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A Bayesian assessment of the mercury and PCB temporal trends in lake trout (*Salvelinus namaycush*) and walleye (*Sander vitreus*) from lake Ontario, Ontario, Canada

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ABSTRACT

Polychlorinated biphenyls (*PCBs*) and total mercury (*THg*) are two of the most prevalent contaminants, resulting in restrictive advisories on consuming fish from the Laurentian Great Lakes. The goal of this study is to examine the temporal trends of the two contaminants in walleye (*Sander vitreus*) and lake trout (*Salvelinus namaycush*) for Lake Ontario. We employed Bayesian inference techniques to parameterize three different strategies of time series analysis: dynamic linear, exponential decay, and mixed-order modeling. Our analysis sheds light on the role of different covariates (length, lipid content) that can potentially hamper the detection of the actual temporal patterns of fish contaminants. Both *PCBs* and mercury demonstrate decreasing temporal trends in lake trout males and females. Decreasing *PCB* trends are evident in walleye, but the mean annual mercury levels are characterized by a “wax and wane” pattern, suggesting that specific fish species may not act as bio-indicators for all contaminants. This finding may be attributed to the shifts in energy trophodynamics along with the food web alterations induced from the introduction of non-native species, the intricate nature of the prey–predator interactions, the periodicities of climate factors, and the year-to-year variability of the potentially significant fluxes from atmosphere or sediments. Finally, a meaningful risk assessment exercise will be to elucidate the role of within-lake fish contaminant variability and evaluate the potential bias introduced when drawing inference from pooled datasets.

1. Introduction

The Laurentian Great Lakes comprise a total of five lakes (Lakes Superior, Michigan Huron, Erie and Ontario), and border eight *US* states and the Canadian Province of Ontario. This interconnected basin represents the largest freshwater basin on Earth, containing 21% of the world's surface fresh water by volume, and one of the most biologically diverse regions worldwide. Namely, the Great Lakes ecosystem contains various types of habitats, such as forests, marshes, wetlands, lakes, and dunes that host more than 3500 species of plants, fish and wildlife (Hales et al., 2008). Lake Ontario, in particular, plays a significant role in both the biodiversity of the Great Lakes eco-region as well as in the income generation for the upstate New York region, the Province of Ontario, the City of Toronto and its affiliated sub-divisions (Marbek et al., 2010). For example, a 15% increase of the fish abundance in Lake Ontario alone may represent economic benefits between \$2.9 and \$5.9 million per year, while a moderate reduction in the number (≈ 11) of beach closure days across all of Lake Ontario's beaches is valued at \$38.4–\$76.8 million annually (Marbek et al., 2010). On the other hand, the highly populated and industrialized nature of the surrounding watersheds combined with the long water residence times made the Great Lakes highly susceptible to anthropogenic disturbances, resulting in the decline of their ecological health during the 20th century.

The consumption of Great Lakes fish has been identified as the main human exposure route to many contaminants (Bhavsar et al., 2010, 2007). Among the toxic, persistent, and bioaccumulative contaminants, *PCBs* and mercury are of particular concern and restrict the use of valuable commercial and recreational fisheries resources in the Great Lakes. Although the two contaminants have contrasting bioaccumulation and biomagnification characteristics, they are both biologically active and mobile through different environments, making their mitigation control at both organismal and ecosystem levels a difficult task (Jones and De Voogt, 1999). Several important physical (e.g., lake area, epilimnetic temperature), chemical (e.g., organic matter, pH, mercury aging, iron levels, balance between sulfate and sulfide) and biological (e.g., type and

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activity of bacteria, food web structure, fish population age and growth rates) factors can potentially modulate the bioavailability and fate of mercury and may dramatically affect the transfer of *Hg* exogenous loads into methyl-mercury in fish (Kidd et al., 2012; Munthe et al., 2007). Methylmercury can biomagnify by approximately 10^6 times in top predators, thereby resulting in significantly higher mercury concentrations relative to the source water, even in areas remote from industrial sources (Fitzgerald et al., 1998). This organic form of *Hg* displays a high affinity to the sulfur-containing groups of host proteins (Wang and Wong, 2003; Hudson et al., 1998). As a result of this biochemical specificity, white muscle tissues characterized by a greater degree of protein content as well as being the most consumed portion of fish are often sampled for mercury detection. The toxic properties of mercury are often manifested as “mercury poisoning”, affecting the nervous system, kidney, liver and reproductive organs of human body. Some reliable symptoms of neurotoxicity in humans include neuronal loss, ataxia, visual disturbances, impaired hearing and death (FAO/WHO, 2003).

*PCB*s are highly lipophilic and more tightly deposit into fatty tissues (Elskul et al., 2005). Depending on their ethology and trophic position, fish typically receive *PCB*s through three specific routes, i.e., gills, epithelial/dermal tissues and gastrointestinal tract (Schlenk, 2005). Lower trophic level fish primarily receive contaminants by the diffusion process through gills and epithelial cells, whereas top predators mainly receive them through dietary uptake of contaminated food. As persistent organic pollutants, *PCB*s are non-ionizable and largely non-polar, while their lipophilic nature allows them to accumulate in fatty tissues and in oil rich organs and glands (Elskus et al., 2005). *PCB*s are known to suppress immune system function (Dallaire et al., 2003), to disrupt thyroid (Brucker-Davis, 1998) and sex hormonal function (Richthoff et al., 2003), and to increase the risk of diabetes (Codru et al., 2007). Long term exposure can cause endocrine system disruptions, leading to poor cognitive development in children born to mothers who had prolonged contact (Faroon et al., 2000).

Contaminant levels in Great Lakes fish have been routinely monitored for more than four decades, allowing the development of extensive datasets (e.g., Bhavsar et al., 2011; Gewurtz et al., 2011a,b). Statistical analyses of this long-term information provided evidence that the implementation of regulatory actions in the 1970s resulted in decreased levels of most contaminants in the Great Lakes environment through the 1980s. However, after the mid-1990s, the decline rates have been reported to be slower, stagnant, or even to have switched to increasing rates in recent years (Bhavsar et al., 2010, 2007). Before putting these trends into perspective, it is important to note that many retrospective analysis studies have not considered important influential factors of the perceived spatiotemporal trends, such as trophic level, fish age, size, gender, growth and lipid content (Sadraddini et al., 2011a,b; Stow et al., 1997). Variations across monitoring programs in the type of sampling procedures and the different statistical methods used may also impede the impartial detection of contaminant trends (Sadraddini et al., 2011a,b; Gewurtz et al., 2011a,b). It is thus imperative to embrace more robust statistical frameworks when undertaking such retrospective analyses, in order to draw inference about the actual contaminant trends with acceptable confidence. To this end, a central feature of recent statistical modeling work has been the adoption of Bayesian inference techniques as a means for critically assessing spatiotemporal contaminant trends in fish communities. The advantage of the Bayesian approach primarily stems from its capacity to explicitly accommodate model (structural and parametric) uncertainty, measurement error, data gaps, and variant degrees of prior knowledge about the ecological questions examined (Azim et al., 2011a,b; Sadraddini et al., 2011a,b; Mahmood et al., 2013a,b).

In this study, we attempt to evaluate the historical and current levels of *PCB*s and mercury in Lake Ontario fish, building upon recent Bayesian modeling work in this line of research. Specifically, we use Bayesian inference techniques to parameterize three different strategies of time series analysis: dynamic linear, exponential decay, and mixed-order modeling. Because the latter two models postulate monotonic decrease of the contaminant levels, we include first-order random walk terms in our statistical formulations to accommodate non-monotonic temporal patterns. Our analysis aims to elucidate the role of different covariates (length, lipid content) that can potentially impede the detection of the actual temporal patterns of fish contaminants. Our study focuses on lake trout and walleye due to their popularity for consumption, economic importance, and/or ecological significance for the Lake Ontario ecosystem. We also examine separately the temporal patterns between males and females of the two species. The analysis of the temporal trends of *PCB*s and mercury concentrations in Lake Ontario fish is important in an ecological as well as in a public health risk context. Based on 40 years of contaminant data, our study offers a unique opportunity to tease out the temporal variability of mercury and *PCB*s in two important members of the Lake Ontario food web and to shed light on significant ecological mechanisms that may be driving these trends.

2. Methods

2.1. Dataset description-Chemical analysis

Our study used fish contaminant data from the Ontario Ministry of the Environment and Climate Change (*OMOECC*) Sport Fish Contaminant Monitoring Program, which routinely collects samples of a wide range of fish species and analyzes contaminant levels mainly in the dorsal skinless–boneless fillet portions. This information is then used to develop fish consumption advisories. In our analysis, we selected lake trout and walleye based on their role as biological indicators (or tracers) of contaminant variability. Being a more oily species, lake trout would better display the degree of *PCB* contamination, whereas walleye could be analyzed to better identify mercury levels due to its role as a higher trophic species (as mercury experiences a greater degree of biomagnification). These species are also popular with the angler community as they tend to grow relatively large in size, thereby providing a challenging and worthwhile catch. All samples were collected from various locations from Lake Ontario (Fig. 1) and correspond to a time span of approximately four decades (1975–2011). The present analysis explicitly considers the likelihood of covariance between contaminant trends and fish gender, length (as a surrogate of age), lipid content.

The *OMOECC* samples were analyzed for *THg* by employing the cold vapor-flameless atomic absorption spectrophotometry (*CV-FAAS*) technique at the *OMOECC* laboratory in Toronto using the *OMOECC* method *HGBIO-E3057*. The method detection limit is $0.01\ \mu g\ g^{-1}$. This method is 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). 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

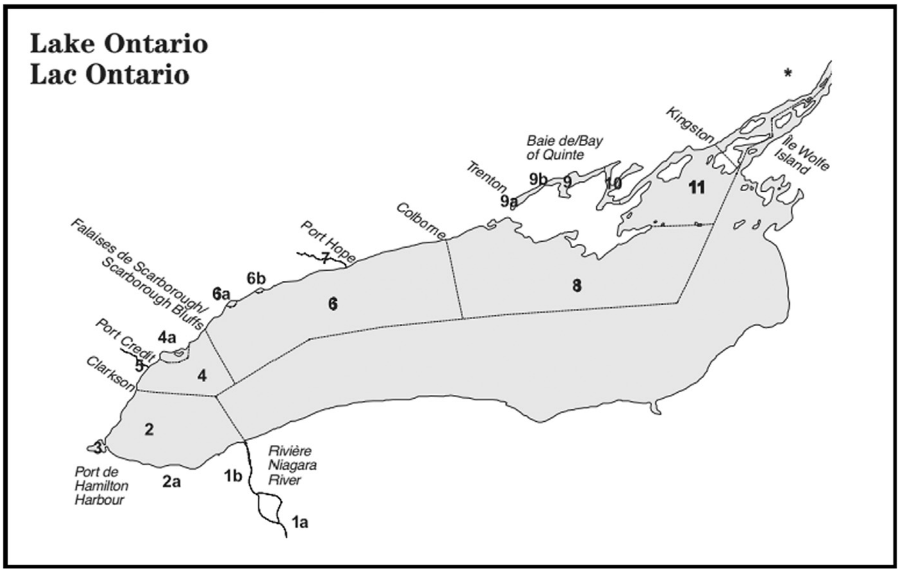

Fig. 1. Map of Lake Ontario illustrating the sampling sites used to create the Ontario Ministry of the Environment dataset (OMOE, 2013).

blank and spiked blank matrix sample was processed with each set of samples (20–30). The method performance is monitored through laboratory intercalibration studies (the Northern Contaminants Program and Quality Assurance of Information for Marine Environmental Monitoring in Europe).

2.2. Modeling framework

Bayesian inference was used as a means for estimating model parameters due to its ability to include prior information (e.g., literature reviews, expert knowledge, metadata, past parameter estimates) in the modeling analysis and to explicitly handle model structural/parametric uncertainty as well as data gaps and measurement errors (Gelman et al., 2004). Bayesian inference treats each parameter θ as random variable, and uses the likelihood function to express the relative plausibility of obtaining different values of this parameter when particular data have been observed:

$$\pi(\theta|data) = \frac{\pi(\theta)L(data|\theta)}{\int_\theta \pi(\theta)L(data|\theta)d\theta} \tag{1}$$

where $\pi(\theta)$ represents our prior statements regarding the probability distribution that more objectively depicts the existing knowledge on the θ values, $L(data|\theta)$ corresponds to the likelihood of observing the data given the different θ values, and $\pi(\theta|data)$ is the posterior probability that expresses our updated beliefs on the θ values after the existing data from the system are considered. The denominator in Eq. (1) is the expected value of the likelihood function, and acts as a scaling constant that normalizes the integral of the area under the posterior probability distribution.

2.3. Exponential decay and mixed-order modeling

The first step of the analysis was based on the exponential decay and mixed-order models (Stow et al., 2004):

$$\left[THg_t/PCB_t\right] = \left[THg_0/PCB_0\right]e^{kt} + \varepsilon \tag{2}$$

$$\left[THg_t/PCB_t\right] = \left\{\left[THg_0/PCB_0\right]^{1-\varphi} - kt(1-\varphi)\right\}^{(1/1-\varphi)} + \varepsilon \tag{3}$$

where $[THg_t/PCB_t]$ is the contaminant concentration in year t; $[THg_0/PCB_0]$ is the contaminant concentration at $t=0$; k is the decay coefficient; and φ is the order of the reaction. The Bayesian configuration of the single exponential and mixed-order models was based on informative normal priors for $[THg_0/PCB_0]$, parameterized such that 95% of the respective values were lying within the minimum and maximum concentrations in the first year examined. By contrast, a practically flat (non-informative) prior was assigned to the k coefficient of the single exponential model, constrained to sample negative values, i.e., $k \in [-\infty,0]$. In a similar manner, the two parameters of the mixed-order model were assigned flat priors constrained within realistic ranges, i.e., $k \in [0,1]$ and $\varphi \in [1,4]$. Detailed sensitivity analysis of the prior parameter specifications to the posterior patterns of the single exponential and mixed-order models has been presented in several recent studies (Azim et al., 2011a; Sadraddini et al., 2011a).

A fundamental weakness of the two models is the postulation of a monotonic decrease of the contaminant levels, and therefore their inability to capture systematic deviations from this trend. In this study, to accommodate possible non-monotonic patterns in the time series data, we included (zero mean) random error terms δ_t representing the annual deviations from the trajectory delineated by the single exponential decay equation:

$$\left[THg_t/PCB_t\right] = \left[THg_0/PCB_0\right]e^{kt} + \delta_t + \varepsilon \tag{4}$$

To reflect the prior belief that these annual discrepancies are correlated, we assumed a first-order random walk prior specified as (Shaddick and Wakefield, 2002; Azim et al., 2011a; Sadraddini et al., 2011a):

$$p(\delta_t|\delta_{-t},\omega^2) \sim \begin{cases} N(\delta_{t+1},\omega^2) \text{for } t=1 \\ N\left(\dfrac{\delta_{t-1}+\delta_{t+1}}{2},\dfrac{\omega^2}{2}\right) \text{for } t=2,\ldots,T-1 \\ N(\delta_{t-1},\omega^2) \text{for } t=T \end{cases} \tag{5}$$

where δ_{-t} denotes all elements of δ_t except from the error associated with a particular year t, ω^2 is the conditional variance and the prior densities $p(\omega^{\mathbf{2}})$ were based on conjugate inverse-gamma (0.001, 0.001) distributions. Our conditional autoregressive approach implies that the first-order differences of the annual contaminant levels are smooth, and that the probability of sudden jumps between consecutive years is unlikely. The ε term represents the measurement error and is assumed to follow a Gaussian distribution, $N(0, \sigma_\varepsilon^2)$. Contrary to the time variant random error terms δ_t, the measurement error does not depend on time and the prior density $p(\sigma_\varepsilon^2)$ was again based on a conjugate inverse-gamma (0.001, 0.001) distribution. Following the typical practice in the literature, the exponential decay and mixed-order models were applied to prespecified length ranges for the two fish species, i.e., 55–65 cm and 45–55 cm for lake trout and walleye, respectively (Bhavsar et al., 2007; 2010).

