

### **Contaminants in Whole Fish**

Indicator #121

### **Overall Assessment**

Status:	Mixed
Trend:	Improving
Rationale:	Since the late 1970s, concentrations of historically regulated contaminants such as PCBs, DDT and mercury have generally declined in most monitored fish species. The concentrations of other contaminants, currently regulated and unregulated, have demonstrated either slowing declines or, in some cases, increases in selected fish communities. The changes are often lake-specific and relate both to the characteristics of the substances involved and the biological composition of the fish community.

### Lake-by-Lake Assessment

PCB and DDT levels are measured in lake trout and walleye while only smelt samples have recent mercury trend data available.

### Lake Superior

Status:	Fair
Trend:	Improving
Rationale:	Concentrations of total PCBs show little change, total DDT shows fluctuating concentrations, while mercury concentrations continue to decline. Total PCB concentrations remain above GLWQA criteria while total DDT and mercury remain below. Contaminants in Lake Superior are typically atmospherically-derived. The dynamics of Lake Superior allow for the retention of contaminants much longer than in any other Great Lake.
Lake Michiga	n
Status:	Fair
Trend:	Improving
Rationale:	Concentrations of total PCBs and total DDT are declining. Total PCB levels remain above GLWQA criteria and total DDT levels remains below. 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 such as Asian carp are also of major concern to Lake Michigan due to the connection of Chicago Sanitary and Ship Canal and the danger the carp pose to the food web.
Lake Huron	
Status:	Fair
Trend:	Improving
Rationale:	Both total PCBs and DDT show general declines in concentrations while mercury displays a flux in concentration. Total PCB concentrations remain above GLWQA criteria while total DDT and mercury remain below. Contaminant loading to Saginaw Bay continues to be reflected in fish tissue contaminant levels.
Lake Erie	
Status:	Fair
Trend:	Improving
Rationale:	Total PCBs and DDT concentrations show a pattern of annual increases linked to changes in invasive
	species populations, such as zebra and quagga mussels. Aquatic invasive species are of major
	concern to Lake Erie because the pathways and fate of persistent toxic substances will be altered,
	resulting in differing accumulation patterns, particularly near the top of the food chain. Mercury concentrations are the highest ever recorded in Lake Erie. Total PCB concentrations remain above GLWQA criteria while total DDT and mercury remain below.

### Lake Ontario

	Status:	Fair
	Trend:	Improving
Rationale: Both total PCBs and DDT concentrations show a pattern of		Both total PCBs and DDT concentrations show a pattern of decline while mercury concentrations
		show little change. Total PCB concentrations remain above GLWQA criteria while total DDT and
		mercury remain below. Historic point sources of mirex and OCS in Lake Ontario have resulted in
		the highest concentration of these contaminants than in any of the other Great Lakes. The presence
		of contaminants of emerging concern, such as PBDEs and PFOS, continues to raise alarm in Lake
		Ontario, due to their continuing increases in concentration over time.

### Purpose

- To describe temporal and spatial trends of bioavailable contaminants in representative open water fish species from throughout the Great Lakes
- To infer the effectiveness of remedial actions related to the management of critical pollutants
- To identify the nature and severity of emerging problems

### **Ecosystem Objective**

Great Lakes waters should be free of toxic substances that are harmful to fish and wildlife populations and the consumers of this biota. Data on status and trends of contaminant conditions, using fish as biological indicators, support the requirements of the Great Lakes Water Quality Agreement (GLWQA, United States and Canada 1987) Annexes 1 (Specific Objectives), 2 (Remedial Action Plans and Lakewide Management Plans), 11 (Surveillance and Monitoring), and 12 (Persistent Toxic Substances).

### State of the Ecosystem

### Background

Long-term (greater than 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 the U.S. Environmental Protection Agency (U.S. EPA) Great Lakes National Program Office (GLNPO) through the Great Lakes Fish Monitoring Program, and Environment Canada, through the Fish Contaminants Surveillance Program, to determine the effects of contaminant concentrations on wildlife and to monitor trends. Environment Canada reports annually on contaminant burdens in similarly aged lake trout (4+ - 6+ year range), walleye (Lake Erie), and in smelt. GLNPO 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. Details of the program can be found at, <u>http://www.epa.gov/glnpo/glindicators/fish.html</u>. Differences between the U.S. and Canadian

programs, including collection site differences and varying species collections, inhibit the direct comparison of results from the two programs.

