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Spatial and length-dependent variation of the risks and benefits of consuming Walleye (*Sander vitreus*)

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ABSTRACT

Restricted fish consumption due to elevated contaminant levels may limit the intake of essential omega-3 fatty acids, such as eicosapentaenoic (EPA; 20:5n − 3) and docosahexaenoic (DHA; 22:6n − 3) acids. We analyzed lake- and length-specific mercury and EPA + DHA contents in Walleye (*Sander vitreus*; Mitchell 1818) from 20 waterbodies in Ontario, Canada, and used this information to calculate the theoretical intake of EPA + DHA when the consumption advisories are followed. The stringent consumption advisory resulted in decreased EPA + DHA intake regardless of the EPA + DHA content in Walleye. Walleye length had a strong impact on the EPA + DHA intake mainly because it was positively correlated with the mercury content and thereby consumption advisories. The highest EPA + DHA intake was achieved when smaller Walleye (30–40 cm) were consumed. The strong relationship between the consumption advisory and EPA + DHA intake enabled us to develop a more generic regression equation to estimate EPA + DHA intake from the consumption advisories, which we then applied to an additional 1322 waterbodies across Ontario, and 28 lakes from northern USA for which Walleye contaminant data are available but fatty acid data are missing. We estimate that adequate EPA + DHA intake (> 250 mg day^{-1}) is achieved in 23% of the studied Ontario lakes, for the general population, when small (30–40 cm) Walleye are eaten. Consumption of medium- (41–55 cm), and large-sized (60–70 cm) Walleye would provide adequate EPA + DHA intake from only 3% and 1% of the lakes, respectively. Our study highlights that mercury contamination, which triggers consumption advisories, strongly limits the suitability of Walleye as the sole dietary source of EPA + DHA to humans.

1. Introduction

Fish consumption provides both significant health benefits as well as potential risks for humans. Adequate intakes of eicosapentaenoic (EPA; 20:5n − 3) and docosahexaenoic (DHA; 22:6n − 3) acids, omega-3, long-chain polyunsaturated fatty acids (LC-PUFA) found in fish, are linked with decreased inflammation, cardiovascular disease, major depression and autoimmune diseases, and also promotes optimal pre- and postnatal development of the brain (Kris-Etherton et al., 2002; Calder, 2015). However, the range of contaminants that may be present in fish can also pose significant health risks (Domingo, 2016). For example, large predatory freshwater fish in the northern hemisphere, often have elevated mercury, specifically methylmercury, levels, which has prompted various national and international agencies worldwide to issue fish consumption advisories (OMOECC, 2015; NYDH, 2016; WHO, 2007). Mercury is a potent toxicant that damages the nervous system, and negatively affects gastrointestinal, renal, and cardiovascular systems (Tchounwou et al., 2003).

When the same food item contributes to both health hazards and positive health outcomes, an overall risk-benefit assessment is necessary to provide balanced consumption advisories to the public (Domingo et al., 2007; Vilavert et al., 2017). The different contaminants (e.g. mercury and PCBs) and nutrients (e.g. EPA and DHA, vitamin D) found in fish have multiple and complex health effects and we still do not have adequate data and knowledge to gain a comprehensive and conclusive understanding of the net outcome of every possible combination of contaminants and nutrients on human health (Mozaffarian, 2009). Whether the risks associated with fish consumption exceed the benefits is still debated, partly due to conflicting results obtained from epidemiological studies (Mozaffarian and Rimm, 2006; Stern, 2007). One reason for the conflicting results is that there is intra- and interspecific variation in both contaminant and EPA + DHA levels in fish (Stern, 2007).

In case of mercury the variation is relatively well established

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(Depew et al., 2013). Fish mercury contents vary as a function of species, age, size (length and body mass), and location (Depew et al., 2013). Local contamination level and environmental conditions, such as the lake water chemistry and catchment characteristics, result in significant spatial variation in fish mercury levels (Evans et al., 2005; Gandhi et al., 2014). The large variation in fish mercury contents are widely acknowledged and thus localized and species-specific monitoring of mercury contents is common and necessary to provide fish consumption advisories for the public (Gandhi et al., 2016). Methylmercury is biomagnified through the food web (Mathers and Johansen, 1985), consequently high mercury levels are often found in large predatory fish (Depew et al., 2013). Although, length-dependent increase in the risk (in terms of mercury) are well documented, much less is known about the benefits (e.g. EPA + DHA). As such, it is still uncertain if the benefits also increase with fish length, possibly compensating for the risks caused by increasing mercury levels.

Interspecific variation in EPA + DHA contents in fish muscle is increasingly well described (Neff et al., 2014; Williams et al., 2017). However, fine-scale information on intraspecific and spatial variability in EPA + DHA contents in freshwater fishes is still fragmented and missing for most species (Turyk et al., 2012; Williams et al., 2017). For selected species that have been more closely studied, variation in EPA + DHA content have been linked with fish size, as well as environmental factors, such as lake water chemistry and catchment characteristics (Strandberg et al., 2016; Williams et al., 2017).

Risk avoidance takes precedence over benefits, hence we here evaluate if recommended EPA and DHA intake from fish (benefits) can be achieved when following the consumption advisories (representing the risks). Our goal was to quantify the variability in the risks versus the benefits associated with eating variably-sized sport fish, Walleye (*Sander vitreus*; Mitchell 1818) collected from temperate and boreal lakes in the Province of Ontario, Canada. Ontario is over 1 million km^2 in area, and our study area represents a range of different environmental conditions, providing a comprehensive overview of risk-benefits of consuming Walleye from northern temperate and boreal areas. Ontario also has an active recreational fishing community with nearly 1.3 million resident and non-resident anglers in 2010 (DFO, 2012). Walleye, a perciform fish common throughout Canada and northern USA, was chosen to represent the predatory fish because it is a preferred species for consumption for recreational anglers and subsistence fishers in North America (Awad, 2006), and is known to contribute to mercury expose to humans, particularly in First Nation communities (Juric et al., 2017). Additionally, little is known about size-specific and spatial variability in Walleye EPA + DHA contents. Our study provides a framework to allow consumers to make informed decisions as to the risks vs benefits associated with eating walleye; a predatory, high trophic level, freshwater fish that often has elevated mercury levels in Ontario lakes.

2. Material and methods

2.1. Fish collections and tissue processing

Walleye samples (n = 216) were collected, using gill nets, from 20 waterbodies located across the Province of Ontario, Canada (Supplement 1) by Ontario Ministry of the Environment and Climate Change (MOECC) in partnership with Ontario Ministry of Natural Resources and Forestry (OMNRF). Weight (g) and total length (cm) of fish were measured before harvesting and subsequently freezing skinless fillets. Fish were separated into three length categories: small (30–40 cm, n = 90), medium (41–55 cm, n = 97) and large (> 60 cm, n = 29). The fillet samples were homogenized using a grinder and stored at − 20 °C prior to the chemical analyses (EPA, DHA, mercury).

