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Estimation of omega-3 fatty acid (EPA + DHA) intake from Lake Ontario fish based on provincial consumption advisories



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ABSTRACT

Elevated contaminant levels in the North American Great Lakes have resulted in restrictive fish consumption advisories. Avoiding the risks associated with fish consumption may also decrease the intake of omega-3 fatty acids, eicosapentaenoic (EPA; 20:5n-3) and docosahexaenoic (DHA; 22:6n-3) acids; compounds that have been associated with human health benefits. We evaluated whether eating Lake Ontario fish, following the consumption advisories, would result in a sufficient intake of EPA + DHA. Fatty acids and contaminants known to be elevated in fish muscle tissue were analyzed for 282 Lake Ontario fish, representing 21 species. Salmonids had the highest EPA + DHA content among the analyzed species, but the calculated EPA + DHA intake for humans following the consumption advisory was not the highest due to the stringent advisory reflecting elevated contaminant levels, specifically polychlorinated biphenyls. The intake of EPA + DHA from lean fish (with correspondingly lower EPA + DHA contents) could reach values comparable to fatty fish because of the less stringent advisories. In general, the intake of EPA \pm DHA was more influenced by the consumption advisory than the EPA \pm DHA content. We suggest that final advisories should be formulated such that they promote maximizing EPA + DHA intake. We found that people, who generally do not consume large amounts of fish from Lake Ontario, may benefit from choosing salmonids, such as lake whitefish, which maximizes the EPA + DHA intake per meal. People who frequently consume Lake Ontario fish may benefit more from choosing panfish like black crappie or yellow perch. Crown Copyright © 2017 Published by Elsevier B.V. on behalf of International Association for Great Lakes Research. All rights reserved.

Introduction

Fish accumulate a wide variety of toxins. In the Laurentian Great Lakes, the substances of concern include polychlorinated biphenyls (PCBs), mercury, dioxins, and pesticides (Bhavsar et al., 2007, 2008, 2010). PCBs and mercury are currently the main contaminants causing fish consumption advisories in the Great Lakes (NYDH, 2016; OMOECC, 2015). The concentrations of PCB peaked in the 1960s and early- to mid-1970s, and began to decline in the late 1970s after the production of PCBs was banned in North America (Baumann and Whittle, 1988). PCB concentrations in Lake Ontario fish have declined by as much as an order of magnitude and continue to decrease today, albeit more gradually (Bhavsar et al., 2007; Carlson et al., 2010; Visha et al., 2015). Continued research on PCB toxicology through the 1980s to 2000s improved the general knowledge on adverse effects and associated concentrations of PCB, which resulted in about 20 times lower advisory benchmarks (OMOECC unpublished data). As such, current PCB concentrations are still elevated enough for authorities in Canada and the USA to issue consumption advisories for many Lake Ontario fish, and decline of PCB in fatty fish below the unrestricted advisory benchmarks may be decades away (OMOECC, 2015; NYDH, 2016; Kaur et al., 2012). Mercury is a potent neurotoxicant, affecting the central nervous system as well as the cardiovascular, renal and immune systems (Zahir et al., 2005). High mercury concentrations are mainly found in large predatory fish, but in contrast to highly lipophilic PCBs, mercury also accumulates in lean fish (Bhavsar et al., 2010). Mercury concentrations are declining in lake trout but appear to be unchanged in walleye in Lake Ontario (Bhavsar et al., 2010; Visha et al., 2015). Other contaminants, such as dioxins and furans, toxaphene, mirex, and photomirex are also monitored, but they typically do not result in consumption advisories for Lake Ontario fish (Bhavsar et al., 2011; Gandhi et al., 2014, 2015; OMOECC, 2015; NYDH, 2016).

Inadequate fish consumption may have significant health effects such as a higher incidence of cardiovascular disease (Kris-Etherton et al., 2002). Risks of perinatal mortality, growth retardation, child cognitive deficits and reduced immune function have also been linked with insufficient fish consumption (Kris-Etherton et al., 2002; Golden et al., 2016). The protective role of fish consumption against cardiovascular disease has been mainly attributed to high levels of eicosapentaenoic (EPA) and docosahexaenoic acid (DHA). Consequently, nutritional guidelines emphasize the importance of fish in the human diet

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(for a review see Kris-Etherton et al., 2009). The recommended intake of EPA + DHA varies depending on the regulatory body, but is typically in the range of 250–500 mg/day for the general population and which equates, depending on the type of fish consumed, to a few fish meals per week (Kris-Etherton et al., 2009). Higher intake of omega-3 fatty acids, specifically DHA, is recommended for children and pregnant women (Kris-Etherton et al., 2009). Health Canada (2009) recommends that women should continue to eat fish during pregnancy, but should also pay attention to fish consumption advisories and choosing fish species that are low in contaminants (Health Canada, 2009).

The amount of EPA + DHA in fish is highly variable (Budge et al., 2002; Hixson et al., 2015; Colombo et al., 2016). Recent studies on EPA + DHA contents in fish from the Great Lakes have emphasized that more data are needed to characterise the variability in fish EPA + DHA contents (Turyk et al., 2012; Neff et al., 2014; Williams et al., 2014). Algae are the main producers of EPA and DHA in aquatic environments (Taipale et al., 2013; Galloway and Winder, 2015; Strandberg et al., 2015a). These molecules are subsequently transferred up in the food chain to fish (Müller-Navarra et al., 2000; Strandberg et al., 2015b); and, as such, the EPA + DHA content in fish flesh is strongly dependent on dietary intake (Tocher, 2010). Various environmental factors, including climate change, affect the production, distribution, and transfer of EPA and DHA in the food chain, potentially influencing the EPA + DHA content in fish (Brett and Müller-Navarra, 1997; Strandberg et al., 2015b, 2016; Hixson and Arts, 2016; Taipale et al., 2016). Temporal variation, due to the reproductive cycle, over-wintering strategies, and resource availability have also been shown to affect fatty acid composition in fish (Guler et al., 2008; Røjbek et al., 2014). Additionally, fatty acid composition varies according to the lipid content and size of the fish (Budge et al., 2002; Strandberg et al., 2017).

