Environmental Pollution xxx (2015) 1–12

Contents lists available at ScienceDirect

Environmental Pollution

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

Invited paper

Concentration and distribution of contaminants in lake trout and walleye from the Laurentian Great Lakes (2008–2012)☆

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ARTICLE INFO

Article history:
Received 24 September 2015
Received in revised form
30 November 2015
Accepted 9 December 2015
Available online xxx

Keywords:
Environmental contaminants
Great lakes
Fish
Biomonitoring

ABSTRACT

Biomonitoring programs for persistent, bioaccumulative, and/or toxic chemicals of concern in fish tissues have been operated by the governments of Canada and the United States in the Great Lakes since the 1970's. The objectives of these programs are to assess concentrations of harmful chemicals in whole body top predator fish as an indicator of ecosystem health and to infer potential harm to fish and fish consuming wildlife in the Great Lakes Basin. Chemicals of interest are selected based upon national and binational commitments, risk assessment, and regulation, and include a wide range of compounds. This review summarizes all available data generated by Environment Canada and the United States Environmental Protection Agency for chemicals measured in whole body homogenates of Lake Trout (*Salvelinus namaycush*) and Walleye (*Sander vitreus*) for the time period spanning 2008 to 2012 from each of the five Great Lakes. The summary shows that concentrations of legacy compounds, such as, POPs listed in the Stockholm Convention and mercury continue to dominate the chemical burden of Great Lakes fish. This assessment, and others like it, can guide the creation of environmental quality targets where they are lacking, optimize chemical lists for monitoring, and prioritize chemicals of concern under agreements such as the Great Lakes Water Quality Agreement and the Stockholm Convention.

1. Introduction

Formal biomonitoring programs for persistent, bioaccumulative, and/or toxic chemicals in fish tissues have been operated by the governments of Canada and the United States in the Great Lakes basin since the 1970's (Gewurtz et al., 2011a; McGoldrick et al., 2010). At their onset, these programs directly supported commitments made in the Great Lakes Water Quality Agreement between Canada and the United States of America (GLWQA) to protect human health and the environment through cooperative and coordinated measures to reduce or eliminate releases of anthropogenic chemicals into the waters of the Great Lakes (GLWQA, 1978). The objectives are to assess concentrations of harmful chemicals in fish as an indicator of ecosystem health and to infer potential harm to fish and fish consuming wildlife in the Great Lakes Basin. These commitments were reaffirmed under Annex 3 (Chemicals of Mutual Concern) of the 2012 protocol amending the GLWQA (GLWQA, 2012).

Lake Trout (*Salvelinus namaycush*) and Walleye (*Sander vitreus*) are targeted for biomonitoring. These species are large bodied and long lived piscivorous fish which occupy the highest trophic levels where they are found and thus tend to accumulate higher levels of persistent and bioaccumulative contaminants. These two species of fish are also valuable components of the commercial and recreational fishing industry in the Great Lakes Basin (Baldwin et al., 2009).

The monitoring programs were historically focussed on measuring organochlorine pesticides (OCs) and polychlorinated biphenyls (PCBs). The measurements of these compounds in fish and other environmental compartments in the Great Lakes preceded the establishment of the Stockholm Convention on Persistent Organic Pollutants (POPs) by several decades and the data generated in the Great Lakes Region contributed to the establishment of the first list of substances, the so-called “dirty dozen”, covered by the convention. These chemicals are well known in the public sphere and are the subject of many scientific publications from the Great Lakes (ex. Borgmann and Whittle, 1991; Carlson and Swackhamer, 2006; Chang et al., 2012; De Vault et al., 1996; Huestis et al., 1996; Morrison et al., 2002; Muir et al., 2004;

☆ This paper has been recommended for acceptance by Jay Gan.
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http://dx.doi.org/10.1016/j.envpol.2015.12.019

Whittle and Fitzsimons, 1983; Whittle et al., 2000; Wong et al., 2003; Xia et al., 2012; Zananski et al., 2011).

During the last decade, initiatives in both Canada, under the Chemicals Management Plan (CMP), and the United States, under the Great Lakes Restoration Initiative (GLRI), have expanded the list of chemicals being monitored. Routine monitoring and surveillance of contaminants in fish now include additional contaminants which have emerged or are of emerging concern to cause harmful impacts in the environment. Examples of these compounds include flame retardants, such as polybrominated diphenylethers (PBDEs), surfactants, such as perfluorooctane sulfonate (PFOS), and many other chemicals being used in a variety of consumer goods and personal care products.

As a result of increased monitoring in recent years, additional information on many chemical compounds have been generated. Some of these data have been reported in independent manuscripts and/or environmental indicator reports (See Supplemental Information). This review summarizes all available data generated by Environment Canada and the United States Environmental Protection Agency for chemicals measured in whole body homogenates of Lake Trout and Walleye for the time period spanning 2008 to 2012 from each of the five Great Lakes, with the aim to provide an integrated picture of the current status of contaminant burden of upper trophic level fish in the Great Lakes Basin. The data summary presented can help to evaluate ongoing mitigation measures, guide the creation of environmental quality targets and inform decisions relating to what chemicals are of concern under agreements such as the GLWQA and Stockholm Convention.

