Polybrominated Diphenyl Ethers (PBDEs) in Wisconsin Fish: 2002-2012

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Abstract — The Wisconsin Department of Natural Resources (WDNR) has been tracking bioaccumulating pollutants in fish that are consumed by wildlife, anglers, and anglers' families since the 1970s. Beginning in 2002 and using funds from grants or special projects, this effort has included monitoring levels of polybrominated biphenyl ethers (PBDEs) in Wisconsin sport fish from the Great Lakes and inland waters. The WDNR also has access to PBDE data from fish collected as part of the United States Environmental Protection Agency's 2003 National Lake Fish Tissue Study and 2010 National Coastal Condition Assessment Great Lakes Human Health Fish Tissue Study. This report summarizes the concentrations of total PBDEs and proportions of PBDE congeners found in 26 fish species from 19 inland waters, Lake Michigan, and Lake Superior and explores the factors affecting PBDE accumulation in fish fillets. We found that PBDE contamination was spatially heterogeneous, and species with higher lipid content contained higher total PBDEs. Congener BDE-47 made up the highest proportion of total PBDEs in fillets of all species tested. We also found that total PBDEs in fish sampled from the Great Lakes did not generally exhibit temporal variability but that proportions of congener types changed consistently through time, suggesting a possible shift in Great Lakes' PBDE origins. Using currently available reference doses, species/location combinations were evaluated to determine risk based on consumption of fish tested. Total PBDE levels in most fish from most locations were not high enough to trigger exceptions to our statewide advice. Where more restrictive advice was warranted, the current advice due to PCB contamination was not superseded. We suggest continued monitoring of PBDEs in Wisconsin sport fish, as the fate of PBDEs in the environment is unclear.

Polybrominated diphenyl ethers (PBDEs) are a class of persistent, bioaccumulative, toxic chemicals that have been in production since the 1970s. They function as flame retardants and were primarily used in the manufacture of foams (such as those found in couch cushions), consumer electronics, and fabrics (Costa and Giordano 2007). There are 209 possible congeners, and commercial formulations (known as Penta, Octa, or Deca) contain varying concentrations of these congeners. Because PBDEs are *added* to products and are not part of their chemical structure, they dissipate as the foam degrades or as the product is heated. PBDEs are lipophilic, meaning that they accumulate in fatty tissues, and thus biomagnify up the food chain along similar exposure routes as polychlorinated biphenyls (PCBs; Stapleton and Baker 2003; Hahm et al. 2009).

Studies showing the toxicological effects of PBDEs in exposed animals began to arise in the 1990s (Andersson and Wartanian 1992, Sellström et al. 1993). Research investigating PBDEs' effects on both animals and humans have proliferated in the years since, linking PBDE exposure to problems with human infant neurological development (Costa and Giordano 2007) and thyroid hormone disruption (Talsness 2008). Lower brominated congeners, particularly penta-BDEs, appear to be more toxic to humans than higher brominated congeners (Darnerud 2003). Because of the aforementioned health effects,

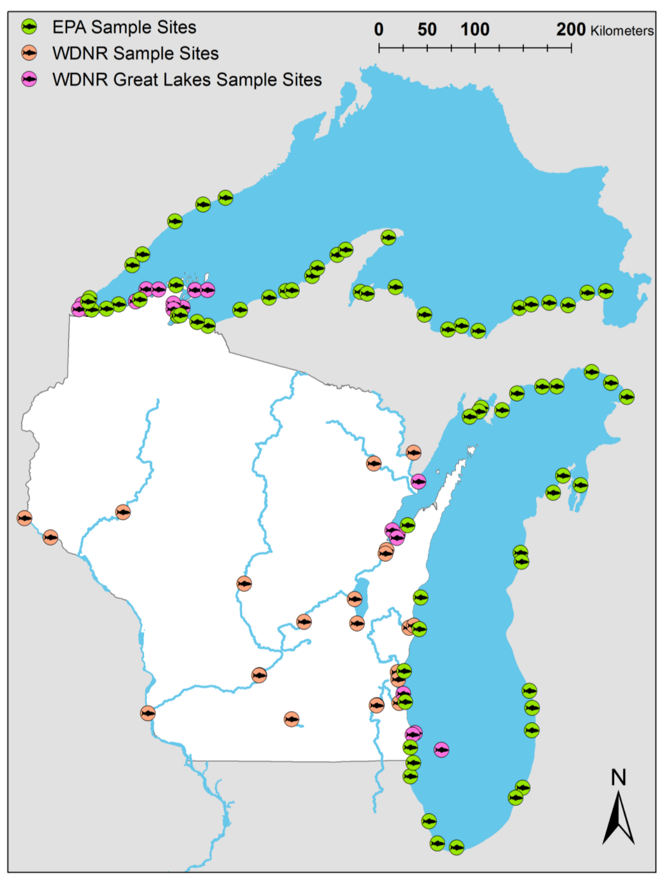

Figure 1. Locations from which fish were sampled for PBDE analysis.

the United States ceased to manufacture and import commercial mixtures of Penta- and Octa-BDEs in 2004, and principal commercial manufacturers agreed to voluntarily cease production and import of Deca-BDE by 2013 (USEPA 2009). However, due to their ubiquity before the ban, the fact that higher brominated mixtures were still produced until 2008, and because higher brominated congeners break down into lower brominated and more toxic congeners, there is still a high probability of exposure to humans and animals alike (Siddiqi et al. 2003, Turyk et al. 2008).

In the present study, it was expected that different fish species would contain different total PBDE (hereafter ΣPBDEs) concentrations and different proportions of congeners, reflecting their position in the food chain, lipid content, and the location from which they were caught (Figure 1). These expectations are consistent with what others have found with regard to PBDEs in North American freshwater fish: fattier or bottom dwelling fish species have higher ΣPBDEs on average than leaner or predator species (Kuo et al. 2010; Stahl et al. 2013) and PBDE contamination is spatially heterogeneous (i.e. point source contamination, Hale et al. 2001; Gewurtz et al. 2011; Skinner 2011).

Sampling and analysis
Prior to 2010, Wisconsin fish samples (skin-off fillets: channel catfish; whole fish: bloater chub, rainbow smelt, and gizzard shad; skin-on fillets: all other species) were analyzed for nine congeners. Starting in 2010, eight additional congeners were quantified as lab capabilities increased (Table 1), although these additional congeners made up <1% of the total concentration in post-2010 samples. BDE-85 was detected in only 1 sample and BDE-156 was not detected in any sample. In this report, ΣPBDEs refers to BDEs 28, 47, 49, 66, 85, 99, 100, 138, 153, and 154 for samples taken prior to 2010 and all congeners listed in Table 1 for samples taken after 2010.

A list of species and locations sampled can be found in Tables 2 and 3 (see Appendices I and II for detailed sample information). Due to cost constraints, the initial approach was to survey many water bodies and obtain information on the range of PBDE concentrations found in a small number of bottom-feeding fish from each location. Subsequent testing focused on additional species from the Great Lakes and Areas of Concern. However, we acknowledge that small sample sizes of many species (often ≤2) from many waters makes statistical comparisons difficult. As such, spatial distribution of ΣPBDEs found in all species is qualitatively represented in map form (Figures 3, 5, and 7). Analyses of variances (ANOVAs) were conducted only with species where >2 samples were collected from one location at one time (Figure 10). Additionally, previous research helps to inform our knowledge of PBDE fate and distribution.

Table 1. PBDE congener analysis information for all samples, including those collected as part of EPA studies.

Congener	Chemical Formula	Year(s) Analyzed
BDE-28	2,4,4'-tri	2002 - 2012
BDE-47	2,2',4,4'-tetra	2002 – 2012
BDE-49	2,2',4,5'-tetra	2002 – 2012*
BDE-66	2,3',4,4'-tetra	2002 – 2012
BDE-85	2,2',3,4,4'-penta	2002 – 2012
BDE-99	2,2',4,4',5-penta	2002 – 2012
BDE-100	2,2',4,4',6-penta	2002 – 2012
BDE-138	2,2',3,4,4',5'-hexa	2002 – 2012
BDE-153	2,2',4,4',5,5'-hexa	2002 – 2012
BDE-154	2,2',4,4',5,6'-hexa	2002 – 2012
BDE-156	2,3,3',4,4',5-hexa	2010 – 2012
BDE-183	2,2',3,4,4',5',6-hepta	2010 – 2012
BDE-196	2,2',3,3'4,4',5,6'-octa	2010 – 2012
BDE-197	2,2',3,3',4,4',6,6'-octa	2010 – 2012
BDE-206	2,2',3,3',4,4',5,5',6-nona	2010 – 2012
BDE-207	2,2',3,3',4,4',5,6,6'-nona	2010 – 2012
BDE-209	decabromodiphenyl ether	2010 – 2012

*Prior to 2010, BDE-49 was analyzed in only 6 of 83 samples

All samples collected by the WDNR were analyzed at the Wisconsin State Laboratory of Hygiene using a method developed in-house (ESS Org Method 1410). Briefly, homogenized fillets or whole fish (depending on the species) were further ground with dry ice and, after standing overnight, combined with sodium sulfate and extracted with dichloro-methane. Lipids were removed by gel-permeation chromatography, and additional clean up was conducted as needed according to United States Environmental Protection Agency (USEPA) Method 3620C, 3630C, or 3640A. Final extract(s) were analyzed using a gas chromatograph and electron-capture detector (pre-2011) or mass spectrometer (post-2011).

