Limnol. Oceanogr., 51(6), 2006, 2794–2807

Long-term changes in legacy trace organic contaminants and mercury in Lake Ontario salmon in relation to source controls, trophodynamics, and climatic variability

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Abstract

We used long-term (20+ yr) datasets to determine how the sum of 209 polychlorinated biphenyl congeners ([ΣPCB]), dodecachloropentacyclodecane ([mirex]), para-para dichlorodiphenyltrichloroethane ([*p,p′*-DDT]), and total mercury ([Tot-Hg]) concentrations have changed in Lake Ontario chinook salmon (*Oncorhynchus tshawytscha*, 1983–2003) and coho salmon (*Oncorhynchus kisutch*, 1976–2003). Exponential decay models best describe temporal reductions of persistent organic pollutant concentrations [POPs], including [ΣPCB], [mirex], and [*p,p′*-DDT], in chinook ($r^2 = 0.68$–0.77, $p < 0.001$) and coho ($r^2 = 0.68$–0.87, $p < 0.001$) salmon over the record. In comparison, declines in [Tot-Hg] were slight, with linear models best describing trends ($r^2 = 0.49$–0.50, $p = <0.001$–0.001). Rapid declines of [POPs] from the mid-1970s through the early 1980s were attributed mostly to Canada–United States bans on usage and sedimentation; subsequent concentration oscillations were linked to salmonine stocking and nutrient abatement programs, climatic cycles, and alewife (*Alosa pseudoharengus*) population dynamics.

A major challenge of the Canada–United States Boundary Waters Treaty (1906) and Great Lakes Water Quality Agreement (1972, 1978) has been to control persistent organic pollutant (POP) loadings to the Great Lakes. In North America, POP production began in 1929 with the synthesis of polychlorinated biphenyls (PCBs), which were common components of heat transfer and lubricating fluids, and some pesticides, paints, adhesives, sealants, and plastics. PCB use in North America peaked during the mid- to late-1960s (Oliver et al. 1989; Wong et al. 1995). The pesticide dichlorodiphenyltrichloroethane (DDT) was first produced in the early 1940s, but the highest application rates in North America occurred in the early 1960s (Oliver et al. 1989). Dodecachloropentacyclodecane (mirex) was first produced in 1959, with maximum use occurring in the mid-1960s (Oliver et al. 1989; Wong et al. 1995). It was used in the United States as a pesticide and fire retardant and for the production of some plastics and entered Lake Ontario via the Niagara and Oswego rivers (Makarewicz et al. 2003). The use of PCBs and DDT was banned in Canada and the United States in the early 1970s, with mirex use being banned since 1977–1978. Given their stability and potential for vaporization and atmospheric transport (e.g., Sun et al. 2006), these so-called legacy POPs remain a concern in the Great Lakes region.

Mercury (Hg) occurs naturally at trace concentrations in soils, water, air, and biomass and is also ubiquitous in the Great Lakes. In North America, pre-1900 Hg emissions were primarily associated with gold and silver mines. with loads from these mines peaking at $>1.5 \times 10^6$ kg yr^{-1} in the 1870s (Pirrone et al. 1998). Although loads from gold and silver mines were considerably reduced by the 1930s, emissions from North American smelters, incinerators, landfills, fossil fuel combustion, and other sources increased rapidly to about 0.3×10^6 kg yr^{-1} from the early 1900s to the 1970s, with one-third being produced in the Great Lakes region (Pirrone et al. 1998). When released to the atmosphere, Hg is primarily in its elemental form (Hg^0), which has a long atmospheric residence time (Gbor et al.

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Acknowledgments

We thank the Ontario Ministry of Natural Resources biologists who provided the tissue samples that formed the basis of this study, and Emily Awad (Ontario Ministry of Environment, MOE) for maintaining the contaminant databases. Robert O'Gorman (United States Geological Survey) is thanked for directing us to his published data on Lake Ontario alewife abundance indices. Dave Dolan (University of Wisconsin-Green Bay, Natural and Applied Sciences) kindly provided us with phosphorus loading estimates for Lake Ontario. Eric Reiner and Rusty Moody (MOE Laboratory Services) provided information regarding analytical methods. Our manuscript was greatly improved by the constructive comments of Charles Driscoll (Syracuse University) and those of an anonymous reviewer.

Funding was provided by MOE, NSERC awards to L.M.C. and D.A.J., and Queen's Graduate Awards to T.D.F.

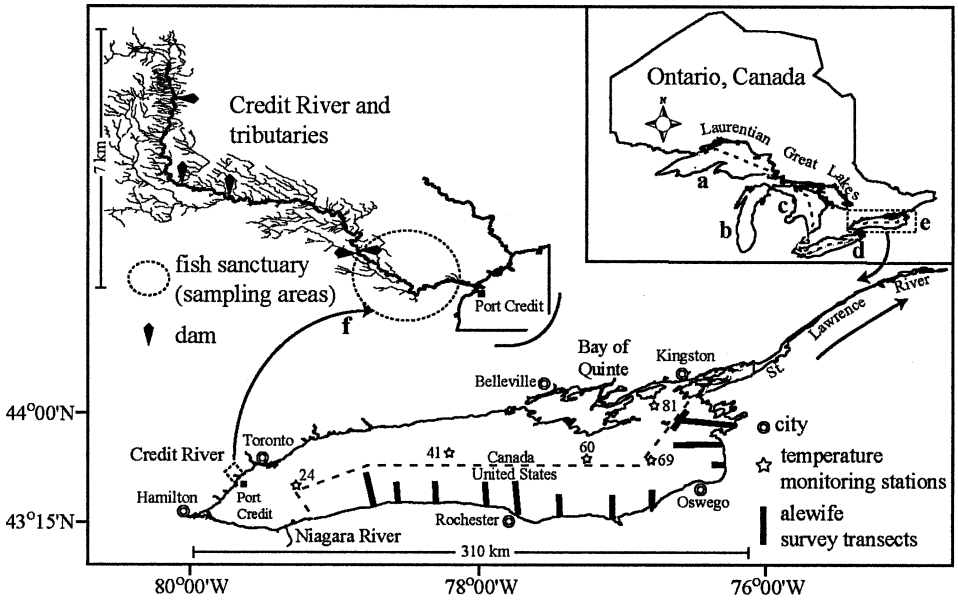

Fig. 1. The Laurentian Great Lakes (*inset*), showing the connectivity of Lakes (a) Superior, (b) Michigan, (c) Huron, (d) Erie, and (e) Ontario. Lake Ontario (expanded scale) shows O'Gorman et al.'s (1997, 2004) alewife survey transects, Environment Canada's water temperature monitoring stations, and Bay of Quinte (at Belleville municipal water intake) temperature monitoring station. Salmon were sampled from the lower reaches of the (f) Credit River.

2006). Thus, Hg depositional rates in the Great Lakes region, like those of some POPs, are a function of local, regional, and global emissions. Liquid effluents, such as those from chlor-alkali (until 1995) and sewage treatment plants, also contribute significant quantities of Hg to the Great Lakes (Pirrone et al. 1998; Trip et al. 2004). There are indications that total Hg loads are declining in the Great Lakes region (e.g., Trip et al. 2004); however, recent studies indicate that North American and global atmospheric loads have stabilized at high levels (Blanchard et al. 2002; Temme et al. 2004) and have even increased in some regions (Donahue et al. 2006).

We used long-term (20+ yr) datasets to determine how the sum of 209 polychlorinated biphenyl congeners (ΣPCB or total PCBs), mirex, para-para DDT (*p,p′*-DDT), and total Hg (Tot-Hg) concentrations have changed in Lake Ontario coho salmon (*Oncorhynchus kisutch*, 1976–2003) and chinook salmon (*Oncorhynchus tshawytscha*, 1983–2003) over time. Published data on forage fish abundance, zooplankton density, and chlorophyll *a* concentration ([Chl *a*]) were used to determine whether changes correlated with ecosystem trends, and climatological data were used to identify linkages between contaminant burdens and El Niño (warming) and La Niña (cooling) events. Comparatively, the legacy POPs represent bioaccumulative compounds that have had limited use during the past 30 years and Tot-Hg those that are still released to the environment at high rates.

Materials and methods

Study area—Lake Ontario (18,600 km^2) is the smallest of the Laurentian Great Lakes, which collectively hold nearly 20% of the world's fresh surface water (Fig. 1). More than 33 million people reside within the 521,830-km^2 Great Lakes watershed, with 25% living in the Lake Ontario sub-basin. Being furthest downstream, the water and sediment quality of Lake Ontario are also affected by human activities in the Superior, Michigan, Huron, and Erie sub-basins. In the Great Lakes, coho and chinook salmon are high trophic-level predators that can live for 3–5 yr; thus, they are ideal species to use in pollution and biomagnification studies.

