Ecological Informatics 18 (2013) 131–148

Contents lists available at ScienceDirect

Ecological Informatics

journal homepage: www.elsevier.com/locate/ecolinf

Fish contamination in Lake Erie: An examination of temporal trends of organochlorine contaminants and a Bayesian approach to consumption advisories

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ARTICLE INFO

Article history:
Received 25 February 2013
Received in revised form 3 July 2013
Accepted 1 August 2013
Available online 9 August 2013

Keywords:
Organochlorines
Bayesian inference
Dynamic linear modeling
Lake Erie
Fish contamination
Consumption advisories

ABSTRACT

When examining environmental levels of organic contaminants, much of our focus has been placed on fish due to their greater potential to bioaccumulate and their direct linkage with human as a staple of their diet. Contaminant levels in Great Lakes fish communities have been closely monitored over the last few decades, and the resulting information has been indispensable in guiding consumption advisories. In this study, we first conducted an analysis of temporal trends of three organochlorines (hexachlorobenzene, octachlorostyrene, and α-hexachlorocyclohexane) in five Lake Erie fish species using dynamic linear modeling, while explicitly considering fish length and lipid content as covariates. Our results indicate that the levels of the three compounds have been steadily decreasing during the late 1970s to 2007, although there were instances in which the fish organochlorine contents exhibited fluctuations through time. The second part of our analysis focused on the development of a Bayesian framework to update fish consumption advisories. We present a methodology that incorporates the uncertainty in contaminant predictions and the natural variability in fish length and lipid content, while remaining flexible for different human weights and diet patterns. We then illustrate our Bayesian framework for two important contaminants in the Great Lakes region, mercury and PCBs. We established thresholds for each contaminant based on the tolerable daily intake (TDI) values and made predictive statements about the probability of exceedance of these critical levels. Our study also discusses how failure to account for model uncertainty/error can have profound implications for the credibility of the predictive risk assessment statements derived. The proposed Bayesian approach to fish consumption advisories can serve as a valuable framework for year-specific, highly customizable risk assessment statements that also account for the inherent variability in natural systems.

1. Introduction

The ecological health of the Laurentian Great Lakes declined during the 20th century as a result of extensive anthropogenic activities, leading to issues such as impairment of the resilience of native fish communities, widespread eutrophication, and the ubiquitous presence of toxic chemicals (Johnson et al., 1999). The highly populated and industrialized nature of the surrounding watersheds combined with the long residence times of the receiving waterbodies made the Great Lakes highly susceptible to anthropogenic disturbances (DeVault et al., 1996; Johnson et al., 1999). Growing concerns about the deteriorating quality of the system led to the ratification of the cross-border Great Lakes Water Quality Agreement in 1972 (revised in 1978), aiming to restore the "integrity of the waters," especially through the reduction of harmful contaminants (IJC, 1978; Johnson et al., 1999). Even though the curtailment of external emissions generally led to pollutant declines, the persistent nature of contaminants was translated into lingering concentrations within the aquatic food web, particularly in top predators (Bhavsar et al., 2007, 2008; Carlson et al., 2010).

Fish communities have historically been used as ecosystem health indicators, given their trophic position in aquatic food webs and their critical link to human consumers (Bentzen et al., 1999). Since the 1970s, contaminant levels in fish have been routinely monitored in the Great Lakes, with the resulting information being used to determine fish consumption advisories (e.g., Bhavsar et al., 2011; OMOE, 2011). However, despite the valuable insights gained into contaminant dynamics through the extensive datasets developed, many studies fail to consider important causal factors that can influence the perceived spatiotemporal trends, such as fish age, size, trophic level, growth and lipid content (Sadraddini et al., 2011a, 2011b; Stow et al., 1997). Variations across monitoring programs in the type of sampling procedures and the different statistical methods used may also impede the robust assessment of contaminant trends (Bhavsar et al., 2007, 2010; Carlson et al., 2010). It is thus essential to strive for more flexible statistical frameworks when undertaking such retrospective analyses, in order to ascertain that the actual contaminant trends are being revealed.

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http://dx.doi.org/10.1016/j.ecoinf.2013.08.001

To this end, a central feature of recent work in Lake Erie was the adoption of Bayesian inference techniques as a means for critically assessing spatiotemporal contaminant trends in fish communities over the last four decades (Azim et al., 2011a, 2011b; Sadraddini et al., 2011a, 2011b). The advantage of the Bayesian approach when addressing ecological questions primarily stems from its ability to explicitly accommodate model structural and parametric uncertainty (Arhonditsis et al., 2007; Dorazio and Johnson, 2003; Ellison, 1996, 2004; Sommerfreund et al., 2010). In particular, temporal trends of total mercury (THg) in Lake Erie fish were first evaluated using Bayesian configurations of the single exponential, double exponential, and mixed-order decay models to assess the presence and magnitude of statistically significant THg trends (Azim et al., 2011a). This analysis revealed instances of species-specific increase in THg concentrations in recent years, suggesting a causal association with the changes in the trophodynamics induced by the invasion of round gobies and dreissenid mussels into the system. A similar study on the polychlorinated biphenyl (PCB) concentrations using exponential decay models indicated nearly monotonic declining or sometimes stabilizing trends across the study period, with the main exception being the recent rise in the walleye PCB levels (Sadraddini et al., 2011a). To discern whether these walleye trends are still manifested if we explicitly account for fish length as a covariate, a follow-up study by Sadraddini et al. (2011b) utilized a dynamic linear modeling (DLM) analysis. It was found that the increasing walleye trend disappeared when using length-corrected predictions, and was thus a reflection of the biases introduced by the local sampling procedures (Sadraddini et al., 2011b). These results reinforce the necessity of accounting for potentially important causal factors when conducting trend analyses, and also highlight the usefulness of DLMs as robust hindcasting tools.

In this study, we present a two-pronged Bayesian DLM approach to address the issue of lingering contaminants in fish and their potential impacts on human consumers. In the first step, we complete our modeling work in Lake Erie by examining temporal contaminant trends of three organochlorines: 1) hexachlorobenzene, a persistent and bioaccumulative pesticide that severely impacts humans and wildlife (ATSDR, 2002); 2) octachlorostyrene, a persistent by-product of industrial processes (CGLI, 1999; Norheim and Roald, 1985); and 3) α-hexachlorocyclohexane (α-HCH), a dominant congener in the banned pesticide technical-HCH (ATSDR, 2005). The second part of this paper aims to broaden our scope and cohesively link together the entire body of the work conducted in Lake Erie to date; our attention is shifted towards relating the derived spatiotemporal contaminant trends to the application of fish consumption advisories. The task of establishing a general framework for fish consumption advisories is a challenging process, given the wide array of both known and unknown factors that can conceivably shape the detected contaminant trends. We also emphasize the issue of model uncertainty that has been profoundly neglected by various fish consumption advisory frameworks. In this regard, we propose a Bayesian DLM strategy that is suitable to explicitly account for all sources of uncertainty, such as model adequacy, parametric uncertainty as well as sampling bias and variability in fish characteristics. In this study, we illustrate the capacity of the proposed approach to develop comprehensive advisories by generating customizable risk statements of the probability of exceedance of critical THg and PCB levels in the human body through the consumption of fish of different lengths and lipid contents.

2. Methods

2.1. Organochlorine trends in Lake Erie fish

Our study used fish contaminant data from the Ontario Ministry of the Environment (OMOE) 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 filet (SBF) portions. This information is then used to guide biennial fish consumption advisories. In our analysis, we selected fish species based on data availability and/or the species' commercial importance. For each contaminant, we examined five fish species: channel catfish (*Ictalurus punctatus*), common carp (*Cyprinus carpio*), coho salmon (*Oncorhynchus kisutch*), rainbow trout (*Oncorhynchus mykiss*) and white bass (*Morone chrysops*). All samples were collected from various locations on the Canadian side of Lake Erie and correspond to a time span of approximately three decades (1976–2007). The collected samples were analyzed at the OMOE Toronto laboratories, where their organochlorine concentrations were determined through gas liquid chromatography-electron capture detection (GLC-ECD), in accordance with OMOE method PFAOC-E3136 (see OMOE, 2007).

2.1.1. Modeling framework

We developed a series of *DLM*s to examine temporal trends of the three organochlorine compounds. We explicitly account 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 (Gewurtz et al., 2011a,b). To compare the relative influence of each of these covariates, we ran the *DLMs* for each congener-fish species combination a total of four times: without any covariates ("random walk"), using the fish length or lipid content alone or both fish length and lipid content as covariates. We thus ran a total of 60 models (5 fish species × 3 compounds × 4 covariate combinations) over the course of this study. Unlike static regression models that have fixed parameters, *DLM*s 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 inter-annual 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, *DLM*s 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. For the sake of brevity, we outline the model for hexachlorobenzene that considers both fish length and lipid content as covariates:

Observation equation:

$$\ln[HCB]_{ti} = level_t + \beta_{t1} \ \ln[length]_{ti} + \beta_{t2} \ \ln[lipid]_{ti} + \psi_{ti} \quad \psi_{ti} \sim N[0, \Psi_t] \tag{1}$$

System equations:

$$level_t = level_{t-1} + rate_t + \omega_{t1} \qquad \omega_{t1} \sim N[0, \Omega_{t1}] \tag{2}$$

$$rate_t = rate_{t-1} + \omega_{t2} \quad \omega_{t2} \sim N[0, \Omega_{t2}] \tag{3}$$

$$\beta_{t1} = \beta_{t1-1} + \omega_{t3} \qquad \omega_{t3} \sim N[0, \Omega_{t3}] \tag{4}$$

$$\beta_{t2} = \beta_{t2-1} + \omega_{t4} \qquad \omega_{t4} \sim N[0, \Omega_{t4}] \tag{5}$$

$$\begin{array}{ll} 1/\Omega_{tj}^2 = \zeta^{t-1} \cdot 1/\Omega_{1j}^2, \ \ 1/\Psi_t^2 = \zeta^{t-1} \cdot 1/\Psi_1^2 & t>1 \ \ and \ \ j = 1 \ \ to \ \ 4 \\ level_1, \ \ rate_1, \ \ \beta_1 \sim N(0, 10000) & t = 1 \\ 1/\Omega_{1j}^2, \ \ 1/\Psi_1^2 \sim G(0.001, 0.001) & \end{array} \tag{6}$$

where $ln[HCB]_{ti}$ is the observed *HCB* concentration at time t in the individual sample i; $level_t$ is the mean *HCB* 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 and ω_{tj} are the error terms for year t sampled from normal distributions with zero mean and variances Ψ_t^2 and Ω_{tj}^2, respectively; the discount factor ζ represents the aging of information with the passage of time; $N(0, 10{,}000)$ 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 $1/\Psi_1^2$ 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 discounting 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. The likelihood of bias due to multiple measurements below the detection limit was considered using a Tobit dynamic linear modeling approach (Amemiya, 1973). Specifically, our model used a bounded distribution for the measurements, where the upper bound was equal to either the detection limit or a very large (arbitrary) number, depending on whether the measurement fell below the detection limit or not (Fig. 1; see also the model code in Appendix A). The determination of the most parsimonious model for each fish species/congener combination was based on the use of the Deviance Information Criterion (*DIC*) values, a Bayesian measure of model fit and complexity, where models with

