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Examination of temporal DDT trends in Lake Erie fish communities using dynamic linear modeling

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ABSTRACT

The industrial pesticide dichlorodiphenyltrichloroethane (DDT) was initially heralded for its effectiveness against malaria and agricultural pests, but was eventually banned in North America during the 1970s due to growing concerns about its detrimental impacts on wildlife. Despite the successful termination of its commercial application, the persistent and bioaccumulative nature of DDT has resulted in lingering concentrations in aquatic food webs, particularly in upper trophic-level fish species. In this study, we used dynamic linear modeling to examine temporal trends of four DDT compounds (*p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDE, and *p,p′*-DDD) in nine fish species in Lake Erie from 1976 to 2007, while considering both fish length and lipid content as covariates. Our results indicate that the levels of both *p,p′*-DDT and *o,p′*-DDT have been decreasing, often to the detection limit, with slowing decline rates during the second half of the study period. The *p,p′*-DDE levels were much more variable, exhibiting large fluctuations through time (though usually with a net downward trajectory), with the annual rates of change of the corresponding concentrations remaining negative or (more recently) near zero. Similarly, *p,p′*-DDD levels fluctuated (though to a lesser degree) over time, with gradually slowing decline rates in many fish species, such as smallmouth bass and freshwater drum. The results are in agreement with our understanding that DDE and DDD are degradation products of *p,p′*-DDT, and thus continue to be produced, as DDT is broken down. Declining trends observed for nearly all congeners and fish species indicate reduction of DDT risks in the Lake Erie fish communities.

Introduction

The organochlorine pesticide dichlorodiphenyltrichloroethane (DDT) is marked by its environmental stability, high potential to bioaccumulate in fatty tissues, and slow degradation time in natural systems (Turusov et al., 2002; Wang and Wang, 2005). Even though the production and release of this "persistent organic pollutant" (POP) has now been widely banned, it was initially heralded as a deterrent against typhus in the Second World War, as an effective means for malaria control in susceptible countries, and later as a popular agricultural insecticide (Seagren, 2005; Turusov et al., 2002). Starting in the 1950s, however, concerns arose about the lingering environmental impacts of the compound, including thinning avian eggshells and reproductive failure (Beard, 2005), endocrine disruption in various species (de Solla et al., 1998), and its role as a possible carcinogen (Turusov et al., 2002). The lipophilicity and stability of DDT and its primary metabolite dichlorodiphenyldichloroethylene (DDE) were responsible for significant biomagnification patterns and increased health risks for top predators, such as piscivorous fish, birds, and humans (ATSDR, 2002; International Agency for Research on Cancer, IARC/WHO, 1991; Ssebugere et al., 2009). Within biological organisms, DDE is also an extremely stable compound, and typically represents the predominant congener stored in tissues as we proceed up the food chain (NRC, 1977).

The mounting evidence against the commercial use of DDT prompted its ban in the 1970s, with some exceptions made under the 1991 Stockholm Convention for disease control in vulnerable countries (Turusov et al., 2002; USEPA, 2009a). Within North America, the Great Lakes system has been particularly susceptible to inputs of persistent contaminants, given the long hydraulic residence times (DeVault et al., 1996). DDT and its degradation compounds DDE and DDD were detected early on in fish and wildlife of the region (Carlson et al., 2010). Lakes Michigan, Ontario and Huron have historically exhibited the highest fish DDT levels, due to the increased loading from the surrounding agricultural and industrial areas (Hickey et al., 2006). Growing concerns about these elevated DDT concentrations and the observed impacts on Great Lakes wildlife (e.g., the bald eagle) prompted a ban by the USEPA in 1972, with target DDT concentrations established for water and fish under the revised Great Lakes Water Quality Agreement in 1978 (IJC, 1978).

Despite the successful curtailment of external emissions, DDT and its metabolites still linger in the Great Lakes ecosystem, particularly in top predators (Carlson et al., 2010). Fish have historically been

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used as indicators of system health, given their higher position in the food web as well as their important link with human consumers (Bhavsar et al., 2007; Carlson et al., 2010). The consumption of contaminated Great Lakes sport fish has been considered the most significant route of POP exposure in humans (Kearney et al., 1999) and has been linked to significant DDE human body burdens (Hanrahan et al., 1999; Hovinga et al., 1992). Even though DDT is not acutely toxic to humans, there is evidence of neurobehavioral and reproductive impacts, endocrine disruption, and possible carcinogenicity (Turusov et al., 2002). As such, contaminant levels in Great Lakes fish have been routinely monitored since the 1970s, with species- and location-specific fish advisories regularly produced to ensure safe sport fish consumption (Bhavsar et al., 2011). These monitoring programs have been invaluable in providing long-term datasets to examine contaminant trends and to assess the success of our remedial efforts (Bhavsar et al., 2007, 2008; Carlson and Swackhamer, 2006).

In general, the implementation of bans lowered the contaminant levels in Great Lakes biota during the 1980s, but the rates of decline have been slowing in recent years, particularly in Lake Erie (Azim et al., 2011a,b; Bhavsar et al., 2010, 2007; Carlson et al., 2010; Sadraddini et al., 2011a,b). Possible explanations for these shifting trends include the food web alterations induced by invasive species (Hogan et al., 2007), the impacts of global warming (French et al., 2006), and the role of sediments (Hickey et al., 2006; Stow et al., 2004). This general pattern also holds true for total DDT in Great Lakes biota, as the initial rapid response to the ban was followed by reduced rates of decline by the end of the 1980s (Baumann and Whittle, 1988). Total DDT, when applied as a pesticide, is composed of up to fourteen compounds, including *p,p′*-DDT (the predominant form), *o,p′*-DDT, *p,p′*-DDE and *p,p′*-DDD (Metcalf, 1995). The latter two compounds are the primary metabolites and degradation products in the environment (ATSDR, 2002). As such, several studies have revealed higher fish concentrations of *p,p′*-DDE in recent years compared to the previously dominant *p,p′*-DDT form (ATSDR, 2002; Carlson and Swackhamer, 2006). High DDE levels in organisms are generally considered to be indicative of long-term exposure to DDT/DDE, while high DDT levels suggest recent exposure (Mariën et al., 1998). Recent studies have also shown variations in DDT trends among the five Great Lakes, based on differences in individual lake dynamics, external loading conditions, and ecological attributes (Hickey et al., 2006; USEPA, 2011).

The presence of inter-lake variability on DDT trends highlights a key aspect of retrospective analyses that is often overlooked. Namely, although the temporal trend assessment alone can offer valuable insights into contaminant dynamics within a water-body, it is also vital to consider the causal factors that may obfuscate the impartial assessment of patterns in both time and space (Azim et al., 2011a,b; Sadraddini et al., 2011a,b). In particular, such trends may be influenced by contaminant properties or fish characteristics such as age, diet, trophic level, reproductive status, growth, and lipid content (Sadraddini et al., 2011a,b; Stow et al., 1997). Data collection over time has not always been consistent (Bhavsar et al., 2011), and so variations in sampling procedures and the type of statistical analyses selected may interfere with a comprehensive assessment of contaminant trends (Sadraddini et al., 2011b). Hence, it is imperative to select quantitative approaches that explicitly account for these causal factors, thereby ensuring that the true contaminant patterns are revealed. One such statistical approach is dynamic linear modeling (*DLM*), which employs an adaptive structure that allows time variant parameters and provides forecasts influenced by recent rather than distant data (Lamon et al., 1998; Pole et al., 1994). The evolving nature of the *DLM* analysis allows greater insights into cause–effect relationships (Lamon et al., 1998), and thus offers an excellent tool for assessing complex ecological patterns.

Here, we use *DLM* to conduct retrospective analysis of the temporal variability of DDT and its metabolites. In particular, our aim is to offer a post-audit exercise that is not intended for forecasting purposes, but rather an attempt to take a look back at fish contaminant changes that have already occurred and draw inference about the efficiency of restoration strategies in the Great Lakes area. The specific objective of our analysis is to critically examine temporal trends of four DDT congeners (*p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDE and *p,p′*-DDD) in nine Lake Erie fish species over an approximately thirty-year period (1976–2007). In our modeling framework, we consider both fish length and lipid content as covariates, thus accounting for the fact that fish of different lengths and lipid content may have been sampled over time (Bhavsar et al., 2011). Following our examination of compound-specific trends, we consider ecological mechanisms that may have played a role in modulating contaminant variability within the Lake Erie ecosystem.

Methods

Dataset description

Our study uses Lake Erie fish DDT concentrations from the Ontario Ministry of the Environment (OMOE), which routinely collects fish contaminant data in partnership with the Ontario Ministry of Natural Resources for a wide range of fish species. The information in this dataset, covering a time span of approximately three decades (1976–2007), has been used in issuing fish consumption advisories for the Canadian waters of Lake Erie (OMOE, 2013). We selected fish species based on data availability and/or the species' commercial importance. For each DDT congener, we examined nine fish species: channel catfish (*Ictalurus punctatus*), common carp (*Cyprinus carpio*), coho salmon (*Oncorhynchus kisutch*), freshwater drum (*Aplodinotus grunniens*), rainbow trout (*Oncorhynchus mykiss*), smallmouth bass (*Micropterus dolomieui*), walleye (*Stizostedion vitreum*), white bass (*Morone chrysops*) and white perch (*Morone americana*). All samples were skin-less boneless fillets of fish collected from various locations in the Canadian waters of Lake Erie (Fig. 1). The trophic position of fish species and their feeding patterns can impact the levels of DDT bioaccumulation, with this relationship varying depending on the contaminant considered and the specific food web in question. An overview of the Lake Erie food web indicates that walleye, smallmouth bass, white bass and rainbow trout are characterized as higher level piscivorous fish, while channel catfish, common carp, freshwater drum and white perch are classified as forage fish.

Chemical analysis

The samples were analyzed for DDT, DDE, and DDD content at OMOE laboratories in Toronto using gas liquid chromatography-electron capture detection (GLC-ECD), in accordance with OMOE method PFAOC-E3136 (OMOE, 2007). Results for these compounds (ng/g) and lipid content (%) are expressed on a wet weight basis.

Modeling framework

We developed a series of *DLM*s to examine temporal trends of the four DDT 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. To compare the relative influence of each of these covariates, we ran the *DLM*s 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 144 (9 fish species × 4 compounds × 4 covariate combinations) models 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"

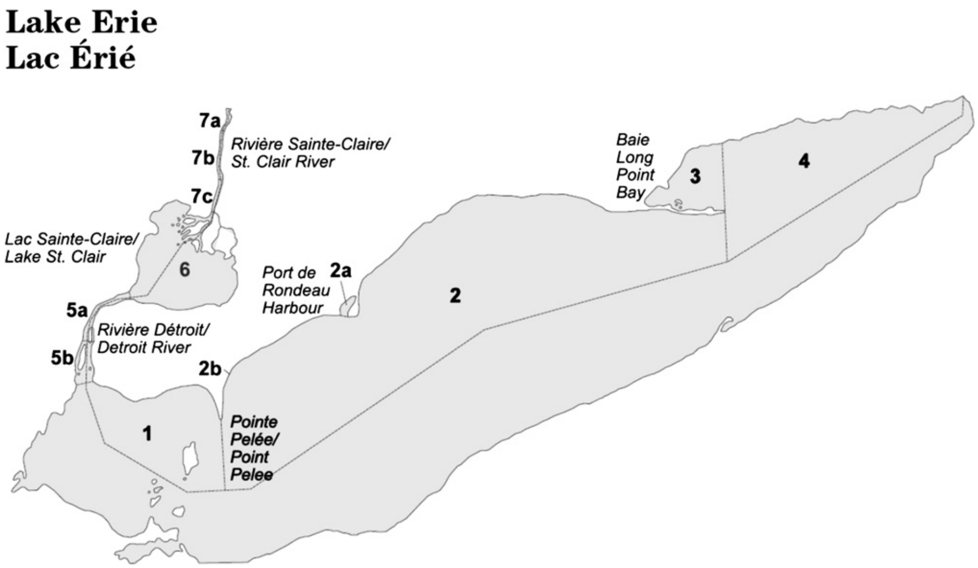


Fig. 1. Map of Lake Erie with the four sampling sites: 1: Western Basin, 2: Central Basin, 3: Long Point Bay, and 4: Eastern Basin. **[Taken from OMOE, 2013.]**

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 (Lamon et al., 1998). An important feature 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; Stow et al., 2004). *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; Stow et al., 2004). Furthermore, *DLM*s minimize the impact of outliers and easily handle missing values or unequally spaced data (Pole et al., 1994). Parameters in these models are time-specific, but are also related to one another stochastically by virtue of an error term (Stow et al., 2004).