Samples collected for the EPA were analyzed according to USEPA Method 1614, described in Stahl et al. (2013). The EPA method is a high-resolution technique, and thus had lower MDLs

Table 2. Samples collected from Lakes Michigan and Superior. Parenthetical numbers represent the pre-composite *n*, if any samples were composited prior to analysis.

Waterbody	Species	*n*
Lake Michigan (includes Green Bay)	Bloater chub	2(60)
	Brown trout	1
	Channel catfish	2
	Chinook salmon	18(20)
	Coho salmon	5
	Common carp	2
	Freshwater drum	1
	Gizzard shad	3(90)
	Lake sturgeon	3
	Lake trout	9(15)
	Lake whitefish	3
	Northern pike	1
	Rainbow smelt	3(30)
	Rainbow trout	13
	Round whitefish	1
	Smallmouth bass	1
	Walleye	9(15)
	White sucker	1
	Yellow perch	6
Lake Superior (includes the St. Louis River/ Superior Harbor)	Bloater chub	2(10)
	Brown trout	3
	Chinook salmon	6
	Cisco / lake herring	4(40)
	Coho salmon	3
	Common carp	2
	Lake trout	30(94)
	Lake whitefish	6
	Longnose sucker	5(15)
	Northern pike	4
	Round whitefish	6(24)
	Siscowet lake trout	6
	Splake	1
	Walleye	6
	White sucker	1
	Yellow perch	4(6)

Table 3. Samples collected from inland locations. Parenthetical numbers represent the pre-composite *n*, if any samples were composited prior to analysis.

Waterbody	Species	*n*
Badfish Creek (Dane County)	Common carp	2
Chippewa River: Holcombe to Mississippi River	Common carp	2
Fox River: DePere to Green Bay	Common carp	4
	Walleye	4
Fox River: Little Lake Butte des Morts to DePere	Common carp	2
Fox (IL) River	Common carp	6
Keyes Lake	Walleye	1
Lake Winnebago system	Common carp	5
Menominee River: Piers Gorge to Lower Scott Flowage	Common carp	3
Milwaukee River Estuary: Estabrook Falls to harbor mouth	Common carp	5
	Smallmouth bass	2
	Walleye	6
Milwaukee River: Grafton to Estabrook Falls	Black crappie	3
Milwaukee River: above Grafton to Newberg	Common carp	1
Mississippi River: Pool 3	Common carp	3
Mississippi River: Pool 4	Channel catfish	2
Mississippi River: Pools 10, 11, & 12	Channel catfish	2
Peshtigo River: High Falls Flowage	Black crappie	3
	Walleye	4
Pewaukee Lake	Largemouth bass	1
Rainbow Flowage	Northern pike	1
Sheboygan River: Sheboygan Falls to mouth	Common carp	4(6)
Spirit River Flowage	Black crappie	1
Upper Fox River	Common carp	3
Whitefish Lake	Walleye	1
Wisconsin River: Nekoosa to Petenwell	Common carp	2
Wisconsin River: Prairie du Sac to Mississippi River	Bighead carp	1
Wisconsin River: Wisconsin Dells to Prairie du Sac	Common carp	2
Wolf Lake	Largemouth bass	1

than the WSLH method. However, sample prep was the same between the two methods and similar relative proportions of congener types, as well as total PBDEs, were detected in species analyzed by both methods.

Comparison of WI sport fish PBDE congener proportions with industrial formulations

Because PBDEs are not naturally-occurring compounds, concentrations and proportions of congeners found in fish tissue can potentially be traced back to the commercial formulation(s) from which they originated. The top row of Figure 2 shows the congener makeup of four commercial PBDE formulations (Rayne and Ikonomou 2002). DE-71 and 70-5DE are known as Penta-BDE mixtures and were used primarily in fire retardant foam materials found in furniture (Sjödin et al. 1998). DE-79 and 79-8DE are Octa-BDE mixtures and are used in

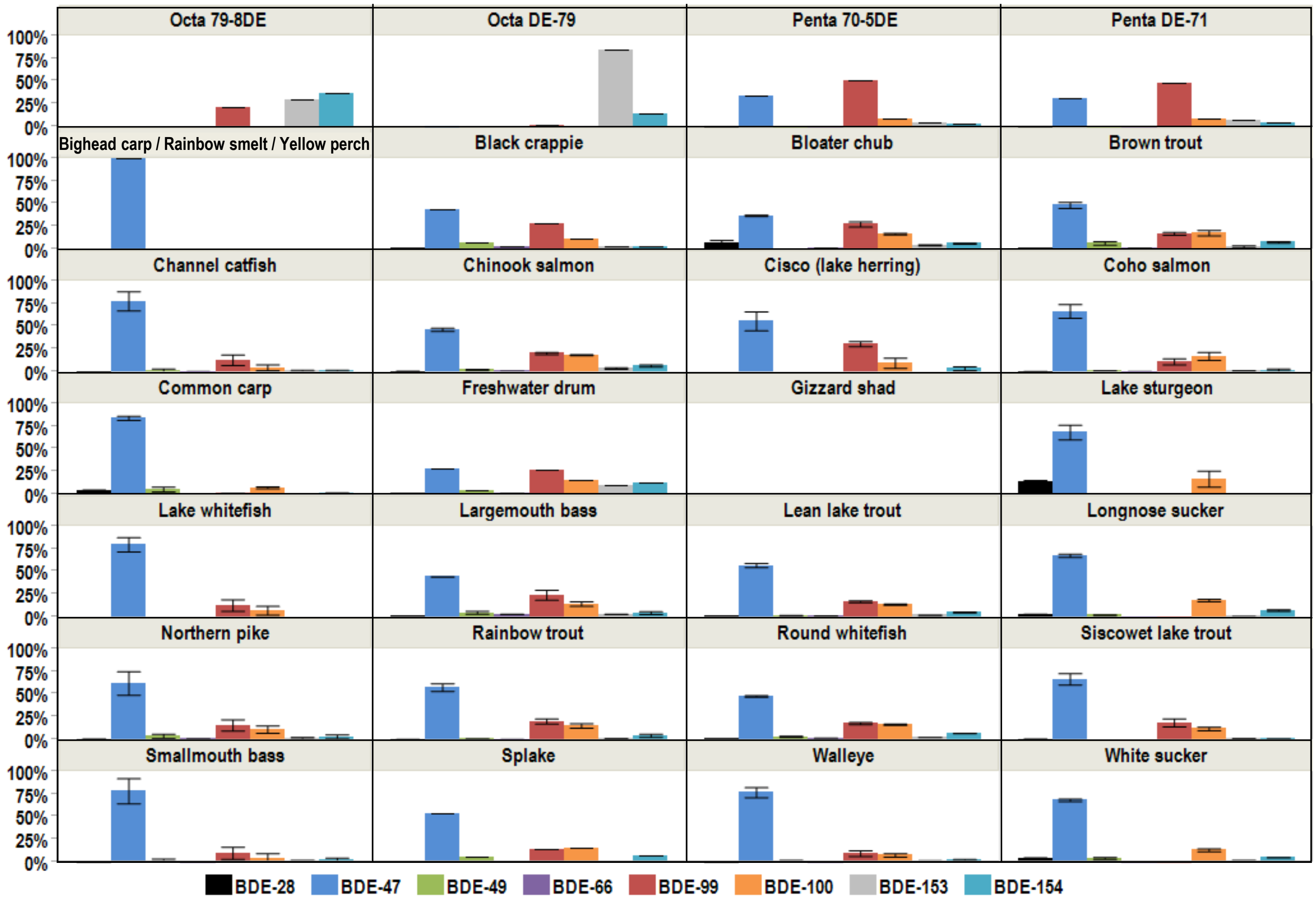

Figure 2. Congener proportions in four commercial PBDE formulations (top row; from Rayne and Ikonomou 2002) and in fish samples (mean ± SE; all sample dates, all waterbodies). Samples of bighead carp, rainbow smelt, and yellow perch contained 100% BDE-47; PBDEs were below the detection limit for all samples of gizzard shad.

plastics and small household appliances (Costa and Giordano 2007).

