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An overview of policies for managing polybrominated diphenyl ethers (PBDEs) in the Great Lakes basin

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ABSTRACT

The Great Lakes are an important environmental and economic resource for Canada and the United States. The ecological integrity of the Great Lakes, however, is becoming increasingly threatened by a number of persistent, bio-accumulative and harmful chemicals that enter the Great Lakes ecosystem through fluvial and atmospheric deposition. Polybrominated diphenyl ethers (PBDEs), a class of brominated flame retardant, are among such chemicals, whose concentration in the Great Lakes has greatly increased in recent years. Despite growing concern over the possible health and environmental effects of these compounds, only four of the eight Great Lakes states have enacted regulations to ban/restrict the use of PBDE while the two Canadian Great Lakes provinces are yet to endorse any regulation. Of the three main commercial PBDE mixtures (pentaBDE, octaBDE and decaBDE), penta- and octaBDE are no longer manufactured or imported into the United States and Canada. DecaBDE, however, still finds use in a variety of products.

In the present paper, the authors review the current regulations and policies for managing PBDEs in the Great Lakes jurisdictions and briefly review commercially available non-bromine chemical alternatives to PBDE. As these alternatives are comparatively more expensive than PBDE, future adoption of more eco-friendly flame retardants by the polymer industry will likely depend on stricter legislation regulating the use of PBDE and/or an increased public demand for PBDE-free products.

1. Introduction

The Great Lakes, comprised of Lake Superior, Lake Huron, Lake Michigan, Lake Erie and Lake Ontario (Fig. 1), have a total volume of 23,000 km^3 and a drainage area encompassing 750,000 km^2. Together, these lakes form the largest fresh water surface system on earth, retaining approximately 80% of the total surface water of North America and more than 20% of the world's total freshwater supply (EC and EPA, 1995). The Great Lakes are thus an important environmental and economic resource for Canada and the United States. The basin provides drinking water for millions of people and supports numerous species of terrestrial and aquatic wildlife, including more than 130 globally rare, endangered and at-risk species (USFWS, 2005; COSEWIC, 2007). Economically, the Great Lakes basin sustains more than 50% of Canada's manufacturing output and generates over $330 billion USD annually in Canada–US trade (EC and EPA, 1995; Fields, 2005).

The immediate vicinity surrounding the lakes, however, is one of the most densely populated and highly industrialized areas in North America. Approximately 40 million people live within the boundaries of the Great Lakes basin, accounting for 30% of the total Canadian population and 10% of the US population (EC and EPA, 2003). Associated high levels of urban, agricultural and industrial run-off of toxic contaminants, in addition to atmospheric deposition and fluvial migration of chemicals, are threatening the ecological and economic health of the ecosystem and millions of people living in the Basin. Over 1000 different chemicals have been identified in the Great Lakes, more than 350 of which exist in considerable quantities (EC, 1997; IJC, 2003a). Levels of a few chemicals, such as PCBs, have declined in recent years as a direct result of legislative changes (DeVault et al., 1996; Hickey et al., 2006). However, concerns with respect to the increasing environmental presence of a group of related compounds, Polybrominated diphenyl ethers (PBDE), have recently been raised (Renner, 2000; Darnerud et al., 2001; Rahman et al., 2001; Hites 2004).

PBDEs are a class of brominated flame retardant known to be environmentally persistent and lipophilic that bioaccumulate in animal tissue and biomagnify with increasing trophic distance (De Wit, 2002; Law et al., 2006; Burreau et al., 2006;Voorspoels et al., 2007). Observations that PBDEs have increased exponentially in the Great Lakes in recent years, combined with data indicating adverse effects on human health, suggest that the formulation and implementation of effective policies to rid the Great Lakes basin of these chemicals is urgently needed. In the present paper, the authors have reviewed the current regulations and policies concerning PBDE in the Great Lakes jurisdictions. The efficacy of these policies has been analyzed and prospective legislative and research priorities are

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Fig. 1. The Great Lakes basin.

suggested. In addition, occurrence, geographic and temporal distribution, toxicity, and environmental effects of PBDE are briefly described.

2. Polybrominated diphenyl ethers (PBDE)

Brominated flame retardants (BFRs) are chemical compounds that inhibit the combustion of organic materials by scavenging free radicals that would otherwise encourage the spread of flames (D'Silva et al., 2004; Hites, 2006). These compounds are found in a wide variety of materials including paints, plastics, textiles, furniture and electronics and may be either covalently bonded to the polymer, or additively mixed into the final product. Mounting evidence, however, suggests that the non-reactive BFRs can easily leach into the environment and pose significant environmental and health concerns (Hites, 2006). Despite this, brominated flame retardants are routinely included in the manufacture of household goods and an increasing consumer demand for such products has been reflected in the global BFR production patterns over the past several years (Alaee et al., 2003).

