Contaminants and Ecotoxicology

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Abstract The lake charr *Salvelinus namaycush* is a sentinel of chemical pollution in North America due to its broad distribution and trophic position as an apex predator. We reviewed the extensive literature on contaminants in lake charr, as well as the more limited information on their biological effects. Strong evidence for PCBs and chlorinated dioxins/furans causing reduced hatchability of Great Lakes lake charr embryos emerged in the 1980s. Other stressors, such as thiamine deficiency and parasitism by sea lamprey *Petromyzon marinus* are now thought to be important factors. Measurements of persistent organic pollutants (POPs) and mercury (Hg) in lake charr began in the 1970s. Long-term trends showed major declines in concentrations of PCBs and dioxins/furans in lake charr in the Great Lakes. New contaminants, such as perfluoroalkyl substances and polybrominated diphenyl ethers increased until the early 2000s but are now declining. Similar trends occurred for many of the same POPs in northern large lakes in Canada. Extensive monitoring of Hg in lake charr showed small annual increases from the mid-2000s to 2015 in the Great Lakes and many inland lakes. Understanding future contaminant trends in lake

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A. M. Muir et al. (eds.), *The Lake Charr Salvelinus namaycush: Biology, Ecology, Distribution, and Management*, Fish & Fisheries Series 39,
https://doi.org/10.1007/978-3-030-62259-6_11

charr will require strong ancillary data and information on lake thermal regimes and other climate indicators.

Keywords PCBs · Bioaccumulation · Biomagnification · Climate change · Dioxins/furans · Guidelines · Mercury · Spatial trends · Temporal trends · Toxicity · Toxic equivalents (TEQs) · Toxaphene · Trophic level

1 Introduction

The lake charr *Salvelinus namaycush* has a long history as a sentinel of chemical pollution in North America due to its broad distribution and position as an apex predator in aquatic food webs. Following the discovery during the 1960s of the bioaccumulation of mercury (Hg) in freshwater fishes (Ishikura and Shibuya 1968; Johnels et al. 1968), Hg in lake charr was first measured in the early 1970s (Bache et al. 1971). The lake charr was the first species of fish in which dichlorodiphenyltrichloroethane (DDT)-related compounds were detected as a result of a detailed study of the cause of hatchery losses of lake charr free-embryos (Burdick et al. 1964). Subsequently, DDT was detected in all species of Laurentian Great Lakes (hereafter Great Lakes) fish with lake charr having the highest concentrations (Reinert 1970). Baseline studies for DDT and newly discovered polychlorinated biphenyls (PCBs) started in Lake Michigan in the early 1970s with a focus on lake charr due to their high tissue concentrations (Veith 1975). Lake charr are valuable in recreational, commercial, and sustenance fisheries across their range, so the lake charr was used to monitor contaminant levels and trends in the Great Lakes and many smaller lakes in the USA and Canada over the past 45 years. In addition, adverse effects of contaminants in wild lake charr have been studied to understand population-level effects related to exposure. Tillitt et al. (2008) extensively reviewed toxicological studies related to effects of dioxin-like chemicals on lake charr and synthesized evidence from field studies in the Great Lakes. The purpose of this chapter is to review historical, recent, and ongoing studies of contaminants in lake charr (n.b., almost all contaminants papers use the common name lake trout rather than lake charr). We review research and assessment of a wide range of chemical pollutants or contaminants from Hg, toxic heavy metals such as cadmium, to halogenated organic compounds such as PCBs, DDT, polychlorinated dibenzo-p-dioxins (PCDDs) and –dibenzofurans (PCDFs), polybrominated diphenyl ethers (PBDEs), and poly/perfluoro alkyl substances (PFASs). Generally, our review is focused on a group or total levels rather than individual contaminant congeners, isomers, or isotopes. We also reviewed studies on biological effects of these contaminants on whole ecosystems with lake charr as the top predator, such as those at the Experimental Lakes Area in northwestern Ontario, Canada (Schindler 1990, 2009).

The term “contaminants” is used in the chapter in relation to the presence of chemicals of concern in lake charr. Although the term “pollutants” is often used, contamination is the presence of a substance where it should not be or at concentrations above background, whereas pollution is contamination that results in adverse biological effects (Chapman 2007). Furthermore, chemicals may be more or less bioavailable depending on their chemical form (dissolved, particle bound, ionized, neutral) and on the environmental compartment in which they reside. Appendix 1 provides definitions of acronyms used in the chapter.

An important aspect of contaminant research is the evolution of analytical technology and laboratory methodology for detecting chemical residues in biological samples. Methods will not be reviewed herein, but are important because their evolution was required to advance the field of ecotoxicology. The invention and commercialization of the electron-capture detector for gas chromatography (GC) in the early 1960s (Lovelock 1974) enabled DDT, other chlorinated pesticides, and PCBs to be quantified in fish at part per billion (ng/g) concentrations and set off a huge expansion of measurements worldwide in the late 1960s and early 1970s. Similarly, the use of GC coupled with high-resolution mass spectrometry enabled determination of PCDD/PCDFs and other "dioxin-like" contaminants (polychloronaphthalenes, non-ortho, or mono-ortho substituted PCBs) in biological samples at picogram per gram concentrations. Since the early 2000s, liquid chromatography-tandem mass spectrometry has enabled detection of many relatively water-soluble and ionizable substances, such as PFASs and pharmaceuticals and personal care products (PPCPs) in fish. The development of the automated Hg analyzer (Salvato and Pirola 1996; U.S. EPA 2007) greatly increased the capacity for total Hg analysis in biological samples. The availability of validated, analytical methodology, particularly those promulgated by the U.S. EPA (https://www.epa.gov/measurements/collection-methods) for contaminants in environmental media including fish tissues also facilitated comparison of results among studies. Use of certified reference materials for organic contaminants and trace metals in fish from the National Institute for Standards and Technology (NIST) (Schantz et al. 2009) and the National Research Council of Canada (NRCC 2018) served to check accuracy and precision of data over time. Interlaboratory studies such as those by the U.S. Fish and Wildlife Service (Schmitt et al. 1981), NIST (Reiner et al. 2012), and the Northern Contaminants Program (Tkatcheva et al. 2013) also helped to ensure that results were comparable among labs analyzing contaminants in lake charr.

2 Ecotoxicology Studies

More than 50 substances have been tested for toxicity to lake charr (Table 1; Appendix 1) compared to 239 for rainbow trout *Oncorhynchus mykiss* (see Acute Toxicity Database (Mayer and Ellersieck 1986); https://www.cerc.usgs.gov/data/acute/acute.html). The disparity in testing reflects the commercial availability and wide use of various life stages of rainbow trout for aquatic toxicity testing relative to lake charr. Thus, much less is known about relative sensitivity to chemical exposures of lake charr than rainbow trout. By far the most toxic substance in static acute

Table 1 Aqueous, dietary, intraperitoneal dosing, and exposure studies of lake charr *Salvelinus namaycush* to toxicants based on peer-reviewed studies

Contaminant	Regime	Lowest observable effects[a]	References
Inorganics			
Aluminum	2-day-old eggs static water exposure	200 µg/L Al increased survival at pH 4.2–88% at 6 days	Hutchinson et al. (1989)
Aluminum	sac fry and swim-up fry (i.e., free embryo and post-embryos per Marsden et al. 2021)	>97% survival at pH ≥ 5.1with 200 µg/L Al	Hutchinson et al. (1989)
Arsenic	Adult (2 year) 20 day dietary 0, 100, 1000 µg As/g	Feed refusal, lipid peroxides ↑ at 1000 µg/g, MT induction not observed	Pedlar et al. (2002)
Copper	Embryos and early juveniles exposure at 3–450 µg/L	Embryo mortality at 555 µg/L. Post-hatch early juvenile mortality at >43.5 µg/L	McKim et al. (1978)
Mercury	Fertilized eggs	0.06–14 mg/L produced mortality 0.005–0.014 mg/L did not decrease median hatch time (MHT)	Klaverkamp et al. (1983)
Nickel	Adult (2 year) 18 day dietary	Significant ↑ MT at 1000 µg/g Ni	Ptashynski et al. (2001)
Selenium	Fertilized eggs	200 mg/L Se decreased MHT and delayed mercury-induced morality	Klaverkamp et al. (1983)
H+ ion	2-day-old eggs static water exposure	<50% survival after 4–6 day exposure to pH 4.2	Hutchinson et al. (1989)
H+ ion	Sac fry and swim-up fry	<30% survival after 6 d exposure to pH 4.2	Hutchinson et al. (1989)
Organics			
2,3,7,8-TCDD	Fertilized eggs (48 h static renewal water exposure)	>50% mortality of fertilized eggs at hatching at 100 pg/L or 400 pg/g in eggs. Smaller but significant effect at 40 pg/g in eggs	Spitsbergen et al. (1991)
2,3,7,8-TCDD	Fertilized eggs (48 h static renewal water exposure)	LD50 = 85 (36–210) pg/g egg	Zabel et al. (1995)
2,3,7,8-TCDD	Eggs were injected at 24–50 h post-fertilization with TCDD	CYP1A ↑ expression at TCDD > 88 pg/g egg. Lowest observable effect at 22 pg/g	Guiney et al. (1997)
2,3,7,8-TCDD	Fertilized eggs static-renewal water exposure	Lowest observable effect on sac fry mortality at 51 pg/g in eggs; TCDD at 253 pg/g in eggs induced CYP1A mRNA levels	Guiney et al. (2000)
2,3,7,8-TCDD	Maternally derived TCDD to oocytes	Nonviable oocytes at 233–387 pg/g egg; dose-related sac fry mortality at 50–152 pg/g. No effect of dietary exposure on adults	Walker et al. (1994)

(continued)

Table 1 (continued)

Contaminant	Regime	Lowest observable effects[a]	References
PCB-126	Fertilized eggs (48 h static renewal water exposure)	LD50 = 29,000 (18,000–34,000) pg/g egg	Zabel et al. (1995)
PCB-126	Juvenile ip-injection or dietary gavage (0.7–40 ng/g whole body)	40 ng/g exposure increased thyroid epithelial cell height, plasma T4 dynamics, and T4-glucuronidation. T3 status and fish growth were unaffected	Brown et al. (2004)
PCB-126	Juvenile ip-injection (0.6–25 ng/g whole body)	Hepatic mixed-function oxidase activity (EROD) and oxidative stress indicator (TBARS) significantly ↑ at 6 ng/g and 25 ng/g	Palace et al. (1996)
Toxaphene	Wild adult—5 year after IP injection at 7 μg/g or 3.5 μg/g	Decreased survival 1 year and failure to spawn in the first year following 7 μg/g injection	Delorme et al. (1999)
Erythromycin	Adult water and food exposure	96 h LC50 = 410 mg/L, no effect on growth at 24 mg/g	Marking et al. (1988)
Oxytetracycline	Adult water and food exposure	96 h LC50 <200 mg/L, no effect on growth at 5.5 mg/g	Marking et al. (1988)
Tetracycline	Adult water and food exposure	96 h LC50 = 220 mg/L, no effect on growth at 5.5 mg/g	Marking et al. (1988)
Polyelectrolyte “Superfloc 330”	Juvenile flowing water exposure	LC50 = 0.31 mg/L (11-day exposure)	Biesinger et al. (1976)
Polyelectrolyte “Calgon M-500”	Juvenile static water exposure	LC50 = 2.90 mg/L (2-day exposure)	Biesinger et al. (1976)
Pydraul 50E (organophosphate esters)	Adult flowing water exposure	Survival reduced at 16 μg/L after 90 days, growth and cataracts affected at 5.3 μg/L	Mayer et al. (1993)
Dinoseb	96 h flow-through exposure	96 h LC50 = 79 (67–93) μg/L	Woodward (1976)
Picloram	96 h flow-through exposure	96 h LC50 = 1850 (1630–2100) μg/L	Woodward (1976)

Note that the life stage terminology from the original studies was retained in the table and maybe inconsistent with Marsden et al. (2021)

[a] *MT* metallothionein

toxicity testing of juvenile lake charr was antimycin A, an antifungal antibiotic, with a 24-h LC-50 of 0.053 μg/L. Antimycin A is very toxic and was tested in the 1960s for use to remove fish from lakes (Walker and Lennon 1964). Synthetic pyrethroids (resmethrin and bioethanomethrin) also had LC-50s in static and flow-through testing at or below 1.2 μg/L (Appendix 1). For most other organic and inorganic substances, 96 h LC50s from acute (typically static) or chronic (usually flow through) exposures of juvenile or adult lake charr were in the 100s of μg/L to 100s of mg/L (Table 1 and Appendix 1) and thus at levels unlikely to be reached from typical use in pesticide applications or from emissions due to industrial uses, apart from accidental spills. However, post-hatch age-0, juvenile mortality is a more sensitive indicator of chemical toxicity to lake charr than tests with adult fish. For example, larvae (i.e., post embryos in lake charr) and age-0 juvenile stages of lake charr, and other fish species, were more sensitive to copper than pre-hatch embryos or adults (Table 1; McKim et al. 1978). Little information is available on toxicity of most chemicals to early life stages of lake charr with the important exception of 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) and related compounds (reviewed by Tillitt et al. 2008 and King-Heiden et al. 2012).

Early life stages of vertebrates were known to be the most sensitive to toxicity of TCDD and related planar chlorinated aromatic compounds from results of mammalian and avian research (Gilbertson 1982; McConnell 1980). Thus, the focus of studies of developmental toxicity of TCDD in lake charr and other fishes used fertilized eggs. For example, TCDD toxicity in lake charr (LD-50 = 40–85 pg/g) was quantified for TCDD-injected pre-hatch embryos (fertilized eggs) (Spitsbergen et al. 1991). Induced early life stage mortality in rainbow trout embryos facilitated development of fish-specific TCDD toxic equivalent factors (TEFs) for PCDD/Fs and selected PCB congeners (Walker and Peterson 1991). Subsequent work using a mixture of 2,3,7,8-substituted PCDDs, PCDFs, and non-ortho substituted PCBs showed that these compounds acted via a common mechanism of aryl hydrocarbon receptor (AhR) mediated toxicity to cause lake charr free embryo mortality although their toxicity was not strictly additive (Walker et al. 1996). Of 11 species tested, lake charr was the most sensitive to TCDD, with pre-hatch embryo TCDD LD50s (40–85 pg/g) about threefold less than brook charr *Salvelinus fontinalis* (138–200 pg/g) and fivefold less than rainbow trout (230–488 pg/g) (Tillitt et al. 2008).

Cook et al. (2003) used the TEFs developed from work with individual PCDD/Fs and PCBs on early-life-stage mortality in rainbow trout to estimate 2,3,7,8-TCDD toxicity equivalence in their retrospective risk analysis of dioxin-like compounds in Lake Ontario lake charr. Tillitt et al. (2008) also reviewed the evidence for the extirpation of lake charr in the lower Great Lakes by PCDD/Fs and related dioxin-like chemicals and concluded that a strong case existed for reproductive failure from the mid-1970s to 1980s (Table 2).

Strong evidence for maternally derived PCBs causing reduced hatchability in embryos from southeastern Lake Michigan was available prior to most of the detailed information on the mechanism of AhR mediated toxicity of TCDD (Mac and Edsall 1991). By the early 1990s, TCDD and related compound levels in lake charr pre-hatch embryos in the Great Lakes decreased below levels expected to cause mortality and could not explain observed rates of juvenile lake charr mortality (Harder et al. 2018). In a review of work on early life stage mortality in lake charr, Fitzsimons (1995) concluded that reduced hatching, blue sac disease, and swim-up

Table 2 Criteria and evidence described by Tillitt et al. (2008) to support the relationship between PCDD/Fs and related dioxin-like chemicals and the extirpation of lake charr *Salvelinus namaycush* in the lower Great Lakes (i.e., Erie and Ontario)

Criteria	Evidences
Probability	Lake charr were exposed to elevated concentrations from the initial production and release of these chemicals in the 1930s into the Great Lakes ecosystem until after they were banned in the late 1970s. Concentrations of dioxin toxic equivalents in fertilized eggs of Great Lakes lake charr exceeded these thresholds for toxicity.
Timing	Temporal coincidence of rises in dioxin-like contaminant concentration and increases in reproductive failure.
Strength of association	Geographic distribution of dioxin-like pathologies including blue-sac-related mortality. The term blue sac related mortality or syndrome, was used by Spitsbergen et al. (1991) and in subsequent articles (Walker et al. 1996; Walker and Peterson 1994) to describe the effects produced by TCDD and other dioxin-like chemicals which they concluded were identical to "blue-sac disease," a condition which commonly observed in hatchery-reared salmonines.
Specificity	Presence of blue sac disease in Great Lakes lake charr from the mid-1970s to 1980s was consistent with aryl hydrocarbon receptor (AhR)-related toxicity.
Consistency of association	Geographic distribution of symptoms of dioxin-like toxicity in lake charr was consistent with the relative degree of contamination in the Great Lakes. Signs of toxicity were observed in different species at the same locations.
Predictive performance	Cook et al. (2003) found that sediment core profiles of TCDD and dioxin-like PCBs in Lake Ontario were predictive of the population abundance of lake charr, the signs of sac-fry mortality, and reductions in post-hatch embryo survival. Extracts of lake charr from Lake Michigan produced graded, dose-dependent symptoms of blue sac syndrome in hatchery-raised lake charr embryos (Tillitt and Wright 1997).
Coherence	Lake charr have been found to be the most sensitive species of fish toward dioxin (Spitsbergen et al. 1991; Walker and Peterson 1991). Lake Superior lake charr had significantly lower concentrations of PCBs and PCDD/Fs, were not observed to have symptoms of TCDD-related toxicity, and were not suffering from a lack of recruitment.

syndrome mortality in lake charr eggs were not solely related to the TCDD and other contaminants, and identified pre-hatch embryo rearing conditions, nutrition, poor egg quality, and altered physiology as alternative explanations for early life stage mortality. Subsequent research pointed to the role of thiamine deficiency as a contributing or causal factor in the reproductive problems of lake charr in the Great Lakes from the 1980s onward (Brown et al. 2005; Fisher et al. 1996; Fitzsimons et al. 1995). Tillitt et al. (2008) concluded that thiamine deficiency, parasitism by sea lamprey, and predation on lake charr free embryos by alewife *Alosa pseudoharengus* (an invasive species in the Great Lakes), could be important factors in addition to impacts of contaminants. Clearly, multiple stressors impinged on lake charr populations in the Great Lakes, with the importance of other factors revealed as one stressor was reduced (D. Tillitt, U.S. Geological Survey, personal communication).

2.1 Field-Based Studies of Toxicity to Lake Charr

Early studies suggested that DDT was associated with reproductive failure in lake charr. Elevated DDT-related compounds were correlated with reduced survival of post-hatch embryos in upstate New York lakes with no survival in lake charr post-hatch embryos that contained DDT over 2950 ng/g wet weight (ww) (Burdick et al. 1964). However, Burdick et al.'s methodology did not enable measurements of PCBs, PCDD/Fs, or other chlorinated pesticides. A subsequent study showed that adult lake charr fed diets with 6000 ng DDT per g feed produced 100% mortality in the lake charr post-hatch embryos (Burdick et al. 1972). DDT concentrations in lake charr in Lakes Ontario and Michigan in the 1970s were high enough (means ranging from 5000 to 20,000 ng/g ww) to have contributed to lake charr pre-hatch embryo mortality (Zint et al. 1995).

A series of field experiments involving exposure to toxic contaminants have provided insights into contaminant effects on adult lake charr under natural conditions. Lake charr were treated with a single intraperitoneal injection of toxaphene in Lake 260 in the Experimental Lakes Area (ELA) over a two-year period at nominal doses of 3.5–7 μg/g ww (Delorme et al. 1993, 1999). These doses were similar to concentrations in Great Lakes fishes. Toxaphene is a major organochlorine pesticide contaminant in Great Lakes fishes (Muir et al. 2006; Xia et al. 2012) and its toxicity to fish is well known (Eisler and Jacknow 1985). No threshold has been formally established for adverse effects of toxaphene in lake charr, but treated adult lake charr survival was lower than controls and they apparently failed to spawn in the first year following injection (Delorme et al. 1993, 1999). The percentage of lake charr eggs successfully fertilized from toxaphene-treated female lake charr did not differ from controls and no major changes in growth were evident. Delorme et al. (1993, 1999) concluded that cumulative effects of toxaphene exposure conducted under a multiple stressor environment of a natural lake had the potential to alter lake charr population dynamics.

The biomass of lake charr in Lake 260 at the ELA declined 23–42% during and after addition of 17α-ethynylestradiol (EE2) to the lake, likely an indirect effect from loss of its prey, the fathead minnow *Pimephales promelas*, pearl dace *Margariscus margarita*, and slimy sculpin *Cottus cognatus* (Kidd et al. 2014). Vitellogenin (VTG) was induced up to 18,700 fold in male lake charr compared to reference lakes (Table 3); however, intersex and effects on gonadal development were not detected (Palace et al. 2009). Because EE2 was added only to the epilimnion during summer, EE2 exposure to lake charr occurred after lake turnover during autumn. Exposure of lake charr to EE2 could also have occurred via the diet given the high exposure of lake charr prey, the fathead minnow, and pearl dace, to EE2 in L260.

