BINATIONAL FRAMEWORK FOR IDENTIFYING SUBSTANCES OF POTENTIAL THREAT TO THE GREAT LAKES BASIN

Test Case: Polybrominated Diphenyl Ethers (PBDEs)

I. FEEDERS FOR SUBSTANCE IDENTIFICATION

National Chemical Management Programs

Canada

PBDEs have been assessed CEPA toxic under the Canadian Environmental Protection Act (CEPA, 1999) and were added to CEPA 1999 Schedule 1- List of Toxic Substances.[1]

Brominated flame retardants (including all PBDEs) are included in the monitoring plan for Chemicals Management Plan (CMP) (either Year 1 or Year 2).[2]

Environment Canada is to publish three documents related to PBDEs on March 28th, 2009. These documents are: State of Science on DecaBDE, Performance Agreement for use of DecaBDE in Canada and a Revised Risk Management Strategy for PBDEs.

United States

Decabromodiphenylether (DecaBDE) is a High Production Volume (HPV) chemical.[3]

Great Lakes Monitoring and Surveillance

Yes, there are specific PBDEs (identified as PBDE congeners) substances common with the Great Lakes Screening Project. These include:[4]

PBDE Congener	*CAS No.*
tetrabromodiphenyl ether (tetraBDE)	40088-47-9
pentabromodiphenyl ether (pentaBDE)	32534-81-9
hexabromodiphenyl ether (hexaBDE)	36483-60-0
heptabromodiphenyl ether (heptaBDE)	68928-80-3
octabromodiphenyl ether (octaBDE)	32536-52-0
nonabromodiphenyl ether (nonaBDE)	63936-56-1
decabromodiphenyl ether (decaBDE)	1163-19-5

Other Sources of Information

United States

PBDEs: Laws have been enacted and legislation introduced relating to PBDEs in many U.S. States (The Toxics Steering Group and Polybrominated Diphenyl Ethers Subcommittee 2008), including California, Maine, Michigan, New York, Hawaii, Washington, Maryland, Rhode Island, Oregon and Illinois. These legislative initiatives initially addressed the PentaBDE and OctaBDE commercial mixtures.

DecaBDE: Several states are now proposing restricted uses of DecaBDE as well. Many of the legislative initiatives concerning DecaBDE are still under discussion by the States in question. However, two States, Washington and Maine, have signed into law legislation prohibiting the use of DecaBDE in certain consumer products:

The state of Washington has restricted the manufacture, sale, and use of DecaBDE in mattresses as of January 2008. That restriction has now been extended to televisions, computers and residential upholstered furniture, effective January 2011.

The state of Maine has restricted the use of DecaBDE in mattresses and upholstered furniture as of January 1, 2008. The state will be phasing in that restriction for televisions and other plastic-cased electronics by January 1, 2010.

Europe

PBDEs: The European Community has adopted Directives to minimize PentaBDE, OctaBDE commercial mixtures in electrical and electronic equipment (EEE) (under the Restriction of the Use of Certain Hazardous Substances in Electrical and Electronic Equipment –RoHS Directive) and to manage EEE waste (under the Waste Electrical and Electronic Equipment – WEEE Directive). As of July 1, 2006, specified EEE products are not allowed to be placed on the European Union (EU) market if a product contains more than 0.1%/wt of PentaBDE or OctaBDE.[5] A draft proposal to update to the EU RoHS Directive (Commission of the European Communities 2008) was published in late 2008 which is aimed at increasing the number of product groups affected by the Directive, in addition to changes in labeling requirements and the streamlining of some requirements. There are currently no exemptions affecting PBDEs in the Directive.

The European Community has also adopted a Directive, Directive 2003/11/EC, banning the manufacture or use of PentaBDE and OctaBDE commercial mixtures and the sale of products containing more than 0.1% PentaBDE or OctaBDE.[6]

DecaBDE: The European Community has adopted Directives to minimize DecaBDE commercial mixtures in electrical and electronic equipment (EEE) (under the Restriction of the Use of Certain Hazardous Substances in Electrical and Electronic Equipment –RoHS Directive) and to manage EEE waste (under the Waste Electrical and Electronic Equipment – WEEE Directive). As of July 1, 2008, specified EEE products are not allowed to be placed on the European Union (EU) market if a product contains more than 0.1%/wt of DecaBDE.[7]

Norway

PBDEs: In Norway, products containing more than 0.25% PentaBDE or OctaBDE commercial mixtures are classified as hazardous waste when they are discarded.

DecaBDE: Norway proposed, in late 2007, the PoHS (Prohibition of certain Hazardous Substances) Regulations. Included in the proposed Regulations were restrictions on the use of DecaBDE in all manufactured products, with the exception of transportation applications (the restriction applies to concentrations > 0.1% by weight, same as in EU RoHS). Norway has yet to finalize the regulations. In addition, in Norway, products containing more than 0.25% DecaBDE commercial mixtures are classified as hazardous waste when they are discarded.

Japan

DecaBDE: Japan has a law concerning the reporting of releases of specific chemical substances and promoting improvements in their management. Under this law, yearly reports are required on volumes of DecaBDE commercial mixture imported and used and quantities released to the environment.[8]

China

PBDEs: China's Ministry of Information Industries issued the Management Methods for the Prevention and Control of Pollution Caused by Electronic Information Products (sometimes referred to as CRoHS). The Methods provide a broad regulatory framework for restrictions of "toxic and hazardous substances", including PBDEs.[9] The first phase, in force since March 1, 2007, requires labeling and disclosure of the content of the substances within EEE products, but imposes no restrictions. The second phase, which will include restrictions, has yet to be implemented.

Korea

PBDEs: Korea implemented a law which covers end-of-life and EU RoHS type restrictions on electronic products and vehicles. Exemptions, limit values, and restricted substances are the same as the EU RoHS Directive. As of July 1, 2008 a restriction similar to EU RoHS, including for PBDEs, came into force for a subset of the product types included in EU RoHS

Sweden

DecaBDE: Sweden reversed a ban on DecaBDE in textiles, furniture and some cables. Instead, the Swedish government is seeking a total ban on the use of DecaBDE at the EU level.

