別添8

商業用ペンタブロモジフェニルエーテルの危険性の概要

分解性	蓄積性	人健康影響	動植物への影響
【生分解性】 ・（Tetra，Penta，HexaBDE）難分解性（BIOWIN） ・（PentaBDE） 分解せず（OECD TG 301BでCO$_2$発生なし） 【半減期】 ・大気中：11-19日（EPIWIN） ・水中： 150日（EPIWIN） ・土壌中：半減期150日（EPIWIN） ・好気性底質中： 600日（EPIWIN） ・1970年代初期にﾖｰﾛｯﾊﾟの海洋の底質に沈降したPentaBDE同属体が現在も相当量存在しており、底質中での残留性が高いことが示されている。	【ｵｸﾀﾉｰﾙ/水分配係数】 log KOW=6.5-7.4 【BAF（経鰓及び経口による生物濃縮係数）】 ｾﾞﾌﾞﾗｶﾞｲ：BAF=1.8 【BMF（経口的生物濃縮係数）】 ・ｳﾐﾊﾞﾄ/ﾆｼﾝ：BMF=17 ・ﾊｲｲﾛｱｻﾞﾗｼ/ﾆｼﾝ：BMF=4.3 ・ｻｹ/ﾆｼﾝ：BMF=3.8 ・動物ﾌﾟﾗﾝｸﾄﾝ/底生生物：BMF=7.1 ・ﾎｯｷｮｸﾀﾞﾗ/動物ﾌﾟﾗﾝｸﾄﾝ：BMF=0.04-3.4 ・ﾜﾓﾝｱｻﾞﾗｼ/ﾎｯｷｮｸﾀﾞﾗ：BMF=13.7 ・ﾎｯｷｮｸｸﾞﾏ/ﾜﾓﾝｱｻﾞﾗｼ：BMF=0.3-11 ・多数の調査から、上位捕食者において懸念される濃度のPentaBDEが存在することが示されている。北極圏では、ﾜｼｶﾓﾒ、ﾎｯｷｮｸｸﾞﾏ、ﾜﾓﾝｱｻﾞﾗｼ、ｼﾛｲﾙｶなどの上位捕食鳥類および哺乳類中から高ﾚﾍﾞﾙのPentaBDEが検出されている ・土壌又は底質中のPentaBDEは、容易に食物連鎖に取り込まれ、人など食物連鎖上位者の脂肪組織中に生物濃縮する。	【反復投与毒性】 ラット（90日）：NOEL 2mg/kg/day未満 主な毒性は、肝臓肥大等（DE71） 【生殖毒性】 ラット（妊娠♀単回）：0.06mg/kgで児に自発行動変化（多動性） 0.3mg/kgで児に精巣体積・精子数の低値 （BDE99） 【催奇形性】 ラット（妊娠6日単回）：0.3mg/kgでばく露の母動物（F1）2個体から得られたF2児で、外観・骨格異常 （BDE99） 【その他】 実験動物で甲状腺ホルモン系への影響	【慢性毒性】 ミジンコ *Daphnia magna* ：繁殖阻害が認められた。

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Report of the Persistent Organic Pollutants Review Committee on the work of its second meeting

Addendum

Risk profile on commercial pentabromodiphenyl ether

At its second meeting, the Persistent Organic Pollutants Review Committee adopted the risk profile on commercial pentabromodiphenyl ether, on the basis of the draft contained in document UNEP/POPS/POPRC.2/7. The text of the risk profile, as amended, is provided below. It has not been formally edited.

PENTABROMODIPHENYL ETHER

RISK PROFILE

Adopted by the Persistent Organic Pollutants Review Committee
at its second meeting

November 2006

CONTENTS

Executive Summary

A substantial range of studies on pentabromodiphenyl ether has been identified and the findings summarised in this risk profile. The new findings reported here support the conclusion reached by the Persistent Organic Pollutants Review Committee in 2005 that PentaBDE's properties fulfill the screening criteria in Annex D of the Stockholm Convention.

Commercial pentabromodiphenyl ether (C-PentaBDE) refers to mixtures of bromodiphenyl ether congeners in which the main components are 2,2', 4,4'- tetrabromodiphenyl ether (BDE-47 CAS No. 40088-47-9) and 2,2',4,4',5-pentabromodiphenyl ether (BDE-99 CAS No. 32534-81-9), which have the highest concentration by weight with respect to the other components of the mixture.

Commercial pentabromodiphenyl ether mixtures (C-PentaBDE) are used for flame retardant purposes as additives in consumer products. The commercial mixtures contain brominated diphenyl ether congeners with three to seven bromines in the molecule, but molecules with four and five bromines predominate. The proportion of the different polybromodiphenyl ether (PBDE) congeners in C-PentaBDE varies in different regions of the world.

PentaBDE is released into the environment during the manufacture of the commercial product, in the manufacture of products containing PentaBDE, during their use and after they have been discarded as waste. Even though production of C-PentaBDE is phased out or being phased out worldwide, different products containing it will still be in use in several years to come, resulting in continuing releases to the environment. The products will in the end of their lifetime become wastes with the potential of additional releases.

The main source in North America and Western Europe has been the C-PentaBDE incorporated in polyurethane foam, used in domestic and public furniture. This use is now mainly phased out. The information is too limited to draw conclusions on the importance of other uses, like textiles, electrical and electronic products, building materials, vehicles, trains and aeroplanes, packaging, drilling oil fluid and rubber products. While some representative examples are covered, detailed information on use is lacking for many regions of the world.

Major releases to air are emissions from products during use, through volatilization of PentaBDE and dust-borne PentaBDE. Emissions of PentaBDE can also occur from recycling and dismantling activities such as dismantling of vehicles, buildings and constructions. Emissions can occur from electronic waste recycling plants and shredder plants. Potentially toxic products such as brominated dibenzo-*p*-dioxins and furans might be generated during incineration of articles containing C-PentaBDE.

The releases of PentaBDE are to air, water and soil, but the major part ends up in soil. The distribution between the environmental compartments is: soil>>>water>air. Several studies using sediment cores show that PentaBDE is very persistent in marine sediments, still occurring after 30 years. In the main, PentaBDE in the environment is bound to particles; only a small amount is transported in its gaseous phase or diluted in water but such transport over long periods can be effective in distributing the PentaBDE widely in the environment, especially into Arctic regions. Occurrence in the Arctic environment is demonstrated in several monitoring studies in air and biota.

Due to its high persistency in air, the main route for long-range transport of PentaBDE - as with so many substances that are sufficiently volatile, persistent and bioaccumulative - is through the atmosphere. Modelling and environmental studies indicate that the transport is through a series of deposition/volatilization hops towards the poles but particulate transport is known to be important, too. Long-range transport through water and emigrating animals is also likely.

Several studies show that PentaBDE in soil and sediments is bioavailable, enters the food chain and that it bioaccumulates and biomagnifies in the food webs, ending up in high levels in top predators.

PentaBDE is widespread in the global environment. Levels of components of C-PentaBDE have been found in humans in all UN regions. Most trend analyses show a rapid increase in concentrations of PentaBDE in the environment and in humans from the early 1970s to the middle or end of the 1990s, reaching plateau levels in some regions in the late 1990s, but continuing to increase in others. The levels in North America and the Arctic are still rising. Vulnerable ecosystems and species are affected, among them several endangered species. Some individuals of endangered species show levels high enough to be of concern. Toxicological studies have demonstrated reproductive toxicity, neurodevelopmental toxicity and effects on thyroid hormones in aquatic organisms and in mammals. The potential for the toxic effects in wildlife, including mammals, is evident.

Potential exposure to humans is through food, and through use of products and contact with indoor air and dust. PentaBDE transfers from mothers to embryos and lactating infants. A Canadian assessment of risk quotients suggests that the highest risks accrue to species high in the food chain. Information is lacking on the effects in humans of short-term and long-term exposure, although it is to be expected that vulnerable groups can be pregnant women, embryos and infants. Considerably higher levels are found in humans from North America in general. About 5% of general populations have been found to be subjected to elevated exposure. This, together with the estimates of the long half-life of PentaBDE congeners in humans, raises concern for long-term effects on human health.

Based on the information in this risk profile, PentaBDE, due to the characteristics of its components, is likely, as a result of long-range environmental transport and demonstrated toxicity in a range of non-human species, to cause significant adverse effects on human health or the environment, such that global action is warranted.

1. Introduction

The Stockholm Convention is a global treaty to protect human health and the environment from persistent organic pollutants (POPs), of which twelve are currently listed under the Convention. POPs are chemicals that remain intact in the environment for long periods, become widely distributed geographically, accumulate in living organisms and can cause harm to humans and the environment. Norway, which is a Party to the Stockholm Convention, submitted a proposal in January 2005 to list pentabromodiphenyl ether in Annex A to the Stockholm Convention, and the POPRC agreed that the commercial product 'pentabromodiphenyl ether' ('PentaBDE') – actually a mixture as described below - met the screening criteria of Annex D to the Convention.

1.1 Chemical identity of the proposed substance

Commercial pentabromodiphenyl ether (C-PentaBDE) refers to mixtures of bromodiphenyl ether congeners in which the main components are 2,2', 4,4'- tetrabromodiphenyl ether (BDE-47 CAS No. 40088-47-9) and 2,2',4,4',5-pentabromodiphenyl ether (BDE-99 CAS No. 32534-81-9), which have the highest concentration by weight with respect to the other components of the mixture.

The numbering system for the PBDEs is the same as that used for polychlorobiphenyls (PCBs) (Ballschmiter *et al.* 1993).

The acronym PBDE is used for the generic term polybromodiphenyl ether, covering all congeners of the family of brominated diphenyl ethers. It is sometimes abbreviated to BDE.

1.2 Conclusion of the Review Committee regarding Annex D information

The Committee has evaluated Annex D information at its first meeting in Geneva in November 2005 (UNEP/POPS/POPRC.1/10) and has concluded that the screening criteria have been fulfilled for C-PentaBDE (Decision POPRC-1/3).

1.3 Data sources

This risk profile is elaborated using Annex E information submitted by countries and nongovernmental organizations, national reports from web sites for environment protection agencies in different countries, contact and submissions from Norwegian research institutes, the bromine industry, EMEP and AMAP.

Eleven countries have submitted information (Australia, Brazil, Canada, Japan, Norway, Mexico, Poland, Republic of Lebanon, Spain, Switzerland and United States of America). Seven countries submitted information on production and use. Only one country submitted information on releases; another reported that they did not have release data. All except one country provided monitoring data. There was no information on stock-piles from submitting countries and only a few have submitted information on trade. Two observers submitted information - World Wide Fund for Nature (WWF) and the International POPs Elimination Network (IPEN).

1.4 Status of the chemical under other international conventions

1.4.1 The OSPAR Convention

The Convention for the Protection of the Marine Environment of the North-East Atlantic (the OSPAR Convention) is guiding international cooperation on the protection of the marine environment of the North-East Atlantic. The OSPAR Convention was signed in Paris in 1992 and entered into force on 25 March 1998. The OSPAR Commission is made up of representatives of the Governments of 17 Contracting Parties and the European Commission, representing the European Community. In 1998, the OSPAR Commission placed PBDEs on its “List of Chemicals for Priority Action.” An OSPAR

Commission background document on PBDEs was reviewed by Sweden in 2001. The next full review of this document is not planned before 2008. At the 4th North Sea Conference, it was decided to phase out the use of brominated flame retardants by 2020.

1.4.2 The UNECE Convention on Long-range Transboundary Air Pollution

United Nations Economic Commission for Europe (UNECE) works for sustainable economic growth among its 55 member countries. The UNECE Convention on Long-range Transboundary Air Pollution was signed by 34 Governments and the European Community in 1979 in Geneva. Under it, Parties shall endeavour to limit and, as far as possible, gradually reduce and prevent air pollution including long-range transboundary air pollution. It entered into force in 1983 and has been extended by eight specific protocols. There are today 50 countries that are parties to the Convention. The Protocol for persistent organic pollutants (POPs) was adopted on 24 June 1998 in Aarhus (Denmark). It focuses on a list of 16 substances that have been singled out according to agreed risk criteria, for total ban, elimination at a later stage or restrictive use. C-PentaBDE was nominated as a new POP to the Convention in 2004 by Norway. In December 2005 it was considered by the Executive Body of the Convention to meet the screening criteria for POPs, set out in EB decision 1998/2. They requested that the UNECE Task Force on POPs continue with the review and fuether explore management strategies..

1.4.3. The Rotterdam Convention

The Rotterdam Convention is a multilateral environmental agreement designed to promote shared responsibility and cooperative efforts among Parties in the international trade of certain hazardous chemicals. It is an instrument to provide importing Parties with the power to make informed decisions on which chemicals they want to receive and to exclude those they cannot manage safely.

The text of the Rotterdam Convention on the Prior Informed Consent Procedure for Certain Hazardous Chemicals and Pesticides in International Trade was adopted at the Diplomatic Conference held in Rotterdam on 10 September 1998. The Convention entered into force on 24 February 2004 and became legally binding for its Parties. Today there are 102 states that are parties to the Convention. The EU notified PentaBDE to the Rotterdam Convention in 2003. For it to become a candidate, bans of the substance must be notified by two parties under the Convention.

1.4.4 Other international forums of relevance

The Arctic Council is a high-level intergovernmental forum that provides a mechanism for addressing the common concerns and challenges faced by the Arctic governments and the people of the Arctic. Member states are Canada, Denmark (including Greenland and the Faeroe Islands), Finland, Iceland, Norway, Russia, Sweden and United States of America. Six international organizations representing many Arctic indigenous communities have the status of Permanent Participants of the Arctic Council.