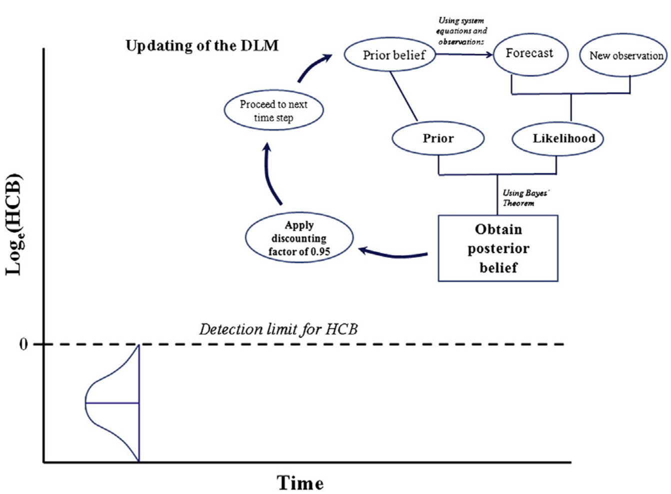

Fig. 1. Basic concepts of Tobit dynamic linear modeling: The sequential flow allows parameter values to vary over time by discounting older observations relative to more recent ones. Values below the detection limit are treated as random draws from a normal distribution parameterized such that 99% of their values are lying within the analytical and actual zero. Our approach uses a bounded distribution for the measurements with an upper bound equal to either the detection limit or a very large (arbitrary) number, depending on whether the measurement falls below the detection limit or not. Thus, the Gibbs sampler samples the observations we have set below the detection limit from the tail of the distribution.

lower *DIC* values are expected to effectively balance between predictive capacity and complexity (Spiegelhalter et al., 2003).

2.1.2. 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. 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). The convergence of the sequences occurred fairly quickly (~1000 iterations) and thus our summary statistics reported are based on the remaining draws. Finally, to ensure the accuracy of our posterior parameter values, we confirmed that the Monte Carlo error for parameters (an estimate of the difference between the true posterior mean and the mean of the sampled values) was less than 5% of the sample standard deviation (Spiegelhalter et al., 2003).

2.2. Statistical framework for fish consumption advisories

The illustration of our Bayesian approach to fish consumption advisories was focused on THg and PCB concentrations in Lake Erie walleye communities, given the high profile of these two contaminants, the popularity of walleye as a sport fish (e.g., Imm et al., 2005), and the consistency of the collected information over time. The proposed strategy involves a DLM framework that incorporates the uncertainty in contaminant predictions and the natural variability in fish length and lipid content, while remaining flexible for different human weights and meal frequencies. We established thresholds for each contaminant based on their tolerable daily intake (TDI) values and were then able to make predictive statements about the probability of exceeding critical levels of that contaminant through consumption of fish of a specific size and lipid content. For the purpose of prediction, it is important to note that the Bayesian approach generates a posterior predictive distribution that represents the current estimate of the value of the response variable (THg and PCB levels in walleye), taking into account both the uncertainty about the parameters and the uncertainty that remains when the parameters are known (Ellison, 2004). Therefore, estimates of the uncertainty of Bayesian model predictions are more realistic (usually larger) than those based on classical procedures. Predictions are expressed as probability distributions, thereby conveying significantly more information than point estimates with regards to uncertainty.

Our analysis is founded upon the THg and PCB dynamic linear models presented by Sadraddini et al. (2011b). We first selected three years in order to examine through our model whether there was a distinct change of the fish contaminant levels over time, e.g., 1986, 1996 and 2006. We then identified a range of human weights and meal patterns to demonstrate the flexibility of our statistical approach to fish consumption advisories. For human weights, we chose 50 kg (lower weight), 75 kg (average weight) and 100 kg (heavier weight), while the fish consumption frequency ranged from one to eight fish meals per month. Similar to the value used by OMOE when producing their established advisories, we used a standard fish meal size of 227 g in our analysis. Our next step was to calculate critical thresholds for each contaminant. The TDI values for THg and total PCBs were obtained from OMOE, which generally receives guidelines from Health Canada and modifies where appropriate (e.g., total PCB). The TDI is defined as the maximum allowable daily intake of a substance that, if consumed over a lifetime, will not lead to adverse health effects (Health Canada, 1996). TDI values are generally expressed for a specific body weight, such as μg per kg of body weight (or kgbw) per day. Specifically, we

Table 1
Summary statistics of hexachlorobenzene concentrations in skinless–boneless filet data (ng/g wet weight) for five fish species in Lake Erie (study period 1976–2007).

Species	N	Mean	SD	Median	IQR	Skewness	Kurtosis	Best model[a]
Channel catfish *Ictalurus punctatus*	533	2.27	2.88	1.00	1.00	4.35	24.18	L + L
Common carp *Cyprinus carpio*	374	3.05	7.64	1.00	1.00	8.44	82.85	L + L
Coho salmon *Oncorhynchus kisutch*	634	2.82	3.7	2.00	2.00	9.32	140.5	L + L
Rainbow trout *Oncorhynchus mykiss*	310	2.16	1.90	1.00	2.00	2.44	6.92	L + L
White bass *Morone chrysops*	1158	1.42	2.32	1.00	0.00	23.13	667.91	L + L

[a] Based on lowest DIC value (LNG = model with length as covariate; LPD = model with lipid as covariate; L + L = model considering both length and lipid as covariates).

used the values of 0.52 μg THg/kgbw per day and 0.09 μg PCB/kgbw per day, and calculated the thresholds for each of the hypothetical scenarios as follows:

$$\text{Threshold} = (\text{human weight [kg]} \times \text{TDI[ng/kgbw/month]})/(\text{meal size} \times \text{meal number}). \tag{7}$$

Having established these critical thresholds for each scenario, our next task was to calculate the corresponding frequency of exceedances, given the predicted contaminant levels for a specific combination of fish length and lipid content.

3. Results and discussion

3.1. Organochlorine temporal trends

For each contaminant, we report the summary statistics of the measured concentrations (ng/g wet weight or ww) of the five fish species examined (Tables 1–3). The high standard deviation values were indicative of the considerable inter- and intra-annual variability in contaminant levels. Further, the positive skewness and kurtosis values suggest right-skewed and leptokurtic distributions; we thus applied a natural log transformation to the data before commencing our DLM analyses. We also found that across all the organochlorines, the most favorable dynamic linear model was the one that considered both fish length and lipid content as covariates (see last columns of Tables 1–3). The only exception was the rainbow trout model for α-hexachlorocyclohexane, in which the use of lipid content as the sole covariate provided the most parsimonious model.

3.1.1. Hexachlorobenzene (HCB)

The persistence of hexachlorobenzene in Great Lakes sediments, biota, and water primarily stems from its chemical stability and high lipophilicity (Burton and Bennett, 1987; Ma et al., 2003; Niimi, 1979), which typically translates into significant levels of biomagnification in fish (Courtney, 1979). HCB is not a naturally occurring compound (ATSDR, 2002) and was primarily used as a fungicide on seed grains, such as wheat and barley (Burton and Bennett, 1987; Courtney, 1979). This pesticide was applied in Canada until 1972, while the U.S. banned its use seven years earlier (Sun et al., 2006). The contaminant also enters the environment as a by-product in the manufacture of several chlorinated solvents (e.g., tetrachloroethylene); other chlorinated compounds (e.g., vinyl chloride); several pesticides, including pentachloronitrobenzene and pentachlorophenol; and with flue gas effluents from municipal incineration (see Bailey, 2001; Courtney, 1979). Because of its persistence and widespread presence, hexachlorobenzene was earmarked as one of 11 “critical Great Lakes pollutants” by the IJC in 1985 (Johnson et al., 1999). Similar regulatory attention worldwide for HCB resulted in global declines in North American and European environments since the 1970s (see Bailey, 2001 for a review) Although not acutely toxic to humans, the substance has been associated with the presence of porphyrins, and elevated concentrations have been measured in human breast milk and adipose tissue (see Burton and Bennett, 1987).

The significant reductions of exogenous HCB discharges have resulted in decreases of the sediment concentrations in Lake Erie over the past two decades (Marvin et al., 2004a). Further, Gewurtz et al. (2010) recently reported that hexachlorobenzene declines in fish from the Lake Huron–Erie corridor, though rates slowed in the 1990s. Our analysis similarly revealed decreasing trends for hexachlorobenzene across the fish species examined. Of the five fish species, the highest average HCB concentrations were recorded in common carp (3.05 ng/g), followed by coho salmon (2.82 ng/g), channel catfish (2.27 ng/g), and rainbow trout (2.16 ng/g) (Table 1). The lowest concentrations of this chemical were observed in white bass (1.42 ng/g). It is also worthwhile to note that the majority of the concentrations in the later sampling years were recorded below the detection limit of 1 ng/g, and thus the median concentrations for four fish species remained at that value; the only exception was coho salmon (median 2 ng/g), a species which was sparsely sampled after the mid-90s and whose median value was thus less impacted by the HCB drop to the detection limit. According

Table 2
Summary statistics of octachlorostyrene concentrations in skinless–boneless filet data (ng/g wet weight) for five fish species in Lake Erie (study period 1981–2007).

Species	N	Mean	SD	Median	IQR	Skewness	Kurtosis	Best model[a]
Channel catfish *Ictalurus punctatus*	497	5.10	8.16	2.00	4.00	4.04	23.71	L + L
Common carp *Cyprinus carpio*	364	5.46	11.85	1.00	4.00	5.21	33.26	L + L
Coho salmon *Oncorhynchus kisutch*	529	2.26	1.58	2.00	2.00	1.85	4.77	L + L
Rainbow trout *Oncorhynchus mykiss*	271	1.57	1.55	1.00	0.00	3.74	16.94	L + L
White bass *Morone chrysops*	1040	1.24	1.16	1.00	0.00	10.10	133.05	L + L

[a] Based on lowest DIC value (LNG = model with length as covariate; LPD = model with lipid as covariate; L + L = model considering both length and lipid as covariates).

Table 3
Summary statistics of α-hexachlorocyclohexane concentrations in skinless–boneless filet data (ng/g wet weight) for five fish species in Lake Erie (study period 1976–2007).