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

Parties to the LRTAP Convention have concluded that c-PentaBDE and c-OctaBDE are persistent organic pollutants (POP). An assessment and management review of c-PentaBDE and c-OctaBDE has been completed by the UNECE's LRTAP. In December 2008, there was consensus on adding c-PentaBDE and c-OctaBDE to Annex I of the POPs Protocol to eliminate their production and use. Related discussions on exempted uses are ongoing.

POP Review Committee under UN Stockholm Convention

Under the convention, the Persistent Organic Pollutants Review Committee (POP RC) has concluded that both c-PentaBDE and c-OctaBDE are persistent organic pollutants. Risk management evaluations for c-PentaBDE and c-OctaBDE have been prepared. The Review Committee decided to recommend to the fourth Conference of the Parties (COP4) that it consider listing c-PentaBDE (including tetra- and penta-BDEs) and c-OctaBDE (including hexa- and hepta-BDEs) in Annex A of the Stockholm Convention, without exemptions. Listing in Annex A obliges the Parties, including Canada, to eliminate the production, use, export and import of the chemical. In May 2009, COP4 is expected to make decisions on these recommendations.

II. CONSIDERATIONS FOR SUBSTANCE SELECTION

Monitoring and Surveillance

Numerous monitoring and surveillance data are available which demonstrates the presence of PBDEs in the Great Lakes Basin. For example:

- Environment Canada 2006, "Canadian Environmental Protection Act, 1999- Ecological Screening Assessment Report on Polybrominated Diphenyl Ethers (PBDEs).[10]
    - Reports concentrations in Great Lake water, sediments, biota, ambient environment & sewage sludge.
    - Luckey *et al.* 2002 measured total (dissolved and particulate phases) PBDE (mono- to heptaBDE congeners) of approximately 9 pg/L in Lake Ontario surface waters in 1999. More than 60% of the total was composed of tetraBDE and pentaBDE, with pentaBDE and heptaBDE congeners each contributing approximately 5 to 8% of the total.
    - Stapleton and Baker 2001 analyzed water samples from Lake Michigan in 1997, 1998 and 1999 and found that total PBDE concentrations ranged from 31 to 158 pg/L.
    - Kolic *et al.* 2004 determined levels of PBDEs in sediments from Lake Ontario tributaries flowing to Lake Ontario. The total PBDEs (tri, tetra, penta, hexa, hepta, and decaBDEs) measured in sediment samples taken from 14 tributary sites (6 reported) ranged from approximately 12 to 430 µg/kg dw. Concentrations of tetra- to hexaBDEs ranged from approximately 5 to 49 µg/kg dw. Concentrations of BDE209 (decaBDE) ranged from 6.9 to 400 µg/kg dw. BDE 47 (hexaBDE), 99 (pentaBDE), and 209 were the predominant congeners measured in sediments.
    - Muir *et al.* (2003) measured concentrations of BDE209 along a north-south transect from southern Ontario/upper New York state to Ellesmere Island. The highest concentrations of BDE209 (up to12 µg/kg dw) occurred in sediments collected from the western basin of Lake Ontario.

Table 4. Measured concentrations of PBDEs in the North American ambient environment and sewage sludge

Medium	Location; year	Total PBDEs	Reference
Air	Alert, Canada; 1994–1995	1–28 pg/m^3	Alaee et al. 2000
Air	Great Lakes; 1997–1999	5.5–52 pg/m^3	Strandberg et al. 2001
Air	Southern Ontario; 2000	10–1300 pg/m^3	Gouin et al. 2002
Air	Ontario; 2000	3.4–46 pg/m^3	Harner et al. 2002
Water	Lake Michigan; 1997–1999	31–158 pg/L	Stapleton and Baker 2001
Water	Lake Ontario; 1999	6 pg/L	Luckey et al. 2002
Sediment	Lake Michigan; 1998	4.2 µg/kg dw	Stapleton and Baker 2001
Sediment	British Columbia; 2001	2.7–91 µg/kg OC	Rayne et al. 2003a
Soil	United States; 2000	<0.1–76 µg/kg dw	Hale et al. 2002
Sewage sludge	Toronto, Canada United States	8280 µg/kg dw 730–24 900 µg/kg dw	La Guardia et al. 2001
Sewage sludge	United States; 2000	3005 µg/kg dw	Hale et al. 2002
Sewage sludge	Southern Ontario	1700-3500 µg/kg dw	Kolic et al. 2004

dw = dry weight; OC = organic carbon

Table 5. Measured concentrations of PBDEs in North American biota

Organism	Location; year	Total PBDEs	Reference
Dungeness crab hepatopancreas	West coast, Canada; 1993–1995	4.2–480 µg/kg lipid	Ikonomou et al. 2002b
Mountain whitefish (muscle)	Columbia River, British Columbia; 1992–2000	0.726–131 µg/kg ww	Rayne et al. 2003a
Heron egg	British Columbia; 1983–2000	1.308–288 µg/kg ww	Wakeford et al. 2002
Murre egg	Northern Canada; 1975–1998	0.442–2.93 µg/kg ww	
Fulmar egg	Northern Canada; 1975–1998	0.212–2.37 µg/kg ww	
Beluga whale blubber	Canadian Arctic	81.2–160 µg/kg lipid	Alaee et al. 1999
Herring gull egg	Great Lakes; 1981–2000	9.4–1544 µg/kg ww	Norstrom et al. 2002
Lake trout	Lake Ontario; 1997	95 µg/kg ww	Luross et al. 2002
	Lake Erie; 1997	27 µg/kg ww	
	Lake Superior; 1997	56 µg/kg ww	
	Lake Huron; 1997	50 µg/kg ww	
Rainbow trout	Spokane River, Washington, USA; 1999	297 µg/kg ww	Johnson and Olson 2001
Mountain whitefish		1250 µg/kg ww	
Largescale sucker		105 µg/kg ww	
Carp	Virginia, USA; 1998-1999	1140 µg/kg ww	Hale et al. 2001

