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# Guiding fish consumption advisories for Lake Ontario: A Bayesian hierarchical approach

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ABSTRACT

We evaluate the temporal trends of total mercury (THg) and polychlorinated biphenyls (PCBs) in walleye (*Sander vitreus*) and lake trout (*Salvelinus namaycush*) based on approximately 40 years of contaminant data from different locations in Lake Ontario. Bayesian inference techniques are employed to parameterize four hierarchical models. Our analysis provides evidence of distinctly declining trajectories for the two contaminants in lake trout. Likewise, walleye demonstrate a decreasing PCB trend, whereas no distinct temporal shifts were found in their THg rates of change. We illustrate the capacity of our statistical framework to aid in formulating fish consumption advisories by generating customizable probability of exceedance of THg and PCB threshold human exposure levels, based on their tolerable daily intake values. Walleye consumption results in 30% lakewide exceedance frequencies of the THg threshold for the sensitive demographic group of children less than 15 years old with an average body weight of 50 kg. Lake trout PCB threshold is frequently exceeded (>80%) in all of the study sites, whereas exposure to THg through lake trout consumption appears to be within the acceptable levels for human health. The overall trends indicate that the reduced contaminant emissions have brought about positive changes in the fish contamination levels in Lake Ontario.



## Introduction

Lake Ontario is the 14th largest lake in the world, with the smallest surface area (18,960 km$^2$) among the North American Great Lakes, mean depth of 85 m, and a drainage basin that covers 64,030 km$^2$. Similar to the rest of the Great Lakes, the lake has experienced a significant degree of anthropogenic stress. Historically, the Niagara River has been a focal source for contaminant loading into Lake Ontario, where the discharged contaminants settle and ultimately accumulate into the three major depositional basins (Niagara, Mississauga, Rochester) (Durham and Oliver, 1983; Marvin et al., 2003). Influenced by industrial activities in the watersheds and along major tributaries, including the Niagara River, levels of various contaminant classes such as polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins and dibenzofurans, organochlorine pesticides, and a host of metals (e.g., THg and Pb) in the sediments of Lake Ontario exceed their Canadian Sediment Quality Probable Effect Levels (PELs) (Marvin et al., 2003). The persistent and biologically active nature of these contaminants can reverberate through the biotic communities of the lake. The presence of these contaminants is expressed as variable amounts within the aquatic food web, most commonly encountered in higher trophic levels, such as top predatory fish species.

Fish communities are perceived as optimal aquatic ecological indicators as well as standard health risk precursors for human consumers in many cases. Among the key features of contaminants like PCBs or many other persistent organic pollutants (POPs) is their high degree of hydrophobicity and bioaccumulation potential (Tilden et al., 1997; Johnson et al., 1999). Even though the production and emission of most of the legacy contaminants in the Great Lakes system have been ceased or curtailed, there is an extended lag time before these contaminants decline to negligible levels due to their persistent nature (Burger and Gochfeld, 2006). Recent studies from Lake Ontario suggest that although fish contaminant levels are generally either still declining or stable, there are some weak increasing trends and considerable seasonal, species- and gender-specific differences (Bhavsar et al., 2007, 2010; Carlson et al., 2010; Madenjian et al., 2010, 2011; Zhang et al., 2012). Reports on potentially ailing health of individuals who consumed large quantities of Great Lakes sport fish have prompted research to shed light on the causal linkages between public health and exposure to contaminants via fish consumption. An insightful study of the blood contaminant levels among Ontario sport fish eaters revealed that THg levels in anglers eating fish from areas of concern (AOC) were higher than those detected in other Great Lakes populations (Cole et al., 2004). Sensitive populations, such as women of child-bearing age, children under the age of 15, who are still experiencing hormonal development, and indigenous native tribes that rely on subsistent fishing from Great Lakes could be at a greater risk of exposure. To protect the public from the harmful effects of ingested toxic substances, fish consumption

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advisories are issued to promote voluntary restriction of potentially tainted fish, while also reminding consumers about the benefits of fish consumption (Tilden et al., 1997).

A typical source of uncertainty underlying the consumption advisories is the adoption of reference concentrations, representing estimated values of the daily human exposure to a particular contaminant that will not result in adverse health effects over a lifetime, e.g., health protection value, tolerable daily intake, and minimal risk level (Scherer et al., 2008). These values emerge from the extrapolation of toxicity data from animals to humans as well as from the challenge to accommodate different tolerance levels in humans as a function of the fish meal sizes, human weights, amount of contaminants remaining in fish after cooking, frequency of consumption, and cancer risk factors (Scherer et al., 2008). The second potential source of error arises from the selection of the optimal statistical/modeling framework to produce the advisories, depending on the amount/quality of data, the regional characteristics, and the selection of the most parsimonious models to delineate the covariance of contaminant trends with the size (or other morphological features/physiological properties) of different fish species. Other unaccounted factors include the partitioning properties, metabolic transformation half-life, and synergistic effects of multiple chemicals in humans (Scherer et al., 2008; Binnington et al., 2014). Acknowledging the ubiquitous uncertainty pertaining to the development of fish consumption advisories, there are several recent attempts to introduce probabilistic methods in the risk assessment paradigm (Roberts et al., 2007; Harris and Jones, 2008; Mahmood et al., 2013a). Despite the aversion of stakeholders and decision makers when confronted with a "range" of values instead of a "fixed" value (Hope et al., 2007), these probabilistic approaches have the ability to accommodate the aforementioned sources of uncertainty or to more faithfully depict the implications of the presence of "outliers" in fish populations (Johnston and Snow, 2007; Mahmood et al., 2013a; Visha et al., 2015).

In this study, we introduce a Bayesian framework founded upon a series of hierarchical models that aim to address two critical issues. First, we attempt to detect the temporal trends of THg and PCBs in two important members of the Lake Ontario food web, walleye and lake trout, and to shed light on significant ecological mechanisms that may be driving the year-to-year variability in different locations of Lake Ontario. Second, our Bayesian framework is illustratively applied to support fish consumption advisories while explicitly accommodating the uncertainty pertaining to the model structure, select parameters, spatial heterogeneity, and variability in fish characteristics. We demonstrate capacity of the proposed approach to aid in formulating advisories by generating customizable probability of exceedance of THg and PCB threshold human exposure levels through the consumption of fish of different lengths and lipid contents. Given the wide array of both known and unknown factors that can conceivably contribute to the detected contaminant trends, the present study aims to elucidate some of the challenges in establishing a general framework for fish consumption advisories.

## Methods

We used fish contaminant data for lake trout and walleye from the Ontario Ministry of the Environment and Climate Change (OMOECC) Fish Contaminant Monitoring Program (Electronic Supplementary Material Section A. The samples were collected from nine (9) locations for lake trout and six (6) sites for walleye in the Canadian waters of Lake Ontario (Fig. 1). The number of years sampled per fish species and location sampled are provided in Electronic Supplementary Material (ESM) Tables S1–S4. The summary statistics for THg and PCB concentrations in walleye and lake trout suggest that walleye had higher THg levels (mean 0.38 and median 0.26 μg $g^{-1}$wet weight or ww) compared to lake trout (0.21 and 0.20 μg $g^{-1}$ww), whereas the higher lipid content of lake trout had considerably higher PCB concentrations (2076 and 1513 ng $g^{-1}$ww) compared to walleye (220 and 100 ng $g^{-1}$ ww) (ESM Table S5 and Figs. 2–5). ESM Tables S6–S7 report the same basic statistics for the lipid content (%) and the length (cm) of the two fish species studied. In particular, we note that the lipid content in lake trout was nearly nine times higher relative to walleye. By contrast, minimal differences were found between the two species with respect to their mean lengths, although walleye was characterized by considerable variability; partly stemming from the fact that walleye females tended to be distinctly longer (≈8 cm) relative to their male counterparts (Visha et al., 2015).

Hierarchical Bayesian modeling was used to detect the temporal THg and PCB trends in fish, while accommodating the variability among the

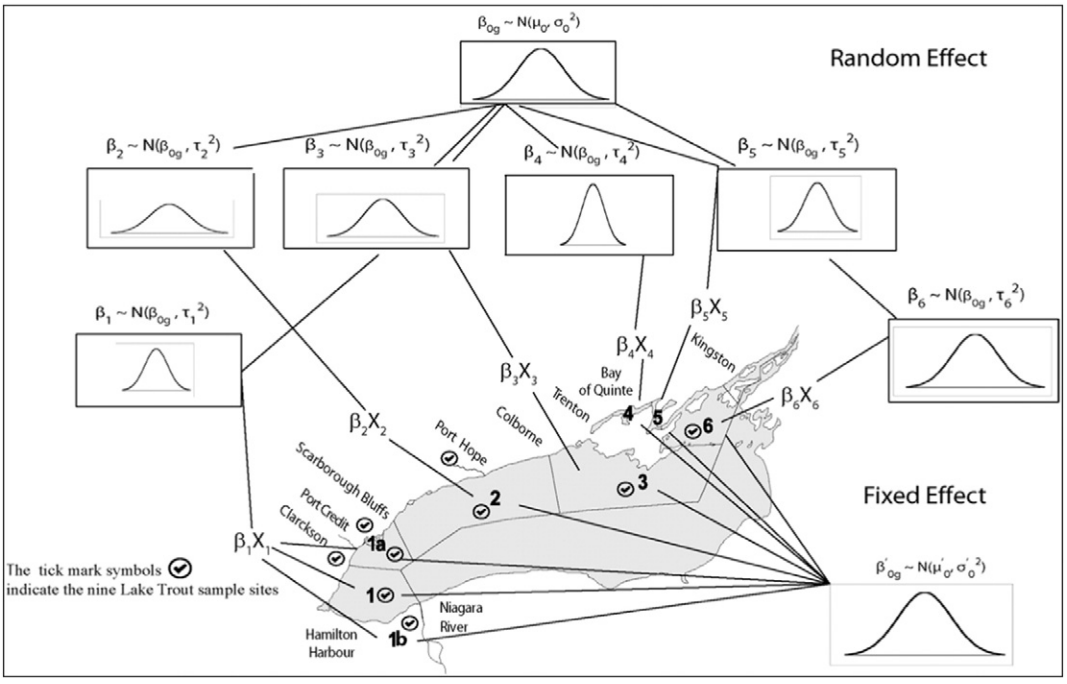


**Fig. 1.** Hierarchical modeling framework and fish sampling locations in Lake Ontario. Fixed parameters are drawn from the same probability distribution over all the locations. Random parameters are assigned location-specific probability distributions which in turn are drawn from a global (lakewide) hyperparameter.

