Are PCB Levels in Fish from the Canadian Great Lakes Still Declining?

Satyendra P. Bhavsar[1,2,*], Donald A. Jackson[1], Alan Hayton[2], Eric J. Reiner[3],
Tony Chen[3], and John Bodnar[3]

[1]*Department of Ecology and Evolutionary Biology*
University of Toronto
Toronto, Ontario M5S 3G5

[2]*Environmental Monitoring and Reporting Branch*
Ontario Ministry of the Environment
125 Resources Road
Toronto, Ontario M9P 3V6

[3]*Laboratory Services Branch*
Ontario Ministry of the Environment
125 Resources Road
Toronto, Ontario M9P 3V6

ABSTRACT. *Long- and short-term levels and trends of polychlorinated biphenyls (PCBs) in lake trout* (Salvelinus namaycush) *and walleye* (Sander vitreus) *from the Canadian waters of the Great Lakes are examined using the bootstrap resampling method in light of the Great Lakes Strategy 2002 (GLS-2002) objective of decrease in concentrations by 25% during 2000–2007. This objective has been set as an indicator of progress toward the long-term goal of all Great Lakes fish being safe to eat without restriction. Lake Superior lake trout and walleye PCB concentrations were almost unchanged between 1990–2006, and the bootstrap analysis suggests that the probability of achieving the GLS-2002 objective is negligible (< 2%). The PCB levels in Lake Huron lake trout and walleye are decreasing; the declines between 2000–2007 are estimated to be 25–35% and 5–30%, respectively. In contrast, Lake Erie walleye concentrations will likely increase by 25–50% between 2000–2007. For Lake Ontario lake trout, achieving the 25% reduction target seems highly probable with a likely decrease of 45–55%; for Lake Ontario walleye, the probability of achieving such a reduction is only 8% with an expected change of –13 to +15%. Although the targeted reduction may not be achieved for walleye from Lakes Superior, Huron, and Ontario, their best projected 2007 PCB levels are below the unlimited fish consumption guideline of 105 ng/g wet weight used by the Ontario Ministry of the Environment. In contrast, although there are high probabilities of achieving the goal for lake trout from Lakes Huron and Ontario, their best projected 2007 PCB levels (160 and 370 ng/g ww, respectively) will continue to result in consumption restrictions. Lake Superior lake trout concentrations may remain unchanged at the current elevated level of 160 ng/g ww. For Lake Erie fish, the projected 2007 concentrations and the increasing trends are both worrisome. Additional measurements beyond 2007 are necessary to confirm these estimates because of the observed periodic oscillations in the concentrations.*

INDEX WORDS: Polychlorinated biphenyls, PCB, chlorinated organic contaminant, Laurentian Great Lakes, long-term trend, fish.

INTRODUCTION

The Great Lakes of North America form the largest fresh-water system on Earth, and are often referred to as inland seas. The lakes supply drinking water to tens of millions of people, act as a major mode of bulk good transport, and are used for recreational purposes. The sport- and commercial-fishing industry represents multi-billion dollars annually. Unfortunately, this vast and important

*Corresponding author. E-mail:
s.bhavsar@utoronto.ca *or* satyendra.bhavsar@ontario.ca

freshwater system has been impaired by elevated concentrations of bioaccumulative, toxic, and persistent organic pollutants (POPs) and other stressors (Environment Canada and U.S.EPA. 2003).

Contamination of the Great Lakes with polychlorinated biphenyls (PCBs) began in the 1930s when PCBs were first synthesized, and peaked in the 1960s–70s (Tanabe 1988). PCBs were used extensively in a wide range of applications, including dielectric fluids for capacitors and transformers, heat transfer and hydraulic fluids, lubricating and cutting oils, and as additives in pesticides, paints, paper, adhesives, sealants, and plastics. In the 1970s, PCB production was effectively banned in North America and the use of PCBs declined. It is well documented that the levels of PCBs in the Great Lakes declined rapidly during the late 1970s and 1980s (Baumann and Whittle 1988, Jeremiason *et al.* 1994, Swackhamer 2005); however, the rate of decrease diminished in the 1990s (De Vault *et al.* 1996, Hickey *et al.* 2006).

Although the current levels of PCBs are low compared to the historical peak values, they remain of concern due to the potential health risk they pose, especially to humans. This is evident from the large number of PCB-driven fish consumption advisories placed for most of the locations in and around the Great Lakes (U.S.EPA 2003, MOE 2005a). As such, it is important to monitor the trends of PCB levels in the Great Lakes and to assess if the system is responding positively to various contamination abatement strategies that are being implemented.

Biomonitors have been used as valuable indicators of ecosystem health. This is because water concentrations of contaminants are highly variable and sediment concentrations are spatially heterogeneous. In contrast, fish integrate temporal and spatial variability over the area they travel and can act as cost-effective surrogates over the long term (De Vault *et al.* 1996). Short-lived fish species such as spottail shiners (*Notropis hudsonius*) are generally used as biomonitors for site-specific assessments (Suns *et al.* 1993, Scheider *et al.* 1998), while top predators such as lake trout (*Salvelinus namaycush*) are monitored to assess lake-wide conditions (Bentzen *et al.* 1999). Although fish concentrations respond slowly to changes in water concentrations and thereby external loadings, they can portray an overall condition of the aquatic system by encompassing both water and sediment concentrations through pelagic and benthic exposure. In addition, fish are the primary link for contaminant transfer from an aquatic system to humans and wildlife. Therefore, long-term biomonitoring programs have been recognized as a valuable tool for assessing potential hazards associated with contaminant levels in aquatic systems, understanding contaminant dynamics, developing predictive models, and determining the effectiveness of regulatory strategies (Mackay 1989, Bentzen *et al.* 1999, Carlson and Swackhamer 2006).

The U.S. Policy Committee (USPC) created the Great Lakes Strategy 2002 (GLS-2002) to advance the restoration and protection of the Great Lakes Basin Ecosystem (U.S.EPA 2002). In order to quantify progress toward the long-term goal of no fish advisories due to toxic substances in the Great Lakes fish, the strategy set a key objective of a 25% decline in lake trout and walleye PCB concentrations between 2000 and 2007 (U.S.EPA 2002). For Lake Michigan lake trout, Stow *et al.* (2004) performed a trend analysis on data collected between 1972–2000 and estimated that the declines in the PCB concentrations between 2000–2007 would be in the range of 5–10%. It is recognized that trend analysis helps in describing past behavior of different processes, understanding the mechanisms behind changes, assessing the impacts of management actions, and extrapolating data for policy guidance. Although monitoring data are essential for verifying projected values, trend analysis aids in making timely decisions.

