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Environmental risk evaluation report: Decabromodiphenyl ether

(CAS no. 1163-19-5)

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Steve Killeen

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Head of Science

Executive summary

Three risk assessment reports for decabromodiphenyl ether (decaBDE) were produced by the Environment Agency under the EU Existing Substances Regulation (ESR). These reviewed the main scientific literature published up to October 2006. A large amount of new information on polybrominated diphenyl ethers (PBDEs) has become available since then, because they remain popular chemicals for academic research. In addition, a new regulatory system – known as REACH – has been introduced in Europe to replace the ESR. This report was therefore commissioned to review the new data relevant to the environmental risks associated with decaBDE with special consideration of the REACH context.

Although decaBDE is no longer permitted to be used in electrical and electronic equipment in Europe, substantial stocks are present in treated articles in the technosphere, and it still has a major use in textiles. Emissions from these sources (e.g. dust arising from textile wear) are likely to occur over a long time frame.

DecaBDE is very persistent and widely dispersed in the environment, including sediment, soil and many types of biota. It is too early to draw any meaningful conclusions about trends in the levels of decaBDE in the samples collected for a major European monitoring programme. However, voluntary point source emission controls have not yet made any significant impact on levels in any of the media sampled from environments away from these point sources.

Whilst a risk arising from direct toxicity of decaBDE has not been identified, and it does not itself meet the Annex XIII criteria of the REACH Regulation, there continue to be concerns related to its presence in food chains (including top predators) and degradation pathways. The latest evidence suggests that detection of hazardous degradation products (e.g. hexaBDE congeners which themselves have very persistent and very bioaccumulative properties, and heptaBDE which has recently been added to the Stockholm Convention on persistent organic pollutants (POPs)) in sediment, sewage sludge and biota may be linked to emissions of decaBDE, although it is still difficult to estimate the rates and amounts of formation.

These factors should be considered in any further prioritisation of decaBDE for regulation under REACH. A number of recent studies have also been performed on its developmental neurotoxicity potential in mammals, but these need to be fully evaluated by appropriate experts before conclusions can be drawn for this endpoint.

Acknowledgements

A draft of this report was circulated to the Bromine Science and Environment Forum (BSEF), KemI (Sweden), the Norwegian Pollution Control Authority (SFT) and the Bureau d'Évaluation des Risques des Produits et agents Chimiques (BERPC, France) in Autumn 2008. Detailed comments were received from BSEF (the suppliers' trade body), and additional references were provided by KemI and the SFT. The Environment Agency would like to thank these organisations for their input.

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1 Introduction

Three risk assessment reports for decabromodiphenyl ether (decaBDE) were produced under the EU Existing Substances Regulation[1] (ESR). The original report was published in 2002 (EC 2002). An updated environmental risk assessment report was agreed in Spring 2004 at the EU Member State technical and policy level and subsequently placed on the website of the European Chemical Bureau (ECB 2004). A further update report, covering both environmental and health risks was agreed in October 2007 (ECB 2007). These previous reports cover the main literature published up to October 2006.

A large amount of new information on polybrominated diphenyl ethers has been published since these reports were completed. In addition, a new regulatory framework, known as the REACH (Registration, Evaluation, Authorisation and restriction of CHemicals) Regulation[2], has been introduced in Europe to replace the ESR. This report therefore reviews the new information relevant to the environmental risks posed by decaBDE and assesses these data in relation to whether there is a need to consider the substance further under the REACH framework.

1.1 Searching methodology

1.1.1 Databases

The published literature has been searched as follows.

- A search of the Chemical Abstracts database has been carried out using the following CAS numbers as search terms.
  - CAS no. 1163-19-5 Decabromodiphenyl ether
  - CAS numbers 68928-79-0, 139749-52-3, 63936-56-1, 64589-00-0, 66169-95-7, 68928-79-0. These CAS numbers all relate to the following substance: benzene, 1,2,3,4,5-pentabromo-6-(tetrabromophenoxy) which relate to nonabromodiphenyl ethers (nonaBDEs).

  This search should identify all the relevant papers where the CAS numbers for decaBDE and nonaBDE have been indexed.
- A search of the PUBMED database for various combinations of the terms flame retardant(s), fire retardant(s), PBDE(s) and BDE(s).
- Journal notification services. Since 2006 we have been regularly scanning the contents and using journal alerting services of the more common journals where publications are likely. This includes publications such as Environmental Science and Technology, the journals published by Elsevier, etc.
- Internet searches for relevant conferences (such as the Dioxin series of conferences) and other publications.

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[1] Official Journal of the European Union (O.J.) No L 084, 05/04/1993 p. 0001 - 0075.

[2] O.J. No L 396, 30/12/2006 p. 0001 - 0849.

This approach is likely to be comprehensive for information on decaBDE and nonaBDE, but may not identify all the published papers on lower brominated congeners (a search of Chemical Abstracts using the relevant CAS numbers for the lower brominated congeners would be needed for this). Therefore there may be papers missing that cover some aspects of polybrominated diphenyl ethers, but these are unlikely to contain significant new information on decaBDE.

The full literature search covers the period November 2006 to 16 June 2008. It is presumed that significant papers published prior to November 2006 were included in ECB (2007) in the lists of reviewed or un-reviewed references. Most of the un-reviewed references in that report have now been considered as part of this project.

The literature found using the strategy above was used as the basis of a draft version of this report. This was circulated to Industry stakeholders for comment, and those comments (BSEF 2009) were taken into account for this final report. Since we estimate that around five to ten papers on polybrominated diphenyl ethers are currently published each week, a further, more targeted, literature search was undertaken using PUBMED covering the period June 2008 until 18 May 2009. The purpose of this targeted strategy was to identify any *significant* additional data that had been published since the main detailed search was performed. The areas targeted included the following (these were identified as the most important areas in the evaluation in the draft report):

- Degradation and metabolism, in particular the potential for formation, and identity of, degradation or metabolic products from decaBDE.
- Studies investigating time-trends of levels of decaBDE in the environment.
- Studies investigating bioaccumulation of decaBDE.
- Studies investigating the ecotoxicity of decaBDE.

Only the most relevant papers from this targeted search were reviewed for this report. Other papers that may contain information on decaBDE are listed in Section 9 as “Other papers that have not been reviewed”.

1.1.2 Screening approach

From a review of the abstracts, the papers were divided into the following main groups:

1. Papers that contain new information on decaBDE or nonaBDEs that is relevant to the environmental risk assessment. These papers have been reviewed in detail for this project, and are provided in the main reference list.
2. Papers that contain new information on decaBDE or nonaBDEs (or where it is thought that they may contain information on decaBDE or nonaBDEs) but that have yet to be reviewed. This includes some conference abstracts that have not yet been formally published, and the bulk of the papers identified in the targeted literature search (up to 18 May 2009) that have not been reviewed in-depth.
3. Papers that contain new information on decaBDE or nonaBDEs (or where it is thought that they may contain information on decaBDE or nonaBDEs) that is relevant to the human health risk assessment. These papers are briefly mentioned in Section 6 and are listed in the reference list under the

title “Human health-related references.” They have not been reviewed for this report.

4. Papers that clearly do not contain information on decaBDE or nonaBDE. These are listed in the reference list as “Other references of limited relevance.” A brief explanation as to why they are not considered for this project is also given there.

Several of the articles that have been reviewed are extended abstracts from conference proceedings and these do not always contain full experimental details. These are marked in the text as “[ABST]”. The results should be treated with caution until the full details have been formally published in the peer-reviewed scientific literature. This is particularly relevant for papers where analytical monitoring for the concentrations of decaBDE have been carried out, because such analyses are difficult and can easily lead to both false positives and negatives, particularly for samples of biota, blood, milk and food (see ECB 2004 and 2007 for further information).

With the exception of decaBDE, the exact identities of the congeners identified in the environmental monitoring data are given by reference to the congener number. This terminology is commonly used in the literature for polybrominated diphenyl ethers. A full list of congener numbers and names is given in the list of abbreviations at the end of this report.

2 Physico-chemical properties

The physico-chemical property data for decaBDE are summarised in EC (2002), ECB (2004) and ECB (2007). New data are summarised below.

Xu et al. (2007) developed quantitative structure-property relationships for polybrominated diphenyl ethers using structural descriptors derived from molecular electrostatic potentials. Using these relationships, estimates for the n-octanol-air partition coefficient (log K_{oa}), the sub-cooled liquid vapour pressure at 298 K and the Henry's law constant were made for all 209 polybrominated diphenyl ether congeners. The estimates obtained for decaBDE and nonaBDE are shown in Table 2.1. The reliability of the values is not known. In addition, the paper gives the values as log values and the underlying units of the vapour pressure and Henry's law constant are not clear.

Table 2.1 Estimates of log K_{oa}, vapour pressure and Henry's law constant.

Congener	log K_{oa}[1]	Sub-cooled liquid vapour pressure at 298 K[2]	Henry's law constant[1, 3]
BDE-206	15.01	1.9×10^{-8}	1.0
BDE-207	14.49	5.1×10^{-8}	3.9
BDE-208	14.78	2.9×10^{-8}	59
DecaBDE	15.73	4.0×10^{-9}	1.2

Notes: [1] The temperature for these values is not clear.

[2] The paper gives the estimates as log values and the underlying units are not clear. It is assumed that the unit is the pascal (Pa).

[3] The paper gives the estimates as log values and the underlying units are not clear. It is assumed that they are Pa m^3/mol. In addition the paper cites an experimental log Henry's law constant of 0.04 (equivalent to a Henry's law constant of 1.1) from a study by Cetin and Odabasi (2005). However that paper was reviewed in ECB (2007) and the Henry's law constant was actually 0.04 Pa m^3/mole. It is therefore not entirely clear if the values reported by Xu et al. (2007) are log values or not.

There are some uncertainties with these data and the results probably do not add significantly to the existing knowledge of the properties of decaBDE itself.

3 Consumption and use

No new information on the current consumption of decaBDE in the UK and the EU has been located.

Xiang et al. (2007) reported that the domestic production of decaBDE in China had increased from 10,000 tonnes/year in 2000 to 25,000 tonnes/year in 2004. A similar estimate is given by Chen et al. (2007) who report an increase from 10,000 tonnes/year in 2000 to 30,000 tonnes/year in 2005. Chen et al. (2007) also reported that the octaBDE and nonaBDE content of two commercial decabromodiphenyl ether products from China was in the range 8.2 to 10.4 per cent. This suggests a higher level of impurity in these products compared with the products typically supplied in the EU (typically ≥97 per cent decaBDE).

Directive 2002/95/EC on the restriction of the use of certain hazardous substances in electrical and electronic equipment (the "RoHS Directive")[3] effectively bans the use of decaBDE in new electrical and electronic equipment in the EU from 30 June 2008. This will clearly affect the future consumption of decaBDE in the UK and the EU. However a substantial amount of decaBDE will remain in existing electrical and electronic equipment until disposal.

The requirements of the RoHS Directive may also have implications for the recycling of plastics containing decaBDE as any recycled plastic used in new electrical and electronic equipment will need to comply with the requirements of the Directive. This is considered further in Section 4.1.2.

The main ongoing European use would therefore appear to be for textile applications, as described in the previous risk assessment reports. It may also still be used in some polymer types not associated with electronic equipment (e.g. hotmelt adhesives), although the amounts are likely to be small (EC 2002).

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[3] O.J. No L 37, 27/01/2003 p. 19 - 23.

4 Environmental exposure

New information on potential environmental exposure to decaBDE is summarised in the following sections.

4.1 Environmental release

4.1.1 Industrial applications

The emissions to the environment from two textile processing sites in the north-west of the United Kingdom are reported in the Environment Agency Pollution Inventory. The data available are summarised in Table 4.1 (some of these data have been considered previously in ECB 2007). Overall, the figures for site A suggest that the emissions to sewer from the site in 2007 are now around one third to one quarter of those in 2002 and 2003.

Table 4.1 Emissions of decaBDE from textile processing sites in the United Kingdom.

Site	Year	Reported emission (kg/year)	
		Air	Sewer
A	2002		1,400[1]
	2003	<100[1]	2,440[1]
	2004		270[1, 3]
	2005		86[1, 3]
	2006		990
	2007		554
B[2]	2004		381[1]

Notes: [1] Emissions are given as bromine in the database. However, as discussed in ECB (2007) it is not always clear if the figures represent the mass of bromine or the mass of decaBDE itself. As decaBDE has a bromine content of around 83% this difference would be small.

[2] Data available for 2004 only.

[3] ECB (2007) gives the 2004 and 2005 figures for this site as 3,260 kg/year and 1,030 kg/year respectively.

Sakai et al. (2005) [ABST] studied the emissions of decaBDE and polybrominated dibenzo-*p*-dioxins and furans from a textile processing facility in Japan. The textile site used both decaBDE and hexabromocyclododecane as flame retardants. Samples of flue gas, waste water, indoor air, ambient air and atmospheric deposition were collected at the facility and a sample of water and sediment was collected both upstream and downstream of the facility. In addition a sample of the textile was collected after processing. The levels of polybrominated diphenyl ethers (and polybrominated dibenzo-*p*-dioxins and furans) measured in the waste water from the plant are summarised in Table 4.2. It should be noted that two other textile processing plants were also sampled. These were reported to use only hexabromocyclododecane but decaBDE was also found in the effluents from these plants (at up to 230,000 ng/l). In addition, lower brominated congeners (di- to octaBDEs) were also found in most effluent streams at low levels.

Table 4.2 Levels of polybrominated diphenyl ethers (and polybrominated dibenzo-*p*-dioxins and furans) in waste water at a textile processing facility.

Substance	**Concentration in waste water (ng/l)**	
	From dyeing[1]	**After treatment**[1]
DecaBDE	150	6,200,000
NonaBDE	3.4	34,000
Polybrominated dibenzo-*p*-dioxins[2]	10	17,000
Polybrominated dibenzofurans[2]	170	150,000

Notes: [1] It is not clear what waste water streams these refer to.
[2] The actual substances found is not given. The analysis was carried out for tetra- to octabrominated congeners.

A mass-balance was also constructed for the textile processing facility (taking into account inputs and outputs from the plant). The batch size at the plant was 300 kg of textile per batch and used 130 kg of decaBDE and 6 kg of hexabromocyclododecane per batch. The water consumption was 21,000 litres per batch. The concentrations in the air emissions were given as 130-6,300 ng/m^3 for polybrominated diphenyl ethers and 0.063-13 ng/m^3 for polybrominated dibenzo-*p*-dioxins and furans. The overall emission factor for decaBDE from textile processing was estimated as 9.0×10^{-7} to air and 0.01 to waste water (it is not clear from the paper whether this relates to a fraction of the decaBDE use or a fraction of textile processed).

For the polybrominated dibenzo-*p*-dioxins and furans, the total output/release from the plant was estimated to be around 148 mg per batch. However the input to the plant (as a result of impurities in the raw materials used at the plant) was estimated to be 94 mg per batch suggesting the possibility of formation of polybrominated dibenzo-*p*-dioxins and furans during textile processing.

Few other details of this study are currently available and it is not always clear how the figures quoted in the paper were derived. However the study appears to show that emissions of decaBDE to water can occur (with an emission factor of 0.01, or 1%). For comparison, ECB (2007) estimated an average emission factor to sewer of 0.0018 (0.18%) from textile processing sites based on a survey of the UK industry in 2005. This emission factor was derived by relating the emissions to sewer occurring over a one week period to the amount of decaBDE used over the same one week period. However, as discussed in ECB (2007) one site accounted for around 75% of the reported emissions to sewer and so the emission factors for some sites in the 2005 survey are likely to be higher than 0.0018.

Buser et al. (2007a) carried out a dynamic substance flow analysis for decaBDE in Switzerland covering the period 1980 to 2020. The analysis predicted that emissions to the environment would increase from 1980 until 1990, reaching a maximum emission of 15 kg/year to the atmosphere, 8 kg/year to the hydrosphere and 55 kg/year to soil. This was then followed by a predicted decrease until the late 2000s after which the emissions were expected to remain fairly constant (at a total emission of around 10 kg/year). A large part of the decrease results from the prohibition of the use of sewage sludge as a fertilizer in Switzerland from the 1st October 2006. The study also found the substance flow to waste (e.g. landfill) increased over the entire study period and highlighted the role of adequate waste management strategies in reducing future emissions of decaBDE.

A similar study for Switzerland was also reported by Buser et al. (2007b) [ABST] and Morf et al. (2007) [ABST]. In this study the maximum emissions were predicted to occur in the mid-1990s, followed by a decrease until the late 2000s. The total emissions estimated at the end of the mid-1990s were about 60 kg/year, with up to 20 kg/year to atmosphere, up to 6 kg/year to the hydrosphere and up to 40 kg/year to soil. The study also estimated the outdoor air concentration for the year 2005 based on the estimated emissions as around 0.5 pg/m^3 (range 0.1 to 2.3 pg/m^3) (estimates were also given for the concentration in atmospheric deposition and sediments but these were displayed graphically only). The estimates were found to be generally lower than the available measured data and so it was thought that atmospheric emissions used in the study may have been underestimated.

4.1.2 Recycling of polymers

A number of recent studies have investigated the potential emissions of polybrominated diphenyl ethers and/or polybrominated dibenzo-*p*-dioxins and dibenzofurans from recycling of polymers.

It has been estimated that more than six million tonnes of waste electrical and electronic equipment are produced annually in Europe and that this amount of waste equipment contains more than one million tonnes of plastics (Schlummer et al., 2006). The waste electrical and electronic equipment (WEEE) Directive[4] sets out recovery and recycling targets for WEEE from the 31 December 2006. Depending on the WEEE, the recovery targets are between 70 and 80 per cent and the recycling targets are between 50 and 75 per cent. Since the plastic content of WEEE is typically between 15 and 30 per cent by weight, Schlummer et al. (2006) considered it unlikely that these targets would be met by metal and glass recycling/recovery alone.

The main source of waste plastic from the recovery/recycling of WEEE is thought to be shredder residues produced by a combination of shredding and metal-removal techniques at WEEE treatment plants (Schlummer et al. 2006). Such plastic waste is thought to consist of more than fifteen different polymer types and will contain brominated flame retardants (including decaBDE) that are restricted by the RoHS Directive. This has implications for the recyclability of such material. Schlummer et al. (2006) investigated a two stage process whereby styrenic polymers could be separated from this plastic waste and the brominated flame retardants (and other brominated compounds) removed. The resulting recycled polymers were shown to comply with the RoHS threshold values (0.1 per cent by weight for polybrominated diphenyl ethers) and also the German Chemikalienverbotsverordnung (for brominated dibenzo-*p*-dioxins and dibenzofurans).

Similarly, Schlummer and Mäurer (2006) demonstrated that styrenic polymers containing brominated flame retardants can be separated form non-bromine containing styrenic polymers using sink and float (density separation) techniques at the shredding stage. The resulting recycled polymers were shown to comply with the RoHS threshold values (0.1 per cent by weight for polybrominated diphenyl ethers) and also the German Chemikalienverbotsverordnung (for brominated dibenzo-*p*-dioxins and dibenzofurans).

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[4] Directive 2002/96/EC of the European Parliament and of the Council of 27 January 2003 on waste electrical and electronic equipment (WEEE). Official Journal of the European Union, 2003, L37, 24-38.

In a further study by Schlummer et al. (2007) the levels of brominated flame retardants and brominated dibenzo-*p*-dioxins and dibenzofurans in waste electrical and electronic equipment were determined. The polymeric samples in the survey included monitor and TV housings and light fractions from shredder processes. The samples were collected from various processing plants within Europe. Housings from nine TV sets and 36 monitors were analysed in all, along with seven samples of housing shredder residues and eight samples of mixed electrical equipment residues. The housings were found to consist of five main polymer types, acrylonitrile-butadiene-styrene (ABS), high impact polystyrene (HIPS), blends of ABS and polyvinylchloride (PVC), blends of ABS and polycarbonate and blends of polyphenylene oxide and polystyrene. HIPS was found to be the predominant polymer in TV housings, whereas ABS and blends of polyphenylene oxide and polystyrene were predominant in the monitoring housings. The brominated flame retardants were found to be associated mainly with ABS and HIPS (phosphorus-based flame retardants were evident in the other polymer types). However it was also noted that TV sets made from HIPS did not contain either brominated flame retardants or phosphorus-based flame retardants. The main brominated flame retardants found in the samples were tetrabromobisphenol-A, decabromodiphenyl ether, octabromodiphenyl ether, 1,2-bis-tribromophenoxyethane and the carbonate oligomer of tetrabromobisphenol-A.

The housing shredder residue samples were also found to contain brominated flame retardants (it was estimated that around 20 to 50 per cent of the plastics in the samples contained brominated flame retardants), with the main brominated flame retardants present being identified as tetrabromobisphenol-A, octabromodiphenyl ether, decabromodiphenyl ether and 1,2-bis-tribromophenoxyethane. The total concentration of brominated flame retardants in the samples was in the range 0.25 to 11 per cent. Decabromodiphenyl ether was found to be present in six out of the seven samples at around 0.2 to 2.2 per cent by weight (values read from a graph) and octabromodiphenyl ether was found to be present in five out the seven samples at around 0.3 to 1.4 per cent by weight (values read from a graph). Five of the housing shredder residue samples were also analysed for the presence of brominated dibenzo-*p*-dioxins and dibenzofurans. These were found to be in the range 2.2 to 33.0 µg/kg for sum 4 (the sum of 2,3,7,8-TeBDF, 2,3,7,8-TeBDD, 2,3,4,7,8-PeBDF and 1,2,3,7,8-PeBDD) or 3.0 to 35.3 µg/kg for sum 5 (sum 4 plus 1,2,3,7,8-PeBDF, 1,2,3,6,7,8-HxBDD, 1,2,3,4,7,8-HxBDD and 1,2,3,7,8,9-HxBDD[5]) were also included in the total. These values were found to be above the limits in the German Chemikalienverbotsverordnung in 100 per cent of the samples when the sum 4 levels were considered (the limit is 1 µg/kg) and in 40 per cent of the samples when the sum 5 levels were considered (the limit is 5 µg/kg).

The eight samples of mixed electrical equipment residues were also found to contain brominated flame retardants in the range 0.05 to 4.9 per cent by weight. Tetrabromobisphenol-A was found to be the predominant brominated flame retardant in these samples (concentration between 0.1 to 1 per cent) but octabromodiphenyl ether was also present in seven out the eight samples at a concentration between 0.08 and 0.44 per cent (in addition non-extractable bromine was present suggesting the presence of reactive brominated flame retardants in the samples). Four of the eight samples were also analysed for the presence of brominated dibenzo-*p*-dioxins and dibenzofurans. These were found to be present at between 1.2 and 11.6 µg/kg (sum 4) and 3.0 to 13.9 (sum 5). These values were found to be above the limits in the German Chemikalienverbotsverordnung in 100 per cent of the samples when the sum 4 levels were considered and in 50 per cent of the samples when the sum 5 levels were considered.

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[5] See the abbreviations list.

Schlummer et al. (2007) considered these data in relation to earlier studies and concluded that over recent years there appears to have been a move from the use of pure polymers containing brominated flame retardants to polymer blends such as ABS and polycarbonate or polyphenylene oxide and polystyrene along with the use of non-bromine-containing flame retardants. Despite this decline in use of polybrominated diphenyl ethers, Schlummer et al. (2007) pointed out that the levels of brominated dibenzo-*p*-dioxins and dibenzofurans in the samples are in the same range as found in older studies carried out in the last decade (e.g. Fluthwedel and Pohle (1996) (data considered in ECB (2000))). Thus there was no clear relationship between the amounts of brominated flame retardant present and the amounts of brominated dibenzo-*p*-dioxins and dibenzofurans in the current samples. In particular it was noted that in several of the samples the levels of brominated dibenzo-*p*-dioxins and dibenzofurans actually decreased as the levels of octabromodiphenyl ether in the sample increased. Schlummer et al. (2007) concluded that factors other than the levels of polybrominated diphenyl ethers in the samples must be an influencing factor on the levels of brominated dibenzo-*p*-dioxins and dibenzofurans in the samples. Schlummer et al. (2007) hypothesised that one factor that may also be important could be the shredder technology employed (there was some evidence that the levels of brominated dibenzo-*p*-dioxins and dibenzofurans in the samples may increase with decreasing particle size in the residues).