2.2. EPA and DHA analysis

Subsample of ground fish fillets was freeze-dried and the moisture % of the tissue was calculated. Moisture factor (MF) was calculated as follows: MF = 1 − moisture %. The MF was subsequently used to convert EPA and DHA mass fractions per dry weight to wet weight. Lyophilized tissue was ground using a mortar and pestle in a small amount of liquid nitrogen. Lipids were extracted twice with chloroform-methanol (2:1 by volume) from the lyophilized tissue (Folch et al., 1957). Prior to extraction an internal standard (5-α-cholestane) was added to each sample. Methyl esters of EPA and DHA were produced by using methanolic sulfuric acid as a catalyst and heating the sample at 90 °C for 90 min. The EPA and DHA were extracted to n-hexane, concentrated, and analyzed with GC (Shimadzu GC-2010 plus) equipped with flame ionization detector. We used a SP-2560 column (100 m × 0.25 mm × 0.2 μm, Agilent). Helium was used as the carrier gas with an average flow of 1.12 ml min^{-1}. We used split injection (50:1) with the following temperature program: 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 and which was then maintained for 15 min. A standard fatty acids methyl ester mix GLC68F (Nu-Chek Prep.) was used for the calibration curve and identification of peaks. Additionally, GLC436 (Nu-Chek Prep.) was used for peak identification.

2.3. Mercury analyses

The samples were analyzed for total mercury content using the MOECC method HGBIO-E3057 (OMOE, 2006). Briefly, 0.2–0.4 g of homogenized tissue was oxidized to its divalent ion form by an overnight acid digestion using 4:1 sulphuric:nitric acid mixture at a temperature of 215–235 °C. The digestion efficiency was checked against two in-house reference materials, which were composite samples of previously analyzed fish fillet samples with mercury concentrations representing low and high values within the desired concentration range. The cold vapor flameless atomic absorption spectroscopy (CV-FAAS) was then used to measure total mercury concentrations. Four mercury calibration standards made from a stock solution traceable to the National Institute of Standards and Technology (NIST) were used. The correlation coefficient of the calibration curve was between 0.990 and 1.00. One sample and one in-house reference material were analyzed in duplicates. Recoveries were monitored by spiking both the sample and reference material, and were on-average about 98.5%.

2.4. Risk-benefit analysis

Elevated mass fractions of multiple contaminants may complicate the assessment of risk associated with fish consumption (Domingo, 2016). A myriad of contaminants were analyzed in Walleye, including organochlorines and other inorganic pollutants, but since mercury was the main cause for consumption advisories (OMOECC, 2015), this study focuses on risks due to mercury. The effect of fish size and sampling location on both mercury and EPA + DHA contents in Walleye were quantified.

Separate consumption advisories were simulated for the general and sensitive population (i.e., women of child-bearing age and children) using the advisory benchmarks from MOECC (Supplement 2). These maximum recommended meals per month were then multiplied by the measured amount of EPA + DHA per fish meal to calculate EPA + DHA (mg) intake per month if fish consumed up to maximum advised meals. This value was then divided by 30 to derive a mean daily intake. We also evaluated which of the two factors, the advisory (i.e., contaminant level) or the EPA + DHA content in Walleye, had a stronger influence on the EPA + DHA intakes. The advisories recommending to limit fish consumption to fewer than 8 meals/month were considered stringent, because such advisories would not allow to

Table 1
Mean mercury concentrations and standard deviations (*SD*) on a wet weight basis (μg g^{-1} ww) in small (30–40 cm, n = 90), medium (41–55 cm, n = 97) and large (> 60 cm, n = 29) sized Walleye from 20 waterbodies across Ontario, Canada (19 lakes and one river*).

Waterbody	Sampling location		Mercury (μg g^{-1} ww)								
			Small			Medium			Large		
	Degrees, minutes, seconds		n	Mean	*SD*	n	Mean	*SD*	n	Mean	*SD*
Attawapiskat*	52°51′45″N	83°52′50″W	5	0.41	*0.19*	5	0.66	*0.26*			
Clear	45°26′28″N	77°11′25″W	5	0.26	*0.07*	5	0.59	*0.14*	2	0.62	*0.09*
Conestogo	43°41′46″N	80°43′53″W	5	0.15	*0.02*	5	0.36	*0.17*			
Doe	45°32′02″N	79°24′51″W	5	0.41	*0.18*	5	0.58	*0.21*	2	1.40	*0.42*
Horwood	47°58′37″N	82°19′34″W	4	0.48	*0.06*	5	1.03	*0.44*	2	1.90	*0.14*
Kagiano	49°18′36″N	86°24′21″W	5	0.46	*0.12*	4	1.14	*0.19*	3	1.97	*0.35*
Kashabowie	48°43′08″N	90°23′39″W	4	0.83	*0.08*	5	1.11	*0.25*	3	2.87	*0.67*
Kwinkwaga	48°48′28″N	85°19′56″W	5	0.88	*0.46*	5	1.04	*0.37*	3	1.57	*0.32*
Lang	51°35′08″N	91°31′28″W	5	0.33	*0.12*	5	1.02	*0.10*			
Lac des Milles	48°50′56″N	90°29′40″W	5	0.49	*0.15*	5	0.46	*0.19*	3	0.52	*0.32*
Nikip	52°53′50″N	91°53′38″W	5	0.27	*0.06*	5	0.43	*0.12*			
Nipigon	49°34′40″N	89°03′25″W	5	0.08	*0.01*	4	0.16	*0.05*	2	0.38	*0.01*
Pakashkan	49°21′20″N	90°16′19″W	5	0.33	*0.10*	4	0.46	*0.22*	3	0.84	*0.49*
Redhead	50°15′35″N	89°57′30″W	4	0.72	*0.23*	5	0.99	*0.40*	2	1.45	*0.21*
Rock	46°26′05″N	83°46′21″W	5	0.47	*0.09*	5	0.83	*0.23*	1	2.70	
Silver	44°05′07″N	81°25′09″W	4	0.34	*0.04*	5	0.57	*0.21*			
Totogan	52°03′47″N	89°11′26″W	4	0.24	*0.04*	5	0.44	*0.04*			
Wabakimi	50°38′36″N	89°46′55″W	3	0.57	*0.17*	5	1.14	*0.47*			
Wasicho	49°25′56″N	80°11′12″W	3	0.48	*0.35*	5	0.63	*0.11*			
White Otter	49°28′29″N	85°32′49″W	4	0.38	*0.09*	5	0.66	*0.12*	3	0.79	*0.11*
Overall			90	0.42	*0.25*	97	0.72	*0.36*	29	1.37	*0.84*

Standard deviations are presented in italics.

