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A Bayesian assessment of polychlorinated biphenyl contamination of fish communities in the Laurentian Great Lakes

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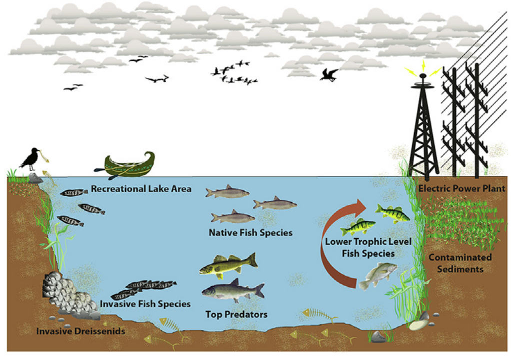
HIGHLIGHTS

- We examine PCB trends in multiple fish species across the Canadian Great Lakes.
- Lake Ontario is characterized by the most severe PCB fish contamination.
- Areas of Concern and bays receiving riverine inputs still display high PCB levels.
- PCB fish concentrations had been decreasing from the 1970s until the early 1990s.
- Deceleration of the PCB declining rates has been experienced after the mid-1990s.

GRAPHICAL ABSTRACT



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ABSTRACT

Polychlorinated biphenyl (PCB) contamination has historically posed constraints on the recreational and commercial fishing industry in the Great Lakes. Empirical evidence suggests that PCB contamination represents a greater health risk from fish consumption than other legacy contaminants. The present study attempts a rigorous assessment of the spatio-temporal PCB trends in multiple species across the Canadian waters of the Great Lakes. We applied a Bayesian modelling framework, whereby we initially used dynamic linear models to delineate PCB levels and rates of change, while accounting for the role of fish length and lipid content as covariates. We then implemented Bayesian hierarchical modelling to evaluate the temporal PCB trends during the dreissenid pre- and post-invasion periods, as well as the variability among and within the water bodies of the Great Lakes system. Our analysis indicates that Lake Ontario is characterized by the highest PCB levels among nearly all of the fish species examined. Historically contaminated local areas, designated as Areas of Concern, and embayments receiving riverine inputs displayed higher concentrations within each of the water bodies examined. The general temporal trend across the Great Lakes was that the high PCB concentrations during the early 1970s followed a declining trajectory throughout the late 1980s/early 1990s, likely as a result of the reductions in industrial emissions and other management actions. Nonetheless, after the late 1990s/early 2000s, our analysis provided evidence of a decline in the rate at which PCB concentrations in fish were dropping, accompanied by a gradual establishment of species-specific, steady-state concentrations, around which there is considerable year-to-year variability. The overall trends indicate that reduced contaminant emissions have brought about distinct beneficial changes in fish PCB concentrations, but past historical contamination along with other external or internal stressors (e.g., invasive species, climate change)

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continue to modulate the current levels, thereby posing potential risks to humans through fish consumption.

1. Introduction

The Great Lakes support a significant recreational and commercial fishing industry. In 2010, more than half of recorded fish harvest in Canada was caught in the Province of Ontario (96 million), with walleye (*Sander vitreus*) being the most popular species amongst anglers. Total direct fishing expenditures for the Province of Ontario in 2010 amounted to more than 900 million dollars (Survey of Recreational Fishing in Canada, 2010). The popularity of fishing as a leisure activity is also closely associated with the health benefits of fish consumption. Fish provide an excellent dietary source of high nutritional quality with easily digestible protein and omega-3 fatty acids (see Cohen et al., 2005; Smith and Sahyoun, 2005; Perhar et al., 2012 for reviews). A wide array of work has been published relating dietary fatty acids to the maintenance of cognitive and nervous system functioning (Minokoshi et al., 2002), and to the improvement of hormonal imbalances and insulin resistance complications (Yamauchi et al., 2001). Longer chain omega-3 fatty acids may also be important in preventing chronic health conditions, such as Alzheimer's disease, type II diabetes, kidney disease, rheumatoid arthritis, high blood pressure, coronary heart disease, alcoholism, and possibly cancer (Mozaffarian and Rimm, 2006). As a result of these advantageous health effects, the American Heart Association provides strong endorsement for regular fish consumption (Oken et al., 2003).

The Great Lakes have experienced varying degrees of pollution impacts, ranging from excessive nutrient loading to toxic contaminant exposure and (more recently) the onslaught of climate change and invasive species. Among the bioaccumulative, toxic and persistent organic pollutants, PCBs are of particular concern and historically have restricted the use of valuable commercial and recreational fisheries in the Great Lakes. PCBs were initially manufactured in the late 1920s by the electric industry as non-flammable additives to oils and industrial fluids, with the purpose of acting as coolers and electrical insulators (Hornbuckle et al., 2006). In their early history, PCBs posed a particular concern among electric utility workers, as excessive occupational exposure could lead to an increase in liver cirrhosis and to the development of malignant melanomas (Loomis et al., 1997; Prince et al., 2006). PCBs make up a group of 209 chlorinated congeners that are highly hydrophobic and therefore tend to tightly deposit into fatty tissues (Elskus et al., 2005). As persistent organic pollutants, PCBs are non-ionizable and largely non-polar, while their hydrophobic nature leads to their accumulation in fatty tissues and in oil rich organs and glands (Elskus et al., 2005). PCBs are known to suppress immune system function (Dallaire et al., 2003), to increase the risk of diabetes (Codru et al., 2007), to cause cardiovascular problems including coronary heart disease (Tomasallo et al., 2010), to be responsible for decreased verbal learning and increased depression (Fitzgerald et al., 2008), neurobehavioral alterations, motor immaturity, hyporeflexia, and lower psychomotor scores (Faroon et al., 2000), and to disrupt thyroid stimulating and sex steroid hormone functions (Turyk et al., 2007).

The presence of PCBs in Great Lakes fish samples was first detected in 1968, in lake trout (*Salvelinus namaycush*) and bloater (*Coregonus hoyi*) chubs caught in Lake Michigan (Veith, 1968). Depending on their trophic position, fish are typically exposed to PCBs through three routes with variant relative importance, i.e., gills, epithelial/dermal tissues, and gastrointestinal tract (Schlenk, 2005). Lower trophic level fish likely receive PCBs by the diffusion process through gills and epithelial cells, whereas top predators mainly receive them through dietary uptake of contaminated food. Responding to increased public pressure for effective elimination of persistent toxic pollutants from the Great Lakes, PCB production was banned in North America in the 1970s. The Great Lakes Water Quality Agreement (GLWQA) between Canada and USA was signed in 1972 and was subsequently revised in 1978 and 2012. The agreement aimed at identifying the spatiotemporal trends of toxic substances in sediments and biota with ultimate goal to protect the environmental integrity of the Great Lakes (International Joint Commission (IJC), 1978, 2006). Contaminant levels in edible fish portions have been monitored, from the 1970s onwards. Species-specific consumption advisories have been issued for the general and sensitive demographic groups by the Ontario Ministry of the Environment, Conservation & Parks (MECP), U.S. Great Lakes states and tribes. Monitoring of fish contamination levels has raised public awareness of potential health risks and has encouraged governmental action. The Government of Ontario introduced the Toxic Reduction Act aiming *(i)* to reduce the use and creation of toxic substances in regulated facilities and *(ii)* to inform Ontarians about toxic substances in the environment through a public open data policy of the facilities operating under the program (https://www.ontario.ca/page/eating-ontario-fish-2017-18).

Implementation of these regulatory actions resulted in reduced levels of most contaminants in Great Lakes fish through the 1980s, but the rate of decrease is reported to have diminished or to have leveled off since the early 1990s (Bhavsar et al., 2007; Sadraddini et al., 2011a,b; Visha et al., 2015). The reasons for these trends are not fully known, but existing mechanistic explanations include the food web alterations induced from invasive species in the Great Lakes (Hogan et al., 2007), as well as shifts in trophodynamics associated with global warming (French et al., 2006). Despite the valuable insights gained into contaminant dynamics through the extensive datasets developed, many studies failed to consider important factors that can modulate the inference drawn, such as fish age, size, trophic level, seasonality, growth and lipid content (Mahmood et al., 2013a,b).Variations across monitoring programs in the type of sampling procedures and different statistical methods used may also impede the robust assessment of contaminant trends (Gewurtz et al., 2011). As a result it is important to develop flexible statistical frameworks, in order to determine correct contamination trends.

To this end, a central feature of recent work in Lakes Erie and Ontario was the adoption of Bayesian inference techniques as a means for critically assessing spatiotemporal contaminant trends in fish communities over the last four decades (Lamon et al., 1998; Azim et al., 2011a,b; Sadraddini et al., 2011a,b; Mahmood et al., 2013a,b; Visha et al., 2015, 2016). The advantages of the Bayesian approach when addressing ecological questions primarily stem from its ability to explicitly accommodate model structural and parametric uncertainty, measurement errors, and data gaps (Dorazio and Johnson, 2003; Ellison, 1996, 2004; Arhonditsis et al., 2007; Blukacz-Richards et al., 2017). The primary goal of the present study was to introduce robust statistical methodologies, such as dynamic linear and Bayesian hierarchical modelling, to address several important questions and issues related to the presence of PCBs in the Great Lakes basin. Our aim is to address the following questions: What are the spatio-temporal PCB trends in fish species with different trophic positions and functional roles in the food web of the Great Lakes? Do contaminant trends differ between the pre- and post-invasion periods (before and after 1995) of dreissenids and round goby in the Great Lakes? How do these trends differ within and among the different water bodies? Eleven fish species spanning multiple trophic levels were selected due to their importance to local anglers and fishing industry. Our study consists of seven major water bodies representing the Canadian portion of the Great Lakes basin: Lake Superior, North Channel, Georgian Bay, Lake Huron main basin, Lake Erie, Lake Ontario, and St. Lawrence River. Not only does our study aim to provide a concrete and complete analysis of PCB contamination trends in the Great Lakes, but it will also shed light on the degree of contamination in Areas of Concern (AOCs) that have not been delisted due to beneficial use impairments associated with their respective fish communities.

2. Methods

Fish Dataset: The fish data used in our study were collected by the MECP Fish Contaminant Monitoring Program. Our analysis is based on dorsal spineless-boneless fillets, which have been extensively used to develop fish consumption advisories. We selected eleven fish species representing different trophic levels of the food web across the lakes as well as playing an important role for recreational and subsistence fisheries. The eleven fish species examined are walleye (*Sander vitreus*), lake trout (*Salvelinus namaycush*), Coho salmon (*Oncorhynchus kisutch*), common carp (*Cyprinus carpio*), freshwater drum (*Aplodinotus grunniens*), lake whitefish (*Coregonus clupeaformis*), yellow perch (*Perca flavescens*), Chinook salmon (*Oncorhynchus tshawytscha*), brown bullhead (*Ameiurus nebulosus*), cisco or lake herring (*Coregonus artedi*), and bloater (*Coregonus hoyi*). All samples were collected from several locations across the Canadian waters of the Great Lakes (Fig. 1). In Fig. SI 1, we also show all 43 Areas of Concerns around the Great Lakes basin. Several of our study sites were these locations, thereby allowing to evaluate the importance of local remedial actions over the past four decades as a confounding factor that shapes the established trends. The number of years sampled varied significantly per fish species and location, but our study overall spans thirty-eight years (1975–2013).

Dynamic linear modelling: We developed a series of Dynamic Linear Models (DLMs) to delineate the temporal PCB trends, while explicitly accounting for the fact that fish length and lipid content typically co-vary with contaminant concentrations. Counter to static regression models that have fixed parameters, our DLMs have an evolving structure (year-specific regression coefficients) that allows parameters to shift through time, whereby non-monotonic patterns in time can be accommodated. An important property of same the strategy is the explicit recognition of structure in the time series; there is a sequential ordering of the data and at each time step the level of the response variable is related to its level at earlier time steps. DLM posterior estimates are influenced by prior and current information (not subsequent data), which is another distinct feature relative to traditional regression analyses [See also Visha et al. (2015) for detailed discussion of the technical strengths of DLMs.]. For the purpose of the present exercise, we pooled together data from all the locations within each of the seven water bodies (Lake Superior, North Channel, Georgian Bay, Lake Huron, Lake Erie, Lake Ontario, and St. Lawrence River) in order to characterize the corresponding temporal PCB trends during the study period. The main DLM components for a given water body are the observation equation and associated system equations.

Observation equation:

$$\ln[PCB]_{it} = level_t + \beta_{1t}\ln[length_{it}] + \beta_{2t}\ln[lipid_{it}] + \psi_{it}$$
$$\psi_{it} \sim N\,[0, \Psi_t] \tag{1}$$

System equations:

$$level_t = level_{t-1} + rate_t + \omega_{1t}\quad \omega_{1t} \sim N\,[0,\Omega_{1t}] \tag{2}$$

$$rate_t = rate_{t-1} + \omega_{2t}\quad \omega_{2t} \sim N\,[0,\Omega_{2t}] \tag{3}$$

$$\beta_{1t} = \beta_{1t-1} + \omega_{3t}\quad \omega_{3t} \sim N\,[0,\Omega_{3t}] \tag{4}$$

$$\beta_{2t} = \beta_{2t-1} + \omega_{4t}\quad \omega_{4t} \sim N\,[0,\Omega_{4t}] \tag{5}$$

$$1/\Omega_{jt}^2 = \zeta^{t-1}\cdot 1/\Omega_{j1}^2,\ 1/\Psi_t^2 = \zeta^{t-1}\cdot 1/\Psi_1^2\ t > 1 \text{ and } j = 1 \text{ to } 4$$

$$level_1,\ rate_1,\ \beta_{11},\ \beta_{21} \sim N(0{,}10000)\ t = 1$$

$$1/\Omega_{j1}^2,\ 1/\Psi_1^2 \sim G(0.001{,}0.001)$$

where $ln[PCB]_{it}$ is the observed *PCB* concentration at time t in the individual sample i; $level_t$ is the mean *PCB* concentration at time t when accounting for the covariance with fish length and lipid content; $\ln[length_{it}]$ is the observed (standardized) fish length at time t in the individual sample i; $\ln[lipid_{it}]$ is the observed (standardized) fish lipid content; $rate_t$ is the rate of change of the level parameter; β_{1t} is a length (regression) coefficient; β_{2t} is a lipid (regression) coefficient; ψ_t, ω_{jt} are the error terms for year t sampled from normal distributions with zero mean and variances Ψ_t^2, Ω_{tj}^2, respectively; the discount factor ζ represents the aging of information with the passage of time. The aging of information in a discount factor is such that older observations are weighted less than newer ones. The discounted posterior becomes the prior for the next time step, and the process is repeated. In this study, we selected a discount factor equal to 0.95, which provided the optimal balance between model performance and the degree of identification (signal-to-noise ratio) of parameters; see Visha et al. (2015) for detailed discussion on the selection of the discount factor value. *N(0, 10000)* is the normal distribution with mean 0 and variance 10000; and *G(0.001, 0.001)* is the gamma distribution with shape and scale parameters of 0.001. The prior distributions for the parameters of the initial year $level_1$, $rate_1$, β_{11}, β_{21}, $1/\Omega_{j1}^2$, and $1/\Psi_1^2$ are considered non-informative. Inference regarding the likelihood of decreasing PCB trends over the span of three decades, i.e., 1980s, 1990s, and 2000s, was based on the odds ratios of the posterior estimates of the rates of change (*rate* parameter) as derived from our DLM analysis. The odds ratio of the rate parameter being below zero in a particular year is the ratio of the probability mass below zero to the mass above zero. The magnitude of the trends were classified such that values > 4 (>80% probability of decrease), 2–4 (66–80% probability of decrease), and 1–2 (50–66% probability of decrease) were indicative of a strong, medium, and weak decrease, whereas odds ratio values < 1 (<50% probability of decrease) are suggestive of a likely increase in the fish contamination levels. Moreover, the exponentiated posterior values of the *rate* parameter offered estimates of the percentage change per year.

Hierarchical modelling framework: Bayesian hierarchical modelling was used to detect the temporal PCB trends in fish, while explicitly accommodating the variability among the water bodies and sampling locations (Cheng et al., 2010). A major strength of the

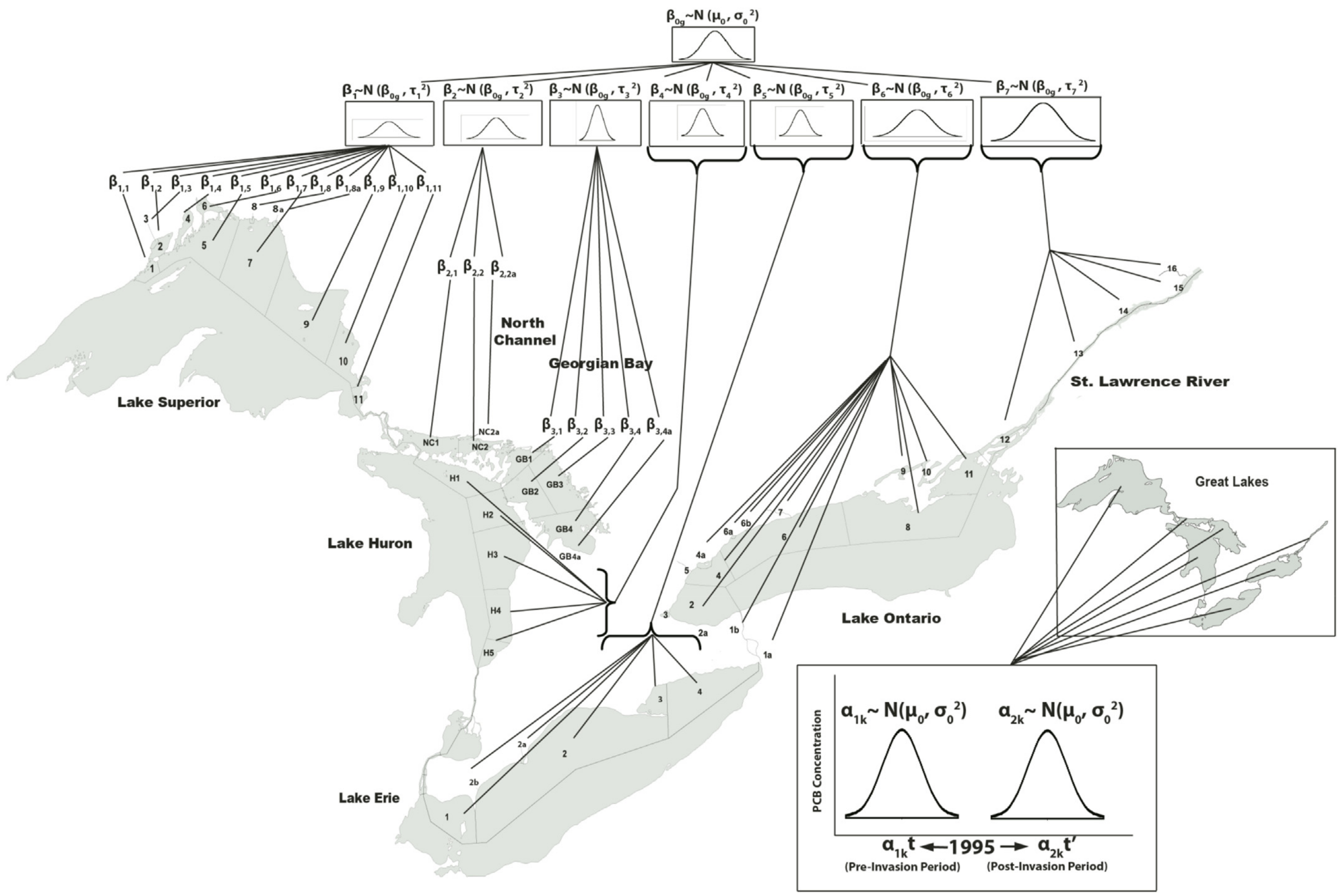

Fig. 1. Bayesian hierarchical modelling framework and fish sampling locations in the Great Lakes. The three hierarchical levels represent (i) the location-specific intercepts nested within the seven water bodies, (ii) the water body-specific intercept terms and the rates of change related to the water body-specific PCB trends before or after the invasion of exotic species, (iii) the global parameters (or hyperparameters) that capture the entire Great Lakes basin. Our hierarchical structure also comprises the fixed-effect parameters: year-specific intercepts, length- and lipid-regression coefficients.