In this study, long-term datasets on tissue contaminants are for adult Credit River coho and chinook salmon populations. About 25,000 chinook salmon return to the river annually to spawn, with the number of returning coho salmon being less than half this number. The Credit River starts above the Niagara Escarpment and empties into Lake Ontario near Toronto (Fig. 1). It is 90-km long with 1,500 km of tributaries. The watershed (1,000 km^2) has a population of 600,000 people, with 85% living in the lower one-third if the watershed. About 65% of the average daily flow (690,000 m^3) comes from groundwater. Flows are affected by five dams (Fig. 1), groundwater extractions, impervious surfaces, and agricultural irrigation. Water and sediment quality have been affected by deforestation, road building, agriculture, storm sewer runoff, and pollution from industrial and municipal sources.

Sampling and laboratory analyses—During the autumn to early winter of each year (September to January, depending on run timing), adult coho (1976–2003) and chinook (1983–2003) salmon migrating up the Credit River to spawn were netted (Fig. 1). Fish were sexed, lengthed

(total) to the nearest 0.1 cm, and weighed to the nearest 1 g. A section of skinless/boneless epaxial muscle was excised from a subsample of fish representing observed size ranges. Muscle sections were wrapped in aluminum foil, frozen, and transported to the Ontario Ministry of Environment (MOE) lab (Toronto), which is accredited by the Canadian Association for Environmental Analytical Laboratories (CAEAL). In the laboratory, muscle sections were partially thawed and homogenized. Homogenates were transferred to 20-mL glass scintillation vials and frozen (−20°C) until analyzed.

Homogenates were thawed and 5.0 ± 0.1 g for %lipid, [ΣPCB], [mirex], and [*p,p′*-DDT] determinations transferred to centrifuge tubes, and 0.5 mL of a 5 : 2 mixture of decachlorobiphenyl and 1,3,5-tribromobenzene was added to each as a surrogate spike. Concentrated hydrochloric acid (18 mL) was added to each tube. After an overnight digestion, 20 mL of a 25% (v/v) dichloromethane in hexane solvent was added to each tube. Solutions were mixed for 45 minutes then racked for 24–48 h. Upper solvent layers were quantitatively pipetted and transferred to 100-mL volumetric flasks and then diluted to 100 mL with solvent. Aliquots (20 mL) of the initial extracts were transferred to preweighed 50-mL beakers and dried for 24 h. After drying, beakers were weighed again and %lipid (detection limit, DL = 0.1%) computed (Eq. 1):

$$\%\text{lipid} = \frac{(B + R) - B}{S \times Va/Ve} \times 100 \qquad \text{(Eq. 1)}$$

where (B + R) is the weight of the beaker with residue, B is the beaker weight before receiving extract, S is the wet weight of original sample, Va is the aliquot volume, and Ve is the total extract volume.

For the analysis of [ΣPCB], [mirex], and [*p,p′*-DDT], initial extracts (1.0 g) were transferred to 200-mL evaporating tubes, and 2 mL iso-octane was added to each. Tube contents were evaporated to 1.0 mL at about 31°C. A 0.5-cm glass wool plug was inserted into the bottom of each chromatographic column, then activated Florisil was packed to 24 cm. Evaporated extracts (1 mL) were added to the columns and drained to the top of the packing. Pure hexane was then added in 1-mL portions until the columns were fully wetted, followed by a 25-mL hexane elution. Column effluents, containing PCB and mirex, were collected in 40-mL graduated tubes. Columns were eluted again with a 25% (v/v) dichchloromethane in hexane solution (25 mL), with effluents containing *p,p′*-DDT collected in 40-mL tubes. Pure iso-octane (1 mL) was added to the fractions, then samples were evaporated to 1 mL. Gas-liquid chromatography was used to determine [ΣPCB] (HP 5890 Series II, Ni^{63} electron capture detectors [ECD], DL = 20 ng g^{-1} wet wt), [mirex] (HP 5890, Ni^{63} ECD, DL = 5 ng g^{-1} wet wt), and [*p,p′*-DDT] (HP 6890 Series Plus, dual column micro Ni^{63} ECD, DL = 5 ng g^{-1} wet wt). A split/splitless injector and helium gas (ultrahigh purity) supply were used in all systems. Column head pressures were 3.5 psi (ΣPCB), 20 psi (mirex), and 35 psi (*p,p′*-DDT), and temperatures at ECDs were 300°C. Standardization and reference stock solutions were purchased from commercial suppliers. Calibration curves were based on six concentrations encompassing the range of tissue concentrations and were accepted if correlation coefficients were ≥0.985.

In Ontario, the analysis of PCBs in fish started in the 1960s. Chromatography techniques used at this time separated the 209 congeners into 23 peaks at most, whereas the capillary columns currently used can separate almost all congeners. Quantification of [ΣPCB] over the record was based on calibrations with Aroclor standards (4 : 1 Aroclor 1254 to Aroclor 1260). To make long-term [ΣPCB] data comparable, the resolution of the current technique was detuned to that of the 1960s (i.e., to differentiate 23 peaks) and [ΣPCB] calculated by summing the concentration equivalents of the 23 peaks. For a positive measure, at least 11 peaks had to be resolved.

To ensure data quality and long-term comparability, the MOE organics lab participates in CAEAL audits. Instrument precision was routinely determined by analyzing 10 replicates of each standard solution. In 1999 for example, relative standard deviations (RSDs) were 9.3% (ΣPCB), 5.1% (mirex), and 4.6% (*p,p′*-DDT). Contamination was assessed by analyzing a blank with each sample run. Within- and between-run precision was regularly determined by duplicate analyses of a single sample. Whereas an acceptance criterion (AC) of ± 25% was applied to duplicates, precision was typically considerably better. For example, within-run duplicates analyzed in June 2000 had RSDs of 4.6% (ΣPCB), 4.9% (mirex), and 15.3% (*p,p′*-DDT). In 1998 to 1999, between-run RSDs were 19% (ΣPCB), 15% (mirex), and 24% (*p,p′*-DDT). Analyte recovery rates were determined for the surrogate spiking solutions and for blanks spiked with standard solutions (AC = 50–150%).

Homogenates (0.2–0.4 g) for [Tot-Hg] determinations were transferred to 50-mL digestion tubes and 5-mL acid (4 : 1 concentrated sulfuric to nitric [v/v]) added to each. Temperatures were maintained at 215°C to 235°C during overnight digestion. Digestates were diluted to 25 mL with pure water, mixed, transferred to culture tubes, and placed in a Gilson autosampler for [Tot-Hg] determinations by cold vapor-flameless atomic absorption spectroscopy (gold-film Jerome Model 511 Hg Analyzer, DL = 0.01 μg g^{-1} wet wt). Wave length was set at 253.7 nm, and 20 μL concentrated sulfuric acid was put in the impinger to dehydrate vapor. Calibration curves were based on five concentrations (made from National Institute of Standards and Technology [NIST] standards) encompassing the range of tissue concentrations and were accepted if correlation coefficients were ≥0.990.

The MOE inorganics lab has participated in interlaboratory comparison programs organized by the Department of Fisheries and Oceans Canada (1972–1998) and the Canadian Food Inspection Agency (1998 onward) since 1972. Generally, four tuna samples were sent to participating laboratories annually, and results were compared. The MOE results have been consistent with those of other laboratories, e.g., [Tot-Hg] measured by MOE during 2000 to 2004 were within ± 8% of the study average with no failures flagged. To test high- and low-range accuracy, two

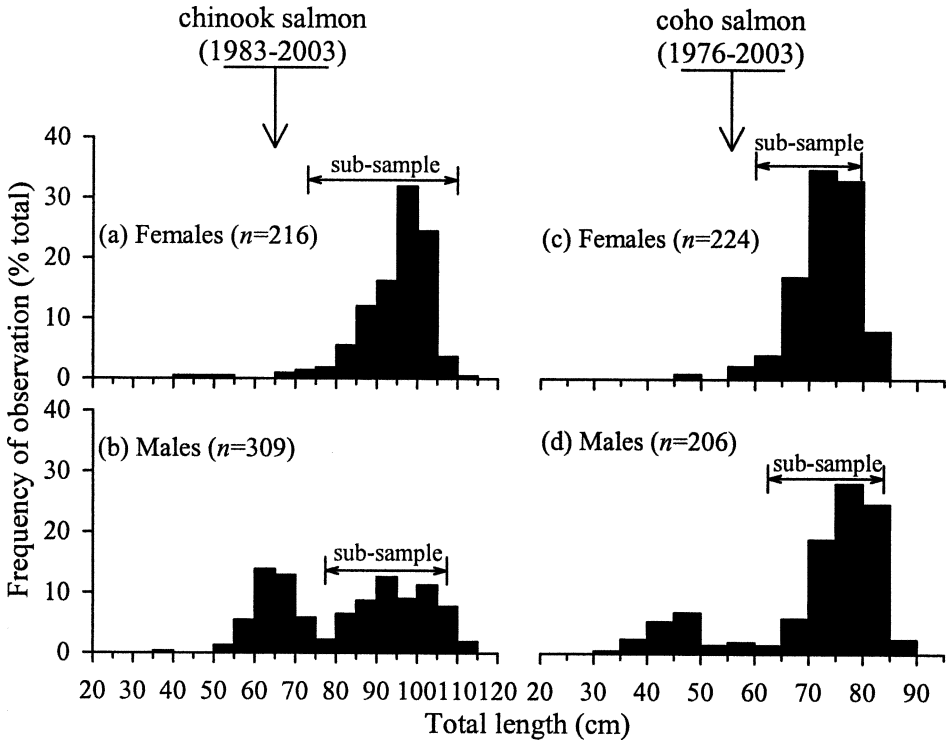

Fig. 2. The size distribution of Lake Ontario (a, b) female and male chinook salmon and (c, d) female and male coho salmon that were analyzed for [POPs] and [Tot-Hg].