2.4. Dynamic linear modeling

We developed a series of *DLMs* to consolidate the temporal contaminant trends, while explicitly accounting for the fact that fish length and lipid content typically co-vary with the contaminant concentrations and that fish of different sizes and lipid compositions may have been sampled over time in Lake Ontario. To compare the relative influence of each of these covariates, we ran the *DLMs* for each congener-fish species-gender combination a total of three times: using the fish length or lipid content alone or both fish length and lipid content as covariates. We thus ran a total of 36 models (2 fish species × 3 gender classifications × 2 compounds × 3 covariate combinations) over the course of this study. Unlike static regression models that have fixed parameters, *DLMs* have an evolving structure that allows parameters to shift through time (Lamon et al., 1998). This "dynamic" feature allows our models to more accurately reflect the intra- and interannual variability of the underlying ecological processes and the level of the response variable. An important property of these models 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 (Lamon et al., 1998; West and Harrison, 1989). *DLM* posterior estimates are influenced only by prior and current information (not subsequent data), which is another distinct feature relative to traditional regression analyses (Azim et al., 2011b). Furthermore, *DLMs* minimize the impact of outliers and easily handle missing values or unequally spaced data. Parameters in these models are time-specific, but are also related to one another stochastically by virtue of an error term (Pole et al., 1994).

The main components of any *DLM* are an observation equation and subsequent system equations.

2.4.1. Observation equation

$$ln[THg/PCB]_{ti} = level_t + \beta_{t1}\,ln[length]_{ti} + \beta_{t2}\,ln[lipid]_{ti} + \psi_{ti}$$
$$\psi_{ti} \sim N[0, \Psi_t] \qquad (6)$$

2.4.2. System equations

$$level_t = level_{t-1} + rate_t + \omega_{t1} \qquad \omega_{t1} \sim N[0, \Omega_{t1}] \qquad (7)$$

$$rate_t = rate_{t-1} + \omega_{t2} \qquad \omega_{t2} \sim N[0, \Omega_{t2}] \qquad (8)$$

$$\beta_{t1} = \beta_{t1-1} + \omega_{t3} \qquad \omega_{t3} \sim N[0, \Omega_{t3}] \qquad (9)$$

$$\beta_{t2} = \beta_{t2-1} + \omega_{t4} \qquad \omega_{t4} \sim N[0, \Omega_{t4}] \qquad (10)$$

$$\begin{array}{ll} 1/\Omega_{tj}^2 = \zeta^{t-1} \cdot 1/\Omega_{1j}^2,\ 1/\Psi_t^2 = \zeta^{t-1} \cdot 1/\Psi_1^2 & t > 1 \text{ and } j = 1 \text{ to } 4 \\ level_1,\ rate_1,\ \beta_1 \sim N(0, 10000) & t = 1 \\ 1/\Omega_{1j}^2,\ 1/\Psi_1^2 \sim G(0.001, 0.001) & \end{array} \qquad (11)$$

where $\ln[THg/PCB]_{ti}$ is the observed *THg/PCB* concentration at time t in the individual sample i; $level_t$ is the mean *THg/PCB* concentration at time t when accounting for the covariance with the fish length and lipid content; $\ln[length]_{ti}$ is the observed (standardized) fish length at time t in the individual sample i; $ln[lipid]_{ti}$ is the observed (standardized) fish lipid content; $rate_t$ is the rate of change of the level variable; β_{t1} is a length (regression) coefficient; β_{t2} is a lipid (regression) coefficient; ψ_t, ω_{tj} 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; $N(0, 10000)$ is the normal distribution with mean 0 and variance 10,000; 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}, $1/\Omega_{1j}^2$, and $1/\Psi_t^2$ are considered "non-informative" or vague.

The sequential updating of a *DLM* makes a forecast for time t based on prior knowledge of the parameters, and then we observe data at time t (Lamon et al., 1998). Based on Bayes' Theorem, our knowledge regarding the parameters is updated using the likelihood of the data and the prior knowledge we have (Congdon, 2003). A discount factor is then applied to this new posterior belief, such that older observations are weighted less than newer ones; the discounted posterior then becomes the prior for the next time step, and the process is repeated. In this analysis, we introduce non-constant and data-driven variances (with respect to time) using a discount factor on the first period prior (West and Harrison, 1989). Discount factors between 0.8 and 1.0 were examined during the specification of our modeling framework. We settled on a value of 0.95 that optimally balances between performance, i.e., deviance ($= -2\log[\text{likelihood}]$) values, and uncertainty of the year-specific estimates of the stochastic nodes considered, i.e., regression coefficients, rates of change, fish contaminant concentrations corrected for the lipid content and fish length variability as well as the error terms. That is, we selected the discount factor that resulted in the highest model performance, while maintaining the best possible identification level, as expressed by the coefficient of variation (and/or the signal-to-noise ratio) values of the corresponding model inputs. The likelihood of bias due to multiple measurements below the detection limit was considered, using a Tobit dynamic linear modeling approach (Azim et al., 2011b; Mahmood et al., 2013a,b). The determination of the most parsimonious model for each fish species/contaminant/gender 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).

2.5. Model computations

Using Markov-chain Monte Carlo (*MCMC*) simulations (Gilks et al., 1998), we obtained sequences of realizations from the model posterior distributions. We used a general normal proposal Metropolis algorithm that is based on a symmetric normal proposal 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). 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).

3. Results

3.1. *THg/PCB* levels

The summary statistics of the two contaminants give a general sense of the species- and gender-specific contamination levels in Lake Ontario over the course of four decades (Table 1). Because these summary statistics are based on pooled data, it should be cautioned that the pairwise comparisons between genders or species can be somewhat misleading, as some of the relative differences have changed in magnitude over time (see also following trend analysis). Taking this point into consideration, we note that walleye was characterized by distinctly higher *THg* concentrations (mean 0.38 and median 0.26 $\mu g\, g^{-1}$wet weight or ww) relative to lake trout (0.21 and 0.20 $\mu g\, g^{-1}$ww). Interestingly, the *THg* levels in lake trout were practically identical between the two genders, whereas the female walleye demonstrated higher concentrations (0.46 and 0.35 $\mu g\, g^{-1}$ ww) relative to their male counterparts (0.33 and 0.23 $\mu g\, g^{-1}$ww). By contrast, lake trout had significantly higher *PCB* concentrations (2076 and 1513 $ng\, g^{-1}$ww) comparing with walleye (220 and 100 $ng\, g^{-1}$ ww) in Lake Ontario (Table 1). For walleye, the males demonstrated higher *PCB* levels (276 $ng\, g^{-1}$ ww for males versus for 175 $ng\, g^{-1}$ ww females) and the opposite was true for lake trout (2101 $ng\, g^{-1}$ ww for females versus 2012 $ng\, g^{-1}$ ww for males). Moreover, the high standard deviation and interquartile range values reflect the substantial inter- and intra-annual variability associated with the contaminant levels of the individual fish species. The positive skewness and kurtosis values for both contaminants suggest right skewed and leptokurtic distributions; thus, the natural log transformation was implemented for the subsequent modeling analysis, effectively imposing a log–normal error structure. In the Supporting Information, Tables 1-SI to 4-SI 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 with no significant differences between the two genders. By contrast, the females sampled tended to be somewhat longer (≈ 8 cm for walleye and ≈ 1 cm for lake trout) relative to their male counterparts.

3.2. Exponential decay and mixed-order modeling

The posterior estimates for the models used to describe the temporal mercury trends in lake trout and walleye are shown in Table 5-SI. The lower *DIC* values render support to the augmented exponential decay model (Eq. (4)), coupled with the first-order temporal smoothing, relative to the conventional formulation (Eq. (2)), expressing continuously decreasing trends in *THg* concentrations at an ever-slowing rate towards a zero concentration. Notably, the mixed-order model consistently fared worse than the other two models. The two configurations of the exponential decay model suggest significantly higher rates of *THg* decrease for lake trout ($k = -0.036 \pm 0.002\ yr^{-1}$ and $k = -0.036 \pm 0.005\ yr^{-1}$) than for walleye ($k = -0.005 \pm 0.004\ yr^{-1}$ and $k = -0.016 \pm 0.006\ yr^{-1}$). Female lake trout ($k = -0.045 \pm 0.004\ yr^{-1}$ and $k = -0.061 \pm 0.006\ yr^{-1}$) were characterized by higher rates of decrease compared to their male counterparts ($k = -0.037 \pm 0.002\ yr^{-1}$ and $k = -0.054 \pm 0.007\ yr^{-1}$). The modeled *THg* concentrations for lake trout decreased until the mid-1990s, then remained more or less constant until the mid-2000s, after when continually decreased at a relatively slow rate (Fig. SI-1a, b and e). The δ_t (random walk) terms were used to detect the systematic errors resulting from the structural inadequacies of the single exponential model. In particular, we note the positive values of the structural error terms during the second half of the survey period with the gender-specific models (Figs. SI-2a and b). Interestingly, the same models predicted higher initial THg_0 values ($0.422 \pm 0.033\ \mu g\, g^{-1}$ ww and $0.421 \pm 0.043\ \mu g\, g^{-1}$ ww) relative to their simpler counterparts ($0.345 \pm 0.019\ \mu g\, g^{-1}$ ww and $0.323 \pm 0.012\ \mu g\, g^{-1}$ ww) as well as higher decay coefficients. When we pooled all the data together, the increasing trend of the random walk terms is not manifested, but rather is replaced by a wax and wane pattern (Fig. SI-2e). Interestingly, the posterior THg_0 and k estimates are practically identical between the two configurations of the exponential decay model. The mean predicted *THg* concentrations for walleye remained essentially unaltered during our study period, as the original decline during the 1980s was counterbalanced by a distinct upward shift in the early/mid-1990s (Fig. SI-3a, b and e).

The *DIC* comparison of the models developed for the two fish species with the *PCB* data is again supportive of the augmented exponential decay formulation to describe the trends (Table 6-SI). According to this model configuration, the *PCB* concentrations have been decreasing with approximately similar rates for lake trout ($k = -0.070 \pm 0.004\ yr^{-1}$) and walleye ($k = -0.064 \pm 0.011\ yr^{-1}$), when we pooled all the data together. However, the gender-specific models predict somewhat higher rates for female lake trout ($k = -0.125 \pm 0.005\ yr^{-1}$ versus $= -0.075 \pm 0.007\ yr^{-1}$ for males) and male walleye ($k = -0.170 \pm 0.014\ yr^{-1}$ versus $= -0.128 \pm 0.022\ yr^{-1}$ for females). The modeled mean *PCB* concentrations in the two fish species decreased until the late 1980s, then remained more or less constant through the early/mid-1990s, after when the *PCB* levels decreased at slower rates until 2010 (Figs. SI-1c, d and f and SI-3c, d and f). The structural error (δ_t) terms of the "pooled" models displayed a wax and wane pattern throughout the survey period (Figs. SI-2f and SI-4f), but there were gender-specific models in which the previously

Table 1
Summary statistics of *THg* ($\mu g/g$ wet weight) and *PCB* (ng/g wet weight) concentrations in lake trout and walleye skinless-boneless fillet data in Lake Ontario.

Species	Gender	N	Mean	Stdev	Median	2.50%	97.50%	Int Quart	Kurtosis	Skewness
Lake Trout	**Female**	339	0.21	0.10	0.21	0.06	0.44	0.13	0.28	0.60
	Male	459	0.21	0.09	0.20	0.06	0.43	0.12	0.12	0.64
	Pooled Samples	1023	0.21	0.10	0.20	0.06	0.44	0.13	0.28	0.67
Walleye	**Female**	351	0.46	0.36	0.35	0.06	1.23	0.58	0.77	1.02
	Male	289	0.33	0.29	0.23	0.04	1.00	0.34	1.84	1.47
	Pooled Samples	745	0.38	0.33	0.26	0.05	1.20	0.41	1.50	1.33
Species	**Gender**	**N**	**Mean**	**Stdev**	**Median**	**2.50%**	**97.50%**	**Int Quart**	**Kurtosis**	**Skewness**
Lake Trout	**Female**	361	2101	1968	1458	180	7000	2048	4.26	1.89
	Male	476	2012	2048	1404	120	6688	2026	16.54	2.98
	Pooled Samples	1030	2076	1984	1513	157	6919	2070	11.70	2.53
Walleye	**Female**	351	175	333	78	20	1151	144	32.56	5.10
	Male	295	276	462	120	20	1585	263	17.95	3.83
	Pooled Samples	754	220	384	100	20	1401	187	24.32	4.37

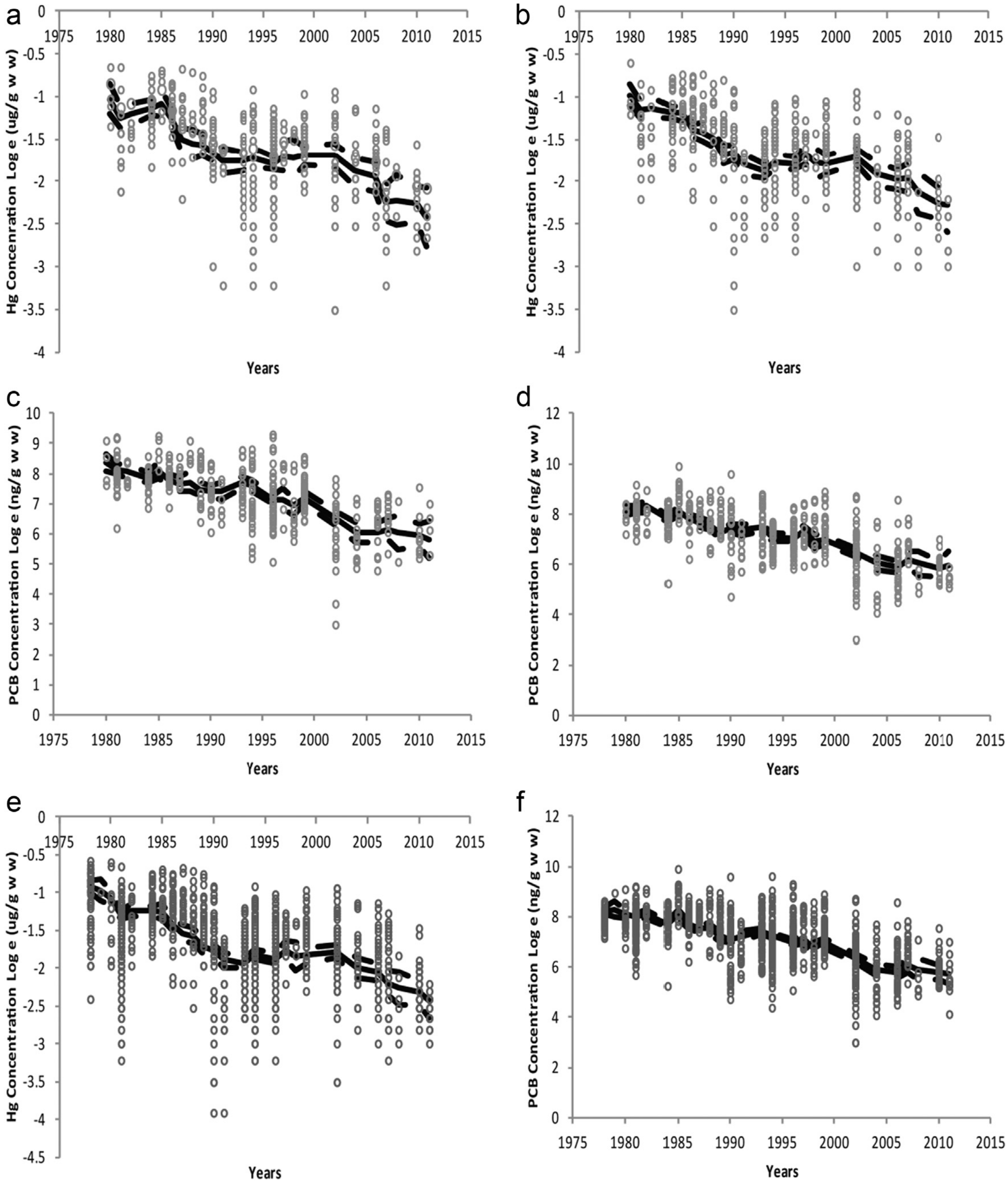

Fig. 2. Dynamic linear modeling analysis for lake trout depicting the actual *THg* (μg/g ww) and *PCB* (ng/g ww) concentrations (gray dots) against the predicted annual trends when accounting for the covariance with fish length and lipid content (black lines). The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted *THg* and *PCB* concentrations for females (a and c), males (b and d) and all data (e and f).

reported increasing trend over time were manifested, i.e., female lake trout (Fig. SI-2c), female walleye (Fig. SI-4c), and male walleye (Fig. SI-4d). Again, it is interesting to note that the same models predicted higher initial PCB_0 values (6980 ± 325 ng g^{-1} ww, 451 ± 128 ng g^{-1} ww, and 1771 ± 349 ng g^{-1} ww) relative to their simpler counterparts (6391 ± 311 ng g^{-1} ww, 162 ± 34 ng g^{-1} ww, and 566 ± 145 ng g^{-1} ww) as well as higher decay coefficients. For a comparison, the predictions of the exponential decay (without the conditional autoregressive terms) and mixed-order models are illustrated in Figs. SI-5 to SI-8.