The use of PBDE gained prominence after the manufacture of polybrominated biphenyls (PBB) was stopped in the United States and Canada in the late seventies (Hites, 2006). PBDEs are a class of additive brominated flame retardants made up of 209 possible congeners containing between 1 and 10 bromine atoms (Alaee et al., 2003). Of these, 23 congeners are of environmental significance, the details of which are given in a review by D'Silva et al. (2004). Until early 2005, three commercial PBDE mixtures were widely distributed for use in North America: Penta-bromodiphenyl ether (pentaBDE), Octa-bromodiphenyl ether (octaBDE) and Deca-bromodiphenyl ether (decaBDE) (Table 1). Worldwide, more than 70,000 metric tons of PBDE have been produced annually, half of which have been used in the US and Canada, including almost all of the pentaBDE manufactured (Renner, 2000; Hites 2006).

Globally, high levels of PBDE have been detected in a variety of environmental media including soil and sediment (Song et al., 2005a, b; Zhu and Hites, 2005), air (Wilford et al., 2004; Hoh and Hites, 2005), house dust (Jones-Otazo et al., 2005; Stapleton et al., 2005) and biological samples (Alaee et al., 1999; Ikonomou et al., 2000; Zhu and Hites, 2006). In a recent meta-analysis of global concentration data, Hites (2004) determined that human PBDE levels had increased by a factor of 100 over the past three decades, with concentration loads doubling every 5 years. North Americans have the highest global body burdens, averaging contaminant levels 20 times that of Europeans.

The potential health hazards of PBDEs are significant. Pregnant women and developing fetuses may be particularly susceptible to their effects (reviewed in McDonald, 2002). DecaBDE has been designated as a possible human carcinogen by EPA on the basis of evidence of cancer for animals (ATSDR, 2004). Animal studies have shown that PBDEs delay hatching, depress rates of swimming and feeding in fish (Timme-Laragy et al., 2006) and induce neural defects and cardiac arrhythmia (Lema et al., 2007). Laboratory experiments using rodents found that neo-natal exposure to PBDEs could impair motor skills, learning and memory (Eriksson et al., 2002; Branchi et al., 2003; Viberg et al., 2007), induce immuno-toxicity (Thuvander and Darnerud, 1999; Martin et al., 2007), disrupt endocrine functioning (Hallgren et al., 2001; Hallgren and Darnerud, 2002) and impair reproductive development (Lichtensteiger et al., 2003; Stoker et al., 2004; Kuriyama et al., 2005). Comparable studies investigating the toxicological effects of PBDEs on humans are extremely few, but recent evidence suggesting that PBDEs have an adverse effect on human health and reproduction has emerged. In particular, the developmental aberrations found in the animal studies discussed above indicate that children may be especially at risk of adverse health effects due to PBDE exposure (Eriksson et al., 2001a; Siddiqi et al., 2003). Transfer of PBDE contamination from maternal to fetal tissue during gestation has been documented (Mazdai et al., 2003; Schecter et al., 2007) and young children may be exposed to additional early PBDE contamination through nursing and house dust (Zuurbier et al., 2006).

The consumption of contaminated food is thought to be a major pathway to human exposure (Watanabe and Sakai, 2003; D'Silva et al., 2004). PBDEs have been detected in a wide variety of meat, fish and dairy products in both North America and Europe (Bocio et al., 2003; Schecter et al., 2004, 2006) and the application of PBDE-laden biosolids to agricultural areas is a potential vector to food contamination (Hale et al., 2001; Sjodin et al., 2003). Correlational 'market basket'

Table 1
Chemical summary and market demand of three major commercial polybrominated diphenyl ether products widely used in North America prior to 2005

Commercial product	Predominant congeners	Applications[a]	Global market demand[b] MT	North American market demand (% global demand)[b] MT
Penta-bromodiphenyl ether	2,2'4,4' — tetrabromodiphenyl ether (BDE-47) 2,2',4,4',5 — pentabromodiphenyl ether (BDE-99) 2,2',4,4',6 — pentabromodiphenyl ether (BDE-100)	Polyurethane foam, textiles, furniture, insulation	7500	7100 (95%)
Octa- bromodiphenyl ether	2,2',3,4,4'5',6 — heptabromodiphenyl ether (BDE-183) 2,2',3,3'4,4'5,6' — octabromodiphenyl ether (BDE-196) 2,2',3,3'4,4',6,6' — octabromodiphenyl ether (BDE-197) 2,2',3,3'4,4'5,6,6' — nonabromodiphenyl ether (BDE-207)	Computers, automobile trim, telephones, household appliances, fax machines	3790	1500 (40%)
Deca-bromodiphenyl ether	2,2',3,3'4,4'5,5',6,6' — decabromodiphenyl ether (BDE-209)	Carpet pads, draperies, television sets, electronic casings, cable insulation, adhesives, textile coating	56100	24500 (44%)

[a] Alaee et al. (2003); Oregon Department of Human Services Environmental Toxicology Program (www.oregon.gov/DHS/ph/envtox/pbde.shtml#why).
[b] Hites (2006); Bromine Science and Environmental Forum (www.bsef.com).

studies between PBDE levels in food and consumers, however, indicate that the total loads recorded in North Americans are likely due to several simultaneous sources of exposure (Schecter et al., 2006; see also De Wit, 2002), rather than a solely nutritional route. Exposure from contact with in-use products containing PBDEs (WHO, 1994), volatization of PBDEs from incinerated and landfill waste (Darnerud et al., 2001), PBDE release from wastewater treatment plants (La Guardia et al., 2007) and inhalation of contaminated house dust are additional sources of exposure. Ambient concentrations of bioavailable PBDE are much higher in urban areas than rural and remote areas (Strandberg et al., 2001; Hoh and Hites, 2005), and a positive relationship exists between ambient levels of PBDEs in the home and in the personal air of inhabitants (Allen et al., 2007). Furthermore, a strong correlation between PBDE levels in house dust, and those in breast milk (Wu et al., 2007) and blood plasma (Karlsson et al., 2007) have been reported.