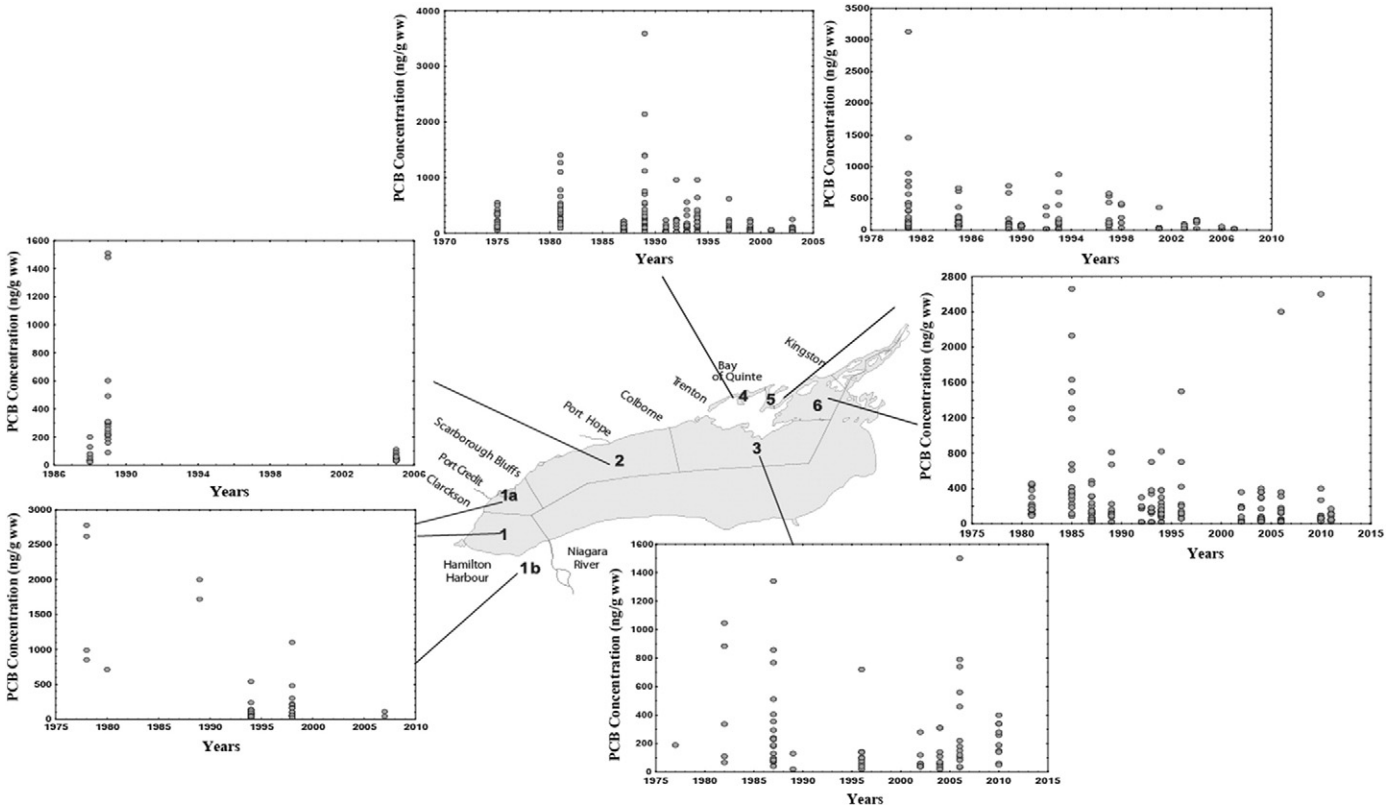


**Fig. 2.** PCB concentrations (ng/g ww) for walleye in six sampling locations in Lake Ontario.

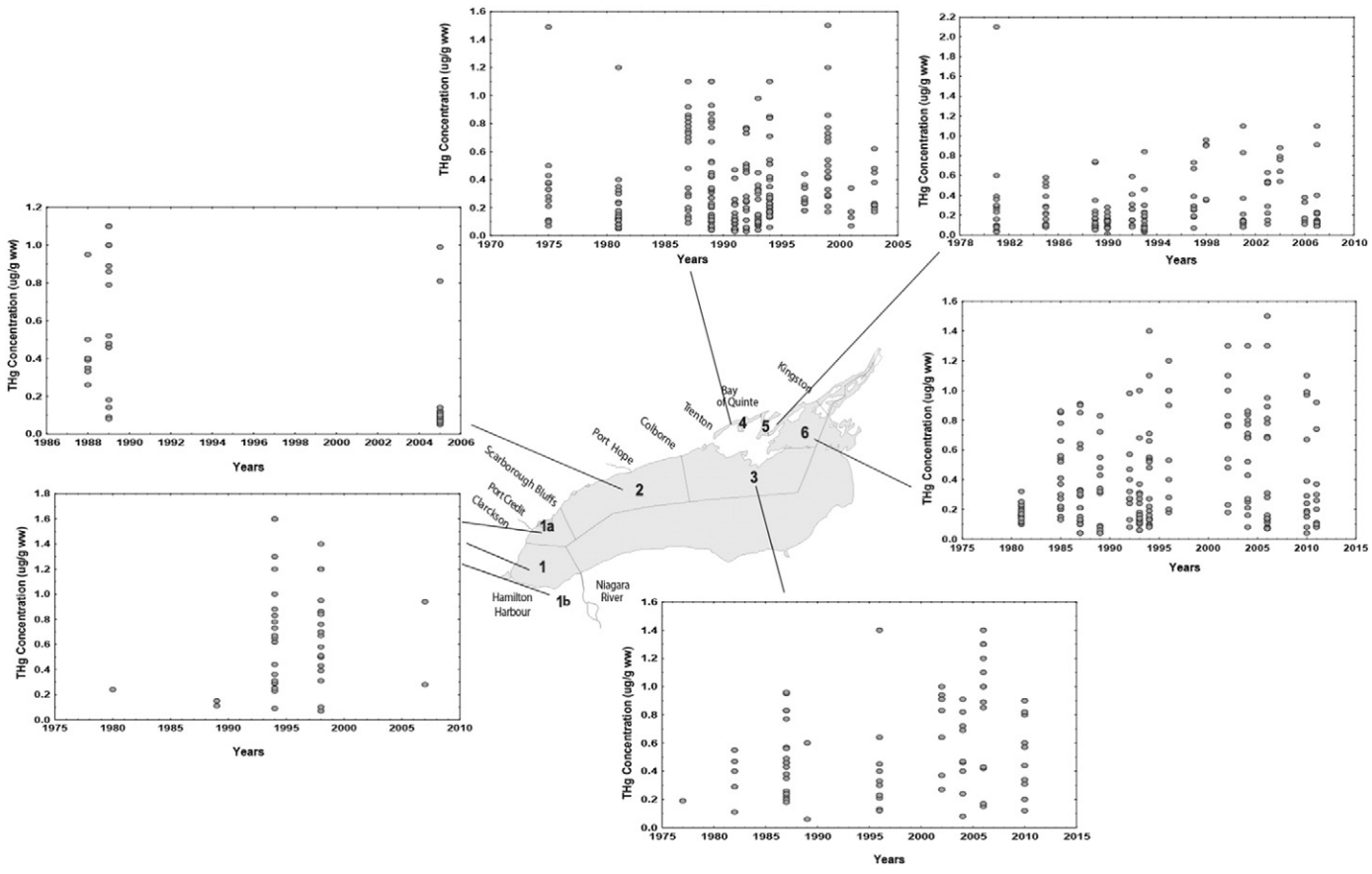


**Fig. 3.** THg concentrations (μg/g ww) for walleye in six sampling locations in Lake Ontario.

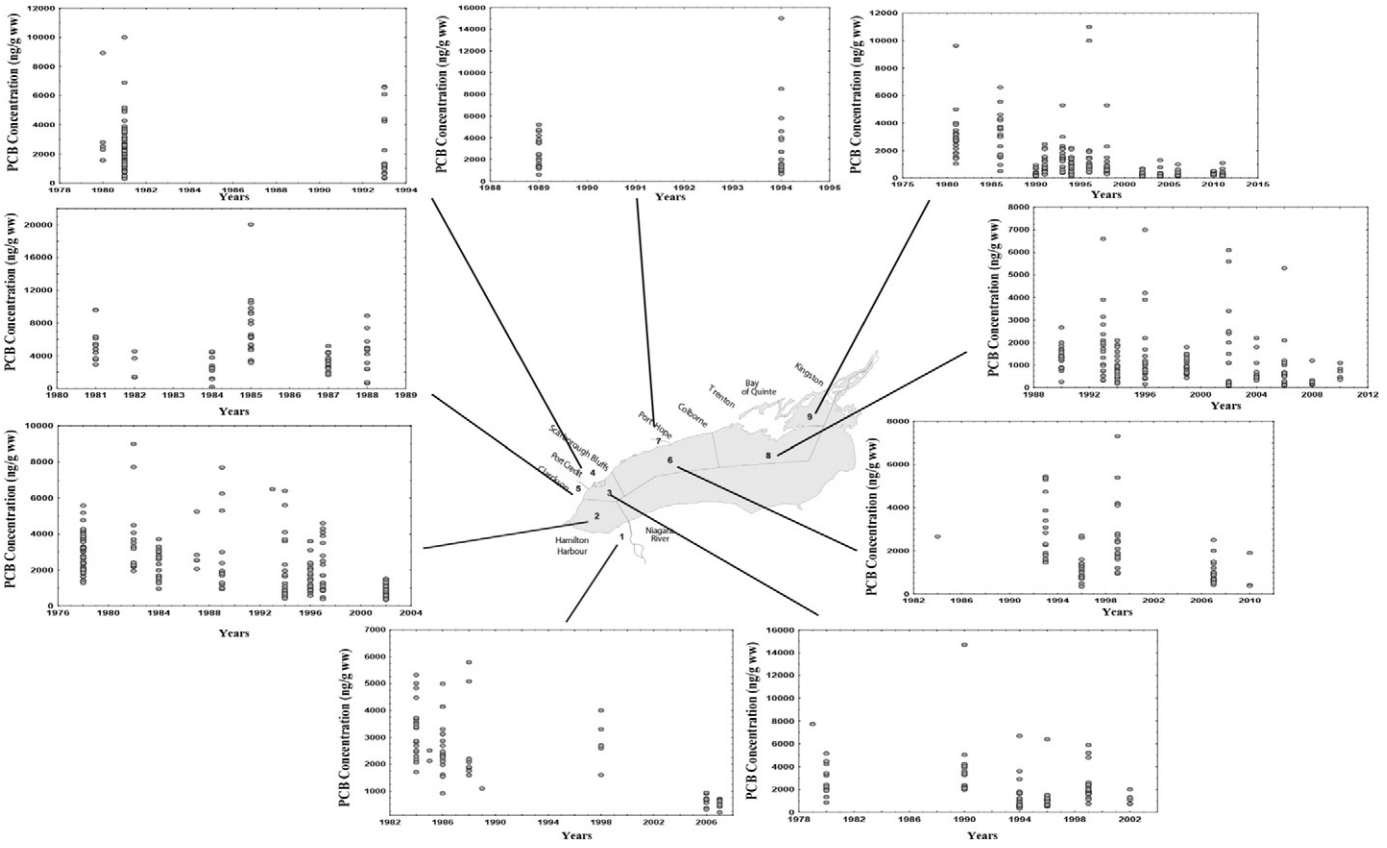


**Fig. 4.** PCB concentration (ng/g ww) for lake trout in nine sampling locations in Lake Ontario.

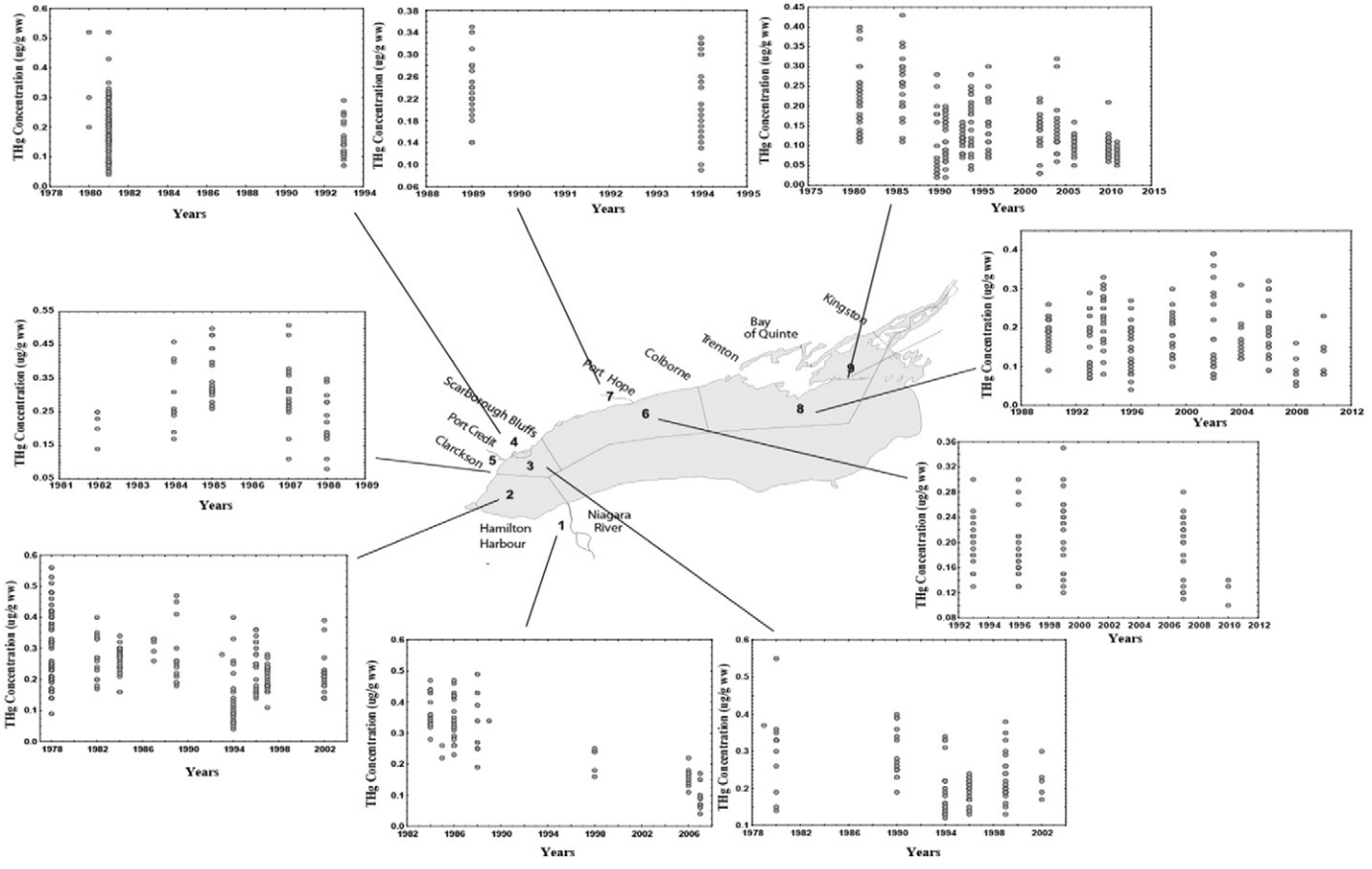


**Fig. 5.** THg concentration (μg/g ww) for lake trout in nine sampling locations in Lake Ontario.

sampling locations (Clark, 2005). Bayesian hierarchical modeling offers an effective methodological framework to exploit disparate sources of ecological information, to disentangle complex ecological patterns, to accommodate tightly intertwined environmental processes operating at different spatiotemporal scales, and to explicitly consider the variability pertaining to latent variables or other inherently immeasurable quantities (Borsuk et al., 2001; Wikle, 2003; Cheng et al., 2010; Liu et al., 2011; Cha and Stow, 2014). Simply put, in the context of regression analysis, a hierarchical model has two distinguishing features: first, the data are structured in groups and the model itself has its own hierarchical configuration, with the parameters of the within-group regressions at the bottom, controlled by the hyperparameters of an upper-level model (Gelman and Hill, 2007). With the hierarchical model structure, we can potentially overcome problems of insufficient group-specific data by "borrowing strength" from well-studied modeled units (Cheng et al., 2010). This feature is particularly important for the present study, as our data set is characterized by understudied locations with extended data gaps and sites that have been sampled consistently throughout the study period.

We introduce four hierarchical structures to evaluate the spatiotemporal contaminant trends in walleye and lake trout while accounting for the potential impact of the invasion of dreissenids along with the covariance of the contaminant concentrations with fish length and lipid content (Table 1; see also mathematical descriptions in ESM Section B). Generally, studies (Bhavsar et al., 2007, 2010) implicitly assume that the contaminant–length relationship reflects the longer exposure to contaminants of older age (and thus larger) fish, but this relationship can be modulated by their diet, behavioural patterns, and growth rate. Fish lipid content stands out as a potentially important covariate that has received considerable attention in the literature. Although there are contradictory results regarding the strength of the causal linkage between fish lipid content and contaminant levels, recent research has rendered support to the hypothesis that within an individual, contaminants accumulate in lipids, but lipid concentration may be unimportant in the mechanism governing contaminant assimilation. In this study, the two covariates were standardized prior to the analysis, and thus the corresponding regression coefficients are comparable and assess the relative strength of the relationships between fish length/lipid content and the two contaminants, while the model intercepts reflect the mean contaminant levels in each location at the beginning (first year) of the study period for a fish individual with average length and lipid content. Similar to the practice followed by other studies (e.g., Stow et al., 2004), the two covariates (fish length and lipid content) were subject to natural logarithmic transformation, and then average and standard deviation values of the ln-transformed data were calculated across all the study years and sampling sites. The series of ln-transformed lengths and lipid content values were standardized, based on these grand means and standard deviations, prior to use as predictors for ln[PCB]/[Hg]. The different assumptions regarding the spatial character (constant or variant) of the four terms considered (i.e., initial contaminant levels for the average fish individual, covariance of contaminants with fish length and lipid content, and temporal contaminant trends) were the primary drivers of the complexity of the four hierarchical configurations (Fig. 1).

The first model (Model 1) aims to evaluate the lakewide THg/PCB temporal trends; namely, the change of the fish contaminant levels over time is postulated to be uniform across the entire Lake Ontario. The same model explicitly considers the spatial differences of the initial contaminant levels (model intercepts) as well as the variability of the relationships between contaminants and fish length/lipid content among the six and nine sites in Lake Ontario, where the walleye and lake trout samples were collected, respectively. The distinct feature of the second model (Model 2) is its capacity to delineate the contaminant temporal trends in each sampling location rather than assuming uniform rates of changes over the entire system. Similar to Model 1, the model intercept maintains its location-specific character, whereas the signature of the fish length/lipid content on contaminant variability is assumed to be spatially constant. The third model (Model 3) evaluates the lakewide fish contaminant trends over time, while explicitly considering the effects of the invasion of dreissenids on the average THg/PCB levels. Food web structural shifts induced by dreissenid mussels have been hypothesized to be responsible for the recent fish contaminant trends in the Great Lakes (Bhavsar et al., 2007). With this model, we assume the influence of the dreissenids on the contaminant levels as an abrupt shift in the average lakewide conditions, based on two spatially constant intercepts for the pre- and post-invasion periods. The fish contaminant–length/lipid relationships are assumed constant across all sampling locations. The fourth model (Model 4) aims to evaluate the lakewide contaminant trends before and after the invasion of dreissenids, while explicitly considering the variations of the initial conditions as well as the covariance with fish length and lipid content in different locations of the lake. The implementation of this model again aims to determine whether the contaminant trends were affected by the presence of dreissenids. By contrast to Model 3 though, this model introduces a second linear trend after the year of the invasion, assumed to be the year of 1995, instead of postulating a distinct shift to the average contaminant levels during the post-invasion period.

We used the WinBUGS software (version 1.4.x) (Spiegelhalter et al., 2003) to obtain Markov chain Monte Carlo (MCMC) simulations and thus generate sequences of realizations from the model posterior distributions (Gilks et al., 1998). We used a general normal proposal Metropolis algorithm that is based on a symmetric normal proposal

**Table 1**
Conceptual foundation of the four hierarchical models used to evaluate the spatiotemporal THg/PCB patterns in walleye and lake trout in Lake Ontario. Sample $i = 1 \ldots N_j$; location $j = 1 \ldots M$; year $t = 1 \ldots T$ where $N_j$, $M$, and $T$ are the total number of samples in location $j$, the total number of sampling locations, and the total number of years, respectively; $t_0$ is the initial year of the analysis; $t_{inv}$ is the year of the invasion of dreissenids, assumed to be the year of 1995.The covariates fish length and lipid content were standardized prior to the analysis, and thus the intercepts reflect the contaminant concentrations at the beginning of our study period for a fish individual with average length and lipid content.

| | | Initial contaminant levels for the average fish individual | Covariance of contaminant levels with fish length | Covariance of contaminant levels with fish lipid content | Temporal contaminant level trends |
|---|---|---|---|---|---|
| Model 1 | Assumption | Location-specific | Location-specific | Location-specific | Constant across Lake Ontario |
| | Parameterization | $\beta_{0j}$ | $\beta_{\text{length}j}\text{length}_{ijt}$ | $\beta_{\text{lipid}j}\text{lipid}_{ijt}$ | $\beta_{\text{year}}(t_{ij} - t_0)$ |
| Model 2 | Assumption | Location-specific | Constant across Lake Ontario | Constant across Lake Ontario | Location-specific |
| | Parameterization | $\beta_{0j}$ | $\beta_{\text{length}}\text{length}_{ijt}$ | $\beta_{\text{lipid}}\text{lipid}_{ijt}$ | $\beta_{\text{year}j}(t_{ij} - t_0)$ |
| Model 3 | Assumption | Two spatially constant intercepts for the pre- and post-invasion periods | Location-specific | Location-specific | Location-specific |
| | Parameterization | $\beta_{0k}$ | $\beta_{\text{length}j}\text{length}_{ijt}$ | $\beta_{\text{lipid}j}\text{lipid}_{ijt}$ | $\beta_{\text{year}j}(t_{ij} - t_0)$ |
| Model 4 | Assumption | Location-specific | Location-specific | Location-specific | Constant across Lake Ontario but different for the periods before and after the invasion of dreissenids |
| | Parameterization | $\beta_{0j}$ | $\beta_{\text{length}j}\text{length}_{ijt}$ | $\beta_{\text{lipid}j}\text{lipid}_{ijt}$ | $\beta_{\text{year}1}t'_{ij} + \beta_{\text{year}2}\,t''_{ij}$<br>$t'_{ij} = t_{ij} - t_0$ if $t_{ij} < t_{\text{inv}}$ else 0<br>$t''_{ij} = t_{ij} - t_{\text{inv}}$ if $t_{ij} > t_{\text{inv}}$ else 0 |

distribution, whose standard deviation is adjusted over the first 4000 iterations, so that the acceptance rate ranges between 20% and 40%. For each analysis, we used three chain runs of 100,000 iterations, keeping every 20th iteration (thin of 20) to minimize serial correlation. Convergence of the MCMC chains was checked using the Brooks–Gelman–Rubin (BGR) scale-reduction factor (Brooks and Gelman, 1998). This diagnostic is based on analyzing multiple simulated MCMC chains by comparing the variances within each chain and the variance between chains. Large deviation between these two variances indicates non-convergence, while values close to 1 suggest that each of the multiple chains has stabilized, and they are likely to have reached the target distribution. The latter condition was met for each of the model parameters considered. 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 (Gilks et al., 1998).

*Fish consumption advisories*

The illustration of our Bayesian approach to fish consumption advisories was based on the predicted contaminant concentrations from our hierarchical models at each study site. We established thresholds for each contaminant based on their tolerable daily intake (TDI) values and were then able to make predictive statements about the probability of exceeding critical levels of that contaminant through consumption of fish of a specific size and lipid content. For the purpose of prediction, the Bayesian approach generates a posterior predictive distribution that represents the current estimate of the value of the response variable (THg and PCB levels), taking into account both parametric uncertainty and structural error (model misfit), and therefore probabilistically depicts the risks associated with fish consumption (Mahmood et al., 2013a,b). Because the apparent error rate (Efron, 1986) or the observed inaccuracy of the fitted model applied to the original data usually underestimates its actual capacity to predict future observations (true error rate), we base our risk assessment analysis on the most parsimonious rather than the highest performing but likely over-fitted model. The determination of the most parsimonious model for each fish species/contaminant combination was based on the use of the deviance information criterion (DIC) values, a Bayesian measure of model fit and complexity, where models with lower DIC values are expected to effectively balance between predictive capacity and complexity (Spiegelhalter et al., 2003).

For illustration purposes, we focused on the sensitive demographic group of children less than 15 years old with an average body weight of 50 kg. Our focus on children under the age of 15 is in compliance with the fish consumption guidelines proposed by the OMOECC, as this cutoff age is often considered the threshold of puberty, and therefore any contaminant related to neurological disorders would be more harmful at this stage of development. We choose 6 meals per month as a regular fish intake. Similar to the value used by OMOECC when producing their established advisories, we used a standard fish meal size of 227 grams in our analysis. Our next step was to calculate critical thresholds for each contaminant. The TDI values for THg and total PCBs were obtained from OMOECC. The TDI is defined as the maximum allowable daily intake of a substance that, if consumed over a lifetime, will not lead to adverse health effects (Health Canada, 1996). TDI values are generally expressed for a specific body weight, such as μg per kg of body weight (or kgbw) per day. Specifically, we used the values of 0.52 μg THg/kgbw per day and 90 ng PCB/kgbw per day and calculated the thresholds for each of the hypothetical scenarios as follows:

$$\text{Threshold} = (\text{human weight[kg]} \times \text{TDI[ng/kgbw/month]}) / (\text{meal size} \times \text{meal number})$$

After establishing critical thresholds, we calculated any changes in the frequency of exceedances of each contaminant, using the observed Lake Ontario contaminant levels in 1985, 1995, and 2005 across the fish length and lipid content values. Recognizing the need to develop advisories that integrate across multiple contaminants, we also conducted a post hoc exercise in which the previously described models were subject to an alternative Bayesian parameter estimation that explicitly accommodates the covariance of the two contaminants in each species. In doing so, we were able to draw inference regarding the joint probability to exceed the critical thresholds for the two contaminants during fish consumption. The statistical characterization of this exercise was based on a bivariate normal likelihood in which the two means were provided by the most parsimonious models as selected by the univariate analysis, i.e., Models 2 (THg) and 4 (PCB) for lake trout and Models 4 (THg) and 3 (PCB) for walleye:

$$\begin{pmatrix} \log e\left(Hg_{MEASijt}\right) \\ \log e\left(PCB_{MEASijt}\right) \end{pmatrix} \sim N\left( \begin{pmatrix} \log e\left(Hg_{PREDijt}\right) \\ \log e\left(PCB_{PREDijt}\right) \end{pmatrix}, \begin{bmatrix} \psi^2_{Hg}, \psi_{Hg,PCB} \\ \psi_{Hg,PCB}, \psi^2_{PCB} \end{bmatrix} \right)$$

$$\psi_{Hg,PCB} = \rho \cdot \psi_{Hg} \cdot \psi_{PCB}$$

$$\psi_{Hg}{}^{-2} \sim G(0.001,\ 0.001) \psi_{PCB}{}^{-2} \sim G(0.001,\ 0.001)$$

$$i = 1....N_j \quad j = 1...M \quad t = 1...T$$

where $Hg/PCB_{MEASijt}$ and $Hg/PCB_{PREDijt}$ represent the measured and predicted contaminant levels in sample i, collected from location j and year $t$; $N$ and $G$ denote the normal and gamma probability distributions; and the correlation coefficient $\rho$ is assumed to be known and thus remained fixed during the model updating. Specifically, we used the correlation values of 0.60 for lake trout and 0.45 for walleye, as derived from the contemporaneous THg and PCB values of the Lake Ontario data set. The rest of the hierarchical configuration was similar to what was described with the single contaminant models. The joint predictive (risk assessment) statements will be illustrated for 2011, which was the last year of our study period.