Figure 2 also shows the percent of ΣPBDEs made up by each congener (mean ± SE for all waterbodies and sample dates per species). Hepta-, octa-, nona-, and deca-BDEs were detected at <1% in all fillets. Among species where more than one congener was detected, BDE-47 made up the highest percentage of the total (an average of 66.2% across all species tested). Congeners 99, 100, and 154 were the second, third, and fourth most common congeners, contributing an average of 12.8%, 12.0%, and 3.7% respectively, to species' total PBDEs in this dataset. The pattern of these four congeners is consistent with previous research (Renner 2000), and mostly closely resembles the makeup of commercial Penta-BDE mixtures DE-71 and 70-5DE (top row, Figure 2). As Penta-BDE mixtures were used in fire-retardant foams and textiles, it is likely that the PBDEs present in Wisconsin sport fish are derived in large part from household products. This finding is also consistent with the fact that Penta-BDE use primarily occurred in North America (Hale et al. 2003).

PBDEs in inland waters

Among inland locations, the highest ΣPBDEs were found in fish fillets from waterbodies with high industrial / manufacturing use or whose flow is highly dominated by effluent, such as the Fox and Sheboygan Rivers, Milwaukee River Estuary, and Badfish Creek (Figure 3). When more than one species was sampled from a location, fillets from fattier species generally contained higher PBDE concentrations (Figure 4). Common carp fillets contained the highest concentration of ΣPBDEs (max concentration 482 ng/g from Badfish Creek, Figure 3). The maximum concentration of ΣPBDEs found in all other inland species was 40.9 ng/g in a channel catfish sample from the Mississippi

River. This accumulation pattern is similar to that found in other lipophilic contaminants like PCBs, where fattier fish contain a higher concentration of contaminants than leaner fish. However, because of the extremely heterogeneous nature of PBDE contamination, sampling location is a better predictor of ΣPBDE concentration than size or lipid content in this dataset.

PBDEs in Lakes Michigan & Superior

In Lake Michigan, the highest concentrations of ΣPBDEs were found in fillets of fish from the southern basin offshore of Milwaukee (WI) and Grand Rapids (MI; Figure 5a). The maximum PBDE concentration was 356.1 ng/g in a lake trout fillet; in contrast, the highest non-salmonid concentration was only 14.1 ng/g in a freshwater drum sample (Figure 6). Partitioning samples into salmonid (Figure 5b) and non-salmonid (Figure 5c) species reveals that the overall trend in ΣPBDEs in Lake Michigan seen in Figure 5a is strongly influenced by PBDEs in fattier salmonid species, which are more likely to accumulate these lipophilic contaminants (Figure 6).

In the US waters of Lake Superior, fillet PBDE concentrations were generally lower than in Lake Michigan, with the exception of siscowet lake trout (not found in Lake Michigan). The

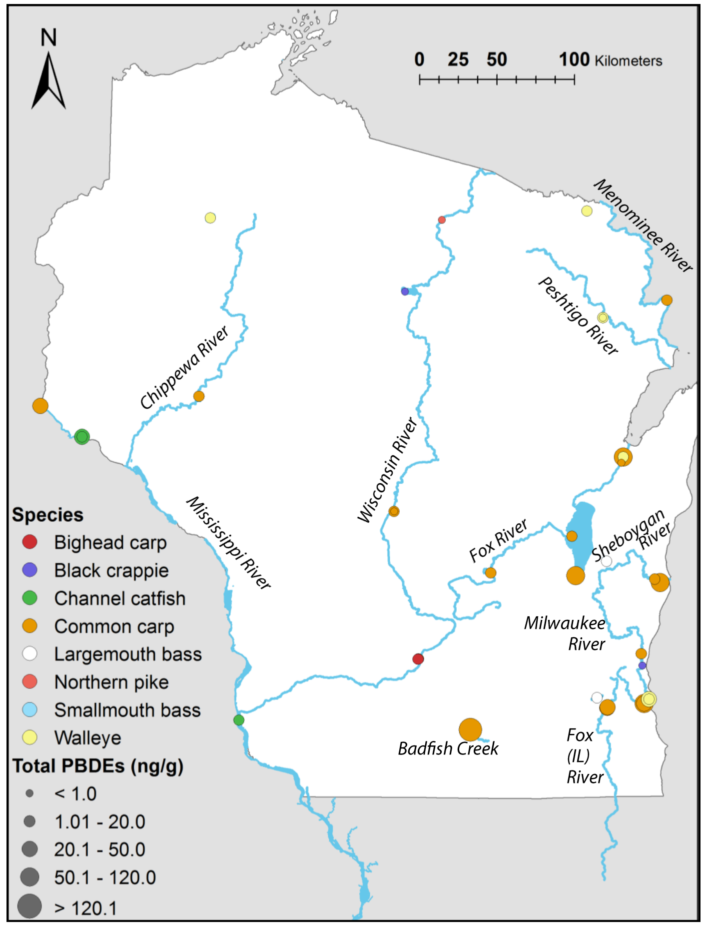

Figure 3. Spatial distribution of ΣPBDEs in fillets of species sampled from inland waters.

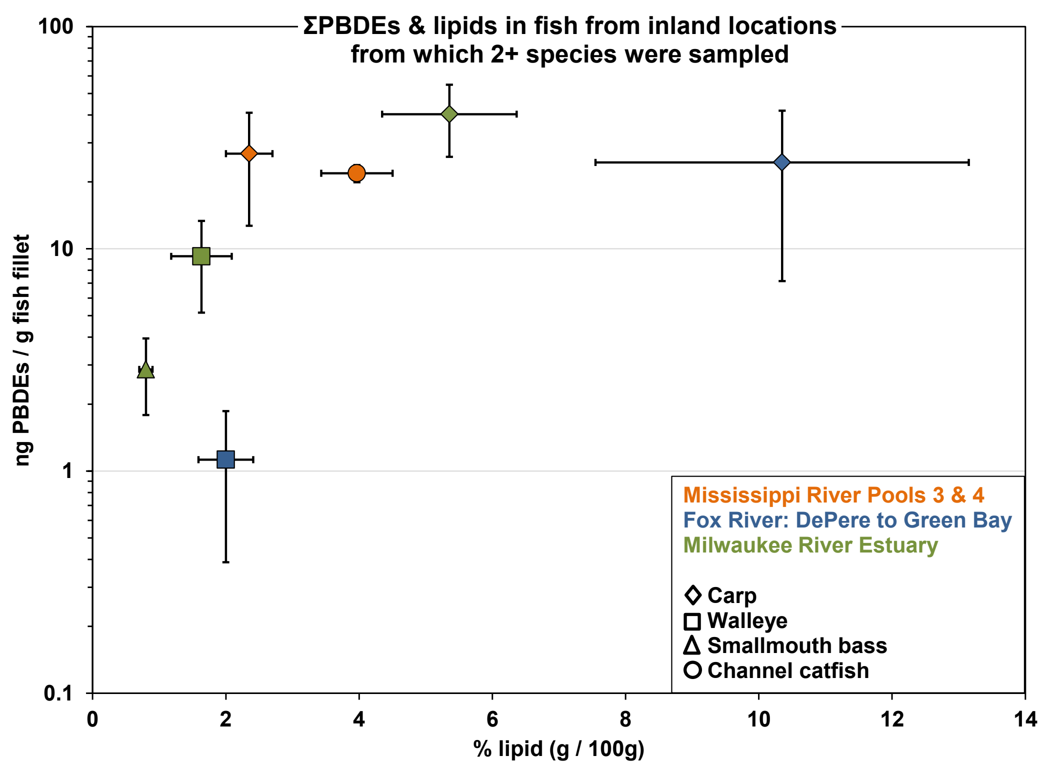

Figure 4. Total PBDE concentrations and lipid content in fish fillets from inland locations where more than one species was sampled.