The main components of any *DLM* are an observation equation and subsequent system equations. For the sake of brevity, we outline below the model considering both fish length and lipid content as covariates:

Observation equation:

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

System equations:

$$\begin{array}{ll}
level_t = level_{t-1} + rate_t + \omega_{t1} & \omega_{t1} \sim N\,[0, \Omega_{t1}] \\
rate_t = rate_{t-1} + \omega_{t2} & \omega_{t2} \sim N\,[0, \Omega_{t2}] \\
\beta_{t1} = \beta_{t1-1} + \omega_{t3} & \omega_{t3} \sim N\,[0, \Omega_{t3}] \\
\beta_{t2} = \beta_{t2-1} + \omega_{t4} & \omega_{t4} \sim N\,[0, \Omega_{t4}] \\
1/\Omega^2_{tj} = \zeta^{t-1} \cdot 1/\Omega^2_{1j},\ 1/\Psi^2_t = \zeta^{t-1} \cdot 1/\Psi^2_1 & t > 1 \text{ and } j = 1 \text{ to } 4 \\
level_1, rate_1, \beta_1 \sim N(0, 10000) & t = 1 \\
1/\Omega^2_{1j}, 1/\Psi^2_1 \sim G(0.001, 0.001) &
\end{array}$$

where $\ln[\mathrm{DDT}]_{ti}$ is the observed DDT concentration at time t in the individual sample i; $level_t$ is the mean DDT concentration at time t when accounting for the covariance with the fish length and lipid content; $\ln[length]_{ti}$ is the observed (standardized) fish length at time t in the individual sample i; $\ln[lipid]_{ti}$ is the observed (standardized) fish lipid content; $rate_t$ is the rate of change of the level variable; β_{t1} is a length (regression) coefficient; β_{t2} is a lipid (regression) coefficient; ψ_t, ω_{tj} are the error terms for year t sampled from normal distributions with zero mean and variances Ψ^2_t, Ω^2_{tj}, respectively; the discount factor ζ represents the aging of information with the passage of time; $N(0, 10000)$ is the normal distribution with mean 0 and variance 10000; 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}, $\beta_{21,}$ $1/\Omega^2_{1j}$, and $1/\Psi^2_1$ 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 (Lamon et al., 1998). In this analysis, we introduce non-constant and data-driven variances (with respect to time) using a discount factor on the first period prior (Congdon, 2003). 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 and 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. 2; see also the model code in the Electronic Supplementary Material or ESM available on-line).

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 20–40%. For each analysis, we used two chain runs of 200 000 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

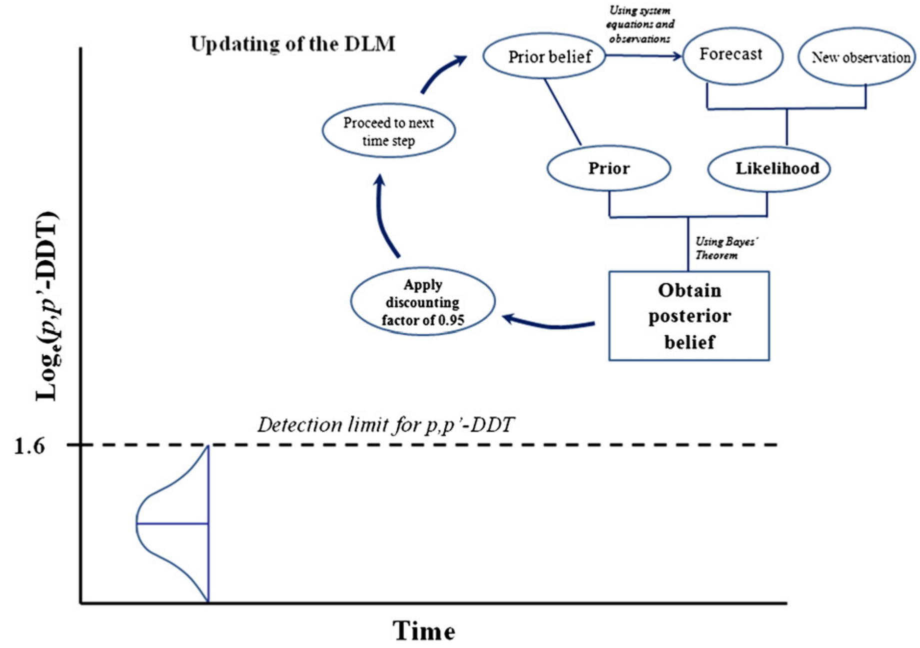


Fig. 2. Basic concepts of Tobit dynamic linear modeling: the sequential flow allows parameter values to vary over time by discounting older observations relative to more recent ones. Measurements below the detection limit are treated as random draws from a normal distribution parameterized such that 99% of their values are lying within the analytical and actual zero. Subsequently, the 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. Thus, the Gibbs sampler samples the observations we have set below the detection limit from the tail of the distribution.

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).

3. Results

Observed DDT levels

For each DDT congener, we report the summary statistics of the observed concentrations (ng/g wet weight or ww) of the nine fish species examined (Table 1). Further, the variability of DDT congener concentrations across four sampling sites for white bass, the most extensively studied species in Lake Erie, is shown in on-line Fig. ESM-1. For the purpose of clarity, we address each congener separately:

p,p′-DDT

The highest concentrations of this DDT constituent were found in coho salmon (mean 15.14 ng/g), followed by rainbow trout (11.07 ng/g) and channel catfish (9.86 ng/g). Conversely, the lowest concentrations were found in white perch (5.05 ng/g), with walleye (5.23 ng/g), common carp (5.81 ng/g) and freshwater drum (5.96 ng/g) which all exhibited similarly low values. It is worthwhile to note that the majority of the concentrations in the recent sampling years were recorded at the minimum detection limit of 5 ng/g; thus, the median concentration of *p,p′*-DDT for all fish species (except coho salmon) is that value. Coho salmon was sparsely sampled after the mid-1990's (merely six observations over two years) and thus the bulk of its data comes from measurements in the more variable earlier years; therefore, its median concentration is less affected by this drop to the detection limit and stands higher than the rest at 8 ng/g.

o,p′-DDT

Similar to *p,p′*-DDT, the highest concentrations of this constituent were found in coho salmon (mean 9.46 ng/g); the order of the next two fish species was reversed, with channel catfish (8.61 ng/g) demonstrating higher values than rainbow trout (8.20 ng/g). The lowest concentrations were again found in white perch (~5.00 ng/g), walleye (5.09 ng/g), common carp (5.73 ng/g), and freshwater drum (5.76 ng/g). As before, the median value for all nine fish species was 5 ng/g; this time, however, the pattern also includes coho salmon, whose *o,p′*-DDT concentrations reached the detection limit well before the sparse data sampling of the later years.

p,p′-DDE

The summary statistics for this congener differed in two ways from the previously examined *ortho*- and *para*-DDT forms (Table 1): the mean values were well above the *p,p′*- and *o,p′*-DDT concentrations, and the medians did not remain at the corresponding detection limit (1 ng/g). For *p,p′*-DDE, the highest concentrations were found in common carp (mean 93.71 ng/g, median 52.5 ng/g), followed by channel catfish (mean 55.92 ng/g, median 33 ng/g) and coho salmon (mean 43.51 ng/g, median 35 ng/g). In contrast, walleye (mean 8.87 ng/g, median 4 ng/g) and white perch (mean 10.72 ng/g, median 9 ng/g), demonstrated the lowest concentrations of this compound (Table 1).

p,p′-DDD

Similar to the other degradation product *p,p′*-DDE, the mean concentrations of *p,p′*-DDD were much higher than the DDT congeners examined (Table 1); the highest values were found in channel catfish (mean 34.88 ng/g, median 15 ng/g), followed by common carp (mean 32.57 ng/g, median 16 ng/g) and coho salmon (mean 25.48 ng/g, median 16 ng/g). Again, the lowest values were found in walleye and white perch, both with mean 5.94 ng/g and median 5 ng/g. The median values for *p,p′*-DDD were variable, ranging from 16 ng/g down to the detection limit of 5 ng/g.

For the majority of the congeners, our results indicate high standard deviation values, which serve as evidence of considerable inter- and intra-annual variability in the DDT levels among the fish species; especially in the earlier years. The generally positive skewness and kurtosis values indicate right-skewed and leptokurtic distributions;

Table 1
Basic statistics in DDT concentrations (four congeners) in skinless–boneless fillet data (ng/g wet weight) for nine fish species in Lake Erie (study period 1976–2007). Best models are based on the 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.

	DDT congener	N	Mean	SD	Median	IQR	Skew-ness	Kurtosis	Best model*
Channel catfish	*p,p′*-DDT	533	9.86	15.64	5.00	1.00	6.06	50.21	L + L
Ictalurus punctatus	*o,p′*-DDT	533	8.61	12.07	5.00	1.00	4.53	23.63	L + L
	p,p′-DDE	533	55.92	72.38	33	53	3.12	13.78	L + L
	p,p′-DDD	533	34.88	48.09	15.00	35.00	2.8	9.84	L + L
Common carp	*p,p′*-DDT	374	5.81	5.27	5.00	0.00	10.42	124.56	L + L
Cyprinus carpio	*o,p′*-DDT	374	5.73	4.97	5.00	0.00	11.54	162.73	LNG
	p,p′-DDE	374	93.71	129.76	52.5	87	3.73	19.00	L + L
	p,p′-DDD	374	32.57	54.75	16.00	27.00	4.67	29.26	L + L
Coho salmon	*p,p′*-DDT	634	15.14	18.81	8.00	11.0	3.18	12.12	L + L
Oncorhynchus kisutch	*o,p′*-DDT	634	9.46	10.43	5.00	3.00	3.57	17.01	L + L
	p,p′-DDE	634	43.51	34.49	35	36	2.29	11.28	L + L
	p,p′-DDD	634	25.48	29.8	16.00	21.00	3.28	14.7	L + L
Freshwater drum	*p,p′*-DDT	359	5.96	5.17	5.00	0.00	7.37	60.52	L + L
Aplodinotus grunniens	*o,p′*-DDT	359	5.76	5.04	5.00	0.00	11.74	164.26	L + L
	p,p′-DDE	360	12.21	17.45	6.5	11	5.11	43.39	L + L
	p,p′-DDD	359	7.78	8.55	5.00	0.00	4.73	26.41	L + L
Rainbow trout	*p,p′*-DDT	307	11.07	15.94	5.00	2.00	3.30	11.49	L + L
Oncorhynchus mykiss	*o,p′*-DDT	308	8.20	9.66	5.00	0.00	3.91	17.46	L + L
	p,p′-DDE	308	41.05	33.10	32.5	34.25	2.21	7.71	L + L
	p,p′-DDD	308	22.47	32.41	10.00	18.00	2.84	8.89	L + L
Smallmouth bass	*p,p′*-DDT	417	8.09	12.32	5.00	0.00	9.51	119.75	L + L
Micropterus dolomieui	*o,p′*-DDT	417	6.90	7.90	5.00	0.00	9.03	112.25	L + L
	p,p′-DDE	417	27.26	34.42	15	26	2.99	12.15	L + L
	p,p′-DDD	417	11.99	17.70	5.00	5.00	4.57	29.36	L + L
Walleye	*p,p′*-DDT	900	5.23	1.63	5.00	0.00	12.39	204.97	LPD
Stizostedion vitreum	*o,p′*-DDT	900	5.09	0.98	5.00	0.00	12.93	178.04	LPD
	p,p′-DDE	900	8.87	29.56	4.00	6.00	13.21	200.96	L + L
	p,p′-DDD	900	5.94	4.14	5.00	0.00	7.98	80.38	L + L
White bass	*p,p′*-DDT	1158	6.96	7.11	5.00	0.00	6.36	51.14	L + L
Morone chrysops	*o,p′*-DDT	1158	6.64	7.04	5.00	0.00	7.84	80.87	L + L
	p,p′-DDE	1158	17.10	15.99	13	15	2.80	512.87	L + L
	p,p′-DDD	1158	12.66	22.72	5.00	6.00	6.46	51.51	L + L
White perch	*p,p′*-DD	348	5.05	0.91	5.00	0.00	13.07	214.7	L + L
Morone americana	*o,p′*-DDT	348	5.02	0.32	5.00	0.00	−4.54	84.74	L + L
	p,p′-DDE	348	10.72	8.19	9.00	7.25	2.68	11.88	L + L
	p,p′-DDD	348	5.94	2.76	5.00	0.00	4.28	22.68	L + L

we thus applied a natural log transformation to the data before commencing our *DLM* analysis.

