TEMPORAL (1983-2005) AND SPATIAL TRENDS OF POLYBROMINATED DIPHENYL ETHERS IN GREAT LAKES RAINBOW SMELT AND LAKE TROUT

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Introduction

Although polybrominated diphenyl ethers (PBDEs) are bioaccumulative and ubiquitous in the environment, North America continues to be a major consumer of different commercial PBDE products[1]. PBDEs have the potential to leach from industrial and manufacturing facilities and can be disseminated easily into the environment from the disintegration, combustion and recycling of PBDE-containing products [2]. As a result of high usage and ease of transport throughout the environment, significantly higher levels of the lower brominated PBDEs have been reported in a variety of biota in North America as compared to Europe [3, 4, 5].

This paper discusses the temporal and spatial trends of selected PBDE congeners in rainbow smelt (*Osmerus mordax*) and lake trout (*Salvelinus namaycush*) in the Great Lakes region of North America. Both of these fish have been extensively used as indicator species in monitoring spatial and temporal trends of many contaminants. Lake trout is a top food web predator, and this species historically has been used to model the likelihood that contaminants, such as PBDEs, will concentrate at higher trophic levels in the Great Lakes region. However, lake trout tend to be less sensitive to temporal changes in PBDE concentrations since the majority of their time is spent in the water column away from contaminated sediment sources and they are a long-lived species. In contrast, rainbow smelt are a shorter-lived species feeding mainly on crustaceans, invertebrates and occasionally small fish, thus making them a sensitive indicator to temporal PBDE changes. Together, contaminant data from the two species can outline the ecological persistence and accumulation of PBDEs in aquatic environments and thus provide a way to track variation over time.

The Great Lakes basin is an important manufacturing region that accounts for 18% of the U.S. and Canadian gross domestic product [6]. This region has been the focus of several studies examining PBDE concentrations in North American aquatic biota and surface water samples. In smelt, total PBDE concentrations in the Great Lakes region have increased exponentially in Lakes Huron, Superior, and Michigan from their first introduction in early1980 to 1999 [7]. Similarly, in lake trout, PBDE concentrations have doubled every 3 to 4 years since 1980, particularly congeners PBDE 47, 99 and 100 [8]. Spatial differences between lakes in the region can be significant, e.g., the highest mean concentrations of total PBDEs in adult lake trout have been found in Lake Ontario (95 ± 22 ng/g wet weight) [9].

Materials and Methods

Lake trout were collected by gill net and rainbow smelt were collected by trawl net from the five Great Lakes as a part of a long term monitoring program by the United States Geological Survey. Archived smelt extracts from 1983 to 1999 collected in Lakes Michigan, Superior and Huron have been analyzed and reported [7]. The same paper describes the protocols for sample processing, gas chromatography/mass spectrometry analysis, quantification and quality assurance used in this study.

Here we expand the dataset to include trout and smelt data for the years 2000 to 2005, and also include Lakes Erie and Ontario, representing a larger region outside the state of Michigan. We report primarily on the more prevalent PBDE congeners, namely, PBDEs 47, 99, 100 and 153. "Total PBDE" is represented as the sum of these four

congeners. The trends and statistics for the most recent data are presented and contrasted to earlier data with goals of examining variation over time and lake-to-lake differences. Simple exponential models are used to fit trend data for each species in two contrasting lakes, Superior and Michigan. In all cases, data presented represent composite samples of 5 to 10 fish.

Results and Discussion

Figure 1 shows the concentration of total PBDEs in smelt in Lake Superior. The three sites sampled within the lake show a fair degree of variation, but the overall trend fits an exponential increase with a doubling time of 5.4 yrs with an $R^2 = 0.61$. Levels currently approach 7 or 8 ng/g in these fish. Figure 2 shows total PBDE levels in lake trout, also from Lake Superior. In comparison to the smelt, the lake trout show concentrations that are approximately 10 times higher (80 ng/g in 2000), and the doubling time is considerably faster, 3.5 yrs. Levels show some variation from the trend line, but the model fit is high ($R^2 = 0.89$).

Figure 1. Trends of total PBDE in rainbow smelt in Lake Superior.

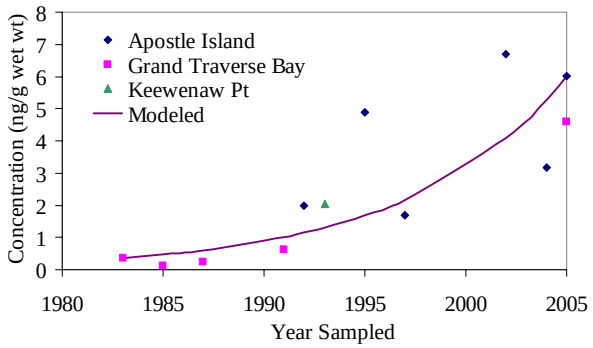

Figure 2. Trends of total PBDE in lake trout in Lake Superior.

