associations. CGLI identifies specific trade associations to begin outreach. EC mails letters to trade initial associations. US EPA mailings to follow.

- WG begins to compile case study reports on reasons why companies remove their PCBs
- WG begins to collect photographs of PCB-containing electrical equipment to assist potential owners with identification of equipment which may contain PCBs
- WG drafts a fact sheet on PCB-containing submersible well pumps to be used for outreach to potential users of wells and servicers of well pumps.
- As of April 2000, approximately 7,500 tonnes of high-level PCBs are either in use or in storage in Ontario; 1,191 active PCB storage sites in Ontario

2001

- WG continues to mail letters to companies and trade associations seeking commitments to phase-out PCBs
- WG prepares case studies submitted by Bethlehem Steel Corporation's Burns Harbor Division and ComEd Energy Delivery, a unit of Chicago-based Exelon Corporation, for posting on the GLBTS Website
- 1/01 PCB federal databases are updated for Canada.
- 5/01 PCB WG progress meeting held in Toronto, Ontario, Canada. WG discusses two reasons that companies are unable to commit immediately to PCB reductions: 1) reduction/replacement is dependent on companies' internal planning and budgeting cycle; 2) reduction/ replacement is tied to market conditions. US EPA and EC will continue mailing out the voluntary reduction and commitment letters to the priority sectors and associations seeking additional commitments to reduce PCBs.
- 5/17/01 WG meeting at the GLBTS Stakeholder Forum in Toronto
- 7/01 US EPA compiles and analyzes data for 1995-1999 submitted by U.S. PCB disposers
- 8/29/01 WG posts photographs of electrical equipment which may contain PCBs (transformers, and capacitors) to GLBTS Website to help increase awareness of the types of equipment that may contain PCBs
- 9/01 In coordination with LaMP activities, EC mails a package of information to all small quantity PCB owners (over 300 owners) in the Lake Superior and Lake Erie Basins to help raise awareness of PCB initiatives underway in support of the GLBTS. The information package contained a copy of PCB Owners Outreach Bulletin, fact sheets, and maps of PCB Storage sites in the Lake Erie and Lake Superior Basins.
- 11/01 PCB WG meeting is held in Chicago, IL. WG discusses the need for more outreach, especially toward small and medium sized companies. Representatives of General Motors outline the company's plan to phase-out all PCB materials from its North American facilities.
- As of April 2001, 80 percent of high-level PCBs (Askarel > 1 percent, 10,000 ppm) had been destroyed in Ontario, Canada; however only 25 percent of low-level PCBs were destroyed, mostly from stored contaminated soil from a contaminated site cleanup in Ontario.
- As of April 2001, approximately 6,000 tonnes of high-level PCBs are either in use or in storage; 992 active PCB storage sites in Ontario.
- 8/30/01 Fact sheet posted to GLBTS Website: PCBs in Submersible Well Pumps
- 11/14/01 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2002

- WG continues to modify BNS-PCB Website based on recommendations received in an email survey conducted by EC and US EPA in November 2001
- 5/02 WG meeting is held at the GLBTS Stakeholder Forum in Windsor, Ontario
- 5/02 Hydro One representative states that the company is free of all high-level PCBs but still has several small stations and other sources of low-level PCBs. Hydro One has introduced a PCB management program that extends to the year 2020.
- 5/02 MOE representative presents a strategy to implement an annual charge for having equipment with PCBs. Amendments for *Regulation 362* are proposed, including the addition of a schedule of destruction targets.

- 10/02 Approx. 400 PCB commitment letters are sent to school boards and other sensitive sites in Ontario.
- 10/02 Canada develops a new (draft) plan of outreach and recognition to try to increase the rate of PCB phase-out in Canada. The main elements of the draft plan are to identify and recognize contributions made by individual companies or their industry associations that go beyond regulatory requirements and to publicize success stories.
- As of April 2002, 84 percent of high-level PCBs (Askarel > 1 percent, 10,000 ppm) had been destroyed in Ontario, compared to 1993.
- As of April 2002, approximately 4,147.4 tonnes of high-level PCBs are either in use or in storage in Ontario; 916 active PCB storage sites in Ontario.

2003

- 5/14/03 WG meeting at the GLBTS Stakeholder Forum in Windsor, Ontario
- 9/11/03 PCB Reduction Recognition Awards presented to Enersource Hydro, Hydro One, Slater Steel, and Stelpipe Ltd.
- 12/16/03 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2004

- 6/17/04 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 6/17/04 PCB Reduction Recognition Awards presented to City of Thunder Bay and Canadian Niagara Power
- 11/30/04 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2005

- 5/17/05 WG meeting in Toronto, Ontario
- 12/06/05 WG meeting in Chicago, IL

2006

- 5/17/06 WG meeting in Toronto, Ontario
- 12/06/06 WG meeting in Chicago, IL. Management Assessment for PCBs finalized.

2007

- 12/12/07 WG meeting in Chicago, IL

2008

- 6/03/08 WG meeting in Burlington, Ontario
- 12/03/08 WG meeting in Chicago, IL

2009

- 12/01/09 WG meeting in Chicago, IL

Other PCB Related Activities

1999 and Earlier

- US EPA finalizes PCB regulations which include a requirement for U.S. owners to register their PCB transformers
- EC and Ontario government hold two workshops on PCB management in the Toronto area
- 10/99 PCB waste collection component of the Cook County (Illinois) PCB/Hg Clean Sweep pilot begins
- U.S. PCB transformer registration database is updated
- Requests for voluntary PCB reduction commitments are mailed to automotive, iron & steel, and municipal electrical power utilities in Ontario

2000

- Region 5 PCB Phasedown Program and pilot phasedown enforcement policy are finalized
- A PBT workgroup continues to work on a National Action Plan for PCBs
- 2/00 EC mails survey to approximately 500 registered owners of in-use PCB equipment in Ontario, requesting updated information
- Cook County PCB/Hg Clean Sweep pilot concludes
- 11/00 Canada mails letter to over 2000 registered PCB waste storage owners/managers in Ontario for a recent update of their stored PCB inventory which will be used to modify federal databases for better tracking and monitoring
- Update and modification of Federal PCB databases started in 2000 and will continue until completion in 2003
- Three Canadian Federal PCB Regulations are being amended: (1) Chlorobiphenyl Regulation; (2) Storage of PCB Material Regulations; (3) PCB Export Regulations
- Extensive Public Consultation is conducted during summer and fall of 2000 and will continue

2001

- 5/2/01 Final Reclassification of PCB and PCB-contaminated Electrical Equipment rule becomes effective
- US EPA finalizes a rule on Return of PCB Waste from U.S. Territories Outside the Customs Territory of the U.S. The rule clarifies that PCB waste in U.S. territories and possessions outside the customs territory of the U.S. may be moved to the customs territory of the U.S. for proper disposal at approved facilities.
- EC updates National PCB In-Service Inventory from survey of registered owners and prepares fact sheet
- EC's regulatory amendment process proposes the strengthening of federal regulations regarding PCB management

2002

- 42 electrical utilities submit voluntary reduction commitment letters to Environment Canada
- Algoma voluntarily commits to eliminate 71,103 kgs (44,400 litres) of PCBs by Dec. 2005
- Approximately 27 school boards and sensitive sites respond to PCB commitment letters; 18 of those companies reported that all PCBs were eliminated from their inventories; 3 reported that all high-level PCBs were eliminated from their inventories

2003

- Amended Canadian PCB regulations are expected to be published in the *Canada Gazette I and II* in 2003. These regulations will target phase-out of high-level PCB use by 2007, low-level PCB use by 2014, and prohibit storage after 2009.

2005

- 06/05 An event report on the May 2005 PCB Award Ceremony is published under the title: "Ontario companies recognized for PCB phase-out" page 8, Canadian HazMat Magazine, June/July 2005, accessible at www.hazmatmag.com.