## Results and discussion

*Spatiotemporal THg/PCB patterns*

The comparison of the four models on the basis of their DIC values indicated that Model 1, site-specific intercepts and contaminant–length/lipid relationships but spatially constant temporal trends, was never the most favorably supported hierarchical configuration from the contaminant/fish species combinations examined (Table 2). Specifically, the second and fourth formulations were the most parsimonious models to depict the THg and PCB trends in lake trout, respectively. Likewise, Models 4 and 3 represented more faithfully the THg and PCB patterns in walleye. [Comparisons between predicted and measured contaminant concentrations for each fish species and location are presented in ESM Figs. S1–S4, while the visualization of the parameter posteriors is

**Table 2**
Determination of the most parsimonious model (bold font) for each fish species/contaminant combination, based on the use of the deviance information criterion (DIC) values, a Bayesian measure of model fit and complexity, where models with lower DIC values are expected to effectively balance between predictive capacity and complexity (Spiegelhalter et al., 2002).

| Lake trout THg | | Lake trout PCBs | |
|---|---|---|---|
| Model 1 | 391.66 | Model 1 | 1567.79 |
| **Model 2** | **273.33** | Model 2 | 1581.33 |
| Model 3 | 415.26 | Model 3 | 1551.98 |
| Model 4 | 284.99 | **Model 4** | **1545.51** |
| **Walleye THg** | | **Walleye PCBs** | |
| Model 1 | 1089.66 | Model 1 | 2003.20 |
| Model 2 | 1098.53 | Model 2 | 2021.52 |
| Model 3 | 1094.59 | **Model 3** | **1995.97** |
| **Model 4** | **1087.14** | Model 4 | 1999.50 |

provided in ESM Figs. S5–S8).] First, we note that the spatial trends delineated by the relative values of the (site-specific) posterior intercepts were qualitatively on par with the spatial variability of the long-term averages recorded in different locations of Lake Ontario (Table 3). The highest intercept value ($-1.092 \pm 0.100$ $\log_e$[μg THg $g^{-1}$ ww]) was derived at the western portion of Lake Ontario (site 1), influenced by Niagara River and extending through St. Catharines to Hamilton Harbour, which displayed the highest mean THg levels in walleye (0.66 μg $g^{-1}$ww), followed by sites 2 (0.43 μg $g^{-1}$ww) and 3 (0.56 μg $g^{-1}$ww) that were located in the northern area of the lake. The Bay of Quinte (sites 4 and 5) was characterized by the lowest mean *THg* levels (≈0.30 μg $g^{-1}$ww), whereas the eastern portion of Lake Ontario – inshore areas near Kingston and the mouth of the St. Lawrence River (site 6) – demonstrated somewhat elevated concentrations (0.41 μg $g^{-1}$ww), which was also reflected in the corresponding posterior intercept ($-1.200 \pm 0.067$ $\log_e$[μg THg $g^{-1}$ ww]). In regards to the contaminant concentrations in lake trout, the area influenced by Port Credit River (site 5) had the highest THg (0.30 μg $g^{-1}$ww) and PCB concentrations (4614 ng $g^{-1}$ww) as well as the highest posterior intercept values ($-0.435 \pm 0.185$ $\log_e$[μg THg $g^{-1}$ ww] and $7.938 \pm 0.083$ $\log_e$[ng PCB $g^{-1}$ ww]). The lowest contaminant values were found in the northeastern portion of Lake Ontario (sites 8 and 9) with average PCB and THg levels approximately lower than 1250 ng $g^{-1}$ww and 0.18 μg $g^{-1}$ww, respectively. Likewise, the corresponding posterior intercepts were comparatively low for both contaminants (<−1.500 $\log_e$[μg THg $g^{-1}$ ww] and <7.350 $\log_e$[ng PCB $g^{-1}$ ww]) in the same locations.

Comparison of the spatial trends of sediment (Marvin et al., 2003) and fish contamination suggests a remarkable consistency with respect to the presence of a distinct west-to-east gradient in Lake Ontario. In particular, existing empirical and modeling evidence shows that the Niagara River is responsible for more than half of the total contaminant loading in Lake Ontario, whereby the contaminated sediments from the industrial activities in the adjacent watersheds are transported and ultimately deposited in the offshore areas of Lake Ontario (Marvin et al., 2004; Ethier et al., 2012). Importantly, despite the substantial improvement in sediment quality over the past four decades, sediment cores collected from the late 1990s showed that there were still frequent THg exceedances of the Canadian Sediment Quality Guidelines in many areas of Lake Ontario (>60%), especially in the three major depositional basins (Niagara, Mississauga, Rochester), where the fine grained sediments tend to accumulate (Marvin et al., 2003). The spatial PCB distribution in the sediments was similar, but none of the stations surveyed in Lake Ontario in 1998 exceeded the Canadian PEL for total PCBs (277 ng/g). Along the same line of reasoning, the high standard deviation and interquartile ranges reported herein reflect the substantial within- and among-year variability associated with the contaminant levels of fish individuals (ESM Table S5; see also Figs. 2–5). Although there is no single explanation for the significant temporal variability, but rather a complex interplay among a number of mechanisms (see also following discussion), one of the possible drivers could be the substantial atmospheric fluxes that conceivably contribute to the year-to-year variations in Lake Ontario (Atkinson et al., 2007). There is also

**Table 3**
Posterior parameter estimates (mean ± standard deviation) of the four hierarchical models developed to examine the spatiotemporal THg and PCB trends in walleye and lake trout from Lake Ontario. Definitions of all model parameters are provided in the Electronic Supplementary Material Section B.

| | Lake trout (THg) | | Lake trout (PCB) | | Walleye (THg) | | Walleye (PCB) | |
|---|---|---|---|---|---|---|---|---|
| | Parameter | Model 2 | Parameter | Model 4 | Parameter | Model 4 | Parameter | Model 3 |
| Baseline conditions for the average fish individual | $\beta_{01}$ | −0.750 ± 0.063 | $\beta_{01}$ | 7.847 ± 0.081 | $\beta_{01}$ | −1.092 ± 0.100 | $\beta_{01\ (1978–1994)}$ | 5.660 ± 0.174 |
| | $\beta_{02}$ | −0.986 ± 0.035 | $\beta_{02}$ | 7.810 ± 0.047 | $\beta_{02}$ | −1.275 ± 0.091 | $\beta_{02\ (1995–2011)}$ | 5.848 ± 0.345 |
| | $\beta_{03}$ | −0.998 ± 0.078 | $\beta_{03}$ | 7.850 ± 0.066 | $\beta_{03}$ | −1.292 ± 0.081 | | |
| | $\beta_{04}$ | −1.047 ± 0.045 | $\beta_{04}$ | 7.870 ± 0.071 | $\beta_{04}$ | −1.349 ± 0.058 | | |
| | $\beta_{05}$ | −0.435 ± 0.185 | $\beta_{05}$ | 7.938 ± 0.083 | $\beta_{05}$ | −1.415 ± 0.063 | | |
| | $\beta_{06}$ | −1.580 ± 0.146 | $\beta_{06}$ | 7.689 ± 0.094 | $\beta_{06}$ | −1.200 ± 0.067 | | |
| | $\beta_{07}$ | −1.600 ± 0.301 | $\beta_{07}$ | 7.454 ± 0.155 | | | | |
| | $\beta_{08}$ | −1.612 ± 0.112 | $\beta_{08}$ | 7.340 ± 0.060 | | | | |
| | $\beta_{09}$ | −1.507 ± 0.042 | $\beta_{09}$ | 7.355 ± 0.056 | | | | |
| Length Effect | $\beta_{length}$ | 0.391 ± 0.013 | $\beta_{length1}$ | 0.273 ± 0.112 | $\beta_{length1}$ | 0.779 ± 0.074 | $\beta_{length1}$ | 0.226 ± 0.140 |
| | | | $\beta_{length2}$ | 0.044 ± 0.063 | $\beta_{length2}$ | 0.955 ± 0.088 | $\beta_{length2}$ | 0.420 ± 0.125 |
| | | | $\beta_{length3}$ | 0.265 ± 0.085 | $\beta_{length3}$ | 0.831 ± 0.079 | $\beta_{length3}$ | 0.395 ± 0.124 |
| | | | $\beta_{length4}$ | 0.197 ± 0.059 | $\beta_{length4}$ | 0.688 ± 0.037 | $\beta_{length4}$ | 0.351 ± 0.059 |
| | | | $\beta_{length5}$ | 0.202 ± 0.079 | $\beta_{length5}$ | 0.666 ± 0.038 | $\beta_{length5}$ | 0.359 ± 0.063 |
| | | | $\beta_{length6}$ | 0.396 ± 0.135 | $\beta_{length6}$ | 0.754 ± 0.035 | $\beta_{length6}$ | 0.551 ± 0.062 |
| | | | $\beta_{length7}$ | 0.306 ± 0.165 | | | | |
| | | | $\beta_{length8}$ | 0.481 ± 0.063 | | | | |
| | | | $\beta_{length9}$ | 0.277 ± 0.036 | | | | |
| Lipid effect | $\beta_{lipid}$ | −0.025 ± 0.011 | $\beta_{lipid1}$ | 0.263 ± 0.072 | $\beta_{lipid\ 1}$ | −0.143 ± 0.099 | $\beta_{lipid\ 1}$ | 0.873 ± 0.158 |
| | | | $\beta_{lipid2}$ | 0.326 ± 0.066 | $\beta_{lipid\ 2}$ | −0.029 ± 0.126 | $\beta_{lipid\ 2}$ | 0.962 ± 0.286 |
| | | | $\beta_{lipid3}$ | 0.592 ± 0.074 | $\beta_{lipid\ 3}$ | 0.079 ± 0.077 | $\beta_{lipid\ 3}$ | 0.930 ± 0.152 |
| | | | $\beta_{lipid4}$ | 0.337 ± 0.061 | $\beta_{lipid\ 4}$ | 0.026 ± 0.035 | $\beta_{lipid\ 4}$ | 0.592 ± 0.068 |
| | | | $\beta_{lipid5}$ | 0.452 ± 0.093 | $\beta_{lipid\ 5}$ | 0.070 ± 0.057 | $\beta_{lipid\ 5}$ | 0.574 ± 0.098 |
| | | | $\beta_{lipid6}$ | 0.670 ± 0.111 | $\beta_{lipid\ 6}$ | −0.043 ± 0.027 | $\beta_{lipid\ 6}$ | 0.201 ± 0.051 |
| | | | $\beta_{lipid7}$ | 0.668 ± 0.208 | | | | |
| | | | $\beta_{lipid8}$ | 0.366 ± 0.064 | | | | |
| | | | $\beta_{lipid9}$ | 0.322 ± 0.034 | | | | |
| Temporal trends | $\beta_{year1}$ | −0.061 ± 0.004 | | | | | $\beta_{year1}$ | −0.058 ± 0.014 |
| | $\beta_{year2}$ | −0.042 ± 0.004 | $\beta_{year1}$ | −0.015 ± 0.003 | $\beta_{year1}$ | −0.005 ± 0.004 | $\beta_{year2}$ | −0.048 ± 0.013 |
| | $\beta_{year3}$ | −0.042 ± 0.005 | $\beta_{year2}$ | −0.127 ± 0.005 | $\beta_{year2}$ | −0.001 ± 0.006 | $\beta_{year3}$ | −0.054 ± 0.013 |
| | $\beta_{year4}$ | −0.054 ± 0.006 | | | | | $\beta_{year4}$ | −0.062 ± 0.012 |
| | $\beta_{year5}$ | −0.112 ± 0.020 | | | | | $\beta_{year5}$ | −0.070 ± 0.012 |
| | $\beta_{year6}$ | −0.011 ± 0.006 | | | | | $\beta_{year6}$ | −0.049 ± 0.011 |
| | $\beta_{year7}$ | −0.012 ± 0.021 | | | | | | |
| | $\beta_{year8}$ | −0.014 ± 0.005 | | | | | | |
| | $\beta_{year9}$ | −0.032 ± 0.002 | | | | | | |
| Model error | $\sigma$ | 0.274 ± 0.008 | $\sigma$ | 0.507 ± 0.036 | $\sigma$ | 0.501 ± 0.015 | $\sigma$ | 0.898 ± 0.030 |

significant seasonal variability in fish contamination stemming from various environmental or physiological factors, such as temperature variability and seasonal dietary shifts; for example, the THg concentrations in the food web of eastern Lake Ontario display distinct seasonality with spring maxima and summer minima (Zhang et al., 2012).

Based on the identification of the posterior regression coefficients, we infer that the THg and PCB concentrations distinctly covary with the fish length in every single sampling location of Lake Ontario (Table 3). The identifiability patterns refer to the degree of delineation of the most likely parameter values, after considering the available data. The statistic used to draw such inference was the coefficient of variation (standard deviation/mean) of the parameter posterior distributions. As previously mentioned, the contaminant–length association presumably reflects the longer exposure to contaminants of older age (and thus larger) fish, although the strength of this relationship can be determined by a wide array of factors (e.g., growth rate, behavioural patterns, diet) affecting contaminant bioaccumulation (Paterson et al., 2006; Trudel and Rasmussen, 2006). The same covariance pattern holds true between lipid content and PCB fish data. Because of the differences in the processes that regulate its fate and partitioning in fish tissues (Gewurtz et al., 2011), the linkage between lipid levels and THg concentrations is weaker with walleye and even has a negative (lakewide) signal with the contaminant levels in lake trout. This result aligns well with the understanding that Hg forms adducts with S-bearing amino acids instead of associating with lipid (Bloom, 1992).

Consistent with recent findings in the literature (Bhavsar et al., 2007, 2010), the posteriors of the regression coefficients related to the temporal contaminant trends are indicative of distinctly declining trajectories of the mean annual levels of the two contaminants in lake trout. Notably, the most accelerated THg declines appear to have occurred in western Lake Ontario, where the highest concentrations are recorded, i.e., from $-0.042 \pm 0.004$ $\log_e$[μg THg $g^{-1}$ ww] $year^{-1}$ in sites 2 and 3 to $-0.112 \pm 0.020$ $\log_e$[μg THg $g^{-1}$ ww] $year^{-1}$ in site 5. Lake trout demonstrated nearly monotonic decrease of their PCB levels in Lake Ontario, although they are still characterized by the highest PCB concentrations relative to the rest of the Great Lakes (Bhavsar et al., 2007). Our analysis is also on par with empirical evidence of a recent acceleration of the decline rates that led lake trout to meet the Great Lakes Strategy 2002 objective of decrease in concentrations by 25% during 2000–2007 (Bhavsar et al., 2007). Namely, the derived temporal trend during the post-dreissenid period ($\beta_{year2} = -0.127 \pm 0.005$ $\log_e$[ng PCB $g^{-1}$ ww] $year^{-1}$) suggests that the average lakewide PCB concentrations in 2011 is 345 ng PCB $g^{-1}$ ww with minimum and maximum levels equal to 65 and 1140 ng PCB $g^{-1}$ ww, respectively; values that are consistent with the measured concentrations.

In a similar manner, walleye demonstrated a decreasing trend with respect to their PCB levels with rates that are fairly uniform across Lake Ontario, from $-0.048$ to $-0.070$ $\log_e$[ng PCB $g^{-1}$ ww] $year^{-1}$. According to our hierarchical model, it is interesting to note that the average PCB levels in walleye during the post-dreissenid period, $\beta_{01} = 5.848$ $\log_e$[ng PCB $g^{-1}$ ww], may appear somewhat higher relative to the average values during the earlier years, $\beta_{02} = 5.660$ $\log_e$[ng PCB $g^{-1}$ ww], but when we consider their corresponding uncertainties, the two coefficients are practically indistinguishable and thus the projected temporal declines are statistically robust. By contrast, the THg model does not show any distinct temporal shifts in the rates of change, with the coefficients corresponding to the pre- and post-invasion periods equal to $-0.005 \pm 0.004$ $\log_e$[ng THg $g^{-1}$ ww] $year^{-1}$ and $-0.001 \pm 0.006$ $\log_e$[ng THg $g^{-1}$ ww] $year^{-1}$, respectively. The mean annual THg levels remained unaltered over the past four decades and their year-to-year variability is apparently subject to a "wax and wane" pattern (Bhavsar et al., 2010; French et al., 2006) see also

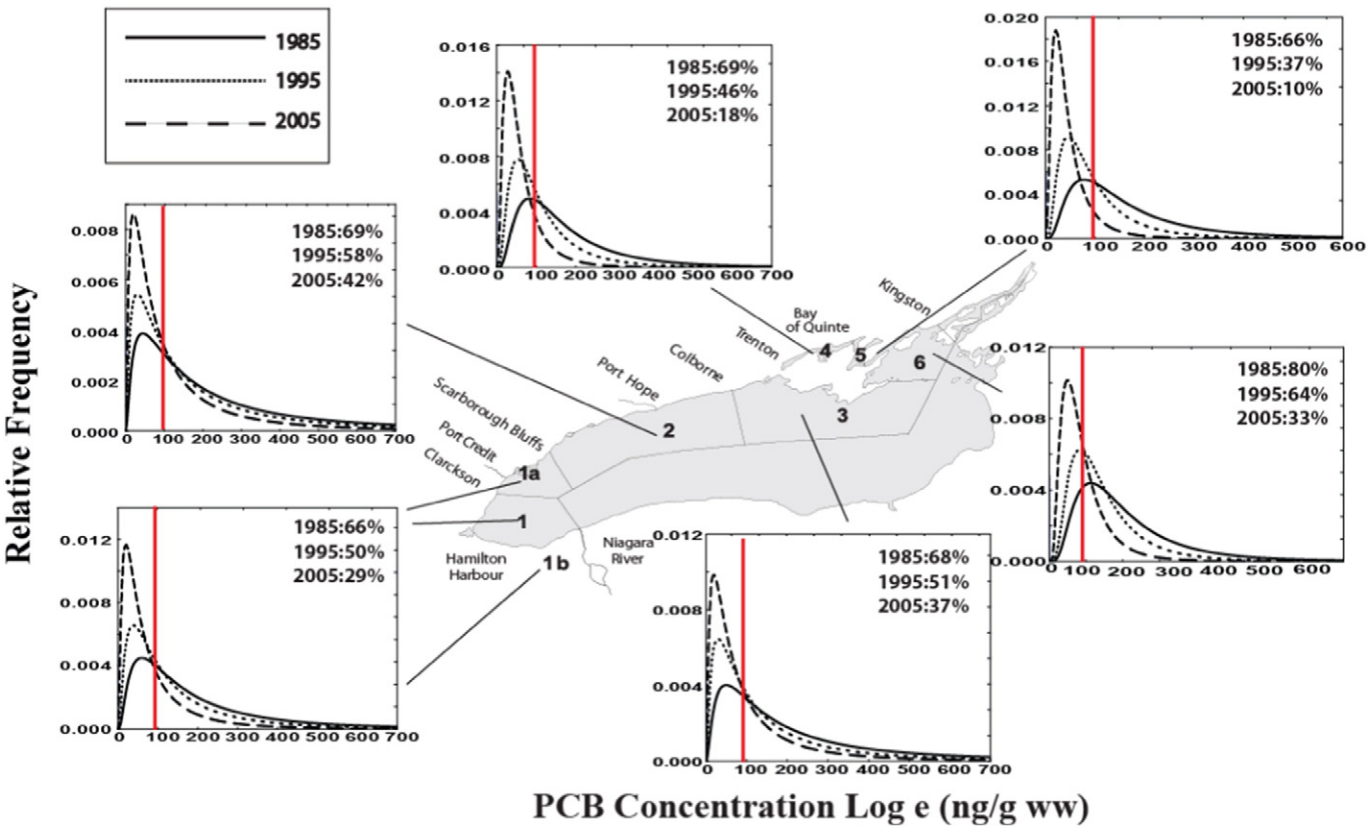


**Fig. 6.** Exceedance frequency of the tolerable daily intake PCB value for walleye during the mid-1980s, 1990s, and 2000s across all the fish length and lipid content values sampled in Lake Ontario. The red bold vertical line is for the PCB threshold of 100 ng $g^{-1}$ww, as calculated for a person of 50 kg who consumes 6 meals per month.

box plots of the decadal variability in Figs. 2–5). Similar oscillations have been reported for different species/contaminant combinations in Lake Ontario and could likely reflect the intricate nature of the prey–predator dynamics and/or the role of climatic forcing (Borgmann and Whittle, 1991; French et al., 2006). Other plausible explanations for the limited response of walleye (and other species) to the reduced contaminant emissions could be the complex interplay among the local variability in exogenous sources, the circulation patterns, the site-specific geochemistry that can profoundly shape the fate of contaminants within fish tissues as well as their spatial transport, and the energy shifts in trophodynamics along with the food web restructuring induced from the invasion of non-native species (Pacyna et al., 2010; Rennie et al., 2010; Gandhi et al., 2014). Regarding the latter mechanism, it has been hypothesized that the invasion of dreissenid mussels and round goby has resulted in a food chain lengthening and therefore in a higher fish contamination through the process of biomagnification, while there is evidence of dietary shifts of the top predators from less contaminated pelagic to more contaminated benthic food sources (Hogan et al., 2007).