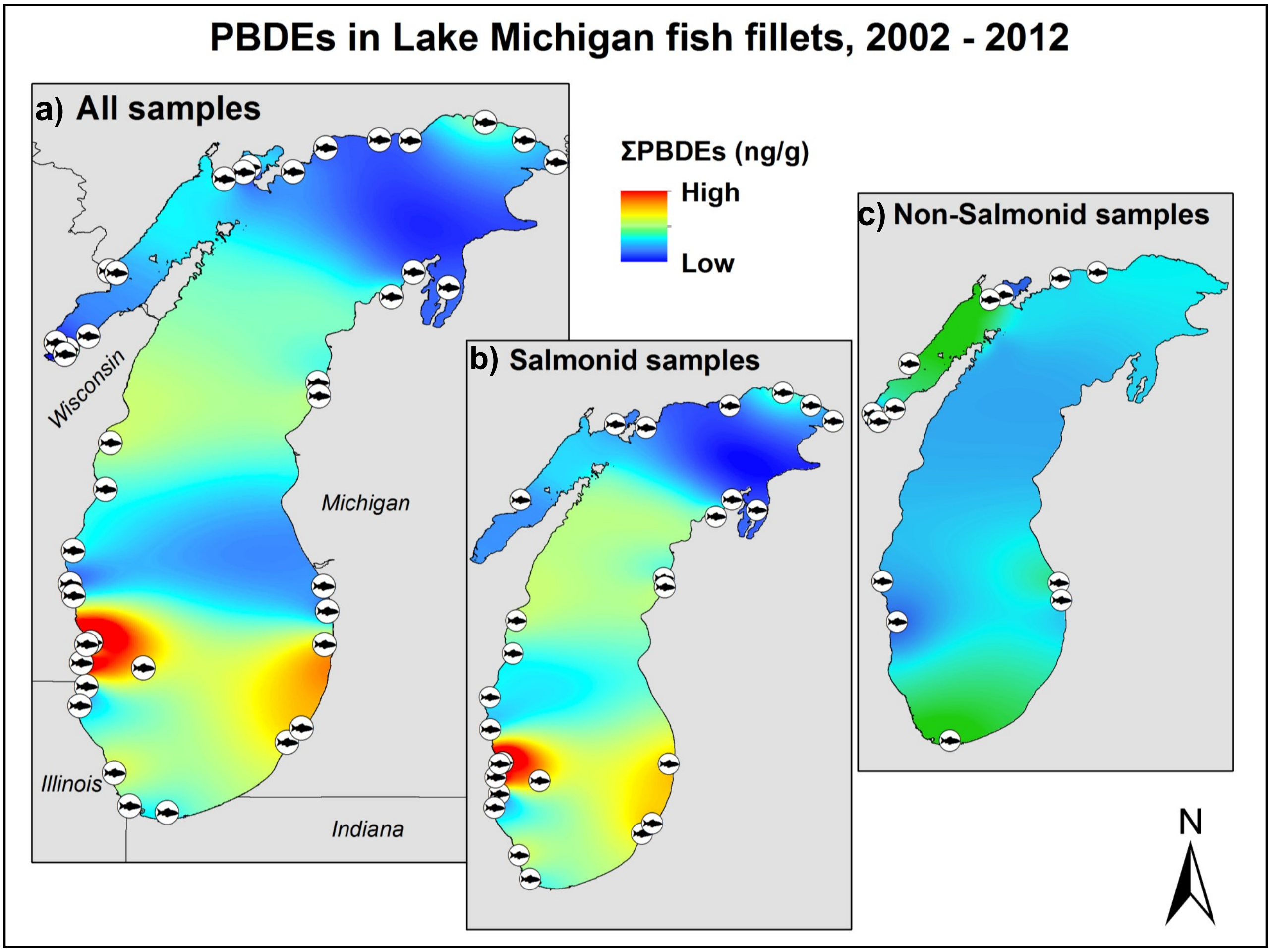

Figure 5. Spatial distribution of PBDEs in fillets of fish sampled from Lake Michigan and Green Bay between 2002 and 2012: a) all samples, b) samples of Salmonid species (bloater chub, salmon species, trout species, and whitefish species), and c) samples of non-Salmonid species (bass, catfish, carp, drum, perch, pike, shad, smelt, sturgeon, sucker, and walleye).

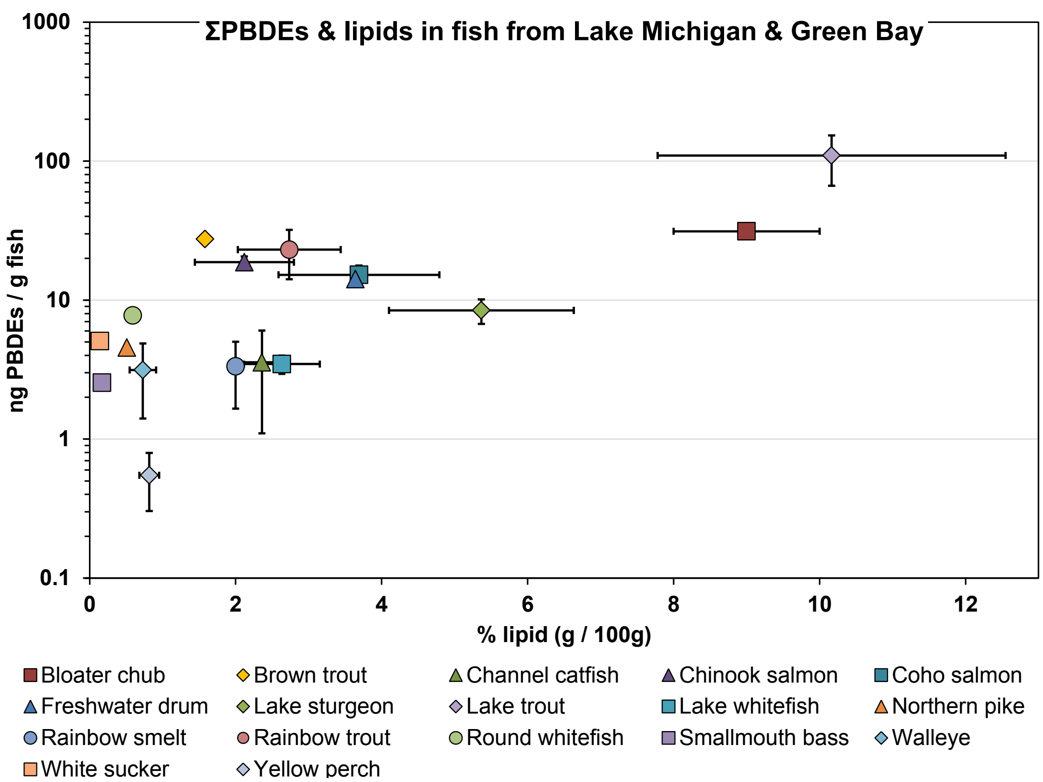

Figure 6. Total PBDE concentrations and lipid content (mean ± SE) in fish sampled from Lake Michigan and Green Bay. PBDES were not found in any sample of carp or gizzard shad.

ΣPBDE represents the sum of 9 congeners for bloater chub and rainbow smelt, and the sum of 17 congeners for all other species.

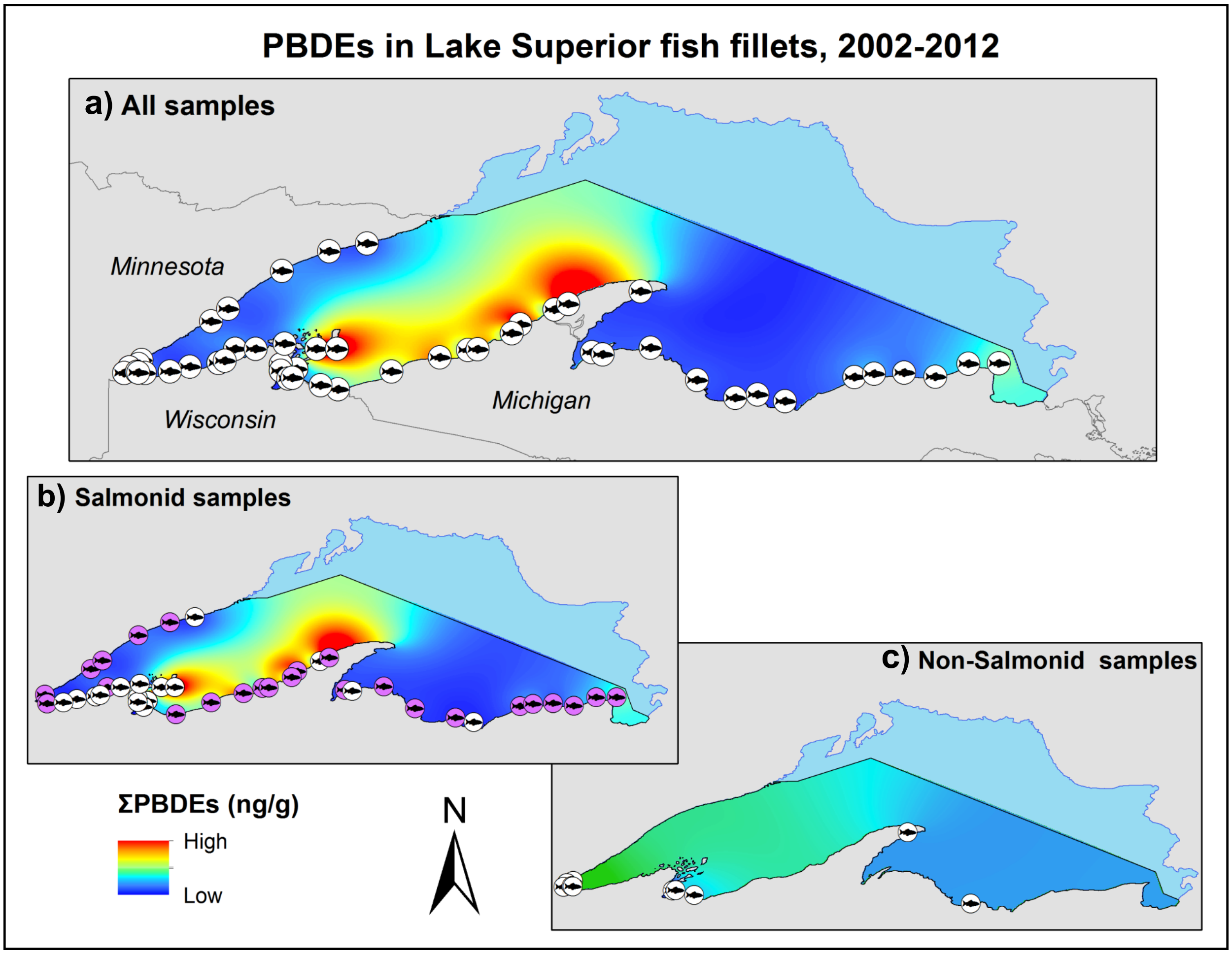

Figure 7. Spatial distribution of PBDEs in fish sampled from Lake Superior between 2002 and 2012: a) all samples, b) samples of Salmonid species (bloater chub, cisco, salmon species, trout species, and whitefish species; purple icons = samples of lake trout, which were the most-widely sampled Salmonid species), and c) samples of non-Salmonid species (carp, perch, pike, sucker, and walleye).