2006

- 11/04/06 Proposed Canadian PCB regulations are published in the *Canada Gazette I*.

2007

- 1/3/07 EC received comments on PCB regulations from 43 stakeholders (following 60-day comment period).
- 10/25/07 EC proposed PCB Regulations Policy Changes to EP Board regarding end-of-use deadlines for lower risk PCBs, criteria for proposed extension system, and implementation approach for proposed extension system.
- 9/20/07 City of Toronto and Dofacso Inc. received PCB Phase-Out Awards for reductions in the number of PCB transformers in use.

2008 and Ongoing

- 9/17/08 Final PCB Regulations are published in the *Canada Gazette II*.
- 9/09 US EPA begins PCBs-in-Building Materials outreach program for schools and childcare facilities.
- 12/09 EC's mandatory phase-out deadline for equipment containing high-level PCBs (over 500 ppm) and low-level PCBs (50 to 500 ppm) in sensitive locations in Canada.

Substance Activities: Dioxins/Furans

GLBTS Workgroup Activities and Reports

1998

- 3/23/98 WG is formed at the first implementation meeting
- 11/16/98 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

1999

- 4/27/99 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 6/1/99 WG Conference call: sources discussions
- 7/7/99 WG Conference call: sources discussions
- 9/7/99 WG Conference call: developing a decision tree source prioritization process
- 10/5/99 WG Conference call: finishing development of a decision tree process
- 11/18/99 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- 12/7/99 WG Conference call: application of the decision tree process

2000

- 1/11/00 WG Conference call: continuing the decision tree process
- 2/1/00 WG Conference call; decision made to initiate a Burn Barrel Subgroup
- 3/7/00 WG Conference call: continuing the decision tree process
- 4/4/00 WG Conference call: continuing the decision tree process
- 4/4/00 Burn Barrel Subgroup has inaugural teleconference
- 4/25/00 Burn Barrel Subgroup teleconference: strategy matrix discussed
- 5/2/00 WG Conference call: continuing the decision tree process
- 5/16/00 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario: decision tree process is completed
- 5/26/00 GLBTS draft *Step 1&2 Sources and Regulations* report is prepared
- 7/11/00 WG Conference call: developing reduction projects for high priority sectors
- 8/1/00 Burn Barrel Subgroup teleconference: discussion Terms of Reference; link to Lake Superior LaMP
- 8/18/00 An addendum to the GLBTS Draft *Sources and Regulations* report is prepared to addressed the newly released U.S. Dioxin Reassessment and the draft report is posted (9/29/00) on the GLBTS Website
- 9/12/00 WG Conference call: developing reduction projects
- 9/12/00 Burn Barrel Subgroup teleconference: discussion of Chisago County "Buyback" program; discussion of survey questions regarding state/local regulatory frameworks, and garbage quantity/quality questions.
- Final GLBTS Step 3 *Reduction Options* report is prepared (9/27/00) and the report is posted (9/29/00) on the GLBTS Website
- 11/14/00 Burn Barrel Subgroup teleconference: outline of a strategy document prepared.
- 11/00 Discussion papers on Landfill Fire and Incinerator Ash Management prepared for workgroup review.

2001

- The WG continues to collect information regarding emissions from steel manufacturing, landfill fires, and incinerator ash management
- 1/16/01 Burn Barrel Subgroup teleconference: Burn Barrel Strategy
- 2/6/01 WG Conference call
- 2/13/01 Burn Barrel Subgroup teleconference: Review presentation for Integration Workgroup
- 3/13/01 Burn Barrel Subgroup teleconference: Status of efforts to prepare regulatory profile
- 4/10/01 Burn Barrel Subgroup teleconference: Proposal for US EPA funding of subgroup activities
- 5/8/01 Burn Barrel Subgroup teleconference: Review Strategy/ Implementation Plan document
- 5/17/01 WG meeting at the GLBTS Stakeholder Forum in Toronto: WG approves Burn Barrel Strategy/ Implementation Plan document; Canadian and US presentations on wood preservation
- 6/12/01 Burn Barrel Subgroup teleconference: Implementation activities for Summer/Fall
- 6/22/01 Burn Barrel Subgroup receives $55k of US EPA PBT funding
- 10/9/01 Burn Barrel Subgroup teleconference: Regional Lake Superior campaign
- 11/6/01 Burn Barrel Subgroup teleconference: Sharing information
- 11/14/01 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- 12/18/01 Burn Barrel Subgroup teleconference: Sharing information

2002

- 2/12/02 Burn Barrel Subgroup teleconference: web page initiation, bylaws/ordinance discussion.
- 3/19/02 Burn Barrel Subgroup teleconference: web page & list serve development, outreach updates
- 4/5/02 Lake Superior Region workshop on household garbage burning issue – Thunder Bay, ON

- 4/16/02 Burn Barrel Subgroup teleconference: web page & list serve development
- 4/24/02 WG Conference call: discussing ash management
- 5/14/02 Burn Barrel Subgroup teleconference: finalize web page, prepare for Windsor GLBTS meeting
- 5/30/02 WG meeting at the GLBTS Stakeholder Forum in Windsor: demonstration of newly launched subgroup Website "Trash and Open Burning in the Great Lakes". The WG meeting was held jointly with the HCB/B(a)P WG due to common issues that are of interest to both workgroups.
- 6/18/02 Burn Barrel Subgroup teleconference: Planned activities for summer, addressing "burners" for sale; purchase Website domain name www.openburning.org
- 7/24/02 WG Conference call: discussing the treated wood issue
- 9/10/02 Burn Barrel Subgroup teleconference: Updates on activities in various jurisdictions
- 11/13/02 WG Conference call: discussing a pilot project on the treated wood issue

2003

- 3/18/03 Burn Barrel Subgroup teleconference: Exploring partnerships with health organizations
- 5/14/03 WG meeting at the GLBTS Stakeholder Forum in Windsor, Ontario
- 6/3/03 Burn Barrel Subgroup teleconference: US EPA Office of Solid Waste outreach materials
- 7/31/03 WG teleconference: Draft two-year workplan
- 9/9/03 Burn Barrel Subgroup teleconference: WDNR's "Air Defenders" kit
- 11/4/03 Burn Barrel Subgroup teleconference: Addressing suppliers of small backyard incinerators
- 11/4/03 WG teleconference: Draft two-year workplan; finalizing the Burn Barrel Strategy
- 12/16/03 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2004

- 3/02/04 WG teleconference: Progress on issue papers
- 3/09/04 Burn Barrel Subgroup teleconference
- 5/11/04 Burn Barrel Subgroup teleconference
- 6/04 Draft issues papers prepared on *Emissions from Agricultural Burning, Structure Fires, Tire Fires, and Wildfires and Prescribed Burning*
- 6/17/04 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 9/14/04 Burn Barrel Subgroup teleconference
- 9/09/04 Burn Barrel Subgroup teleconference
- 10/14/04 WG teleconference: Draft *Management Assessment for Dioxins*
- 11/30/04 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2005

- 05/17/05 WG meeting in Toronto, Ontario
- 12/06/05 WG meeting in Chicago, IL

2006

- 05/17/06 WG meeting in Toronto, Ontario
- 12/06/06 WG meeting in Chicago, IL

2007

- 02/07/07 WG conference call to review management outcomes of framework assessment for dioxins/furans and to discuss the status of the WG
- 03/20/07 Burn Barrel Subgroup teleconference

- 05/29/07 Burn Barrel Subgroup teleconference
- 07/10/07 Burn Barrel Subgroup teleconference
- 09/25/07 Burn Barrel Subgroup teleconference
- 11/13/07 WG conference call to discuss the Dioxin Decision Tree
- 12/12/07 WG meeting in Chicago, IL

