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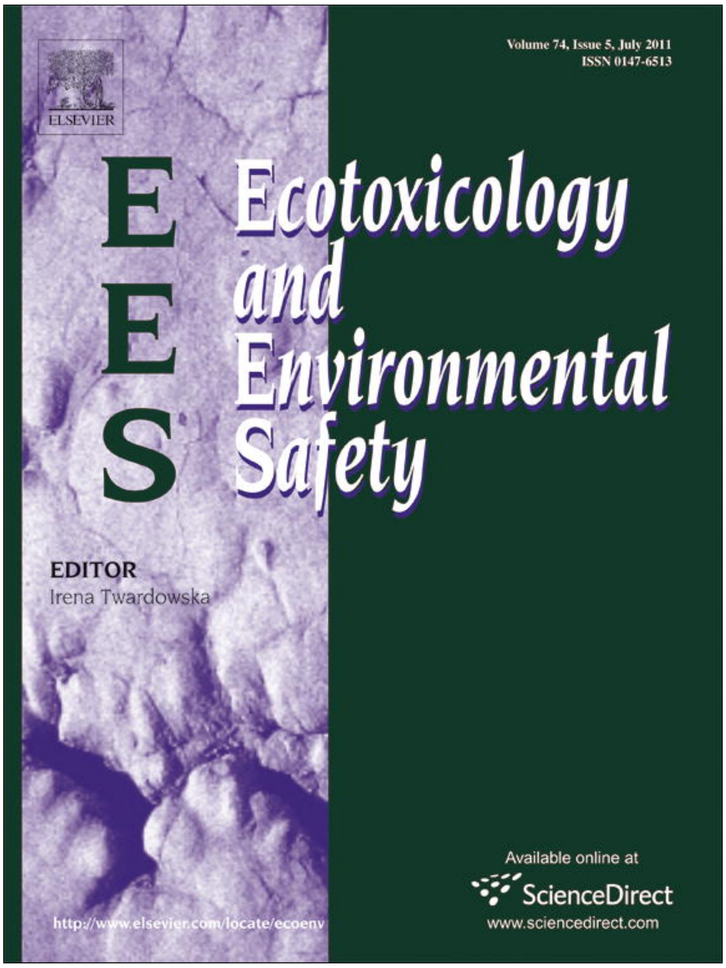

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Detection of temporal trends of α- and γ-chlordane in Lake Erie fish communities using dynamic linear modeling

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ABSTRACT

Dynamic linear modeling (*DLM*) analysis was performed to identify the long-term temporal trends of two toxic components of the technical chlordane pesticide, α- and γ-chlordane, in skinless–boneless muscle tissues of a number of sport fish species in Lake Erie. Our analysis considers the fish length as a covariate of the chlordane concentrations. The α-chlordane models for the coho salmon, channel catfish, rainbow trout, and common carp showed continuously decreasing trends during the entire 30+ year survey period (1976–2007). The γ-chlordane models demonstrated similar trends for the coho salmon, channel catfish, and common carp. These fish species had higher levels of α- and γ-chlordane in their muscle tissues. The α- and γ-chlordane levels in freshwater drum, smallmouth bass, walleye, white bass, whitefish, and yellow perch decreased until the mid-1980s and hovered at levels around the detection limits for the remaining period. The pesticide biotransformation process, the reduction of contaminant emissions to the environment, the feeding habits of the different fish species, and the food-web alterations induced by the introduction of aquatic invasive species are some of the hypotheses proposed to explain the observed temporal trends in different fish species in Lake Erie.

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1. Introduction

Pesticide concentrations in aquatic systems are primarily related to the land use patterns of the surrounding watershed, the local rainfall-runoff characteristics, and the season of the year (Myers et al., 2000). Lake Erie has been historically exposed to the greatest stress from agriculture as compared to the other Laurentian Great Lakes. The 78,000 km^2 basin area is dominated by agriculture (80% in the Canadian part and 63% in the US part) followed by forested (Canada 15% and USA 23%) and residential areas (Canada 4% and USA 12%) (Han et al., 2011). Pesticides applied to agricultural crops, lawns, and gardens in Lake Erie watershed find their way into the system through surface runoff. Growing public concerns and awareness of the water quality problems became the major catalyst for the USA–Canada Water Quality Agreement in 1972. Within this agreement, the Western Lake Erie and Detroit River, which provides 80% of the water flow in Lake Erie, have received special attention (Herdendorf, 1986; Bolsenga and Herdendorf, 1993). Pollution control comprised a range of key regulatory and non-regulatory initiatives, which have significantly contributed to the ecological recovery of Lake Erie over the past decades. This recovery includes improved reproduction and higher abundance of bald eagles, peregrine falcons, walleye, lake sturgeon, lake whitefish, and burrowing mayflies to large areas from which they were extirpated or negatively impacted (Hartig et al., 2009).

Organochlorine pesticides including chlordane were used in high quantities in the Great Lakes basin until the 1970s. Chlordane is a persistent bioaccumulative and toxic chemical that was introduced in North America in 1949 for controlling the insect pests in crops and forests (http://www.ecoinfo.org). Yet, aside from the control of subterranean termites, Canada has suspended its use since 1985. Further, any sale or use of chlordane was effectively banned at the end of 1995, constituting a violation of the Federal Pest Control Products Act of Canada. In a similar manner, the U.S., a major producer and consumer of chlordane, suspended the use in 1983, except for termite control. Although it was completely banned in 1988, the production continued for export until 1997 (Lipnick and Muir, 2000). Currently, the use of pesticides is tightly regulated and their levels in surface waters originating from the Great Lakes basin are being monitored. Generally, while the concentrations for many pesticides appear to be in compliance with the targeted threshold levels, there are cases of pesticides that still exceed the current regulatory criteria (IJC, 2009). In particular, chlordane is detected in fish even in recent years, although it has not been used in North America for the last 15–25 years. The U.S. Food and Drug Administration

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(FDA) has recommended that the level of chlordane in animal fat and fish should not be greater than 100 μg/kg (Abadin et al., 1994). According to FAO/WHO (1995), the acceptable daily intake of chlordane in food is 0.5 μg/kg body weight. Most human health impacts of chlordane exposure are related to the impairment of nervous system (headaches, irritation, confusion, weakness, and vision problem), digestive system (stomach cramps, vomiting, and diarrhea), reproductive system (spermatogenetic dysfunction, and birth deformities), and liver (jaundice) (Abadin et al., 1994; Harry et al., 1998; Reigart and Roberts, 2001; Bolognesi, 2003; Eddleston and Bateman, 2007; Corsini et al., 2008). Recently, chlordane has been further targeted for global elimination under the recently signed Stockholm Convention on Persistent Organic Pollutants (*POPs*) effective from August 26th, 2010 (http://chm.pops.int).

Despite the fact that the levels of contaminants have been significantly reduced in the Great Lakes environment due to the implementation of various preventive and control measures, some legacy contaminants, such as mercury and polychlorinated biphenyls (*PCBs*), have been recently reported to remain stagnant or even to be increased in some fish species (Bhavsar et al., 2007, 2010; Carlson et al., 2010). In particular, recent studies by Azim et al. (2011), Sadraddini et al. (in press) and Sadraddini et al. (submitted) provided clear evidence of such trends in Lake Erie and also reviewed various ecological mechanisms that can conceivably underlie these temporal patterns. As a continuation of our earlier work, the present paper aims to delineate the temporal trends of the organochlorine pesticide chlordane in eleven fish species in Lake Erie over the last three decades.

Our analysis focuses on two toxic residues of chlordane, viz., α-chlordane (or cis-chlordane) and γ-chlordane (or trans-chlordane), known to be accumulated in aquatic biota (Kawano et al., 1986), which make up to 19% and 24% of the technical chlordane, respectively (Simonich and Hites, 1995). Our analysis is based on dynamic linear modeling (*DLM*) due to its evolving structure that enables the elucidation of the role of potentially important cause–effect relationships and supports forecasts that are primarily driven by most recent data while information from the distant past can be discounted (Pole et al., 1994). We selected fish length as a potential covariate of the contaminant concentrations to account for the fact that fish size affects contaminant levels, and different-sized fish may have been sampled over time. The longer exposure time, the dietary shifts with age, the differences in uptake, assimilation and excretion as well as the changes in relative organ size may be some of the reasons that typically result in increased contaminant concentrations with fish size (Evans et al., 1993). Our specific objectives include (i) the comparison of the observed chlordane concentrations among different fish species; (ii) the prediction of the temporal trends when explicitly considering the role of fish length variability; and (iii) the examination of whether the introduction of aquatic invasive species has influenced the contaminant trends. Our study concludes by examining the key causal relationships that may have shaped the chlordane concentrations in Lake Erie over the last three decades.