hierarchical strategy is the ability to overcome problems of insufficient local data by "borrowing strength" from well-studied sites. That is, the hierarchical framework offers the ability to transfer information across sites and support predictions in locations with few observations and limited observational range (Shimoda and Arhonditsis, 2015). Because of the latter advantage, we were able to disaggregate the data and shed light on fish contamination trends within each of the water bodies of the Great Lakes system for all the fish species examined. Our hierarchical framework also considers the potential impact of the invasion of dreissenid mussels along with the covariance of PCB levels with fish length and lipid content. The mathematical notation for the hierarchical model is summarized as follows:

$$log_e(PCB_{mod})_{ijkt} = \beta_G + \beta_{Lj(k)} + \beta_{Wk} + \beta_{Yt} + a_{1k}\,t' + a_{2k}t'' + \beta_1 \text{In}[\text{length}_{ijkt}] + \beta_2 \text{In}[\text{lipid}_{ijkt}] + \psi_{ijkt} \quad (6)$$

$$\text{if } t \le 1995 \text{ then } t' = t_{ijk} - t_0 \quad (7)$$

$$\text{if } t > 1995 \text{ then } t'' = t_{ijk} - 1995 \quad (8)$$

$$\sum_{j=1}^{Mk} \beta_{Lj(k)} = 0 \sum_{k=1}^{K} \beta_{Wk} = 0 \sum_{t=1}^{T} \beta_{Yt} = 0 \quad (9)$$

$\beta_G \sim N(\beta_0, \sigma_0^2)$, $\beta_{Lj(k)} \sim N(\beta_{L0(k)}, \sigma_{L0(k)}^2)$, $\beta_{Wk} \sim N(\beta_{W0}, \sigma_{W0}^2)$, $\beta_{Yt} \sim N(\beta_{Y0}, \sigma_{Y0}^2)$

$a_{1k} \sim N(a_{10}, \sigma_{a10}^2)$, $a_{2k} \sim N(a_{20}, \sigma_{a20}^2)$

$\beta_0 \sim N(\mu_0, \tau_0^2)$, $\beta_{L0(k)} \sim N(\mu_{L0(k)}, \tau_{L0(k)}^2)$, $\beta_{W0} \sim N(\mu_{W0}, \tau_{W0}^2)$, $\beta_{Y0} \sim N(\mu_{Y0}, \tau_{Y0}^2)$

$a_{10} \sim N(\mu_{a10}, \tau_{a10}^2)$, $a_{20} \sim N(\mu_{a20}, \tau_{a20}^2)$

$\mu_0 \sim N(0{,}10000)$, $\mu_{L0(k)} \sim N(0{,}10000)$, $\mu_{W0} \sim N(0{,}10000)$, $\mu_{Y0} \sim N(0{,}10000)$

$\mu_{a10} \sim N(0{,}10000)$, $\mu_{a20} \sim N(0{,}10000)$

$\tau_0^{-2} \sim G(0.001, 0.001)$, $\tau_{L0(k)}^{-2} \sim G(0.001, 0.001)$, $\tau_{W0}^{-2} \sim G(0.001, 0.001)$, $\tau_{Y0}^{-2} \sim G(0.001, 0.001)$

$\tau_{a10}^{-2} \sim G(0.001, 0.001)$, $\tau_{a20}^{-2} \sim G(0.001, 0.001)$

$\sigma_0^{-2} \sim G(0.001, 0.001)$, $\sigma_{L0(k)}^{-2} \sim G(0.001, 0.001)$, $\sigma_{W0}^{-2} \sim G(0.001, 0.001)$, $\sigma_{Y0}^{-2} \sim G(0.001, 0.001)$

$\sigma_{a10}^{-2} \sim G(0.001, 0.001)$, $\sigma_{a20}^{-2} \sim G(0.001, 0.001)$

$\beta_1 \sim N(0{,}10000)$, $\beta_2 \sim N(0{,}10000)$

$\psi_{ijkt} \sim N(0, \sigma_\varepsilon^2), \sigma_\varepsilon^{-2} \sim G(0.001, 0.001)$

$i = 1 \ldots. N_j, j = 1 \ldots M_k, k = 1 \ldots K, t = 1 \ldots T$

where $log_e(PCB_{mod})_{ijkt}$ represents the log-transformed modeled PCB concentration in sample *i*, collected from location *j*, water body *k*, and year *t*; β_G is an overall intercept term; $\beta_{Lj(k)}$ is a location-specific intercept term for all sampling sites *j* nested within the water body *k;* β_{Wk} is a water body-specific intercept term; β_{Yt} is a year-specific intercept term; a_{1k} and a_{2k} are the regression coefficients (or rates of change) related to the water body-specific PCB trends before or after the invasion of exotic species (e.g., dreissenids, round goby), respectively; β_1, β_2 denote the length- and lipid-regression coefficients, respectively; ψ_{ijkt} represents the model error term which is a draw from a normal distribution with a mean equal to zero and error variance σ_ε^2; β_0, $\beta_{L0(k)}$, β_{W0}, β_{Y0}, a_{10}, a_{20} are the global parameters for the global, location-, water body-, year-specific intercept terms, and the PCB rates of change before and after the dreissenid invasion; σ_0^2, $\sigma_{L0(k)}^2$, σ_{W0}^2, σ_{Y0}^2, σ_{a10}^2, σ_{a20}^2 are the variances of the corresponding global parameters; μ_0, $\mu_{L0(k)}$, μ_{W0}, μ_{Y0}, μ_{a10}, μ_{a20} are the means of the hyperparameters; τ_0^2, $\tau_{L0(k)}^2$, τ_{W0}^2, τ_{Y0}^2, τ_{a10}^2, τ_{a20}^2 are the respective variances of the hyperparameters; N_j, M_k, K, and T are the total number of samples in location *j*, the number of sampling locations per water body, the number of water bodies, and the total number of years, respectively; t_0 is the initial year of the analysis, and 1995 is assumed to be the breakpoint for examining distinct changes in the rates of change of PCB levels; *N*(0, 10000) is the normal distribution with mean 0 and variance 10000, and *G*(0.001, 0.001) is the gamma distribution with shape and scale parameters of 0.001. These prior distributions are considered "non-informative" or vague. Finally, it should be noted that the location-, waterbody-, and year-specific intercept terms are constrained to have a zero sum to make the model identifiable.

3. Results

Lake trout (6.3–10.9%), bloater (8.2–8.4%), common carp (5.2–8.0%), and lake whitefish (3.8–8.3%) were characterized by the highest mean lipid content values among the eleven species of our analysis (Table 1). Interestingly, the highest average lipid contents for lake trout and lake whitefish were recorded in Lake Erie, while bloater and common carp had their highest levels in Lake Huron and St. Lawrence River, respectively. By contrast, yellow perch and walleye had the lowest lipid contents among the study species examined, with the lowest mean lipid content values recorded in St Lawrence River (0.6 ± 0.4%) and North Channel (0.9 ± 0.6%), respectively. Fish lipid values in the Lower Great Lakes, tend to be significantly higher than those in the Upper Great Lakes. In Lake Erie, the lipid values for lake trout and lake whitefish have also been shown to be higher in recent years, 2000–2008 (Neff et al., 2012). Chinook salmon and common carp were characterized by the longest mean length values among all of the fish species examined, reaching a peak of 80.3 ± 17.4 cm and 71.9 ± 9.3 cm in Lake Ontario and the St. Lawrence River, respectively. By contrast, yellow perch (21.2–27.9 cm) and bloater (26.9–27.9 cm) displayed the shortest mean body lengths among the eleven fish species examined. Although several fish species (Chinook salmon, lake whitefish, and common carp) were characterized by relatively high PCB concentrations, the highest mean contaminant values were recorded in lake trout samples from Lake Ontario (1994 ± 1989 ng/g wet weight or ww). Chinook salmon (1323 ± 1323 ng/g ww) and common carp (897 ± 1284 ng/g ww) similarly had the highest PCB concentrations among the Lake Ontario fishes included in this study, although the concentrations of the latter species were even higher in the St. Lawrence River (1174 ± 1168 ng/g ww). By contrast, the average PCB concentrations in yellow perch were consistently <100 ng/g ww, with the only exception being the levels in Lake Ontario (111 ± 252 ng/g ww). Likewise, walleye displayed the lowest PCB concentrations in Lake Superior (39 ± 43 ng/g ww), North Channel (53 ± 74 ng/g ww), and Lake Huron main basin (123 ± 279 ng/g ww), whereas the highest PCB concentrations in walleye were recorded in Lake Ontario (209 ± 380 ng/g ww) and St. Lawrence River (227 ± 535 ng/g ww).

According to our hierarchical modelling exercise, the water body-specific intercept (β_W) terms were consistent with the aforementioned spatial PCB patterns (Fig. 2). In particular, the posterior β_W values are on par with the higher PCB levels in Lake Ontario, relative to the rest of the water bodies, with nearly all of the fish species examined. Fairly high posterior β_W values were also derived for St Lawrence River and Lake Erie; especially for walleye and yellow perch. By contrast, our hierarchical model assigned low β_W values to fish species collected from Lake Superior, North Channel, Lake Huron main basin, and Georgian Bay, indicative of a distinct bimodal pattern between the northern and southern portion of the Great Lakes system. Consistent with the measured concentrations, bloater and lake herring displayed the highest β_W values in the main basin of Lake Huron relative to Georgian Bay and Lake Superior.

The location-specific parameters (β_L) offered insights into the spatial patterns of fish contamination within each of the water bodies examined. Specifically, historically industrialized urban areas, designated as AOCs, and embayments receiving riverine inputs displayed high concentrations across all species (Fig. SI 2, Table SI 2). For example, walleye, lake whitefish, and Chinook salmon were generally characterized by higher β_L values in western Lake Ontario (Sites #1 and #2; Queenston to Whirlpool, Lower Niagara River mouth to Lake Ontario, and Niagara Bar), including the Hamilton Harbour AOC (Site #3; Table SI 2), while Chinook salmon also demonstrated elevated PCB levels in Ganaraska River, Main Duck Island, and the area east of Glenora to Kingston (Sites #7 and #9). In Lake Erie, the western basin, including the area surrounding the East Sister Island (Site #1), displayed the highest β_L values. In Lake Superior, walleye was primarily characterized by relatively higher PCB levels in Peninsula Harbour (Site #7), followed by Aquasabon River Mouth, Jackfish Bay, Mobeley Bay, and the Goulais Bay area (Sites #5, #6, and #8). The same pattern held true with lake whitefish, although the highest values were registered in the Peninsula Harbour, South of Marathon (Site #9). Chinook salmon displayed their highest concentrations in samples collected from Thunder Bay, and Inner Harbour (Site #2).

Based on the posterior values of the year-specific (β_Y) intercept terms, we can infer that the general pattern across the Great Lakes was that the high PCB concentrations during the early 1970s preceded a declining trajectory throughout the late 1980s/early 1990s. Reductions in industrial emissions and other management actions, might be responsible for these early declining trends which gradually stabilized after the late 1990s/early 2000s (Fig. 3). The declining trend was more pronounced with lake trout, yellow perch, lake whitefish, and Chinook salmon, whereas the posterior β_Y coefficients for Coho salmon and walleye suggest that their PCB levels exhibit significant year-to-year variability after their original (nearly monotonic) decrease until the early-1990s. Likewise, common carp and freshwater drum were subjected to wax-and-wane cycles with a weaker evidence for a long-term declining trend.

Deviations from the Great Lakes-wide pattern were found in individual water bodies, as depicted by both the water body-specific rates of change of our hierarchical (a_1 and a_2 parameters in Fig. 4) and dynamic linear (*rate* parameter in Fig. 5) models. In particular, the response of walleye to the variability of various stressors (e.g., external emissions, invasive species) was minimal in

Table 1
Descriptive statistics (mean ± standard deviation) for PCB concentrations (ng/g wet weight), lipid content, and length for the eleven fish species and seven water bodies of the Great Lakes system.

Species	Location	N	Concentration	Length	Lipid
Walleye	Lake Superior	182	39 ± 43	46.7 ± 9.2	1.1 ± 0.9
	North Channel	195	53 ± 74	46.6 ± 9.6	0.9 ± 0.6
	Georgian Bay	219	123 ± 279	56.1 ± 12.0	1.0 ± 0.5
	Lake Huron	306	96 ± 125	49.9 ± 9.5	1.4 ± 1.9
	Lake Erie	1115	115 ± 114	52.4 ± 8.7	1.7 ± 1.3
	Lake Ontario	722	209 ± 380	52.1 ± 13.3	1.6 ± 3.2
	St. Lawrence River	443	227 ± 535	52.3 ± 10.3	1.1 ± 0.9
Lake Trout	Lake Superior	2149	505 ± 821	55.5 ± 10.2	8.5 ± 7.5
	North Channel	142	194 ± 134	56.3 ± 7.7	6.3 ± 3.2
	Georgian Bay	303	227 ± 172	59.7 ± 11.9	7.5 ± 4.3
	Lake Huron	605	560 ± 626	59.1 ± 10.9	7.1 ± 4.2
	Lake Erie	142	458 ± 271	58.0 ± 12.5	10.9 ± 4.8
	Lake Ontario	904	1994 ± 1989	62.4 ± 10.6	10.0 ± 5.0
Yellow Perch	Lake Superior	64	23 ± 8	27.9 ± 4.5	0.8 ± 0.7
	North Channel	115	22 ± 20	25.2 ± 3.5	0.8 ± 0.3
	Georgian Bay	154	31 ± 34	23.8 ± 4.5	0.9 ± 2.4
	Lake Huron	226	64 ± 149	24.5 ± 5.6	0.8 ± 0.3
	Lake Erie	438	71 ± 103	24.3 ± 4.8	0.8 ± 0.6
	Lake Ontario	684	111 ± 252	21.2 ± 4.3	1.2 ± 1.7
	St. Lawrence River	521	46 ± 83	22.1 ± 3.3	0.6 ± 0.4
Lake Whitefish	Lake Superior	893	227 ± 735	50.0 ± 6.9	5.1 ± 4.4
	North Channel	129	52 ± 43	51.7 ± 6.9	4.5 ± 3.6
	Georgian Bay	196	110 ± 132	54.7 ± 8.5	5.2 ± 4.3
	Lake Huron	514	125 ± 164	49.7 ± 8.1	3.8 ± 3.3
	Lake Erie	421	230 ± 179	50.0 ± 6.7	8.3 ± 5.8
	Lake Ontario	171	373 ± 528	50.6 ± 7.5	4.8 ± 4.0
Chinook Salmon	Lake Superior	264	225 ± 297	63.6 ± 17.3	2.6 ± 2.9
	North Channel	96	290 ± 274	69.0 ± 11.6	1.9 ± 2.2
	Georgian Bay	173	360 ± 254	79.5 ± 14.4	2.4 ± 2.2
	Lake Huron	339	521 ± 517	68.9 ± 18.3	2.5 ± 2.2
	Lake Ontario	1215	1323 ± 1323	80.3 ± 17.4	3.0 ± 3.2
Brown Bullhead	Lake Erie	82	48 ± 59	28.9 ± 4.4	1.2 ± 0.8
	Lake Ontario	597	236 ± 542	29.4 ± 4.6	1.8 ± 1.3
	St. Lawrence River	258	260 ± 744	27.2 ± 3.8	1.3 ± 0.8
Coho Salmon	Lake Superior	81	56 ± 29	53.1 ± 5.6	1.6 ± 0.8
	Lake Erie	635	468 ± 253	55.8 ± 12.6	4.0 ± 2.7
	Lake Ontario	710	1551 ± 1245	66.5 ± 11.8	3.0 ± 2.4
Common Carp	Georgian Bay	143	799 ± 1077	65.7 ± 10.9	6.2 ± 5.3
	Lake Huron	432	1332 ± 1602	61.3 ± 9.6	6.6 ± 6.0
	Lake Erie	382	723 ± 1110	59.6 ± 10.6	6.3 ± 5.2
	Lake Ontario	472	897 ± 1284	60.4 ± 14.3	5.2 ± 5.1
	St. Lawrence River	195	1174 ± 1168	71.9 ± 9.3	8.0 ± 6.3
Freshwater Drum	Lake Erie	462	224 ± 213	36.7 ± 7.4	3.7 ± 4.0
	Lake Ontario	249	350 ± 551	43.7 ± 8.4	3.2 ± 3.4
Cisco (Lake Herring)	Lake Superior	192	204 ± 448	36.4 ± 5.3	5.0 ± 3.9
	Lake Huron	86	372 ± 453	39.7 ± 4.6	5.4 ± 3.2
Bloater	Georgian Bay	185	342 ± 452	27.9 ± 3.9	8.2 ± 3.7
	Lake Huron	222	337 ± 286	26.9 ± 3.2	8.4 ± 5.3

Lake Superior and Lake Huron, and thus the corresponding PCB levels were relatively stable at low levels throughout our study period. In St. Lawrence River, walleye demonstrated fairly high PCB levels with no apparent decline in the 1980s, which began to decrease more rapidly after the mid-1990s (see also Fig. SI 3). Lake trout from Lake Ontario displayed the most distinct decline in their PCB levels, when compared to the declining trends recorded in other waterbodies (Fig. SI 3). The PCB concentrations in yellow perch remained relatively stable in Lake Superior, North Channel, and Georgian Bay over time, whereas the same fish species in the main basin of Lake Huron, Lake Erie, Lake Ontario, and St. Lawrence River showed a rapid decrease in response to the curtailed emissions during the late 1970s (Fig. SI 3). Lake whitefish, an important commercial and recreational fish species, displayed weakly decreasing trends, after 1995 in Georgian Bay, Lake Erie and Lake Ontario, whereas the corresponding mean PCB concentrations remained relatively unaltered in Lake Erie throughout our study period. PCBs in salmonid fish species (Chinook and Coho salmon) were characterized by a constant decline in Lake Huron and Lake Ontario, whereas the PCB patterns of Chinook salmon in Lake Superior, North Channel, and Georgian Bay provided evidence of wax-and-wane cycles in their tissue content. PCB levels in brown bullhead remained fairly stable after the mid-1980s in Lake Ontario, and St. Lawrence River (Fig. SI 3). Similarly, common carp was also characterized by relatively stable PCB concentrations, with a noticeable decline in St. Lawrence River fishes and less so in Lake Erie only after the mid-2000s. Freshwater drum displayed weakly declining trends in Lake Erie, but was subjected to greater year-to-year variability in Lake Ontario. The recent PCB levels in cisco were distinctly lower in Lake Superior and Lake Huron relative to those registered in the early 1980s. Bloater trends are suggestive of a recent PCB increase in Georgian Bay and less so in Lake Huron.

