Binational Summary Report: Brominated Flame Retardants (PBDEs and HBCD)

1. Overview:

Annex 3 - Chemicals of Mutual Concern commits the Parties to identify and designate, on an on-going basis, Chemicals of Mutual Concern (CMCs) in the Great Lakes, which originate from anthropogenic sources and that are agreed to by both Parties as being potentially harmful to the environment or human health.

As such, the Annex 3 Subcommittee (C3) has charged an Identification Task Team (ITT) with reviewing and critically evaluating relevant existing data and information, in accordance with the Binational *Considerations* developed by the C3, in order to determine which of a suite of seven candidate chemicals / classes should be recommended as CMCs.

This *Binational Summary Report* documents the application of the *Binational Considerations* to the candidate CMCs polybrominated diphenyl ethers (PBDEs, includes seven PBDE homologues, tetra- through deca-) and hexabromocyclododecane (HBCD), both of which are brominated flame retardants. This report was developed with input and review of the entire ITT and the recommendations presented within were reached by 2/3 majority of the ITT.

PBDEs

With respect to PBDEs, there was sufficient data and information available to effectively apply the *Binational Considerations*, and based on their application of the considerations, **the ITT has recommended, by a 2/3 majority decision, that PBDEs be designated as a CMC.**

With respect to PBDEs, the ITT has established that concentrations in top predator fish (e.g. lake trout and walleye), sediment and herring gull eggs exceed relevant guidelines. Furthermore, while temporal trends in some fish species have shown recent declines, beginning in mid-2000, concentrations of some homologues in sediment and herring gull eggs have shown a stable or slightly increasing long-term trend. Additionally, fish consumption advisories exist across the Great Lakes due to PBDE concentrations. Therefore PBDEs have been identified as posing a threat to the environment and human health in the Great Lakes basin.

The ITT has also concluded that while many risk management and science activities are ongoing or forthcoming for PBDEs under federal and provincial / state programs, there are still needs and opportunities for additional activities, many of which would benefit from enhanced coordination and collaboration, for example:

- Implementing and measuring the performance of existing and forthcoming federal actions to reduce the existing uses of PBDEs, with a focus on addressing the issue of PBDEs in products;
- Continuing to undertake monitoring of air, sediment and top-predator fish species in the Great Lakes to: continue tracking long-term trends; provide data to protect human health through provision of fish consumption advice, assess atmospheric transport and loadings to the Great Lakes and measure the performance of ongoing and forthcoming risk management activities;

HBCD

With respect to HBCD, the ITT has concluded that there is insufficient data and/or information available to effectively apply the *Binational Considerations*. Therefore, **the ITT has recommended, by a 2/3 majority decision, that HBCD be identified as insufficient information on which to base a determination.**

With respect to HBCD, data from the Great Lakes is too limited to definitively conclude whether present environmental concentrations exceed relevant benchmarks / guidelines (e.g. draft Federal Environmental Quality Guidelines) or to establish temporal or spatial trends in the Great Lakes environment; however, there is general surveillance data to indicate that HBCD is found ubiquitously in the environment. Furthermore, as federal risk management activities for HBCD have yet to be implemented in Canada or the US, it is unclear how risk management actions will impact levels in the Great Lakes basin.

While a determination could not be reached for HBCD, a number of needs and opportunities for additional activities were identified, many of which could provide the information necessary to reach a determination for example:

- Continuing to undertake monitoring of air, sediment, top-predator fish species and other biota (e.g. herring gulls) in the Great Lakes to establish long-term trends and measure the performance of forthcoming risk management activities. This will also provide additional information regarding the levels and fate of HBCD in biota as well as the information necessary to assess atmospheric transport and loadings to the Great Lakes.
- Undertake activities to gain an understanding of the use and production of HBCD in the US that is comparable to the knowledge of the Canadian status;
- While Great Lakes data was insufficient to reach a determination, the ITT noted that the proposed risk management measures for HBCD in Canada (i.e. PCTSRs) have yet to enter into force and that HBCD is not under federal risk management in the US, and as such, federal actions to manage domestic (i.e. nationwide) sources of HBCD should be finalized in Canada and further explored in the US.

2. Chemical background:

Chemical Identity:

PBDEs

Polybrominated diphenyl ethers (PBDEs) are a class of brominated flame retardants intentionally manufactured to retard the combustibility of treated materials. When fire occurs, the PBDE formulations utilize vapor phase chemical reactions that interfere with the combustion process, thus delaying ignition and inhibiting the spread of fire. PBDEs are used as flame retardants in a number of applications, including textiles, plastics, wire insulation, and automobiles. Historically, PBDEs were used extensively in the U.S. and Canada; however, due to global restrictions, there has been a shift to other substances. .

PBDEs have a common structure of a brominated diphenyl ether molecule that may have anywhere from 1 to 10 bromine atoms attached. Depending on the location and number of bromine atoms, there are 209 possible PBDE compounds; each is termed congener and is assigned a specific brominated diphenyl ether (BDE) number. Historically, the three most common commercial formulations used in consumer products were: Penta-BDE, Octa-BDE and Deca-BDE. Each of these types of PBDEs has different uses and different toxicity. PBDEs have never been manufactures in Canada. North American manufacturers of Penta-BDE and Octa-BDE agreed to voluntarily stop producing these two forms of PBDEs by the end of 2004, which was followed by a phase-out on an international basis. In 2009, the three major producers of Deca-BDE in the US voluntarily committed to stop production, importation and sales of Deca-BDE for all uses by the end of 2013.In Canada, these manufacturers committed to phasing out the export to Canada and sales of Deca-BDE for all uses (including transportation and military) by 2013.

PBDEs have the ability to enter the environment. The dispersion of PBDE congeners in the environment is governed by their respective physical and chemical properties. The atmosphere is the primary transport medium, and soils and sediments are environmental sinks. Transport can occur over relatively long distances, greater than 1,000 km. Evidence for this comes from the presence of PBDEs in polar environments, and in the tissues of deep ocean-dwelling whales and other marine mammals who spend a significant portion of their lives far from anthropogenic sources. PBDEs are lipophilic and hydrophobic compounds and readily bioaccumulate into terrestrial and aquatic food webs. This tendency has resulted in extensive accumulation of PBDEs in a wide variety of birds, fish, insects, and aquatic and terrestrial mammals (EPA 2010).

HBCD

Hexabromocyclododecane (HBCD) is a category of brominated flame retardants, consisting of 16 possible isomers. It has a molecular formula of $C12H18Br6$ and its structure consists of a ring of 12 carbon atoms to which 18 hydrogen and six bromine atoms are bound. HBCD may be designated as a non-specific mixture of all isomers or as a mixture of three main diastereomers. The main use of HBCD is as a flame retardant in expanded polystyrene foam (EPS) and extruded polystyrene foam (XPS) (Weil and Levchik, 2009). HBCD may also be used as a flame retardant in the backcoating of textiles for upholstered furniture, upholstery seating in transportation vehicles, draperies, wall coverings, mattress ticking, and interior textiles such as roller blinds (Morose, 2006; ECHA, 2009).

The maximum concentration of HBCD for use in fabrics and textiles and in rubber and plastic products ranges from 1-30% (US EPA, 2006). The majority of HBCD used in textiles is for upholstered furniture, in order to meet the stringent fire safety laws of the United Kingdom and California (Morose, 2006). However, according to the 2006 TSCA Inventory Update Rule (IUR) data (which includes information for chemicals manufactured and imported in amounts of 25,000 pounds (11,340 kg) or greater at a single site), less than 1% of the total commercial and consumer use of HBCD was used for fabrics, textiles and apparel (US EPA, 2006).

In addition, HBCD is used as a flame retardant in high impact polystyrene (HIPS) for electrical and electronic appliances such as audio-visual equipment, and some wire and cable applications (Morose, 2006 and ECHA, 2009). Less than 10% of all HBCD used in Europe is used in HIPS (ECHA, 2009).

HBCD's use in numerous industrial applications has extended over decades, and is increasing to match an increased demand. This demand may correlate with the decrease in the use of other flame retardants (Polybrominated diphenyl ethers [PBDEs]). As a result, levels of HBCD in the environment have also been increasing, since mid-2000.

HBCD is highly persistent in air, water, soil and sediment and is subject to long-range transport from its source to remote areas, including the Arctic, where concentrations in the atmosphere have been found to be elevated. Along with its persistence, HBCD has been shown to have a strong potential to bioaccumulate and biomagnify up the food chain.

HBCD is very toxic to aquatic organisms. In mammals, studies have shown reproductive, developmental and behavioural effects. Some of these effects, including endocrine disruption, are trans-generational and detectable even in unexposed offspring. Recent studies also indicate its potential to interfere with the hypothalamic-pituitary-thyroid (HPT) axis, to disrupt normal development, to affect the central nervous system, and to induce reproductive and developmental effects. It is found in a variety of Arctic species including benthic organisms, seabirds, walrus, narwhal, beluga whales, and polar bears (UNEP 2006).

3. Review of existing scientific data and a qualitative evaluation of their significance:

Is the candidate chemical present in the Great Lakes ecosystem and does it present a potential threat to ecological or human health in the Great Lakes Basin?

Environmental and Human Health Benchmarks Guidelines:

Federal Environmental Quality Guidelines (FEQGs) FEQGs have been developed in Canada for certain congeners of PBDEs in water, fish tissue, sediment, wildlife (and bird eggs) to assess the ecological significance of levels of PBDEs in the environment. Draft FEQGS have also been developed for HBCD. These FEQGs are benchmarks for aquatic ecosystems that are intended to protect all forms of aquatic life (vertebrates, invertebrates, and plants) from direct adverse effects for indefinite exposure periods via the water column. They are based on studies that directly link exposure to PBDEs and HBCD to adverse effects in animals.

Homologue*	Congener	Water (ng/L)	Fish Tissue (ng/g ww)	Sediment (ng/g dw)	Wildlife Diet (ng/g ww food source)	Bird Eggs (ng/g ww)
triBDE	total	46	120	44	-	-
tetraBDE	total	24	88	39	44	-
pentaBDE	total	0.2	1	0.4	3 (mammal) 13 (birds)	29
pentaBDE	BDE-99	4	1	0.4	3	-
pentaBDE	BDE-100	0.2	1	0.4	-	-
hexaBDE	total	120	420	440	4	-
heptaBDE	total	17	-	-	64	-
octaBDE	total	17	-	5600	63	-
nonaBDE	total	-	-	-	78	-
decaBDE	total	-	-	19	9	-

*****FEQG for triBDE (tribromodiphenyl ether), tetraBDE (tetrabromodiphenyl ether), hexaBDE (hexabromodiphenyl ether), heptaBDE (heptabromodiphenyl ether), nonaBDE (nonabromodiphenyl ether) and decaBDE (decabromodiphenyl ether) are based on data for the congeners: BDE-28, BDE-47, BDE-153, BDE-183, BDE-206, and BDE-209, respectively unless otherwise noted.

Table 1: Canadian Federal Environmental Quality Guidelines for PBDEs (EC, 2013**)**

Water (µg/L)	Sediment* (mg/kg dw)	Mammalian Wildlife Diet (mg/kg food ww)
0.56	1.6	40
*Normalized to 1% organic carbon dw = dry weight; ww = wet weight		

Table 2: Draft Canadian Federal Environmental Quality Guidelines for HBCD (EC, 2014)

The use of FEQGs is voluntary unless prescribed in permits or other regulatory tools. Thus FEQGs, which apply to the ambient environment, are not effluent limits or "never-to-be-exceeded" values but may be used to derive effluent limits (Environment Canada, 2014).

Great Lakes Monitoring and Surveillance Data:

PBDEs:

Temporal trends for ΣBDE_5 in trout from the Great Lakes recorded during 1980–2009 have shown decreases in concentration after 2000–2001 in lakes Huron, Michigan and Ontario, while concentrations seem to have stabilized in lakes Erie and Superior, but have not begun to decline significantly (Crimmins et al., 2012).

In a study on temporal and geographical patterns of PBDEs in US fish related to industrial usage, the authors note that the recognition of the persistent, bioaccumulative, and toxic properties of PBDEs has prompted reductions in their use (Chen et al., 2011).

In general, penta-BDE concentrations in a range of environmental media (air, sediment, , landfill effluent, aquatic biota and birds) reported from Canada increased until approximately 2000, when levelling off or decreasing trends were observed (Backus et al, 2010). Levels also increased in wastewater effluent.

A more recent study that measured concentrations of a large group of legacy organic chemicals in the Great Lakes waters provides additional information as to the levels of PBDEs in the waters of the Great Lakes (Venier et al., 2014). In this study, samples were collected in the spring of 2011 and 2012 at 18 stations throughout all the Great Lakes as shown on the map and analyzed for brominated and organophosphate ester (OPE) flame retardants, as well as PCBs, organochlorine pesticides, and PAHs.

Key findings from Vernier et al. (2014) include:

- The highest concentrations of brominated flame retardants were generally measured in Lakes Erie or Ontario. Total PBDE concentrations were the highest in Lake Ontario, (average of 227 ±

75 pg/L), and lowest in Lake Superior, (average of 34 ± 11 pg/L). PBDE concentrations in these two lakes were significantly higher than those in lakes Superior, Huron and Michigan (Figure 1).