2. Methods

2.1. Dataset description and chemical analysis

The fish samples were collected and analyzed by the Sport Fish Monitoring Program of the Ontario Ministry of the Environment (*MOE*). Chlordane was measured in the dorsal muscles without skins and bones (called skinless–boneless fillet, *SBF*, herein) for the purpose of fish consumption advisories. The dataset spans about 32 years (from 1976 to 2007) of chlordane concentrations measured in eleven and ten fish species for α- and γ-chlordane, respectively. The fish species examined were selected on the basis of the data availability and/or their commercial importance. The fish samples were collected from a number of locations on the Canadian side of Lake Erie and were classified in four regions, viz., Western Basin including Point Pelee; Central Basin including Rondeau Bay, Port Stanley and Wheatley Harbor; Long Point Bay; and Eastern Basin. The chlordane levels were measured at the MOE laboratory in Toronto through gas chromatography with Ni^{63} electron capture detector (*GLC–ECD*)—(the MOE method E3136; MOE, 2007).

2.2. Modeling framework

Dynamic linear modeling (*DLM*) analysis was used to examine the chlordane temporal trends, while explicitly accounting for the fish length as covariate, thereby accounting for the fact 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. *DLM*s easily handle missing values/unequally spaced data, and minimize the effect of outliers (Pole et al., 1994). All *DLM*s consist of an observation equation and system equations (West and Harrison., 1989). In particular, the *DLM*s used herein were specified as follows:

Observation equation

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

System equations:

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

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

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

$$1/\Omega^2_{tj} = \zeta^{t-1} \cdot 1/\Omega^2_{1j}, 1/\Psi^2_t = \zeta^{t-1} \cdot 1/\Psi^2_1 \quad t > 1 \quad \text{and} \quad j = 1 \ \ to \ \ 3$$

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

$$1/\Omega^2_{1j}, 1/\Psi^2_1 \sim gamma(0.001, 0.001)$$

where $\ln[chlordane]_{ti}$ is the observed α- or γ-chlordane concentrations at time t in the individual sample i; $level_t$ is the mean α- or γ-chlordane concentrations 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^2_t, Ω^2_{tj}, 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 $gamma(0.001, 0.001)$ 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, $1/\Omega^2_{1j}$, and $1/\Psi^2_1$ 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 the 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. This discounted posterior knowledge becomes prior knowledge for time $t+1$, and the process is repeated.

2.3. 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% and 40% (Arhonditsis et al., 2007, 2008). We used two 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 (≈ 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

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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 5% of the sample standard deviation.

3. Results

The summary statistics of the α- and γ-chlordane concentrations in different fish species are shown in Tables 1 and 2, respectively. The highest α-chlordane concentrations were recorded in coho salmon (mean 12.2 and median 9 ng/g wet weight or ww), followed by channel catfish (9.4 and 8 ng/g ww), rainbow trout (6.7 and 2 ng/g ww) and common carp (6.4 and 2 ng/g ww). The remaining seven species had lower median concentrations ranging from 2 to 4 ng α-chlordane/g ww. Likewise, coho salmon had again the highest γ-chlordane concentrations (mean 8.6 and median 4 ng/g ww), followed by channel catfish (7.6 and 2 ng/g ww), rainbow trout (5.9 and 2 ng/g ww), and common carp (4.4 and 2 ng/g ww). The median concentrations ranged between 2 and 3.5 ng γ-chlordane/g ww for the remaining fish species. It should also be noted that more than 50% measurements for all species except channel catfish and coho salmon for α-chlordane and coho salmon for γ-chlordane were below the detection limit. Moreover, the high standard deviations along with the positive skewness and kurtosis values reflect the substantial intra- and inter-annual variability associated with the contaminant levels of the individual fish species for both the contaminant components. Therefore, natural log transformation was implemented for the subsequent modeling analysis, effectively imposing a log-normal error structure.

The *DLM* analysis identified three distinct patterns regarding the rates of change of the α-chlordane concentrations in the fish

Table 1
Basic statistics of α-chlordane concentrations (ng/g wet weight) in skinless–boneless fillet samples of different fish species collected from Lake Erie between 1976 and 2007.

Species name	Length (cm)	*N*	Mean	SD	Skewness	Kurtosis
Common carp *Cyprinus carpio*	59 ± 11	494	6.4	11.7	5.2	31.7
Channel catfish *Ictalurus punctatus*	45 ± 12	710	9.4	14.1	3.5	14.2
Coho salmon *Oncorhynchus kisutch*	56 ± 13	655	12.2	12.6	4.6	44.9
Freshwater drum *Aplodinotus grunniens*	36 ± 7	469	3.3	4.5	6.5	54.5
Rainbow trout *Oncorhynchus mykiss*	56 ± 11	441	6.7	10.2	2.9	8.6
Smallmouth bass *Micropterus dolomieui*	35 ± 8	506	3.3	4.5	6.7	58.7
Walleye *Stizostedion vitreum*	53 ± 9	1156	2.5	1.5	4.5	24.2
White bass *Morone chrysops*	30 ± 5	1451	4.2	5.2	4.6	30.3
White fish *Coregonus clupeaformis*	49 ± 7	576	2.8	2.8	6.9	63.6
White perch *Morone Americana*	23 ± 3	500	2.6	1.6	3.4	13.6
Yellow perch *Perca flavescens*	24 ± 5	434	2.3	1.5	8.0	79.4

Table 2
Basic statistics of γ-chlordane concentrations (ng/g wet weight) in skinless–boneless fillet samples of different fish species collected from Lake Erie between 1976 and 2007.

Fish species	Length (cm)	*N*	Mean	SD	Skewness	Kurtosis
Common carp *Stizostedion vitreum*	59 ± 11	369	4.4	5.8	4.3	22.6
Channel catfish *Ictalurus punctatus*	44 ± 11	533	7.6	12.7	4.1	22.1
Coho salmon *Oncorhynchus kisutch*	56 ± 13	649	8.6	10.3	2.3	5.3
Freshwater drum *Aplodinotus grunniens*	35 ± 7	359	3.1	5.6	8.1	79.9
Rainbow trout *Oncorhynchus mykiss*	57 ± 11	312	5.9	9.5	3.2	11.6
Smallmouth bass *Micropterus dolomieui*	34 ± 8	416	3.5	5.5	5.7	39.3
Walleye *Stizostedion vitreum*	52 ± 9.0	874	2.2	1.4	10.1	120.0
White bass *Morone chrysops*	30 ± 5.0	1160	3.4	5.7	10.6	181.6
Whitefish *Coregonus clupeaformis*	49 ± 7.0	350	2.3	0.9	5.9	46.4
Yellow perch *Perca flavescens*	23 ± 5.0	368	2.3	1.4	10.7	146.3

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species examined (Fig. 1). First, except for whitefish, yellow perch, and white perch (Fig. 1i–k), eight species were characterized by negative rates of change of the α-chlordane concentrations during the survey period (Fig. 1a–h). Further, freshwater drum, smallmouth bass, and common carp showed relatively strong, and walleye and white bass showed weakly negative rates of change during the first half of the survey period. For the remaining period, the rates of change gradually minimized and eventually got stabilized around zero. By contrast, channel catfish, coho salmon, and rainbow trout did not show any remarkable changes in their rates of concentration change during the survey period. The aforementioned negative rates of change are reflected in the decrease of the corresponding predicted mean α-chlordane concentrations, when accounting for the covariance with the fish length (Fig. 2a–h). We also note the major trough in the temporal patterns of white bass in 1978 (Fig. 2h), when all 40 samples were collected from Long Point Bay and Central Basin and the measured values were under the detection limit. Second, white perch demonstrated a slightly positive rate of change until the 1990, after which the rates stabilized around zero until the end of the study period (Fig. 1j). Consequently, this trend is primarily manifested as an increase of the length-corrected annual