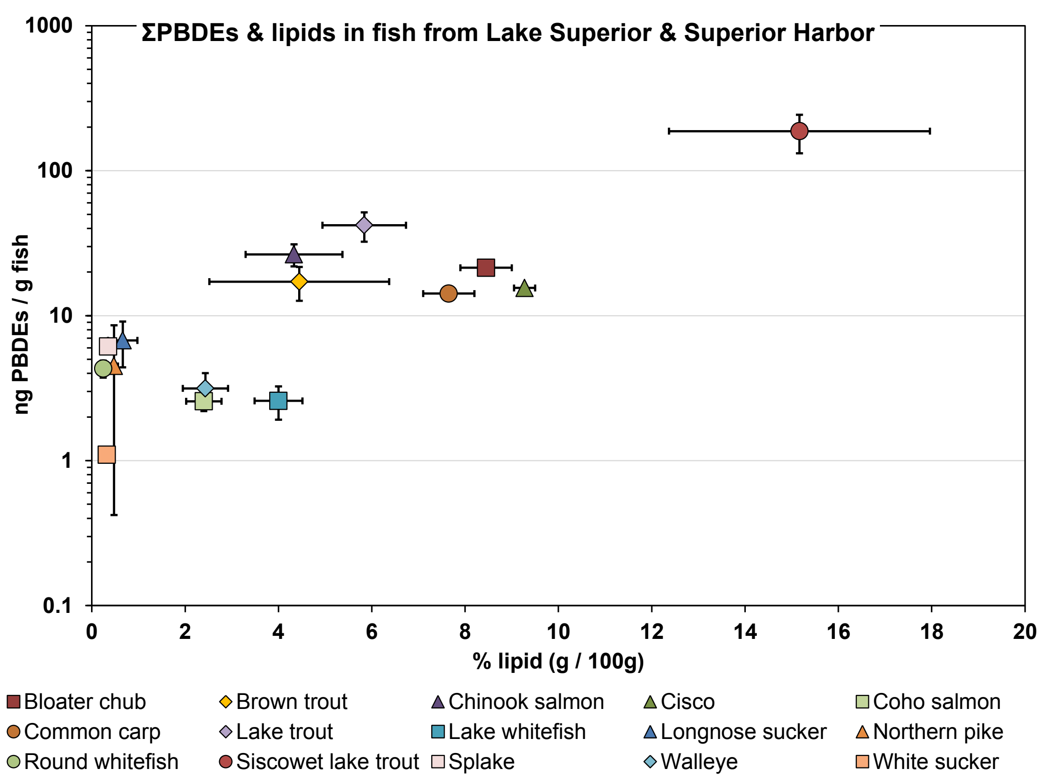

Figure 8. Total PBDE concentrations and lipid content (mean ± SE) in fish sampled from Lake Superior. PBDEs were not found in any yellow perch sample.

ΣPBDE represents the sum of 9 congeners for bloater chub and cisco, and the sum of 17 congeners for all other species .

highest concentrations of ΣPBDEs were found in fish sampled offshore of the Apostle Islands (WI) and Keweenaw Peninsula (MI, Figure 7a). The maximum concentration found was 398.9 ng/g in a siscowet lake trout sample, whereas the highest non-salmonid concentration was 16.7 ng/g in a northern pike sample (Figure 8). As with Lake Michigan fish, ΣPBDEs generally increased with lipid content (Figure 8). It should be noted that Lake Superior samples are composed mainly of salmonid fillets, particularly lake trout (purple icons, Figure 7b), and this likely influenced the overall trend in PBDE concentrations seen in Figure 7a compared to non-salmonid samples (Figure 7c). However, the fact that concentrations varied spatially among lake trout supports the idea that PBDE contamination is heterogeneous in Lake Superior waters.

Temporal trends in ΣPBDEs and BDE congeners in Great Lakes fish

There were 5 Lake Superior and 3 Lake Michigan fish species for which multiple samples were taken over multiple years, allowing for the investigation of temporal trends in both proportions of congeners (Figure 9) as well as ΣPBDE concentrations (Figure 10). Hepta– through deca–BDEs were not measured prior to 2010 and accounted for <1% of the total. They are therefore not included in Figure 9, although their concentrations are included in the measurement of ΣPBDEs shown in Figure 10.

In species sampled multiple times from Lakes Michigan or Superior, there was < 2% change in the proportion of total PBDEs made up by tri-BDE #28 through time (Figure 9). Almost all species displayed a decline in the proportion of tetra-BDEs (#47, #49, #66), with a maximum reduction of 43% between 2005 and 2011 in Lake Superior lake trout. Conversely, there was an overall trend of both penta- (#99, #100) and hexa-BDEs (#138, #153, #154) making up an increasing proportion of total PBDEs through time. For tri-, tetra-, and penta-BDEs, the magnitude of change was lower for Lake Michigan species than for those sampled from Lake Superior (Figure 9).

Given that Great Lakes species' proportions of congeners displayed consistent temporal changes, it would be reasonable to expect that ΣPBDE concentrations would also vary temporally. However, ΣPBDEs were not significantly different between time points for almost any Great Lakes species (Figure 10). The exception was rainbow trout from Lake Michigan, which contained significantly lower ΣPBDE concentrations in 2010 and 2012 than in 2004 (Figure 10), although these differences should be interpreted with caution due to differences in size (or possibly gender, see Madenjian et al. 2012) of fish collected in each year (Appendix I). It is also possible that samples of the other species were not collected adequately far enough apart to see distinct trends in ΣPBDEs.

This report documents increases in penta- and hexa-BDEs, decreases in tetra-BDEs, no change in tri-BDEs, and no significant overall changes in ΣPBDEs through time in fish sampled from Lakes Michigan and Superior. The temporal trends in this report were compared with trends found in a study by Crimmins et al. (2012), who investigated PBDE trends in whole lake trout from the Great Lakes between 1980-2009. Crimmins et al. (2012) found a statistically significant difference in PBDE accumulation trends in Lake Michigan fish, with concentrations increasing before 2000-2001 and decreasing or remaining the same thereafter. Similar, though not statistically significant, patterns were found in Lake Superior fish. It appears that this study continues to document the trend of slowing PBDE accumulation in the Great Lakes observed by Crimmins et al. (2012), although more long-term monitoring would be needed to fully document this phenomenon.

These data also document temporal patterns in congener proportions suggestive of a shift from Penta-BDE-dominated sources to Octa-BDE-dominated sources (see also Zhu and Hites 2004, and Batterman et al. 2007). We hypothesize two possible mechanisms for this apparent shift in PBDE source, and recognize that the two mechanisms may be happening concurrently. First, although production of both Penta– and Octa– formulations ceased in 2004, it is possible that trends documented here reflect differences in the breakdown times of polyurethane foams (containing Penta-mixtures) compared to electronics (containing Octa- mixtures). As foams are more easily broken down, fish sampled temporally closer to the PBDE ban might experience inputs from household and office furniture foams, while those sampled several years later may experience inputs from computers and appliances. Second, Deca-BDE mixtures were not phased out of production until 2013 (Gewurtz et al. 2011), and Deca-BDE can be

