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

M. Ekram Azim [a], Michelle Letchumanan [a], Azzam Abu Rayash [a], Yuko Shimoda [a], Satyendra P. Bhavsar [b], George B. Arhonditsis [a,*]

[a] Ecological Modeling Laboratory, Department of Physical and Environmental Sciences, University of Toronto, 1265 Military Trail, Toronto, Ontario, Canada M1C 1A4
[b] Ontario Ministry of Environment, Environmental Monitoring and Reporting Branch, Toronto, Ontario, Canada M9P 3V6,

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ABSTRACT

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

1. Introduction

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

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

* Corresponding author. Fax: +1 416 287 7279.
E-mail address: georgea@utsc.utoronto.ca (G.B. Arhonditsis).

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

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

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

2. Methods

2.1. Dataset description and chemical analysis

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

2.2. Modeling framework

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

Observation equation

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

System equations:

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

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

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

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

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

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

where $\ln[chlordane]_{ti}$ is the observed α- or γ-chlordane concentrations at time t in the individual sample i; $level_t$ is the mean α- or γ-chlordane concentrations at time t when accounting for the covariance with the fish length; $\ln[length]_{ti}$ is the observed (standardized) fish length at time t in the individual sample i; $rate_t$ is the rate of change of the level variable; β_t is a length (regression) coefficient; ψ_t, ω_{tj} are the error terms for year t sampled from normal distributions with zero mean and variances Ψ_t^2, Ω_{tj}^2, respectively; the discount factor ζ represents the aging of information with the passage of time; $N(0, 10{,}000)$ is the normal distribution with mean 0 and variance 10,000; and $gamma(0.001, 0.001)$ is the gamma distribution with shape and scale parameters of 0.001. The prior distributions for the parameters of the initial year $level_1$, $rate_1$, β_1, $1/\Omega_{1j}^2$, and $1/\Psi_1^2$ are considered "non-informative" or vague. The *DLM* process makes a forecast for time t based on prior knowledge of the parameters, and then we observe data at time t. Using the Bayes' Theorem, our knowledge regarding the parameters is updated using the likelihood of the data and our prior knowledge (Gelman et al., 2004). In this study, we introduce non-constant and data-driven variances (with respect to time) using a discount factor on the first period prior (Congdon, 2003). We examined different discounts between 0.8 and 1.0 (i.e., the static regression model) and the results reported here are based on a discount value of 0.95. This discounted posterior knowledge becomes prior knowledge for time $t+1$, and the process is repeated.

2.3. Model computations

Sequence of realizations from the model posterior distributions were obtained using Markov Chain Monte Carlo (MCMC) simulations (Gilks et al., 1998). Specifically, we used the general normal-proposal Metropolis algorithm as implemented in the WinBUGS software; this algorithm is based on a symmetric normal proposal distribution, whose standard deviation is adjusted over the first 4000 iterations such as the acceptance rate ranges between 20% and 40% (Arhonditsis et al., 2007, 2008). We used two chain runs of 80,000 iterations and samples were taken after the MCMC simulation converged to the true posterior distribution. Convergence was assessed using the modified Gelman–Rubin convergence statistic (Brooks and Gelman, 1998). Generally, we noticed that the sequences converged very rapidly ($\approx$1000 iterations), and the summary statistics reported in this study were based on the last 75,000 draws by keeping every 20th iteration (thin=20) to avoid serial correlation. The accuracy of the