Species	N	Mean	SD	Median	IQR	Skewness	Kurtosis	Best model[a]
Channel catfish *Ictalurus punctatus*	533	2.72	4.63	1.00	1.00	3.86	16.74	L + L
Common carp *Cyprinus carpio*	374	1.3	1.49	1.00	0.00	7.49	64.23	L + L
Coho salmon *Oncorhynchus kisutch*	634	2.86	3.88	1.00	2.00	5.77	56.38	L + L
Rainbow trout *Oncorhynchus mykiss*	308	2.16	2.80	1.00	1.00	3.17	10.95	LPD
White bass *Morone chrysops*	1158	1.39	1.12	1.00	0.00	3.83	18.04	L + L

[a] Based on lowest DIC value (LNG = model with length as covariate; LPD = model with lipid as covariate; L + L = model considering both length and lipid as covariates).

to our DLM analysis, both common carp and rainbow trout demonstrated an increase until the mid-80s and a downward oscillatory pattern thereafter (Fig. 2b,d). The mean annual rates of change of the HCB levels for the two species were weakly positive in the early years and nearly zero ever since (Fig. 3b,d). In particular, the odds that the rate parameter was positive is on average 1.3:1 for common carp and 2.2:1 for rainbow trout until the mid-80s, but fall close to a 50% probability (even odds) for the rest of the study period. [Note that the odds ratio of the rate parameter being above zero in a particular year is the ratio of the probability mass above zero to the mass below zero.] Channel catfish demonstrated an overall decreasing trend, although there were fluctuations in the mean HCB levels through time (Fig. 2a). The rates of change of the annual concentrations for this species remained fairly close to zero throughout the study period (Fig. 3a). Similarly, coho salmon demonstrated minor fluctuations early on in the sampling period, but the mean trends projected after the mid-90s should be interpreted with caution due to the sparse data available (Fig. 2c). The corresponding growth rates for coho began weakly positive but switched to very weakly negative towards the end of the years studied (Fig. 3c). White bass was characterized by relatively stable mean concentrations around the detection limit (Fig. 2e), and the corresponding rates of change hovered around zero (Fig. 3e). On a final note, recent studies suggest that Lake Erie may have switched to a source of HCB to the atmosphere through volatilization of the compound out of the lake (Hoff et al., 1996; Kelly et al., 1991; Marvin et al., 2004b). The latter possibility along with the continued release of HCB as unintended by-product could perhaps explain the fluctuating levels reported in our study; however, HCB is generally not considered to be of concern in current fish consumption advisories (Bhavsar et al., 2011; Gewurtz et al., 2010).

3.1.2. Octachlorostyrene (OCS)

First identified in wildlife of the Netherlands (Koeman et al., 1969; Kuehl et al., 1981; ten Noever de Brauw and Koeman, 1972/73), octachlorostyrene concentrations were detected in fish of the lower Great Lakes during the mid-1970s (Kuehl et al., 1976). The discovery of this compound in environmental systems was initially a perplexing phenomenon, given the apparent lack of evidence for a causal association with anthropogenic activities (Chu et al., 2003; Kaminsky and Hites, 1984). It was eventually deduced that OCS was a by-product formed from high temperature industrial processes involving chlorine, such as the electrolytic production of chlorine gas or magnesium, the chlorination and distillation processes, and the refining and degassing of an aluminum smelt (CGLI, 1999; Kaminsky and Hites, 1984; Norheim and Roald, 1985). The rapid growth of the chlorine industry prior to the 1970s resulted in mounting OCS concentrations in the Great Lakes sediments, but the subsequent shift to metal electrodes in the early 1970s soon resulted in marked declines (see Kaminsky and Hites, 1984 for a review). Within the Lake Erie system (including the Huron–Erie corridor), elevated levels of this persistent and toxic pollutant were observed in fish from around the mouth of the Ashtabula River tributary (Kaminsky and Hites, 1984; Kuehl et al., 1981), while St. Clair River sediments and fish from Lake St. Clair also demonstrated high OCS concentrations (Pugsley et al., 1985; Suns et al., 1985). Octachlorostyrene is a persistent substance (likely due to its chemical structure [Norheim and Roald, 1985]) and studies examining concentration differences between water and fish liver have shown high "bioconcentration and adsorption potential" (Ernst et al., 1984; Pugsley et al., 1985). Furthermore, while our understanding of the eco-toxicology of this pollutant remains unclear, there were instances of increased urinary porphyrins in workers exposed to octachlorostyrene, and some studies suggest OCS may have a half-life twice as long as hexachlorobenzene (see Chu et al., 2003 for a review).

In particular, we found that common carp exhibited the highest concentrations of this contaminant (mean 5.46 ng/g), followed by channel catfish (5.10 ng/g), coho salmon (2.26 ng/g) and rainbow trout (1.57 ng/g) (Table 2). White bass again had the lowest concentrations, with a mean value of 1.24 ng/g. Similar to the pattern reported for the hexachlorobenzene levels, the median values for common carp, rainbow trout and white bass remained at the detection limit of 1 ng/g. Exceptions were the coho salmon (for the reason discussed earlier) and channel catfish (median 2 ng/g), whose OCS concentrations were subjected to wider fluctuations in the later sampling years. It is also important to note that the octachlorostyrene monitoring in our dataset began at various points in the 1980s, and thus the true maxima of this contaminant may have not been captured. Channel catfish, common carp, coho salmon and rainbow trout were characterized by decreasing trends with fluctuations until the mid-1990s, followed by nearly monotonic decline to the detection limit since then (Fig. 4a–d). The corresponding rates of change were negative throughout the study period (Fig. 5a–c), but with slowing decline rates for rainbow trout over time (Fig. 5d). In particular, the odds that the rate parameter was negative are on average 3.0:1 for channel catfish, 4.1:1 for common carp, 3.1:1 for coho salmon, and 5.8:1 for rainbow trout, but fall close to 1.2:1 for rainbow trout during the rest of the study period. Finally, white bass showed a minor increase in the 1980s and stable levels around the detection limit ever since (Fig. 4e). The rates of change for this species reflect the initial peak and subsequent decline in the mid-1980s, followed by practically zero rates until the end of the study period (Fig. 5e). Generally, our analysis suggests decreasing OCS trends through time, which is on par with CGLI's (1999) assertion that the levels of this compound have been substantially declining in the Great Lakes over the past two decades. For example, studies of spottail shiners in the lower Niagara River indicated falling OCS concentrations from the mid-1980s down to the detection limit during the 1990s (see CGLI, 1999 for a review). Similar declines were also reported in fish from the Huron–Erie corridor (Gewurtz et al., 2010). The attention paid to octachlorostyrene as a toxic effluent into the lakes (e.g., St. Clair RAP Team, 2006) along with the aforementioned shift to non-OCS producing metal electrodes is likely to have contributed to the reported decrease of fish OCS levels.

3.1.3. α-Hexachlorocyclohexane (α-HCH)

Highly similar to hexachlorobenzene is our final compound of consideration, hexachlorocyclohexane (HCH), or more specifically, the

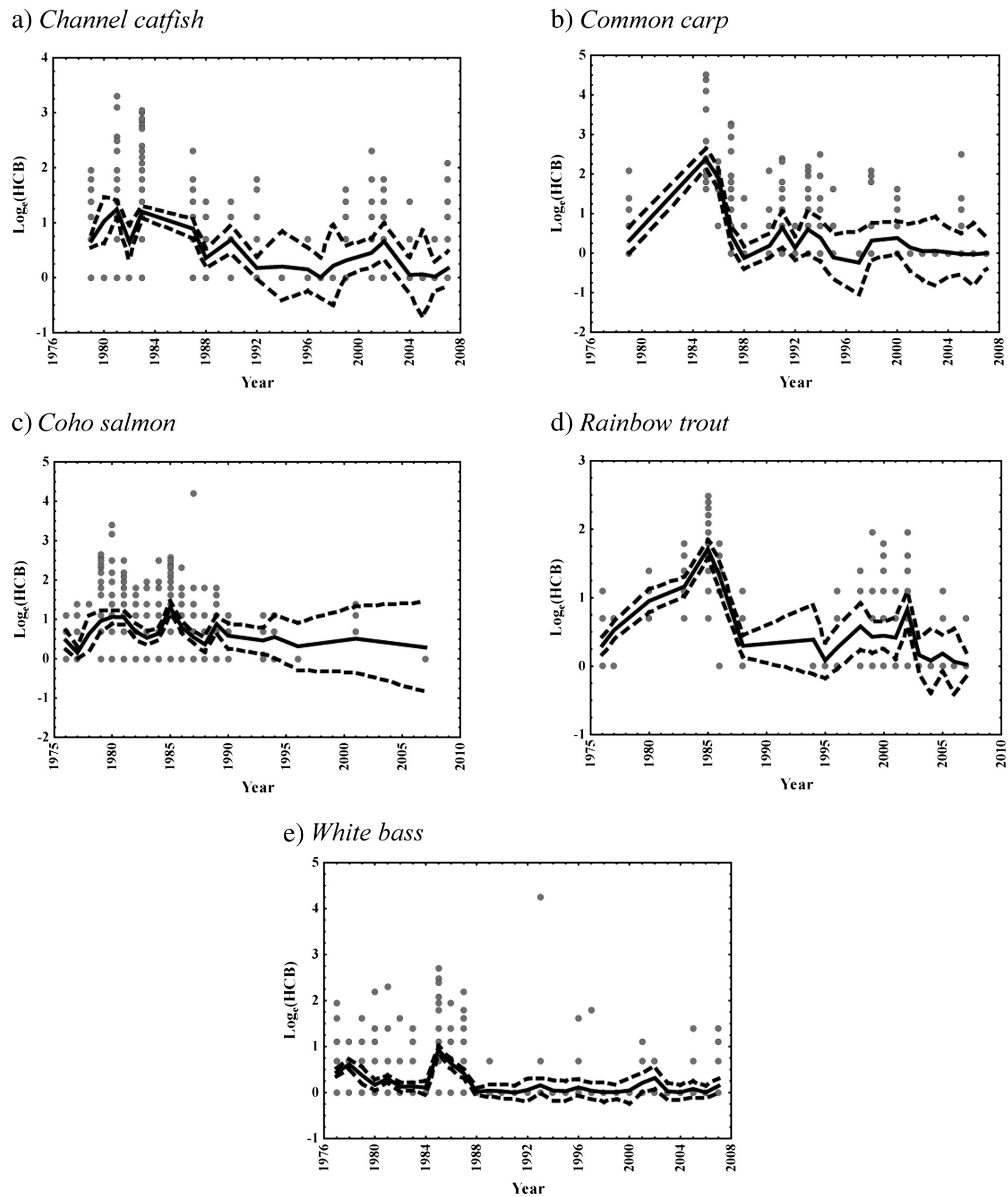

Fig. 2. Dynamic linear modeling analysis depicting the actual hexachlorobenzene concentrations (ng/g wet weight) (gray dots) against the predicted annual hexachlorobenzene trends when accounting for the covariance with the fish length and lipid content (black lines) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1976–2007). The solid and dashed lines correspond to the median and 95% posterior predictive intervals of the *level*$_t$ term in Eq. (1), respectively.

alpha-isomer α-HCH; this substance is often erroneously called "benzenehexachloride" (Willett et al., 1998). Composed of eight isomers, HCH was generally applied as the pesticide technical-HCH (in which the α-congener was dominant) and later as "lindane", which in turn primarily consists of γ-HCH (ATSDR, 2005; Kutz et al., 1991; Safe, 1993). Similar to other persistent organic pollutants, HCH