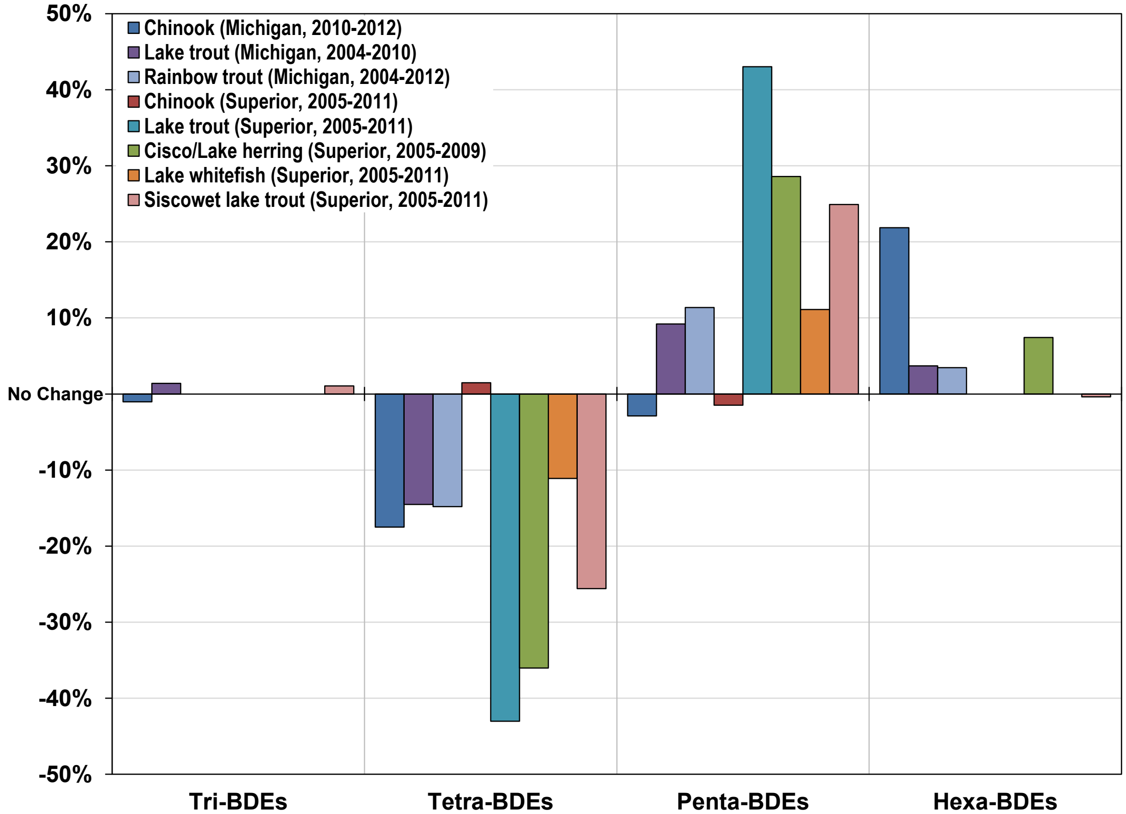

Figure 9. Trends in the proportion of total PBDEs made up by congeners in fish species that were sampled through time from Lakes Michigan or Superior.

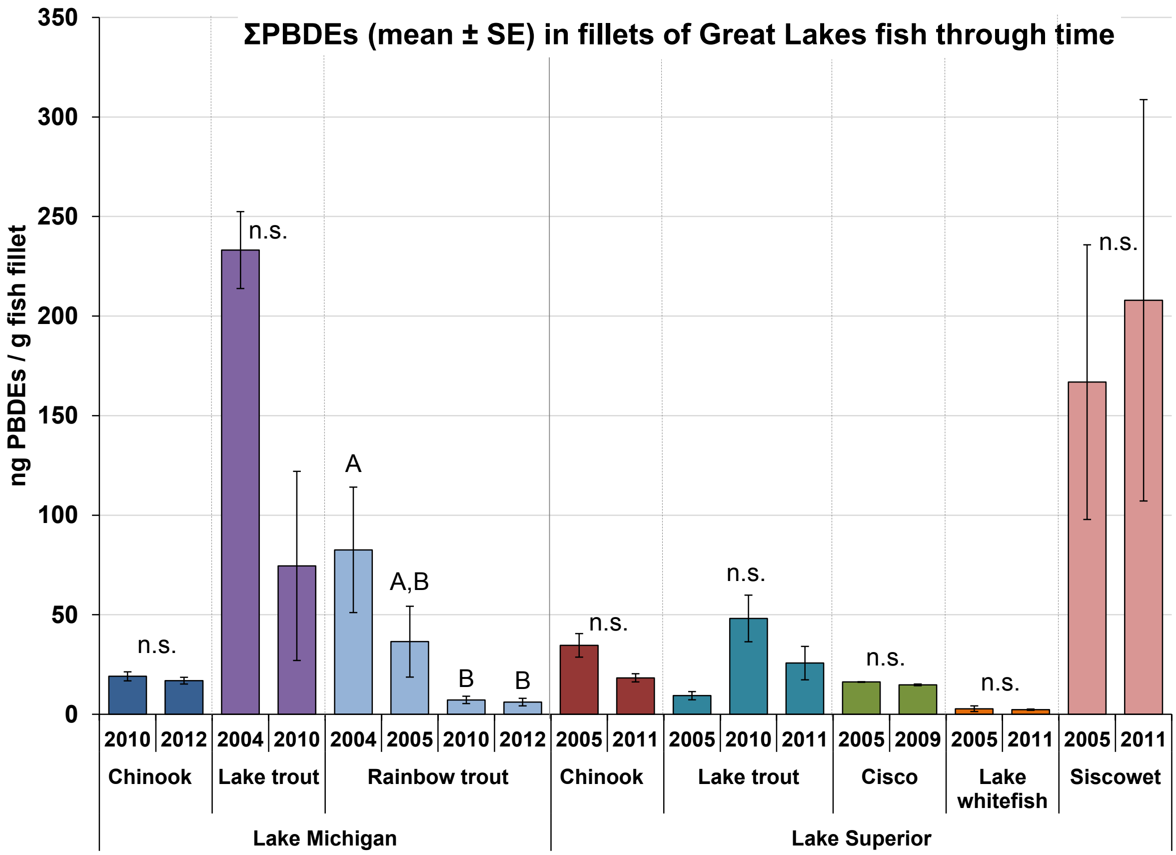

Figure 10. Total PBDEs found in fillets of Great Lakes fish species that were sampled multiple times. Letters indicate levels of significance as determined by an ANOVA with post-hoc Tukey's HSD test (n.s. = difference between years is not significant).

debrominated in the food web to (hexa) BDE-153 (Stapleton et al. 2004), which is a major component of Octa-BDE commercial mixtures. It may therefore only appear as though Octa-BDE commercial mixtures are becoming the source of PBDEs in the Great Lakes, while instead this trend reflects the gradual breakdown of Deca-BDE mixtures.

Possible implications for Wisconsin fish consumption advisories

Assessing risk based on ΣPBDEs is difficult, since lower brominated congeners are more toxic than higher brominated congeners and varying concentrations of each congener are found in different species from different locations. As such, only a few references doses (RfDs) are currently available upon which consumption recommendations could be based: California's Environmental Protection Agency determines risk based on ΣPBDEs by calculating Advisory Tissue Levels (Klasing and Brodberg 2011), while the USEPA has formulated RfDs for four congeners: 47 and 99 at 1×10^{-4} mg/kg/day, 153 at 2×10^{-4} mg/kg/day, and 209 at 7×10^{-3} mg/kg/day (IRIS 2008).

For this report, we calculated the mass (mean ± SE) of each congener that would be present in an 8oz serving of each species sampled from a given water (see Appendix III) and compared that to the RfD for a 150lb adult. We also placed fish into theoretical meal categories based on both the California EPA's Advisory Tissue Levels and the Great Lakes Consortium for Fish Consumption Advisory's mercury meal categories (Anderson et al. 2007), since the lowest RfD (for BDE-47, which is generally present in highest concentrations in fish), is equal to the RfD for mercury.

We found that a serving of most fillets from most locations would not exceed the RfD (values in bold exceed the RfD for that congener in Appendix III). For BDE-47, exceptions include lake trout from the Great Lakes and carp from Badfish Creek in Dane County, the Milwaukee River Estuary, and the Sheboygan River. For BDE-99, only a serving of siscowet lake trout from Lake Superior exceeded the RfD. RfDs for BDE-153 or BDE-209 were not exceeded for a serving of any fish from any location.

Most waters sampled as part of this study already carry exceptions to Wisconsin's general statewide consumption advice due to concerns about PCB and/or mercury contamination, and it is likely that future advisories will continue to be based on those contaminants rather than on PBDEs. Even though some fish from some locations exceeded the RfD for individual congeners, ΣPBDE levels in most fish were not high enough to trigger exceptions to our statewide advice. In fact, all fillet samples except those from one location would fall into the CA EPA's "3 8-oz. servings/week" advice category and the Consortium's "unrestricted" advice category based on their ΣPBDE levels. The lone exception, carp from Badfish Creek, would fall into the CA EPA's "2 8-oz. servings/week" advice category and the Consortium's "1 meal/week" advice category, whereas Wisconsin's current advice due to PCB contamination puts these fish into a "1 meal/month" category. However, there is a continued need for monitoring of contamination patterns, particularly given that PCB concentrations have been declining in some types of fish from Lakes Michigan and Superior (Bhavsar et al, 2007; Rasmussen et al. 2014).

