

Format for submitting pursuant to Article 8 of the Stockholm Convention the information specified in Annex E of the Convention

Introductory information	
Name of the submitting Party/observer	Norway
Contact details (name, telephone, e-mail) of the submitting Party/observer	<p>The Norwegian Pollution Control Authority (SFT) Phone: +47 22 57 34 00 - Fax: +47 22 67 67 06 Address: Postboks 8100 Dep, NO-0032 Oslo, Norway Web: http://www.sft.no State of the Environment Norway: http://www.environment.no</p> <p>Contact person: Liselott Säll, Senior engineer E-mail: liselott.sall@sft.no</p>
Chemical name (as used by the POPS Review Committee (POPRC))	<p>Commercial PentaBDE is a technical mixture and does not have a Chemical Abstracts Service number. Its identified individual components have the following Chemical Abstracts Service numbers:</p> <p>(a) Pentabromodiphenyl ether (CAS No. 32534-81-9) 50–62% w/w; (b) Tetrabromodiphenyl ether (CAS No. 40088-47-9) 24–38% w/w; (c) Tribromodiphenyl ether (CAS No. 49690-94-0) 0–1% w/w; (d) Hexabromodiphenyl ether (CAS No. 36483-60-0) 4–12% w/w; (e) Heptabromodiphenyl ether (CAS No. 68928-80-3) trace,</p> <p>Pentabromodiphenyl ether (BDE-99) Tetrabromodiphenyl ether (BDE-47) Hexabromodiphenyl ether (BDE-100)</p>
Date of submission	

(a) Sources, including as appropriate (provide summary information and relevant references)	
(i) Production data:	No production
Quantity	
Location	
Other	
(ii) Uses	<p>Use banned since 1.7.2004</p> <p>But still in articles in use before this date, and in some imported articles:</p> <p>Different polyurethane (PUR) applications (PUR foam in furnitures and upholstery in automotive industry and domestic furnishing, PUR foam packing)</p> <p>Historic uses in textiles (clothing and PUR coating under carpets)</p> <p>Historic uses in electric and electronic equipment (PUR elastomers (instrument casings), epoxy resins and phenol resins (electric and electronic appliances).</p> <p>Probable still in some electric and electronic equipment imported from Asia.</p> <p>From 1 January 2004, products containing more than 0.25 % is classified as hazardous waste when they are discarded.</p> <p>During the summer of 2004, the target of collecting 80 % of discarded electrical and electronic equipment was reached; cf. the provisions concerning EEE waste in the Norwegian Hazardous Waste Regulations.</p>
(iii) Releases:	minor
Discharges	

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For reasons of economy, this document is printed in a limited number. Delegates are kindly requested to bring their copies to meetings and not to request additional copies.

Losses	
Emissions	
Other	

(b) Hazard assessment for endpoints of concern, including consideration of toxicological interactions involving multiple chemicals (provide summary information and relevant references)

Nomination report

In vivo rat studies indicate that the liver is the main target organ affected by pentaBDE (summarised in COM 2000) with a NOAEL of 1 mg/kg/d. Other in vivo studies have found i.a., developmental neurotoxicity (Eriksson et al. 1998; Branchi et al. 2001, Eriksson et al. 2001). Eriksson et al. (1998) found behavioural effects already after a single dose of 0.8 mg/kg BDE-99 to 10 days old mouse pups.

BDE-47 was shown to be acutely toxic for a copepod *Acartia tonsa* in a standard 48 h study by Breitholz et al. (2001). Moreover, BDE-47 caused disturbances in larval development in much lower levels. The EC50 in a 5 day study was determined as 13 µg/l.

Tjärnlund et al. (1998) observed significant inhibition of EROD activity in the liver of rainbow trout (*Oncorhynchus mykiss*) when fed with food containing BDE-47. Also other metabolic alterations were observed. Effects on rainbow trout fry sack growth were found at the level of 16 µg/l in the study by Wildlife International (2000).

In vitro studies have shown i.a., the thyroxin competing potential of hydroxylated metabolites and other metabolites (Meerts et al. 1998; Brouwer et al. 2001), an ability to activate the Ah-receptor (e.g., Meerts et al. 1998; Bunce et al. 2001) and possible genotoxicity (intragenic recombination) (Helleday et al. 1999). Immunotoxicity for the major pentaBDE congeners has been shown in mice but not in rats (Darnerud and Thuvander 1998). Also antiestrogenic response has been shown in vitro to be caused by several pentaBDE congeners (Brouwer et al. 2001).

New information:

A recent study, soon to be published (Källqvist et al. 2006), performed by Norwegian Institute for Water Research, showed that BDE-47 caused growth inhibition in colonies of the plankton algae *Skeletonema costatum*. (The growth inhibition test was carried out according to International Organization for Standardization, ISO 10253.)

NOEC = 6,6 µg/L

EC50 = 70 µg/L

The Norwegian study also showed a depression on reproductive output of the zooplankton *Daphnia Magna*. (The effect of BDE-47 on the parthenogenetic reproduction of *Daphnia magna* was investigated according to the OECD (Organization for Economic Co-operation and Development) Test Guideline 211.)

NOEC = 14 µg/L

Sublethal toxicity of BDE-47 was observed at low µg/L levels. But the authors conclude that the documented environmental water concentrations are many orders of magnitude lower, suggesting that BDE-47 is of minor risk to these organisms through direct water exposure.

Blubber biopsy and blood samples were collected from weaned grey seal (*Halichoerus grypus*) pups and juveniles during 1998 and 1999 (Hall et al., 2003). 54 post-weaned pups and 55 first year juveniles were studied, of which 13 were recaptured post-weaned pups. The median concentrations of ΣBDE (14 congeners) were 0.17 and 0.46 µg kg⁻¹ 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.

Health risk assessments

An assessment of the potential health risks to children and prospective parents associated with exposure to the commercial pentaBDE product was conducted by Great Lakes Chemical Corporation (GLCC) in accordance with United States Environmental Protection Agency (USEPA) Voluntary Children's chemical evaluation programme pilot in 2002. The result of the first phase of the assessment was a preliminary screening evaluation and was published in 2003. It comprised four components – hazard, exposure, risks and data needs. The information available was very limited and the results indicated that further evaluation was needed to fully characterize the risks. Phase 2 and 3 will provide a more detailed analysis.

Information from a review article: Toxic effects of brominated flame retardants in man and wildlife. By Per Ola Darnerud (2003).

Effects in mammals:

PentaBDE gave a low acute toxicity in experimental animals (rats, rodents) and the oral LD-50 in rat was in the range 0.5–5 g/kg body wt. (IPCS, 1994a). The clinical signs were reduced growth, diarrhoea, piloerection, reduced activity, tremors of forelimbs, red staining around eyes and nose, and continuous chewing. The porphyrin activity was relatively high according to studies in which the concentration of porphyrins increased considerably after oral dosing with the commercial pentaBDE product DE-71 (mixture of tetra-, penta- and hexaBDE) at 100 mg/kg body wt./day for 13 weeks (IPCS, 1994a). No mutagenic potency was observed in Ames test models using several different Salmonella strains with and without microsomal activation (IPCS, 1994a). After repeated dosage with PentaBDE products, several morphological effects were observed, such as changes in hepatic and thyroid size and histology. In two studies of commercial PentaBDE mixtures in rats, these effects appeared at the 10 mg/kg body wt. dose (Great Lakes Chem.; IPCS, 1994a). Immunological effects were suggested in mice after exposure to commercial DE-71 mixture (72 mg/kg body wt. for 14 days); suppression of the anti-SRBC response was observed, as well as a decreased thymus weight (Fowles et al., 1994). The PBDE congener BDE 47 markedly reduced the splenocyte numbers in mice (C57BL) after daily oral administrations of 18 mg/kg body wt. for 14 days (Darnerud and Thuvander, 1999). In the same study, Bromkal 70 reduced IgG antibody production from pokeweed-stimulated mouse splenocyte cultures *ex vivo*, whereas no immunological effects were seen in rats. Commercial PBDEs affected thyroid hormone homeostasis, and both technical products and pure tetra- and penta congeners (the latter at doses of 10–18 mg/kg body wt./day for 2 weeks) produced effects on serum thyroxin levels in rats and mice (Great Lakes Chem., IPCS, 1994a; Hallgren et al., 2001; Zhou et al., 2001; Hallgren and Darnerud, 2002). In one study on mice, effects on thyroxin were observed already at single dose of 0.8 mg/kg (Fowles et al., 1994), although with lack of dose–response effects relationship.

For PentaBDEs, the critical effects among the available studies seem to be developmental neurotoxicity and, 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, PBDEs as well as other BFRs, were capable to induce cell death of cerebellar granule cells in culture (Reistad et al., 2002). To summarise, the LOAEL value for PentaBDE could be set to 0.6–0.8 mg/kg body wt., based on the most sensitive effect observed, neurobehavioural effects during early development.

Effects in wildlife:

In a study on rainbow trout, BDE-47 (tetra) and -99 (penta) were given in the feed to rainbow trout (Tjærnslund et al., 1998). The result was a reduction in GSH reductase, haematocrit and blood glucose values. The effects of the congener BDE-47 on the calanoid *Acartia tonsa* were studied by Breitholz et al. (2001). In the 48-h acute toxicity test and larval developmental test, the 2- and 5-days LC-50 values were 2.4 and 0.013 mg/l, respectively. In addition, in reports not generally available, toxic effects of a tetra- to hexa-BDE mixture induced toxicity in a *Daphnia* test model. The NOEC values in a 48-h acute toxicity test and a 21-day life-cycle study were in both cases about 5 Ag/l (CITI, 1882; Dottar and Kreuger, 1998; both in Peltola and Ylä-Mononen, 2001).

In a study of male fish-eaters from the Baltic region, consuming 0–32 meals/month, a number of hormones were measured in the blood and compared to the levels on some selected environmental contaminants, including the PBDE congener BDE 47 (Hagmar et al., 2001). After adjustment for age, there was a significant association (negative correlation) between plasma BDE-47 and TSH. The authors stated that some significant correlation will occur from pure chance, and it was concluded that high consumption of organohalogen-polluted fish may not appear to affect plasma levels concentrations of pituitary, thyroid, or testosterone hormone levels in male adults.

To conclude, 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 technical PentaBDE products seem to cause effects at the comparably lowest dose. The critical effects of PentaBDEs are those on neurobehavioural 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).

Information from: In vivo and in vitro anti-androgenic effects of DE-71, a commercial polybrominated diphenyl ether (PBDE) mixture. By Stoke et al., 2005.

Recently we showed that the PBDE mixture, DE-71 (containing BDE-47, -99, -100, -153, -154) delayed puberty and suppressed the growth of androgen-dependent tissues in male Wistar rat following a peri-pubertal exposure. These effects suggested that DE-71 may be either inducing steroid hormone metabolism or acting as an androgen receptor (AR) antagonist. In a pubertal exposure study we observed a dose-dependent delay in PPS (preputial separation) with 60 and 120 mg/kg/day of DE-71 (4 and 5 days) and a corresponding suppression of ventral prostate (VP) and seminal vesicle growth at both doses. Adult males exposed to 60 mg/kg DE-71 for 3 days resulted in a significant increase in luteinizing hormone and a non-significant increase in testosterone, androstenedione and estrone. DE-71 also tested positive for anti-androgenic activity in an immature rat Hershberger assay, with decreases in mean VP and seminal vesicle weight following doses of 30-240 mg/kg.

Information from: Polybrominated diphenyl ethers (PBDEs): new pollutants – old diseases. By Siddiqi et al., 2003.

Evidence to date suggests that tetra- and penta-BDEs are likely to be the more toxic and bioaccumulative of the PBDE compounds, compared to octa- and deca-congeners. The toxicology of PBDEs is not well understood, but PBDEs have been associated with tumors, neurodevelopmental toxicity and thyroid hormone imbalance. The neurotoxic effects of PBDEs are similar to those observed for PCBs. Children exposed to PBDEs are prone to subtle but measurable developmental problems. It is presumed that PBDEs are endocrine disrupters, but research in this area is scant.

Information from: Ultrastructural changes observed in rat ovaries following in utero and lactational exposure to low doses of a polybrominated flame retardant. By Talsness et al., 2005.

We evaluated the effects of environmentally relevant concentrations (low doses) of 2,2',4,4',5-pentabromodiphenyl ether (PBDE-99) on the female reproductive system. A single dose of either 60 microg or 300 microg PBDE-99/kg body weight BW was administered on gestation day 6 to gravid Wistar rats. Ultrastructural 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. Mating of the F1 females with untreated males revealed resorption rates in the PBDE groups greater than the limits considered normal for our controls. External and skeletal anomalies were detected in offspring (F2) from two different dams (F1) with early developmental exposure to 300 microg PBDE-99/kg BW. Exposure to PBDE-99 resulted in female reproductive tract changes in the F1 generation which were apparent at adulthood.

Information from: Developmental exposure to low dose PBDE-99: effects on male fertility and neurobehavior in rat offspring. By Kuriyama et al., 2005.

In utero exposure to a single low dose of 2,2,4,4,5-pentabromodiphenylether (PBDE-99) disrupts neurobehavioral development and causes permanent effects on the rat male reproductive system apparent in adulthood. In the present study we assessed the effects of developmental exposure to one of the most persistent PBDE congeners (PBDE-99) on juvenile basal motor activity levels and adult male reproductive health. The exposure to low-dose PBDE-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 microg/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.

Information from: Investigation of strain and/or gender differences in developmental neurotoxic effects of polybrominated diphenyl ethers in mice. By Viberg et al., 2004.

The present study shows that neonatal exposure to 2,2',4,4',5-pentabromodiphenyl ether 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. Neonatal male and female mice were orally exposed on day 10 to 0.4, 0.8, 4.0, 8.0 or 16 mg PBDE-99/kg bw. Spontaneous behaviour (locomotion, rearing, and total activity) was observed in two-, five- and eight-month-old mice.