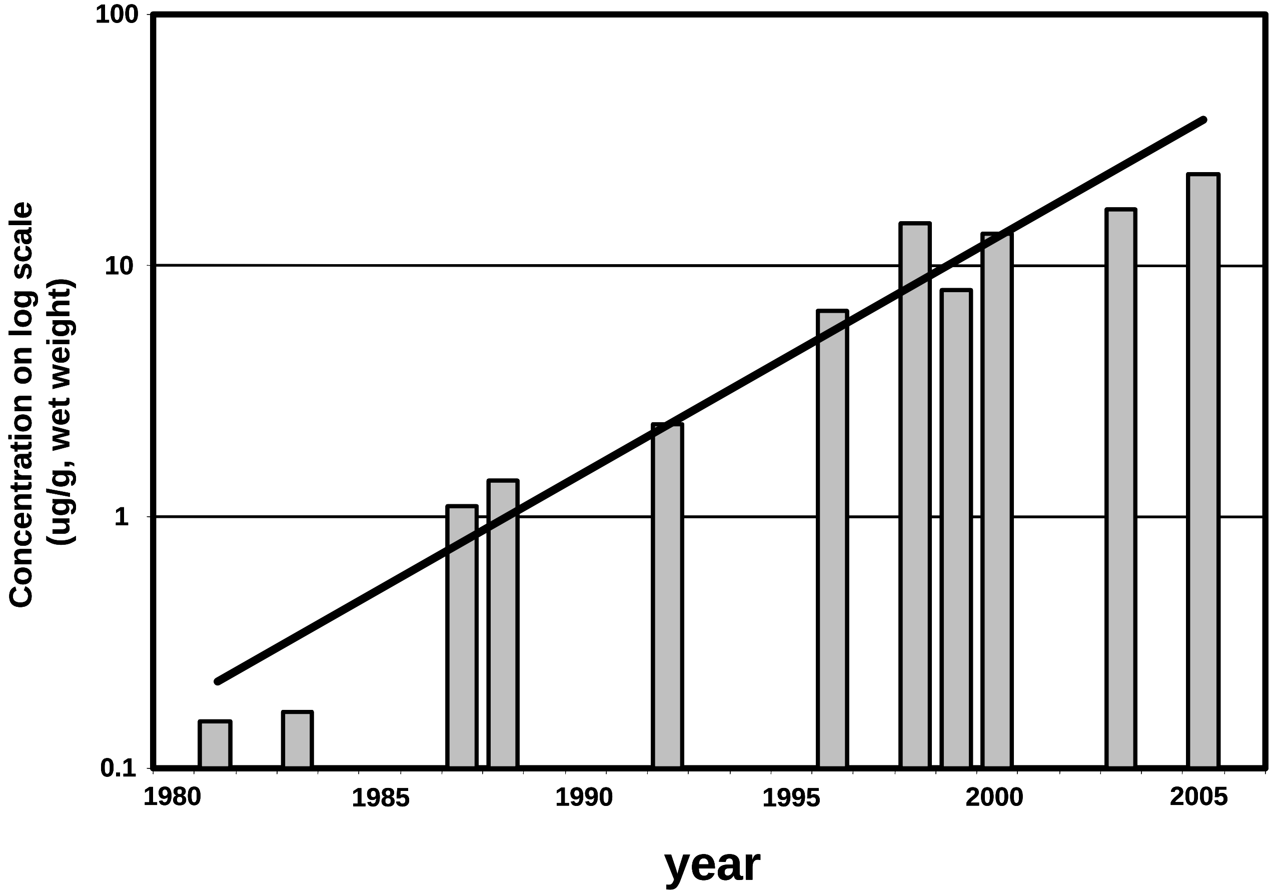
- Report available at: http://www.ec.gc.ca/CEPARegistry/documents/subs_list/PBDE_SAR/PBDEs_SAR_EC_June_2006_(en).pdf.

Environmental Levels and Trends

Some trends identified in Environment Canada's 2006 Ecological Screening Assessment Report on Polybrominated Diphenyl Ethers (PBDEs) are as follows:

- Concentrations of PBDEs in herring gull eggs have increased exponentially between 1981 and 2000 at Lake Ontario, Huron and Michigan sampling sites (see figure below).
- Concentrations of PBDEs (predominantly tetra- and pentaBDE congeners) have also increased exponentially between 1981 and 2000 in Arctic male ringed seals.[11]
- PBDEs have been detected in all environmental media as well as sewage sludge, and there is evidence that their levels in the North American environment are increasing.[12]

A summary of studies of PBDEs in the Great Lakes environment is presented in Appendix A.