The contaminant levels in Great Lakes' sport fish (i.e., species that are regularly caught and consumed by anglers) are routinely monitored, and the data are used to issue species- and lake-specific consumption advisories (OMOECC, 2015; NYDH, 2016). However, comprehensive studies on both the risks and benefits of eating Great Lakes fishes, with specific recommendations for maximizing the benefits while minimizing the risks are scarce (Turyk et al., 2012; Neff et al., 2014; Williams et al., 2014). The World Health Organization (WHO), and Food and Agriculture Organization of the United Nations (FAO) recommend member states to improve existing databases on nutrients and contaminants in fish, as well as develop communication strategies to minimize the risks and maximize the benefits of fish consumption (FAO/WHO, 2011). In the current study, we investigated if consumption of fish from Lake Ontario can provide the recommended intake of EPA + DHA when the advisories (assigned by the Ontario Ministry of the Environment and Climate Change, OMOECC) are followed. We further evaluated how size- and species-selective fish consumption advisories may be applied so as to increase the relative benefits while, at the same time, decrease the risks associated with fish consumption.

Material and methods

Fish samples (n = 245) were collected from various locations in Lake Ontario between late April and early November in 2012, 2013 and 2014, and analysed for fatty acid and contaminant levels in 2016. We supplemented our data with previously analyzed samples (n = 37) collected in 2008–2009 (analyzed in 2010). Ontario Ministry of the Environment and Climate Change (OMOECC), in partnership with Ontario Ministry of Natural Resources and Forestry, collected fish using a variety of field methods including gill netting and electrofishing. The length and weight of the fish were measured, after which fish were kept on ice for the duration of transport to the laboratory, and where fish were filleted and stored at $-20\,^{\circ}\mathrm{C}$ until homogenisation. Skinless fish fillets were thawed and ground using a Buchi grinder, and a subsample was stored at $-20\,^{\circ}\mathrm{C}$ before the contaminant and fatty acid analyses. Details on the fish length, weight, and gender of the fish, as

well as sampling locations and season are provided in (Electronic Supplementary Material (ESM) Tables S1 and S2).

Fatty acid analysis

Fish muscle tissues were freeze-dried, and the % moisture was calculated as follows: (fresh weight — dry weight) / fresh weight (ESM Table S3 gives the species-specific means of moisture %). Moisture factor (MF) was calculated from the moisture fraction (MF = 1 - moisture percentage), and used to convert fatty acid mass ratios per dry weight to wet weight. Freeze-dried muscle tissues were homogenized by grinding with pestle and mortar in liquid nitrogen. Lipids were extracted twice with chloroform:methanol (2:1 by volume), modified from Folch et al. (1957), and non-lipid components were removed by washing with 0.88% KCl. Organic phases were pooled and the solvent was evaporated to dryness under N2 stream after which hexane was immediately added to the samples. Fatty acids were transmethylated using methanolic sulfuric acid (1%) as a catalyst and heating the samples at 90 °C for 90 min. The FAME were extracted to hexane, concentrated to known volume (750 µl) and analyzed with a gas chromatograph (Shimadzu GC-2010 plus) equipped with a flame ionisation detector. We used SP-2560 column (100 m \times 0.25 mm \times 0.2 μ m, Agilent). Helium was used as the carrier gas with average flow 1.12 ml min⁻¹, with split injection (50:1) and the following temperature program: the initial temperature 140 °C was maintained for 5 min, after which the temperature was increased at the rate of 4 °C min⁻¹ to 240 °C that was maintained for 15 min. Tricosanoic acid (23:0, Nu-Chek Prep Inc.; catalog# N-23-M) was used as internal standard, and standard fatty acids methyl ester mix (Nu-Chek Prep Inc.; catalog# GLC-68F) was used for the calibration curve and quantification of FAMEs. For peak identification we also used 37 component FAME mix (Sigma-Aldrich; catalog# 47885-U). The mass ratios of fatty acids were calculated per 100 g of tissue (w.w.), additionally the EPA + DHA content was calculated per meal using 8 oz. or 227 g (w.w.) meal size utilized by OMOECC for a 70 kg adult (OMOECC, 2015).

Contaminant analyses

Fish muscle tissues were analysed for a variety of contaminants including PCB, mercury, dioxins/furans and selected organochlorine pesticides using previously described accredited methods (Bhavsar et al., 2007, 2010; Gewurtz et al., 2011; Gandhi et al., 2015). Instead of doing risk-benefit analysis of separate contaminants, we chose to use advisories, because they combine information on several contaminants and thus provide a wider estimate of the risks associated with fish consumption. The consumption advisories used in this study were developed, and used by the OMOECC (Bhavsar et al., 2011; Gandhi et al., 2017). Briefly, concentrations were calculated at 5-cm fish size intervals from a power series regression of the fish length against contaminant concentrations for each location/ species/contaminant combination, and these concentrations were standardised to various lengths and compared against the benchmarks to derive recommended meals/month for each 5-cm size category. Then, for each location, species, and 5-cm size interval, the most conservative number of advised meals/month was selected considering the risk from all analyzed contaminants (Bhavsar et al., 2011).

Calculation of risks and benefits

The risk of fish consumption is expressed as the maximum number of fish meals that can be safely consumed per month based on contaminant concentrations. Consumption advisories were assigned for each species; for both the sensitive population (i.e., women of childbearing age and children under the age of 15) and the remaining general population. The mean EPA + DHA content (mg) in a fish meal (227 g or 8 oz. of fish fillet) was multiplied by the consumption advisory (i.e., the number of fish meals that is consider safe to consume in a month) to derive the average EPA + DHA (mg) intake per month if the species is

consumed at the advised maximum number of meals. The value was then divided by 30 to have a daily intake estimate.