Hirai et al. (2007) [ABST] carried out a lifecycle assessment of the back covers from televisions containing brominated flame retardants in Japan. The study considered a number of impact categories, including climate change (CO_2 emissions), landfill consumption and toxicity to humans from emissions of polybrominated diphenyl ethers. Two different weighting methods were used (preventative cost method and damage cost method). The methodology was based on a 25-inch television that was used for ten years (it was assumed that 3.8 million of such units were recycled) and the lifecycle stages covered the stages from the manufacture of the polystyrene to the final disposal or recycling of the plastic from the back cover. After disassembly of the TV and shredding of the TV back cover, five options of disposal or recycling were considered including material recycling to video cassette cases, thermal recovery (waste incineration), feedstock recycling (in coke ovens used in the steel industry (the plastic waste replaces coal in the coke oven)), landfilling and landfilling followed by 10% burning in open fires. In each scenario it was assumed that the appliance recycling plant used the same processes up to shredding and separation of the back cover and that 9,500 tonnes of shredded plastic containing 855 tonnes of decaBDE were generated.

The analysis found that the scenario for feedstock recycling in coke ovens had the lowest overall impacts of the options considered. This resulted largely from a reduction in CO_2 owing to the large amount of power generated from by-product gas and a relatively low potential for exposure to polybrominated diphenyl ethers. However, Hirai et al. (2007) noted that the capacity in the steel industry in Japan for such recycling is limited owing to restrictions on the halogen content of plastic waste for such applications. The second best option was material recycling, although it was indicated that this had one of the largest potentials for exposure to polybrominated diphenyl ethers out of the options considered.

Sakai et al. (2007) [ABST] investigated the emissions of polybrominated diphenyl ethers and polybrominated dibenzo-*p*-dioxins/dibenzofurans at a full-scale municipal waste shredder plant in Japan. The plant handled a range of bulky municipal waste including household electrical appliances and waste furniture made of wood, plastics and metals. The plant operated both a hammer shredder (throughput 20 tonnes/hour) and blade shredder (throughput 2 tonnes/hour) during the measurements. The gases surrounding each shredder unit were connected to a bag filter and activated carbon filter and the waste gases emitted directly from the shredders were treated in a cooling tank, dehumidification tower, mist separator and activated carbon filter (steam was also injected in order to prevent explosions). The levels of polybrominated diphenyl ethers (only the total concentration is given), polybrominated dibenzo-*p*-dioxins/dibenzofurans, and other brominated flame retardants (both tetrabromobisphenol-A and hexachlorocyclododecane were determined) found in the various waste stream are shown in Table 4.3.

Table 4.3 Brominated compounds present in waste streams from a municipal shredding plant.

Substance	**Concentration (pg/Nm^3)[1]**			
	Shredding gas		**Final air after treatment**	
	Shredding	**Shredding with steam injection**	**Shredding**	**Shredding with steam injection**
Polybrominated diphenyl ethers (total)	88,000	120,000	350	18,000
Tetrabromobisphenol-A	21,000	73,000	200	7,500
Hexabromocyclododecane	26,000	24,000	<400	<600
Polybrominated dibenzo-*p*-dioxins (total)	Not detected	Not detected	Not detected	Not detected
Polybrominated dibenzofurans (total)	130	940	Not detected	100

Notes: [1]Nm^3 = cubic metre at normal temperature and pressure (293 K and a pressure of 101,325 Pa)

There was also a waste water stream from the system with steam injection. The concentration of polybrominated diphenyl ethers in this stream was determined to be 180 ng/l and the concentration of polybrominated dibenzo-*p*-dioxins and dibenzofurans in this stream was 1.2 ng/l (2.4 pg TEQ/l).

Petreas et al. (2005) investigated the levels of polybrominated diphenyl ethers in autoshredder and electronic waste in California. The results are displayed graphically only but decaBDE appears to have been present at a concentration of around 500 ppm (mg/kg) in four 'CPU' samples (presumably central processing units from computers) and around 10,000 ppm (mg/kg) in four video cassette recorder samples. Only traces of decaBDE were evident in samples of microwave ovens.

4.1.3 Pyrolysis and combustion

Several recent publications have investigated ways of removing bromine from flame retarded plastic waste in order to facilitate reuse of the plastics. One of the more studied methods involves pyrolysis to give oils that can be reused, for example, as feedstocks in the petrochemical industry, fuels, or raw materials (Hall and Williams 2006 and Bhaskar et al. 2006). Several of these studies are briefly summarised below.

Brebu et al. (2007) investigated the effect of antimony trioxide on the thermal degradation of samples of high impact polystyrene (HIPS) containing decaBDE (total bromine content of the HIPS was 10.8 per cent by weight) at 450°C. The experiments were carried out in glass reactors in which the sample was heated at a rate of 10°C per minute up to 360°C (the decomposition of HIPS started at this temperature) and then a slower rate of 5°C per minute up to 450°C under a nitrogen or in some cases ammonia atmosphere. The decomposition products were categorised into residue, oil and gases. The main constituent of the oil was found to be styrene monomer and other styrene derivatives (accounting for around 60 per cent by weight of the oil) and styrene dimers (accounting for around 8 to 12 per cent of the oil). Phenyl- and phenyl-methyl naphthalenes were also thought to be present along with several phenols and dibenzofuran. A number of bromine-containing organic compounds were also tentatively identified in the oils. These included bromophenols, brominated benzene derivates and dibromodiphenyl ether.

Hall and Williams (2006) investigated the pyrolysis of HIPS containing decabromodiphenyl ether using a fluidised bed reactor at temperatures between 450 and 550°C. The fluidised bed consisted of sand (300-425 µm diameter) supported on a distributor plate through which a flow of nitrogen was maintained. The reactor was heated to the required temperature and then 35 g of the plastic was fed into the reactor at a rate of 1.5 g/minute using a screw feeder. The pyrolysis gases and oils were collected and analysed using a range of techniques. During the experiment ≥89.9 per cent of the polymer was converted to oil and >98 per cent of the bromine was present in the oil. Analysis of the oil revealed the presence of around 257 compounds, 110 of which were thought to contain halogens. Benzene, toluene, ethylbenzene, styrene and cumene made up over one third of the oil but compounds with two of more benzene rings were also thought to be present (e.g. naphthalene, diphenylmethane, 1,2-diphenylethane, 1-phenylnapthalene and 1,3,5-triphenylbenzene). Several brominated compounds were also thought to be present including 1,2-dibromobenzene, 4-bromophenol and (1-bromoethyl) benzene. In addition, the amounts of polybrominated diphenyl ethers in the oil was also determined. At 450°C <0.1 per cent of the initial amount of decaBDE was evident in the pyrolysis oil and no decaBDE could be detected in the pyrolysis oil at 550°C. Several tri- to heptaBDEs were tentatively identified to be present (the amounts present were not given).

Broadly similar findings were found in another study by Hall et al. (2007) using a fixed bed reactor at 430°C.

Mitan et al. (2008) carried out a detailed investigation of the pyrolysis of HIPS containing decaBDE. The HIPS samples used contained decaBDE (total bromine content of the final polymer was 10.8 per cent by weight) and the experiments were carried out both with and without antimony trioxide (a concentration of 5 per cent by weight was used). The HIPS sample (1 g) was mixed with polyethylene, polypropylene and polystyrene[6] (3 g each) and then loaded into a batch glass reactor (glass tube of 35 cm length and 3 cm internal diameter) with a flow of nitrogen gas and heated in two steps. In step one, the sample was heated from ambient temperature to 330°C at a rate of 5°C per minute and then held at that temperature for 2 hours. The second step consisted of further heating of the sample from 330°C to 430°C at a rate of 15°C per

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[6] Earlier work has suggested that these polymers may be effective at removing halogens from the final pyrolysis oil.

minute and then holding at that temperature for around ten hours. The vapours and oils generated during both steps of the pyrolysis experiment were collected and subjected to a number of analytical techniques.

The study found that antimony trioxide affected the thermal decomposition of the sample by lowering the decomposition temperature of decaBDE to around 330°C. Thus the formation of liquid products from the pyrolysis experiment with antimony trioxide occurred earlier than in the experiment without antimony. The major bromine-containing compounds were found in the pyrolysis products from step 1 rather than the step 2 oil (gas chromatographic analysis using an electron capture detector revealed around 203 to 211 compounds in the step 1 oil compared with 96 to 111 in the step 2 oil). The yield of liquid products from the two steps was around 4 to 5 per cent for step 1 and 71 to 74 per cent for step 2. The yield of gaseous products was 15 to 16 per cent and the yield of residues was 9.3 per cent in the presence of antimony trioxide and 4.3 per cent in the absence of antimony trioxide.

The main components of the step 1 oil from the experiments with antimony trioxide were found to be ethyl benzene, cumene and 1,3-diphenyl propane and the major components of the step 2 oil were styrene and ethylbenzene. The major proportion of the antimony was found in the step 1 oil (as antimony tribromide) and partly in the residues.

The presence of polybrominated diphenyl ethers in the various pyrolysis oils was also investigated. In the experiments in the presence of antimony trioxide, no decaBDE was found to be present in the step 1 or step 2 oil. A number of lower brominated congeners (ranging from mono- to octabrominated diphenyl ethers) were tentatively identified in both the step 1 and step 2 oils. A smaller number of lower brominated congeners were present in the oils from the experiments carried out without antimony trioxide, and decaBDE (at 12 mg/kg) was detectable in the step 1 oil in this case.

Mitan et al. (2008) concluded that careful control of the pyrolysis temperature in a two step process can result in removal of the major proportion of the brominated hydrocarbons in the lower temperature first step and allows a pyrolysis oil relatively low in bromine content to be formed in the higher temperature second (pyrolysis) step.

A study by Bhaskar et al. (2006) investigated the use of a calcium hydroxide-carbon composite and an iron oxide-carbon composite to affect dehalogenation of the liquid products obtained from the pyrolysis of a mixed waste containing poly(vinylidene chloride) (1 g), HIPS (1 g) containing decaBDE, polyethylene (2 or 3 g), polypropylene (3g) and polystyrene (2 g) both with and without poly(ethylene terephthalate) (1 g). The pyrolysis experiments were carried out at 430°C under a nitrogen atmosphere using a semi-batch procedure. The hot gases from the pyrolysis reactor were fed to another reactor containing either the calcium hydroxide-carbon composite alone or as a mixture with an iron oxide-carbon composite maintained at 350°C (to facilitate dehalogenation by vapour-phase contact) before being condensed.

The yield of pyrolysis oil products was around 65 per cent by weight in the absence of poly(ethylene terephthalate) and 53 per cent by weight in the presence of poly(ethylene terephthalate). The presence of poly(ethylene terephthalate) was also found to have other effects on the pyrolysis; in particular, it resulted in an increased amount of gaseous products formed (around 29-30 per cent in the presence of poly(ethylene terephthalate) compared with 24-27 per cent without) and also led to the formation of a waxy residue.

The calcium hydroxide-carbon composite was found to remove the major proportion of the halogen content from the pyrolysis oils in the experiments without poly(ethylene terephthalate) (for example the chlorine content of the oil was reduced from

3,540 mg/kg to 44 mg/kg and the bromine content was reduced from 730 mg/kg to not detectable levels) but was less effective in the experiments with poly(ethylene terephthalate). In this case the chlorine content of the oil was reduced from 3,820 mg/kg to 450 mg/kg and the bromine content was reduced from 1,410 mg/kg to 490 mg/kg (using the calcium hydroxide-carbon composite alone) or reduced to a chlorine content of 280 mg/kg and a bromine content of 190 mg/kg (using a mixture of calcium hydroxide-carbon composite with an iron oxide-carbon composite). Thus it was also concluded that the presence of poly(ethylene terephthalate) resulted in higher elevation of the levels of halogenated hydrocarbons in the pyrolysis oils.

Bhaskar et al. (2008) investigated the use of microwave irradiation of HIPS containing decaBDE with a triethylene glycol solvent at 250°C as a method for removal of bromine from waste plastics. It was found that this system enhanced debromination and removal of bromine over conventional pyrolysis systems leading to a bromine-free plastic.

4.1.4 Landfills

Danon-Schaffer et al. (2005) [ABST] have investigated the leaching of polybrominated diphenyl ethers from landfill sites in the Canadian Arctic. The paper reports the results of a preliminary study where samples of soil and sediment were collected at designated solid waste disposal sites (Cambridge Bay, Yellowknife and Iqualuit) during the summer of 2004. Background samples were also collected. DecaBDE was found in soil/sediment at all three landfills sites at concentrations ranging from 12 µg/kg to 1,030 µg/kg (the results are not given for soil and sediment separately).

An investigation into the potential for release of polybrominated diphenyl ethers and other brominated products such as brominated dibenzo-*p*-dioxins and dibenzofurans from landfill fires has been carried out by Hirai et al. (2005) [ABST]. Laboratory-scale experiments were carried out simulating landfill fires. The tests were carried out using a stainless steel bowl filled with 15 kg of soil (10 cm deep) on top of which was placed 3 kg of waste. The bowl was then covered with a hood and the waste was ignited (using a torch for ten minutes). Sampling of the flue gas from the hood was started twelve minutes after the sample was first ignited. Two types of waste were studied. The first was refuse-derived fuel made from municipal waste consisting of 51.8 per cent paper and textile, 32 per cent plastics and leather, 5.3 per cent wood and grass, 9.5 per cent garbage, 0.4 per cent non-combustibles and 1.0 per cent others. The second was a refuse-derived fuel mixed with three brominated flame retardants (decaBDE plus tetrabromobisphenol-A and hexabromocyclododecane). Analysis of the waste indicated that the refuse-derived fuel contained 8,100 µg/kg of decaBDE, 1,800 µg/kg of nonaBDE and 23 µg/kg of octaBDE, with minor amounts of lower brominated congeners. The concentration in the refuse-derived fuel mixed with brominated flame retardants were 890,000 µg/kg decaBDE, 540,000 µg/kg nonaBDE and 3,900 µg/kg octaBDE. The concentrations of decaBDE, nona- and octaBDE in flue gas from the simulated fire were all very low (3.1-23, <3-5.1 and 2.1-20 ng/Nm^3 respectively) in the experiment using refuse-derived fuel alone. In addition, only minor amounts of lower brominated congeners (the concentration of mono- to heptabromodiphenyl ether congeners were all <20 ng/Nm^3) and brominated furans (5.3-19 ng/Nm^3) were present. However, in the experiment with fuel mixed with brominated flame retardants, the concentration of decaBDE, nona- and octaBDE in the flue gas were markedly higher (130-680, 1,100-2,100 and 8,400-240,00 ng/Nm^3) and substantial amounts of hepta- (up to 18,000 ng/Nm^3), hexa- (up to 14,000 ng/Nm^3), penta- (up to 7,200 ng/Nm^3) and tetrabrominated congeners (up to 3,400 ng/Nm^3), along with brominated dibenzofurans (up to 45,000 ng/Nm^3; the congeners present are not clear) were also found in the flue gas. The emission factor for decaBDE (defined as the total polybrominated diphenyl ethers emitted to flue gas as a percentage of the decaBDE added to the waste) was estimated to be 9.1 per cent.

Overall this study suggests that landfill fires where decaBDE is present could be a significant source of emissions of lower brominated diphenyl ether congeners and brominated dibenzofurans into the environment. However it should be noted that few details of the analytical methodology used in this study are currently available and, in particular, it is not clear which brominated dibenzofurans were found. It should also be noted that flame retardants other than decaBDE were present in the starting material for some of the experiments.

4.1.5 Other sources

Alonso et al. (2007) [ABST] estimated the emissions of several polybrominated diphenyl ethers from urban sources to waste water treatment plants based on the measured concentrations found in sewage sludge at the plants and the size of the plant. The mean emission of decaBDE from urban sources was estimated to be 6.16 mg per inhabitant per year. The mean emission of nonaBDEs was estimated at 0.23 mg per inhabitant per year for BDE-206 and 0.21 mg per inhabitant per year for BDE-207.

Assuming that a similar emission factor holds for the UK and the EU as a whole (i.e. that the per capita use of products containing decaBDE in Spain is similar to that in the UK and the EU as a whole, which may not necessarily be the case) then the total aquatic emission of decaBDE from uncontrolled urban sources can be estimated at around 370 kg/year in the UK (population ~60 million) and 3,060 kg/year in the EU (population ~497 million).

Chernyak et al. (2007) [ABST] investigated the emissions of polybrominated diphenyl ethers in a newly constructed building. The investigation took place over a seven-month period in a mixed-use seven-floor office building containing offices, classrooms, cafeterias, computer rooms and offices. Samples of dust (collected by vacuuming a 1 m^2 area of floor) and air (both vapour and particulates were sampled separately over a one week period) were collected at intervals in an office suite located on the sixth floor. The sampling was started just prior to occupancy in August 2006 and was continued every two to three months until February 2007. The levels of decaBDE in the dust were around 100 µg/kg at the start of the study but were found to increase rapidly reaching around 600 µg/kg by December 2006 and around 10,000 µg/kg by February 2007. The concentrations of lower brominated congeners (BDE-47, BDE-99 and BDE-100) remained relatively stable throughout the study period (concentration <45 µg/kg). The polybrominated diphenyl ethers present in the vapour phase in the air samples were dominated by the lower brominated congeners and were found to increase over the course of the study. DecaBDE does not appear to have been detected in the vapour phase. The mass of airborne particulates collected in the study was low and insufficient material was available to allow analysis of the concentration of decaBDE present. Lower brominated congeners were detected in some of the airborne particulate samples (concentrations up to 70 pg/m^3) but the levels found were variable.

It is interesting to note that although high levels of decaBDE were present in dust, there appeared to be very little decaBDE present in the air. This implies that the main source of decaBDE in dust is likely to be from wear of articles containing the substance rather than by volatilisation from the articles. Also relevant is the fact that the concentration in dust appears to increase with time, suggesting that the emission from such sources may also increase with time. Thus it is likely that indoor sources of decaBDE (e.g. dust generation) are important sources of decaBDE in the outdoor environment through washing of surfaces, etc.

Allen et al. (2008) developed a methodology for linking the presence of polybrominated diphenyl ethers in household dust to their presence in consumer products based on X-ray fluorescence. Based on measurements in several homes, the study found a strong association between the bromine content of foam furniture and the levels of pentaBDE in household dust and that the levels of decaBDE in household dust were associated with the bromine levels in televisions (this latter association was also dependent on the number of residents in a home which was taken to be a surrogate for TV usage).

4.1.6 Summary and evaluation

A number of new studies are available on the environmental release of decaBDE, and possible breakdown products, to the environment. It should, however, be noted that several of these studies are currently available only in abstract form, and important experimental details (e.g. on the analytical methodologies used) are not yet available for some of them.

The available data confirm that potential sources of release of decaBDE to the environment are textile processing facilities, landfills and household/office dust. Similar sources were also identified in the previous risk assessment reports on decaBDE.

Some of the new studies have also investigated the potential formation or release of polybrominated dibenzofurans and dibenzo-*p*-dioxins from the use of decaBDE in, for example, textile treatment facilities, plastic shredding plants and from landfill fires. In terms of the potential risks to the environment, polybrominated dibenzofurans and dibenzo-*p*-dioxins with bromine substituents in the 2,3,7,8-positions are generally considered to be the most toxic congeners. However, for some of these studies few details of the analytical methodologies used are available and the identities of the polybrominated dibenzofurans and dibenzo-*p*-dioxins found are not always clear.

The recent work by Schlummer et al. (2006 and 2007) and Schlummer and Mäurer (2006) on the levels of polybrominated diphenyl ethers and polybrominated dibenzofurans and dibenzo-*p*-dioxins in plastic shredder waste is of importance given the requirements of the WEEE and RoHS Directives. This work found that the levels of polybrominated diphenyl ethers in some samples of plastic shredder waste were above the RoHS threshold values and the levels of polybrominated dibenzofurans and dibenzo-*p*-dioxins in some samples were above the requirements of the German Chemikalienverbotsverordnung. However, techniques are available that allow such waste to be recycled and the recycled polymer to comply with both the RoHS Directive and the German Chemikalienverbotsverordnung. Interestingly, this study found no correlation between the levels of polybrominated dibenzofurans and dibenzo-*p*-dioxins in the shredder waste and the levels of polybrominated diphenyl ethers in the waste.

The pyrolysis behaviour of plastics containing decaBDE (and other brominated flame retardants) has been investigated recently as a possible recycling process for waste plastics. Recent research has focused on the separation of the bromine-containing compounds from the pyrolysis oils, allowing the oils to be used as a raw material or feedstock. However, few of these studies have investigated the amounts of polybrominated dibenzofurans and dibenzo-*p*-dioxins generated during the process.

Much of the earlier available information on the formation of polybrominated dibenzofurans and dibenzo-*p*-dioxins from the use and pyrolysis of decaBDE has been summarised in the previous risk assessment reports, particularly EC (2002). The new data are generally consistent with the previous information. Although it is clear that polybrominated dibenzofurans and dibenzo-*p*-dioxins are present at low levels in some samples of plastic shredder waste, and have the potential to be formed during combustion or pyrolysis processes involving plastic waste, it is difficult to ascribe their presence or formation solely to the use of decaBDE (other brominated flame retardants and sources of bromine are likely to be present in the waste polymers). Therefore it is not possible to quantify the risks from decaBDE for these sources. It should also be noted that the requirements of the RoHS Directive effectively mean that decaBDE can no longer be used in new polymers for electrical and electronic equipment in the EU. This also means that recycling methods would need to effectively remove the polybrominated diphenyl ethers from any recycled polymer for such use. As reported above, methods have been developed to effectively remove the brominated compounds from such waste streams before recycling. Therefore the amount of decaBDE present in waste polymer streams in the UK and EU should gradually reduce with time, although this could take many years.

4.2 Environmental concentrations

4.2.1 Surface water, effluent streams and sewage sludge

Conclusion (i) monitoring programme

As a result of the risk assessment of decaBDE carried out under the ESR (ECB 2004 and 2007) a ten-year monitoring programme (known as the conclusion (i) monitoring programme) was requested by the EU Member States under Commission Regulation (EC) No. 565/2006[7] to investigate the long-term trends in the levels of decaBDE in sewage sludge, sediment, air and birds' eggs (see also Section 4.2.2, 4.2.4 and 4.2.5). Under this programme sewage sludge samples from a total of twelve sites in the EU are being analysed with three samples being collected at each site over a one week period in each sampling year. The sampling points at the sites include secondary sludge at all sites, sometimes in combination with primary sludge. The sampling sites and the levels determined to date are summarised in Table 4.4. Some of these data have been reported previously in ECB (2007) and the most recent data are taken from Leslie et al. (2008). In future years it is intended that the sites are sampled on a biannual basis.

As data are only currently available for two sampling campaigns it is not possible to draw any meaningful comparisons or conclusions with regards to trends in the levels of decaBDE in the samples.

Levels of certain lower brominated congeners in sewage sludge are also reported by Leslie et al. (2008) for some of the samples collected in 2007, and it is useful to consider some of these data in relation to possible debromination of decaBDE. One of the lower brominated congeners (BDE-126, a pentabromodiphenyl ether) was included in the study because it has been reported that it is formed during the degradation of decaBDE under anaerobic reducing conditions by zerovalent iron. It is therefore a possible marker congener to determine if such reactions occur in the environment (see

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[7] O.J. No L 99, 07/04/2006 p. 003 - 005.

ECB (2007) for further details). This congener was not detectable (concentrations <0.01 µg/kg wet weight) in the samples analysed in the 2006 survey (ECB, 2007). For the 2007 survey, the sewage sludge samples from two of the sites were analysed for BDE-126, and this congener was found to be present in these samples at a concentration of 0.10 µg/kg dry weight at one site and 0.11 to 0.15 µg/kg dry weight at the second site. This finding is important because, although the levels are low, it potentially indicates that debromination of decaBDE to lower brominated congeners is occurring in the environment (and it is only one of a number of such substances that could be formed).