fish control standards with known [Tot-Hg] were analyzed with each sample run: a high (0.600–1.000 μg g^{-1}) and low (0.100–0.400 μg g^{-1}) concentration standard. Accuracy was also tested by analyzing certified reference materials (e.g., the National Research Council of Canada's DORM-2) with each run. Within- and between-run duplicates were analyzed to test precision. Similarly, a series of precision tests were undertaken when methods were modified to ensure that data are comparable over time, i.e., results of original and modified methods compared. An AC of ± 3 SD was used in precision and accuracy tests. A blank (4 : 1 sulfuric to nitric acid mixture) and undigested NIST standard (diluted with 15% sulfuric acid rinsing solution) were analyzed in each sample run to test for contamination. Recovery rates (AC = 78–115%) were determined by analyzing a 1 mg Hg L^{-1} NIST standard solution and a spiked (0.2 mL of 1 mg L^{-1} standard) fish control standard with each sample run.

Data handling and temporal trends—Linear and non-linear regression techniques (SPSS 12.0) were used to determine whether within-year contaminant concentrations were significantly (α = 0.05) correlated with %lipids and fish size (weight and length). Analysis of variance was used to determine whether concentrations were sex dependent. Although [ΣPCB], [mirex], [*p,p'*-DDT], and [Tot-Hg] were often very low or high in the smallest and largest fish, respectively, concentrations were not sex dependent or correlated with size and %lipids within the size ranges: 72–110 cm = 3.77–14.50 kg (♀ chinook salmon); 77–107 cm = 4.20–13.69 kg (♂ chinook salmon); 60–80 cm = 1.50–5.68 kg (♀ coho salmon); and 62–84 cm = 2.50–6.72 kg (♂ coho salmon). Therefore, temporal trends in contaminant burdens were assessed by regressing average concentrations against year for fish within these size ranges, and data for males and females were pooled (Fig. 2). Contaminant concentrations in fish are often correlated with fish size (e.g., Borgmann and Whittle 1991). The absence of significant within-year correlations between size and concentration in the noted size ranges (Fig. 2) likely reflects the life-stage and ecological consistency of sampled individuals, i.e., all fish included in temporal analyses were adult spawners (the same age within species) and from the same population (Credit River). During the study period, 388 chinook salmon and 352 coho salmon within the considered size classes were analyzed for contaminants (Table 1).

Trophodynamic links—In the Great Lakes, coho and chinook salmon aged 2 yr and older feed almost exclusively on alewife (*Alosa pseudoharengus*) and most selectively on the largest individuals (Stewart et al. 1981; Jude et al. 1987; Jones et al. 1993). Because predatory fish incorporate contaminants mostly from their food (Borgmann and Whittle 1991), we hypothesized that temporal variations in contaminant burdens would be a function of alewife population dynamics. Age-specific abundance indices for Lake Ontario alewife, based on bottom trawls along 11 transects running perpendicular to the southern shoreline (Fig. 1), were published by O'Gorman et al. (2004) for the period 1978–2000. To determine whether the contaminant burdens of Lake Ontario coho and chinook salmon

Table 1. Total number of Lake Ontario coho and chinook salmon sampled for [ΣPCB], [mirex], [*p,p′*-DDT], and [Tot-Hg] over the periods 1983–2003 (chinook salmon) and 1976–2003 (coho salmon).

Year	Coho salmon	Chinook salmon
1976	24 (*n* Tot-Hg=0)*	0
1977	19 (*n* Tot-Hg=18)*	0
1978	8 (*n* Tot-Hg=0)*	0
1979	20	0
1980	0	0
1981	12	0
1982	19	0
1983	17	19
1984	27	24
1985	18	17
1986	18	18
1987	20	20
1988	20	20
1989	14	20
1990	27	43
1991	18	42
1992	15	41
1993	19	10
1994	0	16
1995	0	15
1996	0	11
1997	4	12
1998	0	9
1999	2	11
2000	10	5
2001	6	13
2002	5	9
2003	10	13
N	352	388

* Total Hg concentrations were not measured as often as [POPs] from 1976 to 1978.

correlated with alewife dynamics, relative abundance indices for large (>149 mm) alewife were regressed against average annual contaminant concentrations. Alewife population dynamics were, in turn, correlated with indicators of trophic-level change: (1) average (June through October) epilimnetic zooplankton density (1981–1995) (Johannsson et al. 1998), (2) average (June through October) [Chl *a*] (1978–1995) (Johannsson et al. 1998), (3) spring overturn total phosphorus concentrations ([Tot-P]) (1978–1995) (O'Gorman et al. 2004), and (4) annual Tot-P loading rates (1978–1992) (Dr. D. Dolan, International Joint Commission, Windsor, ON, unpubl. data presently with the University of Wisconsin-Green Bay, Natural and Applied Sciences). Zooplankton densities (not including veligers or nauplii) and summer [Chl *a*] were based on pooled data from sites 41 and 81 (Fig. 1).

Climate cycles—Field observations (Rand and Stewart 1998; Wurster et al. 2005) and bioenergetic simulations (Rand et al. 1994) have shown and predicted, respectively, that salmonine feeding rates increase with increasing ambient temperature. Because Lake Ontario coho and chinook salmon feed preferentially on the largest available alewife, average alewife size would be expected to decrease with increasing ambient temperature if predation has a significant role in determining the size structure of the alewife population. Indeed, Jones et al. (1993) have shown that the average size of Lake Ontario alewife decreases in response to increased predation by salmon. Given that ambient temperature can affect the size structure of the alewife population by affecting predation rates and that alewife contaminant burdens are a function of size (O'Gorman and Schneider 1986; Madenjian et al. 1995), we hypothesized that contaminant levels in Lake Ontario coho and chinook salmon should be a function of summer (growing season) epilimnetic water temperatures.

Representative water temperature data were not available for the main basins of Lake Ontario; therefore, direct correlations between interannual variations in contaminant concentrations and summer water temperature could not be made. The Bay of Quinte is Lake Ontario's largest embayment (Fig. 1), and epilimnetic temperatures have been measured continuously in the embayment at the Belleville municipal water intake (3.2-m depth, on average) for >60 years (Casselman 2002). However, epilimnetic temperatures in the embayment would not be expected to be directly representative of those in the more open, and much deeper, main basins of Lake Ontario where coho and chinook salmon are most abundant. Climatic cycles in the Lake Ontario region have also been monitored directly from daily measurements, including air temperature, taken at Environment Canada's Toronto (Pearson International Airport since the 1950s) and Kingston (Norman Rogers Airport since the 1930s) meteorological stations.

To determine whether epilimnetic temperatures in the main basins of Lake Ontario were correlated with those in the Bay of Quinte, measures from the Bay of Quinte were paired by sampling date with available epilimnetic temperatures (Environment Canada data) from sites 24, 41, 60, 69, and 81 (Fig. 1). Linear and nonlinear regression techniques were used to develop best-fit models describing the relationships between Bay of Quinte summer temperatures and temperatures at the five Lake Ontario sites. Average summer (June through August) epilimnetic temperatures in the Bay of Quinte were then correlated with average summer air temperatures at Kingston to determine the relationship between air and epilimnetic temperatures for the period 1976–2003. Air temperatures at Kingston were downloaded from Environment Canada's on-line (http://www.climate.weatheroffice.ec.gc.ca/Welcome_e.html) National Climate and Information Archive. Average summer epilimnetic temperatures in Lake Ontario (outside the Bay of Quinte) were assumed to be a function of average summer air temperature if the correlations between temperatures in the Bay of Quinte and the Lake Ontario sites and those between air temperature and Bay of Quinte temperature were significant.

Thus, in the absence of having suitable long-term epilimnetic temperatures for the main basins of Lake Ontario, average summer air temperature was used as a correlative surrogate. Since coho and chinook salmon gain most biomass in the lake during their last two summers of life, temporal changes in contaminant concentrations were quantified in relation to the average of daily summer air

temperatures during the current year (spawning year for each cohort) and the previous year (i.e., an integration of thermal conditions across two summers). Daily summer air temperature was calculated as the average of temperature readings at Toronto and Kingston, which are located at opposite ends of Lake Ontario (Fig. 1).