3.3. Dynamic linear modeling

The *DIC* comparison suggests that the dynamic linear models collectively considering fish lengths and lipids outperformed those accounting for the two covariates individually (Tables 7-SI and 8-SI). The identifiability patterns ($\hat{\beta}/\sigma_\beta$) of the regression coefficients associated with the fish length are indicative of its strong signature to both *THg* and *PCB* variability. The same trend holds true with the lipid content and the *PCB* fish data. Because of the differences in the processes that regulate its bioaccumulation, the causal association between lipid levels and *THg* concentrations is weaker with walleye and even has a negative relationship with lake trout. The same patterns are evident when we focus on the gender-specific data (Tables 9-SI to 12-SI). The *DLM* analysis demonstrated a nearly monotonic decrease of the two contaminant mean annual levels in lake trout males and females, when

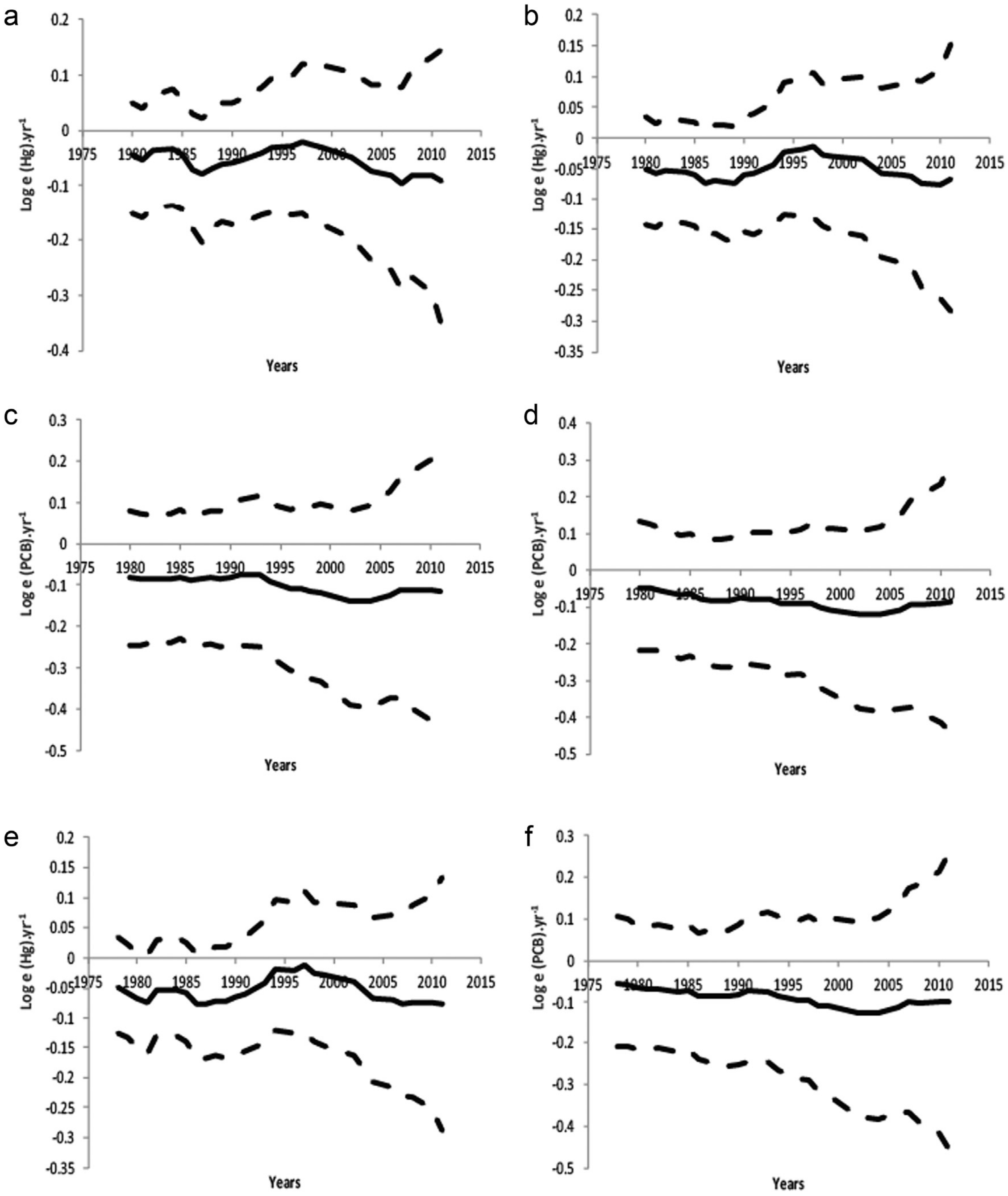

Fig. 3. Dynamic linear modeling analysis depicting the annual rates of changes for *THg* (μg/g ww) and *PCB* (ng/g ww) concentrations in lake trout. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted *THg* and *PCB* concentrations for females (a and c), males (b and d) and all data (e and f).

accounting for the covariance with fish length and lipid content (Fig. 2). In particular, when pooling all the samples together, the probability distributions that represent our knowledge of the rate parameters indicate average odds of 5.20:1 for *THg* and 2.76:1 for *PCBs* that the corresponding rates of changes were negative throughout the study period (Fig. 3). Notably, the values of the odds ratio did not differ between the two genders. [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.] Further, a careful inspection of the contaminant time series is indicative of a temporal regularity/oscillatory behavior within the overall downward trends which could be associated with the population dynamics of prey items (see following discussion). In a similar manner, both walleye males and females displayed a decreasing trend with respect to their *PCB* levels (Fig. 4). The corresponding values of the odds ratio were somewhat lower than those reported for lake trout, suggesting that the odds of a negative rate of change have been on average 1.63:1 during the study period (Fig. 5). On the other hand, the *THg* models do not suggest any distinct temporal shifts in the rates of change, which revolved around the zero level throughout the study period (Fig. 5). Consequently, the mean annual *THg* levels remained practically unaltered and were mainly characterized by (moderate) year-to-year variations (Fig. 4).

4. Discussion

In a recent evaluation of the long-term trends for the Province of Ontario, Canada, Gandhi et al. (2014) showed that the fish

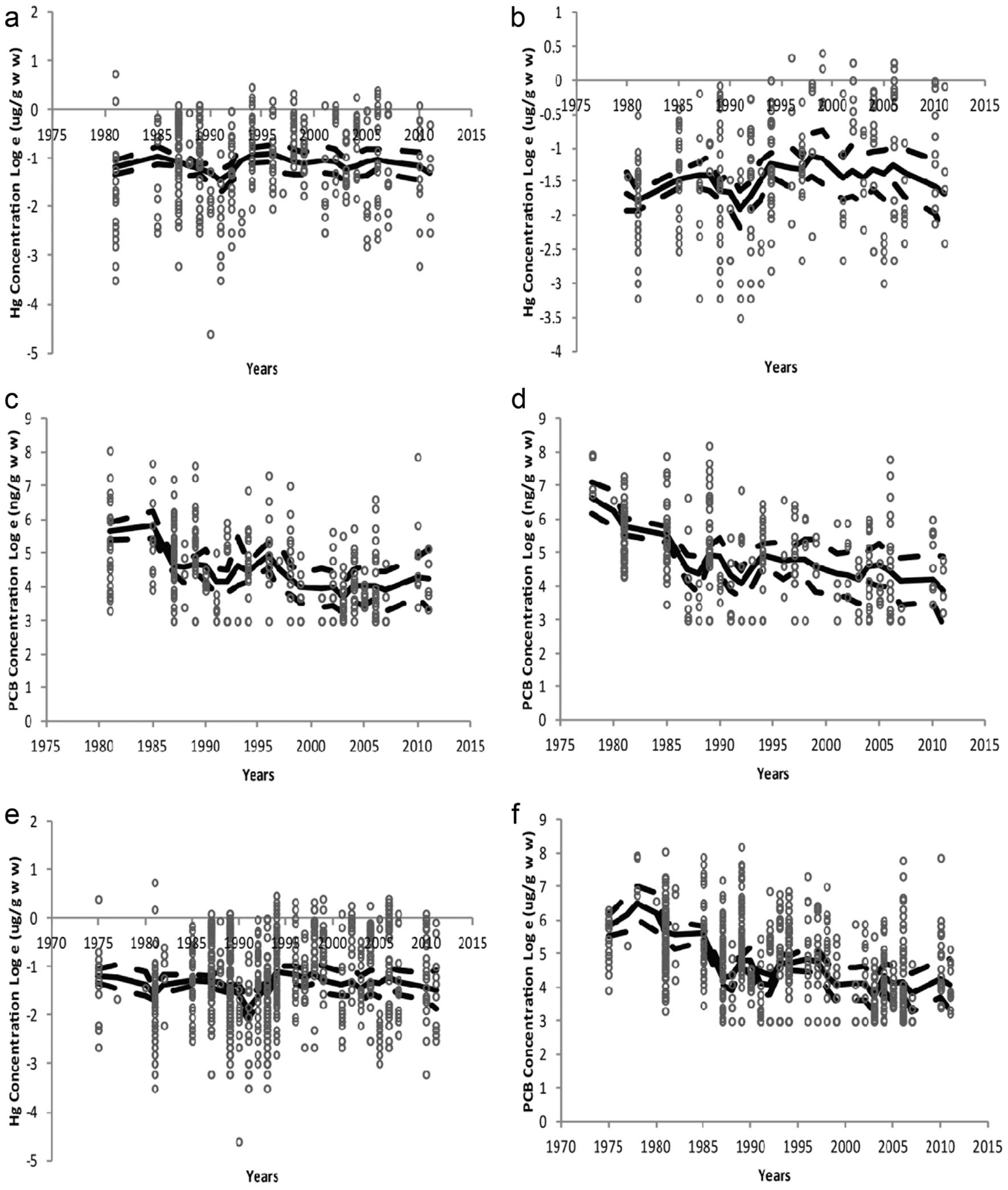

Fig. 4. Dynamic linear modeling analysis for walleye depicting the actual *THg* (μg/g ww) and *PCB* (ng/g ww) concentration (gray dots) against the predicted annual trends when accounting for the covariance with fish length and lipid content (black lines). The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted *THg* and *PCB* concentrations for females (a and c), males (b and d) and all data (e and f).

mercury levels have generally decreased between the 1970s and 2012. However, these decreasing trends mainly occurred during the 1970s/1980s, whereas fish mercury appears to be lately increasing at a significant number of locations, especially in northern Ontario. The same study also argued that despite the recent emission reductions in North America and gradually lowering atmospheric concentrations, other factors such as invasive species, local geochemistry, climate change, and regional variability in exogenous sources may shape mercury fate and transport in fish communities (Gandhi et al., 2014; Pacyna et al., 2010; Rennie et al., 2010; Seigneur et al., 2004). Likewise, the spatiotemporal patterns of total *PCB* levels represent the integration of many abiotic and biotic processes, some with large stochastic components, acting on a number of congeners with distinct chemical properties; thus, the *PCB* trends often appear to be system and fish species specific (Bhavsar et al., 2007). The delineation of contaminant trends may be confounded by many other factors, such as the seasonality, lack of explicit consideration of important covariates (e.g., fish size, lipid content, feeding habits, behavioral patterns, reproductive status and growth), type of statistical analysis performed, type of samples (skinless–boneless fillet versus whole fish portions) or even data pooling across the different locations of a lake (Gewurtz et al., 2011a,b). Dependence upon incomplete information and biased statistical analysis can conceivably result in a misinterpretation of the actual fish contaminant trends, and therefore our study employed a robust statistical framework to tease out any non-monotonic patterns of behavior, while explicitly accommodating the role of important covariates, such as fish length, lipid

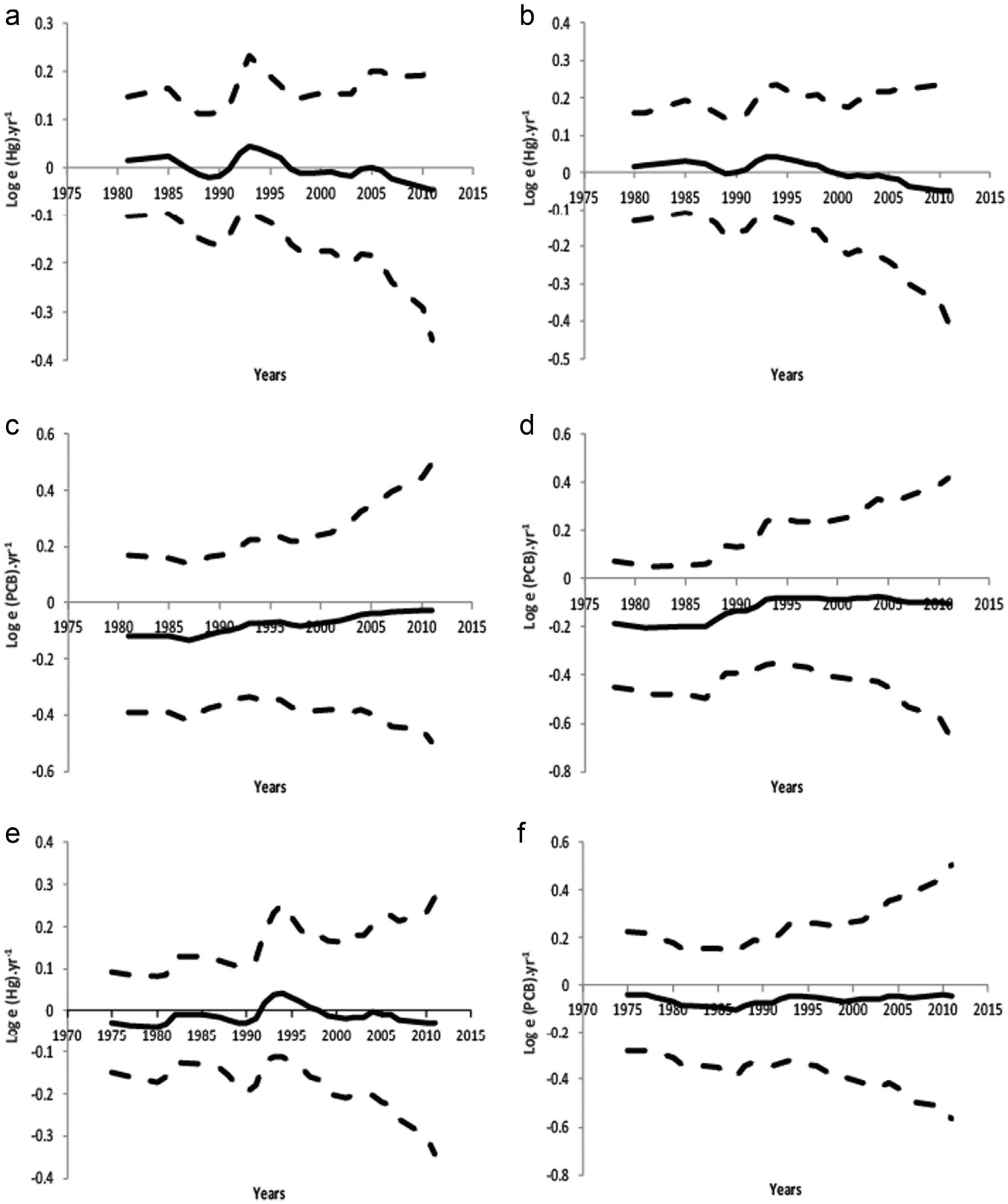

Fig. 5. Dynamic linear modeling analysis depicting the annual rates of changes for THg (μg/g ww) and *PCB* (ng/g ww) concentration in walleye. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted *THg* and *PCB* concentrations for females (a and c), males (b and d) and all data (e and f).

content, and gender.