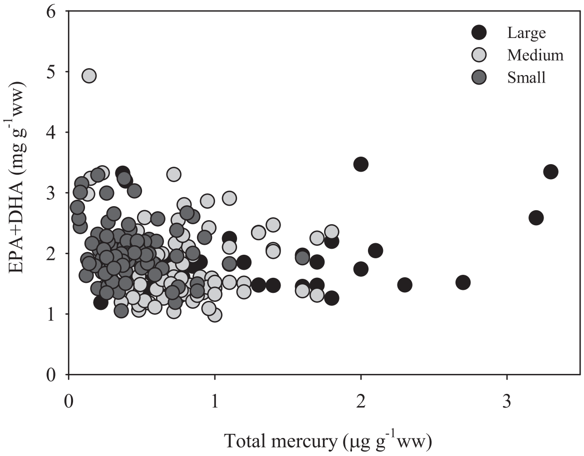

Fig. 1. Mercury and EPA + DHA concentrations in small (30–40 cm), medium (41–55 cm), and large (60–70 cm) sized Walleye.

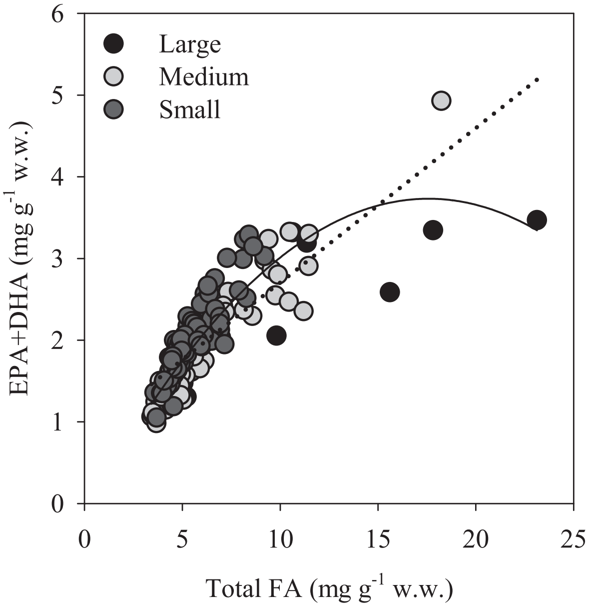

Fig. 2. Linear and quadratic regression (see text for details) between the total fatty acid and EPA + DHA content in small (30–40 cm), medium (41–55 cm), and large (60–70 cm) sized Walleye.

meet the dietary recommendation of two fish meals/week (for a review see Kris-Etherton et al., 2009). The American Dietetic Association and Dietitians of Canada recommend 500 mg EPA + DHA per day from two servings of fatty fish per week (Kris-Etherton and Innis, 2007). The World Health Organization recommends one to two servings 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 selected a daily intake of 250 mg as a benchmark for adequate intake of EPA + DHA and as a representative of the range of reported recommendations.

2.5. Statistical analysis

Correlation between Walleye length and mercury content was analyzed with Spearman correlation (r_s). Bootstrapping (1000 bootstrap iterations) was applied to estimate the 95% confidence interval (biased corrected). Linear and quadratic regressions between the EPA + DHA and total fatty acid content were calculated and the fit between the two models were evaluated.

Possible differences in the mercury and EPA + DHA contents between waterbodies (location) were evaluated using permutational ANOVA (PERMANOVA). Prior to analyses, the concentrations of mercury and EPA + DHA were log(x + 1) transformed. In the PERMANOVA design, location was assigned as a random factor with 20 levels and length was used as a covariate because of the well-known correlation between Walleye length and mercury content. Because we used a covariate, permutation of residuals was run under a reduced model using TYPE I (sequential) sum of squares. The number of

Table 2

Mean and standard deviation (SD) of EPA + DHA content in one meal (mg 227 g^{-1}), the median advisory (and SD) for the general population (gen), and mean (and SD) EPA + DHA daily intake when the maximum advisory is followed over a month for three size ranges of Walleye from 20 waterbodies across the Province of Ontario, Canada (19 lakes and one river*).

Waterbody	Small						Medium						Large					
	EPA + DHA		Advisory(gen)		Intake(gen)		EPA + DHA		Advisory(gen)		Intake(gen)		EPA + DHA		Advisory(gen)		Intake(gen)	
	Mean	*SD*	Median	*SD*	Mean	*SD*	Mean	*SD*	Median	*SD*	Mean	*SD*	Mean	*SD*	Median	*SD*	Mean	*SD*
Attawapiskat*	405	*76*	12	*4.4*	160	*74*	289	*28*	8	*3.3*	70	*38*						
Clear	643	*114*	16	*1.8*	323	*56*	583	*134*	8	*2.2*	119	*30*	381	*120*	6	*2.8*	82	*60*
Conestogo	423	*44*	32	*8.8*	356	*110*	371	*49*	12	*4.9*	149	*65*						
Doe	352	*49*	12	*4.4*	133	*52*	357	*78*	8	*3.3*	85	*39*	422	*124*	3	*1.4*	45	*33*
Horwood	467	*24*	8	*2.0*	140	*27*	432	*139*	4	*0.9*	49	*12*	577	*411*	1	*1.4*	10	*13*
Kagiano	559	*84*	8	*2.2*	178	*46*	477	*56*	4	*1.0*	56	*18*	392	*56*	0	*1.2*	10	*17*
Kashabowie	562	*43*	4	*0.0*	75	*6*	623	*63*	4	*1.1*	68	*28*	654	*201*	0	*0.0*	0	*0*
Kwinkwaga	431	*41*	8	*2.8*	86	*43*	445	*76*	4	*2.4*	57	*31*	447	*45*	2	*1.2*	39	*15*
Lang	397	*33*	16	*3.6*	190	*47*	314	*42*	4	*0.0*	42	*6*						
Lac des Milles	411	*39*	8	*4.4*	119	*54*	386	*87*	12	*3.6*	123	*46*	314	*79*	8	*6.1*	90	*48*
Nikip	414	*114*	16	*1.8*	214	*74*	347	*21*	12	*3.3*	131	*44*						
Nipigon	632	*67*	32	*0.0*	674	*71*	860	*176*	32	*8.0*	808	*308*	769	*21*	12	*0.0*	307	*8*
Pakashkan	464	*42*	16	*4.4*	199	*73*	358	*63*	12	*6.0*	138	*91*	344	*27*	8	*3.5*	69	*42*
Redhead	393	*84*	4	*4.0*	85	*72*	310	*16*	4	*0.9*	37	*10*	333	*4*	2	*0.0*	22	*0*
Rock	503	*58*	8	*2.8*	132	*44*	333	*67*	4	*1.8*	55	*29*	344	–	0	–	0	–
Silver	347	*33*	12	*2.0*	151	*30*	369	*61*	8	*3.3*	85	*30*						
Totogan	382	*68*	16	*0.0*	204	*36*	333	*51*	8	*1.8*	98	*23*						
Wabakimi	438	*20*	8	*4.0*	117	*62*	394	*74*	4	*1.1*	42	*14*						
Wasicho	398	*68*	12	*6.1*	146	*88*	271	*35*	4	*2.2*	49	*17*						
White Otter	497	*69*	10	*3.8*	184	*72*	434	*36*	4	*1.8*	68	*21*	400	*30*	4	*0.0*	53	*4*
Overall	459	*103*	12	*7.4*	200	*147*	409	*146*	4	*5.9*	110	*162*	447	*171*	2	*4.1*	59	*78*

Asterisk marks river.
Standard deviations are presented in italics.

permutations was 9999. The PERMDISP test was conducted to examine the homogeneity of multivariate dispersions of the data set. PERMANOVA is sensitive to differences in the dispersions, and thus PERMDISP was used for the 'location' factor on the basis of distances to centroids, with P-values obtained from permutations (Anderson et al., 2008). It should be noted that PERMDISP can detect small differences in dispersion, which does not necessarily affect PERMANOVA adversely (Anderson et al., 2008).

