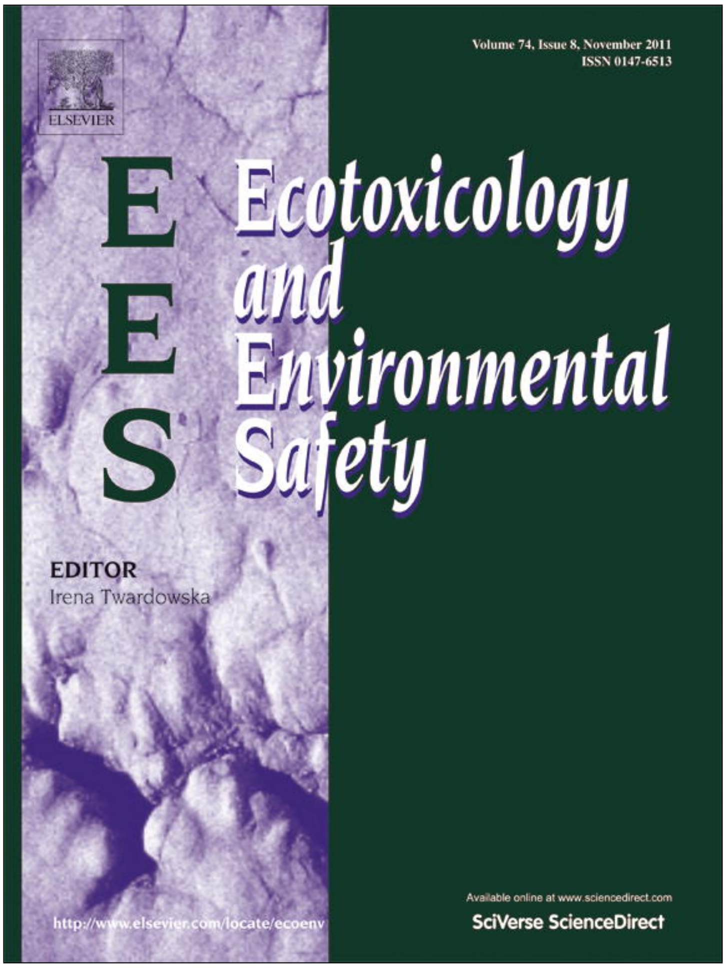
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Temporal PCB and mercury trends in Lake Erie fish communities: A dynamic linear modeling analysis

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ABSTRACT

We performed dynamic linear modeling analysis on fish contaminant data collected from the Ontario Ministry of the Environment and Environment Canada to examine long-term trends of total mercury (THg) and polychlorinated biphenyls (PCBs) in Lake Erie. Several sport fish species (walleye, smallmouth bass, rainbow trout) with differences in their diet habits, food competition strategies and foraging patterns are characterized by weakly increasing trends of their THg levels in Lake Erie after the mid- or late 1990s. Similarly, our analysis shows that the decline rates of the PCB body burdens in white bass, smallmouth bass, freshwater drum and whitefish have slowed down or have switched to weakly increasing rates over the last decade. Our analysis also provides evidence that the rainbow trout and coho salmon PCB concentrations have been decreasing steadily but the associated rates were fairly weak. The systematic shifts in energy trophodynamics along with the food web alterations induced from the introduction of non-native species, the new role of the sediments as a net contaminant source, and the potentially significant fluxes from the atmosphere stand out as some of the hypotheses proposed to explain the limited Lake Erie response in recent years to the various contamination mitigation strategies.

1. Introduction

Among the toxic, persistent and bioaccumulative contaminants, methylmercury (MeHg) and polychlorinated biphenyls (PCBs) are of particular concern and restrict the use of valuable commercial and recreational fishery resources in the Great Lakes. The two contaminants have contrasting bioaccumulation characteristics. MeHg is an intracellular organo-metallic contaminant with high affinity to the sulfite group of host proteins that efficiently deposits into muscle tissue of organisms (Hudson et al., 1998; Wang and Wong, 2003). Existing evidence suggests that several important physical (e.g., lake area, epilimnetic temperature), chemical (e.g., organic matter, pH, Hg aging, iron levels, balance between sulfate and sulfide) and biological (e.g., type and activity of bacteria, food web structure, fish population age and growth rates) factors can potentially control MeHg bioaccumulation and dramatically affect the transfer of Hg load into MeHg in fish (Munthe et al., 2007). PCBs are highly lipophilic and more tightly deposit into fat tissues (Elskus et al., 2005). Depending on their ethology and trophic position, fish are likely to receive PCBs through three specific routes, viz., 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.

High concentrations of the two contaminants were recorded in the Great Lakes in the late 1960s/early 1970s (Fimreite et al., 1971), and soon thereafter their toxic effects were reported in wildlife (Grier, 1968; Gilbertson et al., 1976) and humans (Lonky et al., 1996; Cole et al., 2004). Consumption of Great Lakes sport fish has also been reported to be one of the significant contaminant sources in the human body (Humphrey, 1988; Tee et al., 2003; Cole et al., 2004). MeHg is a potent neurotoxicant, which impairs brain development (Rice, 2008) and increases the risk of cardiovascular diseases (Guallar et al., 2002; Goncharov et al., 2008). PCBs 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 be associated with increased risk of diabetes (Codru et al., 2007).

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The public pressure for virtual elimination of persistent toxic pollutants resulted in the Great Lakes Water Quality Agreement (GLWQA) signed between Canada and USA in 1972 and subsequently revised in 1978. The agreement included a call for monitoring and research programs to identify the spatiotemporal trends of the toxic substances in sediments and biota (IJC, 1978, 2006). The Ontario Ministry of the Environment together with several US states and tribes have monitored contaminant levels in edible fish portions since the 1970s and regularly issue species-specific consumption advisories (OMOE, 2008). The continuous monitoring of the fish contaminant levels has been invaluable in raising public awareness of the potential health risks and in encouraging governmental action to prohibit activities that further produce noxious chemicals. For example, the Government of Ontario recently introduced the Toxic Reduction Act to protect the environment and health of Ontarians by reducing toxic substances in air, land, water and consumer products while fostering the green economy (http://www.ene.gov.on.ca/en/toxics/index.php).

Generally, the implementation of various regulatory actions resulted in decreased levels of most contaminants in the Great Lakes fish through the 1980s, but the rates of decrease are reported to have diminished or even to have switched into increasing trends in the 1990s; especially in Lake Erie (Bhavsar et al., 2007, 2010; Carlson et al., 2010; Azim et al., 2011). The reasons for these unexpected trends are not fully known, but existing hypotheses suggest direct causal associations with the food web alterations induced by aquatic invasive species in the Great Lakes area (Morrison et al., 1998; Hogan et al., 2007; French et al., unpublished data) as well as indirect links with the global warming (French et al., 2006). The invasion of zebra mussels and round gobies has resulted in a food chain lengthening and therefore in a higher fish contamination through the process of biomagnification (Cabana et al., 1994; Campbell et al., 2003), 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). Despite the considerable progress in our understanding of contaminant dynamics in the Great Lakes fish populations, the rigorous assessment of fish contaminant trends in space and time can be influenced by a wide range of factors, such as the type of contaminant and the biological characteristics of the fish itself like size, age, sex, food habits, trophic level, reproductive status, growth and lipid content (Stow et al., 1997). The uncertainty is also associated with the sampling procedures and data analysis including the type of statistical analysis performed, data pooling across locations, type of samples (whole fish vs. fillet portions), sample size and season (Gewurtz et al., unpublished data). Therefore, quantitative approaches that explicitly incorporate all the causal factors are essential to determine the actual patterns of contaminants and to explore the underlying ecological mechanisms.

To this end, dynamic linear modeling (DLM) is an important statistical approach with an evolving structure that enables the elucidation of the role of potentially important cause–effect relationships and supports forecasts primarily driven by most recent data while information from the distant past can be discounted (Pole et al., 1994). The primary objective of this study is to present a dynamic linear modeling analysis based on a long-term fish contaminant dataset for Lake Erie. We consider two important fish contaminants, viz., total mercury (THg) and polychlorinated biphenyls (PCBs) and nine intensively studied fish species for each contaminant. We also selected fish length as a potential covariate of the contaminant concentrations to account for the fact that different sized fish may have been sampled over time. In particular, the Ontario Ministry of the Environment's Fish Contaminant Monitoring Program initially sampled edible portions of wider fish size ranges, but subsequently the focus shifted on larger size classes. By contrast, Environment Canada's program reflects an ecosystem/wildlife health point of view that mainly considers whole fish samples representing all size classes (Bhavsar et al., 2010). Thus, our aim herein is to examine the trends of observed Hg and PCB concentrations in whole fish and skinless boneless fillet of different fish species when explicitly accounting for the fish length variability. We then discuss plausible ecological mechanisms that can conceivably modulate the species-specific contaminant concentrations and thus shape the corresponding temporal patterns.

2. Methods

2.1. Dataset description

The present study is based on the provincial (Ontario Ministry of the Environment; OMOE, Canada) dorsal skinless boneless fillet (SBF) measurements for a wide range of species for fish consumption advisories, and the federal (Environment Canada; EC) whole fish (WF) walleye measurements to assess overall environmental contamination and risk to fish and fish-consuming wildlife (Bhavsar et al., 2010). The two datasets approximately span a 40-year period of contaminant concentrations (1977–2007). The fish species considered were selected on the basis of the data availability and/or their commercial importance. Specifically, we examined the contaminant trends for OMOE SBF data for walleye (*Stizostedion vitreum*), smallmouth bass (*Micropterus dolomieui*), lake trout (*Salvelinus namaycush*), coho salmon (*Oncorhynchus kisutch*), common carp (*Cyprinus carpio*), white bass (*Morone chrysops*), rainbow trout (*Oncorhynchus mykiss*), northern pike (*Esox lucius*), freshwater drum (*Aplodinotus grunniens*), channel catfish (*Ictalurus punctatus*), whitefish (*Coregonus clupeaformis*) and EC WF for walleye. The OMOE SBF samples were collected from several sites on the Canadian side, while EC WF samples were collected from Pelee Island in the western part of Lake Erie (Fig. 1 in the Supplementary material).

2.2. Chemical analysis

The OMOE samples were analyzed for THg by employing the cold vapor-flameless atomic absorption spectrophotometry (CV-FAAS) technique at the OMOE laboratory in Toronto using the OMOE method HGBIO-E3057. The EC samples were analyzed for THg with the cold vapor absorption spectrometry procedure (VVAAS) at the National Laboratory for Environmental Testing using NLET method 2801. These methods are described in supporting material of Bhavsar et al. (2010). Total-PCB analysis on the OMOE and EC samples was performed through gas chromatography with ^{63}Ni electron capture detector (ECD) as described by Bhavsar et al. (2007) and Borgmann and Whittle (1983), respectively. Quantification was carried out using the 23 largest "Aroclor" peaks obtained in the pseudo packed column technique. For lower level samples, a minimum of 11 peaks was required for a positive identification. The areas of the peaks detected were summed and compared to the summed areas of the 4:1 mixture of Aroclor 1254:1260. 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. A blank and spiked blank matrix sample were 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).

2.3. Modeling framework

Dynamic Linear Modeling analysis was used to examine the THg/PCB temporal trends, while explicitly accounting for the fact that the fish length covaries with the contaminant concentrations and that different fish sizes may have been sampled over time. The main advantage of the DLMs is the explicit recognition of structure in the time series, i.e., the data are sequentially ordered and the level of the response variable at each time step is related to its levels at earlier time steps in the data series (Lamon et al., 1998; Stow et al., 2004). In contrast with regression analysis, in which each observation contains information on each parameter, DLM parameter estimates are influenced only by prior and current information, not by subsequent data. Parameter values are dynamic and reflect shifts in both the level of the response variable and the underlying ecological processes. DLMs easily handle missing values/unequally spaced data, and minimize the effect of outliers (Pole et al., 1994). All DLMs consist of an observation equation and system equations (West and Harrison, 1989). In particular, the DLMs used herein were specified as follows:

Observation equation:

$$\ln[THg/PCB]_{ti} = level_t + \beta_t \ln[length]_{ti} + \psi_{ti}, \quad \psi_{ti} \sim N[0, \Psi_t]$$

System equations:

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

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

$$\beta_t = \beta_{t-1} + \omega_{t3}, \quad \omega_{t3} \sim N[0, \Omega_{t3}]$$

$$1/\Omega_{tj}{}^2 = \zeta^{t-1} 1/\Omega_{1j}^2, \quad 1/\Psi_t{}^2 = \zeta^{t-1} 1/\Psi_1^2 \quad t > 1 \text{ and } j = 1 \ 3$$

$$level_1, rate_1, \beta_1 \sim N(0, 10000) \quad t = 1$$

$$1/\Omega_{1j}^2, \quad 1/\Psi_1^2 \sim gamma(0.001, 0.001)$$

where $\ln[THg/PCB]_{ti}$ is the observed Hg/PCB concentration at time t in the individual sample i; $level_t$ is the mean Hg/PCB concentration at time t when accounting for the covariance with the fish length; $\ln[length]_{ti}$ is the observed (standardized) fish length at time t in the individual sample i; $rate_t$ is the rate of change of the level variable; β_t is a length (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 $gamma(0.001, 0.001)$ 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_1, $1/\Omega_{1j}^2$, and $1/\Psi_1^2$ are considered "non-informative" or vague. The DLM process makes a forecast for time t based on prior knowledge of the parameters, and then we observe data at time t. Using Bayes' Theorem, our knowledge regarding the parameters is updated using the likelihood of the data and our prior knowledge (Gelman et al., 2004). In this study, we introduce non-constant and data-driven variances (with respect to time) using a discount factor on the first period prior (Congdon, 2003). We examined different discounts between 0.8 and 1.0 (i.e., the static regression model) and the results reported here are based on a discount value of 0.95. Discounts were selected by conducting a model search, in which models with different discount factors were compared on the basis of their difference in $\log_e$likelihoods (Lamon et al., 1998). This discounted posterior knowledge becomes prior knowledge for time $t+1$, and the process is repeated.