Various health organizations have issued guidelines and recommendations for adequate intake of EPA + DHA; these guidelines were used to evaluate whether hypothetical consumers, following the consumption advisories, would achieve levels of EPA + DHA sufficient to realize the benefits of fish consumption. The recommended intake levels of EPA + DHA vary depending on the organization and country (for a review see Kris-Etherton et al., 2009). In the United States the Institute of Medicine provide recommendations for Adequate Intake (AI) of α -linolenic acid, but not for EPA and DHA (Institute of Medicine, 2005). The American Dietetic Association and Dietitians of Canada recommended 500 mg EPA + DHA a day, provided by two serving of fatty fish per week (Kris-Etherton and Innis, 2007). The World Health Organization recommends one to two serving of fish per week: each serving should provide the equivalent of 200–500 mg EPA + DHA (WHO/FAO, 2003). Similarly the American Heart Association recommends two servings of fish per week (Kris-Etherton et al., 2002) We chose to use a daily intake of 250 mg as a conservative benchmark for adequate intake of EPA + DHA; i.e. a compromise between variable recommendations.

Because only one sample of redhorse sucker and smallmouth bass was analysed, data for these two species are presented, but were excluded from the discussion due to the poor sample sizes. Possible influences of sampling season and spatial differences within Lake Ontario on the contaminant and EPA + DHA levels were considered by comparing fish collected from different locations and times. Despite these efforts, there were limitations on the extent of seasonal and geographic coverage preventing a comprehensive analysis.

We present an overall species-specific evaluation of the risk and benefits of consuming 21 species of sport fish from Lake Ontario. Size-specific differences in monthly intake of EPA + DHA were further evaluated for six species, with adequate sample size, representing both lean (northern pike, walleye, yellow perch) and fatty (brown trout, Chinook salmon, lake trout) fish. The selected species are also among the favorite species for human consumption.

Statistical analyses

Non-metric multidimensional scaling (NMDS) was used to visualize the distribution of EPA + DHA, PCB and mercury in fish. In total 256

samples containing contaminant concentrations above the detection limits were included in the analysis. The data was $\log(x+1)$ transformed prior to the analysis, and Bray-Curtis similarity was used for the resemblance matrix. For clarity, the samples were labelled based on the fish order instead of species.

We investigated which of the two factors used in the maximum intake calculations (the consumption advisory or the EPA + DHA content in a fish meal) was more important in determining the EPA + DHA intake per month. Regression analysis was conducted using both the consumption advisory and EPA + DHA content as independent variables and the maximum monthly EPA + DHA intake as a dependent variable for each species. Spearman correlation coefficient between EPA + DHA and total FA content was analyzed across all samples to evaluate if the fattiness of the flesh is related to the amount of EPA + DHA. Bootstrapping (1000 bootstrap iterations) was applied to estimate the 95% confidence interval (biased corrected) for each correlation coefficient.

The size-specific differences in EPA + DHA intake for northern pike, walleye, yellow perch, brown trout, Chinook salmon, and lake trout) were evaluated by plotting the intake of each fish (calculated from fish specific EPA + DHA content and advisory) against the length. Spearman correlations and 95% confidence intervals were also calculated.

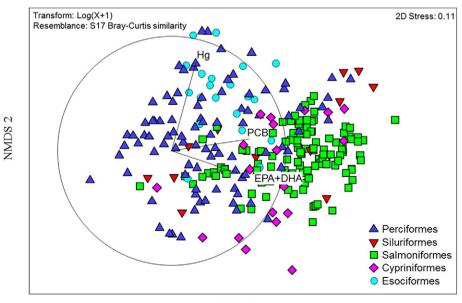
The coefficient of quartile dispersion of EPA + DHA contents (mg 100 g $^{-1}$) was also calculated for different species (Eq. (1)), to evaluate the within species variability in EPA + DHA content. Specifically we wanted to study if the relative variability of EPA + DHA content differed between lean and fatty fish.

Coefficient of quartile dispersion =
$$\frac{Q_1 - Q_3}{Q_1 + Q_3}$$
 (1)

where Q_1 is the 1st quartile, and Q_3 is the 3rd quartile of EPA + DHA content (mg 100 g⁻¹) in different species. Species that had only one sample were omitted from the calculation.

Results

The content of EPA + DHA and contaminants (PCB and mercury) in the muscle tissue was highly taxa-specific (Fig. 1. And ESM Table S4). Salmoniformes and channel catfish had the highest PCB concentrations, while Esociformes (northern pike) had the highest mercury



NMDS 1

Fig. 1. Non-metric multidimensional scaling of fish samples, based on the content of EPA + DHA, PCB and mercury. Vectors indicate the strength and direction of correlation of EPA + DHA, PCB and mercury with the dimensions.

Table 1
Mean content (mg 100 g $^{-1}$ w.w. \pm SD) of n-6 PUFA: linoleic (LIN), and arachidonic (ARA), and n-3 PUFA: α-linolenic (ALA), eicosapentaenoic (EPA), and docosahexaenoic (DHA) acids in Lake Ontario fish species. The sum of EPA and DHA (EPA + DHA) is also presented.

