CONTAMINANTS IN LAKE ERIE
FISH COMMUNITIES:
A BAYESIAN EVALUATION

by

Maryam Mahmood

A thesis submitted in conformity with the requirements
for the degree of Master of Science
Graduate Department of Ecology and Evolutionary Biology
University of Toronto

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2012

Abstract

Increasing awareness about the presence and ecological ramifications of toxic, persistent and bioaccumulative contaminants within the Great Lakes system spurred the implementation of numerous bans and emissions restrictions over the past few decades. Due to their high trophic status in food webs and the critical link they serve with human consumers, fish species have historically been monitored to assess the relative success of such remedial efforts within the region and to simultaneously ascertain the current risks posed to local humans. Using Bayesian dynamic linear modelling, this project first aimed to evaluate temporal trends of various organochlorine contaminants within Lake Erie fish communities, the results of which generally indicated decreasing trends through time. The second half of this study used a similar Bayesian approach to propose a framework for updating fish consumption advisories, with specific attention paid to the acknowledgment of uncertainty and natural variability when producing such consumption guidelines.
Acknowledgments

As with everything else in my life, I first extend my deepest gratitude to Allah (God) for the countless blessings and opportunities I have been showered with, particularly in this past year. I have relied on my faith to keep me grounded through the highs and lows of this journey, and I am so grateful to Allah for the gifts of health, patience and assistance in times of need (cue dramatic montage of late nights and frantic typing).

As I sit here writing at my desk today, surrounded by overly-bright motivational posters/cartoons and an equally lurid array of folders and notebooks, I can’t help but be amazed that this year has come and gone so quickly. I remember starting my degree, fresh out of teachers’ college, excited to finally enter the shiny research world yet feeling slightly apprehensive about the task that lay ahead (which of course translated into days of frenzied reading in an attempt to cram information into my brain). I wish I could go back in time and reassure myself that things would be okay, that this would be an incredibly rewarding learning experience and that I would be happily sitting here at the end of it, feeling confident about the future and grateful for everyone who made this year so memorable.

One of the main reasons for any progress I made is my amazing supervisor, Dr. George Arhonditsis; it would be hard to sum him up in a few words, but suffice it to say that his hilarious personality, vast knowledge about ecological modelling, tireless hours spent advising and the overall warmth he exuded combined to make him an outstanding mentor. Though I started the year with limited knowledge about the modelling field (hence my early cramming sessions), his encouragement, approachability and hours of one-on-one help gave me the tools to succeed and the confidence to keep learning (and it helped that he encouraged my healthy habit of gorging on chocolate as I worked). His open-door policy and the humorous environment he created in our lab made this an enjoyable place to research in, and I am grateful for all his support and the risks he encouraged me to take (e.g. IAGLR conference). His tireless dedication to his research and advising was inspiring, and I truly felt myself grow as a student and researcher under his watch. George’s advice, expertise and jokes made the year go by much more easily, and I look forward to future years (and basketball conversations!) as his student.

A big part of why I loved walking into the office every morning were my lab-mates (10+ of them!), who have been an incredible source of inspiration, encouragement and laughter through it all. The conversations throughout the day (everything from research to sports to babies), meals we shared, assistance everyone was always willing to give one another, random umbrella hanging from the ceiling, frequent seminars, joyous runs to get free food and all the advice and support they gave me over the past months made this an extremely memorable experience. I feel so lucky to have worked alongside such brilliant and thoughtful people, who continually inspire me with their work ethic and have opened my mind to so much.
I am also very grateful to both Dr. Satyendra Bhavsar and Dr. Roberta Fulthorpe for their support and advice throughout the entire research process. Dr. Bhavsar's expertise regarding fish consumption advisories was a valuable asset and his willingness to sit down and explain concepts to me was truly appreciated. I am grateful for all his encouragement over the past year and his dedication to ensuring a sound and well-rounded manuscript. My gratitude to Dr. Fulthorpe extends back to my year as an undergraduate researcher, where her guidance (and calming presence when things went awry) helped me get through my first independent research experience. I am grateful for her support this past year, her advice regarding the writing of my paper and her assistance in preparing me for my defence.

I would also like to thank both Dr. Charles K. Minns and Dr. Maydianne Andrade for agreeing to sit on my exam committee. I am aware of how hectic their schedules must be and am thus very grateful for their willingness to read through this dissertation and participate in my defence.

There are many members of the larger University of Toronto community I’d like to thank, more than I could name here. Thanks to Professor Mary Olaveson for her willingness to assist me regardless of her workload and Dr. Carl Mitchell for his mentorship during my first teaching assistantship. I’m also indebted to the administrative staff at both EEB and DPES for their helpful responses to my emails and their patience whenever I frantically flew into their offices with questions, as I had a tendency to do.

I would also like to acknowledge that funding was received for this study from the Ontario Ministry of the Environment Best in Science Research Program (Grant Funding Agreement 89002), though this is not indicative of Ministry endorsement of the contents of my dissertation.

Last, but certainly not least, an enormous air-hug to all my family and friends, who’ve supported and encouraged me every step of the way. Thanks for the endless love and concern, for bolstering me when I needed it, for dragging me out to lunch when you felt I needed a break. Thanks for being my own personal cheer squad, for making me laugh hysterically, for buying me chocolate and ice cream when required (which was always), for all the sleepovers and movie nights and trips. Thanks also for understanding when I brought my laptop or articles along on said events, for sitting with me at night as I worked, for being so gracious when I had to re-schedule because of a deadline. Thank you to everyone for keeping me sane throughout it all, for reminding me to stop and smell the roses, for giving me a sense of balance and hope. Thank you to my parents for being my support team, for teaching me what’s important in life, and for being so patient with my increasingly messy room as the months rolled on. Thank you to my brother for driving me to Cornwall (and back) without complaining. Thank you to everyone I love for your constant support, the texts, the emails, for saying the words I needed to hear before I even realized I needed them. I can’t thank God enough for surrounding me with people like you. You are my sunshine.
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## List of Abbreviations

*Organizations/agreements:*

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<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tbody>
<tr>
<td>GLWQA</td>
<td>Great Lakes Water Quality Agreement</td>
</tr>
<tr>
<td>GLBTS</td>
<td>Great Lakes Binational Toxics Strategy</td>
</tr>
<tr>
<td>IJC</td>
<td>International Joint Commission</td>
</tr>
<tr>
<td>OMOE</td>
<td>Ontario Ministry of the Environment</td>
</tr>
<tr>
<td>USEPA</td>
<td>United States Environmental Protection Agency</td>
</tr>
<tr>
<td>WHO</td>
<td>World Health Organization</td>
</tr>
<tr>
<td>ATSDR</td>
<td>Agency for Toxic Substances and Disease Registry</td>
</tr>
</tbody>
</table>

*Contaminants:*

<table>
<thead>
<tr>
<th>Contaminant</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>DDT</td>
<td>Dichlorodiphenyltrichloroethane; banned pesticide</td>
</tr>
<tr>
<td>DDE</td>
<td>Breakdown component of DDT; by-product of technical-DDT</td>
</tr>
<tr>
<td>DDD</td>
<td>Breakdown component of DDT; by-product of technical-DDT</td>
</tr>
<tr>
<td>HCB</td>
<td>Hexachlorobenzene</td>
</tr>
<tr>
<td>OCS</td>
<td>Octachlorostyrene</td>
</tr>
<tr>
<td>α-HCH</td>
<td>α-hexachlorocyclohexane</td>
</tr>
<tr>
<td>PCBs</td>
<td>Poly-chlorinated biphenyls</td>
</tr>
<tr>
<td>THg</td>
<td>Total mercury</td>
</tr>
</tbody>
</table>

*Methodology:*

<table>
<thead>
<tr>
<th>Methodology</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>DLM</td>
<td>Dynamic linear model</td>
</tr>
<tr>
<td>DIC</td>
<td>Deviance Information Criterion</td>
</tr>
<tr>
<td>TDI</td>
<td>Tolerable Daily Intake</td>
</tr>
</tbody>
</table>
Copyright Note

The two chapters of this thesis were prepared for journal submission (thus the use of plural possessive throughout the text). Permission has been granted from both Dr. George Arhonditsis and Dr. Satyendra Bhavsar (my co-authors on both papers). The papers are still in preparation.