2.4. Model computations

Sequence of realizations from the model posterior distributions were obtained using Markov chain Monte Carlo (MCMC) simulations (Gilks et al., 1998). Specifically, we used the general normal-proposal Metropolis algorithm as implemented in the WinBUGS software; this algorithm is based on a symmetric normal-proposal distribution, whose standard deviation is adjusted over the first 4000 iterations such as the acceptance rate ranges between 20 percent and 40 percent. We used three chain runs of 80,000 iterations and samples were taken after the MCMC simulation converged to the true posterior distribution. Convergence was assessed using the modified Gelman–Rubin convergence statistic (Brooks and Gelman, 1998). Generally, we noticed that the sequences converged very rapidly ($\approx$1000 iterations), and the summary statistics reported in this study were based on the last 75,000 draws by keeping every 20th iteration (thin=20) to avoid serial correlation. 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; see Spiegelhalter et al., 2003) for all parameters was less than five percent of the sample standard deviation.

3. Results

3.1. THg/PCB levels

The summary statistics of the total mercury (THg) concentrations in the fish species examined are shown in Table 1. The highest concentrations were recorded in northern pike (mean 0.237 and median 0.180 μg g^{-1} wet weight or ww), followed by walleye (0.199 and 0.160 μg g^{-1} ww), freshwater drum (0.183 and 0.140 μg g^{-1} ww), smallmouth bass (0.173 and 0.140 μg g^{-1} ww) and common carp (0.154 and 0.150 μg g^{-1} ww). By contrast, coho salmon (0.100 and 0.090 μg g^{-1} ww) and rainbow trout (0.104 and 0.090 μg g^{-1} ww) demonstrated the lowest concentrations. For walleye, the THg concentrations were more than 60 percent higher in skinless boneless fillet relative to whole fish portions (0.122 and 0.107 μg g^{-1} ww). Table 2 presents the same basic statistics for the observed total-PCB concentrations in nine fish species in Lake Erie. The PCB levels are nearly twelve times higher in walleye whole fish (WF) relative to skinless boneless fillet (SBF). Walleye SBF was also characterized by the lowest concentrations (115 and 80 ng g^{-1} ww), whereas the average PCB values in freshwater drum and

Table 1
Basic statistics of THg concentrations in walleye whole fish data, and skinless–boneless fillet data (μg/g wet weight) for nine fish species in Lake Erie (study period 1977–2007).

Species	*N*	Mean	SD	Median	Inter-quartile range	Skewness	Kurtosis
Walleye (WF), *Stizostedion vitreum*	756	0.122	0.072	0.107	0.070	2.526	10.538
Walleye (SBF), *Stizostedion vitreum*	1839	0.199	0.132	0.160	0.140	2.549	10.914
Smallmouth bass, *Micropterus dolomieui*	714	0.173	0.123	0.140	0.120	2.375	7.824
Lake trout, *Salvelinus namaycush*	126	0.118	0.047	0.110	0.070	0.775	0.506
Coho salmon, *Oncorhynchus kisutch*	729	0.100	0.052	0.090	0.050	1.485	4.479
Common carp, *Cyprinus carpio*	379	0.154	0.082	0.150	0.100	1.314	4.636
White bass, *Morone chrysops*	1651	0.146	0.104	0.120	0.100	3.146	18.841
Rainbow trout, *Oncorhynchus mykiss*	233	0.104	0.062	0.090	0.070	1.782	5.927
Northern pike, *Esox lucius*	286	0.237	0.161	0.180	0.170	1.674	3.239
Freshwater drum, *Aplodinotus grunniens*	621	0.183	0.155	0.140	0.130	2.564	8.396

Table 2
Basic statistics of PCB concentrations in walleye whole fish data, and skinless–boneless fillet data (ng/g wet weight) for nine fish species in Lake Erie (study period 1977–2007).

Species	*N*	Mean	SD	Median	Inter-quartile range	Skewness	Kurtosis
Walleye (WF), *Stizostedion vitreum*	969	1329	987.7	1100	900	2.417	9.658
Walleye (SBF), *Stizostedion vitreum*	899	114.6	119.3	80	105	2.797	1.928
Channel catfish, *Ictalurus Punctatus*	556	1042.7	1194.3	590	1100	2.259	6.515
Coho salmon, *Oncorhynchus kisutch*	694	463.0	250.9	410	311	1.184	2.871
Common carp, *Cyprinus carpio*	374	748.6	1149.7	350	837	3.458	15.730
Freshwater drum, *Aplodinotus grunniens*	360	236.2	290.4	160	200	4.702	37.247
Rainbow trout, *Oncorhynchus mykiss*	302	399.4	290.7	326	350	1.582	5.140
Smallmouth bass, *Micropterus dolomieui*	417	220.3	282.6	112	202	2.579	7.501
White bass, *Morone chrysops*	1165	309.3	245.3	240	264	1.842	4.376
Whitefish, *Coregonus Clupeaformis*	350	247.3	312.8	165	200	8.190	104.351

smallmouth bass were lying in somewhat higher levels ($\approx 230\ \mathrm{ng\,g^{-1}}$ ww). On the other hand, channel catfish had the highest PCB concentrations (1043 and 590 $\mathrm{ng\,g^{-1}}$ ww) followed by common carp (mean 749 and median 350 $\mathrm{ng\,g^{-1}}$ ww), coho salmon (mean 463 and median 410 $\mathrm{ng\,g^{-1}}$ ww), and rainbow trout (mean 399 and median 326 $\mathrm{ng\,g^{-1}}$ ww).

The differences in THg and PCB concentrations between walleye whole fish and skinless boneless fillet samples are unusually high relative to what has been reported (or assumed) in the literature (Stow and Carpenter, 1994; Jackson and Schindler, 1996; Amrhein et al., 1999). Yet, we caution that the fillet and whole-fish concentrations were not derived from concurrent samples and therefore these discrepancies partly stem from the different philosophies (and associated sampling practices) of the two datasets used. Moreover, the high standard deviation and inter-quartile range values reflect the substantial inter- and intra-annual as well as the spatial variability associated with the contaminant levels of the individual fish species. [Note that the summary statistics of THg and PCB concentrations per species and location are provided in the Supplementary Tables S1 and S2.] 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.

3.2. THg trends

The DLM analysis identified several patterns regarding the rates of change in the THg levels in the fish species examined (Fig. 1). Walleye (both WF and SBF data) was characterized by negative rates of change during the initial years, which were gradually diminished or even switched to weakly positive ones after the second half of the 1990s (Fig. 1a,c). In particular, the probability distribution that represents our knowledge of the rate parameter indicates a <50 percent probability (greater than 1:1 odds or even odds) that this rate of change is still negative in walleye SBF data after 1998, while a similar distribution derived from 1985 suggests that the odds were 9.2:1. [Note that 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.] Importantly, the walleye SBF model also predicts that the odds of the rate parameter to be positive in 2008 are 2.1:1. The implications of the latter pattern are reflected in the moderate increase of the predicted average THg levels (when adjusting for the length size sampled) during the second half of our study period (Fig. 2a,c). Similarly, coho salmon showed distinctly negative rates of change during the earlier years of the survey period, which were minimized by the late 1980s (Figs. 1d and 2d). Further, smallmouth bass and northern pike initially demonstrated relatively stable THg concentrations, followed by weakly positive rates of change approximately after the mid-90s and/or throughout the 2000s (Fig. 1e,i). Similar inference can be drawn from the rainbow trout model, although this species' THg burdens from the first half of our study period are missing (Fig. 1h). These recent trends are manifested as a minor (smallmouth bass and northern pike) or moderate (rainbow trout) increase of the length-corrected annual average THg levels (Fig. 2e,h,i). White bass, freshwater drum, common carp, and lake trout do not show any profound temporal shifts in their rates (Fig. 1b,f,g,j), which remained nearly zero throughout the study period resulting in minor changes of the corresponding THg concentrations during the time span examined (Fig. 2b,f,g,j).

3.3. PCB trends

Three main patterns of variability of the PCB rates of change were also identified by the DLM analysis (Fig. 3). First, the walleye (both WF and SBF data), channel catfish, and common carp models do not suggest any distinct temporal shifts in the rates, which remained unaltered throughout the study period (Fig. 3b,c,e,g), and therefore the variability associated with the corresponding level parameters primarily stems from the year-to-year corrections of the length covariance (Fig. 4b,c,e,g). Second, weakly negative rates of change were apparent for coho salmon and rainbow trout (Fig. 3d,j), indicative of a continuous decrease in the PCB concentrations during the time period examined (Fig. 4d,j). The odds that the rate parameter has been negative are on average 2.4:1 for coho salmon and 2.3:1 for rainbow trout during the study period. Third, the rates of change of the white bass, smallmouth bass, freshwater drum, and whitefish PCB concentrations have switched from weakly negative to nearly zero during the 2000s (Fig. 3a,f,h,i), and thus the predicted length-adjusted mean PCB values of the four species appear to have been stabilized after a net decrease during the earlier years of the survey period (Fig. 4a,f,h,i). For example, the probability distribution of the smallmouth bass rate parameter indicates a <50 percent probability that the rate of change is still negative after 1998, while a similar distribution derived from 1985 suggests that the odds were 5.76:1.

A synoptic illustration of the relative Hg and PCB trends for the fish species examined is provided in Fig. 5, in which the relative strength of the trends (arrows) is based on the median values of the corresponding rate parameters while the mid-90s are used as a reference point to distinguish between pre- and post-invasion periods of the Lake Erie food web by exotic species (see also following discussion). We also examined the sensitivity of the trends delineated by the dynamic linear models when using annual averages rather than individual samples. Notably, our re-examination showed that the inference remains practically unaltered regardless of the data manipulation adopted prior to the modeling analysis (Figs. 2 and 5 in the Supplementary material). Finally, the intra- and inter-annual variability of the fish lengths sampled along with the species-specific lipid concentration distributions are provided in Figs. 6 and 7 in the Supplementary material. The former plots suggest that there are cases with systematic increase (e.g., walleye filet data) and/or substantial interannual variability of the median fish length sampled, and therefore a rigorous assessment of fish contaminant spatiotemporal trends should explicitly consider the importance of all the possible covariates (e.g., weight, gender, lipid content, age, season, length).

4. Discussion

Following the historical contamination maxima in the late 1960s-early 1970s, both the federal and provincial/state Canadian and US governments have acted to reduce (or to virtually eliminate) the contaminant levels in the Great Lakes through the legislative establishment of stringent emission standards and the effective ban of their production and/or use in many consumer items (Mohapatra et al., 2007). Yet, there has been considerable controversy regarding the signature of these mitigation measures on different aspects of the Lake Erie ecosystem, while the integrity of the local biotic communities arguably remains at risk. For example, while Painter et al. (2001) showed the lakewide sediment concentrations to have significantly decreased over the course of a 30-year period, they have also delineated extended areas of non-compliance with the Canadian Sediment Quality Guidelines, where frequent exceedences of the THg/PCB threshold effect levels occurred (see their figures 7 and 14). Namely, driven by the predominant influence of the Detroit River and the prevailing circulation patterns, Lake Erie