Species			n-6 PUI	FA			n-3PUF	A						
Common name	Scientific name	n	LIN		ARA		ALA		EPA		DHA		EPA + D	НА
			Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
Black crappie	Pomoxis nigromaculatus	10	12.8	6.3	49.7	20.4	10.9	7.0	16.7	6.4	105.9	40.0	122.6	46.0
Brown bullhead	Ameiurus nebulosus	4	17.1	11.0	39.7	6.3	17.2	13.8	49.2	25.6	62.4	8.8	111.5	34.3
Brown trout	Salmo trutta	23	155.5	100.8	145.0	72.9	125.9	107.3	188.3	104.4	415.2	226.8	603.5	327.1
Channel catfish	Ictalurus punctatus	9	207.7	299.2	120.4	102.9	104.8	96.5	97.8	82.3	203.1	172.9	300.9	238.5
Chinook salmon	Oncorhynchus tshawytscha	31	86.9	144.5	70.1	63.1	67.8	119.0	78.3	69.7	203.0	97.4	281.3	162.7
Coho salmon	Oncorhynchus kisutch	8	91.0	56.9	61.7	34.2	72.0	41.7	68.6	32.3	206.3	58.4	274.9	89.6
Common carp	Cyprinus carpio	10	108.8	119.8	65.1	45.3	54.3	81.0	66.5	72.4	53.4	58.4	120.0	127.2
Freshwater drum	Aplodinotus grunniens	16	24.4	34.7	71.4	31.2	14.1	21.5	52.8	34.0	56.7	76.8	109.5	108.2
Lake trout	Salvelinus namaycush	21	266.1	213.8	245.5	154.2	220.8	169.5	320.2	214.0	698.7	384.6	1018.9	596.8
Lake whitefish	Coregonus clupeiformis	10	119.1	63.6	95.4	45.6	44.5	21.7	184.1	90.2	309.2	125.0	493.3	190.9
Largemouth bass	Micropterus salmoides	12	11.7	4.0	37.2	5.9	7.0	3.1	19.2	5.0	65.7	16.2	84.9	18.3
Northern pike	Esox lucius	21	10.0	4.4	30.6	8.8	5.8	3.5	25.3	8.7	116.0	24.0	141.4	31.5
Rainbow trout	Oncorhynchus mykiss	19	155.8	97.2	121.3	72.7	122.0	89.4	155.1	97.6	377.8	196.8	532.9	288.5
Redhorse sucker	Moxostoma carinatum	1	42.4	_	32.5	_	24.6	_	150.0	_	101.9	_	251.9	_
Rock bass	Ambloplites rupestris	8	8.6	2.1	35.5	3.8	4.5	2.0	16.1	4.8	96.0	20.2	112.1	23.0
Smallmouth bass	Micropterus dolomieu	1	11.3	_	35.0	_	5.2	_	30.6	_	102.4	_	133.0	_
Walleye	Sander vitreus	33	11.5	7.6	38.8	9.9	8.8	6.3	33.3	10.6	128.5	36.3	161.7	44.0
White bass	Morone chrysops	6	51.0	24.0	92.0	27.0	44.7	23.6	89.7	39.6	187.1	63.1	276.8	102.1
White perch	Morone americana	10	46.5	49.7	74.6	49.5	34.6	30.2	88.4	62.0	87.8	48.4	176.1	97.9
White sucker	Catostomus commersonii	10	50.7	45.5	39.2	8.2	34.2	36.8	77.7	33.3	134.2	23.7	211.9	49.9
Yellow perch	Perca flavescens	19	8.7	5.3	41.5	17.9	4.8	3.6	36.0	14.3	115.6	46.8	151.5	55.9

concentration (Fig. 1). The EPA + DHA content in the muscle was strongly correlated with the fattiness of the fish (i.e., the total fatty acid content of the muscle tissue $r=0.8,\,95\%$ CI: 0.8–0.9. The highest EPA + DHA concentrations were found in lake trout, brown trout, rainbow trout, and lake whitefish, while the lowest concentrations were found in lean fish species, such as the largemouth bass and freshwater drum (Table 1, Fig. 2). Intraspecific specific variation in the EPA + DHA content of the muscle (expressed as coefficients of quartile dispersion) was generally largest in salmonids, although also the common carp had high variability (Fig. 3). The mean n-3/n-6 ratio was >1 for most species (Table 2).

The median consumption advisory for the general population was eight meals per month or higher for brown bullhead, walleye, freshwater drum, smallmouth bass (only one sample), largemouth bass,

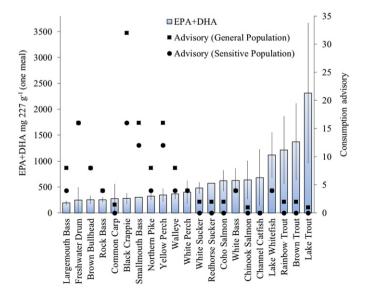


Fig. 2. Species-specific means and standard deviations of EPA + DHA concentrations per meal (227 g), and the median consumption advisories (number of 227 g meals per month) for Lake Ontario fish. The consumption advisories are presented for both the general (triangles) and sensitive population (dots).

northern pike, yellow perch, and black crappie (Fig. 2, ESM Table S5). In general, the median recommended number of meals was lower for fatty fish in comparison to leaner fish although there were exceptions (Fig. 2). The most notable exception was the common carp; a lean fish that had low recommended number of meals per month (2 meals per month) for the general population and *do not eat* advisory for the sensitive segment of the population. The recommended meals were the same for both the general and sensitive population for brown bullhead, channel catfish, freshwater drum, lake whitefish, rock bass, white perch, and white bass; for other cases, the recommended meals were lower for the sensitive population.

 $Maximum\ EPA\ +\ DHA\ intake\ and\ consumption\ advisory$

Fish that contained the highest amount of EPA + DHA also had the lowest number of advised meals, but species with low EPA + DHA content had variable advisory (Figs. 2 and 4A). The maximum EPA + DHA intake per month (mg month⁻¹) varied by species; and, in general, correlated with the recommended maximum number of meals (Fig. 4B).

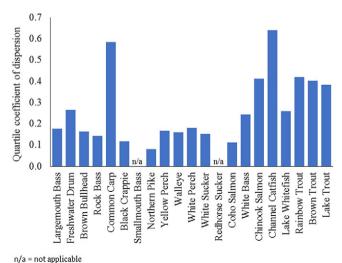


Fig. 3. Coefficients of quartile dispersion of EPA + DHA (mg 100 g $^{-1}$) for Lake Ontario fish.