Information from: Assessment of DE-71, a commercial polybrominated diphenyl ether (PBDE) mixture, in the EDSP male and female pubertal protocols. By Stoker et al., 2004.

Rats were gavaged daily with 0.3, 30, or 60 mg/kg DE-71 in corn oil from postnatal day (PND) 23-53 in the male or PND 22-41 in female. Serum T4 was significantly decreased at 30 and 60 mg/kg following the 5-day exposures and in the 21-day exposed females. Doses of 3, 30 and 60 mg/kg decreased T4 in 31-day exposed males. Serum T3 was decreased and TSH elevated by 30 and 60 mg/kg in the 31-day exposed males only.

Information from: Effects of perinatal exposure to a polybrominated diphenyl ether (PBDE-99) on mouse neurobehavioral development. By Branci et al., 2002.

We investigated the effects of perinatal PBDE exposure on mouse neurobehavioral development. 2,2',4,4,5-pentabromodiphenylether (PBDE-99:0.6, and 30 mg/kg per day) was administered daily to CD-1 Swiss females by gavage from gestational day (GD) 6 to postnatal day (PND) 21. Sensori-motor development analysis (PNDs 2-20) revealed a delayed appearance of climbing response in the PBDE-99 high-dose group. On PND 11, the homing test revealed a trend for treated animals to be more active than controls. PBDE-99 treated mice tended to be hypoactive. These findings showed that perinatal exposure to PBDE 99 produces several behavioural alterations.

Information from: Neonatal exposure to the brominated flame retardant 2,2',4,4',5-pentabromodiphenyl ether causes altered susceptibility in the cholinergic transmitter system in the adult mouse. By Viberg et al., 2002.

The present study indicates that the cholinergic system, in its developing stage, may be target of and sensitive to PBDEs. Neonatal exposure of male NMRI mice on postnatal day 10, to 2,2',4,4',5-pentaBDE (8mg/kg bw) was shown to alter the response to a cholinergic agent, nicotine, at an adult age. The nicotine-induced behaviour test revealed a hypoactive response to nicotine in PBDE 99-treated animals, whereas the response of controls was an increased activity. These findings show similarities to observations from neonatal exposure to PCBs and nicotine, compounds shown to affect cholinergic nicotine receptors. This indicates that PBDE 99 can affect the cholinergic system and might thereby interact with other environmental toxicants.

Information from: Developmental exposure to brominated diphenyl ethers results in thyroid hormone disruption. By Zhou et al., 2002.

Primiparous Ling-Evans rats were orally administered DE-71 (0, 1, 10 and 30 mg/kg/day) in corn oil from gestation day (GD) 6 to postnatal day (PND) 21. There were no significant effects of treatment on maternal body weight gain, litter size, or sex ratio, nor were there any effects on any measures of offspring viability or growth. Serum T(4) was reduced in a dose-dependent manner in fetuses on GD 20 (at least 15 %) and offspring on PND 4 and PND 14 (50 and 64 % maximal in the 10 and 30 mg/kg/day groups, respectively), but recovered to control levels by PND 30. Reduction in serum T(4) was also noted in GD 20 dams (48 % at highest dose), as well as PND 22 dams (44 % at highest dose). Increased liver to body weight ratios in offspring were consistent with induction of EROD, PROD or UDPGT. Induction of PROD was similar in both dams and offspring; however, EROD and UDPGT induction were much greater in offspring compared to dams. These data support the conclusion that DE-71 is an endocrine disrupter in rats during development.

Information from: In vitro estrogenicity of polybrominated diphenyl ethers, hydroxylated PBDEs, and polybrominated bisphenol A compounds. By Meerts et al., 2001.

In this study, we investigated the (anti)estrogenic potencies of several PBDE congeners and three hydroxylated PBDEs. The results indicate that several pure PBDE congeners, but especially HO-PBDEs are agonists of both ER alpha and ER beta receptors, thus stimulating ER-mediated luciferase induction in vitro. These data also suggest that in vivo metabolism of PBDE may produce more potent pseudoestrogens.

(c) Environmental fate (provide summary information and relevant references)				
Chemical/physical properties	BDE-47 (Tetrabromodiphenyl ether)	BDE-99 (Pentabromodiphenyl ether)		
	Molar mass (Da)	485,8	Molar mass (Da)	564,7
	Vapour pressure (Pa)	8,2 X 10 ⁻⁵	Vapour pressure (Pa)	7,6 x 10 ⁻⁶
	Water solubility (µg/L)	9,3	Water solubility (µg/L)	1,0
	Log Kow	6,7	Log Kow	7,4
		Palm et. al 2002		
Persistence	<u>Half-life values (Palm 2001) in the Nomination report:</u>			
	Estim. Halflife in soil (d)	150	Estim. Halflife in soil (d)	150
	Estim. Halflife in sediment (d)	600	Estim. Halflife in water (d)	600
	<u>Environmental data from the Nomination report:</u>			
	PentaBDE congeners deposited in marine sediments a few decades ago are still present in clearly quantifiable amounts (Nylund et al. 1992; Zegers et al. 2000) indicating high persistency in sediment. Also data from remote regions indicate high persistency to degradation in the environment (TemaNord 2001:579).			
	<u>New estimated values on halflife in different environmental compartments (Palm et al. 2002):</u>			
BDE-47 (Tetrabromodiphenyl ether)	BDE-99 (Pentabromodiphenyl ether)			

	<table> <tr> <td>Estim. Half-life in air (d)</td> <td>11</td> <td>Estim. Half-life in air (d)</td> <td>19</td> </tr> <tr> <td>Estim. Half-life in water (d)</td> <td>150</td> <td>Estim. Half-life in water (d)</td> <td>150</td> </tr> </table>	Estim. Half-life in air (d)	11	Estim. Half-life in air (d)	19	Estim. Half-life in water (d)	150	Estim. Half-life in water (d)	150														
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<p>How are chemical/physical properties and persistence linked to environmental transport, transfer within and between environmental compartments, degradation and transformation to other chemicals?</p>	<p>The results from a modeling exercise utilizing the European variant (EVn) BETR multimedia environmental fate model are presented for selected polybrominated diphenyl ethers (PBDEs) of the technical penta- (Pe-) bromodiphenyl ether (BDE) product (Prevedouros et al. 2004). The objectives of this study were to test PeBDE emission estimates from the literature for Europe by investigating the consistency between model predictions and ambient measurements to address the ability of the model to predict spatial variability and differences between congeners. Concurrently sampled and analyzed passive sampling air data, together with soil and grass data, were used as key model validation tools. The model steady-state simulations gave generally good agreement with measured data for BDE-47 and -99 with greater discrepancies for heavier congeners (e.g., BDE-153). To predict future atmospheric concentration trends, the model was used in its fully dynamic mode over the period 1970-2010. It was predicted that atmospheric concentrations peaked around 1997, declining with an overall “disappearance” half-life of 4.8 years. Soil and grass levels were underestimated by the model; possible reasons for differences with measurement data are further explored. Finally, the importance of temporally and spatially resolved environmental data sets is highlighted, while improved quantification of degradation half-lives is essential to better understand and predict the behavior of BDE congeners in PeBDE.</p> <p>A study performed in 2003 in Canada considers polybrominated diphenyl ethers physical chemistry, environmental partitioning and considerations regarding potential for long-range atmospheric transport (LRAT) (Gouin and Harner 2003). Internally consistent physical-chemical property data are presented for five representative congeners (PBDE-15, -28, -47, -99, -153) and used in a multimedia modelling approach. Results of the Level II model 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 that are not well known. Model results indicate that only a small proportion of PBDEs exist in air and water, suggesting that these compounds have limited LRAT potential. However, we 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/volatilisation hops, otherwise known as the “grasshopper” effect. Our understanding of this process and the overall environmental fate of PBDEs would benefit from process studies on air- foliage and air-soil exchange as well as reliable measurements of temperature dependant physical-chemical and reactivity properties.</p>																						
<p>Bio-concentration or bio-accumulation factor, based on measured values (unless monitoring data are judged to meet this need)</p>	<p><u>New information on BCF and BMF</u></p> <table> <tr> <td colspan="2">BDE-47 (Tetrabromodiphenyl ether)</td> <td colspan="2">BDE-99 (Pentabromodiphenyl ether)</td> </tr> <tr> <td>BCF</td> <td>Cyprinus carpio</td> <td>66 700</td> <td>BCF</td> <td>Cyprinus carpio</td> <td>17 700</td> </tr> <tr> <td>BMF</td> <td>Guillemot/herring</td> <td>19</td> <td>BMF</td> <td>Guillemot/herring</td> <td>17</td> </tr> <tr> <td>BMF</td> <td>Salmon/sprat</td> <td>11</td> <td>BMF</td> <td>Salmon/sprat</td> <td>10</td> </tr> </table> <p>CITI, 1982 De Wit, 2000</p>	BDE-47 (Tetrabromodiphenyl ether)		BDE-99 (Pentabromodiphenyl ether)		BCF	Cyprinus carpio	66 700	BCF	Cyprinus carpio	17 700	BMF	Guillemot/herring	19	BMF	Guillemot/herring	17	BMF	Salmon/sprat	11	BMF	Salmon/sprat	10
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(d) Monitoring data (provide summary information and relevant references)Evidence on bioaccumulation from monitoring studies in the Nomination Report:

Concentrations of the major pentaBDE congeners increase as the trophic level rises in the biota of the same region, showing that pentaBDE is biomagnified (results from Baltic Sea sprat, herring and salmon by Burreau et al. 1999 and from Atlantic biota by Burreau et al. 2000).

According to results from pooled Baltic Sea herring samples (Haglund et al. 1997) and sprat samples (Strandman et al. 1999), concentrations of pentaBDE congeners increase with the age of marine fish, indicating bioaccumulation and high resistance to metabolic transformation.

Tetrabrominated and pentabrominated diphenyl ethers show the highest biomagnification potential of all PBDEs studied. Increasing levels of pentaBDE congeners with rising trophic position can be observed for the data in biota from around the world (TemaNord 2001:579).

New information on BAFs

BDE 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 (Stäket, northern inflow to Lake Mälaren) and transplanted in baskets to other downstream sites in Lake Mälaren (west side of Stockholm at Riddarfjärden), Saltsjön (east side of Stockholm in 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 BDE congeners (BDE47, BDE99, BDE100, BDE153 and BDE154) were determined. Σ BDE concentrations were low and ranged from 0.4 to 0.79 $\mu\text{g kg}^{-1}$ wet weight (19 to 39 $\mu\text{g kg}^{-1}$ lipid weight). The congener pattern was dominated by BDE47 and BDE99 and was similar to the penta-mix PBDE technical product. Comparison on a lipid weight basis showed that BDE concentrations were higher close to Stockholm than at the background site and that there was no difference between concentrations on the west or east side of the city centre.

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 highest BAF value obtained was for BDE154 and lowest was for BDE47. In other compound groups studied, BAFs ranged from 0.1 (*p,p'*-DDD) to 0.5 (CB153). This is in agreement with other studies in blue mussels showing higher bioaccumulation potential for the lower brominated BDEs when compared to CBs with similar octanol-water partitioning coefficients (Gustafsson et al., 1999; Booij et al., 2002).

New information on BMFs

Studies of the biomagnification of tri- to deca -BDEs were carried out in three different food chains, two in the Baltic Sea (roach, perch, pike (Burreau et al., 2004); zooplankton, sprat, herring, salmon) and one in the Atlantic Ocean (zooplankton, small herring, large herring, salmon) (Burreau et al., in preparation). Samples were analysed for 13 BDE congeners (BDE28, BDE35, BDE47, BDE49, BDE66, BDE99, BDE100, BDE153, BDE154, BDE155, BDE183, BDE203 and BDE209) as well as stable nitrogen isotopes.

Roach, perch, pike food chain in the Baltic Sea

When compared on a lipid weight basis, BDE47 was the major congener found in perch (median: 10 $\mu\text{g kg}^{-1}$ lipid weight) and pike (71 $\mu\text{g kg}^{-1}$ lipid weight) (Burreau et al., 2004). However, the major congener found in the omnivorous roach was BDE209 (48 $\mu\text{g kg}^{-1}$ lipid weight), and the median BDE99 concentration was particularly low in roach. Low BDE99 levels have been seen previously in several other fish species (bream, carp)

(Sellström et al., 1993; Dodder et al., 2002; Stapleton et al., 2004a) and indicate significant metabolism of this congener.

Regression analyses between lognormalised

concentrations of each BDE congener and the $\delta^{15}\text{N}$ values were used to study

food chain biomagnification. Biomagnification was described using the following equation

$$c = A \cdot e^{(B \cdot \delta^{15}\text{N})}$$

where c is the concentration of a substance measured in organisms in which $\delta^{15}\text{N}$ is also measured. The term A is a constant and the B value describes the biomagnification potential of a substance. A positive B value means that the concentration of a certain substance is higher at a higher trophic level and that biomagnification has occurred.

All the tri- to hepta-BDE congeners biomagnified, but showed a maximum biomagnification for the penta-BDEs (log Kow values 7.24 - 7.32).

Zooplankton, sprat, herring, salmon in the Baltic Sea and comparison to the Atlantic Ocean food chain (zooplankton, small herring, large herring, salmon)

When compared on a lipid weight basis, BDE47 was the major congener found in zooplankton (median: 2.3 $\mu\text{g kg}^{-1}$ lipid weight), sprat (3 $\mu\text{g kg}^{-1}$ lipid weight), herring (4.5 $\mu\text{g kg}^{-1}$ lipid weight) and salmon (22 $\mu\text{g kg}^{-1}$ lipid weight) (Burreau et al., in preparation). As for the roach, perch, pike food chain, the tri- to hepta-BDEs biomagnified, again showing maximum biomagnification for the penta-BDEs.