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

### Chemical Concentrations in Whole Great Lakes Fish

Since the late 1970s, concentrations of historically regulated contaminants such as PCBs, DDT and mercury have generally declined in most monitored fish species. The concentration of other contaminants, currently regulated and unregulated, have demonstrated either slowing declines or, in some cases, increases in selected fish communities. The changes are often lake-specific and relate both to the characteristics of the substances involved and the biological composition of the fish community.

The GLWQA, first signed in 1972, renewed in 1978, and amended in 1987, expresses the commitment of Canada and





Lake Trout = 600-700 mm size range. \*Fish collected between 1972 and 1982 were collected at even year sites only. Walleye = 450-550 mm size range.

Source: U.S. Environmental Protection Agency

the United States to restore and maintain the chemical, physical and biological integrity of the Great Lakes basin ecosystem. When applicable, contaminant concentrations are compared to GLWQA criteria.

### Total PCBs

Total PCB concentrations in Great Lakes top predator fish have continuously declined since their phase-out in the 1970s. However, rapid declines are no longer observed and concentrations in fish remain above the U.S. EPA wildlife protection value of 0.16 ppm and the GLWQA criteria of 0.1 ppm. Concentrations remain high in top predator fish due to the continued release of uncontrolled sources and their persistent and bioaccumulative nature.



**Figure 3.** Total PCBs in 4 to 6 year old individual whole Environment Canada Lake Trout, collected 1977 through 2005, µg/g wet weight. Source: Fisheries and Oceans Canada



**Figure 5.** Total DDT in Even Year whole EPA Lake Trout composites (Walleye in Lake Erie), 1972 - 2000.  $\mu$ g/g wet weight +/- 95% C.I., composite samples.

Lake Trout = 600-700 mm size range. \*Fish collected between 1972 and 1982 were collected at even year sites only. Walleye = 450-550 mm size range. Source: U.S. Environmental Protection Agency



**Figure 2.** Total PCBs in Odd Year whole EPA Lake Trout composites (Walleye in Lake Erie),  $1991 - 2003 \mu g/g$  wet weight +/- 95% C.I., composite samples. Lake Trout = 600-700 mm size range. Walleye = 450-550 mm size range. Source: U.S. Environmental Protection Agency



**Figure 4.** Total PCBs in composite Environment Canada rainbow smelt, collected 1977 through 2005, µg/g wet weight. Source: Fisheries and Oceans Canada

### Total DDT

Total DDT concentrations in Great Lakes top predator fish have continuously declined since the chemical was banned in 1972. However, large declines are no longer observed. Rather, very small annual percent declines are seen, indicating near steady state conditions. It is important to note that the concentrations of this contaminant remain below the GLWQA criteria of 1.0 ppm. There is no U.S. EPA wildlife protection value for total DDT because the PCB value is more protective.

#### Mercury

Concentrations of mercury are similar across all fish in all lakes. It is assumed that concentrations of mercury in top predator fish are atmospherically driven. It is important to note that current concentrations in GLNPO top predator fish in all lakes remain



**Figure 6.** Total DDT in Odd Year whole EPA Lake Trout composites (Walleye in Lake Erie), 1991 - 2001. μg/g wet weight +/- 95% C.I., composite samples. Lake Trout = 600-700 mm size range. Walleye = 450-550 mm size range Source: U.S. Environmental Protection Agency



Figure 8. Total DDT in composite Environment Canada rainbow smelt, collected 1977 through 2005,  $\mu$ g/g wet weight.

Source: Fisheries and Oceans Canada



**Figure 10.** Mercury in 4 to 6 year old individual whole Environment Canada Lake Trout, collected 1977 through 2005, µg/g wet weight. Source: Fisheries and Oceans Canada



Figure 7. Total DDT in 4 to 6 year old individual whole Environment Canada Lake Trout, collected 1977 through 2005,  $\mu$ g/g wet weight.