Results

Temporal patterns—Based on average annual concentrations, exponential decay models best described changes in [POPs] in Lake Ontario coho and chinook salmon, such that declines occurred most rapidly during the first few years that measurements were taken and more slowly with time (Fig. 3–c; Table 2 Eqs. 2–4, 6–8). Total PCB concentrations decreased from maxima of 4,217 ± 1,674 (average ± 1 SD) ng g^{-1} in 1976–1977 (coho salmon) and 4,139 ± 1,188 ng g^{-1} in 1983–1984 (chinook salmon) to minima of 324 ± 110 ng g^{-1} (coho salmon) and 432 ± 101 ng g^{-1} (chinook salmon) in 2002–2003 (90–94% decrease; Fig. 3a). [Mirex] reductions were similar, with concentrations decreasing 90–95% over the record (Fig. 3b). Reductions of [*p,p′*-DDT] in coho salmon decreased from a maximum of 158 ± 86 ng g^{-1} in 1976–1977 to a minimum of 5 ± 0 ng g^{-1} in 2002–2003 (97% decrease), with those in chinook salmon decreasing from 91 ± 47 ng g^{-1} in 1983–1984 to 9 ± 2 ng g^{-1} in 2002–2003 (90% decrease; Fig. 3c). Regressions of annual variability (SD) versus year showed linear decreases of ΣPCB (r^2 = 0.60–0.68; $p < 0.001$), mirex (r^2 = 0.35–0.47; p = 0.001–0.004), and *p,p′*-DDT (r^2 = 0.61–0.62; $p < 0.001$) variability in coho and chinook salmon over the record (Fig. 3a–c). In comparison, declines in average annual [Tot-Hg] in Lake Ontario coho and chinook salmon were very slight, although statistically significant, with linear models best describing changes over the record (Fig. 3d; Table 2 Eqs. 5, 9). While among-year variability in [POPs] declined linearly in coho and chinook salmon over the record, variability in average [Tot-Hg] remained constant for both species.

Closer examinations of the trend lines revealed that [POPs] and [Tot-Hg] oscillated upward and downward at approximately the same frequency through the record. Using [ΣPCB] in chinook salmon as an example (Fig. 3a), concentrations declined to a minimum from 1988 to 1990, then increased to a maximum from 1990 to 1992. After the 1990 to 1992 maximum, concentrations declined to a minimum from 1993 to 1995. The next concentration maxima occurred in 1996–1997 and in 2000, with the next minima occurring in 1998–1999 and in 2000 to the end of the record (Fig. 3a). Another conspicuous trend was that the amplitude and duration of the upward and downward oscillations decreased as time progressed (Fig. 3). These concentration oscillations were subsequently linked to trophodynamic and climatological factors.

Trophodynamic links—Average annual [POPs] in Lake Ontario coho and chinook salmon tracked the abundance of large alewife during the portion of the record extending from 1980 to 2003 (Fig. 3a–c). During this period, linear models showed that alewife abundance accounted for 44% to 55% (p = 0.002–0.010) of variations in average annual concentrations in coho salmon, and 39% to 56% (p = 0.001–0.007) of those in chinook salmon (Table 2 Eqs. 10–12, 14–16). Associations between average annual [Tot-Hg] and alewife abundance were not as strong (r^2 = 0.26–0.29); however, they were statistically significant (p = 0.036–0.048) (Fig. 3d; Table 2 Eqs. 13,17). The association between alewife abundance and salmon contaminant burdens did not hold for years prior to 1980 when [POPs] and [Tot-Hg] in coho salmon were highest despite the very low numbers of large alewife (Fig. 3, coho salmon panels).

It appears that bottom-up trophic changes have driven the overall downward trend of Lake Ontario alewife abundance and associated reductions in coho and chinook salmon contaminant burdens. Although they sometimes prey on small fishes, alewife feed primarily on zooplankton. Based on epilimnetic samples, zooplankton densities in Lake Ontario have declined substantially since the early 1980s, and the alewife decline tracked this trend (r^2 = 0.51, p = 0.003; Fig. 4a). In turn, the zooplankton decline tracked reductions in average summer [Chl *a*] (r^2 = 0.24, p = 0.064; Fig. 4b), with the reduction in [Chl *a*] having tracked reductions in spring overturn [Tot-P] (r^2 = 0.36, p = 0.011; Fig. 4c). Reductions in spring overturn [Tot-P] in Lake Ontario from the early 1970s to the mid-1980s were attributable to reduced external Tot-P loadings (r^2 = 0.64, p = 0.013); however, [Tot-P] continued to decline from the mid-1980s to the early 1990s despite there being years with elevated Tot-P loading (Fig. 4d). The breakdown of the positive correlation between spring overturn [Tot-P] and Tot-P loading occurred in conjunction with the proliferation of zebra (*Dreissena polymorpha*) and quagga (*Dreissena bugensis*) mussels (Fig. 4d, inset).

Climatic cycles—Average summer air temperatures at Kingston and Toronto were highly variable during the study period, with average temperatures during the last three years of record (2001–2003) being 2.4°C warmer than those during the earliest three years (1976–1978) (Fig. 5). Upward and downward oscillations in summer temperatures were linked to strong El Niño (warming) and La Niña (cooling) cycles and to the Mt. Pinatubo (Philippines) eruption (cooling) (Fig. 5c). Summer epilimnetic temperatures at the Lake Ontario sites can be approximated from those in the Bay of Quinte with models given in Table 3 (Eqs. 26–29). Linear regression, in turn, showed that average summer epilimnetic temperatures in the Bay of Quinte were positively correlated with average summer air temperature over the record (Table 3 Eq. 30). Thus, it was concluded that average summer epilimnetic temperatures at the Lake Ontario sites are at least partially dependent on summer air temperatures. The temperature associations suggest that average summer epilimnetic temperatures in Lake Ontario have changed in an upward trajectory over the record.

Log_{10} of average annual [POPs] in Lake Ontario coho (r^2 = 0.43–0.56, $p < 0.001$) and chinook (r^2 = 0.50–0.51, $p <$ 0.001) salmon decreased with increasing average summer

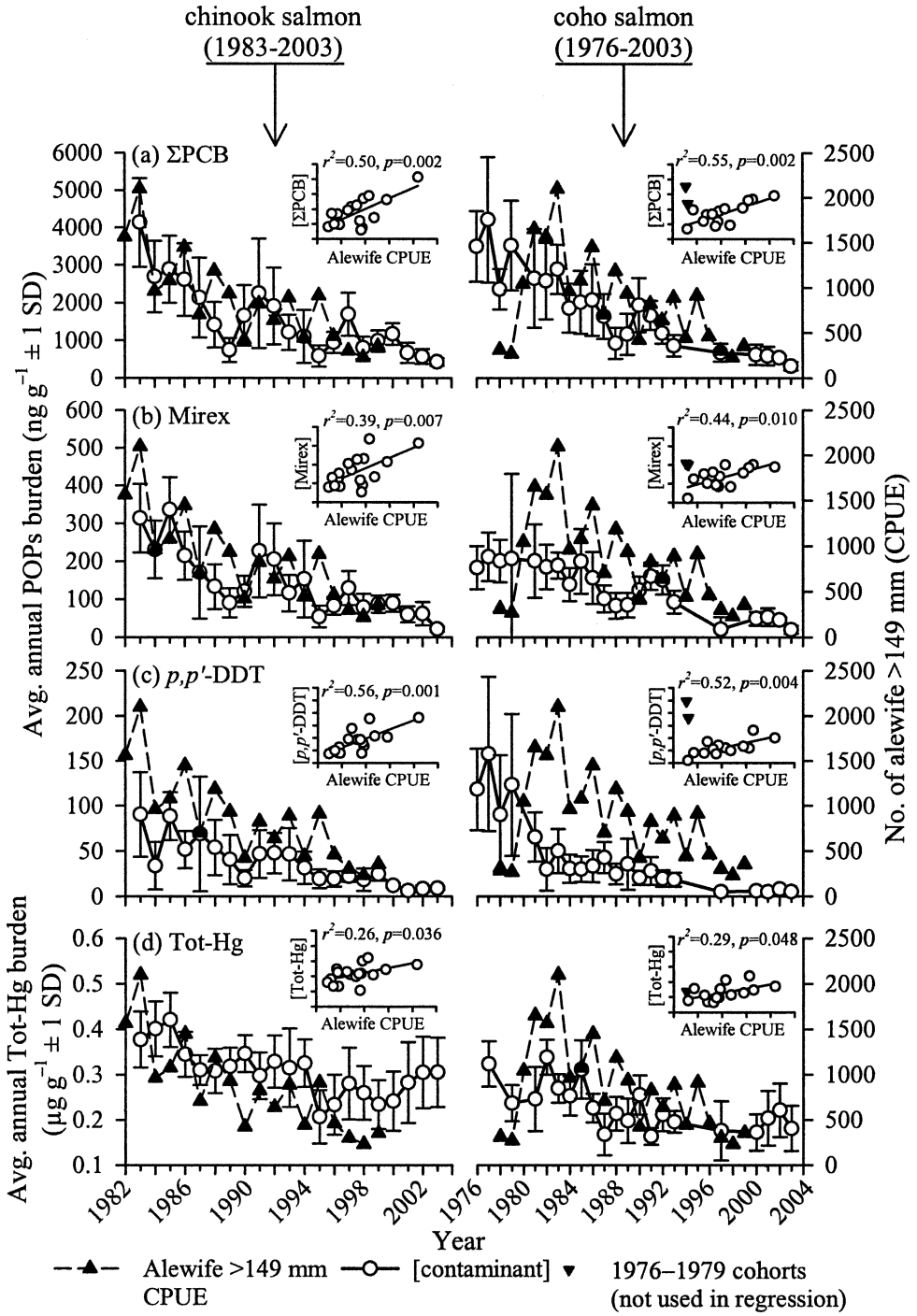

Fig. 3. Temporal trends in (a) [ΣPCB], (b) [mirex], (c) [*p,p'*-DDT], and (d) [Tot-Hg] in Lake Ontario chinook and coho salmon (Table 2 Eqs. 2–9) and association between concentration and large (>149 mm) alewife catch-per-unit-effort (CPUE, *insets*) (Table 2 Eqs. 10–17). Data for large alewife CPUE are from O'Gorman et al. (2004). Data collected before 1980 were not included in regression lines (insets).