A curvilinear regression was also plotted between the advisory and the $\log_{10}$ transformed maximum daily intake of EPA + DHA due to the unequal variances. "*Do not eat*" advisories were excluded from the model development, as this would always lead to zero intake of EPA + DHA. The regression model utilized for the estimation of EPA + DHA intake (y) was:

$$y = 1.38 + 0.0854x - 0.0013x^2 \quad (1)$$

where x is the advisory (Fig. 3B). The lower and upper 95% predictions (Eqs. (2) and (3), respectively) for the estimate were calculated as follows:

$$y = 0.56x + 1.36 \quad (2)$$

$$y = 1.79x - 4.25 \quad (3)$$

where x is the estimated EPA + DHA intake.

The regression was then used to estimate the EPA + DHA intake by consuming Walleye from additional 1322 Ontario inland waterbodies for which advisories have been issued by MOECC based on the measured mercury and other contaminant contents but fatty acid measurements are lacking. Walleye from the Great Lakes were excluded because these are fundamentally different environments. To facilitate evaluation of the data, EPA + DHA intake values were transformed back from the $\log_{10}$ values. Altogether, 18,999 Walleye advisories specific to 5 cm size intervals (9406 for the general population, and 9593 for the sensitive population) from additional 1322 waterbodies throughout the Province of Ontario were included in the expanded EPA + DHA intake estimates. It is important to note that the estimates do not account for the potential waterbody-specific differences in the EPA + DHA content. Although significant waterbody-specific variation in EPA + DHA content was observed in this study, the stronger effect of the advisory on the monthly intake of EPA + DHA validates our application to the larger Walleye advisory dataset. Also, the waterbody-specific differences in EPA + DHA content are considered in the 95% prediction intervals, calculated for the estimates.

We also calculated intakes for Walleye from select northern USA lakes (n = 28) using the regression model (Eq. (1)) developed for Walleye from the 20 Ontario lakes. The USA Walleye data were acquired from the National Lake Fish Tissue Study conducted between 2000 and 2003 (Stahl et al., 2009), and included lakes from 8 states: Indiana, Michigan, Minnesota, New York, North Dakota, South Dakota, Utah and Wisconsin.

3. Results

3.1. Mercury and EPA + DHA

The mean moisture percentage in Walleye was 78 ± 1%, and therefore, a conversion factor of 0.22 was applied to all the samples to convert the fatty acid mass fractions per dry weight to a per wet weight basis. The mean total mercury content increased with fish length ($r_s = 0.549$, 95% CI: 0.443–0.645, $P < 0.001$). The mean mercury content for small, medium and large Walleye across all lakes were: 0.4 ± 0.3, 0.7 ± 0.4, and 1.4 ± 0.8 μg g^{-1} wet weight (ww), respectively (Table 1). The corresponding mean EPA + DHA contents were 2.0 ± 0.5, 1.8 ± 0.6, and 1.9 ± 0.7 mg g^{-1} ww, respectively (Fig. 1). A weak negative, albeit statistically significant, correlation was found between mercury and EPA + DHA contents ($r_s = -0.157$, 95% CI: -0.297 to -0.007, $P = 0.021$). However, this result should be interpreted with caution as the sample number was limited at the upper end of mercury concentrations.

The multivariate PERMANOVA analysis show that Walleye EPA + DHA and mercury contents differed between locations even after the effect of fish length was considered (Pseudo-F = 20.434, $P_{perm} = 0.0001$, Supplement 3). There was also significant interaction

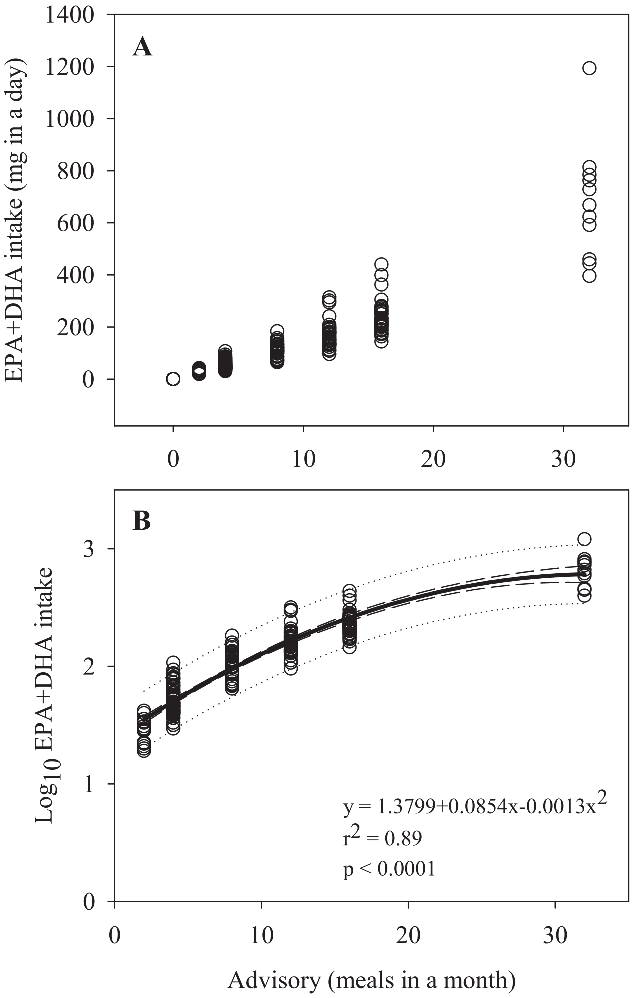

Fig. 3. A) Relationship between the advisory and the daily intake of EPA + DHA when the maximum advisory is followed over a month. Panel B) Nonlinear regression between the consumption advisory and the $\log_{10}$ transformed EPA + DHA intake when the maximum advisory is followed. Dashed and dotted lines indicate the 95% confidence and prediction lines (respectively). This regression was used to estimate EPA + DHA intake from the additional 1322 waterbodies (see the text for details).

effect between the fish size and location, indicating that the effect of fish length on EPA + DHA and mercury contents is not the same across all the lakes (Pseudo-F = 4.698, $P_{perm} = 0.0001$, Supplement 3). The dispersions of the samples between locations were not significantly different (PERMDISP, $F_{19,196} = 1.834$, $P_{perm} = 0.056$). However, considering the strong effect of differences in the dispersion in the PERMANOVA (Anderson et al., 2008), we explored the possible dispersion differences in detail. Large-sized Walleye were caught in only 12 of our study lakes. The absence of large-sized fish samples from the other 8 lakes likely affected the dispersions, because Walleye length was strongly correlated with the mercury content. Note that in the PERMANOVA, this was accounted for by using the length as a covariate, indicating that the PERMANOVA results are not confounded by the limited number of large-sized Walleye samples, and possible differences in the dispersions of the samples between locations. Thus, we are confident that the EPA + DHA and mercury contents in Walleye differ among locations.