 Table 2

 Total fatty acids content (Σfatty acids, expressed as mg 100 g $^{-1}$ w.w. \pm SD), and proportions (% of total fatty acids) of saturated fatty acids (ΣSFA), monounsaturated fatty acids (ΣMUFA), polyunsaturated fatty acids ΣPUFA, n-6 and n-3 polyunsaturated fatty acids (ΣPUFA n-3 and ΣPUFAn-6), as well as the n-3/n-6 PUFA and ΣPUFA/ΣSFA ratios.

Species	Σ fatty acids (mg 100 g ⁻¹)		ΣSFA		ΣMUFA		ΣΡυγΑ		ΣPUFA n-6		ΣPUFA n-3		n-3/n-6 PUFA		ΣPUFA/ΣSFA	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
Black crappie	487.0	209.3	31.9	1.9	19.4	3.1	48.7	2.1	16.2	1.1	32.5	2.2	2.0	0.2	1.5	0.1
Brown bullhead	742.2	403.6	30.4	0.4	31.5	10.6	34.0	9.7	11.4	4.2	22.5	5.6	0.5	0.1	1.1	0.3
Brown trout	4530.3	2355.4	26.0	2.5	43.2	4.6	30.0	4.8	9.4	2.0	20.6	3.5	1.8	0.8	1.2	0.3
Channel catfish	4181.6	3999.0	25.9	2.5	45.2	10.2	28.8	8.5	12.0	3.1	16.8	6.3	1.4	0.4	1.1	0.3
Chinook salmon	2192.8	2355.7	27.8	3.1	27.6	11.3	43.5	14.4	11.9	2.7	31.7	12.5	2.4	1.2	1.6	0.7
Coho salmon	1632.6	861.6	26.1	3.0	33.8	6.4	40.1	8.2	11.9	1.0	28.2	7.7	2.4	0.6	1.6	0.5
Common carp	1930.9	2155.5	26.9	3.6	43.4	5.8	26.6	9.0	14.6	6.1	12.0	3.9	0.9	0.3	1.0	0.5
Freshwater drum	951.8	1568.6	30.3	2.6	27.4	11.3	42.3	9.8	20.4	6.5	22.0	4.1	1.2	0.4	1.4	0.3
Lake trout	6847.1	4400.3	22.9	1.7	44.1	6.2	33.0	6.0	10.3	0.9	22.7	5.5	2.2	0.4	1.4	0.3
Lake whitefish	3084.4	1270.5	25.1	1.7	44.8	6.0	30.1	6.1	9.3	1.6	20.7	6.1	2.3	0.9	1.2	0.3
Largemouth bass	347.9	88.8	28.8	2.4	20.4	3.8	50.2	4.5	17.9	2.6	32.4	3.4	1.6	0.6	1.8	0.2
Northern pike	368.8	109.5	26.7	1.7	14.6	3.4	58.2	3.7	14.0	2.6	44.2	3.9	2.9	1.3	2.2	0.2
Rainbow trout	3106.3	2133.6	26.5	3.2	33.7	6.6	39.9	7.6	12.4	2.2	27.5	5.9	2.2	0.3	1.5	0.4
Redhorse sucker	1115.3	-	27.4	-	30.2	-	36.6	-	7.5	-	29.1	-	0.3	-	1.3	-
Rock bass	369.2	79.8	28.9	1.9	16.3	2.4	53.9	4.7	15.9	2.9	37.9	3.0	1.5	1.0	1.9	0.3
Smallmouth bass	505.9	-	27.0	-	23.3	-	47.6	-	12.9	-	34.7	-	0.4	-	1.8	-
Walleye	514.4	203.6	28.4	2.4	21.2	6.6	50.3	5.1	13.0	1.8	37.3	4.8	2.9	0.6	1.8	0.2
White bass	1390.1	503.9	26.0	2.0	35.3	1.8	38.7	2.6	13.2	1.2	25.5	1.5	1.9	0.1	1.5	0.2
White perch	1253.8	1061.0	28.6	3.1	37.2	7.0	34.2	5.7	12.4	1.1	21.9	4.8	1.8	0.3	1.2	0.2
White sucker	1123.2	667.6	26.7	1.8	31.6	9.1	39.4	10.0	10.9	2.0	28.5	8.4	1.6	1.2	1.5	0.4
Yellow perch	443.6	157.9	29.4	2.2	16.1	3.3	54.3	4.5	15.3	1.7	39.0	4.1	2.2	0.9	1.9	0.3

For the general population, the maximum monthly EPA + DHA intake (mg) depended more on the consumption advisory ($r^2 = 0.619$, P < 0.001) (Fig. 4B), than the EPA + DHA content of the fish ($r^2 = 0.037$, P = 0.404) (Fig. 4C). The fish that had the highest concentration of EPA + DHA did not yield the highest monthly intake because of the stringent consumption advisory (Table 3). These fish will not be an important source of EPA + DHA because of elevated contaminant levels. This is particularly evident in scenarios for the sensitive population (ESM Table S5). Most fatty fish with high EPA + DHA content would have a consumption advisory of no >2 meals per month (general population), or *do not eat* (for the sensitive population).