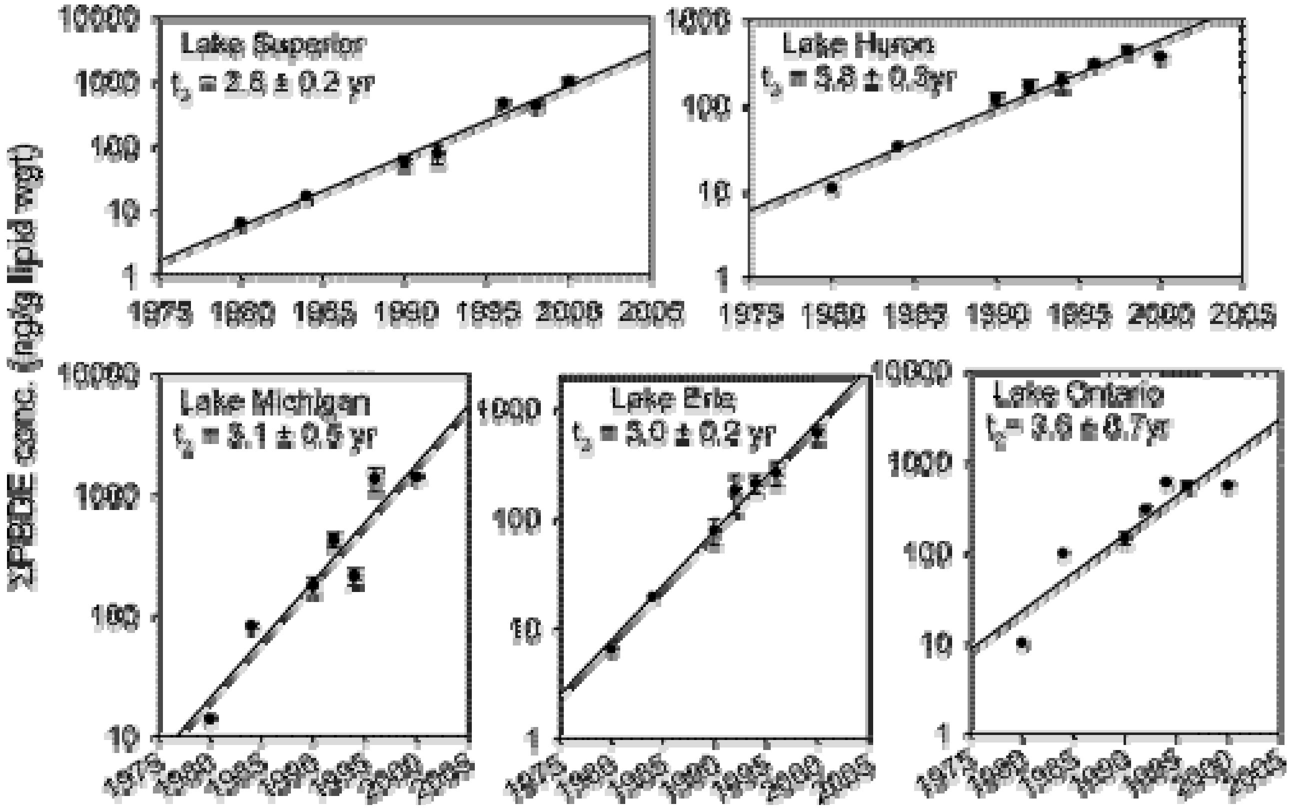


PBDE in Herring Gull Eggs, Gull Island, Lake Michigan, 1981-2005. Source: Great Lakes Binational Toxics Strategy 2006 Annual Progress Report.

A spatial distribution of PBDEs in lake trout from the Great Lakes and temporal trends in lake trout in Lake Ontario along with biomagnification of PBDEs in Lake Ontario food web were accomplished under the supervision of Dave Sergeant and Mike Whittle. PBDE levels ranged between 22 ng/g-wet weight for Lake Erie and 84 ng/g-wet weight for Lake Ontario. The levels of PBDEs in lake trout from Lake Ontario increased from 0.12 ng/g-wet weight in 1978 to 148 ng/g-wet weight in 1998. Biomagnification factors for PBDEs in Lake Ontario food web ranged between 0.6 for transfer of BDE-99 from diporeia to sculpin, and 5.7 for transfer of BDE-100 from plankton to mysid.[13]

A more recent analysis indicates that concentrations of PBDEs in lake trout from Lake Ontario have increased exponentially from 0.54 ng/g in 1988 to 190 ng/g wet weight in whole fish samples collected in 2002 (Whittle et al., 2004). From archived lake trout samples from the five Great

Lakes, total PBDE concentrations increased exponentially with time, doubling every 3 to 4 years (see figure below).[14]



Temporal Trend of Total PBDE Concentrations in Fishes from the Great Lakes, 1975-2005. Source: Great Lakes Binational Toxics Strategy 2006 Annual Progress Report.

In 2008, Venier and Hites reported that concentrations of PBDEs in air at IADN sites on the Great Lakes during 2005-2006 were highest at the urban sites in Chicago and Cleveland (65±4 and 87±8 pg/m^3, respectively), and lowest at the remote site in Eagle Harbor (5.8 ± 0.4 pg/m^3). Concentrations of BDE-209 (decaBDE) were not decreasing at any of the five IADN sites analyzed. Concentrations of BDE-47 and BDE-99 in the atmosphere were decreasing rapidly, except at the Chicago site, with half-lives of ~2 years.[15]

Venier and Hites also estimated the atmospheric deposition of PBDEs into the Great Lakes during 2005-2006. Total PBDEs measured in precipitation in this study ranged from 94 ± 19 ng L^{-1} at Chicago to 0.65 ± 0.14 ng L^{-1} for the rural Sturgeon Point site. The study findings also suggested that Chicago may be a source of BDE-47 to Lake Michigan and that Cleveland may be a source of BDE-209 to Lake Erie.[16]

Source/Use/Release/Exposure Information

Source/Use

PBDEs: PBDEs are not manufactured in Canada.

The only United States manufacturer of PentaBDE and OctaBDE commercial mixtures voluntarily ceased production in December 2004.[17]

DecaBDE: The DecaBDE commercial mixture is imported into Canada for addition to various intermediate and finished products, such as computer housings, household appliances, furniture, automotive/aircraft seating and interiors, and a variety of electrical and electronic components. [18]

DecaBDE is manufactured in the United States. Trends in releases of DecaBDE are being followed by the United States through the Toxics Release Inventory. [19]

Release

PBDEs: PBDEs are potentially released to the environment throughout their lifecycle, from the chemicals themselves as well as from products containing them. PBDEs may enter the environment through treated or untreated municipal or industrial wastewater discharges to surface water and also through leachate from landfills and municipal incineration when products and materials containing these substances are sent for final disposal. PBDEs may also be released directly to air, land and surface water when products containing PBDEs are manufactured and during their use.[20]

It is estimated that releases from the commercial mixture PentaBDE are primarily associated with in-service use of polyurethane foam and to a lesser extent disposal of the foam at the end of its lifecycle. The majority of releases from the use of the OctaBDE commercial mixture are estimated to be associated with product disposal and materials handling before resin compounding.

DecaBDE: Releases of the DecaBDE commercial mixture from plastics are mainly associated with product disposal; whereas releases of the DecaBDE commercial mixture associated with textile applications appear to be spread relatively evenly throughout the lifecycle – with most releases being associated with textile processing/finishing and releases during product service life. Releases of DecaBDE commercial mixture during service life of textiles would occur during washing and would be to wastewater.[21]

Exposure:

It is difficult to derive meaningful estimates of exposure to individual congeners or congener groups. However, exposure is related to releases.[22]

Pathways of exposure include pathways of exposure food, house dust, and indoor air. Research has suggested that house dust may be a substantial contributor to PBDE exposure. To further examine exposure pathway issues, EPA and the USDA are collaborating on a study of the bioavailability of PBDEs in house dust.[23]