Table 3 Mean vitellogenin induction in male fathead minnow *Pimephales promelas*, pearl dace *Margariscus margarita*, lake charr *Salvelinus namaycush*, and white sucker *Catostomus commersonii* in Lake 260 of the Experimental Lakes Area of Ontario treated with 17a-ethynyloestradiol (EE2) compared to reference lakes (Palace et al. 2009)

Year	Season	Fathead minnow (homogenate)	Pearl dace (homogenate)	Lake charr (plasma)	White sucker (plasma)
2001	Fall	22200	5900	1900	22
2002	Spring	3800	3000	11500	24
	Fall	8700	3800	2100	62
2003	Spring	6500	15900	18700	94
	Fall	24200	2300	4600	118

Food-web collapse during a lake acidification experiment resulted in lower lake charr survival over a 7-year period in which the epilimnetic pH was lowered from 6.49 to 5.13 (Mills et al. 1987; Schindler et al. 1985). Condition factor ($CF = 100 \times \text{weight}/\text{length}^3$) of lake charr in Lake 223, ELA gradually declined from 1977 (pH 6.13) to 1983 (pH 5.13; Mills et al. (1987). In 1983, lake charr condition in Lake 223 was less than at any time in any reference lake. By 1983, lake charr were emaciated due to losses of prey organisms. The threshold for lake charr recruitment failure in Lake 223 was pH 5.59, similar to observations for lake charr populations in New York State Adirondack lakes and in a central Ontario survey (Beggs et al. 1985; Schofield 1982).

A whole-lake experimental addition of cadmium (Cd) to ELA Lake 382 did not result in observable effects on lake charr. The study investigated the fate and effects of low concentrations of Cd, not exceeding the Canadian Water Quality Guideline (CWQG) at the time of 200 ng/L (Malley 1996). Over a 6-year period of addition of $CdCl_2$ to the epilimnion, Cd concentrations in water ranged 20–240 ng/L (Lawrence et al. 1996). Cd concentrations in the posterior kidney of lake charr reached 9.5 μg/g ww, a sixfold increase above background. After five years of Cd additions, concentrations in lake charr skeletal muscle remained at background levels of 4 to 8 ng/g ww (Malley, 1996).

Whole lake studies at the ELA illustrate the need to study effects of contaminants at the whole ecosystem level (Blanchfield et al. 2009). While not practical for all chemical contaminant issues due to the need to restore altered habitat to its original state (as required at ELA), ELA experiments yielded unique insights into effects of environmental contaminants on lake charr.

2.2 *Bioaccumulation Studies of Organic Contaminants*

Early studies of PCBs and DDT-related compounds in lake charr, forage fishes, zooplankton, and water from western Lake Superior demonstrated biomagnification of these contaminants in the food web (Veith et al. 1977) and led to the question of the relative importance of water versus dietary uptake pathways for contaminants.

Juvenile lake charr had a bioconcentration factor (BCF) of 53000 for the uptake of p,p′-DDT from water, which was exceeded by p,p′-DDT uptake from food during a 90-day exposure (Reinert et al. 1974). Both water and dietary uptake routes were important for dieldrin, which was eliminated more rapidly following transfer of fish to uncontaminated water (Reinert et al. 1974). Lake charr exposed to p,p′-DDT, p,p′-DDE (100 ng/L), and PCBs as Aroclor 1254 (100 and 500 ng/L) in a flow-through system over a 12-week period, increased tenfold in concentration for both DDT and PCB related compounds, but did not reach equilibrium (Hesselberg and Nicholson 1981). Laboratory exposures by Reinert et al. and Hesselberg and Nicholson represent the only bioconcentration studies (i.e., uptake) from water only, for organic contaminants in lake charr. However, the question of the importance of waterborne vs. dietary exposure was addressed by many other studies with closely related species, especially rainbow trout. In a long-term (96 days) exposure of rainbow trout to 34 organohalogen compounds, BCFs were much lower than field-derived bioaccumulation factors (BAFs) particularly for chemicals with long half-lives in fish (Niimi and Oliver 1985).

Simultaneous measurements of concentrations in lake charr, water, and sediment demonstrated very high BAFs for persistent and bioaccumulative substances in lake charr. Using data from Metcalfe and Metcalfe (1997), who measured PCB congeners in lake charr and water from north-central Lake Ontario, BAFs (concentration in fish lipid normalized; C_F) divided by concentration in water (dissolved) ranged $0.13–4.07 \times 10^6$ for PCB congeners based on lipid weight (Arnot and Gobas 2006). Based on dissolved PCBs and PBDEs in Lake Michigan, BAFs ranged $0.316–316 \times 10^6$ for PCBs and $0.501–31 \times 10^6$ for PBDE congeners 47, 66, 99, and 100 (Streets et al. 2006). BAFs were correlated with log octanol–water partition coefficient (log K_{ow}) (Fig. 1) and therefore could be predicted relatively accurately from basic physical–chemical properties of the compounds (Streets et al. 2006). Thus, selected congeners could be used for monitoring these contaminants in lake charr and dissolved concentrations in water could be inferred.

BAFs for lake charr in the Great Lakes have also been reported for perfluoro alkyl substances (PFASs) (Furdui et al. 2007), toxaphene (Muir et al. 2004), and Hg (Raymond and Rossmann 2009). While BAFs illustrate the tremendous bioaccumulation potential of persistent organic pollutants (POPs) and could be used to compare lakes and food webs assuming that lake charr were at the same trophic level (Streets et al. 2006), they do not provide information on pathways of bioaccumulation.

Biota–sediment accumulation factors (BSAFs) have been used to assess exposure of invertebrates and fishes, including lake charr, to sediment-associated organic contaminants (Burkhard et al. 2012). BSAFs are calculated by dividing the contaminant concentration (lipid normalized) in whole fish by the concentration in sediment (organic carbon normalized). For fishes, especially those not in direct contact with sediments, BSAFs incorporate a range of pathways and processes including biomagnification, sediment-water column chemical exchange, food-web characteristics, the fish's home range, and transformation of the chemical within the fish and its food web. Burkhard et al. (2004) developed a BSAF data set for PCBs, PCDDs,

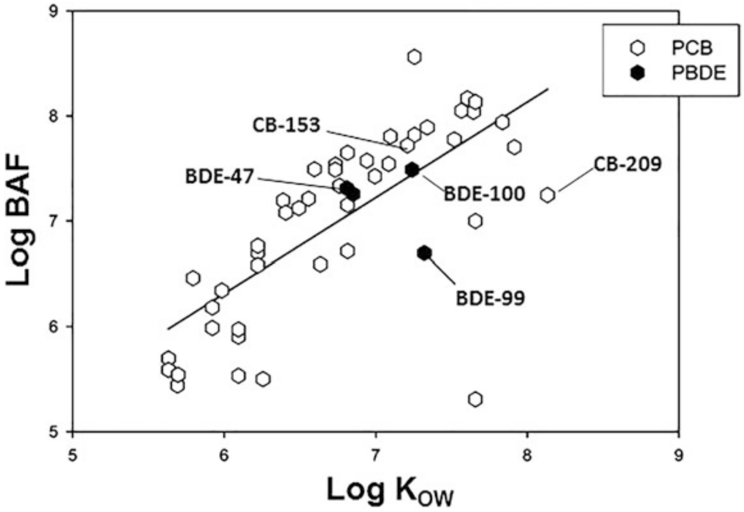

Fig. 1 Bioaccumulation factors versus octanol–water partition coefficients (log BAF vs log K_{ow}) for PCB (open hexagons) and PBDE (closed hexagons) congeners for lake charr *Salvelinus namaycush* in Lake Michigan (Streets et al. 2006)

and PCDFs in southeastern Lake Michigan lake charr. The top 1–2 cm layer was used for determining concentrations in sediment on the basis that it would best represent the exposure history of the lake charr. BSAFs for PCBs and PCDD/Fs for year classes 2–9 were remarkably similar despite known differences in diet. Differences in lipid fraction helped explain a similar range from 5% in 2-year-old lake charr to 18.5% in 9-year-old fish. Variation of BSAFs with log K_{ow} (Fig. 2) for the 6-year-old age class was lower than predicted BSAF based on the Gobas food chain model (described below). The authors concluded that the generally lower observed than predicted BSAFs for PCBs were mainly due to metabolism of some congeners by lake charr. This conclusion was also apparent from the fact that PCDD/Fs had very low BSAFs (0.001–0.2) compared to PCBs (0.1–11), which was consistent with much more rapid elimination and metabolism.

BSAFs were used to relate measurements of PCDD, PCDF, and PCB congeners in Lake Ontario sediments to lipid-normalized concentrations in lake charr unfertilized eggs (Cook et al. 2003). BSAFs for lake charr female and pre-hatch embryos ranged range from >10 to 0.3 for PCBs and from 0.27 to <0.001 for PCDD/Fs. This range was almost identical to BSAF values in southern Lake Michigan lake charr (Burkhard et al. 2004). BSAFs for PCBs in lake charr from Lake Michigan were also strongly correlated with BSAFs for other species, such as adult alewife and walleye in Green Bay ($P<0.003$), and smallmouth bass *Micropterus dolomieu* and yellow perch *Perca flavescens* ($P<0.0001$) in the Hudson River, although absolute values differed (Burkhard et al. 2005). Nevertheless, the relative values of BSAFs for a wide range of PCBs were similar for lake charr and

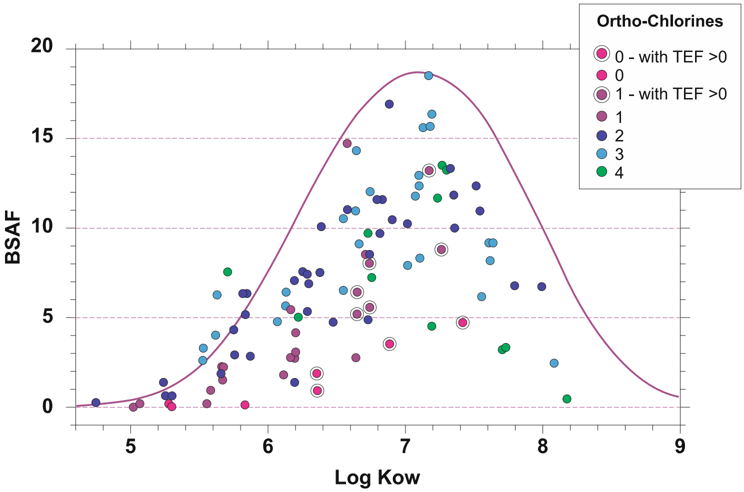

Fig. 2 Biota-sediment accumulation factors (BSAFs) for PCBs in Lake Michigan lake charr *Salvelinus namaycush* (redrawn from Burkhard et al. 2004). Red, purple, blue, turquoise, and green circles represent PCB congeners with 0, 1, 2, 3, and 4 ortho-chlorines, respectively. Double circles are congeners with toxic equivalent factors (TEFs)>0. The curved line illustrates the expected relationship of BSAFs with K_{ow} predicted by use of the Gobas food-web model (Gobas 1993) assuming no metabolism

other fishes across ecosystems. Thus, prevailing concentrations of bioaccumulative chemicals in sediments could be inferred from monitoring of lake charr.

Almost all dietary absorption, uptake rates, and elimination half-lives ($t_{1/2}$) of chemicals in salmonids have been developed using rainbow trout in the 1970s and 1980s (Niimi 1987). However, dietary absorption, or net trophic transfer efficiency (γ), of 75 PCB congeners by lake charr in a laboratory experiment using bloater *Coregonus hoyi* (fed ad libitum) as the source of dietary PCBs was defined as (Madenjian et al. 2012b, 2014):

$$\gamma = \Delta\text{PCB body burden (ng)} \div \text{amount of PCB ingested (ng)}$$

where ΔPCB body burden is the increase during the experiment (determined using average weight of the lake charr × average concentration in whole fish). Increasing the feeding rate caused γ to decrease, whereas the degree of chlorination of the PCB congeners was unrelated to γ, which averaged 0.66 across all congeners (Madenjian et al. 2012b, 2014). However, γ decreased significantly as log K_{ow} increased from 6.0 to 8.2 (Fig. 3), which suggested lower bioavailability of more hydrophobic, highly chlorinated, congeners (Madenjian et al. 2012b, 2014). Activity level did not have a significant effect on γ (Madenjian et al. 2012b, 2014). In a review of dietary bioaccumulation studies of organic contaminants, one (Tomy et al. 2004) of 98 dietary exposure studies on 477 discrete organic chemicals used lake charr (Arnot and

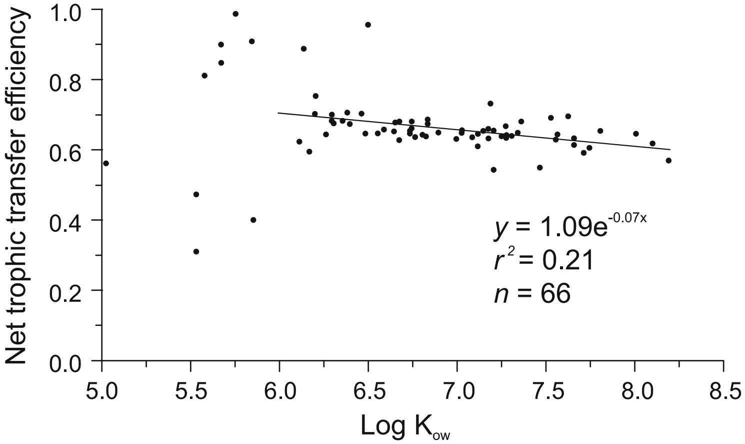

Fig. 3 Estimate of net trophic transfer efficiency (γ) of PCB congeners to lake charr *Salvelinus namaycush* from its prey as a function of the log K_{ow}. Estimates were based on a laboratory experiment, during which lake charr were fed bloater *Coregonus hoyi*. Fitted regression line for congeners with log K_{ow} greater than 6 is also displayed (Madenjian et al. 2012b)

Quinn 2015). Tomy et al. (2004) examined dietary update of polybrominated diphenyl ethers (PBDEs) in juvenile lake charr during a 56-day feeding phase and a 112-day depuration phase (on clean food), along with effects on cytochrome P450 (CYP 1A) enzyme activity, measured as EROD, and free tri-iodothyronine (T3). Half-lives of four PBDE congeners (47, 99, 100, 183) in a low dose exposure (1.1–2.1 ng/g dw food) ranged from 39 to 87 days (Tomy et al. 2004). A higher dose exposure with the same four PBDE congeners were distinctly greater $t_{1/2}$ (173–346 days). Two other BDEs (28, a tribromo-congener and 209, decabromo-) had shorter $t_{1/2}$ (58 and 26 days, respectively; Tomy et al. 2004). The absorption efficiency of very hydrophobic and high molecular weight congener BDE209 in lake charr was much lower (5%) than tribromo- to heptabromo (23–53%) (Tomy et al. 2004). Dietary bioaccumulations of several other BDE congeners (e.g., BDE-66, -77, -153, and -154) by lake charr were much longer than anticipated, likely because these congeners were formed via debromination of other congeners, in addition to being accumulated from food (Tomy et al. 2004). Debromination of PBDE congeners by carp *Cyprinus carpio* (Stapleton et al. 2004) and common sole *Solea solea L.* (Munschy et al. 2011) has also been reported.

Half-lives and absorption efficiencies reported for PBDEs in lake charr can be compared to other species (Appendix 2) for which results of studies of similar design (feeding rates, exposure times, and depuration times) were available from the database for dietary bioaccumulation (Arnot and Quinn 2015). However, concentrations of food varied greatly among studies. Whole-body $t_{1/2}$ of BDE 47 (a tetrabromo congener) were similar in lake charr, carp, and sole (39, 30, and

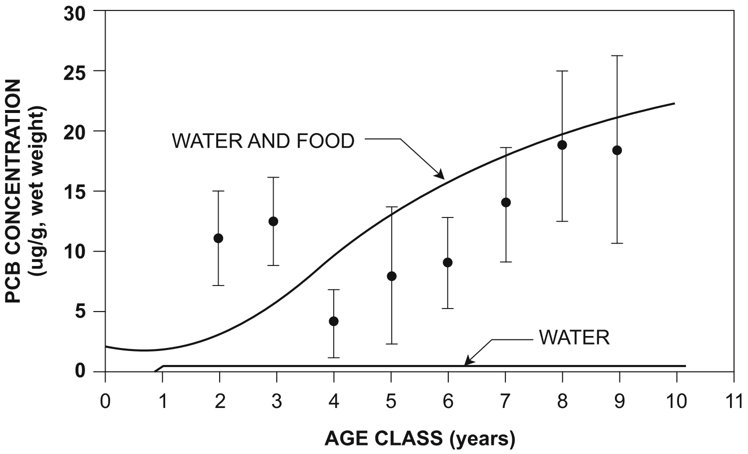

Fig. 4 Predicted versus observed concentrations of PCBs in various lake charr *Salvelinus namaycush* age classes from Lake Michigan from the Thomann and Connally (1984) food chain model. Estimated concentrations from water only exposure and water + food exposure are shown. *Reprinted with permission*

37 days). Half-lives of BDE 28, 100, and 209 in lake charr were within a factor of two of those for sole. Zebrafish had much shorter $t_{1/2}$ for BDE 28, 183, and 209 than lake charr. Fish size and exposure temperature likely influenced some differences (e.g., zebrafish were held at 26 °C versus 12 °C for lake charr). Biotransformation of PCBs to hydroxy-PCBs was positively related to water temperature in rainbow trout (Buckman et al. 2007), although applicability to rates of debromination of PBDEs is uncertain. In vitro studies using liver microsomal incubations showed that debromination rates of PBDE congeners were generally 10–100 times faster in carp than in rainbow trout and Chinook salmon *Oncorhynchus tshawytscha* (Roberts et al. 2011). Further studies of the biotransformation capacity of lake charr for organic contaminants would be useful.

During the 1970s, considerable debate occurred over whether chemicals in fish could be estimated simply from water concentrations using a BCF or log K_{ow} (Hamelink and Spacie 1977). Simple correlative approaches worked well for a wide range of chemicals based on laboratory studies with the fathead minnow (Mackay 1982; Veith et al. 1979), but the approach failed for top predators like the lake charr. Simple empirical correlations between laboratory-derived log K_{ow} and BCFs for PCBs failed to reproduce observed concentrations in lake charr or alewife, even after accounting for fraction of lipid (Thomann and Connolly 1984); Connolly and Pedersen 1988; Thomann 1989). The first food-chain model for PCBs that included 13 age classes of lake charr and predator–prey bioenergetic information for each age class was able to reproduce age-dependent trends and levels of PCB in Lake Michigan lake charr (Thomann and Connolly 1984; Fig. 4). The bioenergetics component accounted for respiration and body weight, metabolic rate, and food assimilation efficiency, based on earlier modelling of PCB accumulation by yellow perch (Norstrom et al. 1976). Connolly and Pedersen (1988) hypothesized that biomagnification occurred due to the development of a chemical gradient in the gut of the fish from preferential lipid breakdown. This concept was subsequently supported by fundamental physical chemistry principles based on diffusion of chemicals from compartments of high fugacity (GI tract) to lipid pools of lower fugacity within the organism (Campfens and Mackay 1997; Gobas et al. 1999).

A bioenergetics model evaluation of PCB accumulation in Lake Michigan lake charr based on knowledge of PCB concentration in lake charr prey consumption (Stewart et al. 1983) indicated that PCB accumulation in lake charr was driven by food consumption, whereas direct uptake of PCBs from the water was negligible (Weininger 1978). Further, lake charr PCB concentration was functionally independent of its lipid content (Weininger 1978). Similarly, a modeling comparison of direct uptake of PCBs by lake charr in Lake Michigan versus uptake of PCBs from food consumption suggested that PCB accumulation was independent of lipid content of the lake charr (Madenjian et al. 1993). In the latter study, bioenergetic dietary accumulation further accounted for individual variation of PCBs in lake charr using a predator–prey encounter submodel that assigned a prey encounter rate that varied with lake charr age and foraging season (Madenjian et al. 1993).

A simplified approach to evaluating PCB dynamics in food webs used a spreadsheet model that incorporated multiple feeding interactions within benthic and pelagic food chains (Fig. 5; Gobas 1993). The Gobas model was validated using concentrations of PCBs in the most important invertebrates and fish species in Lake Ontario from a study of 90 individual chlorinated organics in lake charr and three other species of salmonids (coho salmon *Oncorhynchus kisutch*, brown trout *Salmo trutta*, and rainbow trout) in water, sediment, forage fish, amphipods, oligochaetes, and plankton in Lake Ontario (Oliver and Niimi 1988). Unfortunately, results for lake charr were not reported separately, so no species-specific BAFs or biomagnification factors (BMFs) could be developed. Nevertheless, data were useful for validating the Gobas model, which included a generic salmonid compartment (Fig. 5). The Gobas model was later revised to include aquatic plants and non-lipid organic matter fractions in the diet (Arnot and Gobas 2003, 2004). Later, quantitative structure–activity relationships (QSAR) were developed for predicting biotransformation rates in fish (Arnot et al. 2009). Prediction of BAF and biotransformation rates was included in the U.S. Environmental Protection Agency EPISuite software package (U.S. EPA 2011) and is now widely used to predict organic contaminant concentrations in top predator fishes including lake charr. However, bioaccumulation parameters for dietary assimilation efficiency, uptake from water, biotransformation, and elimination rates in these models do not include data for lake charr, but rather are generally based on data for rainbow trout or other salmonids. Adult lake charr are difficult to work with because of their large size, long time to maturity, and restrictive water quality requirements (Mac and Edsall 1991). These challenges have limited the number of studies and ultimately impacted the accuracy of modeled predictions of lake charr exposure to contaminants.