4.1. Temporal trends and ecological mechanisms of fish contaminant variability in Lake Ontario

Historically, lake trout has the highest *PCB* concentrations in Lake Ontario relative to the rest of the Canadian waters of the Great Lakes, although the among-lake differences appear to be decreasing (Bhavsar et al., 2007). Our study demonstrated a nearly monotonic decrease of the *PCB* levels in lake trout males and females during the study period. Likewise, Bhavsar et al. (2007) used linear regression analysis to derive long (1978–2006) and short (1990–2006) term decline rates of −0.037 and −0.0428 $\log_{10}[PCB] \cdot yr^{-1}$ in 55–65 cm lake trout, respectively (see their Figs. 2 and 4). Our posterior median rates of change from the dynamic linear modeling analysis with the pooled dataset (after the conversion from natural to base 10 logarithms) were practically equal to those reported by the earlier work, rendering validation to their projections that lake trout achieved the Great Lakes Strategy 2002 objective of decrease in concentrations by 25% during 2000–2007. Interestingly, the recent acceleration of the decline rates reported by Bhavsar et al. (2007) extends into the 2007–2011 period, suggesting that lake trout will likely undergo further decrease in their *PCB* levels. Similarly, both walleye males and females were characterized by a declining trend of their *PCB* levels, although the corresponding rates of change were somewhat lower than those reported for lake trout. The mean *PCB* concentrations decreased until the late 1980s, then remained fairly constant (or even increased) through the early/mid-1990s, after when the *PCB* levels decreased with slower rates until 2011. Notably, employing linear regression with suggests that the *PCBs* in 45–55 cm walleye have levelled off since the early 1990s (Bhavsar et al., 2007); a result that differs somewhat from the projections of our exponential decay model, which are still indicative of negative (but slower) rates. Comparing with Lake Erie, where the *PCB* concentrations in walleye are the highest recorded in the Great Lakes, the temporal trends appear to differ in that the earlier declines of Lake Ontario were not manifested, and thus the rates of change remained almost consistently around zero during the time span examined (Sadraddini et al., 2011a,b).

Lake trout females displayed higher *PCB* levels relative to their male counterparts. The latter finding appears to deviate from the results reported by Madenjian et al. (2010), which found that, on average, males were 22% higher in *PCB* concentrations than females in samples collected from Lake Ontario in August and September of 1986. [It is worth noting though that relative to the skinless–boneless fillets used in our study, Madenjian et al. (2010) focused on whole fish that was homogenized in a commercial blender prior to laboratory testing.]. Interestingly, the corresponding *PCB* concentrations for lake trout males and females in our dataset were 2995 and 2690 ng g^{-1} ww during the 1986–87 period, as oppose to their decadal averages of 3219 and 3493 ng g^{-1} ww, respectively. This (probably) coincidental agreement with the Madenjian et al. (2010) study stems from a short term dip in the mid-1980s that was distinctly sharper for female lake trout. Acknowledging the likelihood of a sampling artifact, Madenjian et al. (2010) attempted to offer mechanistic insights into the sexual differences recorded in Lake Ontario, reporting *PCB* levels in gonads and somatic tissues non-supportive of the hypothesis that the shedding of the gametes is responsible for the higher *PCB* concentration in male lake trout. Thus, it was concluded that the gross growth efficiency (the amount of fish growth divided by the amount of food consumption needed to produce that growth) may be the most plausible explanation for the gender-specific *PCB* concentrations of lake trout (Madenjian et al., 2010). Male fish often have higher contaminant levels than females due to their lower gross growth efficiency, differences in habitat utilization, and losses of contaminants by females during spawning (Madenjian et al., 2009; Rypel et al., 2007). Other studies have found that females can bioaccumulate higher amounts due to their greater food consumption in an effort to build metabolic reserves necessary for spawning (Rypel et al., 2007). There are also reports of no significant differences between genders (Burger and Gochfeld, 2007; Pandelova et al., 2008; Rypel et al., 2007), which is on par with the *THg* levels found in our lake trout data. Generally, according to our analysis, the sexual differences tend to be both species- and contaminant-specific, and may be manifested as differences in the actual contaminant levels, the rates of change over time, or even their year-to year variability.

Empirical and modeling evidence suggests that the Niagara River, and to a lesser extent atmospheric deposition, are the predominant sources of contaminants in Lake Ontario (Ethier et al., 2012; Marvin et al., 2004; Thompson et al., 1993). The Niagara River represents between 75% and 85% of the total amount of water that enters Lake Ontario, contributing two-thirds of the total contaminant loadings on an annual basis. Importantly, Marvin et al. (2003) reported frequent *THg* exceedances of the Canadian Sediment Quality Guidelines Probable Effect Level (*PEL*) guidelines in many areas of Lake Ontario, despite the substantial improvement in sediment quality over the past four decades. The observed spatial patterns in sediment contamination were the highest in the three major depositional basins (Niagara, Mississauga, Rochester) of Lake Ontario, while the inshore areas were characterized by relatively lower contamination levels (Marvin et al., 2003). These trends likely reflect the impact of industrial activities in the adjacent watersheds (including the Niagara River), whereby the contaminated sediments transported by the major tributaries deposit in the offshore areas of Lake Ontario (see Fig. 3 in Marvin et al., 2003). Levels of *THg* in atmospheric deposition are reported to be as high as 2.60 ng m^{-3} (Atkinson et al., 2007), and thus can conceivably contribute to the year-to-year variability in Lake Ontario (Ethier et al., 2012). In this context, we highlight the nearly monotonic decrease of the mercury mean annual concentrations in lake trout males and females throughout the study period. The current *THg* levels in lake trout are lower than the values measured in Lakes Superior and Huron, but still distinctly higher than those in Lake Erie (see Fig. 1 in Bhavsar et al., 2010). While the latter pattern suggests that there is still considerable space for improvement, we note that Lake Erie supports a relatively low lake trout population mainly located at its eastern basin. Compared with the western basin, eastern Lake Erie does not receive large inputs of chemical contaminants and its greater depth also reduces the sediment–water column interactions, which is reflected in the fairly low lake trout *THg* concentrations, e.g., 0.12 ng g^{-1} ww (Saddradini et al., 2011b; Carter and Hites, 1992).

Consistent with recent work in the Great Lakes (Azim et al., 2011a; Saddradini et al., 2011b; Bhavsar et al., 2010), we found that the mean annual *THg* levels in walleye remained practically unaltered and were mainly characterized by a “wax and wane” pattern. Similar oscillations have been reported in other Great Lakes time series for several species which have been associated with the food web dynamics and/or the climatic factors (e.g., French et al., 2006; Scheider et al., 1998; Borgmann and Whittle, 1991). Given that the *THg* burdens in fish are mainly driven by their diet, these annual variations could stem from the population dynamics of prey species. Along the same line of reasoning, the recent stabilized *THg* trends in walleye may be driven by the shifts in feeding dynamics after the establishment of non-indigenous species. The resulting food web alterations likely induced diet shifts that decreased the growth rates and subsequent growth dilution (Paterson et al., 2009). There is also evidence of dietary shifts of the top predators from less contaminated pelagic to more contaminated benthic food sources (Hogan et al., 2007). Specifically, dreissenid mussels are fairly reliable sentinels of the bioavailability of contaminants that can facilitate their transfer through the food web (Kwon et al., 2006). The introduction of dreissenids and (concomitantly) round goby (*Neogobius melanostomus*) has resulted in a food chain lengthening and consequently a higher fish contamination through the process of biomagnification (Campbell et al., 2003). Round goby are known to have colonized extensive areas of the western and eastern portions in Lake Ontario and gradually represent a prominent item of the diet of top predators of the system (Dietrich et al., 2006). Although round goby have the capacity to switch to more profitable prey food items when they become available, their diet is dominated by dreissenids, especially in shallow areas, and thus tend to accumulate contaminants which are then transferred to benthic-oriented fish (Walsh et al., 2007). Finally, the detected *THg* temporal trends could also be modulated by seasonal variations in fish due to a number of environmental or physiological factors, such as temperature variability, seasonal dietary shifts, and within-lake methylation rates (Fowlie et al., 2008; Murphy et al., 2007). In particular, the seasonal variation in mercury concentrations and food web structure was assessed for eastern Lake Ontario and tended to be highest in the spring and lowest in the summer (Zhang et al., 2012).

4.2. *Optimizing the trend analysis of fish contaminants*

Among the models historically used to delineate the temporal contaminant trends, the single exponential decay (or first-order) model was an effective strategy to represent the initial declines in the 1980s, following the emission cuts stipulated by the Great Lakes Water Quality Agreement (IJC, 1978, 2006). Nonetheless, the more recent year-to-year variability evidently does not follow the first-order approximation, as the rates of decline in many systems appear to have slowed more than expected (Bhavsar et al., 2007, 2010). To address this problem, Stow et al. (1995, 1999) showed that the first-order model with a non-zero asymptote, the double exponential model (the sum of two first-order processes), and the mixed-order model offer better alternatives. In particular, the mixed-order model can capture a much wider range of dynamics, such as the rapid initial decreasing trends, followed by slower secondary declines, but without the unrealistic long-run constraint of an asymptotic boundary (Stow et al., 1999). Nonetheless, a fundamental weakness of these models is the postulation of a monotonic trajectory of the contaminant levels and their inability to capture systematic deviations from this pattern. Like in Lake Erie, the presence of non-monotonic trends in the fish contaminant time series of Lake Ontario reinforces the notion that the existing rigid model structures should be revisited (Azim et al., 2011a; Sadraddini et al., 2011a). The introduction of the temporally variant random error term δ in the statistical formulation has enabled the detection of non-monotonic trends in our data set, and therefore can ameliorate the structural inadequacies of models that have been traditionally adopted in the context of fish contaminant trend analysis.

Interestingly, consistent with the findings of Azim et al.'s (2011a) sensitivity analysis (see Table 1 in their Electronic Supplementary material), the augmented exponential decay model displays sensitivity to the assumptions made regarding the THg_0/PCB_0 priors, manifested as a positive covariance between the posterior estimates of the exponential decay term and the initial contaminant concentration. Our study revealed a similar overfitting problem with several gender-specific models, in which the predicted higher initial contaminant values were accompanied by higher decay coefficients relative to their simpler counterparts. Because the higher decreasing rates postulated by this parameter specification were not supported by the recent contaminant concentrations, the resulting discrepancy was offset by the positive values of the random walk terms during the second half of the survey period (e.g., Fig. SI-2a and b). Thus, to avoid getting “good results for the wrong reasons”, the specification of the prior probability distributions for the initial contaminant concentrations should be selected with caution among the different specifications proposed in the Bayesian literature (Azim et al., 2011a; Sadraddini et al., 2011a); especially when working with subsets (e.g., gender-specific) that may contain fewer observations during the initial years.

To discern whether the recent increases in the *PCB* levels of walleye in Lake Erie represent a statistically significant pattern, Sadraddini et al. (2011a) used dynamic linear modeling analysis to show that this trend disappeared when using length-corrected predictions, and was thus a reflection of the biases introduced by the local sampling procedures. This finding reinforced the necessity of accounting for potentially important causal factors when conducting trend analysis, and also highlighted the efficiency of *DLM*s as robust hindcasting tools. Generally, studies implicitly assume that the contaminant–length relationship reflects the older age of larger fish, which have longer exposure to contaminants (Trudel and Rasmussen, 2001). However, this relationship can be modulated by a wide array of factors (e.g., growth rate, behavioral patterns, diet) affecting contaminant bioaccumulation (Paterson et al., 2006; Trudel and Rasmussen, 2001). In particular, the relationship between concentration and fish length is fairly well established for mercury (Sadraddini et al. 2011b; Sonesten, 2003), but less so for total *PCB*s (Szlinder-Richert et al., 2009; Bhavsar et al., 2010, 2007; Weis and Ashley, 2007). In this study, our *DLM* analysis provided evidence that the fish length has a universally strong signature to both *THg* and *PCB* interannual variability in Lake Ontario (Gewurtz et al., 2011a,b). [Counter to the sampling practices in Lake Erie though, we have not found a collection bias in favor of larger fish during the recent years of our study period.]

Fish lipid content stands out as one of the possible covariates that have received considerable attention in the literature (Amrhein et al., 1999; Ewald and Larsson, 1994). Generally, there are contradictory results regarding the strength of the causal linkage between fish lipid content and contaminant levels (Larsson et al., 1996; Amrhein et al., 1999). For example, Stow et al. (1997) identified weak *PCB*:lipid relationships for five species of Lake Michigan salmonids, but also reported a stronger association when focusing on samples collected during the spawning period (see their Fig. 5). Amhrein et al. (1999) found that lipid normalization does not efficiently control within-species variability, but can be useful for shedding light on the differences among species. In the Lake Erie dataset, the strength of the *PCB*:lipid relationship among individuals was moderately weak, but the most parsimonious dynamic linear models considered only the fish length as covariate (Sadraddini et al. 2011b). In Lake Ontario, we found that the lipid content had a fairly strong signature to *PCB* variability in both walleye and lake trout. Bioaccumulation of mercury though is driven by different processes and tends to be more closely related to the protein matrix (Amlund et al., 2007). Thus, the lipid levels had a weaker relationship with the *THg* concentrations in walleye and were also characterized by a (presumably) puzzling negative signal in lake trout. Previous researchers have also indicated a minimal contaminant:lipid lake trout relationship, while Borgmann and Whittle (1991) concluded that although lipid might have some effect on contaminant excretion, lipid concentrations were not the major factor controlling contaminants in Lake Ontario lake trout. The counterintuitive signs of the posterior slope coefficients could have occurred from the confounding (collinear) effects of lipid with fish length ($r \approx 0.30$ in our dataset). However, a weak negative relationship ($r \approx -0.10$) also appears in the residual plots when we partial out the covariance between *THg* and the fish length, indicating that our *DLM* estimates were not a statistical artifact (Fig. SI-9). Thus, our results render support to the hypothesis that within an individual, contaminants may accumulate in lipids, but lipid concentrations are likely unimportant in the mechanisms governing contaminant assimilation (Stow et al., 1997).