Significant monitoring and assessment of pollution in the Arctic is performed under the auspices of the Arctic Council (The Arctic Monitoring and Assessment Programme, AMAP). This work is important in identifying pollution risks, their impact on Arctic ecosystems and in assessing the effectiveness of international agreements on pollution control, such as the Stockholm Convention on Persistent Organic Pollutants (POPs). AMAP has shown that PentaBDE is one of the important pollutants of the Arctic.

In the autumn of 2004, the Arctic Council adopted a new Arctic project concerning the reduction of brominated flame retardants. The project will be managed by Norway.

2. Summary information relevant to the risk profile

2.1 Sources

2.1.1. Production and use

Based on the last information on total market demand of C-PentaBDE presented at the Bromine Science and Environmental Forum (BSEF), the estimated cumulative use of C-PentaBDE since 1970 was 100 000 metric tons (tones). The total market demand decreased during the later years of this period, for example from 8,500 tons in 1999 to 7,500 tons in 2001 (BSEF, 2001).

Table 2.1. C-PentaBDE volume estimates: Total market demand by region in metric tons (BSEF, 2001).

	America	**Europe**	**Asia**	**Rest of the world**	**Total**
1999	8,290	210	-	-	8,500
2001	7,100	150	150	100	7,500

These consumption figures need to be seen in the context of the global demand for polybrominated flame retardants of all types, which vastly outweighs the demand for C-PentaBDE. Thus, world totals of PBDE were 204,325 (1999), 203,740 (2001), 237,727 (2002) and 223, 482 (2003) tonnes (BSEF 2006).

C-PentaBDE has been produced in Israel, Japan, U.S. and EU (Peltola *et al.* 2001 and van der Goon *et al.* 2005). Since 2001 actions to regulate or voluntarily phase-out C-PentaBDE have been conducted in several countries.

Production in EU ceased in the former EU (15) in 1997 (EU 2000). Usage in the EU (15) has been declining during the second half of the 1990s and is estimated to be 300 metric tonnes in 2000 (used solely for polyurethane production) (EU 2000). The use of PentaBDE was banned in the EU (25) in 2004. Use in electrical and electronic appliances ceased on 1 July 2006.

In the United States of America, in June 2006, the U.S. Environmental Protection Agency (EPA) issued a significant new use rule on tetra-octaBDE and any combinations of these chemicals resulting from a chemical reaction, which requires persons to notify EPA before commencing manufacture or import for any use. C-PentaBDE will be banned in the state of California from 2008. The sole US manufacturer voluntarily ceased production, but use may be continuing and will cease only when stocks are fully exhausted. Although a patent on production of C-PentaBDE was taken out in China as recently as 1999 for a PBDE mixture that differs from the traditional penta-mix, the substance is being phased out in that country. Remaining production in China is estimated as less than 100 MT/year and will cease in 2007 when the substance is banned in that country.

A major bromine producer in Israel, Israel Chemicals and Industrial Products (formerly the Dead Sea Bromine Group), declares in a public statement on its web site that their products do not contain PentaBDE. This aligns the producer with the ban in the EU, which is an important market for the company's flame retardants.

There is today no production in Japan. The use of C-PentaBDE was voluntarily withdrawn from the Japanese market in 1990 (Kajiwara et al. 2004). Some developing countries around the East China Sea are potential “hot spots” releasing PentaBDE into the marine environment (Ueno *et al.* 2004). Many industrial manufacturers of computers, television sets and other electric household equipment are situated in the coastal areas of Asian developing countries (Ueno *et al.* 2004). There are indications on a phase-out of C-PentaBDE in manufacture of new electrical and electronic products in the Asian region, although uses there were always subsidiary to the major uses in polyurethane foams. The extent of this is uncertain. Waste electric products used in developed countries have been exported to Asian developing countries, such as China, India and Pakistan. This waste material has been recycled for recovery of valuable metals (Ueno *et al.* 2004) and continuation of this trade can remain a source to PentaBDE releases. No restrictions have so far been implemented in developing countries in the Asia Pacific and the southern hemisphere.

The release of 'banked' PentaBDE during recycling of foam products has its parallel in the release of CFCs and other ozone depleting substances which have similarly remained in the foam during its useful lifetime.

Results from a survey of Canadian industries regarding certain substances on the country's Domestic Substances List conducted for the year 2000 indicated that no PBDEs were manufactured in Canada, but approximately 1300 tonnes of C-PentaBDE (for incorporation into finished articles) was imported into the country (Environment Canada 2003). Based on quantities reported, C-PentaBDE was the PBDE imported in greatest volume, followed by the commercial decabromodiphenyl ether product. A very small amount of octabromodiphenyl ether was imported in 2000. The volumes reported do not include quantities imported in finished articles. In 2004, it was proposed that PentaBDE be added to the Virtual Elimination list in Canada.

In the U.S. the sole producer voluntarily ended their production of C-PentaBDE in 2004. In 2001 alone, almost 70,000 metric tons of PBDEs were produced globally, almost half of which was used in products sold in the US and Canada. Before the phase-out in U.S. the majority of C-PentaBDE formulation produced globally was used in North America (>97 %). At the end of 2004 in the US, approximately 7.5% of the more than 2.1 billion pounds of flexible polyurethane foam produced each year in the US contained the C-PentaBDE formulation (Washington State 2005).

In Australia in 2004, the National Industrial Chemicals Notification and Assessment Scheme (NICNAS) advised that all importers were phasing out imports of PentaBDE by the end of 2005, and this was reconfirmed by the major importers in mid-2005.

C-PentaBDE is used or has been used in the following sectors (Alaee *et al.* 2003, Danish EPA 1999, EU 2000, Prevedouros *et al.* 2004b, Swiss Agency for the Environment 2002, Birnbaum and Staskel, 2004):

- Electrical and electronic appliances (EE appliances) – computers, home electronics, office equipment, household appliances and other items containing printed circuit laminates, plastic outer casings and internal plastic parts such as small run components with rigid polyurethane elastomer instrument casings.
- Traffic and transport – cars, trains, aircraft and ships containing textile and plastic interiors and electrical components.
- Building materials – foam fillers, insulation boards, foam insulation, piples, wall and floor panels, plastic sheeting, resins etc.
- Furniture – upholstered furniture, furniture covers, mattresses, flexible foam components.
- Textiles – curtains, carpets, foam sheeting under carpets, tents, tarpaulins, work clothes and protective clothing.
- Packaging – polyurethane foam based packaging materials.

The most common use, accounting for 95-98% of C-PentaBDE since 1999, has been in polyurethane foam (Hale *et al.* 2002). This foam may contain between 10 and 18% of the C-PentaBDE formulation. Polyurethane foam is mainly used for furniture and upholstery in domestic furnishing, automotive and aviation industry. Other uses are in rigid polyurethane elastomers in instrument casings, in epoxy resins and phenolic resins in electrical and electronic appliances, and construction materials. For some years now, the more highly brominated Deca-BDE has been preferred in these applications. C-PentaBDE has also been incorporated in minor amounts in textiles, paints, lacquers, in rubber goods (conveyer belt, coating and floor panels) and in oil drilling fluids. Levels range from 5-30% by weight. Up to the early 1990s, C-PentaBDE was used in printed circuit boards, usually FR2 laminates (phenolic resins) in Asia. Such FR2 laminates are used in household electronics (television, radio, video), vehicle electronics, white goods (washing machines, kitchen appliances, for example). In the early 1990s the amount C-PentaBDE used in textile treatment was 60 % of total use in the EU, but this application is now banned.

C-PentaBDE has been identified as an additive flame retardant in textiles in national substance flow analyses in the ECE region (Danish EPA 1999). Manufacturers of furniture textiles have stated that the textile contained 0.45% PentaBDE in a Norwegian flow analysis reported in 2003. Stringent rules on flammability apply to textiles used in the public sector, the transport sector and business sector, but rules for domestic use are less consistent.

According to information obtained from the bromine industry the use of C-PentaBDE as hydraulic fluid (as a component of a mixture) in petroleum borings and mining was discontinued 10-20 years ago.

Australia has reported uses in manufacture of polyurethane foams for refrigerators and packaging, and in epoxy resin formulations supplied into aerospace market and for use as potting agents, laminating systems and adhesive systems. The US has reported use of C-PentaBDE in the aircraft industry. There is no use of C-PentaBDE in newer aircraft, and thus no exposure of the public, but C-PentaBDE is still used in military aircraft.

2.1.2 Global demands for brominated flame retardants in the future

According to a market analyst consulting company, the global demand for flame retardants is expected to grow at 4.4% per year, reaching 2.1 million metric tons in 2009, valued at $4.3 billion. Growth will largely be driven by gains in developing countries in Asia (China, in particular), Latin America and Eastern Europe. Strong increases are forecast for most of the flame retardants. Globally, demand will be greatest for bromine compounds, due mainly to strong growth in China. Electrical and electronic uses will grow fastest. Higher value products will continue to make inroads as substitutes for less environmentally friendly compounds, especially in Western Europe, and chlorine compounds will begin to be replaced in China by bromine- and phosphate-based and other flame retardants (Fredonia Group 2005).

After a severe falloff in demand in 2001, electrical and electronic applications will continue to recover. Demand growth for flame retardants will be strongest in such applications. As electronic circuits become smaller, and more densely packed electronics are subjected to ever higher temperatures, the need for flame retardants will increase. Construction markets will be the second fastest growing globally, but in China second place will be held by motor vehicles, followed by textiles, both of which industries are growing rapidly in that country. Plastics will continue to replace other materials such as metals and glass in a wide range of products, in order to lower both cost and weight and to allow improved design and more flexible production. Plastic usage is already widespread and growing in fields such as transportation, building products and electronics. Plastics must be made flame retardant for many applications, and as a result some 75% of all flame retardants are used in plastics (Fredonia Group 2005).

Environmental restrictions vary by region. In Western Europe, Japan and to a lesser extent in North America, such restrictions will especially limit growth of chlorinated compounds. A ban on some brominated flame retardants in Western Europe is not expected to spread substantially to other regions, but it will drive the development of alternatives in electrical and electronic equipment for sale on the world market. Dozens of Asian, European and US companies announced in 2005 that they have developed or are developing electrical and electronic equipment that does not contain C-PentaBDE. In Asia, 51% of electronic manufacturers already make products compliant with the ban on PentaBDE in the EU, and 42% expected to have products that are compliant by 1 July 2006. Officials from electronics companies and industry consultants expected that the difficulty of keeping product streams separate would ensure that most electronic equipment sold on the world market would be compliant by 2005 (International Environment Reporter 2006).

2.1.3 Releases to the environment during production

PentaBDE is released into the environment during the manufacturing process, in the manufacture of products, during their use and after they have been discarded as waste. In addition to working towards a manufacturing process that does not cause emissions, it is also important to consider the contributions of emissions from products during use as well as after they have been discarded. Most of the PentaBDE is released as diffuse pollution during and after the service life of articles incorporating C-PentaBDE and as small-scale point source pollution from the waste management chain of the end products.

PentaBDE is synthesised from diphenyl ether by brominating it with elemental bromine in the presence of a powdered iron Friedel-Craft catalyst. The producers of PentaBDE have reported that the major routes of PentaBDE from this process to the environment are filter waste and rejected material, both of which are disposed of in landfills. Waste water releases of PentaBDE may also occur from spent scrubber solutions (Peltola *et al.* 2001).

According to the EU risk assessment of PentaBDE, the emissions in polyurethane production are assumed to occur prior to the foaming process, when handling the additives (discharges to water) and during the curing (emissions to air). Releases to air may occur during the curing phase of foam production, during which the foam stays at elevated temperature for many hours, depending on the production block size. Emission to air at this stage is estimated to be 1 kg/tonne PentaBDE, but it is assumed that some of the volatilized PentaBDE condenses in the production room and ends up in the waste water. The EU risk assessment concludes that 0.6 kg of PentaBDE is released in this way, and 0.5 kg into air, for each tonne of C-PentaBDE used in polyurethane foam production.

Table 2.2 Global production and use of C-PentaBDE in polyurethane foam production, and estimation of associated releases in 2000 (foam containing 10-18% PentaBDE).

Polyurethane foam production	**Quantity of PentaBDE**	**Release of PentaBDE into waste water**	**Emissions of PentaBDE to air during production**
150,000 tonnes/year	15,000-27,000 tonnes/year	9,000-16,200 kg/year	7,500-13,500 kg/year

An important source of release has been associated with the use of liquid flame retardant additives such as C-PentaBDE in production of polymer foams. Approximately 0.01% (that is, 100 g /tonne) of the raw material handled during mixing is estimated to be released to wastewater. There is also potential for release due to volatilization during the curing phase as described above, since foam reaches temperatures of 160°C for several hours. Wong *et al.* (2001) examined the atmospheric partitioning characteristics of BDEs 47, 99 and 153, and predicted that tetra- and pentabromo-congeners will become gaseous at warmer air temperatures. Therefore, although the low measured vapour pressure values for the PBDEs indicate that volatilization is minimal at normal air temperatures, there is potential for release to air at the elevated temperatures reached during curing (European Communities 2001). The European Communities (2001) study estimates the overall release of PentaBDE to be approximately 0.11%, with about one half of this going to air and the other half to wastewater.

2.1.4 Releases to the environment during product use

C-PentaBDE is used solely as an additive in physical admixture with the host polymer, and can thus migrate within the solid matrix and volatilize from the surface of articles during their life cycle (EU 2000). Approximately 3.9 % of the PentaBDE present in articles was estimated to be released each year through volatilization during their assumed service life of 10 years in the EU risk assessment, but each congener will have its own characteristic migration and volatility coefficients. Based on the quantities of shown in Table 2.2, and the 3.9% loss rate, it can estimated that 585-1053 tonnes of PentaBDE enters the environment in this way each year.

