CONCENTRATIONS OF PBDES IN WALLEYE AND ESTIMATED DIETARY INTAKE

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Introduction

Although a number of studies have described concentrations of PBDEs in predatory freshwater fish sampled from U.S. and Canadian waters, no studies to date have estimated dietary intake from consumption of these fish.[1-8] Such estimates will likely be useful in developing consumption advisories to help reduce a potentially significant route of human exposure in certain regions, such as the Great Lakes, where the typical diet commonly includes fish, particularly for sport anglers and their families.[9,10] At the 2013 BFR conference in San Francisco, we described concentrations of PBDEs in wild-caught walleye fillets imported from Canada that were collected as part of a Minnesota market basket survey.[11] Here, we compare those findings to the results of an angling study we conducted in the same state and estimate dietary intake of PBDEs associated with consumption of imported and domestic wild-caught walleye.

Materials and Methods

Collection, preparation and laboratory analysis of fish tissue samples has been previously described in detail by Scott et al.[11] As part of an angling and market basket survey of walleye in Minnesota, walleye fillets imported from Canada (*n* = 21) were purchased from eight seafood markets and/or grocery stores and one on-line fish retailer located throughout the state. Wild-caught walleye were also harvested from one Minnesota lake and one river (*n* = 11) by anglers using hook and line fishing gear and one Minnesota lake using fishing nets (*n* = 7). Of these 18 samples, five individual fish were filleted by local anglers beginning at the mid-dorsal line, including the belly flap, to provide flesh from head to tail. Four of the remaining fillet samples and two whole samples were fish confiscated from anglers by the Minnesota Department of Natural Resources (MN-DNR) in February 2009 and provided to the authors. The remaining fillets were processed at a local fishery in northern Minnesota. Liver tissue from each of the five fish collected from the Rainy River was composited for pooled analysis.

Tissue samples were analyzed by Vista Analytical Laboratory (El Dorado Hills, CA) for 20 PBDEs according to U.S. EPA Method 1614, and lipid content was determined via U.S. EPA Method 8290. Because previous analyses of the market basket data demonstrated the limits of detection had little effect on summed PBDE concentrations, samples below the limit of detection (LOD) were only assigned one surrogate value which was equal to the LOD divided by the square root of two. Summary statistics were calculated for Σ10 PBDE concentrations and selected individual congeners. All summed PBDE concentrations included BDEs 17, 47, 49, 66, 85, 99, 100, 153, 154 and 183/176. Differences between/among groups were examined using the Wilcoxon rank-sum test and an alpha level of 0.05.

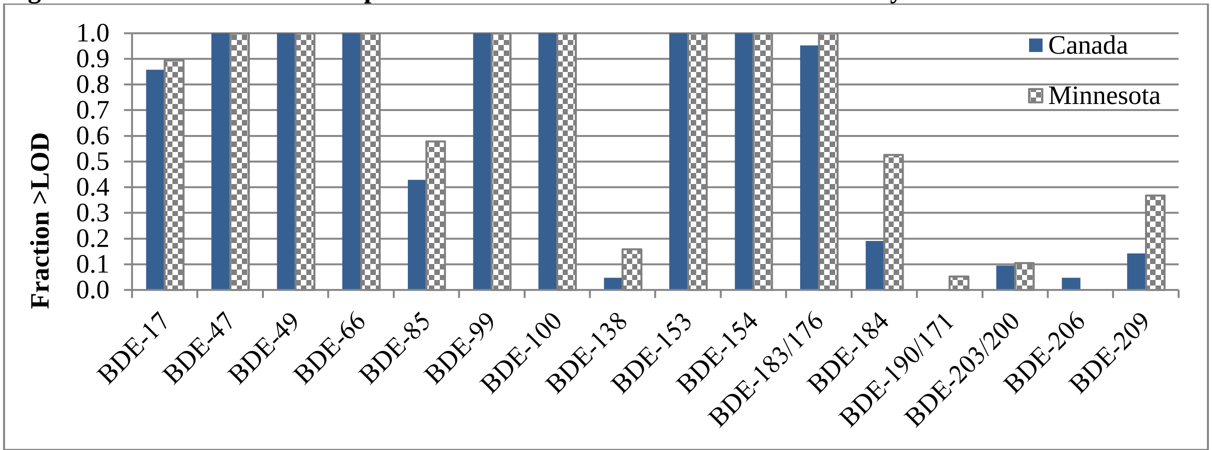
Estimated Daily Intake was calculated using Equation 1. Here, C is the median Σ10 PBDE wet weight concentration, and IR is the fish ingestion rate based on various consumption rates for Minnesota residents identified in the U.S. EPA Exposure Factors Handbook.[12] An exposure frequency (EF) of 350 days/year was specified for all populations. Exposure duration (ED) was assumed to be 70 years for adults and 10 years for children aged 1-14 years. A body weight (BW) of 70 kg was used for adults and 30 kg for children. Averaging time (AT) was set at 25,550 days for adults and 3,650 days for children.