Regarding the likelihood of an increase in fish PCB concentrations during the post-dreissenid invasion period, our analysis showed that there was some evidence of increased bioaccumulation for yellow perch, common carp and bloater from Georgian Bay (Figs. 4 and 5). In particular, the probability for yellow perch to have experienced a PCB increase in their tissues was

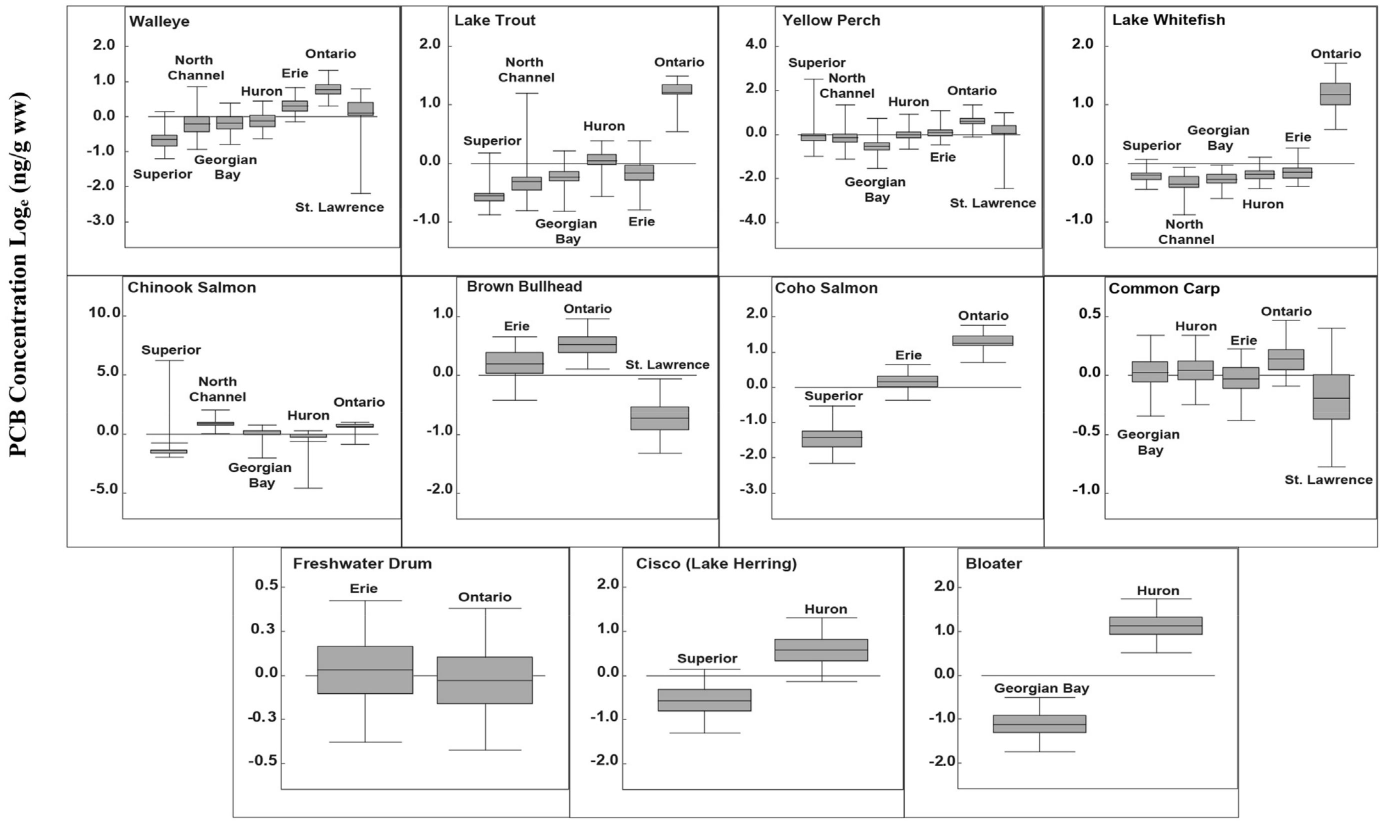

Fig. 2. Box-and-whisker plots depicting the posterior samples for the water body specific intercept terms of the Bayesian hierarchical model. The whisker edges of the box plots represent the 2.5% and 97.5% percentiles, the solid black line at the approximate centre of each box represents the posterior mean values, and the lower and upper edges of each box correspond to 25% and 75% percentiles.

approximately 58%, while the likelihood of increase for common carp, and bloater was 59%, and 70%, respectively. To put these values into perspective, we note that the probability values of a declining trend during the 1990s in yellow perch, common carp, and bloater were 60%, 64%, and 72%, respectively. In Lake Ontario, the probability of a declining trend throughout the 1980s/early 1990s varied from greater than 70% with walleye, lake trout, Chinook salmon, Coho salmon, and brown bullhead, to 60–70% with the rest of the fish species examined. However, these probability levels have consistently decreased (40–80%) depending on the fish species examined over the recent years, reflecting the previously reported slowing down of the PCB decline rates. Likewise, the decline rates of walleye (73% probability of decrease or 8% per year), common carp (68% or 10% per year), and yellow perch (65% or 6.5% per year) in Lake Erie during the early 1990s, have slowed down to 47% (<1% increase per year), 53% (<2% decrease per year) and 53% (<2% decrease per year) during the 2000s, respectively. Similar evidence in support of a deceleration of the PCB declining rates accompanied by a gradual establishment of a steady-state (i.e., species-specific stable concentrations around which some year-to-year variability occurs) was found for the rest of the systems and majority of the fish species examined (Fig. 5).

4. Discussion

PCB Contaminant Sources and Transport in the Great Lakes: Our analysis provided abundant evidence that fish species in the Upper Great Lakes, Lake Superior and Lake Huron, displayed distinctly lower PCB levels regardless of their trophic position in the food web. Among the mechanisms that typically modulate PCB inputs, atmospheric deposition (gas exchange, dry particle, and wet deposition) has been identified to be relatively important in the Upper Great Lakes. Generally, it is believed that atmospheric PCB sources tend to be higher in heavily populated urban and industrial sites. Nonetheless, the magnitude of these sources in urban air is unknown, as emission inventories that have offered reliable estimates for other pollutants (mercury, furans, and dioxins) are inadequate for PCBs (Hornbuckle et al., 2006). Extensive analysis based on air trajectories for sites in the Integrated Atmospheric Deposition Network indicated that Chicago is an important source area for the western Great Lakes region, while the heavily populated East Coast is a source for the eastern portion of the Great Lakes (Hafner and Hites, 2003). PCBs in air are enriched in the less chlorinated congeners, while the higher molecular weight compounds can also be detected but (typically) have low concentrations. Notwithstanding the presence of strong year-to-year variability, PCB levels in the Great Lakes basin have decreased by a factor of 7–10 since their production was banned in 1977. In particular, Buehler et al. (2002) showed that the half-lives for gas-phase $\sum$PCB varied between 6 and 10 years in the Great Lakes, and Lake Superior was lying closer to the lower end of that range. In a similar manner, half-life PCB estimates in water and biota of Lake Superior were on average 3 and 11 years, respectively (Smith, 2000; Offenberg and Baker, 2000).

Albeit their comparatively low PCB levels, our analysis showed that the decline rates in most of the fish species from Lake Superior slowed down after the early 1980s, and this trend could be attributed to a complex interplay among the atmosphere, water column, and sediments. Earlier work by Jeremiason et al. (1994) predicted that volatilization is the dominant PCB loss mechanism, and the

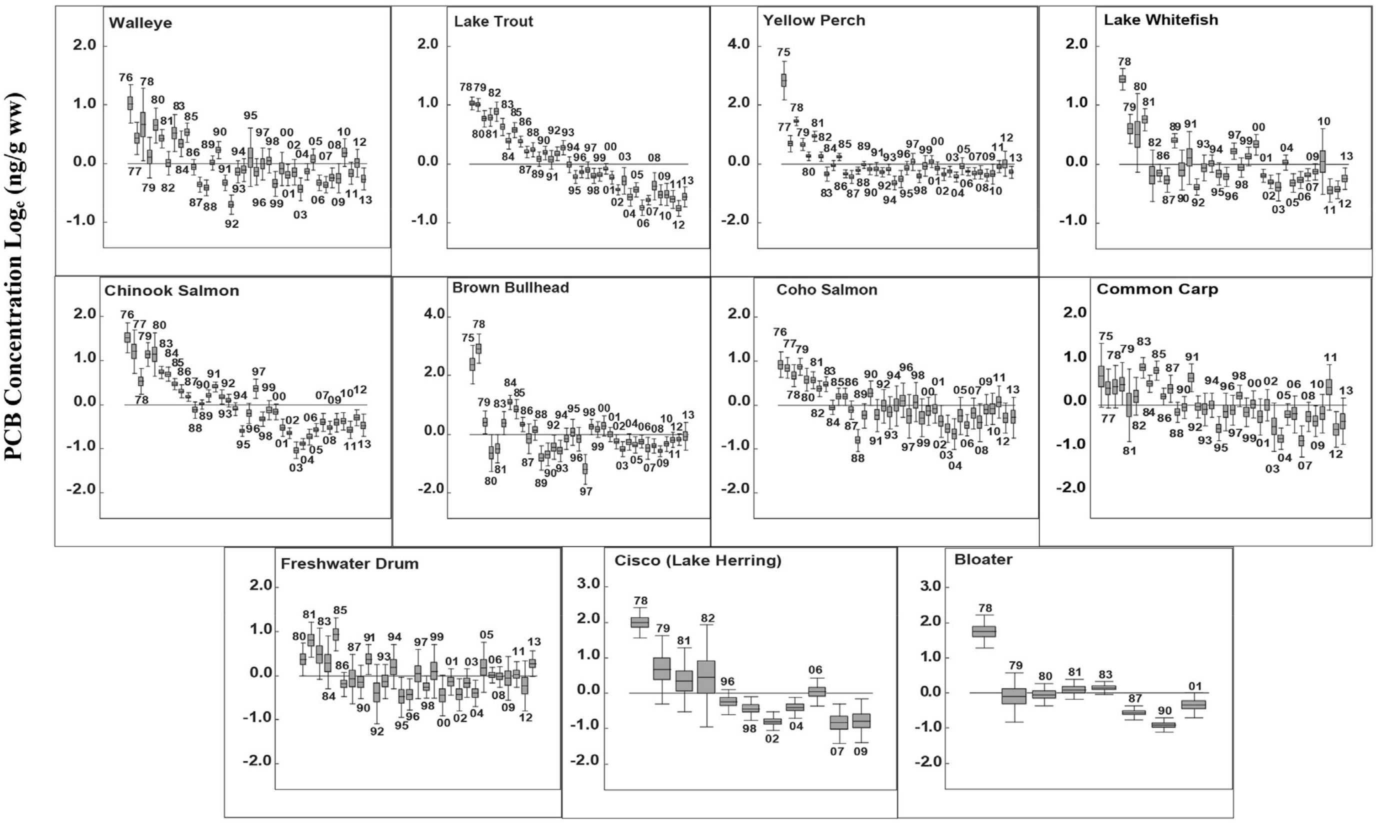

Fig. 3. Box-and-whisker plots depicting the posterior samples for the year specific intercept terms of the Bayesian hierarchical model. The whisker edges of the box plots represent the 2.5% and 97.5% percentiles, the solid black line at the approximate centre of each box represents the posterior mean values, and the lower and upper edges of each box correspond to 25% and 75% percentiles.

associated rate was estimated to be -0.24 yr^{-1}. On the other hand, despite the large fraction (>50%) of PCBs transported by settling particles within 5 m of the lake bottom, PCB loss due to permanent sediment burial was identified to be an insignificant removal mechanism in Lake Superior. Less than 10% of settling PCBs appear to accumulate in bottom sediments, as the benthic food web acts as a pathway that reintroduces them into the water column and thus prevents their complete burial (Jeremiason et al., 1998; Hornbuckle et al., 2006; see also following paragraph). The latter "recycling" process increases contaminant exposure to the lake biota, and may be partly responsible for the moderate response of the PCB levels in the studied fish species examined in our study. Similar to Lake Superior, atmospheric deposition plays a role in the introduction of PCBs in Lake Huron, resulting from emissions in storage facilities and evaporations from contaminated soils in urban regions (Song et al., 2005). Following the chemical ban, local sources have become the most relevant input mechanisms, and the PCB spatial distribution in Lake Huron has been primarily shaped from historic loadings in Saginaw Bay, Spanish River, and Black River (Stevens and Neilson, 1989; Anderson et al., 1999; Hornbuckle et al., 2006).

Unlike the Upper Great Lakes, Lake Erie and Lake Ontario exhibited high PCB levels for most of the fish species examined. Both systems have had a longer history of direct loadings from anthropogenic contamination, particularly from the Detroit and Niagara Rivers. The contamination in Lake Erie collectively reflects the impact of loadings from the area referred to as "Canada's Chemical Valley", located in the city of Sarnia along the shores of St. Clair River, as well as from Lake St. Clair and Detroit River (McCoy et al., 2014). PCB accumulation in the sediments of the shallow western basin of Lake Erie is not a straightforward function of downward settling of falling particles onto the more quiescent depositional basins, as contaminants can be recycled back to the water column via physical and biological processes occurring at the sediment–water interface (Drouillard et al., 2003). Namely, the adsorptive properties of PCBs are primarily regulated by their hydrophobicity along with the particle organic carbon content, which in turn is closely related to the clay or fine-sized particles. PCBs may also partition into pore water or bind to colloidal organic matter and get transported via diffusion within and from the sediments. PCB burial can also be controlled by sediment resuspension and bioturbation (Whittle et al., 2003; Heidtke et al., 2006). Considering the multitude of processes that can alter the depositional contaminant history and determine the residence time in the ecosystem, it is important to note that existing reports suggest a 70% lake-wide contaminant reduction in Lake Erie between 1971 and 1997, stemming from the bi-national agreements and associated remediation efforts (Marvin et al., 2004).

In Lake Ontario, existing empirical and modelling evidence shows that the Niagara River represents 75–85% of the hydraulic and >50% of the total contaminant loading, which is transported and ultimately deposited onto the sediments, thereby forming a distinct west-to-east gradient (Marvin et al., 2004; Ethier et al., 2012). Importantly, despite the elevated PCB concentrations in the three major depositional basins (Niagara, Mississauga, Rochester), none of the sediment cores collected in the late 1990s displayed exceedances of the probable effect level (PEL) for total PCBs (277 ng/g); a benchmark stipulated by the Canadian Sediment Quality Guidelines to define the concentration above which adverse biological effects are expected to occur frequently (Marvin et al., 2003). Overall, the surficial sediment 75th percentiles in Lake

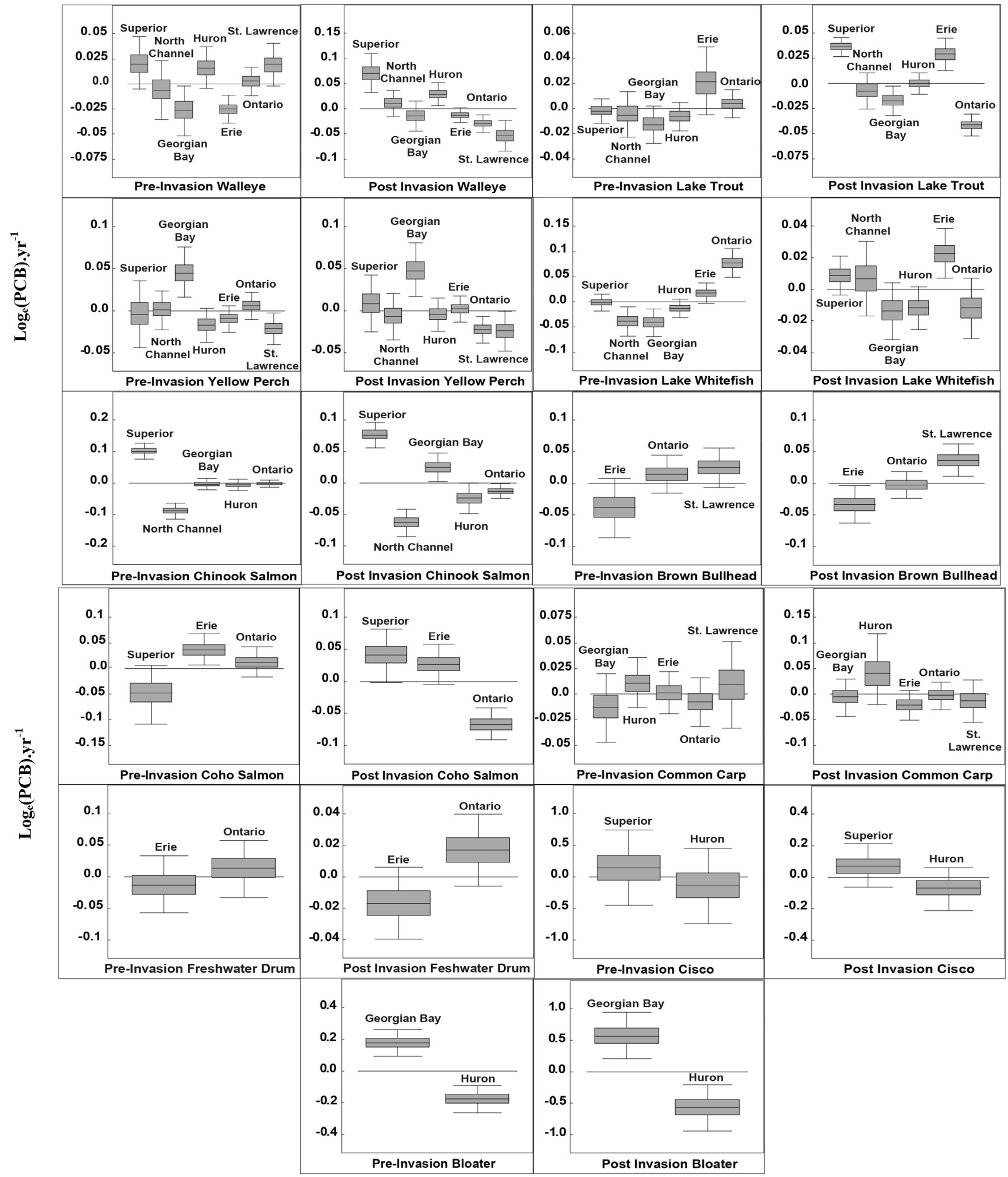

Fig. 4. Box-and-whisker plots depicting the posterior samples of the regression coefficients related to the water body-specific PCB trends before or after the invasion of exotic species in the Great Lakes. The whisker edges of the box plots represent the 2.5% and 97.5% percentiles, the solid black line at the approximate centre of each box represents the posterior mean values, and the lower and upper edges of each box correspond to 25% and 75% percentiles.