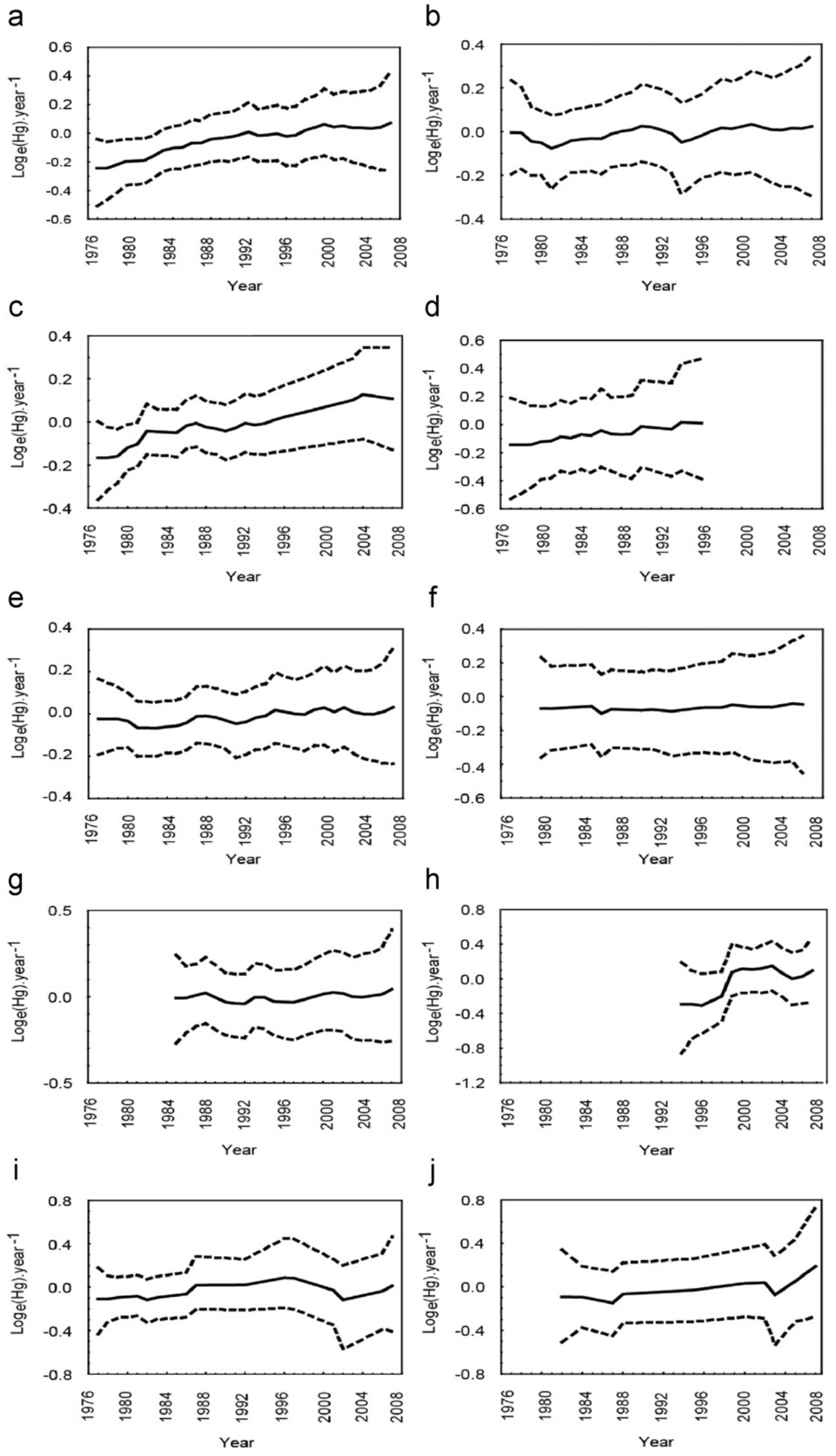


Fig. 1. Dynamic linear modeling analysis depicting the annual rates of change of Hg concentrations (μg/g wet weight) in (a) walleye skinless–boneless fillet, (b) white bass, (c) walleye whole fish, (d) coho salmon, (e) smallmouth bass, (f) freshwater drum, (g) common carp, (h) rainbow trout, (i) northern pike and (j) lake trout in Lake Erie (study period 1977–2007). The solid and dashed lines correspond to the median and the 95 percent posterior predictive intervals, respectively.

demonstrates a spatial trend in increasing sediment contamination from the eastern to the western basin as well as from the north-central to the south-central basin (Painter et al., 2001). Likewise, existing evidence suggests that the chemical burden of the local fish populations has only been moderately ameliorated (Bhavsar et al., 2007, 2010). Acknowledging the multitude of exogenous stressors and intrinsic variables underlying contaminant dynamics in fish communities, our aim herein was (i) to perform retrospective analysis of long-term trends in different fish species while accounting for the confounding effects of an

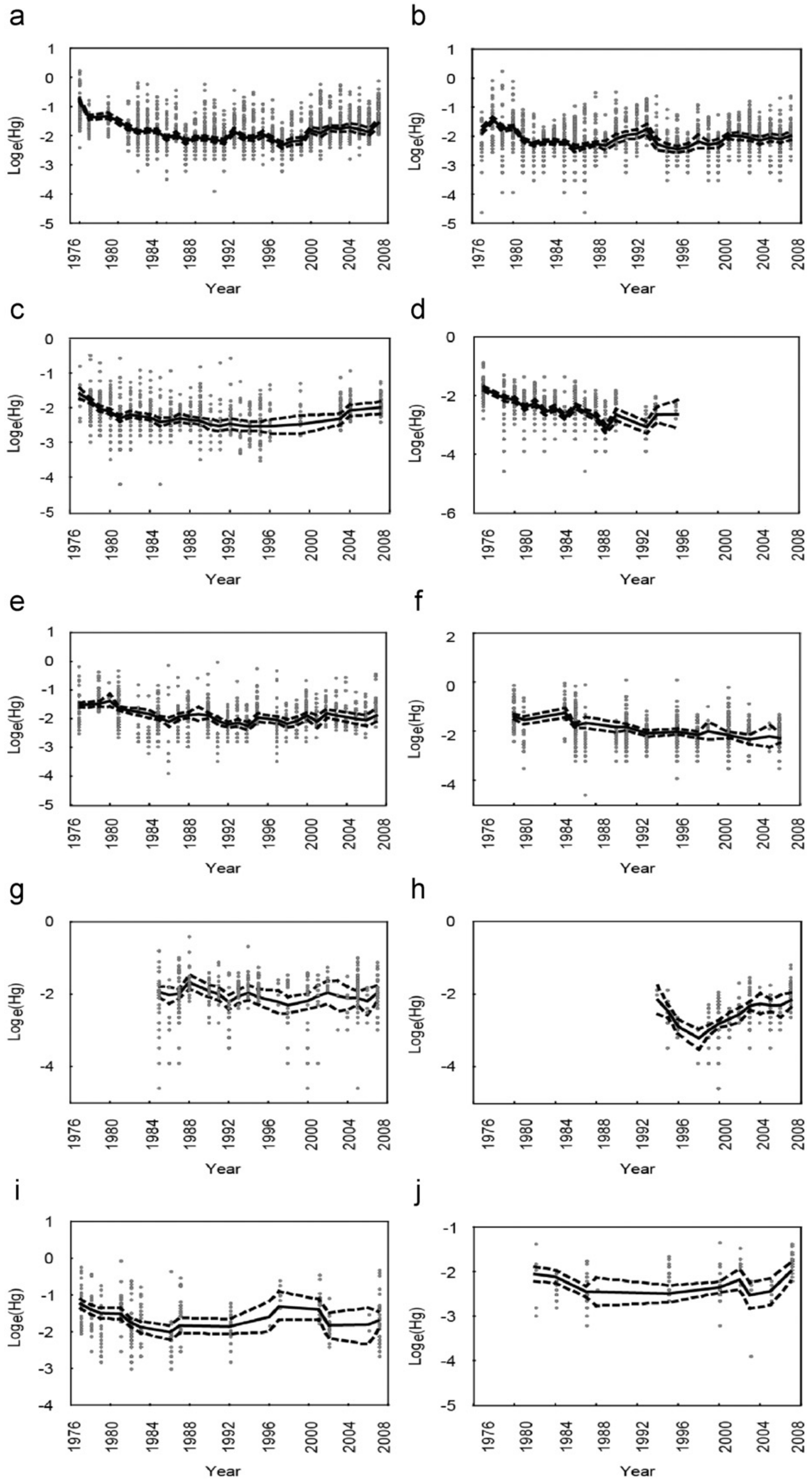


Fig. 2. Dynamic linear modeling analysis depicting the actual Hg concentrations (μg/g wet weight) (gray dots) against the predicted Hg trends when accounting for the covariance with the fish length (black lines) in (a) walleye skinless–boneless fillet, (b) white bass, (c) walleye whole fish, (d) coho salmon, (e) smallmouth bass, (f) freshwater drum, (g) common carp, (h) rainbow trout, (i) northern pike and (j) lake trout in Lake Erie (study period 1977–2007). The solid and dashed lines correspond to the median and the corresponding 95 percent posterior predictive intervals, respectively.

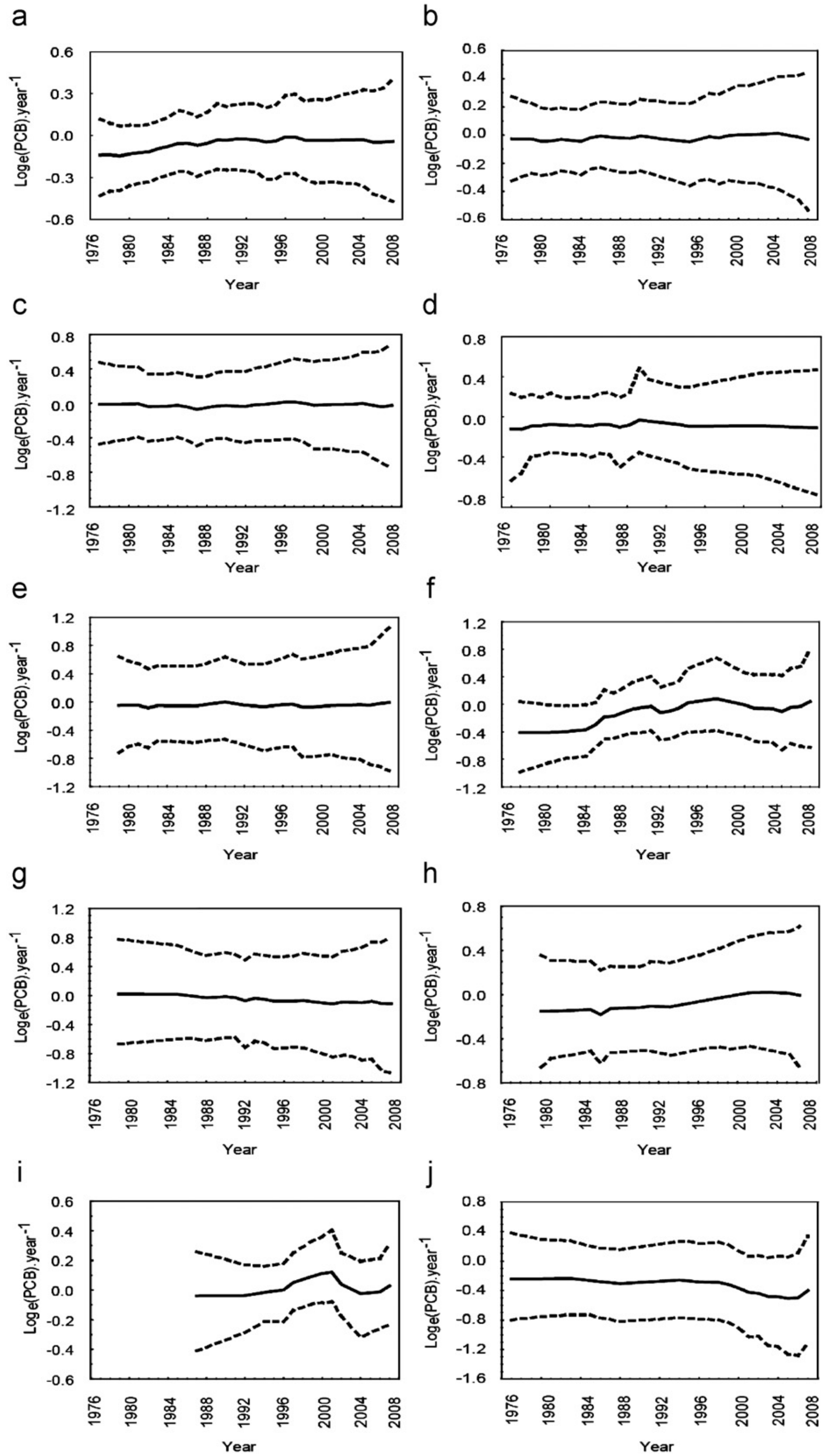


Fig. 3. Dynamic linear modeling analysis depicting the annual rates of change of PCB concentrations (ng/g wet weight) in (a) white bass, (b) walleye whole fish, (c) walleye skinless–boneless fillet, (d) coho salmon, (e) channel catfish, (f) smallmouth bass, (g) common carp, (h) freshwater drum, (i) whitefish and (j) rainbow trout in Lake Erie (study period 1977–2007). The solid and dashed lines correspond to the median and the 95 percent posterior predictive intervals, respectively.

important covariate (i.e., fish length), and (ii) to speculate whether the dietary patterns of each species along with its behavioral habits can serve as reliable latent predictors in future modeling frameworks.