air temperatures over the record (Fig. 6a–c; Table 2 Eqs. 18–20, 22–24). Similarly, [Tot-Hg] decreased with increasing summer temperature over the record (r^2 = 0.30–0.31, p = 0.008–0.013) (Fig. 6d; Table 2 Eqs. 21, 25). On a finer temporal scale, Fig. 5 shows (*see* vertical bars) that downward shifts in contaminant concentrations observed after 1980 were often associated with very warm summers (El Niño cycles), with upward shifts often being associated with cool summers (La Niña cycles and Mt. Pinatubo eruption).

Table 2. Average annual contaminant concentrations in Lake Ontario chinook and coho salmon over time (year of record) and in relation to adult (>149 mm) alewife abundance and average summer air temperature. Methods used to calculate variables given in text.

Predicting average annual contaminant concentration in Lake Ontario chinook (I) and coho (II) salmon*					
From year of record†	Eq.	From adult alewife abundance‡	Eq.	From summer air temperature§	Eq.
I. Chinook salmon					
$[\Sigma PCB] = 3{,}207 \times e^{-0.082(yr)}$ ($r^2 = 0.68$, $p < 0.001$)	(2)	$[\Sigma PCB] = 1.4X + 593$ ($r^2 = 0.50$, $p = 0.002$)	(10)	$\log([\Sigma PCB] + 1) = -0.23X + 7.70$ ($r^2 = 0.50$, $p < 0.001$)	(18)
$[Mirex] = 310 \times e^{-0.088(yr)}$ ($r^2 = 0.69$, $p < 0.001$)	(3)	$[Mirex] = 0.12X + 72$ ($r^2 = 0.39$, $p = 0.007$)	(11)	$\log([Mirex] + 1) = -0.25X + 6.95$ ($r^2 = 0.51$, $p < 0.001$)	(19)
$[p,p'\text{-DDT}] = 93 \times e^{-0.109(yr)}$ ($r^2 = 0.77$, $p < 0.001$)	(4)	$[p,p'\text{-DDT}] = 0.04X + 13$ ($r^2 = 0.56$, $p = 0.001$)	(12)	$\log([p,p'\text{-DDT}] + 1) = -0.28X + 6.93$ ($r^2 = 0.50$, $p < 0.001$)	(20)
$[Tot\text{-}Hg] = -0.006(yr) + 0.375$ ($r^2 = 0.50$, $p < 0.001$)	(5)	$[Tot\text{-}Hg] = 6.3\times10^{-5}X + 0.3$ ($r^2 = 0.26$, $p = 0.036$)	(13)	$\log([Tot\text{-}Hg] + 1) = -0.01X + 0.36$ ($r^2 = 0.31$, $p = 0.008$)	(21)
II. Coho salmon					
$[\Sigma PCB] = 4{,}720 \times e^{-0.101(yr)}$ ($r^2 = 0.87$, $p < 0.001$)	(6)	$[\Sigma PCB] = 1.0X + 637$ ($r^2 = 0.55$, $p = 0.002$)	(14)	$\log([\Sigma PCB] + 1) = -0.26X + 8.24$ ($r^2 = 0.56$, $p < 0.001$)	(22)
$[Mirex] = 308 \times e^{-0.091(yr)}$ ($r^2 = 0.68$, $p < 0.001$)	(7)	$[Mirex] = 0.07X + 61$ ($r^2 = 0.44$, $p = 0.010$)	(15)	$\log([Mirex] + 1) = -0.26X + 7.05$ ($r^2 = 0.54$, $p < 0.001$)	(23)
$[p,p'\text{-DDT}] = 153 \times e^{-0.151(yr)}$ ($r^2 = 0.87$, $p < 0.001$)	(8)	$[p,p'\text{-DDT}] = 0.02X + 9$ ($r^2 = 0.52$, $p = 0.004$)	(16)	$\log([p,p'\text{-DDT}] + 1) = -0.33X + 7.75$ ($r^2 = 0.43$, $p < 0.001$)	(24)
$[Tot\text{-}Hg] = -0.006(yr) + 0.292$ ($r^2 = 0.49$, $p = 0.001$)	(9)	$[Tot\text{-}Hg] = 5.6\times10^{-5}X + 0.2$ ($r^2 = 0.29$, $p = 0.048$)	(17)	$\log([Tot\text{-}Hg] + 1) = -0.01X + 0.30$ ($r^2 = 0.30$, $p = 0.013$)	(25)

* Total PCB, mirex, and *p,p′*-DDT concentrations are ng g^{-1} wet wt.; Tot-Hg concentrations are $\mu g\ g^{-1}$ wet wt.

† Equation is best-fit line that describes changes in contaminant concentrations over data record, Fig. 3 (year for chinook = year of interest (1983 to 2003) − 1983 + 1; year for coho = year of interest (1981 to 2003) − 1981 + 1).

‡ X is relative abundance of alewife >149 mm, Fig. 3 insets.

§ X is average of current and previous summer air temperatures.

Discussion

Our analysis indicates that [ΣPCB], [mirex], and [*p,p′*-DDT] in Lake Ontario coho and chinook salmon declined in exponential trajectories over the past 20+ years (Fig. 3a–c; Table 2). Borgmann and Whittle (1991) analyzed >1,900 Lake Ontario lake trout (*Salvelinus namaycush*), another top-predator species, to assess changes in [PCB], [DDE, dichlorophenyl dichloroethylene], and [mirex] over an 11-yr period (1977–1988). They also found that concentrations decreased rapidly during the first few years of record and that rates of decline slowed from the early 1980s to the end of the record. Similar trends have been observed in Lake Michigan, such that [PCB] and [DDT] in coho and chinook salmon, lake trout, rainbow trout (*Oncorhynchus mykiss*), brown trout (*Salmo trutta*), alewife, and bloater chub (*Coregonus hoyi*) declined exponentially from the early 1970s to the early 1990s (DeVault et al. 1986; Stow et al. 1995). Suns et al. (1993) and Scheider et al. (1998) found that [PCB] and [DDT] in spottail shiner (*Notropis hudsonius*) declined rapidly from the mid-1970s to the early 1980s and then more gradually to the early 1990s at several Great Lakes locations. Scheider et al. (1998) showed similar trends in [ΣPCB] and [mirex] in Lake Huron lake trout and Ganaraska River (Lake Ontario) rainbow trout. Thus, there is considerable evidence that restrictions on PCB, mirex, and DDT use implemented in the 1970s have reduced burdens at high trophic levels.