Figure 1 on page 47 (map of Lake Erie) was obtained from the Ontario Ministry of the Environment’s 2011-2012 Guide to Eating Ontario Sport Fish. The front pages of this document state that “this book may be reproduced for non-commercial purposes with appropriate attribution” © Queen’s Printer for Ontario, 2011.
General Introduction

Heralded as containing close to twenty percent of the world’s surface freshwater, the Laurentian Great Lakes have served for centuries as valuable resources for industrial, shipping and recreational activities within the region (Hicks, 1996), with over thirty million people residing in the surrounding basin (GLBTS, 2004). The high occurrence of relatively unchecked anthropogenic activities early on within the waterbodies eventually translated into critical environmental issues manifesting themselves during the 1960s, including widespread eutrophication, fish depletion and, of particular concern, the ubiquitous presence of toxic chemicals (Johnson et al., 1999). Given their considerable residence times and substantial depths, the Lakes are particularly susceptible to inputs of such chemicals (DeVault et al., 1996; Johnson et al., 1999), and thus special attention has been paid over the past few decades to reducing external contaminant inputs and critically assessing the lingering impacts of bioaccumulative compounds within the biota of the system.

One of the largest concerted attempts to halt the further ecological degradation of the Lakes was the Great Lakes Water Quality Agreement, signed in 1972 (with a revision in 1978) by both Canada and the United States (GLBTS, 2004; IJC, 1978; Johnson et al., 1999); the latter revision included an emphasis on striving to “eliminate or reduce…the discharge of pollutants into the Great Lakes system,” with specific target concentrations outlined for persistent contaminants (IJC, 1978). In order to ensure maximal compliance with these critical emission reductions, the treaty highlighted the need for collaborative planning and management practices on both sides of the border (IJC, 1978). As such, a series of Lakewide Management Plans (LaMPs), Remedial Action Plans (RAPs) and Binational Programs were implemented to follow the recommendations laid out by the IJC, with the 2004 Great Lakes Binational Toxics Strategy building on these prior constructs and reiterating the importance of ongoing contaminant initiatives, given the remarkable persistence of many contaminants of interest (GLBTS, 2004).

In addition to addressing these emission legislations from a management standpoint, considerable efforts have focused on continuously monitoring levels of contaminants such as polychlorinated biphenyls (PCBs), total mercury (THg) and DDT within fish species of the Lakes to assess the relative success of our remedial efforts. Fish species are valuable indicators of the ecological health of a system, given their higher position in the food web as well as their crucial link with human consumers (Bhavsar et al., 2007; Carlson et al., 2010). Recent studies have shown that
though these concerted remedial endeavours did initially result in falling contaminant concentrations in Great Lakes biota, 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). In addition, many of these analyses outline the need for more rigorous temporal contaminant analyses, given the failure of most retrospective studies to account for important covariates through time, such as the length of fish sampled (see Azim et al., 2011a,b; Sadraddini et al., 2011a,b). The issue of contamination in fish is thus a highly relevant issue, a fact which is also attested to by the increasing prevalence of fish consumption advisories issued throughout the Great Lakes and across the continental United States (Gewurtz et al., 2011; USEPA, 2004, cited in Burger and Gochfeld, 2006).

As such, this dissertation aims to critically examine long-term trends of specific contaminants in Lake Erie fish (with an eye towards employing a more comprehensive modelling framework) and subsequently broadens its scope to examine the production of fish consumption advisories. The first chapter examines temporal trends of the organochlorine contaminant DDT and its metabolites in Lake Erie fish communities, using the dynamic linear modelling (DLM) approach employed by Azim et al. (2011b) and Sadraddini et al. (2011b). Within this analysis is an explicit consideration of fish length and lipid content as possible covariates with contaminant concentrations, thus accommodating elements of the more rigorous statistical framework called for by many studies. The second chapter opens with a continuation of this DLM approach, now with a focus on three different organochlorine pollutants, and then turns its focus towards relating a larger body of Lake Erie contaminant analyses to the production of fish consumption advisories. Specifically, a Bayesian DLM framework is proposed that remains flexible for the variable characteristics of a target population, in addition to explicitly accounting for uncertainty and the inherent variability in natural systems; the results of the model also shed valuable light on the link between model strength/error and the predictive capacities of risk assessment frameworks, which has broader implications for environmental management practices.
References


Chapter 1
Examination of temporal DDT trends in Lake Erie fish communities using dynamic linear modelling

1 Abstract

The industrial pesticide dichlorodiphenyltrichloroethane (DDT) was initially heralded for its effectiveness against malaria and agricultural pests, but it 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 modelling 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 1976-2007, 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. Conversely, the levels of \( p,p'\)-DDE 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 were decreasing and fluctuated (though to a lesser degree) over time, with gradually slowing decline rates in many fish species, such as smallmouth bass and freshwater drum. We hypothesize that this overall discrepancy among the different DDT components stems from the fact that DDE and DDD are degradation products of \( p,p'\)-DDT, and thus continue to be produced, as DDT is broken down and subsequently propagated throughout the food web. Although levels of total DDT have dropped significantly since the 1970s and do not produce a current cause for alarm, it would be worthwhile to continue
monitoring the fluctuating $p,p'$-DDE and $p,p'$-DDD levels, particularly in fish species regularly chosen for human consumption.

2 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; USEPA, 1975; WHO, 1979). Starting in the 1950s, however, concerns arose about the lingering environmental impacts of the compound, including thinning avian eggshells and reproductive failure (Beard, 2005; Ramade, 1987; Ratcliffe, 1970), endocrine disruption in various species ([de Solla et al., 1998; Keith, 1997], cited in Turusov et al., 2002), and its role as a possible carcinogen (see Turusov et al., 2002 for a review). Furthermore, the lipophilicity and stability of DDT and its primary metabolite dichlorodiphenyl dichloroethylene (DDE) are responsible for significant bio-magnification patterns and increased health risks for top predators, such as birds, piscivorous fish, and ultimately humans (see ATSDR, 2002; IARC/WHO, 1991; Ssebugere et al., 2009).

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 of the waterbodies (DeVault et al., 1996). DDT and its degradation compounds DDE and
dichlorodiphenyldichloroethane (DDD) were detected early on in fish and wildlife of the region (Carlson et al., 2010; Reinert, 1970), with average DDT concentrations exceeding the level of 6 ppm in certain Lake Michigan fish (Reinert, 1970). 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; Reinert, 1970). 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 (Bierman and Swain, 1982), with target DDT concentrations established for water and fish under the revised Great Lakes Water Quality Agreement of 1978 (IJC, 1978; Johnson et al., 1999).

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 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 ([EC, 1991; Van Oostdam and Kagey, 1991], as cited in Kearney et al., 1999) and has been linked to significant DDE human body burdens (Fiore et al., 1989; Hanrahan et al., 1999; Hovinga et al., 1992; Humphrey, 1983; Kreiss, 1985). Even though DDT is not acutely toxic to humans, there is still evidence of neurobehavioural and reproductive impacts, endocrine disruption, and possible carcinogenicity (see 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 (Carlson et al., 2010; OMOE, 2008). These monitoring programs have been invaluable in providing long-term datasets with which to examine contaminant trends and assess the relative success of our remedial efforts ([Bentzen et al., 1999; Mackay, 1989], cited in Bhavsar et al., 2007; Carlson and Swackhamer, 2006).
In general, the implementation of bans did lower 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; Morrison et al., 1998), the impacts of global warming (French et al., 2006), and the role of sediments (Hickey et al., 2006; Morrison et al., 2002; Pearson et al., 1996; 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 decline rates by the end of the 1980s (Baumann and Whittle, 1988). It is important to note that total DDT, the form applied as a pesticide, was 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 (ASTDR, 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 (ASTDR, 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 (Blus et al., 1987; 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, e.g. food chain structures (Hickey et al., 2006; Rowan and Rasmussen, 1992; see USEPA, 2011).