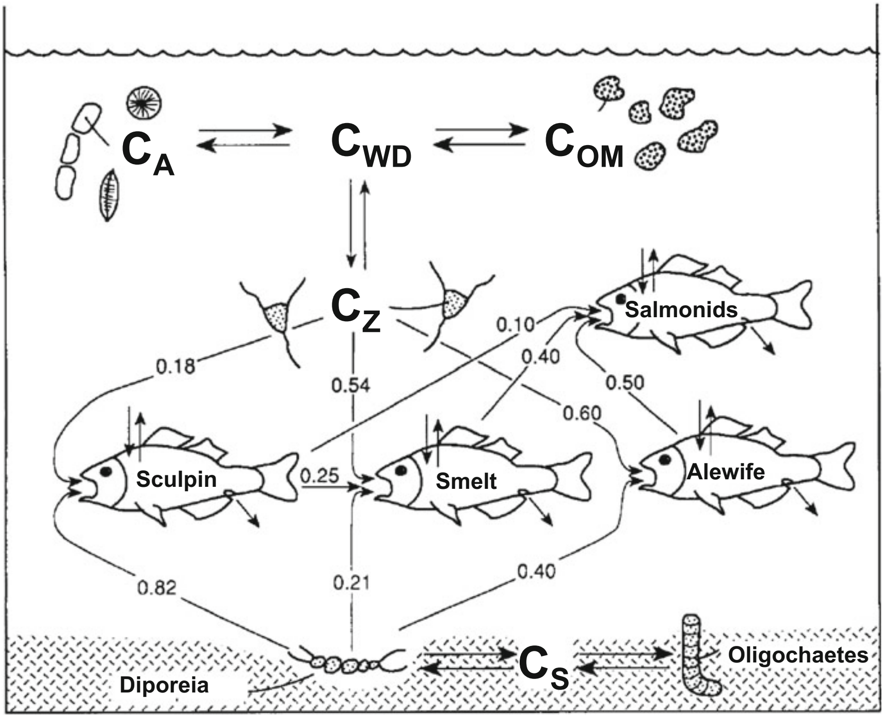

Fig. 5 Schematic illustrating the chemical transfer in the Lake Ontario food-web (Gobas 1993) with salmonids, including lake charr *Salvelinus namaycush* as top predators. C_A = chemical concentration in the organism, C_{WD} = bioavailable (dissolved) concentration in the water, C_{OM} = concentration on organic carbon, C_S = concentration in sediment, and C_Z = concentration in zooplankton

2.2.1 Bioaccumulation of Mercury

A mass balance model was used to predict age-specific Hg concentration in lake charr in Lake Memphremagog (Trudel and Rasmussen 2001). Compared to a bioenergetics approach for modeling food consumption and uptake of organic contaminants by lake charr (Thomann 1989; Madenjian et al. 1993; Luk and Brockway 1997), using a ^{137}Cs method to model food consumption and bioenergetics activity rates (Rowan and Rasmussen 1996) required accurate estimates of feeding rates to predict Hg concentrations in fish (Trudel and Rasmussen 2001). The key to accurately predicting Hg accumulation in fish is to adequately describe activity costs as a mechanism regulating bioenergetics processes in wild fish under natural conditions (Trudel and Rasmussen 2006).

Key pharmacokinetic parameters for modelling inorganic Hg and methyl Hg bioaccumulation in lake charr have not been measured directly, like other studies of organic contaminants. Based on a compilation of studies, the elimination rate of Hg is a function of fish size and water temperature (Trudel and Rasmussen 1997).

Elimination rate of inorganic Hg from fish is 2.8 times greater than that of methyl Hg, which combines with a higher assimilation efficiency of methyl Hg from food to cause a predominance of methyl Hg in fish (Trudel and Rasmussen 1997). However, lack of methyl Hg elimination rate data for lake charr as of the mid-1990s has forced modelers to use estimates for uptake from water and food and elimination rates based on data for other species. For example, modelling efforts assumed an assimilation efficiency (net trophic transfer efficiency) for methyl Hg of 80% (Norstrom et al. 1976; Trudel and Rasmussen 2001, 2006) and 84% (Borgmann and Whittle 1992).

In a large laboratory experiment to determine net trophic transfer efficiency (γ) and elimination rates of methyl Hg and inorganic Hg in adult (1 kg) lake charr fed naturally contaminated bloater from Lake Michigan, γ was 76.6% for methyl Hg and 63.5% for inorganic mercury, and did not differ between active and inactive lake charr (activity level was compared by adjusting water flow and volume for a subset of fish; Madenjian et al. 2012a). The lower transfer of inorganic Hg was likely due to its greater elimination rate, while the elimination rate of methyl Hg (0.000244 day^{-1}, Madenjian et al. 2012a) was 5.5 times lower than predicted by the chronic exposure equation for elimination of methyl Hg (Trudel and Rasmussen 1997). Lake whitefish *Coregonus clupeaformis* eliminated Hg at a rate nearly three times higher than lake charr, even though the two species were roughly equal in size and kept at similar temperatures (Madenjian et al. 2012a). Studies of methyl Hg depuration by northern pike *Esox lucius*, a large top predator following exposure to isotope-enriched methyl Hg in a whole-lake Hg experiment at ELA, showed that elimination rates were 1.2- to 2.7-fold lower than predicted by the Trudel–Rasmussen model (Van Walleghem et al. 2013), which was developed from estimates of methyl Hg elimination rates for fish from the literature published up to the mid-1990s, including rainbow trout, but not lake charr. These more recent experimental data showed that Hg elimination rate varied substantially among species and for adult lake charr was likely much lower than predicted. Further refinement of Hg mass-balance models for fish should account for species-specific elimination rates (Madenjian et al. 2016).

Male lake charr were 8% higher in Hg concentration and 22% higher in PCB concentration than females from Lake Ontario (Madenjian et al. 2010, 2011), because male lake charr ingest Hg at a higher rate than females, but also eliminate Hg at a higher rate than females (Madenjian et al. 2016). Sexual differences in gross growth efficiency (GGE; amount of growth divided by the amount of food eaten) do not explain sexual difference in Hg concentrations because Wisconsin bioenergetics modeling (Stewart et al. 1983) revealed that GGEs did not appreciably differ between sexes of lake charr (Madenjian et al. 2010). The bioenergetics model did not contain sex-specific parameters for standard metabolic rate (SMR) or swimming activity, but higher PCB concentration in male lake charr was likely due to a higher rate of energy expenditure stemming from a higher SMR and swimming activity, which led to a higher rate of food consumption and a higher rate of PCB accumulation (Madenjian et al. 2010). A higher male-to-female ratio of PCB concentration in lake charr in Lake Ontario (1.22) than Hg concentration (1.08) was likely because males eliminated Hg at a greater rate than females, whereas long-term elimination of

PCBs is negligible for both sexes. This higher level of accumulation in males has now been documented in eight species of fish (Madenjian et al. 2016).

2.2.2 Trophic Magnification Studies of Contaminants in Lake Charr Food Webs

In the western Lake Superior food web, PCBs, and DDT biomagnified through zooplankton, *Mysis diluviana* (formerly *M. relicta*), forage fishes (e.g., sculpins *Myoxocephalus thompsonii* and *Cottus cognatus;* rainbow smelt *Osmerus mordax*), and lake charr, although trophic magnification factors (TMFs) were not estimated (Veith et al. 1977). An early attempt to model biomagnification of contaminants in a food web with lake charr as the top predator was based on allometric relationships between body size and trophic level (Borgmann and Whittle 1983). Discovery of the fractionation of heavy isotopes of nitrogen (^{15}N) and carbon (^{13}C) relative to their light isotopes, and the stepwise increase of $\delta^{15}N$ from one trophic level to the next (Peterson and Fry 1987), led to their use in describing contaminant bioaccumulation in lake charr food webs. For example, concentrations of $\delta^{15}N$ and Hg were significantly correlated in lake charr from Canadian Shield lakes (Cabana and Rasmussen 1994) and concentrations of DDT, toxaphene, and hexachlorocyclohexanes (HCH) were significantly correlated with $\delta^{15}N$ in biota from Lake Laberge, Yukon (Kidd et al. 1995). Similarly, $\delta^{15}N$ was strongly correlated with wet-weight concentrations of p,p′-DDE, mirex, and PCBs, and slopes of relationships for individual compounds provided ecological measures of biomagnification in the Lake Ontario food web (Kiriluk et al. 1995). Lack of correlation between $\delta^{15}N$, fish age, or size and the organochlorine concentrations in lake charr indicated that N isotope fractionation was independent of metabolic factors and could be a valid indicator of trophic relationships (Kiriluk et al. 1995). The method used in the above-cited work was later refined by calculating integer-based trophic level (TL) or trophic position (TP) from $\delta^{15}N$ using trophic enrichment factors (fractionation of ^{15}N from prey to predator, called $\Delta^{15}N$; Fisk et al. (2001)) where primary producers and primary consumers included in the calculations occupied discrete trophic levels of 1 and 2, respectively:

$$TL_{\text{consumer}} = \left(\delta^{15}N_{\text{consumer}} - \frac{\delta^{15}N_{\text{primary producer}}}{\Delta^{15}N}\right) + 1$$

The slope of the relationship between Log[Contaminant] and TL represented the average change in contaminant concentration per trophic level (rather than per $\delta^{15}N$) and is analogous to the average biomagnification of a contaminant through the food web (Borgå et al. 2012; Jardine et al. 2006), where TMF = antilog [slope contaminants vs TL]. In a review of literature up to 2013 on TMFs for organic contaminants, 13 of 69 studies were of lake charr food webs, with a wide range of TMFs for eight classes of brominated and chlorinated organics reported (Fig. 6; Walters et al. 2016).

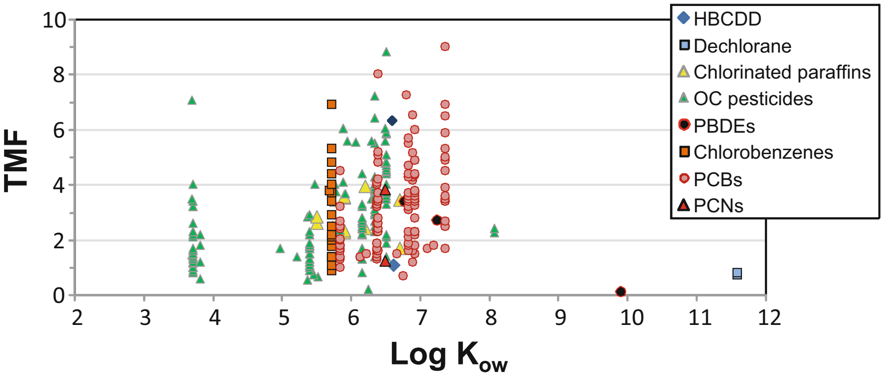

Fig. 6 Trophic magnification factors (TMF) versus log octanol–water partition coefficient (K_{ow}) of brominated and chlorinated organic contaminants in lake charr *Salvelinus namaycush* food webs. Data are from the review by Walters et al. (2016)

For example, HCH isomers (log K_{ow} 3.7) had TMFs from 0.75 to 7 while hexachlorobiphenyls (log K_{ow} 6.8–6.9) had TMFs from 1 to 7 (Fig. 6; Walters et al. 2016). In one of the largest studies of TMFs in lake charr food webs (17 lakes in Canada and the northeastern United States sampled between 1998 and 2001), average TMFs for 14 individual PCBs or organochlorine pesticides (OCPs) were significantly correlated with log K_{ow} (Houde et al. 2008). TMFs of PCBs, hexachlorobenzene, α-HCH, and lindane were weakly correlated with lake area, latitude, and longitude, which suggested that TMFs for PCBs and OC pesticides were only weakly influenced by lake location. Total phosphorus (unfiltered) was correlated to TMFs of PCBs, HCB, and α-HCH, and phytoplankton composition was correlated to HCB, and all parameters were also correlated with latitude and longitude, which confounded the influence of water chemistry (Houde et al. 2008). In summary, a combination of food-web characteristics, such as number of higher and lower trophic level taxa sampled, lake to lake variation in trophic enrichment factors for $\delta^{15}N$, combined with differences in primary productivity and water chemistry may play a role in the wide range of TMFs of chlorinated organics in lake charr food webs (Walters et al. 2016).

Food-chain length affects contaminant concentrations in lake charr. For example, lake charr from Ontario lakes with the shortest food chains (Class 1, $n = 23$, lake charr with small zooplankton, but lacking Mysis and pelagic forage fishes) had the lowest PCB levels and those from lakes with the longest food chains (Class 3; $n = 32$, lake charr with both Mysis and forage fishes, such as smelt and coregonines) had the highest PCB levels (Class 2 lakes lacked only Mysis; Rasmussen et al. 1990). Multiple linear regression showed that PCB levels in lake charr were best predicted by a model including lake class and % lipid as independent variables (Rasmussen et al. 1990). Similarly, PCB and DDT concentrations were each proportional to lipid content in all three lake classes for inland lakes and the Great Lakes, and slopes of relationships of concentrations versus lipid were the same

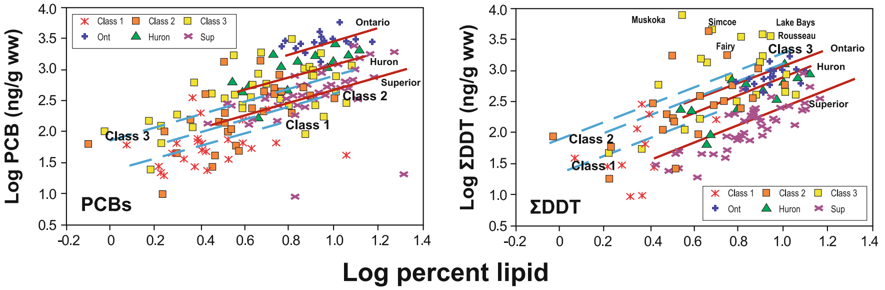

Fig. 7 Relationship between PCBs and total (Σ) DDTs versus percent lipid (all log10 transformed) for PCBs in lake charr *Salvelinus namaycush* from 87 lakes in Ontario (redrawn from (Bentzen et al. 1996). Results for analysis of covariance fit of % lipid, PCB, and ΣDDT are overlaid on lake averages. For the ΣDDT results, lakes that are identified were excluded from the ANCOVA because of possible previous DDT use in their watersheds. Red lines are results for the Great Lakes and blue-dash lines are for the Class 1, 2, or 3 inland lakes

across all lake classes, while intercepts generally differed (Fig. 7; Bentzen et al. 1996).

On the basis of five studies from 24 Canadian lake charr lakes, the slope of log Hg concentration versus δ^{15}N (trophic magnification) TMS exceeded 0, thereby indicating Hg biomagnification in lake charr food webs (Lavoie et al. 2013). The largest dataset was from a study (Kidd et al. 2012) in which Hg TMS in 14 lake charr lakes in Ontario, northern Saskatchewan, and Alberta were best predicted by log lake surface area (positive) and log total phosphorus concentration (positive), which indicated greater Hg biomagnification occurs in larger, more productive systems (Lavoie et al. 2013). However, Hg concentrations in lake charr (size adjusted) that were not correlated with TMS indicated that Hg in lake charr was not directly predicted by the rate of biomagnification through the food web.

In a study of Hg concentrations in lake charr muscle from 96 Ontario lakes, average Hg concentration increased with pelagic food chain length, and ranged 0.03–0.64 μg/g wet wt (ww) among Class-1 lakes, 0.04–1.53 μg/g among Class-2 lakes, and 0.23–3.94 μg/g among Class 3 lakes (Cabana et al. 1994). Lake class and fish weight (wt) both predicted Hg concentration (log Hg (μg/g ww) = −2.72 + 0.21 ± 0.04 (class 1, 2, or 3) + 0.61 ± 0.09 log wt; $r^2 = 0.56$; Cabana et al. 1994). Mean PCBs and Hg in lake charr from the same lake were significantly correlated (Cabana et al. 1994), which illustrates that presence/absence of pelagic forage fishes and Mysis explained elevated concentrations of substances with biomagnification potential in lake charr.

3 Geospatial Trends of Contaminants in Lake Charr

3.1 Mercury

Trends of contaminants in the same species of fish across large numbers of lakes have proven valuable for understanding sources (e.g., atmospheric deposition versus local emissions), biogeochemistry (e.g., mercury methylation), and food-web biomagnification of Hg. For example, the Canadian Fish Mercury Database (Depew et al. 2013; Canadian Mercury Science Assessment, ECCC 2016; http://www.smu.ca/research/fish-mercury-datalayer.html) assessed Hg in fish across Canada based on data during 1967–2010. Lake charr represented 5.6% ($n = 21{,}865$) of individual results in the database and had the highest average concentrations of Hg of all fish species. Highest concentrations were generally found in eastern and northern ecozones (http://ecozones.ca/english/introduction.html), while lowest concentrations were found in Great Lakes lake charr (McGoldrick and Murphy 2016; Fig. 8; Table 11). For example, Hg concentrations averaged 0.80 μg/g ww ($n = 2025$) in the eastern Taiga Shield (northern Quebec) and 0.81 μg/g ww ($n = 473$) in the eastern Boreal Shield (central Quebec). By contrast, total Hg averaged 0.124 μg/g ww ($n = 358$) in Lake Ontario during 2008–2012 (McGoldrick and Murphy 2016). Concentrations depicted in Fig. 8 were not corrected for fish size or food chain length, but mean lengths did not vary widely (range = 44–64 cm; ECCC 2016).

A large study involving composite samples of fish muscle (predators) and whole fish (benthic foragers) from 500 lakes (not including the Great Lakes) selected randomly across the lower 48 states during 2000–2003 included Hg and

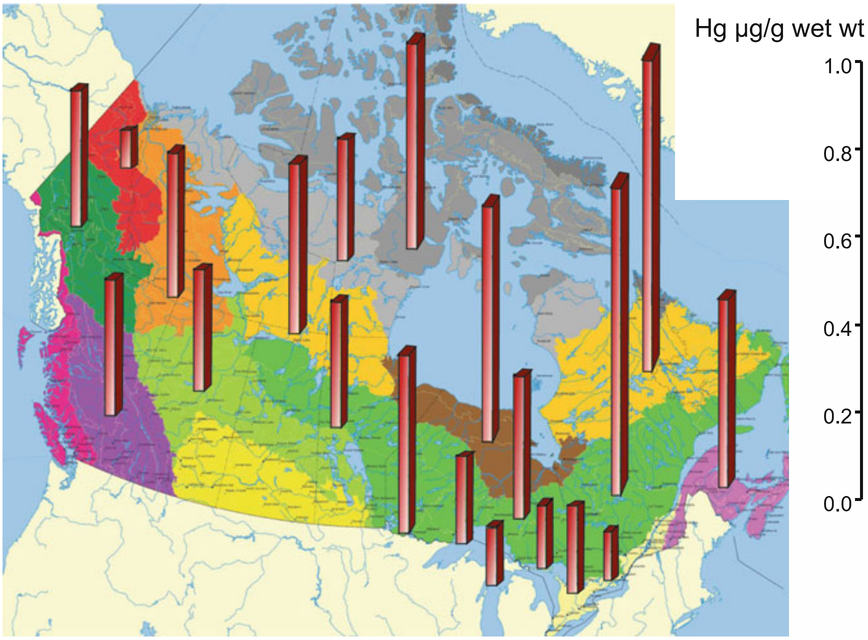

Fig. 8 Total Hg in lake charr *Salvelinus namaycush* muscle grouped by terrestrial ecozone. Bars represent arithmetic means for sample numbers ranging from 5 fish (Taiga Cordillera, northern Yukon) to 6996 (Southern Boreal Shield, central Ontario). Results are from the Mercury Science Assessment Table 10.11 in Chapter 10 (ECCC 2016)

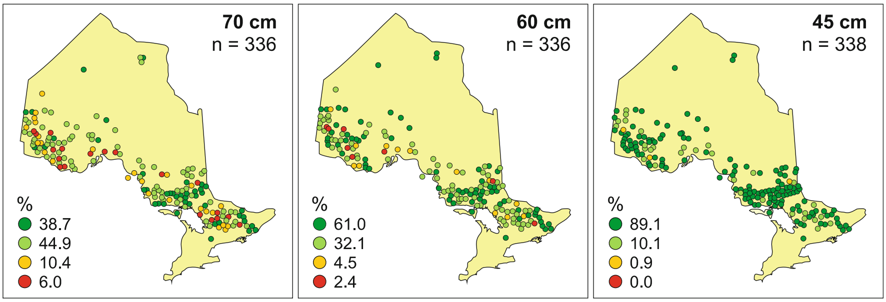

Fig. 9 Mercury concentrations (μg/g wet weight) in skin-off fillets of small, medium, and large-sized lake charr *Salvelinus namaycush* from Ontario lakes collected between 2000 and 2012. Reproduced from Gandhi et al. (2014b) with permission

268 individual chemicals including 209 polychlorinated biphenyl (PCB) congeners (yielding 159 measurements based on co-eluting congeners), five arsenic species, PCDD/Fs, 46 OCPs, and organophosphate (OP) pesticides, and 40 semivolatile organic compounds (mainly polycyclic aromatic compounds) (Stahl et al. 2009; U.S. EPA 2009; https://www.epa.gov/fish-tech/national-lake-fish-tissue-study). Mean Hg concentrations in composite muscle samples of lake charr from 10 lakes in the northern United States ranged 0.012–0.59 μg/g ww, at the low end of the range for lake charr in Canadian lakes, and was highest in Torch Lake, northern Michigan (Stahl et al. 2009).