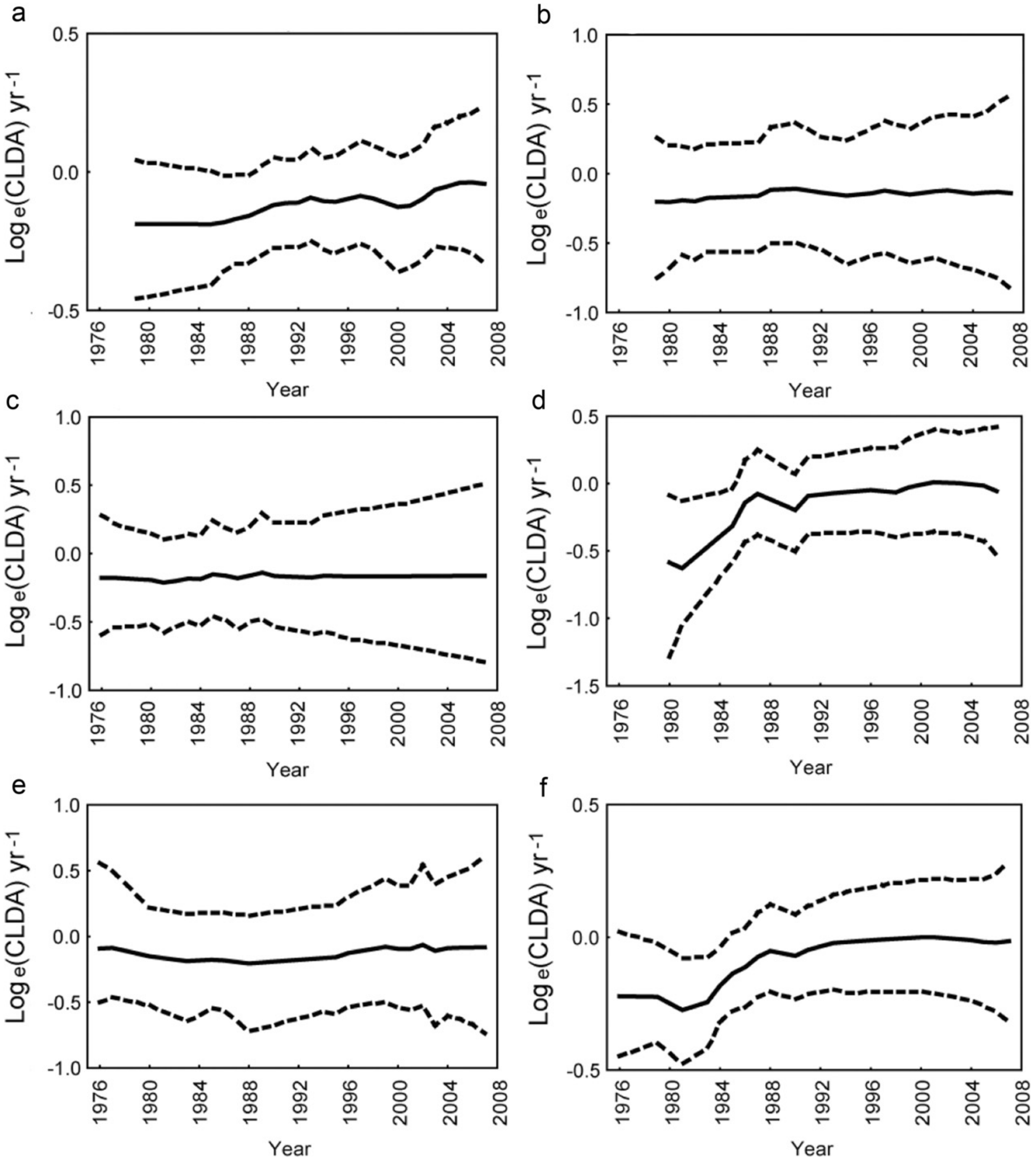

Fig. 1. Dynamic linear modeling analysis depicting the annual rates of change of α-chlordane *(CLDA)* concentrations (ng/g wet weight) in (a) common carp, (b) channel catfish, (c) coho salmon, (d) freshwater drum, (e) rainbow trout, (f) smallmouth bass, (g) walleye, (h) white bass, (i) whitefish, (j) white perch, and (k) yellow perch in Lake Erie. The solid and dashed lines correspond to the median and the 95% posterior predictive intervals, respectively.

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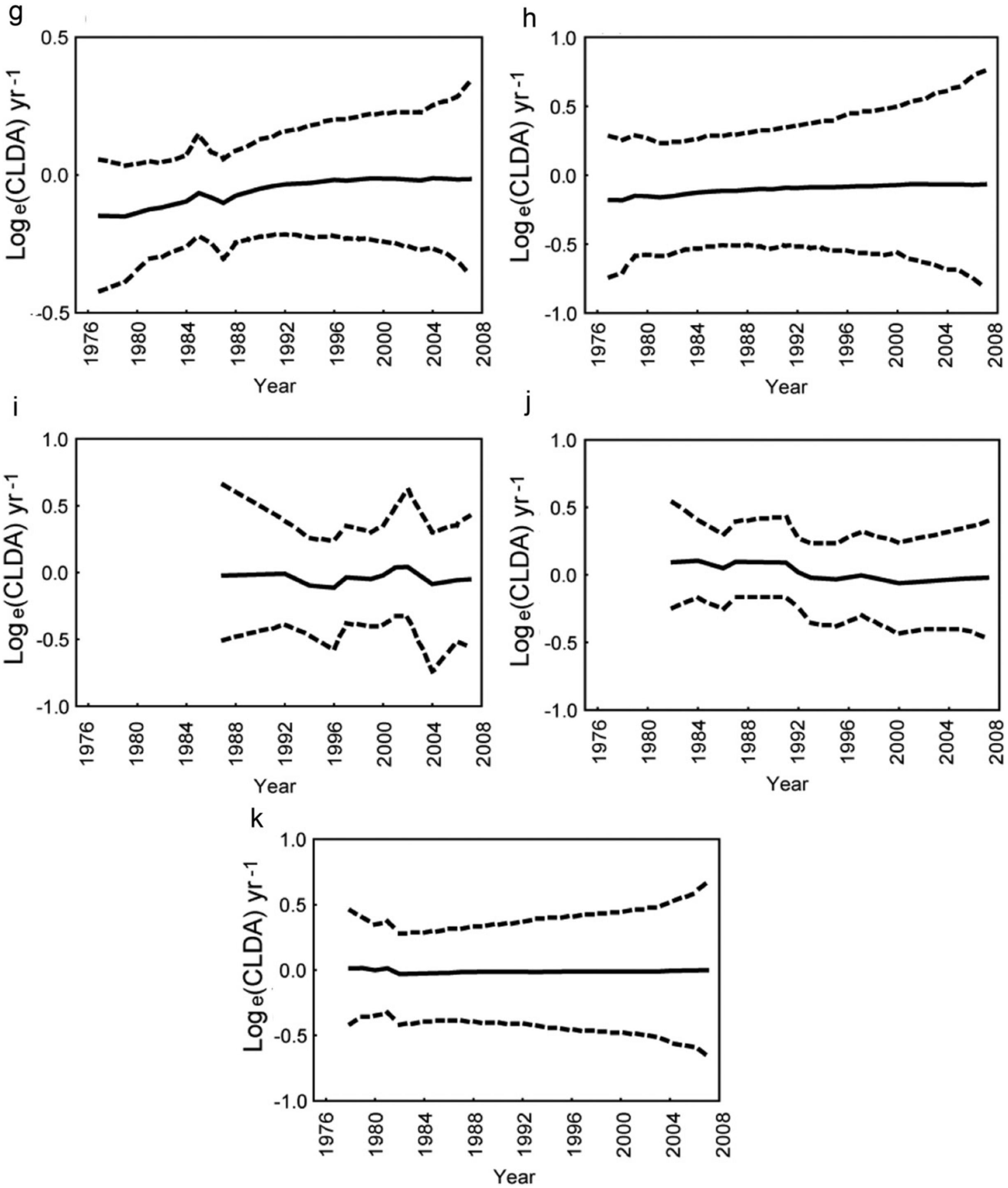

Fig. 1. (*continued*)

α-chlordane concentrations, followed by a decrease and gradual stabilization towards the end of the study period (Fig. 2j). Third, whitefish and yellow perch showed rates of change revolving around zero throughout the study period (Fig. 1i,k), indicating no significant changes in the predicted annual α-chlordane concentrations (Fig. 2i,k). Yet, there were some notably high values for whitefish in 2001 and 2002, which involve samples mainly collected from the Central Basin.

All the fish species examined were characterized by negative rates of change of their γ-chlordane concentrations (Fig. 3). In particular, common carp, channel catfish, and coho salmon exhibited weakly negative rates of change during the years examined (Fig. 3a–c). Therefore, the predicted length-corrected γ-chlordane concentrations for the three fish species decreased continuously at a slow rate until the end of the study period (Fig. 4a–c). Secondly, freshwater drum, rainbow trout, and walleye data were characterized by strongly negative rates of change during the first half of the survey period and then approximately reached zero rates since the early 1990s (Fig. 3d,e,g). Consequently, the predicted length-corrected γ-chlordane levels for the

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three fish species decreased sharply until the late 1980s/early 1990s, and thereafter decreased at significantly slower rates (Fig. 4d,e,g). Similar to *a*-chlordane, we also note that all the observed values for white bass in 1978 were under the detection limits; possibly due to the previously mentioned sampling bias (Fig. 4h). Thirdly, smallmouth bass and white bass demonstrated moderately strong negative rates of change until the late 1980s, and then got stabilized around zero (Fig. 3f,h). These trends were reflected in the predicted mean γ-chlordane concentrations which decreased at a moderately strong rate during first half of the study period and stabilized thereafter (Fig. 4f,h). Finally, yellow perch and whitefish switched from relatively weak negative to nearly zero rates (Fig. 3i,j), which were consequently manifested as a decrease of the length-corrected γ-chlordane concentrations during the earlier years of our study period for both species (Fig. 4i,j).