The influence 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 (Carlson et al., 2010), it is also vital to consider the causal factors that may obfuscate the impartial assessment of spatiotemporal trends (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 (Carlson et al., 2010), 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 modelling (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.

In this study, we use dynamic linear modelling to examine the variability of DDT and its metabolites through time. 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 modelling 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., 2010). 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.
3 Methods

3.1 Dataset description

Our study uses fish contaminant data from the Ontario Ministry of the Environment’s (OMOE) Sport Fish Monitoring Program, which routinely collects samples from a wide range of fish species and analyzes contaminant levels in the dorsal skinless-boneless fillet (SBF) portions. The information in this dataset, covering a time span of approximately three decades (1976-2007), is used to guide biennial fish consumption advisories. 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 dolomieu*), walleye (*Stizostedion vitreum*), white bass (*Morone chrysops*) and white perch (*Morone americana*). Samples were collected from various locations on the Canadian side of Lake Erie (Fig. 1).

3.2 Chemical analysis

The collected samples were sent to the Toronto OMOE laboratory, where their DDT content was analyzed using gas liquid chromatography-electron capture detection (GLC-ECD), in accordance with OMOE method PFAOC-E3136 (OMOE, 2007).

3.3 Modelling framework

In this analysis, we developed a series of dynamic linear models (DLMs) to examine temporal trends of the four DDT compounds. We explicitly account for the fact that fish length and lipid content often 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 DLMs for each congener-fish species combination a total of four times: without any covariates ("random walk"), using the fish length or lipid content alone, or both fish
length and lipid content as covariates. We thus ran a total of 144 models over the course of this study. Unlike static regression models that have fixed parameters, DLMs have an evolving structure that allows parameters to shift through time (Lamon et al., 1998). This "dynamic" feature allows our models to more accurately reflect the intra- and 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, DLMs 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 (West and Harrison, 1989). For the sake of brevity, we outline below the model considering both fish length and lipid content as covariates:

**Observation equation:**

\[
\ln[DDT]_i = level_i + \beta_{i1} \ln[length]_i + \beta_{i2} \ln[lipid]_i + \psi_i \\
\psi_i \sim N[0, \Psi]
\]

**System equations:**

\[
level_i = level_{i-1} + rate_i + \omega_{i1} \\
rate_i = rate_{i-1} + \omega_{i2} \\
\beta_{i1} = \beta_{i1-1} + \omega_{i3}
\]

\omega_{i1} \sim N[0, \Omega_{i1}] \\
\omega_{i2} \sim N[0, \Omega_{i2}] \\
\omega_{i3} \sim N[0, \Omega_{i3}]
\[\hat{\beta}_2 = \hat{\beta}_{2,1} + \omega_n\]

\[\omega_n \sim N[0, \Omega_n]\]

\[
1/ \Omega_y^2 = \zeta^2 \cdot 1/ \Omega_{yj}^2, \quad 1/ \Psi_y^2 = \zeta^2 \cdot 1/ \Psi_{yj}^2
\]

\[t > 1 \quad \text{and} \quad j=1 \text{ to } 4\]

where \(\ln[\text{DDT}]_i\) is the observed DDT concentration at time \(t\) in the individual sample \(i\); \(\text{level}_t\) is the mean DDT concentration at time \(t\) when accounting for the covariance with the fish length and lipid content; \(\ln[\text{length}]_i\) is the observed (standardized) fish length at time \(t\) in the individual sample \(i\); \(\ln[\text{lipid}]_i\) is the observed (standardized) fish lipid content; \(\text{rate}_i\) is the rate of change of the level variable; \(\beta_{ni}\) is a length (regression) coefficient; \(\beta_{n2}\) is a lipid (regression) coefficient; \(\psi_j, \omega_j\) are the error terms for year \(t\) sampled from normal distributions with zero mean and variances \(\Psi_{yj}^2, \Omega_{yj}^2\), respectively; the discount factor \(\zeta\) 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 \(\text{level}_1, \text{rate}_1, \beta_{11}, \beta_{21}, 1/ \Omega_{y1}^2, \) and \(1/ \Psi_{y1}^2\) are considered "non-informative" or vague.

The sequential updating of a DLM makes a forecast for time \(t\) based on prior knowledge of the parameters, and then we observe data at time \(t\) (Lamon et al., 1998). Based on Bayes’ Theorem, our knowledge regarding the parameters is updated using the likelihood of the data and the prior knowledge we have (Gelman et al., 2004). 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). Different discounts were examined between 0.8
and 1.0 (as suggested by West and Harrison, 1989) and we settled on a value of 0.95 for the models in this study.

3.4 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 20\textsuperscript{th} iteration (thin of 20) to minimize serial correlation. Samples were taken after the MCMC simulation converged to the true posterior distribution; convergence was assessed using the modified Gelman-Rubin convergence statistic (Brooks and Gelman, 1998). The convergence of the sequences occurred fairly quickly (~1,000 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).

4 Results

4.1 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 (Tables 1-4). For the purpose of clarity, we address each congener separately:
i) 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) exhibiting 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.

ii) 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.

iii) p,p′-DDE: The summary statistics for this congener differed in two ways from the previously examined ortho- and para- DDT forms (Table 3): 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 ww). 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 3).

iv) \( p,p'-DDD \): Similar to the other degradation product \( p,p'-DDE \), the mean concentrations of \( p,p'-DDD \) were much higher than the first two congeners examined (Table 4); 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. Furthermore, the generally positive skewness and kurtosis values indicate right-skewed and leptokurtic distributions; we thus applied a natural log transformation to the data before commencing our DLM analysis.

4.2 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). 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 favourable model was the one that considered both fish length and lipid content as covariates; exceptions were the walleye models for \( p,p'\text{-DDT} \) and \( o,p'\text{-DDT} \) (lipid content rendered the most parsimonious model) and common carp for \( o,p'\text{-DDT} \) (length model was superior) (see last columns of Tables 1-4).

Generally, the temporal trends of the four DDT congeners followed one of two broader patterns. The \textit{ortho-} and \textit{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 \( p,p'\text{-DDE} \) and \( p,p'\text{-DDD} \) 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.

**\( p,p'\text{-DDT} / o,p'\text{-DDT} \):** The levels of both of these congeners decreased through time down to the detection limit of 5 ng/g ww across all species (Fig. 2). For \( p,p'\text{-DDT} \), 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 (Figs 2a-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. 3). Walleye demonstrated marginally positive rates of change in the latter years, reflecting the small fluctuations in the recent \( p,p'\text{-DDT} \) level (Figs. 2g, 3g). The levels of white perch were generally stable over the sampling years studied (Figs 2i, 3i), although the derived patterns are quite uncertain as the existing information from the system is somewhat sporadic. Similar to \( p,p'\text{-DDT} \), \( o,p'\text{-DDT} \) concentrations generally declined until the mid-1980s and hovered around the detection limit thereafter, with characteristic examples being the channel catfish, common carp, coho salmon, freshwater drum, rainbow trout and white bass (Figs 4a-e, h). The corresponding rates of change followed the same pattern of early
rapid decreases followed by slowing decline rates in the later years (Figs 5). Walleye and white perch generally demonstrated stable levels after the 1980s (Figs 4g,i), and the corresponding rates of change were lying close to zero for most of the study period (Figs 5g,i).

*p,p'-DDE/p,p'-DDD*: Counter 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 (Figs 6a,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 (Figs 7a,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 (Figs 6b,d,i); the net rates of change of the corresponding concentrations remained relatively stable around zero throughout the study period (Figs 7b,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 (Figs 8a,b,c), and the corresponding rates of change remained negative but were slowing over time (Figs 9a,b,c). Freshwater drum, smallmouth bass, walleye, white bass and white perch were characterized by smaller fluctuations, leading to fairly stable concentrations around the detection limit (Figs 8d,f-i). Similarly, the rates of change for these species revealed initial rapid decline rates, followed by values that gradually approached to zero (Figs 9d,f-h); the exception was white perch, whose rates hovered around zero throughout the sampling period (Fig.9i). Lastly, coho salmon demonstrated rapidly downward fluctuating concentrations in the early years, followed by a smooth decrease after the
early 1990s. However, as Fig. 8c clearly shows, data collection became more sparse in the latter period and may be biasing 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 in the remaining years (Fig. 9c).