Lake charr, mainly from Alaska and Canada, were among the fish species with highest Hg concentrations, ranking 7th out of 208 species in a recent survey with a size-adjusted average of 0.27 (0.010–12.3) μg/g ww Hg (285 sites, $n = 4383$) (Eagles-Smith et al. 2016). Fish Hg concentrations (but not specifically lake charr) were weakly correlated with sediment methyl Hg concentrations, which led to the suggestion that factors influencing methyl Hg production may be more important than inorganic Hg loading for determining fish Hg levels. As discussed previously, Hg in lake charr was not directly predicted by the rate of biomagnification (Kidd et al. 2012), while lake class (a measure of food chain length) and fish weight predicted Hg concentrations in lake charr (Cabana et al. 1994). Spatial trends of Hg of three standard fish lengths (45, 60, and 70 cm) for lake charr from 338 lakes in Ontario during 2000–2012 did not differ significantly between northern ($n = 243$) and southern Ontario ($n = 92$-94) locations (Fig. 9; Gandhi et al. 2014b).

Although all studies of spatial trends of Hg in fish reviewed herein were of large geographic scope (western North America, Canadian shield, northern and southern Ontario) and showed substantial variation among lakes, no latitudinal or longitudinal trends were evident for Hg in lake charr. The relatively similar gaseous elemental Hg and total Hg in precipitation over much of Canada (Cole et al. 2014; ECCC 2016) implies similar atmospheric inputs to most lake charr lakes in north-temperate North America. Exceptions would be the lower Great Lakes and some lakes near sources such as smelters located in regions where higher localized atmospheric deposition and/or urban sources have been predicted or observed (ECCC 2016). In a study to assess Hg sources in Great Lakes sediments using Hg isotope ratios included lake charr from Lakes Ontario and Superior, bioaccumulated Hg was isotopically more like atmospherically derived Hg than to the isotope signature in lake sediment, based on mass-independent fractionation (MIF) of ^{200}Hg relative to the major isotope ^{202}Hg (Lepak et al. 2015). Earlier work that showed a lack of MIF for Hg during dietary exposure of lake charr suggested that Hg isotope measurements can provide direct linkages between Hg sources and Hg in fish tissues (Kwon et al. 2012).

Lack of latitudinal trends in lake charr Hg is likely due to the strong influence of food-chain length and complexity (Cabana et al. 1994). For example, invasive species such as non-native smallmouth bass, rainbow smelt, and spiny water flea *Bythotrephes longimanus* have influenced fish diversity and prey abundance for lake charr (Van der Zanden et al. 2004). This food-chain factor may be more influential in lakes nearer population centers that have greater fishing and recreational uses. These factors are further discussed below in relation to temporal trends of Hg in lake charr.

3.2 Legacy POPs

Unlike Hg, lake charr mean PCB concentration declined with latitude (i.e., from south to north) due to higher PCBs in the Laurentian Great Lakes, Michigan, Huron, and Ontario (Rasmussen et al. 1990; Bentzen et al. 1996; $n = 83$ and 87 lakes, respectively). While the main focus of these studies was to understand the effect of food-chain length and percent lipid on PCB concentration in fish, these studies also provided insight into levels of PCBs in remote lakes in northern Ontario and near urban regions (Fig. 10; Bentzen et al. 1996). The model for predicting PCBs in lake charr and other pelagic fishes included lake class, latitude, and % lipid (Rasmussen et al. 1990):

$$\mathrm{Log_{10}PCB\ (ng/g\ wet)} = 1.63 + 0.40(\pm 0.05) \times \mathrm{Lake\ class} - 0.14(\pm 0.02) \times \mathrm{Latitude}\ (^{\circ}\mathrm{N\ of\ 42}) + 0.73(\pm 0.14) \times \mathrm{log\%lipid}\ (r^2 = 0.74; n = 97)$$

The model illustrates a significant negative relationship of declining concentrations of PCBs with increasing latitude in Ontario.

Additional evidence for effects of lipid and lake characteristics on PCBs and OCPs in lake charr comes from a comparison of PCBs and OCPs in lake charr from 14 lakes in British Columbia, Alberta, and Saskatchewan (Donald et al. 1993), with selected data from 14 lakes (Rasmussen et al. 1990). They evaluated food-web length of each lake by identifying key species in lake charr diets and found PCB concentration was positively related to lake area, but not to percent lipid content or lake trophic structure (Fig. 11). Larger catchments received greater loading of PCB and thereby greater concentrations in lake charr. Omitting the Great Lakes (Superior, Huron, and Ontario) results from the model did not change the findings.

Spatial trends of PCBs in lake charr from 23 lakes in Canada and the northeastern United States sampled during 1998–2001were inferred from stable carbon isotope ratios (δ^{13}C), an indicator of littoral feeding because lipid-corrected δ^{13}C was

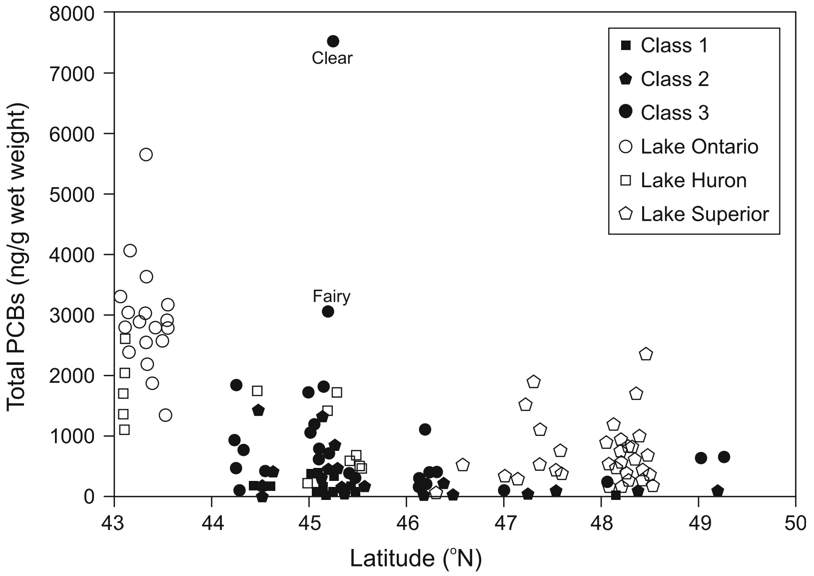

Fig. 10 Lake-averaged PCB concentrations in lake charr *Salvelinus namaycush* muscle versus latitude for the three classes of inland lakes ($N = 87$) and three Great Lakes from Ontario Ministry of Environment data for the late 1980s (Crawford and Brunato 1978). Clear Lake is identified as an outlier due to known PCB contamination reported by Macdonald et al. (1992). Reproduced from Bentzen et al. (1996)

negatively correlated to total (Σ) PCB concentrations (sum of 57 congeners), which supports the hypothesis that increasing access to littoral habitat results in lower concentrations of POPs in lake charr (Guildford et al. 2008). They also found a negative relationship of ΣPCBs with latitude and a positive relationship to lake area:

$$\log_{10}\sum \text{PCB} = 6.222 + 0.158\log_{10}\text{Area} - 0.073 \times \text{latitude}\ (r^2 = 0.733)$$

Lake area may serve as a surrogate for littoral habitat because the proportion of deep water increases and the proportion of shallow water decreases with lake size (Guildford et al. 2008).

Geospatial trends of organic contaminants in lake charr are shown in Fig. 12, which combines lipid normalized results for Σ_{10}PCBs and ΣDDT from samples collected in 2010 from Lakes Kusawa and Laberge, Yukon (Ryan et al. 2013) and in Great Slave Lake, Northwest Territories (Evans et al. 2012; Muir et al. 2013), and from the Great Lakes (Chang et al. 2012). Σ_{10}PCBs (sum of congeners 28, 31, 52, 99, 105, 118, 138, 153,156, 180) were used to enable comparison with the more limited suite of congeners measured in lake charr from Alaskan lakes and the lower 48 states. In Alaska National Parks during 2004–2011 (Ackerman et al. 2008; Flanagan Pritz et al. 2014), concentrations of Σ_7PCBs (sum of 7 congeners), and

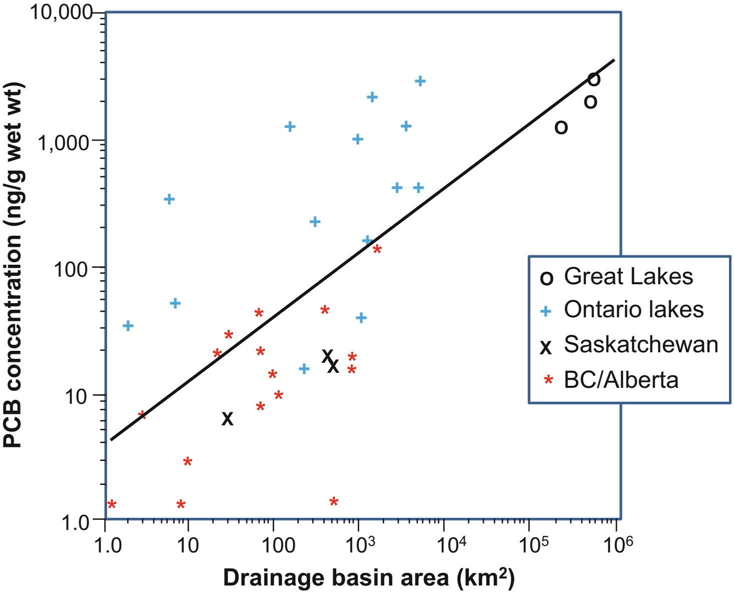

Fig. 11 Relationship between PCB (ng/g wet wt) in lake charr *Salvelinus namaycush* fillets and lake drainage basin area (km^2) for lakes in British Columbia, Alberta, Saskatchewan, and Ontario (Donald et al. 1993)

ΣDDT were highest in Kijik Lake, a small, remote lake in Lake Clarke National Park, Alaska (Fig. 12). Concentrations in Kijik lake charr were higher than in most other remote lake locations, but similar to Lake Laberge, where elevated concentrations of many organochlorine contaminants, especially PCBs and toxaphene, have been attributed mainly to food chain length (Kidd et al. 1998; Ryan et al. 2013). Three- to fourfold higher concentrations of Σ_{10}PCBs and ΣDDT in lake charr from the east arm than from the west basin of Great Slave Lake (Fig. 12) illustrate the effect of food chain and water chemistry on PCB concentration in fishes. Low productivity in the east arm may provide less opportunity for contaminant dilution through fish growth while high suspended particle loadings in the west basin may limit bioavailability (Evans and Muir 2016). Legacy sources from historical use of DDT, which was widely used for biting fly control in the 1950s and 1960s, may explain higher ΣDDT/PCB ratios in Laberge, Brooks, Kekekabic, Upper Two Medicine, and Lewis Lakes than in Kusawa and Great Slave Lakes, although this is not well documented (Fig. 12). Σ_{10}PCBs and ΣDDT concentrations in lake charr (muscle) from six US lakes west of the Great Lakes region were generally similar to those in Alaskan lakes while PCBs were higher in three of the four eastern lakes (Fig. 12). Highest PCBs in lake charr were in Torch Lake (near Rapid City, Michigan), which was designated as an Area of Concern under the 1987 Great Lakes Water Quality Agreement and has three small communities in its catchment

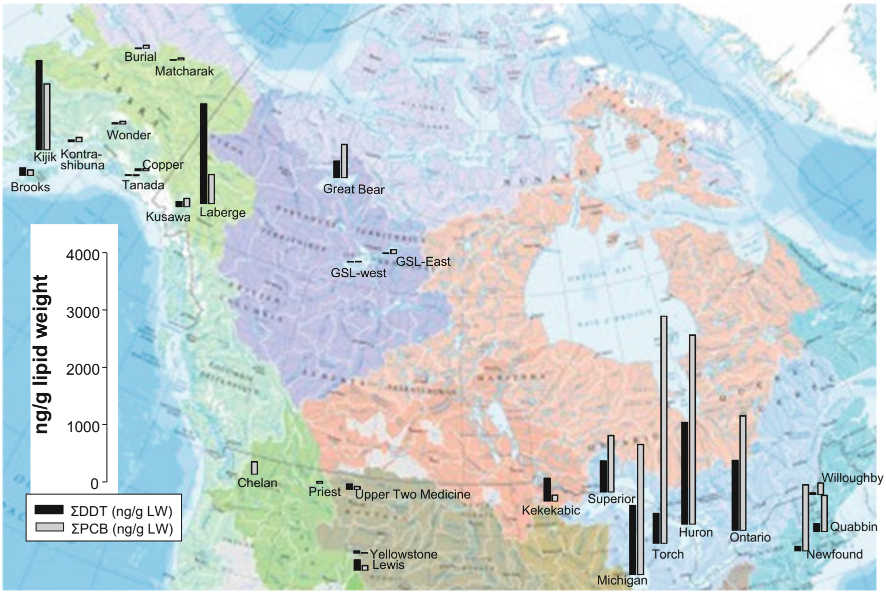

Fig. 12 PCBs (sum of 7–10 congeners) and ΣDDT (6 isomers) in lake charr *Salvelinus namaycush* muscle (or whole fish Great Lakes only). Results (ng/g lipid weight) are from samples collected between 2004 and 2011 in Alaska, 2007 in Great Bear Lake, 2010 in Great Slave Lake, Kusawa Lake and Lake Laberge, and 2009 in four Great Lakes and by the U.S. EPA National Fish Contaminants Study (2000–2003) (U.S. EPA 2009)

and legacy sources of pollution related to mining and industrial operations (U.S. EPA 2018).

Concentrations of legacy contaminants, such as PCBs and OCPs, and more recently measured organic contaminants, in Great Lakes lake charr, ranked by relative concentration in Lake Ontario, were relatively consistent among lakes during 2008–2012 (Fig. 13; McGoldrick and Murphy 2016). First, concentrations of major organic contaminants and Hg in whole fish homogenates indicate that PCBs remain the predominant contaminant in all five lakes despite bans and phaseouts in the late 1970s and 1980s in the USA and Canada (CEC 1996). Next, the combined sum of tetra-, penta-, hexa-, and heptachlorobiphenyls greatly exceed levels of other contaminants (Fig. 13). Next, legacy OCPs, banned in the 1970s and 1980s (ΣDDT, toxaphene, dieldrin, and chlordane-related compounds) are also prominent contaminants, particularly in Lakes Michigan and Superior (Fig. 13). Last, mirex remains a major OCP in lake charr from Lake Ontario, but not in other lakes (Fig. 13).

3.3 *New Organic Contaminants*

Several chemical substances have only been measured since the mid-1990s, including PBDEs, short-chain chlorinated paraffins (SCCPs), nonylphenol-related

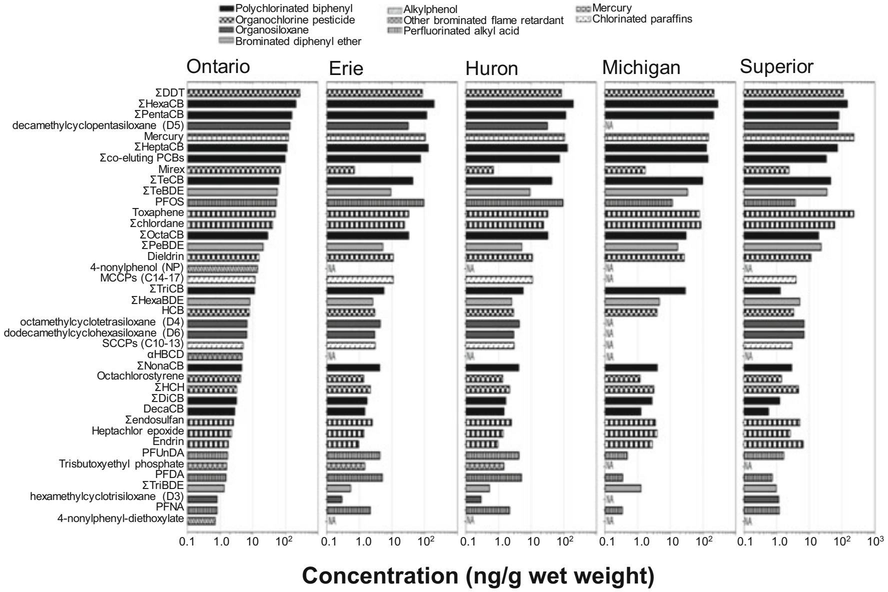

Fig. 13 Mean concentrations of the top 40 compounds (by mass) measured in whole-body homogenates of lake charr *Salvelinus namaycush* from five Great Lakes. Redrawn from McGoldrick and Murphy (2016)

compounds, cyclic methyl siloxanes (D4, D5, D6), medium-chain chlorinated paraffins (MCCPs), perfluorooctanesulfonate (PFOS) perfluorocarboxylates (PFCAs; PFNA, PFDA, PFUnA), and tris(2-butoxyethyl) phosphate (TBOEP; McGoldrick and Murphy 2016). These substances are high production-volume industrial chemicals or persistent transformation products in the case of PFOS and PFCAs that have been in commercial use for the past 30 years or more based on their listing in the chemical inventories of the U.S. EPA and Environment Canada (Howard and Muir 2010). Thus, none are new to commerce but their analysis in lake charr reflects advances in analytical methodology, including availability of appropriate analytical standards, particularly for PFOS and PFCAs and the cyclic siloxanes.

The combined concentrations of cyclic siloxanes D4, D5, D6 places them among the most prominent organic contaminants in lake charr in the Great Lakes (Fig. 13). In lake charr from Lakes Superior, Erie, Huron, Ontario, Athabasca (northern Alberta), and Kusawa (Yukon), cyclic siloxanes were detectable in all lakes, but lowest in Athabasca and Kusawa, the two most northerly lakes (McGoldrick et al. 2014). D5 was the most prominent siloxane with the highest concentration in lake charr collected near the Niagara River outflow into Lake Ontario (45–719 ng/g ww). Higher concentrations of cyclic siloxanes in lake charr from Lakes Ontario, Erie, and Huron were consistent with wastewater effluents and atmospheric emissions from urban areas being the main sources for these compounds (Xu et al. 2014).

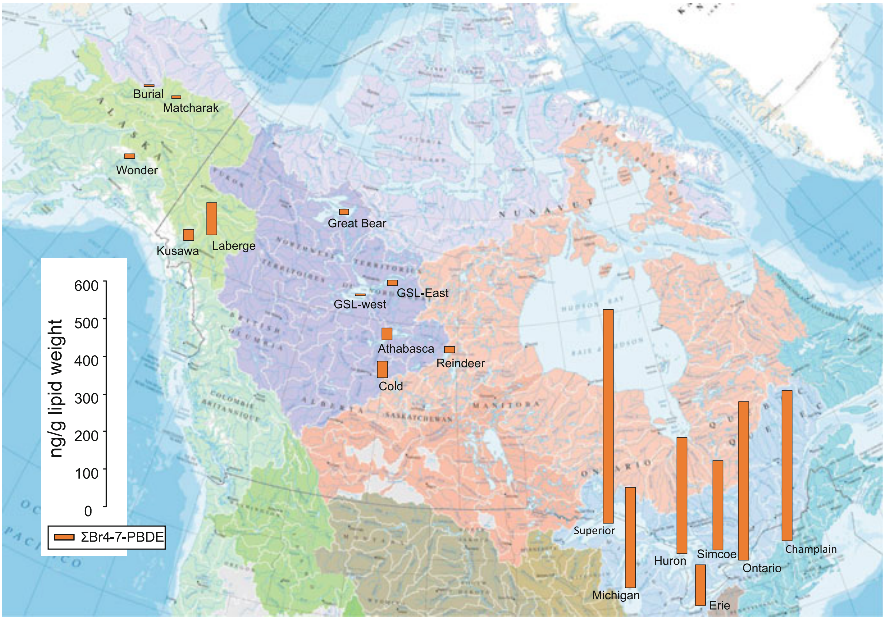

Fig. 14 PBDEs (sum of tetra to heptabromo congeners) in lake charr *Salvelinus namaycush* muscle (Alaska, Yukon, and Northwest Territories) or whole fish (all other sites). Results (ng/g lipid weight) are from samples collected between 2004 and 2011 in Alaska (Ackerman et al. 2008), 2007 Great Bear Lake (Evans and Muir, unpublished data), 2010–2011 in Great Slave Lake, Kusawa Lake, and Lake Laberge (Evans et al. 2012; Muir et al. 2013), and 2009 in four Great Lakes. Data sources are Gewurtz et al. (2011d)

PBDEs are another prominent group of organohalogen compounds in lake charr in the Great Lakes. The combined total for tetra-, penta-, and hexabromo BDEs brings ΣPBDEs to similar levels as siloxanes and PFASs (Fig. 13). Relatively elevated ΣPBDE concentrations in Lake Simcoe and Lake Champlain likely reflect emissions from urban centers on these lakes (Fig. 14). Much lower concentrations of ΣPBDEs in lake charr from Alaskan and northwestern Canadian lakes reflect mainly atmospheric deposition pathways to these lakes (Fig. 14). As with DDT, Lake Laberge stands out as having somewhat higher ΣPBDE than other lakes in the region, possibly reflecting food chain length, although wastewater sources from the city of Whitehorse could also be important.