- The most abundant congeners in all lakes were BDE-47, BDE-99, and BDE-209. BDE-47 and BDE-99 are the two main congeners in the Penta-BDE commercial mixture, which was widely used until 2004, when it was voluntarily withdrawn from the market. BDE-209 was the main congener in the commercial mixture Deca-BDE, which was withdrawn from the market at the end of 2013. A similar pattern of congener abundance has been reported in air samples in the Great Lakes. (Figure 2)
- A high percentage of BDE-209 was found in one station in Lake Huron (located in the Strait of Mackinac, the junction of Lakes Michigan and Huron). This was unexpected, as the shores of the Straits are mainly parks. At the same time, the high values of this particular congener in Lake Michigan are likely driven by the proximity of the sampling sites to urban areas.
- The large imbalance found between air and sediment transfer rates for BDE-209 in Lake Michigan in other papers may suggest the presence of large non-atmospheric sources perhaps localized in urbanized areas. The authors indicate that more data are needed to confirm this hypothesis.
- On the basis of the low vapour pressure of BDE-209, it would be expected to find it mostly in the particulate phase. However, substantial amounts were found in the dissolved phase, which is the case for atmospheric samples in which BDE-209 is found, and for which the ratio of particulate to vapour phase is 8 to 1. In water samples, BDE-209 could be present in the colloidal phase, which was not trapped by our filters. A possible explanation for this is that significant portions of BDE congeners can partition in the colloidal phase and this proportion increases with the degree of bromination. As above, the authors indicate the need for further data to better understand the partitioning behavior of BDE-209.

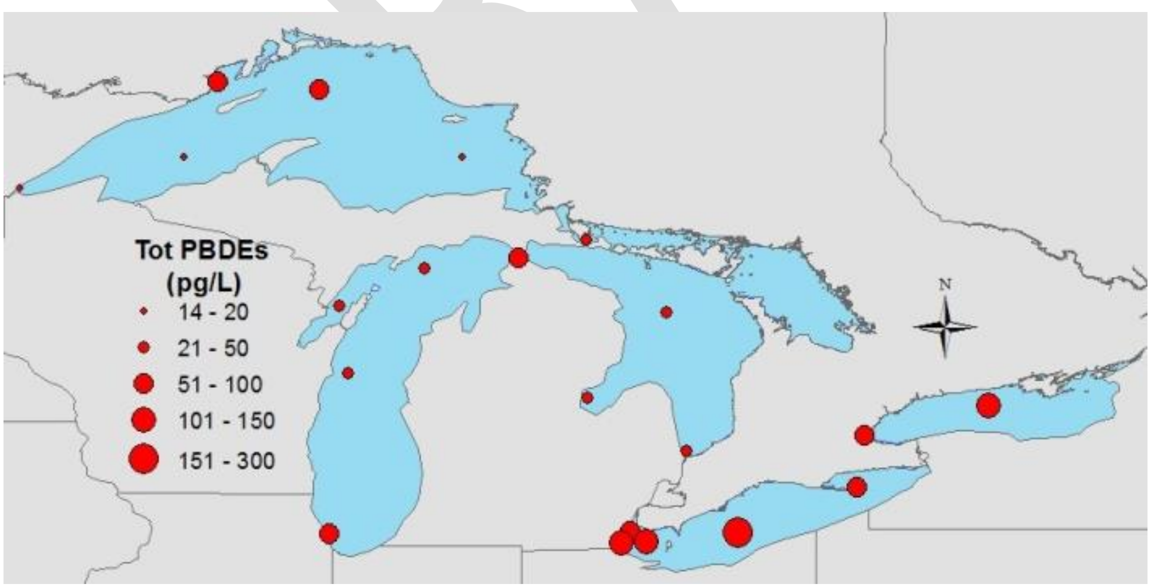


Figure 1: Spatial distribution of total PBDEs (pg/L)

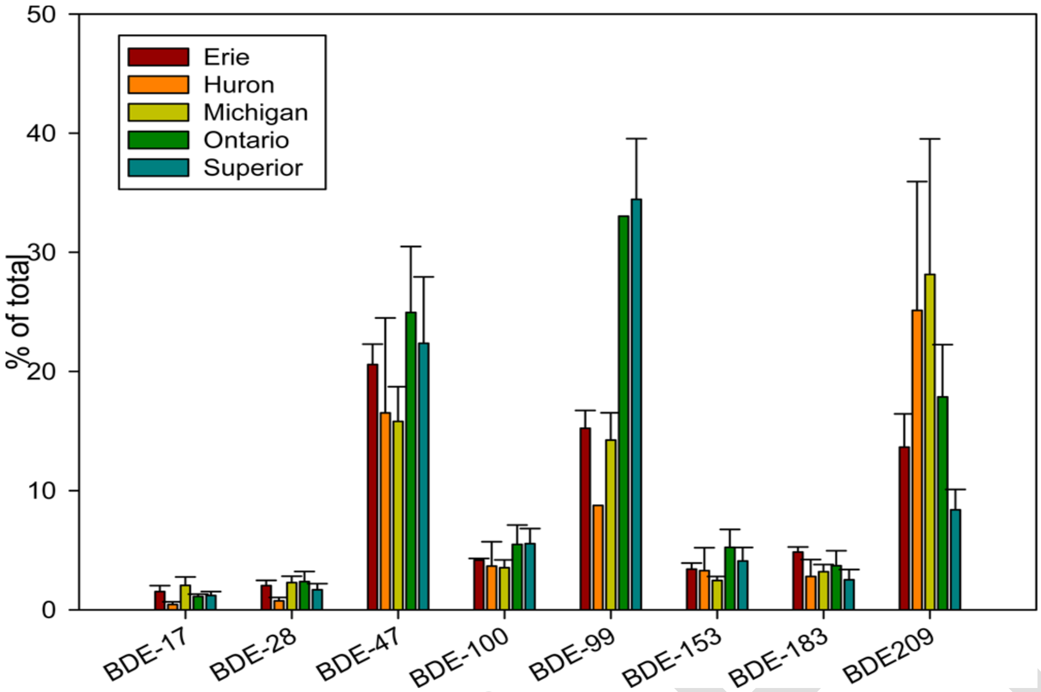


Figure 2: Congener composition of total PBDEs in Great Lakes samples.

Environment Canada monitoring of sediments for PBDES noted moderate exceedances of FEQGs in Lake Ontario, where average pentaBDE concentrations were approximately twice the FEQG (EC, 2011). In a few locations in Lake Ontario, however, values for pentaBDE were 10 – 60 times greater than the FEQG (EC, 2011). The concentration of decaBDE exceeded the FEQG in Toronto Harbor and in six other locations in Lake Ontario (EC, 2011). The temporal trends of average concentrations of PBDEs in sediment (ng/g-dw) in Lake Ontario are presented below in Figure 3 (EC, 2011). The spatial distributions of PBDEs in sediment of the Great Lakes are presented in Figure 4 (EC, 2009).

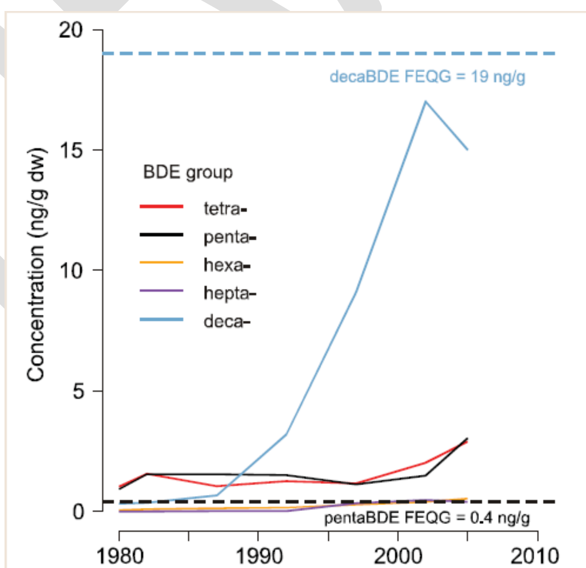


Figure 3: Temporal trends of average concentrations of PBDEs in Lake Ontario sediments. (EC, 2011)

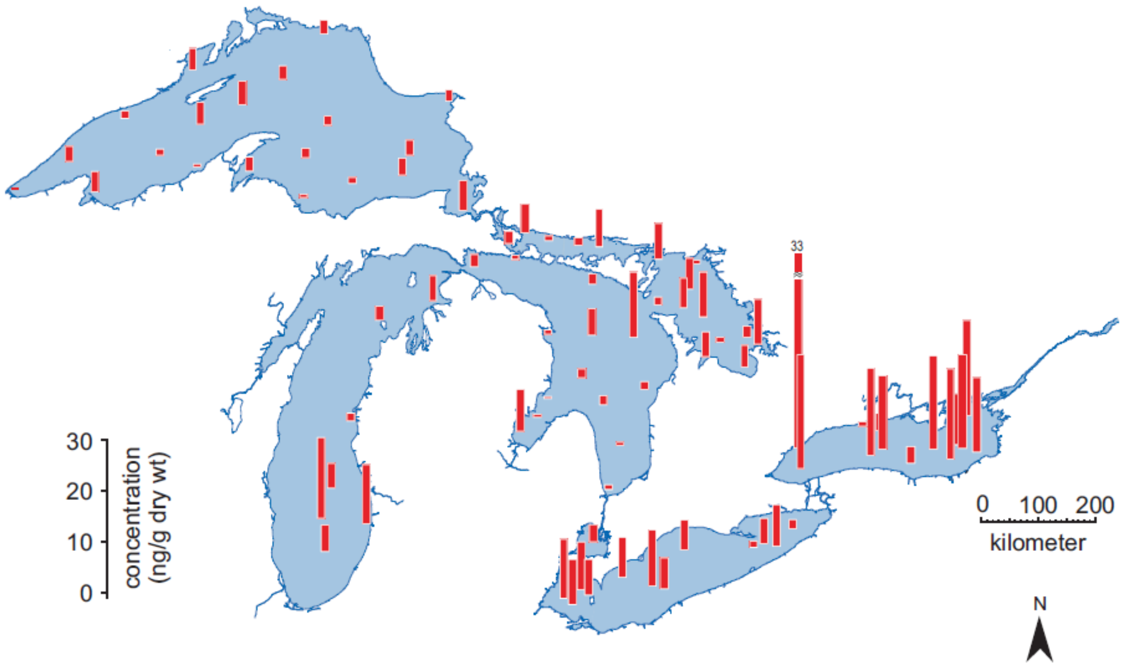


Figure 4: PBDE concentrations in surficial sediments in open waters of the Great Lakes. Source data: Environment Canada and the Ontario Ministry of Environment and Climate Change. (EC, 2009)

In another study on spatial trends of PDBEs in Canadian fish, (Gewurtz et al., 2011), concentrations of the three most abundant PBDE homologue groups (tetra-, penta-, and hexa-PBDEs) were found to be, for the most part, higher in Great Lakes and Lake Champlain fish compared with fish from other systems. For example, the Canadian Federal Environmental Quality Guideline (FEQG) for the penta-homologue was exceeded in 70% of the fish examined. However, virtually no guideline exceedances were found for other congeners. In general, PBDE-47 (a representative lower brominated congener) was significantly and positively correlated with fish length, weight, age, lipid content, and stable isotopes of nitrogen and carbon. The average concentrations of PBDEs in lake trout from Lake Ontario, 1980 to 2009, are presented in the figure below (EC, 2011).

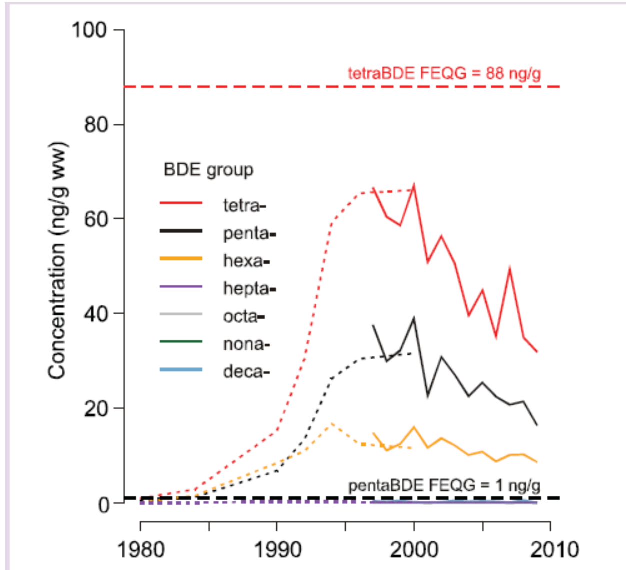


Figure 5: Average concentrations of PBDEs in lake trout (ng/g-ww) from Lake Ontario 1980 to 2009. Source date: US EPA and Environment Canada (EC, 2011)

Routine Canadian and US federal monitoring of PBDEs in whole top predator fish from the Great Lakes (e.g. EC, 2011 and US EPA Great Lakes Fish Monitoring Program reports) combined with retrospective analyses of archived samples by the U.S. EPA and Environment Canada (SOLEC 2013) have provided additional information on PBDE concentrations in the Great Lakes fish (Figures 6, 7, 8 and 9).

The results indicated that concentrations of PBDEs in Lake Trout and Walleye rose continuously up to the early 2000s, and then began to decline (U.S. EPA; Lake Erie), showing significant declining trends of 5.8%/year for tetra-BDEs, 6.4% for penta-BDEs, and 3.4% for hexa-BDEs in Lake Ontario and annual declines of 19% for tetra-BDEs and 17% for penta-BDEs from Lake Michigan. PBDE concentrations in Lakes Superior, Huron, and Erie also appear to be declining as the slopes of the regressions are all negative; however, the slopes are not significantly different from zero at a = 0.05 with a power of 80%. The majority of tetra-BDE and all hexa-BDE concentrations reported for Lake Trout and Walleye in 2009 from all the Great Lakes are below Environment Canada's FEQGs for all congeners, with the exception of penta-BDE, for which concentrations are well above the FEQG of 1.0 ng/g ww.