5 Discussion

Lake Erie has historically been subjected to substantial exogenous stresses stemming from anthropogenic disturbances, with contaminant inflows generally consisting of a mixture of urban and agricultural runoff and point discharges from nearby facilities (Han et al., 2011). The negative repercussions of noxious contaminants in water, sediments, and biota of the Great Lakes spurred numerous emission legislations and chemical bans, with the International Joint Commission (IJC) labelling eleven persistent and toxic substances, including DDT, as "critical Great Lakes pollutants" (Johnson et al., 1999). In recent decades, a growing body of work has centred around assessing the relative success of various remedial efforts within the region, with a special emphasis on detecting long-term trends of critical contaminants (Azim et al., 2011a,b; Bhavsar et al., 2007; Carlson and Swackhamer, 2006; Rowan and Rasmussen, 1992; Sadraddini et al., 2011a,b). In this study, we continue in a similar retrospective vein, yet simultaneously make a concerted effort to employ a more rigorous statistical framework, as reflected by the explicit consideration of causal factors in our models. Our specific aims in this chapter were (i) to rigorously examine long-term trends of an important historical contaminant (DDT and its compounds) in Lake Erie fish communities, while accounting for fish length and lipid content as covariates, and subsequently (ii) to speculate about inter-species and -compound differences that may shed light on the perceived contaminant trends in the system.
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. Although several studies simply analyze total *DDT* concentrations over a historical timeframe, 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 (see ATSDR, 2002; 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 5). In addition to existing as 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 through photochemical reactions (Maugh, 1973) or dehydrochlorination (see Aislabie et al., 1997 for a review), generally under aerobic conditions (Murty, 1986, cited in Robertson and Lauenstein, 1988; Pinkney and McGowan, 2006); conversely, *DDD* formation generally involves reductive dechlorination (Aislabie et al., 1997; Wedemeyer, 1966; see also our Table 5), predominantly in anaerobic environments (Murty, 1986, cited in Robertson and Lauenstein, 1988; Pinkney and McGowan, 2006). 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; Boul et al., 1994; [Spencer et al., 1996, cited in 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).

Given the predominantly agricultural nature of the regions surrounding Lake Erie, large amounts of pesticide runoff likely contributed to the considerable DDT loads in the water (Han et al., 2011). Furthermore, the heavily industrialized Detroit River serves as a significant source of hydrophobic pollutants within the lake, transferring substantial amounts of sediment-bound contaminants within the shallower Western basin (Carter and Hites, 1992). Upon entering the waterbody, the hydrophobic nature of DDT and its metabolites favours 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 lake biota (ATSDR, 2002). In addition to this lingering presence of DDT and its metabolites in the bodies of organisms, lakes may also be subject to the slow release of DDT from sediments (Baumann and Whittle, 1988). The morphological characteristics of Lake Erie suggest that sediment resuspension due to storms and shoreline erosion may also be a significant regulatory process (Mortimer, 1987). As a final route of exposure, DDT can also enter the lake through atmospheric deposition (Eisenreich et al., 1981), aided by the chemical properties of the parent compound and metabolites that make them highly conducive to long-range transport (ATSDR, 2002).

The results of our study can be classified into two general patterns of DDT behaviour, 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 ww in the fish species examined. Second, the levels of the degradation
components $p,p'-\text{DDE}$ and $p,p'-\text{DDD}$ fluctuated through time following a fairly distinct downward trajectory.

The falling $p,p'$- and $o,p'$-DDT trends we observed are on par with findings from other studies that reported similarly declining $\Sigma$DDT trends 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, 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 behaviour was the gradual slowing of decline rates after the second half of our study period, with such reduction in the rates of change also observed in many other fish POP studies (Carlson et al., 2010; DeVault et al., 1996; Hickey et al., 2006; 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 (Carlson et al., 2010). Yet, 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 overwhelming 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, though there are some contradictory studies (see Ricking and Schwarzbauer, 2012 for details). Interestingly, a higher ortho:para \( DDT \) ratio could be indicative of dicofol use in a region (de la Cal et al., 2008; Qiu et al., 2004; Qiu et al., 2005), an effective acaricide that differs from \( DDT \) by a hydroxyl functional group replacing the hydrogen on C-1 (Weem, 2010). Finally, to put our results into perspective, it is important to distinguish between the typically reported trends in total \( DDT \) (\( \Sigma DDT \)) levels and the sharp decline of the \( p,p'-DDT \) levels in Lake Erie fish communities reported herein. Our trend projections are likely more pronounced, given that we do not include the (much higher and variant) concentrations of the \( DDT \) metabolites.

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; Thomas and Frank, 1983], cited in 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 (see Gewurtz and Diamond, 2003). The relative importance of the different pathways of \( DDT \) cycling in the environment remains an area of study, and the true nature of \( DDT \) loss from the system is not always clear (see Robertson and Lauenstein, 1998), i.e., is it due to
settling to the sediments or is it through biotransformation 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 unequivocally explain the fairly wide $p,p'DDE$ and $p,p'DDD$ dynamics reported herein. Unlike its fellow 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 the contribution of both $p,p'$- and $o,p'$-DDE to technical DDT was estimated to be quite 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 coupled with fluctuating levels throughout the study period, slowing or stabilizing decline rates in more recent years, and higher average DDE concentrations as compared to the parent DDT compounds. The generally decreasing trends corroborate studies that show lower DDE concentrations in Great Lakes fish now than in the past (Hanrahan et al., 1999) and declining Lake Erie sediment DDE values as well (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 (Clark et al., 1984; Hesselberg et al., 1990; Stoichev et al., 2007; Suns et al., 1993; [USGS, 1999, cited in ATSDR, 2002]). DDE has been identified as the dominant breakdown congener in most of the Great Lakes (Schmitt et al., 1985; USEPA, 2009b), with DeVault et al. (1988) observing that it made up 80-95% of total DDT in coho salmon from among the Lakes (though Erie had a lower proportion, as will be discussed further on). Importantly, DDE is more persistent than its parent compound in certain media (Boul et al., 1994; [Spencer et al., 1996, cited in Thomas et al., 2008]) and
can have broader ecological ramifications, such as the thinning of avian eggshells and reproductive abnormalities in animals (see Beard, 2005 and Turusov et al., 2002 for a review). 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 due to the potentially destructive ways it might interact with other pollutants (see Turusov et al., 2002 for a review). In any event, the distinct 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 (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 favourable 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). It becomes increasingly clear that the various morphological and ecological lake characteristics can profoundly 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, the multitude of factors that can conceivably shape any trend analysis exercise makes compelling the adoption of more robust hind/forecasting modelling tools. Conducting retrospective trend analyses can be a delicate task due to the considerable disparity of the sampling techniques used over time that make it difficult to draw impartial conclusions (Bhavsar et al., 2007). Our dynamic linear modelling approach is well-suited to explicitly accommodate the role of causal factors that typically underlie the spatiotemporal fish trends (Sadraddini et al., 2011a,b). The fish length was selected as one of our predictor variables due 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, 1993; Rowan and Rasmussen, 1992; Stow, 1995; Stow et al., 1997; Voiland et al., 1991). For example, while Rowan and Rasmussen (1992) found lipid content to be a distinct regulatory factor of the ecological partitioning of organic pollutants across the Great Lakes, Stoichev et al. (2007) reported inconsistent relationships between $\Sigma DDT$ 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. In addition, 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 the ones 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 the residents in the Great Lakes area is the consumption of contaminated sport fish ([EC, 1991; Van Oostdam and Kagey, 1991], as cited in 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; Hicks, 1996; Johnson et al., 1999; Reynoldson, 1987), and thus can still be a serious health concern. A characteristic example is the recent evidence that blood and serum DDE levels, though declining, generally remain higher in frequent sport fish consumers in five Great Lakes states (Hanrahan et al., 1999; Knobeloch et al., 2009). Despite the compelling evidence against indiscriminate DDT spraying, its continuous spread across the globe stems from the extensive use as a defouling paint (Hu et al., 2007; Lin et al., 2009), its efficiency in controlling malaria (Karlsson et al., 2000; Seagren, 2005) or indirectly through the widespread use of the derivative dicofol (Ricking and Schwarzbauer, 2012). 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. However, given the susceptibility of the system to the impacts of contaminants and the
growing knowledge about the remarkable persistence of \textit{DDT} metabolites, we believe that it will be prudent to continue monitoring the various forms of this compound.
6 References


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## Tables

Table 1. Basic statistics of *p,p*-DDT concentrations in skinless-boneless fillet data (ng/g wet weight) for nine fish species in Lake Erie (study period 1976-2007).