Recommendations for future work

PBDE concentrations in Wisconsin sport fish species warrant additional monitoring given their toxicity to fish (Yu et al. 2011), wildlife (Route et al. 2014), and humans (Turyk et al. 2008). Although it might be expected for concentrations of banned substances to decrease though time, the fate of PBDEs, particularly higher brominated congeners, in the environment is uncertain. A standardized temporal monitoring schedule should be endorsed in locations where the highest fish PBDE concentrations are found, including increasing sample sizes where budget and time permit. It may also be valuable for future research efforts to investigate the synergistic or antagonistic effects of exposure to multiple congeners in fish fillets, as synergistic effects have been previously documented in human cells (Tagliaferri et al. 2010). Finally, the EPA Great Lakes National Program Office's monitoring efforts should continue to analyze for PBDEs in fish tissue samples.

Acknowledgments

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Appendix I: Detailed information for samples collected from Lakes Michigan and Superior. Parenthetical numbers represent the pre-composite *n*, if any samples were composited prior to analysis.

Waterbody	Species	*n*	Year	Fish Length (in) mean (min, max)	Fillet Lipid (g/100g) mean ± SE	Fillet ΣPBDEs (ng/g) mean ± SE
Lake Michigan (includes Green Bay)	Bloater chub	2 (60)	2009	9.4 (9.2, 9.5)	9.0 ± 1.0	31.25 ± 0.55
	Brown trout	1	2010	20.1	1.6	27.46
	Channel catfish	1	2010	17.4	2.0	6.04
		1	2011	18.5	2.7	1.10
	Chinook salmon	15 (17)	2010	26.9 (16.3, 35)	2.5 ± 0.79	19.09 ± 2.27
		3	2012	30.8 (28.1, 34.3)	0.46 ± 0.02	16.90 ± 1.71
	Coho salmon	5	2010	24.5 (20.2, 27.0)	3.7 ± 1.1	15.20 ± 2.54
	Common carp	2	2005	23.9 (22.5, 27.0)	3.0 ± 0.65	0 ± 0
	Freshwater drum	1	2010	16.3	3.6	14.15
	Gizzard shad	3 (90)	2005	4.53 (3.9, 5.0)	3.0 ± 0.21	0 ± 0
	Lake sturgeon	3	2011	50.9 (50.5, 51.6)	5.4 ± 1.3	8.43 ± 1.68
	Lake trout	2	2004	31.3 (30.5, 32.0)	17.0 ± 3.0	233.15 ± 19.35
		7 (13)	2010	24.2 (18.3, 29.5)	8.2 ± 2.5	74.54 ± 47.50
	Lake whitefish	3	2012	17.63 (16.5, 18.8)	2.6 ± 0.52	3.47 ± 0.52
	Northern pike	1	2010	25.7	0.51	4.56
	Rainbow smelt	3 (30)	2005	4.67 (4.2, 5.2)	2.0 ± 0.06	3.33 ± 1.68
	Rainbow trout	2	2004	30.2 (30.2, 30.2)	2.2 ± 1.6	82.60 ± 31.50
		2	2005	26.9 (25.1, 28.6)	2.0 ± 0.70	36.50 ± 17.80
		6	2010	26.0 (23.0, 29.8)	1.78 ± 0.92	7.25 ± 1.86
		3	2012	18.8 (14.2, 24.9)	5.5 ± 1.6	6.13 ± 1.94
	Round whitefish	1	2010	19.5	0.59	7.75
	Smallmouth bass	1	2010	17.1	0.17	2.54
	Walleye	2	2005	14.4 (13.0, 15.8)	1.4 ± 0.35	0 ± 0
		3 (9)	2010	23.9 (20.0, 28.3)	0.19 ± 0.07	9.43 ± 2.54
		4	2011	16.7 (15.0, 21.0)	0.83 ± 0.18	0 ± 0
	White sucker	1	2010	16.32	0.14	5.08
	Yellow perch	3	2010	10.6 (10.0, 11.4)	1.1 ± 0.06	1.10 ± 0
		3	2011	13 (11.3, 15.0)	0.53 ± 0.09	0 ± 0
Lake Superior (includes the St. Louis River/ Superior Harbor)	Bloater chub	2 (10)	2009	8.5 (8.4, 8.5)	8.5 ± 0.55	21.40 ± 0.20
	Brown trout	2	2009	21.6 (19.7, 23.4)	6.1 ± 1.9	18.65 ± 7.35
		1	2010	22.9	1.2	14.19
	Chinook salmon	3	2005	30.8 (28.0, 33.7)	5.3 ± 1.9	34.63 ± 5.88
		3	2011	31.6 (27.9, 33.6)	3.3 ± 0.95	18.30 ± 2.04
	Cisco / lake herring	2 (20)	2005	7.7 (7.1, 8.3)	9.6 ± 0.30	16.25 ± 0.15
		2 (20)	2009	9.5 (9.3, 9.7)	9.0 ± 0.05	14.80 ± 0.40
	Coho salmon	3	2011	22.3 (21.2, 23.7)	2.4 ± 0.38	2.57 ± 0.37
	Common carp	2	2012	29.8 (26.0, 33.5)	7.7 ± 0.55	14.20 ± 1.50
	Lake trout	3	2005	20.1 (19.2, 20.7)	10.0 ± 0.52	9.37 ± 2.08
		24 (88)	2010	21.9 (18.3, 27.0)	5.3 ± 1.1	48.15 ± 11.67
		3	2011	24.0 (21.2, 26.9)	6.1 ± 1.2	25.73 ± 8.36
	Lake whitefish	3	2005	19.4 (18.7, 19.8)	3.0 ± 0.38	2.80 ± 1.44
		3	2011	19.0 (17.2, 21.1)	5.0 ± 0.48	2.37 ± 0.32
	Longnose sucker	5 (15)	2010	18.3 (15.5, 19.7)	0.66 ± 0.32	6.75 ± 2.35
	Northern pike	4	2012	26.4 (19.3, 34.0)	0.48 ± 0.09	4.50 ± 4.08
	Round whitefish	6 (24)	2010	15.1 (12.9, 19.8)	0.24 ± 0.04	4.32 ± 0.58
	Siscowet lake trout	3	2005	22.4 (20.0, 24.0)	17.7 ± 5.6	166.83 ± 68.96
		3	2011	26.2 (23.9, 28.3)	12.7 ± 1.2	207.93 ± 100.84
	Splake	1	2010	17.0	0.35	6.13
	Walleye	2	2005	22.0 (21.9, 22.0)	3.0 ± 1.2	2.95 ± 2.95
		4	2012	23.0 (21.2, 25.8)	2.2 ± 0.55	3.25 ± 0.66
	White sucker	1	2010	16.3	0.32	1.10
	Yellow perch	4 (6)	2011	8.9 (7.9, 10.0)	0.23 ± 0.03	0 ± 0

Appendix II: Detailed information for samples collected from inland locations. Parenthetical numbers represent the pre-composite *n*, if any samples were composited prior to analysis.