The other interesting feature of the Leslie et al. (2008) study is that congeners found in the commercial pentabromodiphenyl ether and octabromodiphenyl ether formulations (for example BDE-48, BDE-99, BDE-100, BDE-153 and BDE-183) were also found to be present in sewage sludge samples from Eindhoven and Kralingsveer collected in 2007. As the presence in sewage sludge must represent a relatively recent emission into the waste water treatment plant, this implies that these substances are still being released to waste water despite the fact that their use in the EU in new products has been banned since 2004. This demonstrates a considerable time lag between introducing controls and the cessation of emissions to the environment for substances that are present in articles with a relatively long service life.

An alternative explanation for the continued occurrence of these congeners in sewage sludge is that they are formed by debromination of decaBDE within the waste water treatment plant. Based on the information reported elsewhere (e.g. see EC (2002), ECB (2004) and ECB (2007) as well as this report), this is probably a less likely explanation, but it cannot be totally ruled out as the finding of low levels of BDE-126 in the sewage sludge suggests some degradation could be occurring. In any case, the continued emission of lower congeners from historic uses will continue to obscure any relationships between them and decaBDE for a long time to come.

Other studies

The levels of decaBDE and nonaBDE reported in surface water, effluent streams, sewage sludge and other sludges are summarised in Table 4.5.

Europe

The levels of decaBDE in various effluent streams at an industrial park located on the River Vero, Spain, have been determined by Eljarrat et al. (2007). The effluents sampled included a mixture of sanitary wastewater and effluent from a textile factory, production effluent from the textile factory, effluent from an epoxy resins production site, effluent from a polyamide polymerisation site and the combined effluent from the industrial park. The samples were collected either in November 2005 or March 2006 (some sites were sampled on both occasions). The effluent was filtered (<0.45 µm) prior to analysis and the concentrations of decaBDE in the dissolved and particulate phases were determined separately. DecaBDE was found to be present predominantly in the particulate phase, and the concentrations found were 11 ng/l in the mixed sanitary wastewater/effluent from the textile factory, 5 and 28 ng/l in effluent from the textile factory, 45 ng/l in effluent from the epoxy resin production site, 2,600 ng/l in effluent from the polyamide polymerisation site and 105 and 1,170 ng/l in the effluent from the whole industrial park. These results suggest that the polyamide polymerisation site was responsible for the majority of the decaBDE released to the River Vero from this industrial park. Samples of sediment and fish were also taken from the River Vero and these results are reported in Section 4.2.2 and Section 4.2.5 respectively.

Table 4.4 Levels of decaBDE in sewage sludge from the conclusion (i) monitoring programme.

Location of sewage treatment plant	Treatment plant type	Mean concentration in sludge (± standard deviation)			
		2006		2007	
		µg/kg dry weight	µg/kg organic carbon	µg/kg dry weight	µg/kg organic carbon
UK	Rural domestic wastewater	3,201±216[a]	7,169±426[a]	3,810±2,580	8,530±5,5570
UK	Mainly textile input	3,931±335[a]	12,543±1,065[a]	5,110±1,770	17,910±3,580
UK	Mixed domestic and industrial wastewater	708±843[a]	2,388±2,894[a]	3,000±310	8,240±880
UK	Mixed domestic and industrial wastewater	4,004±259	10,769±450	4,050±631	11,100±1,0070
UK	Urban domestic wastewater	2,985±548[a]	7,028±1,414[a]	5,490±2,890	12,140±6,420
Ireland	Domestic wastewater	7,963±676	20,000±2,562	5,170±989	12,540±2,590
The Netherlands	Mainly domestic wastewater	270±34	755±95	248±145	639±351
The Netherlands	Mainly domestic wastewater	407±6	1,118±49	208±29	600±81
The Netherlands	Domestic, industrial (25%) and rainwater run-off from roads	306±4	880±11	410±17	1,220±105
The Netherlands	Mainly domestic wastewater	399±15	1,482±56	353±28	1,410±105
The Netherlands	Mainly domestic wastewater	381±68	1,002±175	463±35	1,040±85
The Netherlands	Large capacity, including domestic wastewater	309±18	870±41	180±0	473±15

Notes: a - In ECB (2007) these results are reported to be from 2007 based on information given in Leslie et al. (2007). The Leslie et al. (2008) report indicates that these data refer to samples from 2006 and gives further data for 2007.

Table 4.5 Levels of decaBDE and nonaBDE in surface water, effluent streams, sewage sludge and other sludges.

Sample	Comments	Congener	Concentration	Reference
Europe				
Effluents from an industrial park on the River Vero, Spain.	Mixture of sanitary wastewater and effluent from a textile factory, 2005.	DecaBDE	11 ng/l (particulate phase)	Eljarrat et al. (2007)
	Effluent from a textile factory, 2005.	DecaBDE	28 ng/l (particulate phase)	
	Effluent from a textile factory, 2006.	DecaBDE	5 ng/l (particulate phase)	
	Effluent from the whole industrial park, 2005.	DecaBDE	1,170 ng/l (particulate phase)	
	Effluent from the whole industrial park, 2006.	DecaBDE	105 ng/l (particulate phase)	
	Effluent from an epoxy resin production site, 2006.	DecaBDE	45 ng/l (particulate phase)	
	Effluent from a polyamide polymerisation site, 2006.	DecaBDE	2,600 ng/l (particulate phase)	
Landfill leachate, Sweden	Survey of 22 leachate samples from 12 landfills.	NonaBDEs	1.1 µg/l (maximum) 0.023 µg/l (median) 0.29 µg/l (mean)	Öman and Junestedt (2008)
		DecaBDE	4.2 µg/l (maximum) 0.054 µg/l (median) 1.1 µg/l (mean)	
Sewage sludge, Germany	Samples from 11 municipal waste water treatment plants, 2002-2003.	DecaBDE	256 µg/kg dry weight (median) 97-2,217 µg/kg dry weight (range)	Knoth et al. (2007)

Sample	Comments	Congener	Concentration	Reference
Sewage sludge, Spain	Urban waste water treatment plant in Barcelona with a flow of 2,083 m^3/hour, 2005.	BDE-206	<0.4 µg/kg dry weight (not detected)	Eljarrat et al. (2008)
		BDE-207	<0.4 µg/kg dry weight (not detected)	
		BDE-208	<0.4 µg/kg dry weight (not detected)	
		DecaBDE	1,082 µg/kg dry weight	
	Urban waste water treatment plant in Burgos with a flow of 600 m^3/hour, 2005.	BDE-206	6.4 µg/kg dry weight	
		BDE-207	4.3 µg/kg dry weight	
		BDE-208	1.7 µg/kg dry weight	
		DecaBDE	393 µg/kg dry weight	
	Urban waste water treatment plant in Lleida with a flow of 2,917 m^3/hour, 2005.	BDE-206	<0.4 µg/kg dry weight (not detected)	
		BDE-207	<0.4 µg/kg dry weight (not detected)	
		BDE-208	<1.4 µg/kg dry weight (below the quantification limit)	
		DecaBDE	508 µg/kg dry weight	
	Urban and industrial waste water treatment plant in Pampolna with a flow of 4,313 m^3/hour, 2005.	BDE-206	<0.4 µg/kg dry weight (not detected)	
		BDE-207	1.7 µg/kg dry weight	
		BDE-208	<1.4 µg/kg dry weight (below the quantification limit)	
		DecaBDE	356 µg/kg dry weight	
	Urban waste water treatment plant in Tarragona with a flow of 305 m^3/hour, 2005.	BDE-206	<0.4 µg/kg dry weight (not detected)	
		BDE-207	<0.4 µg/kg dry weight (not detected)	
		BDE-208	<1.4 µg/kg dry weight (below the quantification limit)	
		DecaBDE	80.6 µg/kg dry weight	

Sample	Comments	Congener	Concentration	Reference
Sewage sludge, Spain	Small urban waste water treatment plant (<500,000 population).	DecaBDE	87-364 µg/kg dry weight	De la Torre et al. (2007) [ABST]
	Medium urban waste water treatment plant (500,000-1,000,000 population).	DecaBDE	224-715 µg/kg dry weight	
	Large urban waste water treatment plant (>1,000,000 population).	DecaBDE	114-426 µg/kg dry weight	
	Small mixed industrial/urban waste water treatment plant (<500,000 population).	DecaBDE	76-103 µg/kg dry weight	
	Medium mixed industrial/urban waste water treatment plant (500,000-1,000,000 population).	DecaBDE	122-207 µg/kg dry weight	
	Small industrial waste water treatment plant (<500,000 population).	DecaBDE	346-1,136 µg/kg dry weight	
	Large industrial waste water treatment plant (>1,000,000 population).	DecaBDE	185-452 µg/kg dry weight	
Sewage sludge, Sweden	50 sewage treatment plants from Sweden sampled in 2000.	DecaBDE	120 µg/kg dry weight	de Wit et al. (2007a) [ABST] and de Wit et al. (2007b) [ABST]
Compost and digestate from Switzerland	Composts and digestates derived from source-separated green and kitchen wastes.	Total PBDEs (decaBDE accounted for around 72% of the total)	~10 µg/kg dry weight (mean/median)	Brändli et al. (2007) ECB (2007) and Zennegg (2005)

Sample	Comments	Congener	Concentration	Reference
Rest of the world				
Seawater and sea-surface microlayer, Hong Kong, China	Five locations, 2005. Both the dissolved and particulate phases were analysed.	DecaBDE	<0.24 ng/l (dissolved phase) <0.41 ng/l (particulate phase)	Wurl et al. (2006)
Surface water, Pearl River Delta, China	Eight locations sampled over a one year period, 2005-2006.	DecaBDE	0.33-65 ng/l (range)	Guan et al. (2007)
Surface water from San Francisco estuary	A total of 33 samples from 2002 and 2003. Concentrations represent the sum of the particulate and dissolved phases.	BDE-206	0.0006-0.0093 ng/l (range; detected in 36% of samples) 0.0014 ng/l (median)	Hoenicke et al. (2007)
		BDE-207	0.0008-0.014 ng/l (range; detected in 45% of samples) 0.0017 ng/l (median)	
		BDE-208	0.0006-0.0071 ng/l (range; detected in 33% of samples) 0.0014 ng/l (median)	
		DecaBDE	0.012-0.191 ng/l (range; detected in 52% of samples) 0.031 ng/l (median)	
Influent, effluent and sewage sludge	Location of plants and number of plants is not clear	DecaBDE	71 ng/l (influent) 4.6 ng/l (effluent pre-disinfection) 3.5 ng/l (effluent post-disinfection) 650 μg/kg dry weight (sewage sludge)	Kolic et al. (2005) [ABST]
Sewage sludge, Canada	Samples of raw and digested sludge from 3 waste water treatment plants in Ontario, Canada	DecaBDE	200 to 500 μg/kg	Shin et al. (2007)
Sewage sludge, Kuwait	Samples of sewage sludge from three waste water treatment plants in Kuwait.	Total PBDEs (decaBDE accounted for 70-99% of the total)	23-1,559 μg/kg	Gevao et al. (2008)

Sample	Comments	Congener	Concentration	Reference
Sludge from waste water treatment plants in the Antarctic	Samples of waste water sludge from McMurdo research base, 2005.	BDE-206	17 µg/kg dry weight	Hale et al. (2008)
		BDE-207	19 µg/kg dry weight	
		BDE-208	7.6 µg/kg dry weight	
		DecaBDE	1,320 µg/kg dry weight	
	Samples of waste water sludge from Scott Antarctic research base, 2006.	BDE-206	5.4 µg/kg dry weight	
		BDE-207	5.8 µg/kg dry weight	
		BDE-208	3.2 µg/kg dry weight	
		DecaBDE	219 µg/kg dry weight	
Dried dairy manure	Values read from a graph.	BDE-206	~0.07 µg/kg dry weight	Agnihotri et al. (2005) [ABST]
		BDE-207	~0.07 µg/kg dry weight	
		DecaBDE	~1.1 µg/kg dry weight	

Studies by de Wit et al. (2007a [ABST] and 2007b [ABST]) comprised a survey of the levels of decaBDE in samples of sewage sludge from sewage treatment plants in Sweden. A total of 50 sewage treatment plants were sampled covering the major cities and towns and representing different sizes of plant. The samples included both raw and digester sludge. The mean decaBDE concentration found was 120 µg/kg and, with the exception of plants with suspected point sources (e.g. textile industries), the level found did not vary significantly with size of plant indicating a similar input per person equivalent to each plant. No significant difference was found between the levels of decaBDE in raw sludge compared with the levels in digester sludge, suggesting little anaerobic biodegradation of decaBDE was occurring in the plants.

Eljarrat et al. (2008) found decaBDE to be present at a concentration between 80.6 and 1,082 µg/kg dry weight in sewage sludge samples from five waste water treatment plants (four urban and one mixed urban/industrial) in Spain. The samples were collected in 2005. The concentrations of nonaBDEs (BDE-206, BDE-207 and BDE-208) were also determined and the concentrations range between not detected (<0.4 µg/kg dry weight) to 6.4 µg/kg dry weight for BDE-206, not detected (<0.4 µg/kg dry weight) to 4.3 µg/kg dry weight for BDE-207 and not detected (<0.4 µg/kg dry weight) to 1.7 µg/kg dry weight for BDE-208.

Another study on the levels of decaBDE in sewage sludge from waste water treatment plants in Spain found that decaBDE was present in sludge from seven sites at a concentration between 76 to 1,136 µg/kg dry weight (De la Torre et al. 2007) [ABST]. The sites include treatment plants receiving effluent from urban, mixed urban/industrial and industrial sources.

The levels of decaBDE and nonaBDEs (it is not clear which congeners were included)) have been determined in samples of landfill leachate from Sweden (Öman and Junestedt 2008). A total of 22 leachate samples were collected from 12 municipal landfill sites. The landfills sampled ranged in age from a few years to 50 years and they were reported to contain household waste, industrial waste, construction and demolition waste, as well as ash, slag, sludge, lime, sediment and excavation material. The leachate samples were not filtered prior to analysis so the results represent the total (dissolved plus particulate) concentration (the suspended matter content of the samples was in the range 8.7 to 2,100 mg/l with a median value of 32 mg/l). DecaBDE was reported to be present in four samples (it is not entirely clear if this number relates to the total number of samples that were analysed for decaBDE or whether decaBDE was detectable in four out of the 22 samples) and the maximum, median and mean concentration found was 4.2 µg/l, 0.054 µg/l and 1.1 µg/l respectively (the detection limit of the analytical method used was 0.01 µg/l). NonaBDEs were also reported to be present in four samples with a maximum, median and mean concentration of 1.1 µg/l. 0.023 µg/l and 0.29 µg/l respectively (the detection limit for nonaBDEs was 0.002 µg/l)

Knoth et al. (2007) determined the levels of polybrominated diphenyl ethers in sewage sludge samples from Germany. Samples were collected from eleven municipal waste water treatment plants between March 2002 and June 2003. DecaBDE concentrations in the samples ranged between 97 and 2,217 µg/kg dry weight (median concentration 256 µg/kg dry weight) and was the predominant polybrominated diphenyl ether congener present in the samples. No significant change in the polybrominated diphenyl ether congener profile (tri- to heptaBDEs) was seen in sludges from different stages in the treatment process (e.g. primary sludge, secondary excess sludge and dewatered digested sludge) indicating that degradation of decaBDE to these lower brominated congeners was not occurring during the process.

Brändli et al. (2007) reported the levels of decaBDE in compost and digestate derived from source-separated green and kitchen wastes from Switzerland. DecaBDE accounted for around 72% of the total polybrominated diphenyl ether content of such samples (~10 μg/kg dry weight). Similar (or possibly the same) information was included in ECB (2007) and Zennegg (2005) (this latter paper gives the range of decaBDE concentrations as 4.8 to 20 μg/kg dry weight with a mean value of 10 μg/kg dry weight).

Rest of the world

Wurl et al. (2006) determined the levels of polybrominated diphenyl ethers in the dissolved and suspended phases of seawater from Hong Kong. Sample of the sea-surface microlayer and seawater were collected from five samples stations within the coastal waters of Hong Kong in March 2005. The detection/quantification limit of the analytical method used for decaBDE was 240 pg/l in the dissolved phase and 410 pg/l in the suspended particulate matter phase. DecaBDE was not detectable or detectable in only trace amounts (at concentrations below the limit of quantification) in the samples.

An investigation into the levels of decaBDE and nonaBDE in riverine runoff samples from eight major outlets within the Pearl River Delta in China has been carried out by Guan et al. (2007). The samples were collected from either nine or fifteen sampling points at each location at monthly intervals between March 2005 and February 2006. The levels of decaBDE were in the range 0.33-65 ng/l (the concentrations are for the dissolved plus particulate phase). An annual input of decaBDE to the coastal ocean of 1,960 kg/year was estimated through this source. The study also included nonaBDEs (BDE-206, BDE-207 and BDE-208) but the concentrations of these individual congeners are not given (they are however shown graphically and appear to be up to around 0.8 ng/l for BDE-206 and BDE-207 and 0.2 ng/l for BDE-208). A linear correlation between the concentrations of these congeners (particularly BDE-206) and the concentration of decaBDE was apparent.

Hoenicke et al. (2007) analysed 33 samples of surface water from the San Francisco Estuary for polybrominated diphenyl ethers. DecaBDE was detected in 52% of the samples at a concentration between 0.012 and 0.191 ng/l (the concentrations represent the sum of the dissolved and particulate phases). NonaBDEs (BDE-206, BDE-207 and BDE-208) were also included in the study. BDE-206 was detectable in 36 per cent of the samples at a concentration of 0.0006 to 0.0093 ng/l, BDE-207 was detectable in 45 per cent of the samples at a concentration of 0.0008 to 0.014 ng/l and BDE-208 was detectable in 33 per cent of the samples at a concentration of 0.0007 to 0.0071 ng/l. DecaBDE and nonaBDE were reported to be not detectable in sediments from the area.

Kolic et al. (2005) [ABST] investigated the levels of polybrominated diphenyl ethers in influent, effluent and sewage sludge from a waste water treatment plant(s). The location of the plant(s) is not given but was most probably in Canada. DecaBDE was reported to be present at a concentration 71 ng/l in influent, 4.6 ng/l in effluent before disinfection and 3.5 ng/l in effluent after disinfection. The concentration in biosolids was 650 µg/kg dry weight. Based on the difference between the influent and effluent concentration, a removal of around 93-95% can be estimated during waste water treatment.

Shin et al. (2007) determined the levels of decaBDE in samples of raw and digested sludge from 3 waste water treatment plants in Ontario, Canada. The levels found were in the range 200 to 500 µg/kg (it is not clear if these are wet or dry weight concentrations) and the levels in digested sludge were generally higher than in raw or primary sludge.

Gevao et al. (2006) determined the levels of polybrominated diphenyl ethers in samples of sewage sludge from three wastewater treatment plants in Kuwait. The concentration of total polybrominated diphenyl ethers was 23-1,559 µg/kg and decaBDE accounted for 70-99% of the total.

Hale et al. (2008) determined the levels of decaBDE (and nonaBDEs) in samples of waste water sludge from two research bases in the Antarctic (McMurdo and Scott research bases). The levels of decaBDE found in the samples were 1,320 µg/kg dry weight in a sample from the McMurdo research base and 219 µg/kg dry weight in a sample from the Scott research base. NonaBDEs were also present in the samples, with the concentration of BDE-206, BDE-207 and BDE-208 being 17, 19 and 7.6 µg/kg dry weight respectively in the sample from the McMurdo research base and 5.4, 5.8 and 3.2 µg/kg dry weight respectively from the Scott research base. DecaBDE was also found to be present in sediment at the outfall of the waste water treatment plant at the McMurdo research base at a concentration of 3,540 µg/kg organic carbon and the concentrations in sediment were found to decrease with increasing distance from the base.

DecaBDE and two nonaBDEs (BDE-206 and BDE-207) have been detected in a sample of freeze dried manure from a dairy farm (Agnihotri et al. 2005) [ABST]. The levels found were around 1.1 µg/kg dry weight for decaBDE and 0.07 µg/kg dry weight for the two nonaBDEs. The location of the dairy farm was not given in the paper but it may have been in Canada.

Summary of new data

Several new studies have investigated the levels of decaBDE in effluents, water and sewage sludge.

The levels of decaBDE in water are generally very low. Levels up to 2.6 µg/l have been measured in effluents from an industrial (polymer) site in Spain and decaBDE was associated mainly with the particulate phase in these samples. This is consistent with the predicted environmental concentration (PEC) for effluent from a polymer processing site of around 8 µg/l for decaBDE estimated in EC (2002). DecaBDE has also been determined to be present in landfill leachate samples in Sweden at concentrations up to 4.2 µg/l.

DecaBDE is a common contaminant of sewage sludge from waste water treatment plants throughout the world. In general, measured levels are in the region of up to a few milligrams per kilogram dry weight. This is consistent with the previous findings reported in EC (2002) and ECB (2004 and 2007). Where nonaBDEs have been determined in the same samples as decaBDE, the relative proportions of nonaBDEs to decaBDE is similar to that which would be expected based on the nonaBDE content in the commercial decaBDE products (which typically contain ≤3% nonaBDEs). There is therefore little evidence that any debromination of decaBDE to nonaBDEs has occurred in these samples.

The results of the second sampling campaign for sewage sludge under the conclusion (i) monitoring programme are now available. It is difficult to draw any meaningful conclusions in regard to any time trends seen (the levels found at some sites were slightly higher in 2007 compared with 2006 but for other sites the opposite trend was apparent). However, it is important to note that BDE-126, a congener that is being used in the monitoring programme as a potential marker for reductive debromination of decaBDE, was found in the sewage sludge samples from 2007, albeit at low levels (~0.1 µg/kg dry weight at both sites investigated). As noted above, it is only one of several degradation products that could be formed, and the total amounts of these products would be higher than suggested by BDE-126 alone. BSEF (2009) suggest that two further years' monitoring data should be collected before a decision can be made on the importance of this finding.

Although the available evidence, both in this report and the previous risk assessment reports (EC (2002), ECB (2004 and 2007)) suggests that decaBDE is not degraded significantly during waste water treatment processes, the occurrence of BDE-126 in sewage sludge does suggests that some (limited) degradation could be occurring. This congener has also been found in sediment and is considered further in Section 4.2.2.

The detection of high levels of decaBDE in aqueous waste streams at Antarctic research stations shows that point sources may be significant even in supposedly remote regions. This can complicate the assessment of long-range transport potential.

4.2.2 Sediment

Conclusion (i) monitoring study

The conclusion (i) monitoring programme resulting from the ESR assessments (ECB 2004 and 2007) includes the analysis of sediment samples from ten sites in the EU (the programme also includes sewage sludge (see Section 4.2.1) and birds' eggs (see Section 4.2.5)). A total of four composite samples are being collected from each site (future samples will be collected on a biennial basis), with each composite sample consisting of nine sub-samples from an area of approximately 100 m^2. The sediment samples are taken by mini Van Veen grab or box core, ensuring that only the top layer (~ 2 cm) - representing newly deposited sediment - is sampled. The sampling sites and the levels found in the survey to date are summarised in Table 4.6. The sampling sites were chosen based on sufficient sediment rate, lack of disturbance from fishing trawlers and representivity for several EU countries. In addition, several of these locations had been sampled in earlier studies.

The survey results are also shown graphically in Figure 4.1 and Figure 4.2, together with the results of earlier studies in the same areas. These data are summarised in the original risk assessment report (EC 2002) and include the DIFFCHEM study carried out in 1995 (Van Zeijl 1997) and a survey of the levels in samples in 2000 carried out by de Boer *et al*. (2001). No samples were available for the Dublin Harbour site for 2007.

Overall, no firm conclusions on the trends in levels in sediments can be drawn from the conclusion (i) monitoring programme for sediment as yet.

Table 4.6 Levels of decabromodiphenyl ether in sediment from the conclusion (i) monitoring programme.