2008

- 04/15/08 Burn Barrel Subgroup teleconference
- 06/24/08 Burn Barrel Subgroup teleconference
- 10/14/08 Burn Barrel Subgroup teleconference

2009 and Ongoing

- 03/17/09 Burn Barrel Subgroup teleconference
- 08/27/09 Burn Barrel Subgroup teleconference

Other Dioxin/Furan Related Activities

1999 and Earlier

- WLSSD begins multimedia zero discharge pilot / focus on dioxins

- Two Ontario utilities eliminate use of PCP in treated poles

2000

- 1/00 WLSSD report on open barrel burning practices is released
- 2/00 Wood stove changeover pilot programs in Traverse City, MI, and Green Bay, WI
- 6/12/00 draft chapters of the *U.S. Dioxin Reassessment* for external scientific review are released
- 9/28/00 Three draft chapters of the *U.S. Dioxin Reassessment* for SAB review are released

2001

- February 2001, *Release of National Inventory of Releases of Dioxins and Furans, Updated Edition*, by EC
- May 2001, Release of report "Characterization of Organic Compounds from Selected Residential Wood Stoves and Fuels" by EC

2002

- PCP re-registration review proceeding as joint Canada/U.S. endeavor

2003

- 7/18/03 CEC draft Phase One North American Regional Action Plan on Dioxins and Furans, and Hexachlorobenzene available for public comment
- *Ash Characterization Study* in Ontario
- Secondary metal smelter release inventory study in Ontario
- US EPA develops Backyard Trash Burning Website and brochures available at www.epa.gov/nsw/backyard
- Public release of first US National Dioxin Air Monitoring Network (NDAMN) ambient air monitoring data
- Canada-wide Standards for iron sintering and steel manufacturing endorsed in March 2003

- Release of Wisconsin "Air Defenders" Kit for Burn Barrel education
- Dioxin sampler added at an Integrated Atmospheric Deposition Network (IADN site), Burnt Island

2004

- US EPA compiles case studies of open burning reduction efforts

2007

- 1/31; 2/11; 5/22; 10/3/07 US EPA staff conducted open burning outreach presentations at conferences and meetings for local officials in Ohio, Florida, Pennsylvania, and San Diego, among others.

2008 and Ongoing

- 10/14/09 New York (NYSDEC) adopts ban on most open burning, including agricultural plastics burning, statewide.

Substance-Specific Activities: Pesticides

GLBTS Workgroup Activities and Reports

1998

- 3/23/98 WG is formed at the first implementation meeting
- 11/16/98 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- 12/31/98 Draft GLBTS Challenge report for the Level I pesticides is posted on the GLBTS Website

1999

- 4/27/99 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 11/18/99 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2000

- 5/16/00 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- GLBTS U.S. Pesticides Challenge Report: *The Level 1 Pesticides in the Binational Strategy* is finalized (3/1/00) and posted (9/29/00)
- 5/00 EC announces that with the cooperation of PMRA they have reevaluated their position on Level I pesticides, and that based on all available information have met the Level I challenge.

2001

- WG reviews pollution prevention opportunities for Level II pesticides (endrin, heptachlor, lindane and HCH, tributyl tin, and pentachlorophenol) and begins preparing report

Other Pesticide Related Activities

1999 and Earlier

- 10/96 EC prepares report: *Canada-Ontario Agreement Objective 2.1: Priority Pesticides Confirmation of No Production, Use, or Import in the Commercial Sector in Ontario*
- US EPA funding to four existing Clean Sweep programs for pilot data collection efforts for Level I pesticides

2000

- Draft National Action Plan for Level 1 Pesticides under the U.S. National PBT Initiative completed and released for review and public comment
- PBT Pesticides Workgroup reviewing toxaphene remediation in Brunswick, GA
- Level I PBT pesticides (except mirex) are regularly collected by ongoing Clean Sweep programs
- Phase-out of the Level II Pesticides lindane and tributyl tin compounds are the subject of bi-national negotiations through pesticide regulatory agencies in the U.S. and Canada

2001

- Waste pesticide collections (Clean Sweeps) continue
- 10/5/01 Members of the world's primary maritime organization, the International Maritime Organization, adopt the International *Convention on the Control of Harmful Anti-fouling* Systems on Ships. The agreement calls for a global prohibition on the application of organotin compounds by January 1, 2003, and a complete prohibition by January 1, 2008.

2002

- PCP re-registration review proceeding as joint Canada/U.S. endeavor

2004 and Ongoing

- At the end of 2004, lindane use was discontinued in Canada.
- In 2006 U.S. manufacturers agreed to relinquish the remaining registrations for lindane (use will cease in the U.S. in 2009).

Substance-Specific Activities: Hexachlorobenzene (HCB)/Benzo(a)pyrene (B(a)P)

GLBTS Workgroup Activities and Reports

1998

- 3/23/98 WG is formed at the first implementation meeting
- 9/98 & 10/98 Discussions are held with the pesticide manufacturing, chlorinated solvent manufacturing, and petroleum refinery industries regarding their emission levels, and to determine any success stories,

pollution prevention opportunities, and other planned or possible emission reduction actions
- 11/16/98 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

1999

- 4/27/99 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 11/18/99 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- 11/99 Draft GLBTS Step 1&2 *Sources and Regulations* Reports for B(a)P and HCB are posted on the GLBTS Website

2000

- 5/16/00 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- Discussions held with the U.S. Scrap Tire Management Council and scrap tire managers in the Midwest
- 6/15/00 Final drafts GLBTS Step 3 *Reduction Options* reports for B(a)P and HCB are prepared
- 7/12/00 Final drafts GLBTS Step 3 *Reduction Options* reports for B(a)P and HCB are posted on the GLBTS Website
- 9/21/00 WG conference call is held
- 10/00 draft Canadian Steps 1& 2 reports for HCB/B(a)P (PAHs) circulated to stakeholders and workgroup members for comments

2001

- 5/17/01 WG meeting at the GLBTS Stakeholder Forum in Toronto
- 11/14/01 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- Canada implements Strategic Options Processes with steel mills and wood preservers
- Algoma Steel signs an *Environmental Management Agreement* with EC and Ontario MOE to address environmental priorities
- A Woodstove Changeout Program is held in Georgian Bay, Ontario, in conjunction with the Hearth Products Association of Canada

2002

- 5/30/02 WG meeting at the GLBTS Stakeholder Forum in Windsor, Ontario
- Wood stove change-out outreach material in development, a Website may be developed to promote change-outs and share information with stakeholders
- Petroleum refinery B(a)P emissions analysis completed
- Preparation of incentives for scrap tire pile recycling begins
- Status and potential for reduction of newly inventoried primary aluminum B(a)P emissions determined
- Work with Council of Great Lakes Industries (CGLI) and pesticide industry continues to determine pesticide HCB contaminant levels
- Success stories of reductions in HCB TRI releases from the chemical industry are identified
- Outreach activities (e.g., Website development, preparation of consumer information sheets) are conducted to increase public awareness of environmental impacts, safe handling, and applications of used treated wood
- WG seeks to improve linkages and integration of release information and environmental data on persistent toxics
- WG works to fill release data gaps, resolve questions about company NPRI release estimates for Level I substances, and develop reduction projects with stakeholders
- 12/3/02 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2003

- 5/14/03 WG meeting at GLBTS Stakeholder Forum in Windsor, Ontario
- Work with CGLI and pesticide industry, to determine pesticide HCB contaminant levels, continues
- Rubber Manufacturers Assn. provides detailed information on scrap tire management in the Great Lakes Basin
- Resource needs identified to successfully implement a Scrap Tire Outreach Plan
- B(a)P emissions from coke ovens in basin continue to decline as a result of shutdowns and regulations
- Work on more accurate B(a)P inventory (especially for air emissions)
- Several conference calls held on Woodstove Smoke Reduction contract to encourage best practices and develop outreach materials
- Natural Resources Canada *Burn it Smart!* campaign conducts over 300 residential wood-burning workshops across Canada; campaign presentation to be updated to include wood stove change-out and more workshops planned for Ontario
- Initial discussions held with Canadian Vehicle Manufacturers' Association on verification of B(a)P release estimates for the on-road motor vehicle sector
- 12/16/03 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2004

