Chemosphere 210 (2018) 1193–1206

Contents lists available at ScienceDirect

Chemosphere

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

A Bayesian assessment of polychlorinated biphenyl contamination of fish communities in the Laurentian Great Lakes

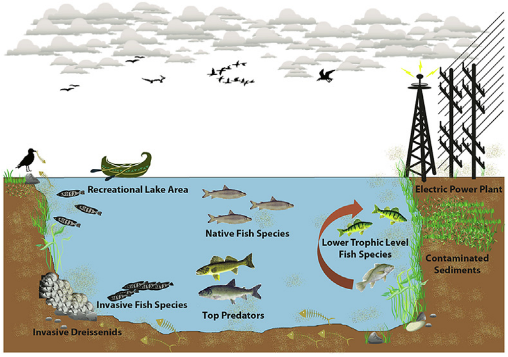
Ariola Visha [a], Nilima Gandhi [a], Satyendra P. Bhavsar [a, b], George B. Arhonditsis [a, *]

[a] *Department of Physical and Environmental Sciences, University of Toronto, Toronto, Ontario, M1C 1A4, Canada*
[b] *Ontario Ministry of Environment, Conservation and Parks, Toronto, Ontario, M9P 3V6, Canada*

HIGHLIGHTS

- We examine PCB trends in multiple fish species across the Canadian Great Lakes.
- Lake Ontario is characterized by the most severe PCB fish contamination.
- Areas of Concern and bays receiving riverine inputs still display high PCB levels.
- PCB fish concentrations had been decreasing from the 1970s until the early 1990s.
- Deceleration of the PCB declining rates has been experienced after the mid-1990s.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 31 March 2018
Received in revised form
12 July 2018
Accepted 13 July 2018
Available online 17 July 2018

Handling Editor: Myrto Petreas

Keywords:
Hierarchical Bayesian modelling
Bioaccumulation
Dynamic linear modelling
Great Lakes
Fish contamination
Invasive species
Polychlorinated biphenyls

ABSTRACT

Polychlorinated biphenyl (PCB) contamination has historically posed constraints on the recreational and commercial fishing industry in the Great Lakes. Empirical evidence suggests that PCB contamination represents a greater health risk from fish consumption than other legacy contaminants. The present study attempts a rigorous assessment of the spatio-temporal PCB trends in multiple species across the Canadian waters of the Great Lakes. We applied a Bayesian modelling framework, whereby we initially used dynamic linear models to delineate PCB levels and rates of change, while accounting for the role of fish length and lipid content as covariates. We then implemented Bayesian hierarchical modelling to evaluate the temporal PCB trends during the dreissenid pre- and post-invasion periods, as well as the variability among and within the water bodies of the Great Lakes system. Our analysis indicates that Lake Ontario is characterized by the highest PCB levels among nearly all of the fish species examined. Historically contaminated local areas, designated as Areas of Concern, and embayments receiving riverine inputs displayed higher concentrations within each of the water bodies examined. The general temporal trend across the Great Lakes was that the high PCB concentrations during the early 1970s followed a declining trajectory throughout the late 1980s/early 1990s, likely as a result of the reductions in industrial emissions and other management actions. Nonetheless, after the late 1990s/early 2000s, our analysis provided evidence of a decline in the rate at which PCB concentrations in fish were dropping, accompanied by a gradual establishment of species-specific, steady-state concentrations, around which there is considerable year-to-year variability. The overall trends indicate that reduced contaminant emissions have brought about distinct beneficial changes in fish PCB concentrations, but past historical contamination along with other external or internal stressors (e.g., invasive species, climate change)

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[*] Corresponding author.
E-mail address: georgea@utsc.utoronto.ca (G.B. Arhonditsis).

https://doi.org/10.1016/j.chemosphere.2018.07.070

continue to modulate the current levels, thereby posing potential risks to humans through fish consumption.

1. Introduction

The Great Lakes support a significant recreational and commercial fishing industry. In 2010, more than half of recorded fish harvest in Canada was caught in the Province of Ontario (96 million), with walleye (*Sander vitreus*) being the most popular species amongst anglers. Total direct fishing expenditures for the Province of Ontario in 2010 amounted to more than 900 million dollars (Survey of Recreational Fishing in Canada, 2010). The popularity of fishing as a leisure activity is also closely associated with the health benefits of fish consumption. Fish provide an excellent dietary source of high nutritional quality with easily digestible protein and omega-3 fatty acids (see Cohen et al., 2005; Smith and Sahyoun, 2005; Perhar et al., 2012 for reviews). A wide array of work has been published relating dietary fatty acids to the maintenance of cognitive and nervous system functioning (Minokoshi et al., 2002), and to the improvement of 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 (Mozaffarian and Rimm, 2006). As a result of these advantageous health effects, the American Heart Association provides strong endorsement for regular fish consumption (Oken et al., 2003).