In order to assess historical as well as recent spatial distributions and temporal trends of PCB levels in lake trout and walleye from the Canadian waters of the Great Lakes, we analyze long-term (1970s–2006) biomonitoring data collected by the Sports Fish Contaminant Monitoring Program (SFCMP) of the Ontario Ministry of the Environment (MOE, Canada). Using trend analysis performed on the long-term (1970s–2006) and recent (1990–2006) fish measurements, we assess whether PCB levels in fish from the Canadian waters of the Great Lakes are still decreasing, have leveled off, or if the trend has been reversed due to increasing concentrations. We then use bootstrap resampling analysis to examine if the GLS-2002 target of 25% decrease in lake trout and walleye PCB concentrations from 2000–2007 is feasible.

MATERIALS AND METHODS

Sample Collection

The SFCMP of MOE has been monitoring contaminant concentrations in Ontario sport fish for ap-

proximately 30 years. As a part of this program, fish samples are collected from the Canadian waters of the Great Lakes generally on an annual basis in collaboration with the Ontario Ministry of Natural Resources (MNR), Canada. Lake trout and walleye (*Sander vitreus*) were collected in varying numbers from several locations in each of Lakes Superior, Huron, Erie and Ontario from 1976–2006 during late summer or early fall using gill nets and electro-fishing. The samples were not necessarily collected from the same location in a lake every year. After collection, fish were stored frozen as whole fish and shipped to MOE laboratories for chemical analysis, where they were measured for length and weight, sexed and filleted (skin-off) before analysis.

Extraction and Analysis

A skinless, boneless fillet of the dorsal muscle of each fish was used to quantify total-PCB levels using the MOE method E3136 (MOE 2005b). This method entails hydrochloric acid digestion of ho-mogenized fish tissue, followed by extraction with hexane/dichloromethane. Complete procedures have been described in detail elsewhere (MOE 2005b). Briefly, fish tissue homogenates were weighed (5.0 g) and transferred to centrifuge tubes. A 0.5 mL aliquot of a 5:2 mixture of decachloro-biphenyl and 1,3,5-tribromobenzene (100 µg/mL) was added to each sample as a surrogate spike, fol-lowed by 18 mL of concentrated, reagent grade hy-drochloric acid and left to digest over night. A 20 mL aliquot 25% (v/v) dichloromethane (DCM) in hexane was added to each sample to extract organic compounds from the acid digestate. Solutions were mixed for 45 minutes on a bench top rotator, and then allowed to stand for 24–48 hours to permit separation of emulsion (if present). The top solvent layer was quantitatively withdrawn and transferred to 100 mL volumetric flasks, and volumes made up to 100 mL with DCM/hexane. Sample extracts 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. A 25 mL aliquot of hexane was used to elute PCBs. Column effluents, containing PCB and mirex (frac-tion 1), were collected in 40 mL graduated tubes and any hexane remaining at the top of the columns was drained off. The remaining compounds (mostly organochlorine pesticides; fraction 2) were then eluted with a 25% (v/v) DCM/hexane and collected in 40 mL tubes. Pure iso-octane (1 mL) was added

to fractions 1 and 2, and the sample extracts were evaporated to 1 mL final volumes.

Gas-liquid chromatography was used to deter-mine total-PCB concentrations using an HP 5890 Series-II gas chromatograph and Ni[63] electron cap-ture detector (ECD). A J&W DB-17 0.53 mm i.d., 0.1 µm film was used. The column head pressure was 3.5 PSI and the temperature program for frac-tion 1 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 6 min. Total–PCBs were determined using a 4:1 mix-ture of Aroclors 1254:1260 for quantification. This ratio of Aroclors best resembled the congener pat-terns detected for most fish samples. The chro-matography was detuned to resemble classical packed column chromatography for classical Aro-clor matching analysis. Quantification was carried out using the 23 largest "Aroclor" peaks obtained in the pseudo packed column technique. For lower level samples, a minimum of 11 peaks was required for a positive identification. The areas of the peaks detected were summed and compared to the summed areas of the 4:1 mixture of Aroclor 1254:1260. A five point calibration curve with sin-gle point continuing calibration was used to quan-tify samples. The method detection limit (MDL) is 20 ng/g. A blank and spiked blank matrix sample was processed with each set of samples (20 to 30). The method performance is monitored through lab-oratory intercalibration studies (the Northern Cont-aminants Program (NCP) and Quality Assurance of Information for Marine Environmental Monitoring in Europe (QUASIMEME)).

Statistical Analysis

In general, the concentrations of PCBs in fish in-crease with fish size (Miller 1994). The size of the fish samples collected varied over a wide range on both an inter- and intra-annual basis. The mean lengths for Canadian Great Lakes lake trout and walleye are approximately 60 and 50 cm, respec-tively (Bhavsar *et al.* In press). To compare the an-nual levels of dioxins and furans, Bhavsar *et al.* (In press) standardized the contaminant concentrations at these mean fish lengths using lake-specific an-nual regressions. However, such a standardization is appropriate when the annual regression slopes of logarithmic concentration versus fish length are parallel (Somers and Jackson 1993). For PCB con-centrations considered in this study, we observed statistically significant differences in the slopes (lake and fish specific *P* values varying from

< 0.001–0.5). As such, instead of standardization, we opted to narrow down the size of fish considered in this study to 55–65 and 45–55 cm for lake trout and walleye, respectively. The selection of these ranges was somewhat arbitrary to include the observed mean lengths of 60 and 50 cm, respectively, to maximize the sample size, and to minimize the influence of fish length on PCB level.

In this study, the measurements for Georgian Bay, a large bay of Lake Huron, were combined with measurements for Lake Huron in order to maximize sample size. We had limited Lake Erie lake trout samples due to their low population in the lake. As such, for Lake Erie, we primarily relied on PCB measurements for walleye rather than for lake trout. Data are reported on a wet weight basis and not on a lipid normalized basis, first, to be consistent with previous studies on temporal contaminant trends in the Great Lakes (e.g., Baumann and Whittle 1988, Huestis *et al.* 1996, Hickey *et al.* 2006), and second, because lipid normalization can be inappropriate and potentially misleading (Hebert and Keenleyside 1995, Stow *et al.* 1997).

The GLS-2002 sets a goal of 25% decrease specifically in "whole fish" rather than fillet concentrations. The primary purpose of the SFCMP of MOE is to issue fish consumption advisories; therefore, the SFCMP measurements are for contaminant levels in fish fillets. A previous study found an almost linear relationship between fillet and whole-fish PCB concentrations for Lake Michigan coho salmon (*Oncorhynchus kisutch*) and rainbow trout (*O. mykiss*) (Amrhein *et al.* 1999). As such, trends in fillet measurements should reflect trends in the corresponding wholefish levels as well (Stow *et al.* 2004).

