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Is mirex still a contaminant of concern for the North American Great Lakes?

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ABSTRACT

Mirex, historically used as a pesticide and fire retardant, was released to Lake Ontario during the 1960s. Even after 35–40 years of cessation of its production and bans on use during the 1970s, mirex is considered a contaminant of concern. In this study, we present a comprehensive view of long-term trends and significance of mirex/photomirex levels in fish from the Canadian waters of the Great Lakes. Majority of measurements (except for Lake Ontario) were below detection, especially in recent years. Concentrations of mirex in Lake Ontario fish decreased by approximately 90% between 1975–2010, and both mirex and photomirex decreased by 75% between 1993–2010. Half-lives of mirex and photomirex for the entire period ranged from 4–10 years, but were lower at 2.5–8 years in recent times indicating expedited recovery possibly in response to remedial actions performed in the 1990s. Simulated fish consumption advisories generated by considering only mirex and photomirex indicated that mirex/photomirex is a minor concern. We predict that within 15 years mirex/photomirex levels in Lake Ontario fish will drop to levels that will result in advisories of at least 8 meals/month. In either case, the presence of other contaminants in Lake Ontario fish contributes to more stringent advisory than generated by mirex/photomirex. It is recommended that the routine monitoring of mirex/photomirex be replaced with periodic surveillance to reduce analytical costs. Dechlorane family compounds (that mirex is a part of) need to be evaluated further for their monitoring needs once in-depth toxicological information becomes available.

Introduction

Mirex is a synthetic organochlorine compound that was used as a pesticide mainly in the southeastern U.S., but was also used as a flame retardant typically under the name Dechlorane (Faroon et al., 1995; Shen et al., 2010). Mirex is very resistant to both biological and chemical degradation, and as such, extremely stable in the environment (Faroon et al., 1995; WHO, 1984). Photodegradation of mirex primarily results in photomirex which is equally stable in the environment (WHO, 1984). Mirex is highly insoluble in water, generally accumulates in sediments and bioaccumulates/biomagnifies through food webs (WHO, 1984). Mirex is toxic for a variety of aquatic biota and is considered a potential human carcinogen (Apeti and Lauenstein, 2006; Faroon et al., 1995; WHO, 1984). Persistent, bioaccumulative, and toxic characteristics of mirex resulted in cessation of its production and a ban on its use in the late 1970s (Faroon et al., 1995; Shen et al., 2010). Mirex was also included in the so-called "dirty dozen" toxic persistent organic pollutants (POPs) that were subjected to global bans or phase out under the Stockholm Convention on POPs in April 2001 (Murphy et al., 2012).

Mirex entered into the North American Great Lakes mainly through large-scale manufacturing related activities by the Hooker Chemical Co. in Niagara Falls, NY, and the Armstrong Cork Company near Fulton, NY (Comba et al., 1993; Velleux et al., 1995). The fugitive emissions of mirex from these companies starting in the late 1950s or early 1960s resulted in the contamination of Lake Ontario via Niagara River and Oswego River, which are the top two tributaries to Lake Ontario (Apeti and Lauenstein, 2006; Pickett and Dossett, 1979; Van Hobe Holdrinet et al., 1978). Elevated mirex levels were observed in the Niagara River and Lake Ontario sediments, and were first discovered in Lake Ontario fish and the food web in 1974 (Kaiser, 1974). In 1976, the Ontario Ministry of the Environment highlighted that virtually all types of fish in Lake Ontario were contaminated with mirex (Kaiser, 1978; Pickett and Dossett, 1979). Studies based on Lake Ontario fish and waterfowl consumption data for New York State and the Province of Ontario concluded that even a low amount of consumption during the 1980s and 1990s would result in significant body burden of organochlorine compounds including mirex (Bloom et al., 2005; Kearney et al., 1999). Although restrictions on consuming Lake Ontario fish due to elevated levels of contaminants including mirex have been issued since the 1970s (Hickey

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et al., 2006; Kaiser, 1978; OMOE, 2013), reports have indicated that human exposure to these contaminants is a concern (e.g., Cole et al., 2002; Fitzgerald et al., 2001; Madden and Makarewicz, 1996). A recent study reported that the past use of mirex may be affecting the health of the current generation of reproductive-age women (Upson et al., 2013). Initial and subsequent observations and modeling simulations indicated that it may take between 60–100 years, if not more, for Lake Ontario to recover from the mirex contamination once the discharges stop (Halfon, 1984; Lum et al., 1987). However, a revised estimate predicted a complete recovery of the Lake Ontario waters by 2010 (Flint and Stevens 1989 in Makarewicz et al., 2003). As such, it is important to examine the status of recent mirex levels in Great Lakes fish, consumption of which is likely the dominant pathway for human and wildlife exposure to mirex in the Great Lakes environment.

A number of studies have investigated mirex and/or photomirex levels in Great Lakes fish based on a limited number of indicator species, sampling locations, in many cases, over restricted time periods (e.g., Carlson et al., 2010; Chang et al., 2012; Dellinger et al., 2014; Makarewicz et al., 2003). In this study, we present a comprehensive view on long-term trends and recent levels of mirex and photomirex in Great Lakes fish using a consistent and robust dataset from the Fish Contaminant Monitoring Program of the Ontario Ministry of the Environment and Climate Change (OMOECC). We then examine the significance of the most recent available mirex/photomirex levels from the perspective of fish consumption by humans. Half-lives of declines in mirex/photomirex levels and a time period required for mirex to fall below the levels of concern in Great Lakes fish were estimated. Based on the findings of these analyses, we then evaluate the appropriateness of continued mirex/photomirex monitoring in light of increased pressure and demand on analytical resources to include newly identified contaminants of concern.

Materials and methods

Data source

Levels of legacy contaminants have been monitored in Great Lakes fish by various national/federal, provincial/state and tribal agencies albeit at varying intensity and frequency (e.g., Bhavsar et al., 2011; Carlson et al., 2010; Dellinger et al., 2014; Gewurtz et al., 2011a,b). For this study, we considered the measurements collected by OMOECC due to the following six major reasons (Bhavsar et al., 2011; OMOE, 2013): 1) fish contaminant levels have been monitored for >40 years and hence long-term information is available, 2) the majority of measurements are for skinless, boneless fillets (SBF) that people generally prefer to consume providing relevance to potential human exposures, 3) a variety of fish species as opposed to one, two or very few indicator species have been monitored, 4) samples were analyzed only at the OMOECC laboratories resulting in a consistent analytical dataset, 5) the monitored area of OMOECC (one agency) covers four out of the five North American Great Lakes, and 6) samples have been collected from a number of locations within each lake providing comprehensive spatial coverage. Because a variety of fish species were monitored from a number of open water regions in each of the four Great Lakes and connecting rivers, the results presented here may also be applicable to the U.S. waters of the Great Lakes, especially for large mobile fish.

Sample collection and analytical method

Fish samples were collected mainly in summer and fall using a variety of methods including gill netting and electrofishing. After collection, fish size (length and weight) was measured and when possible gender was recorded. The dorsal muscle was filleted and the skin was removed. Samples were stored at −20 °C until homogenization and chemical analysis at the OMOECC laboratories. Samples were analyzed for a suite of organic contaminants including mirex and photomirex using the OMOECC method E3136 and adopting quality assurance protocols as described by Bhavsar et al. (2007).

Briefly, 5 g fish tissue homogenates were weighed into centrifuge tubes and fortified with surrogates decachlorobiphenyl (250 ng) and 1,3,5-tribromobenzene (100 ng). Next, 18 mL of concentrated, reagent grade hydrochloric acid, and 20 mL aliquot 25% (v/v) dichloromethane (DCM) in hexane were added to each sample and then left overnight to digest. To extract organic compounds, the acid digested samples were mixed on a bench top rotator for 45 min and then centrifuged at 2500 rpm for 5 min. The upper organic layer was quantitatively transferred to 100 mL volumetric flasks. The extraction of the digested material was repeated and final volume was made up to 100 mL with DCM/hexane. Twenty milliliter sub-samples of the extracts, with 2 mL of iso-octane, were evaporated to 1 mL, added to dry packed Florisil® columns and allowed to drain to the top of the packing. Pure hexane was then added in 1 mL portions until the columns were completely wet. Depending on Florisil activity, approximately 20 mL of hexane was used to elute polychlorinated biphenyls (PCBs), chlorobenzenes (CBs), mirex, photomirex, aldrin, heptachlor, and p,p′-DDE in a 40 mL tubes as Fraction 1. The remaining compounds (mostly organochlorine pesticides) were then eluted with a 25% (v/v) DCM/hexane and collected in 40 mL tubes as Fraction 2. Pure iso-octane (1 mL) was added to the Fractions and the sample extracts were evaporated to 1 mL final volumes. Gas–liquid chromatography was used to determine total-PCB, CB, and the organochlorine pesticides mirex, photomirex, aldrin, heptachlor, and p,p′-DDE using a gas chromatograph and Ni63 electron capture detector (ECD). For Fraction 1 analysis a J&W DB-17 15 m × 0.53 mm × 1.0 μm column was used. The column head pressure was 4 PSI and the temperature program was 80 °C for 1 min; 80 to 180 °C at 10 °C/min; 180 to 260 °C at 5 °C/min; 260 °C for 9.5 min. The method detection limits (MDL) were 20, 5, 4, 1, 1 and 1 ng/g for total-PCB, mirex, photomirex, aldrin, heptachlor, and p,p′-DDE, respectively. A blank and spiked blank matrix sample was processed with each set of samples (20 to 30). The method performance is periodically monitored through laboratory studies such as the Northern Contaminants Program (NCP) and Quality Assurance of Information for Marine Environmental Monitoring in Europe (QUASIMEME).

Data screening for trend analyses

The OMOECC database included a total 46,013 records of mirex and 18,725 records of photomirex (Table 1) concentrations in 57 forage/young-of-the-year/sport fish species sampled at a varying frequency between 1975 and 2011 from approximately 450 different locations in the Canadian waters of the Great Lakes. Monitoring of photomirex began in 1994. We classified the sampling locations into 60 regional blocks used by OMOECC for the purpose of issuing fish consumption advisories (Fig. S1), as well as into lakes and connecting channels. Samples collected from Lakes Superior, Huron and Erie and connecting water channels as well as from the Niagara River upstream of Niagara Falls (i.e., Niagara River — Upper) were mostly (>95%) below detection (Table 1). This is in agreement with reports that historical point discharges in the lower Niagara River were the main source of mirex in the Great Lakes (Kaiser, 1978; Pickett and Dossett, 1979; Van Hobe Holdrinet et al., 1978). As a result, mirex is generally found only in Lake Ontario fish while the majority of measurements for fish from other Great Lakes are below detection, especially in recent years (Carlson and Swackhamer, 2006; Dellinger et al., 2014; Makarewicz et al., 2003). Further, mirex/photomirex measured in fish from the St. Lawrence River is most likely a result of export from Lake Ontario (Comba et al., 1993; Lum et al., 1987; Makarewicz et al., 2003). As such, we focused our study on the lower Niagara River and Lake Ontario. Recent (2000–2011) concentrations of mirex and photomirex in all fish species sampled from various locations in the Niagara River, Lake Ontario and St. Lawrence River have been presented in Electronic Supplementary Material (ESM; Table S1a and b).

Table 1
Summary of mirex and photomirex measurements conducted and detected by the Ontario Ministry of the Environment and Climate Change.

Location	Mirex				Photomirex			
	Period	Measurements	Detected	% Detected	Period	Measurements	Detected	% Detected
Lake Superior	1976–2011	5930	114	2%	1994–2011	2989	14	0%
St. Marys River	1978–2009	235	0	0%	1995–2009	118	0	0%
Lake Huron — North Channel	1976–2010	968	10	1%	1993–2010	558	0	0%
Lake Huron — Georgian Bay	1976–2010	2973	37	1%	1993–2010	1234	13	1%
Lake Huron	1975–2009	4642	103	2%	1993–2009	1442	2	0%
St. Clair River	1977–2010	815	25	3%	1994–2010	546	3	1%
Lake St. Clair	1976–2010	4117	25	1%	1994–2010	828	1	0%
Detroit River	1978–2010	1251	11	1%	1994–2010	665	0	0%
Lake Erie	1977–2010	7720	130	2%	1994–2010	3587	12	0%
Niagara River — Upper	1980–2009	837	31	4%	1994–2009	602	0	0%
Niagara River — Lower	1978–2009	1210	568	47%	1994–2009	687	107	16%
Lake Ontario	1975–2010	11,506	7883	69%	1994–2010	4671	2169	46%
St. Lawrence River	1977–2010	3809	897	24%	1994–2010	798	32	4%
Total		46,013	9834	21%		18,725	2353	13%

Based on recorded mirex measurements for 38 fish species from the Niagara River, Lake Ontario and St. Lawrence River, five species were selected for detailed temporal and spatial trend analysis (ESM Table S2). These species included brown trout (*Salmo trutta*, BT), Chinook salmon (*Oncorhynchus tshawytscha*, CHK), coho salmon (*Oncorhynchus kisutch*, COS), lake trout (*Salvelinus namaycush*, LT) and rainbow trout (*Oncorhynchus mykiss*, RT), and were selected based on their high mirex detection rate (88% for BT, >95% for other species), greater monitoring frequency (time points of 24 for LT, and >30 for other species when all locations combined), and overall wide geographic coverage (8–10 locations for each species in total) (ESM Table S2). For the trend analyses, all non-detect values, which were relatively low (1–5% when locations and years combined by species), were excluded to remove their influence.

Fish contaminant levels can be influenced by fish length (e.g., Gewurtz et al., 2011a,b). Although such an influence is very strong for mercury, fish length has a relatively weak relationship with organic chemicals like PCBs (e.g., Gewurtz et al., 2011a,b). As such, utilization of measurements for a species-specific restricted size range was considered appropriate for this study. The selected size ranges were 70–90 cm for CHS, 60–80 cm for COS, 45–65 cm for BT, 50–70 cm for LT, and 55–75 cm for RT, and were based on previous studies, histograms and boxplots of fish lengths (e.g., Bhavsar et al. 2010, ESM Figs. S2, S3). Further, only those locations/years were considered that yielded at least 5 measurements for the selected size range of each species.