The Great Lakes have experienced varying degrees of pollution impacts, ranging from excessive nutrient loading to toxic contaminant exposure and (more recently) the onslaught of climate change and invasive species. Among the bioaccumulative, toxic and persistent organic pollutants, PCBs are of particular concern and historically have restricted the use of valuable commercial and recreational fisheries in the Great Lakes. PCBs were initially manufactured in the late 1920s by the electric industry as non-flammable additives to oils and industrial fluids, with the purpose of acting as coolers and electrical insulators (Hornbuckle et al., 2006). In their early history, PCBs posed a particular concern among electric utility workers, as excessive occupational exposure could lead to an increase in liver cirrhosis and to the development of malignant melanomas (Loomis et al., 1997; Prince et al., 2006). PCBs make up a group of 209 chlorinated congeners that are highly hydrophobic and therefore tend to tightly deposit into fatty tissues (Elskus et al., 2005). As persistent organic pollutants, PCBs are non-ionizable and largely non-polar, while their hydrophobic nature leads to their accumulation in fatty tissues and in oil rich organs and glands (Elskus et al., 2005). PCBs are known to suppress immune system function (Dallaire et al., 2003), to increase the risk of diabetes (Codru et al., 2007), to cause cardiovascular problems including coronary heart disease (Tomasallo et al., 2010), to be responsible for decreased verbal learning and increased depression (Fitzgerald et al., 2008), neurobehavioral alterations, motor immaturity, hyporeflexia, and lower psychomotor scores (Faroon et al., 2000), and to disrupt thyroid stimulating and sex steroid hormone functions (Turyk et al., 2007).

The presence of PCBs in Great Lakes fish samples was first detected in 1968, in lake trout (*Salvelinus namaycush*) and bloater (*Coregonus hoyi*) chubs caught in Lake Michigan (Veith, 1968). Depending on their trophic position, fish are typically exposed to PCBs through three routes with variant relative importance, i.e., gills, epithelial/dermal tissues, and gastrointestinal tract (Schlenk, 2005). Lower trophic level fish likely receive PCBs by the diffusion process through gills and epithelial cells, whereas top predators mainly receive them through dietary uptake of contaminated food. Responding to increased public pressure for effective elimination of persistent toxic pollutants from the Great Lakes, PCB production was banned in North America in the 1970s. The Great Lakes Water Quality Agreement (GLWQA) between Canada and USA was signed in 1972 and was subsequently revised in 1978 and 2012. The agreement aimed at identifying the spatiotemporal trends of toxic substances in sediments and biota with ultimate goal to protect the environmental integrity of the Great Lakes (International Joint Commission (IJC), 1978, 2006). Contaminant levels in edible fish portions have been monitored, from the 1970s onwards. Species-specific consumption advisories have been issued for the general and sensitive demographic groups by the Ontario Ministry of the Environment, Conservation & Parks (MECP), U.S. Great Lakes states and tribes. Monitoring of fish contamination levels has raised public awareness of potential health risks and has encouraged governmental action. The Government of Ontario introduced the Toxic Reduction Act aiming *(i)* to reduce the use and creation of toxic substances in regulated facilities and *(ii)* to inform Ontarians about toxic substances in the environment through a public open data policy of the facilities operating under the program (https://www.ontario.ca/page/eating-ontario-fish-2017-18).

Implementation of these regulatory actions resulted in reduced levels of most contaminants in Great Lakes fish through the 1980s, but the rate of decrease is reported to have diminished or to have leveled off since the early 1990s (Bhavsar et al., 2007; Sadraddini et al., 2011a,b; Visha et al., 2015). The reasons for these trends are not fully known, but existing mechanistic explanations include the food web alterations induced from invasive species in the Great Lakes (Hogan et al., 2007), as well as shifts in trophodynamics associated with global warming (French et al., 2006). Despite the valuable insights gained into contaminant dynamics through the extensive datasets developed, many studies failed to consider important factors that can modulate the inference drawn, such as fish age, size, trophic level, seasonality, growth and lipid content (Mahmood et al., 2013a,b).Variations across monitoring programs in the type of sampling procedures and different statistical methods used may also impede the robust assessment of contaminant trends (Gewurtz et al., 2011). As a result it is important to develop flexible statistical frameworks, in order to determine correct contamination trends.

To this end, a central feature of recent work in Lakes Erie and Ontario was the adoption of Bayesian inference techniques as a means for critically assessing spatiotemporal contaminant trends in fish communities over the last four decades (Lamon et al., 1998; Azim et al., 2011a,b; Sadraddini et al., 2011a,b; Mahmood et al., 2013a,b; Visha et al., 2015, 2016). The advantages of the Bayesian approach when addressing ecological questions primarily stem from its ability to explicitly accommodate model structural and parametric uncertainty, measurement errors, and data gaps (Dorazio and Johnson, 2003; Ellison, 1996, 2004; Arhonditsis et al., 2007; Blukacz-Richards et al., 2017). The primary goal of the present study was to introduce robust statistical methodologies, such