Concentrations of PFASs in lake charr have a similar geospatial pattern as PBDEs, with highest concentrations in the Great Lakes and nearby lakes with smaller urban centers releasing municipal wastewaters (Peninsula, Champlain) (Fig. 15). In a cross-Canada survey, geometric mean PFOS and ΣPFCAs concentrations were highest in lake charr from Lakes Erie (90 ng/g ww) and Ontario (62 ng/g ww; Gewurtz et al. (2013). In a more detailed analysis of PFAS in Lake Ontario lake charr, PFASs were not related to possible fish covariates, % lipid, length, weight, age, growth rate, $\delta^{15}N$, and $\delta^{13}C$, when using all data from 1997 to 2008 (Gewurtz et al. 2012). In contrast to results for lipophilic compounds such as PCBs,

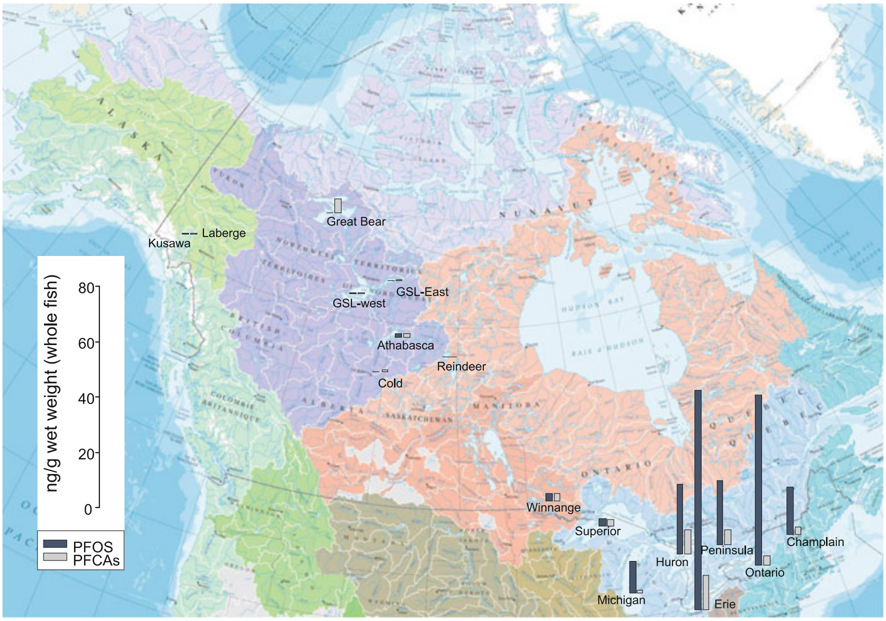

Fig. 15 Concentrations of PFOS and ΣPFCAs (C9–C12) in whole lake charr *Salvelinus namaycush* from the Great Lakes (McGoldrick and Murphy 2016) and lakes across Canada (Gewurtz et al. 2013)

lack of fish covariates may reflect the association of PFASs with proteinaceous tissue and also that uptake from water could be more important for these water-soluble contaminants found at relatively high levels in Lake Ontario (Gewurtz et al. 2013).

Concentrations of SCCPs and MCCPs in lake charr from large lakes in Canada (Kusawa, Athabasca, Superior, Huron, Erie, Ontario), as expected, were higher for MCCPs in the Great Lakes, with highest levels in Lake Huron, but unexpectedly higher for SCCPs from Lake Athabasca and Kusawa Lake, a remote lake in the Yukon (Saborido Basconcillo et al. 2015). Taken together, these results support the hypothesis that atmospheric sources are important for SCCPs in northern large lakes and the upper Great Lakes, while wastewater emissions are the predominant source of MCCPs in the lower Great Lakes.

Among organophosphate ester flame retardant/plasticizers (OPEs) surveyed from the Great Lakes and several large lakes across northwestern Canada, TBOEP and tris (chloroethyl) phosphate (TCEP) were highest in concentration in lake charr (McGoldrick et al. 2014). However, TCEP was only detected in several northwestern lakes (Athabasca, Cold, Kusawa, Great Bear) but not in lake charr from the Great Lakes (McGoldrick et al. 2014). TBOEP was detected in lake charr in all lakes and was the most prominent OPE in Great Lakes waters (Venier et al. 2014). Concentrations of TCEP (geomeans of 6.5–12.8 ng/g lw) were low in Great Lakes lake charr and TCEP and triphenyl phosphate (TPHP) were the most prominent OPEs in Great Lakes lake charr, although TBOEP was not determined (Guo et al. 2017).

4 Temporal Trends

Lake charr were used beginning in the 1970s for monitoring contaminants in fish, particularly in the Great Lakes (Gewurtz et al. 2011a) and since then robust temporal trend studies have built on this early work in programs operated by the U.S. EPA (Great Lakes National Program Office), Environment and Climate Change Canada's Water Quality Monitoring and Surveillance Division (and previously Canada's Department of Fisheries and Ocean (DFO), Great Lakes Laboratory for Fisheries and Aquatic Sciences), and by the Ontario Ministry of the Environment, Conservation and Parks (ECCC and U.S. EPA 2017). Similarly, studies of contaminants in lake charr from the Great Slave Lake, Kusawa Lake, and Lake Laberge began in the 1990s under the Northern Contaminants Program (NCP) of Indigenous and Northern Affairs Canada (NCP 2018), and built on earlier measurements of Hg by DFO's Fish Inspection Division (Lockhart et al. 2005). The time series with greatest statistical power to detect a trend in sample measurements when a trend is occurring, despite "noise" in the data, are from annual sampling (Table 4). Using data for PCBs in lake charr from Lake Ontario, for example, 10–15 years of sampling with 10–15 samples per year are needed to detect a 5% decrease in total PCB concentrations with a 5% significance level (Gewurtz et al. 2011a). This level of sampling had only been achieved for PCBs, OCPs, and Hg in lake charr from Lakes Michigan, Superior, Huron, Erie, and Ontario by the three major monitoring programs. Power analysis on lake charr contaminant data from Great Slave Lake, Kusawa, and Laberge (Macdonald 2014) indicates that these shorter time series had not achieved the NCP objective of 80% power based on 11 to 14 sampling years from the 1990s to 2012, although continued annual sampling would likely achieve it soon.

Lake charr contaminant programs in the Great Lakes have received continued support from environmental agencies because they produced valuable results in support of chemical management and assessment commitments under Canada's Chemicals Management Plan (CMP) and the Great Lakes Water Quality Agreement (GLWQA) related to reducing or eliminating releases of anthropogenic chemicals into waters of the Great Lakes (GLWQA [Great Lakes Water Quality Agreement] 1978). Trends of major contaminants reported in State of the Great Lakes reports (ECCC and U.S. EPA 2017) complement other long-term trend data, such as herring gull egg monitoring and atmospheric measurements (ECCC and U.S. EPA 2017). Contaminant trend data also provide an indicator of aquatic ecosystem health and data for estimating human and fish-eating wildlife exposure to contaminants. These commitments were reaffirmed under Annex 3 (Chemicals of Mutual Concern) of the 2012 protocol amending the GLWQA (GLWQA [Great Lakes Water Quality Agreement] 2012). In 2009, the U.S. EPA eliminated analysis of POPs in sport fish and expanded effort to identify emerging contaminants in whole fish. The program name was changed from the Great Lakes Fish Monitoring Program to the Great Lakes Fish Monitoring and Surveillance Program (ECCC and U.S. EPA 2017).

Table 4 Long-term temporal trend programs for contaminants in lake charr *Salvelinus namaycush*

Program	Water bodies	Frequency	Sample type	Contaminants
Ontario Ministry of the Environment, Conservation and Parks	Lakes Superior, Huron, Ontario, and Erie	Generally on an annual or biennial basis from 1970	Skinless dorsal muscle. Individual fish 60 and 70 cm	Total Hg, 12 metals, PCBs, OCPs, selected new organics
Environment Canada, WQMSD—Great Lakes	Lakes Superior, Huron, Ontario, and Erie	Annual collections since 1977; 2–4 offshore sites per lake	Whole fish (Aged 4–6); individual fish homogenates.	Total Hg, PCBs, OCPs, PBDEs, PFAS, other selected priority organics, and metals
Environment Canada, WQMSD—Chemicals Management Plan	Lakes Kusawa, Great Bear, Athabasca, Reindeer, Cold	Annual collections since 2009	Whole fish; individual fish homogenates	Total Hg, PBDEs, PFAS, non-BDE flame retardants, other selected priority organics, and metals
U.S. EPA GLNPO (Great Lakes Fish Monitoring Program)	Lakes Superior, Michigan, Huron, Erie, and Ontario	Annual collections in US waters from early 1970s; 2 sites per lake on alternate years	Whole fish, (600–700 mm length). 10 composite samples of 5 fish	Total Hg, metals, PCBs, OCPs, PBDEs, PFAS, other chemicals of emerging concern
Northern Contaminants (INAC) and Yukon Contaminants Committee	Kusawa and Laberge	Annual sampling since 2000	Skinless dorsal muscle and liver for PFASs, individual fish	Total Hg, Se, PCBs, OCPs, PBDEs, PFAS
Northern Contaminants (INAC) and Environment Canada	Great Slave Lake	Annual sampling since 1998	Skin on muscle, individual fish	Total Hg, 32 multielement suite, PCBs, OCPs, PBDEs, PFAS

Contaminant time-trend studies in northern lakes also contributed to chemical assessment and management, mainly at an international level. Results for PCBs and OCPs were part of data assembled for the Stockholm Convention on POPs (http://www.pops.int/) and also for the Minamata Convention on Mercury (http://www.mercuryconvention.org/). In addition, results for POPs in lake charr from the Lake Laberge were a key factor in the decision to close the fishery in the lake in 1991–1992 to reduce human exposure. Continued monitoring under the NCP has been in support of that early decision (Ryan et al. 2013).

4.1 Mercury Trends in Lake Charr

4.1.1 Overview

Several recent studies have synthesized spatial and temporal trends of Hg in the Canadian environment, with a focus on long-range atmospheric transport sources (Chételat et al. 2015; Depew et al. 2013; Eagles-Smith et al. 2016; ECCC 2016). Temporal trends in Hg concentrations in lake charr are influenced by several variables. Localized anthropogenic perturbations, such as gold mines, can increase Hg concentrations in lake charr and other fish species, which may decline with a reduction in mining (Armstrong and Scott 1979; Moore and Sutherland 1980; Weech et al. 2004). Reservoir creation that floods the landscape and results in substantial methyl Hg production can result in high Hg concentrations in lake charr that gradually subside as organic matter decomposes (Anderson et al. 1995; Jackson 1991). In the Great Lakes region, while industries such as chlor-alkali plants contributed to localized Hg contamination of the environment and fish, most Hg was atmospheric in source (Bhavsar et al. 2010; Hatch et al. 1987). A recent study using stable Hg isotopes linked Hg signatures of lake charr from all five Great Lakes with precipitation sources, and suggested that methylation of mercury deposited into the upper water column was an important source of mercury to lake charr (Lepak et al. 2018). Mercury emissions in North America and Europe declined in recent decades with improvements in coal-fired power-plant technologies and waste management improvements, while emission rates increased from Asia with expansion in various economic sectors, particularly coal-fired power plants (Durnford et al. 2010; Jaffe and Strode 2008). Global warming, with a longer growing season in many regions of Canada, is resulting in increased primary productivity with a concomitant increase in particulate flux, including Hg, to lake sediments, while methyl-Hg production rates may increase with enhanced productivity (Lehnherr et al. 2018; Outridge et al. 2007; Stern et al. 2012). Increasing productivity may enhance fish growth rates to biodilute Hg in lake charr. Alternatively, an increase in forage fish abundance may result in a richer fish diet and an increase in Hg concentrations. Changes in food-web structure as a result of exotic species and increased fish harvesting pressure changes fish age structure and growth rates that also may result in changes in Hg concentrations in fishes (Lavigne et al. 2010; Rasmussen et al. 1990).

4.1.2 Great Lakes

Annual monitoring of mercury in lake charr from Lakes Superior, Huron, Erie, Ontario, and Michigan by provincial, tribal, state, and federal agencies has shown a substantial decline from the 1970s to 2000, with a further decline, albeit less drastic, during 2000–2015 in most lakes (Bhavsar et al. 2010; Dellinger et al. 2014; Visha et al. 2015; Zhou et al. 2017). Concentration of Hg in lake charr declined significantly in Lakes Superior, Huron, Erie, and Ontario during 1973–2007

(Bhavsar et al. 2010). An apparent increase in Hg concentration in Lake Superior lake charr since 2006 was caused by fish being older at a given length than from 1980 through the 1990s, so fish had more time to accumulate Hg. Declines in Hg were related to decreased Hg emissions from incinerators and smelters while Hg from coal-fired power generation remained relatively constant. Continued Hg inputs from the watershed were inferred as the cause of delayed responses to reduced Hg emissions (Bhavsar et al. 2010). Hg in lake charr from Lakes Superior, Huron, and Michigan did not decline during 1992–2011 (6–7 sampling years) (Dellinger et al. 2014). Concentration of Hg in lake charr from Lake Ontario from the late 1970s to 2011 generally declined, but with short-term increases and decreases in the rate of decline when fish length was considered (Visha et al. (2015). Hg inputs to Lakes Superior and Huron were dominated by atmospheric sources and concentrations in lake charr declining at rates of 5.2–7.8% per year during 2004–2015 (Zhou et al. 2017). Mercury concentrations decreased in Lake Michigan (3.9% per year), increased in Lake Erie (1.2% per year), and exhibited no temporal trend in Lake Ontario over the same period (Zhou et al. 2017). While rates of decline were attributed to decreasing regional Hg emissions, other factors, such as eutrophication, warming, increased local Hg emissions, and food-web changes may have altered trends. In lake charr from Canadian waters of the Great lakes (1977–2015 with gaps from the mid-1990s to mid-2000s), mercury declined at annual rates of 3–5% from the mid-80s until mid-1990s (Blukacz-Richards et al. 2017). From the mid-2000s to 2015, Hg increased slightly in Lakes Superior (2–3%), Huron (2%), Erie (1%), and western Lake Ontario (1%). Studies of temporal Hg trends by Blukacz-Richards et al. (2017) and Zhou et al. (2017) included similar periods (mid-2000s to 2015) and results mostly agreed for Lakes Erie and Ontario, but differed for Lakes Superior and Huron. Blukacz-Richards et al. (2017) used a dynamic linear model incorporating length and lipid whereas Zhou et al. (2017) used age normalization, so different approaches may account for different results for temporal trends of Hg in lake charr from Lakes Superior and Huron.

4.1.3 Ontario Inland Lakes

Since the early 1970s, Hg concentrations in lake charr (also walleye and northern pike) have been measured in more than 100 lakes under programs designed to provide fish consumption advice for the Ontario recreational fishery (Gandhi et al. 2014b, 2015; Tang et al. 2013). Overall, Hg concentrations declined in lake charr at more than half of the locations studied during 1970–2012 (Fig. 16). Rates of decline were particularly large during 1970–1990 in southern Ontario for medium (60 cm length) and large (70 cm) lake charr. However, rates of decline were much lower than those reported from the Great Lakes. Moreover, Hg concentrations increased in some lakes, particularly for medium and large lake charr in southern Ontario during 1970–1990 and 1995–2012, with a weak decline during 1985–2006. Overall findings were consistent with generally observed patterns of decreasing Hg concentrations since the 1970s, with increases in recent years. Recent increases in Asian Hg

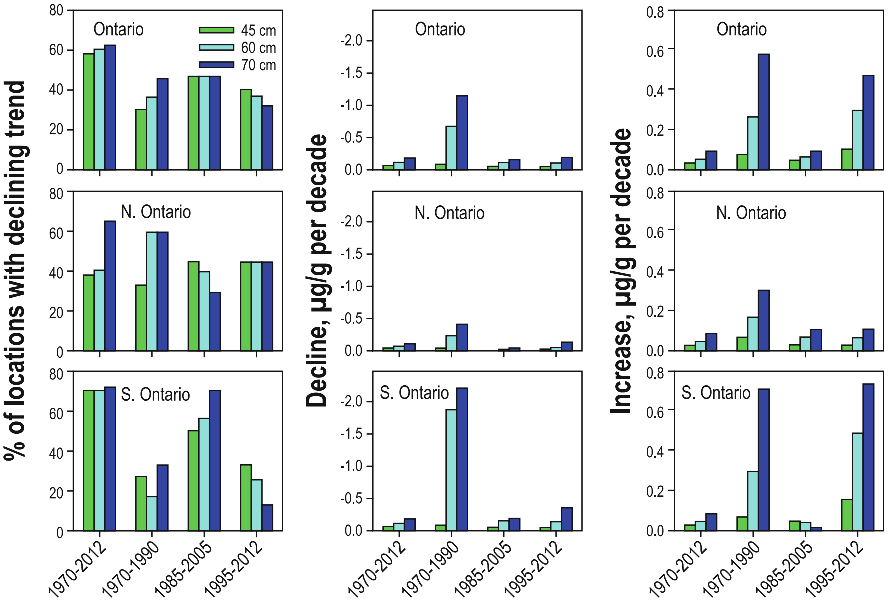

Fig. 16 Spatial and temporal trends in Hg concentrations in lake charr *Salvelinus namaycush* from Ontario lakes. Panels A, B, and C represent % of locations with a declining trend for all of Ontario, northern, and southern Ontario, respectively. To the right of each panel is the decline or increase expressed as µg/g per decade. From Table 1 in Gandhi et al. (2014b)

emissions may have played a role in recent increases in Hg concentrations in Ontario lakes, but warming temperatures may also be important. Few studies reported Hg trends in specific inland lakes, but Hg concentration in lake charr declined significantly in Lake Simcoe, Ontario, between 1970 and 2009 (Gewurtz et al. 2011c).

4.1.4 Other Southern Canada Inland Lakes

Mercury trends have not been monitored in Québec lakes due to the small number of lakes sampled more than three times and the general paucity of lake charr in those lakes, which tend to be walleye and northern pike dominated (Lucotte et al. 2016). Mercury trend studies have not been conducted in Canada's Maritime province lakes (apart from Anderson et al. 1995). Hg concentrations in lake charr measured as part of fish inspection studies showed no temporal trend in Cold Lake, Alberta, and Reindeer Lake, Saskatchewan during 1977–2014 (ECCC 2016). Similarly, Hg concentration showed no detectable trends in lake charr from western Lake Athasbaca during 1978–2009 (Evans and Talbot 2012).

4.1.5 Northern Canada

Mercury concentrations in lake charr have been monitored annually in Great Slave Lake since 1998. More limited data going back to 1977 are available from the west basin commercial fishery and some data from the east arm during the early 1990s (Evans et al. 2013). Mercury increased significantly through time in west basin lake charr and in large (>590 mm) lake charr from the east arm from the early 1990s to 2012, but decreased in small (<590 mm) lake charr from the east arm (Evans et al. 2013) (Fig. 17). Higher Hg concentrations in west basin lake charr were also associated with cooler years and a more positive Pacific North American oscillation index. Cooler years potentially were associated with less productive conditions, slower growth, and a lower condition factor than warmer years. Lake charr Hg concentrations in Lake Laberge and Kusawa Lake were slightly higher in the early 1990s than 2000–2001, particularly in Kusawa lake, although no trends were detected (Chételat et al. 2015).