The introduction of aquatic invasive species, especially zebra mussels and round goby, has been hypothesized to be an important driver of the recent trends of the fish contaminant levels in the Great Lakes (Morrison et al., 1998; Hogan et al., 2007; Sadraddini et al., in press; Sadraddini et al., submitted; French et al., 2011). To examine whether this hypothesis holds true for

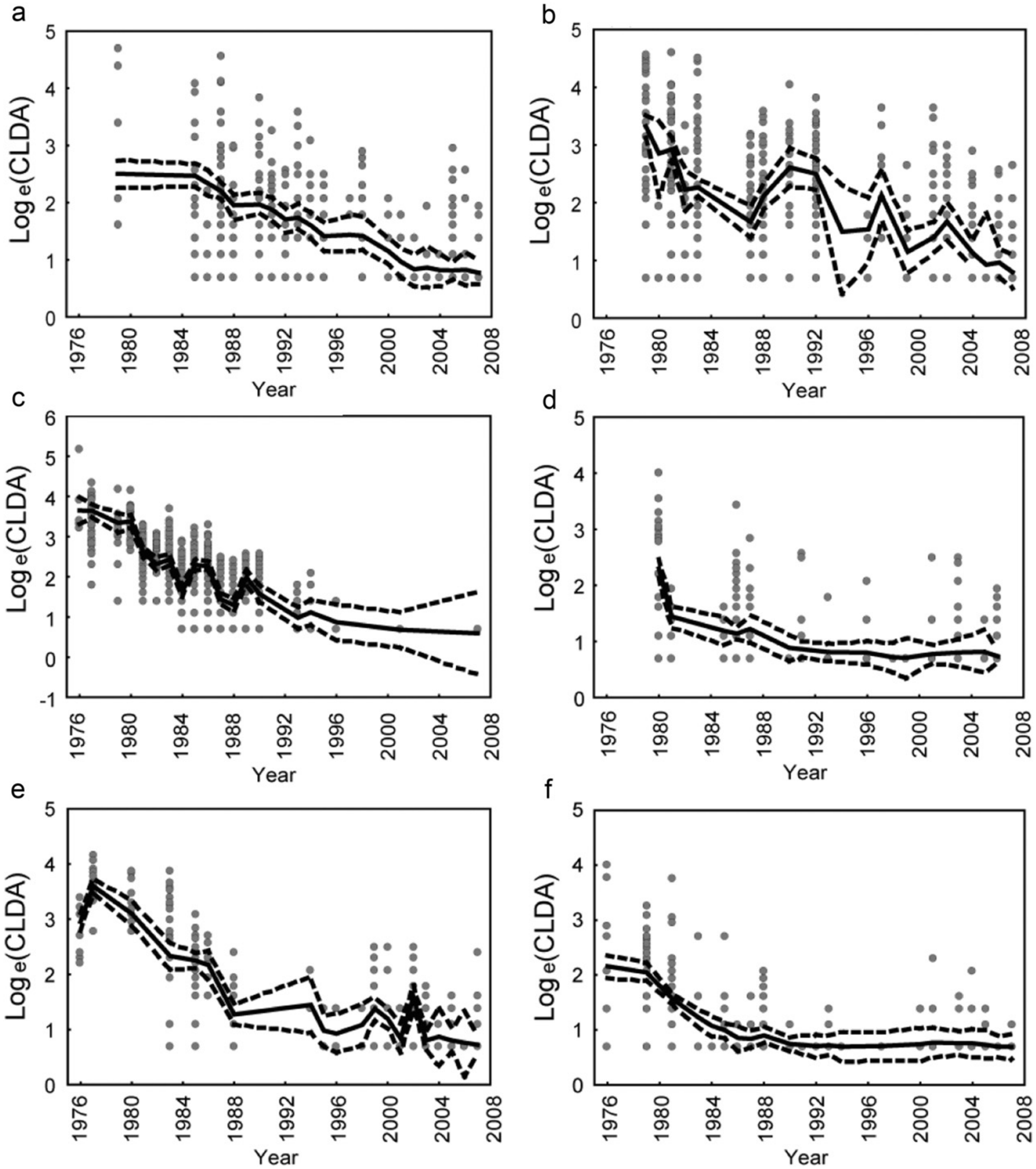

Fig. 2. Dynamic linear modeling analysis depicting the actual α-chlordane *(CLDA)* concentrations (μg/g wet weight) (gray dots) against the predicted *CLDA* trends when accounting for the covariance with the fish length (black lines) in (a) common carp, (b) channel catfish, (c) coho salmon, (d) freshwater drum, (e) rainbow trout, (f) smallmouth bass, (g) walleye, (h) white bass, (i) whitefish, (j) white perch, and (k) yellow perch in Lake Erie. The solid and dashed lines correspond to the median and the 95% posterior predictive intervals, respectively.

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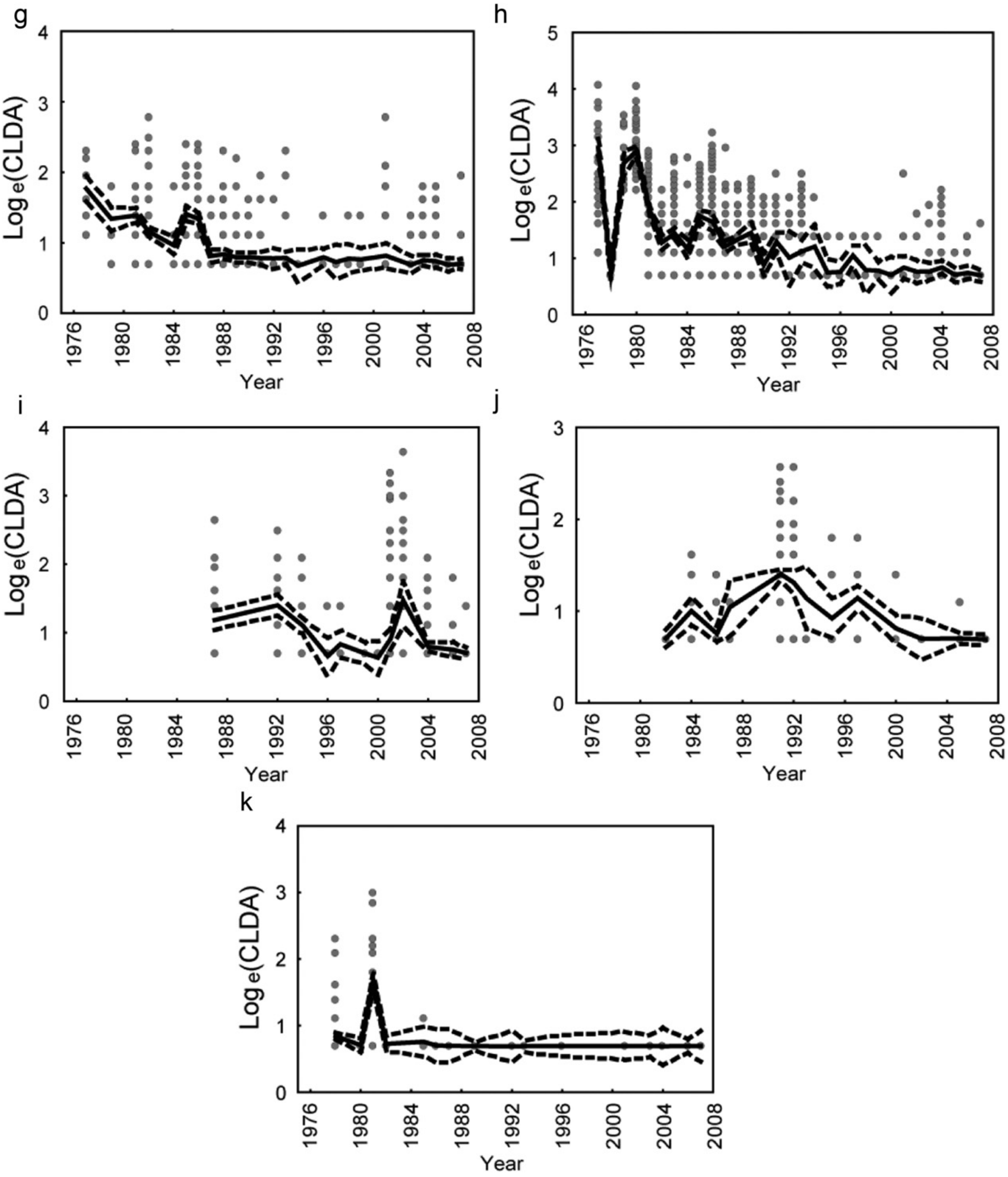