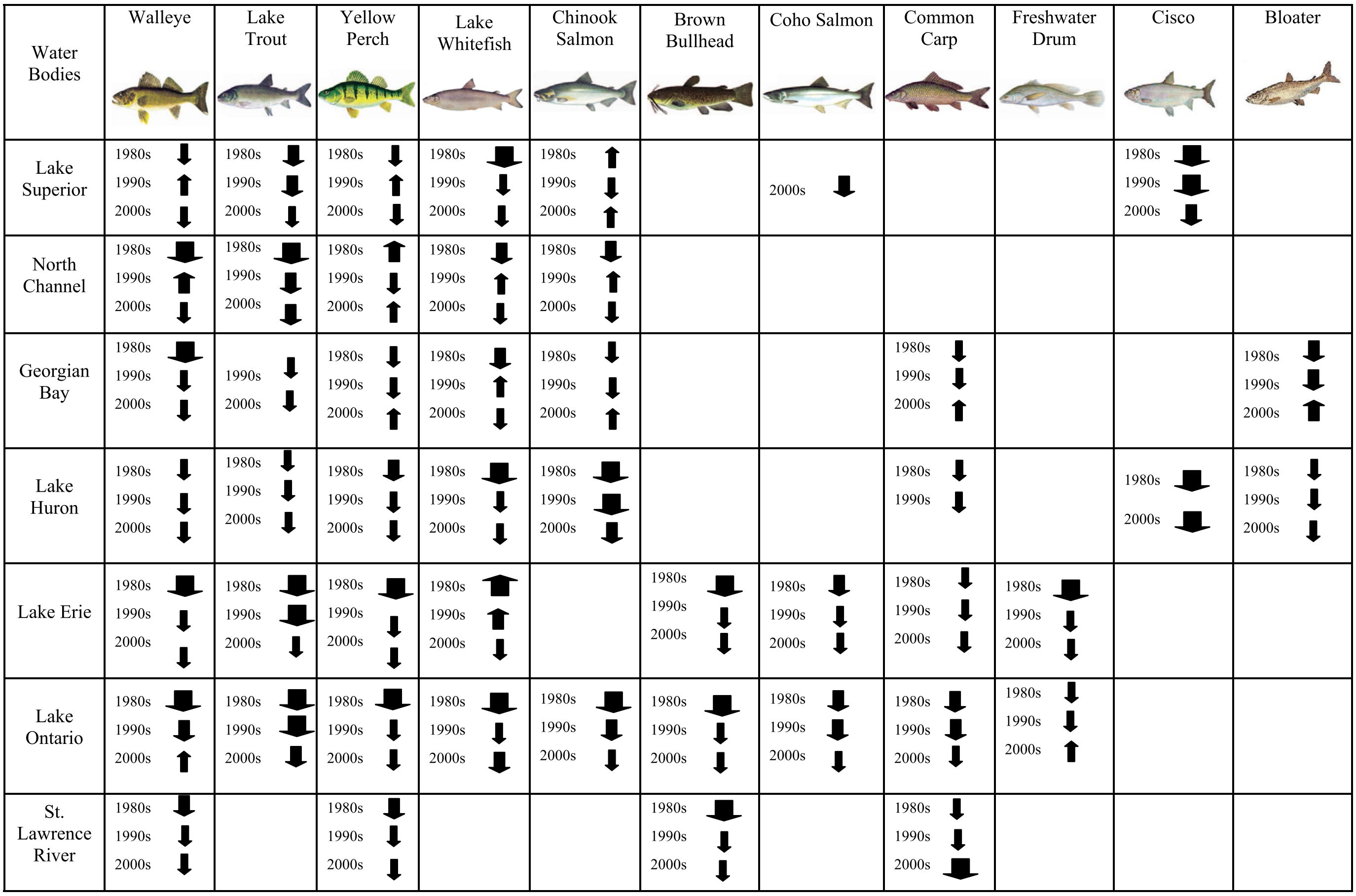

Fig. 5. Rates of change of PCB levels in different fish species and water bodies in the Great Lakes area based on dynamic linear modelling. Direction of the arrows indicates positive (upward) or negative (downward) PCB rates of change, based on the odds ratios of the posterior estimates of the *rate* parameter (eqs (2) and (3)). The odds ratio of the rate parameter being below zero in a particular year is the ratio of the probability mass below zero to the mass above zero. Trend magnitudes (arrow thickness) were classified such that values > 4 (>80% probability of decrease), 2–4 (66–80% probability of decrease), and 1–2 (50–66% probability of decrease) were indicative of a strong, medium, and weak decrease, whereas odds ratio values < 1 (<50% probability of decrease) are suggestive of a likely increase in the PCB levels.

Ontario exhibited lower PCB values than in the western basin of Lake Erie, but distinctly greater than the central and eastern basins (Marvin et al., 2002; see their Table 1). In the same context, our analysis showed that the vast majority of the fish species examined were characterized by the highest PCB levels around the Great Lakes; especially for Chinook salmon, lake trout, and common carp.

Contamination "Hot Spots": After the 2016 assessment (http://publications.gc.ca/collections/collection_2016/eccc/En164-53-1-2016-eng.pdf), there are 14 locations in the Canadian waters still identified as AOCs due to impairment of beneficial uses. Of the 14 identified impairments, PCBs are implicated in seven, including the restrictions on fish and wildlife consumption, degradation of fish and wildlife populations, fish tumors or other deformities, bird or animal deformities or reproduction problems, degradation of benthos, restrictions on dredging activities, and added costs to agriculture or industry (IJC, 2017). Most of these sites may continue to contribute to PCB contamination in the Great Lakes system through tributary flows, erosion, volatilization, and uptake by local fish populations (Hornbuckle et al., 2006). In this context, our study aimed to identify the degree of fish contamination in those sites relative to the (presumably) unimpacted sampling locations within the same water bodies.

In Lake Superior, our modelling analysis was on par with existing empirical evidence that the Thunder Bay AOC exhibits elevated fish contamination levels, although none of the fish species experienced the top quartile of their corresponding concentrations in that location during our study period (Fig. 6). The local watershed is drained by the Kaministiquia River system comprising a number of smaller rivers and creeks. The marsh area of the harbour represents a major portion of wetlands in the Lake Superior basin in Canada, providing habitat for nesting and migrating species of birds and a wide variety of fish. Despite the restrictive consumption advisories, the area supports both commercial and sport fisheries. The City of Thunder Bay, one of Canada's largest shipping ports, is the main population centre in the region. Likewise, relatively high PCB concentrations were found in sampling sites that correspond to the Peninsula Harbour AOC, which includes the harbour, from the peninsula to Ypres Point, and extends about 4 km offshore into Lake Superior past Pebble Beach, southeast of the peninsula (Site #8a). In the same area, there have been continuous efforts to restrict pollution, starting with the Municipal-Industrial Strategy for Abatement that regulated the industrial effluent discharge from several industrial sectors, including the paper/pulp, chemical and mining industries. In 2012, steps were taken to cap contaminated sediments in the area with clean sand, a process known as thin-layer capping (Foster and Ratcliff, 2013). Our results for all fish species studied (Fig. SI 3), are suggestive of a gradual improvement in response to early remediation efforts, but historical contamination still displays a strong signal. Interestingly, the highest PCB levels in lake trout

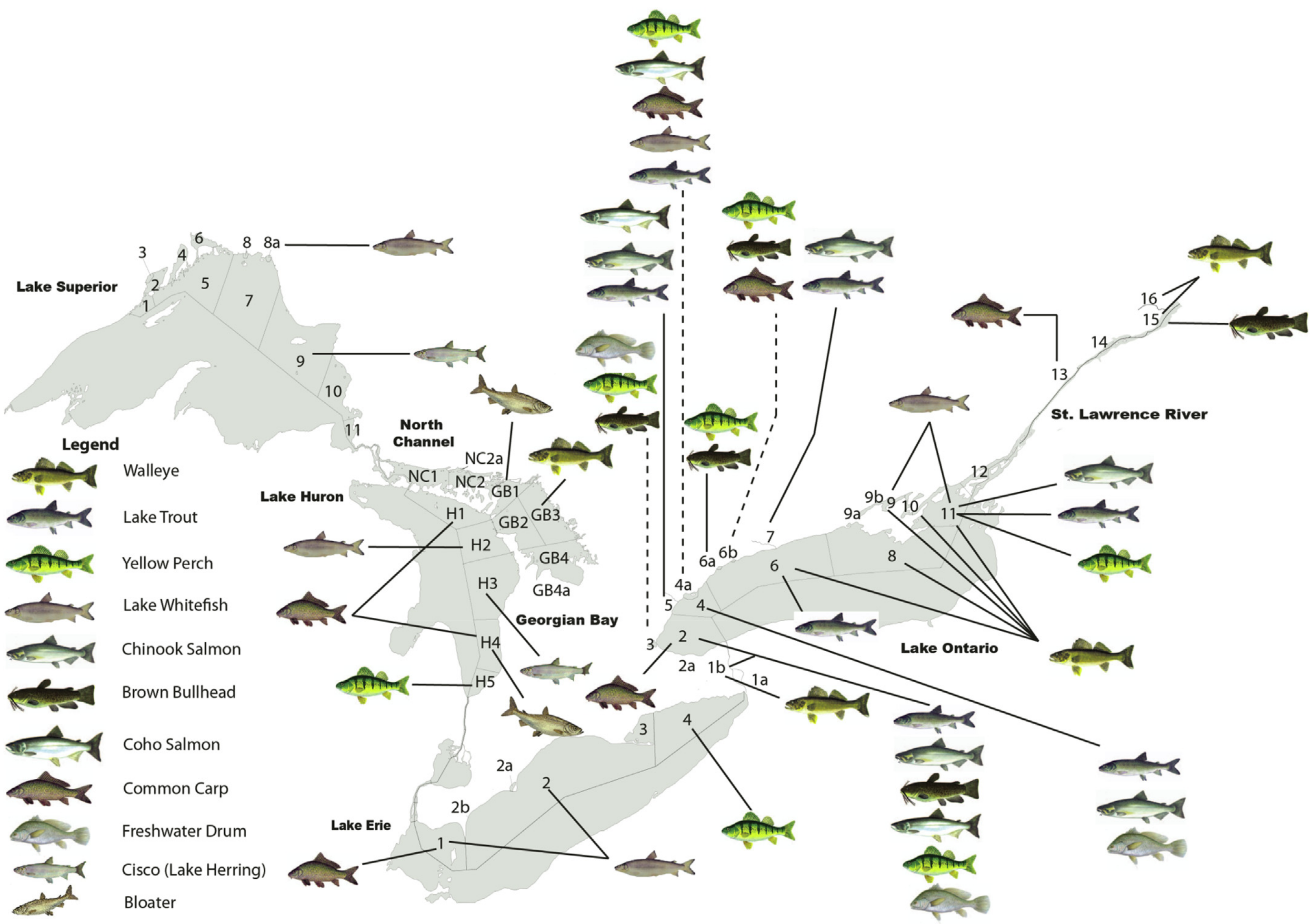

Fig. 6. Hot spots of fish PCB contamination in the Great Lakes. The map shows the locations where each fish species displays the top 25% concentrations across all of the sites sampled during our study period.

(922 ng/g ww) were recorded in eastern Lake Superior around the Michipicoten Bay, Cape Gargantua offshore, and Michipicoten Island area (Site #9) (Table SI 3). These sites have not been reported to have undergone major historical contamination, but the presence of four hydroelectric generating station along the Michipicoten River (which discharges into the Michipicoten Bay) could be contributing factors in explaining the relatively high PCB levels present in the area.

In Lake Huron, the elevated contamination patterns do not spatially overlap with any of the restored or in recovery AOCs. In particular, there are two locations, Collingwood Harbour and Severn Sound, where the impaired beneficial uses have been determined to be in accordance with the Great Lakes Water Quality Agreement, and were thus removed from the list of AOCs in 1994 and 2003, respectively. Likewise, our analysis revealed that PCB levels for the majority of the fish species examined were similar to the values found in other locations of the broader Lake Huron-Georgian Bay area. In a similar manner, the Spanish Harbour, located on the North Channel of Lake Huron, is now classified as an Area in Recovery. Restricted consumption of brown bullhead, white sucker, smallmouth bass, yellow perch, whitefish, channel catfish, and walleye is still advised due to elevated mercury, dioxins, and furans. In addition, total PCB and dioxin-like PCB advisories have also been implemented. Potential risks to humans are mitigated by adhering to provincial fish consumption advisories. Our analysis also showed that lake trout, lake whitefish, and Chinook salmon sampled from the area between Grand Bend to Point Edward displayed relatively high PCB concentrations (Site #H5; Fig. 6). This region is located downstream of Saginaw Bay, which has had a history of contamination and thus has not remained free of PCB inputs, especially if we consider the important role of atmospheric loadings (Guo et al., 2018). Furthermore, our results indicated that PCB concentrations in lake trout were highest in the main basin of Lake Huron, followed by Georgian Bay and North Channel (Table 1 & Fig. 2). There are several possible explanations for this spatial trend. Lake Huron (main basin) and Georgian Bay have lower fish biomass in comparison to North Channel, which also happens to be the smallest basin by volume, and therefore provides a higher prey-density environment (Paterson et al., 2016). Empirical evidence suggests a uniquely high degree of basin fidelity for Lake Huron lake trout throughout their lifetime in addition to highly distinct PCB bioaccumulation profiles, which implies that lake trout most likely allocate more energy to foraging behavior and activity needed for growth in the deeper and less dense prey areas, such as Georgian Bay and the main basin of Lake Huron (Paterson et al., 2016). As a result, the differences in lake trout PCB concentrations observed amongst the three Lake Huron spatial segments could also be reflective of their life history energy expenditures.

Lake whitefish and walleye samples from the western and central basins (East of Point Pelee to West of Long Point Bay; Sites

#1 and #2) in Lake Erie showed high PCB concentrations (Table SI 2, Fig. 6). Both of these basins have experienced excessive PCB loadings from the Detroit River. In particular, the local hydrodynamic (counter-clockwise) patterns tend to move sediment-bound contaminants south from the Detroit River across the Sandusky Basin in the area south of Pelee Island. Elevated PCB levels along the southern shore of the central basin may also have been influenced by inputs from heavily industrialized areas, including Cleveland and Ashtabula as well as agricultural activities from the adjacent sub-watersheds (Frank et al., 1977; Smits et al., 2005). In the same context, Marvin et al. (2004) conducted a sediment survey to characterize spatial trends in contamination, and to assess any improvements in environmental quality since the advent of remedial measures to reduce contamination. Two of their key findings were that lake-wide average sediment PCB levels decreased roughly 70% from 136 ng/g in 1971 to 43 ng/g in 1997, and that there was no evidence that contaminants transported from the western basin and southern areas of the central basin were ultimately deposited in the eastern basin (Marvin et al., 2004). The decrease in sediment loadings during the time-frame stated above is reflective of the declining concentration levels in walleye samples from Lake Erie (Fig. SI 3); especially during the 1980s (Fig. 5). The latter trend was partly attributed to the successful management of sources over the past thirty years along the south-eastern shoreline in New York State in an area that included a number of industrial waste sites (Townsend, 1998). Moreover, none of the stations surveyed in 1997 exceeded the Canadian PEL guideline for the protection of aquatic biota against PCBs, which is indicative of an improvement relative to the frequent exceedances recorded in the area in proximity to the mouth of the Detroit River during the early 1970s (Marvin et al., 2004).

Influenced by industrially-laden watersheds, walleye from the Niagara Falls to Whirlpool, including the Hamilton Harbour AOC, had distinctly higher PCB concentrations (Marvin et al., 2007; see also sites #1–3 in Fig. 6 and SI 1–2). The development of the steel and iron industry in the Hamilton Harbour paved the way for the expansion of an important shipping center. In addition, the harbour has been receiving discharges from waste water treatment plants and urban runoff from the cities of Hamilton and Burlington (Neff et al., 2016). Consumption advisories from western Lake Ontario remain severely restrictive. For instance, Visha et al. (2016) found that the tolerable daily intake (TDI) values from walleye consumption for a given number of meals per month with sensitive demographic groups (e.g., women of child-bearing age, children under the age of 15), were often exceeded during the mid-1980s, 1990s, and 2000s across all the fish length and lipid content values measured in Lake Ontario.

Likewise, the TDI for lake trout (based on fish tissue body burdens) have been frequently exceeded (>95%) in all of the Lake Ontario sites during the three snapshots in time examined (Visha et al., 2016). Chinook salmon similarly exhibited elevated PCB concentrations in several locations, including the Bronte Creek and Niagara Bar (Fig. 6). In a similar manner, lake whitefish from Ashbridge's Bay, Toronto Waterfront, and Humber Bay area displayed distinctly higher PCB levels relative to the rest of the locations sampled in Lake Ontario (Sites #4 and #4a in Fig. 6). Along the same line of evidence, Bhavsar et al. (2018) showed substantial declines in fish contamination in the Toronto and Region AOC, although the PCB levels for the majority of the fish species are still above the benchmarks. Using an empirical exponential decay model, the same study derived PCB half-life estimates to project that "unrestricted" advisories will be achieved for that AOC within the next 8–15 years, depending on fish ecology and feeding patterns (Robinson et al., 2015).