4.2 Temporal Trends of POPs

4.2.1 Overview

Lake charr is one of the main target organisms for basin-wide assessments of POPs in the Great Lakes due to their availability, large home range, and position as a top

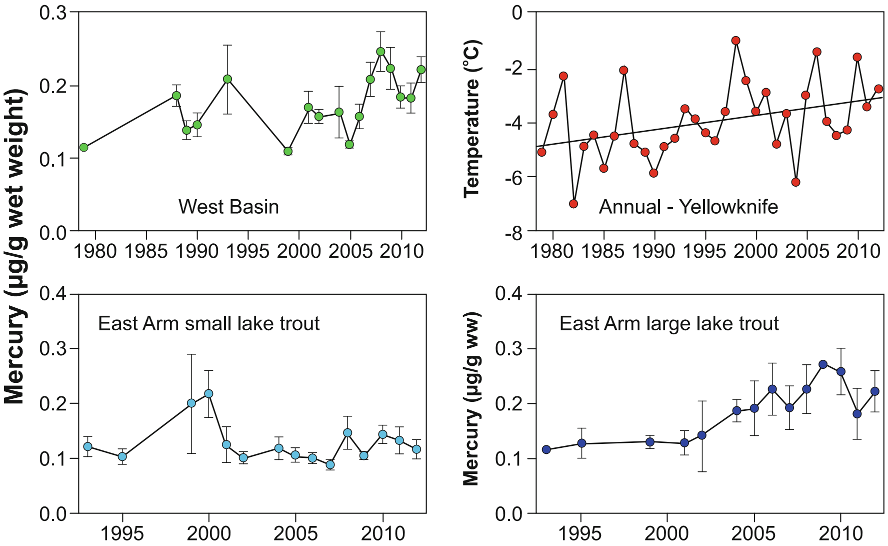

Fig. 17 Trends time in Hg concentrations in lake charr *Salvelinus namaycush* from the west basin and east arm of Great Slave Lake. Also shown is the year-to-year variation in mean annual air temperatures at the city of Yellowknife. Modified from Evans et al. (2013)

predator in the food web (Murphy et al. 2013). Canada and the USA have operated biomonitoring programs to monitor the status and trends of POPs in lake charr tissues from the Great Lakes since the 1970s (Gewurtz et al. 2011a; Murphy et al. 2013). The Province of Ontario and eight Great Lakes states also have long-term monitoring programs related to trends of contaminant concentrations in edible portions of fish (see Section "Human Dietary Exposure and Advisories"). The number of time series of POPs in lake charr is more limited than for Hg due to the high cost of analysis that limits the numbers of samples analyzed and sampling locations.

Since POP monitoring programs began, they have directly supported commitments made in the Great Lakes Water Quality Agreement between Canada and the USA to track progress toward reducing or eliminating releases of organic pollutants in the waters of the Great Lakes (GLWQA [Great Lakes Water Quality Agreement] 1978), and domestic chemical management initiatives. Long-term trend studies of POPs in lake charr began in the Arctic during the early 1990s with studies in Lake Laberge, Yukon, and Great Slave Lake, Northwest Territories (Muir et al. 2013). These studies were initiated because of concerns about human exposure from traditional diets by indigenous peoples.

Long-term monitoring of POPs in lake charr initially focused on PCBs and OCPs in the Great Lakes and Arctic lakes. However, in the early 2000s, attention began to shift toward other classes of POPs that were being reported in environmental media, such as PBDEs and PFASs, as discussed above under "New organic pollutants". The availability of archived tissue samples from Environmental Specimen Banks enabled temporal trends of these new contaminants to be examined retrospectively. The importance of specimen banking was recognized early on during the development of protocols in the Great Lakes (Gewurtz et al. 2011a; McGoldrick et al. 2010) and was part of long-term monitoring of lake charr from lakes in the Yukon and Northwest Territories.

4.2.2 Northern Lakes

Concentrations of POPs were found to be elevated in lake charr from Lake Laberge in the Yukon in the early 1990s (Kidd et al. 1995, 1998), a finding that led to investigative studies in other Yukon lakes intended to ascribe cause. Lake Laberge and nearby Kusawa Lake were later established as long-term POPs (and later for Hg) trend monitoring sites for lake charr under the Northern Contaminant Program. Great Slave Lake, Northwest Territories, was established as a second long-term monitoring site because of its importance to local communities and concern that development in the south, primarily in Alberta, was contributing to significant contaminant loading to the lake (Evans et al. 2005b). An overview of these trends is discussed below.

In 1993, PCBs, DDT, and toxaphene concentrations were substantially higher in Lake Laberge lake charr fillets (averaging 328 ± 121 ng/g, 392 ± 133 ng/g, and 311 ± 62 ng/g, respectively) than in Kusawa Lake lake charr fillets (86 ± 26 ng/g, 44 ± 22 ng DDT/g, and 121 ± 25 ng/g, respectively). While high by northern

Canada standards, these lake's concentrations were lower than those measured in Lake Michigan (at Saugatuck in 1992), where PCBs averaged 3490 ± 450 ng/g, DDT averaged 1160 ± 180 ng/g, and toxaphene averaged 1730 ± 220 ng/g (De Vault et al. 1996). Higher lake charr POPs concentrations in Lake Laberge than Kusawa Lake were attributed to local sources and food chain length, as discussed previously.

Trends in POPs of Yukon lake charr up to 2011 indicate that ΣPCB, ΣDDT, ΣChlordane, ΣHCH, ΣCBz, and toxaphene concentrations declined sharply in Lake Laberge and Kusawa Lake from the 1990s through the early 2000s and then subsequently leveled off (Table 5; Fig. 18; Muir et al. 2013; Ryan et al. 2013). Trends were significant except for ΣDDTs in Lake Laberge and ΣCBz in Kusawa. ΣHCH declined the most (17% per year for Laberge and 14% per year for Kusawa) followed by ΣChlordane (12% per year and 13% per year, respectively) and ΣPCB (7.5% per year and 14% per year). In Lake Laberge, some of the decline in POPs was due to growth dilution, lower lipid content, increased forage fish abundance, and possible shifts in zooplankton community structure (Ryan et al. 2013). However, in the absence of other biological monitoring in Lake Laberge (and Kusawa Lake), a cause of changes in POP concentrations is difficult to confidently ascribe. Other studies in the Great Lakes noted reductions in the rate of POPs decline since the 1990s, with trend reduction related to continued recycling of POPs from sedimentary sinks, inputs from the watershed, and changes in food-web structure (Bhavsar et al. 2007). In Yukon lakes, which have mountain glaciers within their catchments, continued inputs of POPs from glacial melt may also delay reduction (Donald et al. 1999).

In contrast to Yukon lakes, ΣPCBs in lake charr from Great Slave Lake declined slower at a nonsignificant rate (Fig. 18). Initial measurements in 1993 showed that ΣPCBs concentrations averaged 14 ± 7 ng/g in west basin lake charr and 25 ± 19 ng/g in east arm lake charr, like other POPs (Evans and Muir 2016). A closer examination of the data showed that concentrations of other POPs appeared to be declining from the 1990s to 2002, then increased sharply in 2004, and declined thereafter, particularly in recent years. ΣDDT declined significantly in the west basin (8.3% per year) and east arm (6.0% per year). p,p′-DDE became the predominant DDT degradation product in recent years. ΣChlordane declined in the west basin but not east arm lake charr, in contrast to Yukon lakes where concentrations consistently declined, and concentrations are higher in Great Slave Lake than Yukon lakes. ΣHCH declined in both the east arm (10%/year) and west basin 15%/year) at a rate similar to Yukon lakes, although concentrations tend to be higher in Great Slave Lake. Toxaphene declined in lake charr (14%/year) from west basin, Great Slave Lake, although the decline was not significant in east arm lake charr. The lack of a significant decline of ΣPCBs and total (tetra-hexa) chlorobenzenes (ΣCBz) may be related to past sources in urban and industrial areas of the lake and its large watershed. Riverine inputs and atmospheric deposition are important sources of contaminants to Great Slave Lake. The major inflow is the Slave River formed by the confluence of the Peace and Athabasca Rivers. Development, particularly pulp and paper mills but also agriculture, oil sands activities, and urban development

Table 5 Percent annual decline or increase per year[a] for legacy POPs in lake charr fillets from Lake Laberge and Kusawa Lake in the Yukon and the west basin and east arm of Great Slave Lake (GSL). From Chapter 4 of the Canadian Arctic Contaminants Assessment Report (Muir et al. 2013)

Lake	Period	Years	ΣPCBs[b]	ΣDDT	ΣChlordane	ΣHCH	ΣCBz	Toxaphene
Laberge	1993–2011	13	−7.5	3.7 ns	−12	−17	−7.1	−9.6
Kusawa	1993–2011	13	−14	−23	−13	−14	−4.5 ns	−8.5
GSL west	1993–2011	12	−3.9 ns	−8.3	−5.8	−15	+2.0	−14
GSL east	1993–2011	13	−0.1 ns	−6.0	−0.6 ns	−10	+0.9 ns	−9.0 ns

[a] Percent annual change per year was determined from the half-lives ($t_{½}$), estimated from the log mean concentrations versus time (year) using the equation (LN (2)×100/$t_{½}$

[b] NS = not statistically significant ($P > 0.05$)

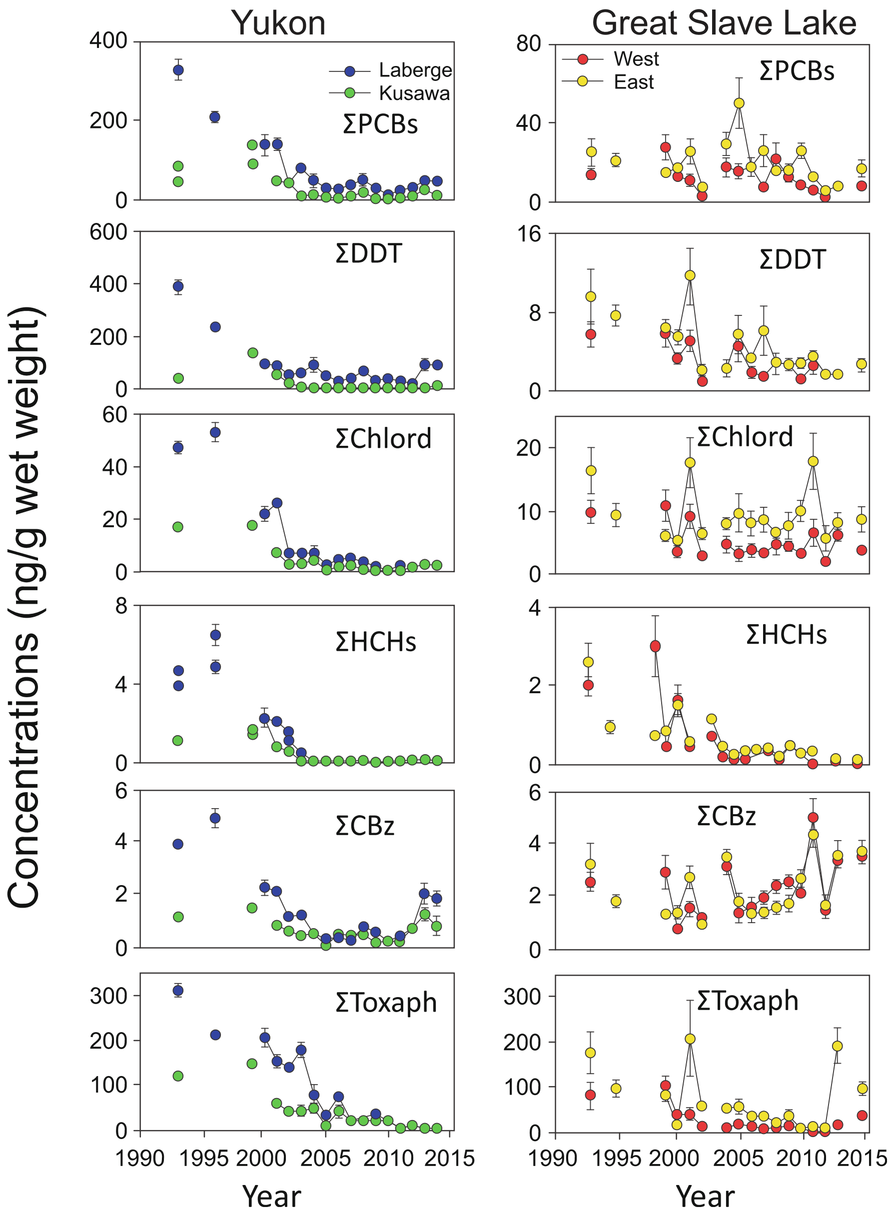

Fig. 18 Temporal trends in persistent organic pollutants (mean and standard error) in lake charr *Salvelinus namaycush* muscle from Lake Laberge and Kusawa Lake in the Yukon and the west basin and east arm of Great Slave Lake. Data from Muir et al. (2013) and Ryan et al. (2013)

fueled concerns that the lake is being adversely impacted by contaminant input via the Slave River and long-range atmospheric inputs (Evans and Muir 2016; Mudroch et al. 1992). The west basin is profoundly affected by the Slave River inflow, while the east arm has clearer and less productive waters that are more strongly influenced by long-range atmospheric inputs.

4.2.3 Great Lakes Region

A large number of studies have examined temporal trends of POPs and Hg in lake charr in the five Great Lakes. Detailed interpretation of annual collections started to be published in the 1980s with studies initially describing the first 10–12 years of data collection (Baumann and Whittle 1988; Borgmann and Whittle 1991; DeVault et al. 1986), and subsequently, temporal trends of POPs from the 1970s to the late 1990s and early 2000s (Hickey et al. 2006); Carlson et al. 2010). As monitoring studies continued, data were incorporated into peer-reviewed publications, government reports, and public documents such as the triennial State of the Great Lakes Reports (SOGLR) (ECCC and U.S. EPA 2017). The Toxic Chemicals in Whole fish sub-indicator of the SOGLR included summaries of the current status and trends for several different contaminants. Here, we describe trends of selected contaminants from the 1970s to 2017, review the most recent temporal trends of lake charr, and examine factors influencing trends. Given the large number of studies, we focus on three legacy contaminants (PCBs, DDT, and PCDD/F) and three new contaminants (PBDEs, PFOS, and HBCDD).

PCBs have been monitored in lake charr from the Great Lakes since the onset of monitoring in 1977. High levels of PCBs were present in Great Lakes fishes and in other environmental media, which led to bans on production and most uses of PCBs in both Canada and the USA by 1979 and globally under the Stockholm Convention in 2004 (although not ratified by the USA). Since the bans, concentrations in lake charr from all Great Lakes declined 2–9% per year (ECCC and U.S. EPA 2017) (Fig. 19). From 1999 to 2014, concentrations of PCBs in lake charr from Lakes Huron, Michigan, Ontario, and Superior, after age normalization, declined 6.0–17.2% per year (Zhou et al. 2018). Declines were smallest in nonindustrial sampling sites on Lake Michigan and most rapid in remote sites on Lake Superior. Despite continued long-term declines, PCBs are still the most predominant class of contaminants measured in lake charr from the Great Lakes (Fig. 13). PCB concentrations remain above the target of 100 ng/g ww in the 1987 amendment to the GLWQA, and are the primary cause of fish consumption advisories in the Great Lakes (Bhavsar et al. 2011; Guo et al. 2017; McGoldrick and Murphy 2016).

DDT and its environmental metabolites DDE and DDD (ΣDDT) are the most abundant organochlorine pesticide in whole-body homogenates of lake charr monitored by ECCC and the U.S. EPA (McGoldrick and Murphy 2016). Like PCBs, DDT was one of the original contaminants added to Annex A of the Stockholm Convention and has been measured since the onset of monitoring in the Great Lakes. Registration and uses of DDT were banned in the USA and Canada in the 1970s and

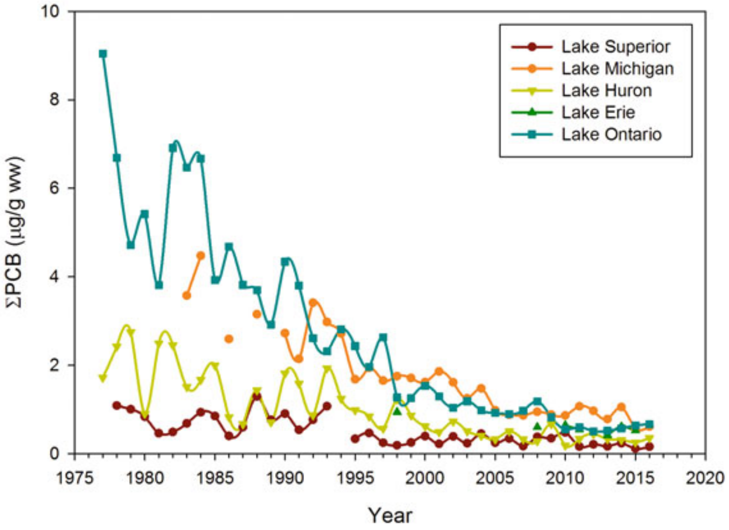

Fig. 19 Annual mean concentrations of ΣPCBs measured by ECCC and U.S. EPA in whole-body homogenates of lake charr *Salvelinus namaycush* from each of the Great Lakes (1977–2016)

all existing stocks of DDT in Canada were to be disposed by 1990. Concentration of ΣDDTs in lake charr declined substantially after the onset of monitoring, with current levels 88–95% lower than the maximum observed concentration in each Great Lake, well below the target of 1.0 μg/g specified in the 1987 GLWQA (Fig. 20). Age-adjusted ΣDDT concentrations in lake charr during 2004–2014 also declined 8.5–18.4%, with the slowest decline in Lake Ontario and the fastest decline in Lake Michigan (Zhou et al. 2018).

Lake Simcoe is a moderately large lake north of Lake Ontario and south of Georgian Bay. Situated relatively close to the urban area of southern Ontario and to intensively farmed areas south of the lake, it presents an interesting case study for temporal contaminant trends. An early study documented elevated ΣDDT concentrations in lake charr averaged 9460 ng/g in 1970 fall-caught lake charr and a rapid decline to 4430 ng/g for fish caught in fall 1975 and 1976 (Frank et al. 1978). Similarly, ΣPCBs concentrations averaged 5050 ng/g in 1970 and 1570 ng/g in fall 1975–1976 and PCBs and ΣDDT continued to decline in lake charr with lipid-based concentrations fitting a first-order exponential decay model over the 36-year period (1970–2006) (Gewurtz et al. 2011c). Mercury concentrations in Lake Simcoe lake charr also declined exponentially from 1970 to 2006, and length was not a significant covariate due to use of a limited size range (55–65 cm lengths) of lake charr (Gewurtz et al. 2011c). Declines of all three of these contaminants (DDT, PCB, and Hg) reflect reductions in local municipal wastewater and agricultural emissions to the lake similar to observations in the Great Lakes.

The final legacy contaminant group to be discussed, PCDD/Fs, are produced as by-products in industrial processes or through combustion of chlorinated

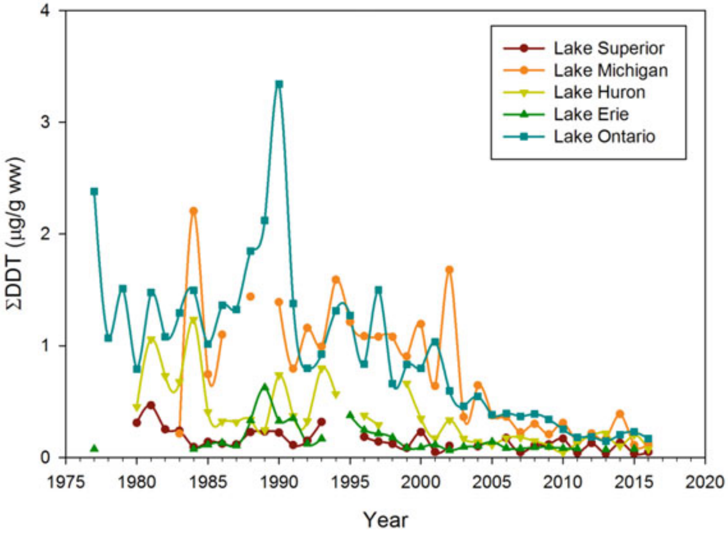

Fig. 20 Annual mean concentrations of ΣDDT measured by ECCC and U.S. EPA in whole-body homogenates of lake charr *Salvelinus namaycush* from each of the Great Lakes (1977–2016)

compounds. These highly toxic contaminants have also been measured and monitored in various media since being detected in Great Lakes biota, including lake charr (Bhavsar et al. 2008; Hallett 1985; Huestis et al. 1997). During 2004–2014, concentrations of 2378-TCDD and 2378-TCDF declined 41–73% across the Great Lakes (Pagano et al. 2018). Despite these declines, toxic equivalents (TEQ) based on PCDD/F exceeded Canadian Tissue Residue Guidelines (TRG) for protection of mammalian consumers of aquatic biota (0.71 pg/g diet). Based on trend models, TEQ based on TCDD/F and non-ortho PCBs in lake charr will remain above the TRG beyond 2050 (Pagano et al. 2018).

In the early 2000s, attention began to shift toward other classes of POPs that were being detected in Great Lakes biota. Unlike many legacy contaminants used in industrial applications or applied for pest control, several POPs were being used as additives to consumer products as flame retardants or as water or stain repellants. The first reports of PBDEs in lake charr from the Great Lakes were published in the early 2000s (Luross et al. 2002; Stapleton and Baker 2003). Subsequent retrospective investigation of trends using archived tissue samples from the U.S. EPA specimen bank demonstrated that concentrations of PBDEs increased exponentially in lake charr from all Great Lakes between 1980 and 2000 (Zhu and Hites 2004). PBDEs were added as routine analytes by ECCC and the U.S. EPA in Great Lakes monitoring programs starting in 2005 and through the additional use of retrospective analysis of archived samples filled in gaps to create a long-term picture of PBDEs in lake charr from the Great Lakes (Zhu and Hites 2004). Levels of all PBDEs appear to have peaked in or prior to 2000, and have since declined with estimated halving times of 3–5 years in Lakes Michigan, Huron, and Ontario, 13 years in Lake

Superior, and although declining, without significant trend in Lake Erie (Crimmins et al. 2012). These declines preceded official industry production phase outs and government regulatory actions, so were likely a result of replacement of PBDEs with other products in anticipation of restriction on use (Alcock et al. 2003; Hardy 1999). The most abundant PBDE congeners measured in lake charr were tetra-brominated BDE-47, penta-brominated BDE-99 and -100, and hexa-brominated BDE-153 and -154 (Gewurtz et al. 2011d). Of these congeners, penta-brominated BDEs are of most concern despite their declines due to the frequency and magnitude of exceedances of Canadian Federal Environmental Quality Guidelines for fish tissues (1.0 ng/g ww) (Fig. 21).