4.1. Species-specific trends

Among the fish species considered in the present analysis, smallmouth bass was characterized by a gradual deceleration of

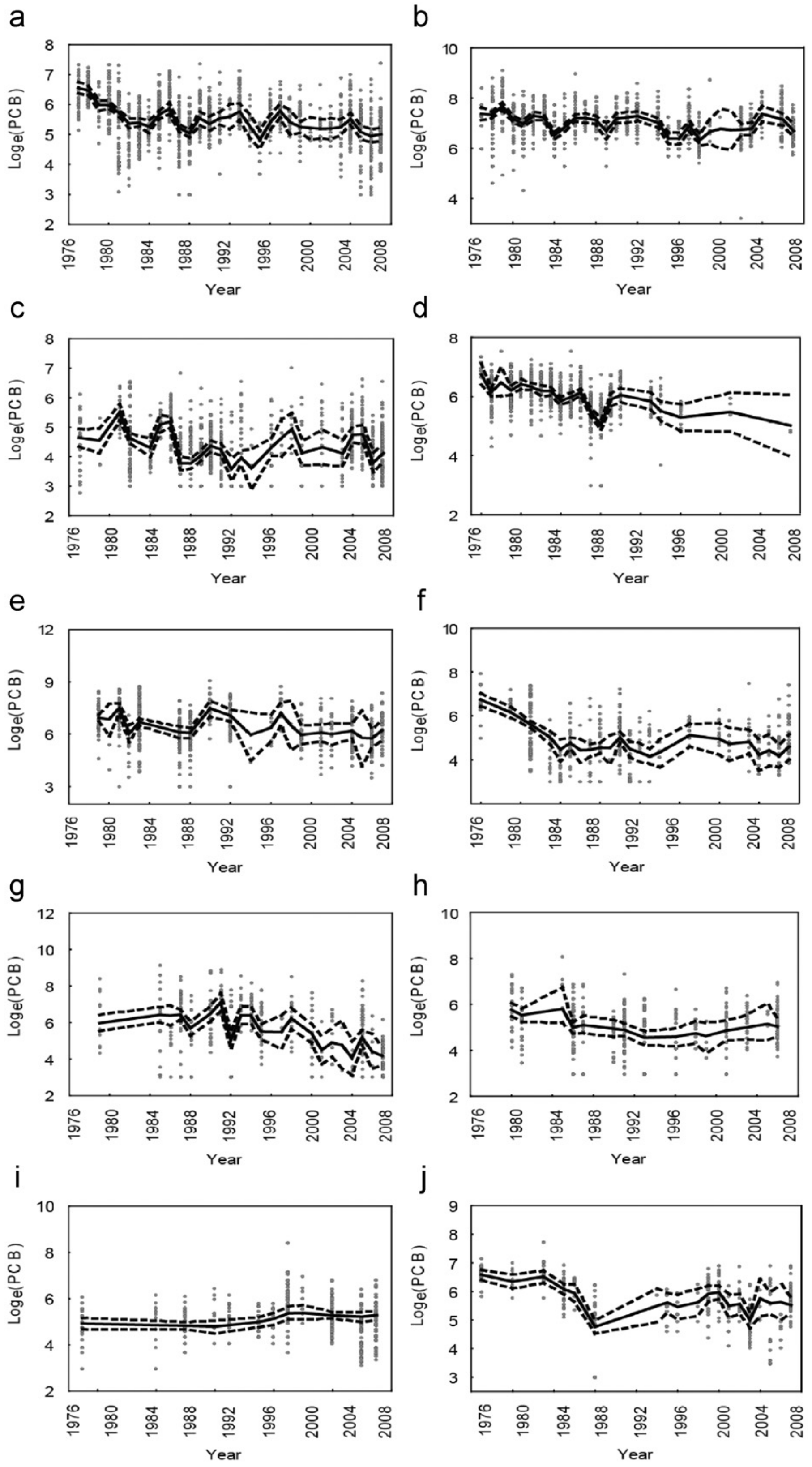
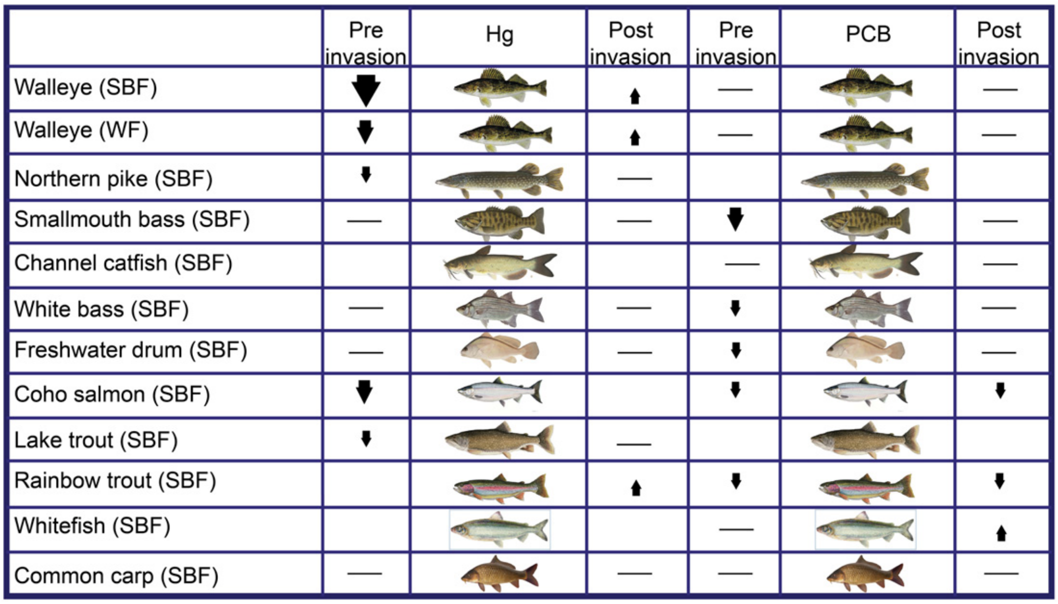


Fig. 4. Dynamic linear modeling analysis depicting the actual PCB concentrations (ng/g wet weight) (gray dots) against the predicted PCB trends when accounting for the covariance with the fish length (black lines) in (a) white bass, (b) walleye whole fish, (c) walleye skinless–boneless fillet, (d) coho salmon, (e) channel catfish, (f) smallmouth bass, (g) common carp, (h) freshwater drum, (i) whitefish and (j) rainbow trout in Lake Erie (study period 1977–2007). The solid and dashed lines correspond to the median and the corresponding 95 percent posterior predictive intervals, respectively.



	Pre invasion	Hg	Post invasion	Pre invasion	PCB	Post invasion
Walleye (SBF)	↓		↑	—		—
Walleye (WF)	↓		↑	—		—
Northern pike (SBF)	↓		—			
Smallmouth bass (SBF)	—		—	↓		—
Channel catfish (SBF)				—		—
White bass (SBF)	—		—	↓		—
Freshwater drum (SBF)	—		—	↓		—
Coho salmon (SBF)	↓			↓		↓
Lake trout (SBF)	↓		—			
Rainbow trout (SBF)			↑	↓		↓
Whitefish (SBF)				—		↑
Common carp (SBF)	—		—	—		—

Fig. 5. Relative Hg and PCB trends for different fish species in Lake Erie. For illustration purposes, the mid-90s are used as a reference point to distinguish between pre- and post-invasion periods of the food web by exotic species (e.g., dreissenids, round goby).

the contaminant decline, which appear to have been stabilized (PCBs) or even to have slightly increased (THg) after the late 1980s. These contaminant temporal trends are on par with the predictions of Hogan et al.'s (2007) conceptual model, which suggests that the invasion of exotic species has shifted the food web from a pelagic-based to a benthic-based one, thereby creating new trophodynamics for contaminant transfer to top predators. Specifically, the invasion of zebra/quagga mussels and round gobies has created a pathway that appears to modulate the contaminant levels in smallmouth bass (Hogan et al., 2007). Further, consistent with Bhavsar et al.'s (2010) recent findings, our analysis provides evidence that the THg concentrations in walleye have also been following an increasing trajectory since the mid-90s, and this pattern was apparent both with the filet and whole-fish data. Yet, the same temporal trends were not manifested with the PCB walleye data, as the rates of change remained almost consistently around zero during the time span examined. The latter result is somewhat different from Bhavsar et al.'s (2007) interpretation of the filet data within the 45–55 cm size range, approximately the mass of data included between the tenth and seventieth percentile of the data used here, which similarly argued that the moderately weak PCB decline rates have leveled off since the late 80s but also suggested a weak upward trajectory after the mid-90s. The difference in our trends after the mid-1990s could be attributed to the consideration of more recent data (for years 2006, 2007), which were among the lowest measurements since the 1990s and the high variability in measurements relative to the overall low PCB levels. While both our and Bhavsar et al.'s (2007) study appear to partial out the fish size effects on PCB bioaccumulation (Amrhein et al., 1999), we draw attention to the systematic increase of the annual median values of the walleye lengths sampled (Fig. 6a,b in the Supplementary material), which may result in discrepancies depending on the statistical method used and/or the assumptions made (e.g., data screening) prior to the analysis. Regarding the likelihood of a causal association between the walleye contaminant trends and the aforementioned trophic pathway involving exotic species, a careful review of the pertinent literature provides contradictory evidence with regards to the walleye reliance on round goby (Bur et al., 2008; Jude et al., 2010). Yet, we cannot rule out the existence of indirect linkages, e.g., through the impact of dreissenids/round gobies on other prey fishes (rainbow smelt, clupeids, shiners), which can presumably shape the contaminant patterns in top pelagic predators.

Our analysis also suggests that freshwater drum is another species that has demonstrated weakly positive rates of change of the PCB levels approximately after the mid-90s and throughout the 2000s. Being closely associated with the sediments and frequently having zebra mussels as a staple in its diet (French and Bur, 1996; Morrison et al., 2000), freshwater drum is likely to have been directly or indirectly influenced by all the alterations in the benthic habitat induced from the colonization and subsequent proliferation of dreissenids (Kuhns and Berg, 1999). Northern pike was similarly characterized by a recent increase of its Hg body burden, although this species' diets are probably closer to those of walleye and the two species are frequently identified as competitors for food resources (Cohen et al., 1993; Fayram et al., 2005). We also caution that the robustness of this increasing pattern remains to be confirmed due to the temporal gaps and the spatial inconsistencies of the information used. That is, the majority of the earlier samples were collected from the eastern basin, while the most recent data were mainly based on sites from the Long Point Bay and the Rondeau Bay (see Fig. 1 in the Supplementary material). Similarly, despite the lack of temporally coherent information for white bass, whitefish, channel catfish, and coho salmon, our modeling analysis suggests that the rates of change of the corresponding THg and/or PCB concentrations have switched from weakly decreasing to nearly zero (or even increasing) during the second half of the survey period. Finally, Lake Erie supports a relatively low lake trout population mainly located at the 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. It has also been estimated that less than 5–10 percent of the sediment-associated pollutants entering the western end of Lake Erie via the Detroit River, ultimately reach the eastern basin (Carter and Hites, 1992). Thus, fish contamination has not been a great concern in the eastern part of the lake, which is also reflected in the relatively low lake trout THg concentrations (mean value $=0.12\ \mathrm{ng\ g^{-1}}$ ww). According to our analysis, the same species is characterized by weakly negative rates of change (not statistically different from zero) of the THg body burdens that have resulted in somewhat reduced (length-corrected) concentrations. Likewise, Bhavsar et al. (2010) has reported weakly negative THg trends for lake trout using simple regression analysis (see their Fig. 2g).

4.2. Possible influencing mechanisms

Generally, our analysis shows that the THg/PCB burdens in a number of fish species with different behavioral patterns/dietary habits appear to have been stabilized or to have increased over the last 10–15 years. Many plausible explanations have been proposed to elucidate the limited response of the Lake Erie fish communities to the various contamination mitigation strategies. As previously discussed, one plausible hypothesis is related to changes in feeding dynamics after the food web restructuring induced by the establishment of non-indigenous species; possibly due to diet shifts in more highly contaminated prey items, decrease in growth rates and subsequent growth dilution and/or switch in food sources that have resulted in energetics-related growth reduction. For example, Paterson et al. (2009) attributed the significant temporal decline in lake trout body mass and energy density to this species *k*-strategist behavior, and thus its inferior competition capacity relative to the *r*-selected non-native salmonids for depleted prey (alewife, rainbow smelt) populations. Further, the introduction of dreissenids has altered the contaminant fluxes and possibly increased the likelihood of trophic transfer through (i) their ability to bioaccumulate by filtering contaminated water and scavenging seston; (ii) the enhancement of the bioavailability of contaminants; (iii) the direct or indirect impact on the diet compositions of other biota; and (iv) the selective removal of particulate organic matter from the water column and the resulting increase of the equilibrium concentrations of the dissolved-phase contaminants which in turn can increase the body levels of many aquatic organisms (Klerks et al., 1997; Nichols et al., 2005; Kwon et al., 2006).

Likewise, among a number of perturbations in the aquatic food web, the invasion of round gobies has caused major shifts in trophic relationships and also increased the growth rates of top predators (Dubbs and Corkum, 1996; Ray and Corkum, 1997; Hogan et al., 2007). In particular, as a benthic fish with diet mainly composed of dreissenids, round goby has the potential to accumulate contaminants and then transfer them to the benthic-oriented smallmouth bass (Johnson et al., 2005). The reportedly high consumption rates of round gobies by smallmouth bass (>70 percent of the diet) as well as their recently documented higher growth rates render support to the strength of dreissenid–goby–smallmouth bass trophic linkage (Johnson et al., 2005; Hogan et al., 2007), and therefore the recent increase (or stabilization) of the smallmouth bass contaminant burdens identified here is a plausible result. Regarding the repercussions of the shift to goby-dominated diets, Johnson et al. (2005) pinpointed the energetic bottleneck stemming from the increased growth response of pelagic predators (e.g., smallmouth bass) despite the low round goby energy density. While it is surmised that this may be due to a reduction in the foraging costs of the goby reliant predators (Johnson et al., 2005), we believe that the elucidation of this causal association may offer insights into the contemporary biomagnification patterns.