Trophodynamics and PCB temporal trends: Generally, our analysis showed that the PCB levels in a number of fish species with different trophic positions and functional roles in the food web showed significant declines throughout the late 1980s/early 1990s, most likely stemming from the reductions in industrial emissions and other contamination mitigation strategies, but appear to have been stabilized over the last 10–15 years. Many plausible explanations have been proposed to elucidate the apparent establishment of a steady state in PCB contamination. One plausible hypothesis is related to changes in trophodynamics after the food web restructuring induced by the establishment of non-indigenous species and a likely diet shift in more highly contaminated prey items. The introduction of zebra (*Dreissena polymorpha*) and quagga (*D. bugensis*) mussels has likely induced major changes in the fluxes of heavy metals within the food webs, because of their ability to bioaccumulate through excessive filtering of contaminated water and scavenging of phytoplankton and particulate matter (Kwan et al., 2003). The capacity of dreissenids to selectively remove particulate organic matter from the water column increases the dissolved-phase fraction of contaminants, which in turn can increase the absorption efficiencies and the body burdens of many aquatic organisms. Dreissenids can thus be good sentinels of the bioavailability of contaminants and may elevate the likelihood of trophic transfer (Kwon et al., 2006). Concomitant to the proliferation of dreissenid mussels, round goby invaded the Great Lakes after the mid-1990s and existing evidence suggests that the rapid proliferation and aggressive behaviour of round goby can alter benthic communities and nutrient cycles (Janssen and Jude, 2001), displace native species through shelter monopolization (Balshine et al., 2005), and voraciously consume eggs of native fishes (Chotkowski and Marsden, 1999). As a benthic fish with diet mainly composed of dreissenids, round goby has the potential to accumulate and subsequently transfer contaminants to the higher trophic levels (Johnson et al., 2005). Consequently, the prospect of an increased reliance upon benthivorous round goby can possibly be one of the factors that have slowed down the earlier declining rates in fish communities around the Great Lakes.

A plausible confounding factor that may drive the recent PCB trends could be the reduced growth rates and subsequent growth dilution, given that the long-term trends of eutrophication-related variables conclusively showed that ambient phosphorus and primary productivity have declined to levels that may undermine pelagic ecosystem integrity in the offshore waters around the Great Lakes (Dove, 2009; Dove and Chapra, 2015). Although it is worth noting that growth dilution occurs when the overall biomass accumulation exceeds contaminant accumulation, and is not necessarily a consequence of lower feeding rates. Another relevant mechanism could be associated with switches to prey items, associated with the recent food-web restructuring (e.g., salmonine restocking, invasive species) that potentially result in energetics-related growth reduction. For example, the significant temporal decline in lake trout body mass and energy density was attributed to their k-strategist behavior, and thus its inferior competition capacity relative to the r-selected non-native salmonids for depleted prey (alewife, rainbow smelt) populations (Paterson et al., 2009). Global warming is also likely to be influencing the trophodynamics of contaminants by altering lake phenologies and biotic community structures (Shimoda et al., 2011). Alongside the identification of the mechanisms that may have led to the deceleration of the PCB declining rates, an additional challenge is the determination of the steady state concentrations that are gradually established in the Great Lakes. The latter species- and system-specific levels are obfuscated by year-to-year variability as well as by the oscillations associated with the nature and relative strength of the different prey–predator interactions within the aquatic food webs and/or the periodicities of climatic forcing (Scheider et al., 1998; French et al., 2006). A characteristic example is the downward trajectory and periodic oscillations of contaminant levels in lake trout, Coho and Chinook salmon from Lake Ontario, which were causally linked to the abundance of alewife biomass (Borgmann and Whittle, 1991; French et al., 2006). A similar example is the study conducted by Stow et al. (1995) in Lake Michigan, linking the increasing PCB trends in Coho and Chinook salmon to growth dynamics driven by changes in the food web; namely, decreased prey availability and/or increased foraging expenditures were likely responsible for decreased growth dilution. Another example is the mayfly (*Hexagenia limbata*) which represents a major prey item of many fish species and tends to be one of the initial organisms introducing PCBs into the food web. Mayfly larvae spend the early stage of their lives burrowing in sediments and feeding off detritus, benthic biofilms and settling particles, thereby accumulating high levels of sediment-affiliated PCBs (Gewurtz et al., 2000; Pitt et al., 2017). The cyclic life patterns and abundance of mayflies can be one such factor that may modulate the inter-annual variability of PCB burdens of different fish species in western Lake Erie (Pitt et al., 2017).

Conclusions: We presented a rigorous assessment of the spatio-temporal PCB trends in multiple species across the Canadian waters of the Great Lakes. Our analysis suggests that Lake Ontario is characterized by the highest PCB levels with nearly all of the fish species examined. The general temporal trend across the lakes was that the high PCB concentrations during the early 1970s declined throughout the late 1980s/early 1990s, as a result of the reductions in industrial emissions and other remedial efforts. However, our analysis provided evidence of a deceleration of the PCB declining rates after the late 1990s/early 2000s, accompanied by a gradual establishment of species-specific stable concentrations, around which there is considerable year-to-year variability. Historically industrialized urban areas and embayments receiving riverine inputs displayed elevated concentrations within each of the water bodies examined. The overall trends indicate that the reduced contaminant emissions have significantly reduced the degree of exposure to PCBs, but historical contamination along with other external or internal stressors (e.g., invasive species, climate change) are potential confounding factors that can modulate the current levels and thus pose human health risks through fish consumption.

Acknowledgements

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.chemosphere.2018.07.070.

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A BAYESIAN ASSESSMENT OF POLYCHLORINATED BIPHENYL CONTAMINATION OF FISH COMMUNITIES IN THE LAURENTIAL GREAT LAKES

[SUPPORTING INFORMATION]

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SECTION A

Chemical analysis

Total PCB analysis on the MECP samples was performed through gas chromatography with ^{63}Ni electron capture detector (ECD) as described by Bhavsar et al. (2007). Quantification was carried out using the 23 largest "Aroclor" peaks obtained in the pseudo packed column technique (OMOE, 2007). 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. This ratio of Aroclors best resembled the congener patterns detected for most fish samples. A five point calibration curve with single point continuing calibration was used to quantify samples. The method detection limit is 20 ng g^{-1}. A blank and spiked blank matrix sample was processed with each set of samples (20 to 30). The method performance is monitored through laboratory inter-calibration studies (i.e., Northern Contaminants Program and Quality Assurance of Information for Marine Environmental Monitoring in Europe).

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SECTION B

Bayesian Modelling

For all of our modelling work, we obtained sequences of realizations from the posterior distributions with Markov-chain Monte Carlo (MCMC) simulations (Gilks et al., 1998). Using the WinBUGS software (Lunn et al., 2000), we implemented a general normal proposal Metropolis algorithm that is based on a symmetric normal proposal distribution. For each analysis, we used three chain runs of 100,000 iterations, keeping every 20th iteration to minimize serial correlation. Convergence of the MCMC chains was checked using the Brooks–Gelman–Rubin (BGR) scale-reduction factor (Brooks and Gelman, 1998). The BGR factor is the ratio of among chain variability to within chain variability. The chains have converged when the upper limits of the BGR factor are close to one. The accuracy of the posterior parameter values was inspected by assuring that the Monte Carlo error (an estimate of the difference between the mean of the sampled values and the true posterior mean) for all parameters was less than 5% of the sample standard deviation (Spiegelhalter et al., 2003).

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Table SI 1: Posterior statistics (mean ± standard deviation) for the global intercept term, the length- and lipid-regression coefficients of the Bayesian hierarchical model used to evaluate the spatiotemporal PCB trends in the Great Lakes.

Species	N	β_G	β_1	β_2
Walleye	3167	4.16 ± 0.05	0.33 ± 0.02	0.40 ± 0.02
Lake Trout	4245	5.84 ± 0.04	0.27 ± 0.01	0.49 ± 0.01
Yellow Perch	2202	3.51 ± 0.07	0.015 ± 0.016	0.11 ± 0.02
Lake Whitefish	2324	4.67 ± 0.04	0.20 ± 0.01	0.45 ± 0.02
Chinook Salmon	2087	5.93 ± 0.24	0.44 ± 0.01	0.18 ± 0.01
Brown Bullhead	937	4.13 ± 0.10	0.25 ± 0.02	0.36 ± 0.02
Coho Salmon	1426	5.25 ± 0.11	0.24 ± 0.02	0.15 ± 0.02
Common Carp	1624	6.03 ± 0.08	0.39 ± 0.03	0.70 ± 0.03
Freshwater Drum	711	5.00 ± 0.07	0.094 ± 0.041	0.56 ± 0.03
Cisco (Lake Herring)	278	4.88 ± 0.19	0.045 ± 0.048	0.46 ± 0.04
Bloater	407	5.35 ± 0.06	0.14 ±0.03	0.23 ± 0.04

Table SI 2: Sampling locations for walleye, lake whitefish, and chinook salmon across the Great Lakes. Numbers in italics correspond to those provided along the X axes in Figure SI 2 (location-specific intercept terms), while numbers in bold font match the numbers used to visualize the sampling locations in Figure 1. Sites highlighted in bold and underlined represent locations with listed and de-listed Areas of Concern (AOCs)

Water Bodies	Walleye		Lake Whitefish		Chinook Salmon	
Lake Superior	*Site # 1*	**1** (Border/Pie Island area, Cloud Bay)	*Site # 1*	**1** (Border/Pie Island area, Jarvis Bay)	*Site # 1*	**2** (Thunder Bay Outer Harbour, Caribou Island)
	Site # 2	**2** (Thunder Bay Outer Harbor)	*Site # 2*	**2** (Thunder Bay Outer Harbour, Welcome Islands)	*Site # 2*	**3** (Thunder Bay Inner Harbour)
	Site # 3	**3** (Thunder Bay Inner Harbor, Mission River Mouth, Current River Mouth)	*Site # 3*	**3** (Thunder Bay Inner Harbour)	*Site # 3*	**4** (Black Bay)
	Site # 4	**4** (Black Bay, Black Sturgeon River)	*Site # 4*	**4** (Black Bay)	*Site # 4*	**5** (No Description)
	Site # 5	**7** (Aquasabon River Mouth)	*Site # 5*	**5** (Rossport, Sturgeon Bay/Black Bay Peninsula)	*Site # 5*	**6** (Nipigon Bay)
	Site # 6	**8** (Jackfish Bay, Mobeley Bay)	*Site # 6*	**6** (Nipigon Bay)	*Site # 6*	**7** (Steel River)
	Site # 7	**8a** (Peninsula Harbor)	*Site # 7*	**7** (Pic & White River area, South of Pic Island, Schreiber Point/Sewell Point)	*Site # 7*	**9** (Michipicoten Bay, Michipicoten River, Michipicoten Island area)
	Site # 8	**11** (Goulais Bay area)	*Site # 8*	**8** (Jackfish Bay, Mobeley Bay)	*Site # 8*	**10** (Agawa Bay, Batchawana Bay)
			Site # 9	**8a** (Peninsula Harbour, South of Marathon)	*Site # 9*	**11** (Goulais Bay, Goulais River)
			Site # 10	**9** (Cape Gargantua offshore, Michipicoten Island area)		
			Site # 11	**10** (Agawa Bay, Batchawana Bay, Montreal River Mouth)		
			Site # 12	**11** (Goulais Bay area)		
North Channel	*Site # 1*	**NC 1** (St. Joseph Island area, St. Joseph Channel, Lake George, St. Marys River, South East. of St. Joseph Island)	*Site # 1*	**NC 1** (Blind River area)	*Site # 1*	**NC 1** (Algoma Mills area)
	Site # 2	**NC 2** (Spanish River, Whalesback Channel)	*Site # 2*	**NC 2** (Heywood Island area, Little Current area, South of Eagle Island)	*Site # 2*	**NC 2** (No Description)
					Site # 3	**NC 2a** (Whalesback Channel: Spanish River Mouth to Wicksteed Point)
Georgian Bay	*Site # 1*	**GB 1** (French River mouth)	*Site # 1*	**GB 1** (No Description)	*Site # 1*	**GB 1** (No Description)
	Site # 2	**GB 3** (Raft Island, Halo Island, Moon River, Byng Inlet)	*Site # 2*	**GB 2** (No Description)	*Site # 2*	**GB 4** (Collingwood area, Owen Sound, Cape Rich, Thornbury, Coldwater River &
	Site # 3	**GB 4** (Sturgeon Bay, Severn Sound, South	*Site # 3*	**GB 3** (Shawanaga Inlet, Byng Inlet)		
			Site # 4			

		of Lion's Head to South of Moon River mouth)		**GB 4** (Mary Ward Shoals, Owen Sound, Cape Rich, Collingwood area, Thornbury, South of Lion's Head to South of Moon River Mouth, Christian Island to Cape Rich)		Matchedash Bay combined, South of Lion's Head to South of Moon River mouth)
Lake Huron	*Site # 1* *Site # 2* *Site # 3*	**H3** (Douglas Point, Saugeen River, South of Stokes Bay to Point Clark) **H4** (No Description) **H5** (Point Edward to Grand Bend)	*Site # 1* *Site # 2* *Site # 3* *Site # 4* *Site # 5*	**H1** (Burnt Island, South of Providence Bay, Cockburn Island to Fitzwilliam Island) **H2** (West of St. Edmunds Township, Dorcas Bay) **H3** (Fishing Islands, Saugeen River, Howdenvale Bay, South of Stokes Bay to Point Clark) **H4** (Bayfield) **H5** (Port Franks, Point Edward to Grand Bend)	*Site # 1* *Site # 2* *Site # 3* *Site # 4*	**H1** (Blue Jay Creek, Burnt Island, Cockburn Island to Fitzwilliam Island) **H3** (Fishing Islands, Saugeen River at Southampton, Douglas Point, South of Stokes Bay to Point Clark) **H4** (No Description) **H5** (Point Edward to Grand Bend)
Lake Erie	*Site # 1* *Site # 2* *Site # 3* *Site # 4*	**1** (East Sister Island, Western Basin) **2** (Central Basin: from East of Point Pelee to just West of Long Point Bay) **3** (Long Point Bay) **4** (Eastern Basin)	*Site # 1* *Site # 2* *Site # 3*	**1** (Western basin) **2** (Central Basin: from East of Point Pelee to just West of Long Point Bay) **4** (Eastern Basin)		
Lake Ontario	*Site # 1* *Site # 2* *Site # 3* *Site # 4* *Site # 5* *Site # 6* *Site # 7* *Site # 8* *Site # 9*	**1b** (Queenston to Whirlpool, Lower Niagara River mouth, Below Falls to Lake Ontario) **2** (Niagara Bar) **3** (Hamilton Harbour) **4a (**Toronto Waterfront Tommy Thompson Park) **6** (Pickering Generating Station) **6a** (Frenchman Bay) **6b** (Whitby Harbor) **8** (Gravelly Bay, Brighton, Northeastern Lake Ontario) **9** (Trent River mouth, Big Bay, Belleville, Telegraph Narrows, Long Reach, south	*Site # 1* *Site # 2* *Site # 3* *Site # 4*	**4a** (Ashbridges Bay, Toronto Waterfront - Humber Bay area) **8** (Northeastern Lake Ontario) **9** (Upper Bay of Quinte, Trent River mouth Trenton to Telegraph Narrows) **11** (Lower Bay of Quinte, Main Duck Island, North of Main Duck Island to Wolfe Island, East of Glenora to Kingston)	*Site # 1* *Site # 2* *Site # 3* *Site # 4* *Site # 5* *Site # 6* *Site # 7* *Site # 8* *Site # 9*	**1b** (Lower Niagara River- Queenston to Whirlpool) **2** (Bronte Creek, Niagara Bar) **3** (Hamilton Harbor) **4** (Toronto Offshore- near Credit River, Scarborough Bluffs, Clarkson to Scarborough Bluffs) **5** (Credit River- Streetsville Dam) **6** (Oshawa Area, East of Scarborough Bluffs to Colborne) **7** (Ganaraska River) **8** (Brighton, Northeastern Lake Ontario) **11** (Nearshore North Channel, Main Duck Island,

	Site # 10 *Site # 11*	shore off Trenton, Upper Bay of Quinte, Salmon River mouth, Trenton to Telegraph Narrows) **10** (Middle Bay of Quinte, Hay Bay, Ram Island, Telegraph Narrows to Glenora, South of Deseronto to Glenora) **11** (Lower Gap, Nearshore North Channel, Adolphus Reach, Keith Shoal, East of Glenora to Kingston, Lower Bay of Quinte, Lyon Island)				East of Glenora to Kingston)
St. Lawrence River	*Site # 1* *Site # 2* *Site # 3* *Site # 4* *Site # 5*	**12** (Lower Thousand Islands, Kingston-Brockville) **13** (Johnstown, Middle Corridor - Brockville to Iroquois) **14** (Upper Canada Migratory Bird Sanctuary, Lake St. Lawrence) **15** (Lake St. Francis, Cornwall Island, East & West of Hamilton Island) **16** (Lake St. Francis at Raisin River Iroquois)				

Table SI 3: Location and species specific PCB averages. The top 25% of the highest averages for each species and location have been highlighted in grey.