*Fish consumption advisories*

We first used the year-specific predictive distributions for each contaminant and fish species to assess the mean frequency of exceedances of the tolerable daily intake values during the mid-1980s, 1990s, and 2000s across all the fish length and lipid content values measured in Lake Ontario. If we assume 6 meals per month as typical fish consumption for a human with an average body weight of 50 kg, the PCB threshold was calculated to be 100 ng $g^{-1}$ww, whereas the THg threshold was 0.57 μg $g^{-1}$ww. When consuming walleye, the probability of exceeding the PCB threshold was fairly high in all of the six locations (Fig. 6). Not surprisingly, the highest exceedance levels were found in 1985 (on average ~70%) with the northeastern part of Lake Ontario (site 6) reaching a mean probability value of 80%. The probability of exceeding the PCB threshold diminished by 10–30% in 1995 and by more than half in the year 2005. The highest exceedance frequency is now encountered in the area extending from Port Hope to Trenton/Prince Edward County (≈40%). On the other hand, although the THg threshold was exceeded from walleye consumption for all of the sites and years examined, the years 1985 and 1995 displayed lower probability values (<20%) relative to 2005 when the exceedance frequencies reach the level of 25% (Fig. 7). In contrast to walleye, the lake trout PCB threshold is greatly exceeded (>95%) in all of the sites during the three snapshots in time examined (Fig. 8). However, the THg threshold in lake trout is marginally exceeded (2–4%) for only 3 locations, with the rest of the sites indicating no exceedances (Fig. 9). These findings are in agreement with a report concluding that PCBs are of greater concern than THg for health risk to humans consuming fish from the Great Lakes (Bhavsar et al., 2011).

Finally, we derived the joint probabilities of exceeding the THg and PCB thresholds, while accounting for the covariance between them within the fish tissues sampled from Lake Ontario (Figs. 10–11). Based on the predictions for 2011, the odds for someone with an average body weight of 50 kg and consuming 6 meals of walleye per month and still remain within the tolerable daily intakes for the two contaminants were approximately equal or greater than 50%. The highest probabilities were found for the two locations of the Bay of

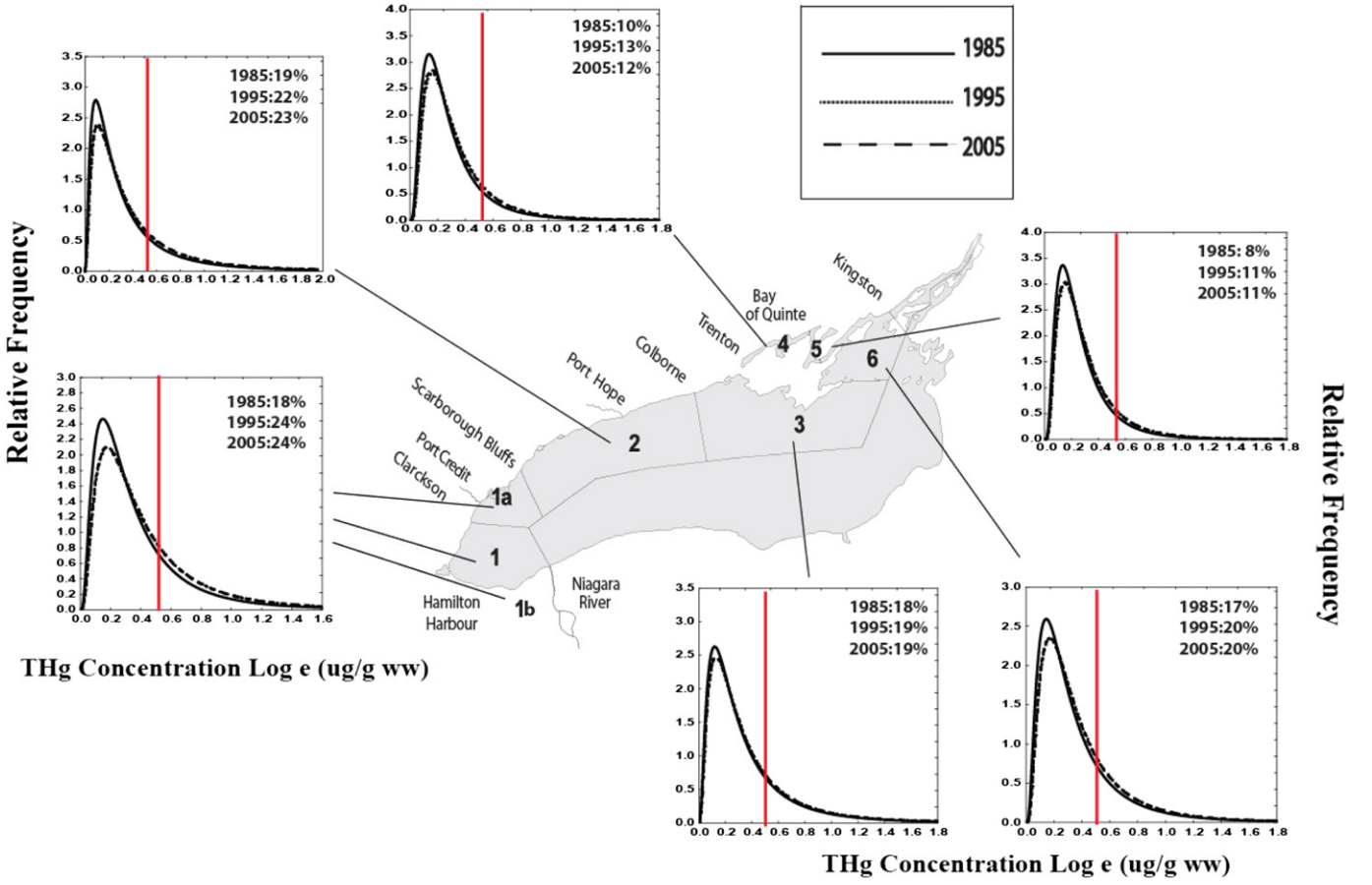


**Fig. 7.** Exceedance frequency of the tolerable daily intake THg value for walleye during the mid-1980s, 1990s, and 2000s across all the fish length and lipid content values sampled in Lake Ontario. The red bold vertical line is the THg threshold of 0.57 μg $g^{-1}$ww, as calculated for a person of 50 kg who consumes 6 meals per month.

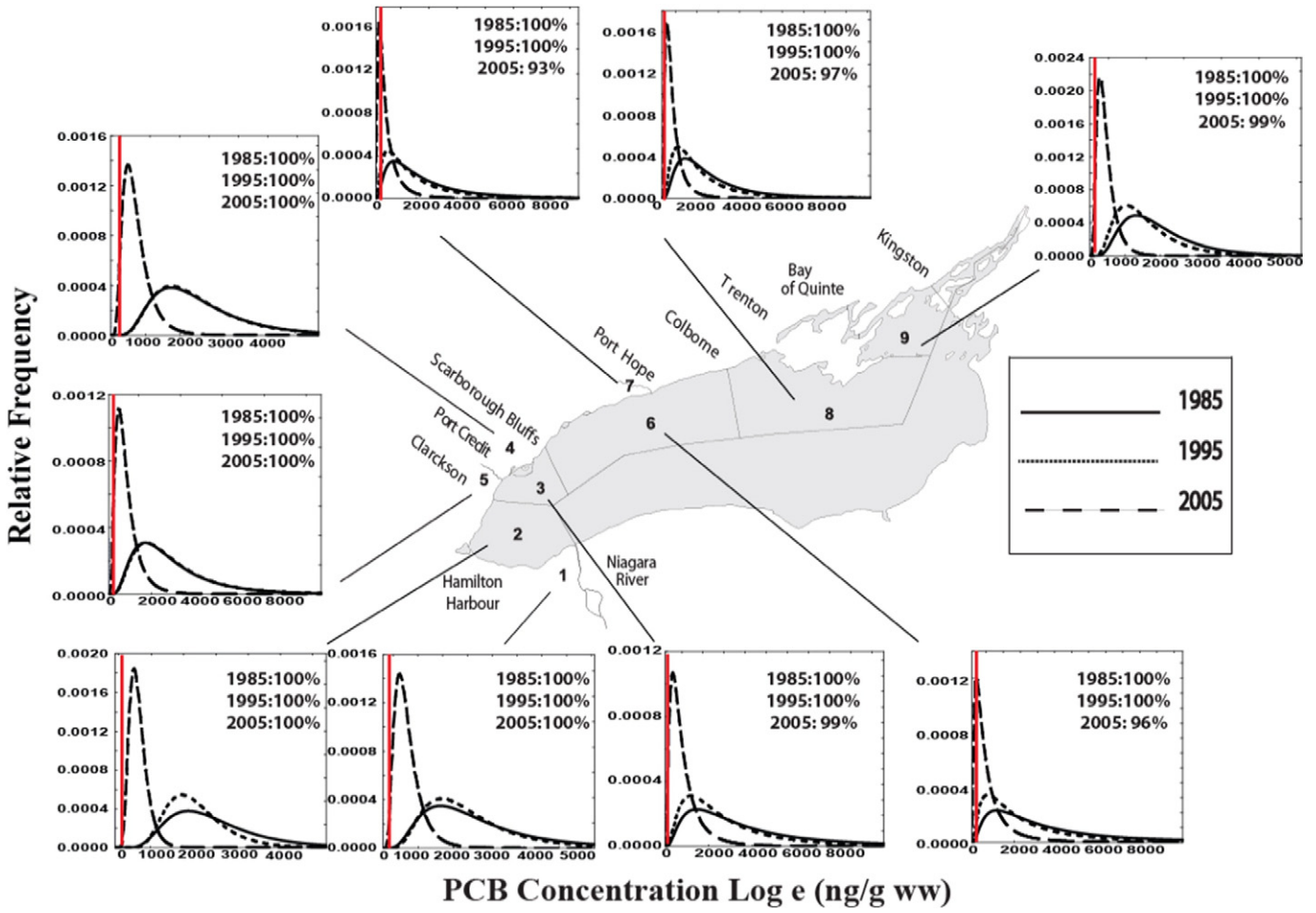


**Fig. 8.** Exceedance frequency of the tolerable daily intake PCB value for lake trout during the mid-1980s, 1990s, and 2000s across all the fish length and lipid content values sampled in Lake Ontario. The red bold vertical line is the PCB threshold of 100 ng $g^{-1}$ww, as calculated for a person of 50 kg who consumes 6 meals per month.

Quinte area (sites 4 and 5) with the lowest THg levels. By contrast, the consumption of 6 meals per month of lake trout appears to be more of a health risk; especially for lake trout from the western part of the lake were the joint probabilities of avoiding exceedances of the thresholds were <10%. Lake trout sampled from the north and northeastern parts of Lake Ontario were characterized by greater (≈20 –30%) joint probabilities of avoiding exceedance of the thresholds, primarily reflecting the substantial progress made with their PCB levels over the past few decades (Ridal et al., 2012).

*Current challenges with fish consumption advisories*

The impartial assessment of contaminant trends in fish communities can be undermined by a number of confounding factors, such as type of statistical analysis performed, type of samples used (data pooling, skinless boneless fillet versus whole fish portions), seasonal variability, and covariance with different facets of fish morphology, physiology, or even ethology (e.g., fish size, lipid content, feeding habits, behavioral patterns, reproductive status, and growth) (Szlinder-Richert et al., 2009; Gewurtz et al., 2011). Recognizing the challenges with the development of fish consumption advisories, the implementation of Bayesian inference techniques has been proposed as a sensible strategy to rigorously quantify and effectively communicate the surrounding uncertainty (Stow et al., 2004; Azim et al., 2011a,b; Sadraddini et al., 2011a,b; Neff et al., 2012; Mahmood et al., 2013a,b). Questioning the efficiency of conventional regression practices, Mahmood et al. (2013b) highlighted the dynamic linear modeling as a better alternative that is also conceptually on par with the recent shift toward probabilistic advisory frameworks. The reasoning for the latter assertion was the fact that dynamic linear models have an evolving structure with time variant parameters; the year-specific predictive fish contaminant distributions are conditioned upon prior and current information, not by subsequent data; the modeling analysis along with the derived fish contaminant trends can be based on individual samples, thereby accommodating both intra- and inter-annual variability; the Bayesian nature of the framework allows both parametric uncertainty and structural error (model misspecification) to be reflected in model predictions. Because of its data requirements though, dynamic linear modeling may not always be a suitable methodology, especially when aiming to detect elevated fish consumption risks in both space and time. Capitalizing upon the capacity of hierarchical models to enable the transfer of information in space, the present study overcomes this problem allowing the effective modeling of locations with limited/inconsistent information.

Generally, our analysis provides evidence of distinctly declining trajectories for both THg and PCBs in lake trout. Walleye demonstrate a similar decreasing PCB trend, whereas no distinct temporal shifts were found with their mercury rates of change. Spatial patterns of fish contamination are indicative of a distinct west-to-east gradient in Lake Ontario, primarily shaped by the contaminant loads transported through Niagara River. Aside from the accommodation of spatiotemporal variability and predictive uncertainty, the estimation of the joint probability of compliance with multiple contaminant thresholds in focal fish

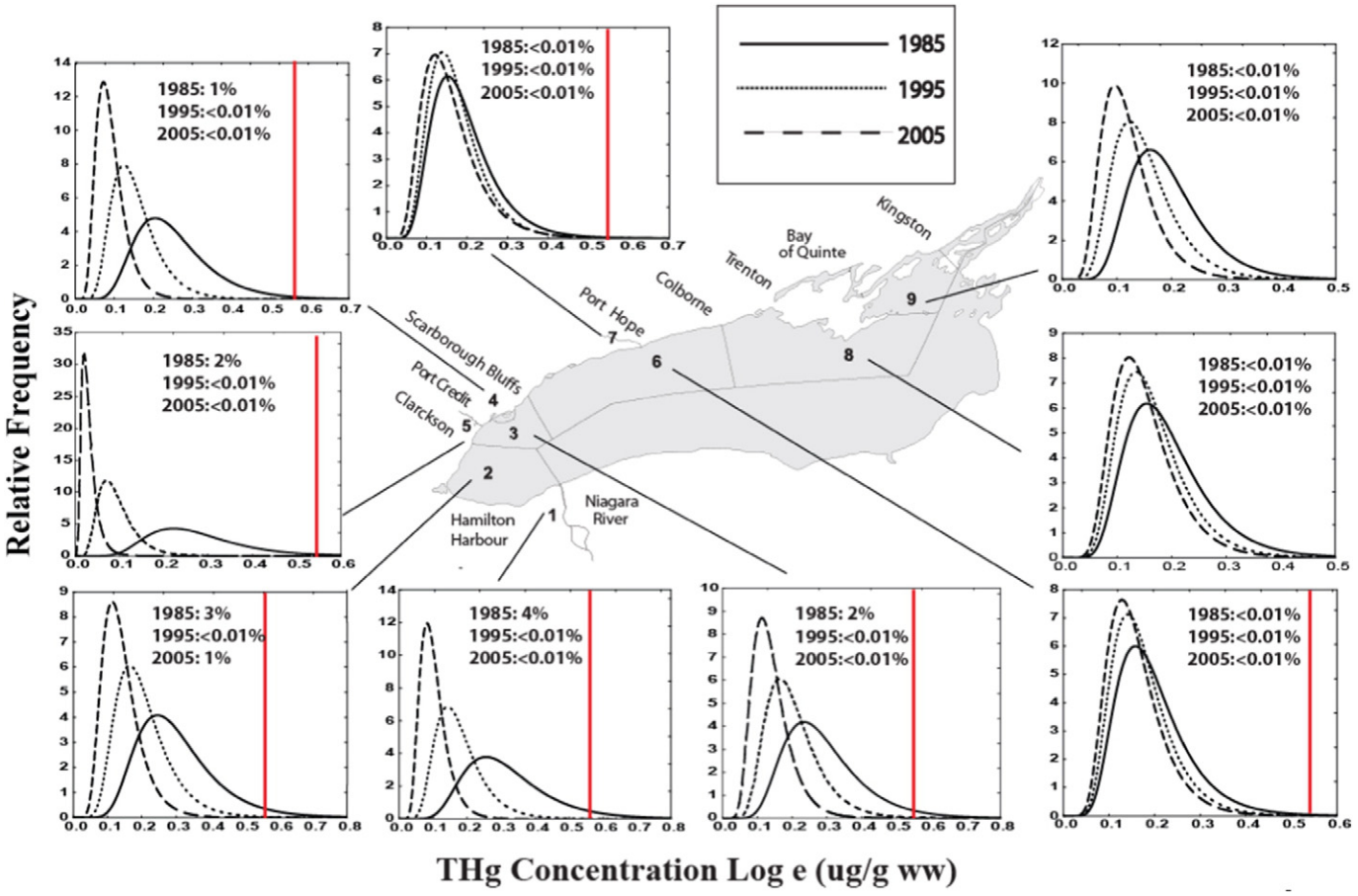


**Fig. 9.** Exceedance frequency of the tolerable daily intake THg value for lake trout during the mid-1980s, 1990s, and 2000s across all the fish length and lipid content values sampled in Lake Ontario. The red bold vertical line is the THg threshold of 0.57 μg $g^{-1}$ww, as calculated for a person of 50 kg who consumes 6 meals per month.

species represents another aspect of the present framework that can assist with the development of defensible integrated advisories (Scherer et al., 2008). Specifically, the consumption of lake trout appears to be particularly a health risk for the sensitive demographic group of children less than 15 years old, as it is characterized by joint probabilities of compliance with the THg and PCBs thresholds that are <10% in the western part of Lake Ontario. The same probabilities with walleye were approximately equal to or greater than 50%. The overall trends contradict the anticipation of a universal improvement across all species and contaminants examined, suggesting that the degree of response to the loading reductions tends to be both species and contaminant specific. In addition, the presence of substantial within-system variability could be an impediment for the robust delineation of the temporal fish contaminant trends and may pose challenges toward the development of fish consumption advisories that effectively balance between overly optimistic and unnecessarily alarmist statements.