Aside from the structural shifts induced in the trophic relationships by the introduction of non-native species, another popular hypothesis suggests that the curtailment of the external contaminant inputs has triggered a change in the historical role of the sediments from a sink to a net contaminant source (Pearson et al., 1996). Consequently, the signature of the sediment fluxes to the body burdens is likely to have increased and it is predicted that we will ultimately reach a chemical equilibrium between sediments and aquatic biota, which may be partly reflected by the plateau-type of pattern (i.e., a decline in the rate of decrease in PCB concentrations in aquatic biota) reported herein and in other Great Lakes sites (Stow et al., 2004; Hickey et al., 2006). The temporal trends of both PCB and THg concentrations in Lake Erie sediments are reported to have declined by approximately 70 percent from 1971 to 1997 (Painter et al., 2001). Yet, there is significant spatial heterogeneity characterizing the sediment contamination, and the western basin of Lake Erie (1997–1998, 0.402 $\mu g\ g^{-1}$) exhibits among the highest mercury levels in the Great Lakes (Marvin et al., 2004a). Further, the sedimentation rates in western Lake Erie (22–160 $g\ m^{-2}\ d^{-1}$) are far greater than in other sites, while the suspended material is predominately characterized by resuspended bottom sediment (Marvin et al., 2004b). There are also several reasons (e.g., extent of the nearshore zone, anoxic conditions) to believe that methylmercury accumulation still occurs in the sediments of Lake Erie, and therefore THg sediment fluxes may accentuate the exposure of the aquatic biota in the overlying waters and conceivably modulate the year-to-year variability in recent years. In this regard, Morrison et al. (2002) supported the notion of a ‘‘two-pronged’’ restoration strategy that alleviates the exogenous inputs, but also enforces the restoration of bottom sediments.

Atmospheric transport and deposition processes may be another potentially important driving factor, as existing evidence suggests that the atmosphere–water exchanges through wet/dry deposition and volatilization can substantially modulate the contaminant fluxes among the various ecosystem components (Jeremiason et al., 1994; Mackay and Bentzen, 1997; Datta et al., 1998). For example, in the case of mercury, a decrease in atmospheric deposition is typically expected to lower fish mercury concentrations, although there could be substantial hysteresis in how lakes respond to decreased loading (Harris et al., 2007). In particular, for all lakes receiving their mercury load from both the watershed and atmosphere, it is predicted that an initial rapid decline in the mercury content of fish, as a result of reduced direct deposition to the lake, will be followed by slower declining rates associated with the complicated (and oftentimes counteracting) processes that are taking place within the watershed context (Harris et al., 2007). Despite the somewhat higher levels relative to other sites in the Great Lakes area, this pattern appears to hold true for the atmospheric PCB concentrations in Lake Erie, which are approaching a steady state after a more rapid decline from 1975 to 1995 (Sun et al., 2006). Further, the net flows of total-PCBs are not significantly different from zero, suggestive of an equilibrium between the corresponding concentrations in the surface water and the overlying atmosphere (Hillery et al., 1997). Yet, the numerous anthropogenic sources associated with urban activities in a local, regional and global scale may cast doubt on the postulation that the recent impact of the atmospheric emissions is consistently minimal. Notably, there is an increasing number of studies pinpointing the significant contribution of global atmospheric emissions to the contaminant deposition in the Great Lakes (Cohen et al., 2004; Trip et al., 2004; Mohapatra et al., 2007).

We emphasize that the present analysis differs from typical dynamic linear modeling approaches in that instead of using annual average concentrations (e.g., Lamon et al., 1998; Stow et al., 2004), we used individual samples to explicitly consider the role of both intra- and interannual variability on the long-term fish contaminant trends (see the walleye filet model code provided in the Supplementary material). To assess the robustness of the projected trends when we partial out the effects of the within-year variability, we implemented the same DLM using the annual means of the natural log-transformed data, while the series of annual average ln-transformed lengths was standardized prior to the analysis (Stow et al., 2004). In this revaluation exercise, both the rates of change and the length-adjusted concentrations suggest that the inference drawn remains unaltered when using annual averages. For example, the DLM still predicts the increasing rates of change (Supplementary Fig. 2a,c) and the U-shaped pattern of the actual THg concentrations (Supplementary Fig. 3a,c) characterizing the walleye SBF and WF data, as well as the nearly constant PCB levels (Supplementary Fig. 5b,c). Aside from the relationship between contaminant concentrations and fish length, the causal link with the fish lipid content could have been another covariate that was not explicitly presented here (Rowan and Rasmussen, 1992; Ewald and Larsson, 1994; Amrhein et al., 1999). Generally, there are contradictory results regarding the strength of this relationship (Larsson et al., 1996; Amrhein et al., 1999), while Stow et al. (1997) noted a stronger PCB:lipid association only when using samples collected during the spawning period. In the Lake Erie dataset, a preliminary examination of the strength of the THg/PCB:lipid relationships among individuals revealed weak covariance for most of the fish species studied in our analysis, and thus the length was the single best covariate for detecting contaminant temporal trends.

5. Conclusions

Several sport fish species (walleye, smallmouth bass, rainbow trout) with differences in their diet habits, food competition strategies and foraging patterns are characterized by weakly increasing trends of their THg levels in Lake Erie after the mid- or late 1990s. Similarly, our analysis shows that the decline rates of the PCB body burdens in white bass, smallmouth bass, freshwater drum and whitefish have slowed down or have switched to weakly increasing rates over the last decade. Our analysis also provides evidence that the rainbow trout and coho salmon PCB concentrations have been decreasing steadily but the associated rates were fairly weak. The systematic shifts in the energy trophodynamics along with the food web alterations induced from the introduction of non-native species, the new role of the sediments as a net contaminant source, and the potentially significant fluxes from the atmosphere stand out as some of the hypotheses proposed to explain the limited Lake Erie response in recent years to the various contamination abatement strategies and predict its future compliance with the goals presented in the Great Lakes Strategy 2002 (US EPA., 2002). The different trends demonstrated by the various fish species or even by different contaminants within the same species stress the importance of embracing more holistic spatial/temporal trend assessment strategies. Future retrospective analysis should focus not only on the total contaminant trends but also on the congener-specific profiles in different sites of the system; such spatially modeling exercise can further advance our understanding of the fish contaminant dynamics and allow the objective assessment of the site-specific risks to the aquatic biota (Batterman et al., 2007).

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Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2011.07.031.

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TEMPORAL PCB AND MERCURY TRENDS IN LAKE ERIE FISH COMMUNITIES: A DYNAMIC LINEAR MODELING ANALYSIS

(Electronic Supplementary Material)

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FIGURES CAPTIONS

Figure 1: Map of Lake Erie with the different sampling sites of the provincial (Ontario Ministry of the Environment; OMOE, Canada) and the federal (Environment Canada; EC) monitoring programs.

Figure 2: Dynamic Linear Modeling analysis with standardised annual average lengths depicting the annual rates of change of *Hg* concentrations (μg/g wet weight) in (*a*) walleye skinless-boneless fillet, (*b*) white bass, (*c*) walleye whole fish, (*d*) coho salmon, (*e*) smallmouth bass, (*f*) freshwater drum, (*g*) common carp, (*h*) rainbow trout, (*i*) northern pike, and (*j*) lake trout in Lake Erie. The solid and dashed lines correspond to the median and the 95% posterior predictive intervals, respectively.

Figure 3: Dynamic Linear Modeling analysis with standardised annual average lengths depicting the predicted *Hg* trends when correcting for the fish length (black lines) against the observed *Hg* concentrations (red dots) in (*a*) walleye skinless-boneless fillet, (*b*) white bass, (*c*) walleye whole fish, (*d*) coho salmon, (*e*) smallmouth bass, (*f*) freshwater drum, (*g*) common carp, (*h*) rainbow trout, (*i*) northern pike, and (*j*) lake trout in Lake Erie. The solid and dashed lines correspond to the median and the corresponding 95% posterior predictive intervals, respectively.

Figure 4: Dynamic Linear Modeling analysis with standardised annual average lengths depicting the annual rates of change of *PCB* concentrations (*ng/g* wet weight) in (*a*) white bass, (*b*) walleye whole fish, (*c*) walleye skinless-boneless fillet, (*d*) coho salmon, (*e*) channel catfish, (*f*) smallmouth bass, (*g*) common carp, (*h*) freshwater drum, (*i*) whitefish, and (*j*) rainbow trout in Lake Erie. The solid and dashed lines correspond to the median and the 95% posterior predictive intervals, respectively.

Figure 5: Dynamic Linear Modeling analysis with standardised annual average lengths depicting the predicted *PCB* trends when correcting for the fish length (black lines) against the observed *PCB*

concentrations (red dots) in (*a*) white bass, (*b*) walleye whole fish, (*c*) walleye skinless-boneless fillet, (*d*) coho salmon, (*e*) channel catfish, (*f*) smallmouth bass, (*g*) common carp, (*h*) freshwater drum, (*i*) whitefish, and (*j*) rainbow trout in Lake Erie. The solid and dashed lines correspond to the median and the corresponding 95% posterior predictive intervals, respectively.

Figure 6: Box plots of the annual sampled length for different fish species in Lake Erie. Extreme values are not included in these plots.

Figure 7: The lipid concentration frequency distributions of different fish species in Lake Erie.

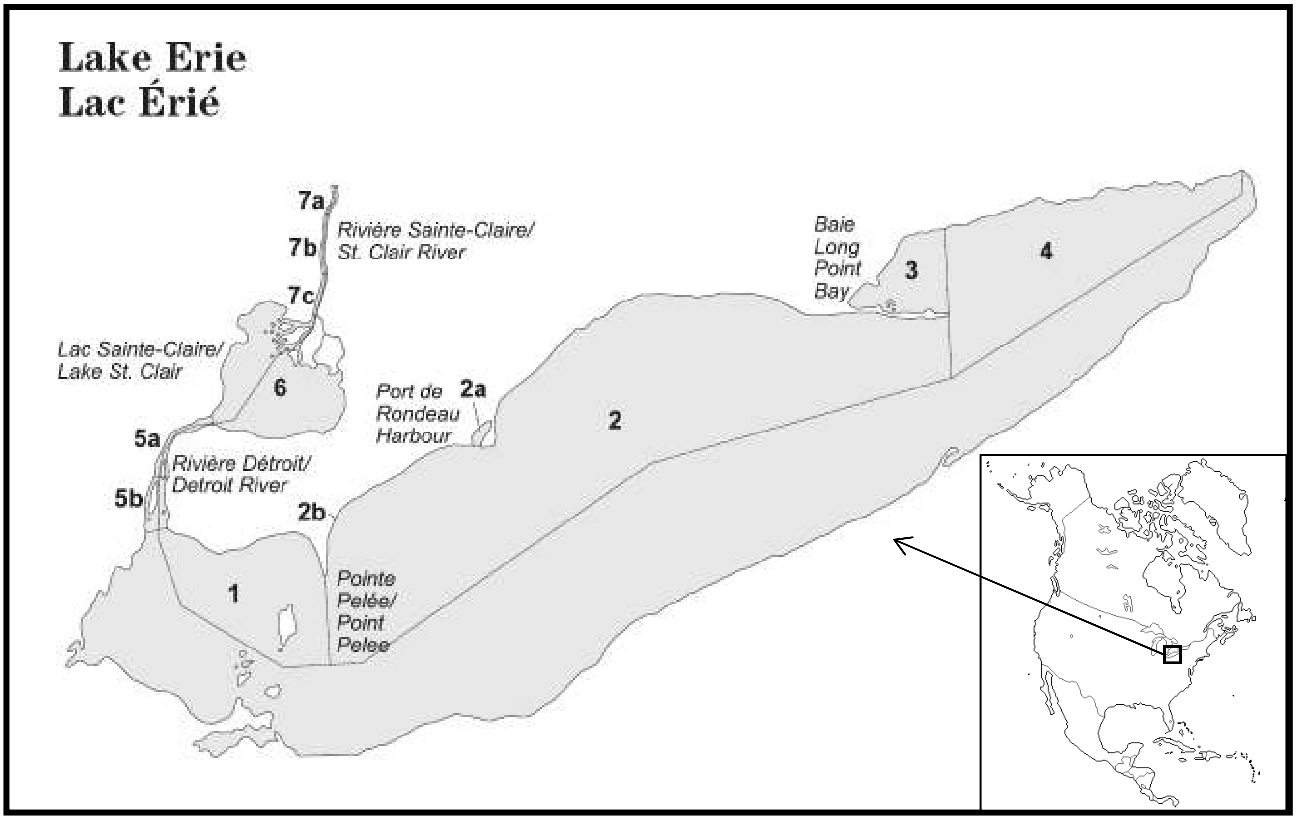
Lake Erie
Lac Érié
7a
7b
7c
Rivière Sainte-Claire/
St. Clair River
Lac Sainte-Claire/
Lake St. Clair
6
5a
Rivière Détroit/
Detroit River
5b
1
2b
Pointe
Pelée/
Point
Pelee
Port de
Rondeau
Harbour
2a
2
Baie
Long
Point
Bay
3
4