Sample location	Sample identity	Concentration					
		2005		2006		2007	
		µg/kg dry weight	µg/kg organic carbon	µg/kg dry weight	µg/kg organic carbon	µg/kg dry weight	µg/kg organic carbon
Mersey estuary, Liverpool Bay, UK	1			757	42,291	736	41,582
	2			779	37,816	764	41,297
	3			734	39,676	691	40,174
	4			794	42,460	785	41,099
	Mean (± standard deviation)			**766±26**	**40,561±2,230**	**744±41**	**41,040±610**
Thames estuary, UK	1			1.2	66	4.7	1,237
	2			1.1	58	10.8	2,455
	3			0.98	44	4.2	1,135
	4			1.6	117	3.0	769
	Mean (± standard deviation)			**1.2±0.27**	**71±32**	**5.7±3.5**	**1,400±730**
Humber estuary (outer), UK	1			<0.25	<417	0.17	155
	2			<0.25	<167	0.17	283
	3			0.95	452	<0.10	<250
	4			<0.25	<167	0.18	200
	Mean (± standard deviation)			**0.33±0.41**[1]	**207±173**[1]	**0.14±0.06**[1]	**191±69**[1]
Tees Bay, UK	1			12	6,000	6.0	349
	2			5.8	4,143	8.8	481
	3			7.4	4,625	8.6	455
	4			5.2	2,899	6.2	380
	Mean (± standard deviation)			**7.6±3.1**	**4,417±1,282**	**7.4±1.5**	**420±62**

Sample location	Sample identity	Concentration					
		2005		2006		2007	
		µg/kg dry weight	µg/kg organic carbon	µg/kg dry weight	µg/kg organic carbon	µg/kg dry weight	µg/kg organic carbon
Western Scheldt, Shaar Ouden Doel, the Netherlands/ Belgium	1	1,340	71,200			570	54,300
	2	1,620	52,400			400	43,500
	3	1,580	54,000			410	46,100
	4	630	35,600			430	46,200
	Mean (± standard deviation)	**1,293±459**	**53,300±14,549**			**543±79**	**47,500±4,690**
Dublin harbour/Liffey estuary, Ireland	1			22	3,607		
	2			14	2,692		
	3			17	3,269		
	Mean (± standard deviation)			**18±4**	**3,189±463**		
Mersey river, Manchester area, UK	1			209 [3]	23,483 [3]	381	57,727
	2			97 [3]	33,448 [3]	239	42,679
	3			932 [3]	73,386 [3]	207	32,857
	4			1,746 [3]	86,010 [3]	372	44,819
	Mean (± standard deviation)			**746±762 [3]**	**54,082±30,299 [3]**	**300±90**	**44,520±10,230**
Elbe estuary, Germany	1	3.5	187			0.61 [2]	95 [2]
	2	2.4	94			0.77 [2]	65 [2]
	3	1.9	67			0.76 [2]	62 [2]
	4	2.1	86			0.79 [2]	71 [2]
	Mean (± standard deviation)	**2.5 ± 0.7**	**109 ± 54**			**0.73±0.09 [2]**	**73±15 [2]**

Sample location	Sample identity	Concentration					
		2005		2006		2007	
		µg/kg dry weight	µg/kg organic carbon	µg/kg dry weight	µg/kg organic carbon	µg/kg dry weight	µg/kg organic carbon
Eems estuary, Germany	1	11.2	267			5.6	212
	2	8.0	208			8.3	257
	3	6.6	147			12	328
	4	9.8	286			6.9	191
	Mean (± standard deviation)	**8.9±2.0**	**227±63**			**8.2±2.8**	**247±61**
Seine estuary, France	1	3.3	418			8.3	535
	2	5.7	399			13	710
	3	13.7	1,370			5.9	440
	4	8.3	643			9.7	539
	Mean (± standard deviation)	**7.8±4.5**	**708±455**			**9.2±3.0**	**556±112**

Notes:

[1] For the estimate of the mean, not detected values were taken to be one half of the detection limit.

[2] Detectable but the concentration was below the limit of reliable quantification. Therefore there is a reduced certainty in the reported concentration.

[3] In ECB (2007) these results are reported to be from 2007 based on information given in Leslie et al. (2007). The Leslie et al. (2008) report indicates that these data refer to samples from 2006 and gives further data for 2007.

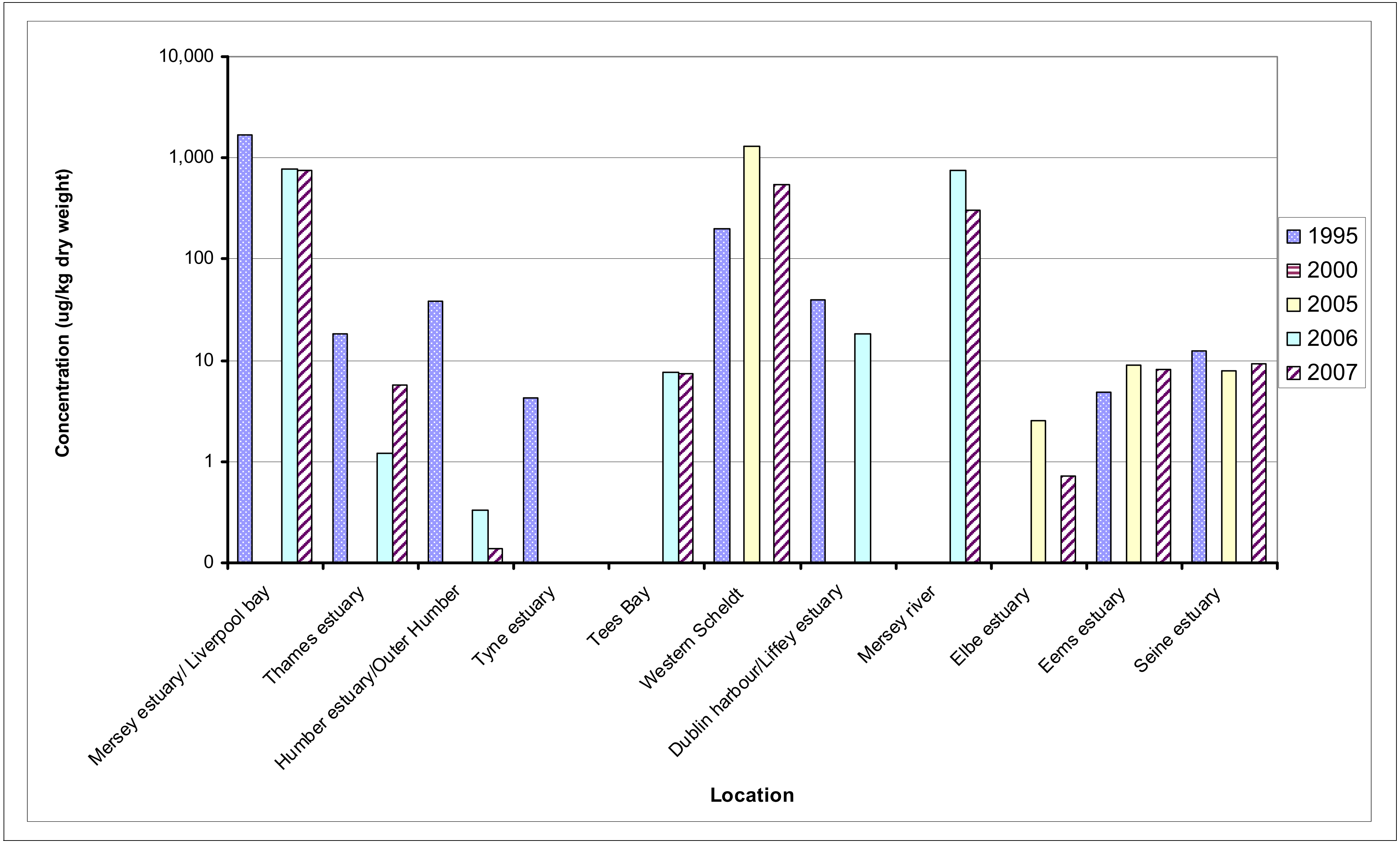

Figure 4.1 Results of the conclusion (i) monitoring study for sediment (concentrations expressed on a dry weight basis).

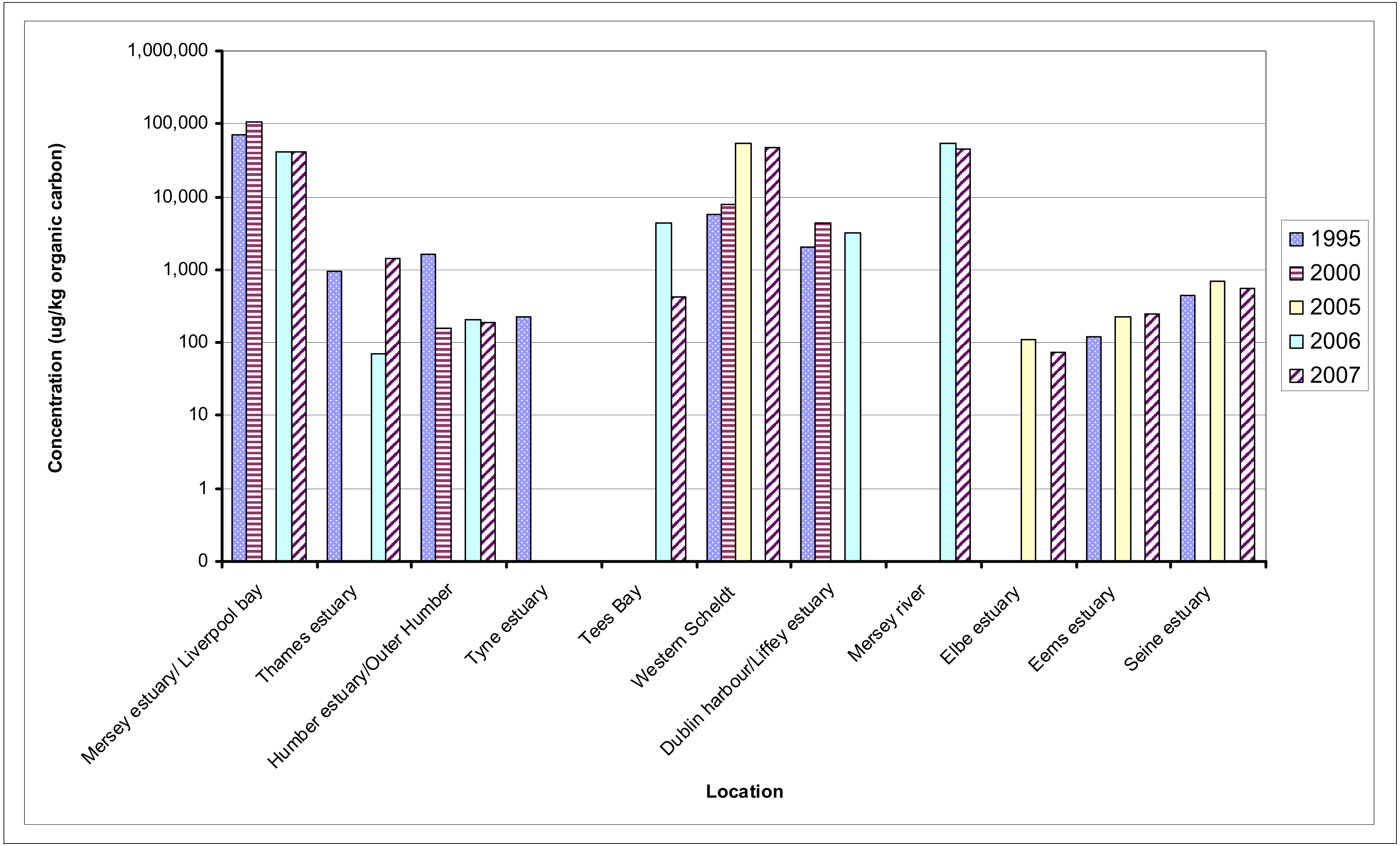

Figure 4.2 Results of the conclusion (i) monitoring study for sediment (concentrations expressed on a organic carbon basis).

As well as decabromodiphenyl ether, some of the samples (two samples each from the Western Scheldt, Elbe, Eems and Seine estuaries) were screened for the presence of lower brominated congeners. In particular, the presence of BDE-126 (3,3',4,4',5-pentabromodiphenyl ether) was determined, as a possible marker congener to determine if degradation reactions under anaerobic, reducing conditions occur in the environment. BDE-126 was found to be present above the limit of detection (0.03 µg/kg dry weight) in five of the eight samples but most of the concentrations could not be accurately quantified (the approximate concentration range was 0.03 to 0.29 µg/kg dry weight). The finding of BDE-126 in the current survey is potentially significant as it indicates that debromination to lower brominated congeners (in this case to a pentaBDE) could be occurring in the environment. Although the concentrations are very low, other degradation products could also be expected to be present as pointed out in Section 4.2.1.

Other data

The levels of decaBDE and nonaBDEs found in sediment are summarised in Table 4.7.

Europe

DecaBDE has been found in 32 sediments from the River Danube and its main tributaries at a median concentration of 3.29 µg/kg dry weight (range not detected to 83.8 µg/kg dry weight; the detection limit was not stated). DecaBDE was detectable in 93% of the samples and the levels found were generally lower in rural regions than in industrialised areas. The highest level (83.8 µg/kg dry weight) was found at the Moson Danuber arm in an industrialised region with textile, electronics, plastics and automotive industries (Sawal et al., 2005) [ABST].

Eljarrat et al. (2007) found elevated levels of decaBDE in surface sediment (0 to 2 cm depth) samples from the Vero River, Spain, downstream of an industrial park. The samples were collected in November 2004 and again in November 2005. The levels of decaBDE found were 7.5 and 26.9 µg/kg dry weight eleven km upstream of the industrial park, 12,459 µg/kg dry weight five metres downstream of the industrial park, 5,395 µg/kg dry weight one km downstream of the industrial park and 1,911 and 7,454 µg/kg dry weight five km downstream of the park. The samples collected in 2005 were also analysed for nonaBDEs (BDE-206, BDE-207 and BDE-208). No nonaBDEs were detectable in the upstream sample (concentration <0.05 µg/kg dry weight). The levels in the sample collected five metres downstream were 375 µg/kg dry weight for BDE-206, 235 µg/kg dry weight for BDE-207 and 169 µg/kg dry weight for BDE-208 and the levels in the sample collected five km downstream were 108 µg/kg dry weight for BDE-206, 100 µg/kg dry weight for BDE-207 and 60.2 µg/kg dry weight for BDE-208. The main source of decaBDE in the samples was thought to be from a polyamide polymerisation site. Samples of effluent from the industrial park and fish from the River Vero were also sampled. The results for these are summarised in Section 4.2.1 and Section 4.2.5 respectively.

Table 4.7 Levels of decaBDE and nonaBDE in sediment.

Location	Comments	Congener	Concentration	Reference
Europe				
River Danube and tributaries	32 Surface sediment samples. DecaBDE detected in 93% (30 samples).	DecaBDE	3.29 µg/kg dry weight (median) 83.8 µg/kg dry weight (maximum)	Sawal et al. (2005)
Sediment core from Lake Greigensee, Switzerland	Concentrations relate to the surface layer corresponding to 2001.	BDE-206	0.12 µg/kg dry weight	Kohler et al. (2008)
		BDE-207	0.070 µg/kg dry weight	
		BDE-208	0.076 µg/kg dry weight	
		DecaBDE	7.2 µg/kg dry weight	
Surface sediments close to an industrial park on the River Vero, Spain	11 km upstream, 2004.	DecaBDE	7.5 µg/kg dry weight	Eljarrat et al. (2007)
	11 km upstream, 2005.	BDE-206	<0.05 µg/kg dry weight (not detected)	
		BDE-207	<0.05 µg/kg dry weight (not detected)	
		BDE-208	<0.05 µg/kg dry weight (not detected)	
		DecaBDE	26.9 µg/kg dry weight	
	1 km downstream, 2004.	DecaBDE	5,395 µg/kg dry weight	
	5 m downstream, 2005.	BDE-206	375 µg/kg dry weight	
		BDE-207	235 µg/kg dry weight	
		BDE-208	169 µg/kg dry weight	
		DecaBDE	12,459 µg/kg dry weight	
	5 km downstream, 2004.	DecaBDE	1,911 µg/kg dry weight	
	5 km downstream, 2005.	BDE-206	108 µg/kg dry weight	
		BDE-207	100 µg/kg dry weight	
		BDE-208	60.2 µg/kg dry weight	
		DecaBDE	7,454 µg/kg dry weight	
Sediment cores, Clyde Estuary, United Kingdom	Six sediment cores collected between November 2002 and February 2003	DecaBDE	1 to 2,337 µg/kg dry weight	Vane et al. (2009)

Location	Comments	Congener	Concentration	Reference
Rest of the world				
Surface sediments, Australia	46 samples from locations across all eight State and Territories collected in 2002/2003 and 2005.	DecaBDE	4.2 µg/kg dry weight (mean) 0.88 µg/kg dry weight (median)	Toms et al. (2007) [ABST] and Toms et al. (2008)
Suspended sediments, Niagara River, Canada	Archived samples of suspended sediment covering the period 1980 to 2004. The concentration reported relates to 2003-2004.	Total PBDEs (decaBDE was the predominant congener present)	2.4-34 µg/kg dry weight	Marvin et al. (2007)
Sediment core, Lake Ontario	Core taken from the central basin of Lake Ontario. The highest concentration was in the surface layer.	DecaBDE	14 µg/kg dry weight (surface layer)	Qiu et al. (2007)
Sediment and flood plain soil from the Saginawa River watershed, Michigan	10 flood plain soils, Shiawassee River.	DecaBDE	0.60 to 41 µg/kg dry weight (range) 11 µg/kg dry weight (mean)	Yun et al. (2008)
	28 sediments, Shiawassee River.	DecaBDE	0.11 to 13 µg/kg dry weight (range) 2.3 µg/kg dry weight (mean)	
	10 flood plain soils, Saginaw River.	DecaBDE	<0.04 to 19 µg/kg dry weight (range) 2.8 µg/kg dry weight (mean)	
	20 sediments, Saginawa River.	DecaBDE	0.08 to 48 µg/kg dry weight (range) 4.8 µg/kg dry weight (mean)	
	6 flood plain soils, Saginaw Bay.	DecaBDE	<0.04 to 2.2 µg/kg dry weight (range) 0.6 µg/kg dry weight (mean)	
	5 sediments, Sagnawa River.	DecaBDE	<0.01 to 5.8 µg/kg dry weight (range) 2.0 µg/kg dry weight (mean)	

Location	Comments	Congener	Concentration	Reference
Surface sediments from Asia	7 coastal sediments from Indonesia, 1998.	DecaBDE	9.9 µg/kg dry weight (mean)[1] 0.03-54 µg/kg dry weight (range)	Sudaryanto et al. (2007b) [ABST]
	7 coastal sediments from Malaysia, 1998.	DecaBDE	8.3 µg/kg dry weight (mean)[1] 0.16-29 µg/kg dry weight (range)	
	11 sediments from the Mekong River Delta, Vietnam, 2003.	DecaBDE	0.60 µg/kg dry weight (mean)[1] <0.03-3.1 µg/kg dry weight (range)	
	6 sediments from the Vientiane Canal, Laos, 2005.	DecaBDE	8.2 µg/kg dry weight (mean)[1] 0.91-20 µg/kg dry weight (range)	
	7 sediments from the Middle Mekong River, Laos, 2005.	DecaBDE	0.22 µg/kg dry weight (mean)[1] <0.03-1.3 µg/kg dry weight (range)	
	14 sediments from the Mekong tributary, Thailand, 2005.	DecaBDE	0.22 µg/kg dry weight (mean)[1] <0.03-1.6 µg/kg dry weight (range)	
	9 sediments from Tokyo Bay, Japan, 2003-2004.	DecaBDE	20 µg/kg dry weight (mean)[1] 0.89-85 µg/kg dry weight (range)	
	27 sediments from coastal locations and rivers, Ehime, Japan, 2005-2006.	DecaBDE	2.4 µg/kg dry weight (mean)[1] Not detected-18 µg/kg dry weight (range)	
	28 sediments from coastal locations, Korea, 2005.	DecaBDE	18 µg/kg dry weight (mean)[1] 0.35-98 µg/kg dry weight (range)	
Sediment cores from the Pearl River Estuary, China	Concentrations related to the layers in three sediment cores corresponding from 1990 onwards.	Octa- and nonaBDEs	Not detected-6.6 µg/kg dry weight (range)	Chen et al. (2007)
		DecaBDE	1.1-61.5 µg/kg dry weight (range)	
Surface sediments, Yangtze River, China	Samples from 13 locations on the lower reaches of the river collected in 2004 and 2005.	BDE-207	~0.23 µg/kg dry weight (mean)	Shen et al. (2006)
		DecaBDE	~0.32 µg/kg dry weight (mean)	

Location	Comments	Congener	Concentration	Reference
Sediments from China	6 bank sediments, Nanyang River, close to an electronic equipment recycling site, 2004.	DecaBDE	17 to 62 µg/kg dry weight (range) 36 µg/kg dry weight (mean)	Luo et al. (2007)
	6 bottom sediments, Nanyang River, close to an electronic equipment recycling site, 2004.	DecaBDE	14 to 54 µg/kg dry weight (range) 31 µg/kg dry weight (mean)	
	5 bottom sediments, Lianjiang River, close to a residential area, 2004.	DecaBDE	15 to 57 µg/kg dry weight (range) 30 µg/kg dry weight (mean)	
	Bottom sediments from a river receiving waste water from a motor vehicle repair workshop, Lo Uk Tsuen, 2004.	DecaBDE	8.1 to 8.8 µg/kg dry weight (range) 6.0 µg/kg dry weight (mean)	
	Bottom sediments from fish ponds in Mai Po Marshes, 2004.	DecaBDE	<1 µg/kg dry weight (not detected)	
Sediment cores, Tokyo Bay, Japan	3 sediment cores. Concentrations relate to the surface layers corresponding to 1998-2000.	DecaBDE	~20 to ~80 µg/kg dry weight (range)	Tanabe et al. (2007) [ABST]
Surface sediments and sediment cores, Tokyo Bay, Japan	3 sediment cores. The concentrations relate to the surface layers corresponding to 1998-2000.	DecaBDE	20 to 85 µg/kg dry weight (range)	Minh et al. (2007)
	6 surface sediments, 2002.	DecaBDE	0.89 to 18 µg/kg dry weight (range)	
Marine sediment from coastal areas of Korea	Samples from 25 locations collected in 2004.	Total PBDEs (decaBDE accounted for >90% of the total)	0.45-494 µg/kg dry weight (range) 27.7 µg/kg dry weight (mean)	Moon et al. (2005) [ABST], Moon et al. (2007a) and Tanabe et al. (2008)

Location	Comments	Congener	Concentration	Reference
Marine sediments from 3 bays in Korea.	Samples from 111 locations in 3 bays collected in 2003-2004.	DecaBDE	2.0-2,249 µg/kg dry weight (range)	Moon et al. (2007c) and Moon et al. (2007d) [ABST]
Marine sediments from Masan Bay, Korea	Samples from 21 locations collected in 2004.	Total PBDEs (decaBDE as >80% of the total)	1.24-20 µg/kg dry weight (range)	Hong et al. (2007) [ABST]

Notes: [1] It is not clear from the paper if these are actually mean or median values. It has been assumed here that they are mean values.

Kohler et al. (2008) investigated the temporal trends of decaBDE and nonaBDE (BDE-206, BDE-207 and BDE-208) in sediments from Swiss lakes. A sediment core was taken from the deepest part (31 m depth) of Greigensee (a small urban lake close to Zürich) in April 2003 and dated sections were analysed for a range of polybrominated diphenyl ethers (precautions were taken during the analysis to prevent photodegradation of the polybrominated diphenyl ethers). The level of decaBDE was found to be 7.2 µg/kg dry weight in the layer corresponding to 2001, decreasing to 6.7, 4.1, 1.9 and 1.1 µg/kg dry weight in the layers corresponding to 1995, 1989, 1982 and 1974 respectively. Similarly the level of nonaBDE (the sum of BDE-206, BDE-207 and BDE-208) was found to be 0.26 µg/kg dry weight in 2001, decreasing to 0.16, 0.12 and 0.03 µg/kg dry weight in the layers corresponding to 1995, 1989 and 1974 respectively (the 1982 layer was not analysed for nonaBDE). A doubling time of approximately 9 years was estimated for decaBDE from these data for the period between 1982 and 2001. The surface flux in 2001 was estimated to be 830 pg/cm^3 per year for decaBDE and 30 pg/cm^3 per year for nonaBDE.