<table>
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<tr>
<th>Species</th>
<th>N</th>
<th>Mean</th>
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<th>Median</th>
<th>IQR</th>
<th>Skewness</th>
<th>Kurtosis</th>
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*Based on lowest DIC value (LNG = model with length as covariate; LPD = model with lipid as covariate; L+L = model considering both length and lipid as covariates)*
Table 2. Basic statistics of *\textit{op-DDT}* concentrations in skinless-boneless fillet data (ng/g wet weight) for nine fish species in Lake Erie (study period 1976-2007).

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*Based on lowest DIC value (LNG= model with length as covariate; LPD= model with lipid as covariate; L+L= model considering both length and lipid as covariates)*
Table 3. Basic statistics of *pp*-DDE concentrations in skinless-boneless fillet data (ng/g wet weight) for nine fish species in Lake Erie (study period 1976-2007).

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*Based on lowest DIC value* (LNG = model with length as covariate; LPD = model with lipid as covariate; L+L = model considering both length and lipid as covariates)
Table 4. Basic statistics of pp-DDD concentrations in skinless-boneless fillet data (ng/g wet weight) for nine fish species in Lake Erie (study period 1976-2007).

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*Based on lowest DIC value (LNG= model with length as covariate; LPD= model with lipid as covariate; L+L= model considering both length and lipid as covariates)*
Table 5. Chemical formulae, structures and fates of the four DDT compounds examined in our study: $p,p'$-DDT, $p,p'$-DDT, $p,p'$-DDD and $p,p'$-DDD.

\[ \text{source: } 1: \text{see ATSDR (2002)} \\
2: \text{see Aislable et al. (1997) and Wetterauer et al. (2012)} \\
3: \text{see Thomas et al. (2008)} \\
4: \text{see Aislable et al. (1997)} \]
8 Figures

8.1 Figure captions

Figure 1: Map of Lake Erie with the four sampling sites: 1: Western Basin, 2: Central Basin, 3: Long Point Bay, and 4: Eastern Basin.

Figure 2: Dynamic Linear Modeling analysis depicting the actual $p,p'-DDT$ concentrations (ng/g wet weight) (grey dots) against the predicted $p,p'-DDT$ 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). The solid and dashed lines correspond to the median and the 95% posterior predictive intervals, respectively.

Figure 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 predictive intervals, respectively.

Figure 4: Dynamic Linear Modeling analysis depicting the actual $o,o'-DDT$ concentrations (ng/g wet weight) (grey dots) against the predicted $o,o'-DDT$ 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).

Figure 5: Dynamic Linear Modeling analysis depicting the annual rates of change of $o,o'-DDT$ 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).

Figure 6: Dynamic Linear Modeling analysis depicting the actual $p,p'$-DDE concentrations (ng/g wet weight) (grey 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).

Figure 7: 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).

Figure 8: Dynamic Linear Modeling analysis depicting the actual $p,p'$-DDD concentrations (ng/g wet weight) (grey 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).

Figure 9: 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).
Figure 1
(g) *Walleye*

![Graph of Walleye data]

(h) *White bass*

![Graph of White bass data]

(i) *White perch*

![Graph of White perch data]

Figure 2
Figure 3
(g) Walleye

(h) White bass

(i) White perch

Figure 4
Figure 5

(g) Walleye

(h) White bass

(i) White perch
(g) **Walleye**

(h) **White bass**

(i) **White perch**

Figure 7
(g) *Walleye*

(h) *White bass*

(i) *White perch*

Figure 8
(g) Walleye

(h) White bass

(i) White perch

Figure 9
Chapter 2
Fish contamination in Lake Erie: an examination of temporal trends of organochlorine contaminants and a Bayesian approach to consumption advisories

1 Abstract

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

2 Introduction

The ecological health of the Laurentian Great Lakes system declined sharply over the past century as a result of extensive anthropogenic activities, leading to compelling management issues, such as impairment of the resilience of native fish communities, widespread eutrophication, and the ubiquitous presence of toxic chemicals (Johnson et al., 1999). The highly populated and industrialized nature of the surrounding watersheds combined with the long residence times of the receiving waterbodies made the Great Lakes highly susceptible to anthropogenic disturbances (DeVault et al., 1996; Johnson et al., 1999). Growing concerns about the deteriorating quality of the system led to the ratification of the cross-border Great Lakes Water Quality Agreement in 1972 (revised in 1978), aiming to restore the "integrity of the waters," especially through the reduction of harmful contaminants (IJC, 1978; Johnson et al., 1999). Even though the curtailment of external emissions generally led to pollutant declines, the persistent nature of contaminants was translated into lingering concentrations within the aquatic food web, particularly in top predators (Carlson et al., 2010). Fish communities have historically been used as ecosystem health indicators, given their trophic position in aquatic food webs and their critical link to human consumers (Bhavsar et al., 2007; Carlson et al., 2010). Since the 1970s, contaminant levels in fish have been routinely monitored in the Great Lakes, with the resulting information being used to determine fish consumption advisories (Carlson et al., 2010; OMOE, 2008). However, despite the valuable insights gained into contaminant dynamics through the extensive datasets developed, many studies fail to consider
important causal factors that can influence the perceived spatiotemporal trends, such as fish age, size, trophic level, growth and lipid content (Sadraddini et al., 2011a,b; Stow et al., 1997). Variations across monitoring programs in the type of sampling procedures and the different statistical methods used may also impede the robust assessment of contaminant trends (Bhavsar et al., 2010, 2007; Carlson et al., 2010). It is thus essential to strive for more flexible statistical frameworks when undertaking such retrospective analyses, in order to ascertain that the actual contaminant trends are being revealed.

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

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

3.1 Organochlorine trends in Lake Erie fish

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

Modelling framework: We used a series of dynamic linear models (DLMs) to examine the temporal trends of the three contaminants in Lake Erie fish communities. Dynamic linear models are useful tools when examining ecological trends due to their flexible structure that allows parameters to vary through time (Lamon *et al.*, 1998). Our modelling analysis explicitly accounts for the fact that fish length and lipid content may co-vary with the contaminant concentrations, and that fish of different sizes and lipid compositions may have been sampled over time (Bhavsar *et al.*, 2010). We evaluate the relative influence of each of these covariates by considering four DLMs for each congener-fish species combination: without any covariates ("random walk"), using the fish length or lipid content alone, or both fish length and lipid content as covariates. We thus ran a total of 60 models over the course of this study. In this analysis, we introduce non-constant and data-
driven variances with respect to time. Discount factors between 0.8 and 1.0 were examined, and we settled on a value of 0.95. For each model, we used two chain runs of 200,000 iterations, keeping every 20th iteration (thin of 20) to eliminate any problems of serial correlation. The determination of the most parsimonious model for each fish species/congener combination was based on the use of the Deviance Information Criterion (DIC) values, a Bayesian measure of model fit and complexity, where models with lower DIC values are expected to effectively balance between predictive capacity and complexity (Spiegelhalter et al., 2003). More information about the dynamic modelling framework used along with the associated WinBUGS codes can be found in Azim et al. (2011b) and Sadraddini et al. (2011a,b), as well as in Chapter 1 of this thesis.