Notwithstanding the contamination risks, fish also provide an excellent dietary source of high quality and easily digestible protein and omega-3 fatty acids. Longer chain omega-3 fatty acids may 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 (Das, 2006). Eicosapentaenoic acid (EPA), for example, is a precursor of eicosanoids (signaling hormones), and eicosanoids derived from EPA tend to impede inflammation associated with many chronic diseases (Perhar et al., 2012). Alpha-linolenic acid (ALA), a precursor of EPA, is hypothesized to support the growth and development of infants. Brain, retina, and sperm are the tissues in the human body with the highest docosahexaenoic acid (DHA) concentrations, and demand for DHA is particularly pressing during the latter stages of pregnancy and early infancy. In this regard, one key challenge involves the capacity of advisories to impartially weigh these tradeoffs and elucidate the conflicting information from "restrictive" advisories and "encouraging" nutritionists (Turyk et al., 2012; Neff et al., 2014). In addition to balancing the risks and benefits of fish, other outstanding issues of consumption advisories revolve around the tone, readability, and effective communication of the associated information to target populations or even the lack of consensus about the definition of what is a "sensitive" population (Oken et al., 2012). In this context, we believe that the proposed Bayesian approach to fish consumption advisories can serve as a valuable framework for highly customizable risk assessment statements that can flexibly incorporate the uncertainty in contaminant predictions, while remaining flexible for different vulnerable subgroups, human weights, and diet patterns.

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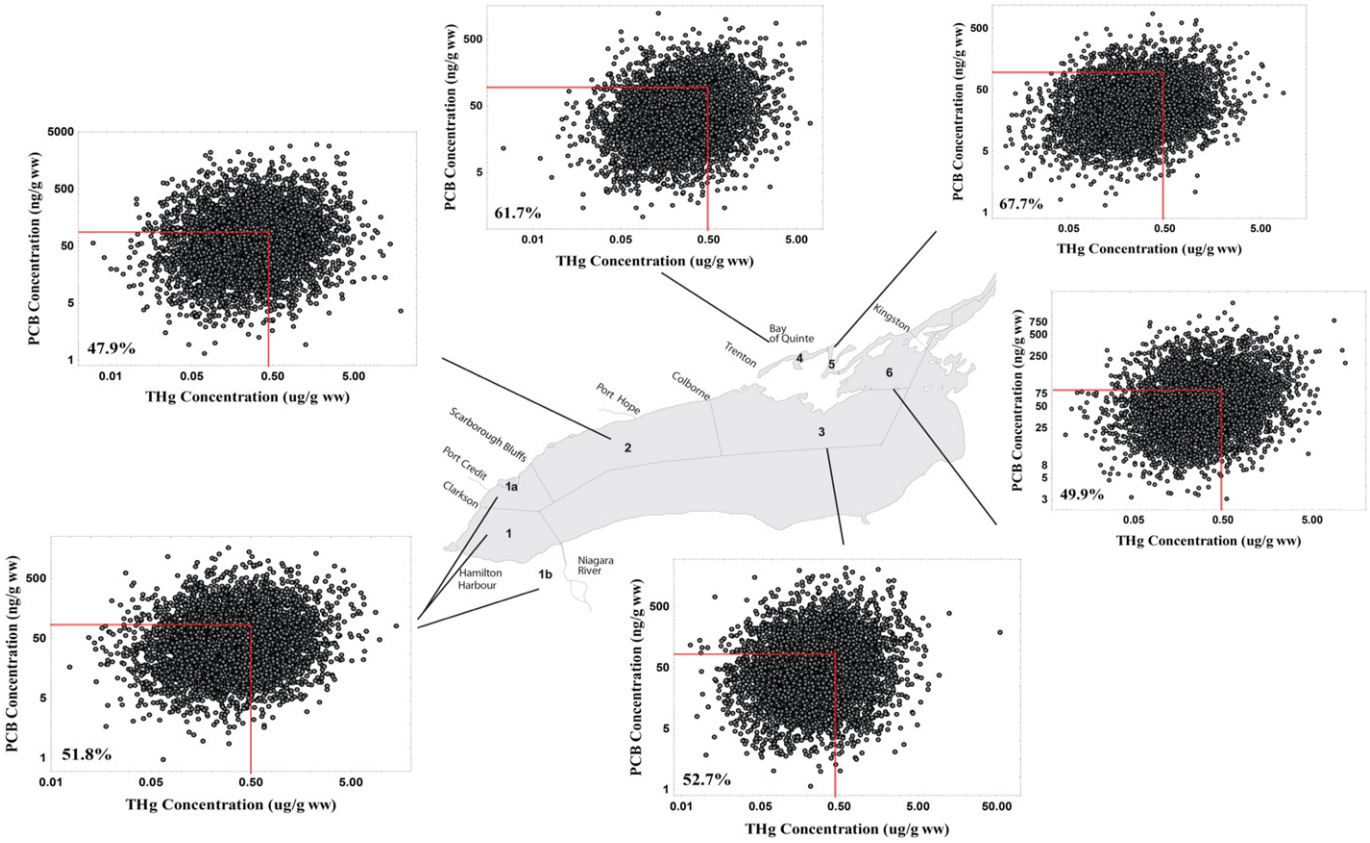


**Fig. 10.** Predicted joint probability of PCB and THg concentrations for Lake Ontario walleye in 2011 across all the fish length and lipid content values. The red horizontal and vertical lines represent the PCB and THg thresholds of 100 ng $g^{-1}$ww and 0.57 μg $g^{-1}$ww, respectively, calculated for a person of 50 kg who consumes 6 meals per month. The area that falls within the boundaries of the two thresholds reflect safe fish consumption and the corresponding probability values are indicated at the bottom left of each panel.

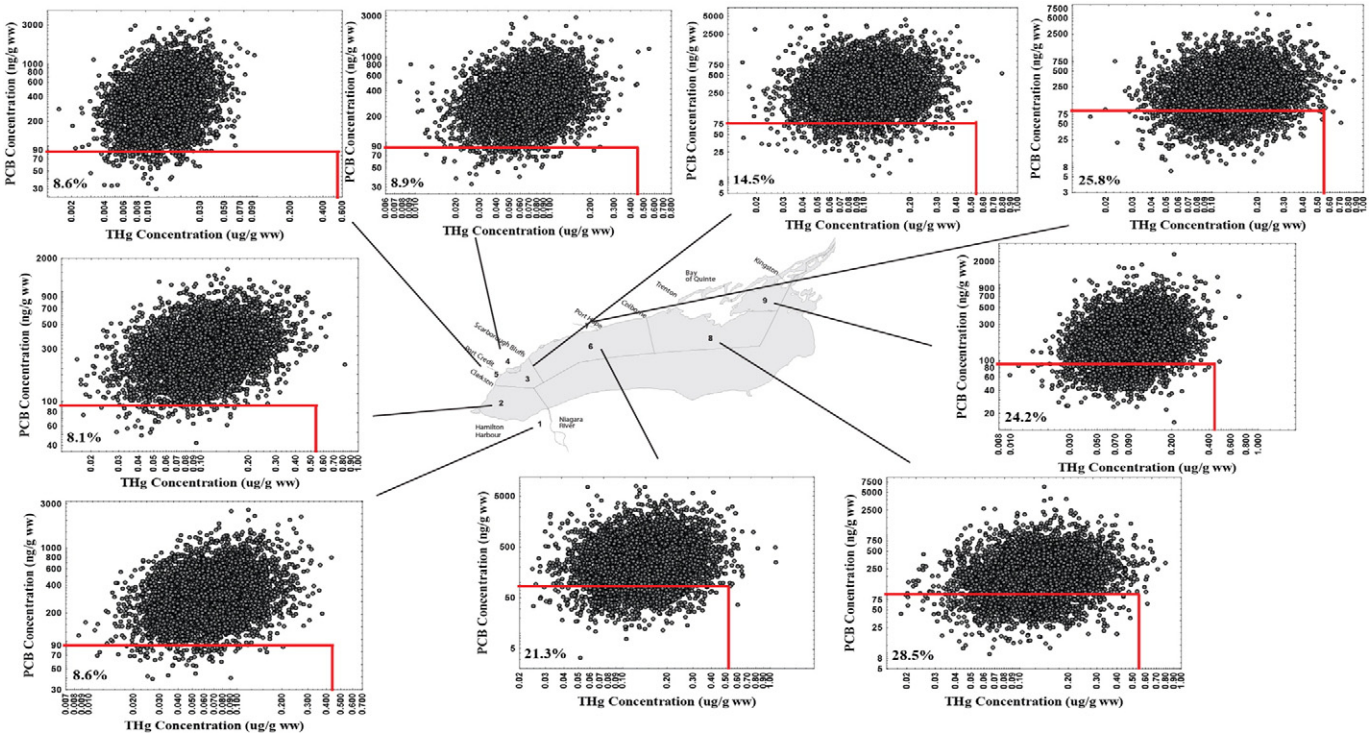


**Fig. 11.** Predicted joint probability of PCB and THg concentrations for Lake Ontario lake trout in 2011 across all the fish length and lipid content values. The red horizontal and vertical lines represent the PCB and THg thresholds of 100 ng $g^{-1}$ww and 0.57 μg $g^{-1}$ww, respectively, calculated for a person of 50 kg who consumes 6 meals per month. The area that falls within the boundaries of the two thresholds reflect safe fish consumption and the corresponding probability values are indicated at the bottom left of each panel.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jglr.2015.11.005.

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# Guiding Fish Consumption Advisories for Lake Ontario:

# A Bayesian hierarchical approach

## [SUPPORTING INFORMATION]

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## SECTION A

### *Dataset description-Chemical analysis*

We used fish contaminant data from the Ontario Ministry of the Environment and Climate Change (*OMOECC*) Fish Contaminant Monitoring Program, which routinely analyzes samples of a wide range of fish species for contaminant levels mainly in the dorsal skinless-boneless fillet (*SBF*) portions. This information is then used to issue updated fish consumption advisories on a biennial basis. In our analysis, we selected lake trout and walleye based on their role as biological indicators (or tracers) of contaminant variability as well as on their commercial importance and popularity among the native and recreational angler community. The samples were collected from various locations in the Canadian waters of Lake Ontario (Fig. 1). There were 9 sampling regions for lake trout samples and 6 regions for walleye. Chemical analyses for the levels of *THg* and *PCB*s were conducted at the *OMOECC* laboratories in Toronto. *THg* was measured with cold vapour-flameless atomic absorption spectrophotometry (*CV-FAAS*) technique using the *OMOECC* method *HGBIO-E3057* as described in Supporting Material of Bhavsar *et al.* (2010). Total-*PCB* analysis on the *OMOECC* samples was performed through gas chromatography with $^{63}$*Ni* electron capture detector (*ECD*) as described by Bhavsar *et al.* (2007).

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## SECTION B

### *Bayesian Hierarchical Modeling*

We introduce four hierarchical structures to evaluate different facets of the spatiotemporal contaminant trends in walleye and lake trout, while accounting for the potential impact of the invasion of dreissenids along with their covariance with fish length and lipid content. The first model (*Model 1*) aims to evaluate the lake-wide *THg/PCB* temporal trends, while considering the differences of the intercepts among the six and nine sites in Lake Ontario, where the walleye and lake trout samples were collected, respectively. The same model accommodates the spatial variability in the relationships between contaminants and fish length/lipid content. Notably, the two covariates were standardized prior to the analysis, and thus the posterior estimates of the intercepts reflect the initial contaminant levels in each location for a fish individual with average length and lipid content. The first hierarchical formulation is summarized as follows:

$$\log_e(C_{MEASijt}) \sim N(\log_e(C_{PREDijt}), \psi^2)$$

$$\log_e(C_{PREDijt}) = \beta_{0j} + \beta_{lengthj} length_{ijt} + \beta_{lipidj} lipid_{ijt} + \beta_{year}(t_{ij}-t_0)$$

$$\beta_{year} \sim N(0, 10000)$$

$$\beta_{0j} \sim N(\beta_{0g}, \tau_{0j}^2) \quad \beta_{lengthj} \sim N(\beta_{lengthg}, \tau_{lengthj}^2) \quad \beta_{lipidj} \sim N(\beta_{lipidg}, \tau_{lipidj}^2)$$

$$\beta_{0g} \sim N(\mu_0, \sigma_0^2) \quad \beta_{lengthg} \sim N(\mu_{length}, \sigma_{length}^2) \quad \beta_{lipidg} \sim N(\mu_{lipid}, \sigma_{lipid}^2)$$

$$\mu_0 \sim N(0, 10000) \quad \mu_{length} \sim N(0, 10000) \quad \mu_{lipid} \sim N(0, 10000)$$

$$\tau_{0j}^{-2} \sim G(0.001, 0.001) \quad \tau_{lengthj}^{-2} \sim G(0.001, 0.001) \quad \tau_{lipidj}^{-2} \sim G(0.001, 0.001)$$

$$\sigma_0^{-2} \sim G(0.001, 0.001) \quad \sigma_{length}^{-2} \sim G(0.001, 0.001) \quad \sigma_{lipid}^{-2} \sim G(0.001, 0.001)$$

$$\psi^{-2} \sim G(0.001, 0.001)$$

$$i=1....N_j \quad j=1...M \quad t=1...T$$

where $C_{MEASijt}$ represents the observed contaminant level in the sample $i$, collected from location $j$ and year $t$; $C_{PREDijt}$ and $\psi^2$ represent the modeled concentrations and model error variance, respectively; $\beta_{year}$ is the regression coefficient related to the lake-wide contaminant trends over time; $\beta_{0j}$, $\beta_{lengthj}$, $\beta_{lipidj}$ denote the site-specific intercepts, length- and lipid-regression coefficients, respectively; $\tau_{0j}^2$, $\tau_{lengthj}^2$, $\tau_{lipidj}^2$ are the corresponding site-specific variances; $\beta_{0g}$, $\beta_{lengthg}$, $\beta_{lipidg}$ are the global parameters; $\mu_0$, $\mu_{length}$, $\mu_{lipid}$ and $\sigma_0^2$, $\sigma_{length}^2$, $\sigma_{lipid}^2$ are the mean and variance of the hyperparameters, respectively; $N_j$, $M$, and $T$ are the total number of samples in location $j$, the total number of sampling locations, and the total number of years, respectively; $t_0$ is the initial year of the analysis; $N(0, 10000)$ 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 “noninformative” or vague.

The distinct feature of the second model (*Model 2*) is its capacity to delineate the contaminant trends in each sampling location, and thus it can be mathematically expressed as follows:

$$\log_e(C_{MEASijt}) \sim N(\log_e(C_{PREDijt}), \psi^2)$$
$$\log_e(C_{PREDijt}) = \beta_{0j} + \beta_{length} length_{ijt} + \beta_{lipid} lipid_{ijt} + \beta_{yearj}(t_{ij}-t_0)$$
$$\beta_{length} \sim N(0, 10000)\ \beta_{lipid} \sim N(0, 10000)$$
$$\beta_{0j} \sim N(\beta_{0g}, \tau_{0j}^2)\ \beta_{yearj} \sim N(\beta_{yearg}, \tau_{yearj}^2)$$
$$\beta_{0g} \sim N(\mu_0, \sigma_0^2)\ \beta_{yearg} \sim N(\mu_{year}, \sigma_{year}^2)$$
$$\mu_0 \sim N(0, 10000)\ \mu_{year} \sim N(0, 10000)$$
$$\tau_{0j}^{-2} \sim G(0.001, 0.001)\ \tau_{yearj}^{-2} \sim G(0.001, 0.001)$$
$$\sigma_0^{-2} \sim G(0.001, 0.001)\ \ \sigma_{year}^{-2} \sim G(0.001, 0.001)$$
$$\psi^{-2} \sim G(0.001, 0.001)$$
$$i=1....N_j \quad j=1...M \quad t=1...T$$

Similar to *Model 1*, the intercept maintains its location-specific character, whereas the signature of the fish length/ lipid content is assumed to be spatially constant; $\beta_{yearj}$, $\beta_{yearg}$, $\tau_{yearj}^2$, $\mu_{year}$, $\sigma_{year}^2$ are the site-specific and global parameters used for the hierarchical characterization of the location-specific temporal trends.

The third model (*Model 3*) evaluates the lakewide contaminant trends, while explicitly considering the effects of the invasion of dreissenids on the average contaminant levels. The fish contaminant:length/lipid relationships are assumed constant across all sampling locations.

$$\log_e(C_{MEASijt}) \sim N(\log_e(C_{PREDijt}), \psi^2)$$
$$\log(C_{PREDijt}) = \beta_{0k} + \beta_{lengthj} length_{ijt} + \beta_{lipidj} lipid_{ijt} + \beta_{yearj}(t_{ij}-t_0)$$
$$\beta_{0k} \sim N(\beta_{0g}, \tau_{0k}^2)\ \beta_{lengthj} \sim N(\beta_{lengthg}, \tau_{lengthj}^2)\ \beta_{lipidj} \sim N(\beta_{lipidg}, \tau_{lipidj}^2)\ \beta_{yearj} \sim N(\beta_{yearg}, \tau_{yearj}^2)$$
$$\beta_{0g} \sim N(\mu_0, \sigma_0^2)\ \ \beta_{lengthg} \sim N(\mu_{length}, \sigma_{length}^2)\ \beta_{lipidg} \sim N(\mu_{lipid}, \sigma_{lipid}^2)\ \beta_{yearg} \sim N(\mu_{year}, \sigma_{year}^2)$$
$$\mu_0 \sim N(0, 10000)\ \mu_{length} \sim N(0, 10000)\ \mu_{lipid} \sim N(0, 10000)\ \mu_{year} \sim N(0, 10000)$$
$$\tau_{0j}^{-2} \sim G(0.001, 0.001)\ \tau_{lengthj}^{-2} \sim G(0.001, 0.001)\ \tau_{lipidj}^{-2} \sim G(0.001, 0.001)\ \tau_{yearj}^{-2} \sim G(0.001, 0.001)$$
$$\sigma_0^{-2} \sim G(0.001, 0.001)\ \ \sigma_{length}^{-2} \sim G(0.001, 0.001)\ \sigma_{lipid}^{-2} \sim G(0.001, 0.001)\ \sigma_{year}^{-2} \sim G(0.001, 0.001)$$
$$\psi^{-2} \sim G(0.001, 0.001)$$
$$i=1....N_j \quad j=1...M \quad t=1...T \quad k=1 \text{ or } 2 \text{ (before and after 1995)}$$

With this model, we emulate the influence of the dreissenids on the contaminant levels as an abrupt shift in the average lakewide conditions; hence, $\beta_{0k}$ are the two spatially-constant intercepts for the pre- and post-invasion periods; $\beta_{0g}$, $\mu_0$, $\sigma_0^2$ are the global parameters used for the hierarchical formulation of the tested shift in baseline conditions.