DLM analysis

As previously mentioned, a total of 144 dynamic linear models were run over the course of this study. We examined the same nine fish species across each of the DDT congeners, running four separate models for each combination ("random walk" or no covariate, fish length or lipid content as the sole covariate, and both fish length/lipid content as covariates). The variability of lipid content (%) for each of the nine fish species examined is shown in Fig. ESM-2 (ESM figures available on-line).

To determine the most parsimonious model for each fish species/congener combination, we compared the Deviance Information Criterion (*DIC*) values of the four combinations; the *DIC* is a Bayesian measure of model fit and complexity, where models with lower *DIC* values are expected to effectively balance between predictive capacity and complexity (Spiegelhalter et al., 2003). Our results suggest that across all the DDT congeners, the most favorable model was the one that considered both fish length and lipid content as covariates; exceptions were the walleye models for *p,p′*-DDT and *o,p′*-DDT (lipid content rendered the most parsimonious model) and common carp for *o,p′*-DDT (length model was superior) (see last column of Table 1). Generally, the temporal trends of the four DDT congeners followed one of two broader patterns. The *ortho*- and *para*-DDT forms were characterized by decreasing levels (often down to the detection limit), with rates of change remaining negative but gradually slowing through time. The two degradation products *p,p′*-DDE and *p,p′*-DDD exhibited downward yet fluctuating levels over the thirty year study period, with decline rates of variant degree depending on the species in question. We thus lumped the congeners into two groups for the following examination of the *DLM* results.

p,p′-DDT/o,p′-DDT

The levels of both of these congeners decreased through time down from 20–50 ng/g ww to the detection limit of 5 ng/g ww across all species (Fig. 3). For *p,p′*-DDT, the sharpest decline in the corresponding levels occurred from the beginning of the sampling period until the mid-1980s, as seen for channel catfish, common carp, coho salmon, rainbow trout, and white bass (Fig. 3a–c,e,h). The associated rates of change for these species indicate faster decline rates in the earlier years, followed by slowing decreasing rates as we progress through time (Fig. ESM-3). Walleye demonstrated marginally positive rates of change in the latter years, reflecting the small fluctuations in the recent *p,p′*-DDT level (Fig. 3g, Fig. ESM-3g). The levels of white perch were generally stable over the sampling years studied (Fig. 3i, Fig. ESM-3i), although the derived patterns are quite uncertain as the existing information from the system is somewhat sporadic. Similar to *p,p′*-DDT, *o,p′*-DDT concentrations generally declined until the mid-1980s and hovered around the detection limit thereafter, with characteristic examples being the channel catfish, common

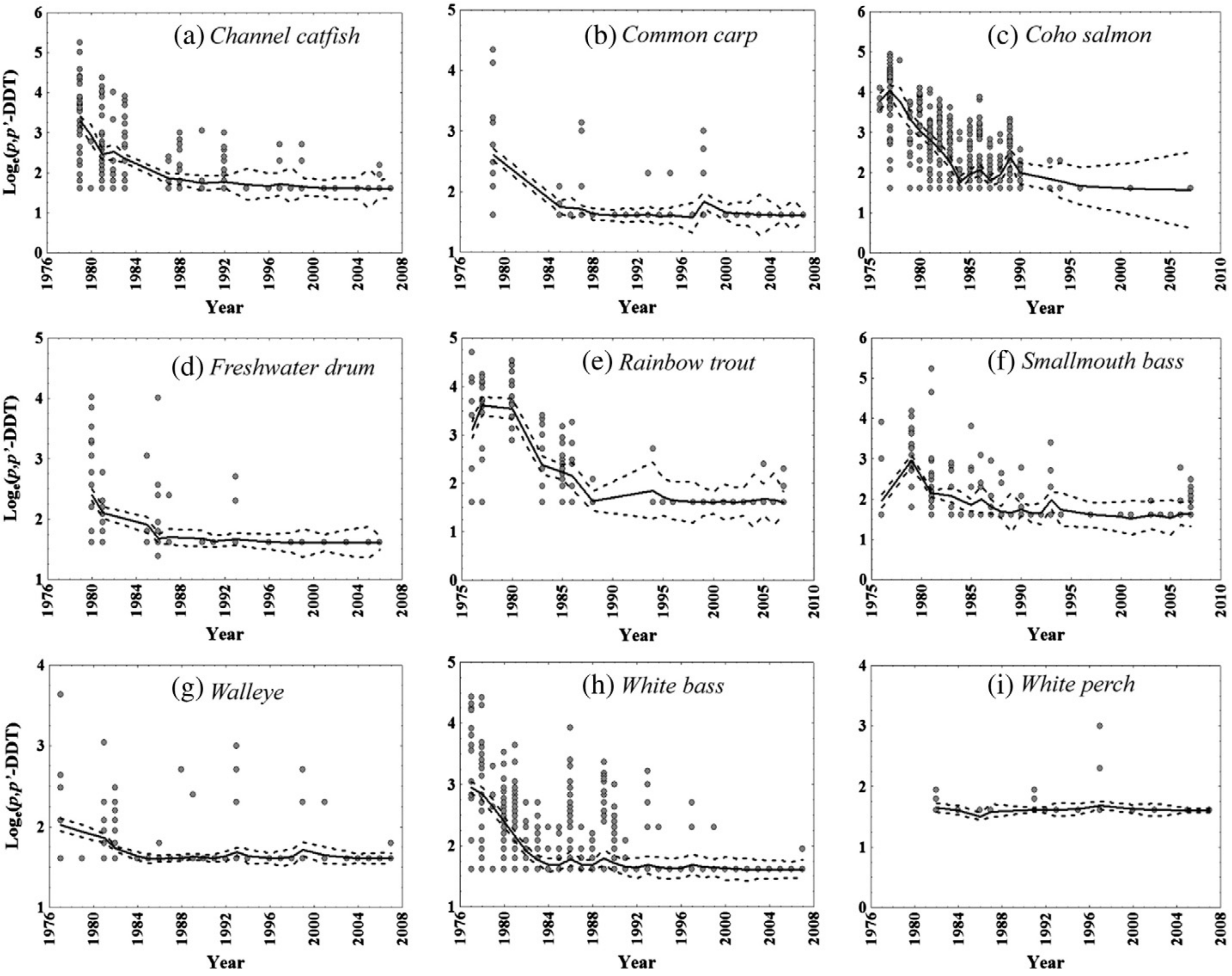


Fig. 3. Dynamic linear modeling analysis depicting the actual *p,p*′-DDT concentrations (ng/g wet weight) (gray dots) against the predicted *p,p*′-DDT trends when accounting for the covariance with the fish length and lipidcontent (black lines) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) freshwaterdrum, (e) rainbow trout, (f) smallmouth bass, (g) walleye, (h) white bass, and (i) white perch 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 the observation equation, respectively.

carp, coho salmon, freshwater drum, rainbow trout and white bass (Fig. 4a–e, h). The corresponding rates of change followed the same pattern of early rapid decreases followed by lower decline rates in the later years (Fig. ESM-4). Walleye and white perch generally demonstrated stable levels after the 1980s (Fig. 4g,i), and the corresponding rates of change were lying close to zero for most of the study period (Fig. ESM-4g,i).

p,p′*-DDE/p,p*′*-DDD*

In contrast to the (nearly) monotonic declining trends of the previous two congeners, the levels of *p,p*′-DDE and *p,p*′-DDD were characterized by large fluctuations during the sampling period (mirroring the scatter in the observed data), although the overall net changes were usually negative. For *p,p*′-DDE, these discernibly variant levels were seen for channel catfish, coho salmon, rainbow trout, smallmouth bass, walleye and white bass (Fig. 5a,c,e–h), with minute initial increases in the concentrations observed for channel catfish, rainbow trout and white bass. The corresponding rates of change for these species were weakly negative and fairly stable through time (Fig. ESM-5a, c,e–h). The remaining species (common carp, freshwater drum and white perch) also demonstrated the same trend of fluctuating levels, but their oscillations were less pronounced (Fig. 5b,d,i); the net rates of change of the corresponding concentrations remained relatively stable around zero throughout the study period (Fig. ESM-5b,d,i).

Our analysis revealed distinctly decreasing trends in the *p,p*′-DDD concentrations, which were also subject to small-amplitude fluctuations. Channel catfish, common carp and rainbow trout exhibited downward fluctuating levels throughout the sampling period (Fig. 6a,b,e), and the corresponding rates of change remained negative but were slowing over time (Fig. ESM-6a,b,e). Freshwater drum, smallmouth bass, walleye, white bass and white perch were characterized by smaller fluctuations, leading to fairly stable concentrations around the detection limit (Fig. 6d,f–i). Similarly, the rates of change for these species revealed initial rapid decline rates, followed by values that gradually approached to zero (Fig. ESM-6d,f–h); the exception was white perch, whose rates hovered around zero throughout the sampling period (Fig. ESM-6i). Lastly, coho salmon demonstrated rapidly downward fluctuating concentrations in the early years, followed by a smooth decrease after the early 1990s. However, as Fig. 6c clearly shows, data collection became more sparse in the latter period and may be compromising our capacity to objectively tease out the *p,p*′-DDD temporal trends. The rates of change for coho salmon slowed until the 1990s, and then stabilized for the remaining years (Fig. ESM-6c).