The B values for the BDE congeners that biomagnify were similar (0.4) to those of CBs (0.3 - 0.4) of similar hydrophobicity. Higher BDE concentrations were found in the Baltic Sea fish than in the Atlantic fish. Biomagnification was more difficult to

discern in the Atlantic Ocean food chain due to confounding body size effects in the fish.

New information on concentrations in organisms in different trophic positions:

Studies performed after the completion of the Nordic report (TemaNord 2001:579) support the reports conclusions for bioaccumulation. Concentrations of BDE-47 and BDE-99 in Lake Ontario pelagic food web shows increasing concentrations with increasing trophic position (Alaee et al. 2002). The dietary uptake of persistent organic pollutants (POPs) is the main route of exposure for animals at upper trophic levels (Kiriluk et al. 1995). Furthermore the bioaccumulation and biomagnification of POPs in top predators is influenced by the length and structure of the food web (Rasmussen et al. 1990). Therefore, in this study the trophodynamics of PBDEs in Lake Ontario pelagic food web was examined. The bioaccumulation and biomagnification factors were compared to other values for other food webs and were compared to other contaminants such as PCBs in Lake Ontario. In this study concentration of PBDEs in archived plankton, *Mysis*, *Diporeia*, alewife, smelt, sculpin and lake trout samples collected in 1993 were determined; and trophodynamics of PBDEs in the Lake Ontario pelagic food web was investigated.

Lake Ontario pelagic food web consists of three trophic levels. Lake trout (*Salvelinus namaycush*) are a top predator fish species in Lake Ontario, which feed 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 netplankton. Previous studies on biota from Lake Ontario showed an increase in the concentration of organochlorine contaminants with each successive level of the food web.

Concentrations of BDE -47, -100, and -153 were increasing at each step up the food chain with biomagnification factors ranging between 7.1 for BDE-99 for biomagnification between netplankton and benthic organisms and 1.7 for BDE-100 for biomagnification between lake trout and forage fish. 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 plankton, *Mysis* and *Diporeia* resembled to the penta-BDE formulation, which indicates that BDE-99 bioaccumulate in the invertebrates and starts to be metabolized by forage fish. A recent study of an arctic food chain shows the same result (Sørmo et al. 2006) as Alaee's study.

Polybrominated diphenyl ethers (PBDEs) were determined in adipose tissue of adult and subadult 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 4 congeners (BDE47, 99, 100, and 153) were consistently identified in all samples. BDE47 was the major PBDE congener representing from 65% to 82% of the sum (Σ) PBDEs. Age was not a significant covariate for individual PBDEs or Σ PBDE. Higher proportions of BDE 99, 100, and 153 were generally found in samples from the Canadian Arctic than from Svalbard or the Bering- Chukchi Sea area of Alaska. Geometric mean Σ PBDE concentrations were highest for female polar bear fat samples collected from Svalbard (50 ng/g lipid weight (lw)) and East Greenland (70 ng/g lw). Significantly lower Σ PBDE concentrations were found in fat of bears from Canada and Alaska (means ranging from 7.6 to 22 ng/g lw).

For the entire dataset, Σ PBDE concentrations were correlated with PCBs. The geographical trends for PBDEs parallel those for PCBs implying similar source regions for long range transport to the Arctic and bioaccumulation pathways in the arctic marine food web. All four major PBDE congeners were found to biomagnify from ringed seals to polar bears. BDE153 showed the greatest (71 \times) biomagnification factors (BMFs) and, on average, had a BMF that was 5.5- fold higher than for PCB congener 153 (13 \times) but similar to PCB congener 194 (73 \times), indicating that it is a highly bioaccumulative compound.

Leonards et al. (2004) studied the transfer of BDEs (congeners BDE28, BDE47, BDE49, BDE66, BDE71, BDE75, BDE77, BDE85, BDE99, BDE100, BDE119, BDE138, BDE153, BDE154, BDE183, BDE190 and BDE209), and HBCD and TBBP-A, using GC-ECNIMS and LC-MS, respectively. The samples comprised sediment, suspended particulate material, invertebrates (e.g. bivalves, shrimps, worms), and fish (e.g. sandeel, flounder, goby, sole, herring, whiting) collected in spring 2003 in the feeding habitats of the common tern (*Sterna hirundo*), the western Scheldt estuary, and harbour seals (*Phoca vitulina*), the Wadden Sea. Tern eggs were collected from the Terneuzen colony, and the blubber of adult male seals collected from stranded animals from the Wadden Sea population. In sediment and SPM, the dominant BFRs were BDE209 and HBCD, and in biota BDE47 and HBCD, followed by BDE49, BDE99, BDE100 and BDE154. For the common tern food chain, BDE concentrations expressed on a lipid basis increased from invertebrates to fish (sandeel) to tern.

The BDE congener patterns in prey species, tern eggs and seal blubber suggest that tern are probably able to metabolise BDE49, which has vicinal H-atoms at the meta-para position.

Seals are able to metabolise both groups of congeners, as has been observed for chlorobiphenyls (Boon et al., 1997), and hence have a greater metabolic capacity than terns.

New information on concentrations in top predators in Europe

Another recent study (Herzke et al. 2005) shows relatively high levels in top predatory eggs of BDE-47 and BDE-99. The highest PBDE level (sum of nine congeners) was found in eggs of white-tailed sea eagle with up to 800 ng/g ww (median sumPBDE: 184 ng/g ww), followed by eggs of peregrine falcon and osprey (median sumPBDE: 155 and 105 ng/g ww, respectively). Golden eagle eggs showed the lowest concentration of all species (median sumPBDE: 3 ng/g ww). The levels in the peregrine falcon are similar to those found earlier in the Baltic region [Lindberg, P., Sellstrom, U., Haggberg, L., Wit, C.A., 2004.]

Data of PBDE concentrations on a lipid weight basis showed that the levels in the white-tailed sea eagle eggs exceed the peak levels for guillemot eggs from the Baltic from the 1980s by a factor of three. This illustrates the relatively high contamination of the white-tailed sea eagle in Norway.

Lindberg et al. (2004) analysed a range of BFRs in peregrine falcon (*Falco peregrinus*) eggs collected in northern and south-western Sweden in 1987-1999. The compounds analysed were constituents of the PBDE penta-mix (BDE47, BDE99, BDE100, BDE153 and BDE154), octa-mix (BDE183), and deca-mix (BDE209) formulations, a hexabrominated biphenyl (BB153) and HBCD (using GC-ECNIMS). The eggs represented females from three different breeding populations, including one captive population raised on a controlled diet of domestic chickens. The northern wild population feeds on waders and ducks and the southern wild population feeds on birds in the terrestrial food web. All BFRs analysed were detected, and concentrations were much higher in wild falcons than in captive birds ($p < 0.001$ for all compounds) (Figure 1). The mean concentrations of the sum of BDE47, BDE99, BDE100, BDE153, BDE154, BDE183 and BDE209 were 3700 and 4500 $\mu\text{g kg}^{-1}$ lipid weight for the southern and northern populations, respectively and 56 $\mu\text{g kg}^{-1}$ lipid weight in the captive population. However, individual ΣBDE concentrations were as high as 39,000 $\mu\text{g kg}^{-1}$ lipid weight, some of the highest concentrations seen in wildlife so far. The BDE congener patterns were dominated by the penta- and hexa-BDEs, BDE99, BDE100 and BDE153, followed by HBCD. BDE47, BDE154 and BDE183 were present in lower, but similar, concentrations, followed by BDE209 and BB153. The captive population had a different congener profile, dominated by BDE153 and BB153, followed by BDE183 and BDE209, then HBCD and BDE154, and low concentrations of BDE47, BDE99 and BDE100. These differences can only be explained by differences in exposure due to diet, although differences in metabolic capacity can also contribute to differences between species.

These congener patterns can be contrasted with those in piscivorous birds, such as guillemots and cormorants (*Phalacrocorax carbo*) (Law et al., 2002), in which BDE47 is the dominant congener. Also, it seems that terrestrial organisms may be more highly exposed to the more highly brominated BDE congeners than are aquatic organisms. In another study, 62 deserted or addled eggs from six species of predatory birds collected between 1991 and 2002 throughout Norway were analysed for BDEs (congeners BDE28, BDE47, BDE99, BDE100, BDE138, BDE153, and BDE154) and brominated biphenyls (congeners BB15, BB49, BB52, BB101 and BB153) (Herzke et al., 2004b submitted). The species investigated were white-tailed sea eagle (*Haliaeetus albicilla*), merlin (*Falco columbarius*), golden eagle (*Aquila chrysaetos*), osprey, peregrine falcon and goshawk (*Accipiter gentilis*), and were selected as representative of different food chains, habitats and migratory behaviour. BDE47, BDE99 and BDE153 were the dominant BDE congeners, with species-dependent compound ratios. BDE153 was the most abundant BDE congener in eggs of peregrine falcon, golden eagle and merlin, and BDE47 in those of the other species. The highest ΣBDE concentration was found in eggs of white-tailed sea eagle, up to 800 $\mu\text{g kg}^{-1}$ wet weight (median value 184 $\mu\text{g kg}^{-1}$ wet weight) followed by peregrine falcon and osprey (median ΣBDE 155 and 105 $\mu\text{g kg}^{-1}$ wet weight, respectively). Golden eagle eggs showed the lowest concentrations. Generally, concentrations were comparable to those reported for peregrines from Sweden (Lindberg et al., 2004). Jaspers et al. (2004) have determined BDEs (congeners BDE28, BDE47, BDE99, BDE100, BDE153, BDE154 and BDE183) in eggs of the little owl (*Athene noctua*) from Charleroi, Belgium. The little owl is a small sedentary predator, and so provides a very suitable biomonitoring species. It feeds on a variety of prey, including mammals, birds, reptiles, amphibians, earthworms and beetles, depending on the season and local availability. ΣBDE concentrations in the eggs ranged from 29 to 572 $\mu\text{g kg}^{-1}$ lipid weight, with a mean value of 108 $\mu\text{g kg}^{-1}$. The congener profile was dominated by BDE99 and BDE153, followed by BDE47.

Voorspoels et al. (2004b) have investigated the levels and distribution of tri- to hepta-BDEs (congeners BDE28, BDE47, BDE99, BDE100, BDE153, BDE154 and BDE183) in muscle, liver, fat and brain from several species of birds of prey from Belgium: (27 buzzards (*Buteo buteo*), 7 sparrowhawks and 8 owls). ΣBDE concentrations in individual birds ranged over almost 4 orders of magnitude. The lowest levels were found in owl brain, while the highest levels were measured in sparrowhawk liver (up to 600 $\mu\text{g kg}^{-1}$ wet weight), and differences observed between the different species could be related to differences in their diets. No significant differences in BDE profiles were observed between different tissues of birds of the same species. However, significant congener profile differences were observed between the three species, suggesting different metabolic capabilities. The contribution of BDE47 and BDE183 to ΣBDE was similar for the three species,

D'Silva et al. (2004) have determined 16 BDE congeners (BDE17, BDE28, BDE47, BDE49, BDE66, BDE71, BDE77, BDE85, BDE99, BDE100, BDE119, BDE126, BDE138, BDE153, BDE154 and BDE183) in eggs and adipose tissue of herons (*Ardea cinerea*) collected from three UK sites in 2002-2003. BDEs were detected in all samples, and concentrations were typically higher in adipose tissue than in eggs from the same location. Congener profiles were similar to those reported previously for cormorant livers from the UK (Law et al., 2002), with BDE47 the dominant congener contributing 40-50% of ΣBDE . ΣBDE concentrations in adipose tissue ranged from 1.4 to 2.9 mg kg^{-1} lipid, and the highest concentration observed in eggs was 4.1 mg kg^{-1} lipid.

Sinkkonen et al. (2004) determined methoxy tri-, tetra- and penta-BDE congeners in guillemots (whole animal) from the Baltic sea and Norwegian west coast using GC-EIMS. The tri- and penta-BDE congeners were not detected, but three MeO-TeBDE congeners were found in one Baltic guillemot sample, at concentrations ranging from 8.3 to 1,100 ng kg^{-1} lipid weight. The profile was similar to that observed previously in a Baltic salmon sample (Haglund et al., 1997). Seven BDE congeners (BDE15, BDE28, BDE47, BDE66, BDE99, BDE100 and BDE153) were also determined, and the concentrations of ΣBDE and BDE47 in the same sample were 332 and 275 ng kg^{-1} lipid weight, respectively. Lower concentrations (118 and 82 ng kg^{-1} lipid weight) were seen in the Atlantic guillemot. The origin of MeO-BDEs in biological samples is unknown. They are often present in high concentrations relative to the BDEs themselves, although they are not known to be produced commercially and some occur as natural products in marine environments.

Guillemot eggs from the Baltic Sea, sampled between 1969 and 2001 (Sellström et al. 2003), were analyzed for tetra- and pentabromodiphenyl ethers (2,2,4,4-tetraBDE (BDE-47), 2,2,4,4,5-pentaBDE (BDE-99), and 2,2,4,4,6-pentaBDE (BDE-100)), and hexabromocyclododecane (HBCD). This temporal trend study indicates that the concentrations of the polybrominated diphenyl ether compounds increased from the 1970s to the 1980s, peaking around the mid- to the late-1980s. These peaks are then

followed by a rapid decrease in concentrations during the rest of the study period, with the concentrations of the major BDE congener below 100 ng/g lipid weight at the end of the period. This corresponds to less than 10% of its peak values.

New information on concentrations in top predators in the Arctic

A recent study (Verreault et al. 2005) on flame retardants and methoxylated and hydroxylated polybrominated diphenyl ethers in two Norwegian Arctic Top predators: glaucous gulls (*Larus hyperboreus*) and polar bears (*Ursus maritimus*), investigated the congener patterns and levels of total hexabromocyclododecane (HBCD), polybrominated biphenyls, polybrominated diphenyl ethers (PBDEs), as well as methoxylated (MeO) and hydroxylated (OH) PBDEs in plasma samples.