The EPA + DHA content was non-linearly correlated to the total fatty acid content (Fig. 2). Although a linear model between the EPA + DHA and total FA content was statistically significant and had a reasonable fit ($r^2 = 0.67$, F = 440.58, $P < 0.001$), adding a non-linear effect in the linear model increased the prediction capacity of the model by about 12 percentage points. Test statistics for the total quadratic model (including both linear and non-linear effects) are $r^2 = 0.79$, F = 397.50, $P < 0.001$ (Fig. 2). Minimal addition in the EPA + DHA content was observed with increasing total fatty acid content beyond 8 mg g^{-1} ww.

3.2. *Risk-benefit*

All Walleye size classes in all the waterbodies provided the recommended EPA + DHA intake (250 mg) in one meal (227 g) (Table 2). However, the EPA + DHA intake over a month was strongly dependent on the consumption advisory (Fig. 3A). The maximum EPA + DHA intake was non-linearly dependent on the advisory, which could explain ~89% of the variation in the EPA + DHA intake (i.e., when the maximum advisory was followed, Fig. 3B). Despite the adequate EPA + DHA content in Walleye muscle, the recommended monthly intake was typically reached only when Walleye would be consumed every day (i.e., no restrictions). However, please note that for Lakes Clear and Nipigon, even fewer meals per month could provide the recommended intake of EPA + DHA (Tables 2 and 3). The consumption advisories for the sensitive population were more stringent than for the general population, and the recommended intake of EPA + DHA was achieved only for small- and medium-sized Walleye from Lake Nipigon (Table 3).

The modelling of EPA + DHA intakes from Walleye for the additional 1322 waterbodies showed no distinct geographical pattern (Fig. 4). Lakes with adequate EPA + DHA intake were distributed throughout the province, although less data was available for the far north. The estimates for the general population showed that the EPA + DHA intake from small-sized Walleye was > 250 mg day^{-1} for 23% of the waterbodies, while an equal intake from large Walleye could be achieved for only 1% of the waterbodies (Fig. 4). For the sensitive population, only ~1% of the waterbodies provided EPA + DHA intake > 250 mg day^{-1} (Fig. 4). Zero EPA + DHA intake indicate *do not eat* advisory, and for the sensitive population, large Walleye from 94% of the waterbodies had *do not eat* advisory. The lower and upper 95% prediction intervals increased with increasing EPA + DHA intake estimate, indicating a better prediction potential for cases with restricted consumption advisories (i.e. 8 or fewer meals per month) (Fig. 3).

4. Discussion

The risks and benefits of consuming Walleye, expressed here as mercury and EPA + DHA contents, respectively, varied among the 20 waterbodies across Ontario. The most important factor affecting the risk versus benefit associated with consuming Walleye was the length of the fish. This is because mercury content, and hence the risk associated with consuming Walleye, increased with increasing fish length. This in accordance with what is known about the bioaccumulation and biomagnification of mercury in fish (Somers and Jackson, 1993; Gewurtz et al., 2011; Clayden et al., 2013). Although EPA + DHA content increased with Walleye length for some lakes, no systematic correlation was observed across all 20 waterbodies. A previous study conducted on Walleye from 33 waterbodies in the northeastern USA reported increasing EPA + DHA contents with increasing fish length, albeit with large lake-specific differences (Williams et al., 2017). In conclusion, the relative risks associated with eating Walleye increased with fish size in our study lakes, but generally the benefits (in terms of EPA and DHA intake) did not.

The spatial differences in EPA + DHA and mercury content in Walleye are presumably due to lake-specific differences in water chemistry and catchment characteristics (Hayer et al., 2011; Monson et al., 2011; Strandberg et al., 2016). Lake-specific differences in mercury contents have been widely noted for various fish species, including Walleye (Wren et al., 1991; Hayer et al., 2011; Monson et al., 2011). Spatial differences in fish mercury are related to the differences in environmental contamination levels, but also the methylation efficiency

Table 3

Mean and standard deviation (SD) of EPA + DHA content in one meal (mg 227 g^{-1}), the median advisory (and SD) for the sensitive population (sen), and mean (and SD) EPA + DHA daily intake when the maximum advisory is followed over a month for three size ranges of Walleye (see Table 2 for details) in 20 waterbodies across the Province of Ontario, Canada (19 lakes and one river*).

Waterbody	Small						Medium						Large					
	EPA + DHA		Advisory(sen)		Intake(sen)		EPA + DHA		Advisory(sen)		Intake(sen)		EPA + DHA		Advisory(sen)		Intake(sen)	
	Mean	*SD*	Median	*SD*	Mean	*SD*	Mean	*SD*	Median	*SD*	Mean	*SD*	Mean	*SD*	Median	*SD*	Mean	*SD*
Attawapiskat*	405	*76*	4	*1.8*	47	*26*	289	*28*	0	*2.2*	16	*23*						
Clear	643	*114*	4	*2.2*	119	*49*	583	*134*	0	*1.8*	13	*29*	381	*120*	0	*0.0*	0	*0*
Conestogo	423	*44*	12	*2.8*	168	*36*	371	*49*	4	*3.3*	60	*42*						
Doe	352	*49*	4	*1.8*	39	*22*	357	*78*	4	*2.2*	31	*29*	422	*124*	0	*0.0*	0	*0*
Horwood	467	*24*	4	*2.0*	47	*32*	432	*139*	0	*0.0*	0	*0*	577	*411*	0	*0.0*	0	*0*
Kagiano	559	*84*	4	*2.2*	48	*45*	477	*56*	0	*0.0*	0	*0*	392	*56*	0	*0.0*	0	*0*
Kashabowie	562	*43*	0	*0.0*	0	*0*	623	*63*	0	*0.0*	0	*0*	654	*201*	0	*0.0*	0	*0*
Kwinkwaga	431	*41*	0	*0.0*	0	*0*	445	*76*	0	*0.0*	0	*0*	447	*45*	0	*0.0*	0	*0*
Lang	397	*33*	4	*1.8*	42	*24*	314	*42*	0	*0.0*	0	*0*						
Lac des Milles	411	*39*	0	*3.6*	32	*47*	386	*87*	4	*1.8*	43	*26*	314	*79*	4	*4.0*	36	*36*
Nikip	414	*114*	4	*2.2*	81	*46*	347	*21*	4	*1.8*	38	*21*						
Nipigon	632	*67*	16	*7.2*	404	*152*	860	*176*	12	*2.0*	317	*95*	769	*21*	4	*0.0*	102	*3*
Pakashkan	464	*42*	4	*1.8*	74	*29*	358	*63*	2	*2.3*	27	*31*	344	*27*	0	*0.0*	0	*0*
Redhead	393	*84*	0	*2.0*	16	*32*	310	*16*	0	*0.0*	0	*0*	333	*4*	0	*0.0*	0	*0*
Rock	503	*58*	4	*1.8*	52	*29*	333	*67*	0	*0.0*	0	*0*	344	–	0	.	0	.
Silver	347	*33*	4	*0.0*	47	*5*	369	*61*	4	*2.2*	28	*26*						
Totogan	382	*68*	6	*2.3*	77	*36*	333	*51*	4	*0.0*	44	*7*						
Wabakimi	438	*20*	0	*2.3*	20	*35*	394	*74*	0	*0.0*	0	*0*						
Wasicho	398	*68*	4	*4.0*	55	*51*	271	*35*	0	*1.8*	6	*14*						
White Otter	497	*69*	4	*0.0*	66	*9*	434	*36*	0	*1.8*	11	*24*	400	*30*	0	*0.0*	0	*0*
Overall	459	*103*	4	*5.0*	75	*99*	409	*146*	0	*2.8*	29	*67*	447	*171*	0	*1.9*	11	*29*