Size-specific differences in monthly intake of EPA + DHA

Across the six species used to evaluate the size-specific difference, the monthly intake of EPA + DHA effectively decreased with increasing fish length (Fig. 5, see ESM Table S6 for the correlation coefficients). The decline was predominately due to more stringent consumption advisories for larger fish. The length dependent differences in EPA + DHA content in each species and size-class are presented in ESM Table S5). This was particularly evident in the case of the sensitive population. For the general public, the strength of the correlation between the monthly maximum intake of EPA + DHA and fish length varied, and was strong in Chinook salmon ($r_s = -0.663$, P < 0.001, 95% CI: -0.87to -0.297), walleye ($r_s = -0.714$, P < 0.001, 95% CI: -0.878 to -0.449), and brown trout ($r_s = -0.513$. P < 0.001, 95% CI: -0.806 to -0.056). For the sensitive population, strong negative correlation was found in Chinook salmon (r = -0.68, P < 0.001, 95% CI: -0.831 to -0.427), walleye (r = -0.881, P < 0.001, 95% CI: -0.968 to -0.701, and also in lake trout (r = -0.585, P < 0.001, 95% CI: -0.79 to $-\,0.335$). In all the cases, the correlation was systematically negative (even if not significant or strong) (Fig. 5).

Discussion

The maximum intake of EPA + DHA from Lake Ontario fish was constrained by the maximum recommended meals based on contaminant levels in fish, not by the EPA + DHA content in the fish. This is logical because even if a fish species has a high EPA + DHA content, it provides little benefits if the advisory recommends a low number of

meals (typically *do not eat* or only one or two meals per month). The consumption advisories for Lake Ontario fish are mainly driven by lipophilic PCBs, while mercury is a secondary driver (Bhavsar et al., 2011; Gandhi et al., 2014). As such, lean fish have less stringent consumption advisories, but also have lower EPA + DHA content, as EPA + DHA content is strongly dependent on the total fat content of the fillet (Strandberg et al., 2017).

On average, the recommended monthly intake of EPA + DHA would not be achieved from consuming any species of Lake Ontario fish, if the consumption advisories were followed. This indicates that reduction of contaminant levels in fatty fish, and subsequent less stringent consumption advisories, would be needed to improve the intake of EPA + DHA from Lake Ontario fish. If the size of the fish is taken into consideration, consumption of smaller sized fish with less stringent advisory could provide the recommended monthly intake of EPA + DHA for the general population. The recommended species for consumption to maximize the intake of EPA + DHA, while following the species-specific consumption advisories, varied based on fish consumption tendencies. People who consume several fish meals in a month should focus on eating lean fish species that have lenient consumption advisories. The overall monthly intake of EPA + DHA from lean fish will be higher when the maximum (or near maximum) number of meals are consumed. However, members of the general public that do not regularly eat fish would benefit more by consuming fatty fish, such as salmonids. Although only few meals are advised to be consumed in a month, the high EPA + DHA content in salmonids maximizes the EPA + DHA intake per

For the sensitive population, the more stringent consumption advisory together with the recommended greater dietary intake of EPA + DHA result in fewer options for meeting monthly EPA + DHA intakes from Lake Ontario fish. In general, fatty fish from the Lake Ontario should not be consumed by this group with the exception of white bass and lake whitefish. For these two species, the median advisory was the same for both the general and sensitive population. The consumption advisories of these species likely reflect the fact that these species feed primarily on lower trophic level resources and thus accumulate less PCBs and mercury, that biomagnify in the food web (Rasmussen et al., 1990; Cabana et al., 1994). Lake whitefish diet consists of zooplankton, macroinvertebrates and molluscs (Pothoven and Nalepa, 2006), and the analyzed white bass (< 40 cm) were not large

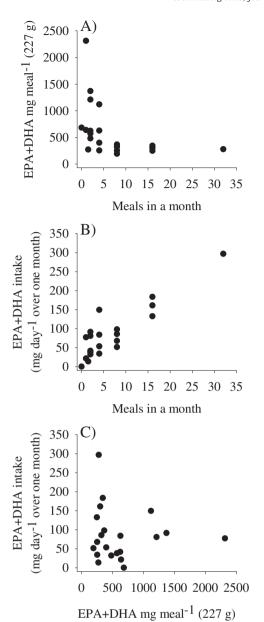


Fig. 4. The species-specific median consumption advisories for general population (i.e. the maximum number of meals recommended to be consumed in a month), and means of EPA + DHA content, and intake across all the fish species. A) The EPA + DHA content in a fish meal (i.e. 227 g w.w.) is related to the consumption advisory (see text for details). B) Fish that contained the highest amount of EPA + DHA also had the most stringent consumption advisories. C) The fish EPA + DHA content is not a good predictor for EPA + DHA intake when advisories are followed.

enough to be predominantly piscivores. It has been estimated that small sized white bass feed mainly on zooplankton and insects, and switch to mainly piscivorous diet at the length of about 50 cm (Olsen, 1996). The sensitive population may also consume the smaller-sized salmonids, but even in these cases, the maximum monthly intake of EPA + DHA would not reach the recommended levels, with the exception of small-sized Chinook salmon.

The recommended EPA + DHA intake varies by agency (Kris-Etherton et al., 2009). Different units used also confound the recommendations; for example, some organizations suggest that that EPA + DHA should contribute certain percentages of daily calories and/or fats, instead of indicating specific amounts. Similarly, some authorities recommend that EPA + DHA intake should be scaled to body weight. In addition, higher recommendations may be assigned for

children and pregnant women mainly because DHA is needed for fetal and neonatal neurological development (Rogers et al., 2013). However, this group is also considered more sensitive with respect to contaminant exposures, posing a special challenge in meeting EPA and especially DHA requirements from consuming Lake Ontario fish.

Most of the risk-benefit studies of fish consumption have been on marine fish, with the general conclusion that health benefits and risks are highly dependent on the fish type (Domingo et al., 2007; Gribble et al., 2016). Additional aspects include the frequency of fish consumption and fishing location (Domingo et al., 2007; Gribble et al., 2016). Results from freshwater fish in the current study are in accordance with the generalizations from the marine environment. A similar study to this one has been previously conducted for Lake Erie fish, using the same consumption advisory thresholds (Neff et al., 2014). The authors concluded that Lake Erie fish can be a good source of omega-3 fatty acids for humans; however, similar to the current study, stringent consumption advisories limited the intake of EPA + DHA, and the upper dietary recommendations of EPA + DHA were not met (Neff et al., 2014). On average, the consumption advisories were more stringent for Lake Ontario compared to Lake Erie fish (Neff et al., 2014).