Figure 1

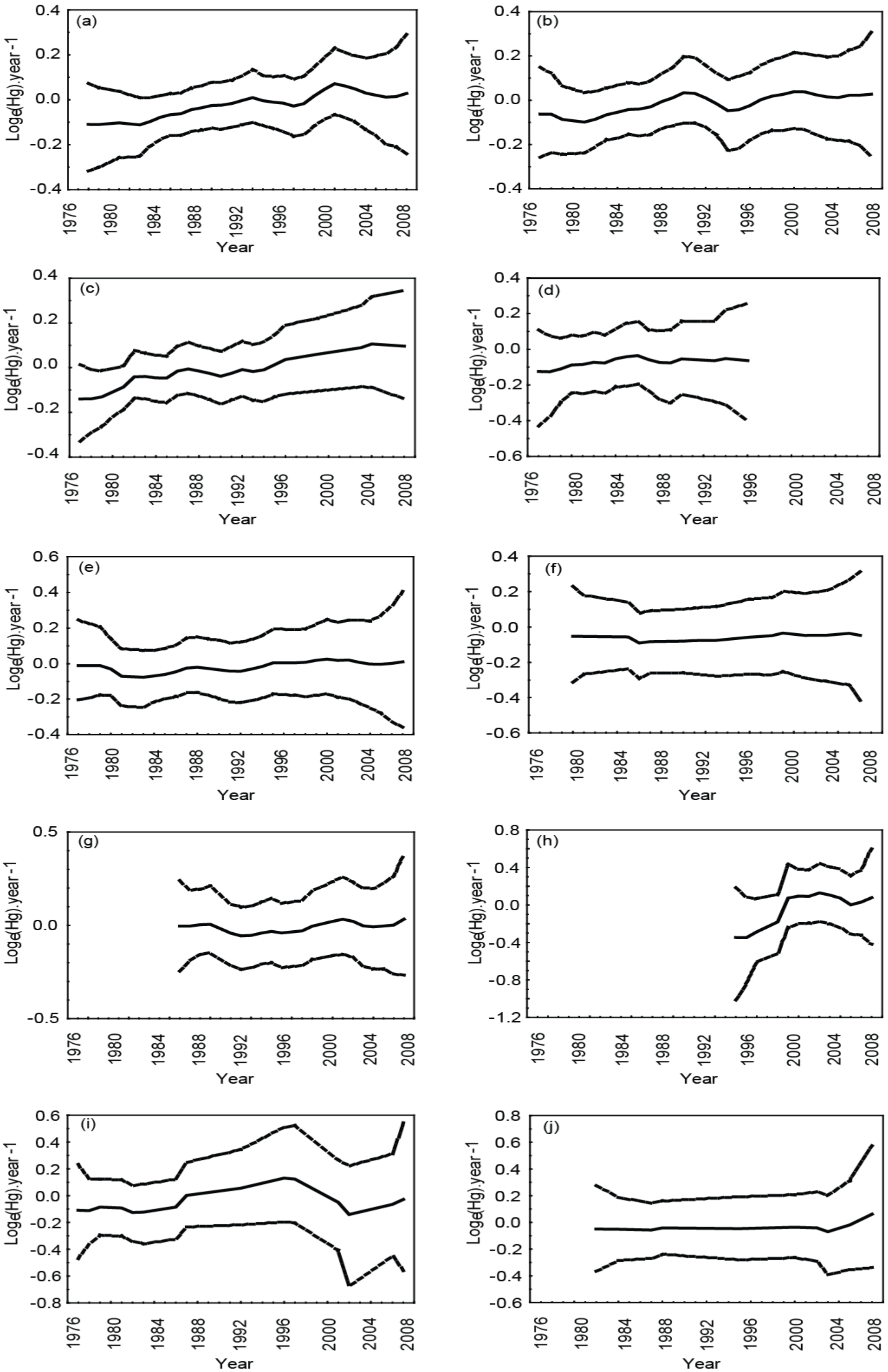
(a)
0.4
0.2
0.0
-0.2
-0.4
Loge(Hg).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(b)
0.4
0.2
0.0
-0.2
-0.4
Loge(Hg).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(c)
0.4
0.2
0.0
-0.2
-0.4
Loge(Hg).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(d)
0.4
0.2
0.0
-0.2
-0.4
-0.6
Loge(Hg).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(e)
0.6
0.4
0.2
0.0
-0.2
-0.4
Loge(Hg).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(f)
0.4
0.2
0.0
-0.2
-0.4
-0.6
Loge(Hg).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(g)
0.5
0.0
-0.5
Loge(Hg).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(h)
0.8
0.4
0.0
-0.4
-0.8
-1.2
Loge(Hg).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(i)
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
Loge(Hg).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(j)
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
Loge(Hg).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year

Figure 2

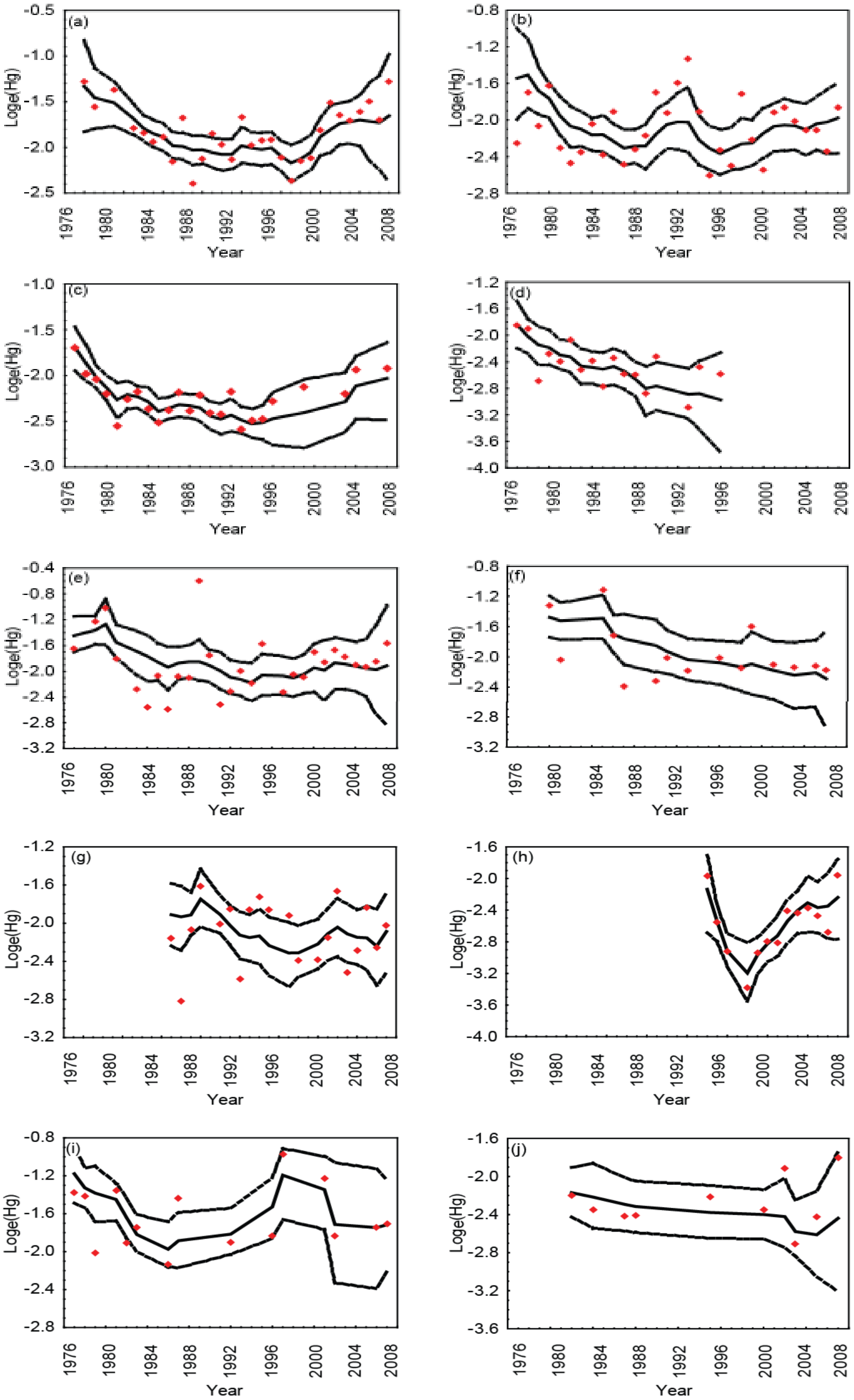(a)
(b)
(c)
(d)
(e)
(f)
(g)
(h)
(i)
(j)
Loge(Hg)
Year
1976
1980
1984
1988
1992
1996
2000
2004
2008
-0.5
-1.0
-1.5
-2.0
-2.5
-0.8
-1.2
-1.6
-2.0
-2.4
-2.8
-1.0
-1.5
-2.0
-2.5
-3.0
-1.2
-1.6
-2.0
-2.4
-2.8
-3.2
-3.6
-4.0
-0.4
-0.8
-1.2
-1.6
-2.0
-2.4
-2.8
-3.2

Figure 3

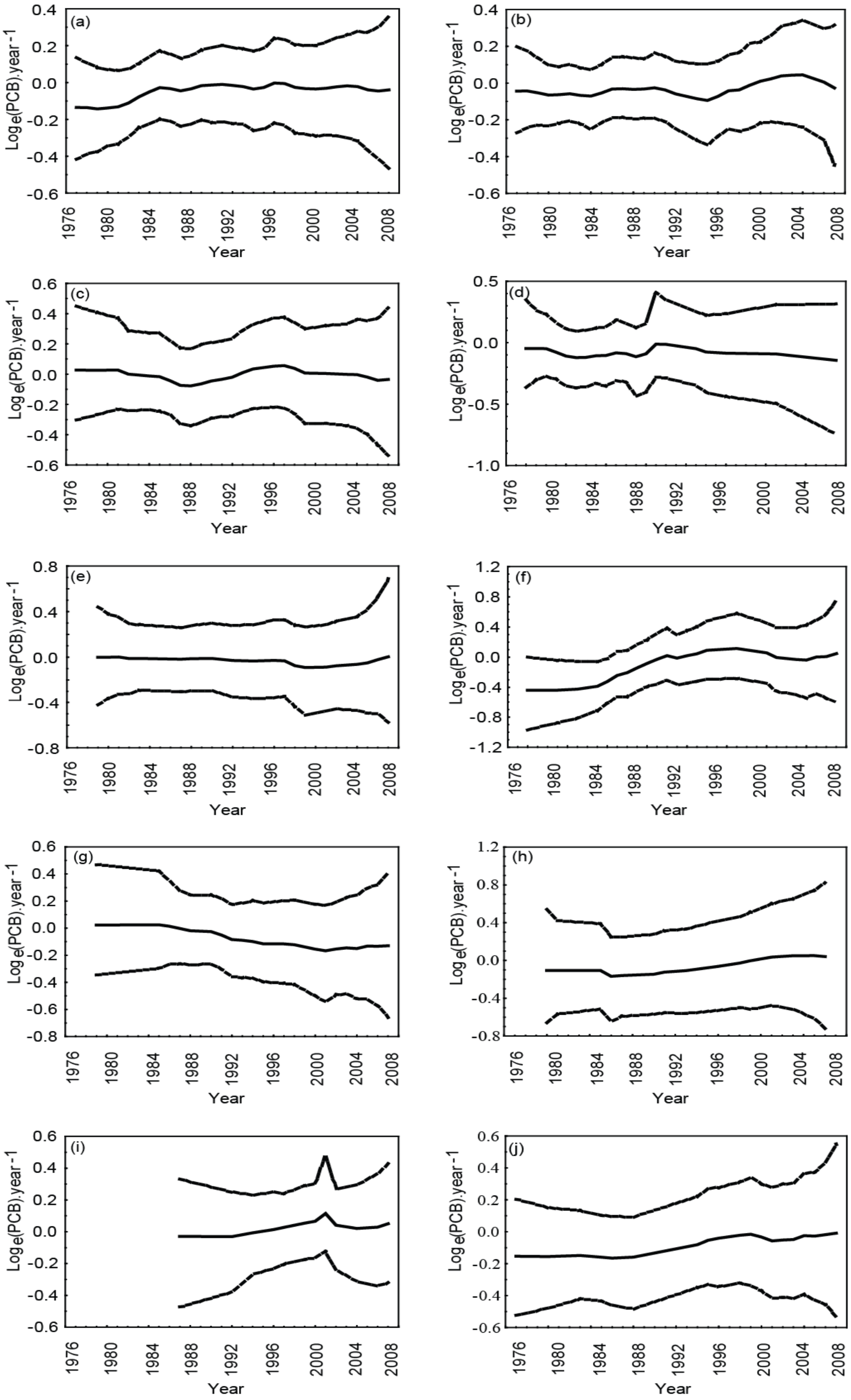
(a)
0.4
0.2
0.0
-0.2
-0.4
-0.6
Loge(PCB).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(b)
0.4
0.2
0.0
-0.2
-0.4
-0.6
Loge(PCB).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(c)
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
Loge(PCB).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(d)
0.5
0.0
-0.5
-1.0
Loge(PCB).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(e)
0.8
0.4
0.0
-0.4
-0.8
Loge(PCB).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(f)
1.2
0.8
0.4
0.0
-0.4
-0.8
-1.2
Loge(PCB).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(g)
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
Loge(PCB).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(h)
1.2
0.8
0.4
0.0
-0.4
-0.8
Loge(PCB).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(i)
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
Loge(PCB).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year
(j)
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
Loge(PCB).year-1
1976
1980
1984
1988
1992
1996
2000
2004
2008
Year