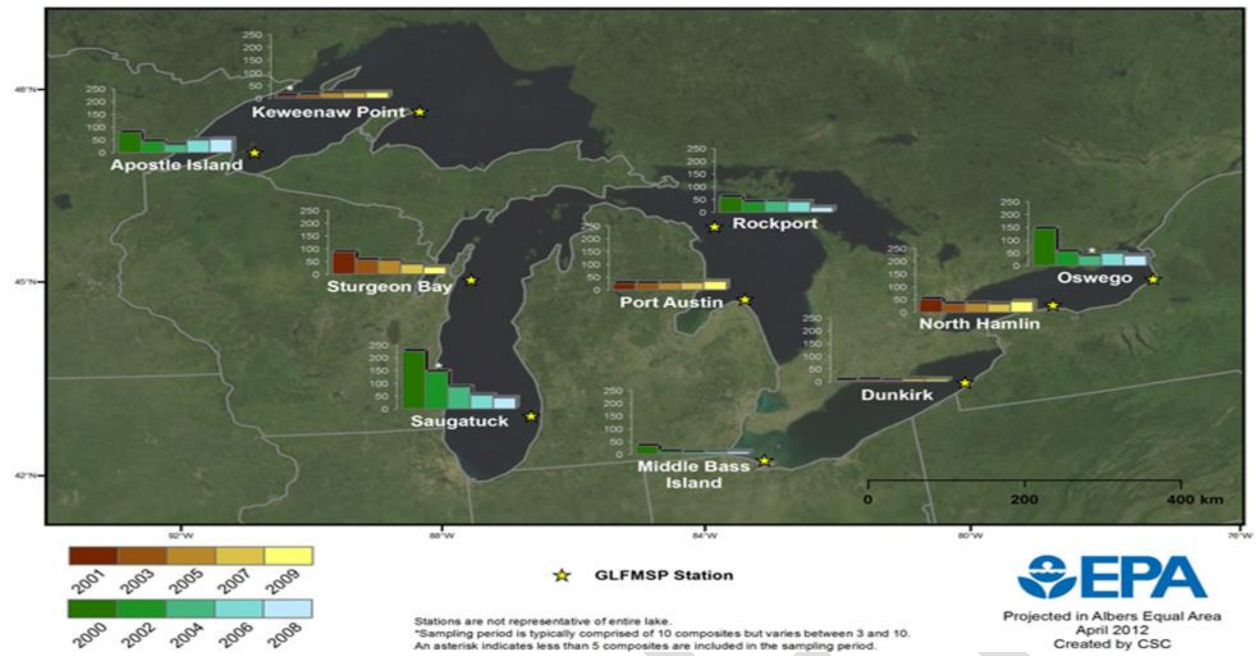


Figure 6: Mean PBDE47 concentration (ppb) in Lake Trout/Walleye from 2000 through 2009. (US EPA GLFMSP)

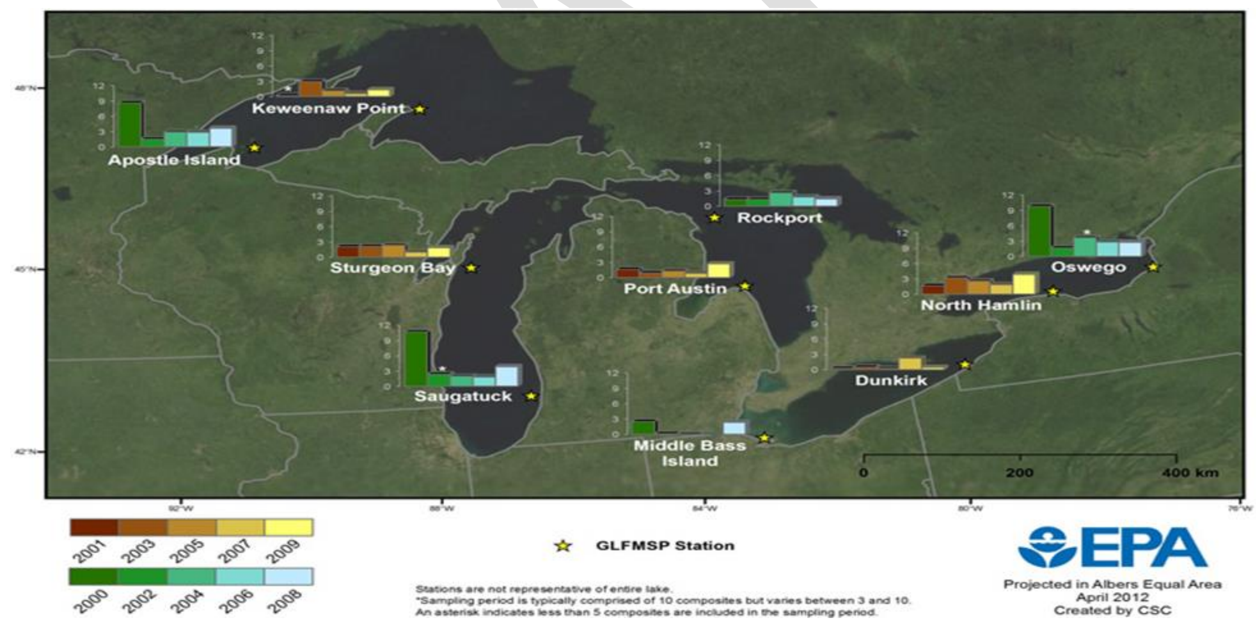


Figure 7: Mean PBDE99 concentration (ppb) in Lake Trout/Walleye from 2000 through 2009. (US EPA GLFMSP)

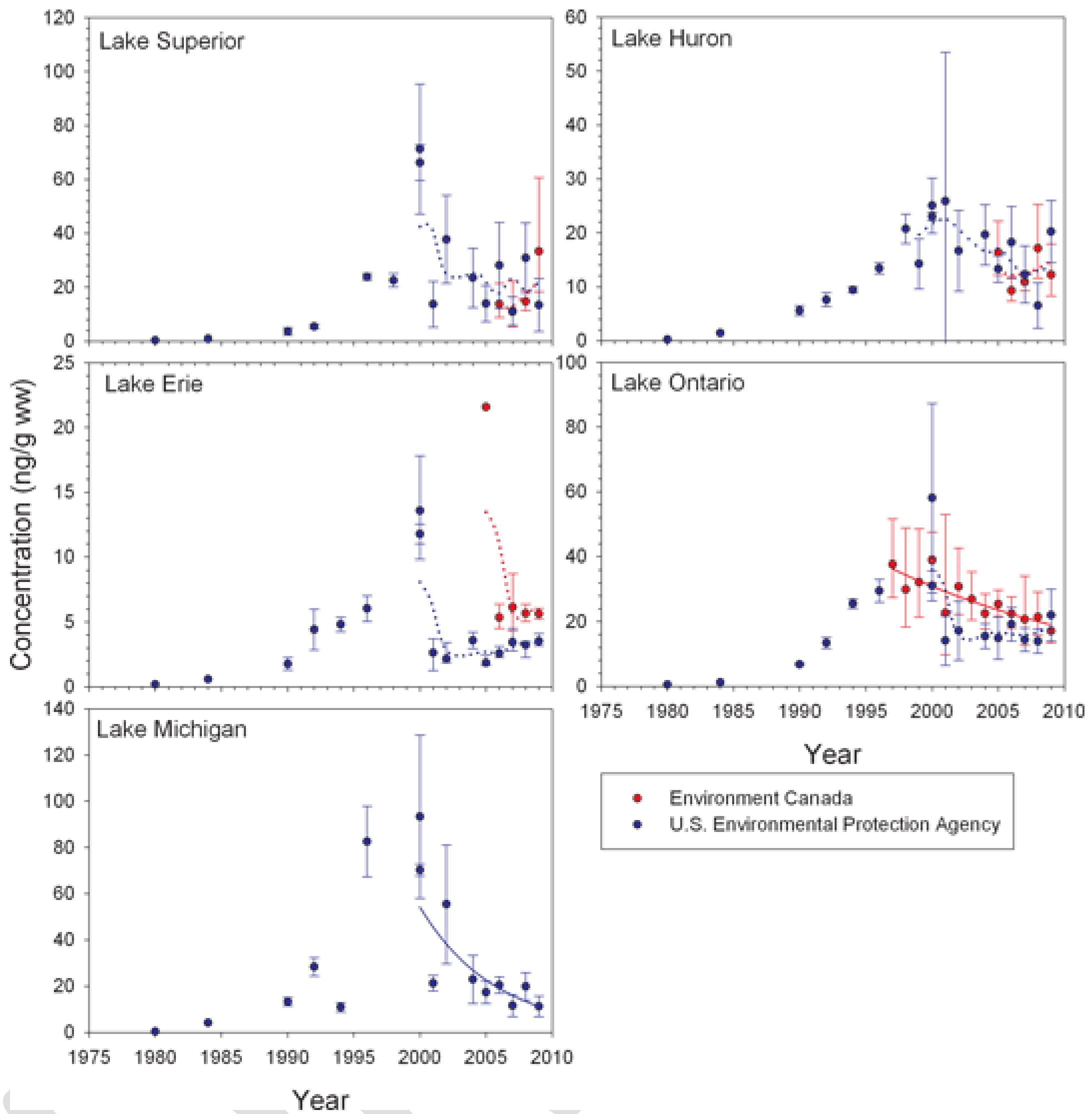


Figure 8: Mean (± standard deviation penta-BDE concentrations in Great Lakes fish measured by Environment Canada, U.S. Environmental Protection Agency and Zhu & Hites (2004). Solid lines denote significant log-linear regressions. Dotted lines denote 3 year moving average when log-linear regression is not significant (SOLEC 2013).

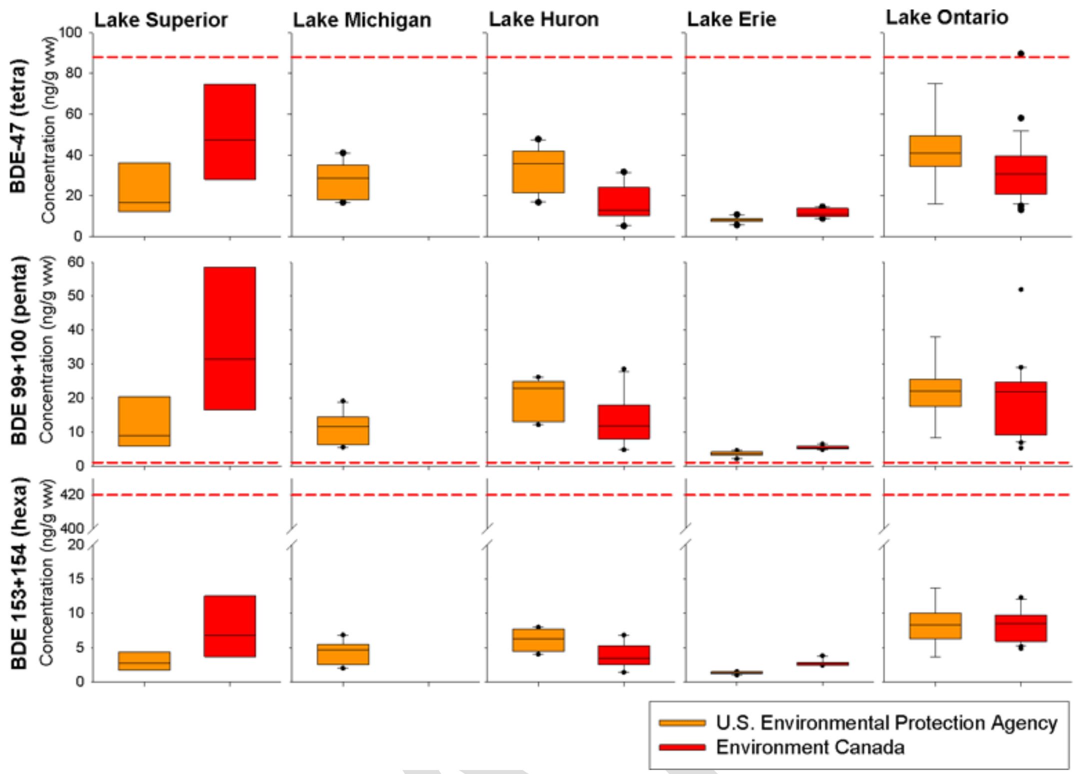
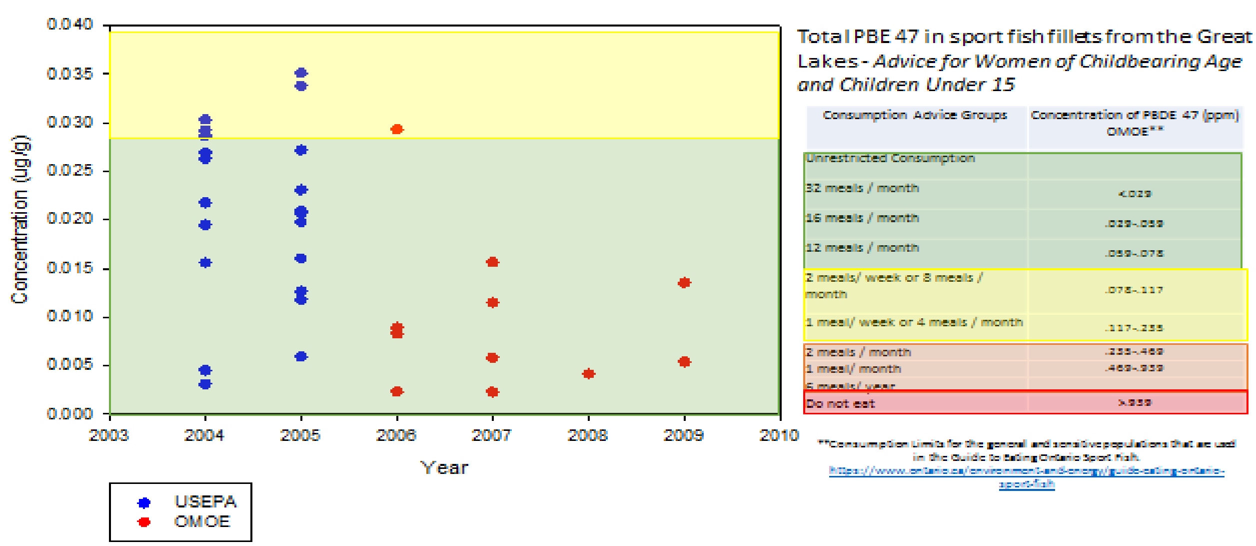
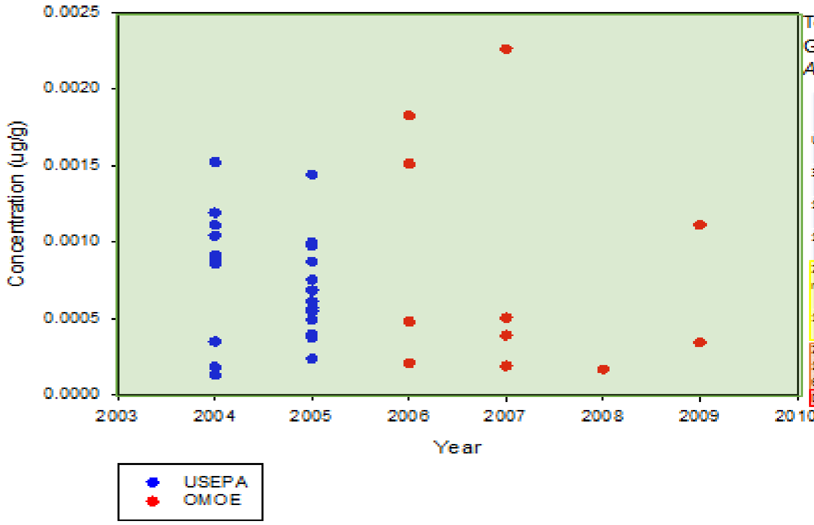