All PBDE congeners monitored were detected in glaucous gull and polar bear samples with the exception of BDE17. In female polar bears, the ^aPBDE concentration was comprised essentially of BDE47 (93%), followed by BDE99 (3%) and traces of several other minor PBDEs. In glaucous gull males and females a lesser proportion of BDE47 (51%) comprised the ^aPBDE concentration, with a nearly equal share of four individual penta and hexa-BDEs, i.e., BDE99, 100, 153, and BDE154/BB153. No significant difference in homologue PBDE profiles was found between male and female glaucous gulls.

Sum (^a) concentrations of the 12 PBDEs monitored in glaucous gulls was in the range: 8.23-67.5 ng/g wet wt

Sum (^a) concentrations of the 12 PBDEs monitored in polar bears was in the range: 2.65-9.72 ng/g wet wt

The presence of MeO- and OHPBDEs in plasma of both species suggests possible dietary uptake from naturally occurring sources (e.g., marine sponges and green algae), but also metabolically derived biotransformation of PBDEs such as BDE47 could be a contributing factor.

A report from 2004 (Verreault et al. 2004) examined the congener patterns and the current levels of new and established organohalogen contaminants and their metabolites in plasma and egg homogenate samples of glaucous gulls breeding at Bear Island (Bjørnøya). Brominated flame retardants (BFRs) were detected in plasma and egg homogenate samples. The congener profile of PBDEs was dominated by BDE-47 that constituted on average 40% of Σ PBDE. In a study of glaucous gulls from Bear Island, BDE-47 and BDE-99 were detected in 15 liver samples, and the concentrations ranged between not-detected to 424 ng/g lipid weight (Herzke et al., 2003). In the present study, the range (14.7–2,037 ng/g lw) for lipid-normalized plasma concentrations of these two compounds was significantly higher.

The sum PBDE level in herring gull eggs collected at the Shelter Island in Lake Huron was reported to be 23, 142 and 633 ng/g (wet weight), for the years 1981, 1990 and 2000, respectively, which corresponds to a doubling in concentration every 5 years (Norström et al., 2002). By contrast, the sum PBDE (BDE-47, -99 and -100) level in guillemot eggs collected at Stora Karlsø in the Baltic Sea has been reported to be 190 ng/g l.w. (23 ng/g wet weight) in 1997 (Sellström, 1999).

Brominated flame retardants (BFRs), PCBs and chlorinated pesticides were investigated in liver and intestinal contents of 15 glaucous gulls from Bjørnøya (Herzke et al. 2003). Of the analysed BFRs only 2,20,4,40-tetra- and 2,20,4,40,5-pentabrominated diphenylethers (PBDE 47 and 99) could be detected. The concentrations ranged between 2 and 25 ng/g ww.

An increase of PBDE levels in guillemot egg between the 1970s and the 1980s, followed by a rapid decrease until 2001, was reported for the Baltic region, with peak levels of the dominating congener BDE 47 of 1000 ng/g lw (Sellstrom et al., 2003.)

Vorkamp et al. (2004a) determined BDEs (BDE17, BDE28, BDE47, BDE49, BDE66, BDE85, BDE99, BDE100, BDE153 and BDE154) in liver and eggs of black guillemot (*Cephus grylle*) from west and east Greenland collected during 1999-2001 using GCECNIMS. Σ BDE concentrations ranged from 1.8 to 3.1 μ g kg⁻¹ wet weight in eggs (west Greenland only) and from 0.7 to 9.5 μ g kg⁻¹ wet weight in livers. Concentrations of congeners BDE17, BDE49, BDE66 and BDE85 were consistently below the detection limits. Higher concentrations of BDEs were seen in livers from east Greenland than west, as for other organochlorines determined in this study. Median concentrations of Σ BDE expressed on a lipid basis were ca. 72 and 25 μ g kg⁻¹ in east and west Greenland, respectively, approximately 2 to 5 times lower than those currently observed in the Baltic guillemot from the preceding study.

Wolkers et al. (2004) analysed BDEs in ringed seals (*Phoca hispida*), beluga whales (*Delphinapterus leucas*) and polar bears (*Ursus maritimus*) from Svalbard, Norway during 1998-2001. In ringed seal, BDE47 accounted for > 90% of Σ BDE. In contrast, beluga whales, feeding on prey similar to that of ringed seals, showed both higher BDE concentrations and a more complex congener pattern. Polar bears contained only BDE47 in relatively small amounts, due to their exceptional metabolic capacity (Norstrom and Muir, 1994). The geometric means observed for BDE47 were 17 μ g kg⁻¹ in ringed seal, 90, 70 and 17 μ g kg⁻¹ in juvenile, adult male and adult female beluga (respectively), and 27 and 46 μ g kg⁻¹ in male and female polar bears, all on a lipid weight basis. A number of methoxylated metabolites of tetra-BDEs were detected in beluga, with two unidentified tetra-BDE metabolites being observed as well as that of BDE47. Σ BDE concentrations (geometric mean; 22 congeners) were 18 μ g kg⁻¹ lipid weight in ringed seal; 234, 161 and 29 μ g kg⁻¹ in juvenile, adult male and adult female beluga; and 27 and 46 μ g kg⁻¹ in male and female polar bears.

Vorkamp et al. (2004b) determined BDEs (congeners and method as listed above) in the blubber of 19 ringed seals from East Greenland (11 male and 8 female; aged 0.5 to 24.5 years) collected in spring 2001. Σ BDE concentrations ranged from 21 to 74 μ g kg⁻¹ lipid weight, with similar concentrations in both adults and subadults. BDE47 accounted for 76% and 71% of Σ BDE in male and female seals, respectively, similar to ringed seal blubber from Arctic Canada.

Covaci et al. (2002c) determined a suite of BDE congeners (BDE28, BDE47, BDE66, BDE71, BDE99, BDE100, BDE153 and BDE154) in the liver of 21 harbour porpoises (*Phocoena phocoena*) stranded on the Belgian coast during 1997-2000. The range of Σ BDE concentrations was 410 to 5,800 $\mu\text{g kg}^{-1}$ lipid weight, with a mean value of 2,180 $\mu\text{g kg}^{-1}$. The median concentrations of BDEs were higher in the adult group ($n = 8$) than in the juveniles ($n = 13$). The BDE profile was, as is common in marine mammals, dominated by BDE47, followed by BDE100, BDE99, BDE154 and BDE153.

Thron et al. (2004) determined BDEs (BDE17, BDE47, BDE49, BDE99, BDE100, BDE119, BDE140, BDE153, BDE154 and BDE183) in the blubber of harbour porpoises from Iceland, Norway, and the Baltic and North Seas collected between 1997-2001. The animals were either stranded or accidentally caught in gill-nets. Σ BDE concentrations ranged widely, from 18 to 5,800 $\mu\text{g kg}^{-1}$ lipid weight. Animals with poor body condition (lower mean blubber thickness) had much higher concentrations, as did males, but no correlation with age was observed. However, females showed decreasing concentrations with age, indicating elimination via transfer from mother to offspring. When animals of similar age, sex and body condition were considered, the concentrations of BDEs were similar in the Baltic and North Sea areas (median = 170 $\mu\text{g kg}^{-1}$ lipid weight) and were significantly higher than those in animals from Iceland (median = 38 $\mu\text{g kg}^{-1}$ lipid weight). Also, BDE concentrations tended to decrease with increasing latitude along the Norwegian coast. The regional differences observed were attributed to the proximity to local sources of PBDEs in Europe.

Kalantzi et al. (2004 in press) tagged grey seal pups on the Farne Islands, NE England, during the 1998 and 1999 breeding seasons. Blubber biopsy samples collected at the same time were then analysed for BDEs (congeners BDE17, BDE28, BDE35, BDE37, BDE47, BDE49, BDE71, BDE75, BDE85, BDE99, BDE100, BDE119, BDE153 and BDE154). Σ BDE concentrations ranged from 45 to 1,500 $\mu\text{g kg}^{-1}$ lipid weight, with a geometric mean of 290 $\mu\text{g kg}^{-1}$ lipid weight. BDE47 dominated the congener profile, followed by BDE100, BDE99, BDE153 and BDE154. No significant differences were found between males and females, probably due to the young age of the animals. A significant decrease was observed in Σ BDE concentrations between newly weaned seals in 1998 and 1999 ($p < 0.01$), but not for the majority of juveniles in 1999 and 2000.

In a recent study, 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 3 months of this period. BDE analysis was undertaken using GC-ECNIMS. A supplement of BDE209 (not present at detectable concentrations in the fish) was included in the diet during the second month of the study. Consistently high absorption (89 - 99%) was observed for all BDE congeners studied (BDE28, BDE47, BDE49, BDE99, BDE100, BDE153, BDE154 and BDE209).

A suite of 37 polybrominated diphenyl ether (PBDE) congeners and all of the homologue groups from mono- to deca-brominated were determined in ringed seal (*Phoca hispida*) blubber collected from subsistence hunts in the Canadian Arctic in 1981, 1991, 1996, and 2000 (Ikonomou et al. 2002). Total PBDE (Σ PBDE) concentrations have increased exponentially over this period in male ringed seals aged 0-15 years. Penta and hexa-BDEs are increasing at approximately the same rate ($t_2 = 4.7$ and 4.3 years, respectively) and more rapidly than tetra-BDEs ($t_2 = 8.6$ years) and tri-BDEs ($t_2 = \infty$) in this age/sex grouping. In contrast to declining PBDE concentrations since 1997 in human milk from Sweden, Σ PBDE concentrations in arctic ringed seals continue to increase exponentially similar to worldwide commercial penta-BDE production. PBDE congener profiles in male ringed seals aged 0-15 years from 1991 to 2000 also differ significantly from other aquatic organisms and semipermeable membrane devices collected from temperate coastal regions of British Columbia. While PBDE concentrations are 50 times lower than those of mono-ortho and non-ortho PCBs, and ~500 times higher than PCDD/Fs, the data indicate that, at current rates of bioaccumulation, PBDEs will surpass PCBs to become the most prevalent organohalogen compound in Canadian arctic ringed seals by 2050.

New information on levels in sediments

de la Cal et al. (2003) determined seven BDE congeners (BDE47, BDE99, BDE100, BDE118, BDE153, BDE154 and BDE183) in two river sediments and five marine sediments collected around Barcelona and Tarragona in Catalonia, NE Spain. Σ BDE concentrations ranged from 0.86 to 2.49 $\mu\text{g kg}^{-1}$ dry weight. In the river sediments BDE99 was present at the highest concentrations, followed by BDE183, whilst in the marine sediments BDE183 was always present at the highest concentration.

Moche and Stephan (2003) determined BDEs (17 congeners) in 13 sediments and 2 samples of suspended particulate matter (SPM) from the River Danube in Austria. Σ BDE concentrations in sediments ranged from 0.29 to 10.4 $\mu\text{g kg}^{-1}$ dry weight, and were 0.38 and 1.1 $\mu\text{g kg}^{-1}$ dry weight in the SPM samples. BDE47 and BDE99 dominated the sediment profiles.

Sawal et al. (2004) determined BDEs (congeners BDE28, BDE47, BDE66, BDE71, BDE75, BDE85, BDE99, BDE100, BDE138, BDE153, BDE154, BDE183, BDE190 and BDE209) in surface sediment samples from the River Elbe, Germany and the Czech Republic, taken in 2002 using GC-ECNIMS. BDE209 was detected at concentrations ranging from 0.5 to 17 $\mu\text{g kg}^{-1}$ dry weight (10 to 230 $\mu\text{g kg}^{-1}$ organic carbon; $n = 29$), representing almost 80% of Σ BDE. Of the other congeners studied, BDE47, BDE85, BDE99, BDE100, BDE153, BDE154 and BDE183 were detected in most samples at low concentrations. In the Czech Republic, a few samples showed higher percentages of tetra- to hexabrominated congeners, suggesting local inputs of the penta-mix PBDE formulation.

Kilemade et al. (2004) determined BDEs (congeners BDE28, BDE47, BDE66, BDE71, BDE75, BDE77, BDE85, BDE99, BDE100, BDE119, BDE138, BDE153, BDE154, BDE183, BDE190 and BDE209) and HBCD (using GC-EIMS) in the $< 63 \mu\text{m}$ fraction of surface sediments from three sites in Cork Harbour, Ireland and a reference site. BDEs were detected at low concentrations at two of the harbour sites, with Σ BDE concentrations of 1.8 and 1.9 $\mu\text{g kg}^{-1}$ dry weight.

Schlabach et al. (2004) determined BDEs (congeners BDE28, BDE47, BDE49+71, BDE99, BDE100, BDE138, BDE153, BDE154, BDE183 and BDE209) and HBCD and TBBP-A in sediments from Lake Mjøsa, Norway, using GC-EIMS. Σ BDE concentrations were in the range 0.6 to 27 $\mu\text{g kg}^{-1}$ dry weight, with the highest concentrations near the inlet at the northern end of the lake and the lowest close to the outlet in the south. Tri- to hexa-BDEs (BDE47, BDE99, BDE100, BDE153 and BDE154) constituted 60-70% of Σ BDE in sediments from the northern part of the lake, whilst elsewhere BDE209 dominated (50-90% of Σ BDE).

Two 50 cm sediment cores were collected from the Scheldt estuary in 2000 and BDEs (BDE28, BDE47, BDE66, BDE85, BDE99, BDE100, BDE138, BDE153 and BDE154) determined with depth (Covaci et al., 2002a). Σ BDE concentrations in the two cores ranged from 1.4 to 179 and 1.4 to 272 $\mu\text{g kg}^{-1}$ dry weight, respectively. The BDE profile matched the composition of the commercial penta-mix formulation, and the concentrations showed a steep increase in the surface layers, probably during the few years immediately prior to sampling, although the cores were not dated.