Kohler et al. (2008) also considered the changes over time in the congener pattern of the nonaBDEs and octaBDEs present in the sediment core. For the nonaBDEs, the congener pattern was broadly similar in all years with BDE-206 ≈ BDE-207 > BDE-208, indicating that once in the sediment there was little evidence for long-term transformation of these congeners. However, the pattern seen did not correspond to either the congener distribution in the commercial decabromodiphenyl ether products (dominated by BDE-206) or the commercial octabromodiphenyl ether products (dominated by BDE-207)[8]. It is likely that the pattern seen in the sediment reflects release from both commercial octa- and decabromodiphenyl ether products[9] but other factors, such as differences in partitioning behaviour or degradation of decaBDE, could also be important.

For the congener profile of the octaBDEs present in the sediment, again the pattern seen was relatively consistent over each year with BDE-197/204 ≈ BDE-193/203 ≈ BDE-196/200 ≈ BDE-201 > BDE-202 > BDE 205 ≈ BDE-194, indicating that no major transformation processes were occurring in the sediments. Again the congener profile did not correspond with the profiles found in commercial decabromodiphenyl ether products (dominated by BDE-196/200) or commercial octabromodiphenyl ether products (dominated by BDE-197/204) suggesting that both commercial products contribute to the levels seen in sediment. Of particular note, however, is that BDE-202 was present in the sediment. This congener was not found to be present in significant amounts in either the commercial decabromodiphenyl ether or octabromodiphenyl ether products analysed suggesting that the occurrence in the sediment samples could result from the degradation of the higher brominated congeners. For example Kohler et al. (2008) indicate that BDE-202 has been found as a degradation product of BDE-208 using anaerobic sewage sludge in the study by Gerecke et al. (2005), as reported in ECB (2007). The identity of this congener was not known at the time of publication of that study. Nevertheless, it should be noted that the supporting information for the

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[8] The actual commercial products analysed included two octabromodiphenyl ether products from Great Lakes (DE-79 and another unknown product) and three decabromodiphenyl ether products (Bromkal 82-ODE, an unknown product from Great Lakes and a technical product of 98% purity from Aldrich). No BDE-202 was detectable in the three decabromodiphenyl ether products or one of the octabromodiphenyl ether products but a trace amount of BDE-202 was found in the other octabromodiphenyl ether product. It should be noted, however, that the BDE-202 content of other products which are currently supplied, or have been supplied in the past, is unknown.

[9] The composition and relative use (and hence input) of these product groups could also vary over time. For example, BSEF (2009) has indicated that the commercial decaBDE provided by the major EU suppliers has had a purity higher than 97 per cent for more than ten years, but the congener profile of historical production, as well as current production by other companies, has not been so well characterised.

paper indicates that BDE-202[10] was present in a sample of “clean” historical sediment (from a sampling depth of 42-43 cm, corresponding to the year 1848) that had been spiked with a commercial decabromodiphenyl ether product. This finding is not commented on in the paper and its significance is unknown.

Vane et al. (2009) investigated the levels of decaBDE in sediment cores from the inner Clyde Estuary, UK. The sediment cores were collected between November 2002 and February 2003 from six sites on the estuary and the levels of decaBDE were determined in samples taken at approximately 10 cm intervals down each core. The limit of detection of the analytical method used was 1 µg/kg dry weight and decaBDE was detected in all six cores at concentrations ranging between 1 µg/kg dry weight and 2,337 µg/kg dry weight. The concentration of decaBDE in four of the six sediment cores showed a generally increasing concentration with decreasing depth in the core, with the highest concentrations being found at or near the surface of the core. However, the plots of the concentrations against depth given in the paper suggest that in three of these cores the concentration at the surface was similar to, or slightly lower than the concentration at 10 cm depth. One core showed little or no variation of the concentration of decaBDE with depth and another core showed a maximum concentration at a depth of around 35 cm with a general trend of decreasing concentration with decreasing depth. Vane et al. (2009) suggested that the pattern seen in these two cores could be due to vertical sedimentary mixing, dredging or sediment movement during storm events amongst others. This makes it difficult to draw firm conclusions as to whether the concentrations of decaBDE are increasing or decreasing in these samples as a whole. The study also investigated the levels of nonaBDEs in the sediment cores but the actual concentrations found are not given (it is reported that they accounted for, on average, around 11 per cent of the total polybrominated diphenyl ethers in the samples).

Rest of the world

A survey of the levels of polybrominated diphenyl ethers in sediments from Australia has been carried out by Toms et al. (2007) [ABST] and Toms et al. (2008). A total of 46 surface sediments from 39 locations across all eight States and Territories were collected in 2002-2003 (six samples were collected in 2005). Five of the sites were in remote areas, two were in remote/agricultural areas, seven were in agricultural areas, eleven were in urban areas, nine were in urban/industrial areas, one was in an industrial/urban/agricultural area, seven were in industrial areas and four were close to waste water treatment plant outfalls. Twenty of the sites were freshwater, twenty five were estuarine and one was marine. Polybrominated diphenyl ethers were detected in samples from 35 out of the 46 sites and the total polybrominated diphenyl ether concentration was up to 60.9 µg/kg dry weight but was less than 1 µg/kg dry weight at 83% of the sites. DecaBDE was the predominant congener in 30 of the samples (contributing 43 to 100 per cent of the total) and the mean and median concentrations of decaBDE were 4.2 µg/kg dry weight and 0.88 µg/kg dry weight respectively. The concentrations of total polybrominated diphenyl ethers in the estuarine sediment samples were higher (around ten times higher) than those in freshwater sediment samples. The estuarine locations were generally close to potential diffuse urban sources and point sources (industry or waste water treatment plants), whereas the

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[10] BSEF (2009) indicate that there is currently no certified analytical standard for BDE-202. Most studies have used samples of BDE-202 (and BDE-196 and BDE-197) that were gifts from a university or other researchers. Little information has been published about the synthesis or confirmation of identity of these samples, thereby introducing some uncertainty into the identification and quantification of this congener. However, the synthesis and characterisation of BDE-202 appears to be given in a paper by Teclechiel et al. (2007) and so this may in practice be less of a concern than indicated by BSEF (2009).

freshwater locations were generally inland and mainly distant from major urban and industrial centres.

Marvin et al. (2007) carried out a temporal trend study of the levels of polybrominated diphenyl ethers in archived samples of suspended sediment from the Niagara River, Canada. Samples were collected between 1982 and 2002 from both the head (Fort Erie) and mouth (Niagara-on-the-Lake) of the river. The levels of decaBDE in the samples from Niagara-on-the-Lake were relatively constant between 1980 and 1988 (concentration in the range 5 to 10 µg/kg dry weight) and showed an increasing trend from 1989 onwards (reaching around 15-20 µg/kg dry weight in the late 1990s early 2000s; the data are only shown graphically and there is some variability between years). DecaBDE was the predominant congener in these samples. In addition to the archived samples, further samples were collected from the two locations in 2003 and 2004. Only total polybrominated diphenyl ether levels are given for these samples and the concentration was reported to be in the range 2.4 to 34 µg/kg dry weight (typically 5 to 20 µg/kg dry weight in samples from Niagara-on-the-Lake and <5 µg/kg dry weight in samples from Fort Erie).

A study of the temporal trends in the levels of polybrominated diphenyl ethers in sediment has been carried out by Li et al. (2005 [ABST] and 2006). A total of 199 sediment samples from sixteen locations within the five Great Lakes were analysed for polybrominated diphenyl ethers (the results of these analyses are considered in ECB (2007)). Based on these measurements Li et al. (2006) estimated that the total amount of decaBDE thought to be accumulated in the Great Lakes was around 92 tonnes in 2002, with the total load in each lake estimated as 3.89 tonnes in Lake Superior, 38.8 tonnes in Lake Michigan, 9.04 tonnes in Lake Huron, 18.5 tonnes in Lake Erie and 21.6 tonnes in Lake Ontario. The annual input flux in 2002 was estimated to be around 0.10 tonnes/year to Lake Superior, 0.75 tonnes/year to Lake Michigan, 0.58 tonnes/year to Lake Huron, 1.62 tonnes/year to Lake Erie and 1.31 tonnes/year to Lake Ontario. The estimates for the input flux to the lakes showed an exponential increase from the 1970s to 2002 with a doubling time of between 7 and >70 years for decaBDE with the doubling time at 13 out of the 16 sampling locations being between 7 and 28 years and being 62 and 74 years at two remaining sites (the doubling time could not be estimated at a further site).

Qiu et al. (2007) determined the levels of decaBDE in a sediment core from the central basin of Lake Ontario. The highest concentration was found in the surface layer (mean concentration 14 µg/kg dry weight). The surface flux was estimated as 0.43 ng/cm^2 per year based on these data.

The levels of polybrominated diphenyl ethers have been determined in over 120 floodplain soil, surface sediment and sediment core samples collected in 2004 from the Shiawassee River and Saginaw Bay, Michigan (Yun et al. 2008). In addition, sediment samples collected from the Pine River and Tittabawassee River in 2002 were also analysed. DecaBDE was the predominant congener found in the samples, accounting for between 68 per cent and 93 per cent of the total polybrominated diphenyl ethers in the samples. The highest concentrations of decaBDE measured were 41 µg/kg dry weight in floodplain soil and 48 µg/kg dry weight in sediment.

A survey of the levels of decaBDE in surface sediments from several Asian countries (including Japan, Korea, Vietnam, Laos, Malaysia and Indonesia) has been carried out by Sudaryanto et al. (2007) [ABST]. The samples were collected between 1998 and 2005 and decaBDE was found to be present in concentrations ranging from <0.03 µg/kg dry weight to 98 µg/kg dry weight. The higher levels were generally found in samples close to human activities (e.g. canals and harbours) and industrial sites. The levels found in individual countries are summarised in Table 4.7. The study also found that nonaBDEs (BDE-206 and BDE-207) were present in the samples but the levels found were only shown graphically in the paper as a proportion of the total PBDEs found.

Chen et al. (2007) investigated time trends in the levels of polybrominated diphenyl ethers in sediment cores from the Pearl River Estuary in South China. The sediment cores were taken from the eastern and western sides of the Pearl River Estuary in April 2004 (two cores) and January 2006 (one core). The cores were sectioned and dated (only two of the three cores could be accurately dated) and the levels of polybrominated diphenyl ethers in the various layers (covering the time period from approximately 1975 onwards) determined. The concentration of decaBDE found in the sediment was found to show a generally increasing trend with time. The levels in the layers corresponding to pre-1990 were in the range 0.43 to 0.63 µg/kg dry weight, 2.0 to 3.3 µg/kg dry weight and 0.1 to 1.6 µg/kg dry weight in the three cores respectively, with the concentrations being relatively constant or showing a slowly increasing trend with time. However, in the layers corresponding to post-1990 a dramatic increase in the concentration was evident, with doubling times of approximately 2.6 years and 6.4 years for the two sediment cores which could be accurately dated (the range of concentrations determined in the post-1990 layers of the three sediment cores were 1.1 to 6.8 µg/kg dry weight, 2.5 to 13.9 µg/kg dry weight and 2.4 to 61.5 µg/kg dry weight respectively). The core collected in 2006 was also analysed for the presence of octaBDEs (BDE-196 and BDE-197) and nonaBDEs (BDE-206, BDE-207 and BDE-208). These were reported to be present at total concentrations up to 6.6 µg/kg in the post-1990 layers and up to 0.3 µg/kg in the pre-1990 layers. Based on these data Chen et al. (2007) estimated the average surface flux of decaBDE to the Pearl River Estuary to be around 29.7 ng/cm^2 per year.

Shen et al. (2006) determined the levels of decaBDE and a nonaBDE (BDE-207) in surface sediments from the lower reaches of the Yangtze River. The results are displayed graphically only and the approximate mean concentrations found (read from the graph) are approximately 0.32 µg/kg dry weight for decaBDE and approximately 0.23 ug/kg dry weight for BDE-207.

The levels of decaBDE have been determined in sediments from the Nanyang River, China close to a site which processed electronic waste (Luo et al. 2007). Samples were collected from six locations along the river bank (sampling depth 0 to 10 cm) and also from the river bottom (sampling depth 0 to 30 cm). In addition bottom sediments (sampling depth 0 to 30 cm) were collected from five locations on the Lianjiang River (close to a residential area) and bottom sediments (sampling depth 0 to 10 cm) were collected from a river receiving waste water from a motor vehicle repair workshop in Lo Uk Tsuen. Sediments (sampling depth 0 to 30 cm) were collected from fish ponds in Mai Po Marshes to serve as a control site. The mean levels of decaBDE found were 36 µg/kg dry weight in the bank samples from the Nanyang River, 31 µg/kg dry weight in the bottom samples from Nanyang River, 30 µg/kg dry weight in the bottom samples from the Lianjiang River, 6 µg/kg dry weight in bottom samples from the discharge in Lo

Uk Tsuen and decaBDE was not detectable (<1 µg/kg dry weight) in the sediments from Mai Po Marshes. Samples of fish from the areas were also analysed but decaBDE was generally not detectable in these samples (it was reported to be detectable only in samples of tilapia (*Oreochromis sp.*) abdomen muscle from the Lianjiang River at a mean concentration of 0.01 µg/kg wet weight).

Tanabe et al. (2007) [ABST] measured the levels of decaBDE in three archived dated sediment cores from Tokyo Bay, Japan. Polybrominated diphenyl ethers first appeared in the samples in the layers corresponding to the mid-1940s and the level of decaBDE showed a continual increase with time with the highest concentrations (~20 to ~80 µg/kg dry weight; values read from graph) occurring in the surface layers corresponding to 1998-2000. The same results also appear briefly in Sudaryanto et al. (2007) [ABST].

Similar (possibly the same) results are given in a paper by Minh et al. (2007). In this study sediment cores were collected from three locations in Tokyo Bay in 2002. The levels of decaBDE in the surface layers of these cores were 20, 46 and 85 µg/kg dry weight. In addition, Minh et al. (2007) analysed the levels of decaBDE in surface sediments from a further six locations in Tokyo Bay. The concentration of decaBDE in these samples was in the range 0.89 to 18 µg/kg dry weight. The concentrations of decaBDE were generally highest in the samples collected close to municipal areas of the bay and showed a decreasing trend towards the mouth of the bay. The sediment cores indicated an increasing trend in the concentration with time with the highest concentrations being present in the surface layers (corresponding to 1998-2000).

Tanabe et al. (2008) considered the sediment core data obtained by Minh et al. (2007) in a review article on the temporal trends of polybrominated diphenyl ethers in Japan and China. Based on the concentrations found in the cores, Tanabe et al. (2008) estimated doubling time for the decaBDE concentration of between 4.6 and 7.9 years. Another review by Law et al. (2008) also concluded that there is evidence of increasing levels of decaBDE in Asia.

The levels of polybrominated diphenyl ether levels in sediment samples from 25 coastal locations in Korea have been determined (Moon et al. 2005 [ABST] and Moon et al. 2007a). The samples were surface sediments (0-3 cm depth) and were collected during February-May 2004. Samples of bivalves were also collected from the same locations and the results for these are given in Section 4.2.5. A total of 20 congeners were found to be present in the sediments and the total polybrominated diphenyl ether levels were 0.45 to 494 µg/kg dry weight (mean concentration 27.7 µg/kg dry weight). DecaBDE was the predominant congener in the samples and accounted for >90 per cent of the total concentration measured. The same results are also reported by Tanabe et al. (2008).

Another survey of the levels of decaBDE in sediments from Korea has been carried out by Moon et al. (2007c and 2007d [ABST]). In this study surface (0-4 cm depth) sediment samples were collected at 111 locations within three industrialised bays (Ulsan Bay, Busan Bay and Jinhae Bay). The samples were collected between February 2003 and March 2004. DecaBDE was found to be present in all samples at a concentration between 2.0 and 2,248 µg/kg dry weight and was found to be the predominant polybrominated diphenyl ether in the samples. The highest levels were found in samples from inner locations (e.g. close to harbours) in Busan Bay. No correlation was found between the levels of decaBDE present and the levels of lower brominated congeners (e.g. tri- to hexaBDE) present in the samples.

Hong et al. (2007) [ABST] investigated the levels of polybrominated diphenyl ethers in sediments from a further bay in Korea. In this study surface (top 2 cm) sediments were collected in February 2004 at 21 locations within Masan Bay (a heavily industrialised are with a high population density). The levels of total polybrominated diphenyl ethers found were in the range 1.24 to 20 μg/kg dry weight and the levels were found to generally decrease along a gradient from the inner areas of the bay to the outer areas of the bay. The one exception to this trend was a location that received effluent from a sewage treatment plant (this location had the highest total polybrominated diphenyl ether level out of all sites). DecaBDE was found to be the predominant congener present, accounting for >80% of the total polybrominated diphenyl ethers present.

Summary of new data

A number of new studies investigating the levels of decaBDE in sediment have been published since the last ESR risk assessment was completed. Several of these have investigated time trends using sediment cores. These studies generally show an increasing trend in the levels of decaBDE in sediments from the Great Lakes and Asia. In addition, one study shows increasing trends in sediment levels in Switzerland. It should be noted that the most recent years included in these sediment core studies are generally up to around 2002 and so it is difficult to infer the current trend in the levels of decaBDE in the UK and EU from these data (since voluntary industry emission reduction activity began in 2004). A sediment core study from the UK covering up to 2003 (Vane et al. 2009) was generally inconclusive as to the current trend in levels.

Further results are also available from the conclusion (i) monitoring study. No clear trend, either upwards or downwards, in the concentrations in sediment in the EU is yet apparent from this study. However, the conclusion (i) monitoring study showed that a marker congener (BDE-126) for possible reductive debromination of decaBDE was present in several sediment samples from 2007, albeit at low levels (up to about 0.3 μg/kg dry weight, although the actual concentrations could not be quantified). This is an important finding as it suggests that some debromination of decaBDE is occurring in the environment. In addition, the sediment core study by Kohler et al. (2008) found evidence for the presence of BDE-202 (an octaBDE) in some of the samples. This congener is not thought to be present in commercial polybrominated diphenyl ether products and has been suggested to be a degradation product of higher brominated congeners.

4.2.3 Soil

The levels of decaBDE and nonaBDE measured in soil are summarised in Table 4.8.

Europe

Eljarrat et al. (2008) determined the levels of decaBDE in samples of soil from Spain. The sites chosen had been used for cultivation of winter crops and the soils had been amended with sewage sludge at least once since 1997. A control soil to which no sludge had been applied was also included. The samples were taken from a depth of 0 to 20 cm in 2005 and the sampling rate was approximately five subsamples per hectare. DecaBDE was found to be present in all soils. The concentrations found in the sewage sludge amended soils were in the range 24.6 to 655 µg/kg dry weight compared with 14.6 µg/kg dry weight in the control soil. The highest concentration was measured in a soil that had received repeated additions of sewage sludge since 1997 and this suggested that decaBDE can build-up in soils following repeated additions of sewage sludge, indicating that decaBDE is persistent in soils. NonaBDEs (BDE-206, BDE-207 and BDE-208) were also found to be present in some samples. The levels found were in the range <1.4 to 7.3 µg/kg dry weight for BDE-206, <1.4 to 6.0 µg/kg dry weight for BDE-207 and <1.4 to 4.5 µg/kg dry weight for BDE-208. NonaBDEs were not quantifiable (concentration <1.4 µg/kg dry weight) in the control soil.

Rest of the world

Wang et al. (2008) analysed two soil samples using a new analytical methodology designed to optimise the simultaneous analysis of both lower brominated congeners (in this case mono- to heptaBDEs) and the higher brominated congeners (e.g. octa- to decaBDEs). The soil samples were collected from the surface layer (depth 0 to 15 cm) from a field where fire-fighter training was undertaken in the United States (furniture and other combustibles were burned at the site). The soil samples were taken at a distance of three feet and six feet from the combustion site.

The level of decaBDE found was found to be 41 or 159 µg/kg dry weight at a distance of three feet from the site and 40 or 221 µg/kg dry weight at a distance of six feet from the site (two different analytical methods were used on each sample). NonaBDEs (BDE-206 and BDE-207) were also present in the samples. The concentration of BDE-206 was 1.3 or 1.5 µg/kg dry weight at a distance of three feet from the site and not detected or 0.72 µg/kg dry weight at a distance of six feet and the concentration of BDE-207 was 1.0 or 2.4 µg/kg dry weight at a distance of three feet and not detected or 0.41 µg/kg dry weight at a distance of six feet.

The levels of polybrominated diphenyl ethers in soil from a rural area of Ansung City, South Korea have been determined by Yeo et al. (2006) [ABST]. The levels of total polybrominated diphenyl ethers were between 2.4 and 1,997 ng/kg dry weight in soil samples. The most abundant congener present was reported to be decaBDE. Pine needles were also analysed from the same area (see Section 4.2.5).

Liu et al. (2008) determined the levels of polybrominated diphenyl ethers in soils from towns in South China close to electronic equipment recycling sites. The soil samples (about 1 m depth) were collected from farmland and roadsides in various locations within a town with an electronic equipment recycling site, a second town close to this town, and two further towns distant from the electronic equipment recycling site (controls). The levels of total polybrominated diphenyl ethers found in the samples were in the range 0.4 to 789 µg/kg dry weight, with the mean values being 142 µg/kg dry weight at the town with the electronic equipment recycling site, 2.5 µg/kg dry weight at the nearby town and 0.57 and 0.48 µg/kg dry weight a the two control sites). The levels of decaBDE are shown only graphically in the paper but decaBDE appears to be the predominant congener present in the sample. Samples of plants and snails were also collected from some locations and decaBDE appears to have been present in some of these samples, particularly from the town with the electronic equipment recycling site (again the levels found are shown only graphically and are not totally clear).

Table 4.8 Levels of decaBDE and nonaBDE in soil.