Figure 9: Concentrations of the dominant PBDE congeners (ηg/g ww) in whole body Lake Trout and Walleye (U.S. EPA; Lake Erie) in each of the Great Lakes measured in 2009 relative to the Federal Environmental Quality Guidelines developed by Environment Canada (red dashed line). (SOLEC, 2013)

Figure 10 below depicts the concentrations of different PBDES in data from the US EPA and OMOECC, compared against current fish consumptions advice (GLFMSP).



(A) Total BDE 47

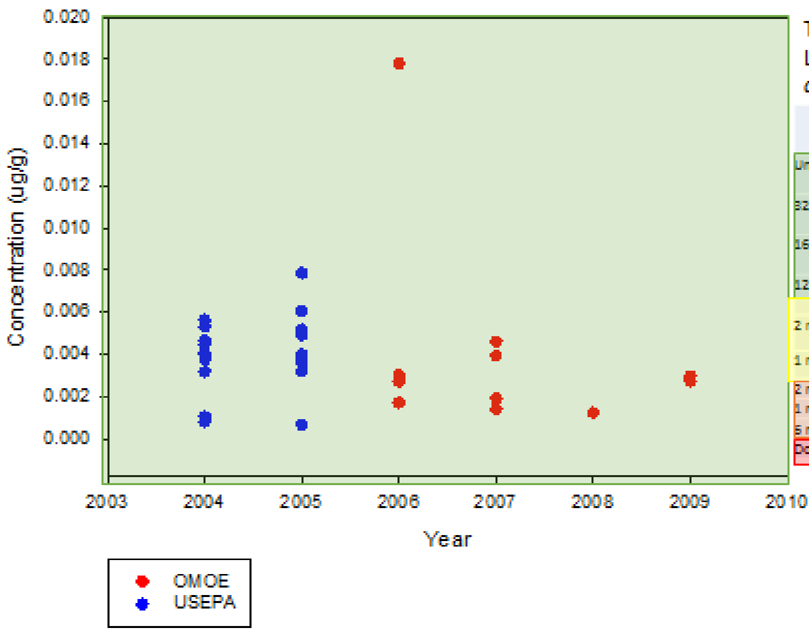


Total BDE 153 in sport fish fillets from the Great Lakes - *Advice for Women of Childbearing Age and Children Under 15*

Consumption Advice Groups	Concentration of PBDE 153 (ppm) OMOE**
Unrestricted Consumption	
32 meals / month	<.059
16 meals / month	.059-.117
12 meals / month	.117-.156
2 meals/ week or 8 meals / month	.156-.235
1 meal/ week or 4 meals / month	.235-.469
2 meals / month	.469-.939
1 meal/ month	.939-1.877
6 meals/ year	
Do not eat	>1.877

**Consumption Limits for the general and sensitive populations that are used in the Guide to Eating Ontario Sport Fish. https://www.ontario.ca/environment-and-energy/guide-eating-ontario-sport-fish

(B) Total BDE 153

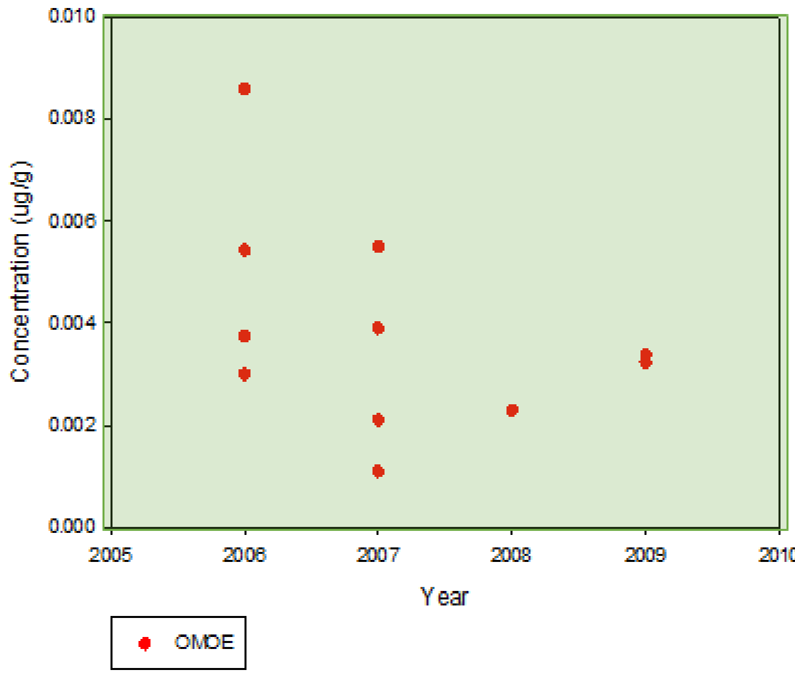


Total BDE 99 in sport fish fillets from the Great Lakes - *Advice for Women of Childbearing Age and Children Under 15*

Consumption Advice Groups	Concentration of PBDE 99 (ppm) OMOE**
Unrestricted Consumption	
32 meals / month	<.029
16 meals / month	.029-.059
12 meals / month	.059-.078
2 meals/ week or 8 meals / month	.078-.117
1 meal/ week or 4 meals / month	.117-.235
2 meals / month	.235-.469
1 meal/ month	.469-.939
6 meals/ year	
Do not eat	>.939

**Consumption Limits for the general and sensitive populations that are used in the Guide to Eating Ontario Sport Fish. https://www.ontario.ca/environment-and-energy/guide-eating-ontario-sport-fish

(C) Total BDE 99



Total BDE 209 in sport fish fillets from the Great Lakes - *Advice for Women of Childbearing Age and Children Under 15*

Consumption Advice Groups	Concentration of PBDE 209 (ppm) OMOE**
Unrestricted Consumption	
32 meals / month	≤2.053
16 meals / month	2.053-4.106
12 meals / month	4.106-5.475
2 meals/ week or 8 meals / month	5.475-8.213
1 meal/ week or 4 meals / month	8.213-16.425
2 meals / month	16.425-32.850
1 meal/ month	32.850-65.701
6 meals/ year	
Do not eat	>65.701

**Consumption Limits for the general and sensitive populations that are used in the Guide to Eating Ontario Sport Fish. https://www.ontario.ca/environment-and-energy/guide-eating-ontario-sport-fish

(D) Total BDE 209

Figure 10 A-D: Concentrations of BDEs in sport fish fillets from the Great Lakes (GLFSMP)

Environment Canada has analyzed the eggs of herring Gulls from monitored colonies across the Great Lakes for PBDEs, in order to obtain long-term trend information on PBDEs in wildlife, presented in Figure 11 (EC, 2011). As observed in fish, tetra-, penta- and hexaBDE are the dominant homologues because of their higher bioaccumulation potential. Although concentrations of tetra-, penta- and hexaBDE are more variable from year to year than in other media, they appear to show a similar temporal trend of increase and subsequent decline to that in fish; however, the peak years and decline occur several years later. In addition, decaBDE concentrations, although relatively low, increased over the entire period from 1980 – 2010. Maximum average values for pentaBDE were up to 20 times higher than the FEQG.

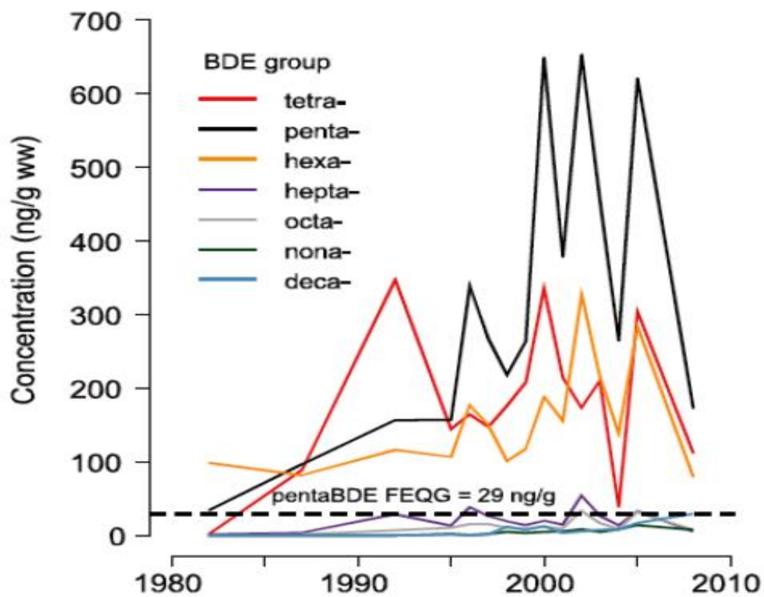


Figure 11: Average concentrations of PBDEs in herring gull eggs (ng/g-ww) in Toronto Harbour, 1982 – 2008. (EC, 2011)

Biomonitoring:

Concentrations of PBDEs were measured in blood-plasma of the Canadian population were evaluated under the first cycle of the Canada Health Measures Survey (CHMS) (HC, 2010). The only PBDE homologue for which > 40% of samples were above the limit of detection (LOD) and therefore a mean value calculated, was PBDE 47 – see table 1 below (HC, 2010)

	n	%<LOD[a]	A.M. 95%CI	G.M. 95%CI	10th 95%CI	25th 95%CI	50th 95%CI	75th 95%CI	90th 95%CI	95th 95%CI
Total, age 20–79	1666	25.27	21.71 17.48 - 25.94	10.04 9.06 - 11.12	<LOD	<LOD	9.71 8.43 - 10.99	20.60 18.04 - 23.16	44.20 39.55 - 48.86	66.60 50.63 - 82.56
20–39	525	21.14	21.34 16.41 - 26.28	10.33 8.77 - 12.17	<LOD	<LOD	10.13 7.94 - 12.31	19.28 16.63 - 21.93	50.18 42.11 - 58.25	74.09 40.89 - 107.29
40–59	596	26.17	21.57 13.92 - 29.23	9.95 8.76 - 11.30	<LOD	<LOD	9.49 7.82 - 11.16	21.54 16.74 - 26.35	39.62 30.99 - 48.26	62.94 44.93 - 80.96
60–79	545	28.26	22.65 15.92 - 29.38	9.70 8.41 - 11.19	<LOD	<LOD	9.29 7.47 - 11.11	21.32 16.94 - 25.70	44.39 31.69 - 57.09	69.81 42.32 - 97.31
Males										
Total, age 20–79	801	26.59	22.12 16.60 - 27.64	9.74 8.55 - 11.10	<LOD	<LOD	9.32 8.01 - 10.63	20.20 16.45 - 23.95	44.08 38.69 - 49.48	63.58 48.77 - 78.39
20–39	240	20.00	20.64 14.98 - 26.30	9.89 7.58 - 12.90	<LOD	<LOD	9.72 7.29 - 12.15	18.61 12.92 - 24.29	45.00 34.84 - 55.16	81.90 27.70 - 136.11
40–59	281	27.40	23.05 10.69 - 35.41	9.68 8.33 - 11.25	<LOD	<LOD	8.65 6.71 - 10.59	19.99 14.73 - 25.24	38.95 24.66 - 53.24	62.77 38.85 - 86.69
60–79	280	31.43	23.04 12.91 - 33.17	9.60 8.07 - 11.41	<LOD	<LOD	9.03 7.07 - 10.99	22.83 14.95 - 30.72	43.76 32.08 - 55.44	75.96 34.38 - 117.55
Females										
Total, age 20–79	865	24.05	21.30 17.12 - 25.49	10.34 8.95 - 11.94	<LOD	<LOD	10.07 7.93 - 12.20	20.92 17.39 - 24.46	44.76 35.73 - 53.79	70.85 48.05 - 93.65
20–39	285	22.11	22.08 14.51 - 29.65	10.83 9.06 - 12.94	<LOD	4.37 <LOD - 6.37	10.98 8.41 - 13.55	19.37 15.95 - 22.78	50.62 38.26 - 62.97	70.98 39.66 - 102.30
40–59	315	25.08	20.10 14.94 - 25.26	10.22 8.53 - 12.24	<LOD	<LOD	9.86 7.55 - 12.16	22.29 15.70 - 28.88	39.52 30.70 - 48.33	67.90 37.26 - 98.55
60–79	265	24.91	22.28 14.63 - 29.93	9.79 7.93 - 12.09	<LOD	3.61 <LOD - 5.25	9.64 7.12 - 12.15	19.88 14.30 - 25.45	45.40 26.65 - 64.15	67.45 43.11 - 91.79