In a study of U.S. exposures to PBDEs, an adult intake dose of total PBDEs was estimated to be 7.7 ng/kg body weight/day, and children's estimated intakes were higher at 49.3 ng/kg/day for ages 1–5, 14.4 ng/kg/day for 6–11, and 9.1 ng/kg/day for 12–19. The much higher dose for the child aged 1–5 was due to the doubling of dust ingestion from 50 to 100 mg/day. Exposures to PBDEs in house dust accounted for 82% of the overall estimated intakes.[24] Huwe et al. 2008 found that PBDEs in household dust are readily bioavailable and that ingestion of PBDE-contaminated dust by rats leads to bioconcentration in the liver.[25]

Concentrations of PBDEs in the blood of the general U.S. population were measured as part of the National Health and Nutrition Examination Survey (NHANES) 2003–2004. A total of 2,062 serum samples, from participants in NHANES aged 12 years and older, were analyzed for PBDEs. The highest serum concentration was found for 2,2´,4,4´-tetrabromodiphenyl ether (BDE-47) with a geometric mean of 20.5 ng/g lipid, followed by 2,2´,4,4´,5,5´-hexaBDE (BDE-153) [5.7 ng/g lipid], 2,2´,4,4´,5-pentaBDE (BDE-99) [5.0 ng/g lipid; a value equal to the highest limit of detection for an individual sample], 2,2´,4,4´,6-pentaBDE (BDE-100) [3.9 ng/g lipid], BB-153 [2.3 ng/g lipid], and 2,4,4´-triBDE (BDE-28) [1.2 ng/g lipid]. In examining those with the highest serum concentrations (above the 95th percentile) of BDE-47, age, but not sex and race/ethnicity, was significantly associated. Senior adults (60 years and older) were two times more likely to have serum concentrations of BDE-47 above the 95th percentile than adults aged 20–59 years and 54% more likely than adolescents to have serum BDE-47 concentrations above the 95th percentile.[26]

Anderson et al. 2008 investigated serum concentrations of PBDEs in a cohort of 508 consumers of sport-caught fish living in five Great Lake states. BDE-47 was found in 98% of samples, and BDE-99 in 62% of samples. Total PBDE levels were positively associated with age, hours spent outdoors, years of sportfish consumption, and catfish and shellfish intake; other dietary components were not predictive of PBDE levels.[27]

Spliethoff et al. 2008 examined PBDE body burdens among 36 sportfish consumers in New York State. The authors found that the number of years of Lake Ontario sportfish consumption between 1980 and 1990, after adjusting for plasma lipids, was a statistically significant predictor of total PBDE blood plasma levels. In addition, the number of meals eaten in the year prior to the study, of Lake Ontario sportfish species known to have high levels of other persistent organic pollutants (POPs), was correlated with total PBDE blood plasma levels.[28]

Concentrations of PBDEs in human breast milk from across Canada ranged between 19.08 ng/g for samples collected from the Maritimes and 2.57 ng/g for samples collected in Ontario. Levels of PBDEs in breast milk rose from a mean of 0.2 ng/g for samples collected across Canada in 1982 to a mean of 42.5 ng/g (25.4 ng/g median) lipid weight for samples collected in the Vancouver area in 2001-2.[29] Schecter et al. 2005 analyzed PBDEs in breast milk in the U.S. The highest 5% of breast milk levels in the U.S. were 6 to 21 ng/g wet weight.[30]

Concentrations of PBDEs in a number of food basket items ranged between 0.04 ng/g for pasta and 1.2 ng/g for wieners; these values resulted in an estimated daily dietary intake of 44 ng, which was higher than that for PCDD/Fs (2.4 ng), and lower than that for PCBs (285 ng).[31]

Environmental Benchmarks

There are no environmental quality benchmark criteria available from the United States.

The Government of Canada is developing federal environmental quality guidelines for PBDEs in the appropriate media as targets for acceptable environmental quality, to assist in the interpretation of monitoring data, and to serve as performance indicators of risk management actions.

Environmental and Health Data

PBDEs: Information on health data can be found from Health Canada's State of the Science Report for a Screening Health Assessment for PBDEs [Tetra, Penta, Hexa, Hepta, Octa, Nona, and Deca] (December 2004).[32]

Chronic toxicity/carcinogenicity studies of BDE-99 (a component of commercial pentaBDE) are not available. There is "inadequate information to assess the carcinogenic potential" of BDE-99. However, the available animal data indicate that the nervous system is a sensitive target. Neurobehavioral developmental toxicity has been identified as the critical endpoint of concern in mice following pre- and neonatal oral exposure to BDE-99.[33]

Herbstman found that BDEs 47, 99, and 100 were associated with lower mental development scores at age 4.[34]

An RfD of 0.1 µg/kg-day was calculated from a BMDL1SD of 0.32 mg/kg-day for effects on spontaneous motor behavior in mice. The database for BDE-99 is sparse for the derivation of an RfD: there are no standard reproductive, developmental, subchronic, or chronic studies in rats or mice nor a much needed developmental neurotoxicity study. In addition, there are several concerns regarding the experimental design of the Viberg et al. (2004a) study used in proposing an RfD. The overall confidence in the RfD assessment is low.[35]

DecaBDE: Johansson et al. 2008 reported that neonatal exposure to DecaBDE (BDE-209) causes dose-response changes in spontaneous behaviour and cholinergic susceptibility in adult mice, and that BDE-209 can be as potent as the lower brominated PBDEs in causing developmental neurotoxic defects.[36]

No information is available on the carcinogenicity of decaBDE in humans.[37] Chronic dietary studies of decaBDE have been conducted in rats and mice. The weight of evidence suggests that decaBDE shows "suggestive evidence of carcinogenic potential."[38]

In Europe the FIRE project (Flame retardants Integrated Risk assessment for Endocrine effects; see www.rivm.nl/fire) completed a 28-day toxicity study of decaBDE in rats. Van der Ven reported results of the 28-day toxicity study, which suggested that the endocrine effects of decaBDE may present a reproductive human health hazard. The study also reported that body burdens in children, with upper values reported as high as “233 ng/g lipid (Fischer et al., 2006) and 23 ng/g blood (Watson, 2005),” suggest that the need for further risk reduction measures be considered.[39]

In addition, a separate study of decaBDE developmental neurotoxicity in mice has been published (Rice et al., Neurotoxicology and Teratology 2007).[40]