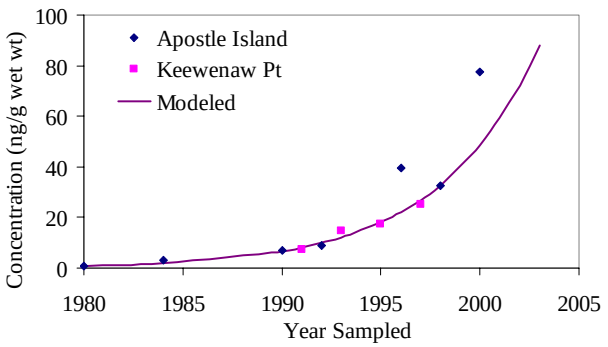

Figure 3 shows the concentration of total PBDEs in smelt in Lake Michigan. As in Lake Superior, concentrations at the sampling sites vary and the exponential model provided moderately good fit ($R^2 = 0.84$). In Lake Michigan, the doubling time was fast, only 1.9 yrs, and concentrations in these fish approach or exceed 100 ng/g, the level found in trout in the Lake Superior. Figure 4 shows trends of total PBDEs in Lake Michigan trout. Here, current total PBDE levels approach 300 ng/g and the doubling time is 2.6 years with a moderately high fit ($R^2 = 0.77$) to the exponential model. However, PBDE trends for both smelt and trout for Lake Michigan (Figures 3 and 4) do not include the most recent (2004-2005) data, which do not appear to fit a simple exponential model. While the most recent PBDE concentrations continue to increase, the more recent (e.g., 1995 to 2005) data suggest a linear trend, rather than a continuation of the exponential trend seen in the earlier period. Many factors can influence PBDE concentrations in both species, including source emissions and food web changes.

Overall, total PBDE levels in lake trout and rainbow smelt have continued to increase exponentially from about the early to mid-1980s to the present in both Lake Superior, the more remote and northernmost lake. Exponential increases to the near present are also observed in Lake Michigan, the heavily urbanized and industrialized area. It should be recognized that PBDE levels currently found in both species in both lakes are hundreds of times higher than found in archived fish samples collected during the early to mid 1980s when usage of PBDEs was limited. The increase noted in both smelt and trout in Lake Superior may largely reflect atmospheric deposition, the effect of which is magnified given the long water retention time in this lake. In contrast, sources affecting Lake Michigan include both effluent discharges and possibly higher rates of air deposition given its proximity to urban and

industrial sources, and the net effect is to significantly increase PBDE concentrations in both fish species by a factor of 5 to 10 times higher than levels in the same species in Lake Superior.

Figure 3. Trends of total PBDE in rainbow smelt in Lake Michigan.

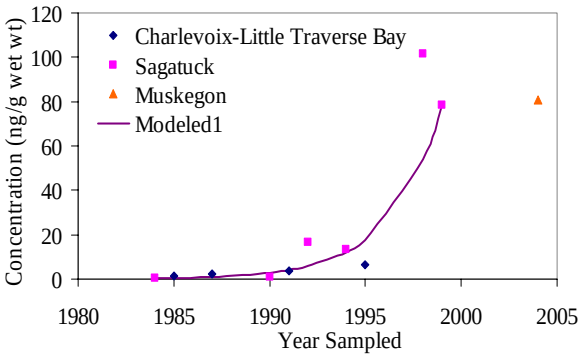

Figure 4. Trends of total PBDE in lake trout in Lake Michigan.

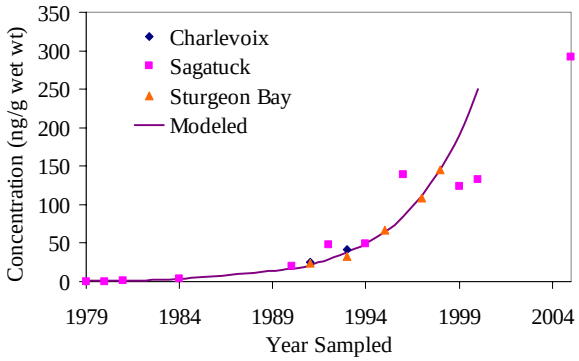

Table 1 shows recent concentrations of the most common PBDE congeners (47, 99, 100 and 153) in trout and smelt collected in the five lakes. The table presents averages of samples collected in 2000 data for trout, and samples collected from 2000 to 2005 for smelt. PBDE 47 is most abundant congener, and trends of each congener generally closely follow those of the total PBDE concentration, as shown elsewhere [8, 9]

Table 1. Statistics of PBDEs in rainbow smelt and lake trout in the five Great Lakes. Trout data uses 2000 average; smelt data use 2000 – 2005 average. Concentrations in ng/g wet weight.

		Lakes				
Species	Congeners	Superior	Erie	Michigan	Huron	Ontario
Lake	Total PBDE	77.39	41.34	132.22	47.73	69.13
Trout	PBDE-47	61.71	28.28	94.18	32.12	54.96
(2000)	PBDE-100	6.02	5.20	13.30	6.54	5.42
n=15	PBDE-99	6.16	5.29	14.00	6.66	5.50
	PBDE-153	3.50	2.57	3.76	2.41	3.25
Rainbow	Total PBDE	4.09	2.39	80.59	11.03	33.37
Smelt	PBDE-47	2.70	1.41	54.59	8.18	27.40
(2000-2005)	PBDE-100	0.66	0.42	11.16	1.78	2.92
n=20	PBDE-99	0.63	0.34	12.32	0.96	2.62
	PBDE-153	0.09	0.22	2.52	0.23	0.43

Lake Michigan fish have by far the highest PBDE concentrations, e.g., PBDE levels in Lake Michigan lake trout are about 2 to 3 times higher than levels found in the four other lakes, where differences are rather small. In contrast, the spatial differences in smelt are much larger, e.g., total PBDE levels in Lake Michigan rainbow smelt are 3 to 20 times higher than levels in fish from the other lakes. PBDE levels in smelt from Lakes Superior and Erie are by far the lowest.

Overall, we have found doubling time of roughly 2 to 5 yrs and levels that depend on lake and fish species. This continuing increase in PBDE concentrations in the two sentinel species warrants continued monitoring and concern.

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