- 6/17/04 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- US EPA wood stove/fireplace initiatives: media outreach package, Website, fact sheets and labeling program promoting EPA-certified stoves and clean/safe wood burning practices.
- Fifty-one *Burn it Smart!* public education workshops delivered in 40 Ontario rural and First Nations communities in 2004
- Work with CGLI and pesticide industry to determine pesticide HCB contaminant levels, continues
- Re-assessment of Ontario HCB/B(a)P releases from use of pentachlorophenol-treated and creosote-treated wood products.
- 11/30/04 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL

2005

- 5/17/05 WG meeting in Toronto, Ontario
- Prepared *Management Assessment Reports for HCB and B(a)P* using the General Framework to Assess Management of GLBTS Level 1 Substances
- 31 *Burn it Smart!* workshops held in various First Nation communities, Ontario communities and 2 U.S. border cities
- Conducted tests on artificial logs to determine emissions
- Worked with CGLI, pesticide industry, and the Pest Management Regulatory Agency of Health Canada to determine HCB releases from pesticide application
- Surveyed 2001 Georgian Bay Wood Stove Changeout and Education seminar attendees to follow-up on changes to their wood burning practices
- Continued to promote scrap tire pile inventory development and mapping, and cleanup initiatives
- 12/06/05 WG meeting in Chicago, IL

2006

- 05/17/06 WG meeting in Toronto, Ontario
- 17 *Burn it Smart!* workshops held in various First Nation and tribal communities, Ontario communities, and two U.S. border cities. Approximately 220 people attended these workshops.
- Initiated a North American HCB modeling project to evaluate long-range transport impacts

- Worked with CropLife Canada and Pest Management Review Agency to improve estimates of Canadian HCB releases from pesticide application.
- New York Academy of Sciences held a conference call in October with stakeholders from both U.S. and Canada to discuss estimates of PAH releases from creosote-treated wood.
- 12/06/06 WG meeting in Chicago, IL

2007

- 09/07 A US EPA gold medal for exceptional service awarded for the production of *Scrap Tire Cleanup Guidebook*
- 12/12/07 WG meeting in Chicago, IL

2008

- 06/03/08 WG meeting in Burlington, Ontario
- 12/03/08 WG meeting in Chicago, IL

2009

- 12/01/09 WG meeting in Chicago, IL

Other HCB/B(a)P Related Activities

1999 and Earlier

- Dow Chemical Company commits to HCB reductions
- Two Ontario utilities eliminate use of PCP in treated poles
- U.S. chlorothalonil manufacturer reduces HCB content through process improvements
- 10/99 Draft Report, *Global HCB Emissions* (Robert Bailey, 1999), is distributed to the WG
- 1/99 wood stove changeover pilot program for Eastern Ontario

2000

- 1/00 WLSSD report on open barrel burning practices is released
- 2/00 Wood stove changeover pilot programs in Traverse City, MI, and Green Bay, WI
- PBT workgroups continue to work on draft *National Action Plans* for HCB/B(a)P
- 5/5/00 Robert Bailey prepares report, *HCB Concentration Trends in the Great Lakes*, for the WG

2001

- 2/01-4/01The Hearth Products Association expands the Great Lakes Great Stove Changeout Program to 12 States
- 6/01 US EPA issues an administrative order requiring Magnesium Corporation of America (Rowley, UT) to ensure proper handling, containment, and disposal of anode dust found to contain high levels of HCB (>12,000 ppm), as well as dioxins, PCBs, and chromium

2002

- Source release information to improve inventories collected through voluntary stack testing
- An emission testing program for wood burning in fireplaces, wood stoves, and pellet stoves developed and implemented with partners to fill information gaps
- PCP re-registration review proceeding as joint Canada/U.S. endeavor

2003

- 7/18/03 CEC draft Phase One North American Regional Action Plan on Dioxins and Furans, and Hexachlorobenzene available for public comment
- A US EPA rule to control emissions (including HCB) from hydrochloric acid production is promulgated
- The "Voluntary Woodstove/Fireplace Smoke Reduction Activities and Outreach Materials" contract awarded by US EPA
- A US EPA rule for the control of coke oven battery stack emissions (including B(a)P) is promulgated
- HCB added to CEPA listing of prohibited toxic substances; proposed regulation published to prohibit products with concentrations greater than 20 ppb

2004

- Twelve Wood Energy Technology Transfer Inc. training workshops held in Ontario
- US EPA *Scrap Tire Pile Mitigation Support Project* underway promoting mapping and cleanup of tire piles
- Scrap tire pile cleanup forum held in Chicago on February 23 – 24, 2004
- Proposed Ontario Tire Stewardship scrap tire diversion program awaiting approval from the Ontario Ministry of the Environment
- Independent third party audits verify Ontario's four metallurgical coke producers meeting reduction goals set out in best practice manual for controlling PAH (includes B(a)P) releases)

2005

- Amendments to U.S. *Air Toxics Standards for Coke Oven Batteries* came out in April 2005.
- US EPA finalized rules on wastewater discharges from iron and steel facilities.
- Developing U.S. best practices Scrap Tire Cleanup Guidebook.
- Partnered with The Home Depot to promote *Burn it Smart!* at six stores in Eastern Ontario.
- Partnered with the Puget Sound Clean Air Agency to conduct more emissions testing on wax firelogs and regular cordwood.
- Commenced Ontario B(a)P mapping project to highlight priority areas.

2006

- US EPA initiated Green Stoves Labeling Program.
- US EPA initiated studies to evaluate Outdoor Wood Boilers.
- EC commenced information gathering exercise with Hearth, Patio and Barbecue Association of Canada on outdoor wood boiler usage in Ontario and Eastern Canada.
- EC completed B(a)P mapping project for the Great Lakes Basin by adding Ontario information
- EC worked with Ontario Ministry of the Environment and initiated other projects to improve the emission inventories of HCB/B(a)P.
- New York Academy of Sciences published an Ecological Assessment and Pollution Prevention Report detailing PAH releases from all sources in New York and New Jersey Harbor
- *Burn-it-Smart!* public education information provided at Cottage Life Shows in Toronto in April and November, at the International Plow Match in Peterborough in September, and the Home Hardware national sales meeting in St. Jacobs (north of Waterloo) in September
- EC produced final report on artificial log study with Puget Sound Clean Air Agency
- EC partnered with Hearth, Patio and Barbecue Association on emission testing of five conventional wood stoves and drafted report
- Ontario Ministry of the Environment announced that the Used Tire Program was deferred beyond the immediate future
- US EPA initiated a Mid-West Clean Diesel Initiative in Region 5 to reduce diesel emissions

2007

- 04/07 Agreement between US EPA and major outdoor wood boiler manufactures takes effect; manufacturers must offer at least one model of wood boiler that will produce 70 percent less emissions, with further reductions in subsequent years.
- 05/07 EC and the Hearth, Patio, and Barbecue Association partnered to conduct a study of conventional wood stoves, results presented at 16th Annual Emission Inventory conference in Raleigh, NC.
- 09/25/07 Comprehensive workshop in Philadelphia on outdoor wood boilers, wood stove change-outs, local air districts' efforts to reduce wood smoke.

2008 and Ongoing

- 10/22/09 US EPA launched its Burn Wise educational campaign (http://www.epa.gov/burnwise) to help reduce wood smoke pollution.