Statistical procedures

Trend analyses

Mean annual mirex and photomirex concentrations were calculated for each species and Lake Ontario block. Temporal trend analysis was conducted using the non-parametric Mann–Kendall (M–K) tests on mean annual concentrations. The M–K test allows identification of the presence of a monotonic increasing or decreasing trends. The slope of the linear trend was then estimated using the non-parametric Sen's slope estimate (Salmi et al., 2002). Percent decrease in concentrations over the monitoring time period was calculated for each species and location.

Half-life ($t_{1/2}$, in years) of mirex and photomirex in each species and location was calculated using the following rate equations:

$$t_{1/2} = \ln(2)/k$$

where k is the rate constant calculated as

$$-k = \ln(C/C_o)/t$$

where C is the final concentration, C_o is the initial concentration for the first sampling event, and t is time between the first and most recent sampling events (in years).

The OMOECC database also included concurrent measurements of fish lipid content for the same samples that were analyzed for mirex/photomirex content. Because lipid can be a major phase for partitioning of mirex/photomirex (Faroon et al., 1995) and changes in fish lipid content over time may have influenced temporal trends of mirex/photomirex, another set of M–K tests were conducted on lipid normalized mirex/photomirex concentrations. Statistical significance was considered at $p < 0.05$.

Fish consumption advisory analyses

One of the major concerns for most legacy contaminants in the Great Lakes environment is risk to human consumers of Great Lakes fish. Fish consumption advisories aimed at guiding people on safe consumption of fish have been issued for the Great Lakes by both Canadian and U.S. agencies (OMOE, 2013; USEPA, 2013). Because OMOECC is the only agency issuing fish consumption advisories for the most part of the Canadian waters of the Great Lakes, the advisories have been consistent from the analytical measurements and advisory benchmarks perspective. The OMOECC advisory benchmarks are generally based on the tolerable daily intakes from the Food Directorate of Health Canada, and have been summarized by Bhavsar et al. (2011). The advisory benchmarks for mirex and photomirex are presented in Table 2. The OMOECC calculates advisories using all available contaminant measurements and issues advisories based on the most restrictive contaminant (Bhavsar et al., 2011). At present, the most restrictive advisories posted are largely due to elevated levels of PCBs, mercury and dioxins/furans (Bhavsar et al., 2011; OMOE, 2013).

Next, we simulated fish consumption advisories solely for mirex and photomirex irrespective of the presence of other contaminants in order to understand their health risk to human consumers of Great Lakes fish. All sport fish mirex/photomirex data for the Canadian waters of the Great Lakes were considered in this analysis. We employed the OMOECC method for this simulation in which a power series regression of fish length vs mirex/photomirex concentration was conducted for each species/location/year combination. Standardized length mirex

Table 2
Fish consumption advisory benchmarks (ng/g wet weight) for mirex and photomirex used by the Ontario Ministry of the Environment and Climate Change for the general (GP) and sensitive (SP) populations for the advisories published in the 2013–2014 Guide to Eating Ontario Sport Fish (OMOE, 2013).

Meals/month	Mirex		Photomirex	
	GP	SP	GP	SP
8	0–82	0–82	0–15	0–15
4	82–164	82–164	15–31	15–31
2	164–329		31–61	
1	329–657		61–122	
0	>657	>164	>122	>31

and photomirex concentrations were calculated from the regressions for every 5-cm size interval. These values were then compared with the OMOECC advisory benchmarks (Table 2), and advisories (in meals/month) were calculated for each 5-cm size interval for the sampled size ranges (Bhavsar et al., 2011). A statistical comparison of simulated advisories with previously published real advisories and the mercury-only and toxaphene-only scenarios was conducted (Bhavsar et al., 2011; Gandhi et al., 2014a). For the comparison purpose, each individual advisory value was categorized into complete restriction (0 meal/month or do not eat), partial restriction (1, 2 and 4 meals/month), and no restriction (i.e., unrestricted = 8 meals/month). The results are presented on a lake-wide basis, as well as advisory block and species basis.

Results

Temporal and spatial trends

During the 1970s and 1980s, mean mirex concentrations typically ranged from 100 to 300 ng/g wet weight (ww) in the restricted size classes of the five indicator fish species (Fig. 1). The concentrations declined at all monitored Lake Ontario blocks for all species with a reasonable availability of long-term (10+ year) datasets (Fig. 1). The mean mirex concentrations in these fish species were <50 ng/g ww during the recent years (i.e., 2000s) (Fig. 1). Overall, mirex concentrations in Lake Ontario fish decreased by about 90% between the mid 1970s and late 2000s. Half-lives of mirex generally ranged between 5–10 years. Marginal spatial differences were evident in the levels of mirex in the indicator fish species, although the detection of trends in space was limited by the available datasets (Fig. 1).

Mean photomirex concentrations in restricted size classes of the five indicator fish species were about 3× lower than the corresponding mirex concentrations, and typically ranged from 20–60 ng/g ww during the 1990s (Figs. 1, 2). Similar to mirex, the photomirex concentrations also declined at all monitored Lake Ontario blocks for all species with a reasonable availability of long-term (10+ year) datasets (Fig. 2). The mean mirex concentrations in these fish during the 2000s were <20 ng/g ww (Fig. 2). Overall, photomirex concentrations in Lake Ontario fish decreased by about 75% between the mid 1990s and late 2000s. Half-lives of photomirex typically ranged between 4–9 years.

The lipid based mirex concentrations differed substantially among the indicator species (ESM Fig. S4). The concentrations were generally greater in salmon species (CHS, COS) and varied dramatically from 4000–20,000 ng/g lipid weight (lw) during the 1970s and 1980s (Fig. S4). In contrast, the concentrations typically ranged from 1500–5000 ng/g lw for the three trout species (BT, LT, RT) (Fig. S4). The species-specific differences for lipid normalized photomirex were similar to those for mirex albeit the levels and differences were lower (ESM Figs. S4, S5). The lipid normalized photomirex concentrations in salmon species (CHS, COS) typically ranged from 1000–6000 ng/g lw during the 1990s (Fig. S4). The corresponding concentrations in the trout species (BT, LT, RT) were lower at about 300–1000 ng/g lw (Fig. S4). Temporal trends of lipid normalized mirex and photomirex concentrations were similar to those from the wet weight based measurements (Figs. 1, 2; ESM Figs. S4, S5). The recent lipid based measurements for the salmon and trout species are generally <3000 and <500 ng/g lw for mirex, respectively, and <1000 and <200 ng/g lw for photomirex, respectively (ESM Figs. S4, S5).

Because a minimal impact of fish lipid content on the mirex and photomirex trends was evident, next we investigated a hypothesis of no major change in fish lipid contents by examining their temporal trends for the limited size classes of the five indicator species selected in this study. As expected, most of the temporal trends for the fish lipid contents were either flat or weakly increasing/decreasing with annual variability in the mean values (ESM Fig. S6). Coho salmon from Lake Ontario block 5 showed a statistically significant decline, which is in agreement with previous reports (Neff et al., 2012).

Advisory simulations

In total, 37 sport fish species from the four Great Lakes were included in this simulation with variable presence at the 59 advisory blocks — see Bhavsar et al. (2011) for more details. Overall, only 1% and 3% of the simulated advisories were restrictive for mirex-only and photomirex-

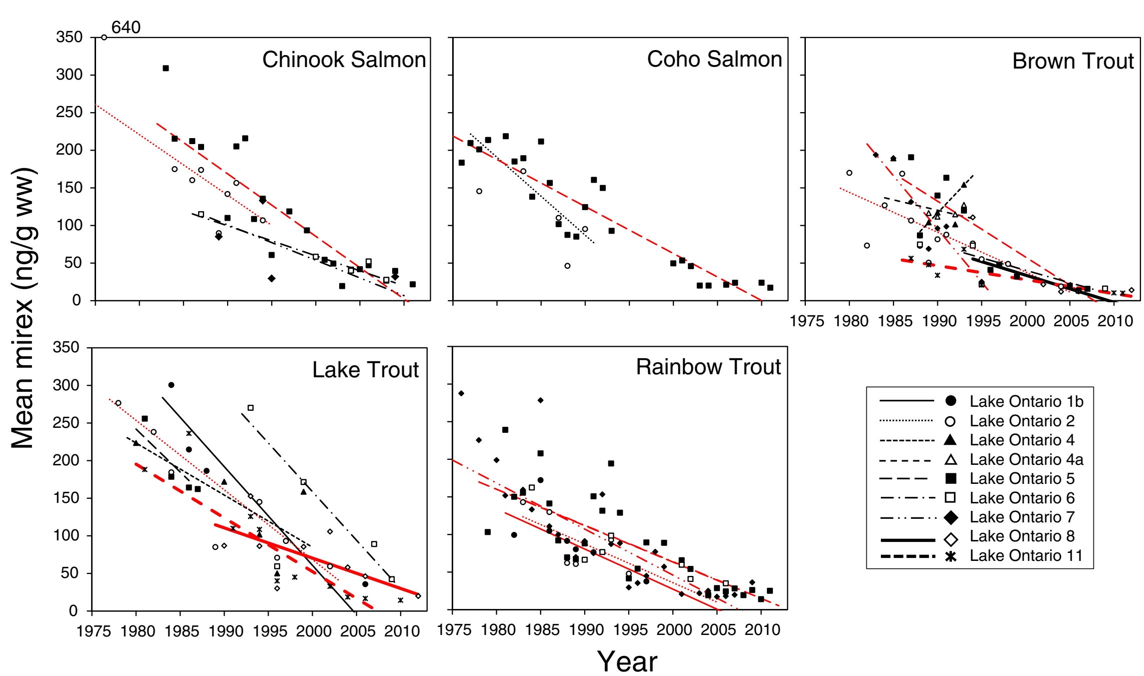

Fig. 1. Mann–Kendall Sen's estimate for mean mirex concentrations (ng/g wet weight) for Chinook salmon, coho salmon, brown trout, lake trout and rainbow trout from various sampling blocks of Lake Ontario. Statistically significant ($p < 0.05$) Sen's estimate regressions are shown in red.

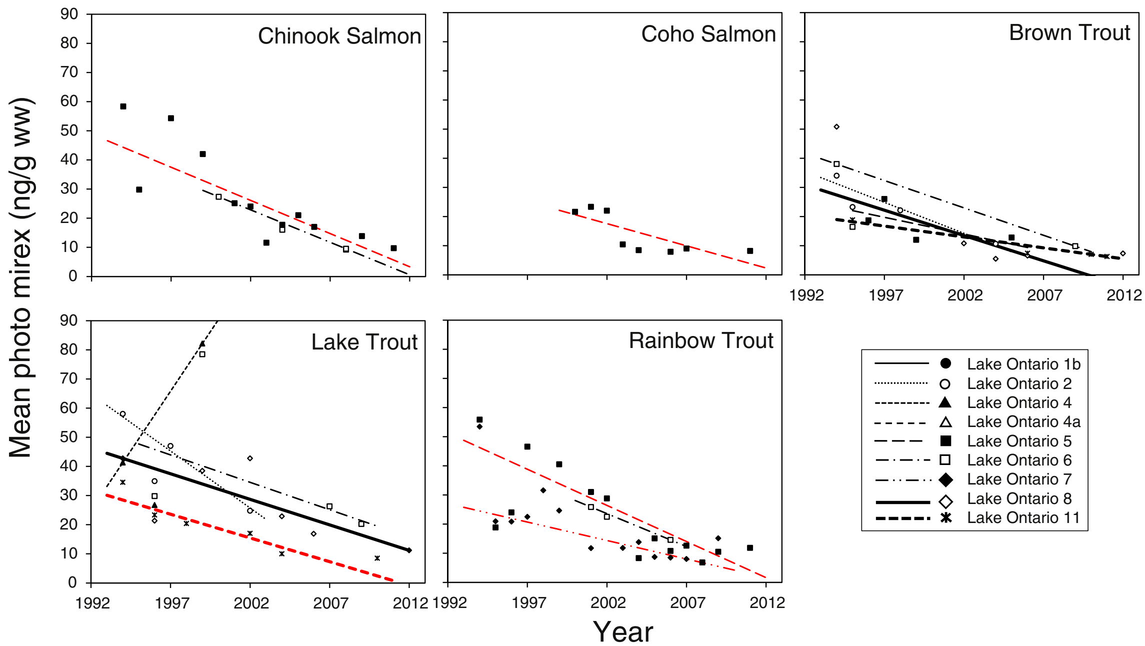

Fig. 2. Mann–Kendall Sen's estimate for mean photomirex concentrations (ng/g wet weight) for Chinook salmon, coho salmon, brown trout, lake trout and rainbow trout from various sampling blocks of Lake Ontario. Statistically significant ($p < 0.05$) Sen's estimate regressions are shown in red.

only scenarios, respectively, when all species- and block/lake-specific values were pooled (results not shown). A lake-wide comparison of real and simulated advisories for both the general and sensitive human populations highlighted that there would be no restrictive advisories under the mirex/photomirex-only scenario for Lakes Superior, Huron and Erie (Fig. 3). For Lake Ontario, about 11% of the mirex/photomirex-only advisories were restrictive, almost all of which were partially restrictive for the general population while about half of those were completely restrictive (i.e., do not eat) for the sensitive population (Fig. 3). The corresponding value for St. Lawrence River was about 3% (Fig. 3).

A species-specific breakdown of the mirex/photomirex-only simulated advisories highlighted that about half of the species present in Lake Ontario and most of the species present in the St. Lawrence River would not have any restrictive advisory under this scenario (ESM Fig. S7). The species with at least 15% of the advisories being restrictive under this scenario would be alewife, American eel, channel catfish, Chinook salmon, coho salmon, lake trout and rainbow smelt (Fig. S7). When examined more closely, it was found that the higher percentage of advisories being restrictive for these species was mostly a result of either low number of advisories due to lack of data (rainbow smelt) and/or use of old measurements for certain size categories and/or locations (all above mentioned species) (results not shown).

A Lake Ontario evaluation showed that all locations would have <15% of mirex-only advisories being restrictive (ESM Fig. S8). In contrast, the photomirex-only scenario resulted in three blocks (Lake Ontario block 2 (LO2), block 4 (LO4) and block 6 (LO6)) with >20% of advisories being restrictive. This higher percentage is due to these blocks being located in open water where the sampling of top predatory fatty fish (e.g. salmon, trout) occurs and these species have a greater percent of restrictive advisories.