3.2 Statistical framework for fish consumption advisories

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

Our analysis is founded upon the THg and PCB dynamic linear models presented by Sadraddini et al. (2011b). We first selected three years in order to examine through our model whether there was a distinct change of the fish contaminant levels over time, e.g., 1986, 1996 and 2006. We then identified a range of human weights and meal patterns to demonstrate the flexibility of our statistical approach to fish consumption advisories. For human weights, we chose 50 kg (lower weight), 75 kg (average weight) and 100 kg (heavier weight), while the fish consumption frequency ranged from one to eight fish meals per month. Similar to the value used by OMOE when producing their established advisories, we used a standard fish meal size of 227 grams in our analysis. Our next step was to calculate critical thresholds for each contaminant. Using data from Health Canada, we obtained the tolerable daily intake (TDI) levels for both THg and total PCBs. Tolerable daily intakes are defined as the maximum allowable daily intake of a substance that, if consumed over a lifetime, will not lead to adverse health effects (Health Canada, 1996). TDI values are generally expressed for a specific body weight, such as μg per kg of body weight (or kgbw) per day. Specifically, we used the values of 0.52 μg THg/kgbw per day and 0.09 μg PCB/kgbw per day, and calculated the thresholds for each of the hypothetical scenarios as follows:

\[
\text{Threshold} = \frac{\text{human weight [kg]} \times \text{TDI [ng/kgbw/month]}}{\text{meal size} \times \text{meal number}}
\]

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

4.1 Organochlorine temporal trends

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

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

The significant reductions of exogenous HCB discharges have resulted in decreases of the sediment concentrations in Lake Erie over the past two decades (Marvin et al., 2004a). Further, Gewurtz et al. (2010) recently reported hexachlorobenzene declines in fish from the Lake Huron-Erie corridor, though rates slowed in the 1990s. Our analysis similarly revealed decreasing trends for hexachlorobenzene across the fish species examined. Of the five fish species, the highest average HCB concentrations were recorded in common carp, 3.05 ng/g, followed by coho salmon, 2.82 ng/g, channel catfish, 2.27 ng/g, and rainbow trout, 2.16 ng/g (Table 1). The lowest concentrations of this chemical were observed in white bass, 1.42 ng/g. It is also worthwhile to note that the majority of the concentrations in the later sampling years were recorded at the minimum detection limit of 1 ng/g, and thus the median concentrations for four fish species remained at that value; the only exception was coho salmon (median 2 ng/g), a species which was sparsely sampled after the mid-90s and whose median value was thus less impacted by the HCB drop to the detection limit. According to our DLM analysis, both common carp and rainbow trout demonstrated an increase until the mid-80s and a downward oscillatory pattern thereafter (Figs 1b,d). The mean annual rates of change of the HCB levels for the two species were weakly positive in the early years and nearly zero ever since (Figs 2b,d). Channel catfish demonstrated an overall decreasing trend, although there were fluctuations in the mean HCB levels through time (Fig. 1a). The rates of change of the annual concentrations for this species remained fairly close to zero throughout the study period (Fig. 2a). Similarly, coho salmon demonstrated minor fluctuations early on in the sampling period, but the mean trends projected after the mid-90s should be interpreted with caution due to the sparse data available (Fig.
The corresponding growth rates for coho began weakly positive but switched to very weakly negative towards the end of the years studied (Fig. 2c). White bass was characterized by relatively stable mean concentrations around the detection limit (Fig. 1c), and the corresponding rates of change hovered around zero (Fig. 2c). On a final note, recent studies suggest that Lake Erie may have switched to a source of HCB to the atmosphere through volatilization of the compound out of the lake (Hoff et al., 1996; Kelly et al., 1991; Marvin et al., 2004b). The latter possibility along with the continued release of HCB as unintended by-product could perhaps explain the fluctuating levels reported in our study; however, HCB is generally not considered to be of concern in current fish consumption advisories (Gewurtz et al., 2010).

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

In particular, we found that common carp exhibited the highest concentrations of this contaminant, mean 5.46 ng/g, followed by channel catfish, 5.10 ng/g, coho salmon, 2.26 ng/g and rainbow trout, 1.57 ng/g (Table 2). White bass again had the lowest concentrations, with a mean value of 1.24 ng/g. Similar to the pattern reported for the hexachlorobenzene levels, the median values for common carp, rainbow trout and white bass remained at the detection limit of 1 ng/g. Exceptions were the coho salmon (for the reason discussed earlier) and channel catfish, median 2 ng/g, whose OCS concentrations were subjected to wider fluctuations in the later sampling years. It is also important to note that the octachlorostyrene monitoring in our dataset began at various points in the 1980s, and thus the true maxima of this contaminant may have not been captured. Channel catfish, common carp, coho salmon and rainbow trout were characterized by decreasing trends with distinct fluctuations until the mid-1990s, followed by nearly monotonic decline to the detection limit since then (Fig. 3a-d). The corresponding rates of change were negative throughout the study period (Figs. 4a-c), but with slowing decline rates for rainbow trout over time (Fig. 4d). Finally, white bass showed a minor increase in the 1980s and stable levels around the detection limit ever since (Fig. 3e). The rates of change for this species reflect the initial peak and subsequent decline in the mid-1980s, followed by practically zero rates until the end of the study period (Fig. 4e). Generally, our
analysis suggests decreasing OCS trends through time, which is on par with CGLI's (1999) assertion that the levels of this compound have been substantially declining in the Great Lakes over the past two decades. For example, studies of spottail shiners in the lower Niagara River indicated falling OCS concentrations from the mid-1980s down to the detection limit during the 1990s (see CGLI, 1999 for a review). Similar declines were also reported in fish from the Huron-Erie corridor (Gewurtz et al., 2010). The cessation of direct inputs of OCS to the region (e.g. St. Clair RAP Team, 2006, cited in Gewurtz et al., 2010) along with the aforementioned shift to non-OCS producing metal electrodes is likely to have contributed to the reported decrease of fish OCS levels.

**α-hexachlorocyclohexane (α-HCH):** Highly similar to hexachlorobenzene is our final compound of consideration, hexachlorocyclohexane (HCH), or more specifically, the alpha-isomer α-HCH; this substance is often erroneously called "benzenehexachloride" (Willett et al., 1998). Composed of eight isomers, HCH was generally applied as the pesticide technical-HCH (in which the α-congener was dominant) and later as "lindane", which in turn primarily consists of γ-HCH ([Kutz et al., 1991; Safe, 1993], cited in ATSDR, 2005). Similar to other persistent organic pollutants, HCH has the potential to bioaccumulate and be transported in long distances (Bhatt et al., 2009). While studies have generally focused more on lindane trends in space and time due to its contemporary use in agricultural practices, concentrations of α-HCH remain in air, precipitation and surface waters, often greater than the γ-isomer (Bhatt et al., 2009). These higher α-HCH environmental levels probably stem from as a result of its increased stability over γ-HCH and/or the γ-HCH degradation to its a-congener (Easton et al., 2002). Generally, a larger proportion of α-HCH in environmental media is thought to be an indication of either recent application of technical-HCH or atmospheric deposition, while higher proportions of the beta-isomer (the most stable) would probably indicate distant application (Willett et al., 1998).
In our study, the highest \(a\)-\(HCH\) concentrations were recorded in coho salmon, 2.86 ng/g, followed by channel catfish, 2.72 ng/g, rainbow trout, 2.16 ng/g, white bass, 1.39 ng/g and finally common carp, 1.3 ng/g (Table 3). The recent decline down to the detection limit translated into recurring 1 ng/g median values. Mirroring the patterns of the observed data, the predicted mean \(a\)-\(HCH\) levels generally showed an initial increase and subsequent decline after the mid-1980s (Fig. 5), with the exception of common carp, which lacked this early peak (Fig. 5b). The rates of change for coho salmon, rainbow trout and white bass switched from weakly positive to negative over time (Fig. 6c-e), while those of channel catfish and common carp started off weakly negative, became slightly more negative in the 1980s and then subsequently slowed to the end of the sampling period (Fig. 6a,b). As the use of technical-\(HCH\) declined worldwide in response to multiple bans, atmospheric \(a\)-\(HCH\) concentrations were expected to follow suit (Li et al., 1998). The \(a\)-\(HCH\) levels have dropped dramatically in Great Lake precipitation levels during the 1990s (Buehler et al., 2002; Chan et al., 2003) and \(a\)-\(HCH\) has also declined significantly in the sediments of Lake Erie (Marvin et al., 2004a). As such, our declining \(a\)-\(HCH\) trajectories in the fish species examined are not surprising.