Wilford *et al.* (2003) conducted controlled chamber experiments in which they passed air through samples of C-PentaBDE -treated foam products containing 12% PBDE w/w. They found that PBDEs volatilized from polyurethane foam at measurable levels. Average total PBDE levels of 500 ng/m^3/g foam were released from the chamber. For BDE-47, BDE-99 and BDE-100 (4,5 and 5 bromines, respectively), the loss rates were 360, 85 and 30 ng/m^3/g foam, respectively. The average temperature range during sampling was 30-34^{o}C.

Given the use of C-PentaBDE in domestic items such as furniture, carpeting and appliances, exposure to indoor air house dust containing PentaBDE has been examined in a number of studies (Shoeib *et al.* 2004, Wilford *et al.* 2005). US researchers (Stapleton *et al.* 2005) report results for a study conducted in 2004 in the Washington, DC, metropolitan area and one home in Charleston, South Carolina. The concentrations of PBDEs in house dust from sixteen homes ranged from 780 ng/g dry mass to 30,100 ng/g dry mass. The dominant congeners were those associated with C-PentaBDE and DecaBDE. It was estimated that young children (1-4 years) would ingest 120-6000 ng/day of PBDEs. For five of the homes, clothes dryer lint was also analyzed, showing PBDE concentrations of 480-3080 ng/g dry mass. The exposures are higher than those observed in Europe, a fact that the researchers attribute to the fact that most markets for C-PentaDBE has been in the United States.

The information in the preceding paragraph highlights the fact that while PentaBDE can volatilize from the products in which it is incorporated, as well as during their whole life-cycle, and during recycling or after disposal, a major route for dissemination of this chemical into the environment will be in the form of particles on which it is absorbed or adsorbed. When emitted from products, the flame retardants are likely to adsorb to particles, and these may adhere to surfaces within appliances or on other surfaces in the indoor environment, or they may spread to the outdoor environment during airing of rooms. Industrial environments where equipment is dismantled may suffer much higher exposures (Danish EPA 1999). There are also releases from products due to weathering, wearing, leaching and volatilization at the end of their service life during disposal or recycling operations (dismantling, grinding or other handling of waste, transport and storage, for example). The annual releases in the EU region from the product life-cycle of polyurethane products were estimated to be distributed among the different compartments as follows: 75% to soil, 0.1% to air and 24.9% to surface water (EU 2000).

The inclusion of C-PentaBDE in materials used for car undercoating, roofing material, coil coating, fabric coating, cables, wires and profiles, and shoe soles can result in slow release to the environment. Emission factors for such releases in the EU risk assessment were judged to be 2-10% during the lifetime of the product, with the higher factors applying to uses with high wear rates such as car undercoating and shoe soles. A further 2% was assumed to be emitted during disposal operations. Taking these into account, the losses in the EU region were estimated to be 15.86 tonnes/year to soil, 5.26 tonnes/year to surface water, and 0.021 tonnes/year to air. No actual measurements were found in the literature with which one might compare these estimates.

Hale *et al.* (2002) demonstrated that flame-retardant treated polyurethane foam exposed to direct sunlight and typical Virginia summer conditions with temperatures up to 30-35^{o}C and humidity of 80% or greater, became brittle and showed evidence of disintegration within four weeks. The authors postulate that the resulting small, low density foam particles would be readily transportable by stormwater runoff or air currents. Such degradation processes may provide an exposure route to organisms via inhalation or ingestion of the foam particles and their associated PentaBDE.

2.1.5 Emissions from waste containing C-PentaBDE

Waste can be generated from production of C-PentaBDE, from processes for manufacture of C-PentaBDE -containing materials, and from end-of-service-life management of products containing PentaBDE.

In production, the C-PentaBDE producers have stated that the major source of release was from filter waste and reject material, but quantities are small to negligible. In general, the waste was disposed of to landfill (EU 2000), although it is noted that waste containing more than 0.25% PentaBDE is classified as 'hazardous waste'.

After curing and cooling, blocks of polyurethane foam generally have to be cut to the required size, although for some applications the foam is produced in a mould of the desired shape so cutting is not required. Some flame retardant is lost in the scrap foam that results from the cutting process. Such foam scrap is often recycled into carpet underlay (rebond), particularly in the United States. Interestingly, the EU exports about 40,000 tonnes/year of scrap foam to the US for such use (EU 2000). In other uses, scrap foam is ground and used as filler in a number of applications such as cars eats or used for addition to virgin polyol in slab foam production. It is also possible that some foam scrap will be disposed of to landfill, or even incinerated.

During the production of printed circuit boards a substantial part of the laminate is cut off and becomes solid waste. In most countries, however, C-PentaBDE is no longer used in this application. There is limited information about waste generated in other applications of C-PentaBDE, such as its use in electrical and electronic appliances. While some such appliances are recycled on account of their metal content, many are burned in municipal waste incinerators and this often the fate of non-metallic portions of this waste stream. In the EU, from December 2006, plastics containing brominated flame retardants must be separated from such waste prior to recovery and recycling.

Used vehicles, often containing solid or foam components with C-PentaBDE are stored outdoors and then dismantled in shredder plants. In some countries, restrictions require that components containing substances like PentaBDE be treated as hazardous waste. Wastes generated from production of building materials, textiles and furniture are disposed of in landfills, or incinerated. This is easy enough for small, easily dismounted components, but most material containing flame retardants is harder to segregate and so these materials end up in the waste from shredder plants and are usually landfilled.

Movement of polymer foam particles containing PentaBDE within the landfill could provide a mechanism for transport of the brominated material to leachate or groundwater. It is not currently possible to assess the significance of such processes. However, given the physico-chemical properties of the substance, it is considered unlikely that significant amounts of PentaBDE will leach from landfills, since it has low water solubility, high octanol-water partition coefficient, and adsorbs strongly to soils (EU 2000). Norwegian screening studies have found levels of PentaBDE of concern in landfill leachates (Fjeld *et al.* 2003, Fjeld *et al.* 2004, Fjeld *et al.* 2005). The quantity of PentaBDE disposed of annually in the EU, and going to landfill or incineration, is estimated to be approximately 1,036 tonnes (EU 2000).

In a Dutch project, the emissions of PentaBDE in the EMEP region were estimated and distribution between sources was as follows: 0.33 tonnes/year from industrial combustion and processes, 9.45 tonnes/year from solvent and product use and 0.05 tonnes/year from waste incineration (van der Gon *et al*. 2005).

At the operating temperatures of municipal waste incinerators almost all flame retardants will be destroyed, but based on experience with other organic compounds, trace amounts could be passing the combustion chamber (Danish EPA 1999). Studies of recipients to municipal solid waste incinerators have detected above-background levels of PentaBDE in both gaseous and particulate fractions in the air in the vicinity of the facility (Agrell *et al.* 2004, Law 2005, ter Schure *et al.* 2004b). Potentially toxic products like brominated dibenzo-*p*-dioxins and dibenzofurans may be produced during incineration of articles containing C-PentaBDE (Danish EPA 1999, Ebert and Bahadir 2003, Weber and Kuch 2003, Birnbaum and Staskel 2004) and possibly released to the environment.

Analyses of dismantled FR2 circuit boards in electrical scrap show that about 35% of the PBDE used was PentaBDE, and for estimation purposes it was assumed that 25% of FR2 laminates in older appliances had been treated with the C-PentaBDE (Swiss agency 2002). Prevedouros *et al.* (2004) estimated production, consumption, and atmospheric emissions of PentaBDE in Europe between 1970 and 2000 based on literature data. According to that study, the flow of PentaBDE in discarded electrical and electronic appliances in Europe is in the range 17-60 metric tons per year for the period 2000-2005. However, a Swiss experimental study of such flow in a modern recycling plant showed values higher than expected on the basis of the literature study. This could mean that the literature has under-estimated the PBDE content of such appliances, and the study acknowledges that companies seldom provide all the information necessary to make accurate estimates (Swiss agency 2002). This same study reported a flow analysis for the life cycles of Penta-, Octa- and Deca-BDE as well as tetrabromobisphenol A (TBBPA). Waste electrical and electronic equipment was the biggest contributor, ahead of automotive shredder residues and construction waste. The plastics in vehicles produced in 1980 contained 0.089 g/kg of PentaBDE (excluding that contained in electrical and electronic components), whereas plastic in those built in 1998 had 0.044 g/kg. At the beginning of this period, almost all unsaturated polyurethane resins were treated with brominated flame retardants, primarily DecaBDE and TBBPA, but also PentaBDE. Even larger quantities, up to 50 g PentaBDE/kg of resin, were used in rail vehicles produced in 1980.

The average concentration of PentaBDE in appliances is estimated to be 34 mg/kg, with the highest concentration – 125 mg/kg – in the plastic fraction (Morf *et al.* 2005). In plants with off-gas filtering, a large proportion of the PentaBDE will be found in the collected fraction (Morf *et al.* 2005). On the other hand, in a facility without an efficient air pollution control device such as that in the modern facility studied, a significant flow of dust-borne PentaBDE may be released to the environment. A case in point was presented by Wang *et al.* (2005), who detected levels of PentaBDE in soil and sediment collected in the vicinity of an open electronic waste disposal and recycling facility located in Guiyu, Guandong, China.

The Swiss study showed that 5% of polyurethane foams produced in 1990 were used in the building industry, and contained up to 220 g/kg of C-PentaBDE. About 10-20% of the thermoplastic sheeting used in construction was treated with brominated flame retardants at levels of 1.3-5% by weight (Danish EPA) but no information about C-PentaBDE content is available. Some polyvinyl chloride sheeting would also have been treated with C-PentaBDE, typically at 49 g/kg. PentaBDE can be assumed to be emitted during dismantling activities but no information is available about the extent of such emissions.

2.2 Environmental fate

2.2.1 Persistence

Estimated half-life values of PDBE in different environmental compartments are scarce in the literature. In table 2.3 half-life estimates found in literature are summarized.

*Table 2.3 Half-lives of PentaBDE (**BDE-99**) in different environmental compartments, estimated with the use of Syracus Corporation's EPIWIN program.*

Environmental compartment	**Half-life estimate (d)**	**References**
Soil	150	Palm 2001, Palm *et al.* 2002
Aerobic sediment	600	Palm 2001, Palm *et al.* 2002
Water	150	Palm 2001, Palm *et al.* 2002
Air	19 11	Palm *et al.* 2002 Vulykh *et al.* 2004

It is noted that caution should be used in relying on half-life estimates derived from this program, now called EPI Suite (http://www.epa.gov/opptintr/exposure/docs/episuite.htm). The EPI Suite's intended use is chemical screening only and may not be appropriate for consideration of substances for global control. Because of interest in this matter, it is likely that half-life data from new studies will be published but the picture provided by existing data seems unlikely to change substantially. The nature of degradation products of the PBDEs is also likely to be elucidated in future, leading to consideration of <u>their</u> toxicity.

With respect to biodegradation, Tetra-, Penta- and Hexa-BDE are predicted to be "recalcitrant" by the BIOWIN program. Using the EPIWIN program, estimated half-lives for PentaBDE are 600 days in aerobic sediment, 150 days in soil, and 150 days in water (Palm 2001). This degree of persistence is supported by the fact that no degradation (as CO_2 evolution) was seen in 29 days in an OECD 301B ready biodegradation test using PentaBDE (Schaefer and Haberlein 1997).

Schaefer and Flaggs (2001) carried out a 32-week anaerobic degradation study using a mixture of ^{14}C-labelled and unlabelled BDE-47 (a TetraBDE) incorporated into sediments. The study showed that <1% of the total radioactivity was recovered as $^{14}CO_2$ and $^{14}CH_4$, indicating that essentially no mineralization had occurred. Overall, the study found that levels of degradation were not statistically significant; however, the HPLC analytical method with radiometric detection indicated that some products had been formed in the 32-week samples. Between one and three such peaks were identified in 26 of 42 samples analyzed. Work is underway to identify these products. It is likely that BDE-47 has the potential to degrade very slowly under anaerobic conditions.

Several studies using sediment cores show that PentaBDE congeners deposited in European marine sediments at the beginning of 1970s are still present in significant amounts, indicating high persistency in sediments (Covaci *et al.* 2002a, Nylund *et al.* 1992, Zegers *et al.* 2000, Zegers *et al.* 2003). The industrial production and use in Europe started in the beginning of the 1970s, with a reduction in more recent years. This is reflected in the sediment core profiles, with no occurrence before this date, and an increase in levels after, with a levelling off in more recent years. In the most recent studies (Zegers *et al.* 2003) sediment cores from Norway, the Netherlands and Germany were studied. Concentrations of PBDEs, normalized to total organic carbon content, were in the range 10-20 μg/g total carbon.

<u>2.2.2 Bioaccumulation</u>

<u>2.2.2.1 Studies on bioaccumulation and biomagnification in local food webs</u>

Several studies have focused on PentaBDE's potential for bioaccumulation and biomagnification. The studies show an increase of concentrations in biota with increasing trophic level in pelagic and Arctic food webs. The calculated bioconcentration factors (BCFs), bioaccumulation factors (BAFs) and biomagnification factors (BMFs) indicate PentaBDE's potential for bioaccumulation and biomagnification. In Table 2.4 the calculated values in the literature are summarized. The octanol/water partition coefficient (log K_{OW}) for PentaBDE in those studies is 6.5 – 7.4. The more recent studies are described in the following text.

Table 2.4 Calculated bioaccumulation factors (BAFs) and biomagnification factors (BMFs) for one PentaBDE (BDE-99) in the literature from environmental studies in pelagic and Arctic food webs. The data are calculated using the mean lipid weight concentrations, except for the study performed by Sørmo et al. 2006, in which the values in brackets are BMFs calculated from mean whole body concentrations.