Table 3: Summary statistics for 2,2',4,4'-Tetrabromodiphenyl ether (PBDE 47) lipid adjusted blood-plasma concentrations for the Canadian population aged 20-79 years. (HC, 2010)

The First Nations Biomonitoring Initiative (FNBI) examined the concentrations of PBDEs in blood-plasma of the Canadian First Nations population in 2011 (AFN, 2013). This analysis included a breakdown of concentrations by ecozones, one of which was the Great Lakes (AFN, 2013). Similarly to the CHMS results, levels of the FNBI samples for all PBDES with the exception of PBDE 47 were below the LOD and therefore not calculated. The PBDE 47 blood-plasma levels observed in the FNBI proper, and specifically in the FNBI Great Lakes ecozone, were significantly lower than levels observed in the CHMS sample (AFN, 2013).

In the Fourth National Report on Human Exposure to Environmental Chemicals (Fourth Report), CDC scientists measured ten different PBDEs in the blood serum (the clear portion of blood) of at least 1,985 participants aged 12 years and older who took part in the National Health and Nutrition Examination Survey (NHANES) during 2003–2004. By measuring PBDEs in blood serum scientists can estimate the amounts of these chemicals that have entered people's bodies. The following PBDEs were detected in greater than 60 percent of participants: BDE-28, BDE-99, BDE-100, and BDE-153. Also, BB-153 was detected in greater than 60 percent of participants.

Biomonitoring studies of serum PBDEs can provide physicians and public health officials with reference values so that they can determine whether people have been exposed to higher levels of PBDEs than are found in the general population. Biomonitoring data can also help scientists plan and conduct research on exposure and health effects.

HBCD:

Monitoring studies document the presence of HBCD in many environmental media, with highest concentrations reported near urban/industrial sources. Analyses of sediment core samples show a clear trend of increasing concentrations of HBCD since the 1970s, confirming stability in deep sediments for periods of more than 30 years. As well, there is evidence of increasing HBCD levels in North American and European biota, both within species and along food chains.

Air:

Concentrations of up to 0.011 ηg/m^3 were measured in the particle phase of air samples collected in 2002 and 2003 at five sites from Lake Michigan through the U.S. Midwest to the Gulf of Mexico (Hoh and Hites 2005). Based on similarities in spatial concentration patterns of HBCD and the brominated diphenyl ether flame retardant BDE-209 (decabromodiphenyl ether), the researchers speculated that the brominated flame retardant market may be shifting from diphenyl ether products to HBCD (Hites and Hoh 2005).

HBCD was detected (0.001 to 0.003 ng/m^3 HBCD; data read from graph) during continuous high-volume air measurements collected at Alert, Nunavut, in the Canadian Arctic between 2006 and 2007 (Xiao et al. 2010).

Precipitation samples collected from the Great Lakes basin contained up to 35 ng/L (Backus et al. 2005). All three major diastereomers were detected, with an average distribution of 77%, 15% and 8% for α-, β- and γ-HBCD, respectively.

Surface Waters

Law et al. (2006a) reported a mean dissolved phase concentration of 0.011 ng/L for α-HBCD in surface water samples collected from the south basin of Lake Winnipeg in 2004. Beta- and γ-HBCD were not detected (detection limit: 0.003ng/L). The researchers commented that detection of only α-HBCD in the samples was consistent with its much greater aqueous solubility (4.88 x 104 ng/L;) relative to that of the β- (1.47 x 104 ng/L) and γ- (2.08 x 103 ng/L) isomers.

Four surficial sediment grab samples collected in 2003 from the same region contained a mean concentration of 0.05 ng/g dw γ-HBCD (Law et al. 2006a). Alpha- and β-HBCD were not detected in the samples (detection limit: 0.04 ng/g for β- and γ-HBCD to 0.08 ng/g dw for α-HBCD). The researchers commented that the results were consistent with γ-HBCD being the most hydrophobic of the three isomers.

Sediment

Marvin et al. (2004, 2006) measured HBCD in suspended sediments collected along the Detroit River from Lake St. Clair to the outflow to Lake Erie, and determined that occurrence of the substance was strongly associated with urban and industrial activities. Annual mean concentrations ranged from 0.012 ng/g to 1.14 ng/g dw, with the highest levels being found downstream of the urban region surrounding the city of Detroit. About two thirds of the samples had isomeric profiles similar to those found in commercial technical mixtures, with a predominance of γ-HBCD, while the remaining samples were dominated by α-HBCD. The β-isomer was present at substantially lower levels, consistent with its lower prevalence in commercial mixtures.

The researchers concluded that distribution of HBCD in the Detroit River appeared to be heavily influenced by HBCD associated with shoreline-based urban and industrial activities. In addition, the widespread occurrence of relatively low concentrations suggested that large urban areas may act as diffuse sources of HBCD.

Medium	Location; year	Concentration	Samples	Reference
Air	Canadian and Russian Arctic; 1994–1995	< 0.0018 ng/m^3	12	Alaee et al. 2003
Air	Alert, Canadian Arctic; 2006– 2007	0.001–0.002 ng/m^3, peak at ~ 0.003 ng/m^3	High volume continuous for 1 year	Xiao et al. 2010[3]
Air	United States; 2002–2003	< 0.0002–0.011 ng/m^3	In 120 of 156	Hoh and Hites 2005
Air	United Kingdom; 2007	0.002–0.04 ng/m^3	5	Abdallah et al. 2008a
Air	The Netherlands; 1999	280 ng/m^3	ns[1]	Waindzioch 2000
Air	Svalbard, Norwegian Arctic; 2006–2007	0.0065 ng/m^3 (2006) 0.0071 ng/m^3 (2007)	Mean values	Manø et al. 2008, as cited by de Wit et al. 2010
Air	Sweden; 1990–1991	0.0053–0.0061 ng/m^3	2	Bergander et al. 1995
Air	Sweden; 2000–2001	< 0.001–1070 ng/m^3	11	Remberger et al. 2004
Air	Finland; 2000–2001	0.002, 0.003 ng/m^3	2	Remberger et al. 2004
Air	China; 2006	0.0012–0.0018 ng/m^3	4	Yu et al. 2008a
Air	China; 2006	0.00069–0.00309 ng/m^3	4	Yu et al. 2008b
Air	Sweden urban and rural	0.002–0.61 ng/m^3	14	Covaci et al. 2006
Precipitation	Great Lakes; no year	nd[2]–35 ng/L	ns	Backus et al. 2005
Precipitation	The Netherlands; 2003	1835 ng/L	in 1 of 50	Peters 2003
Precipitation	Sweden; 2000–2001	0.02–366 ng/m^2·d	4	Remberger et al. 2004
Precipitation	Finland; 2000–2001	5.1, 13 ng/m^2·d	2	Remberger et al. 2004
Water	United Kingdom lakes	0.08–0.27 ng/L	27	Harrad et al. 2009b
Water	Lake Winnipeg, Canada; 2004	α-HBCD: 0.006–0.013 ng/L β-HBCD: < 0.003 ng/L γ-HBCD: < 0.003–0.005 ng/L	3	Law et al. 2006a
Water	United Kingdom; no year	< 50–1520 ng/L	6	Deuchar 2002
Water	United Kingdom; 1999	4810–15 800 ng/L	ns	Dames and Moore 2000b
Water	The Netherlands; no year	73.6–472 ng/g dw[5] (solid phase)	ns	Bouma et al. 2000
Water	Japan; 1987	< 200 ng/L	75	Watanabe and Tatsukawa 1990
Water (solid phase)	Detroit River, Canada - United States; 2001	< 0.025–3.65 ng/g dw	63	Marvin et al. 2004, 2006
Sediment	United Kingdom lakes	0.88–4.80 ng/g dw	9	Harrad et al. 2009b
Sediment	Lake Winnipeg, Canada; 2003	α-HBCD: < 0.08 ng/g dw β-HBCD: < 0.04 ng/g dw γ-HBCD: < 0.04–0.10 ng/g dw	4	Law et al. 2006a
Sediment	Norwegian Arctic; 2001	α-HBCD: 0.43 ng/g dw β-HBCD: < 0.06 ng/g dw γ-HBCD: 3.88 ng/g dw	4	Evenset et al. 2007
Sediment	United Kingdom; no year	1131 ng/g dw	1	Deuchar 2002
Sediment	England; 2000–2002	< 2.4–1680 ng/g dw	22	Morris et al. 2004
Sediment	Ireland; 2000–2002	< 1.7–12 ng/g dw	8	Morris et al. 2004
Sediment	Belgium; 2001	< 0.2–950 ng/g dw	20	Morris et al. 2004
Sediment	The Netherlands; no year	25.4–151 ng/g dw	ns	Bouma et al. 2000
Sediment	The Netherlands; 2000	< 0.6–99 ng/g dw	28	Morris et al. 2004
Sediment	The Netherlands; 2001	14–71 ng/g dw	ns	Verslycke et al. 2005
Sediment	Dutch North Sea; 2000	< 0.20–6.9 ng/g dw	in 9 of 10	Klamer et al. 2005
Sediment	Switzerland; no year	< 0.1–0.7 ng/g dw[3]	1	Kohler et al. 2007
Sediment	Switzerland; 2003	0.40–2.5 ng/g dw	1	Kohler et al. 2008
Sediment	Sweden; 1995	nd–1600 ng/g dw	18	Sellström et al. 1998

(A)

Medium	Location; year	Concentration	Samples	Reference
Sediment	Sweden; 1996–1999	0.2–2.1 ng/g dw	9	Remberger et al. 2004
Sediment	Sweden; 2000	< 0.1–25 ng/g dw	6	Remberger et al. 2004
Sediment	Norway; 2003	α-HBCD: < 0.03–10.15 ng/g dw β-HBCD: < 0.08–7.91 ng/g dw γ-HBCD: < 0.12–3.34 ng/g dw	26	Schlabach et al. 2004a, 2004b
Sediment	Spain; 2002	0.006–513.6 ng/g dw	4	Eljarrat et al. 2004
Sediment	Spain; no year	< 0.0003–2658 ng/g dw	4	Guerra et al. 2008
Sediment	Spain; 2002–2006	nd–2430 ng/g dw	13	Guerra et al. 2009
Sediment	Japan; 1987	nd–90 ng/g dw	in 3 of 69	Watanabe and Tatsukawa 1990
Sediment	Japan; 2002	0.056–2.3 ng/g dw	in 9 of 9	Minh et al. 2007
Soil	United Kingdom; 1999	18 700–89 600 ng/g dw	4	Dames and Moore 2000a
Soil	Sweden; 2000	140–1300 ng/g dw	3	Remberger et al. 2004
Soil	China; 2006	1.7–5.6 ng/g dw	3	Yu et al. 2008a
Landfill leachate	England; 2002	nd	3	Morris et al. 2004
Landfill leachate	Ireland; 2002	nd	3	Morris et al. 2004
Landfill leachate	The Netherlands; 2002	2.5–36 000 ng/g dw (solid phase)	11	Morris et al. 2004
Landfill leachate	Sweden; 2000	3, 9 ng/L	2	Remberger et al. 2004
Landfill leachate	Norway; no year	α-HBCD: nd–0.0091 ng/g ww[3] β-HBCD: nd–0.0038 ng/g ww γ-HBCD: nd–0.079 ng/g ww	ns	Schlabach et al. 2002
STP[4] influent STP effluent Receiving water	United Kingdom; 1999	7.91×10^7–8.61×10^7 ng/L 8850–8.17×10^7 ng/L 528–744 ng/L	3 9 3	Dames and Moore 2000b
STP influent STP effluent STP sludge	United Kingdom; no year	934 ng/L (dissolved phase) 216 000 ng/g dw (solid phase) nd (dissolved phase) 1260 ng/g dw (solid phase) 9547 ng/g dw	ns	Deuchar 2002
STP influent STP effluent STP sludge	England; 2002	nd–24 ng/L (dissolved phase) < 0.4–29.4 ng/g dw (solid phase) < 3.9 ng/L 531–2683 ng/g dw	5 5 5 5	Morris et al. 2004
STP sludge	Ireland; 2002	153–9120 ng/g dw	6	Morris et al. 2004
STP effluent Activated sludge	The Netherlands; 1999–2000	10 800–24 300 ng/L 728 000–942 000 ng/g dw	ns 3	Institut Fresenius 2000a, 2000b
STP influent STP effluent STP sludge	The Netherlands; 2002	< 330–3800 ng/g dw (solid phase) < 1–18 ng/g dw (solid phase) < 0.6–1300 ng/g dw	5 5 8	Morris et al. 2004
STP sludge	Sweden; 1997–1998	11–120 ng/g dw	4	Sellström 1999; Sellström et al. 1999
STP sludge	Sweden; 2000	30, 33 ng/g dw	2	Remberger et al. 2004
STP primary sludge STP digested sludge	Sweden; 2000	6.9 ng/g dw < 1 ng/g dw	1 3	Remberger et al. 2004
STP sludge	Sweden; 2000	3.8–650 ng/g dw	ns	Law et al. 2006c
Plant WWTP[5] influent	United Kingdom; 1999	1.72×10^5–1.89×10^6 ng/L	3	Dames and Moore 2000a