In conclusion, we examined the *THg* and *PCB* levels and temporal trends in walleye and lake trout based on almost 40 years of contaminant data from Lake Ontario. Our study primarily highlighted the ability of dynamic linear modeling to offer a robust hindcasting tool with a flexible structure that effectively accommodates the role of potentially important causal factors. By contrast, the structural inadequacies of the exponential and mixed-order models can be rectified with the inclusion of the conditional autoregressive term, although the identifibiality issue arising from the increased complexity may provide misleading results. To address the latter problem, our analysis suggests that the characterization of the prior distributions assigned to the initial contaminant concentrations should be handled with caution. Regarding the contamination trends in Lake Ontario, walleye was characterized by distinctly higher *THg* levels relative to lake trout, and the opposite was true for the *PCB* concentrations. Female walleye also demonstrated higher *THg* concentrations relative to the male counterparts, which in turn had higher *PCB* levels. Both mercury and *PCB*s demonstrate decreasing temporal trends in lake trout males and females. Decreasing *PCB* trends were also evident in walleye, but the mean annual mercury levels are characterized by a “wax and wane” pattern, suggesting that specific fish species may not act as bio-indicators for all contaminants. The latter finding may be attributed to the shifts in energy trophodynamics along with the food web alterations induced from the introduction of non-native species, the intricate nature of the prey–predator interactions, the periodicities of climate factors, and the year-to-year variability of the potentially significant fluxes from atmosphere or sediments. The identifiability patterns of the *DLM* coefficients depict the strong signature of the fish length to both *THg* and *PCB* variability. By contrast, the lipid content was related only to the *PCB* fish levels, whereas the same causal linkage was weak in walleye and even had a negative relationship with lake trout data. Finally, a meaningful risk assessment exercise will be to elucidate the role of within-lake fish contaminant variability and evaluate the potential bias introduced when drawing inference from pooled datasets.

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Appendix A. Suplementary Information

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.ecoenv.2015.03.022.

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A Bayesian assessment of the mercury and PCB temporal trends in lake trout (*Salvelinus namaycush*) and walleye (*Sander vitreus*) from Lake Ontario, Ontario, Canada

[SUPPORTING INFORMATION]

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FIGURES LEGENDS

Figure 1-SI: Predictions of the exponential decay model, augmented with first order random walk terms, against the actual *THg* (*μg/g ww*) and *PCB* (*ng/g ww*) concentrations (gray dots) in lake trout of 55-65 cm length. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted *THg* and *PCB* concentrations for females (*a, c*), males (*b, d*) and all data (*e, f*).

Figure 2-SI: Temporal variability of the first order random walk terms introduced to relax the monotonic patterns postulated by the exponential decay model. The solid and dashed lines correspond to the median and the 95% credible intervals of the error terms for the *THg* and *PCB* concentrations for lake trout females (*a, c*), males (*b, d*) and all data (*e, f*).

Figure 3-SI: Predictions of the exponential decay model, augmented with first order random walk terms, against the actual *THg* (*μg/g ww*) and *PCB* (*ng/g ww*) concentrations (gray dots) in walleye of 45-55 cm length. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted *THg* and *PCB* concentrations for females (*a, c*), males (*b, d*) and all data (*e, f*).

Figure 4-SI: Temporal variability of the first order random walk terms introduced to relax the monotonic patterns postulated by the exponential decay model. The solid and dashed lines correspond to the median and the 95% credible intervals of the error terms for the *THg* and *PCB* concentrations for walleye females (*a, c*), males (*b, d*) and all data (*e, f*).

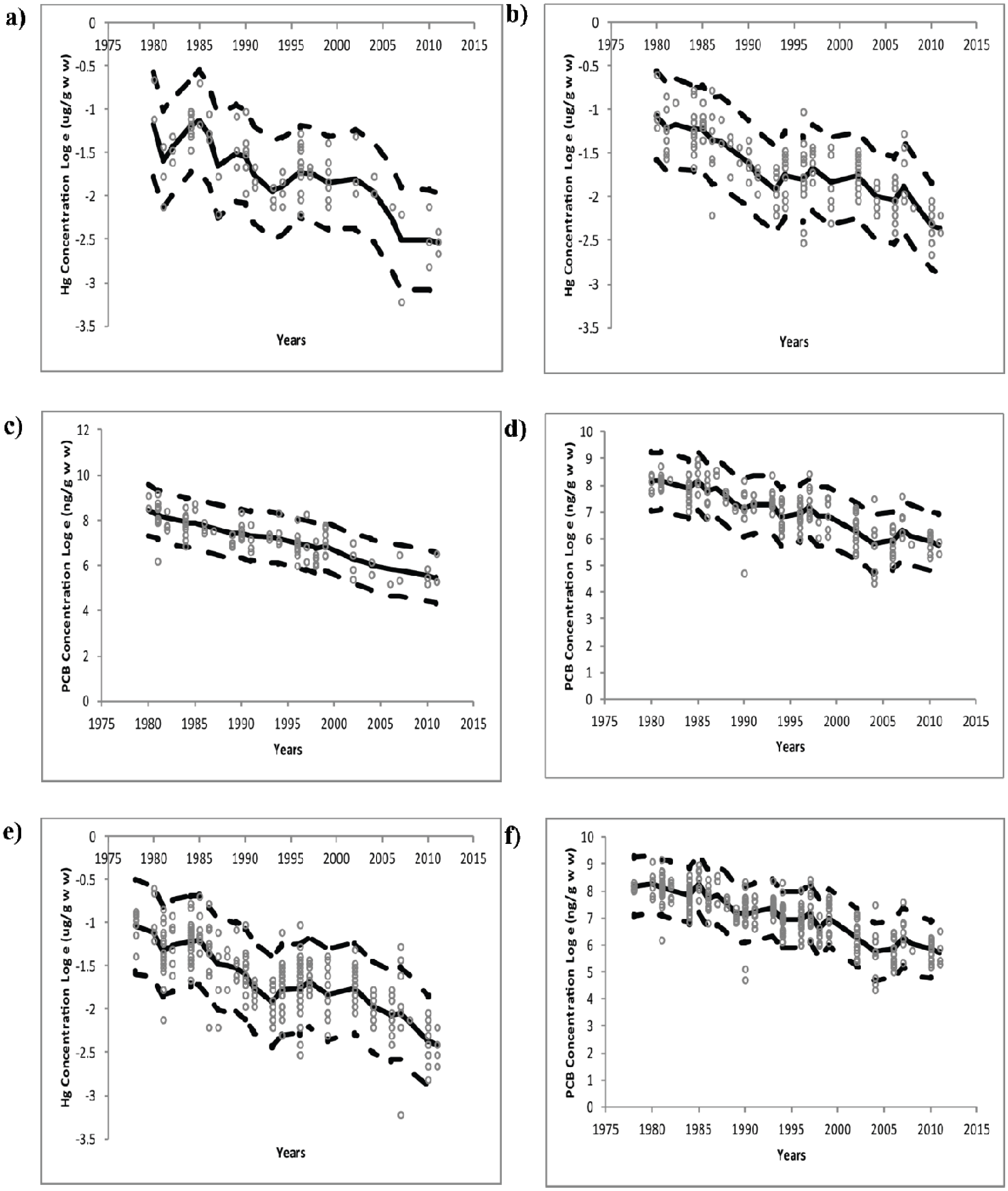
Figure 5-SI: Predictions of the exponential decay model against the actual *THg* (*μg/g ww*) and *PCB* (*ng/g ww*) concentrations (gray dots) in lake trout of 55-65 cm length. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted *THg* and *PCB* concentrations for females (*a, c*), males (*b, d*) and all data (*e, f*).

Figure 6-SI: Predictions of the exponential decay model against the actual *THg* (*μg/g ww*) and *PCB* (*ng/g ww*) concentrations (gray dots) in walleye of 45-55 cm length. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted *THg* and *PCB* concentrations for females (*a, c*), males (*b, d*) and all data (*e, f*).

Figure 7-SI: Predictions of the mixed order model against the actual *THg* (*μg/g ww*) and *PCB* (*ng/g ww*) concentrations (gray dots) in lake trout of 55-65 cm length. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted *THg* and *PCB* concentrations for females (*a, c*), males (*b, d*) and all data (*e, f*).

Figure 8-SI: Predictions of the mixed order model against the actual *THg* (*μg/g ww*) and *PCB* (*ng/g ww*) concentrations (gray dots) in walleye of 45-55 cm length. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted *THg* and *PCB* concentrations for females (*a, c*), males (*b, d*) and all data (*e, f*).

Figure 9-SI: Partial residuals of *THg* concentrations (*μg/g ww*), after accounting for their covariance with fish length, against the lipid content of (*a*) female and (*b*) male lake trout.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Years
b)
Hg Concentration Log e (ug/g w w)
Years
c)
PCB Concentration Log e (ng/g w w)
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10
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Years
d)
PCB Concentration Log e (ng/g w w)
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e)
Hg Concentration Log e (ug/g w w)
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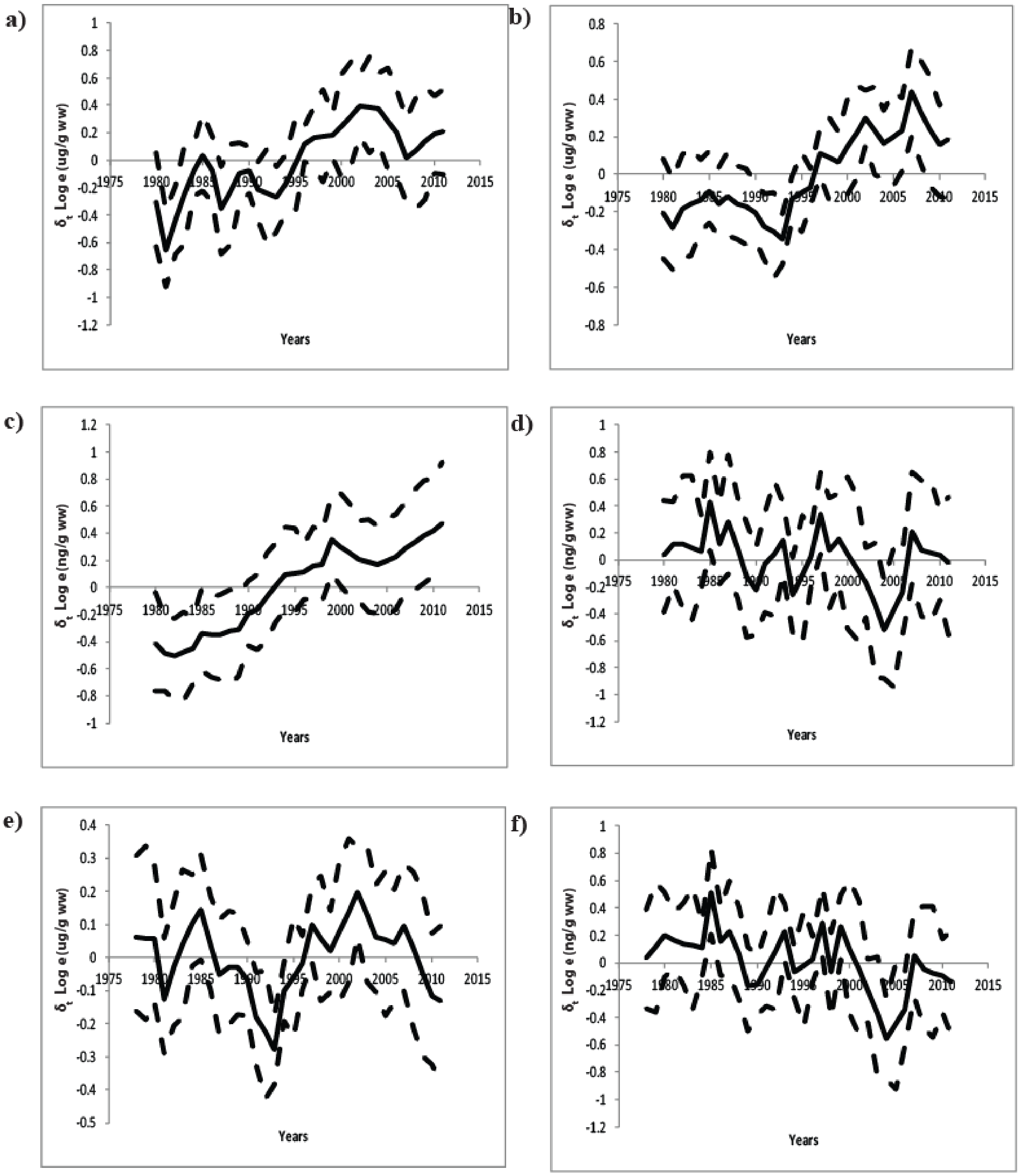
f)
PCB Concentration Log e (ng/g w w)
Years

Figure 1-SI



a)
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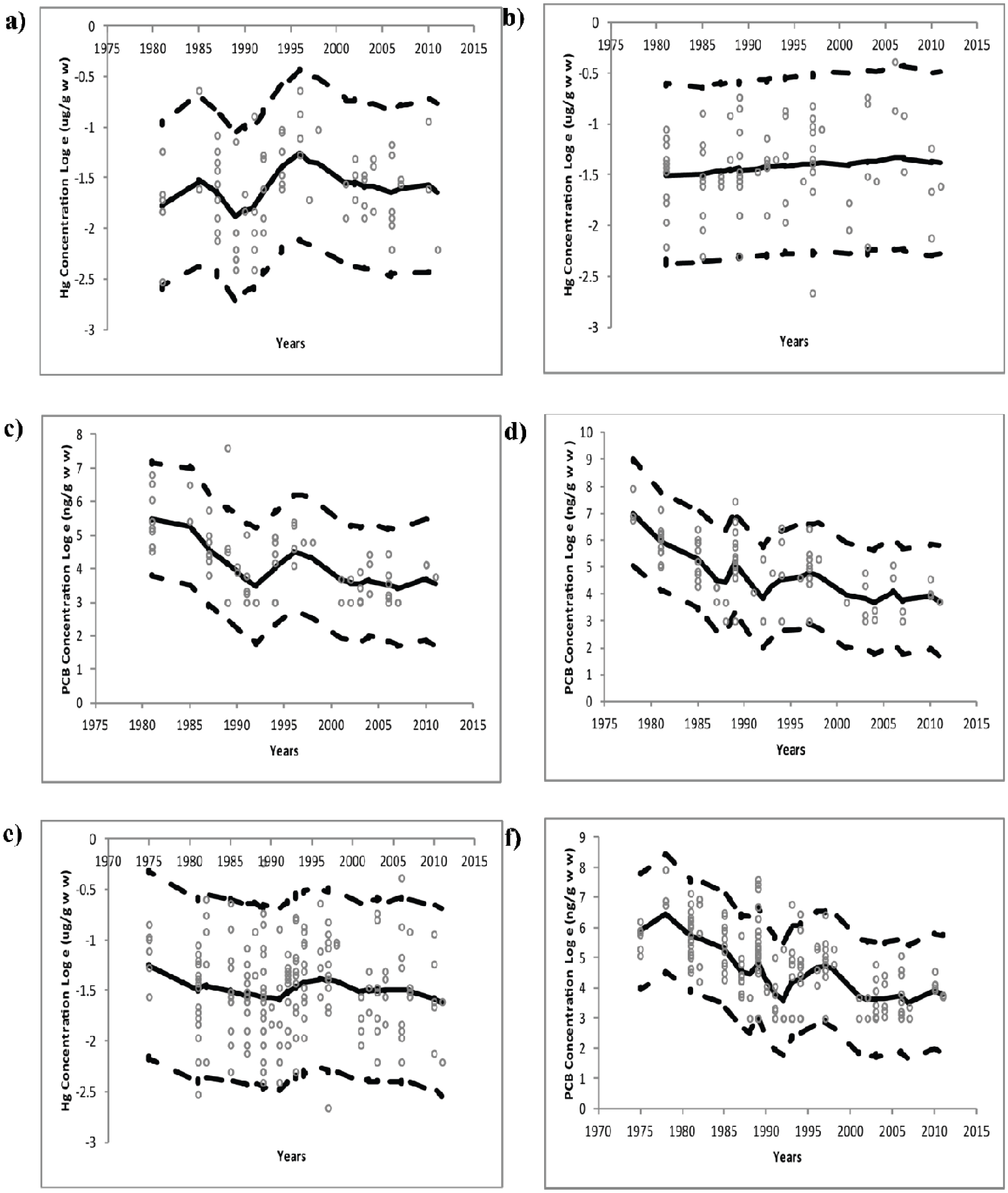
Figure 2-SI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PCB Concentration Log e (ng/g w w)
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Figure 3-SI