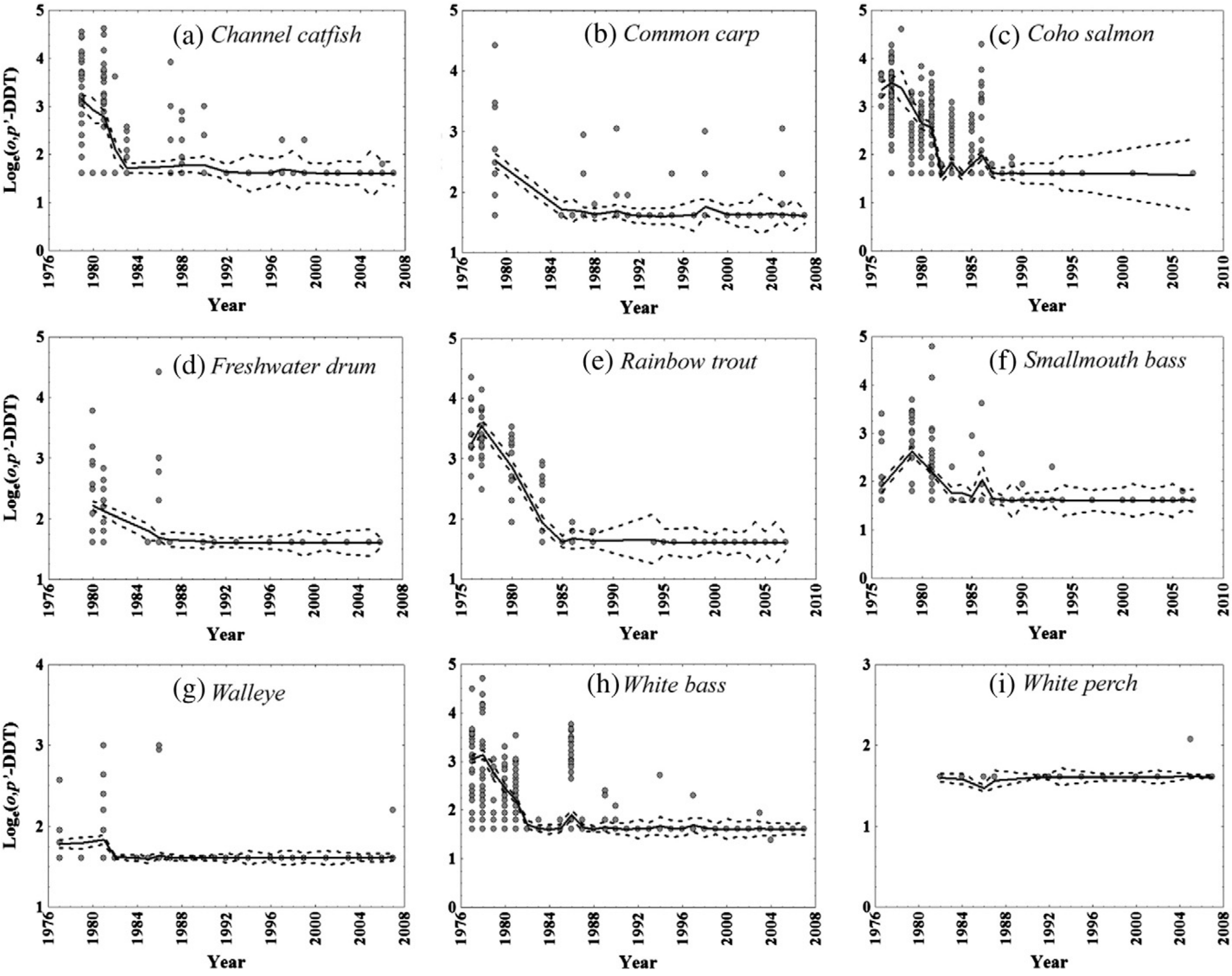


Fig. 4. Dynamic linear modeling analysis depicting the actual *o,p′*-DDT concentrations (ng/g wet weight) (gray dots) against the predicted *o,p′*-DDT trends when accounting for the covariance with the fish length and lipid content (black lines) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) freshwater drum, (e) rainbow trout, (f) smallmouth bass, (g) walleye, (h) white bass, and (i) white perch in Lake Erie (study period 1976–2007).

4. Discussion

The predominantly agricultural nature of the regions around the western end of Lake Erie and the associated large amounts of pesticide runoff likely contributed to the considerable DDT loads in the water (Han et al., 2011). In addition, the heavily industrialized Detroit River also serves as a significant source of hydrophobic pollutants within the lake, transferring substantial amounts of sediment-bound contaminants within the shallower western basin. Upon entering the waterbody, the hydrophobic nature of DDT and its metabolites favors their absorption onto particulate matter and subsequent sedimentation, although a portion of it may volatilize from surface waters (ATSDR, 2002). As these compounds make their way into the lake food web, their persistence and high lipophilicity translate into high levels of bioaccumulation and biomagnification within the biota. In addition to this lingering presence of DDT and its metabolites in the bodies of organisms, the system may also be subject to the slow release of DDT from sediments. The morphological characteristics of Lake Erie suggest that sediment resuspension due to storms and shoreline erosion may also be a significant regulatory process. As a final route of exposure, DDT can also enter the lake through atmospheric deposition, aided by the chemical properties of the parent compound and metabolites that make them highly conducive to long-range transport. In this study, given this complex interplay among watershed, lake water, sediments, and biota, our intent was (*i*) to rigorously examine the long-term trends of DDT and its compounds in fish communities, while accounting for fish length and lipid content as covariates, and subsequently (*ii*) to speculate about inter-specific and -compound differences that may shed light on the perceived contaminant trends in Lake Erie.

The effective monitoring of the DDT trends in any aquatic ecosystem requires both an appreciation for the remarkable persistence of the compound as well as individual testing for the discrete forms it commonly exists in. It has been well-established that DDT exists in various isomeric and metabolic forms within the environment, many of which exceed levels of the parent DDT compound (Ricking and Schwarzbauer, 2012). Technical DDT, the form applied as a pesticide, contains about 65–80% of the active ingredient *p,p′*-DDT, and the remaining forms within the mixture include 15–21% of its isomer *o,p′*-DDT, trace amounts of *o,o′*-DDT, and small amounts of the by-products DDE and DDD (ATSDR, 2002; Metcalf, 1995; see also our Table 2). Aside from contamination products in the formation of technical-grade DDT, DDE and DDD are also important degradation products in natural environments (Aislabie et al., 1997; ATSDR, 2002; Boul, 1995). DDE formation may occur under aerobic conditions through photochemical reactions or dehydrochlorination (Pinkney

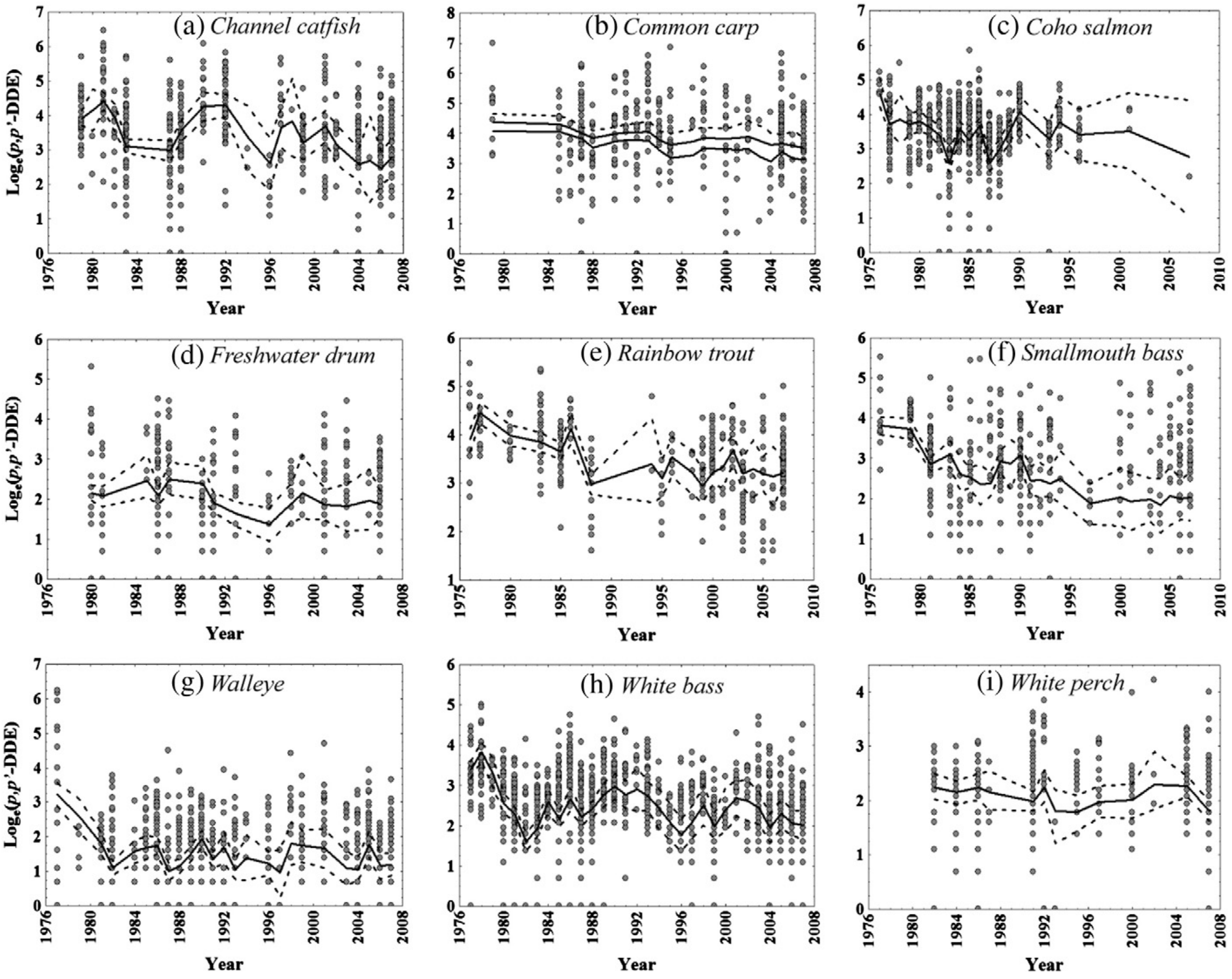


Fig. 5. Dynamic linear modeling analysis depicting the actual *p,p′*-DDE concentrations (ng/g wet weight) (gray dots) against the predicted *p,p′*-DDE trends when accounting for the covariance with the fish length and lipid content (black lines) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) freshwater drum, (e) rainbow trout, (f) smallmouth bass, (g) walleye, (h) white bass, and (i) white perch in Lake Erie (study period 1976–2007).

and McGowan, 2006; see Aislabie et al., 1997 for a review). Conversely, DDD formation mainly involves reductive dechlorination, predominantly in anaerobic environments (Aislabie et al., 1997; see also our Table 2). Notably, DDE has been found to be more persistent than its parent compound, lingering in the environment for decades after the cessation of DDT input (Boul, 1995; Thomas et al., 2008). Both metabolites have been observed in soils within the Lake Erie region, with a Point Pelee analysis revealing high proportions of DDE in sandy soils (Crowe and Smith, 2007), while the Winous Point marshes to the south of the lake mainly contained the reduced DDD metabolite (Spongberg et al., 2004). The continued presence of DDT-associated compounds in soils is a function of their long half-lives as well as their strong absorption to soil particles; DDT and its metabolites generally remain within the top soil layers and are thus susceptible to runoff into waterbodies (ATSDR, 2002).

The results of our study can be classified into two general patterns of DDT behavior, each of which will be examined in the context of previous Great Lakes fish contaminant analyses. First, the so-called “parent” DDT forms (*p,p′*-DDT and *o,p′*-DDT) demonstrated generally negative rates of change throughout the study period, and the corresponding concentrations often reached the detection limit of 5 ng/g in the fish species examined. Second, the levels of the degradation components *p,p′*-DDE and *p,p′*-DDD fluctuated through time while following a fairly distinct downward trajectory. Importantly, high levels of the DDT congeners characterized the piscivorous coho salmon and rainbow trout, an expected result due to their high trophic status in the food web. Likewise, channel catfish and common carp had among the highest DDT concentrations, which may seem somewhat surprising given their lower trophic position. However, these species are primarily known to be benthivorous feeders, thereby providing a plausible pathway for increased bioaccumulation from contaminated sediments through the increased consumption of benthic invertebrates (Azim et al., 2011b).