(B)

Table 4 A-B: HBCD concentrations measures in the ambient environment and waste treatment products. (EC & HC, 2011)

Location; year	Organism	Concentration (ng/g lipid weight)	Samples	Reference
Canadian Arctic; 1976–2004	Ivory gull (*Pagophila eburnea*) egg	2.1–3.8	24	Braune et al. 2007
Canadian Arctic; 1996–2002	 Beluga (*Delphinapterus leucas*) Walrus (*Odobenus rosmarus*) Narwhal (*Monodon monoceros*) Arctic cod (*Boreogadus saida*) Redfish (*Sebastes mentella*) Shrimp (*Pandalus borealis, Hymenodora glacialis*) Clam (*Mya truncate, Serripes groenlandica*) Zooplankton	α-HBCD / Dγ-HBCD < 0.63–2.08 / < 0.07–0.46 nd–0.86 / < 0.12–1.86 2.05–6.10 / < 0.11–1.27 nd–1.38 / nd–0.07 < 0.74–3.37 / < 0.28–1.03 0.91–2.60 / 0.23–1.24 nd–1.03 / < 0.46–5.66 nd–9.16 / 0.13–2.66	 5 5 5 8 5 5 5 5	Tomy et al. 2008
Nunavut; 2007	Ringed seal (*Phoca hispida*)	0.38	10	Morris et al. 2007
Alaska; 1994–2002	Polar bear (*Ursus maritimus*)	< 0.01–35.1	in 2 of 15	Muir et al. 2006
Greenland; 1999–2001	Polar bear (*Ursus maritimus*)	32.4–58.6	11	Muir et al. 2006
Greenland; 1999–2001	Polar bear (*Ursus maritimus*)	41 ng/g ww	20	Gebbink et al. 2008
British Columbia, southern California; 2001–2003	Bald eagle (*Haliaeetus leucocephalus*)	< 0.01 ng/g	29	McKinney et al. 2006
Lake Winnipeg; 2000–2002	 Whitefish (*Coregonus commersoni*) Walleye (*Stizostedion vitreum*) Mussel (*Lampsilis radiate*) Zooplankton Emerald shiner (*Notropis atherinoides*) Goldeye (*Hiodon alosoides*) White sucker (*Catostomus commersoni*) Burbot (*Lota lota*)	α-HBCD / β-HBCD / γ-HBCD 0.56–1.86 / 0.10–1.25 / 0.90–1.19 2.02–13.07 / 0.66–2.36 / 1.65–6.59 6.15–10.09 / < 0.04–2.37 / 6.69–23.04 1.40–17.54 / < 0.04–1.80 / 0.22–1.82 4.51–6.53 / < 0.04–5.70 / 3.66–12.09 7.39–10.06 / < 0.04–2.08 / 3.23–6.95 2.30–5.98 / 0.27–0.90 / 1.53–10.34 10.6–25.47 / 2.29–10.29 / 24.4–47.90	 5 5 5 5 Pooled 5 5 5 5	Law et al. 2006a
Great Lakes; 1987–2004	(ng/g ww) Herring gull (*Larus argentatus*) egg	α-HBCD / β-HBCD / γ-HBCD nd–20 / nd[1] / nd–0.67	41	Gauthier et al. 2006, 2007
Lake Ontario; no year	Whitefish (*Coregonus commersoni*) Walleye (*Stizostedion vitreum*)	92 40	ns[2]	Tomy et al. 2004b
Lake Ontario; 1979–2004	Lake trout (*Salvelinus namaycush*)	α-HBCD / β-HBCD 15–27 / 0.16–0.94 γ-HBCD / ΣHBCD 1.4–6.5 / 16–33	29	Ismail et al. 2009
Lake Ontario; 2002	(ng/g ww) Lake trout (*Salvelinus namaycush*) Rainbow smelt (*Osmerus mordax*) Slimy sculpin (*Cottus cognatus*) Alewife (*Alosa pseudoharengus*) Mysid (*Mysis relicta*) Amphipod (*Diporeia hoyi*) Plankton	α-HBCD / β-HBCD / γ-HBCD 0.37–3.78 / < 0.030 / 0.07–0.73 0.19–0.26 / < 0.030 / 0.03–0.04 0.15–0.46 / < 0.030 / 0.02–0.17 0.08–0.15 / < 0.030 / 0.01–0.02 0.04, 0.07 / < 0.030 / 0.01, 0.02 0.05, 0.06 / < 0.030 / 0.02, 0.03 0.02, 0.04 / < 0.030 / < 0.030, 0.03	 5 3 3 3 2 2 2	Tomy et al. 2004a

Table 5: HBCD Concentrations measured in biota. (EC & HC, 2011)

Biomonitoring Information for HBCD

Because the exposure pathways for HBCD may be varied and exposure estimation uncertain, biomonitoring for HBCD in humans shows promise as a means of determining integrated human exposures to HBCD with lower uncertainty than through estimation of external exposures via multiple pathways. Data from numerous biomonitoring studies of HBCD over the past decade indicate that the central tendency of lipid-adjusted serum and human milk concentrations is approximately 1mg/g lipid, with upper bound levels of approximately 20 mg/g lipid (Aylward, 2011).

Other Factors to Consider:

Levels and trends of PBDEs and HBCDs in the global environment: Status at the end of 2012 (Law et al., 2014)

This paper compiles the most recent literature that has been published from January 2010 to December 2012 on human exposure, environmental distribution, fate and time trends of PBDEs and HBCDs. It summarizes data that bridges the divide between the peak of the first group of flame retardants (the PBDEs and HBCDs) that were used extensively and the rise of their successors, namely, the novel Brominated Flame Retardants (BFRs).

Key findings of this report include:

- While decreasing time trends for penta-mix PBDE congeners were seen in some regions, for example, soils in northern Europe, sewage sludge in Sweden and the USA, carp from a US river, trout from three of the Great Lakes and in Arctic and marine mammals and many birds in the UK, increasing time trends continue in polar bears and some birds at high trophic levels in northern Europe. This may be partially a result of the time delay inherent in long-range atmospheric transport processes.
- In general, concentrations of BDE209 (the major component of the deca-mix PBDE product) are continuing to increase. Of major concern is the possible/likely debromination of the large reservoir of BDE209 in soils and sediments worldwide, to yield lower brominated congeners which are both more mobile and more toxic. This paper has compiled the most recent evidence for the occurrence of this degradation process. Numerous studies reported in this paper reinforce the importance of this concern.
- Time trends for HBCDs are mixed, with both increases and decreases evident in different matrices and locations. Notably, there is increasing occurrence in birds of prey.
- More data on temporal trends of BDE and HBCD concentrations in a variety of matrices and locations are needed before the current status of these compounds can be fully assessed, and the impact of regulation and changing usage patterns among different flame retardants determined.

Specific items in the paper by Law et al. (2014) pertaining to the Binational Summary Report on PBDEs and HBCD are highlighted below:

- Levels in human milk and blood (Section 2.4): Since January 2010, eight published papers reported PBDEs and/or HBCD temporal trends from 1993 to 2009 in human matrices (including human blood serum, dried blood spots and human breast milk). Results were variable. For example, for BDEs, decreasing temporal trends or a peak followed by plateau were identified in the Great Lakes Basin of the USA and other countries (Australia, Italy, Germany in Lower Saxony and Baden-Würtemberg).
- North America (Section 7.2): In archived wastewater sludge (biosolids) samples from Chicago, USA, deca-PBDE concentrations rose from 1995 to 2008, doubling approximately every 5 years, while penta-PBDE concentrations increased and levelled off around 2000. Congener patterns in contemporary biosolids support the contention that the congener BDE209 can be debrominated to yield lesser brominated PBDE congeners (BDE206, BDE207 and BDE208 in particular, in this case).

Conclusions:

PBDES:

The spatial distribution of PBDEs in air, sediment, fish and wildlife across Canada relates largely to levels of urbanization. Higher concentrations were observed near cities, indicating that urban and industrial centres are the primary source of PBDEs in the environment. Lower levels in rural areas and northern Canada are commensurate with ambient environmental levels resulting primarily from atmospheric deposition, although minor inputs from rural populations (e.g., releases from wastewater and consumer products) may also contribute to observed levels in some of these locations.

The following map demonstrates the levels of total PBDEs in air, sediment, wildlife, and fish across Canada in 2008 (EC, 2011). As indicated on the map, the Great Lakes and the St. Lawrence River have the highest levels of total PBDEs in all media.

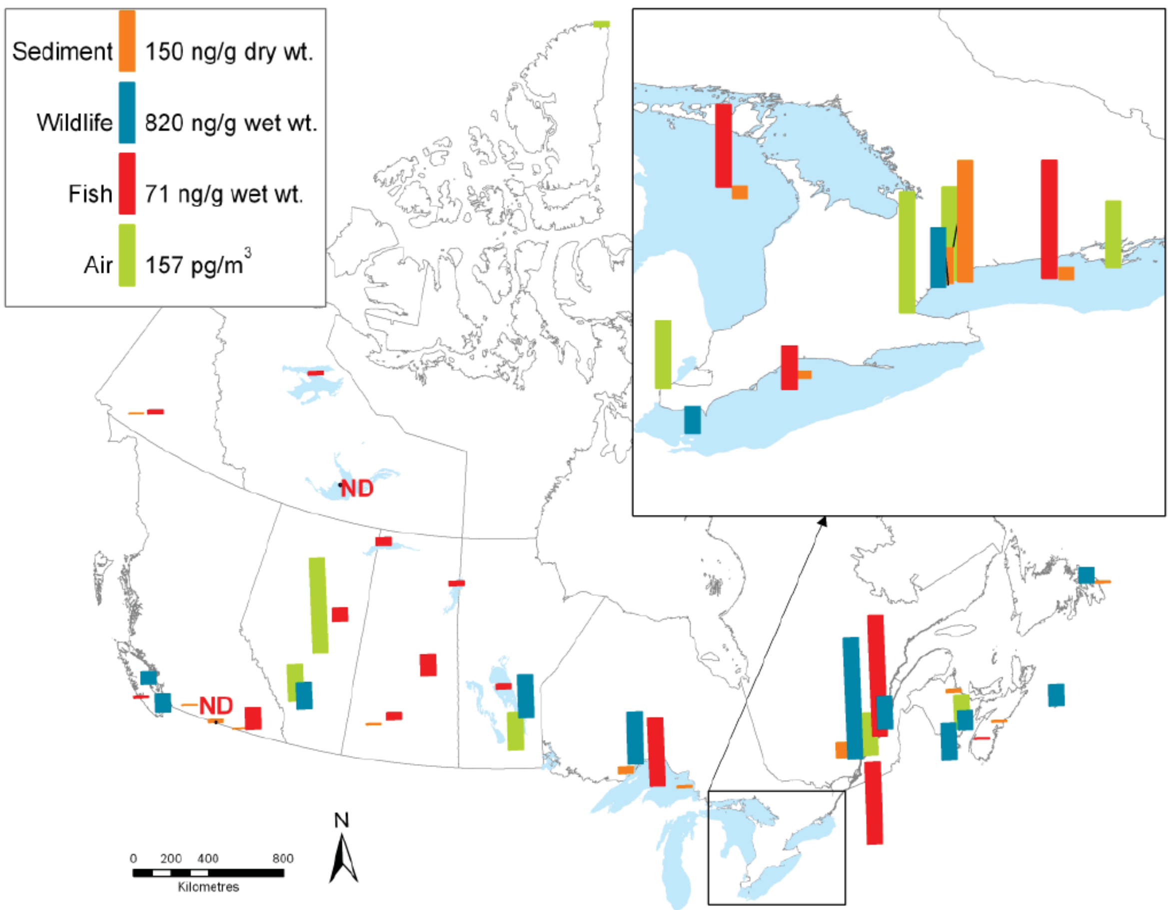


Figure 12: Total PBDEs in sediment, fish, wildlife (gull eggs) and air across Canada in 2008. (EC, 2011)

The maximum values observed for each of these were: Air: 157 pg/m^3 (e.g., Hamilton, Toronto): Sediment: 150 ng/g dry weight (dw); Fish: 70 ng/g wet weight (ww) (St. Lawrence walleye), and 71 ηg/g wet weight (Toronto Harbour, Lake Trout); and Wildlife: 820 ng/g ww eggs of herring gulls, (St. Lawrence River, near Montreal).