Fig. 2. (*continued*)

the temporal trends of chlordane in Lake Erie fish communities, we provide a synoptic illustration of the relative strength of the α- and γ-chlordane rates of change for the fish species examined, in which the early 1990s are used as a reference point to distinguish between pre- and post-invasion periods (Fig. 5). Our analysis indicates that the introduction of invasive species did not have any significant effects on the temporal trends of α-chlordane for common carp, channel catfish, coho salmon, and rainbow trout and the corresponding rates between the pre- and post-invasion periods remained practically unaltered with regards to their sign and magnitude. Similarly, the α-chlordane concentrations in yellow perch and whitefish remained unaffected by the introduction of exotic species. By contrast, the negative rates of change in freshwater drum, smallmouth bass, walleye, and white bass gradually switched to zero, but as shown earlier the timing of this change does not necessarily coincide with the invasion of exotic species. In the case of γ-chlordane, the aquatic invasive species do not have any impact on the rates of change for common carp, channel catfish, and coho salmon. The remaining seven species demonstrated negative trends with varying rates during the pre-invasion period, and have been hovering around zero since the mid-1980s.

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4. Discussion

The present paper provides a comprehensive analysis on temporal trends of the two most toxic components of the pesticide chlordane in eleven fish species in Lake Erie. Our modeling study shows a steady decrease of the two chlordane compounds in a number of sport fish species during the 30+ year survey period (1976–2007). Yet, despite the fact that the use of the pesticide chlordane has been banned in North America for more than 15–25 years ago, we found that the concentrations of both α- and γ-chlordane are still above the detection limits in several fish species. Moreover, the data presented in this paper are based on skinless–boneless muscle tissues with the objective of developing fish consumption advisories by the Ontario Ministry of the Environment (*MOE*). It is known though that the organochlorine contaminants are lipophilic and tend to be stored in fatty tissues (Schlenk, 2005). Thus, it is likely that the chlordane levels in whole fish samples of the same species may be higher. Fish are likely to get exposed to pesticides through branchial, dermal, and oral (e.g., diet) routes. Environmental persistence, physicochemical properties, polarity, and lipid solubility of the different compounds appear to significantly modulate the uptake by the gills and the relative importance

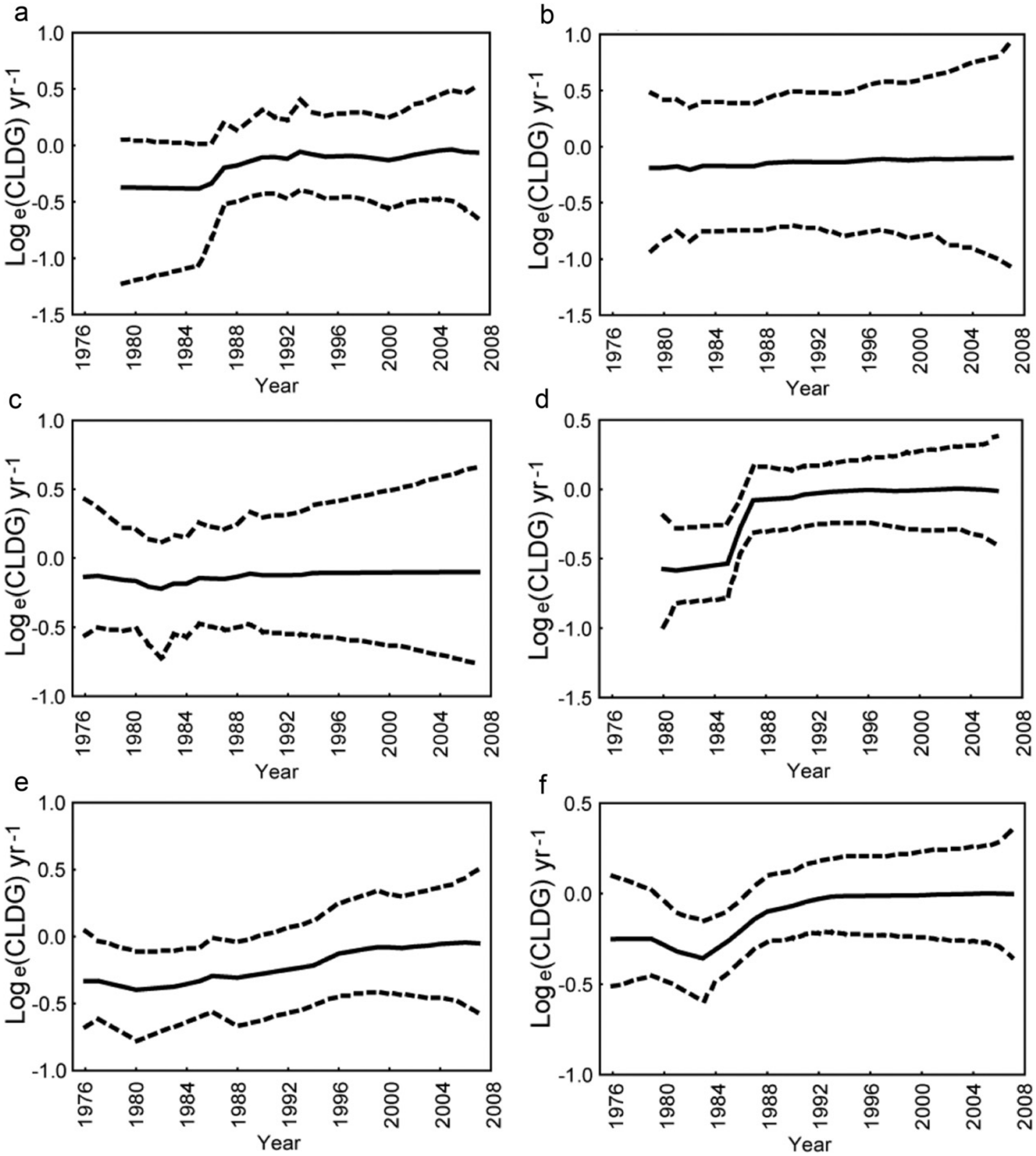

Fig. 3. Dynamic linear modeling analysis depicting the annual rates of change of γ-chlordane *(CLDG)* concentrations (ng/g wet weight) in (a) common carp, (b) channel catfish, (c) coho salmon, (d) freshwater drum, (e) rainbow trout, (f) smallmouth bass, (g) walleye, (h) white bass, (i) whitefish, and (j) yellow perch. The solid and dashed lines correspond to the median and the 95% posterior predictive intervals, respectively.