The fourth model (*Model 4*) aims to evaluate the lakewide contaminant trends before and after the invasion of dreissenids, while explicitly considering the variations of the initial conditions as well as the covariance with fish length and lipid content in different locations of the lake. The model can be described as:

$$\log_e(C_{MEASijt}) \sim N(\log_e(C_{PREDijt}), \psi^2)$$
$$\log_e(C_{PREDijt}) = \beta_{0j} + \beta_{lengthj} length_{ijt} + \beta_{lipidj} lipid_{ijt} + \beta_{year1} t'_{ij} + \beta_{year2} t''_{ij}$$
$$t'_{ij} = t_{ij}-t_0 \text{ if } t_{ij}<t_{inv} \text{ else } 0 \qquad t''_{ij} = t_{ij}-t_{inv} \text{ if } t_{ij}>t_{inv} \text{ else } 0$$
$$\beta_{year1} \sim N(0, 10000)\ \beta_{year2} \sim N(0, 10000)$$
$$\beta_{0j} \sim N(\beta_{0g}, \tau_{0j}^2)\ \ \beta_{lengthj} \sim N(\beta_{lengthg}, \tau_{lengthj}^2)\ \beta_{lipidj} \sim N(\beta_{lipidg}, \tau_{lipidj}^2)$$

$$\beta_{0g} \sim N(\mu_0, \sigma_0^2) \quad \beta_{lengthg} \sim N(\mu_{length}, \sigma_{length}^2) \; \beta_{lipidg} \sim N(\mu_{lipid}, \sigma_{lipid}^2)$$

$$\mu_0 \sim N(0, 10000) \; \mu_{length} \sim N(0, 10000) \; \mu_{lipid} \sim N(0, 10000)$$

$$\tau_{0j}^{-2} \sim G(0.001, 0.001) \; \tau_{lengthj}^{-2} \sim G(0.001, 0.001) \; \tau_{lipidj}^{-2} \sim G(0.001, 0.001)$$

$$\sigma_0^{-2} \sim G(0.001, 0.001) \quad \sigma_{length}^{-2} \sim G(0.001, 0.001) \; \sigma_{lipid}^{-2} \sim G(0.001, 0.001)$$

$$\psi^{-2} \sim G(0.001, 0.001)$$

$$i=1....N_j \quad j=1...M \quad t=1...T$$

The implementation of this model again aims to determine whether the contaminant trends were affected by the presence of invasive species. By contrast to *Model 3* though, this model introduces a second linear trend after the year of the invasion $t_{inv}$, assumed to be the year of 1995, instead of postulating a distinct shift to the average contaminant levels during the post-invasion period.

**Table S1**: Summary of the years with available *PCB* data for lake trout per sampling site.

| Lake Trout PCB | Total # | Years |
|---|---|---|
| Site 1 | 7 | 1984, 1985, 1986, 1988, 1989, 2006, 2007 |
| Site 2 | 10 | 1978, 1982, 1984, 1987, 1989, 1993, 1994, 1996, 1997, 2002 |
| Site 3 | 7 | 1979, 1980, 1990, 1994, 1996, 1999, 2002 |
| Site 4 | 3 | 1980, 1981, 1993 |
| Site 5 | 5 | 1982, 1984, 1985, 1987, 1988 |
| Site 6 | 5 | 1993, 1996, 1999, 2007, 2010 |
| Site 7 | 2 | 1989, 1994 |
| Site 8 | 10 | 1990, 1993, 1994, 1996, 1999, 2002, 2004, 2006, 2008, 2010 |
| Site 9 | 11 | 1981, 1986, 1990, 1991, 1993, 1994, 1996, 2002, 2004, 2006, 2010 |

**Table S2**: Summary of the years with available *Hg* data for lake trout per sampling site.

| Lake Trout Hg | Total # | Years |
|---|---|---|
| Site 1 | 8 | 1984, 1985, 1986, 1988, 1989, 1998, 2006, 2007 |
| Site 2 | 10 | 1978, 1982, 1984, 1987, 1989, 1993, 1994, 1996, 1997, 2002 |
| Site 3 | 7 | 1979, 1980, 1990, 1994, 1996, 1999, 2002 |
| Site 4 | 3 | 1980, 1981, 1993 |
| Site 5 | 5 | 1982, 1984, 1985, 1987, 1988 |
| Site 6 | 5 | 1993, 1996, 1999, 2007, 2010 |
| Site 7 | 2 | 1989, 1994 |
| Site 8 | 10 | 1990, 1993, 1994, 1996, 1999, 2002, 2004, 2006, 2008, 2010 |
| Site 9 | 12 | 1981, 1986, 1990, 1991, 1993, 1994, 1996, 2002, 2004, 2006, 2010, 2011 |

**Table S3**: Summary of the years with available *PCB* data for walleye per sampling site.

| Walleye PCB | Total # | Years |
|---|---|---|
| Site 1 | 6 | 1978, 1980, 1989, 1994, 1998, 2007 |
| Site 2 | 3 | 1988, 1989, 2005 |
| Site 3 | 9 | 1977, 1982, 1987, 1989, 1996, 2002, 2004, 2006, 2010 |
| Site 4 | 12 | 1975, 1981,1987, 1989, 1991, 1992, 1993, 1994, 1997, 1999, 2001, 2003 |
| Site 5 | 13 | 1981, 1985, 1989, 1990, 1992, 1993, 1997, 1998, 2001, 2003, 2004, 2006, 2007 |
| Site 6 | 13 | 1981. 1985, 1987, 1989, 1992, 1993, 1994, 1996, 2002, 2004, 2006, 2010, 2011 |

**Table S4**: Summary of the years with available *Hg* data for walleye per sampling site.

| Walleye Hg | Total # | Years |
|---|---|---|
| Site 1 | 5 | 1980, 1989, 1994, 1998, 2007 |
| Site 2 | 3 | 1988, 1989, 2005 |
| Site 3 | 9 | 1977, 1982, 1987, 1989, 1996, 2002, 2004, 2006, 2010 |
| Site 4 | 12 | 1975, 1981,1987, 1989, 1991, 1992, 1993, 1994, 1997, 1999, 2001, 2003 |
| Site 5 | 13 | 1981, 1985, 1989, 1990, 1992, 1993, 1997, 1998, 2001, 2003, 2004, 2006, 2007 |
| Site 6 | 13 | 1981. 1985, 1987, 1989, 1992, 1993, 1994, 1996, 2002, 2004, 2006, 2010, 2011 |

**Table S5**: Summary statistics of *THg* (*µg/g* wet weight) and *PCB*s (*ng/g* wet weight) in walleye and lake trout skinless-boneless fillet (*SBF*) data from six (6) and nine (9) sampling sites in Lake Ontario, respectively.

| | | ***N*** | ***Mean*** | ***Stdev*** | ***Median*** | ***2.5%*** | ***97.5%*** | ***Int Quart**** | ***Kurt**** | ***Skew**** |
|---|---|---|---|---|---|---|---|---|---|---|
| | | ***Walleye*** | | | | | | | | |
| **Site 1** | *THg* | 45 | 0.66 | 0.41 | 0.65 | 0.09 | 1.58 | 0.57 | -0.45 | 0.54 |
| | *PCB* | 49 | 354 | 643 | 90 | 20 | 2496 | 180 | 6.82 | 2.69 |
| **Site 2** | *THg* | 39 | 0.43 | 0.37 | 0.35 | 0.06 | 1.10 | 0.71 | -1.09 | 0.66 |
| | *PCB* | 39 | 210 | 330 | 90 | 30 | 1482 | 188 | 10.97 | 3.25 |
| **Site 3** | *THg* | 75 | 0.56 | 0.34 | 0.46 | 0.11 | 1.32 | 0.55 | -0.41 | -0.41 |
| | *PCB* | 75 | 258 | 302 | 140 | 20 | 1089 | 241 | 5.20 | 2.22 |
| **Site 4** | *THg* | 220 | 0.33 | 0.29 | 0.24 | 0.04 | 1.10 | 0.31 | 2.40 | 1.56 |
| | *PCB* | 220 | 215 | 367 | 100 | 20 | 1197 | 217 | 36.39 | 5.05 |
| **Site 5** | *THg* | 162 | 0.27 | 0.28 | 0.16 | 0.04 | 0.91 | 0.24 | 11.33 | 2.70 |
| | *PCB* | 174 | 155 | 306 | 60 | 20 | 752 | 118 | 53.06 | 6.20 |
| **Site 6** | *THg* | 194 | 0.41 | 0.33 | 0.27 | 0.06 | 1.22 | 0.52 | 0.33 | 1.08 |
| | *PCB* | 197 | 237 | 410 | 120 | 20 | 1513 | 185 | 17.79 | 3.99 |
| | | ***Lake Trout*** | | | | | | | | |
| **Site 1** | *THg* | 74 | 0.28 | 0.12 | 0.29 | 0.07 | 0.47 | 0.18 | -0.88 | -0.24 |
| | *PCB* | 74 | 2281 | 1390 | 2240 | 361 | 5122 | 2202 | -0.35 | 0.50 |
| **Site 2** | *THg* | 178 | 0.25 | 0.10 | 0.24 | 0.07 | 0.48 | 0.14 | 0.26 | 0.57 |
| | *PCB* | 180 | 2379 | 1521 | 2100 | 459 | 6329 | 1955 | 2.64 | 1.34 |
| **Site 3** | *THg* | 206 | 0.24 | 0.08 | 0.22 | 0.13 | 0.39 | 0.12 | 1.03 | 0.94 |
| | *PCB* | 93 | 2239 | 2026 | 1700 | 500 | 6610 | 1414 | 15.07 | 3.13 |
| **Site 4** | *THg* | 113 | 0.20 | 0.09 | 0.20 | 0.06 | 0.37 | 0.11 | 1.96 | 0.90 |
| | *PCB* | 93 | 2402 | 1821 | 2049 | 373 | 6802 | 1859 | 4.08 | 1.77 |
| **Site 5** | *THg* | 68 | 0.30 | 0.10 | 0.30 | 0.11 | 0.49 | 0.09 | 0.02 | 0.21 |
| | *PCB* | 78 | 4614 | 3052 | 4250 | 723 | 10505 | 3103 | 7.42 | 2.01 |
| **Site 6** | *THg* | 75 | 0.20 | 0.05 | 0.19 | 0.12 | 0.30 | 0.09 | -0.31 | 0.42 |
| | *PCB* | 76 | 1923 | 1445 | 1475 | 405 | 5406 | 1633 | 2.30 | 1.55 |
| **Site 7** | *THg* | 40 | 0.23 | 0.07 | 0.24 | 0.10 | 0.34 | 0.10 | -0.86 | -0.15 |
| | *PCB* | 38 | 3038 | 2645 | 2250 | 665 | 8987 | 2615 | 10.83 | 2.80 |
| **Site 8** | *THg* | 162 | 0.18 | 0.07 | 0.18 | 0.07 | 0.33 | 0.10 | -0.21 | 0.47 |
| | *PCB* | 162 | 1188 | 1180 | 890 | 120 | 5272 | 975 | 8.72 | 2.67 |
| **Site 9** | *THg* | 218 | 0.15 | 0.08 | 0.12 | 0.03 | 0.34 | 0.10 | 1.01 | 1.07 |
| | *PCB* | 236 | 1251 | 1592 | 640 | 80 | 5300 | 5220 | 12.35 | 2.99 |

***Int Quart:*** Interquartile Range*;* ***Kurt:*** Kurtosis***; Skew:*** Skewness

**Table S6**: Summary statistics of the lipid content (%) for walleye and lake trout skinless-boneless fillet (*SBF*) data from six (6) and nine (9) sampling sites in Lake Ontario, respectively.

| | *N* | *Mean* | *Stdev* | *Median* | *2.50%* | *97.50%* | *Int* Quart* | *Kurt** | *Skew** |
|---|---|---|---|---|---|---|---|---|---|
| | | | | ***Walleye*** | | | | | |
| **Site 1** | 49 | 1.27 | 1.34 | 0.90 | 0.32 | 4.26 | 0.70 | 21.74 | 4.23 |
| **Site 2** | 39 | 0.97 | 0.42 | 0.88 | 0.48 | 1.91 | 0.44 | 2.12 | 1.39 |
| **Site 3** | 75 | 1.58 | 1.04 | 1.20 | 0.49 | 4.52 | 1.00 | 3.07 | 1.70 |
| **Site 4** | 220 | 1.08 | 0.71 | 0.90 | 0.11 | 2.90 | 0.80 | 3.37 | 1.44 |
| **Site 5** | 174 | 1.04 | 0.53 | 0.90 | 0.31 | 2.30 | 0.64 | 0.95 | 1.00 |
| **Site 6** | 197 | 2.76 | 5.77 | 1.04 | 0.20 | 25.1 | 1.20 | 12.96 | 3.65 |
| | | | | ***Lake Trout*** | | | | | |
| **Site 1** | 74 | 9.54 | 4.31 | 9.40 | 2.42 | 17.25 | 5.38 | 1.22 | 0.63 |
| **Site 2** | 180 | 9.28 | 4.72 | 8.71 | 2.14 | 20.21 | 4.95 | 2.65 | 1.21 |
| **Site 3** | 206 | 7.72 | 3.89 | 7.42 | 1.92 | 14.77 | 5.92 | 2.71 | 1.07 |
| **Site 4** | 113 | 7.75 | 4.43 | 7.53 | 1.21 | 16.92 | 6.01 | 0.65 | 0.78 |
| **Site 5** | 78 | 13.51 | 5.81 | 13.5 | 5.03 | 25.84 | 6.52 | -0.01 | 0.55 |
| **Site 6** | 76 | 10.31 | 4.52 | 9.55 | 3.61 | 19.25 | 4.13 | 7.63 | 1.92 |
| **Site 7** | 40 | 11.23 | 4.11 | 10.0 | 4.79 | 19.86 | 4.15 | 0.36 | 0.78 |
| **Site 8** | 162 | 9.28 | 4.65 | 8.43 | 2.60 | 19.98 | 5.71 | 1.25 | 0.92 |
| **Site 9** | 236 | 8.45 | 5.02 | 8.12 | 0.49 | 18.22 | 6.32 | 2.25 | 0.96 |

***Int Quart:*** Interquartile Range*;* ***Kurt:*** Kurtosis***; Skew:*** Skewness

**Table S7**: Summary statistics of the length (*cm*) for walleye and lake trout skinless-boneless fillet (*SBF*) data from six (6) and nine (9) sampling sites in Lake Ontario, respectively.

| | *N* | *Mean* | *Stdev* | *Median* | *2.50%* | *97.50%* | *Int Quart* | *Kurt** | *Skew** |
|---|---|---|---|---|---|---|---|---|---|
| | | | | ***Walleye*** | | | | | |
| **Site 1** | 49 | 60.01 | 13.12 | 62.12 | 25.78 | 77.91 | 15.02 | 1.14 | -1.12 |
| **Site 2** | 39 | 51.61 | 15.45 | 52.11 | 30.61 | 76.37 | 27.21 | -1.47 | 0.13 |
| **Site 3** | 75 | 60.19 | 11.69 | 62.51 | 36.82 | 78.64 | 15.81 | -0.25 | -0.47 |
| **Site 4** | 220 | 49.86 | 11.71 | 50.45 | 26.78 | 70.95 | 15.93 | 0.53 | -0.45 |
| **Site 5** | 174 | 46.12 | 13.35 | 45.42 | 22.31 | 69.87 | 19.95 | -0.43 | 0.11 |
| **Site 6** | 197 | 50.97 | 13.93 | 53.11 | 22.19 | 72.82 | 19.01 | -0.58 | -0.42 |
| | | | | ***Lake Trout*** | | | | | |
| **Site 1** | 74 | 66.39 | 6.01 | 66.23 | 55.17 | 76.35 | 6.88 | 0.36 | -0.26 |
| **Site 2** | 180 | 58.16 | 10.93 | 58.81 | 34.85 | 76.12 | 14.25 | -0.21 | -0.42 |
| **Site 3** | 206 | 62.41 | 8.451 | 63.02 | 45.31 | 78.66 | 9.51 | 0.83 | -0.48 |
| **Site 4** | 113 | 47.58 | 11.26 | 48.03 | 24.42 | 68.66 | 10.01 | 0.58 | -0.03 |
| **Site 5** | 78 | 64.75 | 10.42 | 66.25 | 41.04 | 79.81 | 11.31 | 2.09 | -1.18 |
| **Site 6** | 76 | 65.97 | 6.66 | 66.05 | 53.08 | 78.13 | 9.05 | 0.49 | -0.03 |
| **Site 7** | 40 | 69.78 | 8.71 | 71.12 | 52.85 | 83.06 | 12.18 | 0.04 | -0.52 |
| **Site 8** | 162 | 62.38 | 11.47 | 64.45 | 33.32 | 80.14 | 13.18 | 0.55 | -0.82 |
| **Site 9** | 236 | 57.12 | 12.66 | 59.02 | 29.48 | 78.15 | 15.95 | 0.33 | -0.52 |

***Int Quart:*** Interquartile Range*;* ***Kurt:*** Kurtosis***; Skew:*** Skewness

## Figures Legends

**Figure S1**: Box-plots of the differences between observed and predicted *PCB* concentrations in walleye collected from six locations in Lake Ontario during the 1980s, 1990s, and 2000s. Sites 1-6 are provided in Figure 1.

**Figure S2**: Box-plots of the differences between observed and predicted *THg* concentrations in walleye collected from six locations in Lake Ontario during the 1980s, 1990s, and 2000s. Sites 1-6 are provided in Figure 1.

**Figure S3**: Box-plots of the differences between observed and predicted *PCB* concentrations in lake trout collected from nine locations in Lake Ontario during the 1980s, 1990s, and 2000s. Sites 1-9 are provided in Figure 1.

**Figure S4**: Box-plots of the differences between observed and predicted *THg* concentrations in lake trout collected from nine locations in Lake Ontario during the 1980s, 1990s, and 2000s. Sites 1-9 are provided in Figure 1.