Other Reasons for Concern

PBDEs: Evidence suggests that PBDEs have endocrine disrupting properties.[41] Toxicity data indicate potential for thyroid hormone effects (inhibition of tail resorption in tadpoles exposed to a commercial PBDE mixture).[42] Turyk et al. 2008 linked PBDE exposure with changes in thyroid antibodies and thyroid hormone homeostasis in men, reporting that “PBDE exposure, at levels comparable to the general US population, was associated with increased thyroglobulin antibodies and increased thryoxine in adult males.”[43]

The animal model indicates a potential for concern for early lifetime exposure (i.e., fetal or infant exposure) to the chemical. The identification of BDE-99 in human maternal and cord serum, milk, and children’s serum (Mazdai et al., 2003; Schecter et al., 2003; Thomsen et al., 2002) implies humans are exposed to BDE-99 during a period of rapid development of the brain, a critical window of development, indicating a potential for susceptibility. Whether such exposure constitutes a health risk for adverse neurodevelopmental effects in children is not known at this time because of the limited toxicological data base for BDE-99. An association between prenatal or neonatal exposures to BDE-99 and neurobehavioral dysfunction in humans has not been established.[44]

DecaBDE: Riu et al. 2008 found that DecaBDE in late gestation rats is absorbed and reaches the fetus, where it debrominates into OctDBE and other metabolites.[45] These findings suggest potential concern for human fetal exposures to DecaBDE.

III. <u>PRESENT MANAGEMENT STATUS</u>

<u>Canada</u>

PBDEs: *Polybrominated Diphenyl Ethers Regulations* (PBDE Regulations) were published in the *Canada Gazette*, Part II, on July 9, 2008. The Regulations prohibit the manufacture of PBDEs (i.e. tetra-, penta-, hexa-, hepta-, octa-, nona-, and decaBDE congener groups) in Canada. These regulations also prohibit the manufacture, use, sale, offer for sale and import of mixtures, polymers and resins containing those PBDEs that meet the criteria for virtual elimination (tetra-, penta- and hexaBDE congener groups) under the *Canadian Environmental Protection Act, 1999 (CEPA, 1999).* More information on this regulation can be found at: http://www.ec.gc.ca/ceparegistry/documents/regs/g2-14214_r1.pdf.

DecaBDE: The import and use of decaBDE is permitted in Canada.

United States

PBDEs: The EPA has issued a Significant New Use Rule (SNUR) that will require the notification of, and evaluation by, the USEPA of any new use of PentaBDE or OctaBDE commercial mixtures (71 FR 34015 Available at: http://www.epa.gov/oppt/pbde/).[46]

In March 2006, EPA prepared a Project Plan for PBDEs, which outlined EPA's activities regarding PBDEs. Since 2006, EPA has prepared an updated status report on PBDEs annually. EPA's Project Plan and latest status report (March 2008) are available at http://www.epa.gov/opptintr/pbde/. Highlights of the March 2008 status report are presented below.

EPA has formed a Furniture Flame Retardancy Partnership with several industry associations and other stakeholders. The partnership has conducted a screening level hazard assessment of flame retardant chemicals that may be suitable substitutes for pentaBDE. The summary assessment is available at http://www.epa.gov/opptintr/dfe/pubs/projects/flameret/index.htm.

The U.S. Consumer Product Safety Commission (CPSC) has proposed a new mandatory standard to address residential upholstered furniture fires. Under the proposal, manufacturers could meet the performance standard by using smolder-resistant cover fabrics or interior fire resistant barriers to protect the furniture's internal filling material, without requiring the use of fire retardant chemicals.

A program called the Electronic Product Environmental Assessment Tool (EPEAT) was created by a multi-stakeholder group in the US. EPEAT is a procurement tool used to identify desktop computers, notebooks and monitors that satisfy established environmental criteria, including conformance to the European RoHS (i.e. products that may have PBDEs).[47]

DecaBDE: Trends in releases of decabromodiphenyl oxide (decaBDE) are being followed by the US through the Toxics Release Inventory. In June 2008 the U.S. EPA released the final Integrated Risk Assessment Information System (IRIS)1 assessment of decaBDE.

EPA is considering proposing a SNUR under the Toxic Substances Control Act (TSCA) for flame retardants, which would include decaBDE among 16 chemical substances/categories. The proposed SNUR is on hold at EPA pending the CPSC's decision on a new mandatory standard for residential upholstered furniture flammability.

The industry sponsor of decaBDE in the Voluntary Children's Chemical Evaluation Program (VCCEP) committed to conducting a Tier 2 VCCEP assessment for decaBDE in December 2005. New documents will be posted at http://www.epa.gov/oppt/vccep/pubs/chem21.htm.[48]

APPENDIX A
Brominated Flame Retardants in the Great Lakes Environment - Published Studies

1. "Concentrations and spatial variations of polybrominated diphenyl ethers and other organohalogen compounds in Great Lakes air." B. Strandberg, N. G. Dodder, I. Basu and R. A. Hites. Environmental Science & Technology. 2001; 35(6): 1078-1083. PBDEs\Strandberg et. al. 2001.pdf

Summary: This study measured PBDEs in air around the Great Lakes and compares it to well studied compounds such as PCBs and organochlorine pesticides. PBDEs were found in every air sample indicating these compounds are prevalent and well distributed and dispersed in Great Lakes air. Concentrations of PBDEs were similar to that of pesticides like DDT. Spatial trends were similar to PCBs, indicating that gas-particle partitioning is important in understanding the fate of PBDEs.

2. "Geographical Distribution (2000) and Temporal Trends (1981-2000) of Brominated Diphenyl Ethers in Great Lakes Herring Gull Eggs." R.J. Norstrom, M. Simon, J. Moisey, B. Wakeford, and D.V. Chip Weseloh. Environmental Science & Technology.) 2002; 36: 4786-4789. PBDEs \Norstrom et.al. 2002.pdf

Summary: This study measured the concentrations of PBDEs in Herring Gull eggs from 1981-2000, and showed distribution of PBDEs in the Great Lakes for the year 2000. Highest concentrations were found in northern Lake Michigan and Toronto harbor (1000-1400 mg/kg) and lowest in Lake Huron and Lake Erie (192-340 mg/kg). The distribution suggested that input from large urban/industrial areas through air or water emissions contributes local contamination to the herring gull food web. Temporal trends for pentaPBDEs showed an exponential increase. Doubling times for pentaPBDEs were calculated to be 2.6 years in Lake Michigan, 3.1 years in Lake Huron and 2.8 years in Lake Ontario.