When compared to provincially issued advisories, the mercury-only scenario and toxaphene-only scenario, the mirex/photomirex-only advisories are generally much less restrictive (Figs. 3, 4). This is especially evident for Lakes Superior, Huron and Erie, where at present about 52%, 54% and 64% of the advisories are restrictive due to presence of other contaminants, respectively, but there would be no restriction on fish consumption under the mirex/photomirex-only scenario (Figs. 3, 4).

For Lake Ontario, currently there is no restrictive advisory due to mirex/photomirex because more restrictive advisories are in effect due to elevated PCB/dioxins/mercury levels (Fig. 4). However, under the mirex/photomirex-only scenario, there would be about 11% of the advisories being restrictive (Figs. 3, 4). From the perspective of risk to human health through fish consumption, the simulated advisories highlight that mirex/photomirex are more of a concern than toxaphene in Lake Ontario and vice versa for Lakes Superior and Huron (Fig. 4). Both mirex/photomirex and toxaphene are not of a concern for Lake Erie (Fig. 4). These results are in agreement with previous reports (e.g., Carlson and Swackhamer, 2006).

Discussion

Source and discovery

Mirex entered the Great Lakes mainly into Lake Ontario predominantly through industrial discharges during the 1960s (Kaiser, 1978; Pickett and Dossett, 1979; Van Hobe Holdrinet et al., 1978). Subsequently, elevated levels of mirex were observed in various components of the receiving ecosystem (Kaiser, 1978). However, mirex did not become a contaminant of international concern until reports on mirex contamination of seals from Europe and fishes from Lake Ontario were published (Kaiser, 1978). This is likely because fish consumption is generally the major route of human exposure to POPs under most circumstances. This indicates that significance of mirex in Great Lakes fish, especially top-predatory game fish, can be used as a surrogate for overall societal significance of the mirex contamination in the Great Lakes.

Observed declines in mirex/photomirex

In this study, mirex/photomirex concentrations in Great Lakes fish from locations other than the Niagara River – Lake Ontario – St. Lawrence River were only occasionally measured above the detection limits. However, such occasional detections are in agreement with mirex detected in mussels from selected locations throughout the Great Lakes (Apeti and Lauenstein, 2006). Historical use of mirex as a pesticide and associated non-point discharges have been considered potential

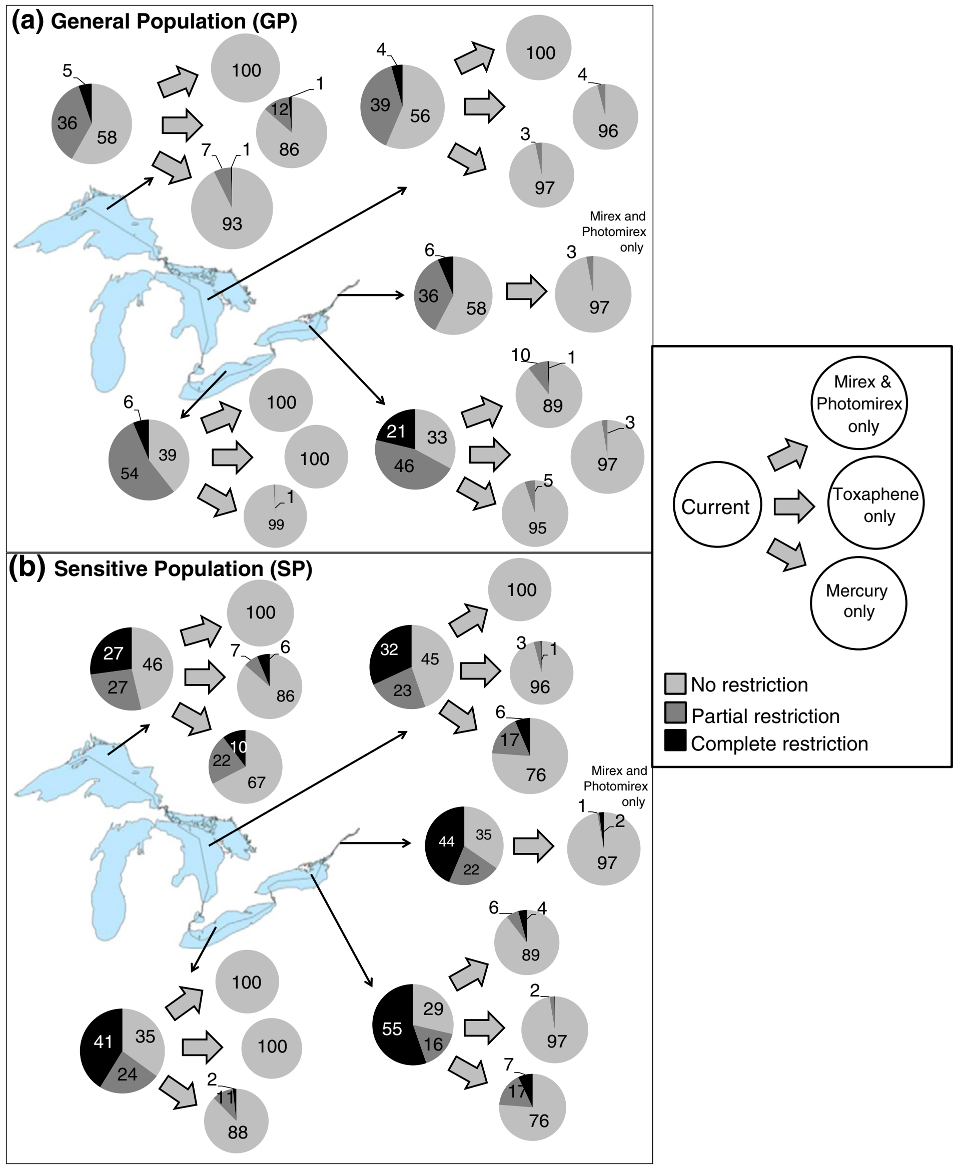

Fig. 3. A comparison of lake-wide current, mirex and photomirex only, mercury-only, and toxaphene-only fish consumption advisories for the general (a) and sensitive (b) populations for Lake Superior, Huron (including North Channel and Georgian Bay), Erie (without the St. Clair River – Lake St. Clair – Detroit River corridor) and Ontario (including Niagara River but excluding the St. Lawrence River). The values are rounded. Results for the mercury-only and toxaphene-only scenarios were taken from Bhavsar et al. (2011) and Gandhi et al. (2014a), respectively.

sources of mirex in the Great Lakes other than Lake Ontario (Apeti and Lauenstein, 2006). Mirex detected in fish collected from Bay of Quinte, Lake Ontario, is considered to be derived from historical contamination in the main basins of Lake Ontario (Gandhi et al., 2014b; Kaiser, 1978; Ridal et al., 2012).

A continued decline in mirex and photomirex was observed in all five fish species studied (Figs. 1 and 2). Lake trout mirex measurements collected by the U.S. Great Lakes Fish Monitoring and Surveillance Program were also consistently above the detection limits only in Lake Ontario fish, and also showed continued decreases between the late 1970s and late 2000s (Carlson et al., 2010; Chang et al., 2012). Similarly, reductions in mirex levels in a variety of fish species including salmonids have been reported (Dellinger et al., 2014; French et al., 2006; Hickey et al., 2006; Makarewicz et al., 2003; Suns et al., 1983). Recently, mirex concentrations in six fish species and invasive, omnivorous rudd (*Scardinius erythrophthalmus*) collected between 2010–2012 from the Buffalo and upper Niagara Rivers, both Great Lakes Areas of Concerns, were below detection (Kapuscinski et al., 2014).

These decreasing mirex concentration trends observed for fish are in agreement with reports on other parts of the Lake Ontario ecosystem. Decreasing mirex levels have been reported for various media of Lake Ontario such as waters (Marvin et al., 2004), sediments (Shen et al., 2011; Yang et al., 2011), mussels (Apeti and Lauenstein, 2006), and herring gull eggs (Norstrom and Hebert, 2006; Pekarik and Weseloh, 1998).

Mechanisms of declines

A number of studies have explored loadings, dynamics and export/removal of mirex from Lake Ontario. Mirex from the Niagara and Oswego Rivers entered into Lake Ontario and a large portion deposited along the southern shore (Pickett and Dossett, 1979; Van Hobe Holdrinet et al., 1978). The hydrological circulation processes distributed the contaminated sediments to other parts of Lake Ontario and increased the area covered by contaminated sediments (Scrudato and Delprete, 1982). Fate and transport simulations have suggested that major loss mechanisms for mirex in Lake Ontario are burial of contaminated

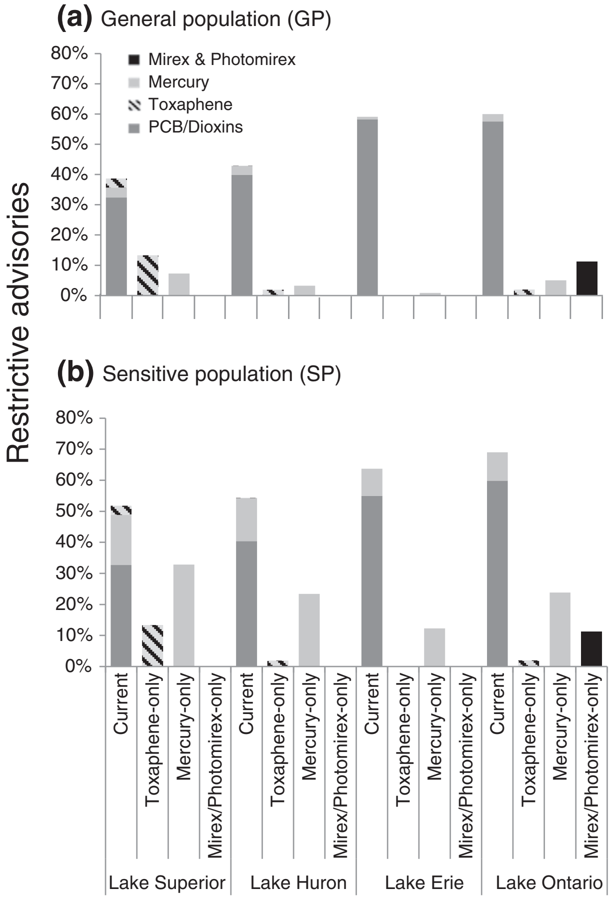

Fig. 4. Percentage of restrictive (partial + complete) fish consumption advisories for the general (a) and sensitive (b) populations with a breakdown of the contaminants causing the restrictions under the current conditions, mirex and photomirex only, toxaphene-only (tox-only), and mercury-only (Hg-only) scenarios. Results for the mercury-only and toxaphene-only scenarios were taken from Bhavsar et al. (2011) and Gandhi et al. (2014a), respectively.

sediments, especially along the southern shoreline, and export via the St. Lawrence River (Comba et al., 1993; Lum et al., 1987). These removal mechanisms would result in lower amounts of mirex in the Lake Ontario environment and thereby availability of mirex for bioaccumulation/biomagnification in fish.

A number of other factors could have contributed to the observed declines in mirex concentrations in Lake Ontario fish. For example, salmon prefer a diet of large alewives, which are generally more contaminated than smaller sizes of the same fish (French et al., 2006; Makarewicz et al., 2003). Consumption by salmon and other factors contributed to the lower abundance of these large alewives likely forcing salmon to rely on relatively less contaminated smaller alewives, and thereby reducing exposure and body burden of POPs including mirex in salmon in the long-term (French et al., 2006; Makarewicz et al., 2003). However, decreases in the size of prey items consumed would also result in an increase in food consumption rates by fish and commensurate increases in foraging costs that would likely compensate or potentially increase actual exposures by fish despite a drop in prey contaminant concentrations. The latter was observed to be the case for PCBs in Lake Ontario lake trout (Paterson et al., 2005, 2009).

Photodegradation of mirex to photomirex would result in lower levels of mirex in the Lake Ontario abiotic and biotic environments; however, this would result in relatively greater abundance of photomirex compared to mirex. Relatively constant ratios of photomirex to mirex and declining trends for both of these compounds observed in this and other studies (Makarewicz et al., 2003; Mudambi et al., 1992) suggest that the major losses of both of these compounds occur by other mechanisms apart from photodegradation. Loss of mirex and photomirex via volatilization has been considered to be minimal due to low volatility from water (Makarewicz et al., 2003). Similarly, loss through migration of fish into stream habitats and fish harvesting has been argued to be minimal (Makarewicz et al., 2003).

The control and removal of contaminated groundwater at the major source site of the former Hooker Chemical Co., Niagara Falls, NY, during the 1990s dramatically reduced concentrations of mirex in Niagara River suspended sediments and is also believed to have aided the recovery of the mirex contamination in Lake Ontario (Makarewicz et al., 2003; Shen et al., 2011). Similarly, control of point source discharges and remediation of contaminated sediments in the Oswego River Area of Concern (AOC) during the 1990s (USEPA, 2014), which resulted in the delisting of the AOC, would have contributed to the improvements in mirex loads to Lake Ontario.

Time period of recovery

Initially it was estimated that it would take between 200–600 years for mirex contaminated sediments in Lake Ontario to be completely buried by clean sediments after major loads of mirex to Lake Ontario were eliminated (Halfon, 1984). This analysis considered burial of the contaminated sediment as the predominant loss mechanism; however, it was shown that export by suspended particulate material in the outflow via the St. Lawrence River (and to certain extent migrating eels) can remove substantial amount of mirex from Lake Ontario (Halfon, 1984; Lum et al., 1987). A revised estimate suggested that it would take about 60–100 years for Lake Ontario to recover from the historical mirex contamination (Lum et al., 1987). These estimates did not account for loss of mirex through other mechanisms such as photodegradation, harvest of fish and turnover of fish populations through reproduction and growth (Paterson et al., 2005, 2009). A revised calculation by Flint and Stevens (in Makarewicz et al., 2003) indicated elimination of mirex from the Lake Ontario waters by 2010 which was much faster than the earlier estimates and also inconsistent with the presence of mirex reported in this study but suggestive of a timeline more representative than the earlier estimates.