Aterisk marks river.
Standard deviations are presented in italics.

of the environment, particularly in the catchment (Wren et al., 1991; Hayer et al., 2011; Monson et al., 2011; Strandberg et al., 2016), as well as biotic factors, such as food web structure and trophic position of fish (McIntyre and Beauchamp, 2007; Johnson et al., 2015). Higher mercury contents in fish are associated with an increased proportion of peatlands in the catchment, which promotes the anaerobic, microbially-mediated, methylation of inorganic mercury to methylmercury following which transportation from the catchment to the lake is facilitated by the export of organic matter (St. Louis et al., 1994). Mercury emissions have declined globally during the past decades in both North America and Europe (Zhang et al., 2016), but despite global declines, fish mercury contents still increased in the Province of Ontario over the period of 1995–2012 (Gandhi et al., 2014, 2015).

Data on the possible effects of environmental conditions on fish EPA + DHA contents are scarce (Strandberg et al., 2016). Ultimately, the availability of EPA and DHA for upper trophic level consumers is dependent on the algal community composition, as only some of the algal taxa produce these fatty acids in appreciable amounts (Taipale et al., 2013; Galloway and Winder, 2015). Possible modifications and selective trophic transfer will also affect EPA + DHA contents in fish (Strandberg et al., 2015a). Water chemistry, particularly nutrient and dissolved organic carbon concentrations, modify the phytoplankton community composition, and thereby the production of EPA and DHA by algae (Brett and Müller-Navarra, 1997; Galloway and Winder, 2015; Strandberg et al., 2015b; Taipale et al., 2016). For example, decreased EPA + DHA content in Eurasian Perch (*Perca fluviatilis*) correlated with increased loading of allochthonous carbon from the catchment (Strandberg et al., 2016), as well as with increasing dominance of cyanobacteria (Taipale et al., 2016). Eutrophication and brownification of lakes, as well as temperature-dependent changes in the algal production of omega-3 fatty acids, may decrease the availability of EPA + DHA to fish and other upper trophic level consumers (Hixson and Arts, 2016; Taipale et al., 2016).

A significant interaction was found between the location and the length of Walleye, indicating that lake-specific conditions differentially influence the size-specific differences in the mercury and EPA + DHA contents. In other words, in some lakes EPA + DHA contents increased with length, but not in all. The reason for this is presumably related to differences in the food web structure and the production of EPA and DHA by algae, as well as the prey selection plasticity of Walleye (Sheppard et al., 2015). For instance, Walleye have been documented to prefer invasive rainbow smelt (*Osmerus mordax*) even if other native, prey species are available (Sheppard et al., 2015). The lake-specific differences in EPA + DHA content may also reflect differences in the nutritional state of Walleye. The EPA + DHA content was strongly dependent on the total fatty acid content of muscle tissue, which in Walleye ranged from 3.5–23.1 mg g^{-1} ww. Across different fish species higher muscle lipid content is typically associated with higher EPA + DHA content, and the highest EPA + DHA contents are thus found in species (such as salmonids) in which muscle tissue is the main energy storage site (Kainz et al., 2017; Strandberg et al., 2017a). Walleye is relatively lean fish and, regardless of mercury levels, solely consuming Walleye would not provide consumers with the recommended EPA + DHA intake unless Walleye was eaten nearly daily; which is not a realistic scenario. In order to meet dietary recommendations fishermen and other individuals consuming Walleye should supplement their diet with other sources of EPA + DHA (e.g., Lake Whitefish that has higher EPA + DHA levels, but low contaminant levels, or alternatively market fish) (Strandberg et al., 2017b).

4.1. Predicted EPA + DHA intake

The EPA + DHA intake over one month could be estimated from the advisory because of the strong relationship between them. The model we used here is simple, and is based on the advisory only, and where the length of Walleye is indirectly considered through its strong correlation with mercury content in Walleye. This approach provides reasonable estimations of EPA + DHA intake when advisories are stringent, whereas location-specific differences in EPA + DHA contents become more important factor in determining the intake when advisories are more lenient.