Consumption advisory

The consumption advisories for most species were related to the fattiness of the muscle tissue and thereby the EPA + DHA content. All the fatty fish, specifically salmonids, with high EPA + DHA content in the muscle had stringent advisories, while lean fish with lower EPA + DHA can generally be consumed more frequently. The major contaminants responsible for the consumption advisory in Lake Ontario fish were PCBs for fatty fish such as salmonids, and mercury for lean fish, such as percids. This is a typical pattern for Great Lakes fish (Bhavsar et al., 2011; Gandhi et al., 2014; OMOECC, 2015). PCB is highly lipophilic compound that accumulates in fish lipids (Bruggeman et al., 1984). There were a few exceptions, most notably the common carp, which has a very stringent advisory, due to elevated PCB levels, for a relatively lean fish. Common carp is a bottom-dwelling fish feeding on benthic organisms, and high degree of interaction with the sediment results in a higher exposure to historically deposited PCBs (Pérez-Fuentetaja et al., 2010).

It has been suggested that the current consumption advisories may not be adequate because of the possible additive effects of chemical mixtures in fish (Gandhi et al., 2017). The consumption advisories are assigned based on the most restrictive contaminant, but a wide array of chemicals is commonly found in fish. In addition to PCBs and mercury, dioxins and furans, toxaphene, as well as mirex and photomirex have been detected in Lake Ontario fish (Bhavsar et al., 2011; OMOECC, 2015). Chemical mixtures may interact additively or synergistically, potentially contributing to the negative health effects for humans (Carpenter et al., 2002). Data on the health effects of chemical mixture are inconclusive, but additive effects have been reported for PCBs and mercury, although not in all cases (Bemis and Seegal, 1999; Piedrafita et al., 2008). Such additive effects could lead to even more stringent consumption advisories (Gandhi et al., 2017), which would be expected to further decrease the importance of Lake Ontario fish as a source of EPA + DHA for humans.

EPA + DHA in lean vs. fatty fish

The EPA + DHA contents in the muscle were strongly dependent on the total fatty acid content, which is related to the lipid storage strategies in different species. Most teleost fish store lipids as triacylglycerols around internal organs (mesenteric fat), in the muscle, and in the liver (Sheridan, 1988). For salmonids, and other fatty fish, the main lipid storage sites are the muscle tissue and mesenteric fat, while in lean fish the main lipid storage depot is the liver, and, to lesser degree the mesenteric fat (Dos Santos et al., 1993). Muscle is not an important storage site of

Table 3 Species-specific approximate EPA + DHA content per meal in relation to advisory. These broad estimates and should be used with caution, detailed data on fish specific EPA + DHA content, advisory and EPA + DHA intake can be found in the ESM. Cells with horizontal stripes indicate those species/size classes that provide the highest intake of EPA + DHA when the maximum advisory is followed (daily intake in this category varies from about 200–750 mg). Downward diagonal striped cells indicate those species/size classes that provide 100–200 mg EPA + DHA day $^{-1}$, and vertical stripes indicate those species/size classes that provide <100 mg EPA + DHA day $^{-1}$, when maximum advisory is followed. Dotted area at bottom of the table marks species/size classes with do not eat advisory. *Data for one fish only.

	EPA+DHA content in a meal (227 g or 8 oz)									
Advisory	< 250 mg	250 – 500 mg	500 – 1000 mg	> 1000 mg						
>16		Black crappie Yellow Perch (< 20 cm)	Chinook Salmon (< 35 cm)							
12 – 16	Freshwater Drum (< 40 cm)	Northern Pike (< 70 cm) Smallmouth Bass* Yellow Perch (> 20 cm) Walleye (< 50 cm)								
4-8	Largemouth Bass Brown Bullhead	Chinook Salmon (< 50 cm) Northern Pike (> 70 cm) Rock Bass White Perch Walleye (> 50 cm)	White bass Channel Catfish (< 35 cm)	Lake Whitefish Lake Trout (< 50 cm)						
1-2	Common Carp (< 80 cm) Freshwater Drum	White Sucker	Chinook Salmon (> 50 cm) Coho Salmon Redhorse Sucker*	Brown Trout (< 70 cm) Lake Trout (50-70 cm) Rainbow Trout						
Do not eat		Common Carp (> 80 cm)	Channel Catfish (> 35 cm)	Brown Trout						

lipids in lean fish. This explains the observed differences in EPA + DHA content between different types of fish. Our study indicates that the intraspecific variation of EPA + DHA contents may be larger for fatty fish than for lean fish (Pantazopoulos et al., 2013; Neff et al., 2014; Williams et al., 2014), most likely reflecting differences in the muscle lipid deposits (Sheridan, 1988; Dos Santos et al., 1993). Muscle lipid storage in fish tissues fluctuate according to the nutritional state and reproductive status, particularly in females. Furthermore, it may be hypothesized that dietary fatty acids have a stronger impact on the muscle fatty acids in fatty fish than in lean fish, and hence the spatial and temporal differences in EPA + DHA content would be bigger for salmonids than, for example, percids like walleye and yellow perch. The common carp was a notable exception in this pattern; it demonstrated a high variation in EPA + DHA content for a relatively lean species. General variation due to size, sampling time and location affect muscle lipids also in lean species (Guler et al., 2008; Mráz and Pickova, 2009; Mráz et al., 2012), and contribute to the intraspecific variation in EPA + DHA content. The coefficient of quartile dispersion is based on the 1st and 3rd quartiles of the sample population and is thus robust to outliers. However, if several/all of the samples represent just the ends of the EPA + DHA concentration spectrum, the coefficient of quartile dispersion would assign a high value. The muscle lipid content in common carp has been noted to vary between 0.8 and 4.5% (Guler et al., 2008; Mráz and Pickova, 2009; Mráz et al., 2012). In the current study most of the samples were within this range, although situated at the lower and upper ends of the range, potentially explaining the variation in EPA + DHA content in these carp. More detailed investigation on the relative variability of different types of fish (lean vs. fatty) across Great Lakes (with larger sample size) is needed to verify if these groups show significantly different variability in the EPA + DHA content, Larger intraspecific variation for fatty fish would have direct implications for the sample sizes for biomonitoring purposes.