2.1. Polybrominated diphenyl ethers in the Great Lakes

Atmospheric deposition is the major source of PBDE to many aquatic systems (Hale, et al., 2003; Wang et al., 2005) including the Great Lakes (Li et al., 2006). In addition to atmospheric deposition, PBDEs enter the Great Lakes through recycling and/or disposal of wastes containing PBDE (Watanabe and Sakai, 2003), manufacturing output and fluvial deposition from tributaries (Samara et al., 2006) and wastewater treatment plants (La Guardia et al., 2007).

Dated sediment cores suggest that approximately 100 metric tons of PBDE are contained in the Great Lakes sediments (Song et al., 2004, 2005a, 2005b; Zhu and Hites, 2005; Li et al., 2006), most of which matches the congener profile for deca-PBDE (BDE-209), a large, relatively immobile molecule that does not likely have the same long-range transportation capabilities as the lower brominated compounds (Gouin et al., 2006). In ambient air samples and biota however, the congener distribution usually reflects the profile of lower brominated penta-PDBE (Table 1), with the most abundant congeners typically being BDE-47, -99, -100, -153 and -154 (Strandberg et al., 2001; Hites, 2004).

PBDEs have been detected in numerous species of fish in the Great Lakes and adjacent rivers (Alaee et al., 1999; Luross et al., 2000, 2002; Manchester-Neesviget et al., 2001; Dodder et al., 2002; Stapleton and Baker, 2003a; Zhu and Hites, 2004), and recent total concentration loads from 1000–3000 ng/g lipid weight have been recorded for slow growing, long-lived predators such as salmon and trout (Hites, 2004) that are favorite catches with anglers and recreational fishers (DFO, 2000). Currently, environmental levels of PBDE in fish are considered to be safe for human consumption (Darnerud et al., 2001; HC, 2006). However, dietary uptake studies conducted on pike (*Esox lucius*) have shown that the uptake efficiency of the lower brominated PBDE congeners from the gastrointestinal tract can be as high as 90% in fish (Burreau et al., 1997) and PBDEs accumulate in animal lipids and muscle (Manchester-Neevig et al., 2001; Voorspoels et al., 2007).

Long-term studies of birds and fish in the Great Lakes have indicated that over the past 20 years PBDE burdens in wildlife have increased exponentially (Norstrom et al., 2002). In general, contaminant levels over this period have doubled approximately every three years, although recent trends indicate that burdens may now be leveling off in some of the lakes (Luross et al., 2000; Zhu and Hites, 2004). But, if the temporal trajectory of PBDE accumulation in the Great Lakes is maintained, levels of PBDEs could soon exceed the upper limit of safe levels of consumption of contaminated fish (Darnerud et al., 2001). Indeed, current levels of PBDE have exceeded those of PCB in some environmental media (Hale et al., 2001a, 2003) and in human blood (Schecter et al., 2005). Furthermore, the data suggest that PBDE may be more easily metabolized than PCB (Stapleton and Baker 2003a). Areas of the Great Lakes adjacent to urbanized and industrialized areas have higher levels of contaminants, and consequently, these areas may be more vulnerable. In 2000, for example, the average concentration of total PBDE recorded in trout from each of the Great Lakes spanned a difference of 1030 ng/g lipid weight, with almost a four fold difference in contaminant levels between Lake Huron and Lake Michigan [Lake Michigan (1400 ng/g lipid), Lake Superior (990 ng/g lipid), Lake Erie (600 ng/g lipid), Lake Ontario (550 ng/g lipid), Lake Huron (370 ng/g lipid)] (Hites, 2006). Thus, focusing future investigative efforts on the most highly contaminated lakes may permit more accurate evaluation of the extent and effects of contamination.

3. PBDE abatement policies

Due to public concerns and economic reasons, the only North American producer of octaBDE and pentaBDE mixtures, Great Lakes Chemical Corporation (now Chemtura) voluntarily stopped producing these chemicals in December 2004 (Tullo, 2003; Hites, 2006). Since then, the use of penta- and octa-BDE products has been effectively eliminated in North America with all affected companies reporting complete phase-out by 2005 (Government of Canada, 2006). While penta- and octaBDE are no longer manufactured or imported into North America, they are present in much of the furniture, mattresses, televisions and office equipment currently in use and remain a significant risk for PBDE exposure. These compounds will continue to enter the environment for many years to come through the disposal of PBDE containing electronics and furniture (Hites, 2006).