**Figure S5**: Box plots of the posterior parameter estimates for lake trout *THg* model.

**Figure S6**: Box plots of the posterior parameter estimates for lake trout *PCB* model.

**Figure S7**: Box plots of the posterior parameter estimates for walleye *THg* model.

**Figure S8**: Box plots of the posterior parameter estimates for walleye *PCB* model.

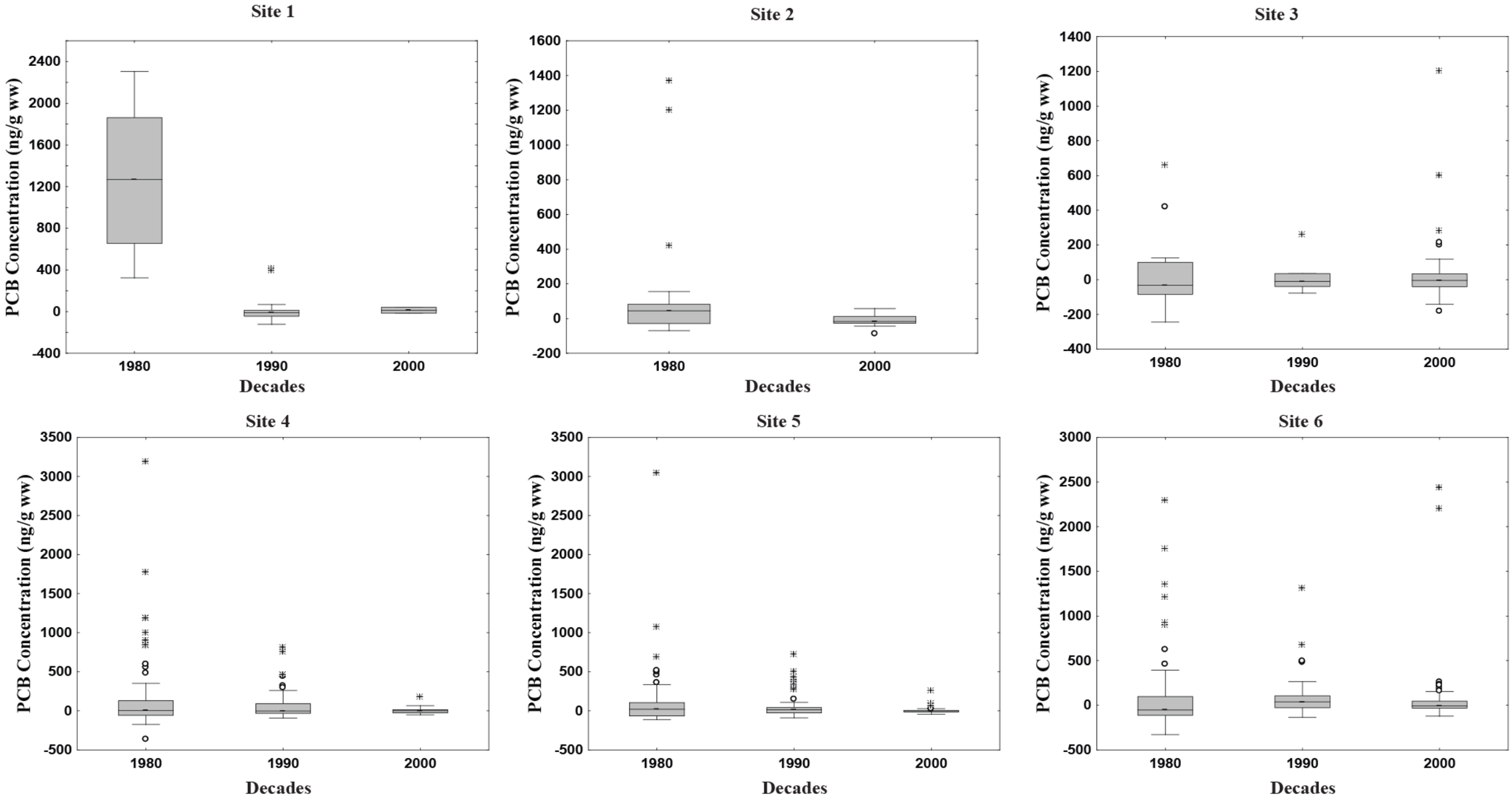

Site 1
PCB Concentration (ng/g ww)
2400
2000
1600
1200
800
400
0
-400
1980
1990
2000
Decades
Site 2
PCB Concentration (ng/g ww)
1600
1400
1200
1000
800
600
400
200
0
-200
1980
2000
Decades
Site 3
PCB Concentration (ng/g ww)
1400
1200
1000
800
600
400
200
0
-200
-400
1980
1990
2000
Decades
Site 4
PCB Concentration (ng/g ww)
3500
3000
2500
2000
1500
1000
500
0
-500
1980
1990
2000
Decades
Site 5
PCB Concentration (ng/g ww)
3500
3000
2500
2000
1500
1000
500
0
-500
1980
1990
2000
Decades
Site 6
PCB Concentration (ng/g ww)
3000
2500
2000
1500
1000
500
0
-500
1980
1990
2000
Decades

**Figure S1**

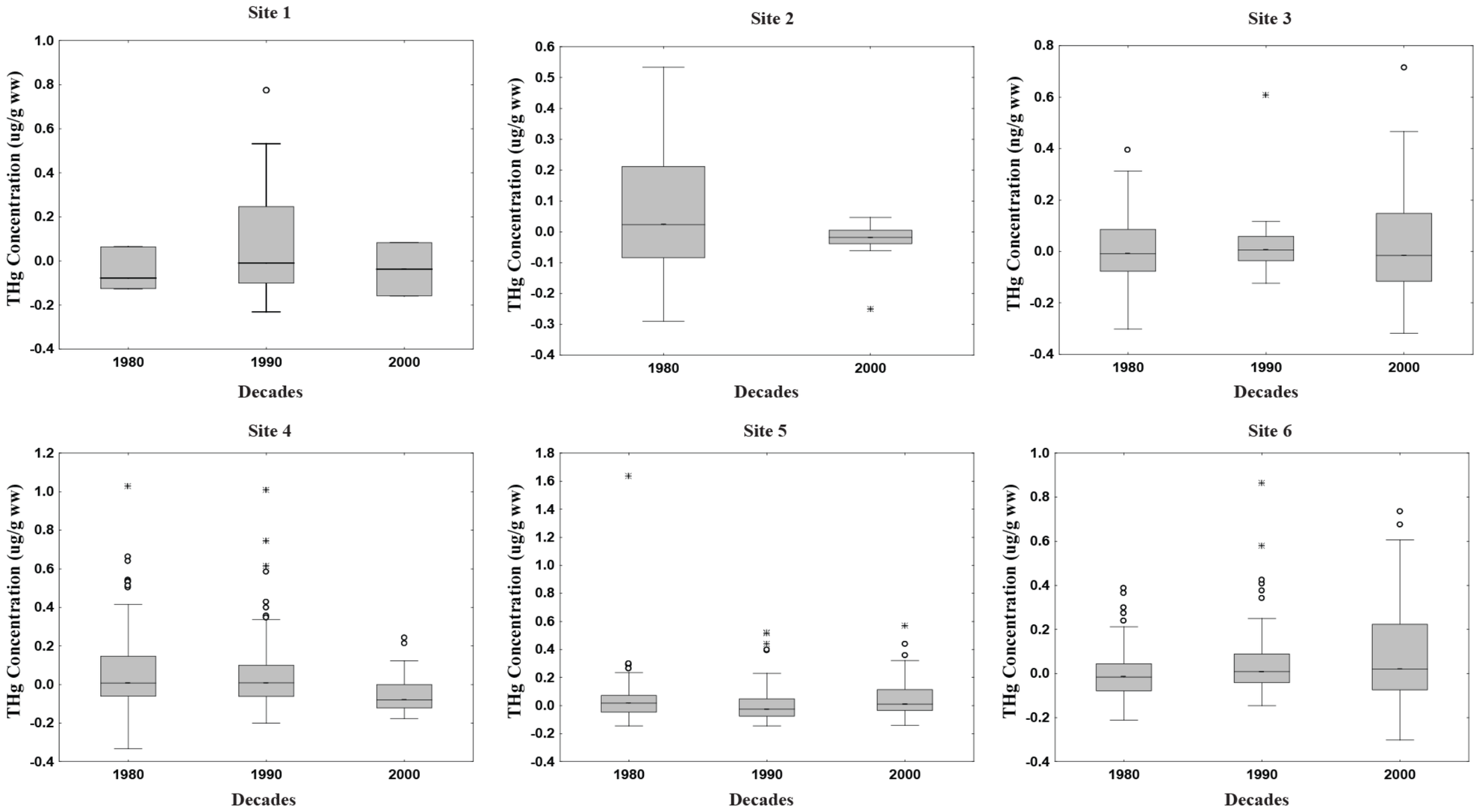

Site 1
THg Concentration (ug/g ww)
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
1980
1990
2000
Decades
Site 2
THg Concentration (ug/g ww)
0.6
0.5
0.4
0.3
0.2
0.1
0.0
-0.1
-0.2
-0.3
-0.4
1980
2000
Decades
Site 3
THg Concentration (ng/g ww)
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
1980
1990
2000
Decades
Site 4
THg Concentration (ug/g ww)
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
1980
1990
2000
Decades
Site 5
THg Concentration (ug/g ww)
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
1980
1990
2000
Decades
Site 6
THg Concentration (ug/g ww)
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
1980
1990
2000
Decades

**Figure S2**

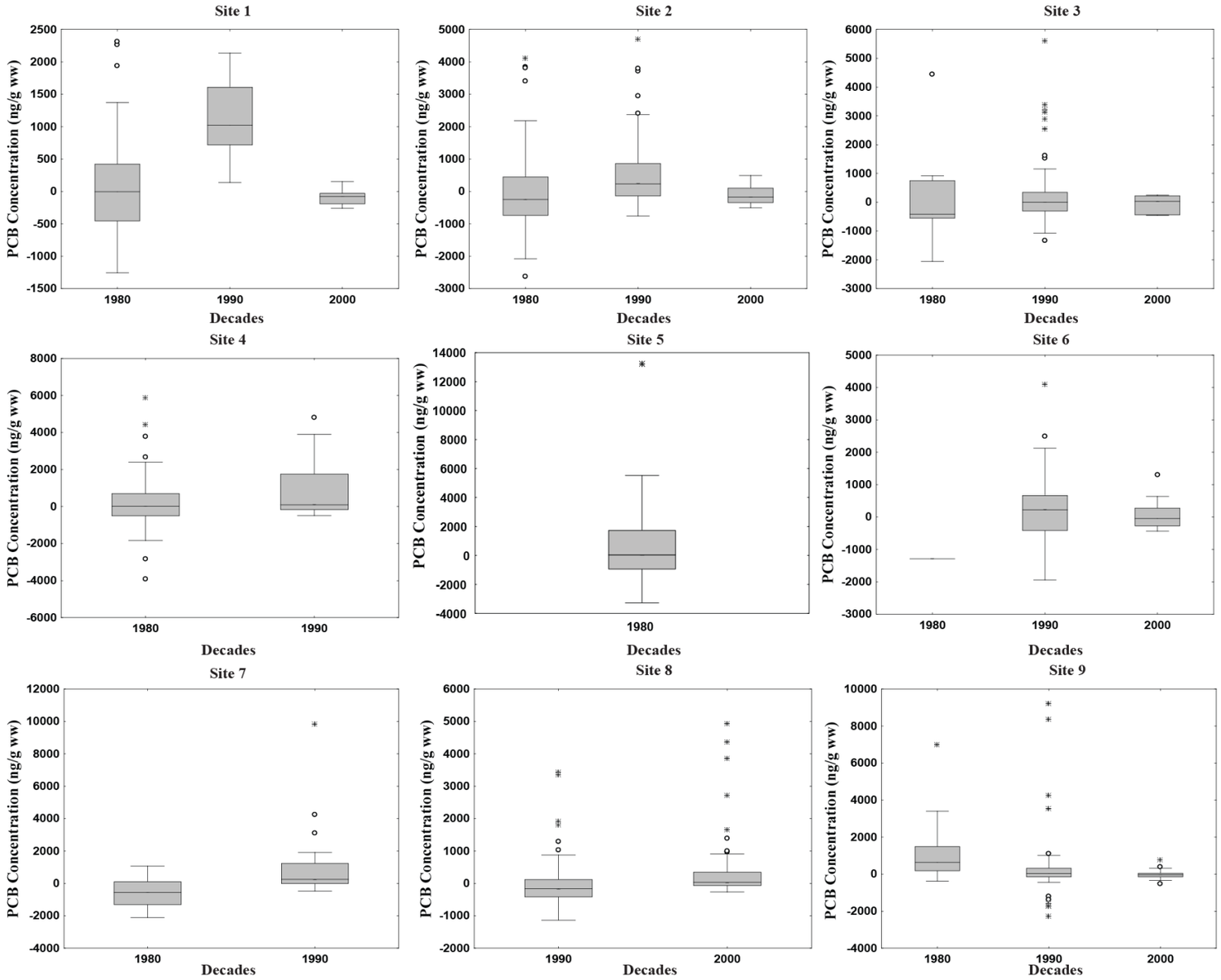

Site 1
PCB Concentration (ng/g ww)
2500
2000
1500
1000
500
0
-500
-1000
-1500
1980
1990
2000
Decades
Site 2
PCB Concentration (ng/g ww)
5000
4000
3000
2000
1000
0
-1000
-2000
-3000
1980
1990
2000
Decades
Site 3
PCB Concentration (ng/g ww)
6000
5000
4000
3000
2000
1000
0
-1000
-2000
-3000
1980
1990
2000
Decades
Site 4
PCB Concentration (ng/g ww)
8000
6000
4000
2000
0
-2000
-4000
-6000
1980
1990
Decades
Site 5
PCB Concentration (ng/g ww)
14000
12000
10000
8000
6000
4000
2000
0
-2000
-4000
1980
Decades
Site 6
PCB Concentration (ng/g ww)
5000
4000
3000
2000
1000
0
-1000
-2000
-3000
1980
1990
2000
Decades
Site 7
PCB Concentration (ng/g ww)
12000
10000
8000
6000
4000
2000
0
-2000
-4000
1980
1990
Decades
Site 8
PCB Concentration (ng/g ww)
6000
5000
4000
3000
2000
1000
0
-1000
-2000
1990
2000
Decades
Site 9
PCB Concentration (ng/g ww)
10000
8000
6000
4000
2000
0
-2000
-4000
1980
1990
2000
Decades

**Figure S3**

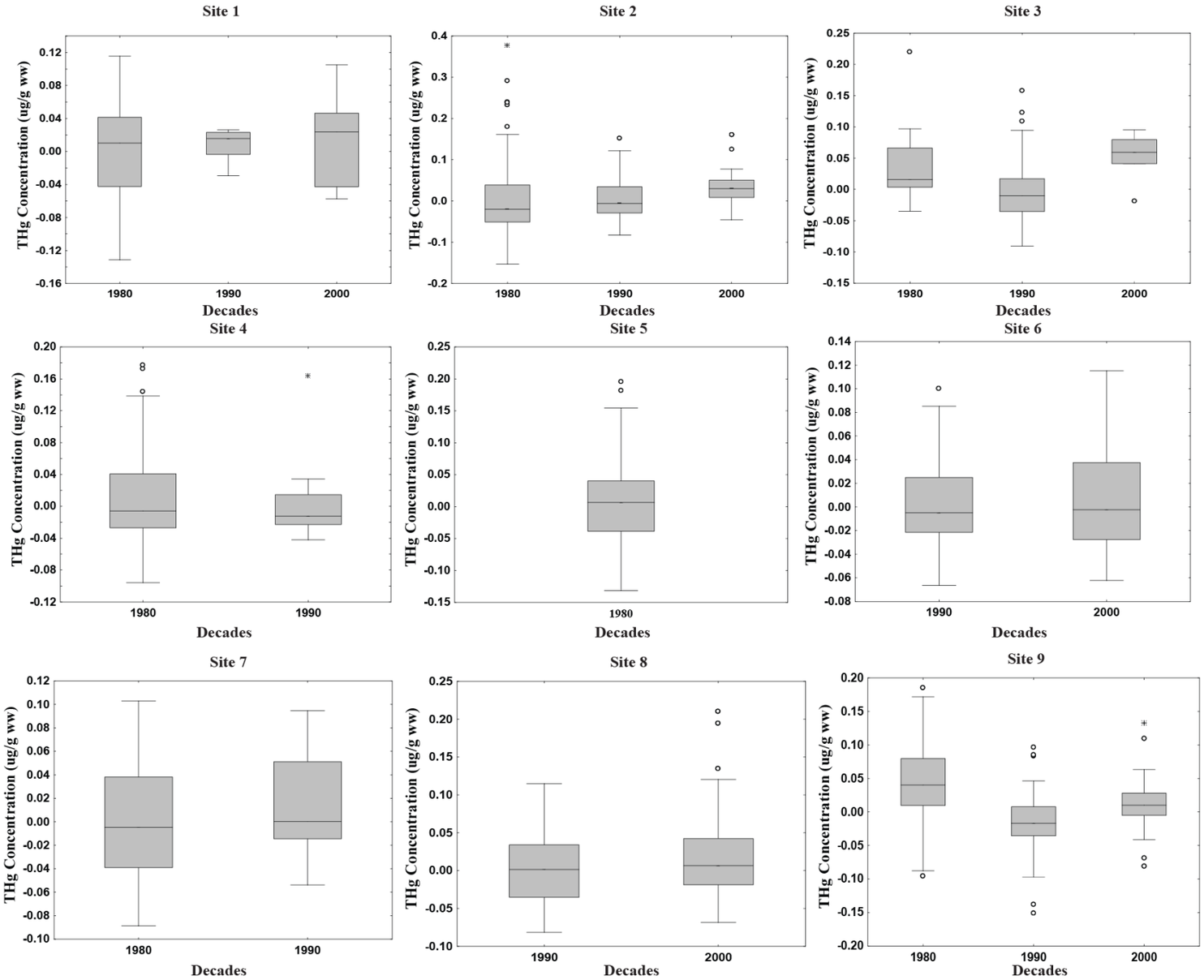
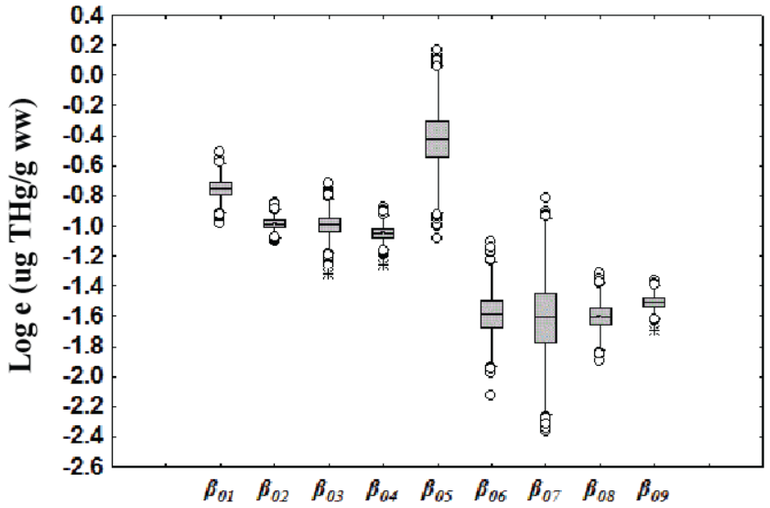
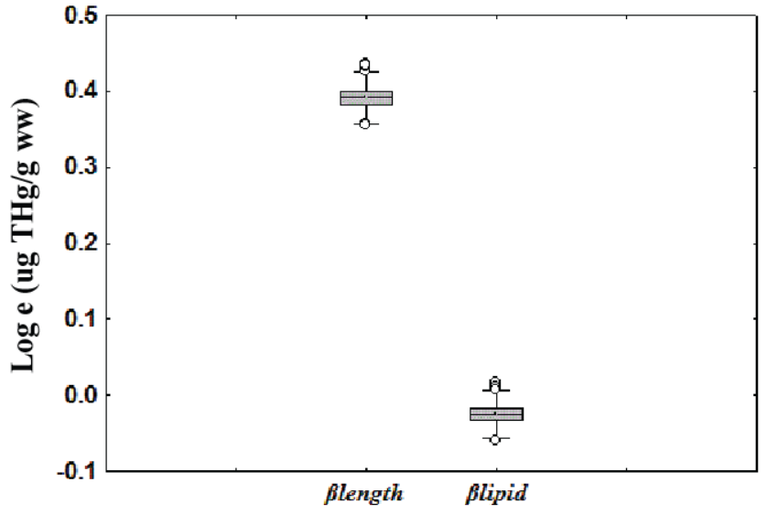