3. "Time Trends (1983-1999) for Organochlorines and Polybrominated Diphenyl Ethers in Rainbow Smelt (Osmerus Mordax) from Lakes Michigan, Huron, and Superior, USA". S.M. Chernyak, C.P. Rice, R.T. Quintal, L.J. Begnoche, J.P. Hickney, and B.T. Vunyard. Environmental Toxicology and Chemistry. 2005; 24,(7):1632-1641. PBDEs\Chernyak et al 2005.pdf

Summary: The USGS Great Lakes Center collected fish from the early 1980's until today, this study measured organochlorines and PBDEs in the fish to understand contaminant trends in the Great Lakes. Concentrations of PBDEs were traced from its entry into the ecosystem in 1980 until

1999, and trends showed PBDE concentrations increasing exponentially at every site with doubling times estimated at 1.58 to 2.94 years.

4. “Dramatic Changes in the Temporal Trends of Polybrominated Diphenyl Ethers (PBDEs) in Herring Gull Eggs from the Laurentian Great Lakes: 1982-2006.” L.T. Gauthier, C.E. Herbert, D.V. Chip Weseloh, and R.J. Letcher. Environmental Science & Technology. 2008; 42(5); 1524-1530. PBDEs\Gauthier et.al. 2008.pdf

Summary: This study measured 43 PBDEs, specifically decaPBDE (209) in Herring Gull eggs from all five Great Lakes. The total PBDE concentration for lower Brominated congeners in the gulls was in the same range as the concentrations for octa and nona PBDEs, indicating that the higher Brominated congeners are accumulating at higher rates than lower brominated ones. PBDE-209 and its products showed increases in concentration over time. Lower Brominated congeners showed rapid increase in the eggs until 2000. Over all trends in pentaPBDEs show a decrease in Lakes Superior, Michigan, and Ontario from 2000-2006, while measurements at Lake Erie showed an increase. Lake Huron had two sites, one showed an overall increase of pentaPBDEs, and the other showed an overall decrease in the concentrations over time. Measurements of PBDE 207 and 197 were used as indicators of metabolic debromination from parent birds to the eggs.

5. “Spatial distribution and temporal trends in PBBs and PBDEs in lake trout from the Great Lakes.” J. M. Luross, C. M. Cannon, M.Alaee, D. B. Sergeant, D. M. Whittle and K. R. Solomon. International Association for Great Lakes Research 44th Conference, Green Bay, WI. 2001.

6. “Comparison of polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in Lake Michigan salmonids.” J. B. Manchester-Neesvig, K. Valters and W. C. Sonzogni. Environmental Science & Technology. 2001; 35(6): 1072-1077. PBDEs \Manchester-Neesvig et. al. 2001.pdf

Summary: PCBs and PBDEs were measured in 21 coho and Chinook salmon taken from Lake Michigan tributaries in 1996 for this study. The study found PBDEs to be found in fish at one-third the concentration of PCBs, but still at the highest reported level for fish worldwide. Implications are that PBDEs are as prevalent in the Lake Michigan water column as PCBs.

7. “Brominated flame retardants in the Great Lakes” Persistent Organic Pollutants in the Great Lakes, Handbook in Environmental Chemistry, Vol. 5, Part N, pp. 328-366, Springer-Verlag: Berlin & Heidelberg, Germany (2006), R. A. Hites, editor.

8. “Identification of brominated carbazoles in sediment cores from Lake Michigan.” L. Y. Zhu and R.A. Hites. Environmental Science & Technology. 2005; 39, 9446-9451. Brominated carbazoles\Zhu 2005.pdf

Summary: This study measured a possible naturally occurring brominated organic a brominated carbazole. The time trend determined by measuring these compounds as a function of depth in the Lake Michigan sediment cores indicated that the concentration of 1,3,6,8-tetrabromocarbazole peaked around 1920-1935. This was the first time that bromocarbazoles have been found in the environment.

9. "Deposition versus Photochemical Removal of PBDEs from Lake Superior Air." J.D. Raff and R.A. Hites. Environmental Science & Technology. 2007; 41: 6725-6731. PBDEs\Raff & Hites 2007.pdf

Summary: This study measured PDBEs from sediment on a remote Lake Superior island as well PBDEs in Lake Superior air. The study show decrease in PBDEs concentrations from air to sediment, except PBDE 209. Study concludes that photolysis is an important process for PBDE degradation in air and could be one reason that PBDE congener profiles in sediment are highly variable.

10. "Brominated flame retardants in sediment cores from Lakes Michigan and Erie" L. Y. Zhu and R.A. Hites. Environmental Science & Technology. 2005; 39, 3488-3494; erratum 39, 5904 (2005). PBDEs\Zhu &
Hites 2005 Lake Erie and Michigan.pdf

Summary: Dated sediment cores were studied to see trends in PBDEs and PBBs (Polybrominated Biphenyls). The study showed higher concentrations in Lake Michigan than Lake Erie, and Lake Superior. The study also showed rapid increase in both PBDEs and PBBs in sediments, concurrent with market increase in demand for these flame retardants. The total burdens of these compounds in the sediment of Lakes Michigan and Erie were 110 and 10 metric tons, respectively. The estimated total burden of these compounds in all of the Great Lakes approximately 200 tons.

11. "Temporal trends and spatial distributions of brominated flame retardants in archived fishes from the Great Lakes." L. Y. Zhu and R.A. Hites. Environmental Science & Technology. 2004; 38: 2779-2784.
PBDEs\Zhu & Hites 2004.pdf

Summary: PBDEs and PBBs were measured in fish from all five Great Lakes. Spatial trends showed Lake Michigan and Ontario had the highest total concentrations of both PBDEs. Concentrations in all fish increased exponentially over time with a calculated doubling rate of 3-4 years. Concentrations of PBBs, banned in the 1970's, remained at a constant with only a slight decrease in fish from Lake Huron.