Source: Fisheries and Oceans Canada



**Figure 9.** Mercury in whole EPA Lake Trout composites (Walleye in Lake Erie), 1999 - 2003,  $\mu$ g/g wet weight +/- 95% C.I., composite samples.

Lake Trout = 600-700 mm size range. Walleye = 450-550 mm size range.

Source: U.S. Environmental Protection Agency



Figure 11. Mercury in composite Environment Canada rainbow smelt, collected 1977 through 2005,  $\mu$ g/g wet weight.

Source: Fisheries and Oceans Canada

above the GLWQA criteria of 0.5 ppm and that Canadian smelt have never been observed to be above the GLWQA criteria.

### Total Chlordane

Concentrations of total chlordane have consistently declined in whole top predator fish since the U.S. EPA banned it in 1988. 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 one of the five components of the technical chlordane mixture, it is the least metabolized and predominates within the food web (Carlson and Swackhamer 2006).



**Figure 12.** Total Chlordane in Even Year whole EPA Lake Trout composites (Walleye in Lake Erie),  $1972 - 2002 \mu g/g$  wet weight +/- 95% C.I., composite samples.

Lake Trout = 600-700 mm size range. \*Fish collected between 1972 and 1982 were collected at even year sites only. Walleye = 450-550 mm size range.

Source: U.S. Environmental Protection Agency



**Figure 14.** Total Chlordane in 4 to 6 year old individual whole Environment Canada Lake Trout, collected 1977 through 2005,  $\mu$ g/g wet weight.

Source: Fisheries and Oceans Canada



**Figure 13.** Total Chlordane in Odd Year whole EPA Lake Trout composites (Walleye in Lake Erie), 1991 – 2003  $\mu$ g/g wet weight +/- 95% C.I., composite samples.

Lake Trout = 600-700 mm size range. Walleye = 450-550 mm size range.

Source: U.S. Environmental Protection Agency



Figure 15. Total Chlordane in composite Environment Canada rainbow smelt, collected 1977 through 2005,  $\mu$ g/g wet weight. Source: Fisheries and Oceans Canada

### Mirex

Concentrations of mirex are highest in Lake Ontario top predator fish due to its continued release from uncontrolled historic sources near the Niagara River.



**Figure 16.** Mirex in Even Year Lake Ontario whole EPA Lake Trout composites,  $1972 - 2002 \mu g/g$  wet weight +/- 95% C.I., composite samples.

Lake Trout = 600-700 mm size range. \*Fish collected between 1972 and 1982 were collected at even year sites only. Walleye = 450 -550 mm size range. Source: U.S. Environmental Protection Agency



Figure 17. Mirex in Odd Year Lake Ontario whole EPA Lake Trout composites ,  $1991 - 2003 \mu g/g$  wet weight +/- 95% C.I., composite samples.

Lake Trout = 600-700 mm size range. Walleye = 450-550 mm size range.

Source: U.S. Environmental Protection Agency



**Figure 18.** Mirex in 4 to 6 year old individual whole Environment Canada Lake Trout, collected 1977 through 2005, µg/g wet weight. Source: Fisheries and Oceans Canada



**Figure 19.** Mirex in composite Environment Canada rainbow smelt, collected 1977 through 2005, μg/g wet weight. Source: Fisheries and Oceans Canada

### Dieldrin

Concentrations of dieldrin in lake trout appear to be declining in all Great Lakes and are lowest in Lake Superior and highest in Lake Michigan. Concentrations in Lake Erie walleye were lower than those in lake trout from the other Great 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.



Figure 20. Dieldrin in Even Year whole EPA Lake Trout composites (Walleye in Lake Erie), 1972 - 2002  $\mu$ g/g wet weight +/- 95% C.I., composite samples.

Lake Trout = 600-700 mm size range. \*Fish collected between 1972 and 1982 were collected at even year sites only. Walleye = 450-550 mm size range.

Source: U.S. Environmental Protection Agency



**Figure 22.** Dieldrin in 4 to 6 year old individual whole Environment Canada Lake Trout, collected 1977 through 2005, µg/g wet weight. Source: Fisheries and Oceans Canada



Figure 21. Dieldrin in Odd Year whole EPA Lake Trout composites (Walleye in Lake Erie), 1991 – 2003  $\mu$ g/g wet weight +/- 95% C.I., composite samples.