Site 1
Site 2
Site 3
Site 4
Site 5
Site 6
Site 7
Site 8
Site 9
THg Concentration (ug/g ww)
Decades
1980
1990
2000

**Figure S4**


Log e (ug THg/g ww)
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
-1.0
-1.2
-1.4
-1.6
-1.8
-2.0
-2.2
-2.4
-2.6
β01 β02 β03 β04 β05 β06 β07 β08 β09


Log e (ug THg/g ww)
0.5
0.4
0.3
0.2
0.1
0.0
-0.1
βlength
βlipid

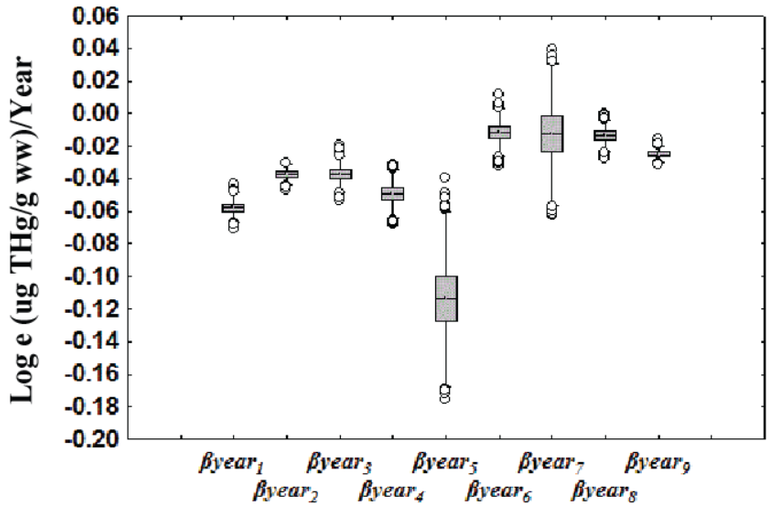
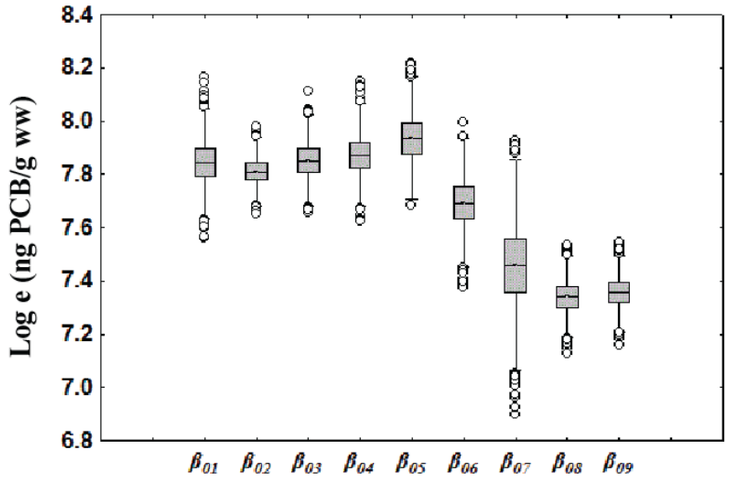
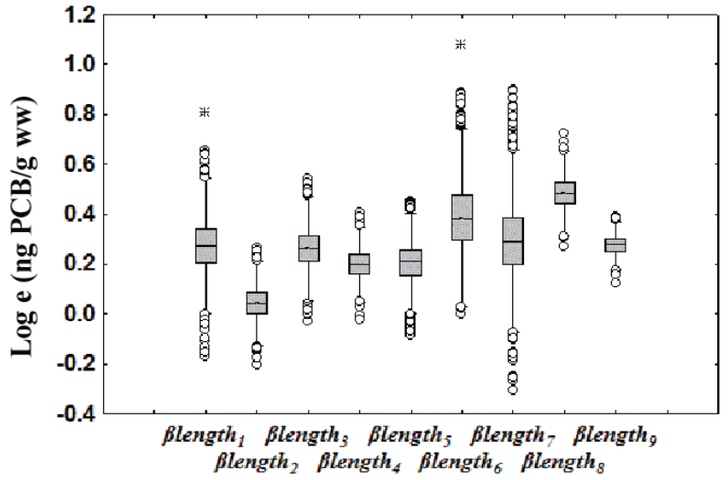
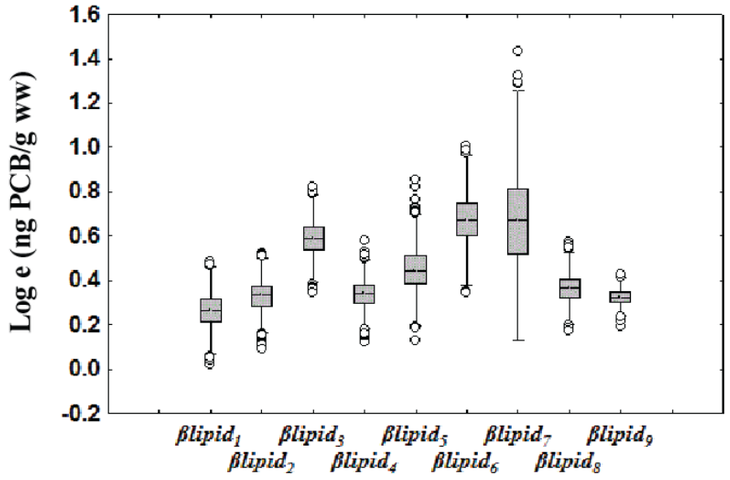

Log e (ug THg/g ww)/Year
0.06
0.04
0.02
0.00
-0.02
-0.04
-0.06
-0.08
-0.10
-0.12
-0.14
-0.16
-0.18
-0.20
βyear1 βyear3 βyear5 βyear7 βyear9
βyear2 βyear4 βyear6 βyear8

**Figure S5**


Log e (ng PCB/g ww)
8.4
8.2
8.0
7.8
7.6
7.4
7.2
7.0
6.8
β01 β02 β03 β04 β05 β06 β07 β08 β09


Log e (ng PCB/g ww)
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
βlength1 βlength3 βlength5 βlength7 βlength9
βlength2 βlength4 βlength6 βlength8


Log e (ng PCB/g ww)
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
βlipid1 βlipid3 βlipid5 βlipid7 βlipid9
βlipid2 βlipid4 βlipid6 βlipid8

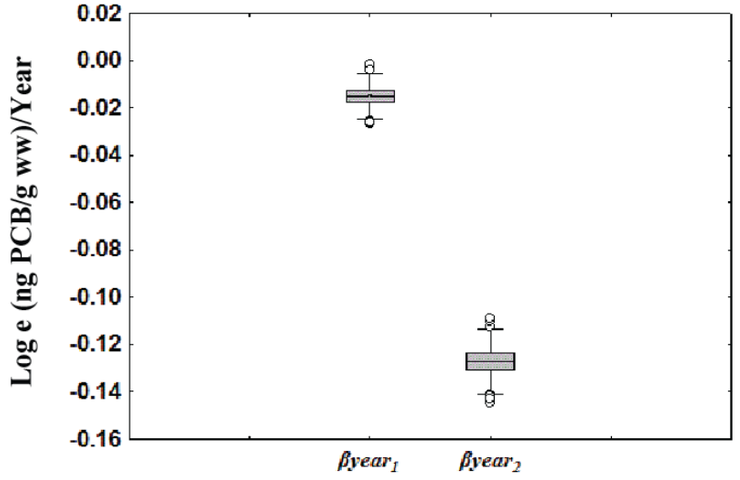

Log e (ng PCB/g ww)/Year
0.02
0.00
-0.02
-0.04
-0.06
-0.08
-0.10
-0.12
-0.14
-0.16
βyear1 βyear2

**Figure S6**

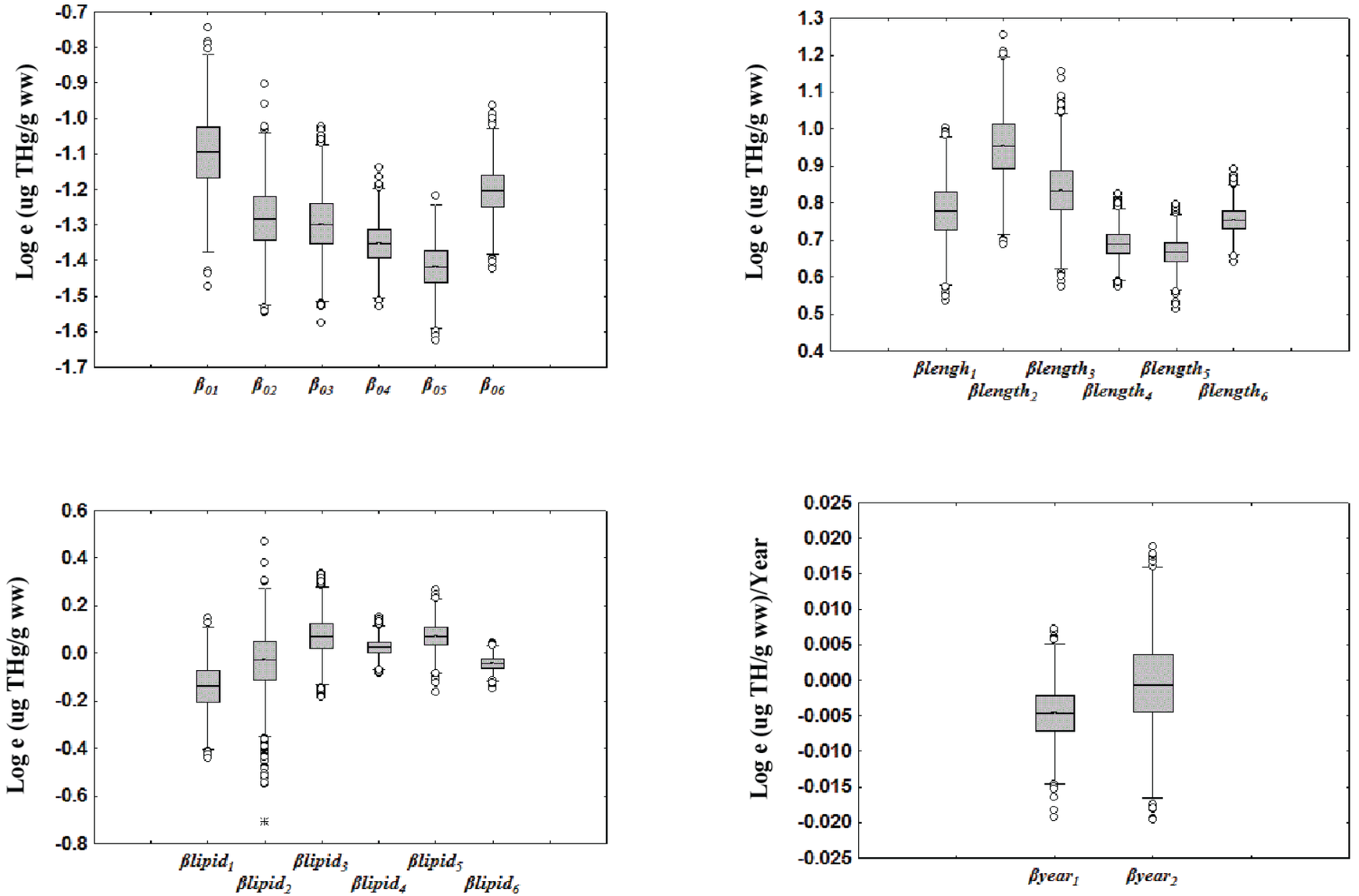
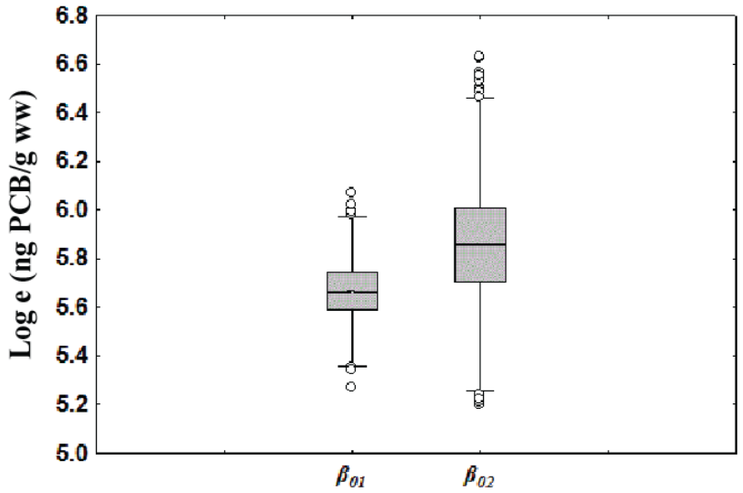
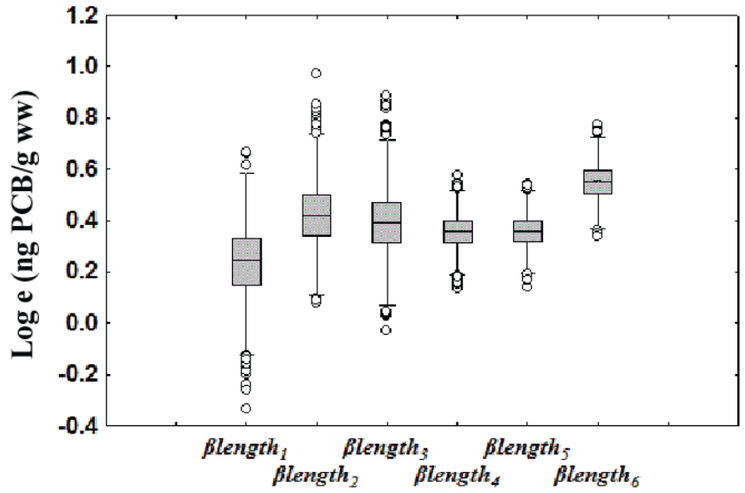
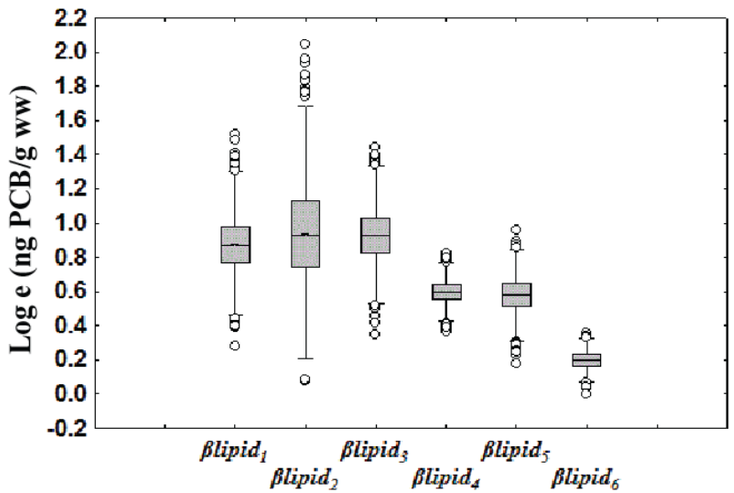

Log e (ug THg/g ww)
-0.7
-0.8
-0.9
-1.0
-1.1
-1.2
-1.3
-1.4
-1.5
-1.6
-1.7
β01
β02
β03
β04
β05
β06
Log e (ug THg/g ww)
1.3
1.2
1.1
1.0
0.9
0.8
0.7
0.6
0.5
0.4
βlength1
βlength2
βlength3
βlength4
βlength5
βlength6
Log e (ug THg/g ww)
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
βlipid1
βlipid2
βlipid3
βlipid4
βlipid5
βlipid6
Log e (ug TH/g ww)/Year
0.025
0.020
0.015
0.010
0.005
0.000
-0.005
-0.010
-0.015
-0.020
-0.025
βyear1
βyear2

**Figure S7**


Log e (ng PCB/g ww)
6.8
6.6
6.4
6.2
6.0
5.8
5.6
5.4
5.2
5.0
$\beta_{01}$
$\beta_{02}$


Log e (ng PCB/g ww)
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
$\beta length_1$
$\beta length_2$
$\beta length_3$
$\beta length_4$
$\beta length_5$
$\beta length_6$


Log e (ng PCB/g ww)
2.2
2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
$\beta lipid_1$
$\beta lipid_2$
$\beta lipid_3$
$\beta lipid_4$
$\beta lipid_5$
$\beta lipid_6$

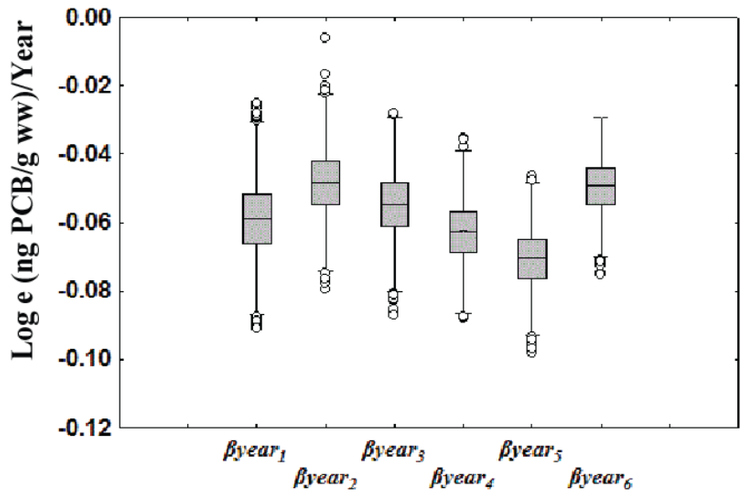

Log e (ng PCB/g ww)/Year
0.00
-0.02
-0.04
-0.06
-0.08
-0.10
-0.12
$\beta year_1$
$\beta year_2$
$\beta year_3$
$\beta year_4$
$\beta year_5$
$\beta year_6$

**Figure S8**