In comparison, [Tot-Hg] in Lake Ontario coho and chinook salmon declined only slightly, if not negligibly, over the record with linear models best describing trends (Fig. 3d; Table 2). While variability about the mean declined with time for the POPs, annual variability in [Tot-Hg] remained constant, which further suggests that concentrations have changed little over the past 20+ years (Fig. 3). Similarly, Borgmann and Whittle (1991) observed gradual, approximately linear declines in [Hg] in Lake Ontario lake trout from 1977 to 1988. For the period 1981 to 1992, Scheider et al. (1998) observed almost constant [Tot-Hg] in Lake Huron and Lake Ontario walleye (*Sander vitreus*). While datasets extending back to the mid-1970s, including ours, indicate that [Hg] in high trophic-level predators have not changed appreciably with time, those extending back to 1970–1971 reveal trends similar to those of the legacy POPs. For example, an analysis by Scheider et al. (1998) showed that [Tot-Hg] in walleye from Lake St. Clair declined exponentially from 1970 to about 1978, with changes being minimal from the early 1980s to 1994. In a time-series (1971–1997) analysis of [Hg] in Lake Erie yellow perch (*Perca flavescens*), Weis et al. (2004) observed a similar exponential decrease in concentration during the early 1970s with concentrations being relatively stable thereafter. Interestingly, [Hg] in Lake St. Clair walleye, smallmouth bass (*Micropterus dolomieui*), and white sucker (*Catostomus commersoni*) followed ∪-shaped (quadratic) trends from 1971 to 1997, such that they declined rapidly from 1971 to the mid-1980s and then increased through to the end of the record (Weis et al. 2004). Concentrations in some Arctic fishes have also increased in recent years (Macdonald et al. 2005). Although we were unable to fit

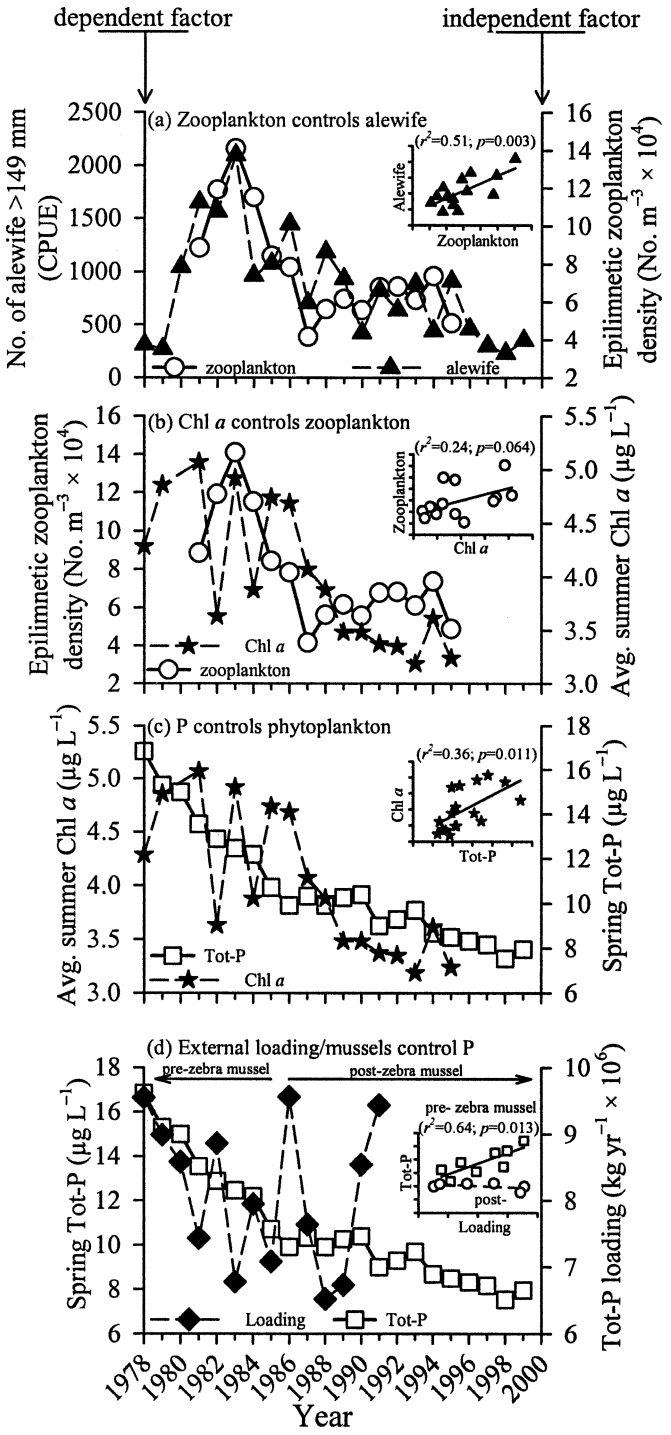

Fig. 4. The decline of large (>149 mm) alewife in Lake Ontario since the early 1980s tracked declines in (a) epilimnetic zooplankton density (not including veligers or nauplii) that were, in turn, associated with declines in (b) average summer [Chl *a*] (not corrected for phaeophytin). Reductions in summer [Chl *a*] tracked decreases in (c) spring-overturn [Tot-P] that were the result of (d) reduced anthropogenic Tot-P loadings. The tight relationship between (d, *inset*) spring overturn [Tot-P] and loading rates broke down following the introduction of zebra and quagga mussels, after which [Tot-P] were considerably lower than those that would be predicted from concurrent loading rates. *See* text for data sources.

significant quadratic curves to our Hg time-series for Lake Ontario coho and chinook salmon, the last five years of data for chinook salmon suggest that [Tot-Hg] are increasing (Fig. 3d).

As described, temporal trends in legacy [POPs] in Lake Ontario coho and chinook salmon were considerably different than those for [Tot-Hg]. Three possible explanations for this are: (1) PCBs, mirex, and DDT are synthetic compounds, so concentrations in highly mobile pools (e.g., water, biota, atmosphere) would be expected to "flush out" of the system after the implementation of use restrictions, (2) once PCBs, mirex, and DDT break down they do not re-form, unlike Hg (an element) that does not degrade and cycles perpetually between organic and inorganic phases, and (3) because Hg is found naturally in virtually all environmental compartments and is readily mobilized by many industrial and domestic activities, releases to the environment are comparatively difficult to control. Thus, there are many modern sources of Hg pollution in the Great Lakes region (Trip et al. 2004) that are, in effect, keeping ecosystem-level concentrations high.

When viewed in relation to production and sales histories, it is clear that the rapid declines of [ΣPCB], [mirex], and [*p,p′*-DDT] in Lake Ontario coho and chinook salmon from the mid-1970s to the early 1980s resulted from Canada–United States bans on usage. From five sediment cores collected along the long-axis of Lake Ontario, Wong et al. (1995) determined that the highest concentrations are in layers corresponding to the 1960s to the early-1970s (ΣPCB, mirex) and the mid-1950s to the early-1960s (ΣDDT). Following the concentration maxima, concentrations in bottom sediments declined exponentially to layers corresponding to the early 1990s (end of record), with the onset of the exponential declines coinciding with the implementation of usage bans (Oliver et al. 1989; Wong et al. 1995). The timing and pattern of concentration declines in sediment profiles appear similar to those observed in coho and chinook salmon tissue (Fig. 3–c), suggesting that sedimentation and burial have had a significant role in the gradual removal of PCBs, mirex, and *p,p′*-DDT from Lake Ontario ecosystems. Concentration declines over the record also reflect the decay-related half lives of the compounds, which have not been determined for the various sinks in natural aquatic systems, and export via the St. Lawrence River, which has not been quantified.

Temporal trends in [Tot-Hg] in Lake Ontario coho and chinook salmon also correspond well with trends in dated sediment profiles, such that [Hg] and deposition rates were highly variable in sediment layers corresponding to the late-1960s through 1998, with overall concentration reductions during the period being minimal with respect to progression toward baseline values (e.g., Marvin et al. 2004). On a finer scale, the exponential declines in [Hg] observed in fish from Lake St. Clair and western Lake Erie during the early 1970s (above) occurred when Hg accumulation rates in Lake Ontario bottom sediments were changing in a downward trajectory (Pirrone et al. 1998). Overall, the cycling of historically released Hg and modern loadings (Trip et al. 2004; Macdonald et al. 2005) are slowing the