Trend analyses presented for Lake Ontario fish in this study indicate that the mirex levels have declined by about 90% between the 1970s and 2010, and both mirex and photomirex concentrations have declined by about 75% between the mid 1990s and 2010. The half-lives typically range from 4–10 years, but were lower by 1–2 years and ranged from 2.5–8 years in the recent times indicating expedited recovery possibly in response to remediation at the contaminated sites in the Niagara and Oswego Rivers during the 1990s (Makarewicz et al., 2003). These findings are in agreement with other estimates (Carlson et al., 2010; Chang et al., 2012). Considering the decreasing mirex/photomirex trends, half-lives of the declines, fish consumption advisory benchmarks used by OMOECC (Table 2) and that mirex- and photomirex-only scenarios resulted in mostly partially restrictive advisories solely for certain species with many old measurements (Figure S7), we predict that Lake Ontario fish mirex and photomirex levels will almost completely drop below the current advisory benchmarks to restrict fish consumption from 4 to 8 meals/month (essentially non-restricted advisory) within 15 years (i.e., by 2030).

Monitoring needs

Mirex has been monitored in various matrices of the Great Lakes, especially in fish from Lake Ontario, by various agencies since the mid 1970s. These monitoring efforts have helped in understanding dynamics of mirex in the Lake Ontario ecosystem and in documenting declines in mirex and photomirex levels throughout the ecosystem. The results presented in this study highlight that the current levels of mirex in Lake Ontario fish would cause relatively marginal restrictions on consuming fish, even if the currently more restrictive contaminants such as PCBs were to decline below their levels of concern (Figs. 3 and 4). Further, based on the current levels and trends, we can expect that some other contaminants such as PCBs and dioxins/furans will remain more of a concern than mirex/photomirex (e.g., Bhavsar et al., 2007, 2008) and will be monitored in Great Lakes fish until they are not among the major contaminants causing restrictive fish consumption advisories for the Great Lakes. As such, regular monitoring of mirex/photomirex can be replaced with periodic surveillance to confirm continued declines in their levels while achieving analytical savings by omission of these contaminants from the routine analytical suite of measurements.

The list of contaminants that need to be monitored in the Great Lakes has been growing since the contamination problem in the Great Lakes was discovered in the 1960s. This list is expected to grow further as new commercial persistent, bioaccumulative and toxic chemicals are identified (Howard and Muir, 2010, 2013). Contaminants of emerging concern continue to exert additional pressure on the current analytical resources available for monitoring of legacy contaminants that were identified as a concern and banned decades ago. Minimizing the ongoing measurement costs of legacy contaminants without compromising the environmental and human health would allow re-allocation of resources towards identifying risks from contaminants of emerging concern and not-yet-detected chemicals. Some of the compounds in the dechlorane family to which mirex (also known as Dechlorane) belongs could be candidates for such monitoring.

Dechlorane family

Dechlorane Plus (DP), and Dechloranes (Dec) 602, 603, and 604 in addition to mirex are dechlorane compounds that have been typically identified in the environmental samples to date (Sverko et al., 2011). These chemicals are similar to mirex in structure and fire retardant properties, were substitutes for mirex and are still in use (Clement et al., 2012; Shen et al., 2010). All of these chemicals were a part of chemical manufacturing at the Hooker Chemical Co. in Niagara Falls, NY, and could have been released into the Niagara River and Lake Ontario (Shen et al., 2010). Atmospheric deposition might have also contributed to the presence of DP in Lake Ontario and other Great Lakes; however, the manufacturing facility in Niagara Falls, NY may be a source of DP in air (Hoh et al., 2006).

As expected, Lake Ontario sediments have 1 to 2 orders of magnitude higher concentrations of mirex, DP, Dec602 and Dec604 than other Great Lakes (Yang et al., 2011). Among these chemicals, DP has been studied more extensively and detected in air, water, sediments, food webs including fish and herring gull eggs from the Great Lakes (Gauthier et al., 2007; Hoh et al., 2006; Qiu et al., 2007; Shen et al., 2010, 2014; Tomy et al., 2007; Venier et al., 2014). Lake Ontario sediment DP concentrations were greater than combined concentrations of the brominated flame retardants (Qiu et al., 2007), some of which have been considered contaminants of emerging concern. However, using the biota-sediment accumulation factors, it was shown that mirex and Dec602 have greater bioaccumulation potentials than DP, Dec604 and PBDEs (Shen et al., 2010, 2011). Further, a recent study identified tribromo-Dec604 analogue in lake trout and lake whitefish at concentrations approximately 50–200 times greater than that of Dec604 (Shen et al., 2014). Investigations of temporal trends of the dechlorane compounds in suspended sediments from the Niagara River, sediment cores from Lake Ontario, and lake trout from Lake Ontario presented a positive outlook with peak concentrations occurring before 2000 and declining trends in the recent years (Hoh et al., 2006; Shen et al., 2011; Yang et al., 2011). Although mirex concentrations in Lake Ontario lake trout are generally highest among the dechlorane family compounds studied to date, dechlorane compounds other than mirex need to be evaluated further for their significance and necessity for monitoring once in-depth toxicological information for those compounds becomes available (Shen et al., 2010, 2011; Sverko et al., 2011).

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Appendix A. Supplementary data

Supplementary data (2 tables, 8 figures) to this article can be found online at http://dx.doi.org/10.1016/j.jglr.2015.09.015.

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Electronic Supplemental Material

Is mirex still a contaminant of concern for the North American Great Lakes?

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Table S1a: Recent (2000-2011) concentrations (ng/g) of mirex in fish collected from various locations in the Niagara River, Lake Ontario and St. Lawrence River. The values are presented as minimum-average-maximum when measured above the detection limit of 5 ng/g. An empty cell indicates no availability of recent data.

	Lake Ontario 1a	Lake Ontario 1b	Lake Ontario 2	Lake Ontario 2a	Lake Ontario 3	Lake Ontario 4	Lake Ontario 5	Lake Ontario 6	Lake Ontario 6a	Lake Ontario 6b	Lake Ontario 7	Lake Ontario 8	Lake Ontario 9	Lake Ontario 9a	Lake Ontario 10	Lake Ontario 11	St. Lawrence River 12	St. Lawrence River 13	St. Lawrence River 14	St. Lawrence River 15	St. Lawrence River 16
Alewife			8-9-10		5-14-25			5-8-12								16-17-18					
American Eel		20-23-25			5-34-120											5-16-38					
Black Crappie													5		5						
Bluegill		5		5	5							5	5		5		5				
Bluntnose Minnow																				5	
Brown Bullhead	5	5-7-21			5-6-20			5	5-7-13	5-8-23		5-5-6	5-5-6	5-7-15	5-5-7	5	5	5	5		
Brown Trout			6-19-36				5-20-96	5-21-85			5-14-37	5-16-45				5-11-35					
Channel Catfish		5			6-45-180								5-25-100		5-51-530	25-101-290					
Chinook Salmon		16-44-66	18-46-73				5-39-120	5-56-140			18-30-44	5-10-53				5					
Coho Salmon							5-25-90	50-78-120													
Common Carp	5-5-13	5-11-80		5-6-13	5-6-13				5-23-69	5-29-140			5-21-74				5-14-89	5-16-36			
Common Shiner	5																				
Emerald Shiner	5	5	5-5-7			5		5			5	5				5		5	5		
Freshwater Drum	5	5-12-37		5-26-90	5-20-71					5-14-30		5-33-85			5-5-8	5-7-45					
Lake Trout		11-34-55	20-68-150			45-99-180		23-83-300				5-93-580				5-19-150					
Lake Whitefish												10-32-100	5-29-69			5-15-75					
Largemouth Bass	5	5			5-5-10					5-5-6			5		5		5			5	
Northern Pike	5	5-5-8							5	5			5		5	5	5	5	5	5	
Pumpkinseed													5		5	5	5			5	
Rainbow Smelt	5	5-7-10	9-10-11		5-11-15																
Rainbow Trout	5-6-9	5-17-38	5-13-35				5-25-85	5-34-110			5-17-66										
Redhorse Sucker	5-6-9	5-6-13																			
Rock Bass	5	5			5				5	5		5	5		5	5	5		5		
Round Goby			5					5								5					
Smallmouth Bass	5	5-8-23	5	5				5				5-9-20	5		5	5-9-55	5		5	5-5-10	
Spottail Shiner	5-5-8	5	5		5	5		5			5	5	5			5		5	5	5	
Walleye		5							5	5-6-13		5-22-130	5-11-35		5-5-9	5-18-230	5		5	5	5
White Perch	5		5		5-9-30								5		5	5					
White Sucker	5				5			5	5	5								5			
Yellow Perch	5	5			5							5	5		5	5	5		5	5	

Table S1b: Recent (2000-2011) concentrations (ng/g) of photomirex in fish collected from various locations in the Niagara River, Lake Ontario and St. Lawrence River. The values are presented as minimum-average-maximum when measured above the detection limit of 4 ng/g. An empty cell indicates no availability of recent data.

	Lake Ontario 1a	Lake Ontario 1b	Lake Ontario 2	Lake Ontario 2a	Lake Ontario 3	Lake Ontario 4	Lake Ontario 5	Lake Ontario 6	Lake Ontario 6a	Lake Ontario 6b	Lake Ontario 7	Lake Ontario 8	Lake Ontario 9	Lake Ontario 9a	Lake Ontario 10	Lake Ontario 11	St. Lawrence River 12	St. Lawrence River 13	St. Lawrence River 14	St. Lawrence River 15	St. Lawrence River 16
Alewife			4-6-7		4-8-12			4-5-6								8-9-10					
American Eel		8-8-8			4-16-52											4-9-19					
Black Crappie													4		4						
Bluegill		4		4	4							4	4		4		4				
Bluntnose Minnow																				4	
Brown Bullhead	4	4-4-8			4-4-8			4	4-4-6	4-5-11		4	4	4-4-7	4	4	4	4	4		
Brown Trout			4-9-16				4-9-33	4-9-34			4-6-17	4-8-20				4-6-16					
Channel Catfish		4			4-9-24								4-13-70		4-24-210	12-45-140					
Chinook Salmon		7-17-26	10-22-34				4-17-68	4-25-68			7-11-18	4-5-22				4					
Coho Salmon							4-10-48	24-39-60													
Common Carp	4	4-6-32		4	4-4-6				4-7-20	4-15-43			4-10-30				4-5-15	4-4-8			
Common Shiner	4																				
Emerald Shiner	4	4	4			4		4			4	4				4		4	4		
Freshwater Drum	4	4-6-16		4-8-20	4-9-34					4-6-12		4-15-32			4	4-4-12					
Lake Trout		4-13-27	8-29-64			20-39-68		10-26-73				4-38-230				4-9-57					
Lake Whitefish												4-12-39	4-16-38			4-6-24					
Largemouth Bass	4	4			4-4-5					4			4		4		4			4	
Northern Pike	4	4							4	4			4		4	4	4	4	4	4	
Pumpkinseed													4		4	4	4			4	
Rainbow Smelt	4	4-4-5	4		4-7-8																
Rainbow Trout	4	4-8-17	4-7-16				4-12-44	4-16-48			4-8-26										
Redhorse Sucker	4	4																			
Rock Bass	4	4			4				4	4		4	4		4	4	4		4		
Round Goby			4					4								4					
Smallmouth Bass	4	4-5-18	4	4				4				4-5-8	4		4	4-5-24	4		4	4-4-8	
Spottail Shiner	4	4	4		4	4		4			4	4	4			4		4	4	4	
Walleye		4							4	4		4-8-44	4-7-24		4	4-9-92	4		4	4	4
White Perch	4		4		4-5-8								4		4	4					
White Sucker	4				4			4	4	4								4			
Yellow Perch	4	4			4							4	4		4	4	4		4	4	

Table S2: Species-specific summary of % of mirex measurements detected and number of monitoring years and blocks (Niagara River, Lake Ontario, St. Lawrence River combined; as well as St. Lawrence River block separately). Species highlighted in bold were selected for detailed temporal and spatial trend analyses.

Species	% Detected	# of Years	# of Blocks
Alewife	84%	5	6
American Eel	74%	14	11
Black Crappie	26%	14	3
Bluegill	0%	8	0
Bluntnose Minnow	0%	1	0
Bowfin	100%	2	2
Brown Bullhead	25%	34	15
Brown Trout	**96%**	**32**	**10**
Channel Catfish	78%	17	8
Chinook Salmon	**96%**	**30**	**9**
Coho Salmon	**97%**	**32**	**8**
Common Carp	53%	30	13
Common Shiner	0%	3	0
Emerald Shiner	0%	8	1
Freshwater Drum	30%	17	9
Gizzard Shad	25%	4	2
Lake Trout	**98%**	**24**	**9**
Lake Whitefish	88%	7	3
Largemouth Bass	4%	20	4
Longnose Gar	100%	1	1
Muskellunge	50%	5	1
Northern Pike	22%	32	12
Pumpkinseed	1%	17	1
Rainbow Smelt	85%	17	9
Rainbow Trout	**88%**	**34**	**9**
Redhorse Sucker	31%	4	2
Rock Bass	13%	19	3
Round Goby	0%	1	0
Round Whitefish	50%	1	1
Smallmouth Bass	41%	33	13
Spottail Shiner	1%	11	1
Sturgeon	100%	1	1
Walleye	37%	32	12
White Bass	61%	10	5
White Crappie	9%	2	1
White Perch	43%	23	6
White Sucker	31%	24	11
Yellow Perch	10%	35	14

Figure S1: Map of the Canadian waters of the Great Lakes classified into the regions used for fish consumption advisory purposes by the OMOECC (Bhavsar et al., 2011; OMOE, 2009).