The falling *p,p′*- and *o,p′*-DDT trends shown herein are on par with findings from other studies that reported similarly declining DDT trends in one or two fish species across the Great Lakes in response to the legislative bans on pesticides (DeVault et al., 1996; Hesselberg et al., 1990; Hickey et al., 2006; Suns et al., 1993). Among the fish species examined, the lack of dramatic declines in white perch for *p,p′*-DDT merely stems from the later start of sampling for that species, and correspondingly, a lower initial concentration to serve as a reference point. We also note that coho salmon was extensively sampled prior to the 1990s, but only had six observations the remaining years, which could have biased the reported temporal trends. Another notable aspect of the parent congener behavior was the gradual slowing of decline rates after the second half of our study period,

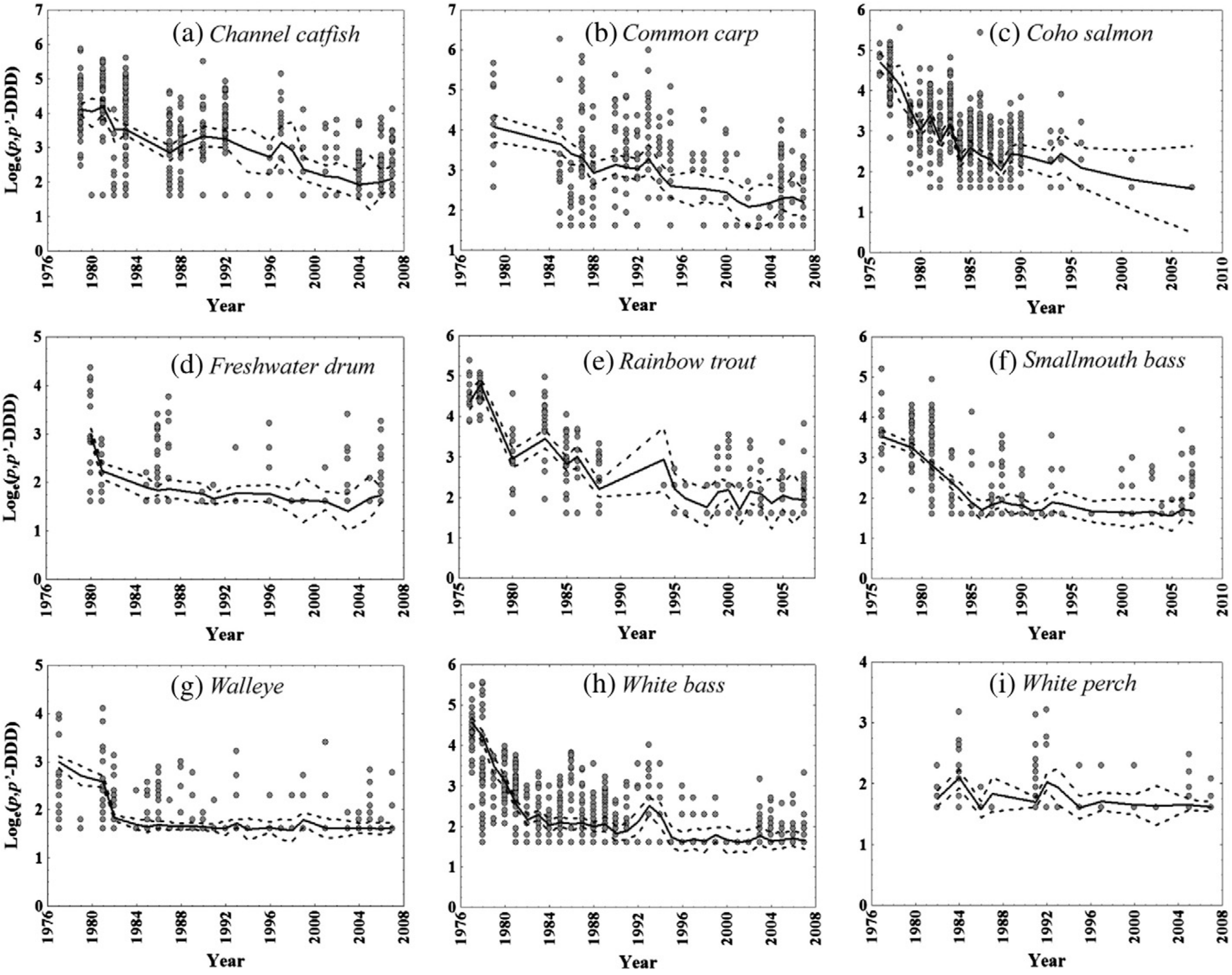


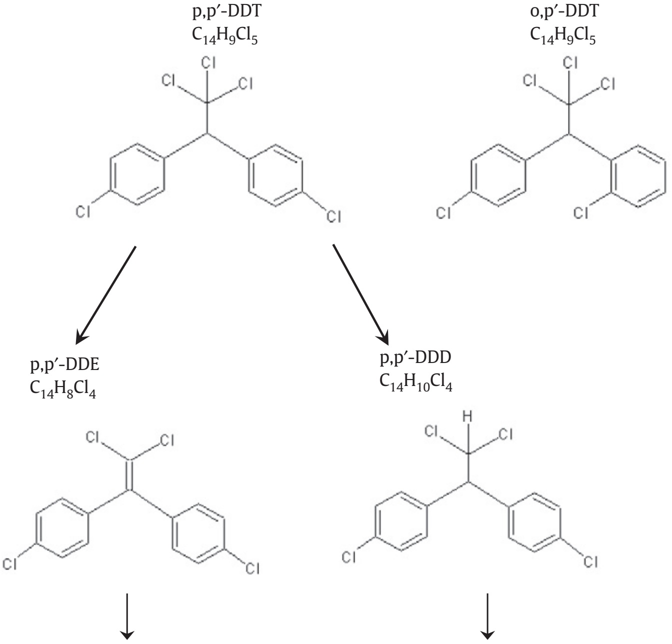
Fig. 6. Dynamic linear modeling analysis depicting the actual p,p'-DDD concentrations (ng/g wet weight) (gray dots) against the predicted p,p'-DDD trends when accounting for the covariance with the fish length and lipid content (black lines) in (a) channel catfish, (b) common carp, (c) coho salmon, (d)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

with such reduction in the rates of change also observed in many other fish POP studies (Bhavsar et al., 2007; Sadraddini et al., 2011a,b). This plateau-type of pattern may reflect earlier predictions that the curtailment of the exogenous contaminant inputs will ultimately bring a chemical equilibrium among atmospheric deposition, sediments, and aquatic biota (Mackay, 1989). However, others have argued that the complicated (and sometimes counteracting) processes that occur within the watershed may decelerate the establishment of such equilibrium, especially in systems like Lake Erie, where substantial watershed–waterbody interactions occur (Harris et al., 2007). Even if the latter assertion holds true though, our analysis provides evidence that it has not been a major impediment to achieve concentrations close to the detection limit for the various DDT compounds examined.

We note here that the para-substituted isomers are generally considered to be of greater interest when examining DDT (Carlson and Swackhamer, 2006), and studies which opt to dissect total DDT trends often preferentially examine the primary metabolites over o,p'-DDT. In Lake Erie, the substantial database on o,p'-DDT offered an opportunity to examine possible differences between the two isomeric forms of the parent DDT compound. As stated previously, there do not seem to be distinct differences in the trends between p,p'- and o,p'-DDT, aside from the (expectedly) lower mean o,p'-DDT concentrations relative to the more prevalent p,p'-DDT form (Hesselberg et al., 1990). Many studies have reported a similarly skewed ratio in fish species, which probably stems from the higher ratio of p,p'-DDT in the technical DDT typically applied to crops and forests; see Ricking and Schwarzbauer (2012) for a review on isomeric DDT forms. The o,p'-DDT form has also been shown to degrade more easily in the environment, likely magnifying this disparity, although there are some contradictory studies (Ricking and Schwarzbauer, 2012). Interestingly, a higher *ortho*:*para* DDT ratio could be indicative of use of dicofol (de la Cal et al., 2008; Qiu et al., 2005), an effective acaricide that differs from DDT by a hydroxyl functional group replacing the hydrogen on C-1 (Weem, 2010).

Historical contamination studies in the Great Lakes have consistently identified Lake Erie as having some of the lowest biotic DDT concentrations (e.g., Clark et al., 1984), although the sediments are just as contaminated as those of other lakes (Allan and Ball, 1990; Gewurtz and Diamond, 2003). To reconcile this seeming disparity, several hypotheses have been proposed that linked the Lake Erie eutrophication levels to the relatively low POP burdens in the biota of the system (Gewurtz and Diamond, 2003). The relative importance of the different pathways of DDT cycling in the environment remains largely unexplored, and the true nature of DDT loss from the system is not always clear (Robertson and Lauenstein, 1998), i.e., is it due to settling of the suspended sediments or is it through biotransformation

Table 2
Chemical formulae[1], structures[2] and fates of the four DDT compounds examined in our study: *p,p′*-DDT, *p,p′*-DDT, *p,p′*-DDT and *p,p′*-DDT.



May undergo further microbial degradation under aerobic conditions[3]

May undergo further degradation under anaerobic conditions (e.g. to DDMU, DDMS)[4]

Sources:
1: See ATSDR (2002).
2: See Aislabie et al. (1997) and Wetterauer et al. (2012).
3: See Thomas et al. (2008).
4. See Aislabie et al. (1997).

within the water column? It is also important to bear in mind that the fate of DDT in fish is still poorly known (Kwong et al., 2008), and this uncertainty/lack of knowledge may be an impediment to explain the fairly wide *p,p′*-DDE and *p,p′*-DDD dynamics reported herein. Unlike the DDD metabolite, DDE was never produced as a pesticide on its own (ATSDR, 2002) and thus its presence in environmental systems is evidence of either contamination with technical DDT or degradation from parent DDT compounds. Importantly, because content of both *p,p ′*- and *o,p′*-DDE in the technical DDT was estimated to be minimal (ATSDR, 2002; Metcalf, 1995), DDE is highly useful as an estimate of the relative age of DDT in the environment. High DDT levels indicate recent exposure to the pesticide, whereas increased DDE levels indicate cumulative exposure (Ssebugere et al., 2009; Strandberg and Hites, 2001).

Our analysis suggests decreasing DDE trends with fluctuating levels throughout the study period, slowing or stabilizing declining rates in more recent years, and higher average DDE concentrations as compared to the parent DDT compounds. The declining trends corroborate studies that show that DDE concentrations in Great Lakes fish are now lower than they were in the past (Hanrahan et al., 1999) and declining Lake Erie sediment DDE levels (Marvin et al., 2004). Furthermore, the higher average DDE concentrations compared to the other congeners reinforce the general pattern of *p,p′*-DDE predominance over other DDT compounds in fish species (ATSDR, 2002; Hesselberg et al., 1990; Stoichev et al., 2007; Suns et al., 1993). DDE has been identified as the dominant breakdown congener in most of the Great Lakes (Schmitt et al., 1985; USEPA, 2009b), comprising up to 80–95% of total DDT in coho salmon (DeVault et al., 1988). Importantly, DDE is more persistent than its parent compound in certain media (Spencer et al., 1996; Thomas et al., 2008) and can have broader ecological ramifications, such as the thinning of avian eggshells and reproductive abnormalities in animals (Beard, 2005). DDE has also been linked to a wide variety of human health issues, due to its role as an androgen receptor antagonist as well as its interactions with other pollutants (see Turusov et al., 2002 for a review). In any event, the fluctuations of the DDE levels may be an evidence of the lingering presence of the DDT residues in Lake Erie fish, suggesting prolonged repercussions of contaminant inputs to freshwater systems, long after external emissions have been curtailed. The actual amplitude of the fluctuations reported herein may be artificially modulated by the variant sampling frequency over time, but could also be a depiction of the role of various perturbations in lake trophodynamics, e.g., invasive species (French et al., 2011; Hogan et al., 2007; Sadraddini et al., 2011a,b).

The metabolite DDD is generally found to be substantially lower than DDE in freshwater fish (Carlson and Swackhamer, 2006), which was also reflected by its relatively lower mean concentrations across most Lake Erie fish species in our analysis. Sediment DDD values have declined across the lake (Marvin et al., 2004), and its reduced presence in fish species has been hypothesized to relate to its reduced lipophilicity and thus lower bioaccumulation potential (Kwong et al., 2008). However, the fluctuating DDD levels in several species in our study paint a somewhat less comforting picture, suggesting that Lake Erie may be associated with elevated DDD formation rates due to the manifestation of hypoxia in late summer (DeVault et al., 1988; Robertson and Lauenstein, 1998). Specifically, the late summer anaerobic conditions in the lake hypolimnion are favorable for DDD production, whereas the fall turnover and the reoxygenation of the water column result in a combination of aerobic and anaerobic conditions that are optimal for further DDD decomposition (DeVault et al., 1988). Recent studies have shown elevated DDD concentrations in two marshes surrounding Lake Erie (Crowe and Smith, 2007; Spongberg et al., 2004) and levels in local mussels were also measured to be relatively high (Robertson and Lauenstein, 1998). Clearly, the various morphological and ecological lake characteristics can alter the ratios of DDT and its metabolites within the water, sediment and biota, and thus careful analyses of different pollutant congeners may be essential tools in freshwater remediation.