Stricter legislation regarding the fate of PBDE containing products is needed. Most PBDE containing products are incinerated or enter landfill facilities, and the small percentage that is recycled is often done so with little concern for human health. The *International Association of Electronics Recyclers* estimates that an average of

400 million consumer electronics is discarded each year (IAER, 2006). Public concerns regarding the enormous amount of e-waste generated in North America have resulted in nine states passing bills regulating e-waste since 2004 and a total of 23 new e-waste bills were introduced in the 2007 legislative session (Computer Take-Back Campaign, 2007). Most of these bills place the burden of responsibility on the industry producer; in Minnesota for example, recent legislation has mandated that electronics manufacturers must collect and recycle 60–80% of the estimated weight of their yearly product sales. Unfortunately, it is estimated that 50–80% of recycled electronics are sent to developing nations for dismantling, where poor labour standards place workers at significant risk of toxic exposure from PBDEs and other compounds (Computer Take-Back Campaign, 2007).

3.1. Canadian policy on the use, sale and import of PBDE

No prohibitive regulations or management policies for PBDE currently exist in Canada at either a federal or provincial level, although regulations on the sale, use and import of PBDEs have recently been proposed. In February 2004 and June 2006, Health Canada and Environment Canada respectively completed screening assessments of the risks of PBDEs to the environment and to human health (HC, 2004; EC, 2006a). These assessments were based on current data, reviews, and comments by industry experts and published reports of their final assessments were released on July 1, 2006. The screening assessments evaluated seven classes of PBDEs (tetra-decaBDE), all of which were determined to be persistent, and to be entering the environment in quantities that could have harmful effects on the environment and/or biological diversity. In addition, tetraBDE, pentaBDE and hexaBDE were found to be bioaccumulative, as defined by the *Persistence and Bioaccumulation Regulations* under the *Canadian Environmental Protection Act (CEPA) 1999,* and both sectors recommended that PBDE be added to the *List of Toxic Substances*. Environment Canada's assessment concluded that wildlife poisoning through the consumption of contaminated prey formed the greatest threat to the environment (EC, 2006a). Similarly, Health Canada determined that current levels of exposure did not pose an immediate human health risk, since the exposure levels of the most highly vulnerable group — that of breast-feeding infants — are currently below the exposure limit for the PBDE critical effect level of 0.8 mg/kg-bw (based on laboratory studies investigating neuro-developmental effects on rodents) (HC, 2004). They cautioned, however, that these assessments did not take into consideration exponentially increasing levels of PBDEs measured in the bodies of Canadians. At current levels, it is suggested that environmental regulations aimed at abating the environmental emission of PBDEs and the uptake into biota will, in consequence, effectively reduce human loads and protect human health (HC, 2004). Following the screening assessments, Environment Canada issued a '*Proposed Risk Management Strategy for PBDEs*' in September 2006. The primary environmental objective outlined in the document was to reduce the environmental concentrations of all seven PBDEs to the lowest level possible by preventing the manufacture of all PBDE classes, prohibiting the use and import of tetraBDE, pentaBDE and hexaBDEs, and reducing the emission of PBDEs into the environment (EC, 2006b).

Based on the screening assessment and recommendations, the Government of Canada added PBDE to the *List of Toxic Substances* on December 7, 2006, as defined under CEPA 1999. Following this, on December 16, 2006 the federal government proposed regulations on allowable classes and uses of PBDE (Government of Canada, 2006). The regulations propose the prohibition of manufacture of all seven PBDE classes, in addition to a complete ban on the sale, use or import of tetraBDE, pentaBDE and hexaBDE in polymers and resins, which would effectively eliminate the use of pentaBDE and octaBDE commercial mixtures. The regulations do not apply to imported finished manufactured articles containing these products, for which additional management strategies would be drawn.

In their current form, the proposed Canadian federal regulations have been criticized for having little practical economic or environmental impact (Boyd and Wallace, 2006). PentaBDE and OctaBDE were phased out of manufacture in the USA in 2005, and PBDEs have never been manufactured in Canada. No restrictions on the only class of PBDEs currently in wide use (decaBDE) have been proposed, although the *Proposed Risk Management Strategy for PBDEs* outlines the development of approaches to minimize decaBDE emissions, in consultation with various stakeholders (EC, 2006b). The approaches appear primarily voluntary and have not, as of yet, been implemented (BSEF, 2007). Several environmental organizations have objected to the proposed regulations and have recommended that the decaBDE commercial mixture, mainly composed of the congener BDE-209, is added to the list of PBDEs slated for virtual elimination (SLDF, 2007).