For the temporal trend assessment, the concentrations were log transformed to stabilize variance before performing regression analyses using SPSS (version 10.1.3, 2001, SPSS Inc., Chicago, IL, USA). A nonparametric Mann-Kendall test (Gilbert 1987) was also performed to assess monotonic temporal trends in the recent PCB levels. A non-parametric Sen's method (Gilbert 1987) was then used to estimate the magnitude of trends. The calculations were performed using an Excel® (version 2003; Microsoft Corp., Redmond, WA, USA) template MAKESENS – <u>M</u>ann-<u>K</u>endall test for trend and <u>Sen's</u> slope estimates (Salmi *et al.* 2002).

The bootstrap method was used to prepare confidence intervals of the standard regression estimated changes in fish concentrations from 2000–2007. The analysis was performed using Resampling Stats for Excel software (version W3.20; Resampling Stats Inc., Arlington, VA, USA) and 10,000 iterations. In this analysis, a new set of regression slope and intercept was prepared for log-transformed concentrations by resampling data points at every iteration with replacement from the original sample. The 10,000 combinations of slope and intercept were then used to estimate fish concentration in year 2007. These estimated 10,000 values for the year 2007 concentrations were compared with the fish concentration for the year 2000, which was calculated using the standard regression from SPSS. The expected percentage changes in the concentrations during 2000–2007 were reported using frequency distribution histograms.

RESULTS AND DISCUSSION

Spatial Differences

During 1970–80s, the inter-lake differences in the Canadian Great Lakes lake trout and walleye PCB concentrations were 2–9-fold with Lake Ontario fish having the highest concentrations (Fig. 1). The historical levels were generally in the order of Lakes Superior < Huron ≈ Erie << Ontario (Fig. 1). PCB concentrations in lake trout from Lake Michigan (Hickey *et al.* 2006), the fifth lake of the Great Lakes, were comparable to those in Lake Ontario. The PCB concentrations in lake trout ranged from 200–1,400 ng/g wet weight (ww) in Lake Superior to 1,000–5,000 ng/g ww in Lake Ontario. The levels in walleye ranged from approximately 25–325 ng/g ww in Lakes Superior, Huron and Erie to 100–1,000 ng/g ww in Lake Ontario. Similar spatial heterogeneity in historical fish concentrations have been reported (Baumann and Whittle 1988, De Vault *et al.* 1996). These inter-lake differences were also reflected in the limited measurements of water concentrations (Stevens and Neilson 1989).

During late 1990s (i.e., 1998–2000) the among-lake differences in the fish PCB levels decreased to 2–6-fold. The median PCB concentrations (ng/g ww) in 1998–2000 were still in the historical order: for lake trout, Lakes Superior (130–160) < Huron (260–330) ≈ Erie (340) << Ontario (570–1,350); for walleye, Lakes Superior (20–50) < Huron (30–100) ≈ Erie (60–100) << Ontario (160) (Fig. 1). This order reported here from the 1998–2000 data collected by SFCMP of MOE is consistent with the observations of the Great Lakes Fish Monitoring Program (GLFMP) administered by the U.S.EPA Great Lakes National Program Office (GLNPO) (Carlson and Swackhamer 2006). However, the re-
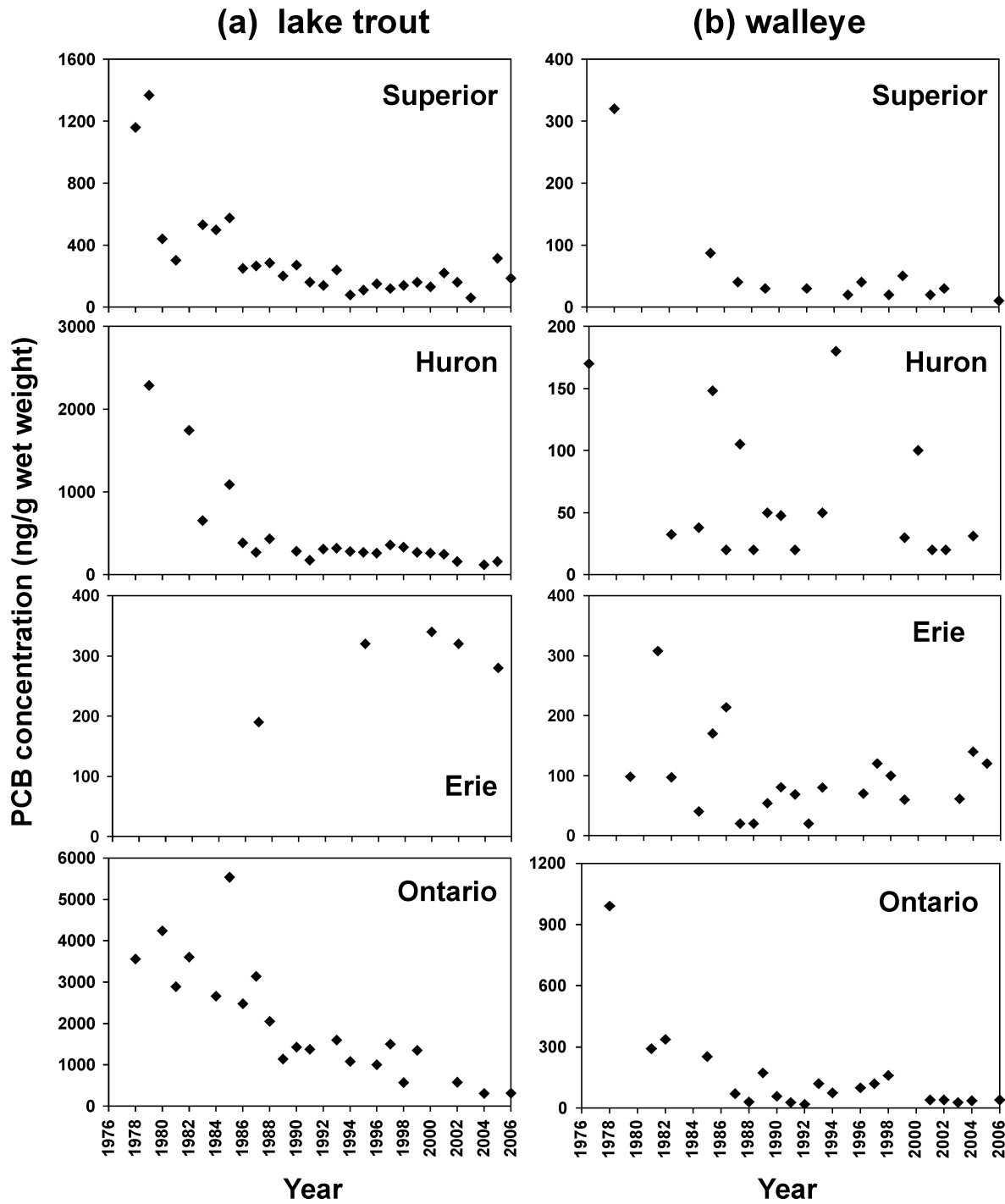
(a) lake trout

(b) walleye

FIG. 1. *Measured PCB concentrations (ng/g wet weight) in skinless fillets of (a) lake trout and (b) walleye collected from Lakes Superior, Huron, Erie, and Ontario between 1976 and 2006. Values are medians of the observed values for fish length of 55-65 and 45-55 cm for lake trout and walleye, respectively. The annual sample sizes were lake and species specific as shown in the data points for Figure 2.*

cent (2000–2006) measurements suggest that although the lake trout median PCB levels (ng/g ww) are still in the historical order of Lakes Superior (60–220) < Huron (120–260) < Erie (280–340) < Ontario (310–580), the walleye median PCB levels (ng/g ww) are now in the order of Lakes Superior (20–30) < Huron (25–45) ≈ Ontario (28–40) < Erie (60–140). The among-lake differences have decreased and are now 2–4-fold.