Voorspoels et al. (2004a) have analysed sediments from the Belgian North Sea (BNS), the Western Scheldt Estuary (SE) and freshwater watercourses from the Scheldt basin for eight BDE congeners (BDE28, BDE47, BDE99, BDE100, BDE153, BDE154, BDE183 and BDE209). Concentrations of summed tri- to hepta-BDEs in the sediments were $< 0.2 \mu\text{g kg}^{-1}$ dry weight for samples from the BNS, whilst in those from the SE this sum ranged from < 0.20 to $0.41 \mu\text{g kg}^{-1}$ dry weight. Compared to the marine and estuarine locations, the sediments from the freshwater watercourses were relatively more polluted with tri- to hepta-BDEs ($< 0.20 - 19 \mu\text{g kg}^{-1}$ dry weight), while concentrations of BDE209 were up to $320 \mu\text{g kg}^{-1}$ dry weight.

Three dated sediment cores from locations in western Europe were analysed 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 pentamix were levelling off in the most recent layers (1995 & 1997 – concentrations normalised to total organic carbon), whereas those in the Drammenfjord were still increasing in 1999. The absence of all BDE congeners in the older (deeper) layers of all 3 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. In the case of the Lake Woserin core, the concentrations of BDE congeners observed are probably due entirely to atmospheric transport.

New information on levels in seafood and fish in Europe

Covaci et al. (2002b) determined seven BDE congeners (BDE28, BDE47, BDE99, BDE100, BDE153, BDE154 and BDE183) in fish from the Danube Delta, a large wetland in Romania. Σ BDE concentrations in eleven species of fish ranged from < 0.1 to $14 \mu\text{g kg}^{-1}$ lipid weight. There was no clear relationship between feeding pattern (benthic, herbivorous, omnivorous or piscivorous), but the highest concentrations were seen in carp. The Σ BDE concentrations in sediments from the same locations as those at which fish were collected were $< 0.1 \mu\text{g kg}^{-1}$ dry weight, while Σ BDE levels between 2.9 and $6.5 \mu\text{g kg}^{-1}$ lipid weight were found in cormorant liver.

Asplund et al (2004) investigated spatial and temporal trends of BDEs, MeO-BDEs and HBCD in herring from the Swedish coast using GC-ECNIMS. The BDE congeners determined were BDE47, BDE99, BDE100, BDE153 and BDE154; and the methoxylated derivatives studied were 6-MeO-BDE47 and 2'-MeO-BDE68. No significant time trends were apparent for any of the compounds determined, and it seems likely that the MeOBDEs are of natural origin. Concentrations of BDE47 (the dominant congener) were similar in fish from both the Baltic and North Seas. This is in contrast to the situation for PCBs, for which concentrations are about five times higher in the Baltic, and suggests that the sources and inputs of these two groups of chemicals are quite different.

Eljarrat et al. (2004b) determined HBCD (by GC-ECNIMS) and BDEs (40 congeners, but only 16 detected) in barbel (*Barbus graellsii*) from 4 sites in the Cinca River, NE Spain, a tributary of the River Ebro. Σ BDE concentrations ranged from 0.2 to 279 and 1.3 to $298 \mu\text{g kg}^{-1}$ wet weight in the same tissues. The congener profile was dominated in most cases by BDE153 and BDE154, and BDE47 was always a minor component. Eljarrat et al.

(2004c) also determined HBCD (by GC-ECNIMS) and BDEs (BDE47, BDE153, BDE154, BDE183 and BDE209) in bleak (*Alburnus alburnus*) from 3 sites in the Cinca River. As for the barbel from the same area, the congener profile from the two most contaminated sites was dominated by BDE153 and BDE154, and BDE47 was a minor component.

Polar cod (*Boreogadus saida*) from Svalbard (Wolkers et al., 2004) yielded a geometric mean BDE47 concentration of $2.1 \mu\text{g kg}^{-1}$ lipid weight, with BDE47 representing ca. 60% of Σ BDE (22 congeners). Spatial and temporal trends of BDEs (congeners BDE28, BDE47, BDE100 and BDE154) and HBCD were studied in polar cod and Atlantic cod from Norwegian waters (Bytingsvik et al., 2004). Polar cod were collected from Bear Island, and Atlantic cod at two sites in south (Hvaler) and central (Froan) Norway in 2003. Cod from Hvaler taken in 1998 were also available for analysis. Concentrations of all compounds determined decreased in the order Hvaler > Froan > Bear Island in 2003. At Hvaler, an increase of 3 – 4 times was seen in wet weight concentrations of all BDE congeners over the period 1998-2003, and an eightfold increase for HBCD. On a lipid weight basis the increases were more modest, 1.5 times and 3 to 4fold, respectively.

Livers from dab (*Limanda limanda*), and flounder (*Plathychthys flesus*) have been analysed for a range of BDE congeners

(BDE28, BDE47, BDE66, BDE71, BDE75, BDE85, BDE99, BDE100, BDE138, BDE153, BDE154, BDE183, BDE190 and BDE209) (Lepom, unpublished data). With the exception of BDE71, BDE190 and BDE209, all congeners were found at detectable concentrations in at least some of the samples. Σ BDE concentrations (sum of the tri- to hexa-brominated congeners) in individual liver samples ranged from 1 to 26 $\mu\text{g kg}^{-1}$ wet weight (mean 1.7 to 15.6) corresponding to 5 to 100 $\mu\text{g kg}^{-1}$ lipid weight (mean 7.5-70.9). BDE congener patterns in the two fish species were similar, with BDE47 as the dominant congener followed by BDE100 and BDE154. BDE99, vulnerable to metabolism in freshwater fish (Stapleton et al., 2004a) was found at much lower concentrations than would be expected from its percentage composition in the technical penta-mix formulation. This suggests that there are similar metabolic pathways for this congener in marine fish species. There were pronounced differences in liver BDE concentrations of flatfishes from various locations (Figure 2). Although the food of dab varies with local conditions, it eats any bottom-living invertebrate which is abundant locally and which is small enough to be captured (small crustacean, molluscs, polychaetes and other worms), the differences seen in BDE concentrations indicated geographical differences in the level of contamination. The highest BDE concentrations were found in the central North Sea (station P01) near a Danish oil and gas production area (Danfield) and off the British coast (stations N04, N06). The lowest BDE concentrations were seen in dab and flounder from the Baltic Sea (stations B1, B11). Voorspoels et al. (2003) reported BDE concentrations in dab liver from the Western Scheldt Estuary (a highly polluted estuary) and from the Belgian North Sea which were in the same order of magnitude as those from sampling sites near the British coast and at Danfield. The Tees estuary has previously been shown to be a major source of BDE congeners deriving from the penta-mix formulation to the North Sea (Boon et al., 2002), but in the light of the data reported here the Danish gas and oil production area has also to be considered as a possible major source of tri- to hexa-BDE congeners to the central North Sea. Lipidnormalised BDE47 concentrations in cod (*Gadus morhua*) liver samples taken at three sites (N04, B01, B11) ranged from 14 to 35 $\mu\text{g kg}^{-1}$ and were much lower than those reported in previous studies in cod from the North Sea (de Boer, 1989; de Boer and Allchin, 2001; Boon et al., 2002). There were no marked differences in BDE levels in cod from the three locations.

Paepke and Herrmann (2004) determined BDEs (congeners BDE17, BDE28, BDE47, BDE66, BDE77, BDE99, BDE100, BDE153, BDE154, BDE183 and BDE209) in fish from the North Sea (herring, *Clupea harengus*) and the North-east Atlantic (plaice, *Pleuronectes platessa*; trout, *Salmo trutta*; and halibut, *Hippoglossus hippoglossus*) using GC-HRMS. The fish were bought from markets in Germany. Σ BDE concentrations in fish muscle were 14, 6.7, 9.7 and 0.4 $\mu\text{g kg}^{-1}$, respectively, expressed on a lipid basis. Hites et al. (2004a) determined BDEs (43 congeners) in wild and farmed salmon collected around the world. Σ BDE concentrations were significantly higher in farmed salmon from Europe (especially Scotland) than in those from North America and Chile, and than those in wild salmon, up to ca. 5 $\mu\text{g kg}^{-1}$ wet weight, similar to concentrations reported earlier by Jacobs et al. (2002). The congener profiles were dominated by BDE47, then BDE99 and BDE100. Bethune et al. (2004) determined BDEs (congeners 28, BDE47, BDE99, BDE100, BDE153 and BDE154) in fish (salmon, mackerel *Scomber scombrus*, herring, cod and halibut), blue mussels and edible crabs *Cancer pagurus* from Norway collected in 2003. Analysis was by GC-ECNIMS. Σ BDE concentrations ranged from 1.1 to 4.5 $\mu\text{g kg}^{-1}$ wet weight in salmon, 1.3 to 1.8 $\mu\text{g kg}^{-1}$ wet weight in mackerel, 1.0 to 3.5 $\mu\text{g kg}^{-1}$ wet weight in herring, 0.3 to 18 $\mu\text{g kg}^{-1}$ wet weight in halibut and 0.02 to 0.04 $\mu\text{g kg}^{-1}$ wet weight in cod, all samples being of muscle. Cod liver contained 5.2 to 9.5 $\mu\text{g kg}^{-1}$ wet weight. Crab muscle contained 0.03 to 0.07 $\mu\text{g kg}^{-1}$ wet weight, and shell meat 0.6 to 7.0 $\mu\text{g kg}^{-1}$ wet weight. Σ BDE concentrations in blue mussels were 0.06 to 0.25 $\mu\text{g kg}^{-1}$ wet weight. BDE47 accounted for 60-70% of Σ BDE, and BDE99 and BDE100 were the next most abundant congeners.

Johansson et al. (2004) determined BDEs (congeners BDE28, BDE47, BDE49, BDE66, BDE77, BDE85, BDE99, BDE100, BDE138, BDE153, BDE154, BDE183 and BDE209) in archived, freeze-dried, blue mussel samples from the Seine estuary, France, using GCECNIMS. Over the period 1982 to 1993, there was an exponential increase in BDE concentrations. Between 1993 and 1997 levels were constant, followed by a further increase to a maximum in 1999 and 2001 (peak Σ BDE concentration 24 $\mu\text{g kg}^{-1}$ dry weight), and by a significant decline after 2002 (Figure 3). If this decline has resulted from the reduction in the usage of the penta-mix PBDE formulation in Europe, then it has occurred much more rapidly than that of PCB levels following their earlier prohibition.

Gama et al. (2004) determined BDEs (congeners BDE28, BDE47, BDE66, BDE71, BDE75, BDE77, BDE85, BDE99, BDE100, BDE119, BDE138, BDE153, BDE154, and BDE190) in riverine fish (*Barbus* spp.) and coastal mussels (*Mytilus galloprovincialis*) from Portugal using GC-ECNIMS. Lower concentrations were observed in mussels than fish. For BDE47, the concentration in mussels was approximately 2 $\mu\text{g kg}^{-1}$ dry weight, whilst in fish muscle and liver the ranges were not detected to 7.9 $\mu\text{g kg}^{-1}$ dry weight and 1.6 to 30 $\mu\text{g kg}^{-1}$ dry weight, respectively.

Covaci et al. (2004) have investigated the levels and distribution of BDEs (congeners BDE28, BDE47, BDE49, BDE66, BDE85, BDE99, BDE100, BDE153, BDE154 and BDE183) in zebra mussels and several freshwater fish species (eel, carp and gibel carp *Carassius gibelio*) at various sites in Flanders, Belgium. At most sites, individual BDE congeners were present at detectable levels in whole zebra mussel tissue, with Σ BDE concentrations ranging from 0.15 to 1.8 $\mu\text{g kg}^{-1}$ wet weight. Except for one site (Blokkeerdijk, Antwerp), where sum BDEs was < 0.1 $\mu\text{g kg}^{-1}$ wet weight in carp muscle, all fish samples from other sites contained detectable levels of BDEs, with the highest concentrations (14 \pm 14 $\mu\text{g kg}^{-1}$ wet weight) being measured in eel liver from Watersportbaan (Ghent). The sampled sites covered a broad concentration range of organohalogenated pollutants with the highest values consistently found in eel liver. Generally, correlations between concentrations of BDEs and of CBs, HCB and *p,p'*-DDE for each species were low ($r < 0.50$) and most were not statistically significant ($p > 0.05$).