2. Methods

All organic contaminant and mercury concentrations in whole body Lake Trout and Walleye generated from long term monitoring in the Great Lakes by Environment Canada (EC) and the United Stated Environmental Protection Agency (USEPA) between the years 2008 and 2012 were compiled (GLENDA [Great Lakes Environmental Data Base]; McGoldrick et al., 2010). Some of these are reported, in whole or in part, in publications which contain specific analytical methodologies for each parameter group. These publications are listed as a bibliography in the supplemental information associated with this manuscript. All contaminants which were detected in less than 10% of measurements made between 2008 and 2012 were excluded from this assessment. The arithmetic means and standard deviations of the concentration for all remaining contaminants over the five year period were calculated. Means and standard deviations for contaminants with measured concentrations below the detection limits were estimated using the non-parametric Kaplan–Meier method (Helsel, 2012). All compounds included in this review are listed in Table 1. Polychlorinated biphenyls (PCB) and polybrominated diphenyl ethers (PBDE) congeners were summed by homologue group denoted by the number of chlorine or bromine atoms in the congener. Co-eluting congeners of PCB with differing number of chlorine atoms were summed together as “Sum(co-eluting PCBs)”. Other chemicals were also grouped into classes for summary purposes and are listed in Table 2. As these programs measure contaminants in biological organisms, monitoring is also restricted to those contaminants which bioaccumulate and persist in fish tissues. The dataset is a compilation of all results for up to ~1800 fish samples generated by the USEPA and Environment Canada. Not all contaminants were measured at all locations in each of the Great Lakes by both agencies. A complete list of chemicals measured, waterbody, and source agency is provided in Table 1. Sample sizes for each contaminant and water body are provided in the supplemental information package (Tables S1–S5). The rankings presented are based on wet weight concentrations and are not weighted for the relative toxicities or effects they may exert on fish.

3. Results and discussion

All contaminants presented are the results of targeted analyses for chemicals and thus are not a complete reflection of the total contaminant burden of fish as there are likely many other known and unknown contaminants present in fish that are not included in the datasets. After excluding contaminants detected in less than 10% of measurements, there were 44 compounds or compound groupings remaining and included in further analyses (Table 1).

3.1. Mercury

Although not an organic pollutant, mercury was included in the analysis as it is a global contaminant of concern and the consumption of fish is an important route of exposure to humans and wildlife. Mercury was the most abundant contaminant in fish from Lake Superior, second most in Lake Huron, fourth in lakes Michigan and Erie, and was the fifth most abundant contaminant in Lake Ontario (Figs. 1–5). Between 2008 and 2012, average concentrations of total mercury were highest in Lake Superior (233 ng/g) followed by Huron (168 ng/g), Michigan (148 ng/g), Ontario (124 ng/g), and Erie (121 ng/g). The pattern is consistent with atmospheric deposition being an important source of mercury to the Great Lakes and the relative importance of atmospheric inputs relative to other contaminant inputs to Lake Superior (Lepak et al., 2015; Jiménez et al., 2015; Monson et al., 2011; Pacyna et al., 2006). Mercury levels in fish from the Great Lakes have generally declined from peak levels; however, the declines appear to have ceased in the 1990s and may be increasing at some locations, particularly in Lake Erie (EC & USEPA, 2014; Bhavsar et al., 2010; Monson et al., 2011). At present, there are no binational targets for mercury concentrations in fish. The observed concentrations of mercury in fish are generally below levels of concern for the health of fish consuming wildlife that were established in the 1987 GLWQA of 500 ng/g (GLWQA, 1987) and are not responsible for the majority of consumption advisories in the Great Lakes basin (Bhavsar et al., 2011).

3.2. Polychlorinated biphenyls (PCBs)

Polychlorinated biphenyls (PCBs) are bioaccumulative, persistent and toxic chemicals that were manufactured and used across the globe primarily as coolants and lubricants in electrical and other equipment. They were also widely used as plasticizers in products such as caulks, adhesives and paints (ATSDR, 2000). The production, importation and most uses of PCBs were banned in both Canada and the USA by 1979 and PCBs were among the 12 initial persistent organic pollutants listed under Annex A of the Stockholm Convention. From 2008 to 2012, the sum of measured PCB congener concentrations (ΣPCBs) exceed all other contaminants in Great Lakes fish by a wide margin in all lakes except in Ontario and Superior (Fig. 6). The average concentrations of ΣPCBs were highest in fish from Lake Michigan (935 ng/g) followed by Ontario (692 ng/g), Huron (653 ng/g), Erie (625 ng/g) and Superior (372 ng/g). Of the 209 PCB congeners, the hexa-chlorinated biphenyls (hexaCB) were the most abundant followed by the penta- and hepta-chlorinated biphenyls (pentaCB & heptaCB) which comprised 65–80% of all PCBs measured in fish. These three PCB homologue groups in addition to co-eluting PCBs were regularly among the 10 most abundant compounds measured in fish from the Great Lakes (Figs. 1–5). HexaCB were the most abundant contaminant group in lakes Michigan, Huron and Erie, second most

Please cite this article in press as: McGoldrick, D.J., Murphy, E.W., Concentration and distribution of contaminants in lake trout and walleye from the Laurentian Great Lakes (2008–2012), Environmental Pollution (2015), http://dx.doi.org/10.1016/j.envpol.2015.12.019