Sample	Comments	Congener	Concentration	Reference
Europe				
Sewage-sludge amended soil samples from Spain	Soil from Tornabous sampled in April 2005. Sludge was applied in 2003 (25 tonnes/ha), 2004 (25 tonnes/ha) and January 2005 (33 tonnes/ha).	BDE-206	3.8 µg/kg dry weight	Eljarrat et al. (2008)
		BDE-207	2.0 µg/kg dry weight	
		BDE-208	<1.4 µg/kg dry weight (below the quantification limit)	
		DecaBDE	332 µg/kg dry weight	
	Soil from Hostalets de Pierola sampled in October 2005. Sludge applications in 2003 (15 tonnes/ha) and September 2005 (15 tonnes/ha).	BDE-206	<1.4 µg/kg dry weight (below the quantification limit)	
		BDE-207	<1.4 µg/kg dry weight (below the quantification limit)	
		BDE-208	<1.4 µg/kg dry weight (below the quantification limit)	
		DecaBDE	24.6 µg/kg dry weight	
	Soil from Pujalt sampled in October 2005. Sludge applications in 1999, 2000 and 2001 (application rate unknown).	BDE-206	<1.4 µg/kg dry weight (below the quantification limit)	
		BDE-207	<1.4 µg/kg dry weight (below the quantification limit)	
		BDE-208	<1.4 µg/kg dry weight (below the quantification limit)	
		DecaBDE	71.7 µg/kg dry weight	
	Soil from Pujalt sampled in October 2005. Sludge application in 2004 (application rate unknown).	BDE-206	2.0 µg/kg dry weight	
		BDE-207	<1.4 µg/kg dry weight (below the quantification limit)	
		BDE-208	<1.4 µg/kg dry weight (below the quantification limit)	
		DecaBDE	161 µg/kg dry weight	

Sample	Comments	Congener	Concentration	Reference
Sewage-sludge amended soil samples from Spain	Soil from Pujalt sampled in October 2005. Yearly sludge applications between 1997 and 2004 (15-20 tonnes/ha) and again in August 2005 (15 tonnes/ha).	BDE-206	7.3 µg/kg dry weight	Eljarrat et al. (2008)
		BDE-207	6.0 µg/kg dry weight	
		BDE-208	4.5 µg/kg dry weight	
		DecaBDE	655 µg/kg dry weight	
	Soil from Pujalt sampled in October 2005. No sludge application (reference site).	BDE-206	<1.4 µg/kg dry weight (below the quantification limit)	
		BDE-207	<1.4 µg/kg dry weight (below the quantification limit)	
		BDE-208	<1.4 µg/kg dry weight (below the quantification limit)	
		DecaBDE	14.6 µg/kg dry weight	
Rest of the world				
Sample close to a fire-fighter training site, United State	Surface sample (0-15 cm depth) taken at a distance of 3 and 6 feet from the site.	BDE-206	Not detected-1.5 µg/kg dry weight	Wang et al. (2008)
		BDE-207	Not detected-2.4 µg/kg dry weight	
		DecaBDE	40-221 µg/kg dry weight	
Soil from South Korea.	Soil samples from a rural area of Ansung City. The levels refer to total polybrominated diphenyl ethers but decaBDE was reported to be the most abundant congener present.	Total BDEs	0.0024-2.0 µg/kg dry weight	Yeo et al. (2006) [ABST]
Soil from towns close to an electronic equipment recycling site, China.	Samples taken from a town with an electronic recycling site, a nearby town and two distant (control) towns in 2005.	Total PBDEs (decaBDE appears to be the predominant congener)	0.40-789 µg/kg dry weight	Liu et al. (2008)
Soil from Guiyu, China, close to an electronic equipment recycling site	Samples from various locations within the town. The highest concentrations were from a site where open burning was occurring.	Total PBDEs (decaBDE accounted for 63-81% of the total)	2-44,473 µg/kg dry weight	Wong et al. (2007)

Sample	Comments	Congener	Concentration	Reference
Soil from Pearl River Delta, China	33 surface soils, 2002 and 2005	DecaBDE	2.4 to 67 μg/kg dry weight (range) 14 μg/kg dry weight (mean)	Zou et al. (2007)
	Surface soils close to three point sources (electronic waste dismantling sites), 2005	DecaBDE	26 to 102 μg/kg dry weight (range)	

Another investigation of the levels of polybrominated diphenyl ethers in soils close to an electronic waste recycling facility in China has been carried out by Wong et al. (2007). In this study a total of 49 surface soil samples (top 10 cm) were collected between June and December 2004 from various locations in the town (Guiyu). The levels of total polybrominated diphenyl ethers were found to be between 2 and 44,473 µg/kg dry weight in the sample and decaBDE accounted for 63 to 81 per cent of the total. The highest concentrations were from a site where open burning of waste electrical equipment was occurring. Sediment samples were also included in the study but the levels of decaBDE in these were not clear from the paper (the total levels were in the general range 4 to 407 µg/kg dry weight but much higher levels were apparent in areas with large amounts of burnt residues).

Zou et al. (2007) determined the levels of decaBDE in 33 surface soils, six soil profiles and three point-source polluted soils (close to electronic waste dismantling sites) from the Pearl River Delta, China. The concentration of decaBDE was between 2.4 and 67 µg/kg dry weight (average 14 µg/kg dry weight) in the surface soils and was between 26 and 102 µg/kg dry weight in the soils near to the point source. The soil profiles showed that the concentration of decaBDE generally decreased with increasing depth.

Summary of new data

New data are available on the levels of decaBDE in soils from Europe, North America and Asia. Elevated levels in soil (up to 44 mg/kg dry weight) have been measured at sites close to electronic equipment recycling sites in China although the relevance of these data to the situation in the UK and EU is unclear. Levels of up to 0.66 mg/kg dry weight have been measured in soil from Spain that receives applications of sewage sludge and these values appear to be in line with the data on levels in soil that have been reported previously in EC (2002) and ECB (2004 and 2007).

4.2.4 Air and dust

The levels of decaBDE and nonaBDE found in air are summarised in Table 4.9. The levels found in both outdoor and indoor air are given. Although the indoor air data are relevant mainly for human exposure they are potentially useful for highlighting possible sources of release to air, and hence ultimately the environment.

Outdoor air

Conclusion (i) monitoring study

As part of the conclusion (i) monitoring programme, a preliminary investigation of decabromodiphenyl ether in air samples at a site in the United Kingdom has been undertaken by Thomas and Jones (2007). The site is at Hazelrigg (a well characterised field and meteorological station around 4 km from Lancaster in north-west England) in a semi-rural location at a height of 94 m above sea level. The levels of decaBDE and nonaBDEs (BDE-206, BDE-207 and BDE-208) were determined in a total of nine two-day air samples collected over a period of two months during May to June 2007. The samples collected represented the total (i.e. particulate plus vapour phase) concentration. The quality control/quality assurance procedures included the routine analysis of field blank samples and exposure to UV-light was minimised during the extraction and clean-up procedure. DecaBDE was found to be present in all samples in the range 6.8 to 89 pg/m^3. The median and geometric mean concentrations of decaBDE found were 15 and 18 pg/m^3 respectively. The same location had been sampled previously in 2005 (see ECB (2007) for a summary of the findings). The geometric mean concentration of decaBDE found in 2007 was 18 pg/m^3, which was not statistically significantly different from that found in 2005 (15 pg/m^3).

For the nonaBDEs, the geometric mean concentrations found in the 2007 survey were 0.43 pg/m^3 for BDE-206, 0.90 pg/m^3 for BDE-207 and 0.27 pg/m^3 for BDE-208. These compare with geometric mean concentrations of 0.42 pg/m^3 for BDE-206, 0.69 pg/m^3 for BDE-207 and 0.49 pg/m^3 for BDE-208 found in the 2005 survey. Again the levels found in 2007 were not statistically significantly different from those found in the 2005 survey.

In contrast to the findings for decaBDE and nonaBDEs, Thomas and Jones (2007) found a statistically significant decrease (95% confidence limit) in the concentration of two octaBDEs (BDE-196 and BDE-197) had occurred in 2007 compared with 2005. The geometric mean concentration in air in the 2007 survey was 0.15 pg/m^3 for BDE-196 and 0.21 pg/m^3 for BDE-197 compared with a geometric mean concentration of 0.29 pg/m^3 for BDE-196 and 0.47 pg/m^3 for BDE-197 in the 2005 survey. This result is of interest as it shows that the levels of octaBDE in air are falling (presumably as a result of restrictions in the EU on the use of the commercial octabromodiphenyl ether products) while the levels of decaBDE and nonaBDE have remained relatively constant over the same time period. This suggests that formation of octaBDE (by photolysis/atmospheric degradation of decaBDE) might not make a significant contribution to the *current* levels of octaBDE found in these air samples. At the same time, this study cannot be used as evidence that this mechanism is not relevant.

Europe – other data

The levels of total PBDEs in ambient air in Vienna have been determined to be in the range 13 to 36 pg/m^3 (Moche et al. 2007 [ABST]). The samples were collected over eighteen consecutive days (six three-day samples were collected) in each of winter and summer 2006 and the samples represented the total (i.e. particulates plus vapour phase) concentration. DecaBDE was the predominant congener found in the winter samples, accounting for 57 per cent to 94 per cent (mean 77 per cent) of the total, and around 40 per cent of the total PBDEs in the summer samples.

Wilford et al. (2008) determined the levels of decaBDE and nonaBDEs (BDE-206, BDE-207 and BDE-208) in the particulate phase of air from a semi-rural site in north-west England. The samples were collected daily between 17 April and 20 May 2004. The quality control/quality assurance procedures included the routine analysis of field blank samples and exposure to UV light was minimised during the extraction and clean-up procedure. The reported concentrations were blank-corrected and the detection limits were set at at least three times the standard deviation of the blank levels. DecaBDE was detectable in eighteen of the twenty eight airborne particulate samples at a median concentration of 13 pg/m^3 (range <0.49 to 100 pg/m^3). NonaBDEs (BDE-206, BDE-207 and BDE-208) were also present in some of the samples. BDE-206 was detected in seventeen of the samples at a median concentration of 1.1 pg/m^3 (range <0.048 to 13 pg/m^3), BDE-207 was detected in twenty three of the samples at a median concentration of 2.5 pg/m^3 (range <0.042-79 pg/m^3) and BDE-208 was detected in twenty three of the samples at a median concentration of 0.93 pg/m^3 (range <0.048-12 pg/m^3).

Wilford et al. (2008) also carried out some investigations into the levels of nonaBDEs and decaBDE in the vapour phase at this site. The vapour phase samples were collected over seven seven-day periods during the study and decaBDE was found in three of these samples (the detection limit was 0.55 pg/m^3; the levels of decaBDE found were not clear from the paper). In addition small amounts of nonaBDEs were found in one of the vapour phase samples. Wilford et al. (2008) speculated that the presence of decaBDE and nonaBDE in the vapour phase samples most probably represented substance adsorbed onto very fine particles (<1 µm diameter) that were not trapped by the glass fibre filters used to collect particulates and so passed through to the polyurethane foam plugs that were used to collected substances in the vapour phase.

An interesting finding in this study was that the nonaBDEs constituted a higher proportion of the total polybrominated diphenyl ether levels in the particulate samples than would be expected based on their reported occurrence in the commercial decabromodiphenyl ether products. For example, the median contribution of decaBDE, BDE-206, BDE-207 and BDE-208 to the total level of polybrominated diphenyl ethers found in the particulate samples was 60 per cent, 4 per cent, 16 per cent, and 6 per cent respectively and the median concentration of total nonaBDE was around 45 per cent of the concentration of decaBDE (whereas 3 per cent may be expected based on the known amounts in current commercial decabromodiphenyl ether products supplied in the EU). Wilford et al. (2008) discussed a number of possibilities to explain this enhancement in the concentration of nonaBDE over that of decaBDE including higher volatility of nonaBDEs than decaBDE leading to a preferential release to the vapour phase, historical use of decaBDE (some older commercial products had a decaBDE content of around 70 per cent with a correspondingly higher nonaBDE content), release of nonaBDEs from the use of commercial octabromodiphenyl ether products and (photo)degradation of decaBDE. The first possibility was ruled out as a result of an analysis of the relationship of the concentration of nonaBDEs with the concentration of particles within selected size bands. This found strong indications that nonaBDEs were associated mainly with the larger size particles (>3 µm diameter) which are associated with windblown dusts from mechanical processes rather than smaller particles formed of soot and condensed vapours. This suggested that nonaBDEs (and decaBDE) were predominantly attached to particles formed by abrasion of articles in use rather than volatilisation from articles.

For the possibility that the elevated levels of nonaBDEs (relative to the decaBDE levels) arise as a result of a contribution of nonaBDE from the commercial octabromodiphenyl ether product, the study also determined the levels of BDE-183 (this is known to be a major congener in the commercial octabromodiphenyl ether product). This congener was detected in eleven of the twenty-eight samples analysed in the range <0.048 to 92 pg/m^3, with the majority of the levels being ≤ 5.6 pg/m^3 (levels higher than this were measured on two occasions: 25 pg/m^3 and 92 pg/m^3). A correlation analysis between the concentrations measured for the various congeners was undertaken. This analysis was carried out using log$_{10}$ transformed values with outliers (tested against the 95% confidence limit on the regression) removed and both including and excluding samples with concentrations higher/lower than most other samples. This showed significant correlations ($p<0.01$) between decaBDE, BDE-208, BDE-207 and BDE-206 but not with BDE-183, suggesting that the nonaBDEs were derived from the same source as decaBDE.

This analysis therefore leads to two likely possible explanations for the congener pattern seen: debromination of decaBDE to nonaBDEs or elevated levels of nonaBDE resulting from the past use of commercial decabromodiphenyl ether products with higher nonaBDE contents. Wilford et al. (2008) did not distinguish between these two possibilities further. As noted in Section 3, nonaBDE impurity levels in the decaBDE products made in China appear to be somewhat higher than that produced elsewhere.

Table 4.9 Levels of decaBDE and nonaBDE in air.

Sample	Comments	Congener	Concentration	Reference
Europe				
Ambient air in Vienna	Sampled for 18 days each in winter and summer 2006.	Total PBDEs (decaBDE accounted for around 77% of the total in winter and around 40% of the total in summer)	0.013-0.036 ng/m^3 (range)	Moche et al. (2007) [ABST]
Outdoor air at a semi-rural site in the United Kingdom	Samples (particulates plus vapour phase) from Hazelrigg as part of the conclusion (i) monitoring programme. A total of nine 2-day samples collected in May-June 2007.	BDE-206	<0.00017-0.00093 ng/m^3 (range) 0.00043 ng/m^3 (geometric mean)	Thomas and Jones (2007)
		BDE-207	<0.00030-0.0020 ng/m^3 (range) 0.00090 ng/m^3 (geometric mean)	
		BDE-208	<0.00007-0.00085 ng/m^3 (range) 0.00027 ng/m^3 (geometric mean)	
		DecaBDE	<0.0068-0.089 ng/m^3 (range) 0.018 ng/m^3 (geometric mean)	

Sample	Comments	Congener	Concentration	Reference
Outdoor air at a semi-rural site in the United Kingdom	Airborne particulates sampled between 17 April and 20 May 2004.	BDE-206	<0.000048-0.013 ng/m^3 (range) 0.0011 ng/m^3 (median)	Wilford et al. (2008)
		BDE-207	<0.000042-0.079 ng/m^3 (range) 0.0025 ng/m^3 (median)	
		BDE-208	<0.000048-0.012 ng/m^3 (range) 0.00093 ng/m^3 (median)	
		DecaBDE	<0.00049 and 0.1 ng/m^3 (range) 0.013 ng/m^3 (median)	
Indoor air, Sweden	Samples collected from 5 homes. DecaBDE detected in only one home.	BDE-206	<0.027 ng/m^3	Karlsson et al. (2007)
		BDE-207	<0.027 ng/m^3	
		DecaBDE	<0.17 to 0.26 ng/m^3	
Rest of the world				
Indoor and outdoor air at a computer laboratory, United States.		DecaBDE	0.002-0.077 ng/m^3 (indoor air) 0.004-0.018 ng/m^3 (outdoor air)	Cahill et al. (2007)
Indoor and outdoor air at a electronic equipment recycling facility, United States	Indoor levels taken in the dismantling hall.	DecaBDE	88-830 ng/m^3 (indoor air) 0.14-11 ng/m^3 (outdoor air)	Cahill et al. (2007)
Outdoor air at a automotive shredding facility, United States		DecaBDE	1.9 ng/m^3 (maximum)	Cahill et al. (2007)

Sample	Comments	Congener	Concentration	Reference
Residential indoor air exposure	20 residents in Boston, USA. Samples collected using personal air samplers. The concentration represents total exposure (particulates + vapour).	DecaBDE	0.174 ng/m^3	Allen et al. (2007b)
Outdoor air at Alert, Canada	Samples collected weekly over the period 2002 to 2004.	DecaBDE	0.000091 to 0.0098 ng/m^3 (range) 0.0016 ng/m^3 (mean)	Su et al. (2007a) [ABST] and Su et al. (2007b)
Indoor and outdoor air from Guangzhou, China	Samples of household, office and outdoor air samples, 2004-2005.	DecaBDE	0.039-11.5 ng/m^3 (indoor residential air) 0.080-13.7 ng/m^3 (indoor office air) 1.1-49.9 ng/m^3 (outdoor air)	Chen et al. (2008a)
Outdoor air at Waliguan Baseline Observatory, China	Samples collected in 2005.	Total PBDEs (decaBDE was the third most abundant congener)	0.002-0.015 ng/m^3 (range) 0.008 ng/m^3 (mean)	Cheng et al. (2007)
Car interiors	DecaBDE was detectable in 23 out of 33 car interiors.	DecaBDE	1.05 ng/m^3 (maximum) 0.15 ng/m^3 (median)	Mandalakis et al. (2008)

Karlsson et al. (2007) determined the levels of decaBDE and two nonaBDEs (BDE-206 and BDE-207) in samples of indoor air (particulate plus vapour phase) from five homes in Sweden. DecaBDE found was not detectable in air from four of the homes (concentration <0.17 ng/m^3) but was present at 0.26 ng/m^3 in one home. Neither BDE-206 or BDE-207 were detected in any of the samples (concentrations <0.027 ng/m^3). Samples of dust were also collected and analysed (see Table 4.10).

Rest of the world

Cahill et al. (2007) determined the levels of polybrominated diphenyl ethers in indoor air at a computer laboratory at the University of California, indoor and outdoor air from an electronics recycling facility and outdoors at an automotive shredding/metal recycling facility in the United States. The samples were collected in 2004 and the values determined represent the total (particulate phase plus vapour phase) concentrations. The levels of decaBDE found in outdoor air the University of California were between 4.4 and 18 pg/m^3 (mean 11 pg/m^3) (samples taken on two consecutive days). The levels found in the computer office were between 2.2 and 77 pg/m^3 (samples collected on eight days), and were in the range 2.2 to 18 pg/m^3 when the computers were off (two sampling occasions) and between 47 and 77 pg/m^3 when the computers were on. The level of decaBDE found inside the electronics recycling facility (dismantling hall) was between 320,000 and 830,000 pg/m^3 when shredding was occurring (two sampling occasions) and was around 80,000 pg/m^3 on one sampling occasion when shredding was not occurring. The levels in outdoor air outside electronics recycling facility were in the range 140 to 11,000 pg/m^3 (three sampling occasions). At the automotive shredding facility the levels of decaBDE were between not detected and 1,940 pg/m^3 in outdoor samples (three sampling occasions).

Allen et al. (2007b) measured the indoor air exposure to decaBDE of 20 residents in Boston, Massachusetts, using personal air samplers. The total personal air concentration (particulate + vapour) determined for decaBDE was 174 pg/m^3. The majority (>70 per cent) of decaBDE was thought to be present in the particulate phase.

Su et al. (2007a) [ABST] and Su et al. (2007b) report the results of the Arctic Monitoring and Assessment Programme (AMAP). This programme started monitoring the levels of polybrominated diphenyl ethers at Alert in Canada in 2002 with the aim of deriving long-term trends in concentration in Arctic air and the current papers consider the inter-annual variations in the concentrations of polybrominated diphenyl ethers over the period 2002 to 2004. Samples were collected weekly (each sample was collected over a seven-day period (representing ~13,000 m^3 air)) and the samples represented the total (i.e. particulate plus vapour phase) concentration in air. The study found that decaBDE was detected frequently in air. The average concentration of decaBDE found in the samples over the two year period was 1.6 pg/m^3 (range 0.091 to 9.8 pg/m^3). The authors also estimated the inter-annual time trend in the concentrations and determined that the levels of decaBDE found increased over the time period with a doubling time around 6 years.

Levels of decaBDE in indoor air (home and workplace) and outdoor air in Guangzhou, China have been determined by Chen et al. (2008a). The samples were collected from October 2004 to April 2005. The concentrations of decaBDE found were 39-11,468 pg/m^3 in air in homes, 80-13,732 pg/m^3 in office air and 1,082-49,937 pg/m^3 in outdoor air.

The levels of decaBDE in air from the Waliguan Baseline Observatory in northwestern China have been determined (Cheng et al. 2007). The observatory is part of the Global Atmospheric Watch network of the World Meteorological Organization and is located on the edge of the northeastern Tibetan Plateau at a height of 3,816 m above sea level. Air samples (both particulate and gaseous) were collected from the site between 2 April and 23 May 2005 and a total of 26 samples were collected (each sample was collected over a 24 hour period). The levels of total polybrominated diphenyl ethers measured at the site were ion the range 2.2 to 15 pg/m^3, with a mean of 8.3 pg/m^3. DecaBDE was reported to be the third most predominant congener in the samples (the actual levels of decaBDE found were not given).

Atmospheric bulk deposition samples from urban, suburban and rural sites in coastal areas of Korea have been analysed for polybrominated diphenyl ethers by Moon et al. (2007b). A total of 20 congeners were found to be present in the samples and the total depositional flux was between 10.1 and 89.0 µg/m^2 per year, with the highest levels being found in urban areas. DecaBDE was the predominant congener found (>93% of the total).

Mandalakis et al. (2008) determined the levels of polybrominated diphenyl ethers (including decaBDE) in air inside cars. The levels of total polybrominated diphenyl ethers were in the range 0.4 to 2,644 pg/m^3, with three congeners (BDE-47 (tetraBDE), BDE-99 (pentaBDE) and decaBDE) accounting for around 70 per cent of the total. DecaBDE was detectable in the interiors of 23 out of 33 cars tested and the highest concentration of decaBDE found was 1,053 pg/m^3 and the median concentration was estimated to be 150 pg/m^3. The concentration measured was found to be significantly correlated with the age of vehicle (higher concentrations in newer vehicles) and interior temperature (higher concentrations at higher temperatures).

Dust

Europe (including comparative studies with the rest of the world)

The levels of decaBDE and nonaBDEs in dust are summarised in Table 4.10. Although these data are relevant mainly for human exposure they are potentially useful for highlighting possible source of release to air, and hence ultimately the environment.

As part of a study to develop an improved methodology for the analytical determination of the concentrations of polybrominated diphenyl ethers, Regueiro et al. (2007) determined the levels in samples in household dust from Spain. DecaBDE was found to be present at a concentration of 58-1,615 µg/kg in the dust samples.

Fabrellas et al. (2005) [ABST] found decaBDE to be the predominant congener present in samples of household dust. A total of 100 samples of household dust (34 from Spain, 32 from Belgium, 22 from Portugal and 12 from Italy) were collected in the winter of 2003-2004 using domestic vacuum cleaners. The median levels of total polybrominated diphenyl ethers found in the samples were 98 µg/kg dry weight in the samples from Spain, 125 µg/kg dry weight in the samples from Belgium, 91 µg/kg dry weight in the samples from Portugal and 286 µg/kg dry weight in the samples from Italy. DecaBDE accounted for >60 per cent of the total polybrominated diphenyl ethers found in each sample (the levels found for decaBDE alone are not reported).

Table 4.10 Levels of decaBDE and nonaBDE in dust.