DecaBDE is the most widely used commercial additive in the polymer industry (Hardy, 2002) and has garnered much recent debate regarding the environmental and health risks of the higher-brominated congeners. While acknowledged to be persistent in the environment, until recently the BDE-209 molecule had been considered too large to bioaccumulate, and thus not a risk to human health or that of wildlife (NRC, 2000; Zhou et al., 2001; Hardy, 2002). Studies published subsequent to the ecological screening assessment have, however, demonstrated that BDE-209 does accumulate in animal tissues (Lindberg et al., 2004; Verreault et al., 2005; Voorspoels et al., 2006; Chen et al., 2007) and has been detected in the blood serum and milk of people (Sjodin et al., 1999, 2001; Jakobsson et al., 2002; Schecter et al., 2003; Thuressen et al., 2005). Furthermore, evidence indicates that decaBDE debrominates in response to organic solvents (Eriksson et al., 2001b; Hua et al., 2003), certain reducing agents (Keum and Li, 2005) and photodegradation (Jafvert and Hua, 2001; Soderstrom et al., 2004). The compound is also metabolically converted to lower brominated congeners in animals (Stapleton and Baker, 2003b; Kierkegaard et al., 2007). Thus, it appears that decaBDE can transform into the more bio-accumulative and toxic lower brominated congeners, and does so readily in nature (La Guardia et al., 2007; but see EC, 2006b). In light of this current research, a legal objection has been filed by several organizations under the *Canadian Environmental Protection Act*, requesting a review of the federal regulations. A governmental response to this objection has not yet been released (SLDF, 2006).

3.2. United States policy on the use, sale and import of PBDEs

The *Toxic Substances Control Act* (TSCA) of 1976 allows the United States Environmental Protection Agency to control the use and import of chemical substances that may pose a risk to environmental or human health. In December 2004, the EPA proposed a federal *Significant New Use Rule* (SNUR) under the Toxic Substances Control Act that required persons or businesses to notify the EPA 90 days prior to the intent to manufacture, use or import pentaBDE or octaBDE products (EPA, 2005a). The SNUR, which came into effect in August 2006, allows the EPA to review proposals for new products containing these compounds and regulate them accordingly.

In March 2006, the EPA released the *Polybrominated diphenyl ethers (PBDEs) project plan*, which outlined EPA's major initiatives concerning PBDEs (EPA, 2006). Several broad objectives were identified, including the identification of PBDE substitutes and developments regarding alternative BFRs, the risk assessment of pentaBDE, octaBDE and decaBDE and the evaluation of decaBDE for continued use. As mentioned in Section 3 above, the only North American producer of pentaBDE and octaBDE (Great Lakes Chemical Corporation) ceased production of these compounds in 2004. DecaBDE, however, is manufactured by several companies in the United States and industry officials strongly object to claims that decaBDE should be listed as a toxic substance and should therefore be restricted or banned (Hardy, 2002; BSEF, 2007). Currently, the EPA is conducting an interim review of all available scientific information concerning decaBDE. Following its completion, EPA will determine whether decaBDE should be subjected to additional assessment and regulation. In addition, EPA has conducted toxicological reviews of BDE-47, BDE-99, BDE-153, and BDE-209 for inclusion in the EPA *Integrated Risk Information System* (IRIS), which consolidates data pertaining to the health effects of exposure to various substances found in the environment. External review drafts of the toxicological reviews were released for public comment in December 2006 on the EPA website. According to a project status report released in March 2007, EPA is currently continuing to review and assess information pertaining to toxicity and environmental exposure of all classes of PBDEs.

Aside from the 2006 SNUR, no US federal regulations pertaining to future restriction or prohibition of any PBDE category have been proposed. However, a number of Great Lakes states have passed bills banning the use or manufacture of products containing pentaBDE and/or octaBDE commercial mixtures (Table 2). Of the eight states bordering the Great Lakes, four have enacted or proposed regulations on the use and distribution of PBDEs. Effective 2006, the use of pentaBDE and octaBDE commercial products was banned in Minnesota, Michigan, Illinois (octaBDE ban effective 2008) and New York, and assessment and/or proposed regulations on the use of decaBDE have been undertaken in all of these states. In addition, several other states in USA have enacted restrictions on PBDE use. Of particular note, in 2003 California became the first state to propose a ban on penta- and octaBDE classes, and in recent bills scheduled to take effect January 1, 2008, Washington and Maine have introduced the first restrictions on the use of decaBDE in certain manufactured items (BSEF, 2007; NCEL, 2007). Because of the long-range transport capabilities of the lower brominated PBDEs however, regulations in all of the federal states may have a direct bearing on the influx of PBDEs in the Great Lakes.

3.3. United States and Canadian bi-national policy in the Great Lakes region

Beyond the prohibition of PBDEs in four of the Great Lakes states, there are no direct regulatory acts that specifically concern the occurrence of PBDEs in the Great Lakes. However, a variety of national and bi-national joint initiatives exist that regulate and manage the Great Lakes waters with respect to toxic contamination. Representatives from both the United States and Canada jointly signed *the Boundary Waters Treaty* in 1909. The purpose of the treaty was to enact a framework within which disputes concerning shared water resources could be mitigated. In 1972, the *Great Lakes Water Quality Agreement* (GLWQA) was enacted in association with the *Boundary Waters Treaty*, with the specific objective of protecting and restoring the Great Lakes. The agreement was subsequently amended in 1987 to include specific actions focused on identifying, monitoring and restoring specific Great Lakes Areas of Concern (AOC), developing lake management plans for each of the Great Lakes, and eliminating toxic pollutants from within the Great Lakes basin (IJC, 1978; amended 1987). With respect to the latter, Canada and the United States developed a framework to reduce or eliminate bio-accumulative persistent toxic substances from the Great Lakes Basin. In 1997, the *Canada–United States Strategy for the Virtual Elimination of Persistent Toxic Substances in the Great Lakes Basin* was signed (EC and EPA, 1997), which makes use of existing programs operating under a variety of legislative acts (Table 3).