Figure 4

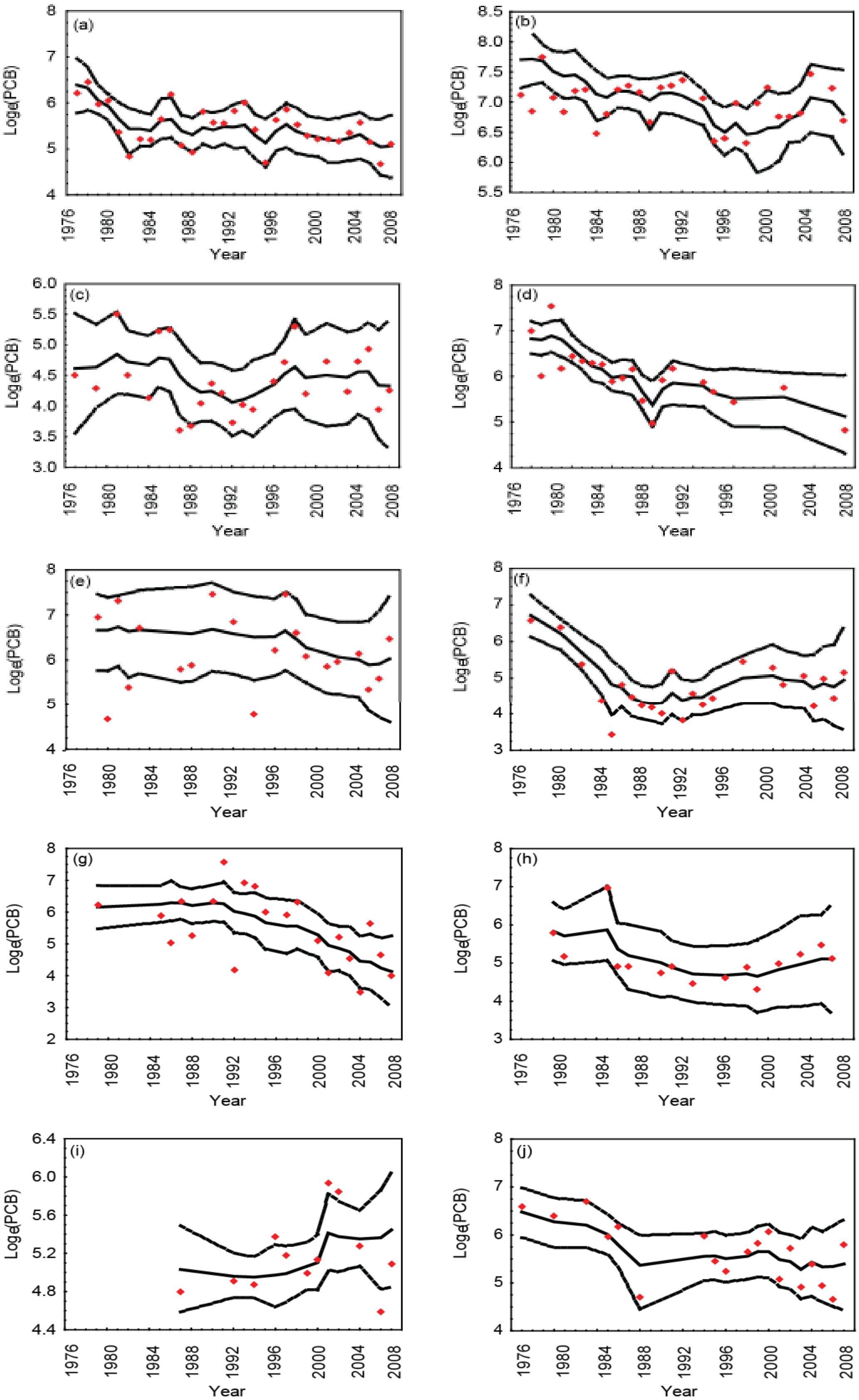
(a)
(b)
(c)
(d)
(e)
(f)
(g)
(h)
(i)
(j)
Loge(PCB)
Year
1976
1980
1984
1988
1992
1996
2000
2004
2008
4
5
6
7
8
5.5
6.0
6.5
7.0
7.5
8.0
8.5
3.0
3.5
4.0
4.5
5.0
5.5
6.0
2
3
4.4
4.8
5.2
5.6
6.0
6.4

Figure 5

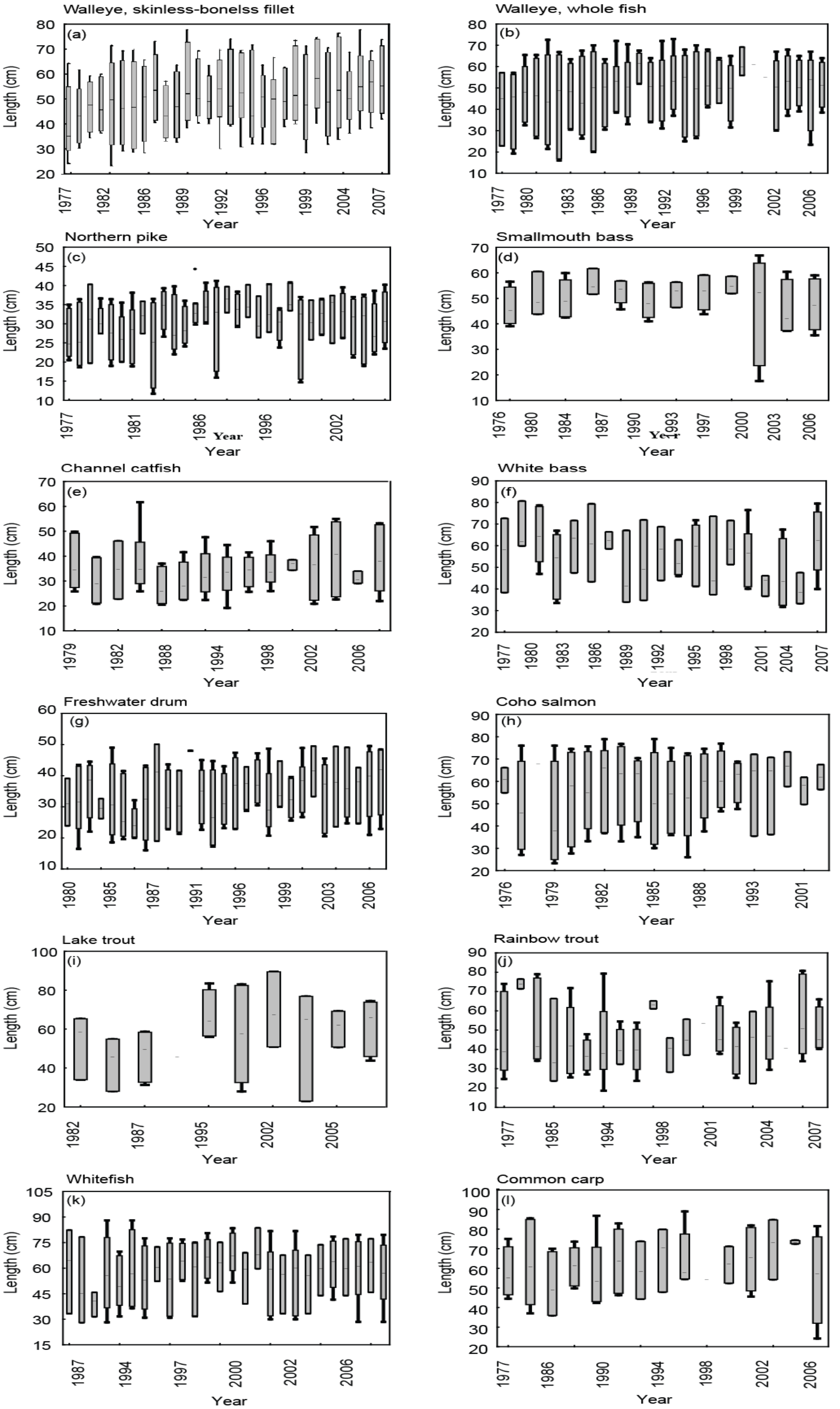
Walleye, skinless-bonelss fillet
(a)
Length (cm)
20
30
40
50
60
70
80
1977
1982
1986
1989
1992
1996
1999
2004
2007
Year
Walleye, whole fish
(b)
Length (cm)
10
20
30
40
50
60
70
80
1977
1980
1983
1986
1989
1992
1996
1999
2002
2006
Year
Northern pike
(c)
Length (cm)
10
15
20
25
30
35
40
45
50
1977
1981
1986
1996
2002
Year
Year
Smallmouth bass
(d)
Length (cm)
10
20
30
40
50
60
70
1976
1980
1984
1987
1990
1993
1997
2000
2003
2006
Year
Year
Channel catfish
(e)
Length (cm)
10
20
30
40
50
60
70
1979
1982
1988
1994
1998
2002
2006
Year
White bass
(f)
Length (cm)
20
30
40
50
60
70
80
90
1977
1980
1983
1986
1989
1992
1995
1998
2001
2004
2007
Year
Freshwater drum
(g)
Length (cm)
10
20
30
40
50
60
1980
1985
1987
1991
1996
1999
2003
2006
Year
Coho salmon
(h)
Length (cm)
20
30
40
50
60
70
80
90
1976
1979
1982
1985
1988
1993
2001
Year
Lake trout
(i)
Length (cm)
20
40
60
80
100
1982
1987
1995
2002
2005
Year
Rainbow trout
(j)
Length (cm)
10
20
30
40
50
60
70
80
90
1977
1985
1994
1998
2001
2004
2007
Year
Whitefish
(k)
Length (cm)
15
30
45
60
75
90
105
1987
1994
1997
2000
2002
2006
Year
Common carp
(l)
Length (cm)
20
30
40
50
60
70
80
90
100
1977
1986
1990
1994
1998
2002
2006
Year

Figure 6

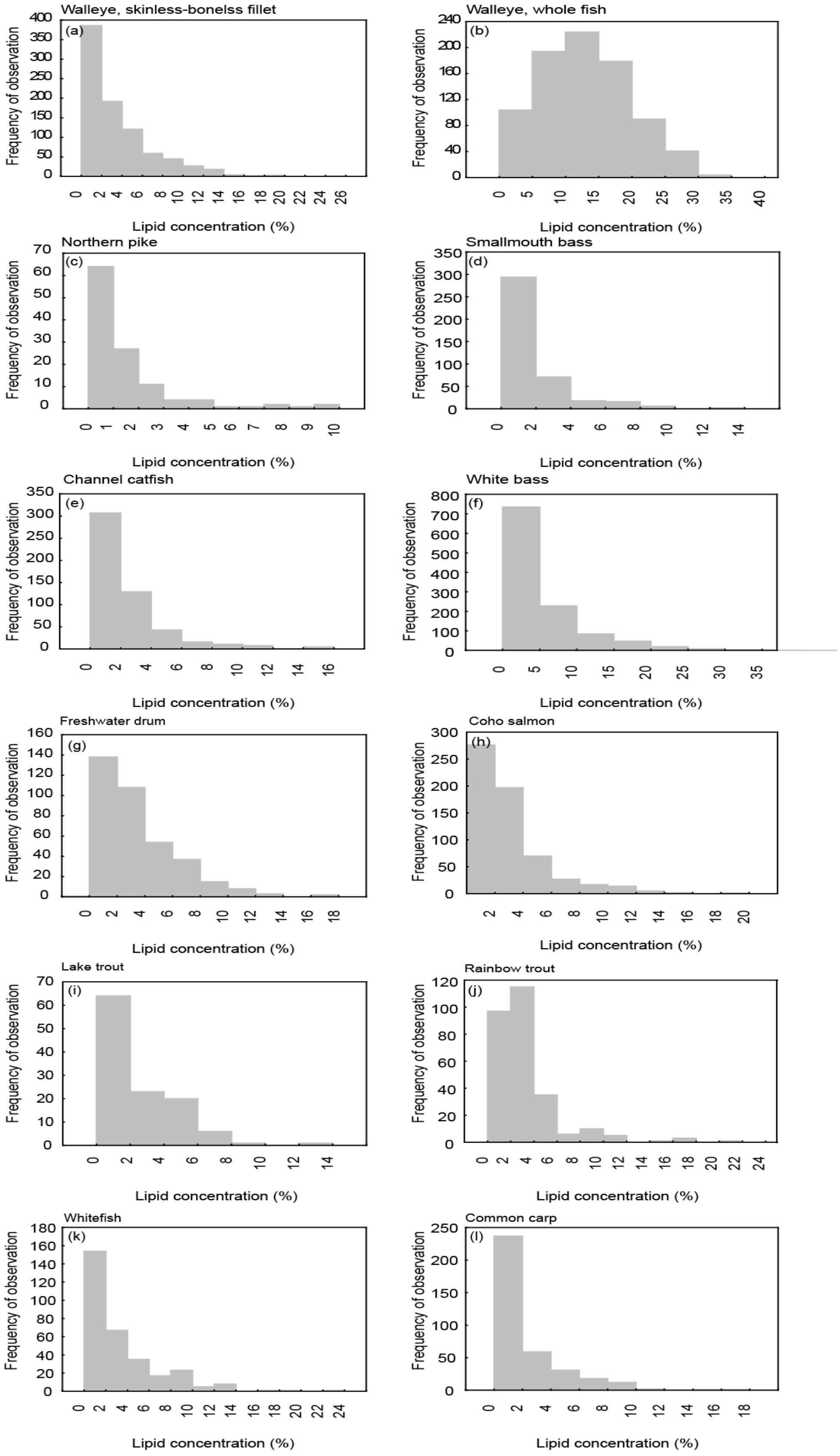
Walleye, skinless-bonelss fillet
(a)
Frequency of observation
0
50
100
150
200
250
300
350
400
0 2 4 6 8 10 12 14 16 18 20 22 24 26
Lipid concentration (%)
Walleye, whole fish
(b)
Frequency of observation
0
40
80
120
160
200
240
0 5 10 15 20 25 30 35 40
Lipid concentration (%)
Northern pike
(c)
Frequency of observation
0
10
20
30
40
50
60
70
0 1 2 3 4 5 6 7 8 9 10
Lipid concentration (%)
Smallmouth bass
(d)
Frequency of observation
0
50
100
150
200
250
300
350
0 2 4 6 8 10 12 14
Lipid concentration (%)
Channel catfish
(e)
Frequency of observation
0
50
100
150
200
250
300
350
0 2 4 6 8 10 12 14 16
Lipid concentration (%)
White bass
(f)
Frequency of observation
0
100
200
300
400
500
600
700
800
0 5 10 15 20 25 30 35
Lipid concentration (%)
Freshwater drum
(g)
Frequency of observation
0
20
40
60
80
100
120
140
160
0 2 4 6 8 10 12 14 16 18
Lipid concentration (%)
Coho salmon
(h)
Frequency of observation
0
50
100
150
200
250
300
2 4 6 8 10 12 14 16 18 20
Lipid concentration (%)
Lake trout
(i)
Frequency of observation
0
10
20
30
40
50
60
70
0 2 4 6 8 10 12 14
Lipid concentration (%)
Rainbow trout
(j)
Frequency of observation
0
20
40
60
80
100
120
0 2 4 6 8 10 12 14 16 18 20 22 24
Lipid concentration (%)
Whitefish
(k)
Frequency of observation
0
20
40
60
80
100
120
140
160
180
0 2 4 6 8 10 12 14 16 18 20 22 24
Lipid concentration (%)
Common carp
(l)
Frequency of observation
0
50
100
150
200
250
0 2 4 6 8 10 12 14 16 18
Lipid concentration (%)