$$\text{Daily Intake} = \frac{\text{C} \times \text{IR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \quad (1)$$

Results and Discussion

Scott and colleagues have previously provided descriptive statistics for the limits of detection.[11] BDEs 47, 49, 66, 99, 100, 153, and 154 were detected in all samples, while BDEs 28/33, 71, 128, and 191 were not measureable in any of the samples. As shown in Figure 1, the largest differences in the fraction of tissue samples with concentrations greater than the LOD were for BDEs 184 and 209.

Figure 1: Fraction of tissue samples with measureable PBDE concentrations by initial location of catch.



Fish collected from MN lakes and rivers had a mean lipid fraction of 0.9% (range: 0.4 – 3.8%) while imported samples purchased at markets and grocers had a mean of 1.9% (range: 0.4-6.0%). The lipid fraction of the liver composite for fish collected from the Rainy River was 4.2%. Overall, the fish collected for this study were noticeably leaner than walleye and other predatory freshwater fish collected from the Great Lakes.[1-3,13]

Three samples imported from Canada and seven of the samples from Minnesota lakes and rivers (four of the five fish collected from the Rainy River and the liver composite for these fish and two of the samples from Lake of the Woods) had measureable concentrations of BDE 209. Concentrations for Minnesota walleye, excluding the liver composite, ranged from 0.09 to 0.28 ng/g (6.21-51.8 ng/g lipid) with a median concentration of 0.17 ng/g (22.2 ng/g lipid). Wet weight concentrations of BDE 209 for tissue samples from Canada were not considerably different (range: 0.10-0.22 ng/g; median: 0.12 ng/g), but lipid normalized concentrations (range: 7.33-9.03 ng/g lipid; median: 8.31 ng/g lipid) were several-fold lower. In general, both wet weight and lipid adjusted concentrations of individual congeners were much lower for imported samples collected from Minnesota markets and grocers (data not shown). Median wet weight concentrations of BDE 47, 99, and 100 measured in walleye from Minnesota lakes and rivers were more than twice those for samples imported from Canada, while lipid normalized concentrations were between five and ten times higher. More detailed comparisons of individual congener concentrations will be described in a later publication.

Σ10 PBDE wet weight and lipid-adjusted concentrations are presented in Table 1. The whole fish provided by the MN-DNR had the highest wet weight and lipid-normalized concentrations of summed PBDEs. While it is possible these results are due to cross-contamination of the samples, concentrations measured in the four fillets that were also confiscated from anglers at the same lake were substantially lower (median: 0.08 ng/g, 14.3 ng/g lipid). Both summed wet weight and lipid weight concentrations for walleye collected from Minnesota lakes and rivers were statistically higher than the market basket fish imported from Canada (w.w.: $p=0.01$; l.w.: $p<0.001$). These differences were not lessened by removing the whole and pan-dressed samples from the analysis (w.w.: $p=0.01$; l.w.: $p=0.001$). As expected, Σ10 PBDE wet weight concentrations measured in the composited livers of fish collected from the Rainy River (4.04 ng/g) were considerably higher than the total concentrations for the fillets from the same fish (range: 0.26-0.78 ng/g). Differences in the lipid normalized concentrations (liver composite: 96.2 ng/g lipid; fillet - range: 68.9-113 ng/g lipid) were not quite so disparate.