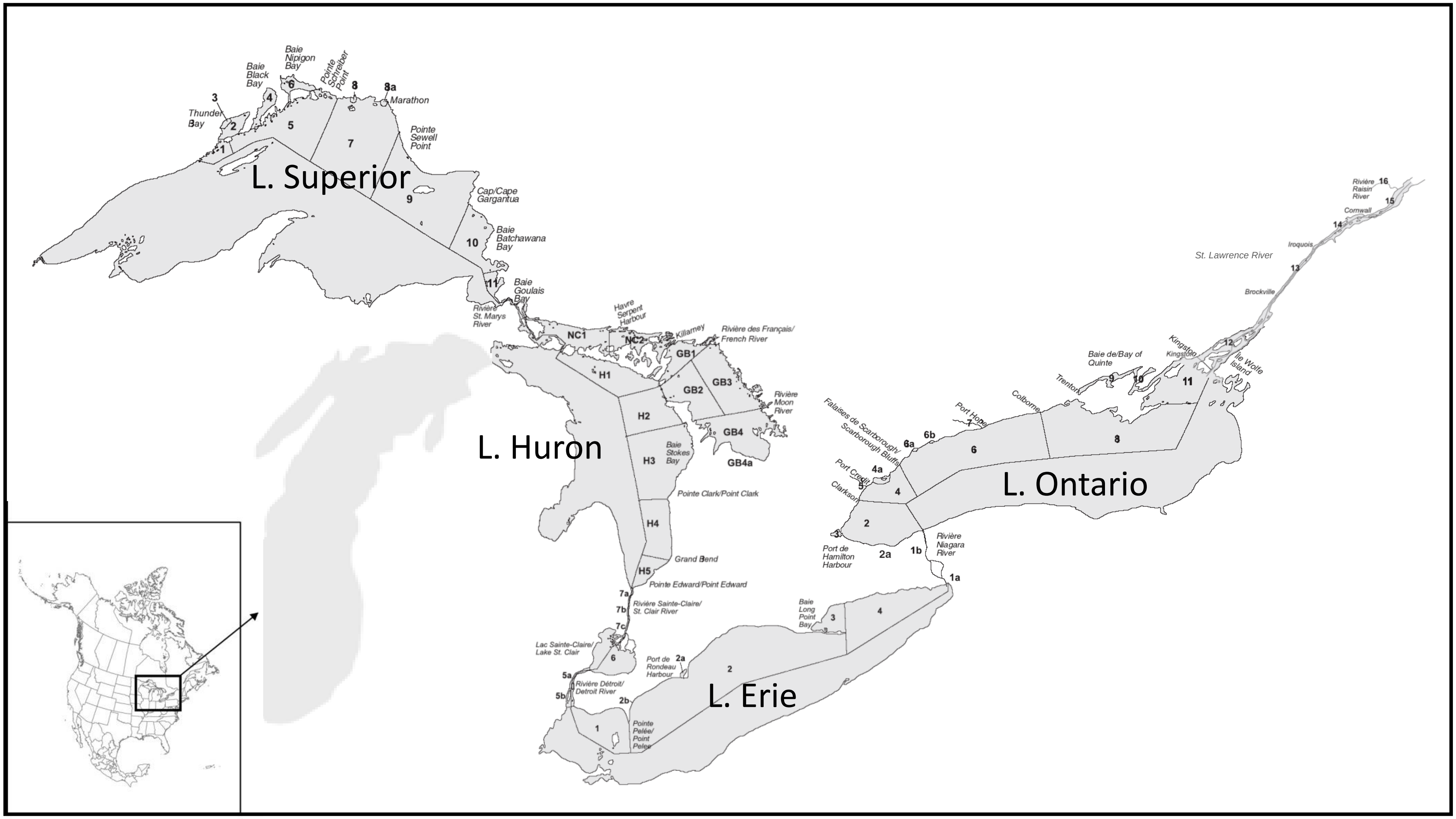


Figure S2: Histograms of length of fish samples collected. The yellow box shows the size range selected for detailed trend analyses.

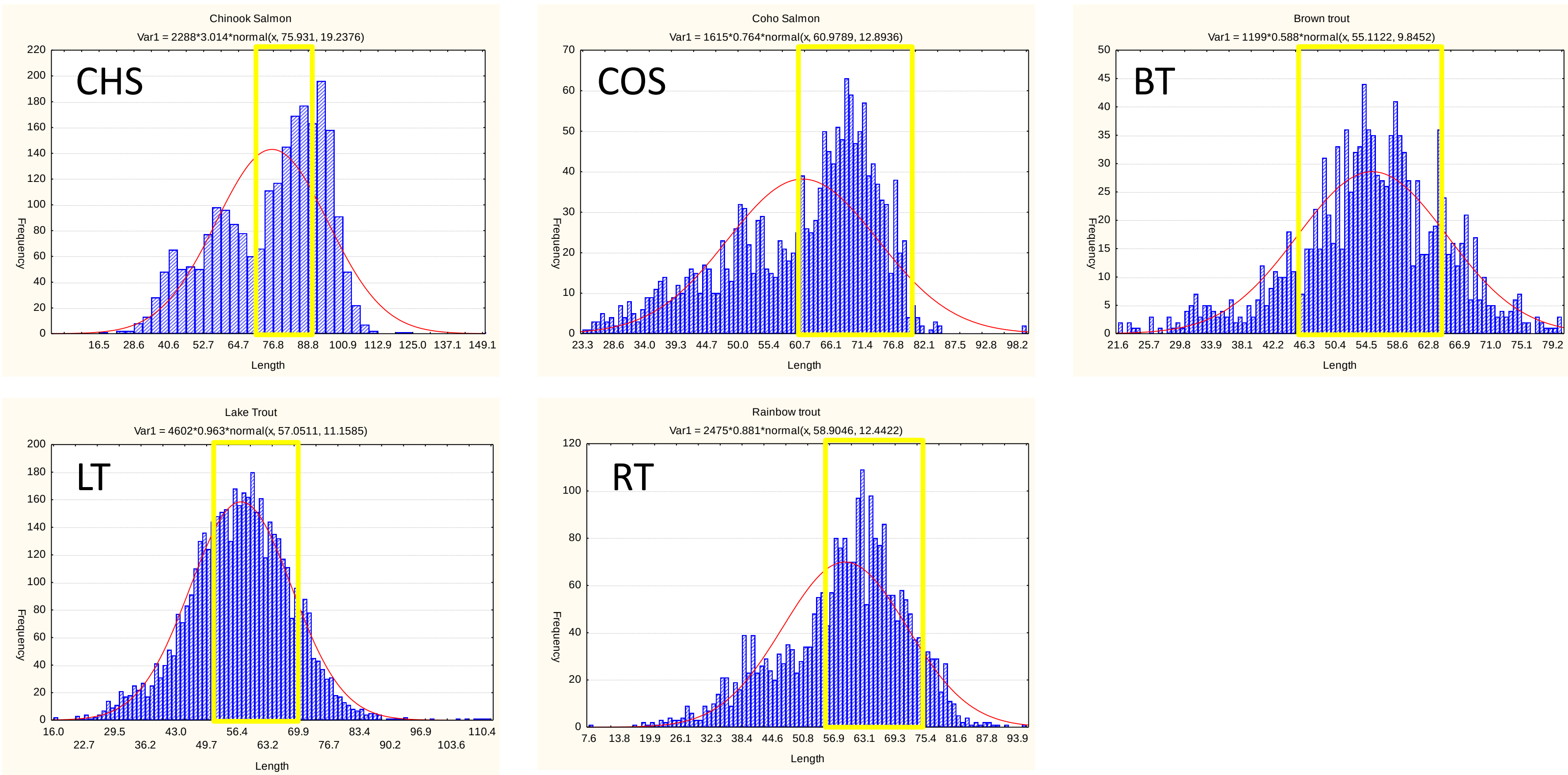


Figure S3: Boxplots of length of fish samples collected over the years from Niagara River, Lake Ontario and St. Lawrence River. The purple box shows the size range selected for detailed trend analyses.

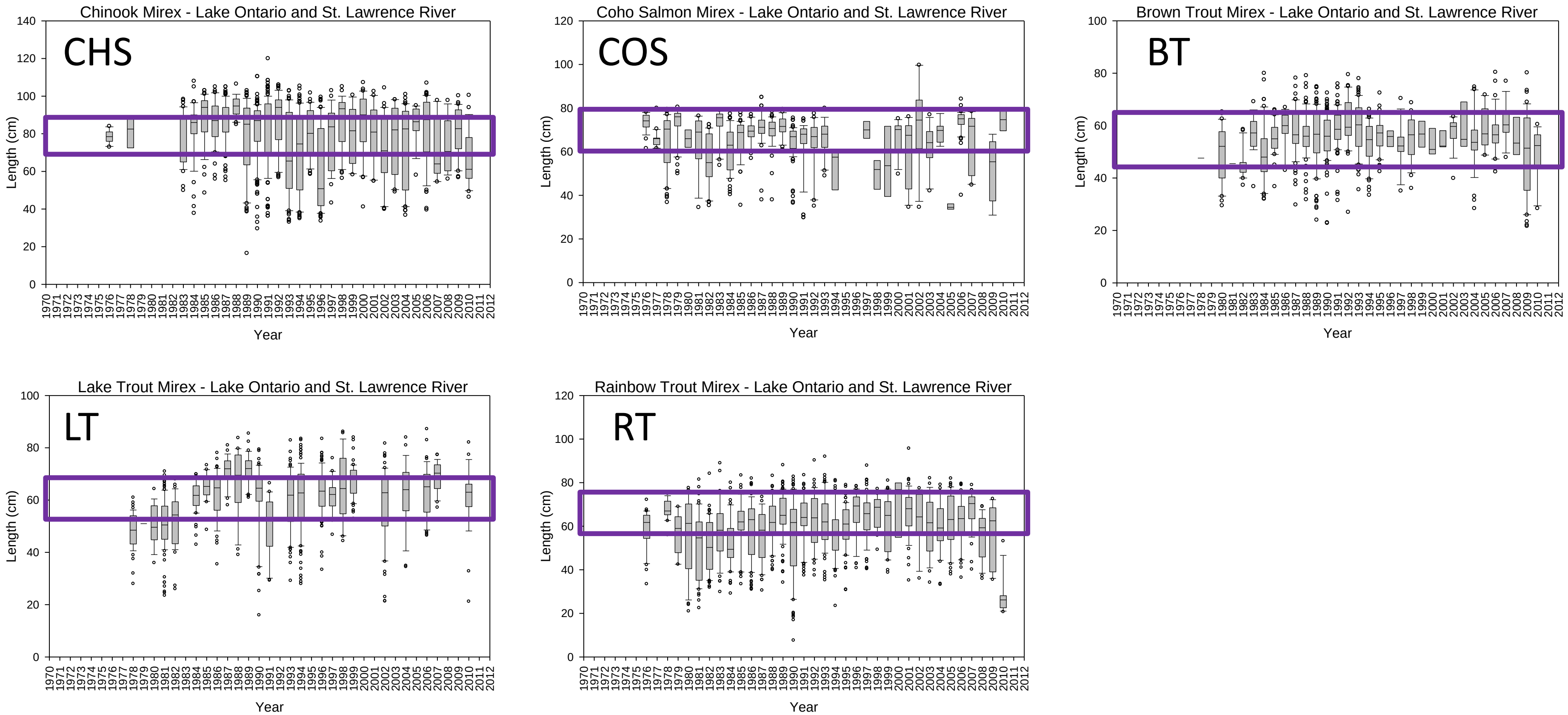


Figure S4: Mann-Kendall Sen's estimate for mean **mirex** concentrations (ng/g **lipid weight**) for Chinook Salmon, Coho Salmon, Brown Trout, Lake Trout And Rainbow Trout from various blocks of Lake Ontario. Statistically significant ($p<0.05$) Sen's estimate regressions are shown in red.

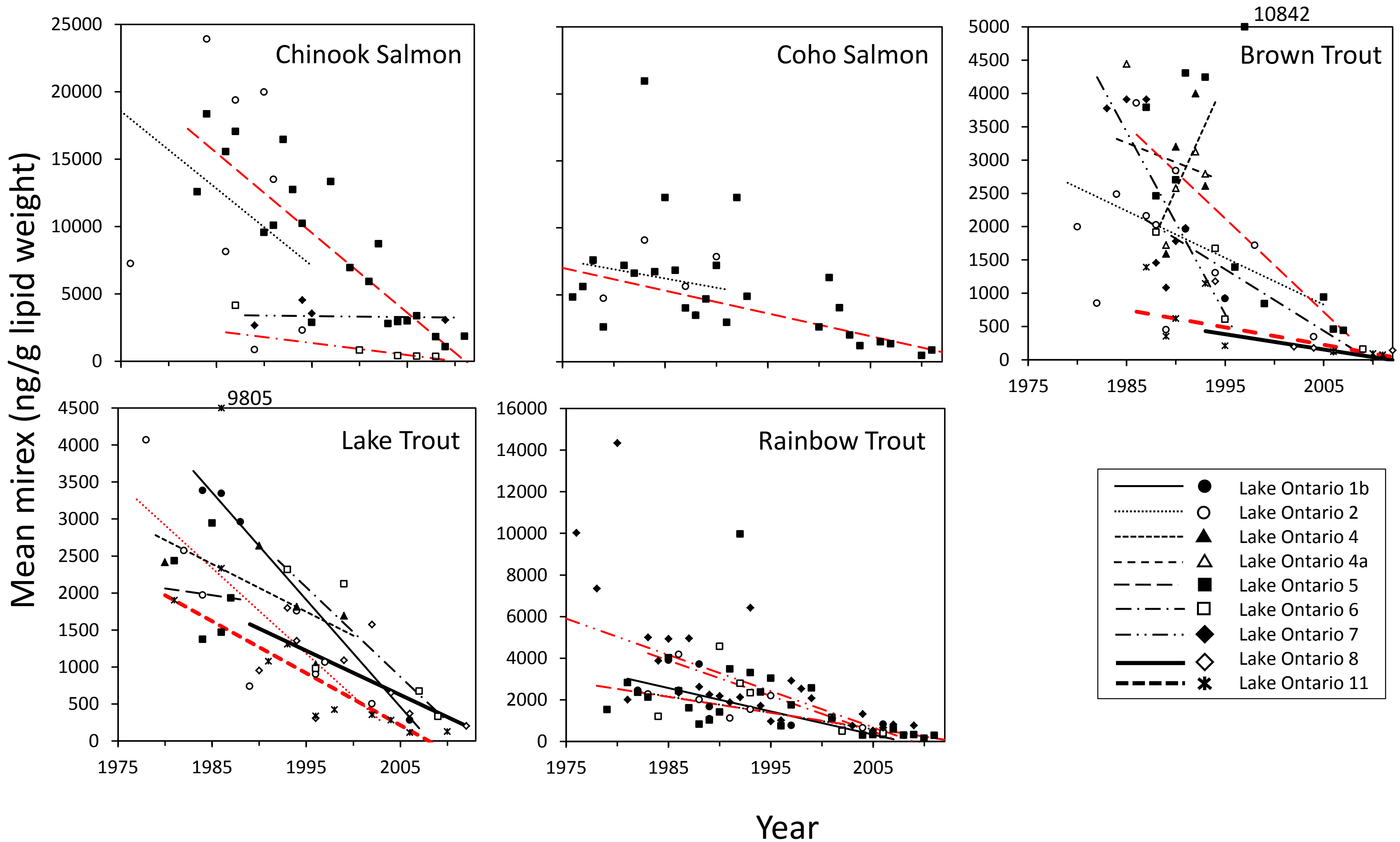


Figure S5: Mann-Kendall Sen's estimate for mean **photomirex** concentrations (ng/g **lipid weight**) for Chinook Salmon, Coho Salmon, Brown Trout, Lake Trout And Rainbow Trout from various blocks of Lake Ontario. Statistically significant ($p<0.05$) Sen's estimate regressions are shown in red.