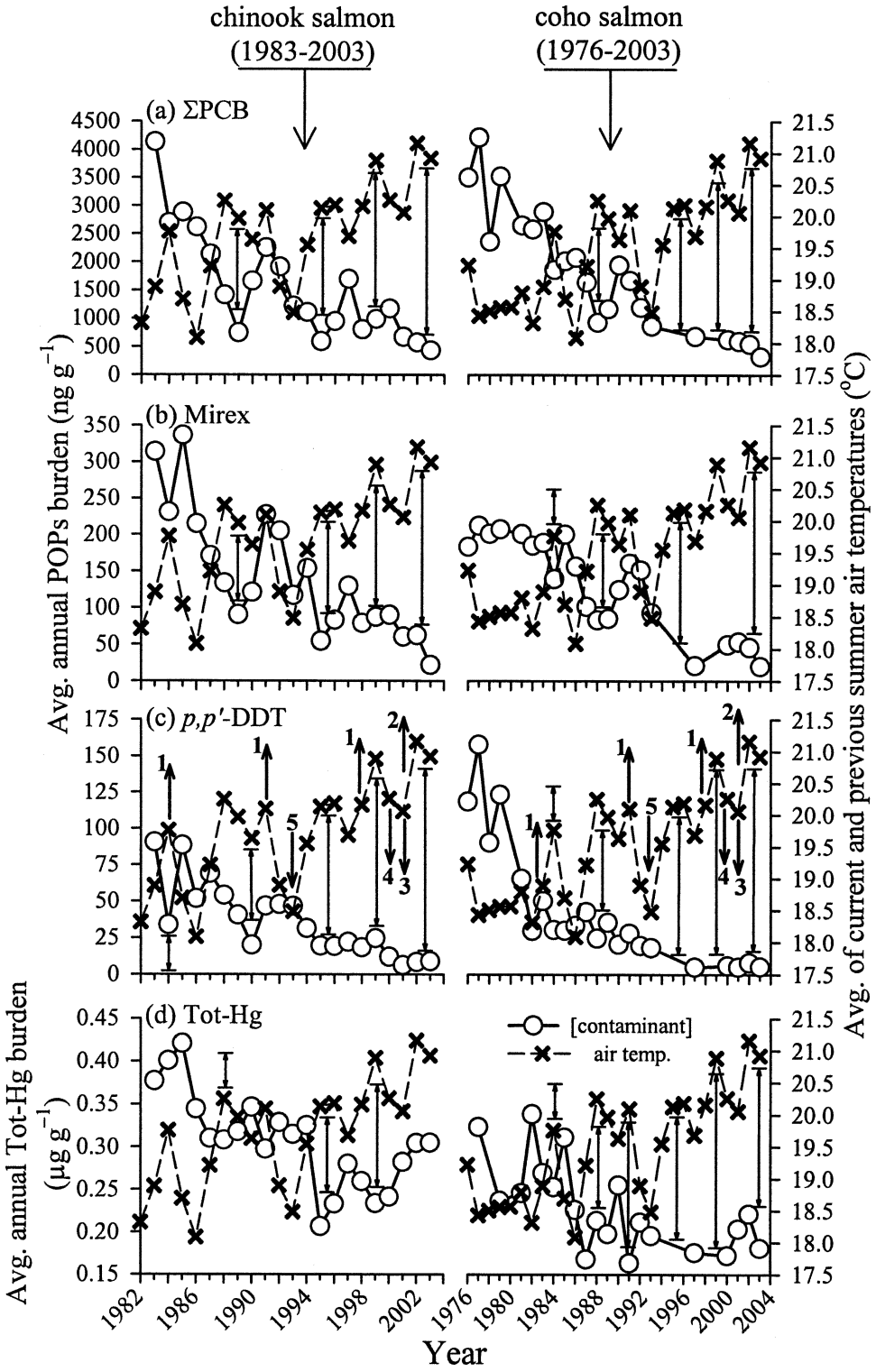

Fig. 5. Following the initial rapid reductions in (a) [ΣPCB], (b) [mirex], (c) [*p,p′*-DDT], and (d) [Tot-Hg] during the late 1970s and the early 1980s, concentrations oscillated in a downward trajectory to 2003. The regularly observed downward oscillations in contaminant concentrations (vertical bars) were often associated with warming El Niño events. Warming (up arrows marked 1 [= strong El Niño] and 2 [= moderate El Niño]) and cooling (down arrows marked 3 [= strong La Niña], 4 [= moderate La Niña], and 5 [= Mt. Pinatubo, Philippines, eruption]) events are indicated on panel (c).

Table 3. Epilimnetic temperatures at four Lake Ontario sites in relation to those in Bay of Quinte (BoQ) and average summer epilimnetic temperatures in BoQ in relation to average summer air temperature (*see* Fig. 1 for site locations). Methods used to calculate variables given in text.

Epilimnetic temperatures at Lake Ontario sites from those in the BoQ*	Eq.	Average summer epilimnetic temperature in BoQ from average summer air temperature†	Eq.
°C at Site 81 = 57.2 × (−874/BoQ) (r^2 = 0.64, $p < 0.001$, n = 36 pairs)	(26)	Avg. BoQ (°C) = 0.5 × Air (°C) + 12.9 (r^2 = 0.58, $p < 0.001$, n = 28 years)	(30)
°C at Site 69 = 70.9 × (−1217/BoQ) (r^2 = 0.57, $p < 0.001$, n = 37 pairs)	(27)	—	—
°C at Site 60 = 70.8 × (−1221/BoQ) (r^2 = 0.62, $p < 0.001$, n = 37 pairs)	(28)	—	—
°C at Site 41 = 63.7 × (−1143/BoQ) (r^2 = 0.63, $p < 0.001$, n = 39 pairs)	(29)	—	—

* BoQ is epilimnetic temperature in Bay of Quinte at Belleville municipal water intake.
† Air (°C) is average summer air temperature at Kingston airport from 1976 to 2003.

rate at which Great Lakes ecosystems can recover from contamination.

After the initial period of rapid decline (1976 to mid-1980s), [ΣPCB], [mirex], and [*p,p′*-DDT] in Lake Ontario coho and chinook salmon oscillated upward and downward at similar frequencies (Fig. 3a–c). Oscillations in [Tot-Hg] were also apparent, although less pronounced (Fig. 3d). Our initial interpretation of these oscillations was that they might have resulted from slight changes in analytical performance and/or sampling bias. However, similar oscillations are also apparent in shorter Great Lakes time-series for several species (e.g., DeVault et al. 1986; Borgmann and Whittle 1991; Scheider et al. 1998), which suggests that they are meaningful. If the declines in legacy [POPs] were attributable only to use restrictions, physical, chemical, and hydrological factors (above), concentration trajectories should have been consistently downward. The temporal regularity and synchronicity of the oscillations within the overall downward trend suggested to us that they were somehow related to food web dynamics and/or climatic cycles. Based on the principle that fish incorporate the bulk of their contaminant burdens from their food, Borgmann and Whittle (1991) were first to suggest that annual variations in contaminant concentrations in Lake Ontario lake trout should be linked to the population dynamics of prey species. Our results indicate that the overall downward trajectory and periodic oscillations of [POPs] and [Tot-Hg] in Lake Ontario coho and chinook salmon from the early 1980s onward were indeed positively correlated with the abundance of large alewife (Fig. 3; Table 2). The correlation did not hold for the years 1976 to 1980 when contaminant concentrations were at an all-time high despite the low numbers of alewife (Fig. 3, coho salmon panels). Alewife abundance was low during this period because of a major die-off in the unusually cold and prolonged winter of 1976–1977, before which alewife were very abundant (O'Gorman et al. 1987).

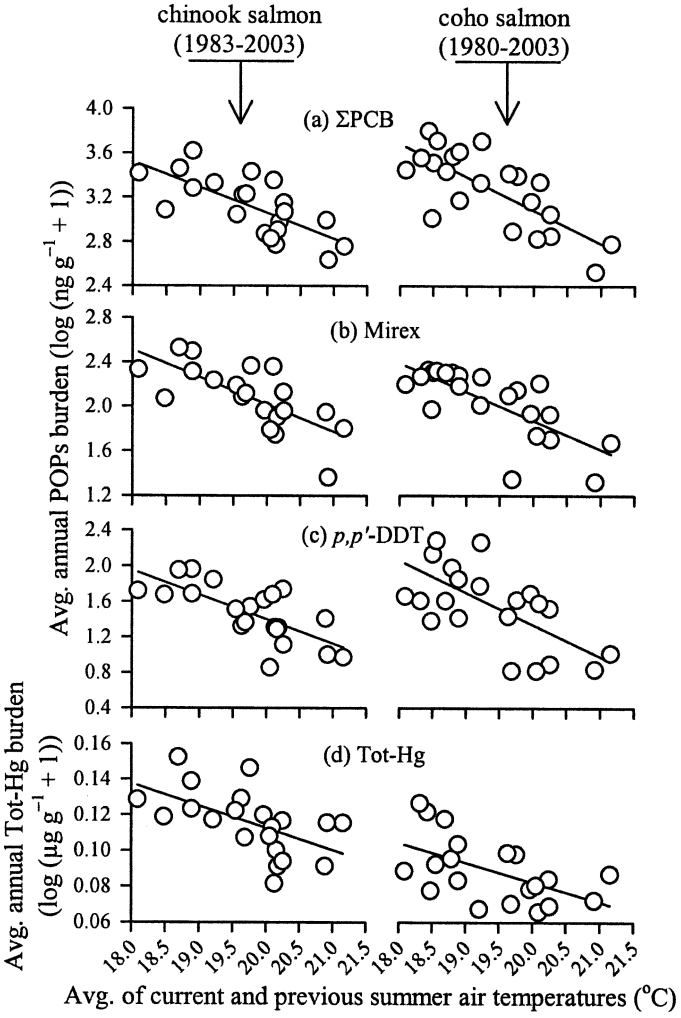

Fig. 6. Log_{10} average annual (a) [ΣPCB], (b) [mirex], (c) [*p,p′*-DDT], and (d) [Tot-Hg] in Lake Ontario coho and chinook salmon were negatively correlated with average summer air temperature (Table 2 Eqs. 18–25).

Assuming that alewife had a mechanistic role in determining contaminant transfer rates to Lake Ontario coho and chinook salmon from 1980 onward (Fig. 3, *insets*), food-web–related events might have also had a role in affecting the decline of contaminant concentrations early in the record when concentrations declined rapidly. Two major food-web–related events took place in the late 1970s: (1) the 1976–1977 alewife winterkill (Fig. 3, coho salmon panels), and (2) the enhancement of salmonine stocking programs. After the 1976–1977 alewife winterkill, there was

a rapid resurgence of the population from 1978 to 1983 when densities of large adults reached a 25-yr maximum (Fig. 4a). Because alewife growth rates are density-dependent, growth rates during this period were very high (O'Gorman and Schneider 1986), and this would have resulted in growth-related contaminant dilution (Borgmann and Whittle 1991, 1992; Madenjian et al. 1995). Therefore, reductions of [POPs] in alewife during this period of fast growth (e.g., Madenjian et al. 1995) would have decreased the exposure of coho and chinook salmon to contaminants. In this view, we suggest that the 1976–1977 alewife winterkill accelerated the initial rates at which contaminant concentrations declined in Lake Ontario coho and chinook salmon.