Location	Walleye	Lake Trout	Yellow Perch	Lake Whitefish	Chinook Salmon	Brown Bullhead	Coho Salmon	Common Carp	Freshwater Drum	Cisco	Bloater
Lake Superior 1	26	635		231							
Lake Superior 2	23	440		169	37					87	
Lake Superior 3	43	181	24	177	420					40	
Lake Superior 4	24	258	21	59	73					116	
Lake Superior 5		317		81	35		42			50	
Lake Superior 6		153	24	114	334		53				
Lake Superior 7	48	426		171	574		46			53	
Lake Superior 8	23	280		88							
Lake Superior 8a	52	564		1034							
Lake Superior 9		922		62	365		67			678	
Lake Superior 10		583	25	48	128		53				
Lake Superior 11	20	117	23	36	247		77			263	
North Channel (NC1)	73	236	31	62	303						
North Channel (NC2)	38	161	21	43	268						
North Channel (NC2a)					293						
Georgian Bay (GB 1)	44	213	20	104	412						710
Georgian Bay (GB 2)		192		85							428
Georgian Bay (GB 3)	185	244	20	209							
Georgian Bay (GB 4)	40	233	41	84	357			865			239
Georgian Bay (GB 4a)			26					373			
Lake Huron (H1)		332	22	74	398			1490		237	313
Lake Huron (H2)		584		285						272	286
Lake Huron (H3)	96	375	27	88	466			1024		480	321
Lake Huron (H4)	62	471	24	190	176			1458			640
Lake Huron (H5)	102	848	102	153	793			1108			248
Lake Erie 1	135		84	244			510	1321	247		

Lake Erie 2	92		31	287			458	545	246		
Lake Erie 2a			34					204	247		
Lake Erie 2b			28					869	136		
Lake Erie 3	102		87				484	414	195		
Lake Erie 4	121	458	100	168			368	80	159		
Lake Ontario 1a			57					498	223		
Lake Ontario 1b	863	2281	92		989	165	1946	661	429		
Lake Ontario 2	140	2074	92		2144	558	1578	1211	475		
Lake Ontario 2a								348	301		
Lake Ontario 3	112		214		480	403		766	792		
Lake Ontario 4		2297	57		1405		1231	788	850		
Lake Ontario 4a	76	2884	269	1096			1537	1654	412		
Lake Ontario 5		4301			1342		1556				
Lake Ontario 6	175	1771			856		950	940			
Lake Ontario 6a	42		542			812		906			
Lake Ontario 6b	56		103			398		1747	224		
Lake Ontario 6c								199			
Lake Ontario 7		2754			1116		426				
Lake Ontario 8	224		31	144	342		743		340		
Lake Ontario 9	215		38	475				788	106		
Lake Ontario 10	148		61						44		
Lake Ontario 11	251	1302	92	344	1378				85		
St. Lawrence 12	96		36					756			
St. Lawrence 13	33		25					1410			
St. Lawrence 14	105		34					826			
St. Lawrence 15	205		70			866		23			
St. Lawrence 16	640										

FIGURES LEGENDS

Figure SI 1: Map of the 43 Areas of Concerns around the Great Lakes basin - 12 are in Canada, 26 are in the United States, and 5 are shared by both countries.

Figure SI 2: Box-and-whisker plots depicting the posterior samples for the location-specific (β_L) intercept terms per waterbody studied for three selected fish species: walleye, lake whitefish, and chinook salmon. The whisker edges of the box plots represent the 2.5% and 97.5% percentiles, the solid black line at the approximate centre of each box represents the posterior mean values, and the lower and upper edges of each box correspond to 25% and 75% percentiles. Numbers along the X axes correspond to the locations provided in Table SI 2

Figure SI 3: Dynamic linear modelling analysis for the eleven fish species examined, depicting the recorded PCB (ng/g wet weight) concentrations (gray dots) against the predicted annual trends when accounting for the covariance with fish length and lipid content (black lines). The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted PCB concentrations.

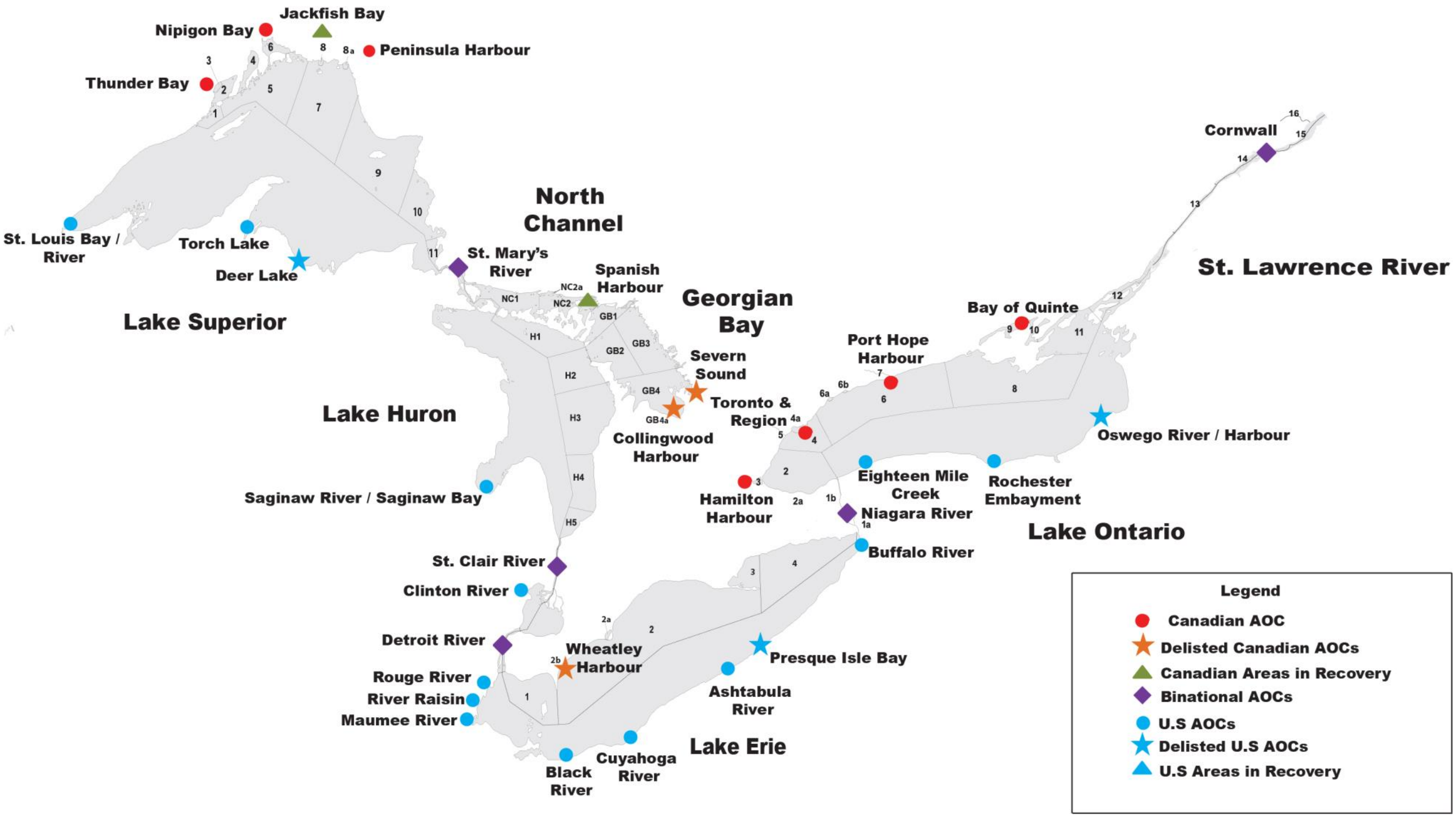

Jackfish Bay
Nipigon Bay
Peninsula Harbour
Thunder Bay
North Channel
St. Louis Bay / River
Torch Lake
Deer Lake
St. Mary's River
Spanish Harbour
Lake Superior
Georgian Bay
St. Lawrence River
Cornwall
Bay of Quinte
Port Hope Harbour
Severn Sound
Toronto & Region
Collingwood Harbour
Lake Huron
Oswego River / Harbour
Eighteen Mile Creek
Rochester Embayment
Saginaw River / Saginaw Bay
Hamilton Harbour
Niagara River
Lake Ontario
Buffalo River
St. Clair River
Clinton River
Detroit River
Wheatley Harbour
Presque Isle Bay
Rouge River
River Raisin
Maumee River
Ashtabula River
Cuyahoga River
Black River
Lake Erie
Legend
Canadian AOC
Delisted Canadian AOCs
Canadian Areas in Recovery
Binational AOCs
U.S AOCs
Delisted U.S AOCs
U.S Areas in Recovery

Figure SI 1

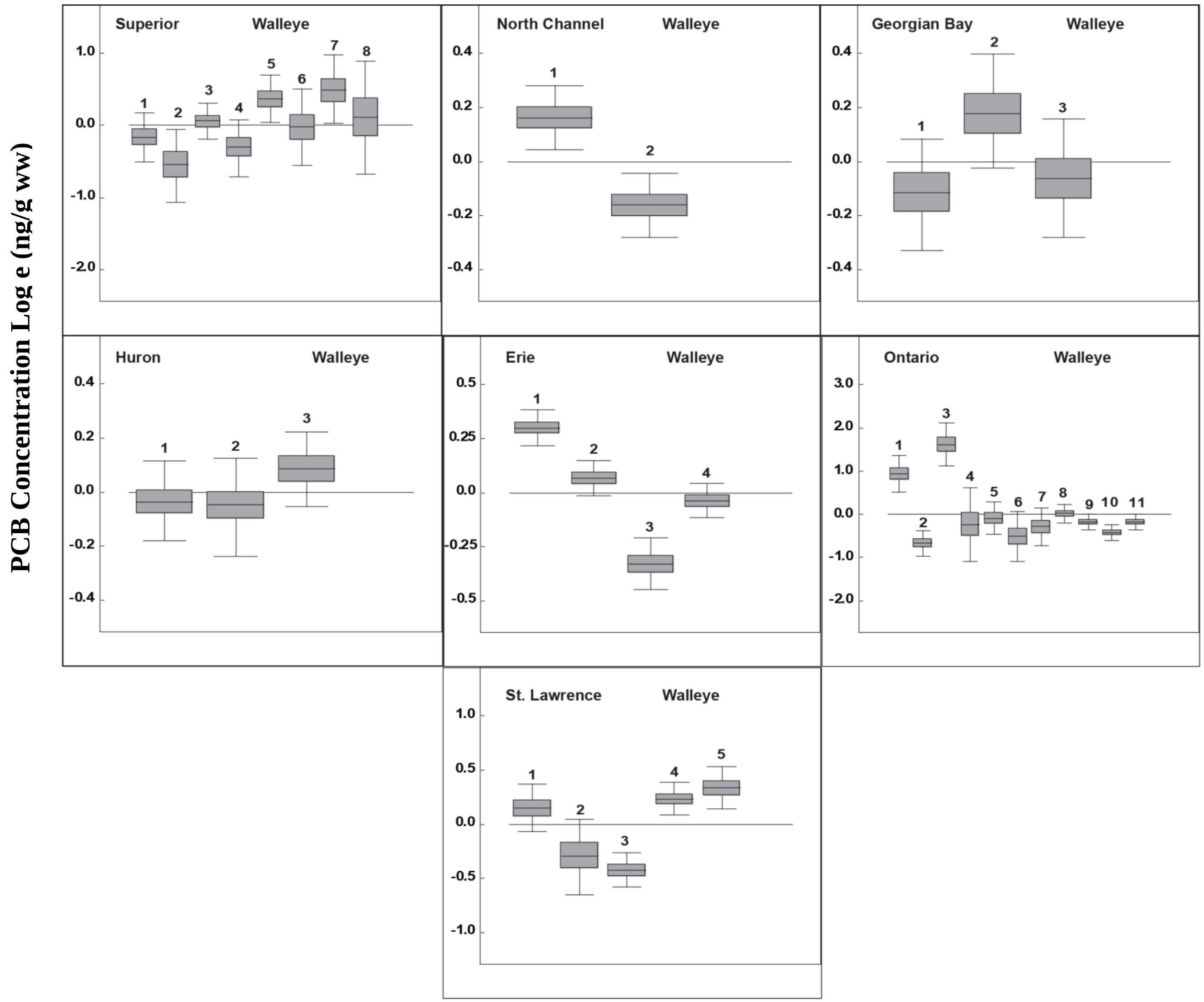
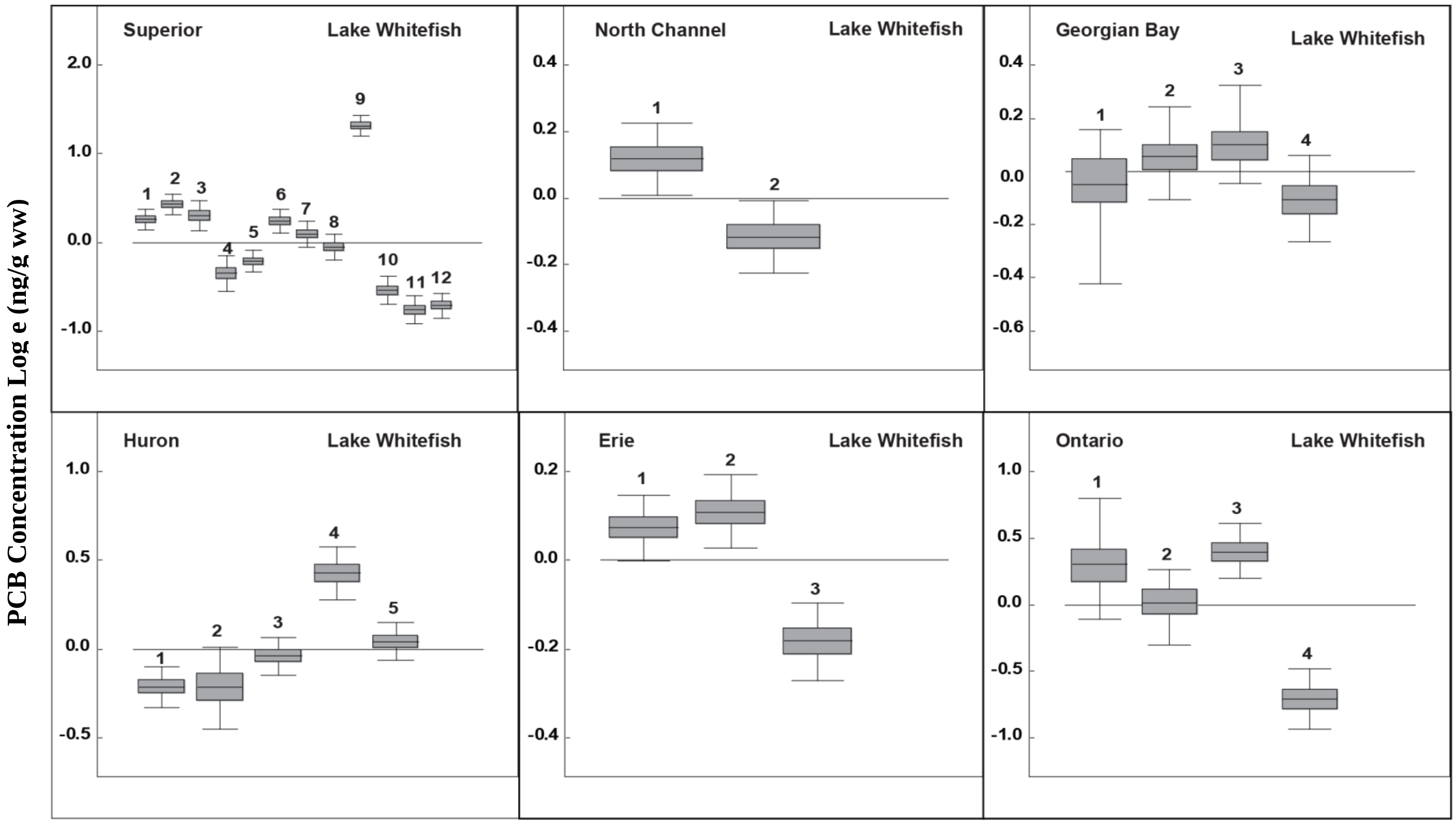

PCB Concentration Log e (ng/g ww)
Superior
Walleye
North Channel
Walleye
Georgian Bay
Walleye
Huron
Walleye
Erie
Walleye
Ontario
Walleye
St. Lawrence
Walleye

Figure SI 2



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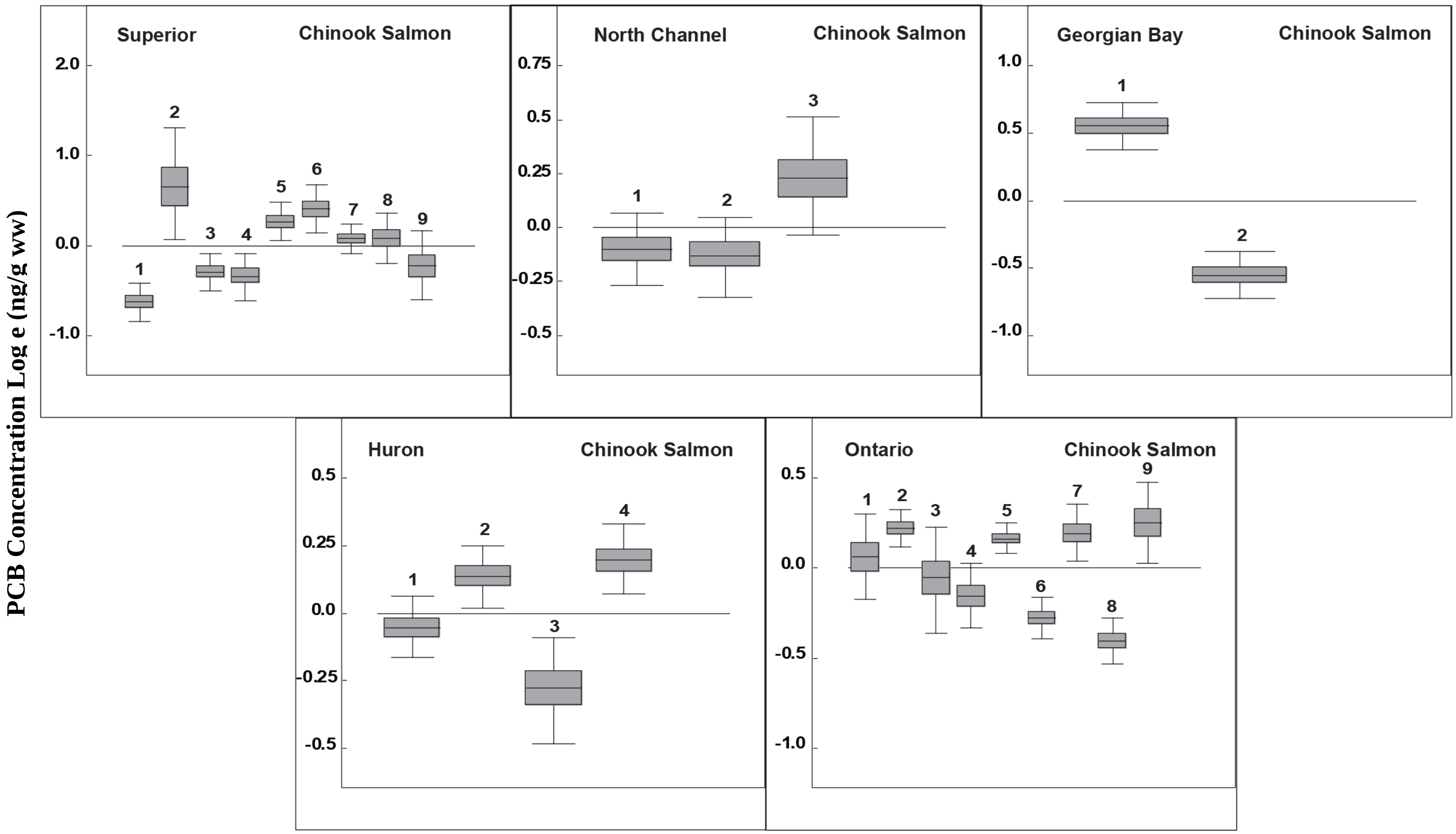