Temporal Trends

In general, PCB levels in lake trout and walleye declined rapidly by about an order of magnitude during the late 1970s and 1980s (Fig. 1) (Borgmann and Whittle 1991, Suns *et al.* 1993, Bentzen *et al.* 1999). A similar rate of decline in Lake Superior water concentrations from 2.4 ng/L in 1980 to 0.18 ng/L in 1992 has been reported and attributed to loss by volatilization (Jeremiason *et al.* 1994). Decreases in Lake Ontario fish concentration can be primarily linked to source reductions in the Niagara River (Durham and Oliver 1983, Whittle and Fitzsimons 1983). This is supported by decreasing lake trout PCB concentrations from west to east in Lake Ontario (Borgmann and Whittle 1991).

The downward trend in concentrations, however, has leveled off to a certain extent since the early 1990s (Fig. 1). Measurements from other monitoring programs and studies have also indicated relatively stable concentrations in the Great Lakes fish since late 1980s (Stow *et al.* 1995, De Vault *et al.* 1996, Hickey *et al.* 2006). Many explanations have been proposed to explain this change in the decline pattern. For example, a decrease in external PCB inputs may have affected the dynamics of contaminants in the systems. Sediments that historically served as a net sink for PCBs may have turned into a net source (Mackay 1989, Pearson *et al.* 1996). Under the new dynamics, PCB cycling between atmosphere and water through wet and dry deposition and volatilization might be playing an important role in PCB movement (Mackay 1989, Jeremiason *et al.* 1994).

Another factor that has affected the PCB trend in lake trout and walleye from the Great Lakes is food web structure. Introduction of many invasive species especially during the late 1980s and early 1990s have been reported (Sprules *et al.* 1990, Mills *et al.* 1993). This may have altered the way contaminants are accumulated in predator fish due to modifications in the structure of the food chains (Morrison *et al.* 1998). Changes in alewife popula-

tion, a major prey of lake trout, have been correlated with PCB levels in lake trout (Borgmann and Whittle 1991). Similarly, a decline in lake trout lipid content also contributed to decreased PCB levels (Borgmann and Whittle 1991). This hypothesis of the effect of changes in food web structure is supported by the leveling off of biota levels despite continued decreases in water concentrations (De Vault *et al.* 1996). It is also likely that PCBs are efficiently recycled within the biota of the food web (Stow *et al.* 2004).

Model Forecast

Figure 2 presents regressions for long-term (1970s–2006) PCB trends. For Lakes Superior, Huron, and Ontario, regressions for the lake trout data provided a better fit (R^2 = 0.24 to 0.49) than for the walleye data (R^2 = 0.1 to 0.25) (Fig. 2). For Lake Erie, the greater scatter in the observed concentrations resulted in lower amounts of variation being explained ($R^2 < 0.01$) for walleye. As mentioned above, PCB concentrations in fish are correlated to fish length. Selection of size ranges (55–65 and 45–55 cm for lake trout and walleye, respectively) rather than standardization to mean fish lengths produced relatively large scatter in the annual data points, i.e., the variation associated with individual fish rather than sample means are being evaluated. Further, it is likely that the year-to-year variation in the contaminant levels is a result of real differences in local populations within a lake because the samples were not necessarily collected from the same location every year (Carlson and Swackhamer 2006). However, temporal trends over the relatively long time period evaluated in this study should portray a good overall picture of contaminant levels in lake trout and walleye due to their movements over large areas.