Variable	**Organism**	**Area**	**Value**	**Reference**
BAF	*Dreissena polymorpha*	Lake Mälaren, Sweden	1.8	Lithner et al. 2003
BMF	Guillemot egg/herring	Baltic sea	17	Sellström 1996
	Grey seal/herring	Baltic sea	4.3	Sellström 1996
	Salmon/sprat	Baltic sea	10	Burreau *et al.* 1999
	Salmon/sprat	Baltic sea	5.9	Burreau *et al.* 2000
	Atlantic Salmon/Small Herring	The Northern Atlantic Sea	3.8	Burreau *et al.* 2000
	Net plankton/Benthic organisms	Lake Ontario, Canada	7.1	Alaee *et al.* 2002
	Benthic organisms/Forage fish	Lake Ontario, Canada	0.8	Alaee *et al.* 2002
	T. libellula/Copepods	Svalbard, Arctic Norway	0.65 (1.3)	Sørmo *et al.* 2006
	G.wilkitzkii/Copepods	Svalbard, Arctic Norway	47.6 (19.0)	Sørmo *et al.* 2006
	Polar cod/Copepods	Svalbard, Arctic Norway	2.1 (1.6)	Sørmo *et al.* 2006
	Polar cod/*T. inermis*	Svalbard, Arctic Norway	1.9 (1.2)	Sørmo *et al.* 2006
	Polar cod/*T. libellula*	Svalbard, Arctic Norway	3.4 (1.3)	Sørmo *et al.* 2006
	Polar cod/*G.wilkitzkii*	Svalbard, Arctic Norway	0.04 (0.1)	Sørmo *et al.* 2006
	Ringed seal/*T. inermis*	Svalbard, Arctic Norway	26.8 (54.5)	Sørmo *et al.* 2006
	Ringed seal/*T. libellula*	Svalbard, Arctic Norway	43.1 (60.0)	Sørmo *et al.* 2006
	Ringed seal/*G.wilkitzkii*	Svalbard, Arctic Norway	0.6 (3.9)	Sørmo *et al.* 2006
	Ringed seal/Polar cod	Svalbard, Arctic Norway	13.7 (56.6)	Sørmo *et al.* 2006
	Polar bear/Ringed seal	Svalbard, Arctic Norway	0.3 (0.29)	Sørmo *et al.* 2006
	Polar bear/Ringed seal	Arctic Canada	3.4	Muir *et al.* 2006
	Polar bear/Ringed seal	Arctic Canada	11	Muir *et al.* 2006
	Polar bear/Ringed seal	Arctic Canada	8.0	Muir *et al.* 2006
	Polar bear/Ringed seal	Greenland	1.0	Muir *et al.* 2006
	Polar bear/Ringed seal	Svalbard, Arctic Norway	5.9	Muir *et al.* 2006

PBDE analyses of zebra mussels (*Dreissena polymorpha*) were included in a larger study undertaken in and around the city of Stockholm, Sweden (Lithner *et al.*, 2003). Mussels were collected from a background site and transplanted in baskets to other downstream sites in Lake Mälaren, Saltsjön and in several small lakes. Freshwater flows from Lake Mälaren, through the middle of Stockholm, then out into the brackish Baltic Sea via Saltsjön. Five PBDE congeners (BDE-47, BDE-99, BDE-100, BDE-153 and BDE-154) were determined. The congener pattern was dominated by BDE-47 and BDE-99 (four and five bromines, respectively) and was similar to the C-PentaBDE. Bioaccumulation factors (BAFs) for the various compounds studied were estimated using data from suspended particulate matter (SPM) collected in sediment traps in 1998-99 at the same sites in Riddarfjärden and Saltsjön (Broman *et al.*, 2001). The concentrations on SPM were assumed to reflect water concentrations. BAFs were calculated using lipid weight concentrations in mussels and organic carbon based concentrations in the SPM.

When compared to other compounds (PCBs, DDTs, HCB), the BDEs had the highest BAFs, ranging from 1 to 2. The BAF (= level in mussel/level in SPM) for PentaBDE was 1.8.

Concentrations of BDE-47 and BDE-99 in Lake Ontario pelagic food web show increasing concentrations with increasing trophic position (Alaee *et al.* 2002). In this study, concentrations of PBDEs in archived plankton, *Mysis*, *Diporeia*, alewife, smelt, sculpin and lake trout samples collected in 1993 were determined. The trophodynamics of PBDEs in the Lake Ontario pelagic food web were also investigated. Lake Ontario pelagic food web consists of three trophic levels. The lake trout (*Salvelinus namaycush*) is a top predator fish species in Lake Ontario, feeding on forage fish including alewife (*Alosa pseudoharengus*), rainbow smelt (*Osmerus mordax*) and slimy sculpin (*Cottus cognatus*); in turn these fish feed on *Mysis* and *Diporeia*, which feed on phytoplankton, and zooplankton sampled as net plankton. Concentrations were increasing at each step up the food chain. The exception to this trend was the biomagnification of BDE-99 from benthic organisms to forage fish, which had a biomagnification factor of 0.8. This is an indication of the breakdown of BDE-99. In fact, the PBDE profile in the plankton; *Mysis* and *Diporeia* resembled the C-PentaBDE formulation, which indicates that BDE-99 bioaccumulates in the invertebrates and starts to be metabolized by forage fish.

Further studies of metabolism involving reductive debromination are discussed in Section 2.3.5.

Whittle *et al.* (2004) conducted surveys of PBDE levels in fish communities of Lake Ontario and Lake Michigan in 2001 and 2002 and evaluated biomagnification in the local pelagic food web (net plankton/*Mysis*/*Diporeia* → forage fish (smelt/sculpin/alewife) → lake trout). Their analysis, which included a total of forty one PBDE congeners, showed that BDE 47, 99 and 100 were prominent at each trophic level. The biomagnification factors (BMFs) representing total PBDEs for forage fish to lake trout ranged from 3.71 to 21.01 in Lake Michigan and from 3.48 to 15.35 in Lake Ontario. The BMF for plankton to alewife as 22.34 in Lake Ontario.

A recent study of an Arctic food chain shows the same result (Sørmo *et al.* 2006) as Alaee´s study. Concentrations of PBDEs were investigated in an Arctic marine food chain, consisting of four invertebrate species, polar cod (*Boreogadus saida*), ringed seals (*Pusa hispida*) and polar bears (*Ursus maritimus*). The most abundant PBDEs, BDE-47 and BDE-99, were found in detectable concentrations even in zooplankton, the lowest trophic level examined in this study. Most of the investigated PBDEs biomagnified as a function of tropic level in the food chain. A noticeable exception occurred at the highest trophic level, the polar bear, in which only BDE-153 was found to increase from its main prey, the ringed seal, indicating that polar bears appear to be able to metabolize and biodegrade most PBDEs. The authors suggested that this discrepancy in the fate of PBDEs among the different species may be related to greater induction of oxidative detoxification activities in the polar bear. Absorption and debromination rates may be more important for bioaccumulation rates of PBDEs in zooplankton, polar cod and ringed seals. BDE-99 showed no biomagnification from pelagic zooplankton to polar cod, probably as a consequence of intestinal or tissue metabolism of BDE-99 in the fish. Also among pelagic zooplankton, there was no increase in concentrations from calanoid copepods to *T. libellula*. Lipid-weight based concentrations (LWCs) and whole-body based concentrations (WBCs) of PBDEs were used to assess biomagnification factors (BMFs). Whole body concentrations gave the most realistic BMFs, as BMFs derived from LWCs seem to be confounded by the large variability in lipid content of tissues from the investigated species. This study demonstrates that PentaBDEs have reached measurable concentrations even in the lower trophic levels (invertebrates and fish) in the Arctic and biomagnifies in the polar bear food chain.

Polybrominated diphenyl ethers (PBDEs) were determined in adipose tissue of adult and sub-adult female polar bears sampled between 1999 and 2002 from sub-populations in Arctic Canada, eastern Greenland, and Svalbard, and in males and females collected from 1994 to 2002 in northwestern Alaska (Muir et al. 2006). Only four congeners (BDE-47, BDE-99, BDE-100, and BDE-153) were consistently identified in all samples. BDE-47 was the major PBDE congener representing from 65% to 82% of the

ΣPBDEs. Age was not a significant covariate for individual PBDEs or ΣPBDE. Higher proportions of BDE-99, BDE-100, and BDE-153 were generally found in samples from the Canadian Arctic than from Svalbard or the Bering- Chukchi Sea area of Alaska. All four major PBDE congeners were found to biomagnify from ringed seals to polar bears. The polar bear-seal BMFs were relatively consistent despite the large distances among sites. The exceptions were the BMFs for BDE-99, BDE-100, and BDE-153 in East Greenland which had lower BMFs than those at all other sites. This may imply differences in the transformation of PBDEs in the marine food web leading to polar bears or to food web differences. Species differences in bioaccumulation and biotransformation of PBDEs have been noted for fish and this could lead to differences in congener patterns in fish-eating mammals and their predators.

Studies of the biomagnification of Tri- to DecaBDEs were carried out in three different food chains, two in the Baltic Sea and one in the Atlantic Ocean (Law 2005). All of Tri- to HeptaBDE congeners biomagnified, but the maximum biomagnification was for the PentaBDEs.

Matscheko *et al.* (2002) investigated the accumulation of seven PBDEs, eight PCBs and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCCD/Fs) by earth worms collected from Swedish soils in spring and autumn 2000. The selected sampling sites were agricultural lands receiving applications of sewage sludge, and a field flooded by a river known to contain the target substances in its sediment. Reference sites were rural and urban soils with no known sources of the target substances other than background. Earthworms (primarily *Lumbricus terrestris*, *Lumbricus spp*, *Aporrectodea caliginosa, A. rosea* and *Allolobophora chlorrotic*) were collected from all field sites, starved for 24 h to clear gut contents, and then analyzed for the presence of the target substances. Biota-soil accumulation factors (BSAFs) were calculated as the ratio of concentration of target substance in worm lipids to that in soil organic matter. BSAFs for BDE-47, BDE-66, BDE-99 and BDE-100 ranged from 1 to 10. They were comparable to those determined for the PCBs but higher than those for PCCD/Fs. BSAFs of greater than 10 were determined at one agricultural site, where factors of 11, 18 and 34 were calculated for BDE 99, 47 and 100 respectively. Data collected for BDE-153, BDE-154 and BDE-183 were not used, as levels in the earthworm blanks were deemed to be unacceptable high.

2.2.2.2 Monitoring results indicating bioaccumulation

A large range of studies show concentrations of concern in top predators. High levels in top predators are usually an indication on the potential of a compound to bioaccumulate in the top predator food chain.

Several studies (Jaspers *et al.* 2004, Herzke *et al.* 2005, Lindberg *et al.* 2004, D`Silva *et al.* 2004, Law *et al.* 2005, Sinkkonen *et al.* 2004, Sellström *et al.* 2003) indicate that PentaBDE is widespread in top predatory birds in Europe, such as peregrine falcon (*Falco peregrine*), merlin (*Falco columbarius*), goshawk (*Accipiter gentiles*), golden eagle (*Aquila chrysaetos*), and buzzard (*Buteo buteo*). High levels are detected in top predatory eggs of white-tailed sea eagle, peregrine falcon, osprey, and golden eagle (Herzke et al. 2005, Lindberg *et al.* 2004). High levels have also been detected in European harbour porpoises (*Phocoena phocoena*) (Thron *et al.* 2004 and Covaci *et al.* 2002).

In the Arctic, C-PentaBDE is detected in high levels in top predatory birds and mammals (Verrault *et al.* 2005, Verrault *et al.* 2004, Norström *et al.* 2002, Herzke *et al.* 2003, Vorkamp *et al.* 2004a and b, Wolkers *et al.* 2004, Thron *et al.* 2004, Thomas *et al.* 2005, Ikonomou *et al.* 2002), such as glaucous gulls (*Larus hyperboreus*), polar bears (*Ursus maritimus*), ringed seals (*Phoca hispida*) and beluga whales (*Delphinapterus leucas*).

2.2.3 Long-range environmental transport

2.2.3.1 Environmental studies on transport and distribution

There are several factors indicating long-range transboundary transport of PentaBDE in the environment. It has a high persistency in air, with a half-life of 11-19 days (Palm *et al.* 2002, Vulykh *et al.* 2004)). Monitoring studies have detected a widespread occurrence in the European atmosphere (ter Shure *et al.* 2004, Lee *et al.* 2004, Jaward *et al.* 2004, Harrad and Hunter 2004, Harrad *et al.* 2004) and Arctic (AMAP 2002 and AMAP 2005, Peltola *et al.* 2001).

Sampling of air in the Great Lakes region of North America was undertaken in 1997-1999 and reported by Strandberg *et al.* (2001). PBDEs, mainly BDE-47 and BDE-99, were detected in all samples from four locations, and there was little variation over the time period. PBDE concentrations ranged from 5 pg/m^3 near Lake Superior to about 52 pg/m^3 in Chicago. At the temperatures of collection, 20±3°C, approximately 80% of the tetrabromo congeners were in the gas phase, but 70% of the hexabromo congeners were associated with particles.

Results for the far-northern Pacific covered particulate matter collected in July-September 2003 from the Bohai Sea to the high Arctic, 37° – 80° N (Xin-Ming Wang *et al.* 2005). The dominant congeners were BDE-47, BDE-99, BDE-100 (all present in the commercial pentamix) and BDE-209, with concentrations falling from mid- to high-latitudes, probably resulting (according to the authors) from dilution, deposition and decomposition of the PBDEs during long-range transport. Total PBDE concentrations were in the range 2.25 – 198.9 pg/m^3 with a mean of 58.3 pg/m^3. The source of the PBDEs is believed to be the North American continent from which they distill to an Arctic 'cold trap'.