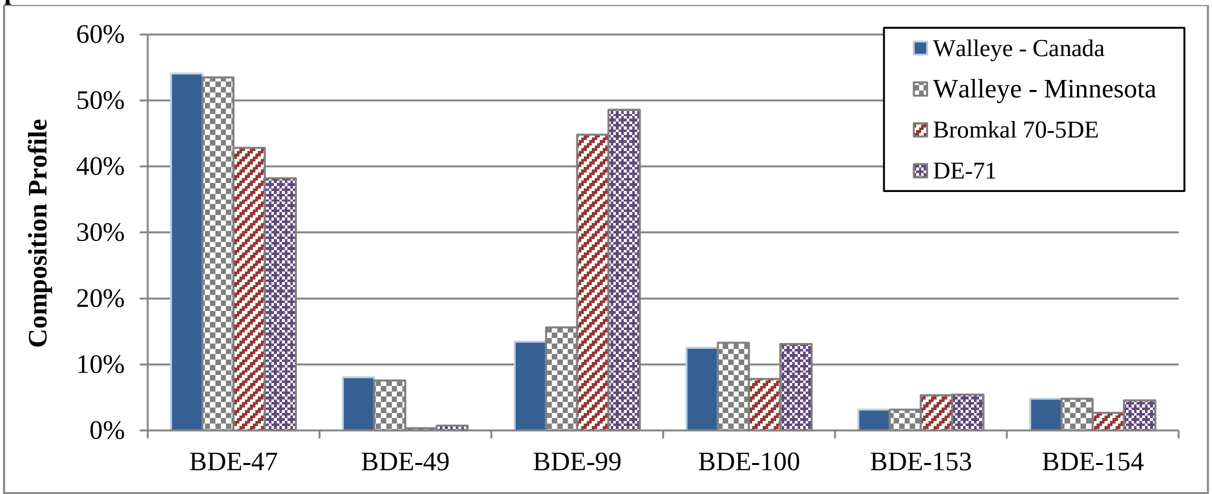
The congener patterns for walleye sampled from Minnesota lakes and rivers and walleye imported from Canada were very similar, with BDEs 47, 49, 99, and 100 accounting for, on average, 90% and 88% of Σ10 PBDE concentrations, respectively. Compared with two technical penta-BDE formulations, wild-caught walleye had higher fractions of BDEs 47 and 49 but a much lower fraction of BDE 99.[14] These differences could, however, be caused by the debromination of BDE 99 in fish tissue.

Table 1: Σ10 PBDE concentrations measured in wild-caught walleye.

		Wet weight (ng/g)			Lipid weight (ng/g lipid)		
	N	Range	P50	P95	Range	P50	P95
All Samples	39	2.54E-02 - 2.25E+00	1.98E-01	2.25E+00	3.92E+00 - 3.07E+02	1.78E+01	1.13E+02
Canada	21	2.54E-02 - 1.94E+00	1.14E-01	7.82E-01	3.92E+00 - 7.24E+01	8.21E+00	6.97E+01
Fillet	19	2.54E-02 - 1.94E+00	1.10E-01	1.94E+00	3.92E+00 - 7.24E+01	7.46E+00	7.24E+01
Pan-dressed	2	3.10E-01 - 4.24E-01	3.67E-01	NC	9.56E+00 - 1.82E+01	1.39E+01	NC
Minnesota	18	1.25E-01 - 2.25E+00	2.45E-01	2.25E+00	1.08E+01 - 3.07E+02	4.22E+01	3.07E+02
Fillet	16	1.25E-01 - 7.78E-01	2.28E-01	7.78E-01	1.08E+01 - 1.13E+02	3.51E+01	1.13E+02
Whole	2	2.25E+00 - 2.25E+00	2.25E+00	NC	5.85E+01 - 3.07E+02	1.83E+02	NC

P50, 50th percentile; P95, 95th percentile; NC, not calculated

Figure 2: PBDE composition profiles for imported and domestic wild-caught walleye and two commercial penta-BDE mixtures.