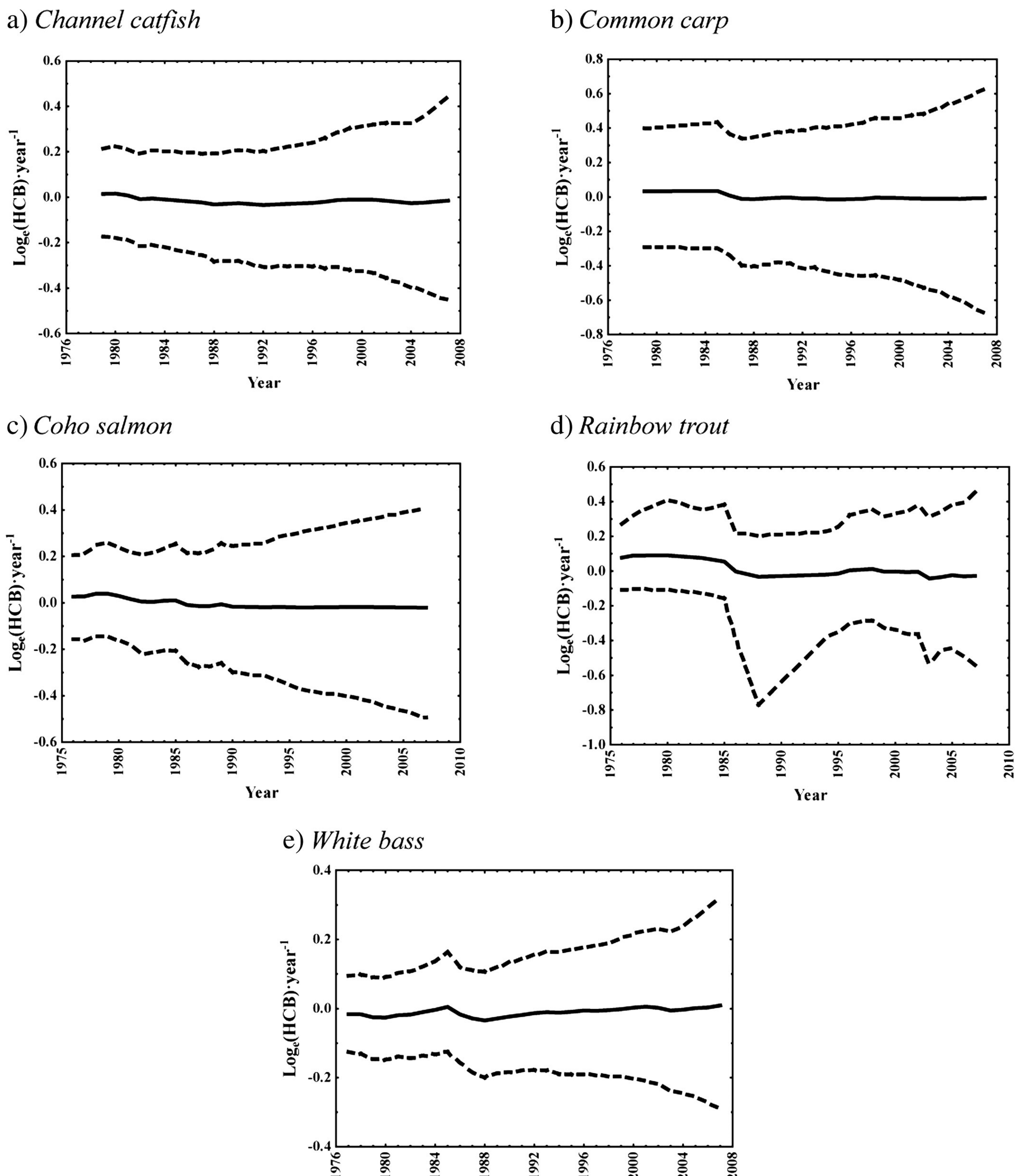

Fig. 3. Dynamic linear modeling analysis depicting the annual rates of change of hexachlorobenzene concentrations (ng/g wet weight) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1976–2007). The solid and dashed lines correspond to the median and the 95% posterior predictive intervals, respectively.

has the potential to bioaccumulate and be transported in long distances (Bhatt et al., 2009). While studies have generally focused more on lindane trends in space and time due to its contemporary use in agricultural practices, concentrations of α-HCH remain in air, precipitation and surface waters, often greater than the γ-isomer (Bhatt et al., 2009). These higher α-HCH environmental levels probably stem from its increased stability relative to γ-HCH and/or the γ-HCH degradation to its α-congener (Easton et al., 2002). Generally, a larger proportion of

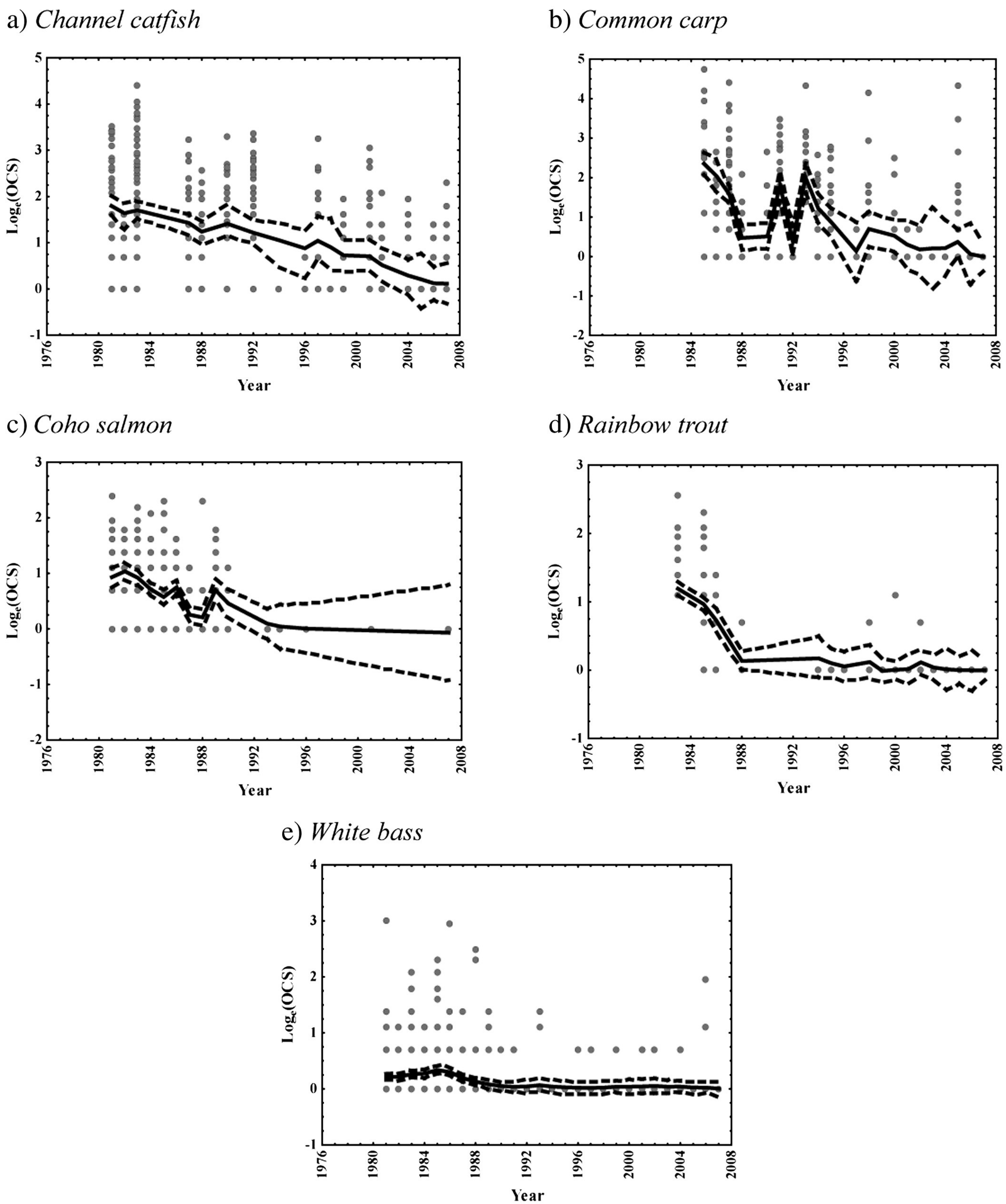

Fig. 4. Dynamic linear modeling analysis depicting the actual octachlorostyrene concentrations (ng/g wet weight) (gray dots) against the predicted annual median octachlorostyrene trends when accounting for the covariance with the fish length and lipid content (black lines) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1981–2007).

α-HCH in environmental media is thought to be an indication of either recent application of technical-HCH or atmospheric deposition, while higher proportions of the beta-isomer (the most stable) would probably indicate distant application (Willet et al., 1998).

In our study, the highest α-HCH concentrations were recorded in coho salmon (2.86 ng/g), followed by channel catfish (2.72 ng/g), rainbow trout (2.16 ng/g), white bass (1.39 ng/g) and common carp (1.3 ng/g) (Table 3). The recent decline down to the detection limit

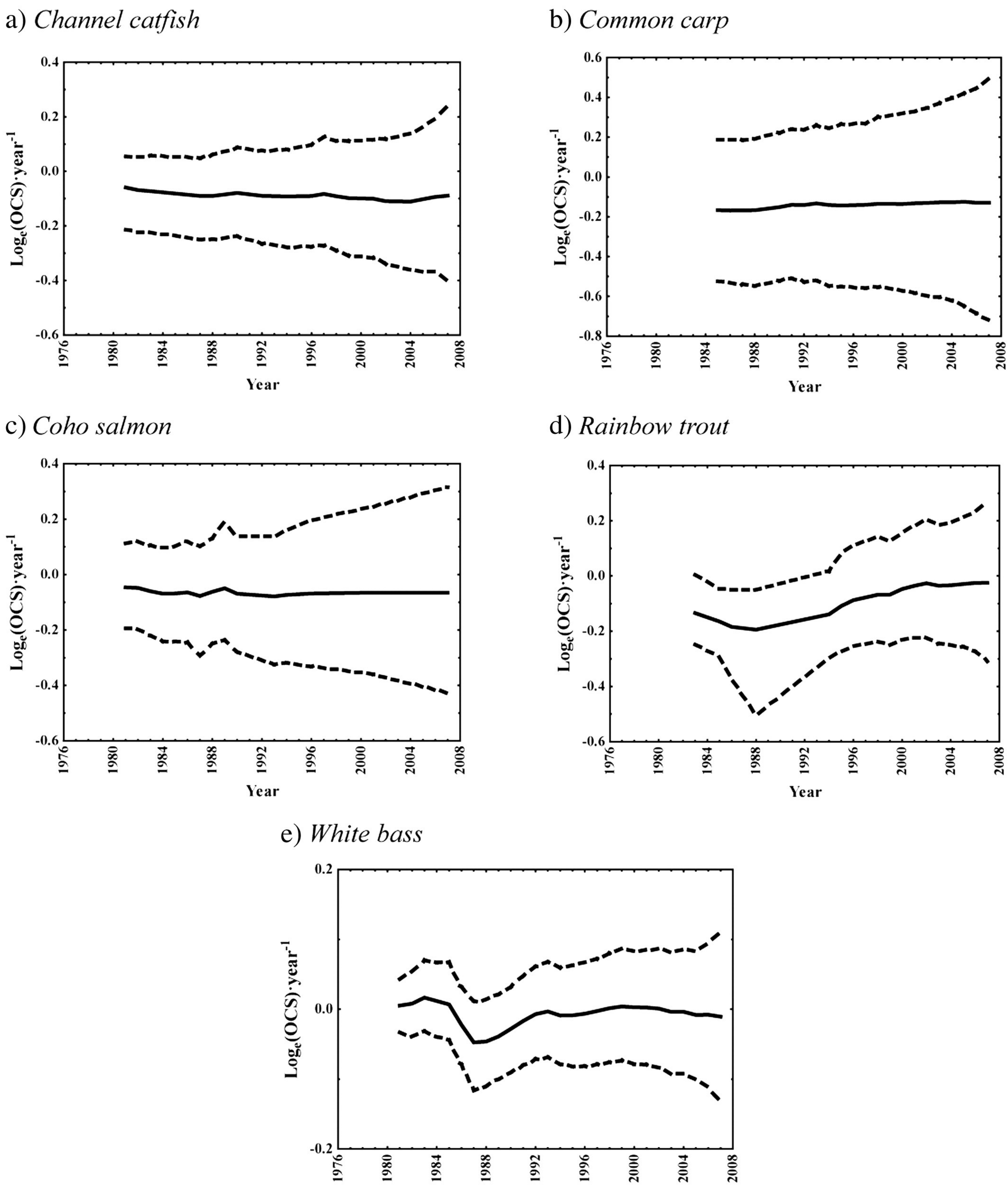

Fig. 5. Dynamic linear modeling analysis depicting the annual rates of change of octachlorostyrene concentrations (ng/g wet weight) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1981–2007).