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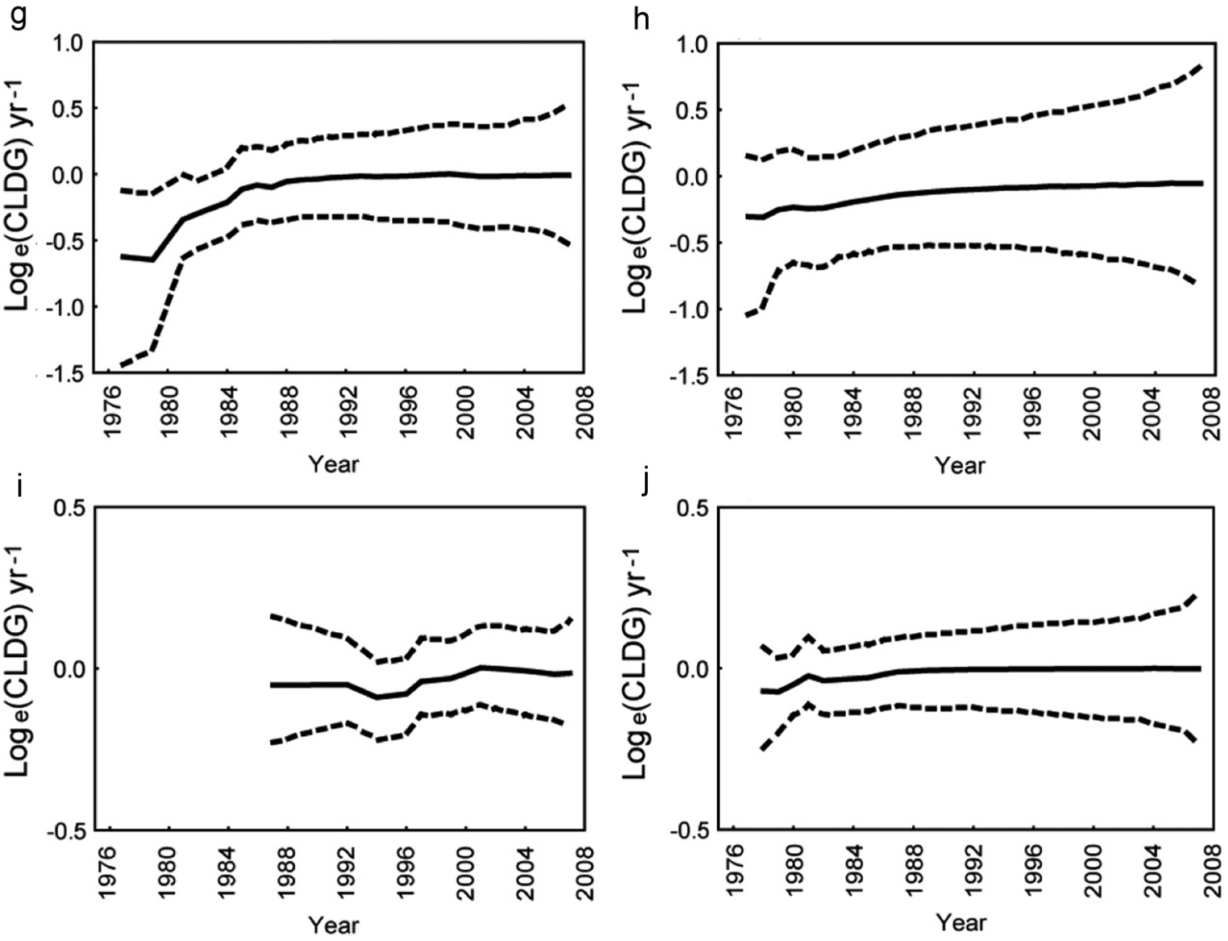

Fig. 3. (*continued*)

of the dermal or dietary exposure (Murty, 1986). We believe that the observed relatively higher concentrations in channel catfish, rainbow trout, and common carp can be related to their feeding habits, and may be further influenced by a range of factors including the life stages of fish, the nature and abundance of available food sources, and their behavioral or physiological patterns (Schlenk, 2005).

The above detection chlordane body burdens of some of the fish species examined can conceivably be attributed to the consumption of higher amounts of benthic invertebrates directly contaminated from the sediments. For example, common carp is a bottom dwelling fish species mainly feeding upon macroinvertebrates (Rahman et al., 2010). Channel catfish is an omnivorous opportunistic feeder, primarily feeding upon Trichoptera, Odonata, filamentous algae, chironomids, and aquatic Lepidoptera (Marsh, 1981; Tyus and Nikirk, 1990). Buktenica et al. (2007) identified five major food items of macroinvertebrate origin (i.e., Coleoptera, Trichoptera, Hymenoptera, Gastropoda, and Chironomids) in the guts of rainbow trout collected from Cater Lake, Oregon. The fate of chlordane in the sediments may be influenced by biodegradation processes as well as physical processes such as sedimentation and sediment mixing. The former process is predominant in depositional environments leading to a natural capping, whereby the contaminated layers can be buried from increasingly cleaner sediments over time (Yang et al., 2007). The sediment mixing (e.g., bioturbation and resuspension by bottom currents) can potentially negate the latter effect through redistribution of the contaminant vertical profiles (Arzayus et al., 2002). Yet, while the levels of different contaminants (mercury, *PCBs*, dioxins, and trace metals) in the Lake Erie sediments are well documented and reported to be declined (e.g., Painter et al., 2001; Marvin et al., 2004), historical data of chlordane levels in the Lake Erie sediments are lacking and therefore the importance of the causal link between the benthic community and the relatively high chlordane levels in some fish species still remains in the realm of speculation.

One possible explanation for the differences in the trajectories followed by the fish species examined may also be their different ability to biotransform lipophilic pesticides into more hydrophilic derivatives in an attempt to enhance polarity and to eliminate them from their body (Pyysalo et al., 1981; Schlenk, 2005). The primary organ for biotransformation in fish is the liver, but kidney, gut, and gill tissues are also responsible for significant extrahepatic activity. Fish have active Phases I and II biotransformation pathways to regulate the fate and toxicity of pesticides (Huckle and Millburn, 1990), but the nature and the role of specific enzymes on the formation of specific metabolites for the different pesticides are not fully known. Generally, Phase I reactions tend to enhance the solubility of the contaminant through the use of a polar functional group, such as the water or the molecular oxygen in monooxygenation (Stegeman and Hahn, 1994; Schlenk, 2005). Yet, Phase I reactions often result into the formation of reactive intermediate metabolites, which can be more biologically active than the original (parent) compounds. Phase II reactions can potentially alleviate this problem through the use of endogenous macromolecules with higher cellular concentrations that can conjugate the reactive intermediates, thereby enhancing water solubility and subsequent elimination from the body. In particular, both α- and γ-chlordane are

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reported to be metabolized into dichlorochlordene and oxychlordane, and these metabolic intermediates are further converted into the relatively steady compounds, 1-exo-hydroxy-2-chlorochlordene and 1-exo-hydroxy-2-endo-chloro-2,3-exo-epoxychlordene (Tashiro and Matsumura, 1977). Moreover, Tashiro and Matsumura (1977) reported another quite effective metabolic route for α-chlordane degradation that comprised more direct hydroxylation reactions. However, without any comparative studies on the chlordane levels and biotransformation rates of different fish species, a reliable conclusion cannot be drawn regarding the capacity of this hypothesis to explain some of the chlordane trends presented herein.

Despite the fact that the top predators are likely to accumulate higher levels of contaminants through the biomagnification processes (Morrison et al., 1998; Schlenk, 2005; Hogan et al., 2007), our study reports lower levels of chlordane in some top