The average concentrations of pentaBDE were largely below or slightly above the FEQG of 29 ng/g ww, with the exception of Toronto Harbour and Deslauriers Island in the St. Lawrence River. At this latter site, "levels in eggs of ring-billed gulls were less than the FEQG, levels in herring gull eggs were three times above, but still within the margin of safety"(Environment Canada 2012).

While levels of PBDEs in sediment, fish and wildlife from Lake Ontario showed a marked increase beginning in the early 1980s, in recent years a decreasing trend has been observed that seems to coincide with the voluntary and regulatory phase-out of the use of pentaBDE and octaBDE commercial formulations.

Tetra-, penta- and hexaBDE are the dominant homologues in fish and wildlife over the entire period, while tetra-, penta- and decaBDE congeners are dominant in air, reflecting the influence of penta-BDE and deca-BDE technical mixtures.

Tetra- and pentaBDE had the highest concentrations in sediment in the beginning of the period, but by the late 1980s, levels of decaBDE had surpassed levels of the other congeners, and remained dominant for the rest of the period.

The average concentrations of pentaBDE in Lake Ontario in sediment, fish and wildlife exceeded the pentaBDE FEQG for the entire period.

Increasing trends in PBDEs in air in northern Canada are indicative of long-range transport of PBDEs. At the same time, declines in Tetra-and decaBDEs have been evident in southern Ontario.

A rapid increase in the accumulation of decaBDE in Lake Ontario sediment was observed between the mid-1980s and the early 2000s, followed by a decline. This was not observed in other homologues. In sediment from Lake St.-Pierrre, Quebec, decaBDE only decreased an average of 30% at the same time that other homologues decreased by about 70%. An increasing trend in levels of decaBDE of > 10 ng/g from 2003 to 2008 was noted at other sites downstream from the Lake St.-Pierre site. (Note: The FEQG for decaBDE is 19 ng/g.)

Other homologues decreased during that period. This may be explained by the increased use of the decaBDE commercial mixture in the 1990s and 2000s and the known high tendency of DecaBDE to bind to sediment and low tendency to degrade within sediment.

Annual measurements of PBDE concentrations in Lake Ontario lake trout made by Environment Canada (1997–2009) were combined with data gathered through the United States Environmental Protection Agency (US EPA) Great Lakes Program (1980–2000). The dominant homologues throughout the period are tetra, penta- and hexaBDE. Tetra- and pentaBDE did show an overall increase over the period of 1980 to 2000, with a decreasing trend beginning thereafter.

HexaBDE showed a stabilization trend and a decrease beginning around 1995. The heavier homologues (hepta - through decaBDE) are not found in fish in substantial quantities due to their lower bioaccumulation potential. However, biotransformation or the breakdown from heavier to lighter homologues within organisms could contribute to the relative dominance of tetra- through hexaBDE. The report noted that levels of pentaBDE in fish tissue were 20–30-fold higher than the FEQG, but did not exceed the margin of safety built into the FEQG (88 ng/g).

Although concentrations of tetra-, penta and hexaBDE in wildlife are more variable from year to year than the other media, they appear to show a similar temporal trend of increase and subsequent decline to that in fish; however, the peak and decline occur several years later. In addition, decaBDE concentrations, although relatively low, increased over the entire period. Maximum average values for pentaBDE were up to 20-fold higher than the draft tissue residue guidelines for the protection of wildlife consumers (i.e. Canadian FEQG). While herring gull populations have been declining on the Great Lakes over the last decade, this is a basin-wide phenomenon and not specific to sites with high PBDEs. It is likely that this species may be less susceptible to the effects of PBDEs than is the American kestrel, upon which the above FEQG is based.

During the treatment of wastewater PBDEs partition to solids during wastewater treatment rather than being removed through biodegradation. The most prevalent homologue groups measured in biosolids were tetra-, penta-,nona- and decaBDE, representing approximately 17%, 19%, 9% and 48% respectively.

HBCD:

Limited data from the Great Lakes exists, and therefore it is difficult to definitively conclude whether relevant guidelines and/or benchmarks are being exceeded and to establish whether any spatial or temporal trends exist at this time.

4. Review of past, present and/ or planned science and risk management actions:

Is there a need for additional risk management and/or science activities and are there resources and/or tools available to support the delivery of such activities?

Canadian Federal Risk Management Activities:

PBDEs:

From: *Environment Canada, 2015, List of Toxic Substances Managed under CEPA 1999 (Schedule 1): Polybrominated Diphenyl Ethers (PBDEs).*

PBDEs were identified as a high priority for action in the Chemicals Management Plan, as announced by the Government in December 2006. Following the screening assessments of the Ministers of the Environment and Health, published on July 1, 2006, PBDEs have been identified as entering the environment in a quantity or concentration or under conditions that have or may have an immediate or long-term harmful effect on the environment or its biological diversity (i.e., “toxic” as defined under paragraph 64(a) of the *Canadian Environmental Protection Act, 1999*). The seven PBDE homologues included in these screening assessments are tetra-(CAS No. 40088-47-9), penta-(CAS No. 32534-81-9), hexa-(CAS No. 36483-60-0), hepta-(CAS No. 68928-80-3), octa-(CAS No. 32536-52-0), nona-(CAS No. 63936-56-1) and decaBDE-(CAS No. 1163-19-5).

In December 2006, Environment Canada and Health Canada published a Risk Management Strategy for PBDEs with the objective of reducing the concentration of PBDEs in the Canadian environment to the lowest level possible. The strategy uses a multi-instrument approach that combines regulatory and voluntary measures, the development of environmental quality guidelines, international co-operation and ongoing monitoring.

In July 2008, the Government of Canada published the *Polybrominated Diphenyl Ethers Regulations* to protect Canada's environment from the risks associated with PBDEs by preventing their manufacture and restricting their use in Canada, thereby minimizing their release into the environment. Specifically, the Regulations prohibit the manufacture of PBDEs in Canada (tetraBDE, pentaBDE, hexaBDE, heptaBDE, octaBDE, nonaBDE and decaBDE congeners); and prohibit the use, sale, offer for sale and import of those PBDEs that meet the criteria for virtual elimination under CEPA 1999 (tetraBDE, pentaBDE and hexaBDE congeners), as well as mixtures, polymers and resins containing these substances.

Since the completion and publication of the Ecological Screening Assessment Report in 2006, a large amount of new information has been published respecting the accumulation of one form of PBDE, decaBDE, in biota and its potential transformation to persistent and bioaccumulative products. This information has been summarized and evaluated in the Ecological State of Science Report on decaBDE (draft published in March 2009, final in August 2010). The outcome of this review and comments received from the public provided justification for the development of additional regulatory controls for this form of PBDE. In August 2010, the Government published a Final Revised Risk Management Strategy for PBDEs. In comparison with the Revised Risk Management Strategy, published in March 2009, the final Strategy broadens controls on all forms of PBDEs, including DecaBDE, for both substances and products containing them.

The Government's Consultation Document on the Proposed Risk Management Measure for PBDEs was published on February 5, 2013 for a 60-day public comment period. Comments received were considered during the development of the proposed risk management action. The proposed risk management measure was that prohibitions would apply to the manufacture, use, sale, offer for sale, import and export of all PBDEs (tetraBDE, pentaBDE, hexaBDE, heptaBDE, octaBDE, nonaBDE and decaBDE) and any resin or polymer containing these substances. Therefore, the Government of Canada proposes to implement regulations to extend the existing PBDE prohibition to prohibit the use, sale, offer for sale, and import to heptaBDE, octaBDE, nonaBDE and decaBDE. As a result, the commercial mixture DecaBDE would be prohibited.

The proposed *Regulations Amending the Prohibition of Certain Toxic Substances Regulations, 2012* include controls on PBDEs and was published in the *Canada Gazette*, Part I on April 4, 2015. The proposed Amendments would expand the scope of the existing prohibition for PBDEs to cover all PBDE substances (including deca-BDE) and products containing them, except manufactured items. The proposed Amendments are subject to a 75-day public comment period from April 4, 2015 to June 18, 2015. Comments received during this period will be considered for the final development of these Regulations. The *Polybrominated Diphenyl Ethers Regulations* will be repealed once the proposed Amendments are finalized and come into force.

The global use of PBDEs has significantly decreased since the 2000s due to numerous initiatives, including the phase-out of pentaBDE and octaBDE commercial mixtures internationally and the prohibition of the use of PBDEs in international agreements. As a result of these actions, it is anticipated that there will be fewer products containing PBDEs introduced into the Canadian market, resulting in declines in the quantity of PBDEs released to the environment. Over time, declines in PBDE levels in the environment are expected to occur.

FEQGs have been developed in Canada for certain congeners of PBDEs in water, fish tissue, sediment, wildlife (and bird eggs) to assess the ecological significance of levels of PBDEs in the environment. These FEQGs are benchmarks for aquatic ecosystems that are intended to protect all forms of aquatic life (vertebrates, invertebrates, and plants) from direct adverse effects.

Canada played an active role in the process of adding PentaBDE (c-PentaBDE) and OctaBDE (c-OctaBDE) to two international agreements: the Stockholm Convention on Persistence Organic Pollutants (POPs) and the Persistent Organic Pollutant (POPs) Protocol of the United Nations Convention on Long-range Transboundary Air Pollution (LRTAP). These chemicals have been added to these conventions as certain PBDEs contained in them have the ability to undergo long-range transport, are persistent and bioaccumulative and are deemed to have sufficient indications that they are likely to cause adverse effects as a result of their long-range transport (EC, 2010).

HBCD:

From: *Environment Canada, 2015, List of Toxic Substances Managed under CEPA 1999 (Schedule 1): Hexabromocyclododecane (HBCD).*

In Canada, t HBCD is found primarily in polystyrene foam (over 90% of its use) used as an insulation material in the building industry. There are also a number of other minor uses as a flame retardant, such as in textiles used for upholstered furniture and upholstered seating in transportation, and in wall coverings and draperies. It is also used in some glues, paints, adhesives and polymers contained in electronic equipment.

The final screening assessment report, published on November 12, 2011, concluded that HBCD is entering the environment in a quantity or concentration or under conditions that have or may have an immediate or long-term harmful effect on the environment or its biological diversity (i.e., meets the criteria of section 64(a) of the *Canadian Environmental Protection Act, 1999* (CEPA 1999)). Furthermore, the available data regarding its persistence and bioaccumulation potential indicate that HBCD satisfies the criteria outlined in the *Persistence and Bioaccumulation Regulations*, under CEPA 1999, and meets all other criteria to implement virtual elimination. The final screening assessment report concludes that HBCD is not a concern for human health at current levels of exposure.

The Risk Management Approach for HBCD, published on November 12, 2011, outlines the proposed risk management actions. Proposed in the Risk Management Approach, are restrictions on the manufacture, use, sale, offer for sale or import of HBCD and products containing the substance.

The Government's Response to Stakeholder's Comments regarding the Risk Management Approach and the Consultation Document on the Proposed Risk Management Measure for HBCD are available. There was a 60-day public comment period on the Consultation Document from October 3, 2012 to December 2, 2012. Comments received were considered during the development of the proposed risk management actions described below.

The proposed *Regulations Amending the Prohibition of Certain Toxic Substances Regulations, 2012* include controls on HBCD and were published in the *Canada Gazette*, Part I on April 4, 2015. The proposed Amendments would prohibit HBCD and certain products containing the substance. Time-limited exemptions are proposed for certain uses to allow industry to phase-out their use of HBCD. The proposed Amendments are subject to a 75-day public comment period from April 4, 2015 to June 18, 2015. Comments received during this period will be considered in the final development of these Regulations.

Furthermore, Canada plays an active role with regards to HBCD in two international agreements related to POPs: the Stockholm Convention and LRTAP – see *International Programs* below.

U.S. Federal Risk Management Activities:

PBDEs:

U.S. Domestic manufacture of c-pentaBDE and c-octaBDE stopped in 2004 when the Great Lakes Chemical Corporation (now Chemtura Corporation) voluntary phased out their production. The U.S. EPA subsequently promulgated a regulation (74 FR 34015, June 13, 2006) which requires that anyone who intends to manufacture or import a chemical substance or mixture containing any of the congeners present in c-pentaBDE or c-octaBDE notify EPA at least 90 days in advance. The notice provides EPA with an opportunity to evaluate the intended new use and, if necessary, take action to limit or prohibit it. The SNUR did not address importation of articles to which c-pentaBDE or c-octaBDE has been added;

In December 2009, EPA received commitments from the principal manufacturers and importers of c-decaBDE to initiate reductions in the manufacture, import and sales of c-decaBDE starting in 2010, with all sales to cease by December 31, 2013.

On April 2, 2012, EPA proposed to amend the PBDE SNUR by (1) designating processing of any combination of the six PBDE congeners contained in c-pentaBDE or c-octaBDE for any use which is not ongoing, as a significant new use (2) designating manufacturing, importing, or processing of decaBDE for any use which is not ongoing after December 31, 2013, as a significant new use and (3) designating the manufacture (including import) or processing of any article to which PBDEs had been added, as a significant new use. Any person who intended to import a PBDE as part of an article for a significant new use would be subject to significant new use reporting. Ongoing uses would be excluded from the SNUR

In January 29, 2014, through its DfE program, U.S. EPA released the final "An Alternatives Assessment for the Flame Retardant Decabromodiphenyl Ether (DecaBDE)".