translated into recurring 1 ng/g median values. Mirroring the patterns of the observed data, the predicted mean α-HCH levels generally showed an initial increase and subsequent decline after the mid-1980s (Fig. 6), with the exception of common carp, which lacked this early peak (Fig. 6b). The rates of change for coho salmon, rainbow trout and white bass switched from weakly positive to negative over time (Fig. 7c–e), while those of channel catfish and common carp started off weakly negative, became slightly more negative in the 1980s and

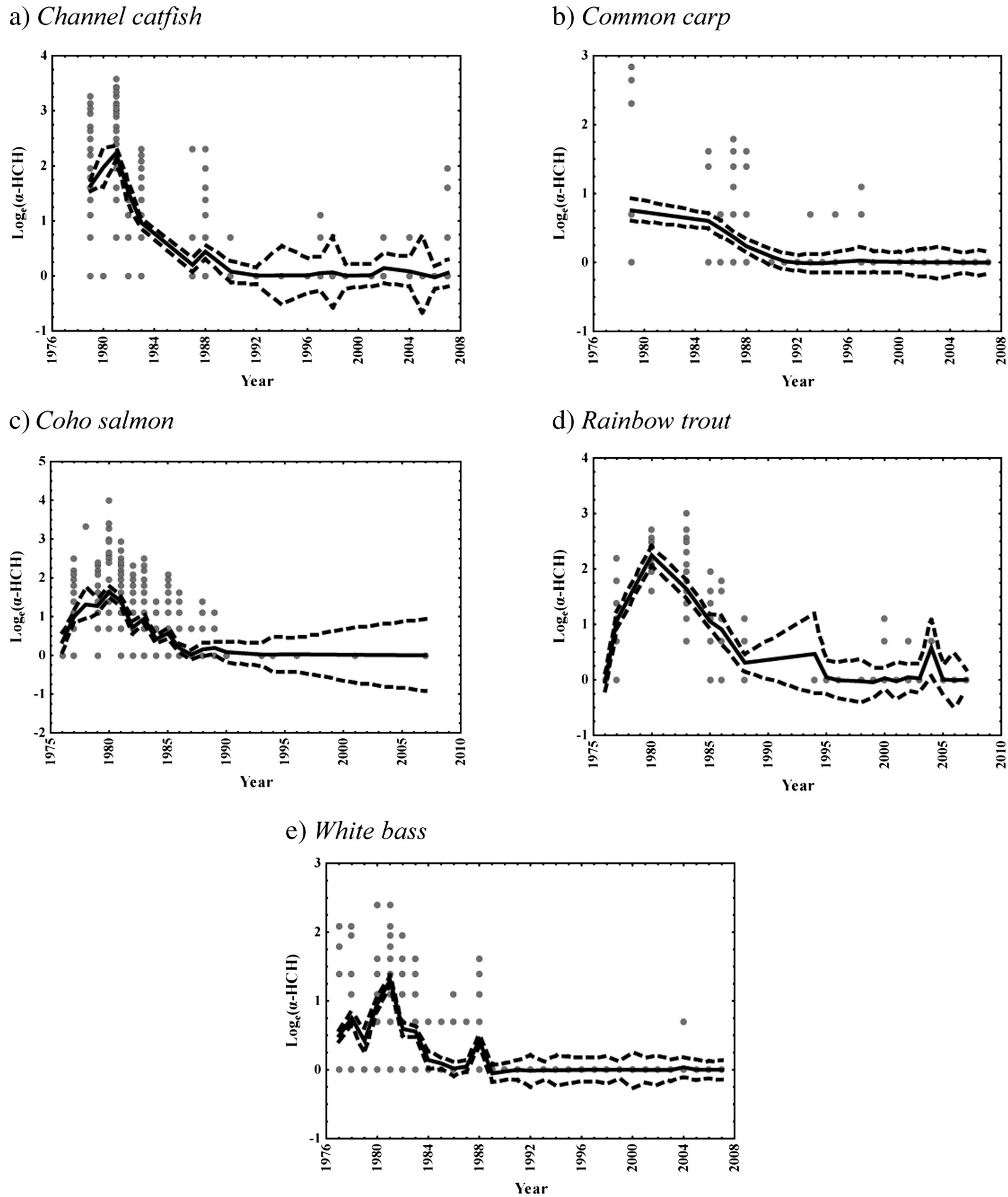

Fig. 6. Dynamic linear modeling analysis depicting the actual α-hexachlorocyclohexane concentrations (ng/g wet weight) (gray dots) against the predicted annual median α-hexachlorocyclohexane trends when accounting for the covariance with the fish length and lipid content (black lines) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1976–2007).

then subsequently slowed to the end of the sampling period (Fig. 7a,b). In particular, we note that the odds that the earlier rates of change were negative are on average 5.7:1 for channel catfish, and 21:1 for common carp, but gradually tend to an even odds ratio after the early 90s. As the use of technical-HCH declined worldwide in response to multiple bans, atmospheric α-HCH concentrations were expected to follow suit (Li

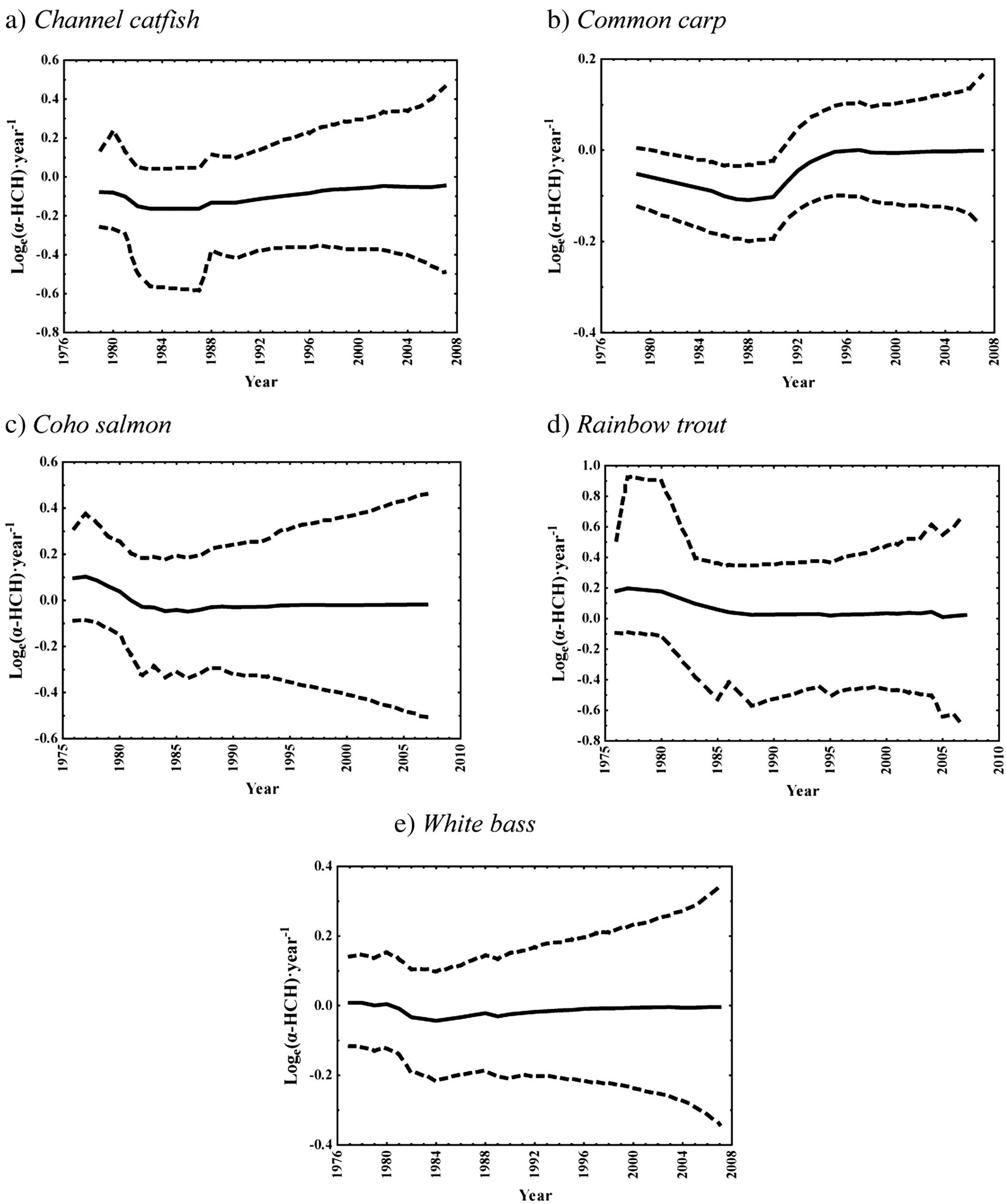

Fig. 7. Dynamic linear modeling analysis depicting the annual rates of change of α-hexachlorocyclohexane concentrations (ng/g wet weight) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1976–2007).

et al., 1998). The α-HCH levels dropped dramatically in Great Lake precipitation levels during the 1990s (Buehler et al., 2002; Chan et al., 2003) and α-HCH has also declined significantly in the sediments of Lake Erie (Marvin et al., 2004a). As such, our declining α-HCH trajectories in the fish species examined are not surprising.