Perfluorooctane sulfonate (PFOS) is a chemical within a large group of per- and polyfluoroalkyl substances (PFAS) that was used as a water, oil, and grease repellant on paper products and fabrics and in firefighting foams. Similar to PBDEs, the primary manufacturer of PFOS voluntarily phased out its manufacture and use in products in 2002 due to concerns surrounding its persistence and potential for causing environmental harm and impending government restrictions and regulations. PFOS levels in lake charr from Lake Ontario increased from 43 ng/g in 1980 to 180 ng/g by 2001 (Martin et al. 2004). Follow-up investigations of trends of several PFAS compounds in lake charr from Lake Ontario that combined current monitoring with analyses of archived tissues from ECCCs specimen bank and previous published studies found the best-fit model for the trend in PFOS was an exponential rise at estimated rates of ~6% per year until levelling off in the 1990s and remaining stable to 2009 (Furdui et al. 2008; Gewurtz et al. 2012). Continued monitoring by ECCC and U.S. EPA in the Great Lakes have since shown high interannual

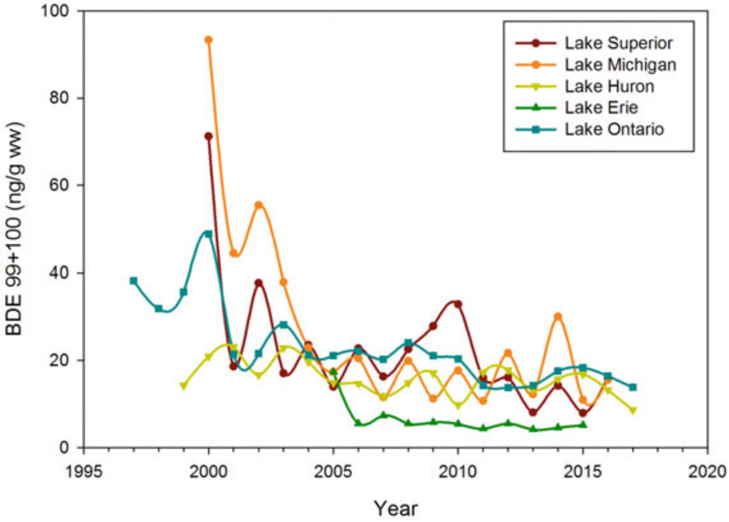

Fig. 21 Annual mean concentrations of BDE99+100 measured by ECCC and U.S. EPA in whole-body homogenates of lake charr *Salvelinus namaycush* from each of the Great Lakes (1997–2017)

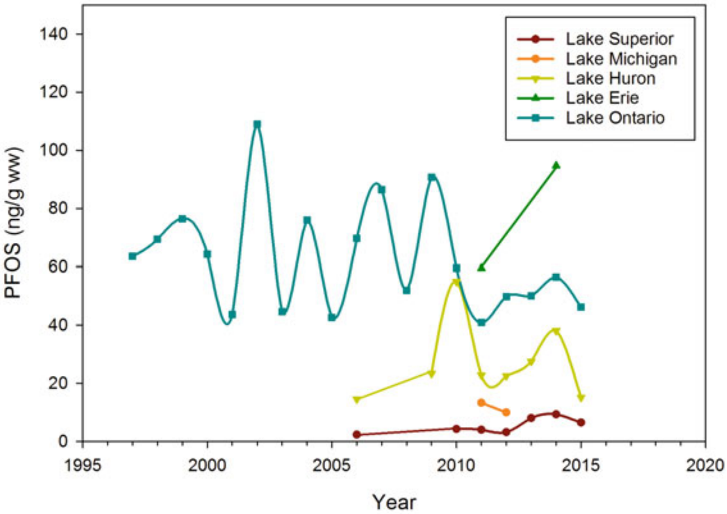

Fig. 22 Annual mean concentrations of PFOS measured by ECCC and U.S. EPA in whole-body homogenates of lake charr *Salvelinus namaycush* from each of the Great Lakes (1977–2016)

variability in the concentration of PFOS in lake charr (Fig. 22). In Lakes Superior, Michigan, Huron, or Erie, PFOS concentration in lake charr exhibited no temporal trends since the phase out of manufacture in 2002, whereas in Lake Ontario PFOS concentrations have started to decline (U.S. EPA and FDA 2017).

Concentrations of hexabromocyclododecane (HBCDD), a chemical widely used as replacement for PBDEs (Koch et al. 2015), especially α-HBCDD, the most abundant HBCDD isomer in lake charr, were fairly constant from 1980 to 2000 and then rose post-2000 through to 2010, after which the increase in concentration ceased and may be decreasing (Su et al. 2018). The rise and plateau of HBCDD in lake charr closely mimicked when PBDEs were phased out of production by government regulation of HBCDD through international and domestic actions. Levels of γ-HBCDD, the primary component of the technical mixture, declined significantly from 1975 to 2015, possibly because new releases of HCBDD to the environment are declining in response to regulatory restrictions (Su et al. 2018).

Long-term monitoring of contaminants in lake charr in the Great Lakes, in conjunction with retrospective analyses of fish tissue from specimen banking programs, identified chemicals that were causing or may cause harm to the environment. The data not only helped support actions to restrict use of these chemicals but also served to track progress of actions to reduce levels in the environment (McGoldrick and Murphy 2016). Concentrations of legacy contaminants in lake charr from the Great Lakes all declined significantly from high levels when first identified in the 1970s. Availability of preserved specimens in ECCC and U.S. EPA specimen banks also allowed for quick determination of trends for new chemicals as they emerged and contributed to decisions by governments and industry that halted increases

(PFOS and HBCDD) and initiated declines (PBDEs). Continuous technological improvements in analytical capabilities, such as recent advancements in nontarget analysis, will allow monitoring programs to adopt new and improved methods to identify contaminants of concern in lake charr from the Great Lakes (Fernando et al. 2018).

4.3 Effects of Changing Climate and Food Webs on Contaminant Temporal Trends

Climate warming may have unexpected effects on contaminant cycling due to the balancing effects of uptake rates and growth dilution in fishes, and because of temperature sensitivities of individual species (Ng and Gray 2011). Increasing trends of mercury in lake charr from lakes in southern and northern Ontario from the early 2000s through 2012 were attributed to remobilization of Hg, accelerated conversion to MeHg, and changes in food-web structure caused by invasive species (Gandhi et al. 2014b). Similar qualitative observations have been made in the Great Lakes. In Lake Ontario, Hg and PCBs in lake charr muscle declined overall but year-to-year variation was related to food-web changes and variation in Hg and PCB fluxes from atmosphere or sediments (Visha et al. 2015). Concentration of Hg in lake charr in Lake Ontario decreased 6.7–7.5% per year from 2004 to 2009 and then increased 3.3–4.6% per year from 2010 to 2015 (Zhou et al. 2017). The increasing Hg trend in Lake Ontario lake charr contrasted with lakes Superior and Michigan, where Hg declined from 2004 to 2015, likely because atmospheric emissions of Hg increased and Hg reservoirs in sediments that could be resuspended by increasing storms and less ice cover were greater in Lake Ontario than the other lakes (Zhou et al. 2017).

Changes in Great Lakes food webs have affected health of Great Lakes' fishes over the past 20 years, including changes in bioaccumulation potential of contaminants in top predators, such as lake charr (Murphy et al. 2018). Food-web structure has been stressed by nutrient availability, invasive species, anthropogenic change, declines in prey availability, increases in predator density, shifts in predator–prey ratios, and density-dependent growth declines resulting in higher chemical concentrations of POPs and mercury (He et al. 2015, 2016; Lake Michigan Lake Trout Working Group 2016; Tsehaye et al. 2014). Consequently, long-term chemical monitoring and surveillance programs in the Great Lakes are incorporating fish age into their assessments of concentrations and trends as a result.

Minor oscillations (within an order of magnitude) in temporal trends of POPs in lake charr from Lakes Ontario, Huron, Michigan and Superior from 1999 to 2009 were attributed to trophodynamic, meteorological, or climatological changes (Chang et al. 2012). Surface water temperature was not correlated to fish POP concentrations in any lakes, thereby suggesting that temperature alone was not directly responsible for the oscillations (Chang et al. 2012). Modelling of the effect of a warming climate on PCB 77 (2,4,2′4′-tetrachlorobiphenyl) uptake by lake charr predicted indirect effects due to changing growth and consumption rates of major prey species that might lead to higher chemical uptake (Ng and Gray 2011).

5 Human and Wildlife Contaminant Exposure

5.1 Human Dietary Exposure and Advisories

In general, fish consumption is an important pathway for human exposure to contaminants. For example, PCB concentration in anglers was positively related to years of Great Lakes sport fish consumption and the number of fish meals consumed (Cole et al. 2002). Similarly, blood Hg levels were higher among anglers consuming fish caught in Great Lakes areas of concern than within the general population (Cole et al. 2004). First-Nations communities in northern Ontario and Manitoba with exposure to PCBs and DDE via fish consumption had higher incidence of type-2 diabetes (Marushka et al. 2018). Lake charr form a significant portion of the fish diet of First Nations communities in Ontario (Marushka et al. 2017) and are a popular sport fish (Gandhi et al. 2014b). Monitoring of lake charr for assessment of human health-related exposure has been conducted primarily by the US Great Lakes states and Provincial and Territorial governments in Canada. These programs have focused on analyzing fillets for Hg and in some cases for other priority chemicals such as PCBs.

Consumption advisories are generally based on risk assessments that consider tolerable daily intakes and exposure rates to contaminants. Tolerable daily intake (TDIs) for human consumers of fish have been developed by health agencies around the world. Many consumption advisories are related to contaminants in lake charr, especially Hg. Provincial, Territorial, and State agencies post advisories on web sites and in fishing regulation guides (http://ec.gc.ca/mercure-mercury/default.asp?lang=En&n=DCBE5083-97AD-4C62-8862). Advisory program managers from U.S. government health, water quality, and fisheries agencies bordering the Great Lakes have developed protocols for issuing fish consumption advice (MDH 2018).

Mercury fish consumption advisories are not evenly distributed across the lake charr range, but are most numerous in areas with strong sport fish monitoring programs, with Hg often measured as part of stock and other assessments. Regions where Hg in fish has not been identified as a wide-spread problem have no or limited Hg monitoring programs, such as Nunavut and British Columbia. For example, lake charr are uncommon in British Columbia and advisories have been issued for only three locations: Williston Reservoir and two lakes near two historical gold mining sites. In the Yukon, advisories are precautionary, nonspecific to any lake and river, and only for women of childbearing years and children less than 12 years old. The advisory states that lake charr under 40 cm can be consumed without limit while fish >60 cm should be consumed only 1–2 times per week. Several Hg advisories have been issued for lake charr (and other predatory fishes) in the Northwest Territories (NWT HSS 2018). Advisories are most strongly associated with small to medium size lakes (<500 km^2), Hg methylation rates are greater, and with older fish populations (Evans et al. 2005a; Lockhart et al. 2005; Stephens 1995). In contrast, hundreds of Hg consumption advisories have been issued for Great Lakes lake charr. Large sport fisheries, relatively easy access for study, and significant local Hg emissions all contribute to the prevalence of Hg advisories in the Great Lakes basin. Consumption advisories typically are based on length categories of lake charr with large fish having more stringent consumption limitations due to bioaccumulation and biomagnification (Bhavsar et al. 2011; Gandhi et al. 2015; Gewurtz et al. 2011b).

The number of Hg advisories for fish consumption is projected to increase in regions such as the Great Lakes where climate is warming. For example, the percentage of lakes in northern Ontario with "do not consume" advisories for lake charr is expected to increase from 0–6% to 0–52% for the general population and from 21–72% to 24–88% for sensitive populations by 2050, based on an apparent slow increase in Hg concentrations (Gandhi et al. 2015). Similarly, the number of health advisories is also predicted to increase in southern Ontario from 0–7% to 0–24% for the general population and 13–66% to 25–95% for sensitive populations (Gandhi et al. 2015).

Consumption advisories based on POPs are fewer in number than for Hg, with more advisories in the five Great Lakes than inland lakes in the region (Dellinger 2004). PCBs are the major driver of consumption advisories, with PCDD/PCDFs of secondary importance, toxaphene, and mirex only occasionally high enough to result in advisories (Gandhi et al. 2014a, 2017). While DDT has been implicated in many measures adversely affecting bird health, the advisory benchmark for human consumption of fish is high (Table 6) relative to environmental concentrations. Average concentrations of PCBs, OCPs, PBDE congeners, and PFOS in lake charr from lakes Superior, Michigan, Huron, and Ontario did not exceed Province of Ontario benchmarks (Dellinger et al. 2014; Table 6), whereas total TEQs for PCDD/Fs and dioxin-like PCB congeners exceeded guidelines (Pagano et al. 2018). In Lake Simcoe, PCBs in large lake charr (>65 cm) exceeded the consumption guideline (211 ng/g) for sensitive populations, while PBDEs (BDE-47, BDE-99, BDE-153-, BDE-209) were below consumption guidelines (Gewurtz et al. 2011c). In the early 1990s, Health Canada issued a consumption advisory for lake charr and burbot *Lota lota* liver from Lake Laberge in the Yukon based on high toxaphene concentrations, but this advisory was discontinued due to declining contaminant levels (Ryan et al. 2013). Lake charr from four northern Québec lakes had PCB and PCDD/PCDF concentrations below Health Canada guidelines (Laliberté and Tremblay 2002).

Consumption advice related to Hg in lake charr and other fishes varies, particularly with respect to subsistence consumers who are more likely to consume fish regularly and in larger quantities than occasional recreational anglers (Table 7). For example, the U.S. Food and Drug Administration (U.S. FDA) set an action level of 1000 ng/g for Hg, while the U.S. EPA set a screening value of 400 ng/g ww for recreational anglers and 49 ng/g for subsistence fishers for total Hg. The U.S. FDA action level used for commercial fish is an indicator of chemical residue levels in fish and shellfish that should not be exceeded for the general population (U.S. EPA

Table 6 "Do not eat" fish consumption advisory benchmarks used by the Province of Ontario and potential health effects for major contaminants found in Great Lakes fishes, including the lake charr *Salvelinus namaycush*. Sensitive population refers to women of childbearing age and children. Modified from Gandhi et al. (2017). Data sources for concentration are provided in the footnote

Contaminant	Units (wet wt)	General population	Sensitive Population	Range of mean concentration[a]	Potential health effects
Mercury	ng/g	>1800	>500	99–159	Neurotoxicant, can also damage immune, digestive, and nervous systems
PCBs	ng/g	>844	>211	53–207	Neurotoxicant, affects reproductive and immune systems, developmental effects, potential carcinogen
Dioxin/furan/dioxin-like PCBs (TEQs)	pg/g	>21.6	>5.4	22–51	Neurotoxicant, affects reproductive, immune, and endocrine systems
Mirex	ng/g	>657	>164	1–7	Can affect stomach, intestines, liver, kidneys, eyes, thyroid, nervous system, reproductive health
Toxaphene	ng/g	>1877	>469	49–173	Potential carcinogen, convulsions, liver and kidney damage
Chlordane	ng/g	>469	>117	–	Affects nervous and digestive systems and liver
DDT	ng/g	>5000	>5000	21–83	Affects nervous system, potential carcinogen, developmental, reproductive effects
HCB	ng/g	>2534	>634	2–6	Affects nervous system, liver, thyroid, possible carcinogen, endocrine disruptor
PFOS	ng/g	>640	>160	0.85–46	Potential carcinogen, endocrine disruptions, oxidative stress
Aldrin + dieldrin	ng/g	>939	>235	–	Potential carcinogen, convulsions, nervous system effects, kidney damage
BDE-47		>939	>235	55–135	Can affect thyroid and liver, behavioral changes, may affect immune system, possible carcinogen, BDE 47 and 99 more toxic than BDE 209
BDE-99		>939	>235	15–29	
BDE-153		>1877	>469	3.8–4.4	
BDE-206		>65,701	>16,425	–	

[a] Range of average concentrations of PCBs and OCPs (except toxaphene) in lake charr *Salvelinus namaycush* reported during 1999–2011 for Lakes Superior, Michigan, and Huron from Dellinger et al. (2014). Range of average concentrations for Lakes Superior, Michigan, Huron, and Ontario for BDE 47, 99, and 153 during 2004–2009 from Crimmins et al. (2012) for PFOS during 2008–2009 from Guo et al. (2012), for toxaphene during 2009 from Xia et al. (2012), and for dioxin/PCB TEQs from Pagano et al. (2018)

2000). As of 2017, the U.S. EPA and U.S. FDA issued joint advice for mercury in fish and shellfish (U.S. EPA and FDA 2017). The National Lake Fish Tissue Study (Stahl et al. 2009; U.S. EPA 2009) used a fish tissue criterion of 300 ng/g based on a criterion for methyl Hg (U.S. EPA 2001). The Great Lakes Fish Consortium, Fish Advisory Workgroup from Great Lake states issued protocols for fish consumption advisories for PCBs and mercury and discussion papers related to chlordane and toxaphene (MDH 2018). Their guidelines for mercury are similar to those issued by the U.S. FDA and U.S. EPA. However, the tissue concentration for unrestricted consumption for PCBs (50 ng/g) is 20-fold higher than the U.S. EPA screening value (Table 7). In northern Canada, an Hg guideline of 200 ng/g was initially considered for subsistence fishers (Lockhart et al. 2005; Wheatley 1979), in addition to the Health Canada guideline of 500 ng/g for commercial sale of fish, but has not been used for advisories. Rather, guidelines generally are issued only for lakes where average Hg concentration in the species exceeds 500 ng/g, with consumption of small fish in greater quantities than larger fish (Health Canada 2007). Concentrations of 370 ng/g (average for walleye or yellow perch) resulted in average consumption of 70% of TDI values for women (60 kg) and 110% for children 1–4 years old (Health Canada Hg risk assessment 2007), and are likely similar for lake charr consumption.

The U.S. FDA action level for PCBs is 2000 ng/g (ww) while U.S. EPA screening values (SV) are 20 ng/g for recreational and 2.45 ng/g for subsistence fishers (Table 7). SVs are defined as the concentration of a chemical in fish tissue that is of potential public health concern and used as a threshold value against which tissue residue levels of the contaminant in fish are compared. Screening values are based on both noncarcinogenic and carcinogenic effects of the chemical contaminant (U.S. EPA 2000). Screening values for dieldrin and heptachlor epoxide are also quite low relative to other OCPs. As a consequence of these stringent guidelines for subsistence fishers, lake charr in several lakes in Alaska exceeded consumption thresholds for dieldrin, chlordane, and p,p′-DDE (Ackerman et al. 2008; Flanagan Pritz et al. 2014). Σ_7PCBs (sum of seven major congeners) in lake charr from Alaskan lakes (Fig. 12) generally did not exceed subsistence consumption values, but likely would have if based on a larger suite of congeners. Lake charr from inland lakes in the Great Lakes region and from Kusawa Lake, Lake Laberge, and Great Slave Lake in northern Canada exceed subsistence limits for PCBs (Fig. 18).

A more recent approach to consumption advice for contaminants is based on risk-benefit assessment and a recognition that fish contain high levels of healthy omega-3 fatty acids (Dellinger 2004; Dellinger and Ripley 2016; Knuth et al. 2003; Reyes et al. 2017; Turyk et al. 2012; Williams et al. 2017). Furthermore, a shift in diet from wild-caught fish to alternate foods that are higher in unhealthy fats, sugars, and salts poses a different health risk. Subsistence fish consumers may also not have the means to purchase or have access to alternate foods. Therefore, an increasingly large number of studies are investigating fatty acids in fish in addition to contaminant levels when developing more tailored consumption advice, such as research to optimize the balance between contaminant and fish fatty acid consumption. Advice will be lake- and fish-size-specific. For example, lake charr in Lake Ontario are high

Table 7 Comparison of FDA Action Levels and Tolerances with U.S. EPA screening values (SV), Health Canada guidelines, and Great Lakes Consortium advisory guidance. Modified from Tables 1 and 2 in the U.S. EPA guidance document for fish advisories (U.S. EPA 2000)

Chemical	Units	FDA action level	EPA SV: recreational fishers	EPA SV subsistence fishers	Health Canada	Great lakes consortium DNE[a]	Great lakes consortium unrestricted consumption
Mercury	ng/g	1000	400	49	500	950	50
Chlordane	ng/g	300	114	14			
Dieldrin	ng/g	300	2.5	0.307			
Heptachlor epoxide	ng/g	300	4.39	0.540			
Mirex	ng/g	100	800	98			
Total DDT	ng/g	5000	117	14			
PCBs	ng/g	2000	20	2.45		1890	50

[a] "Do not eat" values

in eicosapentaenoic (EPA) and docosahexaenoic acid (DHA) fatty acids that can be good food choices (Strandberg et al. 2017), although, Hg and POP concentrations also need to be considered when weighing the merit of consuming lake charr versus other species such as lake whitefish or yellow perch. The lake charr is somewhat lower in fatty acid concentration, but substantially higher in POPs and Hg than other species.