When we performed the bootstrap resampling analysis on these long-term regressions (Fig. 2) to project likely concentrations in the year 2007, the analysis suggested that the GLS-2002 targeted decrease of 25% in PCB levels between 2000 and 2007 is achievable in all lakes except Lake Erie (Fig. 3). The long-term trajectories suggest a very high likelihood to achieve concentration decreases of at least 25% in lake trout from Lakes Superior, Huron, and Ontario (Fig. 3a). For walleye, more than 90% of the bootstrapped values exceeded this decrease for Lake Ontario whereas 75% and 60% of the values showed at least a 25% reduction for Lakes Superior and Huron (Fig. 3b). In contrast, for
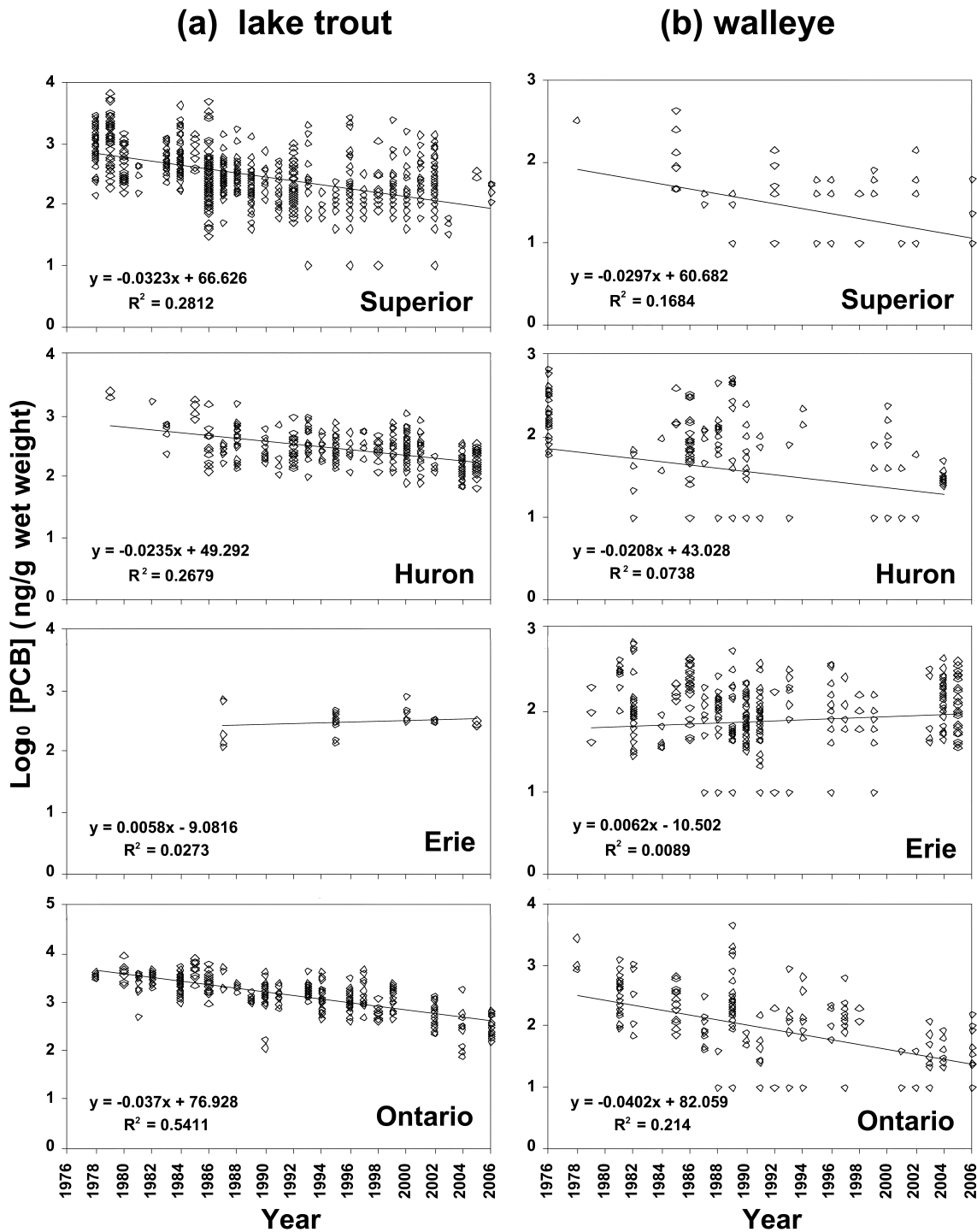
FIG. 2. *Long-term (1976–2006) temporal trends of PCB levels (log-transformed, ng/g wet weight) in (a) 55–65 cm lake trout and (b) 45–55 cm walleye collected from Lakes Superior, Huron, Erie, and Ontario. The lines are estimated linear regressions.*
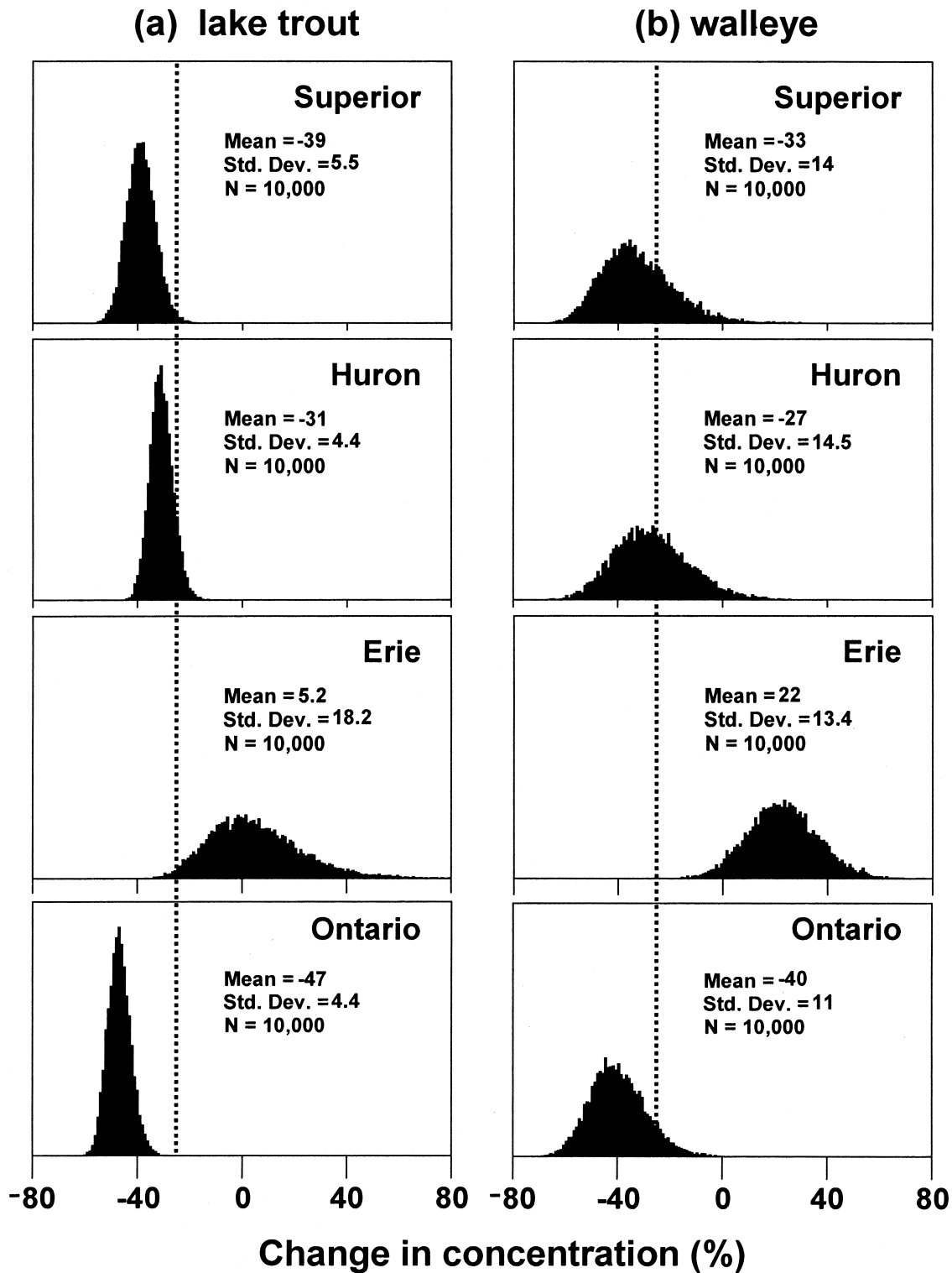
(a) lake trout

(b) walleye

Superior

Mean = -39
Std. Dev. = 5.5
N = 10,000

Superior

Mean = -33
Std. Dev. = 14
N = 10,000

Huron

Mean = -31
Std. Dev. = 4.4
N = 10,000

Huron

Mean = -27
Std. Dev. = 14.5
N = 10,000

Erie

Mean = 5.2
Std. Dev. = 18.2
N = 10,000

Erie

Mean = 22
Std. Dev. = 13.4
N = 10,000

Ontario

Mean = -47
Std. Dev. = 4.4
N = 10,000

Ontario

Mean = -40
Std. Dev. = 11
N = 10,000

-80   -40   0   40   80   -80   -40   0   40   80

Change in concentration (%)

FIG. 3. Frequency distributions of the estimated percentage changes in PCB concentrations between the years 2000 and 2007. The histograms were prepared from 10,000 iterations of bootstrap resampling analysis performed on the long-term (1976–2006) PCB measurements for the Canadian Great Lakes (a) lake trout and (b) walleye. Negative value indicates a decrease in the concentration. The dotted line drawn at –25% represents a key objective of the Great Lakes Strategy 2002 (U.S.EPA 2002). The area under the curve on the left side of the dotted line represents the proportion of bootstrapped estimates achieving the GLS-2002 targeted decrease of 25% in PCB levels.