An earlier study by DeVault et al. (1986) pinpointed data inadequacy as a significant hindrance to fish contaminant trend detection. While the latter issue may not entirely hold true in the Great Lakes area, conducting retrospective trend analyses can be a delicate task due to the considerable disparity of the fish sampling techniques used over time that make it difficult to draw impartial conclusions (Bhavsar et al., 2007). Our dynamic linear modeling approach is well-suited to explicitly accommodate the role of causal factors that typically underlie the spatiotemporal fish trends (Sadraddini et al., 2011a,b). Fish length was selected as one of our predictor variables due to its well-established covariance with pesticide accumulation as well as the reported sampling bias towards larger fish in recent years (Bhavsar et al., 2007; Sadraddini et al., 2011a,b). Our second covariate has undergone considerable debate in the literature, as there is controversial evidence on the direct linkage between fish lipid content and contaminant concentrations (Amrhein et al., 1999; Larsson et al., 1996; Rowan and Rasmussen, 1992; Stow, 1995; Stow et al., 1997). For example, while Rowan and Rasmussen (1992) found lipid content to be a regulatory factor of the ecological partitioning of organic pollutants across the Great Lakes, Stoichev et al. (2007) reported inconsistent relationships between ΣDDT and fat content in various marine fish species in the Black Sea region and Hickey et al. (2006) found the lipid content to be a weak factor in the variation of contaminant concentrations. There are also indirect pathways that may modulate this relationship, i.e. higher organochlorine concentrations in heavier and fattier fish may simply be due to their increased activity and thus increased potential to accumulate pesticides (Hamelink and Spacie, 1977). Nonetheless, our analysis hypothesized that the lipid content is likely to be a major determinant of

DDT accumulation, given the lipophilicity of the compounds and its metabolites and their preferential deposition into fatty tissues (Wang and Wang, 2005). We found that the consideration of both fish length and lipid content almost consistently provided the most parsimonious dynamic linear models, but it is also important to note that the models with the lipid content as the sole covariate outperformed those with the fish length in twenty two (22) out of the thirty six (36) cases examined.

As one of the predominant routes of POP exposure for residents in the Great Lakes area is the consumption of contaminated sport fish (Kearney et al., 1999), analyses such as ours are useful in ascertaining the relative risks for each pollutant. DDT has a substantial ability to bioaccumulate and biomagnify (Aislabie et al., 1997; Johnson et al., 1999), and thus has the potential to be a health concern. Generally, DDT is no longer a “consumption-limiting contaminant” in fish for the Great Lakes region (OMOE, 2013). In our analysis, the distinctly downward trajectories observed with nearly all congeners and fish species examined are indicative of reduced DDT risks in the Lake Erie fish communities, relative to the prevailing conditions in the early 1970s. Our findings corroborate the current view of the status of DDT in Lake Erie waters and, by extension, the relative success of contaminant legislations in the region.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jglr.2013.06.013.

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EXAMINATION OF TEMPORAL DDT TRENDS IN LAKE ERIE FISH COMMUNITIES USING DYNAMIC LINEAR MODELLING

(Electronic Supplementary Material)

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FIGURE LEGENDS

Figure ESM-1: Variability of *DDT* congener concentrations (*ng/g* wet weight) across four sampling sites for white bass, the most extensively studied species in Lake Erie. Minor sample numbers were also taken from Lake Ontario- Fort St. Erie.

Figure ESM-2: Variability of lipid content (%) for each of the nine fish species sampled.

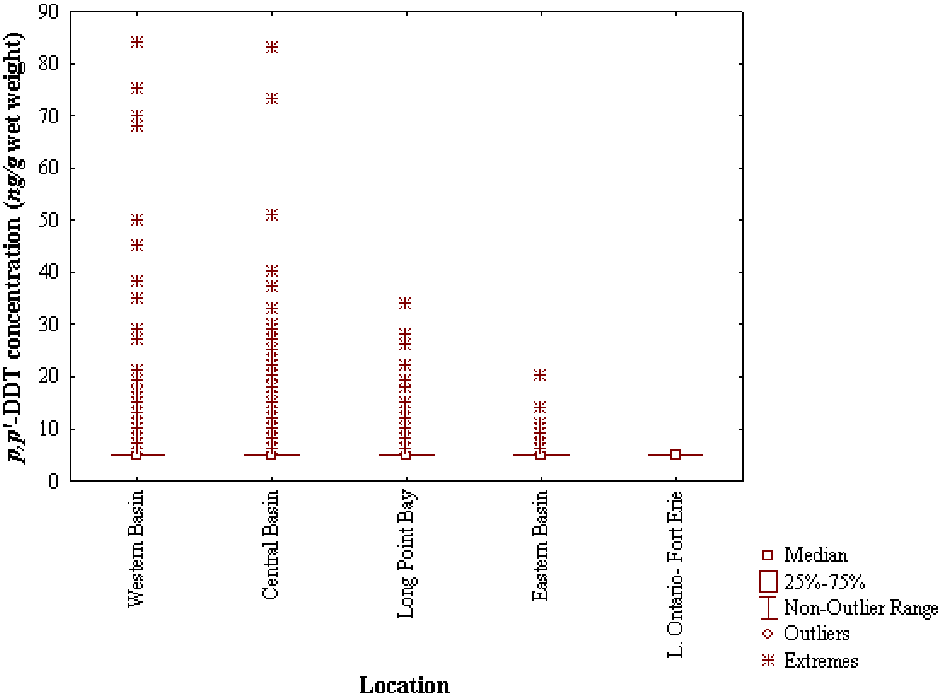
Figure ESM-3: Dynamic Linear Modeling analysis depicting the annual rates of change of *p,p'-DDT* concentrations (ng/g wet weight) in (*a*) channel catfish, (*b*) common carp, (*c*) coho salmon, (*d)* freshwater drum, (*e*) rainbow trout, (*f*) smallmouth bass, (*g*) walleye, (*h*) white bass, and (*i*) white perch in Lake Erie (study period 1976-2007). The solid and dashed lines correspond to the median and the 95% posterior credible intervals, respectively. Green lines indicate zero rate of change.

Figure ESM-4: Dynamic Linear Modeling analysis depicting the annual rates of change of *o,p'*-DDT concentrations (ng/g wet weight) in (*a*) channel catfish, (*b*) common carp, (*c*) coho salmon, (*d)* freshwater drum, (*e*) rainbow trout, (*f*) smallmouth bass, (*g*) walleye, (*h*) white bass, and (*i*) white perch in Lake Erie (study period 1976-2007). Green lines indicate zero rate of change.

Figure ESM-5: Dynamic Linear Modeling analysis depicting the annual rates of change of *p,p'-DDE* concentrations (ng/g wet weight) in (*a*) channel catfish, (*b*) common carp, (*c*) coho salmon, (*d)* freshwater drum, (*e*) rainbow trout, (*f*) smallmouth bass, (*g*) walleye, (*h*) white bass, and (*i*) white perch in Lake Erie (study period 1976-2007). Green lines indicate zero rate of change.

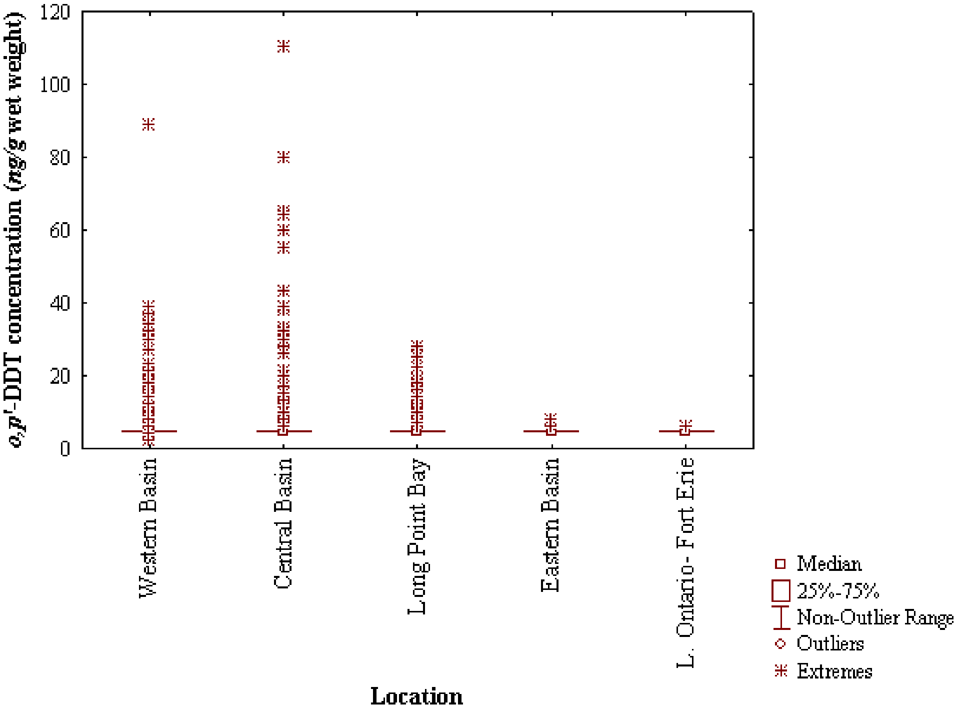
Figure ESM-6: Dynamic Linear Modeling analysis depicting the annual rates of change of *p,p'-DDD* concentrations (ng/g wet weight) in (*a*) channel catfish, (*b*) common carp, (*c*) coho salmon, (*d)* freshwater drum, (*e*) rainbow trout, (*f*) smallmouth bass, (*g*) walleye, (*h*) white bass and (*i*) white perch in Lake Erie (study period 1976-2007). Green lines indicate zero rate of change.

a)



p,p'-DDT concentration (ng/g wet weight)
0
10
20
30
40
50
60
70
80
90
Western Basin
Central Basin
Long Point Bay
Eastern Basin
L. Ontario- Fort Erie
Location
Median
25%-75%
Non-Outlier Range
Outliers
Extremes

b)



o,p'-DDT concentration (ng/g wet weight)
0
20
40
60
80
100
120
Western Basin
Central Basin
Long Point Bay
Eastern Basin
L. Ontario- Fort Erie
Location
Median
25%-75%
Non-Outlier Range
Outliers
Extremes