5.2 Comparison to Guidelines for Piscivorous Fish and Fish-Eating Wildlife

Sandheinrich et al. (2011) proposed a toxicity reference value (TRV) for Hg of 300 ng/g wet weight (whole body) for piscivorous fishes (not lake charr) and concluded that fish inhabiting inland lakes were at more risk than species inhabiting the Great Lakes, based on lowest observable effect levels for sublethal effects of mercury on freshwater fish, including changes in reproductive health (Dillon et al. 2010; Sandheinrich and Wiener 2011). Based on an assessment of the large OMECC database, 70% of lake charr populations in northern Ontario and 57% in southern Ontario exceeded a TRV for Hg of 470 ng/g in fillet (or 330 ng/g ww whole body) for mature lake charr, with exceedances projected to increase to 76–92% for northern and 79–89% in southern lakes by 2050 due to slowly increasing contaminant concentrations (Gandhi et al. 2015). Similarly, Hg in lake charr in northern Quebec and in many lakes in Northwest Territories also exceeded the 470 ng/g TRV (Laliberté and Tremblay 2002).

Tissue residue guidelines (TRG) for protection of fish eating wildlife are available for selected POPs and Hg (CCME 1999; U.S. EPA 1995), but do not specifically consider lake charr as prey. Because adult lake charr inhabit cold deep waters (Marsden et al. 2021), they are unlikely prey for most fish-eating mammals and birds except during spring when they forage in shallow nearshore waters or at the surface over deep waters (Vinson et al. 2021). Nevertheless, a comparison of guidelines is useful to illustrate the relevance of contaminant measurements. For example, total TEQ (PCDD/F +dioxin-like PCBs) in Great Lakes lake charr (Table 6) that exceeded Canadian TRGs (CCME, 2001) for fish-eating mammals and birds indicate the potential for adverse effects if consumed (Pagano et al. 2018). Based on an assessment of U.S. EPA wildlife contaminant health thresholds, chlordane concentrations in lake charr from three of eight Alaskan lakes exceeded the health threshold for kingfisher (4.5 ng/g), but not for mink (830 ng/g) or river otter (1140 ng/g), and dieldrin and pp-DDE thresholds were not exceeded for any wildlife species (Table 8; Flanagan Pritz et al. 2014; Ackerman et al. 2008).

Table 8 Wildlife contaminant thresholds and tissue residue guidelines (ng/g wet weight). Data sources provided in the footnotes

Chemical	Kingfisher[a]	Mink[a]	River Otter[a]	Fish	CCME TRG for fish consuming wildlife[b]
Chlordane	4.5	830	1140		–
Dieldrin	360	20	30		–
Total DDT	490	360	490		14
PCBs	440	130	180		0.00079–0.0024 (TEQs)
PCDD/Fs	–	–	–		0.00071–0.00475 (TEQs)
Toxaphene	–	–	–		6.3
Methyl Hg	–	–	–	300[c]	33

[a] Ackerman et al. (2008)
[b] CCME (http://st-ts.ccme.ca/en/index.html); (CCME 1999)
[c] Sandheinrich et al. (2011)

6 Conclusions

The contaminants literature for lake charr is substantial, and encompasses trend measurements, bioaccumulation, and adverse biological effects. Nonetheless, many data gaps are evident. For adverse effects, lake charr have not been extensively studied as test organisms (with much more work on rainbow trout), which increases the uncertainty of risk assessments of contaminant exposure. Notably, lake charr were the most sensitive of 11 species to TCDD in tests of free embryo mortality (Walker et al. 1996). Most of the 1980s and 1990s and new or emerging contaminants (outlined by McGoldrick and Murphy 2016) have not been tested for their toxicity to lake charr. The combined effects of exposures to chemical mixtures are also unknown and remain a major knowledge gap. Another major knowledge gap is the extent to which lake charr populations are vulnerable to the combined effects of contaminants with other stressors, such as increased primary production, prey abundance, and invasive species all of which are changing in respect to a changing climate.

Long-term monitoring of POP and Hg concentrations in lake charr has been invaluable in assessing the effectiveness of domestic regulatory programs for chemical management, like Canada’s CMP and the binational Great Lakes Water Quality Agreement, and for judging progress on global bans on POPs through the Stockholm Convention. Continuing these trend monitoring studies is important because they become more statistically powerful with each new sampling year. Long-term trend data are increasingly being interpreted with consideration of climate change impacts on food webs and contaminants (e.g., greater methylation of Hg). Climate warming is already emerging as a serious concern for Hg in lake charr, with concentrations increasing in Ontario (Gandhi et al. 2014b, 2015) and in Great Slave Lake (Evans et al. 2013). However, compensating factors, such as increased growth rates and increased fishing pressure as lakes become more accessible, may counteract this. Bioenergetics modelling of effects of climate warming on PCBs in lake charr have illustrated the complexity of this issue (Ng and Gray 2011). Understanding future contaminant trends and possible adverse effects will require strong ancillary data, such as data on prey fish populations, stomach contents, stable isotope ratios, other indicators of diet and food-web relationships, measures of primary productivity, and climate parameters such as lake thermal regimes and ice out times.

The suite of contaminants measured in Great Lakes lake charr is extensive, but more limited for other lakes across Canada and the northern USA, except Hg. Concentrations of POPs and new contaminants such as chlorinated paraffins and volatile methyl siloxanes are likely lower in lake charr across Canada than in the Great Lakes (Gewurtz et al. 2011d, 2013; Saborido Basconcillo et al. 2015), but perhaps not for all organic pollutants, especially where local sources are available (Ackerman et al. 2008) or where long food chains lead to lake charr (Kidd et al. 1998; Rasmussen et al. 1990; Ryan et al. 2013). Thus, carefully revising and possibly expanding the number of contaminants is important to keep monitoring programs relevant. This has begun under the U.S. EPA Great Lakes, Environment and Climate Change Canada monitoring programs, and Northern Contaminants Program. The number of contaminants measured should also be expanded under provincial, state, tribal, and territorial programs to more fully assess human exposure, especially from subsistence fish consumption. Maintenance and availability of archived samples in formal specimen banking programs, such as by ECCC and the U.S. EPA, for assessing new contaminants is also important and requires ongoing commitment from funding agencies for long-term storage costs.

Acknowledgments We thank Dr. Don Tillitt, Columbia Environmental Research Center, U.S. Geological Survey (Columbia MO) and Dr. Charles Madenjian, Great Lakes Science Center, U.S. Geological Survey (Ann Arbor, MI) for helpful reviews of this chapter.

Appendix 1

Acronyms and chemical terminology

AhR	Aryl hydrocarbon receptor
Aroclor	Commercial name for PCB mixtures, e.g., Aroclor 1254 (54% Chlorine)
BAF	Bioaccumulation Factor (concentration in field collected biota (usually lipid adjusted)/dissolved concentration in water)
BCF	Bioconcentration Factor (concentration in lab exposed biota (usually lipid adjusted)/dissolved concentration in water)
BSAF	Biota-Sediment Accumulation Factor (concentration in field collected biota (usually lipid adjusted)/concentration in sediment (usually organic carbon adjusted)
DHA	Docosahexaenoic acid - a long-chain polyunsaturated fatty acid: 22:6n-3

(continued)

AhR	Aryl hydrocarbon receptor
Dieldrin	Insecticide with a hexachloro-1,3-cyclopentadiene ring. Also, a degradation product of Aldrin
DDT	Insecticide. Chemical name dichlorodiphenyltrichloroethane. Has p,p′ and o,p′ isomers. Also with degradation products DDD and DDE
D4, D5, D6	Cyclic methyl siloxanes. Widely used silicon-based chemicals
EPA	Eicosapentaenoic acid—a long-chain polyunsaturated fatty acid: 20:5n-3
EROD	Ethoxyresorufin o-deethylase. A measure of Hepatic mixed-function oxidase activity, specifically induction of cytochrome p4501A1
GC	Gas chromatography
HBCDD	Hexabromocyclododecane. A flame retardant and replacement for PBDEs
HCH	Hexachlorocyclohexane (isomers α-HCH, lindane or γ-HCH)
log K_{ow}	Logarithm of the octanol–water partition coefficient
MCCPs	Medium-chain chlorinated paraffins (C14–C17 chlorinated n-alkanes)
MeHg	Methyl mercury. The toxic and bioaccumulative form of mercury (Hg)
mirex	Insecticide based on a chlorinated cyclopentadiene caged ring structure
net trophic transfer efficiency (γ)	$\gamma = \Delta$PCB body burden (ng) ÷ amount of PCB ingested (ng)
Non-ortho PCBs	PCB congeners lacking chlorine substitution in the ortho or 2,6 positions on the phenyl ring. Also referred to as “dioxin-like PCBs”
OCPs	Organochlorine pesticides. DDT, chlordane, HCH, dieldrin, mirex, aldrin, toxaphene, etc.
OPEs	Organophosphate ester flame retardant/plasticizers
PBDEs	Polybrominated diphenyl ethers. Tetra-brominated BDE-47, penta-brominated BDE-99 and -100, and hexa-brominated BDE-153 and -154
PCB congener	One of 209 possible structural isomers having 1–10 chlorines on the biphenyl rings
PCBs	Polychlorinated biphenyls
PCDDs	Polychlorinated dibenzo-p-dioxins
PCDFs	Polychlorinated dibenzofurans
PFASs	Poly/perfluoro alkyl substances
PFCAs	Perfluorocarboxylates. Typically consist of perfluorinated 4 carbon to 16 carbon chains
PFOS	Perfluorooctane sulfonate. Contains a perfluorinated 8 carbon chain
POPs	Persistent organic pollutants. Includes PCBs, OCPs. Refers to the chemicals listed by the Stockholm Convention on POPs
PPCPs	Pharmaceuticals and personal care products
SCCPs	Short-chain chlorinated paraffins (C10–C13 chlorinated n-alkanes)
TBARS	Thiobarbituric acid reactive substances. An oxidative stress indicator
TBOEP	Tris(2-butoxyethyl) phosphate, a phosphorus-based flame retardant
TCDD	2,3,7,8-tetrachlorodibenzo-p-dioxin. The most toxic dioxin congener
TCEP	Tris (chloroethyl) phosphate
TEFs and TEQs	TCDD toxic equivalents or factors. Toxicity relative to 2,3,7,8-TCDD = 1.

(continued)

AhR	Aryl hydrocarbon receptor
TL	Trophic level = $TL_{consumer} = ((\delta^{15}N_{consumer} - \delta^{15}N_{primary\ producer})/\Delta^{15}N) + 1$
TMF	Trophic Magnification Factor. TMF = antilog [slope contaminants vs Trophic Level of individual food-web organisms]
TMS	Trophic magnification slope. TMS = antilog [slope contaminants vs $\delta^{15}N$ of individual food-web organisms]
Toxaphene	Insecticide also called polychlorocamphene. Complex mixture of chlorinated bornanes
TP	Trophic position = $TP_{consumer} = \delta^{15}N_{primary\ producer} + (\delta^{15}N_{consumer} - \delta^{15}N_{primary\ producer})/\Delta^{15}N)$
TPHP	Triphenyl phosphate
TRG	Tissue Residue Guidelines
TRV	Toxicity reference value. A toxicological index is used to qualify or quantify a risk of adverse effects to human or wildlife health.
T3	Tri-iodothyronine. Thyroid hormone
VTG	Vitellogenin. A precursor protein of egg yolk used as a biomarker in vertebrates of exposure to estrogenic substances
$\Delta^{15}N$	Trophic enrichment factors (fractionation of ^{15}N from prey to predator)
$\delta^{13}C$	Carbon stable isotope ratio ($^{13}C/^{12}C$). Expressed as parts per thousand (‰) relative to a standard
$\delta^{15}N$	Nitrogen stable isotope ratio ($^{15}N/^{14}N$). Expressed as parts per thousand (‰) relative to a standard

Appendix 2

Aquatic toxicity of 39 pesticides and selected industrial chemicals to lake charr (i.e., Lake trout) *Salvelinus namaycush*. Data extracted from the acute toxicity database (https://www.cerc.usgs.gov/data/acute/acute.html.)

Chemical	Units (/L)	Use	CAS RN	Temp (°C)	Hardness	Type	Duration (hrs)	N	Average 96 hr LC50	Range
2,4-D	mg	Herbicide	94-75-7	5	44	Static	96	6	58.1	44.5–105
2,4-D Butyl ester	µg	Herbicide	94-08-4	10	44	Static	96	14	1040	500–2800
2,4-D Propylene Glycol butyl ether ester	µg	Herbicide	1928-45-6	5	44	Static	96	14	1151	390–2930
Antimycin A	ng	Pesticide	1397-94-0	12	44	Static	24	1	53	45–63
Captan	µg	Fungicide	133-06-2	12	44	Static	96	3	54.4	49–63.2
Carbaryl	µg	Insecticide	63-25-2	12	40	Static	96	5	783	690–2300
Carbofuran	µg	Insecticide	1563-66-2	12	314	Flow-through	96	1	164	119–226
Chlorpyrifos	µg	Insecticide	29921-88-2	12	44	Static	96	6	148	73–227
Coumaphos	µg	Insecticide	56-72-4	12	162	Static	96	1	593	416–846
Diazinon	µg	Insecticide	333-41-5	12	162	Static	96	1	602	400–906
Dichlorvos	µg	Insecticide	62-73-7	12	162	Static	96	2	185	183–187
Dicofol	µg	Acaricide	115-32-2	12	162	Static	96	1	87.0	53–142
Dinitramine	µg	Herbicide	29091-05-2	12	44	Static	96	1	920	776–1090
Dinoseb	µg	Herbicide	88-85-7	10	162	Flowthrough	96	12	202	2.2–1440
Diuron	mg	Herbicide	330-54-1	15	44	Static	96	13	3.0	1.1–11.5
D-Trans Allethrin	µg	Insecticide		12	40	Static	96	2	16.7	16–17.3
EPTC	mg	Herbicide	759-94-4	10	44	Static	96	8	13.7	11.5–1450
Fenthion	µg	Insecticide	55-38-9	12	44	Static	96	3	1573	1370–1900
Flit Mlo	mg	Mosquito Larvicide		12	162	Static	96	1	>700	–
Folpet	µg	Fungicide	133-07-3	12	44	Static	96	2	55.3	24–86.5

(continued)

Chemical	Units (/L)	Use	CAS RN	Temp (°C)	Hardness	Type	Duration (hrs)	N	Average 96 hr LC50	Range
Lethane 384	mg	Insecticide	112-56-1	12	162	Static	96	2	4.0	3.9–4.1
Lindane	µg	Insecticide	58-89-9	12	44	Static	96	2	28.0	24–32
Malathion	µg	Insecticide	121-75-5	12	162	Static	96	2	109	76–142
Methoxychlor	µg	Insecticide	72-43-5	12	162	Static	96	2	16.5	16–17
Methyl parathion	µg	Insecticide	298-00-0	12	162	Static	96	2	3570	3360–3780
Mexacarbate	mg	Insecticide	315-18-4	12	162	Static	96	1	8.2	3.69–18
Naled	µg	Insecticide	300-76-5	12	162	Static	96	1	87.0	53–142
Parathion	µg	Insecticide	56-38-2	12	162	Static	96	2	1705	1490–1920
PCB Aroclor 1016	µg	Industrial	12674-11-2	10	170	Static	96	2	685	480–890
Picloram	mg	Herbicide	19180-02-1	10	44	Static	96	13	3.9	1.6–16.8
Pydraul 50e	mg	Hydraulic Fluid		10	170	Static	96	3	2.4	1.5–2.9
Pyrethrum	µg	Insecticide	8003-34-7	12	44	Static	96	2	28.4	19.7–37
Resmethrin	µg	Insecticide	10453-86-8	12	44	Static	96	2	1.2	0.74–1.68
Ronnel	µg	Insecticide	299-84-3	12	162	Static	96	2	378	265–490
Ru-11679 (bioethan-omethrin)	µg	Insecticide	51202-40-5	12	44	Static	96	1	0.20	0.14–0.20
Ru-11679 (bioethan omethrin)	µg	Insecticide	51202-40-5	12	314	Flowthru	24	1	0.27	0.25–0.30
Temephos	mg	Insecticide	3383-96-8	10	40	Static	96	4	2.7	1.05–4.8
Thanite	µg	Insecticide	115-31-1	12	162	Static	96	2	116	109–123
Trichlorfon	µg	Insecticide	52-68-6	12	162	Static	96	2	790	550–1030

Appendix 3

Half-lives and absorption efficiencies for PBDE congeners in lake charr (i.e., lake trout) *Salvelinus namaycush* and other fish species based on data from the Arnot and Quinn (2015) database for dietary bioaccumulation studies

PBDE Congener	Species	Temp. (°C)	Feeding rate[a] (g/g BW/d)	Exposure Duration (d)	Depuration (d)	Growth rate constant (/d)	Food concentration (ng/g)[b]	Tissue analyzed[c]	Half-life ± SE (d)	Assimilation efficiency	References[d]
BDE 47	sole	19	0.008	84	149	0.008	82.2 ww	M + L	59 ± 21	0.15	1
BDE 47	Common carp	22	0.033	60	100	0.009	174 ww	WB − GIT	30 ± 12	0.93	2
BDE 47	Lake charr	12	0.015	56	112	0.013	2.1 dw	WB −(L & GIT)	39 ± 8	0.22	3
BDE 47	Lake charr	12	0.015	56	112	0.015	11.4 dw	WB −(L & GIT)	346 ± 173	0.43	3
BDE 99	sole	19	0.008	84	149	0.008	85.7 ww	M + L	37 ± 7	0.13	1
BDE 99	Common carp	22	0.033	60	100	0.009	490 ww	WB − GIT	N/A	0.00	2
BDE 99	Lake charr	12	0.015	56	112	0.013	1.1 dw	WB −(L & GIT)	87 ± 11	0.31	3
BDE 99	Lake charr	12	0.015	56	112	0.015	10.8 dw	WB −(L & GIT)	346 ± 173	0.42	3
BDE 100	sole	19	0.008	84	149	0.008	93.1 ww	M + L	62 ± 24	0.14	3
BDE 100	Lake charr	12	0.015	56	112	0.013	1.1 dw	WB −(L & GIT)	63 ± 17	0.42	3
BDE 100	Lake charr	12	0.015	56	112	0.015	6 dw	WB −(L & GIT)	173 ± 173	0.53	3
BDE 183	Zebrafish	26	0.020	42	14	NR	86700 ww	WB	10	0.09	4
BDE 183	Zebrafish	26	0.020	42	14	NR	635.8 ww	WB	15	0.09	4

(continued)

PBDE Congener	Species	Temp. (°C)	Feeding rate[a] (g/g BW/d)	Exposure Duration (d)	Depuration (d)	Growth rate constant (/d)	Food concentration (ng/g)[b]	Tissue analyzed[c]	Half-life ± SE (d)	Assimilation efficiency	References[d]
BDE 183	Lake charr	12	0.015	56	112	0.013	2.1 dw	WB −(L & GIT)	69 ± 14	0.23	3
BDE 183	Lake charr	12	0.015	56	112	0.015	15.8 dw	WB −(L & GIT)	346 ± 173	0.33	3
BDE 209	Lake charr	12	0.015	56	112	0.015	27.5 dw	WB −(L & GIT)	26 ± 5	0.05	3
BDE 209	Rainbow trout	12	0.010	150	0	0.003	940 ww	WB	N/A	0.04	5
BDE 209	sole	19	0.008	84	149	0.008	184 ww	M + L	43 ± 8	0.01	1
BDE 209	Zebrafish	26	0.020	42	14	NR	69055 ww	WB	6.5	0.01	4
BDE 28	Common carp	22	0.033	60	100	0.009	96 ww	WB − GIT	36.5 ± 17	0.20	2
BDE 28	Lake charr	12	0.015	56	112	0.015	12.6 dw	WB −(L & GIT)	58 ± 10	0.53	3
BDE 28	Lake charr	12	0.015	56	112	0.013	1.3 dw	WB −(L & GIT)	NR	0.48	3
BDE 28	sole	19	0.008	84	149	0.008	84.4 ww	M + L	86 ± 27	0.16	1
BDE 28	Zebrafish	26	0.020	42	14	NR	313 ww	WB	23	0.97	4
BDE 28	Zebrafish	26	0.020	42	14	NR	28076 ww	WB	25	1.20	4

[a] Feeding rate = g-ww-food/g-ww-body weight (BW) fish/day

[b] dry wt (dw) or wet wt (ww)

[c] Tissue analyzed: *M* muscle, *L* liver, *GIT* gastrointestinal tract, *WB* = whole body

[d] References: 1—Munschy et al. (2011); 2—Stapleton et al. (2004); 3—Tomy et al. (2004); 4—Nyholm et al. (2009); 5—Stapleton et al. (2006)

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