3.2. Framework for fish consumption advisories

3.2.1. Background

The prevalence of persistent, toxic and bioaccumulative substances within the Great Lakes system has been the focal point of numerous legislations and analyses over the past half-century, with concerns raised not only for preserving the ecological health of the waters but also for minimizing the potential ramifications for consumers of local fish. Among the hallmarks of contaminants like PCBs or the pesticide dichlorodiphenyltrichloroethane (DDT) is their high degree of lipophilicity and thus their potential to progressively biomagnify up the food chain (Johnson et al., 1999; Tilden et al., 1997). Even though the emissions of many contaminants have been banned or significantly curtailed, there is often an extended lag time before these results are reflected in the biota (Burger and Gochfeld, 2006). In an effort to protect the public from the harmful impacts of ingested contaminants, fish consumption advisories were instated to encourage voluntary restriction of potentially tainted fish (in a manner that maximizes angler compliance), while also reminding consumers about the benefits of fish consumption (Buchanan et al., 2005; see Tilden et al., 1997 for an overview). The earliest advisories were produced for PCBs in the 1970s (Buchanan et al., 2005) and were gradually expanded to include contaminants such as mercury, chlordane, and DDT (see Burger, 2000). To date, the entire Great Lakes region is under advisories, with the total number in the US increasing by 125% since 1993 (Burger and Gochfeld, 2006; USEPA, 2004). On the Canadian side of the Great Lakes, the OMOE has been regularly issuing biennial fish advisories since the 1970s, with a site- and species-specific approach based on extensive contaminant databases (Bhavsar et al., 2011; OMOE, 2011).

The development of fish consumption advisories differs among regions, although there have been concerted efforts in recent years to establish protocols and region-wide guidelines to ensure consistency. The first step involves the establishment of a reference concentration, representing an estimate of the daily human exposure to a contaminant that will not result in adverse health effects over a lifetime, e.g., USEPA reference dose, health protection value, tolerable daily intake, and minimal risk level (Dourson and Clark, 1990; HPTF, 2004). Calculation of these reference levels may consider uncertainty factors to account for the extrapolation of toxicity data from animals to humans or to accommodate different tolerance levels in humans (USEPA, 2000). The next step in developing advisory guidelines is to identify standard values for fish meal sizes, human weights, amount of contaminants remaining in fish after cooking, frequency of consumption, and cancer risk factors (GLC, 2007; WVITC, 2007). Depending on the amount/quality of data and the region characteristics, the production of the advisories may be based on regression models, mean/median concentrations or frequency distributions (GLC, 2007). Regression models are used to relate contaminant data to the size of various fish species, postulating a relationship between the two variables across the fish sizes sampled, while the selection of the best-fit model (e.g., original or logarithmic scale) is typically based on the coefficient of determination (r^2) values (GLC, 2007). If no relationship exists with the fish length or the data are inadequate to conduct regression analysis, data pooling may be used to obtain species-specific average concentrations (GLC, 2007).

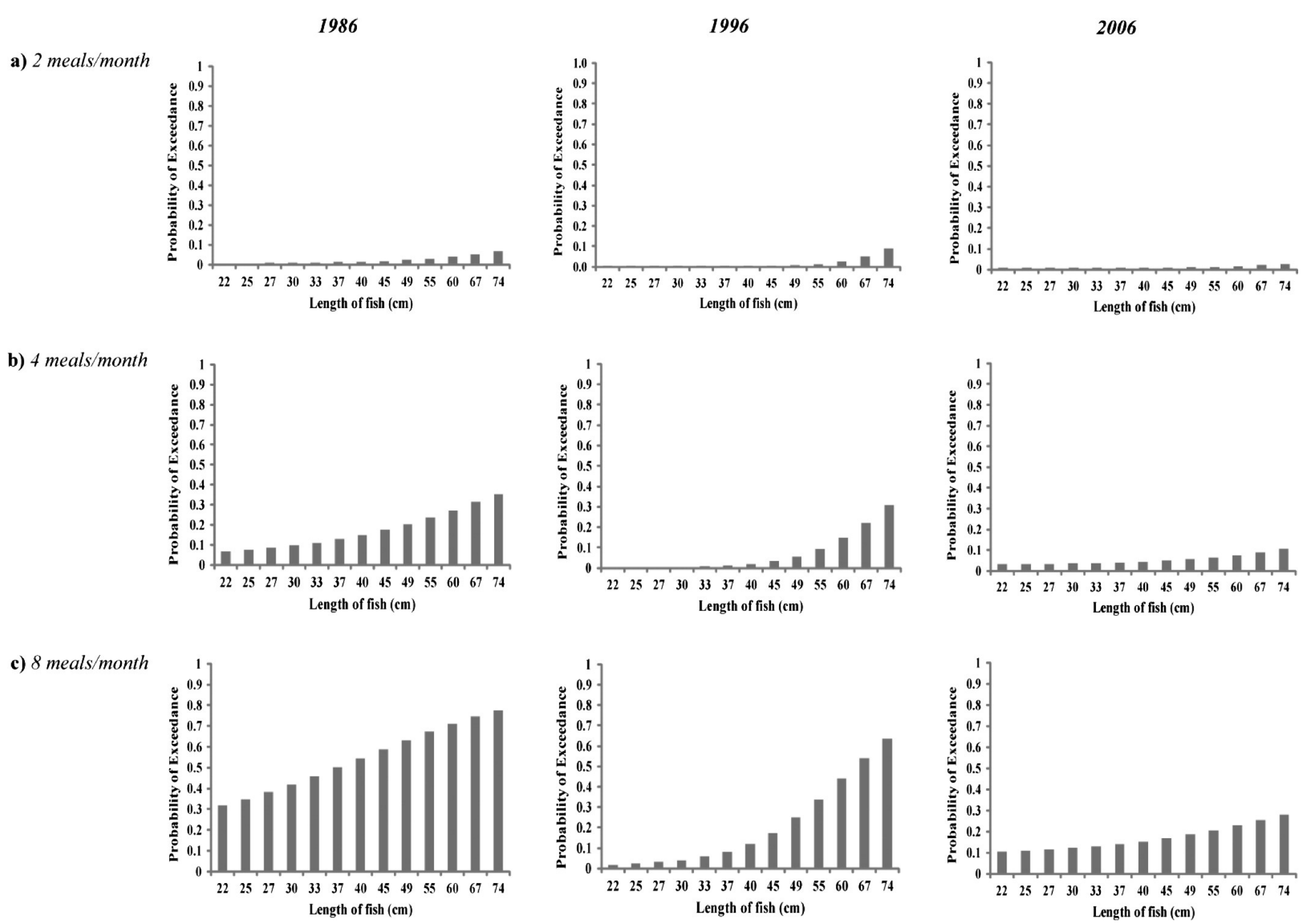

Fig. 8. Probability of exceedance of the PCB tolerable daily intake through the consumption of walleye from a 75-kg person: 2, 4, and 8 fish meals/month. Results are presented for different fish lengths consumed (with the lipid content set equal to 1.2%) in 1986, 1996, and 2006.

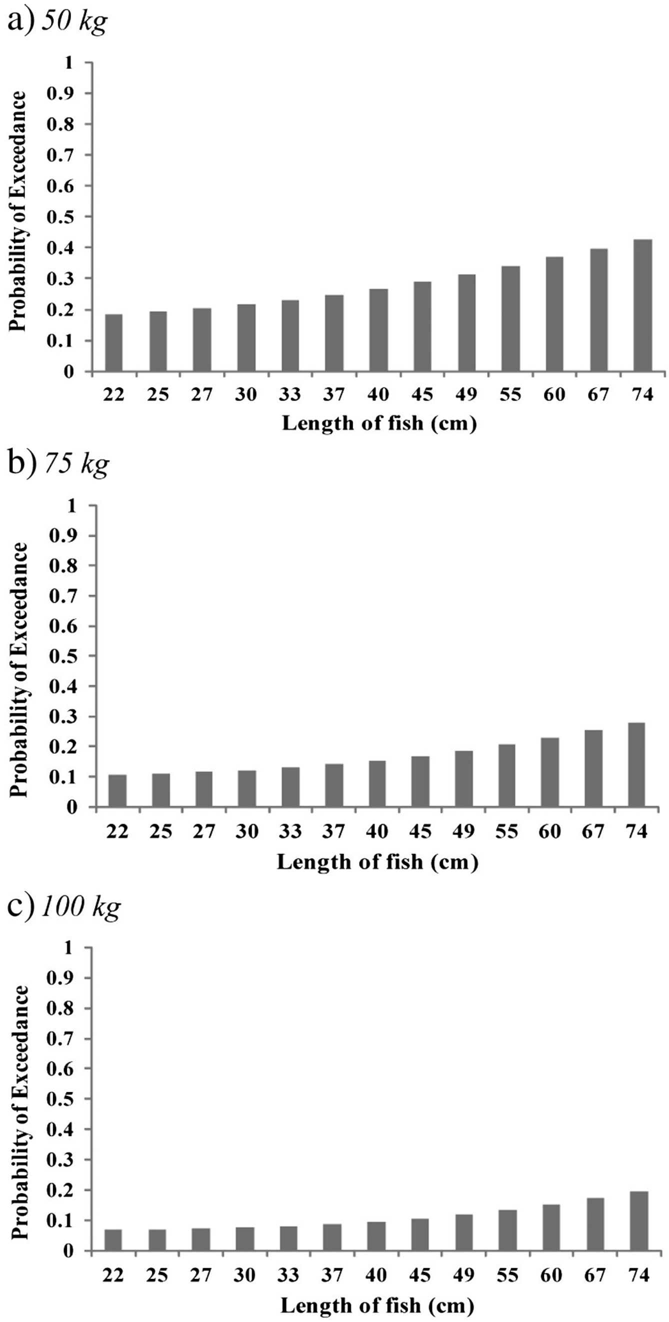

Fig. 9. Probability of exceedance of the *PCB* tolerable daily intake through the consumption of walleye from different human weight categories: 50, 75, and 100 kg. Results are presented for different fish lengths (with the standard lipid content of 1.2%) and a fixed consumption of 8 meals per month in 2006.

Further, despite the aversion of stakeholders and decision makers when confronted with a "range" of values instead of a "fixed" value (Hope et al., 2007; Tannenbaum et al., 2003), there has been a gradual emergence of probabilistic methods in the risk assessment paradigm (e.g., Antonijevic et al., 2007; see Bilau et al., 2007; Harris and Jones, 2008; Roberts et al., 2007; Wilson et al., 2001; Zhang et al., 2009), due to their ability to accommodate the associated uncertainty or more faithfully depict the risk of "outliers" in a fish population (Johnston and Snow, 2007; Sioen et al., 2008).

3.2.2. Bayesian approach to fish consumption advisories

Our analysis is conceptually on par with the aforementioned shift towards probabilistic advisory frameworks in the context of fish consumption advisories. Compared with the conventional regression modeling practices underlying fish consumption advisories, our DLM approach has five distinct features: (i) the models have an evolving structure that allows parameters to vary over time; (ii) 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; as such, the year-specific predictive fish contaminant distributions are conditioned upon prior and current information, not by subsequent data; (iii) instead of using annual average concentrations, the long-term fish contaminant trends are based on individual samples to explicitly accommodate both intra- and interannual variability; (iv) the Bayesian nature of the framework allows both parametric uncertainty and structural error (model misfit) to be reflected in the model predictions; and (v) bearing in mind that the apparent error rate (sensu Efron, 1986) or the observed inaccuracy of the fitted model applied to the original data usually underestimates its actual capacity to predict future observations (true error rate), we base our retrospective analysis on the most parsimonious rather than the highest performing but likely overfitted model.