Figure 7

Table S1: Summary statistics of THg concentrations (ng/g wet weight) for skinless-boneless fillet data for nine fish species in four locations in Lake Erie. Missing years across all sites (not site-specific) are also identified.

Species	Central Basin	Eastern Basin	Long Point Bay	Western Basin
Walleye (SBF) *Stizostedion vitreum*				
Count	*519*	*415*	*121*	*784*
Mean	0.154	0.227	0.256	0.205
Standard Deviation	0.099	0.133	0.169	0.135
Missing years:	*1978, 1980 (Range: 1976- 2007)*			
Smallmouth Bass *Micropterus dolomieui*				
Count	*6*	*74*	*469*	*165*
Mean	0.159	0.234	0.158	0.190
Standard Deviation	0.043	0.152	0.117	0.118
Missing years:	*1978, 1989, 1982, 1996 (Range: 1977- 2007)*			
Lake Trout *Salvelinus namaycush*				
Count	*0*	*126*	*0*	*0*
Mean	-	0.118	-	-
Standard Deviation	-	0.047	-	-
Missing years:	*1983, 1985, 1986, 1988, 1989, 1990, 1991, 1992, 1993, 1994, 1996, 1997, 1998, 1999, 2001, 2004, 2006 (Range: 1982-2007)*			
Coho Salmon *Oncorhynchus kisutch*				
Count	*193*	*82*	*194*	*261*
Mean	0.087	0.104	0.113	0.100
Standard Deviation	0.039	0.046	0.045	0.063
Missing years:	*1978, 1991, 1992, 1995 (Range: 1977-1996)*			
Common Carp *Cyprinus carpio*				
Count	*134*	*3*	*127*	*115*
Mean	0.141	0.15	0.162	0.159
Standard Deviation	0.084	0.114	0.083	0.078
Missing years:	*1989, 1996, 1999 (Range: 1985-2007)*			
White Bass *Morone chrysops*				
Count	*602*	*248*	*241*	*560*
Mean	0.155	0.118	0.123	0.158
Standard Deviation	0.114	0.085	0.068	0.107
Missing years:	*None (Range: 1977-2007)*			

Rainbow Trout				
Oncorhynchus mykiss	*116*	*81*	*36*	*0*
Count	0.099	0.074	0.127	-
Mean	0.044	0.045	0.041	-
Standard Deviation				
Missing years:	*1997 (Range: 1994-2007)*			
Northern Pike				
Esox lucius				
Count	*92*	*30*	*164*	*0*
Mean	0.243	0.298	0.212	-
Standard Deviation	0.164	0.189	0.138	-
Missing years:	*1980, 1984, 1985, 1988, 1989, 1990, 1991, 1993, 1994, 1995, 1996, 1998, 1999, 2000, 2003, 2004, 2005 (Range: 1977-2007)*			
Freshwater Drum				
Aplodinotus grunniens				
Count	*213*	*70*	*177*	*161*
Mean	0.141	0.265	0.205	0.150
Standard Deviation	0.120	0.200	0.158	0.114
Missing years:	*1982, 1983, 1984, 1988, 1989, 1992, 1994, 1995, 1997, 2000, 2002, 2004 (Range: 1980-2006)*			

Table S2: Summary statistics of PCB concentrations (ng/g wet weight) for skinless-boneless fillet data for nine fish species in four locations in Lake Erie. Missing years across all sites (not site-specific) are also identified.

Species	Central Basin	Eastern Basin	Long Point Bay	Western Basin
Walleye (SBF) *Stizostedion vitreum*				
Count	*254*	*271*	*75*	*299*
Mean	91.319	109.266	111.36	139.883
Standard Deviation	100.286	105.524	111.329	141.489
Missing years:	*1978, 1980, 1983, 1995, 2000, 2002 (Range: 1977-2007)*			
Channel Catfish *Ictalurus Punctatus*				
Count	*199*	*6*	*83*	*268*
Mean	805.487	483.333	507.446	1397.160
Standard Deviation	973.337	487.552	678.577	1360.96
Missing years:	*1984, 1985, 1986, 1989, 1991, 1993, 1995, 1998, 2000, 2003, 2005 (Range: 1979- 2007)*			
Coho Salmon *Oncorhynchus kisutch*				
Count	*173*	*74*	*194*	*243*
Mean	456.618	373.486	494.284	498.165
Standard Deviation	236.099	246.651	257.228	266.570
Missing years:	*1978, 1991, 1992, 1995, 1997, 1998, 1999, 2000, 2002, 2003, 2004, 2005, 2006 (Range: 1976- 2007)*			
Common Carp *Cyprinus carpio*				
Count	*134*	*3*	*117*	*120*
Mean	580.829	46.667	500.376	1195.558
Standard Deviation	1000.425	30.551	1094.992	1244.84
Missing years:	*1980, 1981, 1982, 1983, 1984, 1989, 1996, 1999 (Range: 1979-2007)*			
Freshwater Drum *Aplodinotus grunniens*				
Count	*138*	*20*	*96*	*106*
Mean	230.493	460.6	193.146	240.358
Standard Deviation	205.581	757.668	232.198	262.912
Missing years:	*1982, 1983, 1984, 1988, 1989, 1992, 1994, 1995, 1997, 2000, 2002, 2004 (Range: 1980- 2006)*			
Rainbow Trout *Oncorhynchus mykiss*				
Count	*124*	*76*	*83*	*19*
Mean	382.048	183.263	600.542	498

Standard Deviation	226.975	128.804	347.848	212.2249854
Missing years:	*1978, 1979, 1981, 1982, 1984, 1987, 1989, 1990, 1991, 1992, 1993, 1997 (Range: 1977-2007)*			
Smallmouth Bass *Micropterus dolomieui*				
Count	*6*	*28*	*297*	*87*
Mean	97.833	255.536	150.380	552.368
Standard Deviation	43.398	217.024	178.506	509.973
Missing years:	*1978, 1980, 1982, 1995, 1996, 1998, 1999, 2002 (Range: 1976-2007)*			
White Bass *Morone chrysops*				
Count	*418*	*152*	*158*	*437*
Mean	306.110	190.309	260.69	371.417
Standard Deviation	239.654	184.25	223.809	257.322
Missing years:	None (Range: 1977-2007)			
Whitefish *Coregonus Clupeaformis*				
Count	*92*	*104*	*0*	*154*
Mean	407.848	155.740	-	213.325
Standard Deviation	530.005	95.305	-	166.371
Missing years:	*1988, 1989, 1990, 1991, 1993, 1995, 1998, 2003, 2005 (Range: 1987-2007)*			

Dynamic Linear Modeling

The WinBUGS codes associated with the dynamic linear model for the total mercury concentrations in walleye skinless boneless fillet data is as follows:

```
model {
# Specification of the observation equation
for (i in 1:N) {
LogHgm[i]<-level[time[i]+1]+beta[time[i]+1]*length[i]
LogHg[i]~dnorm(LogHgm[i],mtau[time[i]+1])
LogPredHg[i]~dnorm(LogHgm[i],mtau[time[i]+1])
PredHg[i]<-exp(LogPredHg[i])}
# Specification of the system equations for the second year until the end of the study period
for (t in 2:29) {
beta[year[t]]~dnorm(beta[year[t-1]],btau[year[t]])
rate[year[t]]~dnorm(rate[year[t-1]],gtau[year[t]])
levelm[year[t]]<-level[year[t-1]]+rate[year[t]]
level[year[t]]~dnorm(levelm[year[t]],ltau[year[t]])
# Specification of the discount factors for the second year until the end of the study period
ltau[year[t]]<-ltau.in*pow(0.95,year[t]-1)
lsigma[year[t]]<-sqrt(1/ltau[year[t]])
btau[year[t]]<-btau.in*pow(0.95,year[t]-1)
bsigma[year[t]]<-sqrt(1/btau[year[t]])
gtau[year[t]]<-gtau.in*pow(0.95,year[t]-1)
gsigma[year[t]]<-sqrt(1/gtau[year[t]])
mtau[year[t]]<-mtau.in*pow(0.95,year[t]-1)
msigma[year[t]]<-sqrt(1/mtau[year[t]])
}
# Specification of the system equations for the first year
beta[year[1]]~dnorm(beta[1],btau[year[1]])
rate[year[1]]~dnorm(rate[1],gtau[year[1]])
levelm[year[1]]<-level[1]+growth[year[1]]
level[year[1]]~dnorm(levelm[year[1]],ltau[year[1]])
# Specification of the discount factors for the first year
ltau[year[1]]<-ltau.in*pow(0.95,year[1]-1)
lsigma[year[1]]<-sqrt(1/ltau[year[1]])
btau[year[1]]<-btau.in*pow(0.95,year[1]-1)
bsigma[year[1]]<-sqrt(1/btau[year[1]])
gtau[year[1]]<-gtau.in*pow(0.95,year[1]-1)
gsigma[year[1]]<-sqrt(1/gtau[year[1]])
mtau[year[1]]<-mtau.in*pow(0.95,year[1]-1)
msigma[year[1]]<-sqrt(1/mtau[year[1]])
# Prior distributions for the parameters of the first year
beta[1]~dnorm(0,0.0001)
rate[1]~dnorm(0,0.0001)
level[1]~dnorm(0,0.0001)
ltau.in~dgamma(0.001,0.001)
```

```
ltau[1]<-ltau.in
btau.in~dgamma(0.001,0.001)
btau[1]<-btau.in
gtau.in~dgamma(0.001,0.001)
gtau[1]<-gtau.in
mtau.in~dgamma(0.001,0.001)
mtau[1]<-mtau.in

}
```

Inference Data

```
list(N=1839,
year=c(2,4,6,7,8,9,10,11,12,13,14,15,16,17,18,19,20,21,22,23,24,25,26,27,28,29,30,31,32),
time=c(paste time.dat here),
LogHg=c(paste walleyeHg.dat here),
length=c(paste length.dat here),
```

Initial values 1

```
list(beta=c(0,0,NA,0,NA,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0),
     rate=c(0,0,NA,0,NA,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0),
     level=c(0,0,NA,0,NA,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0),
     mtau.in=0.2, ltau.in=1, btau.in=1, gtau.in=1,
     LogPredHg=c(paste walleyeHg.dat here))
```

Initial values 2

```
list(beta=c(1,1,NA,1,NA,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1),
     rate=c(1,1,NA,1,NA,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1),
     level=c(1,1,NA,1,NA,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1,1),
     mtau.in=0.32, ltau.in=0.32, btau.in=0.32, gtau.in=0.32,
     LogPredHg=c(paste walleyeHg.dat here)
```