The emphasis on any assessment of the dispersal of PentaBDE into the environment has to be on long-range transport, specially to Arctic regions, but there also is a growing body of data on dispersal of the substance and related congeners within regions. Air sampling in Southern Ontario in the Spring of 2000, before bud burst, showed PBDE concentrations of 88-1250 pg/m^3, with the lighter congeners (DBE-17, -28 and -47) dominating (Gouin *et al.* 2002). The concentrations fell to 10-20 pg/m^3, a change that the researchers attributed to, firstly, enhanced levels caused by expiration from the winter snowpack, followed by possible sorption by emergent foliage. Other studies in Ontario (Harner *et al.* 2002) found air levels of total PBDE in the range 3.4-46 pg/m^3. In later work, organic films on indoor and outdoor windows in Southern Ontario were examined for their content of PBDEs by Butt *et al.* (2004). While the PBDE content was dominated by BDE-209 from the decabromo mixture, there were significant quantities of congeners deriving from the C-PentaBDE. Back calculation gave total PBDE concentrations in outdoor air of 4.8 pg/m^3 and 42.1 pg/m^3 for indoor air.

Jaward *et al.* (2004a) studied a total of 71 passive air samples using semi permeable membrane devices (SPMDs) for eight BDE congeners (BDE-28, BDE-47, BDE-49, BDE-75, BDE-99, BDE-100, BDE-153 and BDE-154) during a six week period in 2002 at remote/rural/urban locations across 22 countries in Europe. BDEs were detected in approximately 50% of the samples, and the equivalent ΣBDE air concentrations estimated from the passive sampler data ranged from 0.5 to 250 pg m^3. The focus of the most elevated concentrations was the UK, which has a history of PBDE production and has also been a major user of PBDE formulations due to stringent fire regulations within the country. The UK is clearly a regional source for BDEs to the European atmosphere and, in contrast, levels reaching Europe from the west (over the Atlantic Ocean) are low. Other high values were detected in urban centres in mainland Europe – samples from Athens, Bilthoven (Netherlands), Geneva, Milan and Seville, for example. Non-detectable/very low values occurred in remote/background sites, especially in Iceland, Ireland, Norway and Sweden, and values in Eastern Europe were generally low. BDE-47 and BDE-99 contributed ca. 75% to ΣBDE, similar to their proportion in the Bromkal 70-5DE C-PentaBDE.

In the US, high-volume samplers were used to examine concentrations of gaseous and particulate PBDEs at five sites (urban, semi-urban, agricultural and remote) from the Midwest to the Gulf of Mexico, every twelve days during 2002-2003 (Hoh and Hites 2005). The mean concentration of total PBDEs at the Chicago site was 100±35 pg/m^3, some 3-6 times higher than those at other sites and significantly higher than measurements made in 1997-1999 (Strandberg *et al.* 2001). The mean concentration of PentaBDE was 31 pg/m^3 at the Chicago site, some 2-4 times the values for other sites.

Fugacity model results indicate that PBDEs will largely partition to organic carbon in soil and sediment and that their persistence will be strongly influenced by degradation rates in these media (although these are not well known). Only a small proportion of PBDEs exist in air and water. If this is the case, it suggests that these compounds have limited LRAT potential (Prevedouros *et al.* 2004a, Gouin and Harner 2003). This corresponds with PentaBDE's affinity for carbon, low solubility in water (1.0 µg/L) and low vapour pressure (7.6 x 10^{-6} Pa). However, Gouin and Harner (2003) suggest that because of their physical–chemical properties, PBDEs may experience active surface–air exchange as a result of seasonally and diurnally fluctuating temperatures. Subsequently, this may result in the potential for LRAT of the PBDEs through a series of deposition/volatilization hops, otherwise known as the ‘‘grasshopper’’ effect. This assumption is supported by environmental data. Lee et al. (2004) detected atmospheric concentrations of BDEs at two rural/semirural sites in England, and one remote site on the west coast of Ireland in 2001 and in 2000, respectively. ΣBDE concentrations at Mace Head, Ireland, were 0.22 to 5.0 pg/m^3 with a mean of 2.6 pg/m^3 and were controlled primarily by advection. ΣBDE concentrations at Hazelrigg (NW England) were 2.8 to 37 pg/m^3 with a mean of 12 pg/m^3, and at Chilton (SW England) were 3.4 to 33 pg/m^3 with a mean of 11 pg/m^3. The congener profile was, on average, similar to that of the C-PentaBDE. At the two English sites in the summer, PBDE concentrations were strongly influenced by temperature, indicating that land/air exchange processes play an important role in determining atmospheric concentrations.

The concentrations of PBDEs were determined in soil samples collected along a latitudinal transect through the UK and Norway, at remote/rural woodland (both coniferous and deciduous) and grassland sites (Hassanin *et al.* 2004). Concentrations for ΣBDE ranged from 65 to 12,000 ng/kg dry weight. BDE congeners BDE-47, BDE-99, BDE-100, BDE-153 and BDE-154, covering the major constituents of the C-PentaBDE, dominated the average congener pattern in the soils. This was interpreted as evidence that transfer of the congeners from materials treated with the commercial product from source to air to soil occurs with broadly similar efficiency, and that there is little degradation of the congeners by processes acting either during atmospheric transport or within the soils themselves. There was evidence of latitudinal fractionation of the BDE congeners, with the relative amounts of BDE-47 and the lighter congeners increasing to the north (with increasing distance from source areas) while the proportion of BDE-99 and the heavier congeners decreased. Plots of BDE congener concentrations against percentage soil organic matter yielded different slopes for different congeners. Steeper slopes were generally observed for lighter congeners such as BDE-47, indicating that they have undergone some air-surface exchange (“hopping”), whilst those of heavier congeners such as BDE-153 were close to zero, indicating that they are retained more effectively by soil following deposition. A Japanese study detected seasonal variations in the partitioning of PBDEs between the gas and particulate phase. The fraction of particulate PBDEs was higher in samples collected in winter than those in the summer (Hayakawa *et al.* 2004). PentaBDE is expected to be transported in the environment mostly by being absorbed onto particles due to its low volatility, low solubility and high affinity for carbon compounds. There are results from environmental studies which indicate that PBDEs are transported on air borne particles, and that they are susceptible to wet deposition (ter Schure *et al.* 2004a, ter Schure and Larsson 2002). Further transport depends on the fate of the particles. Fate after depositions on land depends on the level of wind erosion, that can vary with the season. Fate after deposition into the sea depends on oceanographic processes, such as water layering and transport by currents in the surface layers.

Ter Schure *et al.* (2004a) collected air and atmospheric bulk deposition samples on the island of Gotska Sandön in the Baltic Proper during a 10 week period in autumn 2001. The sampling site was chosen because of its central position in the Baltic Sea, and because of the absence of local point sources of pollution. Ten PBDE congeners were determined (BDE-17, BDE-28, BDE-47, BDE-85, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209). The median ΣBDE concentration (ΣBDE is the sum of the concentrations of the congeners determined in each study) was 8.6 pg/m^3, and the BDEs were mainly associated with particles. A comparison to levels of PCB in the atmosphere indicated that, as PCB concentrations in Baltic air have been declining, the input of BDEs by atmospheric deposition to the Baltic Proper now exceeds that of the PCBs by a factor of almost 40 times.

BDEs were determined in precipitation falling in southern Sweden during a two week period in 2000 (ter Schure and Larsson, 2002). The particle-associated and "dissolved" phases were separated during sampling and 65 ± 18% of ΣBDE was found to be particle-associated. The volume weighted mean concentration of ΣBDE (nine congeners) in rain was 209 pg/l, and the total deposition rate was 2 ± 1 ng ΣBDE/m^2/day. The congener profile in both phases of the total deposition was dominated by BDE-209, and thereafter BDE-47, BDE-99 and BDE-183, representing inputs from all three commercial PBDE formulations. The authors found that particle associated BDEs are effectively removed during small precipitation episodes, and that particle scavenging was an important mechanism for the wet deposition of BDEs.

A model assessment of potential for long-range transboundary atmospheric transport and persistence of PentaBDE have been carried out by EMEP (Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe). The values of LRTP were considered to be strongly influenced by environmental processes, such as degradation, deposition, gas/particle partitioning, and gaseous exchange with underlying surface. The main process of removal from the atmosphere for the two congeners BDE-47 and BDE-99 was found to be deposition to land and seawater, 78% to land and 15% to sea for BDE-47 and 77% to land and 21% to sea for BDE-99. Only 7% of BDE-47 and 2% of BDE-99 was degraded. The calculated half-life in air was 7 days for BDE-47 and 11 for BDE-99. The findings showed a spatial distribution of BDE-47 that covers the Arctic, Europe, the Mediterranean Sea and northern Africa. BDE-99 spreads over longer distances and spreads to the Arctic, Atlantic Ocean, Asia and Africa. Transport distances (TD) were calculated for the two congeners. The TD was 2300 km for BDE-47 and 2800 km for BDE-99 (EMEP 2004).

Wania and Dugani (2003) examined the long-range transport potential of PBDEs using a number of models – TaPL3-2.10, ELPOS-1.1.1, Chemrange-2, and Globo-POP-1.1 – and various physical and chemical properties – for example, solubility in water, vapour pressure, log K_{OW}, log K_{oa}, log K_{aw}, and estimated half-lives in various media. They found that all models yielded comparable results, with tetrabromodiphenyl ether showing the greatest atmospheric transport potential and decabromodiphenylether the lowest. The researchers estimated a characteristic transport distance (CTD) ranging from 1113 to 2483 km for the tetrabromo, 608 to 1349 for the pentabromo, 525 to 854 for the hexabromo, and 480 to 735 for the decabromo congener. The CTD was defined as the distance a parcel of air has travelled until 1/*e* (approximately 63%) of the chemical has been removed by degradation or deposition processes (Gouin and Mackay 2002).

The EU risk assessment (EU 2000) concluded that the major part of releases end up in soil. From soil, PentaBDE can be expected to be moved mainly through leaching with water in the suspended solids fraction or through wind erosion where it occurs. A small part in the soil can be volatilized, especially in the warm season, and so may be considered a plausible alternative mechanism for transport in addition to volatilization and advective transport of vapor identified in the literature. Although PentaBDE has low water solubility, it has been detected in lakes and seas, and can be transported with water in the soluble and particle phases (Peltola *et al.* 2001). Occurrence in migratory birds and fish indicate the possibility of transport by migration of animals, but the main route seems to be through the atmosphere.

2.2.3.2. Levels in remote areas

The detected levels in the Arctic atmosphere, biota and environment are strong indicators of the PentaBDEs potential for long-range transport (Verreault *et al.* 2005, Verreault *et al.* 2004, Norstrøm *et al.* 2002, Herzke *et al.* 2003, Vorkamp *et al.* 2004a and b, Wolkers *et al.* 2004, Thron *et al.* 2004, Thomas *et al.* 2004, Ikomomou *et al.* 2002, Christensen *et al.* 2002, de Wit *et a*l. 2004, AMAP 2002 and AMAP 2005).

There are several studies showing the occurrence of PentaBDE in remote areas in Europe as well (Vives *et al.* 2004, Hassanin *et al.* 2004 and Zenegg *et al.* 2003). Levels in remote regions are considered to be an indication on long-range transport.

PentaBDE (as total BDE) has been detected in Canadian and Russian Arctic air at concentrations up to 28 pg/m^3 (Alaee *et al.* 2002). Strandberg *et al.* (2001) reported concentrations of total PBDE (BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-190 and BDE-209) in air from the Great Lakes area during the period 1997-1999. Average concentrations based on four samples from each of four locations ranged from 4.4 pg/m^3 near Lake Superior in 1997 to 77 pg/m^3 in Chicago in 1998. The average air concentration of total PBDEs (1997, 1998 and 1999) for the sampling sites ranged from 5.5 to 52 pg/m^3. Tetra- and pentabromo congeners accounted for approximately 90% of the total mass of PBDE in this study. At 20±3°C, about 80% of the tetrabromo congeners and 55-65% of the pentabromo congeners were in the vapour phase while about 70% of the hexabromo congeners were associated with the particulate phase.

A larger study was performed detecting BDEs in trout (three species) from eleven high mountain lakes in Europe (566 to 2,485m altitude) (Vives *et al.*, 2004). These lakes were selected as being far from local pollution emission sources, and it was considered that the only source of BDEs to these lakes was as a result of atmospheric transport and deposition. The major congeners identified (of 39 determined) were BDE-47 and BDE-99, followed by BDE-100, BDE-153, BDE-154 and BDE-28, and these congeners were found in all samples analysed. The highest concentrations of ΣBDE in fish muscle and liver were found in Lochnagar, Scotland, 1.2 and 11 µg/kg wet weight, respectively (177 and 366 µg/kg on a lipid basis). No correlation was observed between the occurrence of these compounds and altitude, latitude or temperature, and the authors inferred that the environmental distribution of the BDEs has not, as yet, reached a steady-state.

2.3 Exposure

2.3.1 Levels

PentaBDE has spread widely in the global environment. A large quantity of monitoring data exist with detected levels in marine and terrestrial birds, sea and terrestrial mammals, sediments, soil, seafood and fish. A global study by Ueno *et al.* (2004) of PentaBDE in skipjack tuna (*Katsuwonus pelamis*) shows a wide spread occurrence in the offshore waters of various regions in the world. Table 2.5 gives an overview over the levels found in different parts of the world.