Substance-Specific Activities: Alkyl-lead

GLBTS Workgroup Activities and Reports

1998

- 3/23/98 WG is formed at the first implementation meeting
- 11/16/98 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- 12/31/98 Draft GLBTS Challenge report for alkyl-lead is posted on the GLBTS Website

1999

- 1/99 EC prepares *Alkyl Lead Inventory Study - Sources, Uses and Releases in Ontario, Canada: A Preliminary Review*, and posts report on the GLBTS Website. The report concludes that the Canadian challenge of reducing alkyl-lead use by 90 percent between 1988 and 2000 has been exceeded.
- 9/8/99 GLBTS and PBT workgroups meet with National Motor Sports Council to discuss voluntary phase-out of leaded gasoline
- 10/29/99 draft GLBTS *Sources, Regulations and Options* (Steps 1, 2 & 3) Report for Alkyl-Lead is posted on the GLBTS Website

2000

- GLBTS *Sources, Regulations, and Reduction Options* (Step 1, 2 & 3) report for alkyl-lead is finalized (6/00) and posted (9/29/00) on the GLBTS Website
- GLBTS U.S. Challenge on Alkyl-lead: *Report on the Use of Alkyl-lead in Automotive Gasoline* is finalized (6/00) and posted (9/29/00) on the GLBTS Website

2001

- The U.S. meets the challenge of confirming no use of alkyl-lead in automotive gasoline. The US EPA PBT Program takes the lead for the U.S. in coordinating stakeholder efforts to reduce remaining alkyl-lead releases

Other Alkyl-lead Related Activities

1999 and Earlier

- Work begins on a draft *National PBT Action Plan* for Alkyl-lead

2000

- 8/25/00 A Draft *PBT National Action Plans* for alkyl-lead is posted on the PBT Website for public review and comment
- Auto racing industry expresses interest in working with US EPA to find lead-free gas substitutes

2001

- US EPA begins working with NASCAR to permanently remove alkyl-lead from racing fuels used, specifically, in the Busch, Winston Cup, and Craftsman Truck Series

Substance-Specific Activities: Octachlorostyrene (OCS)

GLBTS Workgroup Activities and Reports

1998

- 3/23/98 WG is formed at the first implementation meeting
- 6/16/98 Background Paper and Draft Action Plan for OCS posted on GLBTS Website
- 11/16/98 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- 12/31/98 Draft GLBTS Challenge report for OCS is posted on the GLBTS Website

1999

- 4/27/99 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 11/18/99 WG meeting at the GLBTS Stakeholder Forum in Chicago, IL
- Data on OCS trends in fish is assessed by the WG

2000

- 5/16/00 WG meeting at the GLBTS Stakeholder Forum in Toronto, Ontario
- 6/30/2000 EC draft report on Octachlorostyrene Sources, Regulations and Programs for the Province of Ontario 1988, 1998, and 2000 forwarded to interested stakeholders
- 9/22/00 Draft GLBTS Stage 3 report for OCS is distributed at the 9/22 Integration Workgroup meeting and e-mailed to the OCS Workgroup
- 12/00 US EPA and EC convene a meeting of North American magnesium producers to promote sharing of lessons regarding methods for preventing and managing OCS and other chlorinated hydrocarbon wastes

2004

- 8/04 Draft *Management Assessment for OCS* (Step 4) Report prepared

Other OCS Related Activities

1999 and Earlier

- 3/10/99 CGLI report, OCS and Suggested Industrial Sources: A Report to the GLBTS Workgroup, is submitted to the workgroup

2000

- 8/25/00 A Draft PBT National Action Plan for OCS is posted on the PBT Website for public review and comment

2002

-4/02 Toxics Release Inventory data for 2000 is made available to the public

Substance/Sector Workgroup Activities

2007

- 11/30/07 Introductory meeting of Substance/Sector Group (joint meeting conducted by teleconference) to review draft terms of reference for the new groups.

2008

- 06/02/08 to 06/03/08 WG meeting in Burlington, Ontario
- 07/08/08 to 07/09/08 WG leaders conference, Toronto, Ontario
- 08/07/08 WG teleconference
- 09/25/08 WG meeting in Chicago
- 12/02/08 to 12/03/08 WG meeting in Chicago

2009

- 03/31/09 WG meeting in Toronto
- 12/02/09 WG meeting in Chicago
- 12/30/09 US EPA released action plans for phthalates, long-chain perfluorinated chemicals (PFCs), polybrominated diphenyl ethers (PBDEs) in products, and short-chain chlorinated paraffins.

Sediments

Canadian and U.S. Activities

1998 and Earlier

- 6/15/98 PCB WG requests that the IG develop a strategy on sediments
- 6/19/98 Integration WG discusses sediments challenge
- US EPA provides guidance to workgroups on how to deal with sediments within chemical-specific workgroups

1999

- 1/26/99 Overview and presentation of IJC SedPAC Activities given at Integration WG meeting
- 2/99 Integration WG members develop a draft charge for a sediments subgroup
- 4/28/99 Draft Sediments subgroup charge presented at Integration WG meeting

2000

- 2/15/00 US EPA and EC present a draft sediment reporting format at the Integration WG meeting. The proposed format will map progress and report annually on

sediment remediation in the Great Lakes Basin using 1997 as the baseline year

- 5/16/00 At the Stakeholder Forum, US EPA and EC present the draft sediment reporting format and commit to hold a sediment technology workshop

2001

- 4/24/01 US EPA and EC host a two-day workshop on "Removing and Treating Great Lakes Contaminated Sediment," presenting sediment remediation technologies and case studies

2002 and Ongoing

- Ongoing assessments and remediations in both the U.S. and Canada within the Great Lakes watershed (see Section 7.0)

Related Sediment Activities

1998 and Earlier

- 11/97 The IJC's Sediment Priority Action Committee (SedPAC) issues draft white paper *Overcoming Obstacles to Sediment Remediation in the Great Lakes Basin*
- 12/1-2/98 IJC SedPAC holds "Workshop to Evaluate Data Interpretation Tools Used to Make Sediment Management Decisions" in Windsor, Ontario

2002

- 1/02 The second National Sediment Quality Survey report to Congress, *The Incidence and Severity of Sediment Contamination in Surface Waters of the United States, National Sediment Quality Survey: Second Edition*, is released for review by US EPA.

2004

- Work under The Great Lakes Legacy Act begins.

2008

- 9/28/08 Congress passed the Great Lakes Legacy Act of 2008, which extends the Legacy Act for two years at a funding level of $54 million per year.

Long-Range Transport (LRT) Activities

1999

- 11/19/99 EC presents the status of their LRT effort at the Integration WG meeting.

2000

- 3/27/00 EC prepares report: *Long-Range Transport of Persistent Toxic Substances to the Great Lakes: Review and Assessment of Recent Literature* (Ortech Environmental)

2001

- Several studies are undertaken in the U.S. and Canada to characterize global transport processes.

2003 and Ongoing

- 9/16/03 - 9/17/03 EC and US EPA sponsor LRT Workshop in Ann Arbor, MI, with support of the CEC, the IJC, and the Delta Institute.
- 9/03 LRT workshop background paper, the workshop program, presentations, and draft summary document are posted on the Internet at http://delta-institute.org/pollprev/lrtworkshop/_workshop.html
- Research into long-range transport of persistent toxic substances to the Great Lakes continues.