Lake Erie, the best estimated average change (i.e., mean boostrapped value) in Lake Erie lake trout and walleye PCB concentrations are increases of 5% and 22%, respectively (Fig. 3). Therefore, based on the long-term trends, it appears that Lake Erie lake trout and walleye will most likely not achieve the 25% PCB reduction goal of the GLS-2002.

As discussed in the *temporal trend* section, many studies have found that the original first-order declining trend of PCB concentrations in fish from the Great Lakes has slowed since the late 1980s (Stow *et al.* 1995, De Vault *et al.* 1996, Hickey *et al.* 2006). Therefore, it is more appropriate to project future concentrations using recent temporal trends. We recognize that estimates from such a trend analysis can be highly sensitive to the time period selected. Therefore, to avoid the influence of short-term (3–6 years) oscillations observed in the annual fish concentrations (French *et al.* 2006), to attain a moderate number of data points, and to harmonize the time frame with possible significant changes in the system dynamics as discussed earlier, we selected the 1990–2006 data for the recent trend analysis.

The regression equations prepared using the 1990–2006 observations (Fig. 4) portrayed a different scenario compared to the long-term analyses for possible changes in the Lakes Superior fish concentrations between 2000–2007 (Figs. 3 and 5). In contrast to the more than 30% decline in PCB concentrations in Lake Superior lake trout and walleye estimated by the long-term trends (Fig. 3), the short-term trends suggest that the concentrations will remain almost unchanged or may even slightly increase between 2000–2007 (Fig. 5). When we performed bootstrap analysis on these recent measurements, only < 2% and 15% of the bootstrap estimates achieved the targeted 25% reduction in PCB levels of Lake Superior lake trout and walleye, respectively (Fig. 5). It has been suggested that a large portion (> 50%) of PCB loading to Lake Superior is transported to the lake sediments; however, only 2–5% of this settled PCB is retained in the sediments and most of the PCB is recycled in the benthic region. This leads to sustained high PCB levels in the Lake Superior fish via the benthic food web (Jeremiason *et al.* 1998).

For Lake Huron, the short-term data present a slightly less optimistic outlook about the decrease in fish concentrations compared with long-term trends. According to the short-term trends, the declines in Lake Huron lake trout and walleye con-

centrations between 2000–2007 will likely be 30% and 18%, respectively, with 85% and 38% of the respective bootstrap estimates suggesting the declines below the 25% reduction target (Fig. 5).

For Lake Erie, the short-term trends also reflect the conclusion from the long-term analysis that there is little possibility of achieving the GLS-2002 target (Fig. 5). The best estimated average change in the Lake Erie lake trout concentration between 2000–2007 is +0.7% (Fig. 5); however, this estimate is based on low number of the data points and therefore may be less reliable. In contrast, Lake Erie walleye concentrations are expected to increase by 38% (Fig. 5b). These results contribute further to the mixed-deteriorating state of the Lake Erie ecosystem which is also impacted by continuing introduction of non-native species, the re-emergence of oxygen-depleted areas, and the ongoing habitat degradation (Environment Canada and U.S.EPA 2003).

For Lake Ontario lake trout, the 2007 projections from the long- and short-term trends did not differ much. The short-term trend corresponded with the conclusion of the long-term trend of virtually 100% probability of Lake Ontario lake trout achieving the GLS-2002 target (Fig. 5a). In contrast, the short-term trend is less optimistic about the decrease in Lake Ontario walleye PCB levels and suggested almost no change (2.3% increase, Fig. 5b) compared to the –40% from the long-term trend (Fig. 3b). Less than 10% of the bootstrap estimated values for Lake Ontario walleye achieved the GLS-2002 targeted decrease of 25% (Fig. 5b) compared to the > 90% from the long-term trend (Fig. 3b).

We performed a Mann-Kendall test and Sen's analysis to further examine the recent PCB concentration trends (1990–2006) for lake trout from Lakes Superior, Huron, and Ontario and walleye from Lake Erie (Fig. 6). In the Mann-Kendall test, presence of a statistically significant trend is evaluated using a Z value in conjunction with a P value. A positive (or negative) Z value indicates an upward (or downward) trend whereas a lower (or higher) P value suggests higher (or lower) statistical significance of the result. Sen's nonparametric method estimates strength of the linear trend in terms of slope Q. A low positive Z value (0.23) with low positive Q value (0.001) and high P value (> 0.1) for Lake Superior lake trout suggest a very weak increasing trend in the PCB levels (Fig. 6). In comparison, high negative Z values (–2.18 and –2.49), high negative Q values (–0.017 and –0.042), low P values (< 0.05), and narrow angles between