Contamination of the environment and biota in remote regions can be a threat to vulnerable species and ecosystems. In the Arctic, together with other pollutants of concern, PentaBDE is detected in high levels in top predatory birds and mammals (Verreault *et al.* 2005, Verreault *et al.* 2004, Norstrøm *et al.* 2002, Herzke *et al.* 2003, Vorkamp *et al.* 2004a and b, Wolkers *et al.* 2004, Thron *et al.* 2004, Thomas *et al.* 2004, Ikomomou *et al.* 2002) showing that the Arctic food webs are seriously affected. Wolkers *et al.* (2004) detected levels of PentaBDE in beluga whales (*Delphinapterus leucas*) in the Arctic, a species protected by the Convention on migratory species (the Bonn convention). ΣBDE concentrations (geometric mean; 22 congeners) were 234, 161 and 29 µg/kg in juvenile, adult male and adult female beluga.

In fact, there are detected high levels of PentaBDE in several species, with populations of concern protected by the Bonn convention. Several studies (Jaspers *et al.* 2004, Herzke *et al.* 2005, Lindberg *et al.* 2004, D`Silva *et al.* 2004, Law *et al.* 2005, Sinkkonen *et al.* 2004, Sellström *et al.* 2003, Kannan *et al.* 2005, Ramu *et al.* 2005 and Wolkers *et al.* 2004) indicate that PentaBDE is widespread in peregrine falcon (*Falco peregrine*), merlin (*Falco columbarius*), goshawk (*Accipiter gentiles*), golden eagle (*Aquila chrysaetos*), buzzard (*Buteo buteo)*, beluga whales (*Delphinapterus leucas*), irrawaddy dophins (*Orcaella brevirostris*), and Indo-Pacific humpback dolphin (*Sousa chinensis*), all protected by the Bonn convention. High levels of PBDEs are also detected in peregrine falcon eggs in Sweden (Lindberg *et al.* 2004), for which individual ΣBDE concentrations were as high as 39,000 μg kg^{-1} lipid weight, some of the highest concentrations seen in wildlife so far.

The populations of harbour porpoises (*Phocoena phocoena*) in the North and Baltic seas are protected through the Bonn Convention. Studies have detected high levels in those populations (Thron *et al.* 2004 and Covaci *et al.* 2002). In a study by Thron *et al.* (2004) animals with poor body condition (lower mean blubber thickness) had much higher concentrations than other individuals. Only females showed decreasing concentrations with age, indicating elimination via transfer from mother to offspring.

The harbour porpoise is, together with peregrine falcon and merlin, also on the list for strictly protected (endangered) species in the convention on the conservation of European wildlife and natural habitats (the Bern Convention). The white-tale sea eagle is on the list for endangered species in the Bern Convention. Levels of concern are detected in both individuals and eggs (Herzke *et al.* 2005). Beluga whales and irrawaddy dolphins are on list for protected (vulnerable) species. High levels are found in white-beaked dolphin (*Lagenorhyncus albirostris*), another endangered species. The parties of this convention undertake to take appropriate measures to ensure the conservation of endangered and vulnerable species and their habitats.

*Table 2.5 Levels of PentaBDE (**BDE-99**) in different parts of the world (LW=Lipid weight, DW=Dry weight).*

Country/Region	**Organism/compartment**	**Levels of PentaBDE**	**References**	**Comments**
Europe	Atmosphere Gas phase	10-120 pg/m^3	Jaward *et al.* 2004	22 countries
Japan	Atmosphere Particulate Gas phase	0.05-0.9 pg/m^3 0.05-19’ pg/m3	Hayakawa *et al.* 2004	Measured in the summer
Sweden	Sediments	<0.7-51.4 ng/g DW	Palm *et al.* 2002	Rivers at point source
United Kingdom	Soil	78 – 3200 pg/g DW	Hassanin *et al.* 2004	
Western Europe	Sediments	<0.2-6.9 ng/g DW	Palm *et al.* 2002	Estuaries
Japan, Osaka	Sediments	9-28 ng/g DW	Palm *et al.* 2002	
North Pacific Ocean	Skipjack tuna	0.18-2.1 ng/g LW	Ueno *et al.* 2005	
Japan	Skipjack tuna	1.1-1.7 ng/g LW	Ueno *et al.* 2005	Offshore waters
East China Sea	Skipjack tuna	2.4-4.7 ng/g LW	Ueno *et al.* 2005	
Philippines	Skipjack tuna	2.1 ng/g LW	Ueno *et al.* 2005	Offshore waters
Brazil	Skipjack tuna	1.9 ng/g LW	Ueno *et al.* 2005	Offshore waters
Canada	Atlantic tomcod	77 ng/g LW	Law *et al.* 2003	
Chilika Lake, India	Irrawaddy dolphin	0.12-0.78 ng/g LW	Kannan *et al.* 2005	Endangered species
Hong Kong, China	Indo-Pacific humpback dolphin	33.6-720 ng/g LW	Ramu *et al.* 2005	Coastal waters 12% of ΣPBDEs
United Kingdom	White beaked dolphin	1480 ng/g LW	Law *et al.* 2003	Endangered species
Hong Kong, China	Finless porpoises	27.6-117.6 ng/g LW	Ramu *et al.* 2005	Coastal waters 12% of ΣPBDEs
Japan	Northern fur seal	2.64-4.56 ng/g LW	Kajiwara *et al.* 2004	Pacific coast 12% of ΣPBDEs

Svalbard, Arctic Norway	Polar bear	0.7-4.7 ng/g LW	Gabrielsen *et al.* 2004	
Canadian Arctic	Polar bear	1.04-11.3 ng/g LW	Muir *et al.* 2006	
Bjørnøya, Arctic Norway	Glacous gulls	0-7.9 ng/g LW	Herzke *et al.* 2003	
Norway	White-tailed sea eagle	6-184 ng/g LW	Herzke *et al.* 2005	In eggs. Endangered Species
Sweden	Peregrine falcons	110-9200 ng/g LW	Lindberg *et al.* 2004	Endangered species
Australia	Melon-headed whale	4.8 ng/g LW	Law *et al.* 2003	
Canada	Beluga whale	108 ng/g LW	Law *et al.* 2003	Vulnerable species
Netherlands	Mussels	0.3-11 ng/g LW	Law *et al.* 2003	Marine+freshwater
Sweden	Frog	5.6 ng/g LW	De Wit *et al.* 2004	
Canada	Zooplankton	0.46 ng/g LW	Law *et al.* 2003	

2.3.2 Trends

Most trend analysis show an increase in concentrations of PBDEs in the environment and in humans from the beginning of the 1970s, with a peak around the mid-1990s and a stabilisation or subsequent levelling off in Europe (Covaci *et al.* 2002, Fängström *et al.* 2005, Thomsen *et al.* 2005 and Knudsen *et al.* 2005), but with a continuous increase in the Arctic (Vorkamp *et al.* 2005, AMAP 2002 and AMAP 2005). PentaBDEs are reported in the studies to follow the same trend as ΣPBDEs. This increase has also been seen in North America, in air, soil and sediment, and wildlife, but insufficient data exist to allow comment on trends in the human population.

In the Asia-Pacific region a study on northern fur seals on the Pacific coast of Japan shows an increase of PBDEs to about 150 times between 1972 and 1994, and then levels decreased to about 50% in 1998 (Kajiwara *et al.* 2004). The reduction in PBDEs values was assumed to be due to the voluntary phase out of C-PentaBDE in Japan in 1990. BDE-99 levels showed the same pattern as ΣPBDEs.

Analysis of archived herring gull eggs (sampled in 1981, 1983, 1987, 1989, 1990, 1992, 1993, 1996, 1998, 1999 and 2000) enabled Norstrom *et al.* (2002) to establish temporal trends in PBDE concentrations over the period 1981-2000. At Lake Michigan, Lake Huron and Lake Ontario sampling sites, concentrations of tetra- and pentabromodiphenyl ethers (that is, BDE-47, BDE-99 and BDE-100) increased by 71-112-fold over these two decades (from 4.7 to 400.5 µg/kg ww at Lake Ontario; from 8.3 to 927.3 µg/kg ww at Lake Michigan; from 7.6 to 541.5 µg/kg ww at Lake Huron). These increases were found to be exponential at all three locations ($r^2 = 0.903 – 0.964$, $p< 0.00001$).

Wakeford *et al.* (2002) undertook sampling of eggs of the great blue heron in 1983, 1987, 1991, 1996, 1998 and 2000 in southern British Columbia and found that total PBDE concentrations (sum of tetra-, penta- and hexabromo-congeners) increased from 1.31 to 287 µg/kg ww between 1983 and 1996, but then dropped slightly to 193 µg/kg ww in 2000. They also undertook sampling of the eggs of thick billed murre in the Canadian North in 1975, 1987, 1993 and 1998, and observed a trend of gradually increasing PBDE concentrations (sum of tetra-, penta- and hexabromo-congeners) in these eggs from 0.43-0.89 µg/kg ww in 1975, to 1.83-3.06 µg/kg ww in 1998.

PBDEs have been detected in a variety of marine mammals. Alaee *et al.* (1999) reported average PBDE (di-to hexaBDE) concentrations in the blubber of marine mammals from the Canadian Arctic as 25.8 µg/kg lipid in female ringed seals (*Phoca hispida*), 50.0 µg/kg lipid in male ringed seals, 81.2 µg/kg lipid in female beluga (*Delphinapterus leucus*) and 160 µg/kg lipid in male beluga. BDE-47, a tetrabromodiphenyl ether, was the predominant congener, followed by the pentabromo BDE-99. Ikonomou *et al.* (2000, 2000b) reported PBDE concentrations in biota samples from the west coast and Northwest Territories of Canada. The highest concentration of of total PBDE residues, 2269 µg/kg lipid, was found in the blubber of a harbour porpoise form the Vancouver area. With a concentration of about

1200 µg/kg, one congener, BDE-47, accounted for slightly more than half of the total PBDE in the sample. Ikonomou *et al.* (2002a) analyzed temporal trends in Arctic marine mammals by measuring PBDE levels in the blubber of Arctic male ringed seals over the period 1981-2000. The mean total concentrations increased exponentially, from 0.572 µg/kg lipid in 1981 to 4.622 µg/kg in 2000, a greater than eightfold increase. They determined that Penta- and HexaBDEs are increasing at approximately the same rate (doubling time 4.7 and 4.3 years, respectively), more rapidly than TetraBDEs, for which the doubling time was 8.6 years. Once again, BDE-47 was predominant, followed by BDE-99 and BDE-100.

A marked increase in tissue PBDE levels was also evident in blubber samples collected from San Francisco Bay harbour seals over the period 1989 to 1998 (She *et al.* 2002). Total PBDEs (the sum of BDEs 47, 99, 100, 153 and 154) rose from 88 µg/kg lipid to a maximum of 8325 µg/kg lipid over this short period. Stern and Ikonomou (2000) examined PBDE levels in the blubber of make SE Baffin Bay beluga whales over the period 1982-1997, and found that the levels of total PBDEs (tri-to hexa-congeners) increased significantly, Mean total PBDE concentrations were about 2 µg/kg lipid in 1982, and reached a maximum value of about 15 µg/kg lipid in 1997. BDE-47 was the dominant congener, with a mean concentration of approximately 10 µg/kg lipid in 1997. Total PBDE residues (concentrations for individual congeners not provided) in the blubber of St Lawrence estuary belugas sampled in 1997-1999 amounted to 466 (±230) µg/kg ww blubber in adult males, and 655 (±457) µg/kg ww blubber in adult females. These values were approximately twenty times higher than concentrations in beluga samples collected in 1988-1990 (Lebeuf *et al.* 2001).

The results from a modelling exercise utilizing the European variant (EVn) BETR multimedia environmental fate model were presented for the C-PentaBDE product by Prevedouros *et al.* (2004). To predict future atmospheric concentration trends, the model was used in its fully dynamic mode over the period 1970-2010. It predicted that atmospheric concentrations would have peaked around 1997, and then declined with an overall "disappearance" half-life of 4.8 years. The model steady state simulations gave generally good agreement with measured data for BDE- 47 and BDE-99. The empirical data for North America presented above, however, show continuing increases in concentrations, at least up the year 2000, and so while the model results match some European data with fair agreement, they are not in accord with data from North America.

Three dated sediment cores from locations in Western Europe were analyzed for 14 BDE congeners (Zegers *et al.*, 2003). Cores from the Drammenfjord (Norway), the western Wadden Sea (The Netherlands) and Lake Woserin (Germany) showed a time dependent pattern in the distribution of BDEs since the start of production of PBDE formulations. Two of the three commercial formulations could be distinguished. The penta-mix formulation is clearly present from the beginning of the 1970s. This is in agreement with data for the industrial production of this formulation. In the cores from the Netherlands and Germany, concentrations of BDE congeners associated with the C-PentaBDE were levelling off in the most recent layers (1995 & 1997), whereas those in the Drammenfjord were still increasing in 1999. The absence of all BDE congeners in the older (deeper) layers of all three cores, as well as in several 100 to 150 million year old layers of clay from Kimmeridge, UK, indicated that these BDE congeners are not produced naturally.

Human exposure to polychlorobiphenyls and PBDEs in Japan in 1980 and 1995 showed that levels of the latter had increased substantially over the twenty-year period, although there was great variation between regions. The main congeners detected in serum were BDE-47 and BDE-99. Most total PBDE levels had more than doubled, and in one area increased twenty-fold, with 1995 values falling in the range 0.6 – 41.4 ng/g lipid Koizumi *et al.* 2006).

2.3.3. Bioavailability

Environmental studies on bioavailability have detected uptake of PentaBDE in soil organisms (Matscheko *et al.* 2002), sediment dwelling organisms (Magnusson *et al.* 2003) and aquatic organisms (Lithner *et al.* 2003, Voorspoels *et al.* 2003, Marsch *et al.* 2004, Kierkegaard *et al.* 2004, and Sinkkonen *et al.* 2004), making PentaBDE's way into the food webs evident. Subsequent bioaccumulation and biomagnification of the compound has been detected and described in Section 2.2.2.