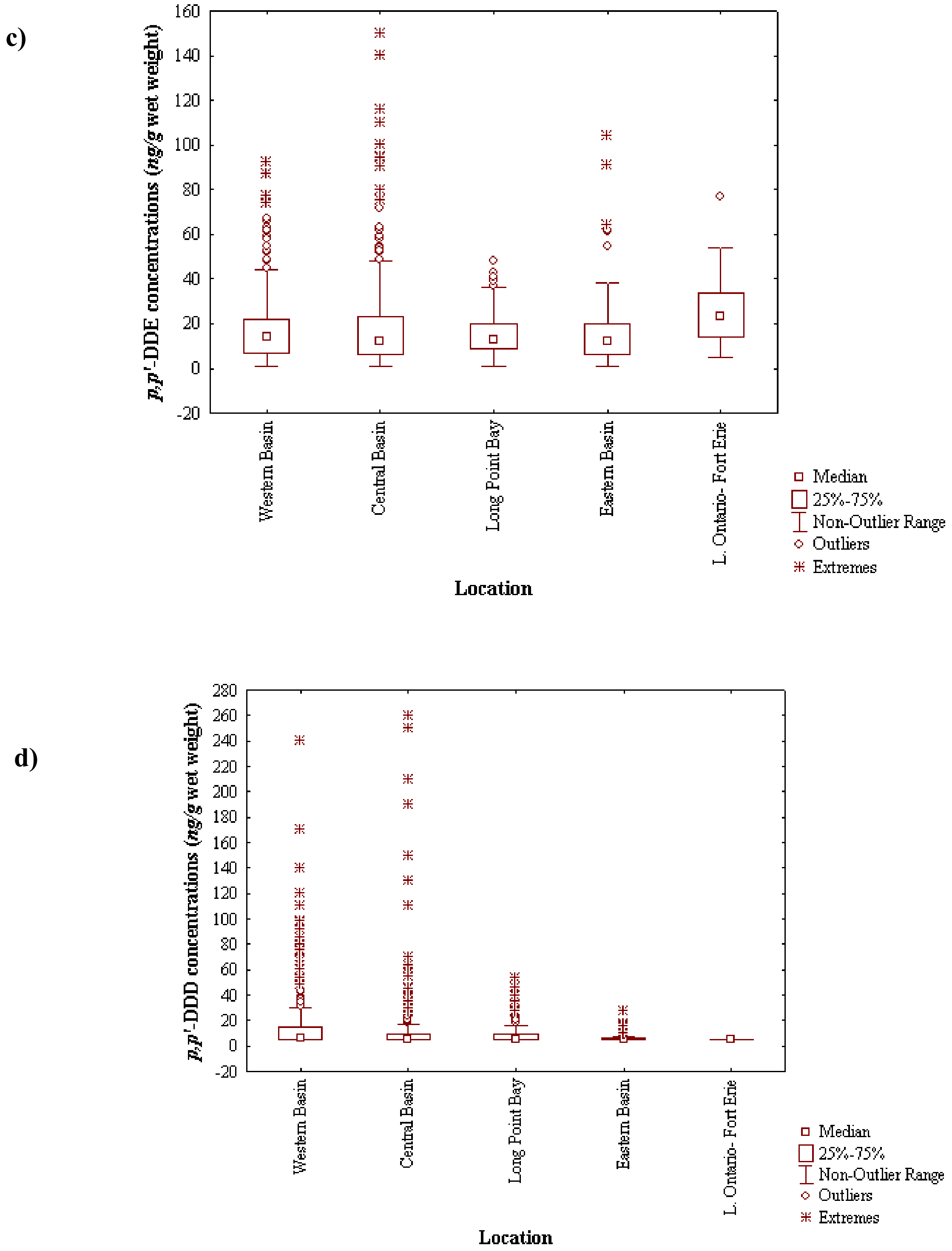
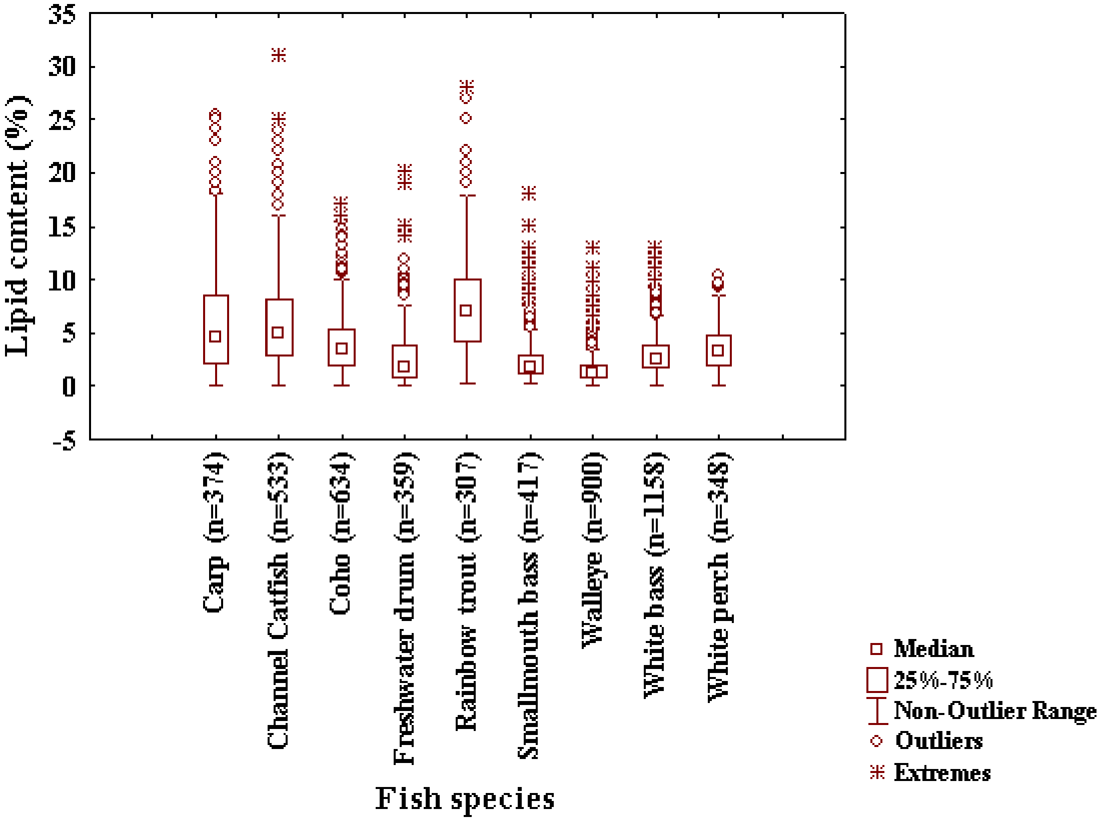

c)
p,p'-DDE concentrations (ng/g wet weight)
160
140
120
100
80
60
40
20
0
-20
Western Basin
Central Basin
Long Point Bay
Eastern Basin
L. Ontario- Fort Erie
Location
Median
25%-75%
Non-Outlier Range
Outliers
Extremes
d)
p,p'-DDD concentrations (ng/g wet weight)
280
260
240
220
200
180
160
140
120
100
80
60
40
20
0
-20
Western Basin
Central Basin
Long Point Bay
Eastern Basin
L. Ontario- Fort Erie
Location
Median
25%-75%
Non-Outlier Range
Outliers
Extremes

Figure ESM-1



Lipid content (%)
35
30
25
20
15
10
5
0
-5
Carp (n=374)
Channel Catfish (n=533)
Coho (n=634)
Freshwater drum (n=359)
Rainbow trout (n=307)
Smallmouth bass (n=417)
Walleye (n=900)
White bass (n=1158)
White perch (n=348)
Fish species
Median
25%-75%
Non-Outlier Range
Outliers
Extremes

Figure ESM-2

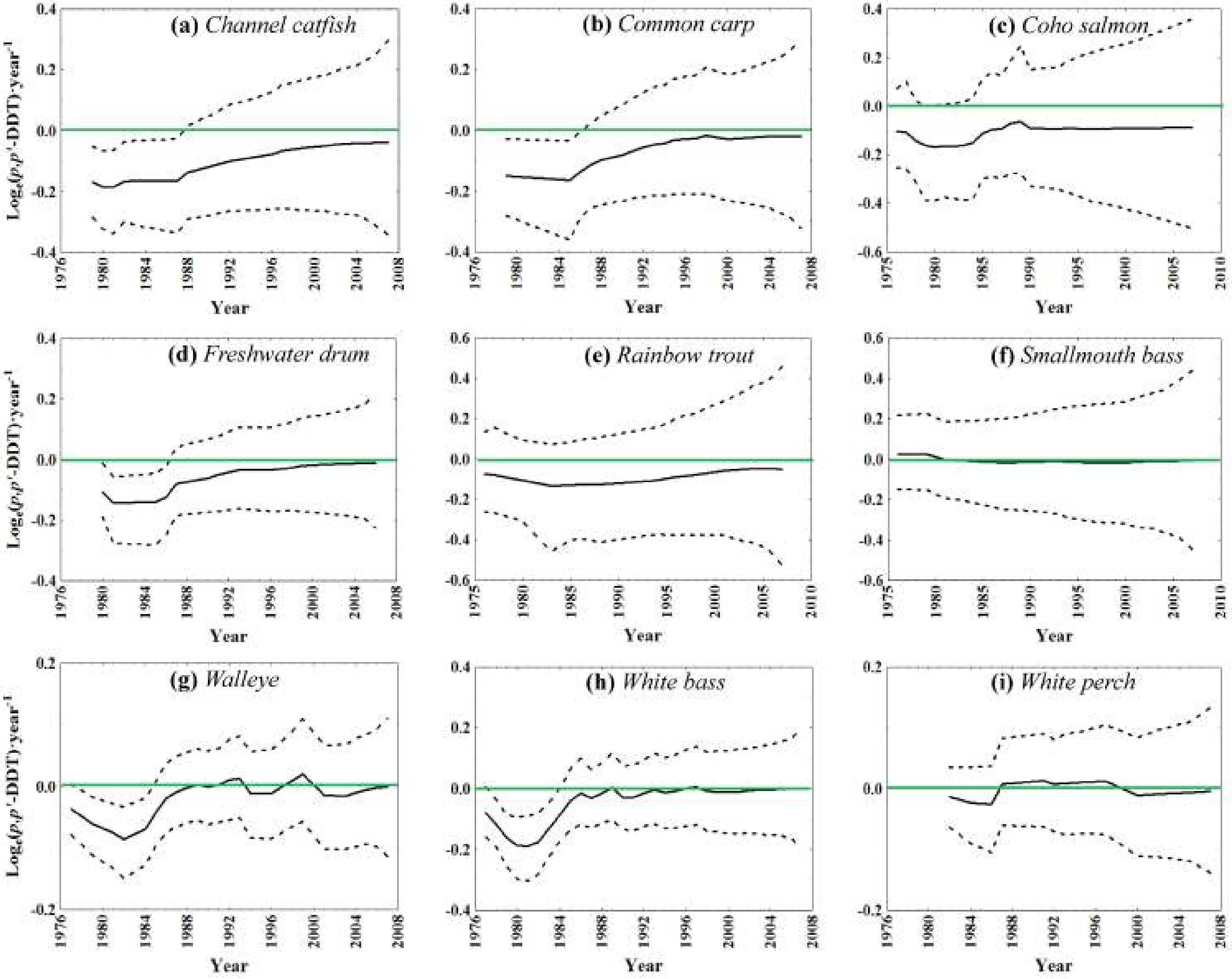

(a) Channel catfish
(b) Common carp
(c) Coho salmon
(d) Freshwater drum
(e) Rainbow trout
(f) Smallmouth bass
(g) Walleye
(h) White bass
(i) White perch
Log_e(p,p'-DDT)·year^-1
Year
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
1976
1980
1984
1988
1992
1996
2000
2004
2008
1975
1985
1990
1995
2005
2010