4.2 Framework for fish consumption advisories

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

The development of fish consumption advisories differs among regions, although there have been concerted efforts in recent years to establish protocols and region-wide guidelines to ensure consistency. The first step involves the establishment of a reference concentration, representing an estimate of the daily human exposure to a contaminant that will not result in adverse health effects over a lifetime, e.g., USEPA reference dose, health protection value, tolerable daily intake, and minimal risk level (Dourson and Clark, 1990; HPTF, 2004). Calculation of these reference levels may consider uncertainty factors to account for the extrapolation of toxicity data from animals to humans or to accommodate different tolerance levels in humans (USEPA, 2000). The next step in developing advisory guidelines is to identify standard values for fish meal sizes, human weights, amount of contaminants remaining in fish after cooking, frequency of consumption, and cancer risk factors (GLC, 2007; WVITC, 2007). Depending on the amount/quality of data and the region characteristics, the production of the guidelines may be based on regression models, mean/median concentrations or frequency distributions (GLC, 2007). Regression models are used to relate
contaminant data to the size of various fish species, postulating a linear relationship between the two variables across the fish sizes sampled, while the selection of the best-fit model (e.g., original or logarithmic scale) is typically based on the coefficient of determination ($r^2$) values (GLC, 2007). If no relationship exists with the fish length or the data are inadequate to conduct regression analysis, data pooling may be used to obtain species-specific average concentrations (GLC, 2007). Further, despite the aversion of stakeholders and decision makers when confronted with a "range" of values instead of a "fixed" value (Tannenbaum et al., 2003, cited in Hope et al., 2007), there has been a gradual emergence of probabilistic methods in the risk assessment paradigm (e.g., Antonijevic et al., 2007; see Bilau et al., 2007; Harris and Jones, 2008; Roberts et al., 2007; Wilson et al., 2001; Zhang et al., 2009), due to their ability to accommodate the associated uncertainty or more faithfully depict the risk of "outliers" in a fish population (Johnston and Snow, 2007; Sioen et al., 1998).

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

For illustration purposes, we used the dynamic linear models originally developed by Sadraddini et al. (2011b) to detect \textit{THg} and \textit{PCB} temporal trends in walleye. The optimal model (lowest \textit{DIC} value) for the former case had the fish length as the sole covariate, whereas the latter one was based on both fish length and lipid content. We selected the predictive distributions for three years, e.g., 1986, 1996 and 2006, to examine how the intra- and inter-annual variability in the walleye contaminant levels as well as the total model (structural and parametric) uncertainty shape the risk assessment statements related to human fish consumption. Our first example presents the probability of exceedance of \textit{PCB} tolerable daily intake through the consumption of Lake Erie walleye for a 75-kg person in different fish consumption scenarios: 2, 4, and 8 fish meals/month (Fig. 7). The results are presented for different fish lengths consumed, 22 to 74 cm, while the standard lipid content was set equal to 1.2\%. Our analysis (plausibly) suggests that increasing the number of meals per month translates to a greater risk of exposure. In particular, while two meals per month resulted in negligible exceedances across all fish lengths and years examined, four and (in particular) eight meals can significant increase the risk of violation of the \textit{PCB} tolerable daily intake. The risk of exceeding the critical \textit{PCB} level has distinctly decreased over time, as seen from the reduced probabilities of exceedance from 1986 to 2006. Further, when we consume fish of greater length, the probability of exceeding safe \textit{PCB} levels rises as well; especially consumption of fish longer than 50 cm appears to be associated with more than 20\% probability of exceedance of the \textit{PCB} tolerable daily intake even in 2006. While these predictions were aimed at the average human weight, our analysis also suggests that the risk of \textit{PCB} exposure in individuals with weight 50 kg (or less) can exceed the level of 30\%, when fish with length greater than 50 cm is consumed twice per
week (Fig. 8). The risk is clearly lower for heavier adults, and can drop below 20% when we consider individuals with body weights of 100 kg or more.

As a follow-up exercise, we examined how model performance can influence our capacity to obtain reliable risk assessment statements. In particular, we compared the mean predicted walleye THg concentrations as derived from three DLMs: the model that considers the covariance between THg and fish length; the one with the lipid content as the sole covariate; and the more complex structure with both fish length and lipid content as covariates (Fig. 9a). The three models were updated with the parameter posteriors for 2006, and the corresponding predictions are based on an assortment of fish length and lipid content values falling within the range observed over the last four decades in Lake Erie. In doing so, our intent was to reproduce the broadest range of THg concentrations that could potentially be measured in walleye, given the parameterization of the model for that particular year. The dotted line in Fig. 9a represents the lowest threshold value to avoid harmful intake of THg for a 50 kg person (e.g., adult with low body weight) that has 4 fish meals per month. First, we note that all the mean model predictions fall well below the critical level, indicating that THg does not currently produce a major cause for alarm. Second, the lipid model clearly underestimates the potential range of THg concentrations in the walleye community, indicative of its inferior performance or the weak covariance between THg and fish lipid content (Fig. 9b; \( r^2 < 0.07 \)). On the other hand, the length and length/lipid models are equally proficient at recreating the potential range of fish contamination levels in the system. Notably, the discrepancy between the observed THg levels in 2006 and the predicted ones from the length and length/lipid models primarily stems from the broader range of fish lengths used for our predictive exercise relative to the systematically larger fish sampled in recent years (Bhavsar et al., 2007; Sadraddini et al., 2011a). The higher predictive capacity of the length and length/lipid models is also reflected by the
significantly higher $r^2$ values (>0.64) between measured concentrations and predicted median values (Fig. 9b).

Contrary to the inability of the mean predictions from the lipid model to capture the within-year $THg$ variability in walleye, the consideration of the total uncertainty associated with the corresponding predictions paints a different picture. In particular, when using the average lipid content of 1.2%, the higher median prediction of the lipid model along with the wider 95% predictive interval (higher model error) result in a marginal exceedance of the threshold value (Fig. 10a). Although the use of logarithmic scale is somewhat misleading, the predictive uncertainty of the lipid model suggests that 95% of the $THg$ concentrations fall within the 0.04-1.0 ng/g range, whereas the corresponding uncertainty zone for the other two models lies between 0.03 and 0.6 ng/g. More solid evidence for the ramifications of the higher predictive uncertainty of an inferior model is presented in Fig. 10b, where the 95% predictive intervals of the lipid model against the observed $THg$ concentrations are two- to five times broader than the (generally overlapping) points for the other two models. Hence, the adoption of probabilistic statements does not conceal the weaknesses of a model, but rather the inflated error/uncertainty makes the corresponding predictions practically uninformative for risk management. Simply put, the selection of an erroneous model in our example resulted in unjustifiably comforting risk statements when our retrospective analysis is solely based on the mean predictions, and in overly alarmist (or otherwise uninformative) projections when we also consider the underlying predictive error.

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

In addition to balancing the risks and benefits of fish, a lengthy list of additional challenges plagues the production of fish consumption advisories (e.g., see Oken et al., 2012). One of the primary issues revolves around ensuring effective communication, as many anglers may not be aware of the guides or opt to downplay the advisories due to preconceived notions, such as optimism about their catch, failure to understand the guides or distrust of the government (see Burger, 2000;
Burger and Gochfeld, 2006; Oken et al., 2012; Pflugh et al., 1999; Tan et al., 2011). As such, a number of studies have primarily focused on working with target populations to identify possible ways of enhancing guides and improving the tone and readability of the advisories (Connelly and Knuth, 1998; Jardine, 2003; Tan et al., 2011; Velicer and Knuth, 1994). Other communication challenges include the failure to reach vulnerable subgroups, such as pregnant women, women of childbearing age, children under 15, Hispanics and African-Americans (Anderson et al., 2004; Lepak et al., 2009; Scherer et al., 2008; Shimshack et al., 2007); disconnect with Native populations (see Donatuto and Harper, 2008); and lack of consensus about the definition of what is a "sensitive" population (Lepak et al., 2009). Aside from these "communication-centric" issues, the production of fish consumption advisories is also made inherently more complex by the synergistic effects among multiple contaminants (Clark et al., 1987; Scherer et al., 2008), and the differential impacts of cleaning and cooking fish on different contaminants (GLSFATF, 1993).