General Activities Related to Reductions in GLBTS Substances

US EPA Regulatory Determinations

1998 and Earlier

- 12/95 Maximum Available Control Technology (MACT) rules for large Municipal Waste Combustors (MWC) are promulgated
- 9/97 MACT rules for Medical Waste Incinerators (MWI) are promulgated
- 4/15/98 Pulp, Paper, and Paperboard Cluster Rule is promulgated
- 6/29/98 Amendments to the PCB Disposal Regulations are finalized
- 11/12/98 Federal Plan for MACT Implementation for large MWCs is finalized

1999

- 5/28/99 An Advance Notice of Proposed Rulemaking is released for the RCRA LDR for Mercury-Bearing Hazardous Wastes
- 7/6/99 Federal Plan for MACT Implementation for MWI is proposed
- 8/30/99 MACT for small MWCs are proposed (expected to be final in 2000)
- 9/30/99 Final Standards for Hazardous Air Pollutants for HWC are promulgated
- 10/29/99 TRI Amendments: new PBT reporting thresholds

2000

- 12/00 Compliance deadline for large MWC MACT
- 9/02 Compliance deadline for MWI MACT
- 1/1/00 New TRI reporting thresholds for PBTs become effective

2001

- US EPA finalizes the Reclassification of PCB and PCB-contaminated Electrical Equipment rule and a rule on Return of PCB Waste from US Territories Outside the Customs Territory of the US

2002

- PCP re-registration review proceeding as joint Canada/U.S. endeavor
- 4/02 the first year of data reported under TRI PBT rule become available
- 2/14/02 President Bush announces Clear Skies Initiative to cut mercury emissions from power plants by 70 percent

2005

- 5/18/05 US EPA publishes Clean Air Mercury Rule

2006

- 6/06/06 US EPA reaffirms Clean Air Mercury Rule

2007

- 9/20/07 US EPA publishes a Proposed Rule under 40 CFR Part 63 on Electric Arc Furnace Steelmaking Facilities, regarding a MACT standard for controlling emissions of mercury when such facilities use steel scrap that contains auto switches and other devices that contain mercury (72 FR 53814-53836).

US EPA Activities

1999 and Earlier

- 6/97 *Deposition of Air Pollutants to the Great Waters: Second Report to Congress* is released
- 12/97 *Mercury Report to Congress* is released
- 4/98 *Final Emission Inventory Data for Section 112(c)(6) Pollutants* is released
- 11/16/98 US EPA's Multimedia PBT Strategy is announced
- 11/16/98 Under the PBT Strategy, a draft *National Action Plan for Mercury* is released
- PBT Strategy grant awarded to WLSSD to work on reducing open trash burning
- U.S. PCB transformer registration database is updated
- Sample collection begins for the National Study of Chemical Residues in Fish
- U.S. GLBTS workgroup leaders participate in development of Draft National Action Plans of part of PBT Strategy

2000

- 6/00 *Deposition of Air Pollutants to the Great Waters: Third Report to Congress* is released
- 6/12/00 draft chapters of the *U.S. Dioxin Reassessment* for external scientific review are released
- 9/00 US EPA's 1996 National Toxics Inventory is released

- 9/28/00 Three draft chapters of the *U.S. Dioxin Reassessment* for SAB review are released
- PBT workgroups continue to work on National Action Plans for HCB, B(a)P, the Level I pesticides, and PCBs
- US EPA's Office of Air and Radiation and Office of Water collaborate on an Air-Water Interface Workplan to address atmospheric deposition of toxics and nitrogen to U.S. water bodies.

2001

- 5/23/01 U.S. signs the United Nation's global treaty on Persistent Organic Pollutants (POPs)

2002

- 1/02 *The Incidence and Severity of Sediment Contamination in Surface Waters of the United States, National Sediment Quality Survey: Second Edition* is released for review
- 7/23/02 Final PBT National Action Plan for Alkyl-lead published
- Preliminary data from first year of National Study of Chemical Residues in Lake Fish Tissue released

2004

- 5/18/04 Great Lakes Interagency Task Force created by U.S. Executive Order

2009

- 05/07/09 EPA releases 2010 budget, which includes $475 million for the Great Lakers Restoration Initiative

EC Regulatory Determinations

1999 and Earlier

- *Canadian Environmental Protection Act* is renewed

2000

- Canada-Wide Standards (CWS) (release limits) are developed for mercury, particulate matter, ozone, and benzene, and are being developed for dioxins/furans.
- Canadian Strategic Options Processes (SOPs) are under development for the Iron and Steel Manufacturing sector and finalized for the Wood Preservation sector
- 6/19/00 EC solicits public comments on proposed amendments to the PCB regulations under CEPA

2001

- 2/19/01 Canada announces $120.2 million in new regulatory and other measures to accelerate action on clean air
- 7/7/01 A notice with respect to Polychlorinated Biphenyls in Automotive Shredder Residue is published in the Gazette, Part I, for automobile shredding facilities that generated PCB-contaminated residue during 1998, 1999, or 2000.
- EC proposes amendments to the Chlorobiphenyl Regulations and Storage of PCB Material Regulations promulgated in 1977 and 1992, respectively
- Canada's PCB Waste Export Regulations (SOR/97-108) are being amended

2005

- 6/05 CCME accepts in principle a draft CWS for the coal-fired electric power generation sector. Final endorsement of the CWS is expected prior to the end of 2005.

2006

- 11/04/06 Proposed Canadian PCB regulations are published in the *Canada Gazette, Part I*.
- 11/21/06 to 1/20/07 Province of Ontario collected public comments on a risk-based decision-making framework for contaminated sediments completed under the 2002-2007 Canada-Ontario Agreement Respecting the Great Lakes Ecosystem.
- 11/29/06 Final regulatory amendments to include Pentachlorobenzene, and Tetrachlorobenzene on the Prohibition of Certain Toxic Substance Regulations, 2005 were published in *Canada Gazette, Part II*
- 12/08/06 Canada announces intention to commit $300 million over four years to implement the Chemicals Management Plan.
- 12/13/06 Hexachlorobutadiene (HCBD) was added to the *Virtual Elimination List* with a level of quantification in chlorinated solvents.

2007

- 12/9/06 Environment Canada published a Proposed Notice under CEPA 1999: requiring the preparation and implementation of pollution prevention plans for mercury (Hg) releases from mercury switches in end-of-life vehicles processed by steel mills.

2008

- 9/17/08 Final PCB Regulations are published in the *Canada Gazette II*.

EC Activities

1999 and Earlier

- Ontario "Drive Clean" program
- 1/99 The Canadian *Dioxins and Furans and Hexachlorobenzene Inventory of Releases* is finalized.
- EC upgrades and digitizes its National PCB database

2000

- Draft HCB, B(a)P (PAH), and OCS release inventories for Ontario are updated and circulated for review
- EMA with Algoma Steel being finalized.
- EC, in coordination with the Hearth Products Association, conducts testing of conventional and US EPA-certified wood stoves to investigate releases of dioxins/furans, PAHs, HCB, and particulate matter

2006

- 12/06 Canada's Chemicals Management Plan (CMP) is launched to protect Canadians and the environment by ensuring any risks posed by chemicals are assessed and managed properly.

2007

-02/07 CMP Challenge initiative is launched to collect information on the properties and uses of ~200 chemical substances identified as high priorities for action. The information will be used to make decisions regarding the best approach to protect Canadians and their environment from any risks these substances might pose.

2009

-05/09 The Canadian Great Lakes Chemical Priorities Working Group is charged with providing directions and recommendations regarding Canada's priorities for chemicals in the Great Lakes Basin for federal, joint-jurisdictional and binational programs.