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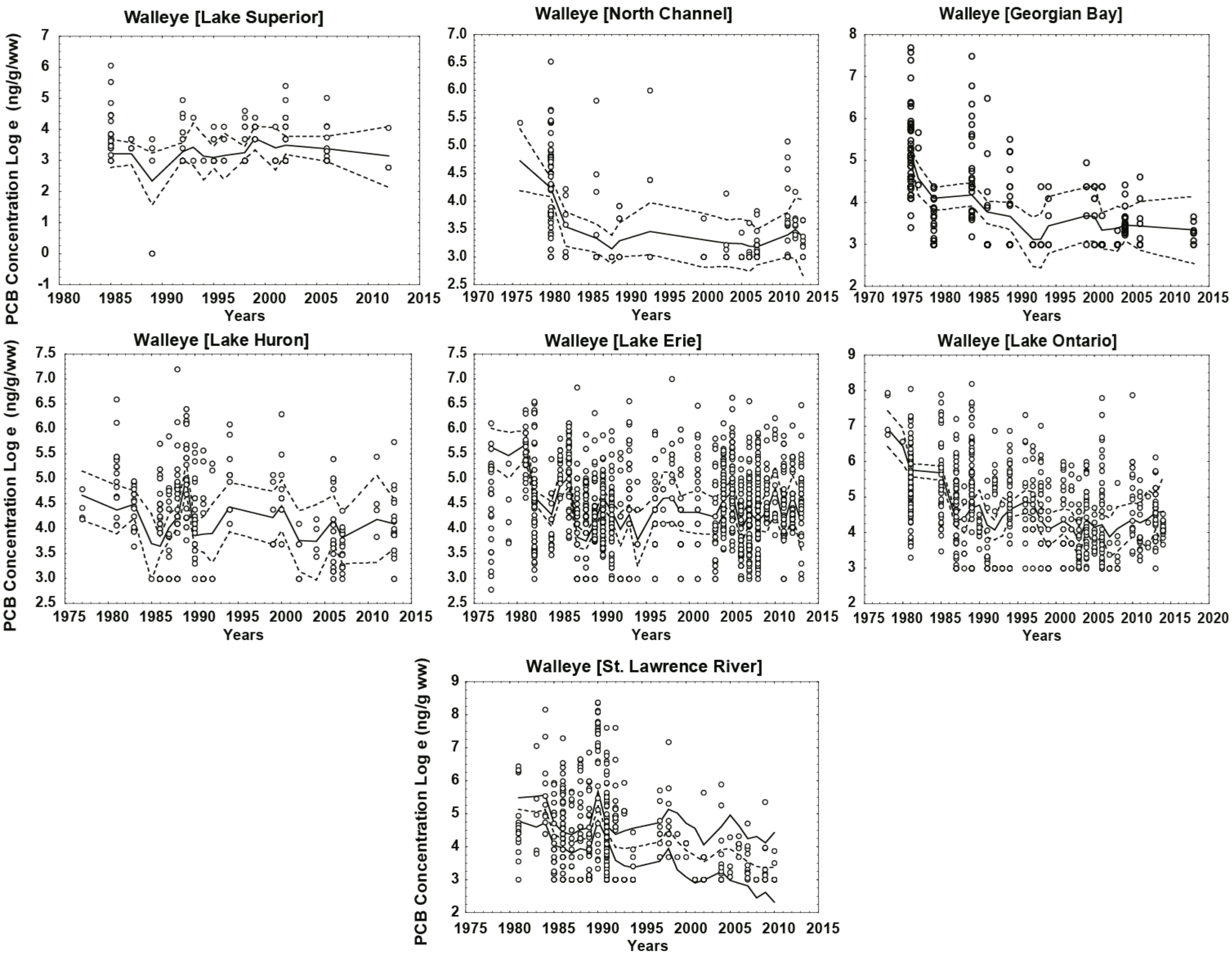
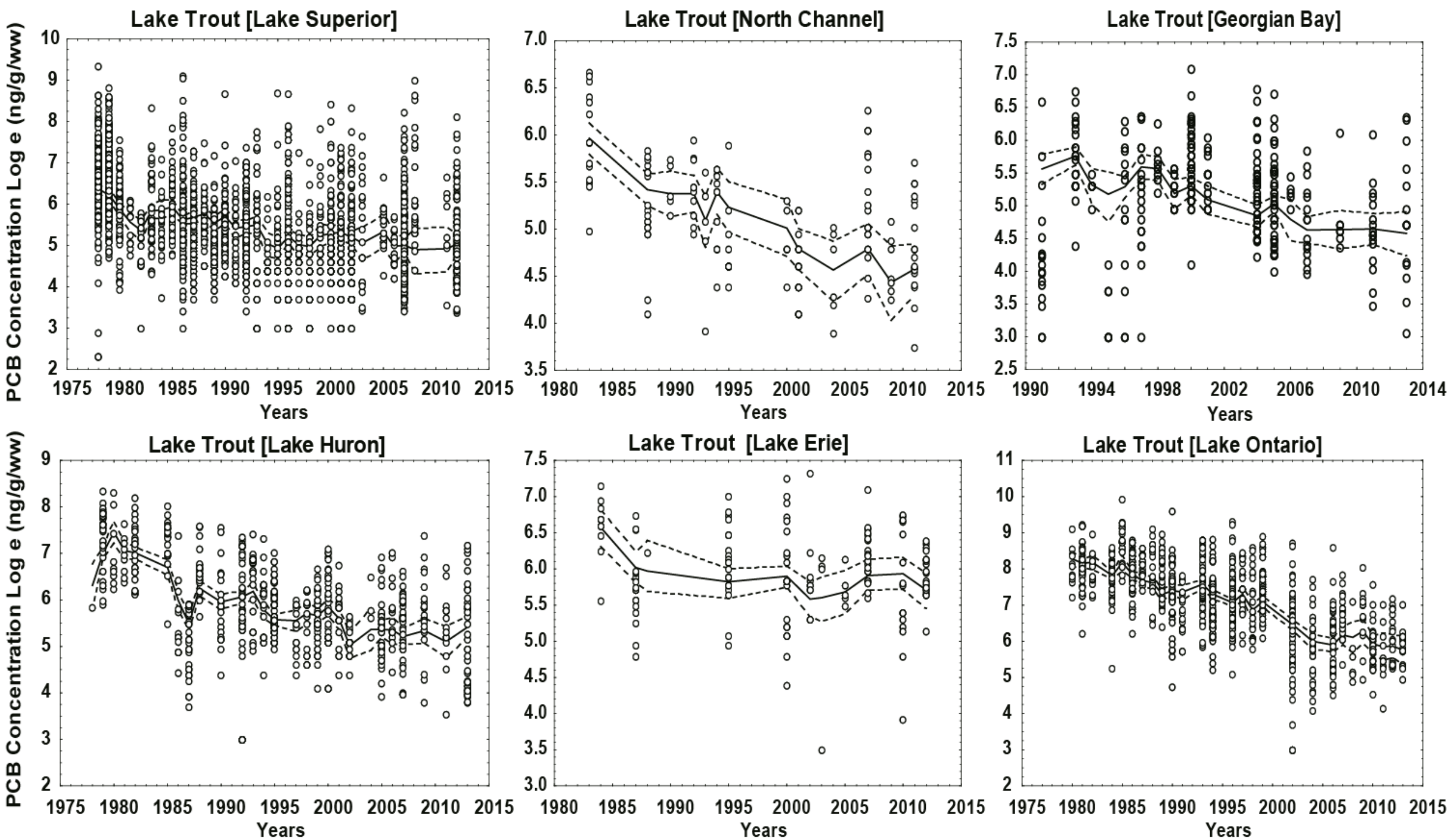


Walleye [Lake Superior]
Walleye [North Channel]
Walleye [Georgian Bay]
Walleye [Lake Huron]
Walleye [Lake Erie]
Walleye [Lake Ontario]
Walleye [St. Lawrence River]
PCB Concentration Log e (ng/g/ww)
PCB Concentration Log e (ng/g ww)
Years

Figure SI 3



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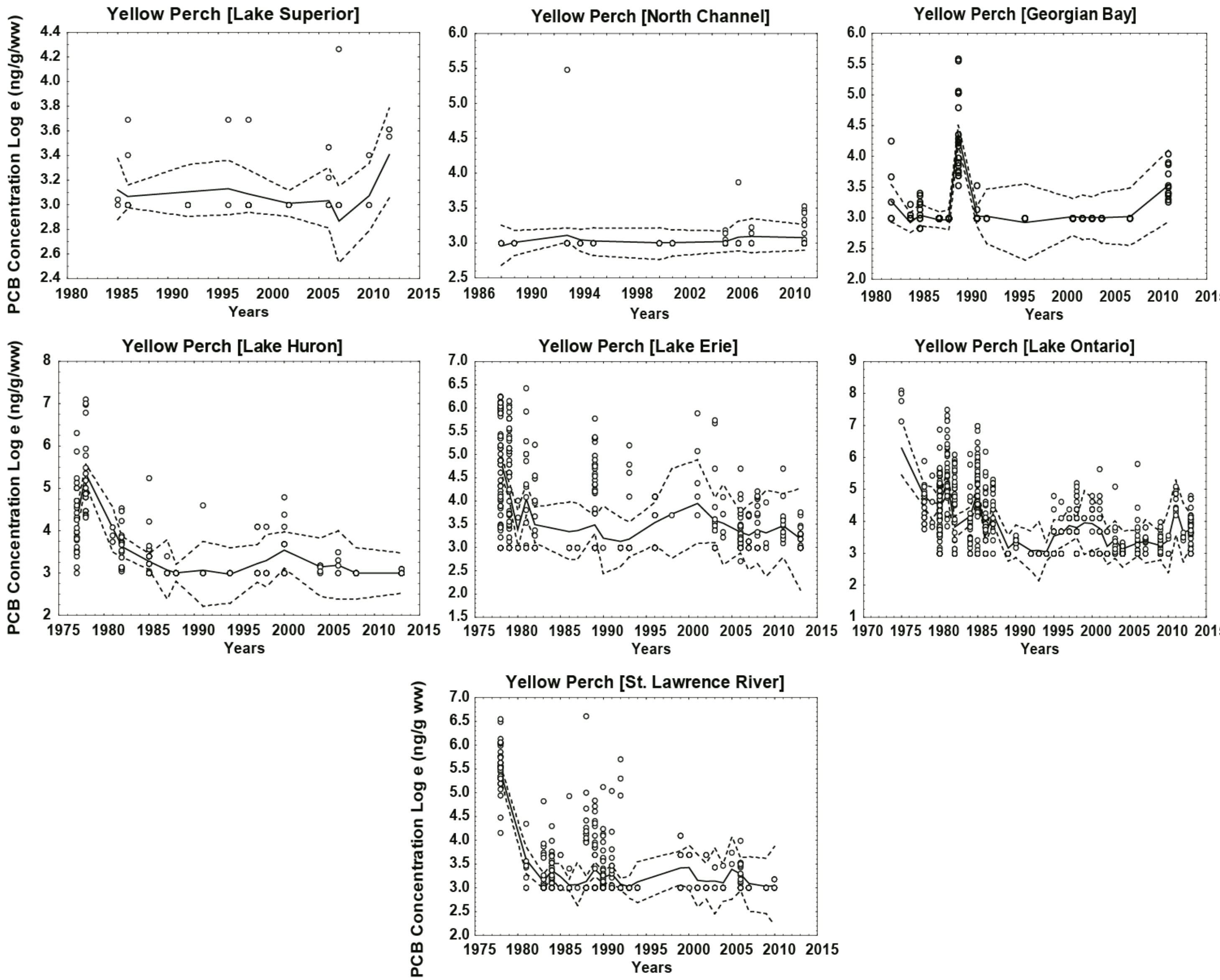

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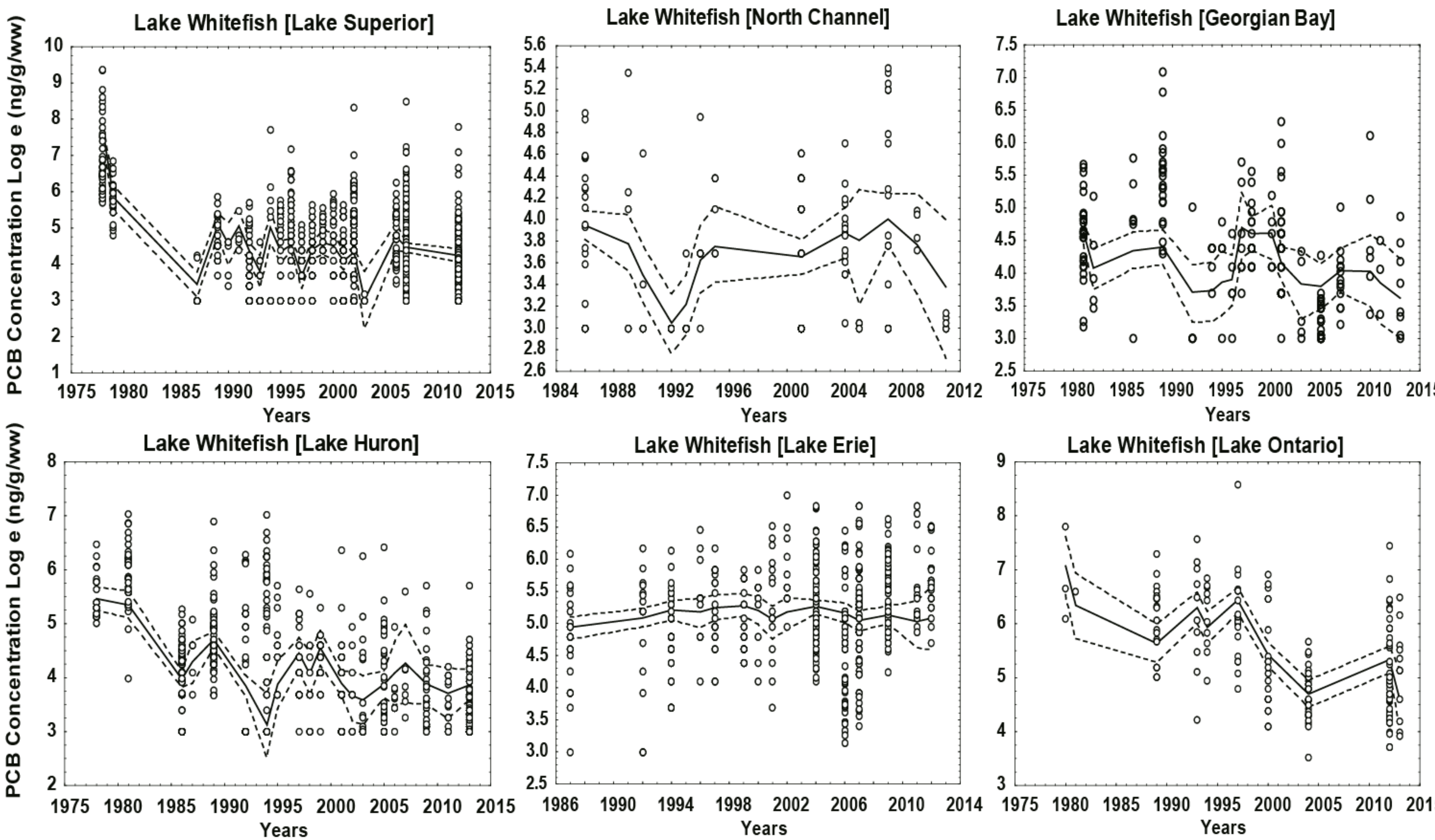

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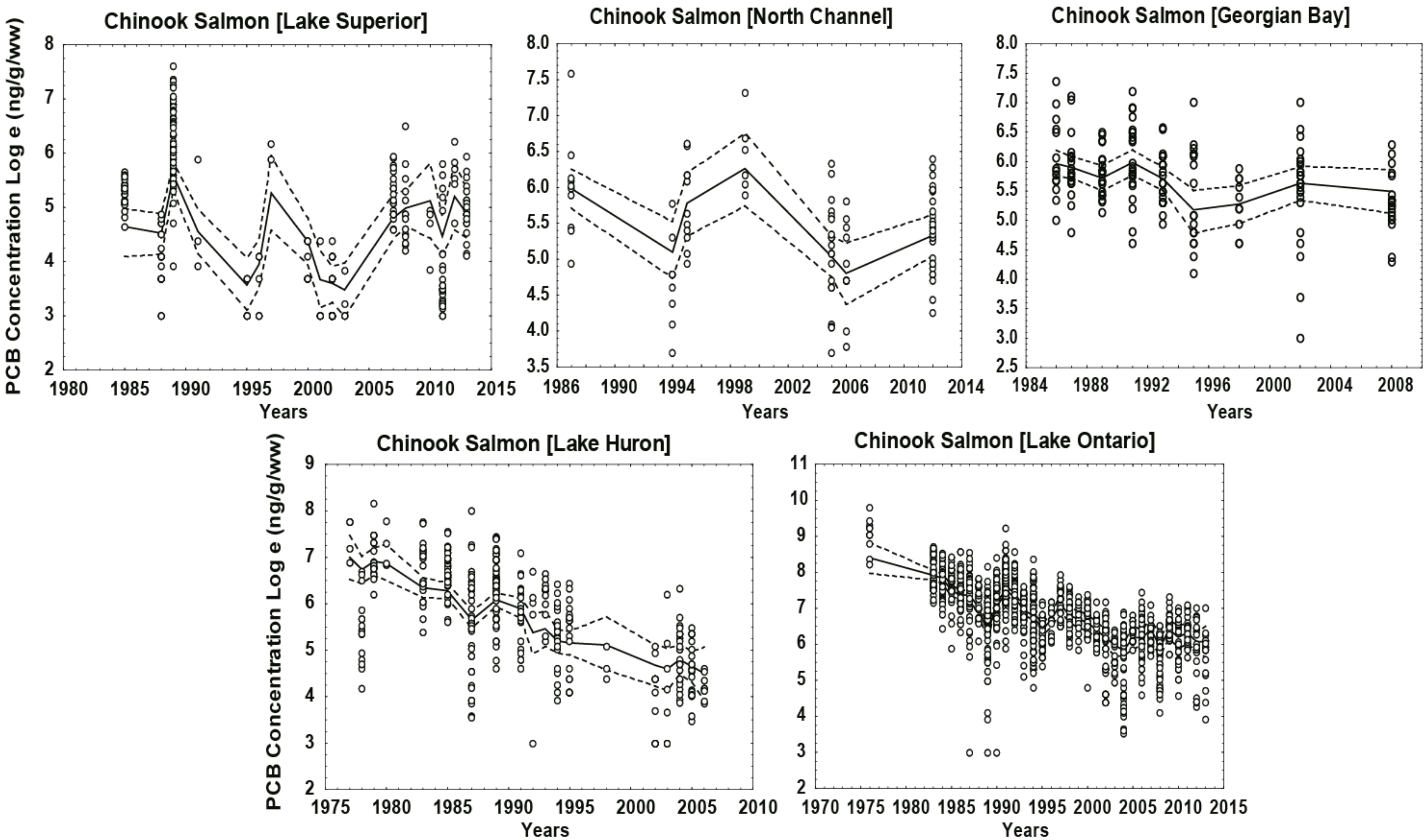