Estimates of dietary intake associated with the consumption of walleye are presented in Table 2. For both adults and children, the highest estimates of dietary PBDE intake was associated with any consumption of whole walleye by consuming anglers and high consumption levels (≥95th percentile) of whole sport-caught walleye for the general population. Ingestion of ≥95th percentile of pan-dressed fish also increased dietary intake. There was no to very little difference in dietary intake estimates from consumption of walleye fillets from MN Lakes and rivers and imported fillets purchased at local markets and grocery stores for the general Minnesota population. In contrast, dietary intake of PBDEs for consuming child anglers who ingest walleye at the 95th percentile was almost three times higher compared to children in the general population who consume only sport-caught or purchased walleye, suggesting consuming child anglers may be particularly susceptible to exposure via diet. For adult consuming anglers who ingest walleye at the 95th percentile, dietary intake of PBDEs was less than that for consumption of all walleye by the general population and just slightly higher for consumption of only sport-caught and only purchased fish by the general population.

Although PBDE concentrations vary substantially between imported and domestic wild-caught walleye, dietary intake associated with consumption of this fish primarily differs only between children in the general population and recreational child anglers, particularly those who ingest significant quantities of fish. We expect that the estimates of intake presented here will be useful in conducting preliminary risk assessments and developing initial fish consumption advisories and environmental policies for PBDE exposure in both children and adults. Nonetheless, the model used here to estimate intake assumes PBDEs are continuously present in all walleye at an average level, thereby overestimating exposure from consumption of walleye. To address this and other differences in consumption, body weight, exposure of sensitive subpopulations such as Native Americans, etc., PBDE dietary intake will also need to be estimated using probabilistic methods.

Table 2: Daily intake (ng/kg-d) of PBDEs from consumption of walleye for children (A) and adults (B).

(A)

IR (g/day)	Sport-caught Fish*		Purchased Fish*		Total*		Consuming Anglers†	
	P50 = 1.2	P95 = 13.7	P50 = 3.6	P95 = 31.3	P50 = 6.9	P95 = 38.1	Mean = 14	P95 = 37
All Samples	---	---	---	---	0.04	0.24	---	---
Canada	---	---	0.01	0.11	---	---	---	---
Fillet	---	---	0.01	0.11	---	---	---	---
Pan-dressed	---	---	0.04	0.37	---	---	---	---
Minnesota	0.01	0.11	---	---	---	---	0.11	0.29
Fillet	0.01	0.10	---	---	---	---	0.10	0.27
Whole	0.09	0.98	---	---	---	---	1.01	2.66

(B)

IR (g/day)	Sport-caught Fish*		Purchased Fish*		Total*		Consuming Anglers†	
	P50 = 2.8	P95 = 28.9	P50 = 6.6	P95 = 43.2	P50 = 12.3	P95 = 64.5	Mean = 14	P95 = 37
All Samples	---	---	---	---	0.03	0.18	---	---
Canada	---	---	0.01	0.07	---	---	---	---
Fillet	---	---	0.01	0.07	---	---	---	---
Pan-dressed	---	---	0.03	0.22	---	---	---	---
Minnesota	0.01	0.10	---	---	---	---	0.05	0.12
Fillet	0.01	0.09	---	---	---	---	0.04	0.12
Whole	0.09	0.89	---	---	---	---	0.43	1.14

IR, ingestion rate; P50, 50th percentile; P95, 95th percentile
*Fish consumption for general population of Minnesota residents
†Freshwater recreational fish consumption for Minnesota residents

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