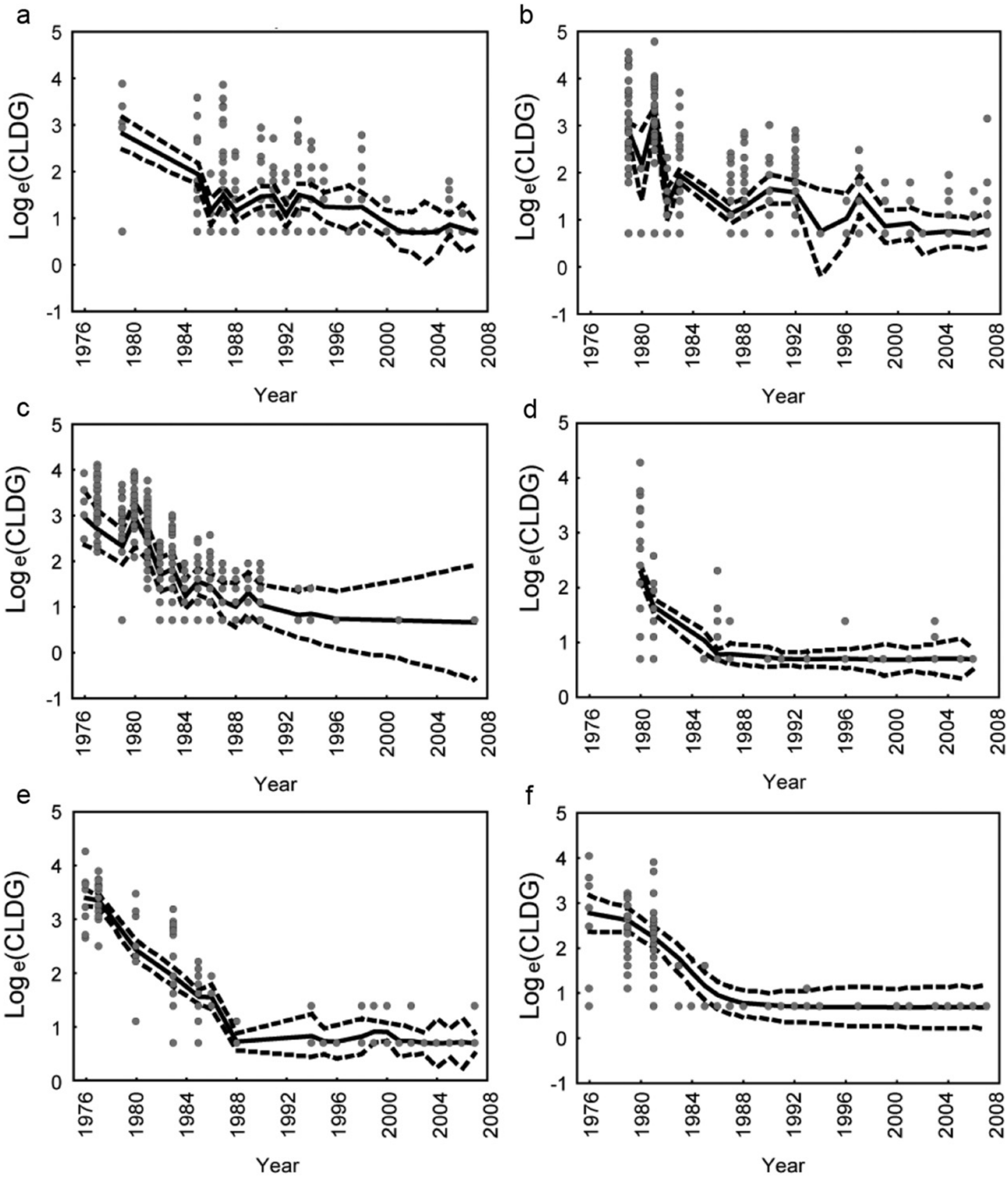

Fig. 4. Dynamic linear modeling analysis depicting the actual γ-chlordane *(CLDG)* concentrations (μg/g wet weight) (gray dots) against the predicted *CLDG* trends when accounting for the covariance with the fish length (black lines) in (a) common carp, (b) channel catfish, (c) coho salmon, (d) freshwater drum, (e) rainbow trout, (f) smallmouth bass, (g) walleye, (h) white bass, (i) whitefish, and (j) yellow perch in Lake Erie. The solid and dashed lines correspond to the median and the 95% posterior predictive intervals, respectively.

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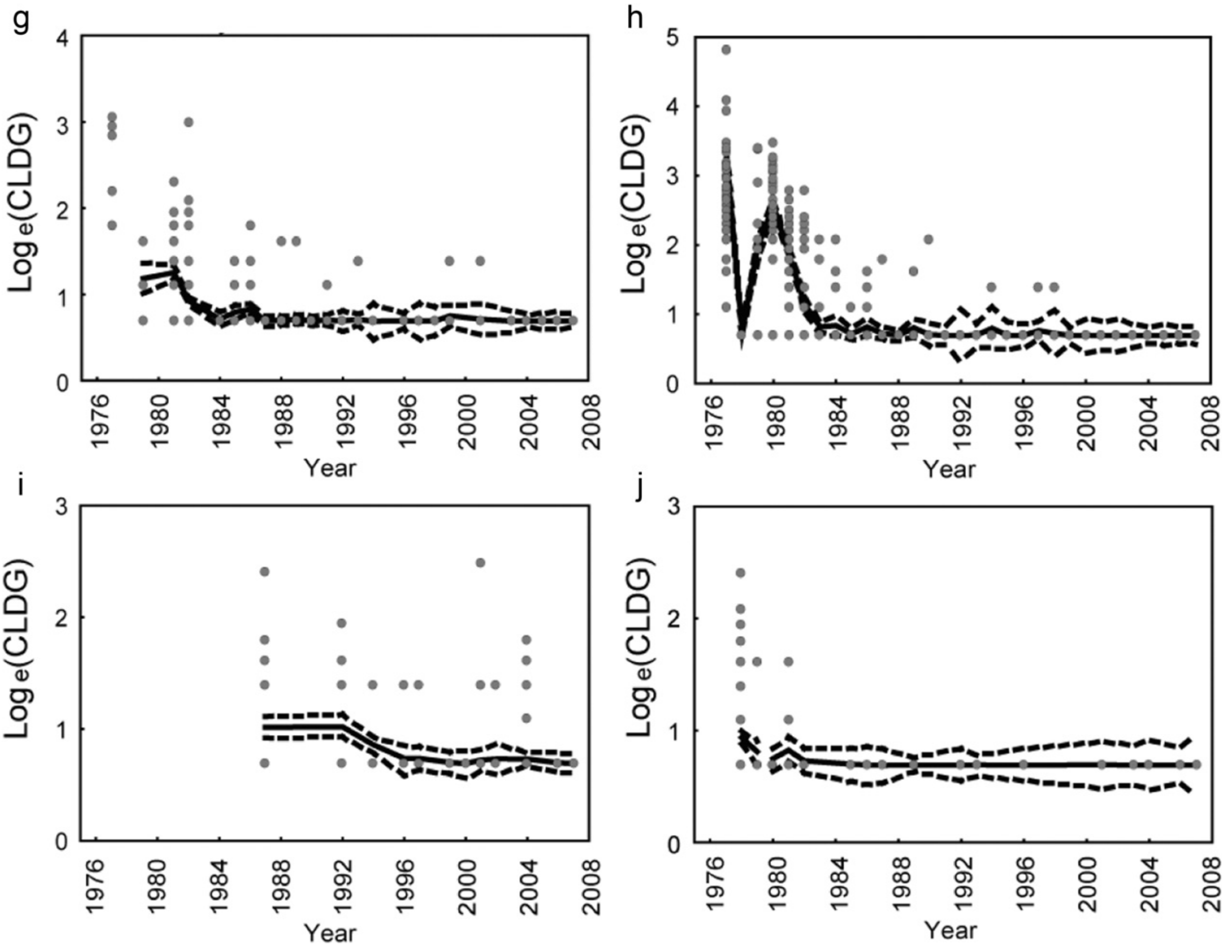

Fig. 4. (*continued*)

	Pre invasion	CLDA	Post invasion	Pre invasion	CLDG	Post invasion
Walleye	⬇		—	⬇		—
Yellow perch	—		—	↓		—
Smallmouth bass	⬇		—	⬇		—
Channel catfish	↓		↓	↓		↓
White bass	⬇		—	⬇		—
Freshwater drum	⬇		—	⬇		—
Coho salmon	↓		↓	↓		↓
White perch	↑		↓		—	
Rainbow trout	↓		↓	⬇		—
Whitefish	—		—	↓		—
Common carp	⬇		⬇	⬇		⬇

Fig. 5. Relative α- and γ-chlordane trends for different fish species in Lake Erie. For illustration purposes, the early 1990s are used as a reference point to distinguish between pre- and post-invasion periods of the food-web by exotic species (e.g., dreissenids and round goby).

predators, such as the walleye. This finding is on par with Zabik et al. (1995) low levels (30 ng/g ww) of the chlordane complex in skin-on walleye samples, which represented the mean size of fish caught in recreational fishing. One possible reason for the lower walleye burdens of chlordane may be the absence of highly contaminated food sources from their diet. Depending on the

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food availability, season of the year, and presence of other competitors, recent gut content analyses revealed that the principal food items identified in walleye include juveniles gizzard shad (*Dorosoma Cepedianum*), rainbow smelt (*Osmerus mordax*), and insect larvae (Bur et al., 2008; Wuellner et al., 2010). Although their prey gizzard shad are primarily known to be detritivores, there is evidence that they preferentially feed upon – the less contaminated – zooplankton when its abundance increases, which in turn may be associated with the reduced chlordane walleye concentrations (Yako et al., 1996; Schaus et al., 2002). Stable sulfur isotope (De Brabandere et al., 2009) and stoichiometric studies (Pilati and Vanni, 2007) consolidated the ontogenetic changes in the diet composition of gizzard shad with zooplankton during the larval and with detritus in the later stage. Interestingly, we note the recent occurrence of several α-chlordane values above the detection limit (i.e., the period after 2000 in Fig. 2g), although walleye appears to rarely eat the invasive – and supposedly more contaminated – round gobies (Bur et al., 2008).

In this regard, our modeling analysis also shows that the introduction of aquatic invasive species did not have any discernible influence on the temporal trends of both α- and γ-chlordane in Lake Erie. The lack of gut content data does not allow us to unequivocally conclude whether the fish species examined have changed their feeding patterns by incorporating invasive zebra mussels and/or round gobies into their diet. Yet, the continuously decreasing trends in fish species with high body burdens (e.g., coho salmon, channel catfish, rainbow trout, and common carp) suggests that even if such dietary shifts have occurred, they do not appear to predominantly shape their chlordane levels. Of equal importance is that several of the fish species (e.g., freshwater drum, smallmouth bass, and yellow perch) have experienced fast decline rates and ultimately reached close to detection values prior to the invasion of exotic species. Contrary to the recent increase in the *PCB* and mercury concentrations (Bhavsar et al. 2007, 2010; Azim et al., 2011; Sadraddini et al., in press; Sadraddini et al., submitted), the latter trend may also explain the lack of similar signals with chlordane in species that have been surmised to be affected by the invasion of round gobies (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% of the diet) as well as their well-documented higher growth rates have generated a hypothesis on a strong dreissenid–goby-smallmouth bass trophic linkage (Johnson et al., 2005; Hogan et al., 2007), which, however, does not appear to have significantly affected the smallmouth bass chlordane burdens. Likewise, Fernie et al. (2008) also did not find any effects of invasive species on the pesticides levels in water snakes collected from Lake Erie between pre-invasion (1990) and post-invasion (2003) periods. The same study reported significant declines or steady levels in heptachlore epoxide, oxychlordane, dieldrin, technical chlordane, and *DDE* concentrations from 1990 to 2003.