Sample	Comments	Congener	Concentration	Reference
Europe (including comparative studies with the rest of the world)				
Household dust samples from Spain		DecaBDE	58-1,615 μg/kg	Regueiro et al. (2007)
Household dust samples from Spain, Italy, Portugal and Belgium	34 samples from Spain collected in Winter 2003-2004.	Total PBDEs (decaBDE accounted for >60% of total)	98 μg/kg (median) 2,047 μg/kg (max)	Fabrellas et al. (2005) [ABST]
	12 samples from Italy collected in Winter 2003-2004.	Total PBDEs (decaBDE accounted for >60% of total	286 μg/kg (median) 3,986 μg/kg (max)	
	22 samples from Portugal collected in Winter 2003-2004.	Total PBDEs (decaBDE accounted for >60% of total	91 μg/kg (median) 2,720 μg/kg (max)	
	32 samples from Belgium collected in Winter 2003-2004.	Total PBDEs (decaBDE accounted for >60% of total	125 μg/kg (median) 711 μg/kg (max)	
Indoor dust samples from Belgium, Romania and Spain	8 samples from Antwerp, Belgium, 2004.	DecaBDE	8-307 μg/kg (range)	Dirtu et al. (2008)
	1 sample from Romania, 2004.	DecaBDE	27 μg/kg	
	1 sample from Spain, 2004.	DecaBDE	140 μg/kg	
Household dust samples from Sweden	Samples collected from 5 homes.	BDE-206	2.1 to 44 μg/kg	Karlsson et al. (2007)
		BDE-207	1.9 to 42 μg/kg	
		DecaBDE	44 to 1,560 μg/kg	

Sample	Comments	Congener	Concentration	Reference
Household dust samples from Australia, Germany, United Kingdom and United States	10 samples from Australia.	DecaBDE	730 μg/kg (median) 23-13,000 μg/kg (range)	Sjödin et al. (2008)
	10 samples from Germany.	DecaBDE	63 μg/kg (median) <6-410 μg/kg (range)	
	10 samples from United Kingdom.	DecaBDE	10,000 μg/kg (median) 910-54,000 μg/kg (range)	
	10 samples from the United States.	DecaBDE	2,000 μg/kg (median) 120-21,000 μg/kg (range)	
Household dust samples from UK, Canada and United States	Samples from Amarillo and Austin, Texas.	DecaBDE	1,300 μg/kg (median) 3,300 μg/kg (max)	Ibarra et al. (2007) [ABST] and Harrad et al. (2008)
	Samples from Birmingham, UK.	DecaBDE	2,800 μg/kg (median) 520,000 μg/kg (max)	
	Samples from Toronto, Canada.	DecaBDE	560 μg/kg (median) 1,100 μg/kg (max)	
Rest of the world				
Indoor dust samples from research bases in Antarctica.	McMurdo research base, 2005.	BDE-206	201 μg/kg	Hale et al. (2008)
		BDE-207	510 μg/kg	
		BDE-208	163 μg/kg	
		DecaBDE	4,160 μg/kg	
	Scott Antarctic research base, 2006.	BDE-206	56 μg/kg	
		BDE-207	69 μg/kg	
		BDE-208	43 μg/kg	
		DecaBDE	1,650 μg/kg	
Household dust from the United States	Samples of household dust from 11 homes in Greater Boston, Massachusetts. DecaBDE detected in 45% of the samples.	DecaBDE	<0.5 to 9.0 mg/kg (range)	Wu et al. (2007)

Sample	Comments	Congener	Concentration	Reference
Household dust samples, Boston, United States	20 living rooms, 2006.	BDE-206	87.4 µg/kg (geometric mean)	Allen et al. (2007a) [ABST]
		BDE-207	40.2 µg/kg (geometric mean)	
		BDE-208	27.9 µg/kg (geometric mean)	
		DecaBDE	4,602 µg/kg (geometric mean)	
	20 bedrooms, 2006.	BDE-206	43.2 µg/kg (geometric mean)	
		BDE-207	19.2 µg/kg (geometric mean)	
		BDE-208	9.2 µg/kg (geometric mean)	
		DecaBDE	1,667 µg/kg (geometric mean)	
	20 vacuum cleaner bags, 2006.	BDE-206	41.7 µg/kg (geometric mean)	
		BDE-207	25.2 µg/kg (geometric mean)	
		BDE-208	26.3 µg/kg (geometric mean)	
		DecaBDE	1,846 µg/kg (geometric mean)	
Household dust from Hawaii		BDE-206	5.5 µg/kg	Wang et al. (2008)
		BDE-207	0.95 µg/kg	
		DecaBDE	40 µg/kg	
Household dust samples from Singapore.	31 samples collected from various locations in Singapore.	DecaBDE	1,000 µg/kg (median)	Tan et al. (2007)
Household dust samples from Japan.	13 samples from 9 homes	BDE-206	<7-330 µg/kg (range)	Kono et al. (2007) [ABST]
		BDE-207	<10-180 µg/kg (range)	
		DecaBDE	14-3,200 µg/kg (range)	

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DecaBDE has also been reported to be present in indoor dust samples from Belgium by Dirtu et al. (2008). In this study eight samples of indoor dust were collected from Antwerp in 2004 and the level of decaBDE in the samples was in the range 8 to 307 µg/kg. In addition samples of indoor dust from Romania and Spain were also analysed. The decaBDE content of these samples was 27 µg/kg (Romania) and 140 µg/kg (Spain).

The levels of decaBDE and two nonaBDEs (BDE-206 and BDE-207) have been determined in samples of household dust from five homes in Sweden (Karlsson et al. 2007). The levels of decaBDE found were in the range 44 to 1,560 µg/kg, and the levels of BDE-206 and BDE-207 were in the range 2.1 to 44 and 1.9 to 42 µg/kg respectively. Samples of indoor air were also collected but the levels present were low (see Table 4.9).

Sjödin *et al.* (2008) also carried out a comparison between the levels in household dust in various countries. A total of forty samples (ten samples from each country) were collected from Australia (Brisbane), Germany (Hamburg, Harburg, Konstanz, Muenster, Berlin and Freiberg), United Kingdom (Newcastle upon Tyne) and the United States (Atlanta). The samples were collected from vacuum cleaner bags from private households. The median (and range) concentrations of decaBDE in the <2 mm particle fraction of the dust were 730 µg/kg (23-13,000 µg/kg) in the samples from Australia, 63 µg/kg (<6-410 µg/kg) in the samples from Germany, 10,000 µg/kg (910-54,000 µg/kg) in the samples from the United Kingdom and 2,000 µg/kg (120-21,000 µg/kg) in the samples from the United States.

A comparison of the levels of decaBDE in domestic indoor dust from Canada, the United Kingdom and the United States has been carried out by Ibarra et al. (2007) [ABST] and Harrad et al. (2008). Samples of indoor dust from ten to twenty eight homes in each of Toronto, Canada, Birmingham, United Kingdom and Amarillo and Austin, Texas, United States were collected. The median and maximum concentrations found were respectively 560 µg/kg and 1,100 µg/kg in the samples from Toronto, 2,800 µg/kg and 520,000 µg/kg in the samples from Birmingham and 1,300 µg/kg and 3,330 µg/kg in the samples from Amarillo and Austin. Two of the samples from Birmingham were found to have very high levels of decaBDE (100,000 µg/kg and 520,000 µg/kg) but the levels found in the remaining samples were generally similar to those found in the samples from the United States. However, the levels of decaBDE were found to be significantly higher in the samples from the United Kingdom compared with the samples from Canada.

Rest of the world

Hale et al. (2008) determined the levels of decaBDE (and nonaBDEs) in samples of indoor dust from two research bases in the Antarctic (McMurdo and Scott research bases). The levels of decaBDE found in the dust samples were 4,160 µg/kg in a sample from the McMurdo research base and 1,650 µg/kg in a sample from the Scott research base. NonaBDEs were also present in the samples, with the concentration of BDE-206, BDE-207 and BDE-208 being 201, 510 and 163 µg/kg respectively in the sample from the McMurdo research base and 56, 69 and 43 µg/kg respectively from the Scott research base.

Wu et al. (2007) found decaBDE to be present at concentrations up to 9.0 mg/kg in samples of household dust from Greater Boston, Massachusetts. A total of eleven samples were surveyed and decaBDE was detectable in five (45 per cent) of these samples. The detection limit was around 0.5 mg/kg.

Another study of the levels of decaBDE and nonaBDE in household dust in Boston, United States, has been carried out by Allen et al. (2007a) [ABST]. Samples were collected from the main living room, bedroom and from the household vacuum cleaner. The levels found for decaBDE (geometric mean) were 4,602 µg/kg in the living rooms, 1,667 µg/kg in the bedrooms and 1,846 µg/kg in the vacuum cleaner bags. The geometric mean levels of nonaBDEs were between 41.7 and 87.4 µg/kg for BDE-206, 25.2 and 40.2 µg/kg for BDE-207 and 9.2 and 27.9 µg/kg for BDE-208. The study also investigated the correlation between the levels of decaBDE found in dust and those found in air (the air levels were not reported in the paper) but no correlation was found.

Wang et al. (2008) analysed a sample of household dust from Hawaii using a new analytical methodology designed to optimise the simultaneous analysis of both lower brominated congeners (in this case mono- to heptaBDEs) and the higher brominated congeners (e.g. octa- to decaBDEs). The level of decaBDE found was 40 µg/kg dry weight. NonaBDEs (BDE-206 and BDE-207) were also present at a concentration of 5.5 µg/kg dry weight and 0.95 µg/kg dry weight respectively.

A survey of the levels of decaBDE in house dust from Singapore has been carried out by Tan et al. (2007). Samples were collected from thirty one homes and polybrominated diphenyl ethers were found to be present in all samples. DecaBDE was present at a median level of 1,000 µg/kg and contributed around 88% of the total polybrominated diphenyl ethers present. No significant correlation was found between the concentration present and factors such as number of TVs and computers, floor area or flooring material.

Kono et al. (2007) [ABST] found BDE-206, BDE-207 and decaBDE to be present in the range <7-330 µg/kg, <10-180 µg/kg and 14-3,200 µg/kg respectively in thirteen samples of house dust from nine homes in Japan. The samples were collected from around televisions, the upper parts of fluorescent lamps, air-conditioning filters, vacuum cleaner contents, the outside of washing machines, fan heater filters and floor surfaces.

Takigami et al. (2007) [ABST] investigate the relationship between the levels of polybrominated diphenyl ethers in household dust with those in indoor air in two houses in Japan. However the paper only reports the results as total polybrominated diphenyl ethers and does not given an indication as to the proportion that decaBDE or nonaBDE contributed to these levels.

Summary of new data

The available new data show that decaBDE occurs widely in indoor and outdoor air and dust. Levels up to 520 mg/kg have been measured in indoor dust from the United Kingdom. In air samples, decaBDE appears to be associated mainly with the particulate phase rather than the vapour phase, and the concentrations of decaBDE in outdoor air are generally <1 ng/m^3 in Europe. Higher levels have been measured in outdoor air from China (up to 50 ng/m^3) and from close to electronic equipment recycling facilities in the United States (up to 11 ng/m^3).

Of particular interest to this assessment are the data from the Wilford et al. (2008) study. This study provides evidence that the decaBDE found in indoor air is predominantly associated with particles formed by abrasion of articles (particle size >3 µm diameter), although a small fraction was associated with fine particulates (particle size <1 µm diameter) or the vapour phase. This indicates the importance of abrasion of articles as an emission source for decaBDE. The finding that decaBDE is mainly associated with the larger particulate fractions is also relevant in terms of the long-range transport potential for decaBDE (this is considered further below). Furthermore, the congener profile found in this study is suggestive of debromination of decaBDE to nonaBDEs in these samples, although as discussed above, other explanations are also possible.

Another interesting point is that decaBDE has been determined in outdoor air at monitoring stations in remote regions (e.g. Alert, Canada and the Waliguan Baseline Observatory in China). Although the levels found are generally very low (up to about 10 pg/m^3 and decaBDE was not detected in all samples analysed) the results do suggest that long-range transportation of decaBDE can occur. However, given the widespread use of articles containing decaBDE, local sources (e.g. the research stations themselves) cannot be totally ruled out. In addition, the data from Alert, Canada, suggest that the concentration in air at this location is increasing with time.

4.2.5 Biota and food

Aquatic species

The levels of decaBDE and nonaBDE in aquatic species (including marine mammals) are summarised in Table 4.11.

Europe

Eljarrat et al. (2007) determined the levels of decaBDE in fish from the River Vero, Spain, close to an industrial park. The samples analysed included barbel (*Barbius graellsii*) and carp (*Cyprinus carpio*). The samples were collected at a site eleven km upstream of the industrial park and five km downstream of the industrial park (samples from both sites were collected in November 2004 and again in November 2005). Muscle samples of the fish were analysed for decaBDE. The quality assurance procedures used included the routine analysis of procedural blanks and no decaBDE was found in these blank samples. DecaBDE was not detectable (concentration <0.019 µg/kg wet weight) in samples of barbel collected from the upstream site in both 2004 (a total of six samples were analysed) and 2005 (a total of eight samples were analysed). The mean (±standard deviation) found in the downstream samples were 66.5±89.2 µg/kg lipid in barbel collected in 2004 (eight samples), 79.5±22.4 µg/kg lipid in carp collected in 2004 (two samples) and 195±287 µg/kg lipid in barbel collected in 2005 (five samples). Effluent and sediment samples from the River Vero were also analysed for decaBDE. The results of these analyses are given in Section 4.2.1 and Section 4.2.2 respectively.

Table 4.11 Levels of decaBDE and nonaBDE in aquatic species (including marine mammals).

Sample	Comments	Congener	Concentration	Reference
Europe				
Mussel (*Mytilus edulis*)	10 pooled samples from coast of Scotland, 2006.	DecaBDE	0.032-0.094 µg/kg wet weight (range)	Fernandes et al. (2008)
Scallops (*Pecten maximus*)	10 pooled samples from coast of Scotland, 2006. The gonad and adductor samples were analysed separately.	DecaBDE	0.010-0.044 µg/kg wet weight (range in gonad) 0.006-0.027 µg/kg wet weight (range in adductor)	Fernandes et al. (2008)
Oysters (*Crassostrea gigas, Ostrea edulis*)	5 pooled samples from coast of Scotland, 2006.	DecaBDE	0.006-0.018 µg/kg wet weight (range)	Fernandes et al. (2008)
Zebra mussel (*Dreissena polymorpha*)	Pooled samples of 30-40 individuals from 14 sites at Lake Maggiore, Italy, 2005.	DecaBDE	4.6-84.6 µg/kg lipid (detected in 11 out of 14 samples)	Binelli et al. (2008)
Barbel (*Barbius graellsii*) and Carp *(Cyprinus carpio*)	Samples from 11 km upstream and 5 km downstream of a industrial park on the River Vero, Spain, 2004 and 2005.	DecaBDE	<0.019 µg/kg wet weight (not detected in upstream samples) 66.5-195 µg/kg lipid (range of mean values in downstream samples)	Eljarrat et al. (2007)
Common sole (*Solea solea L.*) muscle	Pooled samples of ten individuals from three locations off the coast of France, 2003 and 2004.	DecaBDE	1.76 µg/kg lipid (maximum) (not quantifiable in 4 out of the 6 samples)	Munschy et al. (2007) [ABST]
Harbour seal (*Phoca vitulina*) and harbour porpoise (*Phocoena phocoena*)	Blubber samples from 28 seals and 35 porpoise from the southern North Sea collected between 1999 and 2004.	DecaBDE	<10 µg/kg lipid (not detected)	Weijs et al. (2007)[ABST]
Ringed seal (*Phoca hispida*)	57 samples from 1982-2006.	DecaBDE	<0.91-5.6 µg/kg lipid (detected in 8 samples only)	Vorkamp et al. (2008)
Striped dolphin (*Stenella coeruleoalba*) skin biopsies	200 samples from three locations in the Mediterranean Sea, 2006.	DecaBDE	not detected	Jiménez et al. (2007) [ABST]

Sample	Comments	Congener	Concentration	Reference
Rest of the world				
Bivalves (*Mytilus edulis*, *M. coruscus* and *Crassostrea gigas*)	25 coastal locations in Korea collected in 2004.	Total PBDEs (decaBDE accounted for >60% of the total)	0.38 to 9.2 µg/kg wet weight 2.9 µg/kg wet weight (mean)	Moon et al. (2005) [ABST], Moon (2007a) and Tanabe et al. (2008)
Blue mussel (*Mytilus edulis*)	20 locations around the coast of Korea, 2005	DecaBDE	detectable in samples from eleven locations	Ramu et al. (2007a)
Blue mussel (*Mytilus edulis*) and green mussel (*Perna verdis*)	Samples from several locations in Asia, 2003-2005.	DecaBDE	<2-56 µg/kg lipid	Ramu et al. (2005) [ABST] and Ramu et al. (2007b)
Carp (*Cyprinus carpio*), snakehead (*Chana* sp.) and tilapia (*Oreochromis mossambicus*) muscle	10 snakehead from a location affected by a municipal waste water treatment plant in Laos, 2005.	BDE-206	0.017 µg/kg lipid (mean) <0.040-0.15 µg/kg lipid (range)	Sudaryanto et al. (2007a) [ABST]
		BDE-207	<0.13 µg/kg lipid (not detected)	
		DecaBDE	<0.57 µg/kg lipid (not detected)	
	5 snakehead from a rural area in Laos, 2005.	BDE-206	<0.040 µg/kg lipid (not detected)	
		BDE-207	<0.13 µg/kg lipid (not detected)	
		DecaBDE	<0.57 µg/kg lipid (not detected)	
	5 tilapia from an intensive aquaculture area of Laos, 2005.	BDE-206	<0.040 µg/kg lipid (not detected)	
		BDE-207	<0.13 µg/kg lipid (not detected)	
		DecaBDE	<0.57 µg/kg lipid (not detected)	
	7 tilapia from a lake/lagoon in an urban area in Laos, 2005.	BDE-206	<0.040 µg/kg lipid (not detected)	
		BDE-207	<0.13 µg/kg lipid (not detected)	
		DecaBDE	<0.57 µg/kg lipid (not detected)	
	3 carp from near a municipal waste dump site, Laos, 2005.	BDE-206	<0.040 µg/kg lipid (not detected)	
		BDE-207	<0.13 µg/kg lipid (not detected)	
		DecaBDE	<0.57 µg/kg lipid (not detected)	
Lake trout (*Salvelinus namaycush*)	Sample from Lake Ontario, Canda, collected in 1979, 1983, 1988, 1993, 1998 and 2004. 4-5 individuals collected in each year.	DecaBDE	2.3 to 12 µg/kg lipid (mean values) 0.27 to 1.3 µg/kg wet weight (mean values)	Ismail et al. (2009)

Sample	Comments	Congener	Concentration	Reference
Snapping turtle (*Chelydra serpentine*) eggs	Samples from 15 locations on Lake Erie, Lake Ontario, Detroit River and St. Lawrence River, 2001-2004.	DecaBDE	<0.01 µg/kg wet weight (not detected)	De Solla et al. (2007)
Harbour seal (*Phoca vitulina concolor*)	Blubber samples from eight seals from the northwestern Atlantic coast (United States) collected between 1991 and 2005.	DecaBDE	1-8 µg/kg lipid (detected in 4 out of 8 samples)	Shaw et al. (2007a) [ABST]
Harbour seal	Sample from Kodiak, Alaska, 2000.	BDE-206	<0.05 µg/kg (not detected)	Wang et al. (2008)
		BDE-207	<0.05 µg/kg (not detected)	
		DecaBDE	<0.05 µg/kg (not detected)	
Killer whale (*Orcinus orca*)	Blubber samples from 9 individuals collected off the coast of Japan in 2005.	DecaBDE	<0.5 µg/kg lipid (not detected)	Kajiwara et al. (2006)
Melon-headed whale (*Peponocephala electra*)	Archived blubber samples collected from mass strandings in Japan in 1982 (10 samples), 2001 (12 samples), 2002 (28 samples) and 2006 (5 samples).	DecaBDE	<0.5 µg/kg lipid (not detected)	Kajiwara et al. (2008b)

The levels of decaBDE in zebra mussels (*Dreissena polymorpha*) from Lake Maggiore, Italy have been determined (Binelli et al. 2008). Mussels were collected from fourteen sites within the lake in April 2005 and a pooled sample consisting of thirty to forty individuals from each site was analysed. DecaBDE was found to be present in procedural blank samples analysed alongside the mussel samples but the amounts of decaBDE present in the blank samples were well below ten per cent of those in the mussel samples. The detection limit of the analytical method is not entirely clear but appears to have been around 0.5 µg/kg wet weight. DecaBDE was detected in mussels from eleven of the fourteen sites investigated at concentrations ranging between 4.6 and 84.6 µg/kg lipid. The mussels do not appear to have been depurated prior to analysis and so the levels measured could have been influenced by the presence of contaminated sediments, etc., in the gut contents.

The levels of decaBDE in samples of shellfish from locations around the coast of Scotland have been determined by Fernandes et al. (2008). The samples were collected between January and March 2006 and included mussels (*Mytilus edulis*), oysters (*Crassostrea gigas, Ostrea edulis*) and scallops (*Pecten maximus*). Pooled samples were analysed and decaBDE was found to be present in all samples at a concentration of 0.032 to 0.094 µg/kg wet weight in mussels, 0.006-0.018 µg/kg wet weight in oysters, 0.010-0.044 µg/kg wet weight in scallop gonad and 0.006-0.027 µg/kg wet weight in scallop adductor. The shellfish do not appear to have been depurated prior to analysis. The quality assurance/quality control procedures used included frequent analysis of procedural blanks and reference materials.

Munschy et al. (2007) [ABST] reported that decaBDE was present in muscle samples of juvenile sole (*Solea solea* L.) from coastal areas of France. The samples were collected from three locations (Seine estuary in the English Channel and the Vilaine estuary and the Pertuis Breton in the Bay of Biscay) in both 2003 and 2004. The samples analysed were pooled from ten individuals. DecaBDE was detected in all samples analysed but the levels found did not exceed the levels found in procedural blank samples except for two samples (from the Seine estuary in 2003 and the Vilaine estuary in 2003) where the levels found were 1.76 µg/kg lipid and 0.73 µg/kg lipid respectively (these concentrations were reported to be >4 times the blank levels and were not blank corrected). NonaBDEs (BDE-207 and BDE-208) were also reported to be present at low levels but again the levels found were not always significantly different from the levels in the procedural blanks.

Weijs et al. (2007) [ABST] investigated the levels of decaBDE in blubber samples of 28 harbour seals (*Phoca vitulina*) and 35 harbour porpoises (*Phocoena phocoena*) stranded in the southern North Sea between 1999 and 2004. DecaBDE was not detectable in any sample (detection limit 10 µg/kg lipid).

Vorkamp et al. (2008) surveyed the levels of decaBDE in samples of blubber from ringed seals (*Phoca hispida*) from central west Greenland. The samples analysed included nine samples from 1982, nine from 1986, ten from 1994, nine from 2000 and twenty from 2006. The quality assurance/quality control procedures used included routine analysis of procedural blanks and internal reference materials. DecaBDE was found to be present in small amounts in some of the blanks and the detection limit was set above these blank values (the actual detection limit for decaBDE varied with the sample but was between 0.91 and 1.36 µg/kg lipid). DecaBDE was not detectable in the majority of the samples, except for one individual from 1982 (3.3 µg/kg lipid), one individual from 1994 (5.6 µg/kg lipid), one individual from 2000 (2.0 µg/kg lipid) and five out of twenty samples from 2006 (0.93-3.3 µg/kg lipid).

DecaBDE was not detectable in skin biopsies collected from two hundred striped dolphin (*Stenella coeruleoalba*) from three areas of the Mediterranean Sea (Gibraltar Straits, Pelagos Sanctuary (Ligurian Sea) and the West Ionian Sea (Sicily) in 2006 (Jiménez et al. 2007) [ABST]. Few other details are currently available.

Rest of the world

Moon et al. (2005 [ABST] and 2007a) determined the levels of polybrominated diphenyl ethers in bivalve samples (including mussels (*Mytilus edulis* and *M. coruscus*) and oysters (*Crassostrea gigas*)) from 25 coastal locations in Korea (the study also investigated the levels in sediment and these are summarised in Section 4.2.2). The samples were collected between February and May 2004. A total of 20 congeners were found to be present and the total levels present were between 0.38 and 9.2 µg/kg wet weight in bivalves. DecaBDE was the predominant congener present and accounted for >60% of the total in bivalves. The same results are also reported by Tanabe et al. (2008).

Ramu et al. (2007a) determined the levels of polybrominated diphenyl ethers in samples of blue mussels (*Mytilus edulis*) from twenty locations around the coast of Korea. DecaBDE was reported to be present “at variable levels” in samples from eleven locations. The mussels were not depurated prior to analysis so the detection of decaBDE could be due to the presence of sediment in the gut of mussels.

Another study of the levels of decaBDE in mussels has been carried out by Ramu et al. (2005 [ABST] and 2007b). In this study samples of green mussel (*Perna verdis*) and blue mussel (*Mytilus edulis*) were collected between 2003 and 2005 from various locations in Asia, including coastal locations in India, Indonesia, China, Hong Kong, Japan, Cambodia, Vietnam and the Philippines. DecaBDE was reported to be found in the mussel samples from China, Hong Kong and Indonesia (and on the occasional sample from other regions) and the levels in these mussels were in the general range <2 to 56 µg/kg lipid. In addition, the paper indicates that the mussels were not depurated prior to analysis and so the detection of decaBDE could be due to the presence of sediment in the gut of mussels.

DecaBDE was found to be not detectable (concentration <0.57 µg/kg lipid) in muscle samples of three species of fish (carp (*Cyrpinus carpio*), snakehead (*Channa* sp.) and tilapia (*Oreochromis mossambicus*) from Laos (Sudaryanto et al. 2007a) [ABST]. The samples were collected in both urban and rural areas, and also from an area with intensive aquaculture and an area close to a municipal waste dump. The study also investigated the concentrations of two nonaBDEs (BDE-206 and BDE-207). BDE-207 was not detectable (concentration <0.13 µg/kg/lipid) and BDE-206 was detected only in the samples of snakehead from an urban area (concentration range found was <0.040 to 0.15 µg/kg lipid with a mean concentration of 0.017 µg/kg lipid).