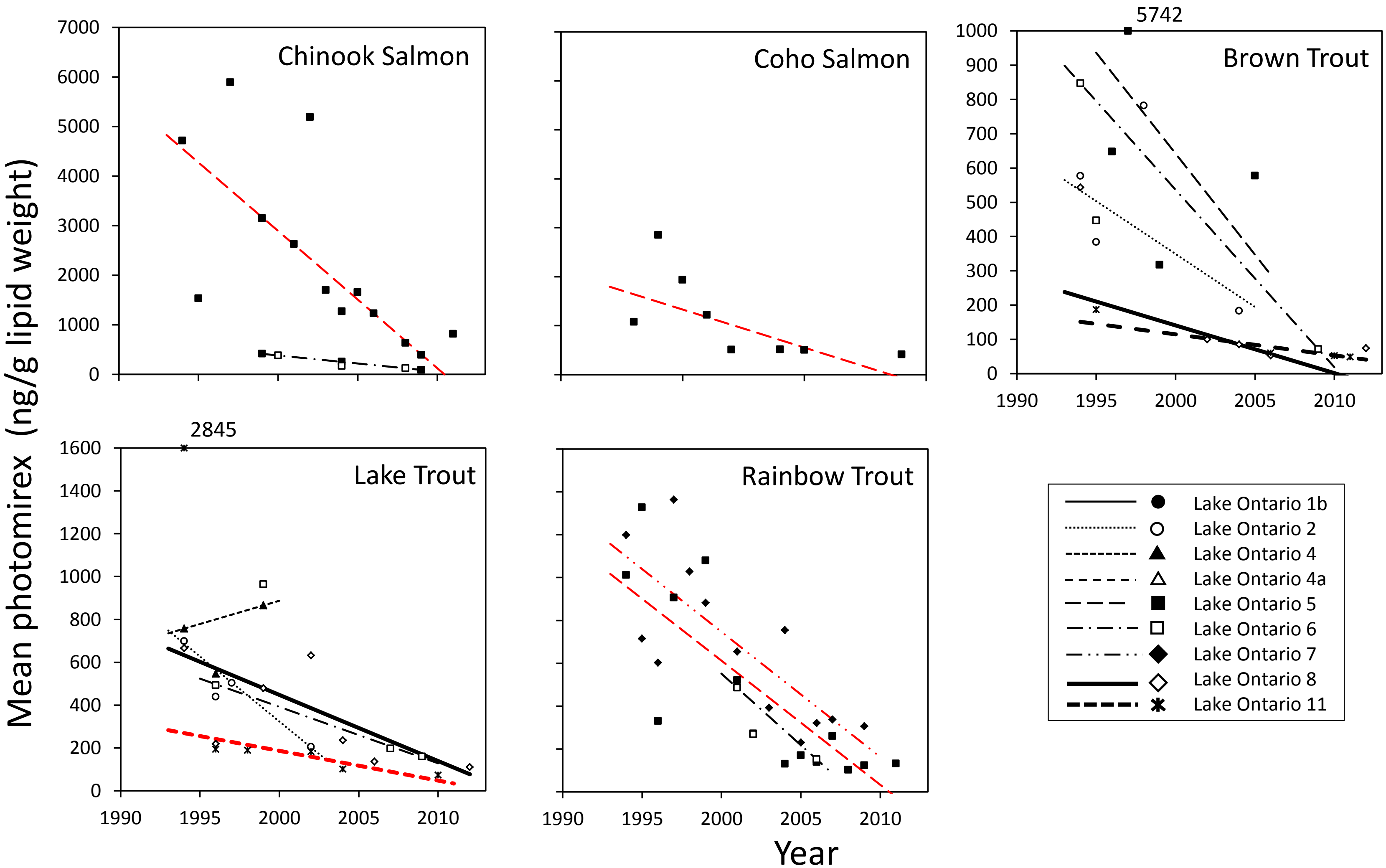


Figure S6: Mann-Kendall Sen's estimate for mean **lipid** concentrations for Chinook Salmon, Coho Salmon, Brown Trout, Lake Trout And Rainbow Trout from various blocks of Lake Ontario. Statistically significant (p<0.05) Sen's estimate regressions are shown in red.

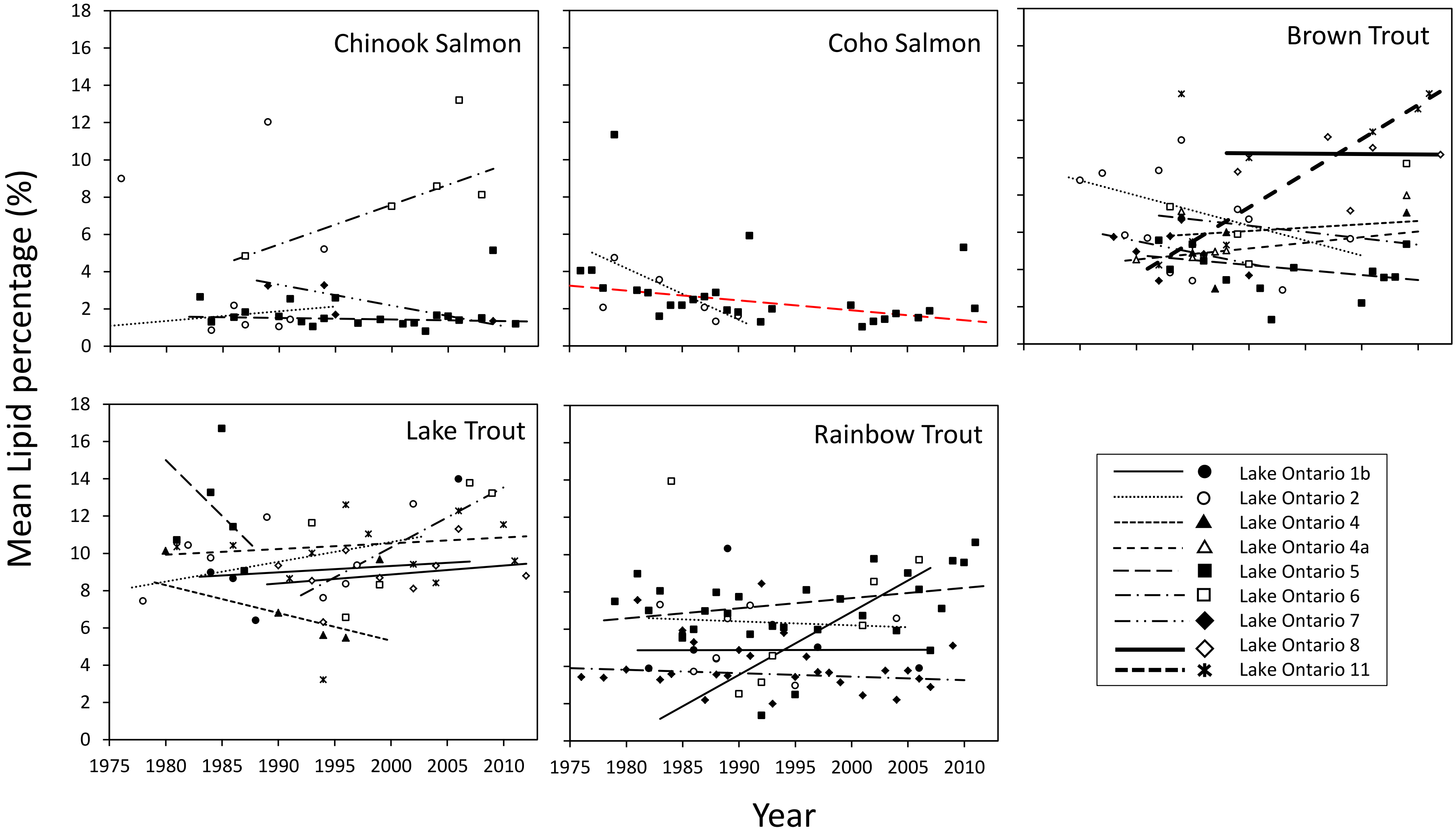


Figure S7a-a. Species-specific breakdown of fish consumption advisories for the **mirex-only** scenario for the **general population** for the combined blocks of Lakes Superior, Huron (including North Channel and Georgian Bay), Erie (excluding the St. Clair River -Lake St. Clair-Detroit River corridor) and Ontario (including the Niagara River but excluding the St. Lawrence River). The absence of data bars indicates the unavailability of advisories.

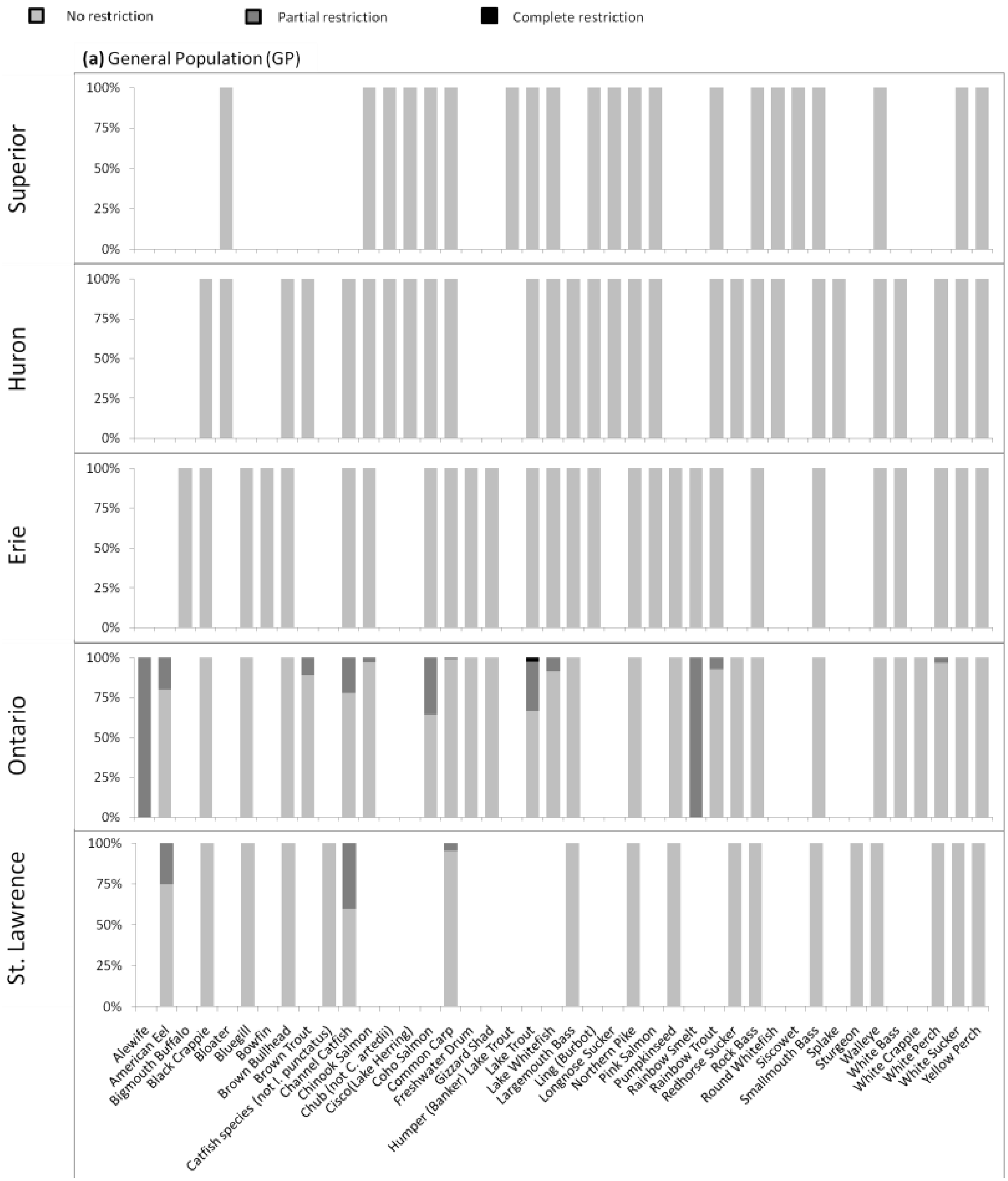


Figure S7a-b. Species-specific breakdown of fish consumption advisories for the **mirex-only** scenario for the **sensitive population** for the combined blocks of Lakes Superior, Huron (including North Channel and Georgian Bay), Erie (excluding the St. Clair River -Lake St. Clair-Detroit River corridor) and Ontario (including the Niagara River but excluding the St. Lawrence River). The absence of data bars indicates the unavailability of advisories.