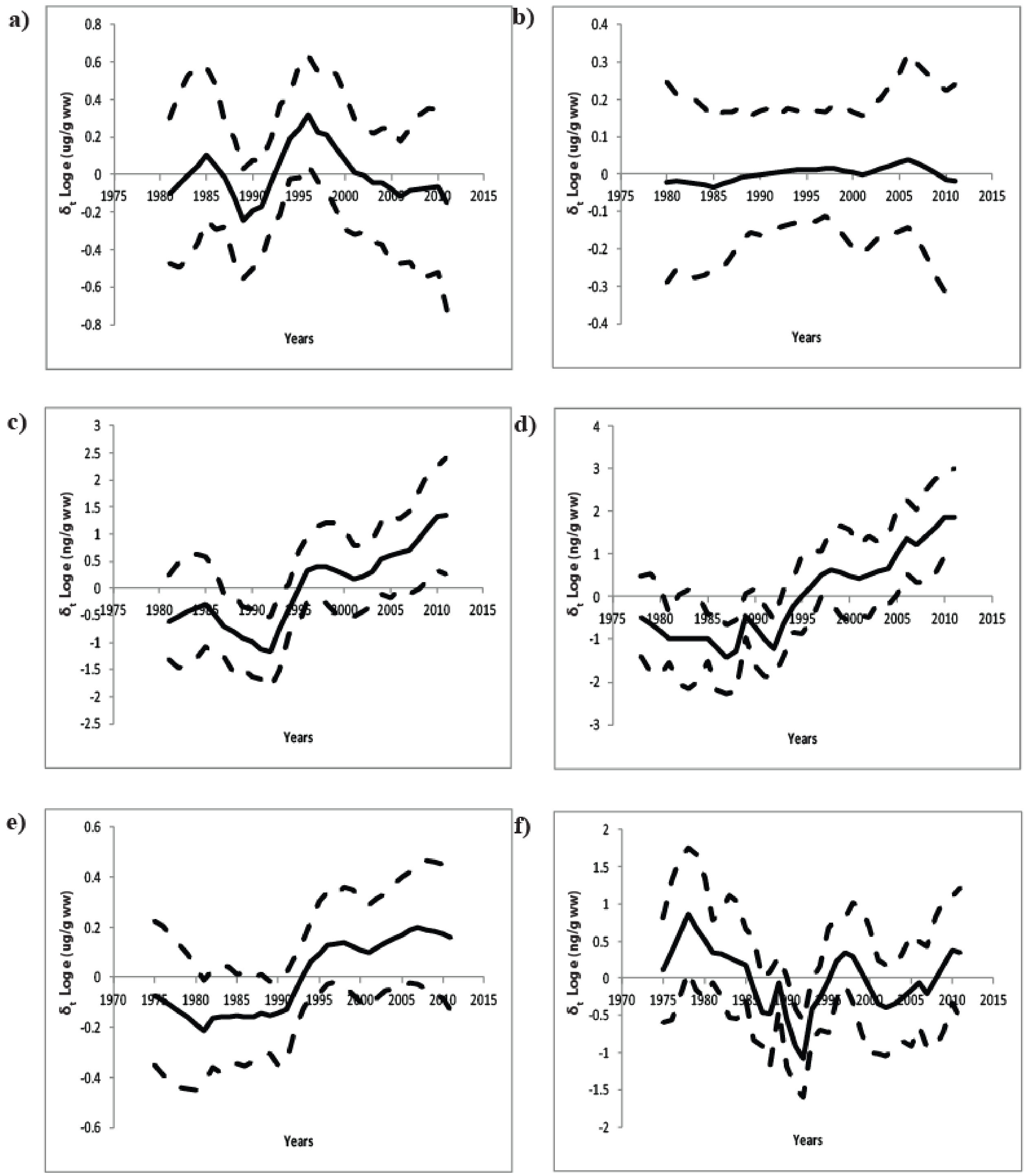
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Figure 4-SI

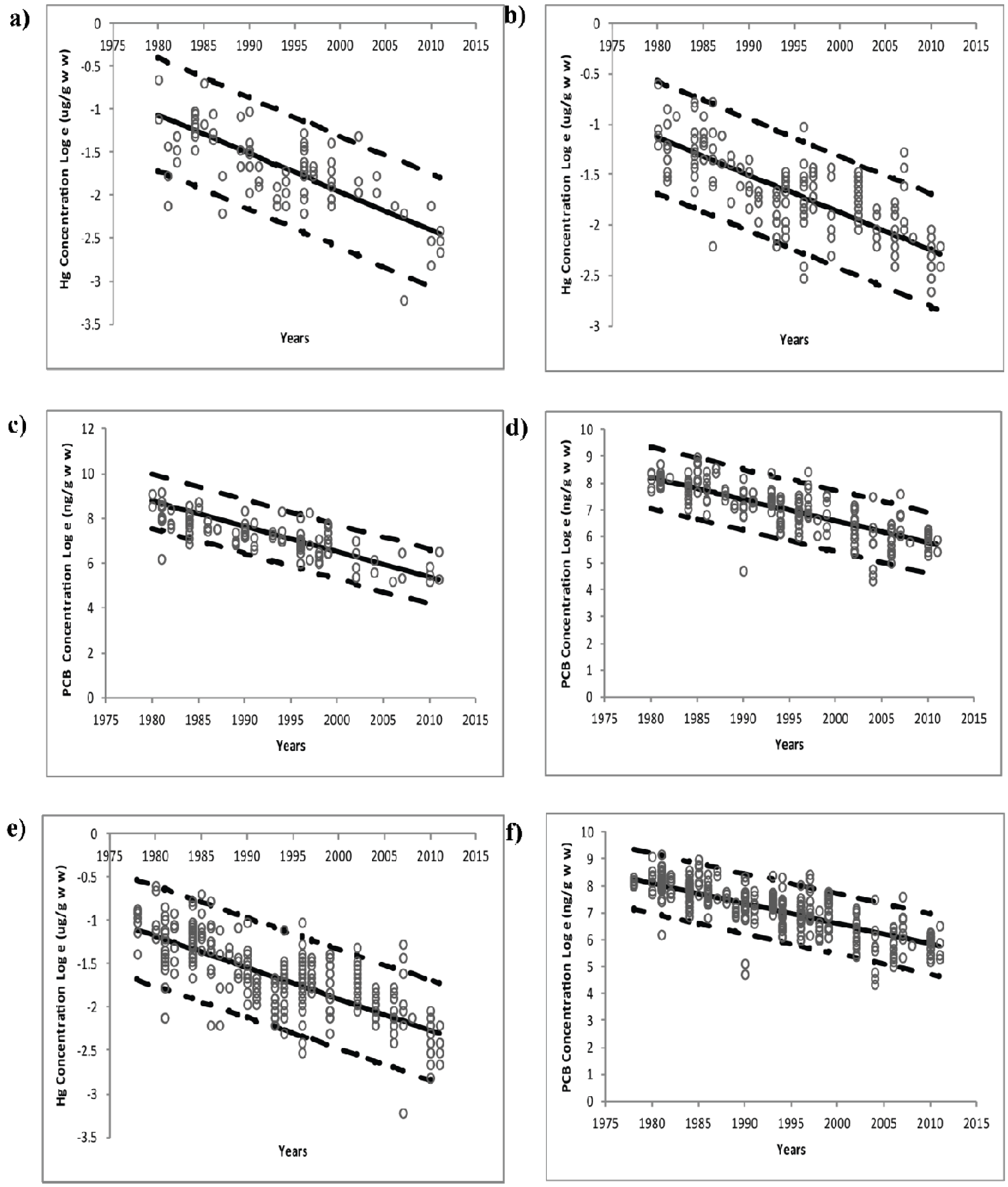
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Figure 5-SI

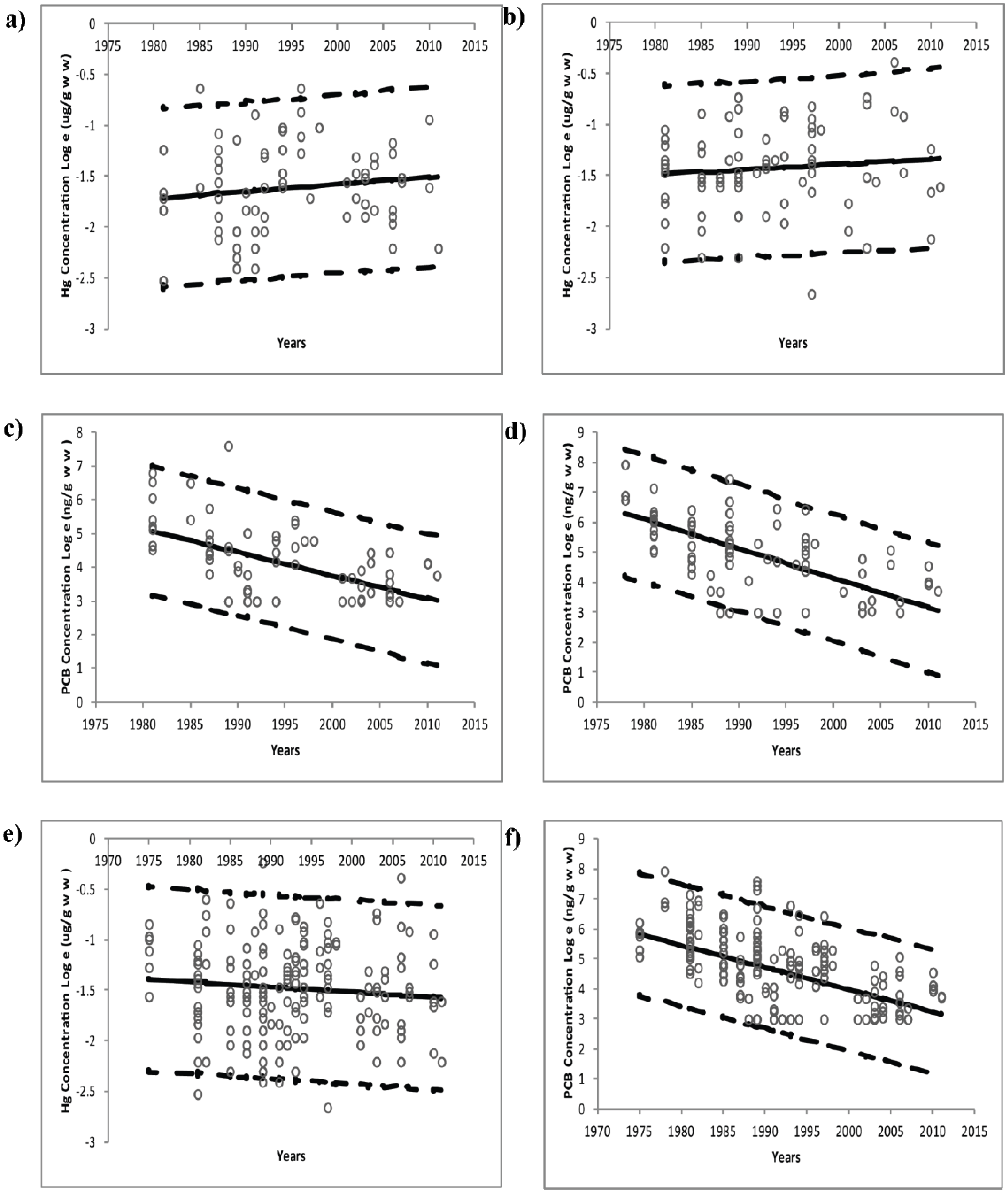
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Figure 6-SI