Soil exposed to PBDEs in various ways was analyzed for BDE-47, BDE-66, BDE-99, BDE-100, BDE-153, BDE-154 and BDE-183 (Matscheko *et al.*, 2002). Earthworms collected at all soil sampling sites were analyzed as well. The BDE congener profile in all soil samples was dominated by BDE-47 and BDE-99. Accumulation of the compounds in earthworms from the sites yielded a direct relationship between the concentrations in the soil and concentrations in the worms. The biota-soil accumulation factors (BSAFs) of BDE congeners BDE-47, BDE-99 and BDE-100 were around 5 (organic matter/lipids). Thus, earthworms living in contaminated soils will accumulate tissue BDE concentrations and, as these animals represent the base of the terrestrial food chain for many organisms, this form a pathway for the accumulation of BDEs in organisms at higher trophic levels.

The western Scheldt estuary is subject to a variety of suspected PBDE sources, such as a brominated flame retardant manufacturing plant, Antwerp harbour, and the textile industry located further upstream. PBDE concentrations in samples of biota, including crab, shrimp, starfish, benthic fish (such as dab, goby, plaice and sole) and gadoid fish (such as bib and whiting) from the estuary were compared to those in samples from the Belgian North Sea beyond the mouth of the estuary (Voorspoels *et al.*, 2003). Eight BDE congeners (BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209) were determined. Concentrations observed in the estuarine samples were up to 30 times higher than in those from the Belgian North Sea, with an increasing gradient towards Antwerp. Concentrations in the North Sea ranged from 0.02 to 1.5 µg/kg wet weight in benthic invertebrates and goby, from 0.06 to 0.94 µg/kg wet weight in fish muscle, and from 0.84 to 128 µg/kg wet weight in fish liver. The corresponding ranges in samples from the estuary were from 0.2 to 30, 0.08 to 6.9, and from 15 to 984 µg/kg wet weight, respectively. The ratio BDE-99/BDE-100 was found to be highly location- and species-dependent, possibly relating to differences in metabolism. In shrimp, the value of this ratio (4:1) was very similar to that observed in the Bromkal formulation and in estuarine sediment, and was similar in shrimp from both the North Sea and the estuary, implying both that these congeners are readily bioavailable and that shrimp lack the ability to metabolize either congener. On a lipid weight basis, concentrations of BDE-47 ranged from 3 to 108 µg/kg lipid weight in samples from the North Sea, and from 8 to 1,550 µg/kg lipid weight in estuarine samples. BDE-47 was the most abundant congener in all samples, comprising 43 to 75% of ΣBDE.

Thomas *et al.* (2004) conducted an input-output balance study of BDEs on three captive, juvenile grey seals. The animals were fed a diet of herring for six months, and the study was performed during the last three months of this period. BDE analysis was undertaken using GC-ECNIMS. Consistently high absorption (89 - 99%) was observed for all PBDE congeners studied (BDE-28, BDE-47, BDE-49, BDE-99, BDE-100, BDE-153, BDE-154 and BDE-209).

2.3.4 Human exposure

Studies, assessments and reviews referred to in this section have shown that the main routes for human exposure are food, and exposure to dust in indoor air at home and workplaces due to levels in products like furniture and electronic devices. Fish and agriculture products are the main food sources of PentaBDE for humans, and mother's milk for the nursing child. Fatty fish from contaminated areas are a major source (Sjödin *et al.* 2003). PentaBDE has been detected in various foods (VKM 2005, Burniston *et al.* 2003 and Bocio *et al.* 2003) as well as in indoor dust (Shoeib *et al.* 2004 and Wilford *et al.* 2005). Levels in foods in the US have been reported by Schecter *et al.* (2004), Schecter *et al* 2006, and Huwe *et al.* (2005). There are several hazard assessments in EU and US, looking into the exposure of humans

(VCCEP 2003, COT 2004, VKM 2005). They conclude that the available hazard or exposure information is inadequate to fully characterize the risks.

About 5% of the individuals in general populations have been found to be subjected to elevated exposure (Thomsen *et al.* 2005 b). This, together with estimates of the half life of C-PentaBDE congeners in humans, raises concern for long-term effects on human health. The half-lives for these congeners in humans have been estimated to be 1,040 days (BDE-99) and 573 days (BDE-100) (Geyer *et al.* 2004).

Domestic house dust is likely to be a significant source where furniture, carpet or appliances contain C-PentaBDE. This has been discussed in Section 2.1.1. It is not clear which sources are the greatest, and there could be wide variations depending on lifestyle and diet.

Several studies have detected levels of PentaBDE in sewage sludge (Matscheko *et al.* 2002, Fabrellas *et al.* 2004, Motche and Tanner 2004 and Sjödin *et al.* 2003, Hale 2002). Sewage sludge is considered to be one of the main sinks for PBDEs. The application of sewage sludge to agricultural land is one of the reasons for detected levels of PentaBDE in food products. This can explain the detected levels in vegetables and root crops in experimental studies. Levels in fish and root crops can be the source of exposure to domestic animals like chickens and pigs, and the source of PBDEs in meat products for human nourishment.

A Canadian global study showed that PentaBDE is widespread in human milk in populations all over the world (Ryan 2004). There are data on levels in human blood serum and milk from USA, Canada, Mexico, Japan, the EU region, the Arctic region and Scandinavia. A meta-analysis by Hites (2004), using data published up to mid-2003, showed that serum and milk levels in the US were much higher than those in Europe - ~35 ng/g *vs* ~ 2 ng/g lipid – and were doubling on average every 4-6 years. BDE-47 and BDE-99 were the major congeners detected. Considerably higher levels are found in humans from North America in general. About 5% of general populations have been found to be subjected to elevated exposure. Thus, together with estimates of the half-life of PentaBDE congeners in humans, raiss concern for long-term effects on human health (Thomsen *et al.* 2005b).

Levels increasing from the 1980s to the 2000s have been observed in mother's milk from Sweden as well as in blood from Germany and Norway (Sjödin *et al.* 2003). A more recent study in Sweden (Fängström *et al.* 2005) assessed the temporal trends of polybrominated diphenyl ethers (PBDEs), in mothers' milk in the Stockholm area. The pooled samples were covering the time period 1980 to 2004, with emphasis on samples from the last ten years. Concentrations of BDE-47, BDE-99 and BDE-100 reached a peak in the mid-1990s and are now clearly showing decreasing levels. The concentrations are however still much higher than in 1980.

The objective of a recent Norwegian study was to complete and extend a previous study on time trends of PBDEs in Norwegian pooled serum samples (Thomsen *et al.* 2005a) and put together an overview of the PBDE body burden in the general population from 1977 to 2004. The temporal trend of the sum of seven PBDEs (BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154 and BDE-183) in the pooled serum from the present study are in close agreement with the levels found in a previous study by the same authors. In general, for similar time periods the levels in breast milk seem to be somewhat lower than in the serum, but the same overall trend is observed. This confirms that the PBDE body burdens in these regions have risen rapidly from 1977 to about 1997, but now seem to have stabilized or even to have decreased. This is in accordance with the trends observed in Swedish breast milk, as an indicator of the European situation, but may not be true of levels in North America. The PBDE level was previously found to be about twice as high in a serum pool from infants up to four years of age compared to serum pools from elderly persons. This finding was confirmed in the Norwegian study. However, in 2002, children between the ages of 5 and 14 years showed higher levels of PBDEs than the average adult.

Contemporary PBDE concentrations in Europe and Asia are remarkably similar, with low median values on a lipid basis for all countries and relatively small variations. The situation in North America is completely different with median values for individual studies in the range of 20-50 ng/g LW (Ryan 2004). However, in parallel with the regional differences that were reported above for biota, the levels in breast adipose tissue taken from women living in San Francisco Bay area in 2000 were almost two orders of magnitude higher than what has been reported in human milk from Sweden (Sjödin *et al.* 2003). A more recent study of levels in human adipose tissue in New York was published by Johnson-Restrepo *et al.* (2005). The study of 40 males and 12 females of a range of ages and ethnicities showed wide variations in lipid PBDE concentrations, with mean values substantially higher than the medians. Median concentrations were: BDE-47, 29.3 ng/g lipid; BDE-99, 10.3 ng/g lipid; BDE-100, 12.0 ng/g lipid.

In a preliminary screening of PBDEs in plasma and milk samples from Mexican women, the levels were well above European levels of PBDEs reported so far (López *et al.* 2004). The mean level of PBDEs (with BDE-209 excluded) in Mexican women living in urban areas was approx. 20 ng/g LW in plasma. The levels in women living in rural areas in Mexico were however comparable with women living in rural areas in Sweden. (BDE-209 levels were only detected in women living in the Mexican city).

Ryan (2004) detected a big individual variation in levels in the general population in a study from Canada. The values span more than three orders of magnitude, with a few values showing a much greater level. Levels detected in the Canadian Arctic in Ryan's study (2004) were increasing. Values in human milk from the Faroe Islands showed the same trend (Fängström *et al.* 2004).

Two studies in Australia indicated that levels of PBDEs in Australian breast milk and blood serum are higher than those in Europe but lower than those found in North America (Harden *et al.* 2004 and 2005).

*Table 2.6 Data on mean levels of PentaBDE (**BDE-99**) (ng/g LW) in humans from different parts of the world.*

Data	Country/region	Levels	References	Year	Comments
blood	The Netherlands	0.8	Weiss *et al.* 2004	unknown	
blood	Norway	1.0	Thomsen *et al.* 2004	1999	
blood	Mexico	2.0	López *et al.* 2004	2003	Urban population
blood	Australia	2.3	Harden *et al.* 2004	2003	
milk	Germany	0.2	Harden *et al.* 2004	2000	
milk	Sweden	0.3	Fängström *et al.* 2005	2003	Urban population
milk	Mexico	0.6	López *et al.* 2004	2003	Rural population
milk	Sweden	0.5	López *et al.* 2004	2003	Rural population
milk	United Kingdom	0.9	Harden *et al.* 2004	?	median
milk	Faroe Islands	1.0	Fängström *et al.* 2004	1999	Rural population
milk	Australia	1.9	Harden et al. 2005	2002/2003	
milk	Canada	4	Ryan *et al.* 2002	2002	Rural population
milk	USA	28	Päpke *et al.* 2001	2000	Urban population

Although they are less relevant that environmental data, results from occupational studies bear out the facility with which the PBDEs are taken up by human bodies. In Sweden, occupational exposure to PBDE has been identified among electronics recycling personnel (Sjødin *et al.*, 1999) and in technicians responsible for repair and maintenance of computers (Jacobsson *et al.,* 2002) as well as in nearby soil and sediment (Wang *et al.* 2005). Also workers in industry manufacturing C-PentaBDE, or polyurethane foam and electronic equipment containing it can be exposed to PentaBDE. There is an extensive literature on such exposures.

2.3.5 Debromination

There is growing interest in the fate of PBDEs in the environment. In experiments reported by Stapleton *et al.* (2004), carp were fed food spiked with individual BDE congeners for 62 days, and tissue and excreta were examined. At least 9.5±0.8% of BDE-99 in the gut was reductively debrominated to BDE-47 (one less bromine) and assimilated in carp tissues. Similarly, 17% of the heptabromo congener BDE-183 was reductively debrominated to hexabromo congeners. The authors noted that body burdens of PBDEs may thus reflect direct uptake from exposure as well as debromination of more highly brominated congeners. Highly selective reductive microbial debrominations were observed in experiments reported by He *et al.* (2006). Hepta- and Octa-BDEs were produced in cultures of *Sulfurospirillum multivorans* to which DecaBDE had been added, but OctaBDE was not attacked in a similar system. Cultures of an alternative organism, *Dehalococcoides sp.*, failed to attack the DecaBDE but an OctaDBE mixture was extensively changed, yielding a mixture of Hepta- through Di-BDEs which included the PentaBDE, BDE-99. The authors draw attention to the potential for conversion of higher congeners in the environment to more toxic congeners with fewer bromine substituents. Further studies particularly environmental monitoring studies focussing on congeners for which the primary source is likely to be debromination reactions, are required to clarify the role of debromination in determining the final mix of PBDE congeners in the environment.

Hydroxylated BDEs (OH-BDEs) have been detected and identified as metabolites in several species after exposure to specific BDE congeners but have also been found to occur as natural products in marine sponges and ascidians (Marsch *et al.* 2004). Methoxylated BDEs (MeO-BDEs) have also been reported as natural products present in marine sponges and green algae. It would seem that the origin of these substances can be natural, anthropogenic or both. Nine OH-BDEs and six MeO-BDEs were identified in blood of Baltic Sea salmon (*Salmo salar*) using newly synthesized standards (Marsch *et al.*, 2004). All of the identified OH- and MeO-BDEs were substituted with four or five bromine atoms and five of them also had one chlorine substituent. Fourteen have the methoxy or hydroxy group substituted in the position *ortho*-to the diphenyl ether bond. The structures of several of the compounds support natural rather than anthropogenic origins. However, at least one of the OH-BDEs (4’-OH-BDE-49) may be a hydroxylated metabolite of BDE-47. Estrogenic activity of some hydroxylated PBDEs has been reported by Meerts *et al.* (2001).

Other studies of metabolism of PBDEs are summarized in Section 2.2.2.1.