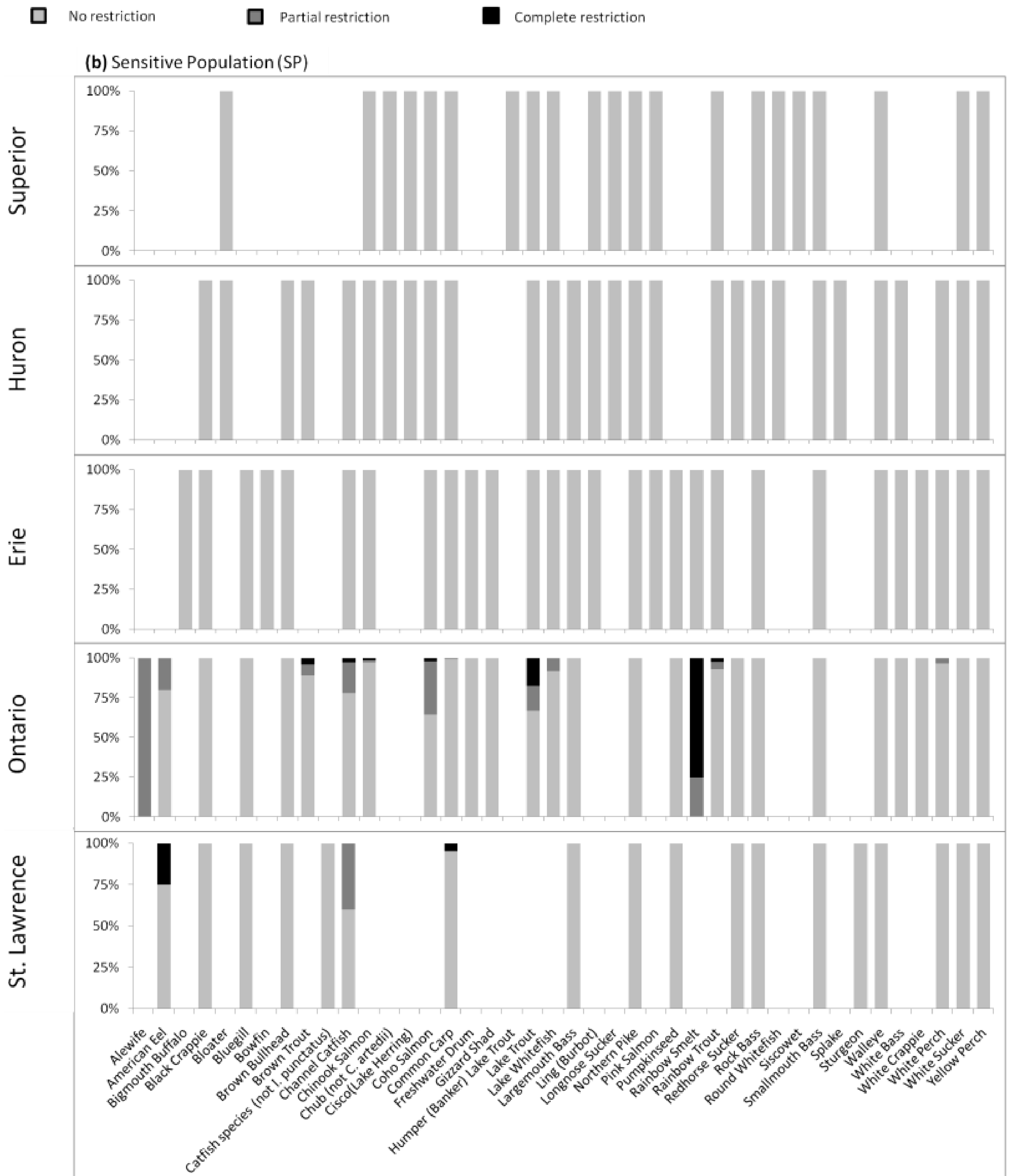


Figure S7b-a. Species-specific breakdown of fish consumption advisories for the **photomirex-only** scenario for the **general population** for the combined blocks of Lakes Superior, Huron (including North Channel and Georgian Bay), Erie (excluding the St. Clair River -Lake St. Clair-Detroit River corridor) and Ontario (including the Niagara River but excluding the St. Lawrence River). The absence of data bars indicates the unavailability of advisories.

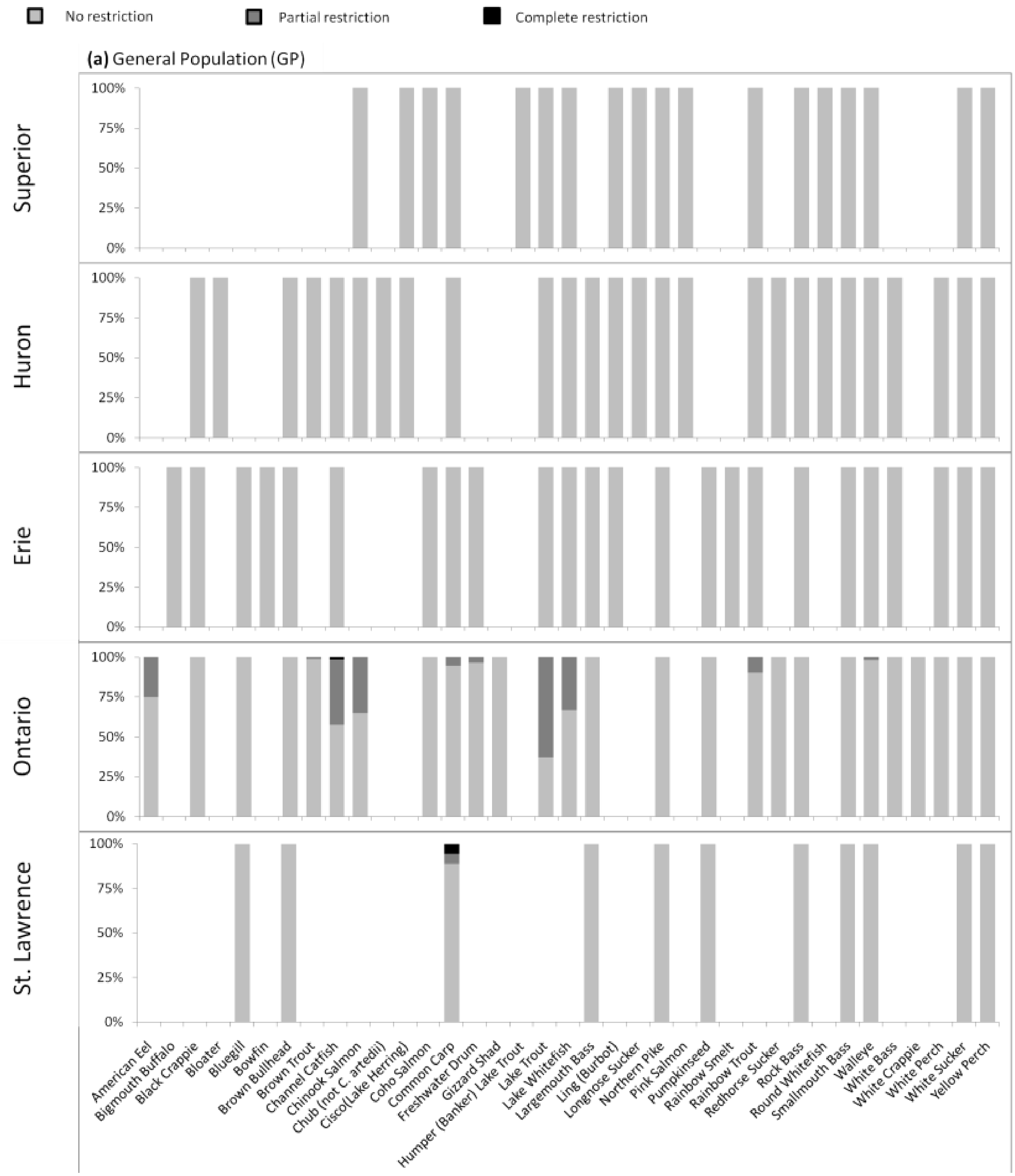


Figure S7b-b. Species-specific breakdown of fish consumption advisories for the **photomirex-only** scenario for the **sensitive population** for the combined blocks of Lakes Superior, Huron (including North Channel and Georgian Bay), Erie (excluding the St. Clair River -Lake St. Clair-Detroit River corridor) and Ontario (including the Niagara River but excluding the St. Lawrence River). The absence of data bars indicates the unavailability of advisories.

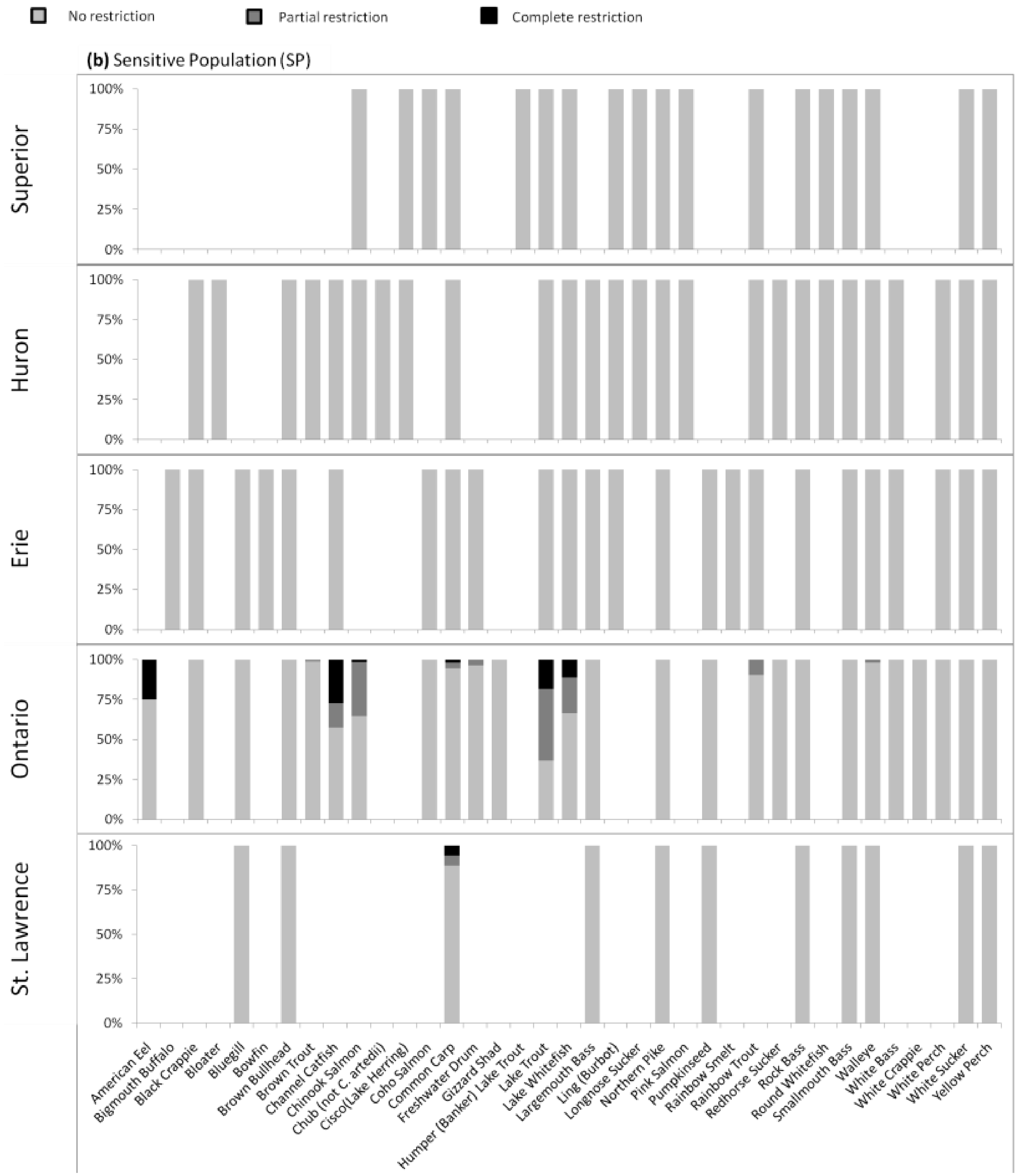


Figure S7c-a. Species-specific breakdown of fish consumption advisories for the **mirex/photomirex-only** scenario for the **general population** for the combined blocks of Lakes Superior, Huron (including North Channel and Georgian Bay), Erie (excl the St. Clair River - Lake St. Clair-Detroit River corridor) and Ontario (including the Niagara River but excluding the St. Lawrence River). The absence of data bars indicates the unavailability of advisories.