12. "Polybrominated diphenyl ethers in the sediments of the Great Lakes. 3. Lakes Ontario and Erie." W. Song, J.C. Ford, A. Li, N.C. Sturchio, K.J. Rockne, D.R .Buckley, and W.J. Mills. Environmental Science & Technology. 2005; 39,(15):5600-5. PBDEs\Song et. al. 2005 Lakes Ontario and Erie.pdf,

Summary: Sediment cores from both Lake Ontario and Erie were analyzed for PBDE and PCB concentrations Lake Ontario. Results showed a dramatic increase in PDBE concentrations, while PCB concentrations peaked and then showed slow decrease. Lake Erie sediments showed no temporal trends due to high rates of sediment mixing. Overall congener profiles of PBDEs showed that PBDE-209 is the highest single congener concentration making up 90-95% of total PBDE concentration.

13. "Polybrominated diphenyl ethers in the sediments of the Great Lakes. 2. Lakes Michigan and Huron." W. Song, A . Li, J.C. Ford, N.C. Sturchio, K.J. Rockne, D.R .Buckley, and W.J. Mills.. Environmental Science & Technology. 2005; 39, (10):3474-9. PBDEs\Song et.al. 2005 Lakes Michigan & Huron.pdf

Summary: PBDEs and PCBs were measured in sediments from both Lake Huron and Lake Michigan. PBDE concentrations increase over time at all sampling sites. Spatial distribution and PBDE congener profiles implied that both lakes concentrations appeared to be dependent upon latitude and the sites proximity to populated areas. It was suggested that north-bound air plumes from urban areas are the major sources of PBDEs found in the lake sediments at locations away from the shores.

14. "Polybrominated diphenyl ethers in the sediments of the Great Lakes. 1. Lake Superior." W. Song, J.C. Ford, A. Li, W.J. Mills, K.J. Rockne, and D.R .Buckley. Environmental Science & Technology,. 2004; 38(12):3286-93. PBDEs\Song et. al. 2004 Lake Superior.pdf

Summary: Measurements of PBDE and PCB concentrations in Lake Superior showed that PCBs concentrations in sediments are slowly declining, while PBDEs are increasing with time. The load of total PBDEs to Lake Superior was estimated to be 2-6 metric tons, and the calculated loading rate was 80-160 kg per year.

15. "Polybrominated diphenyl ethers in the sediments of the Great Lakes. 4. Influencing factors, trends, and implications." A. Li, W. Song, J.C. Ford, N.C. Sturchio, K.J. Rockne, D.R .Buckley, and W.J. Mills. Environmental Science & Technology. 2006 Dec 15; 40 (24):7528-34. PBDEs\Li et. al. 2006.pdf

Summary: Sediment cores from all five Great Lakes were analyzed for PBDEs. Spatial trends indicated that major individual PBDE congeners showed strong dependence on the latitude of sampling sites. The study suggested that data reflected both the influence of urbanization, which showed south-to-north gradient in the region, and the general direction of long-range transport of airborne pollutants in the northern hemisphere. Doubling times of PBDEs in sediment showed a longer time than reported doubling times for humans or fish, indicating a slower response in sediments to emissions of PBDEs. Regressions applied to the data showed that latitude, year of deposition, and organic matter were all independent variables to PBDE concentration.

16. "Partitioning and Bioaccumulation of PBDEs and PCBs in Lake Michigan". S. Streets, S. Henderson, A. Stoner, D. Carlson, M. Simcik, and D. Swackhamer. Environmental Science & Technology. 2006; 40: 7263-7269. PBDEs\Streets et al 2006.pdf

Summary: Water samples from Lake Michigan were analyzed for PBDEs and PCBs. Concentrations of PBDEs were similar to PCBs in the water column, and data suggested that partitioning for PBDEs between particulates and dissolved phase behaves similar to PCBs partitioning. Fish from all the Great Lakes were also analyzed for PCBs and PBDEs, specifically

BDE-99. PCBs and PBDEs did not follow the same trend of accumulation in fish, and this paper suggested that biotransformation of BDE-99 are occurring.

17. "Polybrominated Diphenyl Ethers and Hydroxylated and Methoxylated Brominated and Chlorinated Analogues in the Plasma of Fish from the Detroit River." K. Valters, H. Li, M. Alaee, I. D'Sa, G. Marsh, A. Bergman, and R.J. Letcher. Environmental Science & Technology. 2005; 39: 5612-5619. PBDEs\Valters et.al 2005.pdf

Summary: This study measured concentrations of PBDEs and their metabolites-hydroxylated PBDEs (OH-PBDEs), methoxylated-PBDEs (MeO-PBDEs), as well as the antimicrobial triclosan and its metabolites-OH-trichlorodiphenyl ether, and methylated (MeO) triclosan in fish from the Detroit River. The study found that the sum of PBDE to sum of OH-PBDE concentration ratios ranging from 0.0005 to 0.02. OH-PBDEs are likely derived in these freshwater species as metabolites of precursor PBDEs and are subsequently retained in the blood, although they could have external origins and be picked up and accumulated by fish. Anthropogenic triclosan concentrations ranged from 750 to >10,000 pg/g wet weight and was clearly a bioaccumulative halogenated phenolic compound in these fish. MeO-triclosan concentrations were considerably lower.

18. "Spatial distribution of polybrominated diphenyl ethers and polybrominated biphenyls in lake trout from the Laurentian Great Lakes." J. M. Luross, M. Alaee, D. B. Sergeant, C. M. Cannon, D. D.M.Whittle, K. R. Solomon and D. C. G. Muir. Chemosphere. 2002; 46, (5):665-672. PBDEs\Luross et.al. 2002.pdf

Summary: This study measured PBDE and PBB concentrations in trout from all five Great Lakes. Concentrations were highest at Lake Ontario and lowest at Erie for PBDEs. Lake Huron had the highest concentrations of PBBs, with the lowest concentrations being at Lake Superior. This study also found that the three most prominent PBDEs (47, 99,100) in fish are all components of the commonly used penta-BDE found in flame retardants.

Note: Appendix A compiled by Carolyn Peerson, University of Iowa, for International Joint Commission Chemicals of Emerging Concern Workgroup, July, 2008.

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