Size-related differences in maximum monthly EPA + DHA intake

Because of a more stringent consumption advisory, the monthly intake of EPA + DHA decreased with fish size. Indicating that within any of the analyzed species, choosing smaller sized fish will be more beneficial. Large sized fish are not recommended for consumption and thus cannot be important source of EPA + DHA even if the tissue concentrations would be high. Although sample size restricted the number of species used for the evaluation of size-specific differences, both fatty and lean fish were represented. We did not discern a difference in the size-specific trend in EPA + DHA intake between lean and fatty fish. For both groups, any correlation between the variables was either negative or flat.

The mercury levels in fish are known to be strongly correlated with the fish size (Somers and Jackson, 1993; Gewurtz et al., 2011). Larger older predatory fish have longer time to accumulate contaminants, and accordingly the consumption advisory typically becomes more restrictive with fish size (Somers and Jackson, 1993; Gewurtz et al., 2011). The correlation between PCBs and fish size show species-specific variation, with strongest correlations have been found in species that have moderate to high muscle lipid content, such as lake trout and Chinook salmon (Gewurtz et al., 2011). The relationship between PCB and fish size is weaker for lean fish; for instance in yellow perch, the muscle PCB concentration is not related to the length of the fish (Gewurtz et al., 2011). The weak or absent correlation between PCBs and fish length in lean fish can be attributed to the low muscle lipid content. In lean fish,

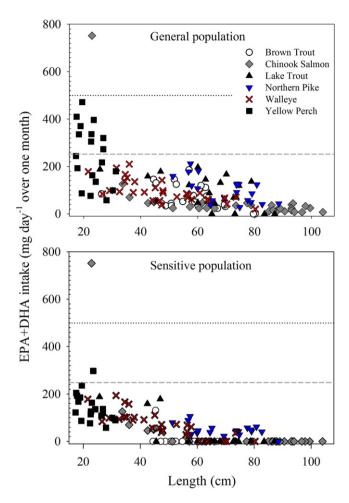


Fig. 5. Correlation between the maximum monthly intake of EPA + DHA (expressed on a daily basis) and the length for 3 salmonids and 3 lean fish species from Lake Ontario: brown trout (n = 23), Chinook salmon (n = 31), lake trout (n = 21), northern pike (n = 21), walleye (n = 33), yellow perch (n = 19). See text for the correlation coefficients. The lines mark two threshold intakes of EPA + DHA (250 mg day $^{-1}$ and 500 mg day $^{-1}$).

the lipophilic PCBs tend to accumulate in more lipid rich tissues, such as liver and gonads (Monosson et al., 2003).

Lipid content and fatty acid composition in fish has also been correlated with fish size (Budge et al., 2002; Breck, 2014). In the current study, lake trout was the only species that exhibited size-specific differences in the EPA + DHA content. Note, that the positive correlation between the fish length and the EPA + DHA content (mg $100\,\mathrm{g}^{-1}$) did not extend to the monthly intake of EPA + DHA because of the stronger positive correlation between contaminants and fish length that resulted in stringent consumption advisories for larger fish.

Other sources of EPA + DHA

In the USA the average consumption rate of finfish in adults (≥21 years) is about 12.8 g day⁻¹ (raw weight, edible portion) (USEPA, 2014). However, most of the consumed fish is store-bought, only about 7% (about 4.2 million) of the adults residing in the Great Lakes Area in USA consumed fish from the Great Lakes (Imm et al., 2005). Avid sport-fishers were the highest consumers of Great Lakes fish (Imm et al., 2005). Lake Ontario does not have significant commercial fishing (OMNRF, 2017), indicating that the consumption advisories are most beneficial for sport-fishers. The EPA + DHA content in store bought fish can be highly variable, depending on the diet and environmental factors (marine, estuarine, freshwater), as well as whether the

fish was cultured or wild (see Kris-Etherton et al., 2002), making it challenging to estimate the relative importance of store-bought fish in relation to Lake Ontario fish.

Conclusion

Fish muscle EPA + DHA content was highly species-specific and the highest contents were found in salmonids and the lowest contents in lean fish such as smallmouth bass and freshwater drum. However, the muscle EPA + DHA content was not a good predictor for the overall monthly intake of EPA + DHA if the consumption advisories were followed. Consumption of Lake Ontario salmonids (all size categories) was restricted to only few meals in a month, which was not enough to provide the recommended dietary intake of EPA + DHA (Fig. 3). If only small-sized fish were considered, the monthly intake reached the dietary guidelines for EPA + DHA for in few species, mainly yellow perch, but also for small-sized walleye and Chinook salmon (Fig. 3) This was mainly due to more lenient consumption advisories for smaller-sized fish. Similarly, the consumption advisory could predict the maximum monthly intake of EPA + DHA across the analyzed species. Recommendations for consuming Lake Ontario fish depend on fish consumption habits. The general population who infrequently consume fish from Lake Ontario would benefit from choosing lake whitefish or lake trout, as these fish will maximize the EPA + DHA intake per meal. Black crappie or yellow perch can also be recommended for people who frequently consume Lake Ontario fish. These species have less stringent consumption advisory, and thus maximizing EPA + DHA intake over a month. The declining trend of major contaminants, including PCBs, in Lake Ontario fish provides an optimistic prospect with respect to the importance of Lake Ontario fish in providing EPA + DHA to humans.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jglr.2017.08.009.

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