No clear geographic pattern in the predicted EPA + DHA intake

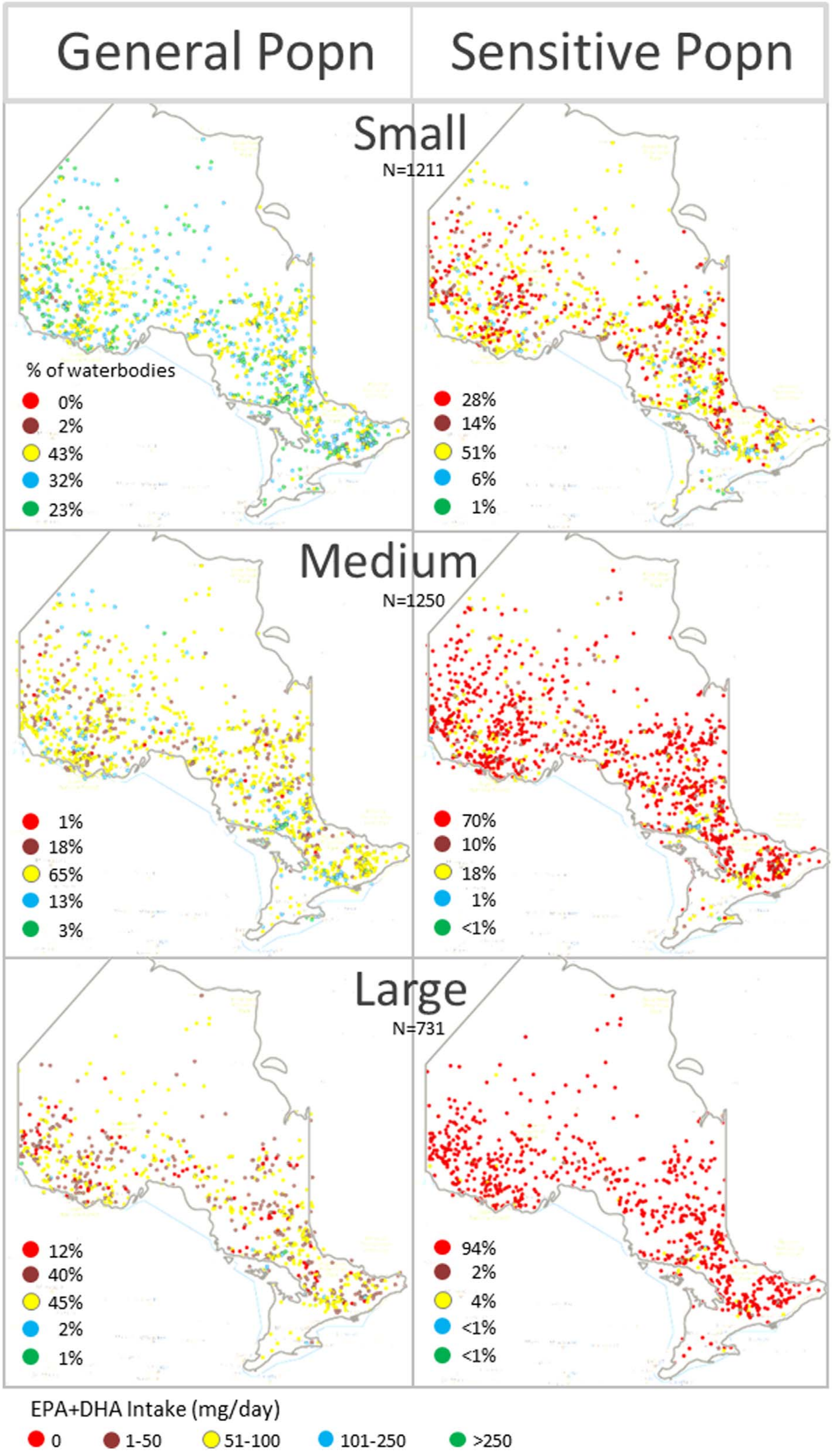

Fig. 4. Estimates of EPA + DHA intake from eating small, medium and large Walleye from total 1322 waterbodies across Ontario. Estimates are calculated separately for the general and the sensitive populations. Percentage values represent the percentage of waterbodies from which a population can obtain the corresponding levels of EPA + DHA intake, indicated by color. Lower and upper 95% prediction intervals for the EPA + DHA intake estimates were as follows: 50: 29–85 mg day^{-1}, 100: 57–174 mg day^{-1}, and 250: 141–442 mg day^{-1}.

were detected; although the data are biased towards southern and central Ontario, with fewer waterbodies from far north Ontario (Fig. 4). The advisory, and subsequently the EPA + DHA intake, are more likely driven by differences in contaminant levels, which are affected by waterbody-specific differences in water chemistry and catchment characteristics (Hayer et al., 2011; Monson et al., 2011; Strandberg et al., 2016). About a quarter of the waterbodies would provide adequate EPA + DHA intake for the general population if consuming only small-sized Walleye, but the proportion drop as the length of Walleye increased. For the sensitive population, the adequate intake could be achieved for only ~1% of the waterbodies, regardless of the size of fish consumed. This indicates that Walleye may not be regarded as an adequate source of EPA + DHA for the sensitive population.

The Walleye mercury levels in the USA dataset were lower than in

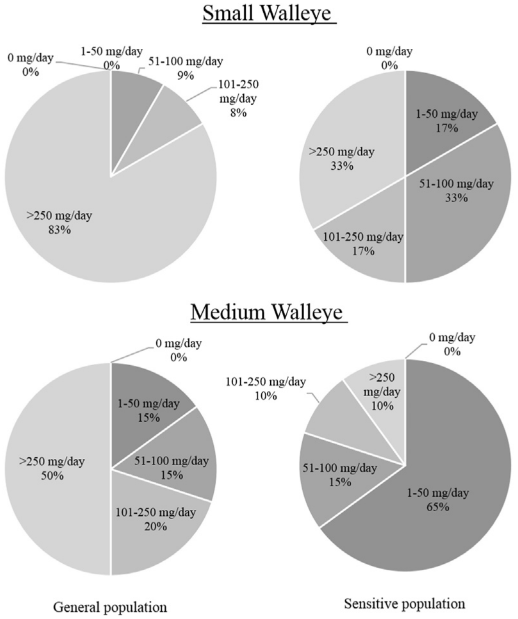

Fig. 5. Percentage of lakes in northern USA with simulated EPA + DHA intake from small (n = 12) and medium (n = 20) sized Walleye. Charts on the left represent values for the general population and charts on the right for sensitive population. The calculations of the intake are based on the model described in the present paper and the data are from Stahl et al. (2009).

the present study, possibly due to different local geochemistry. Elevated mercury levels in fish from shield areas, characterized by Precambrian metamorphic rocks, compared to no-shield areas have been observed (Neff et al., 2013). The Ontario waterbodies considered in the regression model application are mostly in the shield region, while the USA locations are located in the non-shield region. This could explain the lower fish mercury levels in the USA waterbody and correspondingly higher simulated EPA + DHA intakes. Adequate intake could be obtained for the general population in 50–83% of the analyzed USA lakes, depending on the length of the fish. Particularly noteworthy is that the USA lakes did not have any "*do not eat*" advisories (Fig. 5).

Other sources of EPA + DHA may include store-bought seafood (e.g. finfish, shellfish) and/or recreational fishing for other fish species. Interspecific comparison of Walleye with other wild freshwater fish from North America and Europe show that Salmoniformes in general have higher EPA + DHA contents making them valuable sources of EPA + DHA providing that the contaminant levels do not exceed set guidelines (Table 4). Within the Salmoniformes order the Coregonid species often feed lower down on the food chain than many of the large salmonines, but still accumulate lipids, including EPA and DHA, in their muscle tissue (Strandberg et al., 2017a). Thus, species, such as Lake Whitefish (*Coregonus clupeaformis*) and Cisco (Lake Herring, *Coregonus artedi*), may be a better choice than salmon and trout due to their high EPA + DHA content and typically more lenient consumption advisories (Bhavsar et al., 2011; Strandberg et al., 2017b). The average EPA + DHA content in Walleye in the present study was comparable with previous study on Walleye in inland lakes (Williams et al., 2017), but the mercury levels in our study were higher than Canada-wide averages (Table 4). Similarly, the EPA + DHA in Walleye from the present study were comparable with Pike-Perch (*Sander lucioperca*) from Europe, a close relative to the Walleye (Table 4). EPA + DHA and mercury in common store-bought marine or cultured fish are typically reported as long term averages without specification of fish length or detailed fishing location (see Mozaffarian and Rimm, 2006 and

Table 4
Mean EPA + DHA (mg 100 g^{-1} ww) and mercury (μg g^{-1} ww) contents in the muscle of selected freshwater fish from North America (northern USA and Canada) and Europe (central and northern Europe). The number of samples is given in brackets. Walleye from the present study is presented separately (bolded and marked with *). All the fish in this table were caught from freshwater, even if the species has marine or brackish water life-stages or populations.