Figure ESM-3

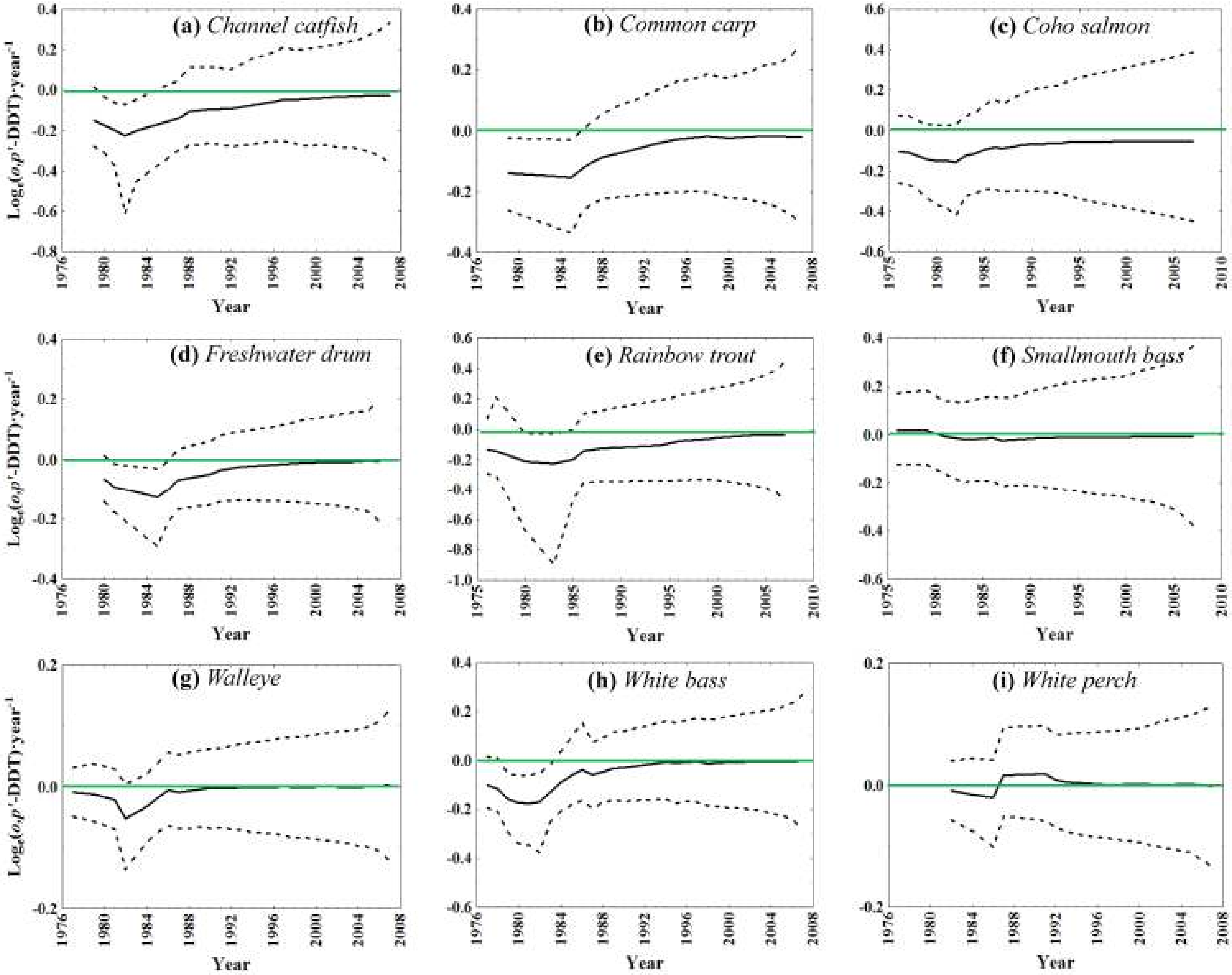

(a) Channel catfish
(b) Common carp
(c) Coho salmon
(d) Freshwater drum
(e) Rainbow trout
(f) Smallmouth bass
(g) Walleye
(h) White bass
(i) White perch
Log$_e$(o,p'-DDT)·year^{-1}
Year
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
-1.0
0.6
1975
1976
1980
1984
1985
1988
1990
1992
1995
1996
2000
2004
2005
2008
2010

Figure ESM-4

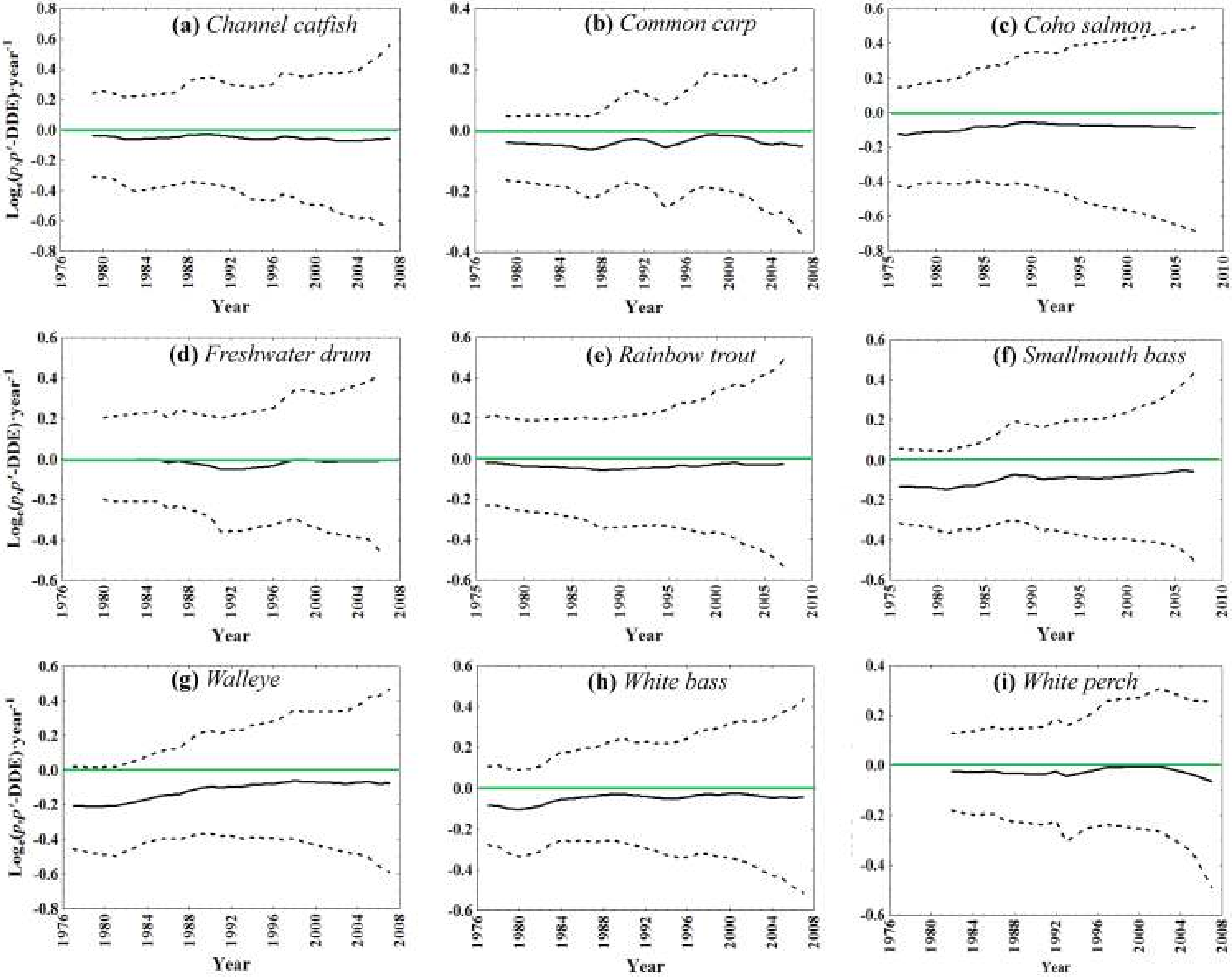

(a) Channel catfish
(b) Common carp
(c) Coho salmon
(d) Freshwater drum
(e) Rainbow trout
(f) Smallmouth bass
(g) Walleye
(h) White bass
(i) White perch
Log$_e$(p,p'-DDE)·year^{-1}
Year
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
1976
1980
1984
1988
1992
1996
2000
2004
2008
1975
1985
1990
1995
2005
2010

Figure ESM-5

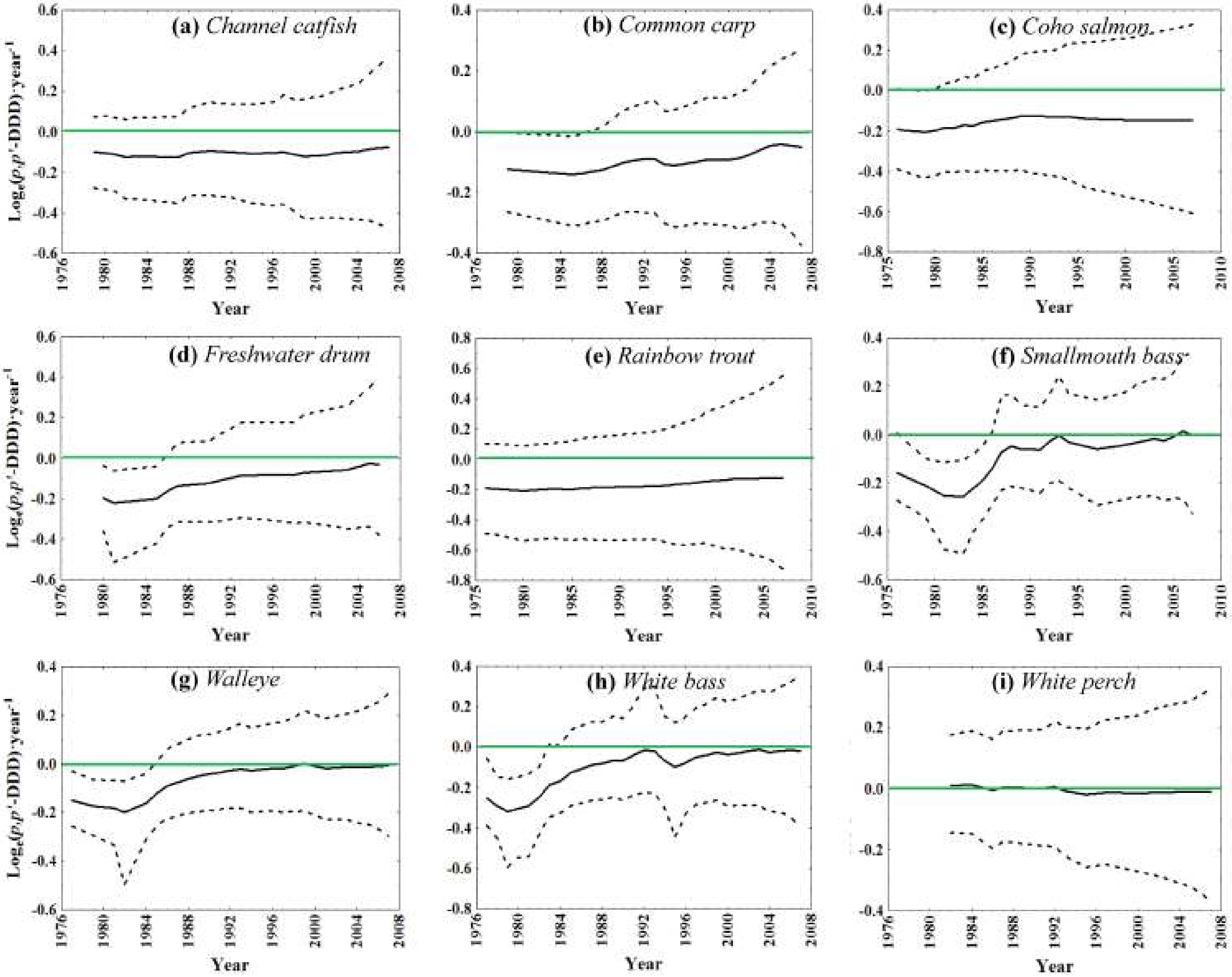

(a) Channel catfish
(b) Common carp
(c) Coho salmon
(d) Freshwater drum
(e) Rainbow trout
(f) Smallmouth bass
(g) Walleye
(h) White bass
(i) White perch
Loge(p,p'-DDD)·year-1
Year
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
1975
1976
1980
1984
1985
1988
1990
1992
1995
1996
2000
2004
2005
2008
2010

Figure ESM-6

Tobit dynamic linear modelling approach

The WinBUGS code associated with the dynamic linear model for the *p,p’*-DDT 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(1.609437912-LogPPDDT[i])
DETLIM[i]~dnorm(-3.8,0.226)
lengthstdev[i]<-(length[i]-3.395819885)/0.182811676
lipidstdev[i]<-(lipid[i]-0.921975)/0.58858
LogPPDDTm[i]<-level[time[i]+1]+beta1[time[i]+1]*lengthstdev[i]+beta2[time[i]+1]*lipidstdev[i]
LogPPDDT[i]~dnorm(LogPPDDTm[i],mtau[time[i]+1])I(,upper.lim[i])
LogPredPPDDT[i]~dnorm(LogPPDDTm[i],mtau[time[i]+1])
PredPPDDT[i]<-exp(LogPredPPDDT[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=10000,
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),
LogPPDDT=c(paste whitebassPPDDT.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,
LogPredPPDDT=c(paste whitebassPPDDT.dat here))
```

Initial values 2

```
list(beta10=0.25,beta20=0.25growth0=0.25,level0=0.25,
beta1=c(0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.2
5,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25),
beta2=c(0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.2
5,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25),
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(0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.2
5,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25,0.25),
mtau.in=0.5,ltau.in=0.5,btau1.in=0.5, btau2.in=0.5,gtau.in=0.5,
LogPredPPDDT=c(paste whitebassPPDDT.dat here))
```