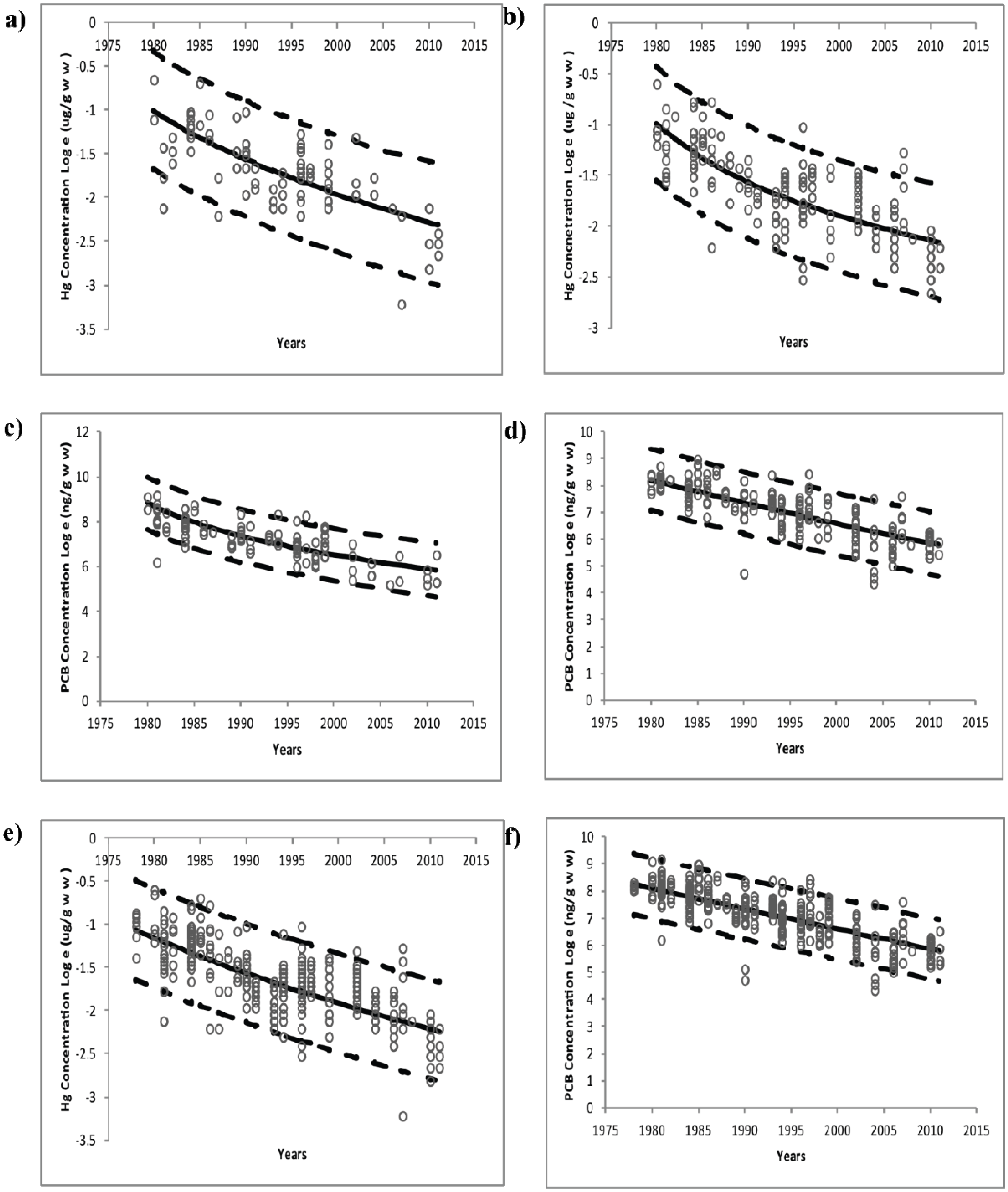
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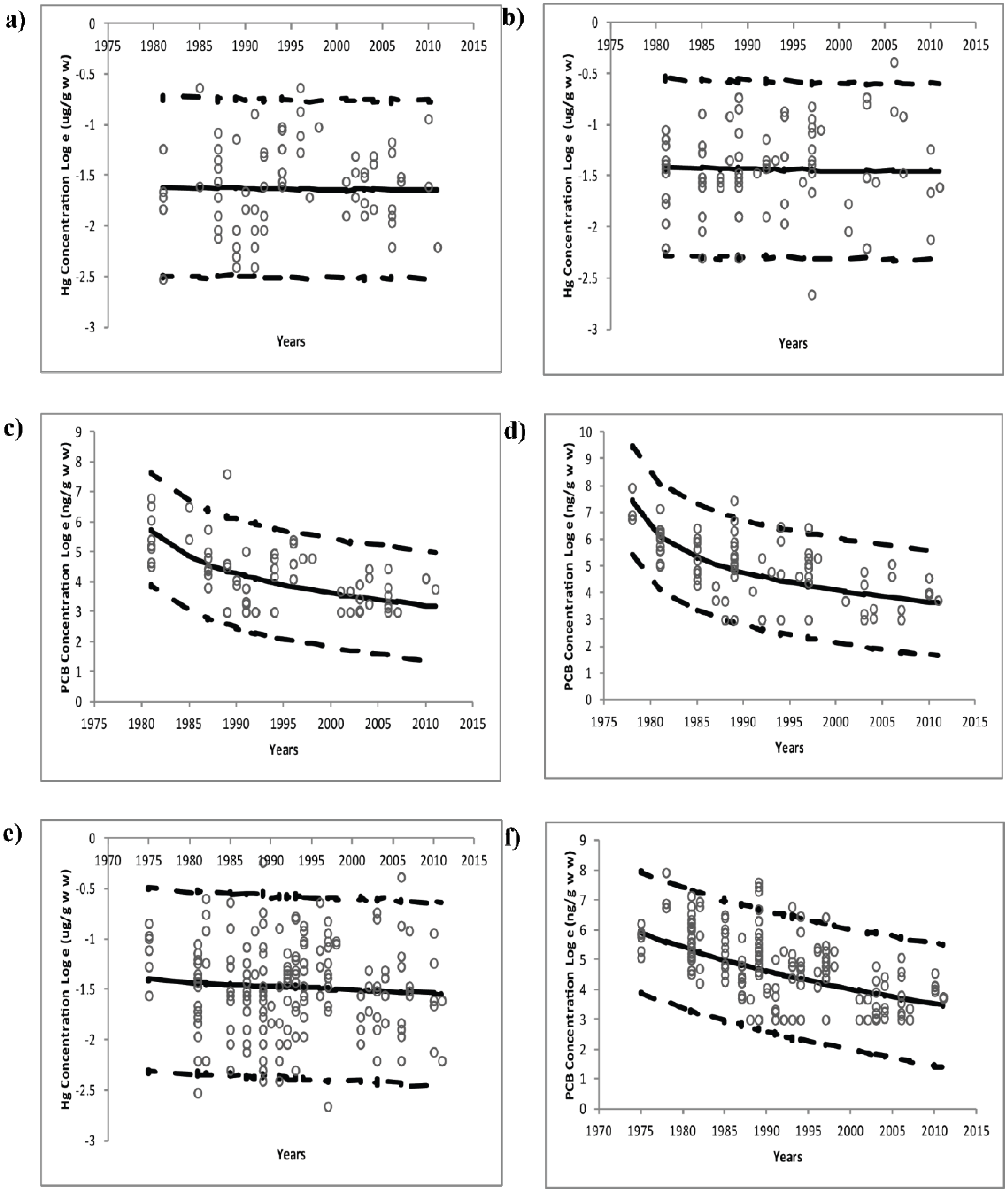
Figure 7-SI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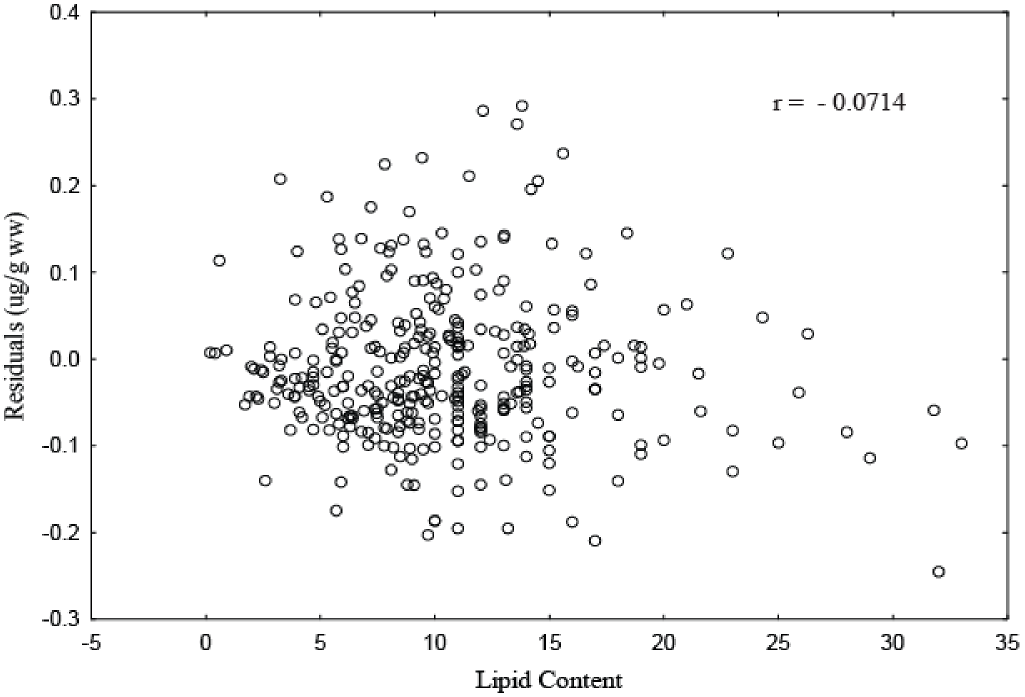
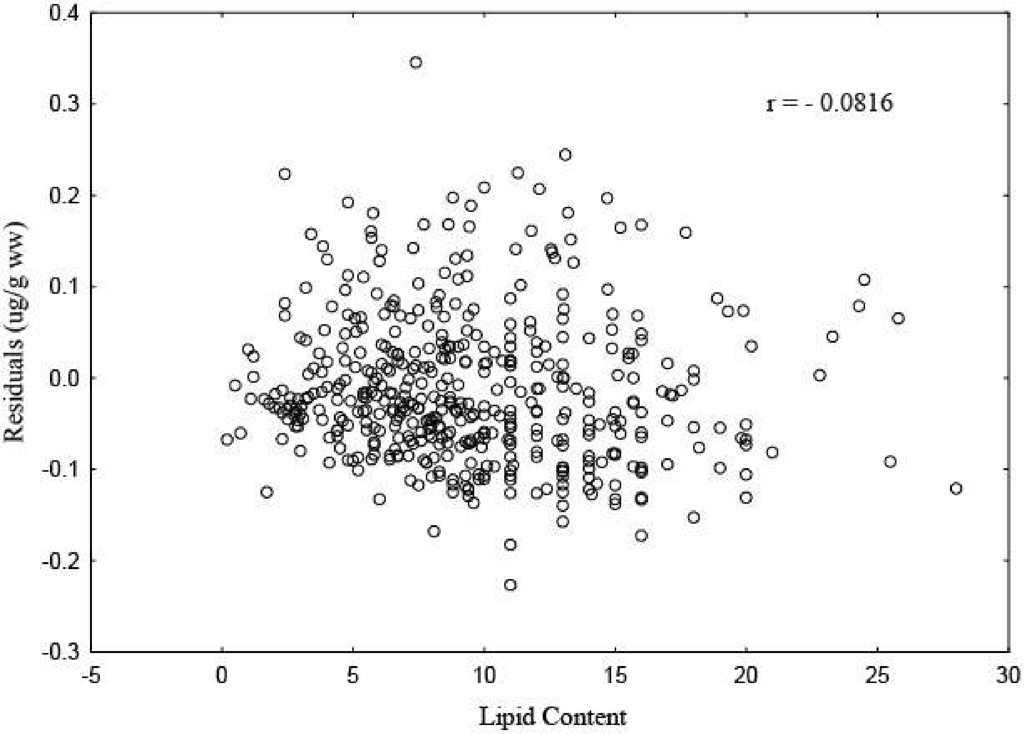
1975 1980 1985 1990 1995 2000 2005 2010 2015
Years
b)
Hg Concentration Log e (ug/g w w)
0
-0.5
-1
-1.5
-2
-2.5
-3
1975 1980 1985 1990 1995 2000 2005 2010 2015
Years
c)
PCB Concentration Log e (ng/g w w)
9
8
7
6
5
4
3
2
1
0
1975 1980 1985 1990 1995 2000 2005 2010 2015
Years
d)
PCB Concentration Log e (ng/g w w)
10
9
8
7
6
5
4
3
2
1
0
1975 1980 1985 1990 1995 2000 2005 2010 2015
Years
e)
Hg Concentration Log e (ug/g w w)
0
-0.5
-1
-1.5
-2
-2.5
-3
1970 1975 1980 1985 1990 1995 2000 2005 2010 2015
Years
f)
PCB Concentration Log e (ng/g w w)
9
8
7
6
5
4
3
2
1
0
1970 1975 1980 1985 1990 1995 2000 2005 2010 2015
Years

Figure 8-SI



r = - 0.0714
Residuals (ug/g ww)
0.4
0.3
0.2
0.1
0.0
-0.1
-0.2
-0.3
-5
0
5
10
15
20
25
30
35
Lipid Content



r = - 0.0816
Residuals (ug/g ww)
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0.3
0.2
0.1
0.0
-0.1
-0.2
-0.3
-5
0
5
10
15
20
25
30
Lipid Content

Figure 9-SI

Table 1-SI: Summary statistics of lipid content values (%) in relation to *THg* concentrations in lake trout and walleye skinless-boneless fillet data in Lake Ontario.

Species	Gender	N	Mean	stdev	Median	2.50%	97.50%	Int Quart	Kurt	Skew
Lake Trout	**Female**	339	10.38	5.31	9.70	2.25	23.72	6.15	2.61	1.18
	Male	459	9.48	4.75	8.80	2.11	20.00	6.56	0.63	0.71
	Pooled Samples	1023	9.24	4.95	8.64	1.66	20.56	6.22	1.86	0.99
Walleye	**Female**	351	1.60	3.50	1.00	0.29	5.20	0.74	56.24	7.32
	Male	289	1.78	3.14	1.07	0.30	10.38	0.90	31.59	5.42
	Pooled Samples	745	1.57	3.12	1.00	0.20	6.20	0.90	55.34	7.10

Table 2-SI: Summary statistics of lipid content values (%) in relation to *PCB* concentrations in lake trout and walleye skinless-boneless fillet data in Lake Ontario.

Species	Gender	N	Mean	stdev	Median	2.50%	97.50%	Int Quart	Kurt	Skew
Lake Trout	**Female**	361	10.35	5.18	9.70	2.30	23.00	5.90	2.86	1.21
	Male	476	9.53	4.73	8.80	2.18	20.00	6.45	0.61	0.71
	Pooled Samples	1030	9.32	4.92	8.70	1.70	20.42	6.10	1.88	0.99
Walleye	**Female**	351	1.61	3.50	1.00	0.29	5.20	0.76	56.17	7.32
	Male	295	1.77	3.12	1.06	0.30	9.91	0.90	31.99	5.44
	Pooled Samples	754	1.57	3.10	1.00	0.20	6.20	0.90	55.82	7.12

Table 3-SI: Summary statistics of length values (*cm*) in relation to *THg* concentrations in lake trout and walleye skinless-boneless fillet data in Lake Ontario.

Species	Gender	N	Mean	stdev	Median	2.50%	97.50%	Int Quart	Kurt	Skew
Lake Trout	**Female**	339	63.62	10.98	65.60	34.79	78.83	11.35	1.64	-1.13
	Male	459	61.88	10.10	62.50	37.61	79.76	11.75	0.93	-0.64
	Pooled Samples	1023	59.91	12.11	61.90	30.83	79.05	16.30	0.32	-0.68
Walleye	**Female**	351	56.38	13.16	58.10	28.58	77.28	-114.10	-0.49	-0.46
	Male	289	48.27	11.01	47.20	27.68	67.76	16.20	-0.77	0.01
	Pooled Samples	745	51.10	13.71	51.70	24.38	73.94	20.40	-0.48	-0.24

Table 4-SI: Summary statistics of length values (*cm*) in relation to *PCB* concentrations in lake trout and walleye skinless-boneless fillet data in Lake Ontario.

Species	Gender	N	Mean	stdev	Median	2.50%	97.50%	Int Quart	Kurt	Skew
Lake Trout	**Female**	361	63.17	10.91	65.20	36.00	78.50	12.00	1.38	-1.02
	Male	476	62.07	10.16	62.80	37.99	80.39	11.80	0.92	-0.60
	Pooled Samples	1030	60.36	11.69	62.20	32.75	79.03	15.60	0.39	-0.67
Walleye	**Female**	351	56.38	13.16	58.10	28.58	77.28	19.10	-0.49	-0.46
	Male	295	48.23	11.05	47.50	27.54	67.73	16.05	-0.68	-0.04
	Pooled Samples	754	51.06	13.69	51.85	24.20	73.81	20.38	-0.46	-0.25

Table 5-SI: Deviance information criterion, posterior mean values, 2.5% and 97.5% percentiles (italicized numbers) of the stochastic nodes of the models used to describe the temporal trends of *THg* concentrations (*µg/g* wet weight) in lake trout and walleye.