North America				Europe			
Species	Order	EPA + DHA (mg 100 g^{-1})	Mercury (μg g^{-1})	Species	Order	EPA + DHA (mg 100 g^{-1})	Mercury (μg g^{-1})
Common Carp	Cypriniformes	59 (7)	0.17 (3573)	Bleak	Cypriniformes	327 (2)[b]	0.09 (5)[h]
Black Crappie	Perciformes	111 (50)	0.11 (1480)	Bream	Cypriniformes	287 (6)[bc]	0.12 (98)[ehjk]
Bluegill	Perciformes	94 (29)	0.08 (801)	Brown Trout	Salmoniformes	158 (5)[b]	0.10 (16)[d]
Brown Bullhead	Siluriformes	101 (1)	0.1 (3890)	Common Carp	Cypriniformes	99 (3)[b]	0.06 (6)[dh]
Brown Trout	Salmoniformes	365 (3)	0.17 (1315)	Common Roach	Cypriniformes	244 (9)[bc]	0.13 (427)[a]
Chain Pickerel	Esociformes	70 (1)	0.57 (82)	Eurasian Perch	Perciformes	257 (82)[bcf]	0.18 (106)[efghi]
Channel Catfish	Siluriformes	128 (15)	0.2 (1741)	European Eel	Anguilliformes	1590 (6)[b]	0.22 (57)[ei]
Cisco	Salmoniformes	569 (14)	0.14 (4576)	Northern Pike	Esociformes	263 (2)[b]	0.23 (15)[eik]
Northern Pike	Esociformes	137 (49)	0.38 (52881)	Pike-Perch	Perciformes	192 (4)[bc]	0.19(14)[ek]
Lake Whitefish	Salmoniformes	653 (12)	0.09 (16779)	European Catfish	Siluriformes	360 (3)[b]	0.27(47)[el]
Largemouth Bass	Perciformes	154 (54)	0.29 (3453)				
Lake Trout	Salmoniformes	741 (94)	0.41 (21437)				
Pumpkinseed	Perciformes	128 (1)	0.09 (1643)				
Rainbow Trout	Salmoniformes	516 (12)	0.11 (3014)				
Sunfish family	Perciformes	62 (1)	0.22 (121)				
Smallmouth Bass	Perciformes	145 (25)	0.33 (10436)				
Splake	Salmoniformes	410 (10)	0.17 (357)				
Walleye	Perciformes	185 (179)	0.41 (64898)				
Walleye*		**193 (216)**	**0.84 (216)**				
White Crappie	Perciformes	109 (5)	0.08 (150)				
Yellow Perch	Perciformes	144 (23)	0.14 (12557)				
Total				Total	Anguilliformes	1590	0.22
	Cypriniformes	59	0.17		Cypriniformes	239	0.10
	Esociformes	104	0.48		Esociformes	263	0.23
	Perciformes	136	0.14		Perciformes	225	0.19
	Salmoniformes	543	0.14		Salmoniformes	158	0.10
	Siluriformes	115	0.15		Siluriformes	360	0.27

Citations for North American samples: Mercury data (Depew et al., 2013), EPA + DHA data (Williams et al., 2017) Great Lakes samples excluded from the EPA + DHA data.
Citations for European samples: [a]Sonesten, 2001, [b]Yamada et al., 2014, [c]Joordens et al., 2014,[d]Djedjibegovic et al., 2012, [e]Noël et al., 2013, [f]Strandberg et al., 2016, [g]Orban et al., 2007, [h]Petkovšek et al., 2012, [i]Yamaguchi et al., 2003, [j]Farkas et al., 2002, [k]Kenšova et al., 2012, [l]Squadrone et al., 2015.

references within). Walleye and lean marine fish like Atlantic Cod (*Gadus morhua*), Skipjcak Tuna (*Katsuwonus pelamis*) and Mahimahi (*Coryphaena hippurus*) show comparable EPA + DHA levels, but the mercury values are higher in Walleye (Mozaffarian and Rimm, 2006). Atlantic herring (*Clupea harengus*) and anchovies (several species from the Engraulidae family) have over ten times higher EPA + DHA levels that Walleye and low contaminant, including mercury, levels (Mozaffarian and Rimm, 2006).

Comparison of Walleye in this study with Walleye from Lake Ontario revealed that despite Lake Ontario being heavily burdened with contaminants such as PCBs (Strandberg et al., 2017b), their mean mercury levels were nonetheless lower than in the Walleye from the lakes in the present study (0.30 μg g^{-1} and 0.84 μg g^{-1}, respectively). Lake Ontario Walleye showed similar size-dependent increase in muscle mercury contents as Walleye caught from the smaller inland lakes, but the mass fractions of mercury were lower in Lake Ontario Walleye (Strandberg et al., 2017b). This intraspecific variation is presumably related to differences in environmental contaminants levels, methylation efficiency, or alternatively, to differences in food web structure, particularly food chain length which has been shown to affect contaminant levels in top predators (Mathers and Johansen, 1985, St. Louis et al., 1994, Ullrich et al., 2001, McIntyre and Beauchamp, 2007).

Compared to mercury, much less is known about the spatial and size-dependent variation in muscle EPA and DHA content in Walleye or other fish species (but see Neff et al., 2014; Strandberg et al., 2017b; Williams et al., 2017 for Great Lakes fish). Interestingly, the mean EPA + DHA content in Lake Ontario Walleye (n = 33, all size classes) was ~367 mg in a meal (227 g), less than the overall mean in the current study 459 mg 227 g^{-1} (Table 3; Strandberg et al., 2017b). This indicates that although size is a dominant factor in risk-benefit assessment, spatial differences in EPA + DHA contents also play a role. Correspondingly, the predicted EPA + DHA intake in Walleye shows large variation, which, presumably, can be reduced once the reasons for lake-specific differences in Walleye EPA + DHA content (e.g. differences in lake water chemistry, food web structure and catchment characteristics) are better understood (Strandberg et al., 2016).

5. Conclusions

Elevated mercury contents were observed in Walleye from Ontario lakes; particularly in large-sized fish. All Walleye length classes across all the 20 waterbodies studied provided the recommended EPA + DHA intake (250 mg) in one meal (227 g). However, if the consumption advisories were followed, the intake of EPA + DHA from eating Walleye would typically be inadequate. The potential intake of EPA + DHA was more influenced by the consumption advisory than the fish EPA + DHA content. No distinct geographic patterns in the Walleye as the source of EPA + DHA were observed likely because the waterbody-specific differences are driven by local differences in the environmental mercury levels and methylation potentials. As expected, the EPA + DHA intake estimates decline with increasing Walleye length; for large-sized (> 60 cm) Walleye only 1% of the waterbodies provided the adequate EPA + DHA intake. However, consumption of small-sized (30–40 cm) Walleye provided an adequate EPA + DHA intake (250 mg day^{-1}) for the general population in 23% of the waterbodies. Overall, Walleye is a relatively poor source of EPA + DHA compared to e.g. Lake Whitefish, and consumers should not solely rely on 1–2 Walleye meals in a week to obtain the EPA + DHA.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2017.12.029.

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