Lake Trout = 600-700 mm size range. Walleye = 450-550 mm size range.

Source: U.S. Environmental Protection Agency



Figure 23. Dieldrin in composite Environment Canada rainbow smelt, collected 1977 through 2005,  $\mu$ g/g wet weight.

Source: Fisheries and Oceans Canada

### 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).

### PBDEs

Both U.S. EPA and Environment Canada analyze for polybrominated diphenylethers (PBDE) in whole top predator fish. Retrospective analyses of archived samples have demonstrated the continuing increase in concentrations of PBDEs and are confirmed by present day concentrations in top predator fish. It is important to note that the concentration of most other persistent organic pollutants in top predator fish have declined, while PBDEs continue to increase.

### Other Contaminants of Emerging Interest

One of the most widely used brominated flame retardants (BFR) is hexabromocyclododecane (HBCD). Based on its use pattern as an additive BFR, it has the potential to migrate into the environment from its application site. Recent studies have confirmed that HBCD isomers do bioaccumulate in aquatic ecosystems and do biomagnify as they move up the food chain. Recent studies by Tomy *et al.* (2004) confirmed the food web biomagnification of HBCD isomers in Lake Ontario (Table 1).

Perfluoroctanesulfonate (PFOS) has also been detected in fish throughout the Great Lakes and has also demonstrated the capacity for biomagnification in food webs. PFOS is used in surfactants such as water repellent coatings (e.g., Scotchguard <sup>TM</sup>) and fire suppressing foams. It has been identified in whole

Species	ΣHBCD (α+γ isomers) (ng/g wet wt ± S.E.)
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

 Table
 1.
 Lake
 Ontario
 food
 web

 bioaccumulation of HBCD isomers.
 Source: Tomy et al. (2004)
 Source: Tomy et al. (2004)
 Source: Tomy et al. (2004)

lake trout samples from all the Great Lakes at concentrations from 3 to 139 ng/g wet weight (Stock *et al.* 2003). In addition, retrospective analyses of archived lake trout samples from Lake Ontario have identified a 4.25-fold increase (43 to 180 ng/g wet weight, whole fish) from 1980 to 2001 (Martin *et al.* 2004).

### Pressures

### Current

The impact of invasive nuisance species on toxic chemical cycling in the Great Lakes is still being investigated. The number of non-native invertebrates and fish species proliferating in the Great Lakes basin continues to increase, and they continue to spread more widely. Changes imposed on the native fish communities by non-native species will subsequently alter ecosystem energy flows. As a consequence, the pathways and fate of persistent toxic substances will be altered, resulting in different accumulation patterns, particularly at the top of the food web. Each of the Great Lakes is currently experiencing changes in the structure of the aquatic community, hence there may be periods of increases in contaminant burdens of some fish species.

A recently published, 15-year retrospective Great Lakes study showed that lake trout embryos and sac fry are very sensitive to toxicity associated with maternal exposures to 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) and structurally related chemicals (Cook *et al.* 2003). The increase in contaminant load of TCDD may be responsible for declining lake trout populations in Lake Ontario. The models used in this study can be used in the other Great Lakes.

### <u>Future</u>

Additional stressors in the future will include climate change, with the potential for regional warming to change the availability of Great Lakes critical habitats, change the productivity of some biological communities, accelerate the movement of contaminants from abiotic sources into the biological communities, and effect the composition of biological communities. Associated changes in the concentration of contaminants in the water, critical habitat availability and reproductive success of native and non-native species are also factors that will influence trends in the quantity of toxic contaminants in the Great Lakes basin ecosystem.

### **Management Implications**

Much of the current, basin-wide, persistent toxic substance data that is reported focuses on legacy chemicals whose use has been previously restricted through various forms of legislation. There are also a variety of other potentially harmful contaminants at various locations throughout the Great Lakes that are reported in literature. A comprehensive, basin-wide assessment program is needed to monitor the presence and concentrations of these recently identified compounds in the Great Lakes basin. The existence of long-term specimen archives (greater than 25 years) in both Canada and the United States could allow retrospective analyses of the samples to determine if concentrations of recently detected contaminants are changing. Further control legislation might be needed for the management of specific chemicals.

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