(e) Exposure in local areas (provide summary information and relevant references)	
- general	<p><u>New Information:</u></p> <p><u>Exposure around municipal solid waste incineration facilities</u></p> <p>A recent study has investigated whether release of BDEs takes place at a municipal solid waste incineration (MSW) facility (Agrell et al., 2004; ter Schure et al., 2004b). The rationale was that MSWs handle large quantities of waste and that the amounts of collected electronic waste are steadily increasing, and that plastic products may contain up to 32% by weight of PBDE (WHO, 1994). During the year 2000, 2.35 Mt of household and industrial waste were incinerated at 22 plants in Sweden, and the volume of waste is increasing at approximately 2,500 tpa. The MSW plant studied, in Malmö, received 200 kt of waste in 2002, of which 2 kt was electronic waste. BDE congeners BDE28, BDE47, BDE66, BDE100, BDE153, BDE154, BDE183 and BDE209 were determined using GCECD. Data obtained were compared with similar measurements made at an urban reference site, where manufacture of concrete and asphalt is conducted, since this type of industry makes no use of brominated flame retardants. ΣBDE concentrations (gaseous + particulate phase concentrations) ranged from 2.2 and 21.3 pg m⁻³ at the MSW site, with a median value of 6.3 pg m⁻³ (n = 17) and from 0 to 26.7 pg m⁻³ at the reference site, with a median value of 3.5 pg m⁻³ (n = 19) (Agrell et al., 2004). Median values of BDE47 and BDE209 were 2.1 and 10.4 pg m⁻³ at the MSW and 1.7 and 6.5 pg m⁻³ at the reference site. Compared to other studies, the concentrations observed were relatively low, possibly as a result of sampling during October-March when temperatures and so evaporation are relatively low. Atmospheric bulk deposition samples were also collected and analysed (ter Schure et al., 2004b). Between 75 and 95% of the deposited BDEs consisted of BDE47, BDE99 and BDE209. Concentrations in rain were significantly higher at the MSW than at the reference site, with median dry and wet deposition fluxes for ΣBDE of 18.4 and 21.6 ng m⁻² day⁻¹; and 7.0 and 6.8 ng m⁻² day⁻¹, respectively. The authors suggest that treatment of waste is currently a source of “old” PBDEs to the environment, whereas the observed concentrations of BDE209 represent diffuse leakage from products still in current use. The emission of BDEs to the atmosphere from waste treatment should be considered as an important source when assessing the transport of these substances in the environment (Agrell et al., 2004).</p> <p>Seven BDE congeners (BDE28, BDE47, BDE99, BDE100, BDE153, BDE154 and BDE183) were determined in large (2 to 6 kg weight) brown trout (<i>Salmo trutta</i>) from five lakes in SE Norway (Mariussen et al., 2003). The mean ΣBDE concentrations in trout from Lake Mjøsa were much higher than in those from the other four lakes, 353 μg kg⁻¹ wet weight compared to 3.6 to 18 μg kg⁻¹ wet weight in trout from the other lakes (equivalent to 5,280 and 161 to 616 μg kg⁻¹ on a lipid basis). The highest ΣBDE concentration observed was 1,120 μg kg⁻¹ wet weight (17,400 μg kg⁻¹ on a lipid basis). In whole-body homogenates of burbot (<i>Lota lota</i>), ΣBDE concentrations up to 2,270 μg kg⁻¹ wet weight (45,100 μg kg⁻¹ on a lipid basis) were observed. Lake Mjøsa is located in a relatively densely populated and industrialised area, and the study suggests that there is a local source of congeners deriving from the commercial penta-mix formulation. HBCD was detected in fish of a number of species from Lake Mjøsa and further downstream at concentrations from 90 to 800 μg kg⁻¹ lipid (Schlabach et al., 2004).</p> <p>Allchin & Morris (2003) determined HBCD (by LC-MS) and BDEs (BDE28, BDE47, BDE99, BDE100, BDE153 and BDE154) in edible fish (brown trout and eel <i>Anguilla anguilla</i>) from the Rivers Skerne and Tees in NE England, downstream of a brominated flame retardant manufacturing plant. BDEs were detected in fish from all sites, and the lowest concentrations were found in the two upstream Tees sites above the Skerne confluence. Mean ΣBDE concentrations in trout at these sites were 4.9 and 5.3 μg kg⁻¹ wet weight. The highest concentrations (mean and maximum ΣBDE concentrations 118 and 197 μg kg⁻¹ wet weight) were observed just downstream of the factory in which PBDE formulations had previously been manufactured (Allchin et al., 1999), and declined with distance downstream to 23 μg kg⁻¹ wet weight at Croft-on-Tees, below which trout could not be caught. Mean ΣBDE concentrations in eels sampled at 4 sites further downstream in the River Tees, as far as the tidal barrage, ranged from 130 to 235 μg kg⁻¹ wet weight. The congener profiles, in which BDE47 dominated, reflected exposure primarily to pentamix congeners. Despite the cessation of production of PBDE formulations in the late-1990s, BDE concentrations remain high in edible fish from the Rivers Skerne and Tees.</p>

The results of this study are in line with the earlier studies which indicated widespread contamination of the Rivers Skerne and Tees, Tees Bay, and the western North Sea with BDEs (Allchin et al., 1999; Allchin and de Boer, 2001; de Boer and Allchin, 2001; Boon et al., 2002).

Levels in sewage sludge samples

Relatively few studies have been conducted of the contamination of sewage sludge in Europe, despite the fact that sewage sludge is considered to be one of the main sinks for polybrominated diphenyl ethers. The application of sewage sludge to agricultural land in many countries also increases the possibility of subsequent remobilization of these compounds.

The Swedish Environmental Protection Agency initiated a larger study to determine the extent of the contamination in sewage sludges from STPs across Sweden. As a result, sewage sludge samples were collected in 2000 from 50 STPs and were analyzed for BDE47, BDE99, BDE100, BDE153, BDE154, BDE209, HBCD (determined using GC-ECNIMS), TBBP-A and BB209. Samples represented major cities (Stockholm, Gothenburg, Malmö), towns with possible point sources and STPs of differing sizes randomly scattered around the country. BDE209 was the compound usually found at the highest concentrations in most sludges. The congener profile of congeners BDE47, BDE99, BDE100, BDE153 and BDE154 in all sludges was similar to that of the penta-mix PBDE technical product Bromkal 70-5DE, which is probably the original source. Concentrations of the lower brominated BDEs were fairly similar in all sewage sludge samples, indicating diffuse leaching of these from products into wastewater streams (Table 2). The lack of point sources for this product is probably a result of the phasing out of the penta-mix product in the EU and the fact that Sweden no longer imports such products.

Christensen et al. (2003) determined BDEs (BDE17, BDE28, BDE47, BDE49, BDE66, BDE85, BDE99, BDE100, BDE153, BDE154, BDE183 and BDE209) in a sewage sludge sample from Bjergmarken STP in Roskilde, Denmark. The Σ BDE concentration (all congeners except BDE209) was $238 \pm 23 \mu\text{g kg}^{-1}$ dry weight, and of BDE209 was $248 \pm 81 \mu\text{g kg}^{-1}$ dry weight.

Sewage sludge from 6 STPs in Spain collected during 2002 were analysed for BDEs (congeners BDE7, BDE15, BDE17, BDE28, BDE47, BDE49, BDE66, BDE85, BDE99, BDE100, BDE119, BDE153, BDE154 and BDE209) by Fabrellas et al. (2004). All congeners except BDE209 were determined using GC-EIMS/MS, and BDE209 using GC-ECNIMS. In all cases the dominant congener was BDE209 (93-99%) with minor quantities of BDE47, BDE99, BDE100, BDE153 and BDE154. Σ BDE concentrations ranged from 844 to 18,100 $\mu\text{g kg}^{-1}$ dry weight, and the highest concentration was associated with an industrial area with mainly textile manufacture.

Moche and Thanner (2004) determined BDEs (BDE11, BDE17, BDE25, BDE28, BDE47, BDE49, BDE77, BDE99, BDE100, BDE116, BDE138, BDE140, BDE153, BDE154, BDE155, BDE166, BDE181 and BDE183) in effluents and sludges from 36 STPs and wastewater treatment plants in Austria. The dominant congeners were BDE47, BDE99, BDE100, BDE153 and BDE183, and no significant differences were observed between urban and industrial effluents, with the single exception of one effluent from a textile factory. This showed much higher concentrations (Σ BDE 186 ng l⁻¹, exceeding the maximum for the other samples by approximately a factor of 4 times) ca. 40% of which was contributed by BDE183.

New information on levels in soil and sediment from an electronic waste recycling facility

Soil and sediment collected in the vicinity of an open electronic waste disposal and recycling facility located in Guiyu, Guangdong, China, were analyzed for the levels of common polybrominated diphenyl ethers (PBDEs) by using soxhlet extraction and gas chromatography/mass spectrometry (Wang et al. 2005). The PBDEs were detected in the soil and sediment samples at levels of 0.26–824 ng/g (dry weight). The obtained isomer profiles of PBDEs from the two soil samples collected from different contamination sites were found to be similar to various technical formulations of fire retardant products. Concentrations of PBDEs detected in sediment and soil samples (ng/g, dried weight):

Penta-BDE	sediment	soil 1	soil 2
BDE-85	nd	nd	nd
BDE-100	0.89	2.70	89.4
BDE-99	6.87	13.3	615
BDE-119	nd	nd	nd
BDE-126	nd	nd	nd

the concentrations of total penta-BDEs in Soil 2 were approximately 20 and 40 times, respectively, higher than those in Soil 1. Soil 2 was collected in the vicinity of a site used for the dumping and dismantling of printer rollers. The high levels of the PBDE congeners in this soil sample probably derived from the commercial penta-BDE product used in the fire retardants applied to printers. In contrast, Soil 1, was collected adjacent to the site used for burning plastic materials.

Levels of total PBDEs in the soil samples collected near the e-waste dumps were higher than those in the sediment taken from the nearby Lianjiang River. However, it is apparent that the river sediment was contaminated from e-waste processing activities such as the dumping, dismantling and burning of waste materials. Uncontrolled recycling and disposal of e-wastes by simple dismantling, acid treatment, and open burning have resulted in severe soil contamination and migration of PBDEs into the river system. However, data is limited and sample collection at the study sites was restricted.

Information from: A review on human exposure to brominated flame retardants – particularly polybrominated diphenyl ethers.
By Andreas Sjødin et al. 2003.

Increasing levels have been observed in mother's milk from Sweden as well as in blood from Germany and Norway. The levels are in general lower than PCB levels. However, the PBDE concentrations found in the North America are considerably higher compared to European subjects. The PBDEs are dominated by BDE-47 (tetra). While the lower and medium brominated diphenyl ethers are persistent, BDE-209 (deca) has fairly short half-life of approximately 2 weeks.

Brominated flame retardant (BFR) make their way to human population primarily via food intake in cases when the compounds are persistent enough to be biomagnified in the food web similarly to other persistent chemicals as has been shown for BDE-47 in humans consuming large quantities of Baltic Sea fish (Sjødin et al., 2000). This means that fatty fish from contaminated areas are a major source (Sjødin et al., 2000, Sjødin 2000) and mother's milk is a source (Meironyte et al., 1999, Meironyte Guvenius, 2002) for the nursing child. Swedes who did not consume fish had a median PBDE level of 0.4 ng/g l.w. compared to 2.2 ng/g l.w. for the high consumers of fish who ate between 12 and 20 meals of fatty Baltic Sea fish per month (Sjødin et al., 2000).

The concentration of BDE-47 (the dominating congener in non-occupationally exposed subjects) increased in the general European population from the early 1970s until the mid-1990s. Interestingly, one of these studies (Meironyte Guvenius, 2002) shows a decrease in concentration starting in the latter part of the 1990s. Finding a decreasing trend of PBDEs in Sweden is encouraging news and is further supported by data on Guillemot eggs from Stora Kalsø in the Baltic proper (Sellström, 1999), which also show decreasing levels of PBDEs. However, the trend-line on Guillemot eggs starts to decrease in the mid-1980s, indicating a potential lag-time of a decade before a positive effect in the environment was translated to decreasing levels in the human population. The levels of breast adipose tissue taken from women living in San Francisco Bay area (She et al., 2002) in 2000 are almost two orders of magnitude higher than what has been reported in human milk from Sweden (Meironyte et al., 1999).

It has been reported that PBDEs are present in chickens originating from North America. The levels in poultry were reported to be 1.8 -3.9 ng/g l.w for the sum of tri- to decaBDE (Huwe et al., 2002). Tri- to hexaBDE levels in spinach, potato and carrot have been reported in Japan to be 134, 47.6, and 38.4 ng/g fresh weight (Ohta et al., 2002), respectively. The same article also reported 63.4 ng/g in pork, 16.2 ng/g in beef and 6.25 ng/g in chicken. The total daily intake of PBDEs has been estimated to be 44 ng/g in Canada by compiling PBDE concentrations data from 40 common commercial food items and food consumption data (Ryan and Patry, 2001).

Eleven samples from North America of biosolids destined for land-application had levels of 1100-2290 ng/g of tetra- to hexa-BDE (Hale et al., 2001). A potential route of human exposure to PBDE is the application of biosolids in agricultural areas; this route of exposure should be investigated further.

Also, the presence of persistent BFRs in ambient air in indoor work and/or home environment may cause a no negligible exposure of these chemicals in humans.

Occupational exposure:

In Sweden, occupational exposure to PBDE has been identified among electronics recycling personnel (Sjodin et al., 1999), technicians responsible to repair and maintain computers (Jacobsson et al., 2002), workers adding decaBDE during rubber manufacturing (Turesson et al., 2002), and employees working at facilities manufacturing electric cables using the same rubber (Thureson et al., 2002). Findings demonstrate that there was occupational exposure at recycling plants to BDE-183 and BDE-209 and to a lesser extent exposure to BDE-47, BDE-153 and BDE-154.

Other articles on human exposure

It has been performed a study in Sweden (Fängström et al. 2005) to assess 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. BDE-47, -99 and -100 concentrations reached a peak in the mid 1990's and are now clearly showing decreasing levels. The concentrations are however still much higher than in 1980.

In a Canadian study (Wilford et al. 2005) following up a previous investigation into indoor air levels of PBDEs. House dust was analyzed from the family vacuum cleaners of 68 of the same 74 randomly selected homes, in Ottawa, Canada during the winter of 2002- 2003. Correlations were found between pentamix congener levels in dust and in air from the same homes. Exposure scenarios are presented for mean and high dust ingestion rates, and compared against exposures from other pathways, for both adults and toddlers (6 months-2 years). Assuming a mean dust ingestion rate and median dust and air concentrations, adults would be exposed to ca. 7.5 ng ^aPBDE d-1 via the dust ingestion pathway, which represents ~14% of total daily exposure when compared to diet (82%) and inhalation (4%). However, for toddlers the equivalent intakes would be 99 ng d-1, representing 80% of their daily PBDE exposure. At high dust ingestion rates these values increase to 180 ng d-1 (80% daily intake) for adults and 360 ng d-1 (89% daily intake) for toddlers. The data give a clearer picture of sources of PBDE exposure in the home environment and suggest that dust could be a significant exposure pathway for some individuals, particularly children.

High-volume samples of PeBDE was collected for indoor and outdoor air to (Shoeib et al. 2004). Mean indoor air concentrations (pg/m³) were 630 (ΣPeBDE). The ratios of concentrations between indoor and outdoor air were 15 for ΣPeBDE.