Table 2
Summary of enacted and proposed PBDE state and federal legislation in the Great Lakes region

State/province	PentaBDE/OctaBDE	Year effective	DecaBDE
Illinois	Restricted/banned	2006/2008	Proposed ban
Indiana	No restrictions	n/a	No restrictions
Michigan	Banned	2006	Proposed ban
Minnesota	Banned	2006	Proposed ban
New York	Banned	2006	Under assessment
Ohio	No restrictions	n/a	No restrictions
Pennsylvania	No restrictions	n/a	No restrictions
Wisconsin	No restrictions	n/a	No restrictions
Ontario[a]	Proposed Ban	Unspecified	No restrictions
Quebec[a]	Proposed Ban	Unspecified	No restrictions

[a] Proposed regulations are federal, rather than provincial.

In 2003, the International Joint Commission (IJC), composed of three Canadian and three US commissioners that address and mediate issues of bi-nationally shared water under the *Boundary Waters Treaty,* requested a collaborative meeting of the scientific advisory board and associated bodies to identify and discuss emerging issues in the Great Lakes, including prevalent new chemicals (Boyer et al., 2006). General resulting recommendations of the IJC included a re-evaluation of the *Great Lakes Water Quality Agreement* (IJC, 2003b; 2006a) and a systematic increase in scientific information collection (IJC, 2003b). Expert consultants further recommended water quality criteria that included a broader range of chemicals, including PBDEs (Muir et al., 2006).

The IJC has stressed the need for accountability and an action-oriented restoration framework, so that existing recommendations, programs and policies may effectively protect and restore the Great Lakes (IJC, 2006b). In Canada, the *Canada–Ontario Agreement* (COA) (EC and OMOE, 2007) pledged support and protection of the Great Lakes Basin Ecosystem in fulfillment with Canada's requirements under the *Great Lakes Water Quality Agreement*. Under the *Harmful Pollutants* Annex II, Canada and Ontario proposed to eliminate all Tier 1 persistent bio-accumulative toxic compounds and to address and investigate how substances of emerging concern recently identified in the Great Lakes may be impacting human health or the environment. PBDEs are not specifically mentioned in the list of Tier 1 or Tier 2 chemical substances identified for priority assessment and action in any of the above documents pertaining to toxic pollutants in the Great Lakes Basin, although the recent addition of this group of compounds to Schedule 1 of the *List of Toxic Chemicals* warrants their consideration under these policies.

In the United States, the Council of Great Lakes Governors established nine priorities for restoring and maintaining the integrity and health of the Great Lakes (CGLG, 2003). These priorities formed the basis for the establishment of the Great Lakes Regional Collaboration, a partnership between the Great Lakes Interagency Task Force and the Great Lakes States, as well as local communities, tribes and non-governmental organizations (US Presidential Executive Order, 2004). Eight strategy teams were created to address the priorities of the Council of Governors, and in December 2005, the Great Lakes Regional Collaboration released an action plan for the protection and restoration of the Great Lakes (GLRC, 2005). Several of the priorities — non-point source pollution, persistent bio-accumulative toxins and coastal health — affect PBDE contamination of the Great Lakes. Wastewater treatment plants are a significant threat to coastal ecosystems and tri- through deca-BDEs have been identified in sludge from these facilities (Hale et al., 2001; La Guardia et al., 2007). In the action strategy report, brominated flame retardants are specifically identified as a non-point source contaminant of emerging concern and in 2007, the Great Lakes Regional Collaboration released the *Toxic Pollutants Initiative*, which launched an educational outreach program aimed at promoting safe disposal of electronic wastes in the Great Lakes states. Additionally, a variety of conservation programs under state authority that address general non-point sources of pollution exist that could potentially help manage PBDE influx into the Great Lakes (GLRC, 2005; Table 4).

3.4. International policy and initiatives

In August 2004, The European Union (EU) brought into effect legislation prohibiting the marketing and/or use of products

Table 3
Summary of Canadian and US legislative regulations affecting PBDE use and disposal in the Great Lakes Basin