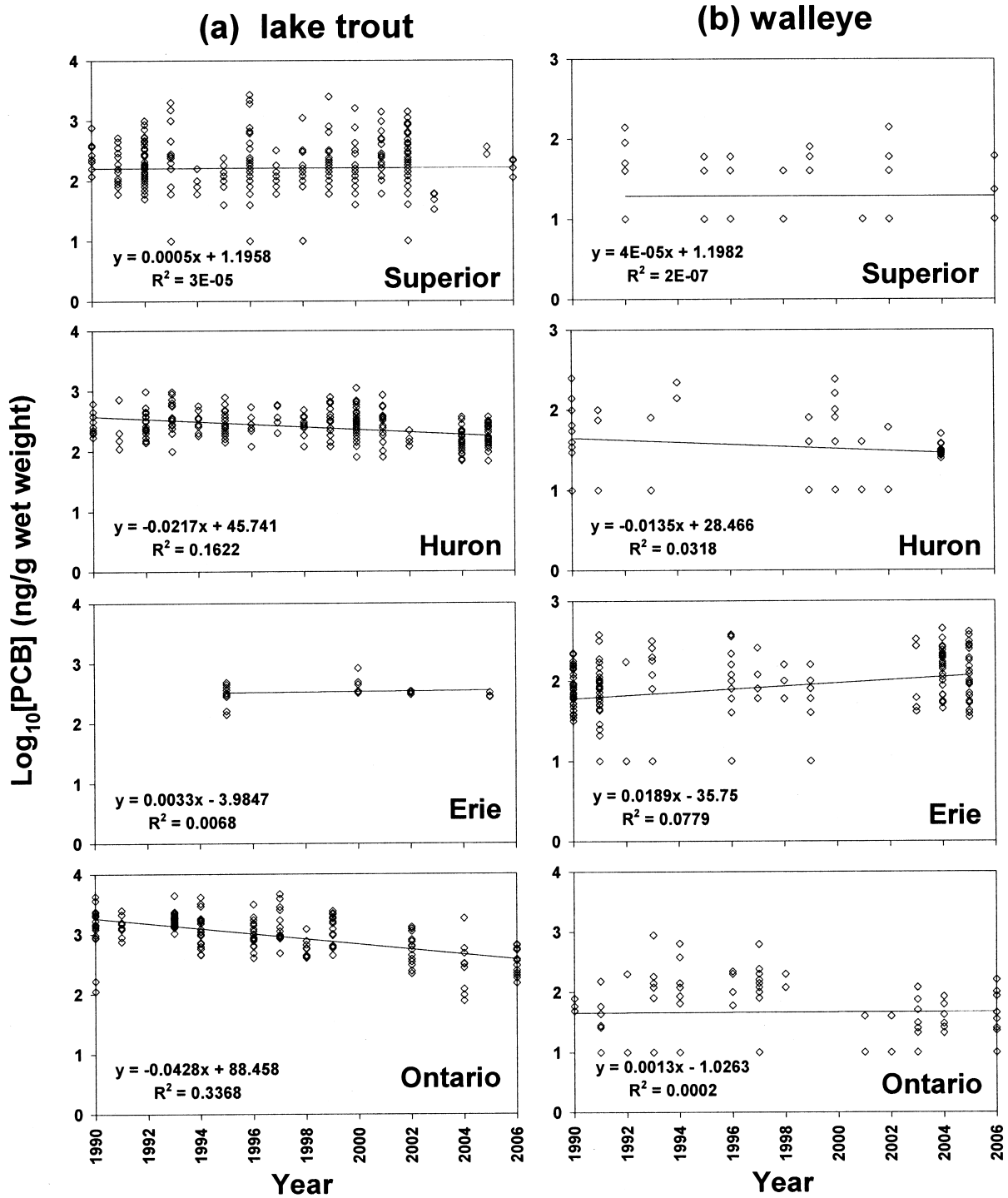## (a) lake trout (b) walleye

FIG. 4. *Recent (1990–2006) temporal trends of PCB levels (log-transformed, ng/g wet weight) in (a) 55–65 cm lake trout and (b) 45–55 cm walleye collected from Lakes Superior, Huron, Erie, and Ontario. The lines are estimated linear regressions.*
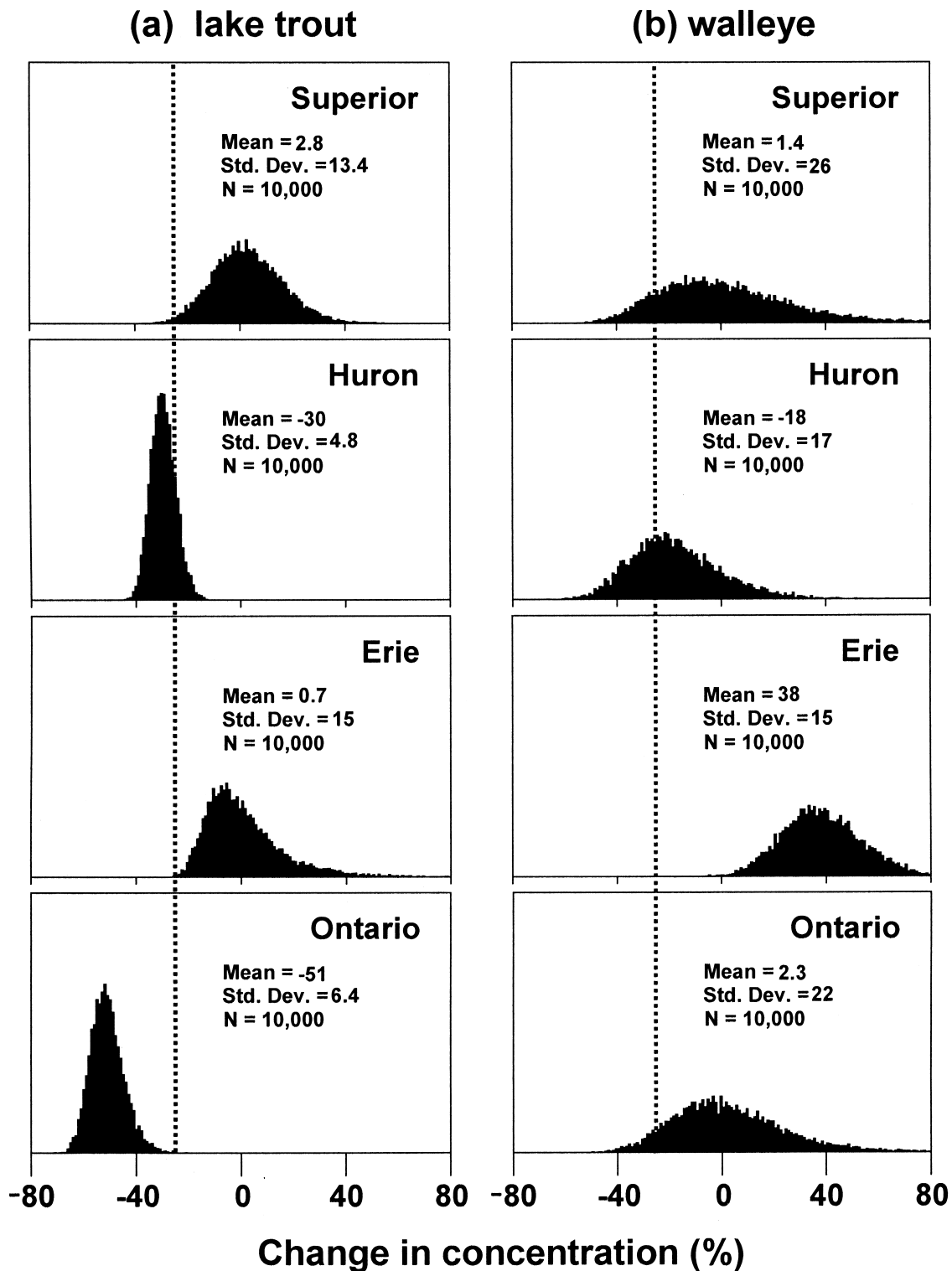
FIG. 5. Frequency distributions of the estimated percentage changes in PCB concentrations between the years 2000 and 2007. The histograms were prepared from 10,000 iterations of bootstrap resampling analysis performed on the recent (1990–2006) PCB measurements for the Canadian Great Lakes (a) lake trout and (b) walleye. Negative value indicates a decrease in the concentration. The dotted line drawn at –25% represents a key objective of the Great Lakes Strategy 2002 (U.S.EPA 2002). The area under the curve on the left side of the dotted line represents the proportion of bootstrapped estimates achieving the GLS-2002 targeted decrease of 25% in PCB levels.
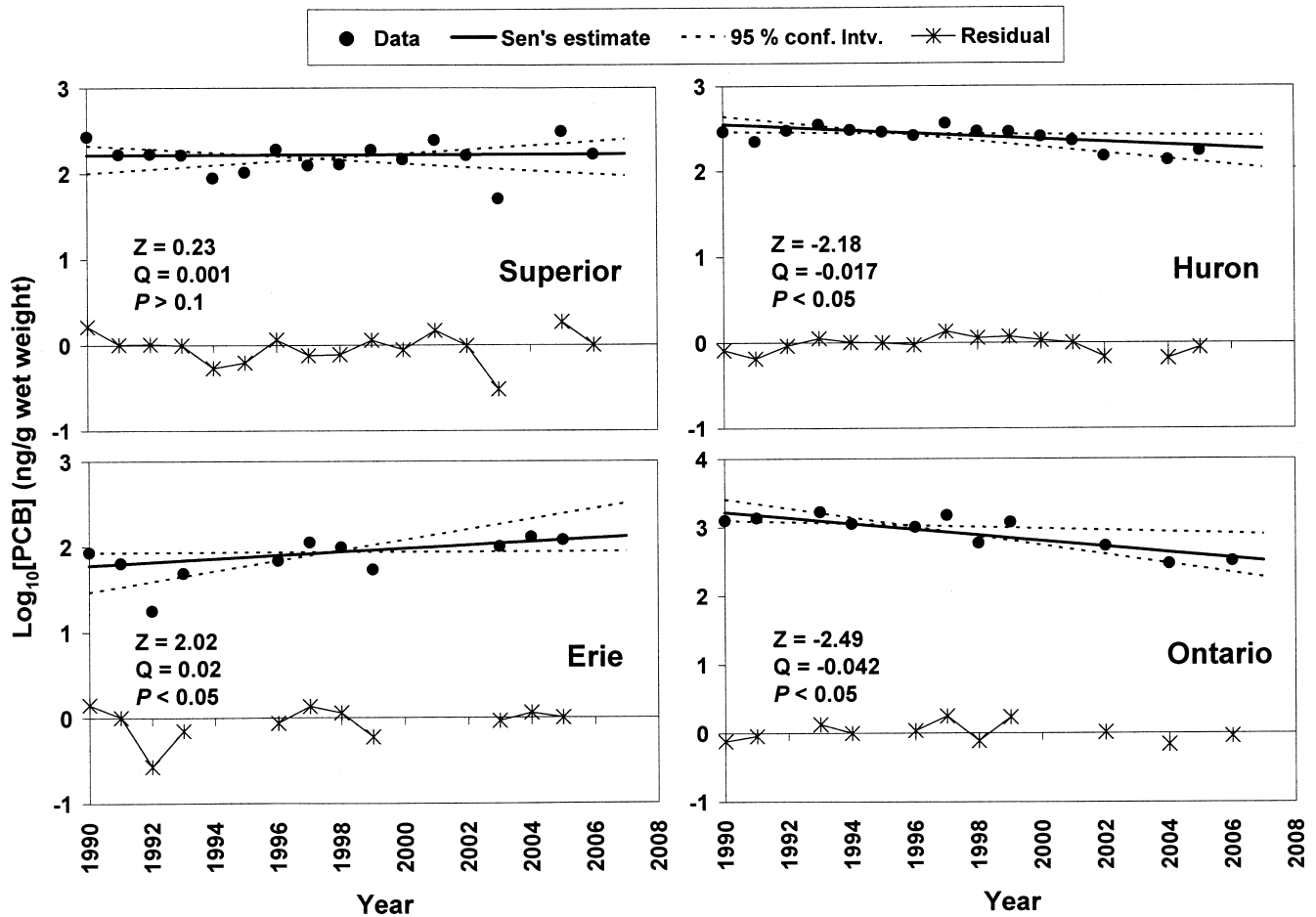
FIG. 6. *Time series (1990–2007) and Sen's estimates for annual mean values of log transformed PCB concentrations (ng/g wet weight) in lake trout (55–65 cm) from Lakes Superior, Huron, and Ontario, and walleye (45–55 cm) from Lake Erie. Residual is the difference between the Sen's estimate and data point. Z is Mann-Kendall test statistic, Q is Sen's slope, and P is level of statistical significance.*