	Lake Trout				**Walleye**			
Models	**Parameters**	**Female**	**Male**	**Pooled Samples**	**Parameters**	**Female**	**Male**	**Pooled Samples**
Exponental Decay Model	*DIC*	62.9	59.4	124.3	*DIC*	95.5	96.6	242.7
	k	-0.045	-0.037	-0.036	*k*	0.007	0.005	-0.005
		0.004	0.002	0.002		0.006	0.005	0.004
		-0.052	*-0.042*	*-0.040*		*-0.004*	*-0.005*	*-0.012*
		-0.038	*-0.033*	*-0.033*		*0.018*	*0.016*	*0.002*
	Hg0	0.345	0.323	0.330	*Hg0*	0.181	0.226	0.250
		0.019	0.012	0.010		0.015	0.017	0.017
		0.310	*0.299*	*0.311*		*0.154*	*0.194*	*0.218*
		0.382	*0.348*	*0.350*		*0.211*	*0.260*	*0.285*
	σ_ε	0.328	0.281	0.292	σ_ε	0.440	0.434	0.460
		0.024	0.015	0.011		0.036	0.035	0.024
		0.286	*0.254*	*0.270*		*0.375*	*0.372*	*0.415*
		0.380	*0.311*	0.315		*0.517*	*0.510*	*0.510*
Mixed Order Model	*DIC*	83.1	73.5	140.5	*DIC*	112.1	112.6	255.6
	k	0.180	0.334	0.080	*k*	0.028	0.021	0.093
		0.166	0.188	0.038		0.066	0.047	0.153
		0.046	*0.086*	*0.038*		*0.001*	*0.001*	*0.001*
		0.701	*0.812*	*0.179*		*0.202*	*0.146*	*0.572*
	Hg0	0.364	0.373	0.345	*Hg0*	0.198	0.241	0.246
		0.025	0.024	0.014		0.010	0.012	0.018
		0.320	*0.330*	*0.319*		*0.180*	*0.219*	*0.219*
		0.417	*0.423*	*0.375*		*0.219*	*0.265*	*0.290*
	σ_ε	0.335	0.279	0.291	σ_ε	0.442	0.434	0.459
		0.025	0.015	0.011		0.036	0.034	0.024
		0.290	*0.252*	*0.270*		*0.378*	*0.372*	*0.416*
		0.389	*0.310*	*0.315*		*0.520*	*0.508*	*0.509*
Exponential Dcaly Model With Random Walk Temporal Smoothing	*DIC*	31.8	17.1	64.4	*DIC*	84.6	98.6	238.5
	k	-0.061	-0.054	-0.036	*k*	0.006	0.004	-0.016
		0.006	0.007	0.005		0.011	0.008	0.006
		-0.071	*-0.066*	*-0.046*		*-0.015*	*-0.011*	*-0.027*
		-0.049	*-0.038*	*-0.025*		*0.031*	*0.020*	*-0.004*
	Hg0	0.422	0.421	0.336	*Hg0*	0.190	0.230	0.308
		0.033	0.043	0.029		0.030	0.027	0.032
		0.356	*0.329*	*0.279*		*0.131*	*0.180*	*0.247*
		0.486	*0.504*	*0.382*		*0.252*	*0.285*	*0.372*
	σ_ε	0.261	0.243	0.260	σ_ε	0.392	0.433	0.449
		0.023	0.013	0.011		0.036	0.035	0.024
		0.221	*0.218*	*0.240*		*0.328*	*0.371*	*0.405*
		0.310	*0.270*	*0.282*		*0.472*	*0.509*	*0.499*

Table 6-SI: Deviance information criterion, posterior mean values, 2.5% and 97.5% percentiles (italicized numbers) of the stochastic nodes of the models used to describe the temporal trends of *PCB* concentrations (*ng/g* wet weight) in lake trout and walleye.

	Lake Trout				**Walleye**			
Models	**Parameters**	**Female**	**Male**	**Pooled Samples**	**Parameters**	**Female**	**Male**	**Pooled Samples**
Exponental decay Model	*DIC*	208.8	347.9	587.4	*DIC*	215.4	253.3	559.9
	k	-0.113	-0.081	-0.075	*k*	-0.069	-0.098	-0.074
		0.005	0.004	0.002		0.013	0.015	0.007
		-0.122	*-0.088*	*-0.080*		*-0.095*	*-0.128*	*-0.087*
		-0.104	*-0.073*	*-0.070*		*-0.043*	*-0.070*	*-0.061*
	PCB0	6391.0	3619.0	3809.0	*PCB0*	162.1	566.7	340.5
		311.3	206.8	130.0		34.4	145.0	37.5
		5782.0	*3225.0*	*3555.0*		*106.3*	*342.7*	*270.5*
		7005.0	*4033.0*	*4069.0*		*241.6*	*909.0*	*416.9*
	σ_ε	0.611	0.583	0.569	σ_ε	0.947	1.060	1.035
		0.043	0.029	0.022		0.079	0.088	0.053
		0.533	*0.528*	*0.528*		*0.808*	*0.907*	*0.937*
		0.701	*0.643*	*0.615*		*1.118*	*1.250*	*1.148*
Mixed Order Model	*DIC*	218.3	365.2	605.1	*DIC*	225.4	257.4	573.6
	k	0.005	0.042	0.060	*k*	0.003	0.002	0.016
		0.008	0.021	0.012		0.003	0.001	0.014
		0.001	*0.009*	*0.032*		*0.001*	*0.001*	*0.002*
		0.028	*0.079*	*0.076*		*0.008*	*0.006*	*0.056*
	PCB0	6746.0	3692.0	3819.0	*PCB0*	321.1	1711.0	372.7
		315.7	225.0	133.4		86.0	305.4	42.5
		6124.0	3269.0	*3562.0*		*176.6*	*1121.0*	*293.1*
		7360.0	4147.0	*4086.0*		*508.7*	*2315.0*	*459.8*
	σ_ε	0.592	0.583	0.571	σ_ε	0.914	0.989	1.027
		0.040	0.030	0.022		0.075	0.079	0.053
		0.519	*0.529*	*0.530*		*0.780*	*0.850*	*0.928*
		0.677	*0.645*	*0.617*		*1.074*	*1.157*	*1.138*
Exponential Decay Model With Random Walk Temporal Smoothing	*DIC*	187.4	323.8	549.2	*DIC*	199.3	234.1	524.9
	k	-0.125	-0.075	-0.070	*k*	-0.128	-0.170	-0.064
		0.005	0.007	0.004		0.022	0.014	0.011
		-0.135	*-0.088*	*-0.078*		*-0.164*	*-0.195*	*-0.083*
		-0.115	*-0.060*	*-0.062*		*-0.077*	*-0.138*	*-0.042*
	PCB0	6980.0	3368.0	3543.0	*PCB0*	451.0	1771.0	329.9
		325.6	341.1	192.2		128.6	349.3	55.4
		6347.0	*2710.0*	*3166.0*		*203.6*	*1085.0*	*221.6*
		7631.0	*4037.0*	*3918.0*		*710.8*	*2455.0*	*438.0*
	σ_ε	0.538	0.526	0.524	σ_ε	0.815	0.892	0.913
		0.039	0.030	0.022		0.072	0.080	0.051
		0.467	*0.473*	*0.484*		*0.687*	*0.751*	*0.819*
		0.622	*0.589*	*0.569*		*0.969*	*1.062*	*1.019*

Table 7-SI: Posterior estimates of the length (*β_1*) and lipid (*β_2*) regression coefficients (mean values ± standard deviations) for the dynamic linear models used to describe the temporal trends of *THg* concentrations (*µg/g* wet weight) in lake trout and walleye.

	Lake Trout Hg Pooled Samples							**Walleye Hg Pooled Samples**					
	DIC L+L		**DIC LNG**	**DIC LPD**				**DIC L+L**		**DIC LNG**	**DIC LPD**		
	361.7		389.7	1043.5				1032.6		1035.1	1873.4		
	$\widehat{\beta}_1$		$\sigma_{\beta 1}$	$\widehat{\beta}_2$		$\sigma_{\beta 2}$		$\widehat{\beta}_1$		$\sigma_{\beta 1}$	$\widehat{\beta}_2$		$\sigma_{\beta 2}$
1978	0.394	±	0.034	-0.097	±	0.039	1975	1.151	±	0.108	0.044	±	0.041
1979	0.388	±	0.039	-0.062	±	0.047	1977	1.091	±	0.153	0.046	±	0.041
1980	0.386	±	0.034	-0.030	±	0.041	1980	1.119	±	0.153	0.052	±	0.037
1981	0.369	±	0.021	-0.015	±	0.025	1981	1.185	±	0.075	0.055	±	0.025
1982	0.357	±	0.034	-0.068	±	0.043	1982	0.968	±	0.141	0.053	±	0.035
1984	0.399	±	0.038	-0.073	±	0.042	1985	0.695	±	0.063	0.051	±	0.038
1985	0.431	±	0.046	-0.065	±	0.042	1987	0.692	±	0.056	0.033	±	0.041
1986	0.457	±	0.048	-0.081	±	0.036	1988	0.662	±	0.128	0.022	±	0.046
1987	0.454	±	0.046	-0.105	±	0.046	1989	0.766	±	0.040	0.015	±	0.045
1988	0.442	±	0.041	-0.142	±	0.050	1990	0.828	±	0.100	0.019	±	0.050
1989	0.444	±	0.041	-0.069	±	0.036	1991	1.121	±	0.132	0.032	±	0.052
1990	0.462	±	0.031	0.004	±	0.043	1992	1.031	±	0.120	0.028	±	0.046
1991	0.456	±	0.038	0.023	±	0.048	1993	0.449	±	0.040	0.016	±	0.037
1993	0.403	±	0.037	-0.032	±	0.036	1994	0.769	±	0.062	0.012	±	0.045
1994	0.439	±	0.029	0.000	±	0.023	1996	0.935	±	0.137	0.014	±	0.056
1996	0.470	±	0.040	-0.100	±	0.038	1997	0.844	±	0.176	0.013	±	0.062
1997	0.465	±	0.056	-0.082	±	0.057	1998	0.750	±	0.097	0.011	±	0.064
1998	0.450	±	0.060	-0.093	±	0.072	1999	0.927	±	0.186	0.026	±	0.068
1999	0.456	±	0.056	-0.094	±	0.062	2001	0.839	±	0.129	0.046	±	0.071
2002	0.432	±	0.046	-0.076	±	0.049	2002	0.968	±	0.168	0.052	±	0.073
2004	0.400	±	0.054	-0.059	±	0.056	2003	0.843	±	0.205	0.063	±	0.075
2006	0.391	±	0.063	-0.010	±	0.061	2004	0.827	±	0.122	0.078	±	0.073
2007	0.333	±	0.075	0.027	±	0.083	2005	0.922	±	0.132	0.091	±	0.079
2008	0.325	±	0.079	-0.006	±	0.095	2006	0.882	±	0.102	0.104	±	0.078
2010	0.306	±	0.081	-0.053	±	0.083	2007	0.778	±	0.137	0.107	±	0.090
2011	0.282	±	0.097	-0.087	±	0.106	2010	0.807	±	0.147	0.114	±	0.100
							2011	0.600	±	0.183	0.110	±	0.120

DIC L+L, DIC LNG, DIC LPD represent the deviance information criterion values for the models that collectively (L+L) or individually consider fish lengths (LNG) and lipids (LPD) as covariates.

Table 8-SI: Posterior estimates of the length (β_1) and lipid (β_2) regression coefficients (mean values ± standard deviations) for the dynamic linear models used to describe the temporal trends of *PCB* concentrations ($\mu g/g$ wet weight) in lake trout and walleye.

	Lake Trout PCB Pooled Samples							**Walleye PCB Pooled Samples**					
	DIC L+L		**DIC LNG**	**DIC LPD**				**DIC L+L**		**DIC LNG**	**DIC LPD**		
	1603.2		1749.3	1844.8				1947.7		2101.9	2173.0		
	$\hat{\beta}_1$		$\sigma_{\beta 1}$	$\hat{\beta}_2$		$\sigma_{\beta 2}$		$\hat{\beta}_1$		$\sigma_{\beta 1}$	$\hat{\beta}_2$		$\sigma_{\beta 2}$
1978	0.205	±	0.064	0.233	±	0.053	1975	0.893	±	0.145	0.335	±	0.115
1979	0.241	±	0.073	0.242	±	0.056	1977	0.919	±	0.155	0.325	±	0.155
1980	0.301	±	0.062	0.249	±	0.051	1978	0.949	±	0.157	0.317	±	0.158
1981	0.303	±	0.039	0.232	±	0.041	1980	0.964	±	0.149	0.210	±	0.148
1982	0.278	±	0.062	0.246	±	0.050	1981	0.988	±	0.128	0.059	±	0.052
1984	0.377	±	0.075	0.272	±	0.052	1982	0.878	±	0.122	0.319	±	0.139
1985	0.437	±	0.097	0.309	±	0.065	1985	0.735	±	0.095	0.529	±	0.108
1986	0.425	±	0.085	0.288	±	0.050	1987	0.779	±	0.084	0.683	±	0.125
1987	0.402	±	0.083	0.272	±	0.055	1988	0.691	±	0.111	0.756	±	0.170
1988	0.438	±	0.082	0.271	±	0.054	1989	0.584	±	0.067	0.792	±	0.128
1989	0.399	±	0.082	0.265	±	0.052	1990	0.437	±	0.129	0.680	±	0.191
1990	0.286	±	0.052	0.265	±	0.056	1991	0.472	±	0.127	0.734	±	0.204
1991	0.324	±	0.066	0.310	±	0.056	1992	0.469	±	0.125	0.663	±	0.152
1993	0.386	±	0.058	0.344	±	0.055	1993	0.318	±	0.067	0.375	±	0.083
1994	0.395	±	0.051	0.308	±	0.036	1994	0.330	±	0.089	0.583	±	0.123
1996	0.531	±	0.073	0.313	±	0.049	1996	0.438	±	0.155	0.732	±	0.203
1997	0.568	±	0.104	0.334	±	0.069	1997	0.431	±	0.174	0.794	±	0.218
1998	0.596	±	0.099	0.323	±	0.075	1998	0.340	±	0.138	0.748	±	0.180
1999	0.592	±	0.100	0.314	±	0.074	1999	0.364	±	0.175	0.574	±	0.214
2002	0.529	±	0.074	0.277	±	0.073	2001	0.344	±	0.165	0.608	±	0.238
2004	0.498	±	0.095	0.324	±	0.076	2002	0.331	±	0.186	0.622	±	0.229
2006	0.541	±	0.113	0.370	±	0.089	2003	0.253	±	0.193	0.659	±	0.239
2007	0.464	±	0.133	0.380	±	0.107	2004	0.200	±	0.152	0.746	±	0.185
2008	0.386	±	0.140	0.356	±	0.114	2005	0.215	±	0.164	0.815	±	0.256
2010	0.314	±	0.141	0.329	±	0.116	2006	0.317	±	0.152	0.928	±	0.188
2011	0.267	±	0.163	0.335	±	0.137	2007	0.268	±	0.176	0.860	±	0.282
							2010	0.360	±	0.204	0.773	±	0.276
							2011	0.283	±	0.253	0.644	±	0.412

DIC L+L, DIC LNG, DIC LPD represent the deviance information criterion values for the models that collectively (L+L) or individually consider fish lengths (LNG) and lipids (LPD) as covariates.

Table 9-SI: Posterior estimates of the length (β_1) and lipid (β_2) regression coefficients (mean values ± standard deviations) for the dynamic linear models used to describe the temporal trends of mercury concentrations ($\mu g/g$ wet weight) in lake trout and walleye females.

[redacted]

DIC L+L, DIC LNG, DIC LPD represent the deviance information criterion values for the models that collectively (L+L) or individually consider fish lengths (LNG) and lipids (LPD) as covariates.

Table 10-SI: Posterior estimates of the length (β_1) and lipid (β_2) regression coefficients (mean values ± standard deviations) for the dynamic linear models used to describe the temporal trends of PCB concentrations ($\mu g/g$ wet weight) in lake trout and walleye females.

[redacted]

DIC L+L, DIC LNG, DIC LPD represent the deviance information criterion values for the models that collectively (L+L) or individually consider fish lengths (LNG) and lipids (LPD) as covariates.

Table 11-SI: Posterior estimates of the length (β_1) and lipid (β_2) regression coefficients (mean values ± standard deviations) for the dynamic linear models used to describe the temporal trends of mercury concentrations ($\mu g/g$ wet weight) in lake trout and walleye males.

[redacted]

DIC L+L, DIC LNG, DIC LPD represent the deviance information criterion values for the models that collectively (L+L) or individually consider fish lengths (LNG) and lipids (LPD) as covariates.

Table 12-SI: Posterior estimates of the length (β_1) and lipid (β_2) regression coefficients (mean values ± standard deviations) for the dynamic linear models used to describe the temporal trends of PCB concentrations ($\mu g/g$ wet weight) in lake trout and walleye males.

[redacted]

DIC L+L, DIC LNG, DIC LPD represent the deviance information criterion values for the models that collectively (L+L) or individually consider fish lengths (LNG) and lipids (LPD) as covariates.