2.4 Hazard assessment for endpoints of concern

Evidence to date suggests that the major congeners of the C-PentaBDE formulation, BDE-47 and BDE-99, are likely to be more toxic and bioaccumulative than other PBDE congeners. Although the toxicology of PBDEs is not completely understood, some studies on PentaBDE have demonstrated reproductive toxicity, neurodevelopmental toxicity and effects on thyroid hormones. The neurotoxic effects of PBDEs are similar to those observed for PCBs and so children exposed to PBDEs are likely to be prone to subtle but measurable developmental problems. It is presumed that PBDEs are endocrine disrupters, but research results in this area are scant (Siddiqi *et al.* 2003).

While further studies follow internationally-accepted guidelines might be needed to make a full risk assessment of the situations of children, there are sufficient data for development of the present risk profile.

It is acknowledged that these conclusions rest to some extent on examination of reviews, rather than reanalysis of primary data, but in general the studies under review have followed internationally accepted experimental protocols. Nonetheless, there is no significant disagreement between some reported results and later analyses, such as that of the US Voluntary Children's Chemical Evaluation Program (VCCEP) (2005).

2.4.1 Ecotoxicity

Recent studies show that exposure to BDE-47 can cause growth inhibition in colonies of the plankton algae (*Skeletonema costatum*) and a depression on reproductive output of the zooplankton *Daphnia magna* (Källqvist *et al.* 2006).

A recent paper by Timme-Laragy *et al.* (2006) showed adverse effects on fish development at low concentrations. However, the endpoints that were affected in this report (behavioural learning) are not usually accepted risk assessment endpoints. Other endpoints that would be acceptable, such as growth or survival, were not affected.

Canada was able to perform a risk quotient analysis for each congener, integrating known or potential exposures with known or potential adverse effects. In its simplest form, the risk quotient may be described by the equation:

$$\text{Risk quotient} = \frac{\text{exposure reference value}}{\text{toxicity reference value}}$$

and it is customary to use conservative values in order to highlight the worst case.

Exposures were estimated local to emission sources including areas receiving urban drainage (wildlife consumers) and downstream of a polymer processing facility (benthic organisms). Adjustment factots of 100-1000-fold were applied to critical toxicity values to reflect extrapolation from laboratory to fielod conditions, intraspecies and interspecies variations in sensitivity, and because compounds are bioaccumulative and persistent.

A risk quotient value >1 signifies the likelihood or potential for adverse effects to occur, while those <1 imply no danger to organisms. The Canadian results shown in Table 3.1 are based partly on Canadian empirical data and partly on surrogate data from Swedish and US sources.

Table 3.1 Risk quotient values for PentaBDE (Environment Canada 2006, Canadian Wildlife Table 8).

Commercial Product	**Pelagic organisms**	**Benthic organisms**	**Soil organisms**	**Wildlife consumers**
C-PentaBDE	4×10^{-3}	45.2	0.13-0.26	149

These values reflect the bioaccumulation of PentaBDE which causes organisms higher in the food chain to be exposed to greater risk.

2.4.2 Effects in mammals

In a review article on toxic effects of brominated flame retardants, Darnerud (2003) drew on a range of primary literature to conclude that exposure to PBDEs gives rise to adverse effects in experimental *in vivo* models, and depending on type of product different effects are seen, occurring at varying dose levels. Generally, the C-PentaBDE products cause effects at the lower dosages. The critical effects of PentaBDE are those on neurobehavioral development and, although somewhat less sensitive, thyroid hormones in offspring (from 0.6 to 0.8 and 6 to 10 mg/kg body wt., respectively) (Darnerud 2003). Note that some data reported in Table 2.7 show levels below these. More recent information, especially for North America, is available in Birnbaum and Staskal (2004).

Blubber biopsy and blood samples were collected from weaned grey seal (*Halichoerus grypus*) pups and juveniles during 1998 and 1999 (Hall *et al.*, 2003). Fifty four post-weaned pups and fifty five first year juveniles (of which thirteen were recaptured post- weaned pups) were studied. The median concentrations of ΣBDE (14 congeners) were 0.17 and 0.46 µg/kg lipid weight in the blubber of the pups and the juveniles, respectively. The study indicated that thyroid hormone levels in the blood of grey seals during their first year of life were significantly, and positively, related to ΣBDE concentrations in blubber, after accounting for the effects of possible confounding variables. Such an association is not, in itself, sufficient evidence for a causal relationship, but is in accordance with the hypothesis that these compounds can act as endocrine disrupters in grey seal pups.

Darnerud (2003) concluded in his review that for PentaBDEs, the critical effects among the available studies seem to be developmental neurotoxicity and, although generally at somewhat higher doses, altered thyroid hormone homeostasis. Regarding the neurotoxicity in mice, no clear mechanism could be defined but effects of the PentaBDEs both via thyroid hormone disruption and directly on signal transmission in brain have been discussed. For example, a number of PBDEs were capable of inducing cell death of cerebellar granule cells in culture (Reistad *et al.*, 2002, Reistad and Mariussen 2005). The LOAEL value for PentaBDE could be set to 0.6–0.8 mg/kg body wt., based on the most sensitive effect observed, neurobehavioral effects during early development (Darnerud 2003, although it is not the task of the POPRC to set a regulatory level, for construction of which resort would need to be made a wider range of data.

In a hazard assessment by the Committee on Food Safety in Norway (VKM 2005) the following toxic effects of exposure to BDE-99 or the C-PentaBDE formulation was reported: neurotoxicity, effects on neurobehavioral development, effects on the thyroid hormone system and hispatological alterations in the tyroid and liver.

Table 2.7 Overview of No Observed Effect level (NOEL) and Lowest Observed Effect Level (LOEL) after oral administration of ***BDE-99*** *congener or C-PentaBDE formulations. Bold values are the lowest LOEL or NOEL detected.**

PentaBDE	**Duration**	**Dose**	**NOEL mg/kg/ day**	**LOEL mg/kg/day**	**Endpoint**	**Species**	**Reference**
BDE-99	s.d	0.8 or 12.0 mg/kg	n.d.	0.8	Neurotoxicity Behaviour, motor activity level and learning	mouse	Eriksson *et al.* 2001
BDE-99	s.d	0.6, 6, or 30 mg/kg	n.d.	0.6	Developmental- and neurotoxicity Behaviour - hypoactive	mouse	Branchi *et al.* 2002
BDE-99	s.d	0.4, 0.8, 4.0, 8.0, or 16 mg/kg	**0.4**	**0.8**	Developmental- and neurotoxicity Behaviour	mouse	Viberg *et al.* 2004 Sand *et al.* 2004
BDE-99	s.d.	0,06 and 0,3 mg/kg to pregnant female	**n.d.**	**0,06**	Developmental- and neurotoxicity Behaviour (increased activity)	rat, F1 gen.	Kuriyama *et al.* 2005
BDE-99	s.d.	0,06 and 0,3 mg/kg to pregnant female	**0,06**	**0,3**	Reduced testis size and number of sperms	rat, F1 gen.	Kuriyama *et al.* 2005

Penta mix DE-71	30 d	0.01, 0.05, 0.1, 0.5, or 1.0 mg/kg/day	**1**	n.d.	Growth, food intake, hematology, histopatology Clinical chemistry	rat	Great lakes Chemical Corporation 1985
Penta mix DE-71	30 d	0, 3, 30, or 60 mg/kg/day	3	30	Liver weight, puberty, reproduction, liver enzymes, T_4-reduction	Male rat	Stoker *et al.* 2004
Penta mix DE-71	30 d	0, 3, 30, or 60 mg/kg/day	n.d.	3	T_4-reduction	Female rat	Stoker *et al.* 2004
Penta mix DE-71	35 d	0, 1, 10 or 30 mg/kg/day	1	10	T_4-reduction Liver enzymes	pregnant rat	Zhou *et al.* 2002, Zhou *et al.* 2001
Penta mix DE-71	90 d	0-0.44 mg/kg/day	n.d.	0.44	Liver enzymes	rat	Carlson 1980
Penta mix DE-71	90 d	0, 2,10, or 100 mg/kg/day	0-2	2-10	Hepatocyto-megali Tyreoidea hyperplasi	rat	Great lakes Chemical Corporation 1984

n.d. = not defined, s.d. = single dose

* Most of the studies are in line with the OECD test guidelines and for those are not, the quality of the study is assessed to be adequate.

The PBDE mixture known as DE-71 (71% bromine by mass, and containing BDE-47, BDE-99, BDE-100, BDE-153, BDE-154) delays the puberty and suppresses the growth of androgen-dependent tissues in male Wistar rat following a peri-pubertal exposure. These effects suggest that DE-71 may be either inducing steroid hormone metabolism or acting as an androgen receptor (AR) antagonist (Stoker *et al.* 2005).

Talsness *et al.* (2005) evaluated the effects of environmentally relevant concentrations (low doses) of BDE-99 on the female reproductive system in rats. Ultra structural changes compatible with altered mitochondrial morphology were observed in the ovaries of the F1 offspring. No statistically significant changes in ovarian follicle counts were observed. External and skeletal anomalies were detected in offspring (F2) from two different dams (F1) with early developmental exposure to 300 µg BDE-99/Ikg BW. Exposure to BDE-99 resulted in female reproductive tract changes in the F1 generation which were apparent at adulthood.

In utero exposure to a single low dose of BDE-99 disrupts neurobehavioral development and causes permanent effects on the rat male reproductive system apparent in adulthood (Kuriyama *et al.* 2005). Also in this study, the effects of developmental exposure to BDE-99 on juvenile basal motor activity levels and adult male reproductive health were assessed. The exposure to low-dose BDE-99 during development caused hyperactivity in the offspring at both time points (postnatal days 36 and 71) and permanently impaired spermatogenesis by the means of reduced sperm and spermatid counts. The doses used in this study of 60 and 300 µg/kg BW are relevant to human exposure levels, being approximately 6 and 29 times, respectively, higher than the highest level reported in human breast adipose tissue. This is the lowest dose of PBDE reported to date to have an *in vivo* toxic effect in rodents and supports the premise that low-dose studies should be encouraged for hazard identification of persistent environmental pollutants. The study by Viberg *et al.* (2004) shows that neonatal exposure to BDE-99 can induce developmental neurotoxic effects, such as changes in spontaneous behaviour (hyperactivity), effects that are dose-response related and worsen with age. The changes are seen in C57/B1 mice of both sexes. Spontaneous behaviour (locomotion, rearing, and total activity) was observed in two-, five- and eight-month-old mice.

2.4.3 Toxicity to humans

Several hazard assessments have been produced in EU and in US. The conclusions in the hazard assessments elaborated are qualified by the lack of sufficient knowledge of the toxicology of PentaBDE to enable assessment of the risk to humans (COT 2004, VKM 2005 and VCCEP 2003). The toxicological importance for humans of detected effects in laboratory animals is not clear. There is still not enough knowledge of the mechanisms, half-life and metabolism of PentaBDE in experimental animals and humans (VKM 2005).

The conclusion in the hazard assessment by the Committee on Food Safety in Norway was that the exposure through food and mother's milk is considerably lower than the observed NOEL in laboratory mammals (VKM 2005). It is believed that long-time exposure to lower doses of PentaBDE can cause health effects, since PentaBDE accumulates in the human body. Since the half-life of PentaBDE in humans is not known it is not possible today to conclude on long-time exposure effects. This is true even for the US situation, where levels may be 10-20 times those observed in Europe, but pharmacokinetics, toxicology, exposure and other critical data are lacking.

Vulnerable groups could however be pregnant women, embryos and infants, because of effects on the thyroid hormone balance, and the embryo's development of the central nervous system. During pregnancy, maintenance of the thyroid hormone balance is a physiological challenge. Embryos and infants are particularly vulnerable for reductions in thyroid hormone levels (VKM 2005). Infants are exposed to PentaBDE through the diets of their mothers’ milk, since PentaBDE is lipophilic and accumulates in the milk (VKM 2005).

3. Synthesis of information

3.1 Summary

PentaBDE meets all of the Annex D screening criteria, and details are included (for the sake of completeness) in Table 3.2, below.

In the absence of production controls, the levels detected in humans, other species and the environment have been observed to rise steeply and this increase is observed in remote locations as well as closer to sites of production and use. In the US, where C-PentaBDE was in high use until recently and where it remains in such materials as polyurethane foam incorporated into consumer products, there has been a build-up in human tissue.

PentaBDE in soil or sediment is readily incorporated into the food chain and bioaccumulates in the fatty tissues of top predators, including humans.

There are toxicological studies of concern that demonstrate neurodevelopmental impacts in animals at low tissue levels that are of relevance to levels observed in populations. Such body burdens remain under close review.

An assessment of the impact of PBDEs on the environment was recently concluded by Environment Canada (2006), taking into account critical studies and lines of evidence that support the conclusion that these commercial substances entering the environment have or may have an immediate or long-term harmful effect on the environment or its biodiversity.

4. Concluding statement

Pentabromodiphenyl ether (C-PentaBDE) is a synthetic mixture of anthropogenic origin with no known natural occurrence. It can be concluded therefore that the presence of components of PFOS in the environment is the result of anthropogenic activities. Long range transport must be responsible for its presence in areas such as the Arctic region, remote from sites of production and release. PentaBDE degrades slowly in the environmernt and can bioaccumulate and biomagnify in mammals and piscivorous birds.

The phase out of C-PentaBDE production and use has led to a reduction in current use but many materials in use, such as polyurethane foams and plastics in electronic equipment, contain PentaBDE which is slowly released to the environment. This release will be accelerated at end-of-life of such materials, especially during recovery and recycling operations.

Although levels of PentaBDE in human blood and milk, and in other environmental species, are falling in Europe, they continue to increase in North America and the Arctic region.

Based on the information in this risk profile, C-PentaBDE, due to the characteristics of its components, is likely, as a result of long-range environmental transport and demonstrated toxicity in a range of non-human species, to cause significant adverse effects on human health and the environment, such that global action is warranted.

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