the 95% confidence trend lines with negative slopes for lake trout from Lakes Huron and Ontario, respectively, suggest strong decreasing trends of PCB levels in these lakes. In contrast, a high positive Z value (2.02), high positive Q value (0.02), and low P value (< 0.05) suggest a strong increasing trend for Lake Erie walleye (Fig. 6). These results are in accordance with the bootstrap resampling analysis discussed above.

Although the results from analyses of the recent measurements suggest low or negligible probabilities of achieving the targeted PCB concentration decrease in walleye from Lakes Superior, Huron, and Ontario, the average projected 2007 PCB levels in these fish are 21, 27, and 50 ng/g ww, respectively (results not shown), which are below the 105

ng/g ww of *unlimited* fish consumption guideline value used by the MOE (MOE 2007). In contrast, although there is a high likelihood of achieving the 25% PCB reduction goal for lake trout from Lakes Huron and Ontario (Fig. 5a), the average projected 2007 PCB levels (160 and 370 ng/g ww, respectively; results not shown) will remain a concern. However, the decreasing trends for the fish from these lakes suggest that further improvements in these ecosystems will occur. Lake Superior lake trout concentrations may remain unchanged at the current elevated level of 160 ng/g ww. For Lake Erie, both the average projected 2007 lake trout and walleye PCB concentrations (440 and 125 ng/g ww, respectively; results not shown) as well as the increasing trends are worrisome.

If we are to evaluate the success of 25% PCB reduction goal of the GLS-2002 based on the measurements in years 2000 and 2007 only, the assessment may result in erroneous conclusions due to periodic short-term (3–6 years) oscillations observed in the concentrations. For example, French *et al.* (2006) found short-term fluctuations that related to short-term climatic cycles such as El Nino effects, and the associated impacts on prey-fish year-class strengths and the associated changes in contaminant transfer within the foodweb. Therefore, it will require a few years of measurements beyond 2007 to confirm the projected 2007 concentrations, as well as whether the concentration on an average basis decreased by 25% between 2000–2007.

In summary, trends of PCB levels in lake trout and walleye from the Canadian Great Lakes are presented using long-term biomonitoring data collected by the SFCMP of MOE. In general, PCB trends are lake and fish-species specific. The PCB fish levels appear to have leveled off in Lake Superior but are decreasing in Lake Huron. The concentrations are increasing in Lake Erie walleye. Conversely, for Lake Ontario lake trout and walleye, PCB levels are decreasing and have leveled off, respectively.

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