Salmonine stocking rates to Lake Ontario increased steadily from 2.5 million fish yr^{-1} in 1978 to 8 million fish yr^{-1} by 1984 (Mills et al. 2003). Since coho and chinook salmon feed preferentially on the largest available alewife, enhanced stocking would have, theoretically, reduced average alewife size. Because contaminant concentrations in alewife are lowest in the smallest individuals (e.g., Madenjian et al. 1995) stocking would be expected to reduce the rate at which contaminants are transferred from alewife to their predators. Supporting this idea, surveys by Rand et al. (1994) found that the average size of Lake Ontario alewife decreased steadily from 1974 to 1990, with direct observations and modeling simulations showing that [mirex] and [PCB] in alewife have decreased substantially over the past 20+ yr (Madenjian et al. 1995; Makarewicz et al. 2003). Growth-related contaminant dilution in alewife would have contributed to the reduction of contaminants in Lake Ontario coho and chinook salmon for a period immediately following the 1976–1977 winterkill, with the relative influence of enhanced salmonine stocking increasing after the recovery of the alewife population in the early 1980s.

As discussed, there is circumstantial evidence that reductions of [POPs] and [Tot-Hg] in Lake Ontario coho and chinook salmon were accelerated by the 1976–1977 alewife winterkill and by the gradual decline of large alewife from 1983 onward (Fig. 3; Table 2). Following the abundance peak of 1983 (Fig. 4a), the alewife population decline was triggered by increased salmonine stocking rates (Jones et al. 1993; O'Gorman et al. 1997; Madenjian et al. 2002). However, although stocking programs have had a major role in controlling the alewife population, alewife numbers continued to decline from 1992 onward even though stocking rates were reduced by an average of 3 million fish yr^{-1} during this period (Fig. 4a). Therefore, factors in addition to salmonine stocking must have contributed to the alewife decline. Canada–United States nutrient abatement programs reduced Tot-P loadings to Lake Ontario by about 15% between 1974 and 1991, with reduced loadings, in turn, resulting in lower spring overturn [Tot-P] (Fig. 4d). As suggested by the bottom-up trophic cascade depicted in Fig. 4, reduced nutrient availability at low trophic levels can at least partially explain the decline of alewife abundance over the record. Interestingly, while nutrient loading rates to Lake Ontario were periodically elevated in the mid-1980s and early 1990s, offshore spring-overturn [Tot-P] continued to decline (Fig. 4d). This breakdown of the loading-spring [Tot-P] relationship (Fig. 4d, *inset*) was presumably caused by a net transfer of phosphorus to inshore areas and benthic cycling by invasive zebra and quagga mussels (e.g., Hecky et al. 2004). We suggest that the overall downward trend of Lake Ontario alewife since the early 1980s has been driven by two major factors: (1) reduced offshore nutrient availability resulting from nutrient abatement programs and inshore, benthic cycling by invasive mussels and (2) salmonine stocking programs. The decline of large alewife has, in turn, reduced the rate at which contaminants are incorporated into coho and chinook salmon tissues via trophic transfer. It has been suggested that Lake Ontario coho and chinook salmon switched their feeding preferences to rainbow smelt (*Osmerus mordax*) and/or slimy sculpin (*Cottus cognatus*) during the period of alewife decline. However, because these species typically have higher contaminant burdens than alewife (Niimi 1996; Tomy et al. 2004), an increase in contaminant concentrations in coho and chinook salmon would have been expected if prey switching had occurred to a significant degree.

Our analysis shows that upward oscillations in [POPs] and [Tot-Hg] were often associated with cooling La Niña cycles (Fig. 5, vertical bars) and that the overall downward trajectories of contaminant concentrations were negatively correlated with summer air temperature (Fig. 6; Table 2). The ecological and physiological responses of alewife and coho and chinook salmon to rising temperatures might explain the negative correlation between contaminant concentrations and temperature. Survival rates of young-of-the-year (YOY) alewife increase with increasing ambient water temperatures (Kellogg 1982; Eck and Wells 1987; Casselman et al. 1999). Increased YOY survival during warm years would increase competition for resources within the cohort, thereby reducing the probability that individuals recruit to larger, more contaminated size classes. Such competition would have been intensified by Canada–United States nutrient abatement programs, the invasion of zebra and quagga mussels, and the ensuing bottom-up trophic cascade (Fig. 4). Supporting this, Rand et al. (1994) determined that the average weight of Lake Ontario alewife aged 2 yr and older decreased from 41 g in 1978 to 19 g in 1989. They also reported that the spring energy density of Lake Ontario alewife decreased from 6,259 J g^{-1} in 1979 to 4,600 J g^{-1} in 1990. Since the bioenergetic demands of salmon increase with increasing temperature over the range observed in Lake Ontario (Wurster et al. 2005), temperature increases over the study period would have increased the rate at which coho and chinook salmon feed on alewife. Declines in the nutritional quality (i.e., energy density) of alewife over the study period would have further increased food intake requirements. Supporting this position, outputs from bioenergetic models suggest that predation rates by Lake Ontario chinook salmon increased nearly three-fold from 1978 to 1990 (Rand et al. 1994), and stomach-content analyses indicate that the reliance of coho and chinook salmon on small, lower-quality alewife increased with time from 1983 to 1993 (Rand and Stewart 1998). Thus, it appears that stocking and nutrient abatement programs, zebra and quagga

mussel invasions, and warming temperatures have worked in concert to reduce the size and energy density of Lake Ontario alewife, with the relative significance of these factors being unknown. As suggested above, decreases in alewife size would have decreased the rate of contaminant transfer to coho and chinook salmon.

Warming rates over the study period (about 2.4°C in 27 years; Fig. 5) far exceeded average rates for the past century and those predicted for the long-term future. Trends in summer air temperature discussed in this article were based on averages of daily temperatures at Toronto (2,500,000 people) and Kingston (120,000 people); thus, it is conceivable that this period of rapid warming was more reflective of localized urban expansion than of regional climatic trends. Since the growth rate of Toronto was significantly greater than that of Kingston over the study period, we would have expected that warming rates at Toronto would have been greater than those at Kingston if trends were strongly influenced by urban heat. To test this possibility, we assessed summer air temperature trends independently for Toronto and Kingston. Average summer temperatures at Kingston were typically 0.5°C cooler than those at Toronto, but warming trends at the two locations were virtually identical. For another comparison, we downloaded daily temperature data for the Cornwall (46,000 people) weather station, which is located near the St. Lawrence River about 200 km northeast of Kingston. During the study period, average summer temperatures at Cornwall were about 1°C warmer than those at Toronto, with warming trends at Cornwall tracking those at Kingston and Toronto nearly perfectly. Therefore, we are confident that presented temperature trends (Fig. 5) are representative of regional trends over the study period. In support of this argument, an analysis of epilimnetic temperatures in the Bay of Quinte (Fig. 1) for the years 1950 to 2000 indicated that warming rates were above average from 1977 onward. Casselman (2002) referred to this period of rapid warming as a climatic "regime shift" and showed that it was attributable to an increasing frequency of strong El Niño cycles and their increasing influence on eastern Ontario. The period of rapid warming observed in eastern Ontario coincides with a continuing warming trend in the Arctic, which has been attributed to the cyclical Arctic Oscillation (Macdonald et al. 2005).

We demonstrated that [ΣPCB], [mirex], and [*p,p′*-DDT] in Lake Ontario coho and chinook salmon have decreased exponentially over the past 20+ years and that [Tot-Hg] have decreased slowly, if not negligibly. The high rate of [POPs] declines during the late 1970s and the early 1980s was attributed mostly to Canada–United States bans on usage and sedimentation. The dynamics of these contaminants in salmon from the early 1980s to 2003 were linked to the combined influences of salmonine stocking and nutrient abatement programs, climatic cycles, and alewife population dynamics. In view of climatic cycles, our analysis shows that contaminant concentrations in coho and chinook salmon were negatively correlated with summer temperatures. It is important to note, however, that the influences of temperature on contaminant transfer to coho and chinook salmon appear to be manifested in the negative effects that warming temperatures have on prey species, such as alewife, that have a preference for cool to cold waters. Given that sedimentation and water flushing rates in Lake Ontario are slow and that contaminants are stored within several environmental compartments, including biota, we predict that further reductions in fish tissue contaminants will continue to be slow.

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Received: 18 December 2005
Accepted: 29 June 2006
Amended: 10 July 2006