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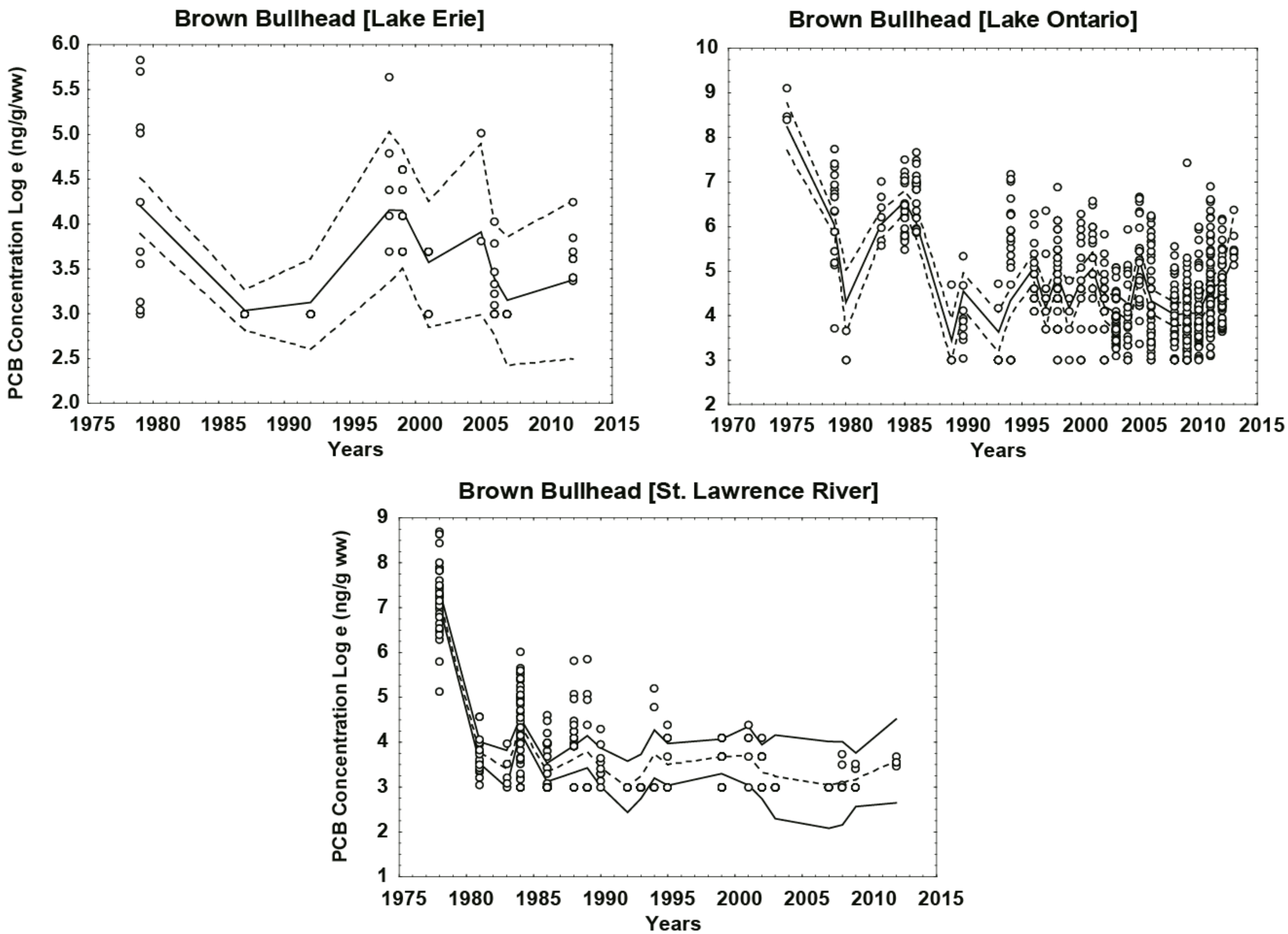

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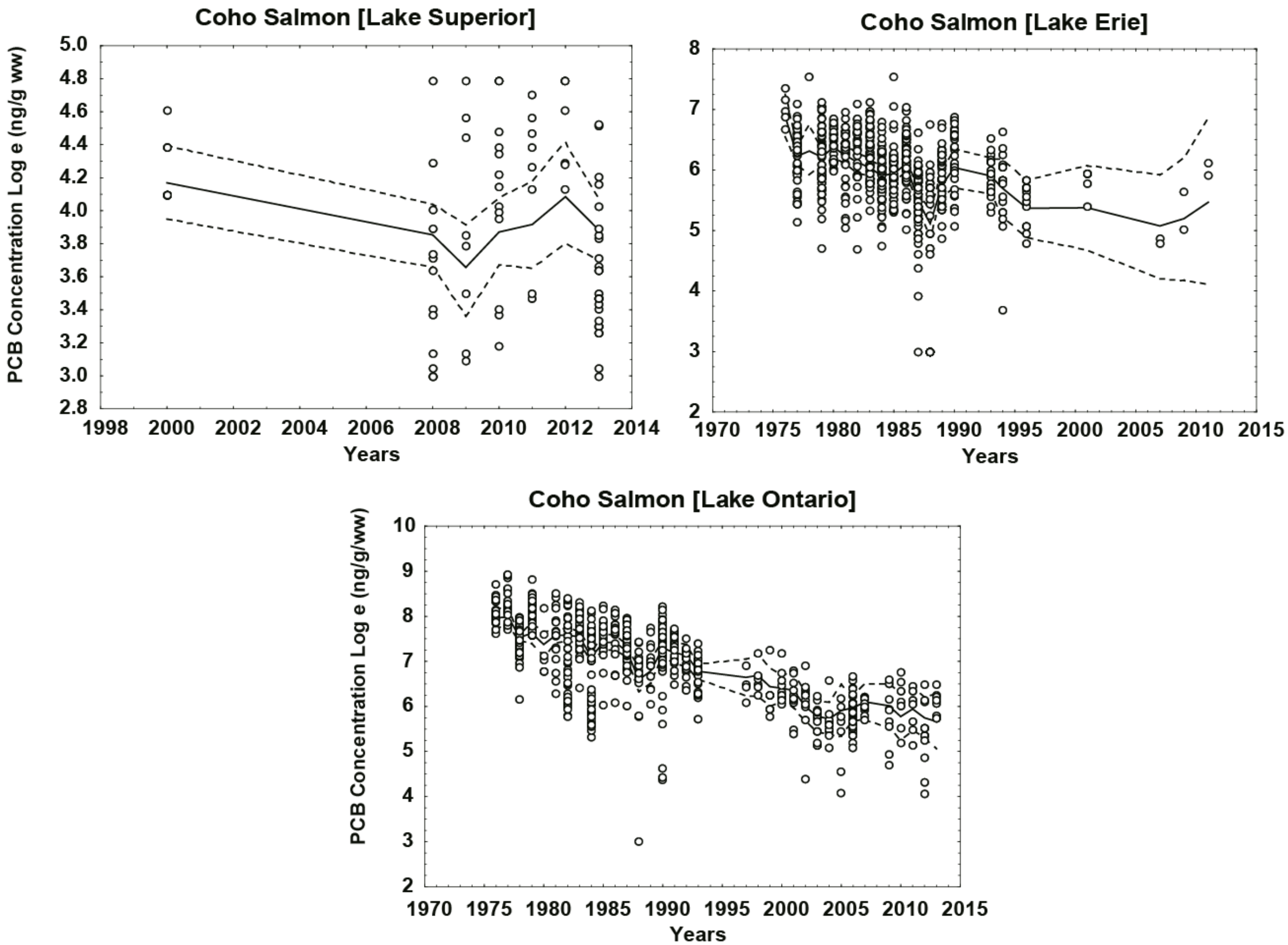

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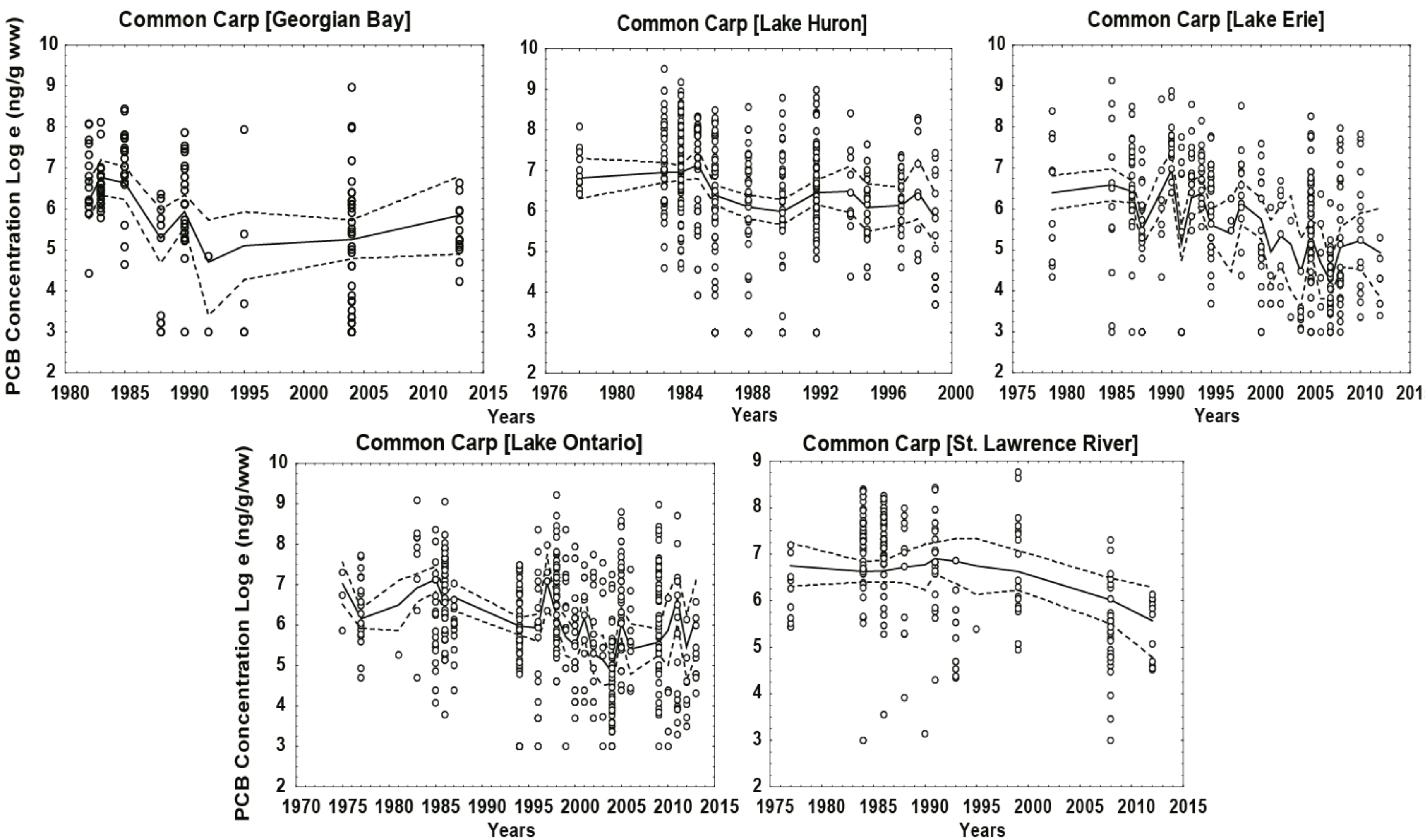

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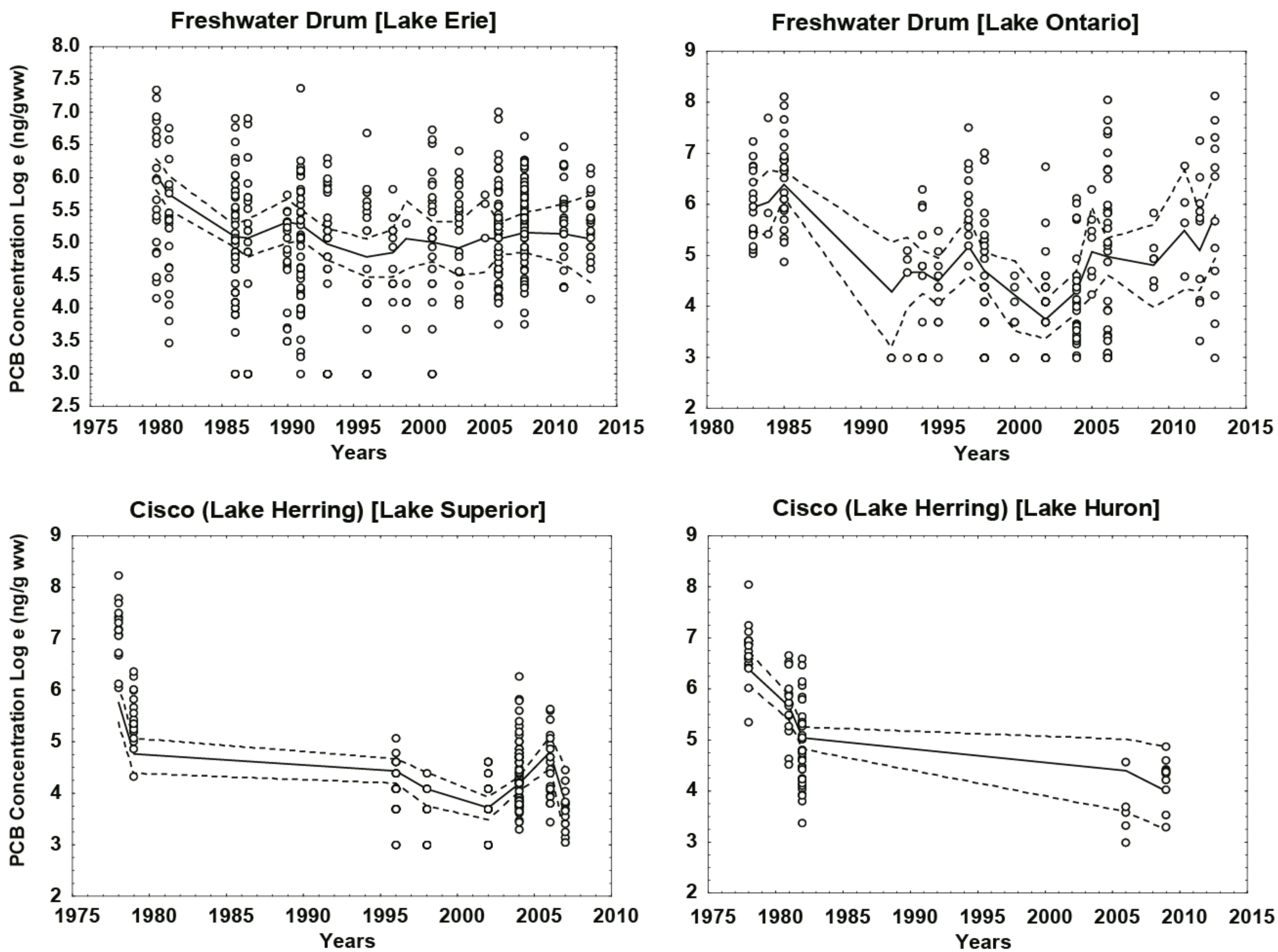

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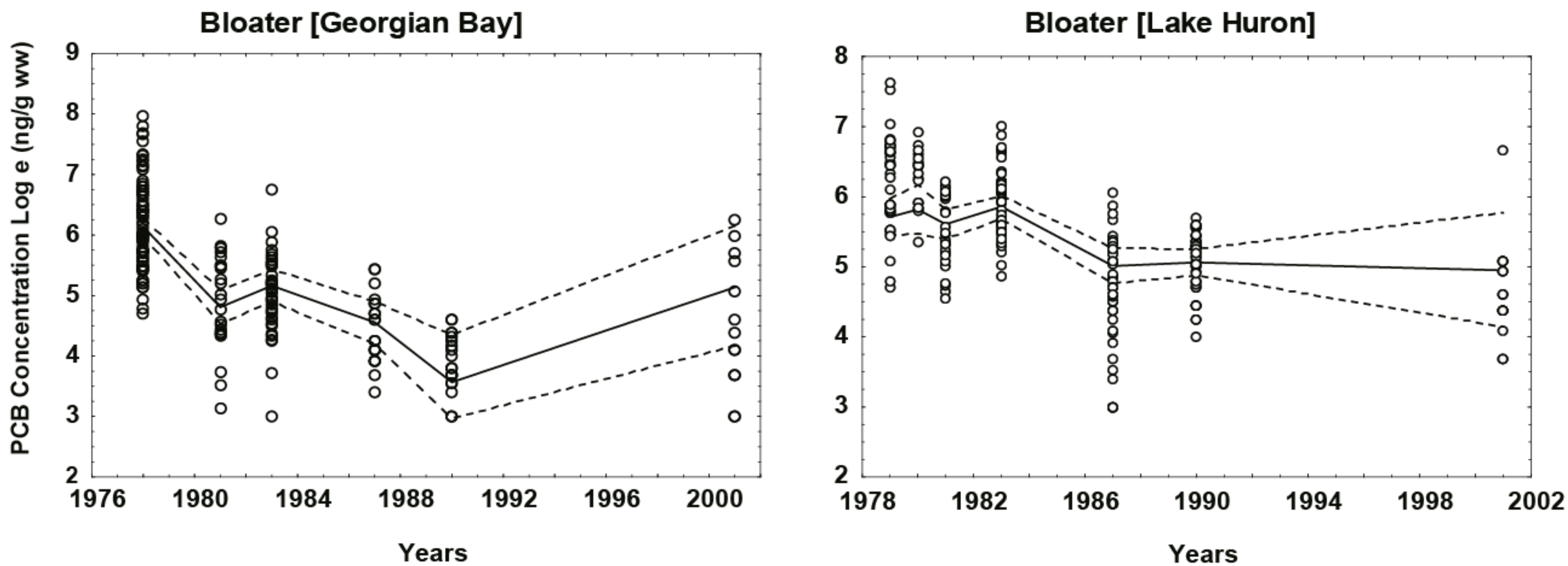

Figure SI 3 (Continued)