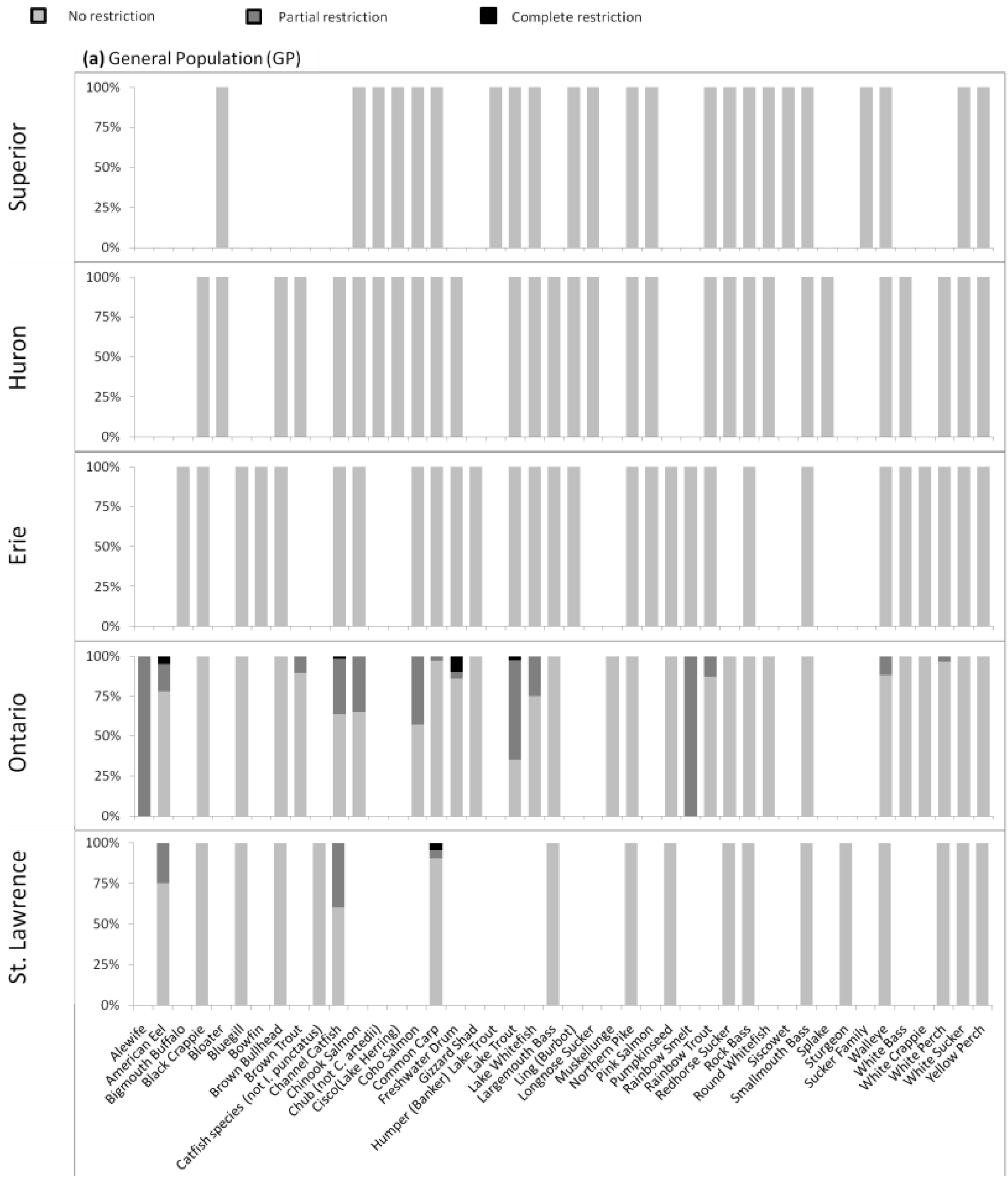


Figure S7c-b. Species-specific breakdown of fish consumption advisories for the **mirex/photomirex-only** scenario for the **sensitive population** for the combined blocks of Lakes Superior, Huron (including North Channel and Georgian Bay), Erie (excl the St. Clair River - Lake St. Clair-Detroit River corridor) and Ontario (including the Niagara River but excluding the St. Lawrence River). The absence of data bars indicates the unavailability of advisories.

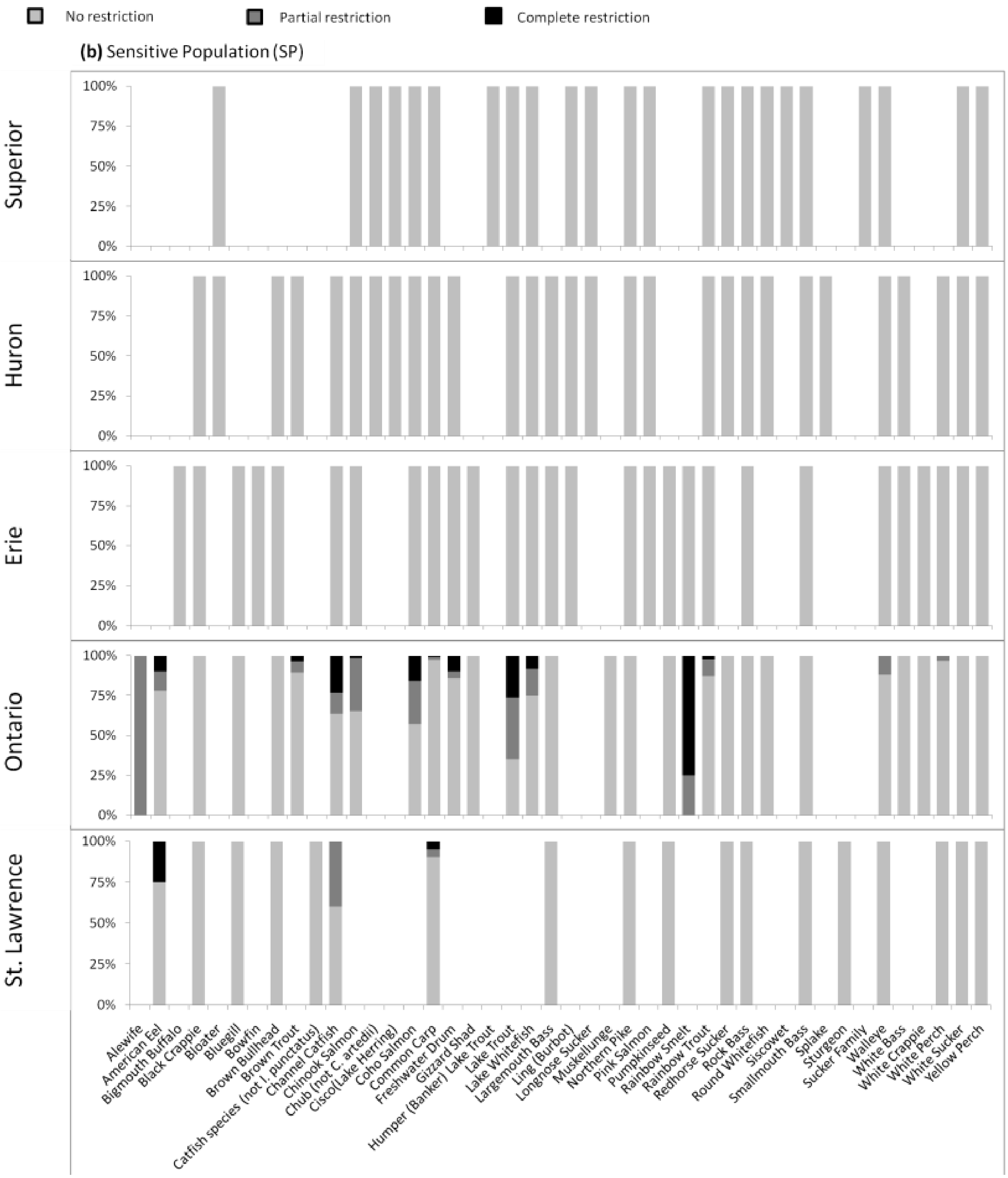


Figure S8a. Block-specific breakdown of **mirex-only** fish consumption advisories for the general (a) and sensitive (b) populations.

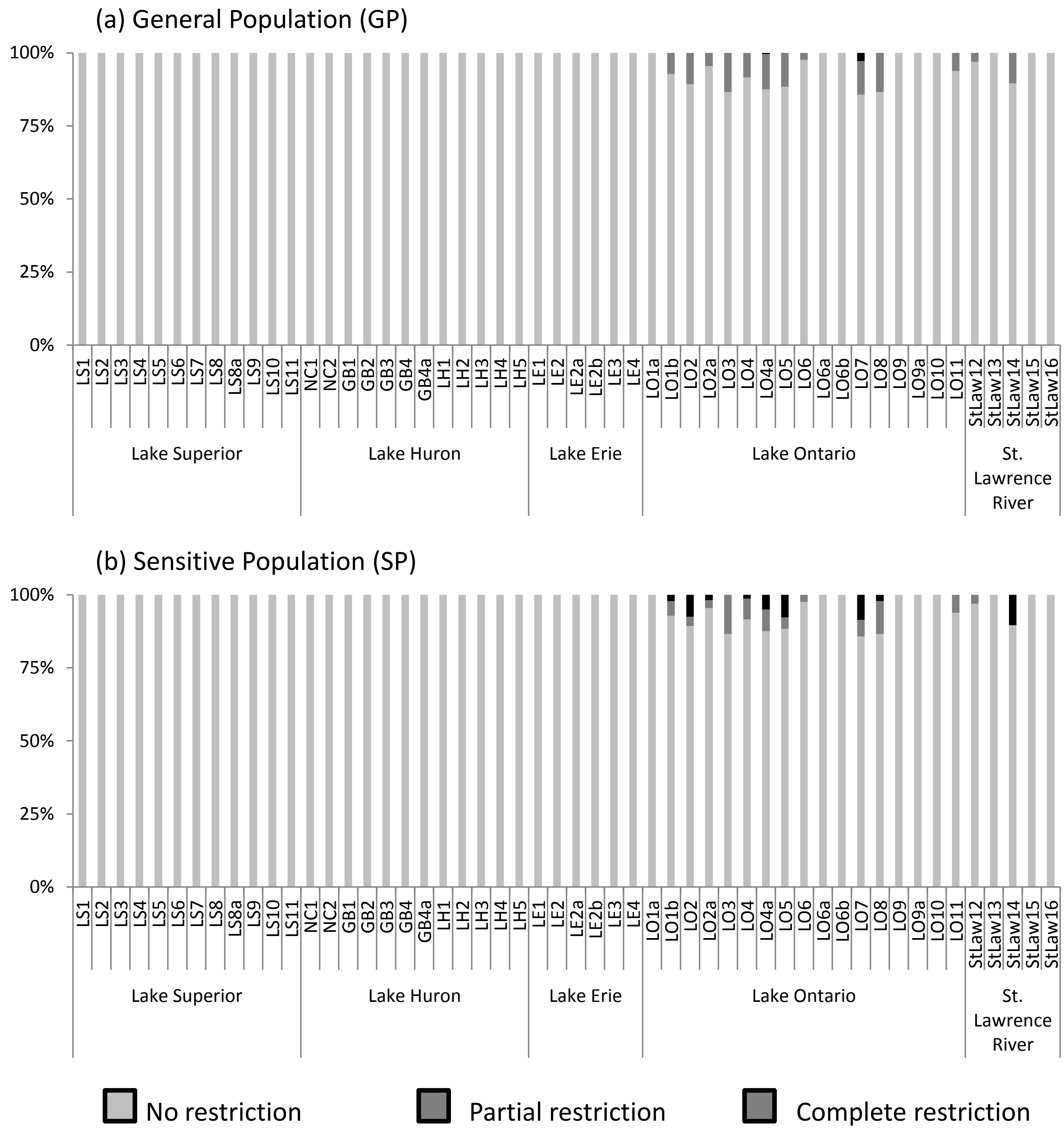
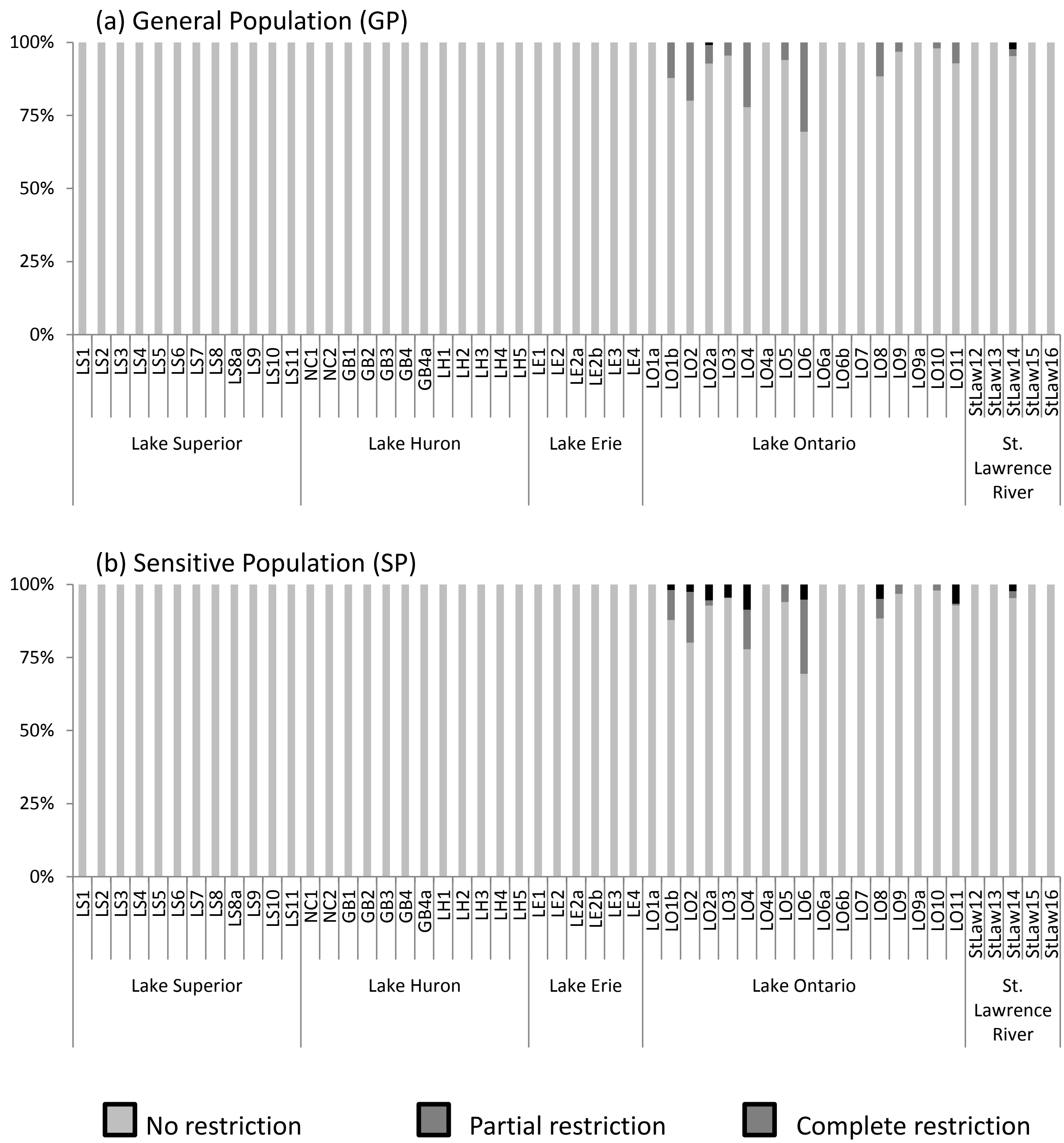


Figure S8b. Block-specific breakdown of **photomirex-only** fish consumption advisories for the general (a) and sensitive (b) populations.



References

Bhavsar, S. P., et al., 2011. Great Lakes fish consumption advisories: is mercury a concern? Ecotoxicology. 20**,** 1588-1598.

OMOE, 2009. 2009-2010 Guide to eating Ontario sport fish. Ontario Ministry of the Environment, Toronto, Ontario, Canada.