Other Activities

1998 and Earlier

- CEC issues Continental Pollutant Pathways Initiative
- 7/98 UNEP POPs negotiations initiated

1999

- Under the GLWQA, The Lake Ontario LaMP Stage 1 report is released
- By the end of 1999, emission control retrofits either completed or underway at all large MWC in the U.S.
- The initial *Great Lakes Regional Air Toxics Emissions Inventory*, using 1993 data, is released
- The Lake Ontario LaMP Update 1999 is released

2000

- Under the GLWQA, Canada and the U.S. work on restoring beneficial uses to 43 AOCs in the Great Lakes Basin through the RAP program
- The Lake Erie, Lake Michigan, and Lakes Superior LaMPs 2000 are released
- The Lake Ontario Lamp Update 2000 is released
- The Lake Huron Initiative Action Plan is released
- Numerous pilot projects and pollution prevention/reduction agreements relevant to toxics of concern are underway with the steel, automobile, and other manufacturing industries and utilities in Ontario and the U.S. Great Lakes States
- 11/8/00 – 11/9/00 Atmospheric deposition workshop held, *Using Models to Develop Air Toxics Reduction Strategies*
- 12/00 Final POPs negotiations
- The 1996 Great Lakes Inventory of Toxic Air Emissions is prepared by the Great Lakes Commission

2001

- 2/01 21st session of the UNEP Governing Council is held: UNEP will undertake a global study on the health and environmental impacts of mercury
- 8/22/01 The IJC issues a Review of Progress under the Canada-United States Great Lakes Binational Toxics Strategy
- Monitoring of air deposition of toxic pollutants in the Great Lakes Basin under IADN

2002

- Monitoring of air deposition of toxic pollutants in the Great Lakes Basin continues under IADN

2003

- 9/19/03 – 9/20/03 IJC 2003 Great Lakes Conference and Biennial Meeting in Ann Arbor, MI
- Monitoring of air deposition of toxic pollutants in the Great Lakes Basin continues under IADN

2004

- 4/23/04 Great Lakes Commission releases 2001 Great Lakes Regional Air Toxic Emissions Inventory, available at www.glc.org/air
- 10/6/04 – 10/8/04 State of Lakes Ecosystem Conference (SOLEC) held in Toronto, Ontario

2006

- 11/01/06 – 11/03/06 State of Lakes Ecosystem Conference (SOLEC) held in Milwaukee, WI
- Monitoring of air deposition of toxic pollutants in the Great Lakes Basin continues under IADN

2007 and Ongoing

- 2/07 NWF issues report, Environmentally Preferable Purchasing in the Great Lakes Region
- 7/16/07 US EPA workshop, Building an Integrated Surveillance System for Emerging Chemicals in the Great Lakes and Nationwide, held in Chicago
- 8/21/07 Montebello Accord – U.S./Canada/Mexico Security and Prosperity Partnership Agreement

APPENDIX B:
INTERNATIONAL JOINT COMMISSION REVIEW OF CHEMICALS OF EMERGING CONCERN AND ANALYSIS OF ENVIRONMENTAL EXPOSURES IN THE GREAT LAKES BASIN

Duluth Entry - North Pier Head Lighthouse at Sunrise, Photograph by Jerry Bielicki

INTERNATIONAL JOINT COMMISSION REVIEW OF CHEMICALS OF EMERGING CONCERN AND ANALYSIS OF ENVIRONMENTAL EXPOSURES IN THE GREAT LAKES BASIN

Submitted by: Gary Klecka, Carolyn Persoon, and Rebecca Currie

The Dow Chemical Company, Midland, MI

The University of Iowa, Iowa City, IA

Goals of the Study

To assess the current status of chemicals of emerging concern in the basin with a focus on water quality through:

- Literature search
- Database of reported concentrations
- Statistical analysis to define current environmental exposures

To develop a preliminary assessment of their potential ecological significance, the concentrations were compared with currently available regulatory standards, guidelines, or criteria.

Introduction

Environmental analysis and monitoring have long been recognized as a means for assessing environmental quality. Within the Great Lakes watershed, the governments of the United States and Canada, together with collaborating agencies, have performed numerous surveys of environmental contaminants in the air, water, sediments, and biota. Environmental monitoring programs are necessary to develop comprehensive descriptions of environmental quality, including at spatial and temporal scales, and to provide a sound basis for effective measures, strategies, and policies to address environmental problems (Calamari et al., 2000). While an important use of monitoring data is to inform environmental risk assessment, information gained from environmental measurements is also important for priority-setting regarding potential hazards of chemical contaminants.

Over the past 10 years, the emphasis on monitoring has shifted from the analysis of so-called legacy pollutants to a wide array of new chemicals being discovered in the environment that are often referred to collectively as "chemicals of emerging concern." While it has been known for over 20 years that compounds such as pesticides, detergents, personal care products, and pharmaceuticals enter the environment, improvements in the instrumentation and analytical methodology for detecting chemical substances in various environmental media (air, water, sediment, biota) have brought increased awareness and concern over the presence and potential risk that these chemicals may pose (Daughton, 2001). Although thousands of chemicals are listed on chemical inventories in both the United States and Canada, very few have regulations governing their release to the environment. The term "chemicals of emerging concern" has come to define the emerging awareness of the presence in the environment of many chemicals used by society that are unregulated or inadequately regulated, along with concern over the risk that these chemicals may pose to the health of humans and ecosystems.

The topic of chemicals of emerging concern is not new to the International Joint Commission Boards and was specifically addressed by the Science Advisory Board with its *Expert Consultation on Emerging Issues of the Great Lakes in the 21st Century* held February 5-7, 2003 at Wingspread, WI. Several papers in the 2003-2005 Priorities Report dealt with the issue. Muir et al. (2006) summarized the various means for tracking, categorizing, and assessing chemicals in commerce, and presented an overview of recent measurements of "new" chemicals in the Great Lakes. Walker (2006) addressed whether currently available tools, such as quantitative SARs, can identify emerging pollutants that will threaten the Great Lakes ecosystem. Fox (2006) discussed the importance of monitoring programs in the context of meeting the requirements of the Great Lakes Water Quality Agreement.

In October, 2007, the International Joint Commission began work on the 2007-2009 Nearshore Framework

Priority. The purpose of this Priority is to assemble and report on the latest scientific, policy, and governance information on the nearshore of the Great Lakes so as to assess the binational implications of nearshore conditions and stressors. Nearshore problems are pressing and have significant social, economic, and environmental impacts. Current nearshore water quality is being adversely impacted by increased human population and problems due to impervious surfaces and fertilizer use. Nearshore water quality is also influenced by land-based discharges from urban and agricultural sources, sediment resuspension, habitat loss and degradation, and atmospheric deposition, as well as by offshore waters. As the population increases, sewage discharges to receiving waters increase and impinge on water quality in the nearshore. Water quality in the nearshore is important to fish, aquatic birds, amphibians, and reptiles, since nearly all fish species spawn, have nursery grounds, and feed in the nearshore at some time in their development. The link between land-based activities and the nearshore has become recognized as the key challenge to protecting and restoring the chemical, physical, and biological integrity of the waters of the Great Lakes Basin Ecosystem.

Within the context of the 2007-2009 Nearshore Framework Priority, the Priority on Chemicals of Emerging Concern will allow a more thorough review of the scientific and policy aspects related to identification, impact, and management. The current challenge is to apply the latest information based on regional, national, and international approaches to the existing binational policy framework(s) for the Great Lakes to identify potential shortcomings or gaps. As a first step, the body of current scientific knowledge on chemicals of emerging concern specific to the Great Lakes watershed will be reviewed, to be followed by an expert consultation to identify and assess opportunities for strengthening actions to protect the Great Lakes. The consultation will include scientists and other experts from governments, industry, and other key stakeholders in order to ensure the process is as inclusive as possible within an expert and informed group of participants.

The objectives of this report were to review and compile all peer reviewed scientific studies and reports since 1997 in relation to chemicals of emerging concern that may pose threats to water quality in the Great Lakes watershed. Emphasis was placed on chemicals discharged to the Great Lakes nearshore waters from wastewater treatment plants as well as from other point and non-point sources of rural and urban pollution. The concentrations of chemicals in various environmental media were assembled into a database, which was statistically analyzed to develop a quantitative understanding of current environmental exposures. To develop an initial assessment of their potential ecological significance, the concentrations were compared with currently available regulatory standards, guidelines, or criteria.