We also highlight that the present analysis differs from the 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. Yet, a recent revaluation exercise verified the robustness of the projected trends when we partial out the effects of the within-year variability, and instead we implement the same *DLM* using the annual means of the natural log-transformed data (Sadraddini et al., submitted). We further examined the sensitivity of the reported results on the high number of measurements below the detection limit using a Tobit modeling approach (Amemiya, 1973). In particular, this model 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 fell below the detection limit or not (see the corresponding model code in Appendix). By doing so, the Gibbs sampler samples the observations we have set below the detection limit from the tail of the distribution. This exercise indicated that the projected trends remain practically unaltered, even if we explicitly account for the impact of the below the detection limit chlordane values.

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. Nevertheless, we compared mean species lipid contents (average of different samples within the same species) and mean chlordane levels and found that the chlordane levels were even negatively correlated with lipid contents ($r=-0.41$). This indicates that the fish species with higher lipid content do not necessarily bioaccumulate pesticides, which somewhat contradicts the results reported by Stow et al. (1997) or – at least – stresses the need for more precise data to confirm this hypothesis. To this end, Elskus et al. (2005) suggested to use lipid class, instead of total lipid, as covariate so that the lipophilic/hydrophobic pollutant could be partitioned among the different lipid classes. In addition, it is uncertain whether the sampling locations have an influence on contaminant trends. The Western Basin is reported to be more polluted than the Eastern Basin, because it receives more contaminants from the Detroit River and its shallow depth also increases the sediment–water column interactions (Carter and Hites, 1992; Morrison et al., 2002). The data collected in the present analysis were also not well-distributed among the four different locations. The data from the Eastern Basin were underrepresented for the common carp (6 observations only in 2001) and channel catfish (6 observations only in 1988).

5. Conclusions

Our analysis showed that fish species (coho salmon, channel catfish, rainbow trout, and common carp) with higher chlordane body burdens demonstrated continuously decreasing trends of contaminants during the entire survey period (1976–2007). The predicted α- and γ-chlordane levels in freshwater drum, smallmouth bass, walleye, white bass, whitefish, and yellow perch decreased until the mid-1980s and remained at levels around the detection limit for the remaining period. The same fish species also exhibited the lower body concentrations of both contaminants. Based on the results presented herein and similar to the results reported for St. Clair River (Gewurtz et al., 2010), the temporal trends of the toxic compounds of the chlordane pesticide are unlikely to reach alarming levels for fish consumption ($>$59 ng/g).

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Appendix A. Dynamic linear modeling approach

The WinBUGS code associated with the dynamic linear model for the γ-chlordane (*CLDG*) concentrations is as follows:

```
model {
for (i in 1:N) {
lengthstdev[i] < -(length[i]-3.947061)/0.17152
LogCLDGm[i] < -level[time[i]+1]
  +beta[time[i]+1]*lengthstdev[i]
LogCLDG[i]~dnorm(LogCLDGm[i],mtau[time[i]+1])
LogPredCLDG[i]~dnorm(LogCLDGm[i],mtau[time[i]+1])
PredCLDG[i] < -exp(LogPredCLDG[i])}
for (t in 2:24) {
beta[year[t]]~dnorm(beta[year[t-1]],btau[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]])
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]])
}
beta[year[1]]~dnorm(beta[1],btau[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*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]])
beta[1]~dnorm(0,0.0001)
growth[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=1156,
year=c(3,5,6,8,9,10,11,12,13,14,15,16,17,18,20,
  21,22,23,25,27,28,29,30,31),
time=c(paste time.dat here),
LogCLDG=c(paste walleyeCLDG.dat here),
length=c(paste length.dat here),
```

Initial values 1

```
list(beta=c(0,NA,0,NA,0,0,NA,0,0,0,0,0,0,0,0,0,0,0,NA,0,
  0,0,0,NA,0,NA,0,0,0,0,0),
growth=c(0,NA,0,NA,0,0,NA,0,0,0,0,0,0,0,0,0,0,0,NA,0,0,
  0,0,NA,0,NA,0,0,0,0,0),
level=c(0,NA,0,NA,0,0,NA,0,0,0,0,0,0,0,0,0,0,0,NA,0,0,0,0,
  NA,0,NA,0,0,0,0,0),
mtau.in=0.2, ltau.in=1, btau.in=1, gtau.in=1,
LogPredCLDG=c(paste walleyeCLDG.dat here))
```

Initial values 2

```
list(beta=c(1,NA,1,NA,1,1,NA,1,1,1,1,1,1,1,1,1,1,1,NA,1,1,1,1,
  NA,1,NA,1,1,1,1,1),
growth=c(1,NA,1,NA,1,1,NA,1,1,1,1,1,1,1,1,1,1,1,NA,1,1,1,1,
  NA,1,NA,1,1,1,1,1),
level=c(1,NA,1,NA,1,1,NA,1,1,1,1,1,1,1,1,1,1,1,NA,1,1,1,1,NA,
  1,NA,1,1,1,1,1),
mtau.in=0.32, ltau.in=0.32, btau.in=0.32, gtau.in=0.32,
LogPredCLDG=c(paste walleyeCLDG.dat here))
```

Tobit Approach

```
model {
for (i in 1:N) {
upper.lim[i]  < - DETLIM*is.detlim[i]
  +UPPERLIM*(1 - is.detlim[i])
is.detlim[i]  < -step(0.693147181- LogCLDG[i])
lengthstdev[i] < -(length[i]-3.947061)/0.17152
LogCLDGm[i] < -level[time[i]+1]
  +beta[time[i]+1]*lengthstdev[i]
LogCLDG[i]~dnorm(LogCLDGm[i],mtau[time[i]+1])
  I(,upper.lim[i])
LogPredCLDG[i]~dnorm(LogCLDGm[i],mtau[time[i]+1])
PredCLDG[i] < -exp(LogPredCLDG[i])}
for (t in 2:24) {
beta[year[t]]~dnorm(beta[year[t-1]],btau[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]])
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]])
}
beta[year[1]]~dnorm(beta[1],btau[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*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]])
beta[1]~dnorm(0,0.0001)
growth[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
}
```

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Inference Data

```
list(N=1156,DETLIM=0.693147181,UPPERLIM=10000,
year=c(3,5,6,8,9,10,11,12,13,14,15,16,17,18,20,21,22,
  23,25,27,28,29,30,31),
time=c(paste time.dat here),
LogCLDG=c(paste walleyeCLDG.dat here),
length=c(paste length.dat here),
```

Initial values 1

```
list(beta=c(0,NA,0,NA,0,0,NA,0,0,0,0,0,0,0,0,0,0,
  NA,0,0,0,0,NA,0,NA,0,0,0,0,0), growth=c(0,NA,0,NA,0,0,
  NA,0,0,0,0,0,0,0,0,0,0,0,NA,0,0,0,0,NA,0,NA,0,0,0,0,0),
level=c(0,NA,0,NA,0,0,NA,0,0,0,0,0,0,0,0,0,0,NA,0,0,0,
  0,NA,0,NA,0,0,0,0,0),
mtau.in=0.2, ltau.in=1, btau.in=1, gtau.in=1,
LogPredCLDG=c(paste walleyeCLDG.dat here))
```

Initial values 2

```
list(beta=c(1,NA,1,NA,1,1,NA,1,1,1,1,1,1,1,1,1,1,NA,1,1,
  1,1,NA,1,NA,1,1,1,1,1),
growth=c(1,NA,1,NA,1,1,NA,1,1,1,1,1,1,1,1,1,1,NA,1,1,1,
  1,NA,1,NA,1,1,1,1,1),
level=c(1,NA,1,NA,1,1,NA,1,1,1,1,1,1,1,1,1,1,NA,1,1,1,1,
  NA,1,NA,1,1,1,1,1),
mtau.in=0.32, ltau.in=0.32, btau.in=0.32, gtau.in=0.32,
LogPredCLDG=c(paste walleyeCLDG.dat here))
```

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