Netherlands institute for fisheries research has monitored Brominated flame retardants in office dust samples in Parliament buildings in 8 European countries. The results demonstrate that the possibility of continuous exposure of these compounds via inhalation of particles in offices is potentially an important route of human exposure.

Information from: Polybrominated Diphenyl Ethers in Indoor Dust in Ottawa, Canada: Implications for Sources and Exposure. By Wilford et al., 2005.

This study follows a previous investigation into indoor air levels of PBDEs. House dust was analyzed from the family vacuum cleaners of 68 of the same 74 randomly selected homes, in Ottawa, Canada during the winter of 2002-2003. PBDEs, comprising on average 42 % bde-209, were found in all samples. The levels were log-normally distributed with a geometric mean Σ PBDE of 2000 ng/g dust. Assuming a mean dust ingestion rate and median dust and air concentrations, adults would be exposed to ca. 7.7 ng Σ PBDE/day via dust ingestion pathway, which represents ~14 % of total daily exposure when compared to diet (82 %) and

<p>- as a result of long-range environmental transport</p>	<p>inhalation (4 %). However, for toddlers the equivalent intake would be 99 ng/day, representing 80 % of their daily PBDE exposure. At high dust ingestion rates these values increase to 180 ng/day (80 % daily intake) for adults and 360 ng/day (89 % daily intake) for toddlers.</p>
	<p>Information from: <u>Levels of brominated flame retardants in human samples from Norway through three decades. By Thomsen et al.</u></p> <p>The objective of this study was to complete and extend a previous study on time trends of PBDEs in Norwegian pooled serum samples. We further compare these levels with levels in other human samples from Norway in order to put together an overview of the PBDE body burden in the general population from 1977 to 2004. The temporal trend of the sum of 7 PBDEs (28, 47, 99, 100, 153, 154 and 183) in the pooled serum from the present study are in close agreement with the levels found in the previous study, except for the pools from 1991 and 2002 which were found to be considerably higher than expected from earlier results of preceding and following years. This was surprising as the pools contained at least 20 individual samples (mean aged 40 – 50 years). In the samples from 2002, the mean of sum 7 PBDEs is 3.8 ng/g lipid (serum from the youngest group excluded) and 3.5 ng/g lipids in men age 25-59 years. 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 have risen rapidly from 1977 to about 1997, but now seems to have stabilized or even to have decreased. This is in accordance with the trends observed in Swedish breast milk. 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 elder persons. This finding was confirmed in the present study. However, it seems that in 2002 also children of age 5-14 years show higher levels of PBDEs than average adult.</p>
	<p><u>New Information on concentrations, distribution and behaviour in the atmosphere, and deposition in local areas:</u></p> <p>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 brominated diphenyl ether (BDE) congeners were determined (BDE17, BDE28, BDE47, BDE85, BDE99, BDE100, BDE153, BDE154, BDE183 and BDE209). 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. ΣBDE concentrations were dominated by BDE209, which still has primary sources within the EU following the phasing-out of the penta-mix formulation, followed by BDE47 and BDE99. These background concentrations may therefore reflect either a change in use of commercial BDE formulations, or the greater affinity of BDE209 for particles relative to BDE47 and BDE99. As PCB concentrations in Baltic air have been declining, the input of BDEs (especially BDE209) by atmospheric deposition to the Baltic Proper now exceeds that of the PCBs by a factor of almost 40 times.</p> <p>Lee et al. (2004) determined atmospheric concentrations of BDEs at 2 rural/semi-rural sites in England, and 1 remote site on the west coast of Ireland in 2001 and in 2000, respectively (n = 108). 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 commercial penta-mix PBDE formulation. At the two English sites in the summer, BDE concentrations were strongly influenced by temperature, indicating that land/air exchange processes play an important role in determining atmospheric concentrations.</p> <p>Jaward et al. (2004a) studied a total of 71 passive air samples using semi-permeable membrane devices (SPMDs) for eight BDE congeners (BDE28, BDE47, BDE49, BDE75, BDE99, BDE100, BDE153 and BDE154) during a six week period in 2002 at remote/rural/urban locations across 22 countries in Europe. BDEs were detected in ca. 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</p>

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. BDE47 and BDE99 contributed ca. 75% to Σ BDE, similar to their proportion in the Bromkal 70-5DE pentamix technical product.

In a subsequent study, Jaward et al. (2004b) determined the same eight BDE congeners in passive air samples collected at 11 sites along a latitudinal transect from the south of the UK to the north of Norway during 2000-2002. Equivalent concentrations of Σ BDE in air at background locations were 2.0 (range 1.1 to 2.5) and 1.1 (range 0.8 to 1.6) $\mu\text{g m}^{-3}$ in the UK and Norway, respectively. The proportion of the lighter BDEs decreased with increasing latitude while the heavier compounds were more evenly distributed, indicating that fractionation of the BDEs was occurring during atmospheric transport.

Harrad & Hunter (2004) determined BDEs (congeners BDE28, BDE47, BDE99, BDE100, BDE153 and BDE154) in passive air samples collected during 2003 using polyurethane foam (PUF) discs at 11 sites in the West Midlands (UK) centred on the city of Birmingham. Atmospheric Σ BDE concentrations ranged from 1.3 to 6.7 $\mu\text{g m}^{-3}$, and higher concentrations were observed in samples from urban/suburban sites than at semirural/rural sites, with about a five-fold difference in concentration. The profiles were dominated by BDE47 and BDE99. In an earlier study, Harrad et al. (2004) determined BDEs (same congeners less BDE28) in 6 outdoor air samples collected in the city in July and August 2002 and April and May 2003. Σ BDE concentrations were in the range 10 to 33 $\mu\text{g m}^{-3}$.

New information on concentrations in precipitation and wet deposition

BDEs were determined in precipitation falling in southern Sweden during a 2 week period in 2000 (ter Schure and Larsson, 2002). The particle-associated and “dissolved” phases were separated during sampling. $65 \pm 18\%$ of Σ BDE was found to be particle-associated. The volume weighted mean concentration of Σ BDE (9 congeners) in rain was 209 $\mu\text{g l}^{-1}$, and the total deposition rate was 2 ± 1 $\text{ng } \Sigma$ BDE $\text{m}^{-2} \text{day}^{-1}$. The congener profile in both phases of the total deposition was dominated by BDE209, and thereafter BDE47, BDE99 and BDE183, representing inputs from all three commercial PBDE formulations. As has been observed for other POPs (persistent organic pollutants), it seems that particle associated BDEs are effectively removed during small precipitation episodes, and that particle scavenging is an important mechanism for the wet deposition of BDEs.

New information on distribution in sediments in remote areas supporting the conclusion on long range transport

Malmquist et al. (2003) analysed sediment cores from seven lakes in west Greenland for BDE47 using GC-EIMS. Concentrations of BDE47 were low, with maximum core concentrations in the range 7 to 51 ng kg^{-1} dry weight. The temporal trend apparent from four of the cores suggests the appearance of BDE47 in the lake sediments around 1950- 1960, with generally increasing concentrations towards more recent years. Atmospheric transport, probably from lower latitudes, was suggested as the source of the organohalogen contaminants detected.

New information on distribution in soil in remote areas supporting the conclusion on long range transport

BDEs 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 Σ BDE ranged from 65 to 12,000 ng kg^{-1} dry weight. BDE congeners BDE47, BDE99, BDE100, BDE153 and BDE154, the major constituents of the commercial pentamix PBDE formulation, dominated the average congener pattern in the soils. This was interpreted as evidence that transfer of the congeners from materials treated with the penta-mix 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 BDE47 and the lighter congeners increasing to the north (with

increasing distance from source areas) while the proportion of BDE99 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 (e.g. BDE47), indicating that they have undergone some air-surface exchange (“hopping”), whilst those heavier congeners (e.g. BDE153) were close to zero, indicating that they are retained more effectively by soil following deposition.

New information on levels in biota in the Arctic

Christensen et al. (2002) analysed BDEs (congeners BDE47, BDE99, BDE100 and BDE153) in blue mussels (*Mytilus edulis*) and the livers of four fish species collected from sites in southern Greenland in 2000 using GC-ECNIMS. Shorthorn sculpin (*Myoxocephalus scorpius*) were collected from three locations, Usuk (no population), Igaliko (population 40) and Qaqortoq (population 3200). All other samples were collected at Usuk. BDE47 was detected in all samples, BDE99 and BDE100 in over half, though close to the limit of detection, and BDE153 in none. Σ BDE concentrations ranged from 0.1 $\mu\text{g kg}^{-1}$ wet weight in mussels to 12 $\mu\text{g kg}^{-1}$ wet weight in uvak (*Gadus ogac* – a predatory fish) from Usuk. In all the fish samples BDE47 represented > 90% of Σ BDE. The BDE47 and Σ BDE concentrations in shorthorn sculpin (a stationary species, likely to have lived most of their lives close to the sampling sites) were Qaqortoq > Igaliko > Usuk, in line with the increasing population. This suggests local sources in addition to longrange transport.

Trout (3 species) from 11 high mountain lakes in Europe (566 to 2,485m altitude) were taken for BDE analysis (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 BDE47 and BDE99, followed by BDE100, BDE153, BDE154 and BDE28, and these congeners were found in all samples analysed (n = 51). The highest concentrations of Σ BDE in fish muscle and liver were found in Lochnagar, Scotland, 1.2 and 11 $\mu\text{g kg}^{-1}$ wet weight, respectively (177 and 366 $\mu\text{g kg}^{-1}$ on a lipid basis). This could suggest higher BDE emissions in the UK than in other countries, as a result of production and use. 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.

Levels in biota because of atmospheric deposition

Zennegg *et al.* (2003) determined BDEs (BDE28, BDE47, BDE99, BDE100, BDE153, BDE154 and BDE183) in whitefish (*Coregonus* sp.) collected from eight lakes and farmed rainbow trout (*Oncorhynchus mykiss*) from four fish-farms in Switzerland. In whitefish muscle, Σ BDE concentrations ranged from 2.0 to 7.4 $\mu\text{g kg}^{-1}$ wet weight (36 to 165 $\mu\text{g kg}^{-1}$ on a lipid basis). The highest concentration was found in Lake Greifen, and the lowest in Lake Thun. A similar congener pattern was observed for all samples, dominated by BDE47 and followed by BDE99 and BDE100. All of the lakes investigated are situated in populated areas and, although none can be considered as background sites, diffuse contamination and atmospheric deposition are the most likely source for the BDEs found in whitefish. Lower Σ BDE concentrations were found in the farmed trout, from 0.7 to 1.3 $\mu\text{g kg}^{-1}$ wet weight (12 to 24 $\mu\text{g kg}^{-1}$ on a lipid basis).

In Australia polybrominated diphenyl ethers have been found in Australian pig fat (Burniston et al. 2003). Congeners BDE 47, BDE 85, BDE 99, BDE 100, BDE 138, BDE 153, and BDE 154 were detected and quantified in all samples analysed. BDE 47 and 99 dominated both in the fat samples and background levels. The mean concentration of BDE 99 and BDE47 in 2002 was 172 and 130 ng/g lipid, respectively. Mean background level was 2.3 ng/g lipid for both compounds.

There has been no production of PBDEs in Australia and relatively small amounts of these BFRs are imported relative to the global consumption, 1.2%, pentaBDE formulations. While this is true in Australia the production of these compounds in Singapore and its use in Southeast Asia make Australia susceptible to their long range atmospheric transport. While the direct use of the PBDEs in Australia is low the large volume of imported articles containing these BFRs could be considered a contributing

<p>- information regarding bio-availability</p>	<p>factor to background levels of PBDEs as they volatilise from their original product.</p>
	<p><u>New information on exposure in local areas and uptake in soil organisms</u> Reference (non-treated) and sewage-sludge amended soils (1 - 3 tonnes dry matter (DM) hectare-1 year-1) from three agricultural research stations in Sweden (Igelösa, Peterstorp, Lanna) were sampled in 2000 and analyzed for BDE47, BDE66, BDE99, BDE100, BDE153, BDE154 and BDE183 (Matscheko et al., 2002). In addition, soil samples were taken from two privately owned farms. At one farm, a total of 614 tonnes of sewage sludge (DM) was used on 25 hectares as fertilizer during 1978-1982, the equivalent of 25 tonnes hectare-1 for this period. At this time, a textile industry that used PBDE formulations in their production was connected to the local STP. At the other farm, fields were flooded the summer before by the river Viskan, which contains sediments which are highly contaminated with BDEs (Sellström et al., 1998). Reference samples were also taken from the farms, and earthworms collected at all soil sampling sites were also analyzed as well. The BDE congener profile in all soil samples was dominated by BDE47 and BDE99. The concentrations in reference soils from the agricultural research stations varied from 29 to 95 ng kg-1 DM for the sum of BDE47, BDE66, BDE99, BDE100, BDE153, BDE154 and -BDE183. For samples from the two farms, the reference concentrations were 110 and 190 ng kg-1 DM for ΣBDE. In plots to which sewage sludge had been applied, soil concentrations at the research stations increased to between 75 and 930 ng kg-1 DM. At the farm at which sewage sludge had been applied, the treated field had soil concentrations of 840,000 ng kg-1 DM for ΣBDE. Soil from the flooded field of the other farm contained 4,000 ng kg-1 DM for ΣBDE. The impact of applying 1 – 3 tonnes hectare-1 year-1 of sludge was determined by calculating the ratios of the concentrations of the compounds in the sludge-treated soil to those in the reference soil from the same site (S/R ratio). The S/R ratios for the individual BDE congeners were greater than 1 (range 3 to 12) indicating that sewage sludge application is a source of BDE contamination in agricultural soils. The largest increase in the S/R ratios (2,100 to 12,000) was found at the private farm, where large amounts of sludge had been applied approximately 20 years ago. This indicates that these substances can have quite long residence times in soils. 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 BDE47, BDE99 and BDE100 were around 5 (organic matter/lipids), similar to those obtained for the ortho-substituted chlorobiphenyls. Thus, earthworms living in highly contaminated soils will accumulate high tissue BDE concentrations and, as these animals represent the base of the terrestrial food chain for many organisms, this forms a pathway for the accumulation of BDEs in organisms at higher trophic levels.</p>
	<p><u>New information on local exposure and uptake in aquatic organisms</u> 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. BDE 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 (SE) were compared to those in samples from the Belgian North Sea (BNS) beyond the mouth of the estuary (Voorspoels et al., 2003). Eight BDE congeners (BDE28, BDE47, BDE99, BDE100, BDE153, BDE154, BDE183 and BDE209) were determined. Concentrations observed in the SE samples were up to 30 times higher than in those from the BNS, with an increasing gradient towards Antwerp. Concentrations in the BNS ranged from 0.02 to 1.5 µg kg-1 wet weight in benthic invertebrates and goby, from 0.06 to 0.94 µg kg-1 wet weight in fish muscle, and from 0.84 to 128 µg kg-1 wet weight in fish liver. The corresponding ranges in samples from the SE were from 0.2 to 30, 0.08 to 6.9, and from 15 to 984 µg kg-1 wet weight, respectively. The ratio BDE99/BDE100 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 SE sediment, and was similar in shrimp from both the BNS and</p>

SE, implying both that these congeners are readily bioavailable and that shrimp lack the ability to metabolise either congener. On a lipid weight basis, concentrations of BDE47 ranged from 3 to 108 $\mu\text{g kg}^{-1}$ lipid weight in samples from the BNS, and from 8 to 1,550 $\mu\text{g kg}^{-1}$ lipid weight in SE samples. BDE47 was the most abundant congener in all samples, comprising 43 to 75% of ΣBDE .