Waterbody	Species	*n*	Year	Fish Length (in) mean (min, max)	Fillet Lipid (g/100g) mean ± SE	Fillet ΣPBDEs (ng/g) mean ± SE
Badfish Creek (Dane County)	Common carp	2	2003	19.0 (17.6, 20.3)	3.4 ± 0.15	466.0 ± 16.0
Chippewa River: Holcombe to Mississippi River	Common carp	2	2003	26.6 (24.6, 28.5)	10.9 ± 2.1	5.35 ± 1.05
Fox River: DePere to Green Bay	Common carp	4	2012	25.5 (22.7, 27.8)	10.4 ± 2.8	26.72 ± 18.97
	Walleye	4	2012	21.4 (16.8, 26.7)	2.0 ± 0.41	1.13 ± 0.74
Fox River: Little Lake Butte des Morts to DePere	Common carp	2	2003	17.5 (17.5, 17.5)	0.30 ± 0	0 ± 0
Fox (IL) River	Common carp	6	2007	19.7 (14.9, 22.8)	2.1 ± 0.49	18.10 ± 6.28
Keyes Lake	Walleye	1	2003	22.0	3.0	3.45
Lake Winnebago system	Common carp	2	2003	25.7 (24.5, 26.8)	12.8 ± 4.2	46.45 ± 21.15
		3	2009	18.1 (17.2, 18.8)	4.4 ± 0.87	1.33 ± 0.15
Menominee River: Piers Gorge to Lower Scott Flowage	Common carp	3	2012	25.8 (21.5, 30.6)	3.8 ± 0.97	3.57 ± 1.17
Milwaukee River Estuary: Estabrook Falls to harbor mouth	Common carp	1	2002	21.6	4.5	38.80
		4	2012	23.1 (22.2, 23.9)	5.6 ± 1.3	40.73 ± 18.61
	Smallmouth bass	2	2012	13.9 (12.1, 15.6)	0.85 ± 0.15	2.87 ± 1.08
	Walleye	6	2012	18.3 (16.5, 21.5)	1.6 ± 0.45	9.25 ± 4.09
Milwaukee River: above Grafton to Newberg	Common carp	1	2002	20.0	4.5	11.00
Milwaukee River: Grafton to Estabrook Falls	Black crappie	3	2012	10.3 (10.0, 10.8)	0.33 ± 0.09	0 ± 0
Mississippi River: Pool 3	Common carp	3	2009	22.9 (22.5, 23.3)	4.0 ± 0.54	21.87 ± 1.93
Mississippi River: Pool 4	Channel catfish	2	2002	22.1 (20.5, 23.6)	2.4 ± 0.35	26.80 ± 14.10
Mississippi River: Pools 10, 11, & 12	Channel catfish	2	2002	21.0 (17.0, 24.9)	3.8 ± 0.60	6.10 ± 3.00
Peshtigo River: High Falls Flowage	Black crappie	3	2011	10.0 (9.5, 10.9)	Not measured	0 ± 0
	Walleye	4	2011	21.8 (19.6, 23.2)	Not measured	0.93 ± 0.93
Pewaukee Lake	Largemouth bass	1	2003	14.2	0.34	1.07
Rainbow Flowage	Northern pike	1	2003	20.9	0.44	0.33
Sheboygan River: Sheboygan Falls to mouth	Common carp	4(6)	2004	21.0 (19.4, 25.0)	3.1 ± 0.57	51.78 ± 27.68
Spirit River Flowage	Black crappie	1	2003	12.3	1.5	0.38
Upper Fox River	Common carp	3	2006	20.1 (20.0, 23.5)	4.5 ± 1.4	1.50 ± 0.30
Whitefish Lake	Walleye	1	2003	16.6	1.3	3.57
Wisconsin River: Nekoosa to Petenwell	Common carp	2	2003	24.4 (23.7, 25.0)	1.2 ± 0.20	3.60 ± 3.60
Wisconsin River: Prairie du Sac to Mississippi River	Bighead carp	1	2011	44.5	6.5	1.40
Wisconsin River: Wisconsin Dells to Prairie du Sac	Common carp	2	2003	26.3 (22.8, 29.7)	9.1 ± 5.9	3.10 ± 3.10
Wolf Lake	Largemouth bass	1	2003	11.7	0.42	1.75

Appendix III: Mass (µg) of congener / 0.5 lb serving of fish (mean ± SE). Values in bold exceed the RfD for a 150lb adult (6.80 µg/kg/day for BDEs 47 and 99, 13.6 µg/kg/day for BDE-153, and 476.3 µg/kg/day for BDE-209).

Waterbody	Species	BDE-47	BDE-99	BDE-153	BDE-209
Badfish Creek	Common carp	**86.18 ± 2.27**	0 ± 0	0 ± 0	0
Chippewa River	Common carp	1.21 ± 0.24	0 ± 0	0 ± 0	0
Fox (IL) River	Common carp	3.50 ± 1.18	0 ± 0	0 ± 0	0
Fox River: DePere to Green Bay	Common carp	4.15 ± 2.95	0.52 ± 0.37	0 ± 0	0
	Walleye	0.26 ± 0.17	0 ± 0	0 ± 0	0
Fox River: Little Lake Butte des Morts to DePere	Common carp	0 ± 0	0 ± 0	0 ± 0	0
High Falls Flowage	Black crappie	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	Walleye	0.10 ± 0.10	0.11 ± 0.11	0 ± 0	0 ± 0
Keyes Lake	Walleye	0.48	0.10	0.024	
Lake Winnebago system	Common carp	3.63 ± 2.37	0 ± 0	0 ± 0	
Menominee River	Common carp	0.55 ± 0.12	0 ± 0	0 ± 0	0 ± 0
Milwaukee River: Grafton to Estabrook Falls	Black crappie	0 ± 0	0 ± 0	0 ± 0	0 ± 0
Milwaukee River: above Grafton to Newberg	Common carp	2.50	0	0	
Milwaukee River Estuary	Common carp	**6.39 ± 2.24**	0 ± 0	0 ± 0	0 ± 0
	Smallmouth bass	0.54 ± 0.14	0.11 ± 0.11	0 ± 0	0 ± 0
	Walleye	1.45 ± 0.57	0.33 ± 0.19	0 ± 0	0 ± 0
Mississippi River Pool 3	Common carp	3.02 ± 0.33	0 ± 0	0 ± 0	
Mississippi River Pool 4	Channel catfish	3.72 ± 1.72	1.55 ± 0.67	0 ± 0	
Mississippi River Pools 10, 11, and 12	Channel catfish	1.38 ± 0.68	0 ± 0	0 ± 0	
Pewaukee Lake	Largemouth bass	0.11	0.05	0.008	
Rainbow Flowage	Northern pike	0.03	0.02	0.002	
Sheboygan River	Common carp	**8.73 ± 4.43**	0 ± 0	0 ± 0	0
Spirit River Flowage	Black crappie	0.04	0.02	0.002	
Upper Fox River	Common carp	0.34 ± 0.07	0 ± 0	0 ± 0	
Whitefish Lake	Walleye	0.41	0.18	0.03	
Wisconsin River: Nekoosa to Petenwell	Common carp	0.70 ± 0.70	0 ± 0	0 ± 0	
Wisconsin River: Prairie du Sac to Mississippi River	Bighead carp	0.32	0	0	0
Wisconsin River: Wisconsin Dells to Prairie du Sac	Common carp	0.82 ± 0.82	0 ± 0	0 ± 0	
Wolf Lake	Largemouth bass	0.177	0.12	0.01	
Lake Michigan (including Green Bay)	Bloater chub	2.72 ± 0	1.60 ± 0.03	0.25 ± 0	0
	Brown trout	2.63	0.90	0.33	
	Channel catfish	0.40 ± 0.15	0.16 ± 0.16	0.04 ± 0.04	0.007 ± 0.007
	Chinook salmon	1.90 ± 0.23	0.74 ± 0.07	0.21 ± 0.04	0.006 ± 0.004
	Coho salmon	1.82 ± 0.24	0.62 ± 0.11	0.06 ± 0.04	0.0007 ± 0.0004
	Common carp	0 ± 0	0 ± 0	0 ± 0	0
	Freshwater drum	0.92	0.84	0.34	0.002
	Gizzard shad	0 ± 0	0 ± 0	0 ± 0	
	Lake sturgeon	1.30 ± 0.26	0 ± 0	0 ± 0	0 ± 0
	Lake trout	**16.56 ± 6.89**	3.10 ± 1.24	0.60 ± 0.23	0.002 ± 0.001
	Lake whitefish	0.44 ± 0.02	0.26 ± 0.02	0 ± 0	0 ± 0
	Northern pike	0.49	0.13	0.04	0.003
	Rainbow smelt	0.76 ± 0.38	0 ± 0	0 ± 0	
	Rainbow trout	3.05 ± 1.28	0.82 ± 0.24	0.08 ± 0.06	0.0008 ± 0.0005
	Round whitefish	0.92	0.25	0.05	0.005
	Smallmouth bass	0.24	0.07	0.03	0.003
	Walleye	0.31 ± 0.17	0.10 ± 0.05	0.04 ± 0.02	0.0007 ± 0.0005
	White sucker	0.80	0.006	0.03	0.003
	Yellow perch	0.12 ± 0.06	0 ± 0	0 ± 0	0 ± 0
Lake Superior (including St. Louis River/Superior Harbor)	Bloater chub	1.76 ± 0.03	1.58 ± 0.03	0.27 ± 0	
	Brown trout	2.00 ± 0.52	0.69 ± 0.17	0.10 ± 0.07	
	Chinook salmon	3.26 ± 0.61	1.69 ± 0.30	0 ± 0	0 ± 0
	Cisco	2.00 ± 0.42	1.08 ± 0.07	0 ± 0	
	Coho salmon	0.49 ± 0.02	0 ± 0	0 ± 0	0 ± 0
	Common carp	2.22 ± 0.27	0.32 ± 0.05	0 ± 0	0 ± 0
	Lake trout	4.85 ± 1.03	2.09 ± 0.54	0.23 ± 0.07	0.003 ± 0.0006
	Lake whitefish	0.55 ± 0.15	0 ± 0	0 ± 0	0 ± 0
	Longnose sucker	1.06 ± 0.39	0.001 ± 0.0007	0.01 ± 0.003	0.003 ± 0.0006
	Northern pike	0.62 ± 0.52	0.27 ± 0.27	0 ± 0	0 ± 0
	Round whitefish	0.46 ± 0.06	0.19 ± 0.03	0.02 ± 0.004	0.013 ± 0.01
	Siscowet lake trout	**28.01 ± 8.25**	**8.17 ± 3.69**	0.80 ± 0.47	0 ± 0
	Splake	0.75	0.19	0.03	
	Walleye	0.68 ± 0.18	0 ± 0	0 ± 0	0 ± 0
	White sucker	0.17		0.004	0.003
	Yellow perch	0 ± 0	0 ± 0	0 ± 0	0 ± 0