5 Conclusions

We used dynamic linear modelling to examine the temporal trends of three organochlorine compounds (hexachlorobenzene, octachlorostyrene, and α-hexachlorocyclohexane) in five Lake Erie fish species. Our analysis indicates that the levels of organochlorines have been decreasing over the last three decades, although there were cases that exhibited fluctuations through time. The present results reinforce the findings of our recent work that the levels of several important contaminants, such as \( THg \) (Azim et al., 2011a; Sadraddini et al., 2011b), PCBs (Sadraddini et al., 2011a,b), chlordane (Azim et al., 2011b), and dichlorodiphenyltrichloroethane (Chapter 1) have been declining in the fish communities of Lake Erie over the past few decades and do not produce major causes for alarm. However, it must be noted that there are fish species (walleye, smallmouth bass, rainbow trout, white bass, freshwater drum) with differences in their dietary habits, foraging behaviour and competition strategies which exhibit weakly increasing trends of their \( THg \) and/or PCB body.
burdens following the mid- or late 1990s (Azim et al., 2011a; Sadraddini et al., 2011b). Thus, it is important to closely monitor these trends, particularly in fish species regularly chosen for human consumption. We also presented a Bayesian framework to update fish consumption advisories that incorporates the uncertainty in contaminant predictions and the natural variability in fish length and lipid content, while remaining flexible for different human weights and diet patterns. Our results demonstrate that the risk of exposure has clearly decreased over time, but there is still substantial likelihood of exceedance of the tolerable daily intake for PCBs in sensitive populations (e.g., children with low body weight) with frequent consumption of large fish. Future augmentations of the present framework need to focus on its capacity to generate integrative statements that impartially weigh benefits and net risks of consuming fish, and special emphasis should be given to the increased sensitivity of vulnerable groups.
6 References


Ontario Ministry of the Environment (OMOE), 2007. The determination of polychlorinated biphenyls (PCBs), organochlorines (OCs) and chlorobenzenes (CBs) in fish, clams and mussels by gas liquid chromatography-electron capture detection (GLC-ECD). OMOE Laboratory Services Branch: Quality Management Unit.


7 Tables

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

<table>
<thead>
<tr>
<th>Species</th>
<th>N</th>
<th>Mean</th>
<th>SD</th>
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<th>IQR</th>
<th>Skewness</th>
<th>Kurtosis</th>
<th>Best model</th>
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*Based on lowest DIC value* (LNG= model with length as covariate; LPD= model with lipid as covariate; L+L= model considering both length and lipid as covariates)
Table 2. Basic statistics of octachlorostyrene concentrations in skinless-boneless fillet data (ng/g wet weight) for five fish species in Lake Erie (study period 1981-2007).

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<tr>
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* Based on lowest DIC value (*LNG*= model with length as covariate; *LPD*= model with lipid as covariate; *L+L*= model considering both length and lipid as covariates)
**Table 3.** Basic statistics of α-hexachlorocyclohexane concentrations in skinless-boneless fillet data (ng/g wet weight) for five fish species in Lake Erie (study period 1976-2007).

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*Based on lowest DIC value* (LNG= model with length as covariate; LPD= model with lipid as covariate; L+L= model considering both length and lipid as covariates)
8 Figures

8.1 Figure captions

**Figure 1**: Dynamic Linear Modeling analysis depicting the actual hexachlorobenzene concentrations (ng/g wet weight) (grey dots) against the predicted hexachlorobenzene trends when accounting for the covariance with the fish length and lipid content (black lines) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1976-2007). The solid and dashed lines correspond to the median and the 95% posterior predictive intervals, respectively.

**Figure 2**: Dynamic Linear Modeling analysis depicting the annual rates of change of hexachlorobenzene concentrations (ng/g wet weight) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1976-2007). The solid and dashed lines correspond to the median and the 95% posterior predictive intervals, respectively.

**Figure 3**: Dynamic Linear Modeling analysis depicting the actual octachlorostyrene concentrations (ng/g wet weight) (grey dots) against the predicted octachlorostyrene trends when accounting for the covariance with the fish length and lipid content (black lines) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1981-2007).

**Figure 4**: Dynamic Linear Modeling analysis depicting the annual rates of change of octachlorostyrene concentrations (ng/g wet weight) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1981-2007).

**Figure 5**: Dynamic Linear Modeling analysis depicting the actual α-hexachlorocyclohexane concentrations (ng/g wet weight) (grey dots) against the predicted α-hexachlorocyclohexane when accounting for the covariance with the fish length and lipid content (black lines) in (a) channel catfish,
(b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1976-2007).

**Figure 6:** Dynamic Linear Modeling analysis depicting the annual rates of change of α-hexachlorocyclohexane concentrations (ng/g wet weight) in (a) channel catfish, (b) common carp, (c) coho salmon, (d) rainbow trout and (e) white bass in Lake Erie (study period 1976-2007).

**Figure 7:** Probability of exceedance of the PCB tolerable daily intake through the consumption of walleye from a 75-kg person: 2, 4, and 8 fish meals/month. Results are presented for different fish lengths consumed (with the lipid content set equal to 1.2%) in 1986, 1996, and 2006.

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

**Figure 9:** (a) Mean predictions of walleye THg concentrations (µg/g) based on the range of fish lengths and lipid contents measured over the last four decades in Lake Erie. The numbered lines correspond to predictions from the dynamic linear models that consider fish length (l), lipid content (2), and both length and lipid values (3) as covariates. The three models were updated with the parameter posteriors for 2006. The predictions are plotted against the historically observed THg values (gray dots), while the dotted line represents the lowest threshold value to avoid harmful THg intake for a 50 kg person that has 4 meals of fish per month. (b) Comparison between the measured and median predicted THg concentrations from the three models.

**Figure 10:** (a) Predicted walleye Hg concentrations (µg/g) for the average fish length and lipid values measured over the last four decades in Lake Erie. The numbered lines correspond to model predictions
with the average fish length of 40 cm (1), the average fish lipid content of 1.2% (2), and both average length and lipid content (3). The central markers on each numbered line correspond to the median model prediction, while the outer markers represent the 2.5 and 97.5 percentiles. The three models were updated with the parameter posteriors for 2006; (b) 95% predictive intervals against observed THg (µg/g) concentrations in 2006, using the three dynamic linear models.
Figure 1

(a) Channel catfish

(b) Common carp

(c) Coho salmon

(d) Rainbow trout

(e) White bass
Figure 2

(a) Channel catfish

(b) Common carp

(c) Coho salmon

(d) Rainbow trout

(e) White bass
Figure 3
(a) Channel catfish

(b) Common carp

(c) Coho salmon

(d) Rainbow trout

(e) White bass

Figure 4
(a) Channel catfish

(b) Common carp

(c) Coho salmon

(d) Rainbow trout

(e) White bass

Figure 5
Figure 6

(a) *Channel catfish*

(b) *Common carp*

(c) *Coho salmon*

(d) *Rainbow trout*

(e) *White bass*
Figure 7

a) 2 meals/month

b) 4 meals/month

c) 8 meals/month
Figure 8

a) 50 kg

b) 75 kg

c) 100 kg
Figure 9

(a) Graph showing Hg concentration (µg/g) against year. The x-axis represents the years from 1975 to 2005, and the y-axis represents Hg concentration. The data points are scattered across the graph, with a dashed line indicating a threshold.

(b) Graph showing median predicted Hg concentrations (µg/g) against observed Hg concentrations (µg/g). The equation for Length is $y = 0.104 + 0.536x; r^2 = 0.636$, for Lipid is $y = 0.189 + 0.077x; r^2 = 0.066$, and for Length and lipid is $y = 0.105 + 0.537x; r^2 = 0.658$. The data points are differentiated by grey, triangle, and black markers, respectively.
Figure 10