For illustration purposes, we used the dynamic linear models originally developed by Sadraddini et al. (2011b) to detect THg and PCB temporal trends in walleye. The optimal model (lowest DIC value) for the former case had the fish length as the sole covariate, whereas the latter one was based on both fish length and lipid content. We selected the predictive distributions for three years, e.g., 1986, 1996 and 2006, to examine how the intra- and inter-annual variability in the walleye contaminant levels as well as the total model (structural and parametric) uncertainty shape the risk assessment statements related to human fish consumption. Our first example presents the probability of exceedance of PCB tolerable daily intake through the consumption of Lake Erie walleye for a 75-kg person in different fish consumption scenarios: 2, 4, and 8 fish meals/month (Fig. 8). The results are presented for different fish lengths consumed, 22 to 74 cm, while the standard lipid content was set equal to 1.2%.

Our analysis (plausibly) suggests that increasing the number of meals per month translates to a greater risk of exposure. In particular, while two meals per month resulted in negligible exceedances across all fish lengths and years examined, four and eight meals can significantly increase the risk of violation of the PCB tolerable daily intake. The risk of exceeding the critical PCB level has distinctly decreased over time, as seen from the reduced probabilities of exceedance from 1986 to 2006. Further, when we consume fish of greater length, the probability of exceeding safe PCB levels rises as well; especially consumption of fish longer than 50 cm appears to be associated with more than 20% probability of exceedance of the PCB tolerable daily intake even in 2006. While these predictions were aimed at the average human weight, our analysis also suggests that the risk of PCB exposure in individuals with weight 50 kg (or less) can exceed the level of 30%, when fish with length greater than 50 cm is consumed twice per week (Fig. 9). The risk is clearly lower for heavier adults, and can drop below 20% when we consider individuals with body weights of 100 kg or more.

As a follow-up exercise, we examined how model performance can influence our capacity to obtain reliable risk assessment statements. In particular, we compared the mean predicted walleye THg concentrations as derived from three DLMs: the model that considers the covariance between THg and fish length; the one with the lipid content as the sole covariate; and the more complex structure with both fish length and lipid content as covariates (Fig. 10a). The three models were updated with the parameter posteriors for 2006, and the corresponding predictions are based on an assortment of fish length and lipid content values falling within the range observed over the last four decades in Lake Erie. In doing so, our intent was to reproduce the broadest range of THg concentrations that could potentially be measured in walleye, given the parameterization of the model for that particular year. The dotted line in Fig. 10a represents the lowest threshold value to

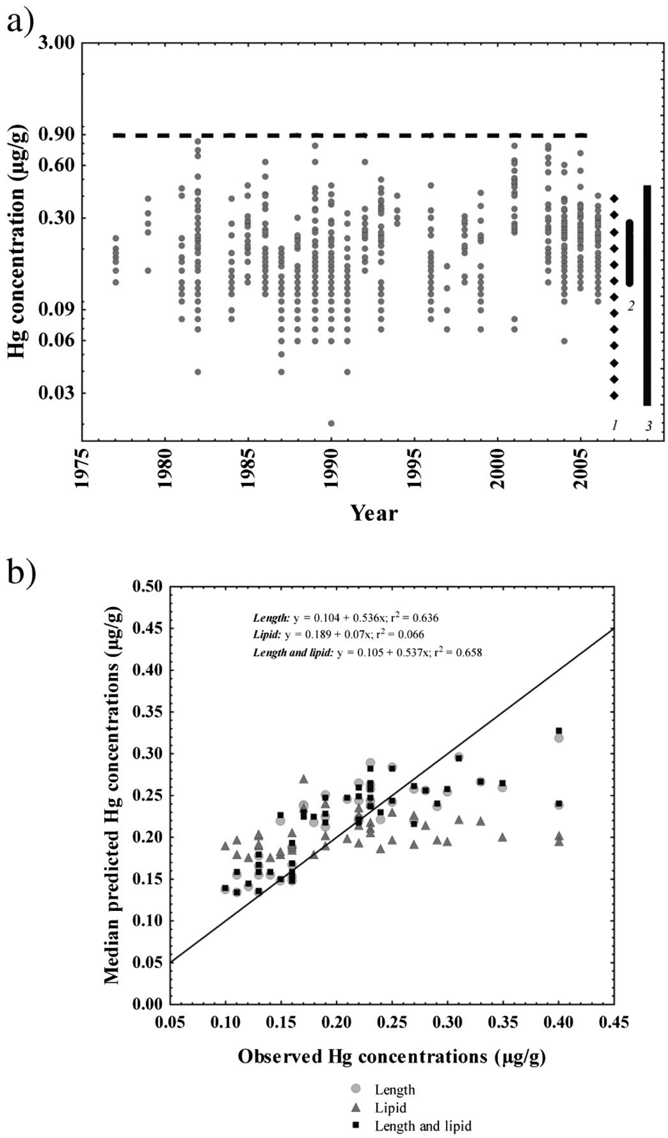

Fig. 10. (a) Mean predictions of walleye THg concentrations (μg/g) based on the range of fish lengths and lipid contents measured over the last four decades in Lake Erie. The numbered lines correspond to predictions from the dynamic linear models that consider fish length (1), lipid content (2), and both length and lipid values (3) as covariates. The three models were updated with the parameter posteriors for 2006. The predictions are plotted against the historically observed THg values (gray dots), while the dotted line represents the lowest threshold value to avoid harmful THg intake for a 42 kg person that has 4 meals of fish per month. (b) Comparison between the measured and median predicted THg concentrations from the three models.

avoid harmful intake of THg for a 50 kg person (e.g., adult with low body weight) that has 4 fish meals per month. First, we note that all the mean model predictions fall well below the critical level, indicating that THg does not currently produce a major cause for alarm. Second, the lipid model clearly underestimates the potential range of THg concentrations in the walleye community, indicative of its inferior performance or the weak covariance between THg and fish lipid content (Fig. 10b; $r^2 < 0.07$), On the other hand, the length and length/lipid models are equally proficient at recreating the potential range of fish contamination levels in the system. Notably, the discrepancy between the observed THg levels in 2006 and the predicted ones from the length and length/lipid models primarily stems from the broader range of fish lengths used for our predictive exercise relative to the systematically larger fish sampled in recent years (Bhavsar et al., 2007; Sadraddini et al., 2011a). The higher predictive capacity of the length and length/lipid models is also reflected by the significantly higher r^2 values (>0.64) between measured concentrations and predicted median values (Fig. 10b).

Contrary to the inability of the mean predictions from the lipid model to capture the within-year THg variability in walleye, the consideration of the total uncertainty associated with the corresponding predictions paints a different picture. In particular, when using the average lipid content of 1.2%, the higher median prediction of the lipid model along with the wider 95% predictive interval (higher model error) result in a marginal exceedance of the threshold value (Fig. 11a). Although the use of logarithmic scale is somewhat misleading, the predictive uncertainty of the lipid model suggests that 95% of the THg concentrations fall within the 0.04–1.0 μg/g range, whereas the corresponding uncertainty zone for the other two models lies between 0.03 and 0.6 μg/g. More solid evidence for the ramifications of the higher predictive uncertainty of an inferior model is presented in Fig. 11b, where the 95% predictive intervals of the lipid model against the observed THg concentrations are two- to five times broader than the (generally overlapping) points for the other two models. Hence, the adoption of probabilistic statements does not conceal the weaknesses of a model, but rather the inflated error/uncertainty makes the corresponding predictions practically uninformative for risk management. Simply put, the selection of an erroneous model in our example resulted in unjustifiably comforting risk statements when our retrospective analysis is solely based on the mean predictions, and in overly alarmist (or otherwise uninformative) projections when we also consider the underlying predictive error.

3.2.3. Outstanding issues with the fish consumption advisories

Aside from the accommodation of observed variability and predictive uncertainty, another key challenge with the development of proper fish advisories involves their capacity to effectively convey the benefits of fish consumption while stressing the associated contaminant risks for public health (Tilden et al., 1997). Although the repercussions of ingesting contaminants through fish consumption are well-established, fish also provide an excellent dietary source of high quality and easily digestible protein and omega-3 fatty acids (see Burger, 2000; Cohen et al., 2005; Smith and Sahyoun, 2005 for reviews). A wide array of work has been published relating dietary fatty acids to everything from cognitive functioning and nervous system maintenance (Minokoshi et al., 2002), to hormonal imbalances and insulin resistance complications (Yamauchi et al., 2001). Longer chain omega-3 fatty acids may also be important in preventing chronic health conditions, such as Alzheimer's disease, type II diabetes, kidney disease, rheumatoid arthritis, high blood pressure, coronary heart disease, alcoholism, and possibly cancer (Das, 2006). In this regard, the American Heart Association has advocated the adoption of two meals of fish per week to capitalize on these advantageous health effects (Kris-Etherton et al., 2002; Oken et al., 2003). It is therefore plausible that the public faces considerable confusion when dealing with these mixed messages from "restrictive" advisories and "encouraging" nutritionists, and so a large body of work has centered around assessing the relative risks of fish consumption and examining how consumers are dealing with this conflicting information (e.g., Burger and Gochfeld, 2006; Mozaffarian and Rimm, 2006; Stern and Korn, 2011). To ensure that guidelines do not completely inhibit fish consumption (see Cohen et al., 2005), advisories selectively recommend that people should avoid eating certain species/sizes of fish caught from certain contaminated locations. Yet, there is still no widely accepted methodology to generate integrative statements that impartially weigh the benefits and net risks of consuming fish, though there are some recent proposals (e.g. Stern and Korn, 2011).