HBCD:

On March 26, 2012, EPA proposed a significant new use rule (SNUR) under section 5(a)(2) of the

Toxic Substances Control Act (TSCA) for two chemical substances: Hexabromocyclododecane

(Chemical Abstracts Service Registry Number (CASRN) 25637-99-4) and 1,2,5,6,9,10-hexabromocyclododecane (CASRN 3194-55-6) (hereafter HBCD). This proposed rule would designate “use in consumer textiles, other than for use in motor vehicles” as a significant new use. This action would require persons who intend to manufacture (including import) or process HBCD for use in covered consumer textiles to notify EPA at least 90 days before commencing that activity. The required notification would provide EPA with the opportunity to evaluate the intended use and, if appropriate, to prohibit or limit that activity before it occurs. For this proposed rule, the general SNUR article exemption for persons who import or process chemical substances as part of an article would not apply. This rule is expected to be final in 2015.

In June 2014 EPA Design for the Environment (DfE) Program released its final report titled: Flame Retardant Alternatives for HBCD. The report identified possible viable flame retardant alternatives to HBCD for use in expanded and extruded polystyrene foam (EPS and XPS) insulation under current manufacturing processes. Based on DfE criteria and guidance, an alternative was identified that is anticipated to be safer than HBCD for multiple endpoints (EPA 2014).

International Programs:

PBDEs:

The Stockholm Convention entered into force in May 2004 and is an international legally binding agreement that has been ratified by 128 countries, including Canada. Under this convention, Parties are bound to take action to prohibit the manufacture and import of the chemicals listed on Annex A and B of the Convention.

In May 2009, the Fourth Conference of the Parties (COP4) decided to list PentaBDE and OctaBDE congeners (tetra-, penta-, hexa- and heptaBDE) to Part 1 of Annex A of the Convention, with specific exemptions for recycling articles, until 2030. COP4 also decided to establish work programs to further understand issues related to recycling and waste associated with the PentaBDE and OctaBDE listing. Listing in Annex A obliges the Parties, including Canada, to eliminate the production, use, export and import of the chemical.

The LRTAP Convention entered into force in 1953 and is an international legally binding agreement that has been ratified by 51 countries, including Canada. The Convention requires all Parties to endeavor to limit and, as far as possible, gradually reduce and prevent air pollution including long-range transboundary air pollution.

Parties to the LRTAP Convention have concluded that PentaBDE and OctaBDE are persistent organic pollutants. An assessment and management review of PentaBDE and OctaBDE has been completed by the UNECE's LRTAP. In December 2009 at the 27th session of the Executive Body, the components of the PentaBDE and OctaBDE commercial mixture (i.e. tetraBDE, pentaBDE, hexaBDE and heptaBDE) were added

HBCD:

United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution (LRTAP):

In December 2009, the Executive Body (EB) of the Commission agreed that HBCD meets the criteria for being a persistent organic pollutant (POP) under the Convention set out in EB decision 1998/2, that is:

HBCD has the potential for long-range transport and is found in remote regions; it has the potential to adversely affect human health and/or the environment; it is persistent and bioaccumulates; and its release into the environment is wide-dispersive (UNEP Task Force 2009).

The Stockholm Convention on POPs:

On June 18th 2008, Norway submitted a proposal to list HBCD as a possible Persistent Organic Pollutant (POP) under Annex A, of the Stockholm Convention. Subsequently, a draft risk profile for HBCD was prepared. [Annex A of the POPs Convention contains chemicals to be eliminated]

Provincial and State - Level Actions:

Restrictions and bans on the manufacture and use of PBDEs exist in California, Maine, Minnesota, Rhode Island, Hawaii, New York, Oregon and Washington.

PBDEs are designated as a chemical on the California Environmental Contaminant Biomonitoring Program list.

HCBD is designated as a chemical on the California Environmental Contaminant Biomonitoring Program list.

It is also included on the State of Washington Chemicals of High Concern for Children list under the Children's Safe Products Act and has been named a Priority Chemical in Minnesota because it is persistent, bioaccumulative, and toxic and it has been found in the home environment.

Identification of Gaps in Management and/or Science Activities

(1) Are environmental levels below applicable benchmarks and are there any discernable environmental trends?

With respect to PBDEs, concentrations in top predator fish (e.g. lake trout and walleye), sediment and herring gull eggs exceed relevant guidelines. Furthermore, while temporal trends in some fish species have shown recent declines, beginning in mid-2000, concentrations of some homologues in sediment and herring gull eggs have shown a stable or slightly increasing long-term trend.

With respect to HBCD, there is very limited Great Lakes data available and therefore it is not possible to definitively conclude whether present environmental concentrations exceed relevant benchmarks or guidelines or to establish temporal or spatial trends in the Great Lakes environment. However, data from other international and North American monitoring and surveillance activities suggest that HCBD is ubiquitous in the ambient environment.

(2) Is the GLB-relevant human health exposure being adequately addressed?

In general, PBDE concentrations in sport-fish fillets do not result in consumption advisories for the general or sensitive populations.

There is insufficient data available to establish whether human health is being impacted by HBCD through Great Lakes relevant sources of exposure:

(3) Are applicable/available objectives for the substances being met?

As noted in the report, concentrations in some environmental media (e.g. lake trout) have begun to show declines which appear to be aligned with the implementation of voluntary and regulatory risk management activities for PBDEs, concentrations of some homologues still routinely exceed benchmarks in sediment, fish and herring gull eggs in the Great Lakes basin.

Limited data from the Great Lakes exists for HBCD, and therefore it is difficult to definitively conclude whether relevant guidelines and/or benchmarks are being exceeded and to establish whether any spatial or temporal trends exist at this time.

(4) If no objectives exist for the substance, is progress being made towards reducing levels in the environment, generating needed data, etc.?

For PBDEs, progress is described in the question above. With regards to HBCD, the data available is insufficient to conclude whether progress is being made to reduce levels in the environment.

(5) If progress is not being made, are actions in place to expect progress (e.g., regulations that have yet to take effect);

There are forthcoming actions in Canada and the US for both PBDEs and HBCD (as described in Section 4 above) which are expected to result in reduced levels in the Canadian and US environment, including in the Great Lakes basin.

(6) Gaps in risk management, research or monitoring for the substance (e.g., ongoing releases of concern, knowledge needs, lack of monitoring data) and possible actions that would fill these gaps:

- Noting that actions to address HBCD and to further address PBDEs are forthcoming in Canada and the US, this should be considered a gap in risk management, until such a time that these actions are in force;

- There is a need / opportunity to address the use and disposal of products containing BFRs, including PBDEs and HBCD;

- For both PBDEs and HBCD, there is a need to initiate / continue activities to monitor long-term trends in air, sediment, and biota (top-predator fish and herring gull egg) in the Great Lakes, in order to establish and continue to track long-term trends; track issue of long range atmospheric transport and deposition; and evaluate the performance of existing and forthcoming risk management activities;
- As the relative contribution of environmental transformation of decaBDE to the total loadings of lower brominated PBDEs in the environment is not known, this could be considered a knowledge gap for future research related to bioaccumulation and persistence;
- More work is needed to understand the effects of debromination of PBDEs, particularly, in the long-term. For example, concentrations of BDE209 (the major component of the deca-mix PBDE product) are continuing to increase. Of major concern is the possible debromination of the potentially large reservoir of BDE209 in sediments, to yield lower brominated congeners which are both more mobile and more toxic;

5. Final Recommendation:

With respect to PBDEs, there was sufficient data and information available to effectively apply the *Binational Considerations*, and based on their application of the considerations, **the ITT has recommended, by a 2/3 majority decision, that PBDEs be designated as a CMC.**

With respect to HBCD, the ITT has concluded that there is insufficient data and/or information available to effectively apply the *Binational Considerations*. Therefore, **the ITT has recommended, by a 2/3 majority decision, that HBCD be identified as insufficient information on which to base a determination.**

While consensus was reached for both decisions, there were minor dissenting views. Given the information available and considering existing and forthcoming management actions:

- Some members felt that PBDEs should be designated not a CMC; and
- Some felt HBCD should be designated as a CMC while others felt it should be designated Not a CMC;

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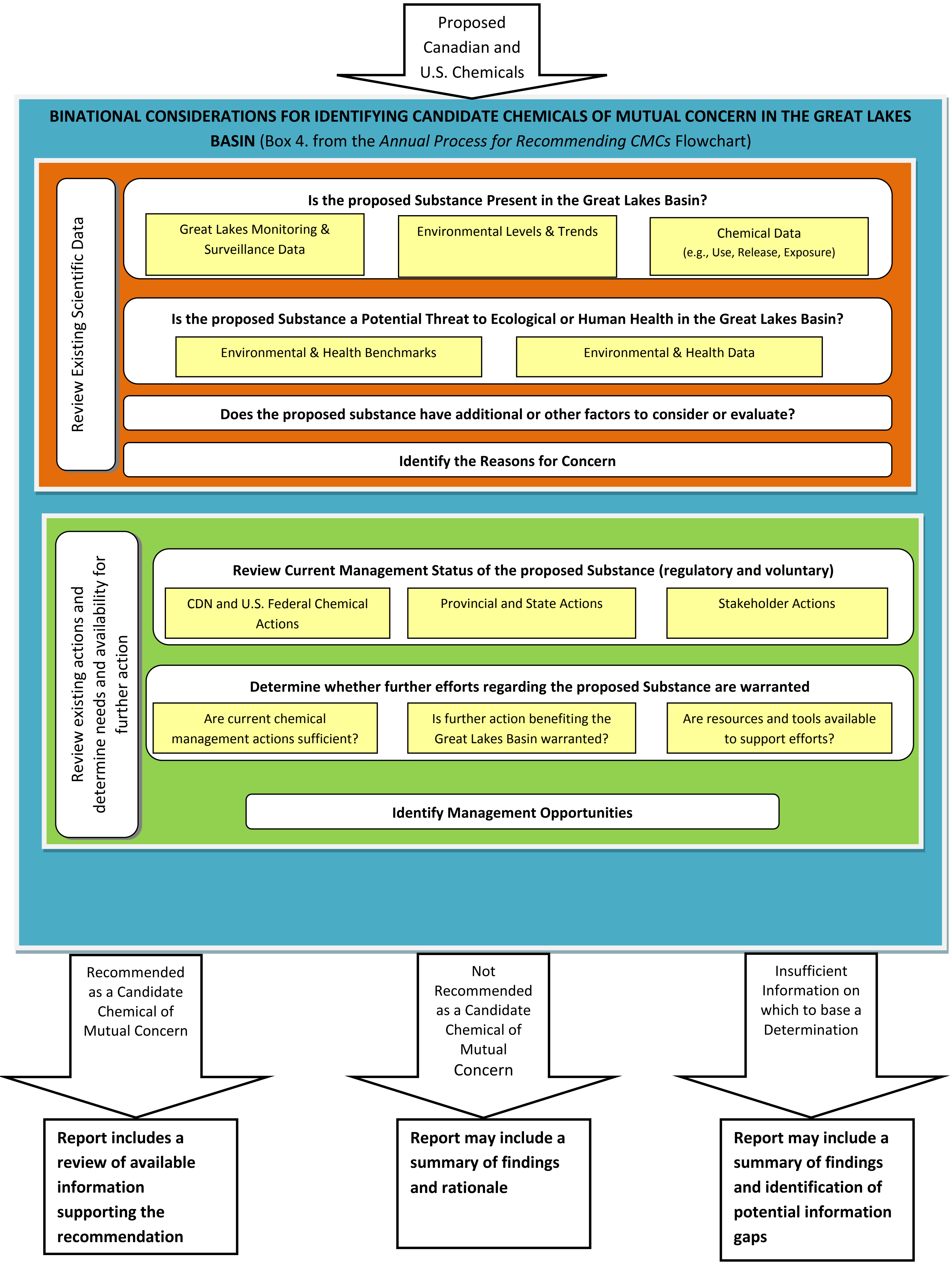
Appendix A:

Binational Considerations When Evaluating Candidate Chemicals of Mutual Concern


Proposed Canadian and U.S. Chemicals
BINATIONAL CONSIDERATIONS FOR IDENTIFYING CANDIDATE CHEMICALS OF MUTUAL CONCERN IN THE GREAT LAKES BASIN (Box 4. from the Annual Process for Recommending CMCs Flowchart)
Review Existing Scientific Data
Is the proposed Substance Present in the Great Lakes Basin?
Great Lakes Monitoring & Surveillance Data
Environmental Levels & Trends
Chemical Data (e.g., Use, Release, Exposure)
Is the proposed Substance a Potential Threat to Ecological or Human Health in the Great Lakes Basin?
Environmental & Health Benchmarks
Environmental & Health Data
Does the proposed substance have additional or other factors to consider or evaluate?
Identify the Reasons for Concern
Review existing actions and determine needs and availability for further action
Review Current Management Status of the proposed Substance (regulatory and voluntary)
CDN and U.S. Federal Chemical Actions
Provincial and State Actions
Stakeholder Actions
Determine whether further efforts regarding the proposed Substance are warranted
Are current chemical management actions sufficient?
Is further action benefiting the Great Lakes Basin warranted?
Are resources and tools available to support efforts?
Identify Management Opportunities
Recommended as a Candidate Chemical of Mutual Concern
Not Recommended as a Candidate Chemical of Mutual Concern
Insufficient Information on which to base a Determination
Report includes a review of available information supporting the recommendation
Report may include a summary of findings and rationale
Report may include a summary of findings and identification of potential information gaps

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