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 (Marsh et al., 2004). Methoxylated BDEs (MeO-BDEs) have also been reported as natural products present in marine sponges and green algae. Thus, the origin of these substances can be natural, anthropogenic or both. Nine OH-BDEs and six MeO-BDEs were identified in Baltic Sea salmon (*Salmo salar*) blood using newly synthesized standards (Marsh et al., 2004). All of the identified OH- and MeO-BDEs were substituted with four or five bromine atoms and five also had one chlorine substituent. Fourteen have the methoxy or hydroxy group substituted in the *ortho*- position 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-BDE49) may be a hydroxylated metabolite of BDE47.

Temporal trends of concentrations of BDE47, BDE99, BDE100, BDE153 and BDE154, as well as of two MeO-BDE congeners (6-MeO-BDE47 and 2'-MeO-BDE68) were determined in pike from Lake Bolmen, Sweden, during the years 1967-2000 (Kierkegaard et al., 2004a). All of the BDE congeners showed increasing trends in concentrations up to the mid-1980s, with a more than 25-fold increase during this period (from 60 to 1600 ng kg⁻¹ wet weight in 1989). After the mid-1980s, the concentrations either levelled off or began to decrease. The decreases of concentrations in pike are much slower than has been seen for guillemots in the Baltic Sea (Sellström et al., 2003) suggesting that there are local sources of BDEs close to Lake Bolmen. For the MeO-BDEs, the concentrations decline during the same period, with the decline for 6-MeO-BDE47 continuing into the 1990s. No correlation was seen between the BDE concentrations and the MeO-BDE concentrations, indicating other sources than the BDEs for the MeO-BDE compounds.

MeO-BDE congeners (tri-, tetra- and penta-BDEs) were also determined in Baltic Salmon and Arctic cod (*Gadus morhua*) (Sinkkonen et al., 2004). The tri- and penta-BDE congeners were not detected, but three MeO-TeBDE congeners were detected in Baltic salmon, and two in cod. Concentrations of those congeners detected ranged from 22 to 5,710 ng kg⁻¹ lipid weight in salmon and 14 to 16,400 ng kg⁻¹ lipid weight in cod. The concentrations of MeO-TeBDEs in the Arctic cod liver samples from years 1992 and 1993 were higher than those seen in both earlier (1987-1991) and later (1994-1995, 1998) years.

New information on uptake in sediment dwelling organisms

Magnusson *et al.* (2003) spiked intact, soft-bottom sediments with ¹⁴C-labelled BDE47, and studied the vertical distribution of the compound within the sediment and its bioaccumulation in 39 different macrofauna taxa over 29 weeks exposure. Throughout the period, 90% of the BDE47-derived radioactivity was observed in the upper 5cm of the sediment, mixed to that depth by bioturbation. Biota-sediment accumulation factors (BSAFs) were significantly higher for surface deposit feeders than for the suspension feeders, subsurface deposit feeders and predators, which yielded similar BSAF values to one another. Trophic level in the foodweb, measured as ¹⁵N enrichment, was not correlated with the degree of bioaccumulation. Hydrophilic metabolites of BDE47 were found in 15 of the species studied, indicating that biotransformation had taken place.

(f) National and international risk evaluations, assessments or profiles and labelling information and hazard classifications, as available (provide summary information and relevant references)**International:**

The use of two types of brominated flame retardants, tris (2,3 dibromopropyl) phosphate and polybrominated biphenyls (PBB), in textiles that are intended to come into contact with skin is prohibited both in Norway and in the EU through Council Directive 76/769/EEC on restrictions on the marketing and use of certain dangerous substances and preparations.

The EU has banned the use of penta- with effect on 15 August 2004. The EU has also included poly-BDE in the list of substances for which the IPPC Directive requires an exchange of information to be organized, and penta-BDE is on the list of priority hazardous substances under the Water Policy Directive. Penta-BDE is on the list of substances for which risk assessments are to be carried out within the EU. Processing of the risk assessments for penta-BDE has been completed.

According to the RoHS (Reduction of Hazardous Substances in EEE) Directive, no new electrical and electronic products may contain PBB or poly-BDE after 1 July 2006.

Brominated flame retardants are included on OSPAR's (The OSLO and PARIS Convention for the Protection of the Marine Environment of the North-East Atlantic) list of chemicals for priority action to protect the marine environment. At the 4th North Sea Conference, it was decided to phase out the use of brominated flame retardants by 2020.

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.

National:

Survey on uses of brominated flame retardants in products in Norway from 2003.

From 1 January 2004, products containing more than 0.25 % is classified as hazardous waste when they are discarded.

According to Norwegian legislation, chemicals containing brominated flame retardants that are hazardous to health or the environment must be classified and labelled before they are placed on the market.

Classification:

R 48/21/22 Harmful: danger of serious damage to health by prolonged exposure in contact with skin and if swallowed.

R64 May cause harm to breastfed babies.

N; R50/53 Very toxic to aquatic organisms may cause long-term adverse effects in the aquatic environment.

A national action plan for brominated flame retardants in 2002 (updated 2005).

The following points in the action plan concerns PeBDE:

Restrictions on use

- The Pollution Control Authority will follow up on the ban against penta- and octa-BDE in imported products in relevant branches. We will also take steps to obtain information, if necessary by issuing orders, on what types of brominated flame retardants other than the banned ones are also contained in the electrical and electronic equipment (EEE) they import. The Norwegian importers will also be required to present plans for reducing or phasing out the use of brominated flame retardants.

There is a pressing need for exact information on the possible content of brominated flame retardants that imported EEE items contain. Experience has shown that many importers lack knowledge of the chemical content of their products. Up to now, their interest and possibility to acquire such knowledge has been limited. The Product Control Act requires Norwegian importers to exercise due care and to

have adequate knowledge of the substances and products they place on the Norwegian market. This check gives a quick indication of a product's bromine content. If products contain brominated flame retardants, the importers will have to draw up plans for reducing the content of such substances or for phasing out their use. The aim will be to eliminate the need for flame retardants wherever possible, or to replace them with flame retardants that are less hazardous to health and the environment.

- The Pollution Control Authority will follow up on the ban on penta- and octa-BDE for Norwegian manufacturers. We will also follow up on the use of brominated flame retardants other than those that have been banned. The producers must be able to document that they are complying with the statutory requirement to apply the substitution principle pursuant to the Product Control Act, and/or that they are implementing measures to reduce emissions.

The Product Control Act requires Norwegian manufacturers to exercise due care and to have adequate knowledge of the substances and products they use in manufacturing processes. This also includes manufacturers of products such as textiles and fittings for vehicles and boats, where there may be special rules relating to fire safety. The requirement means that if substances and products that are hazardous to health or the environment are used, risk assessments shall be carried out and steps taken to reduce the risk. Because the Pollution Control Authority has already included brominated flame retardants on the observation list, we expect active steps to be taken to replace these substances.

- The Pollution Control Authority is following up on the use of brominated flame retardants as well as precisely which substances are being used as substitutes in consequence of the bans against penta- and octa-BDE. We wish to obtain information on the quantities used and the health and environmental effects of these substances.

The ban against penta- and octa-BDE makes it necessary either to alter the design of the product so that the need for flame-retarding properties is reduced/eliminated or to replace the substance with a different one. There is a need to monitor precisely which substances are used as substitutes and to ensure that they represent the smallest possible risk to health and the environment.

Waste treatment

- The Pollution Control Authority will follow up at the industry level such that the agreed-upon target of 80 per cent *recovery of EEE waste* (WEEE) is maintained. For the types of EEE products that contain a lot of brominated flame retardants, SFT is attempting to further increase the degree of recovery via its strategy for increased collection of hazardous waste.

WEEE makes up a substantial proportion of the waste containing brominated flame retardants, and it is important to achieve good control of the way this waste is managed. The Ministry of the Environment has an agreement with the relevant branches of industry in which they take responsibility for the reduction, collection and treatment of EEE waste (the WEEE agreement). The target is to reach a waste recovery level of 80 per cent.

- The Pollution Control Authority is implementing a strategy for the *increased recovery of hazardous waste*. One of the results of this will be that the degree of collection for the 3 prioritised types of hazardous waste that contain a lot of brominated flame retardants will

increase significantly before 2006.

Regulations concerning hazardous waste are causing discarded products containing penta-, octa-, deca-BDE, HBCDD or TBBPA to be considered as hazardous waste. Through its strategy for the increased collection of hazardous waste, the Pollution Control Authority is prioritising 12 types of hazardous waste for measures during the period spanning 2004-2006. In this regard 3 of the types of waste: EEE plastic, expanded and extruded plastic and cellular rubber is being prioritised because they contain brominated flame retardants.

- The Pollution Control Authority is working with county governors and the *waste disposal industry* in order to ensure that emissions of brominated flame retardants become a part of the future environmental monitoring at landfills where relevant. An updated monitoring programme is expected to be able to be launched at all depots by 1 July 2007 at the latest.

Water that flows through a landfill will absorb pollutants from the deposited waste. The waste depositories are thus deemed to be a potential source for emissions of environmental toxins. By strengthening the monitoring, the pollution authority will receive better data on these emissions and the authority can then require that the requisite measures be undertaken.

- The Pollution Control Authority is working with county governors on the on-going follow-ups on the *shredding systems* in order to ensure the responsible management of EEE waste containing brominated flame retardants. A particular emphasis will be placed upon the follow-ups on the shredding facilities concerning the conditions of the discharge permits and whether one in turn finds pollutants from the facilities in the surroundings.

A number of shredding facilities receive and process EEE waste. It is important that the waste be handled in an environmentally responsible manner that prevents emissions of brominated flame retardants into the environment.

Information on the harmful properties of brominated flame retardants

- The Pollution Control Authority will ensure that information is easily available *to the public*. It will explain the serious problems involved, help the public to demand products that do not contain brominated flame retardants, and ensure that products that contain these chemicals are delivered to the recovery system for WEEE.

A good deal of informational material already exists concerning the content of brominated flame retardants in electrical and electronic equipment and the hazardous properties they have with respect to health and the environment. Attempts must be made to see to it that the normal consumer still remembers to not dispose of WEEE with their ordinary household waste. Providing information on the serious health and environmental effects of these chemicals is a key factor in maintaining alterations in people's behaviour with respect to waste disposal as well as their desires to purchase environmentally friendly products.

- We will also ensure that information on the serious hazards to health and the environment posed by brominated flame retardants in various products is readily available to the *relevant industries*. This will be to give the industries and relevant purchasers of the products more information and make it possible for them to demand less hazardous products. SFT is in

close touch with the relevant industries such as electrical and electronic equipment (EEE), automotive, paints, textiles, etc.

Nowadays, a green procurement policy is part of the overall strategy of many companies. But this is difficult to put into practice without adequate knowledge of the problems. Knowledge is also an important basis for bringing about changes in waste management. It thus is important for purchasers that relevant informational material is available concerning the brominated flame retardant content of EEE products and the hazardous properties of such with respect to their health and the environment. The WEEE agreement requires the EEE industry to carry out waste reduction measures, for example by reducing the use of substances that are hazardous to health and the environment in these products. SFT will continue the contacts with other authorities and agencies in order to ensure a continuation of the work on avoiding the use of brominated flame retardants.

Environmental monitoring

- The Pollution Control Authority is continuing its environmental monitoring and is assessing on an on-going basis the need for monitoring projects for brominated flame retardants in Norway.

SFT has performed many monitoring studies in recent years, however it will continue to be important to follow environmental developments concerning brominated flame retardants. Penta-BDE is on the list of priority hazardous substances under the EU Water Framework Directive, and requirements will therefore be laid down for monitoring of the substance in various ecosystems.

More information on:

http://www.sft.no/arbeidsomr/kjemikalier/flammehemmere/brominatedflameretardants_actionplan.htm

(g) Status of the chemical under international conventions

PentaBDE is nominated to the Stockholm Convention under UNEP and the POP-protocol under the Long-range air transport Convention under UNEP-ECE.

PentaBDE is considered a POP under LRTAP Convention and Stockholm Convention.

PentaBDE is nominated by EU to the Rotterdam Convention in 2003 and is on the agenda for the next meeting in 2006.