Legislative act	Source	Year enacted	Description	Reference to PBDE
Canadianlegislation				
Environmental Protection Act	Federal	1999	Pollution prevention and control/elimination of toxic substances, including PBDE	Specific
Fisheries Act	Federal	1985	Regulation of the deposition of deleterious substances in waters	General
Environmental Assessment Act	Federal	1992	Regulation and monitoring of projects potentially causing significant environmental damage	General
Toxic Substances Management Policy	Federal	1995	Management of Track 1 and Track 2 substances under CEPA 1999, including PBDE	Specific
Safe Drinking Water Act	Federal	2002	Regulation, safety and monitoring of drinking water.	General
Ontario Environmental Protection Act	Provincial	1990	Regulation of contaminants, pollutants, waste management and disposal.	General
Ontario Water Resource Act	Provincial	1990	Water quality maintenance and regulation/prohibition of pollution discharge.	General
Ontario Environmental Assessment Act	Provincial	1990	General protection, conservation and management of the environment in Ontario	General
US legislation				
Comprehensive Environmental Response, Compensation and Liability Act	Federal	1980	Regulation and prohibition of hazardous waste sites and regulation of cleanup of hazardous waste.	General
Resource Conservation and Recovery Act	Federal	1976	Control of hazardous wastes and management of non-hazardous wastes.	General
Clean Water Act	Federal	2002 (amended)	Regulation of discharged pollutants into waters.	General
Clean Air Act	Federal	1990 (amended)	Regulation of hazardous air pollutants.	General
Toxic Substances Control Act	Federal	1976	Screening and regulation of chemicals causing environmental or human health hazards.	General
Pollution Prevention Act	Federal	1990	Reduction of pollutant production and use.	General
Safe Drinking Water Act	Federal	1974	Regulates and monitors the safety and quality of drinking water.	General

Source: Government of Ontario. 2007. Environmental Assessment Legislation.
Environmental Protection Agency. 2007. Major Environmental Laws.

containing more than 0.1% pentaBDE or octaBDE and the European Commission prohibited the use of these compounds in any electrical and electronic product under the *Restriction of Hazardous Substances* (RoHS) directive, effective 1 July 2006 (EU, 2003a). The *RoHS* is closely associated with the *Waste Electrical and Electronic Equipment Directive* (WEEE), which regulates the disposal of electrical goods (EU, 2003b). Together these two directives govern the use and disposal of PBDE containing products in European Union member states. As in North America, decaBDE (composed solely of the congener BDE-209) is currently exempt from regulation, following the release of a scientific assessment in 2005 (European Commission, 2005), however, the strict 0.1% weight-by-mass upper limit on banned octa- and penta-congeners means that most commercial decaBDE mixtures, which include these congeners in small quantities, are not permitted. The decaBDE exemption has been legally challenged, by the European Parliament and the Government of Denmark (BSEF, 2007). These bodies have recommended that decaBDE be banned completely. The legal objection, however, is currently awaiting further action.

Two international treaties on toxic pollutants have identified PBDEs as a global concern that warrants multinational cooperation. The *Stockholm Convention on Persistent Organic Pollutants* (POPs) is a global agreement that entered into force in 2004. The primary aim of the convention was to outline obligations for governments regarding the use, export, import and disposal of POPs, and to eliminate or reduce the environmental release and effects of harmful chemicals (EC, 2006c). In November 2005, the Persistent Organic Pollutants Review Committee under the United Nations (UN) Stockholm Convention determined that pentaBDE fulfilled the criteria of a persistent organic pollutant (UN, 2005) and also proposed the inclusion of octaBDE (UN, 2006). In addition, because of the known long-range transportation capabilities of pentaBDE, The United Nations Economic Commission for Europe (UNECE) *Long-Range Transboundary Air Pollution* (LRTAP) convention agreed that the pentaBDE commercial mixture should be considered as a persistent organic pollutant under the 1998 Aarhus Protocol and management strategies are currently being considered (EC, 2006b).

4. Alternatives to PBDE

Despite the availability of alternative flame retardants, deca-BDE remains the most cost-effective, and thus most frequently used, polymer industry additive. However, public concern regarding the health effects of PBDEs coupled with proposed restrictions and an uncertain future have prompted the development of several new flame retardants which have been adopted to various extents by the polymer industry. Environmental and health assessments of some of these alternatives are being undertaken by coalitions such as the *Furniture Flame Retardancy Partnership* (EPA, 2005b), a joint enterprise between several industry stakeholders and the Environmental Protection Agency (EPA) with an aim to find environment friendly alternatives to PBDE. For pentaBDE for example, potential alternative compounds currently under assessment for use in polyurethane foam include: (1) organic phosphate ester, (2) aryl phosphate, (3) tribromoneopentyl alcohol, (4) tris(1,3-dichloro-2-propyl) phosphate, (5) chloroalkyl phosphate, (6) reactive brominated flame retardants, (7) tetrabromophthalate diol diester, (8) halogenated aryl ester and (9) isopropylated triaryl phosphate (EPA, 2005b).

For electronics, decaBDE substitution strategies range from direct substitution of the resin system and flame retardant to a complete redesign of the product. Phosphate and phosphonate compounds and/

Table 4
Programs addressing non-point source pollution and water restoration under Great Lakes US state authority that affect PBDE deposition

Program/initiative	State authority
Illinois Great Lakes Protection Fund	Illinois Environmental Protection Agency
Clean Michigan Initiative	Michigan Department of Environmental Quality
Part 201 Programs	Michigan Department of Environmental Quality
Minnesota Great Lakes Protection Fund	Minnesota Pollution Control Agency
Great Lakes Charter Program	Ohio Department of Natural Resources
Lake Erie Project and Ohio Great Lakes Fund	Ohio Environmental Protection Agency
Pennsylvania Great Lakes Office	Pennsylvania Department of Environmental Protection
Shore Structure Permit Program	Ohio Department of Natural Resources
Great Lakes Harbors and Bay Restoration Fund	Wisconsin Department of Natural Resources
Lakes Charter Program	Ohio Department of Natural Resources
Great Lakes Salmon and Trout Stamp Program	Wisconsin Department of Natural Resources