Some Binational Findings on Chemicals of Emerging Concern in the Great Lakes Basin

- **Current Use Pesticides** – Concentrations of many current use pesticides are below current regulatory criteria. For others (e.g., 2,4-D, metolachlor, and metribuzin), 95th percentile concentrations were below standards, but exceedences were noted for maximum concentrations. Atrazine, azinophos-methyl, chlorpyrifos, diazinon, and parathion exceeded regulatory standards in 6% to 32% of the samples. More information can be found in the full report.
- **Pharmaceuticals** – Detectable concentrations of pharmaceutical compounds were present in 34% of the samples. At present, there are no standards, guidelines, or criteria with which to compare environmental concentrations.
- **Organic Wastewater Contaminants, Personal Care Products, Steroids, and Hormones** – Bis(2-ethylhexyl) phthalate (DEHP) was detected in a single sample at levels which exceeded the US EPA Maximum Contaminant Level for drinking water, the EC Interim Water Quality Guideline, and the European Union (EU) predicted no effect value. The maximum concentration of bisphenol-A exceeded the Canadian predicted no-effect

concentration (PNEC) for water but was below the PNEC for sediment organisms.

- **Synthetic Musks** – Maximum concentrations of musk xylene, musk ketone, acetyl hexamethyl-tetrahydronaphthalene (AHTN), and hexahydrohexamethylcyclopentabenzopyran (HHCB) in environmental media from the Great Lakes indicated that all values were below the PNEC.
- **Alkylphenol Ethoxylates** – None of the samples exceeded the US EPA Water Quality Criterion for nonylphenol (NP); 22% of the samples exceed the NP equivalent Canadian Water Quality Guideline. Sediment concentrations exceeded the NP equivalent Canadian Sediment Guideline in 31% of the samples.
- **Perfluorinated Surfactants** – Risks for secondary poisoning from the ingestion of food were indicated for PFOS and total perfluorinated surfactant concentrations. Concentrations of PFOS and perfluorooctanoic acid (PFOA) in water were below available PNEC and estimated no-effect value (ENEV).
- **Polybrominated Diphenyl Ethers** – Sediment concentrations for various PBDEs were below PNEC and ENEV values. Tetra and penta-brominated congener concentrations were above the Canadian ENEV criteria value for secondary consumers (0.0084 mg/kg food).
- **Chlorinated Paraffins** – All exposures were below the no effect values (ENEV/PNEC).

Some Binational Findings and Recommendations on Policy for Great Lakes Chemicals of Emerging Concern

- Industrial chemicals in the U.S. and Canada are subject to pre-manufacturing notification, review and approval by the federal government.
- International treaties have been developed for the identification, assessment and management of persistent organic pollutants.
- There are voluntary stewardship initiatives in place on both sides of the border that address some chemicals of emerging concern (e.g., U.S. PFOS Stewardship Initiative).
- There are gaps or inadequacies in chemicals assessment and management for certain classes of chemicals that are not subject to TSCA or CEPA regulations, with regard to their potential impact in the environment, including: pharmaceutical compounds, some personal care product constituents, nanomaterials, and chemical constituents in imported goods.
- The U.S. should ratify the Stockholm Convention on Persistent Organic Pollutants (POPs) and the United Nations Economic Commission for Europe Convention on Long Range Transboundary Air Pollution.
- A renewed GLWQA should include a description of the underlying principles and processes by which the Parties would establish priorities, rather than a specific list of substances.
- An emphasis should be placed on moving upstream and adopting sustainable solutions to the design, production and consumption of chemicals of emerging concern in the Great Lakes Basin.
- A pre-manufacturing notification level of review should be conducted for all chemical classes, including grandfathered TSCA substances, pharmaceuticals, personal care product constituents and nanomaterials, as well as constituents in imported goods.
- Adoption of enhanced wastewater treatment technologies to provide improved control and management of chemicals of emerging concern should be implemented. Wastewater treatment is an essential component for controlling a wide diversity of chemicals that are discharged to the Great Lakes, and there is a need for accelerated continuous improvement of existing facilities.

- Strict regulations and enforcement should be put into place for waste and nutrient management practices for all farm operations to protect the Great Lakes. The adequacy of current regulations to mitigate inputs of chemicals of emerging concern needs to be reviewed and strengthened, including assistance and support to encourage compliance from farm operators.
- New policies need to be developed to manage chemicals of emerging concern in the Great Lakes with new and innovative approaches that continue to use sound scientific methods and principles.
- Consumer education should be conducted and incentives should be provided to encourage conservation and consumer choices that can help drive changes in consumer products and create marketplace incentives for manufacturers.
- Further emphasis should be placed on gaining knowledge and understanding of human health effects as they pertain to the major categories of chemicals of emerging concern.

Conclusions

There has been an increasing shift in focus from industrial point sources to dispersed, non-point releases of chemicals and substances, such as those in consumer products and pharmaceuticals that may require new analyses and approaches, including risk management approaches. General conclusions from this project include the following:

- A wide variety of chemicals have been detected in various media within the Great Lakes Basin.
- Our ability to detect chemicals in the environment exceeds our ability to understand the significance of the findings.
- The availability of data varies considerably.
- Some substances have relatively extensive datasets covering broad regions of the basin while other studies focused on more localized areas or regions.
- A limited amount of data was available for many substances, and many concentrations are < 1 ug/L.
- The highest concentrations were found in the vicinity of sources (e.g., wastewater treatment plants, or WWTPs) and declined with increasing distance from sources.
- Low to non-detect levels of many substances were found in open waters.
- Results of comparisons of environmental exposures to regulatory criteria yielded mixed results: For some, levels are below ENEVs, PNECs, and water quality standards (WQSs); for others, current exposures may indicate a potential risk.
- Criteria have not been established for many substances.
- Regulatory and/or voluntary actions to reduce or eliminate emissions are underway for a number of substances included in the analysis.

References

Calamari, D., K. Jones, K. Kannan, A. Lecloux, M. Olsson, M. Thurman, P. Zannetti. 2000. Monitoring as an indicator of persistence and long-range transport. In Evaluation of Persistence and Long-Range Transport of Organic Chemicals in the Environment, G. M., Klecka et al., eds. Society of Environmental Toxicology and Chemistry, Pensacola, FL.

Daughton, C.G. 2001. Pharmaceuticals in the environment: overarching issues and overview. In Pharmaceuticals and Personal Care Products in the Environment: Scientific and Regulatory Issues, Daughton, C.G. and Jones-Lepp, T. (eds.), Symposium Series 791; American Chemical Society: Washington, D.C., pp. 2-38.

Fox, G.A. 2006. Back to the future: rediscovering the requirement for monitoring in the Great Lakes Water Quality Agreement. In Expert Consultation on Emerging Issues of the Great Lakes in the 21st Century. Report of the Great Lakes

Science Advisory Board to the International Joint Commission, November, 2006.

Muir, D., M. Alaee, and B. Scott. 2006. Identifying potential and emerging chemical contaminants in the Great Lakes. In Expert Consultation on Emerging Issues of the Great Lakes in the 21st Century. Report of the Great Lakes Science Advisory Board to the International Joint Commission, November, 2006.

Walker, J.D. 2006. Can QSARs identify emerging pollutants that will threaten the Great Lakes ecosystem? In Expert Consultation on Emerging Issues of the Great Lakes in the 21st Century. Report of the Great Lakes Science Advisory Board to the International Joint Commission, November, 2006.

For more information, contact us:

Alan Waffle, P. Eng.
Head, Great Lakes

Great Lakes Section
Environmental Protection Operations Division
Environment Canada
4905 Dufferin Street
Toronto, Ontario M3H 5T4
CANADA

E-mail: Alan.Waffle@ec.gc.ca

Edwin R. (Ted) Smith
Pollution Prevention &
Toxics Reduction Team Leader

Great Lakes National Program Office
U.S. Environmental Protection Agency
77 W. Jackson Blvd. (G-17J)
Chicago, IL 60604
USA

E-mail: Smith.Edwin@epamail.epa.gov

www.binational.net

Printed on recycled paper with soy ink