In addition to balancing the risks and benefits of fish, a lengthy list of additional challenges plagues the production of fish consumption advisories (e.g., see Oken et al., 2012). One of the primary issues revolves around ensuring effective communication, as many anglers may not be aware of the advisories or may opt to downplay the advisories due to preconceived notions, such as optimism about their catch, failure to understand the advisories or distrust of the government (see Burger,

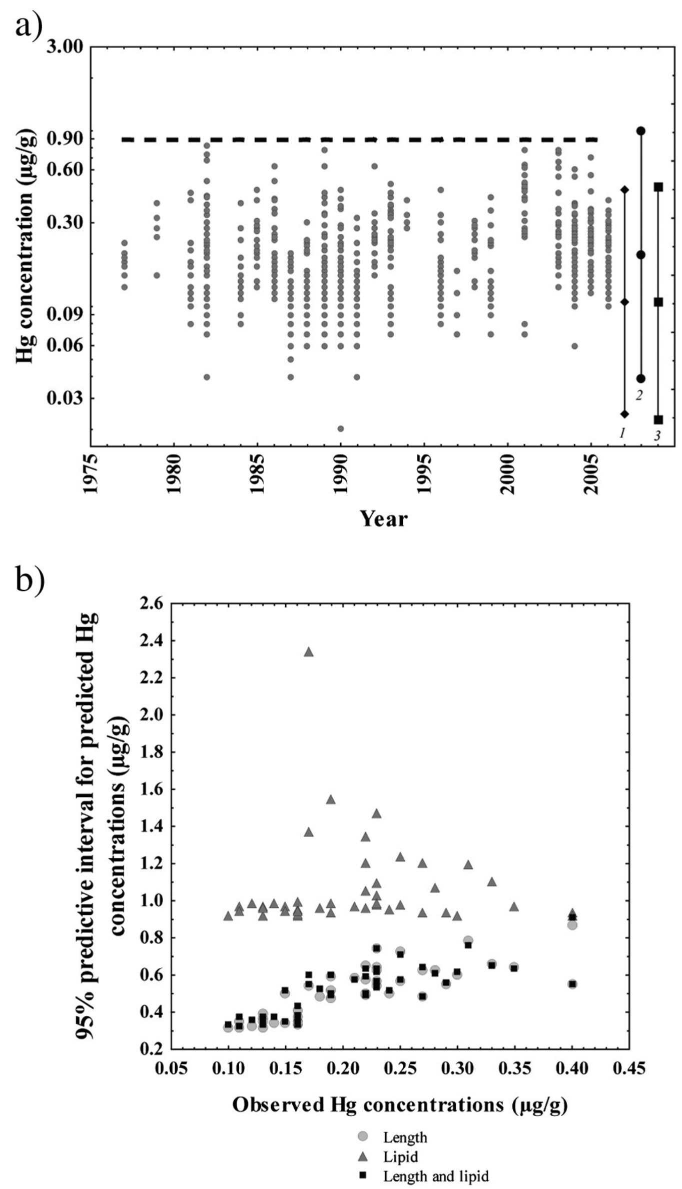

Fig. 11. (a) Predicted walleye Hg concentrations (µg/g) for the average fish length and lipid values measured over the last four decades in Lake Erie. The numbered lines correspond to model predictions with the average fish length of 40 cm (1), the average fish lipid content of 1.2% (2), and both average length and lipid content (3). The central markers on each numbered line correspond to the median model prediction, while the outer markers represent the 2.5 and 97.5 percentiles. The three models were updated with the parameter posteriors for 2006; (b) 95% predictive intervals against observed THg (µg/g) concentrations in 2006, using the three dynamic linear models.

2000; Burger and Gochfeld, 2006; Oken et al., 2012; Pflugh et al., 1999; Tan et al., 2011). As such, a number of studies have primarily focused on working with target populations to identify possible ways of enhancing advisories and improving their tone and readability of the advisories (Connelly and Knuth, 1998; Jardine, 2003; Tan et al., 2011; Velicer and Knuth, 1994). Other communication challenges include the failure to reach vulnerable subgroups, such as pregnant women, women of childbearing age, children under 15, Hispanics and African-Americans (Anderson et al., 2004; Lepak et al., 2009; Scherer et al., 2008; Shimshack et al., 2007); disconnect with Native populations (see Donatuto and Harper, 2008); and lack of consensus about the definition of what is a "sensitive" population (Lepak et al., 2009). Aside from these "communication-centric" issues, the production of fish consumption advisories is also made inherently more complex by the synergistic effects among multiple contaminants (Clark et al., 1987; Scherer et al., 2008), and the differential impacts of cleaning and cooking fish on different contaminants (GLSFATF, 1993).

4. Conclusions

We used dynamic linear modeling to examine the temporal trends of three organochlorine compounds (hexachlorobenzene, octachlorostyrene, and α-hexachlorocyclohexane) in five Lake Erie fish species. Our analysis indicates that the levels of organochlorines have been decreasing over the last three decades, although there were cases that exhibited fluctuations through time. The present results reinforce the findings of our recent work that the levels of several important contaminants, such as THg (Azim et al., 2011a; Sadraddini et al., 2011b), PCB (Sadraddini et al., 2011a, 2011b), chlordane (Azim et al., 2011b), and dichlorodiphenyltrichloroethane (Mahmood et al., in press) have been declining in the fish communities of Lake Erie over the past few decades and do not produce major causes for alarm. However, it must be noted that there are fish species (walleye, smallmouth bass, rainbow trout, white bass, freshwater drum) with differences in their dietary habits, foraging behavior and competition strategies, which exhibit weakly increasing trends of their THg and/or PCB body burdens following the mid- or late 1990s (Azim et al., 2011a; Sadraddini et al., 2011b). Thus, it is important to closely monitor these trends, particularly in fish species regularly chosen for human consumption. We also presented a Bayesian framework to update fish consumption advisories that incorporates the uncertainty in contaminant predictions and the natural variability in fish length and lipid content, while remaining flexible for different human weights and diet patterns. Our results demonstrate that the risk of exposure has clearly decreased over time, but there is still substantial likelihood of exceedance of the tolerable daily intake for PCBs in sensitive populations (e.g., children with low body weight) with frequent consumption of large fish. Future augmentations of the present framework need to focus on its capacity to generate integrative statements that impartially weigh benefits and net risks of consuming fish, and special emphasis should be given to the increased sensitivity of vulnerable groups.

Acknowledgments

This project has received funding support from the Ontario Ministry of the Environment (Best in Science Research Program-Grant Funding Agreement 89002). Such support does not indicate endorsement by the Ministry of the contents of the study. All the material pertinent to this analysis is available upon request from the corresponding author.

Appendix A. Tobit dynamic linear modeling approach

The WinBUGS code associated with the dynamic linear model for the hexachlorobenzene (HCB) concentrations is as follows:

```
model {
for (i in 1:N) {
upper.lim[i] < - DETLIM[i]*is.detlim[i] + UPPERLIM*(1 - is.detlim[i])
is.detlim[i] < -step(0-LogHCB[i])
DETLIM[i] ~ dnorm(-4.605,0.511)
lengthstdev[i] < -(length[i]-3.3958199)/0.1828117
lipidstdev[i] < -(lipid[i]-1.014126)/0.559355
LogHCBm[i] < -level[time[i] + 1] + beta1[time[i] + 1]*
lengthstdev[i] + beta2[time[i] + 1]*lipidstdev[i]
LogHCB[i] ~ dnorm(LogHCBm[i],mtau[time[i] + 1])I(,upper.lim[i])
LogPredHCB[i] ~ dnorm(LogHCBm[i],mtau[time[i] + 1])
PredHCB[i] < -exp(LogPredHCB[i])}
for (t in 2:31) {
beta1[year[t]] ~ dnorm(beta1[year[t-1]],btau1[year[t]])
beta2[year[t]] ~ dnorm(beta2[year[t-1]],btau2[year[t]])
growth[year[t]] ~ dnorm(growth[year[t-1]],gtau[year[t]])
```

```
levelm[year[t]] < -level[year[t-1]] + growth[year[t]]
level[year[t]] ~ dnorm(levelm[year[t]],ltau[year[t]])
ltau[year[t]] < -ltau.in*pow(0.95,year[t]-1)
lsigma[year[t]] < -sqrt(1/ltau[year[t]])
btau1[year[t]] < -btau1.in*pow(0.95,year[t]-1)
btau2[year[t]] < -btau2.in*pow(0.95,year[t]-1)
bsigma1[year[t]] < -sqrt(1/btau1[year[t]])
bsigma2[year[t]] < -sqrt(1/btau2[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]])
}
beta1[year[1]] ~ dnorm(beta1[1],btau1[year[1]])
beta2[year[1]] ~ dnorm(beta2[1],btau2[year[1]])
growth[year[1]] ~ dnorm(growth[1],gtau[year[1]])
levelm[year[1]] < -level[1] + growth[year[1]]
level[year[1]] ~ dnorm(levelm[year[1]],ltau[year[1]])
ltau[year[1]] < -ltau.in
lsigma[year[1]] < -sqrt(1/ltau[year[1]])
btau1[year[1]] < -btau1.in
btau2[year[1]] < -btau2.in
bsigma1[year[1]] < -sqrt(1/btau1[year[1]])
bsigma2[year[1]] < -sqrt(1/btau2[year[1]])
gtau[year[1]] < -gtau.in
gsigma[year[1]] < -sqrt(1/gtau[year[1]])
mtau[year[1]] < -mtau.in
msigma[year[1]] < -sqrt(1/mtau[year[1]])
beta10 ~ dnorm(0,0.0001)
beta20 ~ dnorm(0,0.0001)
growth0 ~ dnorm(0,0.0001)
level0 ~ dnorm(0,0.0001)
ltau.in ~ dgamma(0.001,0.001)
btau1.in ~ dgamma(0.001,0.001)
btau2.in ~ dgamma(0.001,0.001)
gtau.in ~ dgamma(0.001,0.001)
mtau.in ~ dgamma(0.001,0.001)
}
```

Inference data

```
list(N = 1158, UPPERLIM = 10,000,
year = c(1,2,3,4,5,6,7,8,9,10,11,12,13,14,15,16,17,18,19,20,21,
22,23,24,25,26,27,28,29,30,31),
time = c(paste time.dat here),
LogHCB = c(paste whitebassHCB.dat here),
lipid = c(paste lipid.dat here),
length = c(paste length.dat here)),
```

Initial values 1

```
list(beta10 = 0.5,beta20 = 0.5,growth0 = 0.5,level0 = 0.5,
beta1 = c(0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,
0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5),
beta2 = c(0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,
0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5),
growth = c(0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,
0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5),
level = c(0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,
0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5,0.5),
mtau.in = 0.2,ltau.in = 0.2,btau1.in = 1, btau2.in = 1,gtau.in = 1,
LogPredHCB = c(paste whitebassHCB.dat here))
```

Initial values 2

```
list(beta10 = 0.25,beta20 = 0.25,growth0 = 0.25,level0 = 0.25,
beta1 = c(0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,
0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,
0.25,0.25,0.25,0.25,0.25,0.25,0.25),
beta2 = c(0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,
0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,
0.25,0.25,0.25,0.25,0.25,0.25,0.25),
growth = c(0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,
0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,
0.25,0.25,0.25,0.25,0.25,0.25,0.25),
level = c(0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,
0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,
0.25,0.25,0.25,0.25,0.25,0.25),
mtau.in = 0.5,ltau.in = 0.5,btau1.in = 0.5,
btau2.in = 0.5,gtau.in = 0.5,
LogPredHCB = c(paste whitebassHCB.dat here))
```

Initial values 3

```
list(beta10 = 0.05,beta20 = 0.05,growth0 = 0.05,level0 = 0.05,
beta1 = c(0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,
0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,
0.18,0.18,0.18,0.18,0.18,0.18,0.18),
beta2 = c(0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,
0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,
0.18,0.18,0.18,0.18,0.18,0.18,0.18),
growth = c(0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,
0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,
0.18,0.18,0.18,0.18,0.18,0.18,0.18),
level = c(0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,
0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,0.18,
0.18,0.18,0.18,0.18,0.18,0.18),
mtau.in = 0.05,ltau.in = 0.05,btau1.in = 0.05,
btau2.in = 0.05,gtau.in = 0.05,
LogPredHCB = c(paste whitebassHCB.dat here))
```

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