Source: Great Lakes Regional Collaboration Action Plan. 2005. Appendix — non-point sources of pollution.

or a combination of polycarbonate resin with phosphate ester and bioplastics derived from corn and other agricultural substances have been identified for use as alternative flame retardants for electronics (LCSP, 2005). Alternatively, separation of high-voltage components that need greater ignition protection from low-voltage components, or a reduction in the voltage required for operation are product design modifications that could reduce the need for flame-retardant enclosure materials (LCSP, 2005). The inclusion of inherently flame resistant materials in textiles, such as aramids and melamines, can reduce the use of PBDEs in clothing and furniture. Fibers and polymers that are not inherently fire resistant can be made so by coating the fabric and exhausting flame retardants into fibers during the dying process, adding flame retardants into the molten plastic during the spinning process (e.g. organophosphorus added to viscose fibers), or mixing two or more different monomers, polymerizing them to form a more flame-retardant plastic fiber. Several non-brominated substances currently exist on the market, such as ammonium polyphosphate, magnesium hydroxide and melamine phosphate, and are often superior in performance to deca-BDE (LCSP, 2005).

Research on environment-friendly flame retardants has been conducted for more than a decade (Zaikov and Lomakin, 1996) and a variety of non-bromine chemical alternatives to PBDE are readily and commercially available. Environmentally friendly polymeric flame retardants may be broadly be classified as (1) Phosphorus containing flame retardants, (2) Silicon containing flame retardants, (3) Boron containing flame retardant, (4) Nitrogen containing flame retardants and (5) Organometallic and inorganic transition metal compounds (e.g. antimony polymer) (reviewed in Lu and Hamerton, 2002). Additionally, alternative manufacturing processes exist that render materials inherently less flammable (Lu and Hamerton, 2002; LCSP, 2005; Posner and Börås, 2005). Flame-retardant materials are intended, in addition to their ability not to ignite and spread fire, to act as a barrier for underlying layers of materials, so that the fire does not propagate through the whole substance. Swelling, or intumescent systems may be advantageously used as alternatives to other flame-retardant treatments for this purpose. The swelling system is based on the formation of expanded coal tar, which partly acts as an insulating barrier against heat but also as a smoke-fume trap (Posner and Börås, 2005). Intumescent nanocomposites exhibit superior mechanical and anti-flammatory properties, and a single treatment is sufficient to protect most materials. This system supports the development of multifunctional materials and its industrial applications are varied (Bourbigot and Duquesne, 2007).

5. Discussion and conclusion

IJC has repeatedly expressed the concern for increasing concentration of PBDEs in the Great Lakes environment, yet there is no mechanism for regular monitoring of these chemicals in the Basin. Public awareness of PBDEs in the basin is also relatively low compared to other chemicals such as PCBs and mercury. Upgrading the level of consumer awareness about the PBDE containing products is necessary and could be undertaken within existing programmes of the Great Lakes and St. Lawrence Cities Initiative, as well as various non-governmental organizations.

The United States and Canada have pledged to reduce the environmental concentrations of PBDEs by preventing their manufacture and prohibiting their import and use and reducing the emission of PBDEs into the environment. However, there is yet inadequate federal regulation in either country to this effect. In the USA, the EPA is currently continuing to review and assess information pertaining to toxicity and environmental exposure of all classes of PBDEs. In Canada, the approaches appear primarily voluntary. While penta- and octaBDE are no longer manufactured or imported into the United States and Canada, the continued use of long-lived PBDE containing appliances, and the disposal of obsolete electronics and furniture means that high levels of PBDEs will continue to enter the environment for many years to come. The ban of penta- and octaBDE and assessment and/or proposed regulations on the use of decaBDE have been undertaken only in the four states of Minnesota, Michigan, Illinois (octaBDE ban effective 2008) and New York. In addition to banning/restricting the use of PBDE containing products, stricter regulations for recycling and disposal of PBDE containing waste are necessary.

Research on environment-friendly flame retardants has been conducted for more than a decade and a variety of non-bromine chemical alternatives to PBDE are readily and commercially available. These alternative flame retardants are relatively more expensive compared to decaBDE, but alternatives to PBDE are available in the market. The widespread adoption of these alternatives to PBDE by the polymer industry, however, will likely depend on future Government regulatory action and/or an increase in public demand for PBDE- free products. Until then, several avenues for continued scientific research are warranted. First, additional studies investigating the potential toxicological hazards of decaBDE are needed, (Darnerud, 2003) in light of the current lack of regulation of this chemical in North America. Second, a quantitative assessment of the sources, the extent and the pathways of environmental and human exposure from PBDEs (including penta- and octa-BDE) in long-lived, in-use consumer products is essential. Third, more studies investigating long-range transport of PBDEs would be useful to ascertain the extent of aerial transportation from other regions to the Great Lakes basin (MacLeod and Mackay, 2004). Finally, continued research on potential alternatives to PBDEs, their environmental fate and toxicological effects, should be made a priority.

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