Rainey et al. (2007) investigated the levels of polybrominated diphenyl ethers in the edible parts of nineteen salmon (comprising samples of Chinook, Chum and Coho salmon) from the pacific coast of California, Washington and Alaska. The samples were collected between 2005 and 2007. DecaBDE was found to be present in the samples, but the levels found were below the reporting limit, and were not significantly different from the levels historically detected in laboratory blanks at the laboratory in question. Therefore the significance of the results is unclear.

A time trend study of the levels of decaBDE in lake trout (*Salvelinus namaycush*) from Lake Ontario, Canda, has been carried out by Ismail et al. (2009). Archived samples of whole lake trout were used in the study. The fish used were all collected from the same area and a total of four or five individuals from the years 1979, 1983, 1988, 1993, 1998 and 2004 were used in the study. The fish were all between four and five years of age at the time of collection. The analytical method used included routine analysis of procedural blanks and decaBDE was detectable in these blank samples. Therefore the analytical detection limit for decaBDE was set at 0.23 µg/kg wet weight (this value was the mean plus three standard deviations of the blank values). DecaBDE was detectable in most of the individual fish included in the study. The mean (± standard deviation) concentrations found in each year ranged from 2.3±0.5 µg/kg lipid (0.27±0.03 µg/kg wet weight) in 1979 to 12±5.3 µg/kg lipid (1.3±0.8 µg/kg wet weight) in 2004, with an approximate four fold increase in the concentration occurring between 1998 and 2004. It was determined that a statistically significant ($p=0.05$) increase in the concentration had occurred between 1979 and 2004, with an estimated doubling time over the entire period of around 19 years. However, it was noted that considerable variability was evident in the concentration data, particularly for 2004. Ismail et al. (2009) also investigated changes in trophic level of lake trout (based on $\delta^{15}N$-measurements) over the same time period. These data suggested that there may have been changes in trophic position of the lake trout, particularly between 1998 and 2004. However adjusting the measured concentrations of decaBDE for changes in $\delta^{15}N$ did not significantly affect the temporal trend seen for decaBDE. In contrast to decaBDE, lower brominated congeners (tri- to hexaBDEs) showed an increase in concentration from 1979 until 1993 or 1998, followed by a decrease or levelling off of the concentration between 1998 and 2004.

The levels of decaBDE in eggs of snapping turtles (*Chelydra serpentine*) have been investigated by De Solla et al. (2007). Samples of eggs (approximately five eggs per location) were collected from fifteen locations on Lake Erie, Lake Ontario, Detroit River, St. Lawrence River and two inland reference sites. The eggs were collected between 2001 and 2004. DecaBDE was not detectable in the samples from any site (the detection limit was given as 0.01 µg/kg wet weight). The quality assurance/quality control procedures used during the analysis included the routine analysis of procedural blanks and standard reference materials.

Shaw et al. (2007a) [ABST] reported that decaBDE was detectable in blubber samples from four out of eight harbour seals (*Phoca vitulina concolor*) from the northwestern Atlantic coast of America at a concentration between 1 and 8 µg/kg lipid. Few other details are given. Shaw et al. (2007b) [ABST] give similar results (decaBDE was detected in blubber from two out of four harbour seals at a concentration of 1.4 and 7.6 µg/kg) and indicate that these results are preliminary results from a more detailed investigation of the levels present in forty two individuals.

Tanabe et al. (2007) [ABST] determined the levels of polybrominated diphenyl ethers in archived samples of northern fur seals (*Callorhinus ursinus*), melon-headed whales (*Peponocephala electra*), striped dolphin (*Stenella coeruleoalba*) and finless porpoise

(*Neophocaena phocaenoides*) collected from the coast of Japan. The paper indicates that decaBDE was detectable in samples of striped dolphin but gives no information on the levels found, the number of occurrences or the occurrence in the other species studied.

Kajiwara et al. (2008b) found that decaBDE was not detectable in archived blubber samples from 55 melon-headed whales (*Peponocephala electra*) from the coast of Japan. The whales were collected during mass-strandings in January 1982 (ten samples), February 2001 (seven samples), March 2001 (seven samples), February 2002 (28 samples) and January-February 2006 (five samples). The detection limit of the analytical method used was 0.5 µg/kg lipid.

Similarly Kajiwara et al. (2006) found that decaBDE was not detectable in blubber samples of nine killer whales (*Orcinus orca*) that were found dead after being stranded in drifting sea ice on the coast of Japan in February 2005. The detection limit of the analytical method used was again 0.5 µg/kg lipid.

Wang et al. (2008) analysed a sample of blubber from harbour seals (collected from Kodiak, Alaska in 2000) using a new analytical methodology designed to optimise the simultaneous analysis of both lower brominated congeners (in this case mono- to heptaBDEs) and the higher brominated congeners (e.g. octa- to decaBDEs). DecaBDE was not detected in the sample (detection limit was 50 ng/kg). Similarly NonaBDEs (BDE-206 and BDE-207) were also not detectable (detection limits were again 50 ng/kg for both BDE-206 and BDE-207).

Birds and birds' eggs

Conclusion (i) monitoring programme

As a result of the risk assessment of decaBDE carried out under the ESR programme (ECB, 2004 and 2007) a monitoring programme (known as the conclusion (i) monitoring programme) was instigated to investigate the long-term trends in the levels of decaBDE in birds' eggs as well as sewage sludge and sediment (see Section 4.2.1 and 4.2.2). Under this programme eggs of two species are being sampled (Glaucous Gull (*Larus hyperboreus*) and Eurasian Sparrowhawk (*Accipiter nisus*)) and a total of twelve eggs from each species are being collected on an annual basis.

The Glaucous Gull samples are being collected from a colony in Bear Island (Bjørnøya) in northern Norway. The colony inhabits the “top cliff” area of the island and consists of 50-60 nests that have been present since the 1960s. The colony feeds mainly on birds' eggs (such as Common Guillemot *Uria aalge*). The eggs are collected between late May to mid-June or the beginning of July and the samples are the first egg laid where possible.

The Eurasian Sparrowhawk eggs are sampled annually from the same area within the UK (the same site cannot be guaranteed because breeding territories are dynamic and so may shift in location and size). The samples are failed eggs (either unfertilized or unhatched) and are being collected annually between 19 June and 24 July. Assuming that egg laying starts in mid-May, the average number of days between laying and collection is around 53 days, with a minimum of 35 and a maximum of 70 days.

The data available so far from the conclusion (i) programme are summarised in Table 4.12 and Table 4.13 and displayed in Figure 4.3 to Figure 4.6. The levels found in 2005 and 2006 have been reported previously in ECB (2007). The data for 2007 are taken from the project report for 2007 (Leslie et al. 2008).

Out of the twelve Glaucous Gull egg samples collected in 2007, decaBDE was detected in eight samples at a concentration between 0.20 and 0.68 μg/kg wet weight (1.7 to 7.2 μg/kg lipid). The mean concentration (based on the samples where decaBDE was detectable) was 0.37 μg/kg wet weight or 3.4 μg/kg lipid in 2007. The equivalent mean values for the 2005 and 2006 samples were 0.23 μg/kg wet weight or 2.1 μg/kg lipid for 2005 and 0.23 μg/kg wet weight or 2.3 μg/kg lipid for 2006. Thus the mean concentration found in 2007 is higher than in 2005 or 2006[11], although this does not appear to be statistically significant (t-test, $p > 0.05$). In terms of the number of samples in which decaBDE was detectable, this was ten out of twelve in 2005, four out of twelve in 2006 and eight out of twelve in 2007. The differences in the number of samples in which decaBDE was detected do not appear to be statistically significantly different between the years (Fischer's exact test, $p > 0.05$). Overall, the results for the Glaucous Gull eggs are inconclusive.

For the Eurasian Sparrowhawk egg samples, decaBDE was detected in all twelve samples in the 2007 survey at a mean concentration of 2.1 μg/kg wet weight or 45 μg/kg lipid. In 2005, decaBDE was detectable in ten out of the twelve samples at a mean concentration of 1.1 μg/kg wet weight or 12 μg/kg lipid (based on the samples where the concentration was above the detection limit) or around 1.0 μg/kg wet weight or 10 μg/kg lipid (assuming detection limit/2 for the non-detectable samples). Only one sample was available for 2006 and this had a concentration of 0.11 μg/kg wet weight or 2.4 μg/kg lipid.

Based on these data, the concentrations found in 2007 appear to be higher than in 2005. Indeed the highest concentration measured in the 2005 survey was 36 μg/kg lipid and four of the eggs sampled in 2007 have concentrations higher than this value. However if this comparison is done on a wet weight basis only one sample from 2007 has a higher concentration than found in the 2005 survey. Statistically significant differences between the mean concentration in 2005 compared with 2007 have been investigated using the one-tailed t-test. This indicates that the mean concentration in 2007 is statistically significantly higher ($p<0.05$) than in 2005 when based on lipid weight values. However, no statistically significant differences are evident if this comparison is based on the wet weight values. As with the other matrices sampled in this programme, the limited number of sampling occasions (as well as the small number of samples) means that it is too early to make clear statements about trends (either decreasing or increasing).

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[11] A similar finding is also obtained if the mean is calculated assuming the non-detected levels represent a concentration of half the detection limit.

Table 4.12 Levels of decaBDE in Glaucous Gull eggs from the conclusion (i) monitoring programme.

Egg sample	Concentration					
	2005[1]		2006[1]		2007	
	µg/kg wet weight	µg/kg lipid	µg/kg wet weight	µg/kg lipid	µg/kg wet weight	µg/kg lipid
1	0.45	4.3	<0.06	<0.54	<0.16	<1.2
2	0.16	1.2	0.22	2.4	<0.15	<1.5
3	0.36	2.6	<0.05	<0.54	0.20[2]	1.7
4	0.13	1.1	<0.05	<0.45	0.40[2]	3.1
5	<0.06	<0.6	0.29	3.3	0.29[2]	3.0
6	<0.11	<1.1	<0.05	<0.48	0.44[2]	3.2
7	0.12	1.1	<0.05	<0.55	<0.15	<1.3
8	0.11	1.0	0.36	0.31	0.24[2]	2.7
9	0.10	1.2	<0.05	<0.46	<0.15	<1.4
10	0.37	4.1	<0.05	<0.46	0.51	3.6
11	0.12	1.3	<0.06	<0.60	0.68	7.2
12	0.37	3.0	0.057	0.54	0.21[2]	2.4
Mean (± standard deviation)	**0.23 ± 0.14[3]** **0.20 ± 0.15[4]**	**2.1 ± 1.3[3]** **1.8 ± 1.3[4]**	**0.23 ± 0.13[3]** **0.09 ± 0.12[4]**	**2.3 ± 1.3[3]** **0.95 ± 1.22[4]**	**0.37 ± 0.17[3]** **0.27 ± 0.20[4]**	**3.4 ± 1.7[3]** **2.5 ± 1.9[4]**

Notes:
[1] Data taken from ECB (2007).
[2] Detectable but the concentration was below the limit of reliable quantification. Therefore there is a reduced certainty in the reported concentration.
[3] Mean of the concentrations above the detection limit.
[4] Mean of all samples, taking a value of half the detection limit for 'not detectable' (<) concentrations.

Table 4.13 Levels of decaBDE in Eurasian Sparrowhawk eggs from the conclusion (i) monitoring programme.

Egg sample	Concentration					
	2005[1]		2006[1, 2]		2007	
	µg/kg wet weight	µg/kg lipid	µg/kg wet weight	µg/kg lipid	µg/kg wet weight	µg/kg lipid
1	<0.2	<2	0.11	0.24	0.82	10.4
2	0.37	4.6			0.99	12.9
3	<0.2	<2			12	188
4	0.69	11			0.72	9.0
5	0.78	11			1.6	25
6	4.4	36			0.48	7.4
7	0.16	3.0			0.59	94
8	0.2	2.2			1.51	22
9	0.8	7.5			0.48	8.6
10	0.7	6.1			1.62	33
11	2.6	32			2.11	63
12	0.49	4.9			2.22	67
Mean (± standard deviation)	**1.1 ± 1.3[3]** **1.0 ± 1.3[4]**	**12 ± 12[3]** **10 ± 12[4]**	**0.11[2]**	**2.5[2]**	**2.1 ± 3.2[3]** **2.1 ± 3.2[4]**	**45 ± 53[3]** **45 ± 53[4]**

Notes: [1] Data taken from ECB (2007).
[2] Only one egg was analysed owing to a lack of unhatched eggs and breakage of some samples before analysis.
[3] Mean of the concentrations above the detection limit.
[4] Mean of all samples, taking a value of half the detection limit for 'not detectable' (<) concentrations.

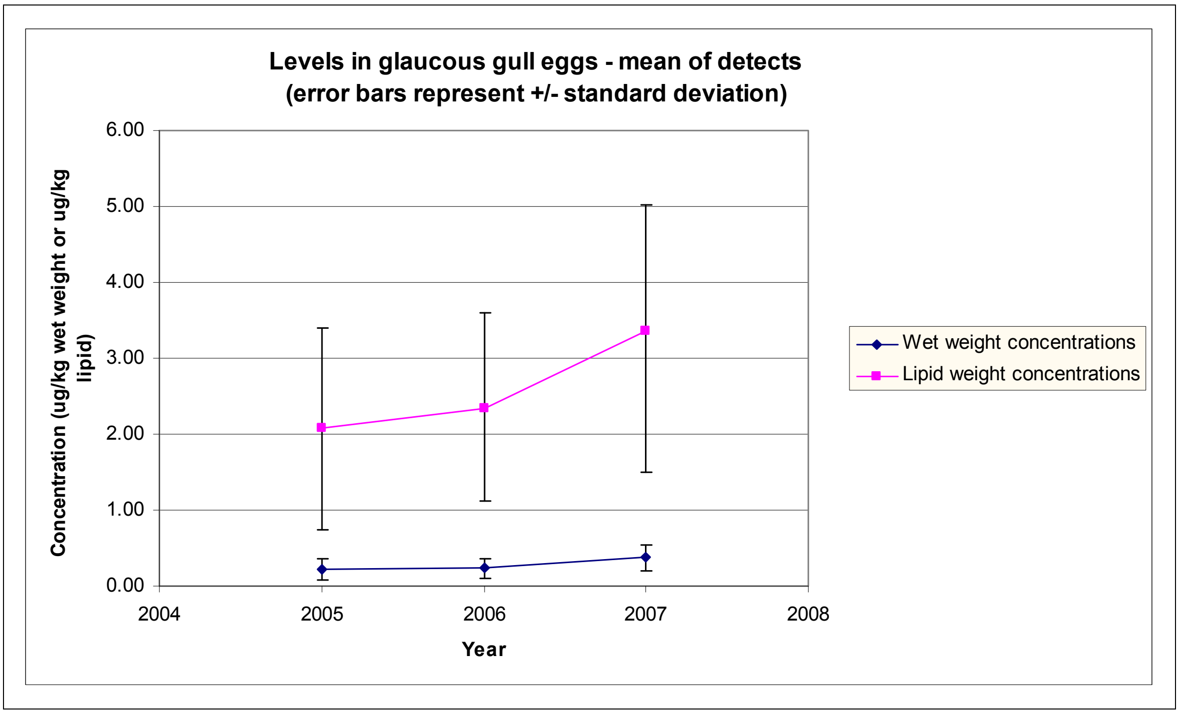

Figure 4.3 Levels in Glaucous Gull eggs (mean of concentrations > detection limit).

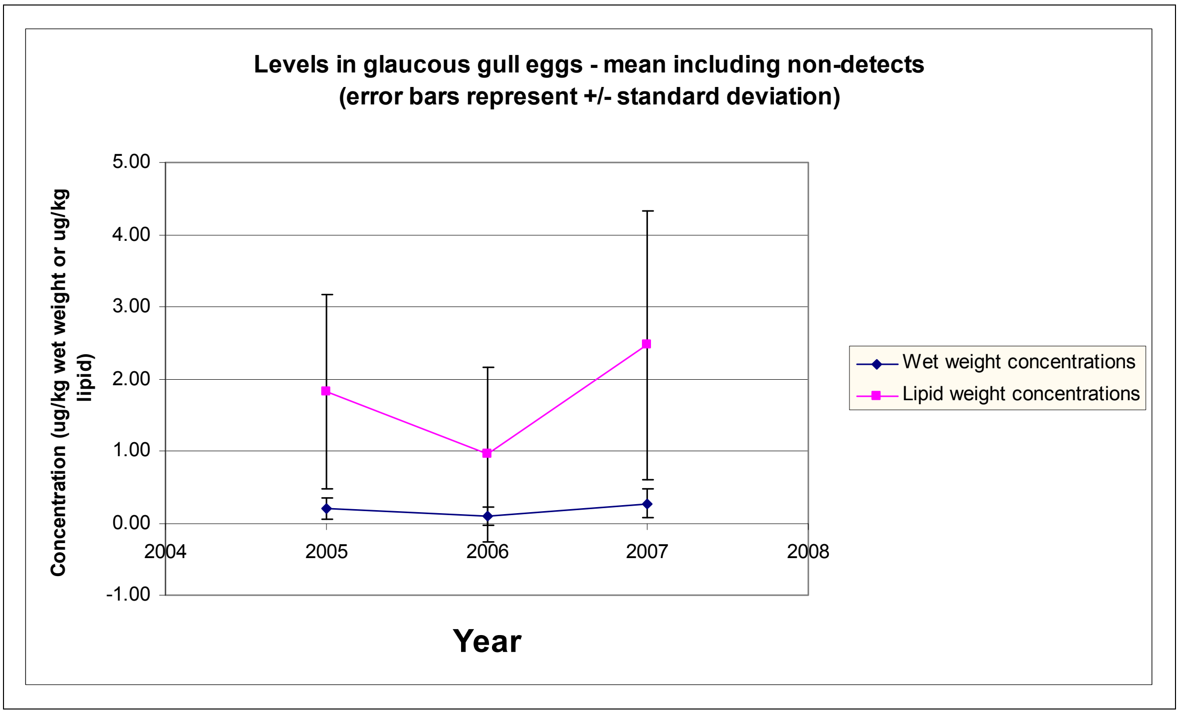

Figure 4.4 Levels in Glaucous Gull eggs including 'not detected' concentrations.

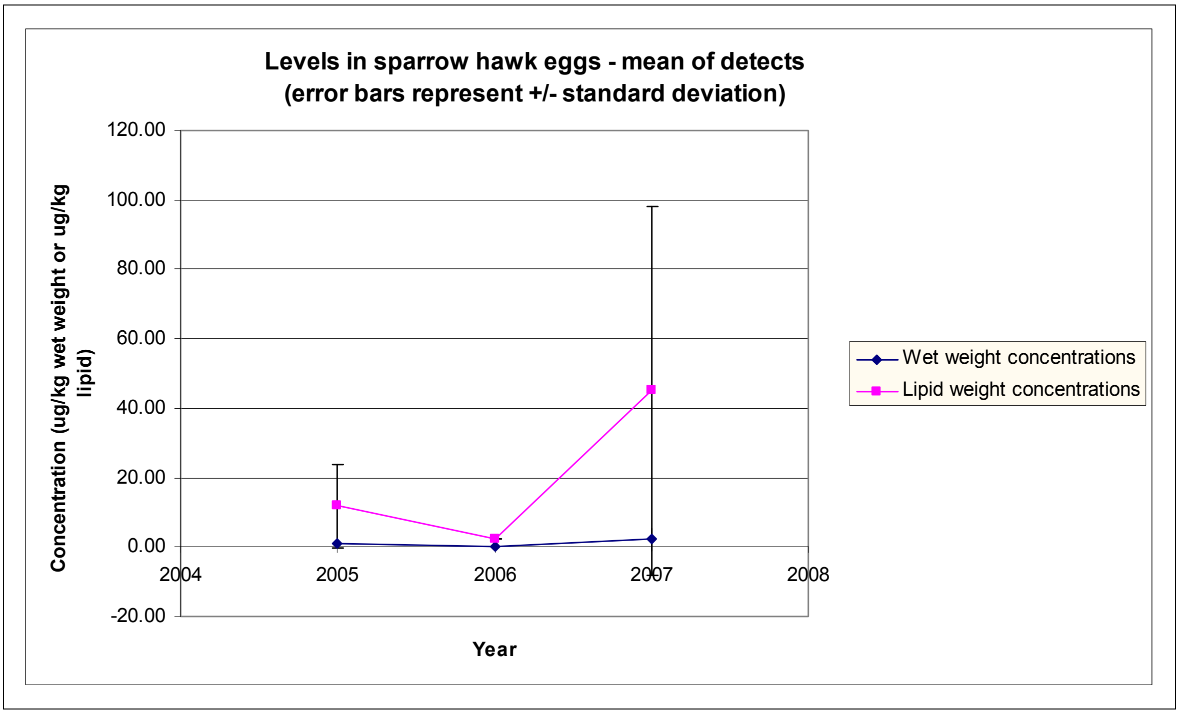

Figure 4.5 Levels in Eurasian Sparrowhawk eggs (mean of concentrations > detection limit).

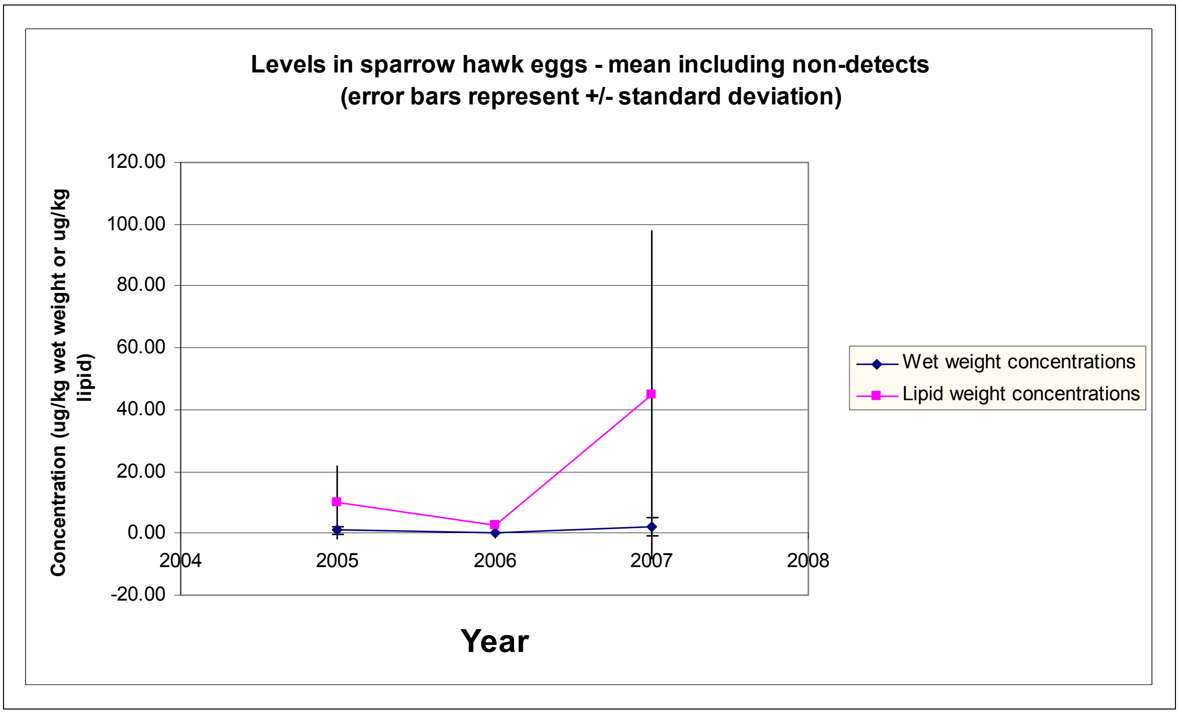

Figure 4.6 Levels in Eurasian Sparrowhawk eggs including 'not detected' concentrations.

Europe – other studies

The levels of decaBDE and nonaBDE found in birds in Europe from other studies are summarised in Table 4.14.

A study of the levels of decaBDE in eggs from White-tailed Sea Eagles (*Haliaeetus albicilla*) in Sweden has been carried out (Nordlöf et al. 2005) [ABST]. The eggs were collected between 1991 and 2004 and included 12 eggs from Lapland, 12 eggs from inland freshwater lakes (middle and south of Sweden), 12 eggs from the southern coast of the Bothnian Sea and 8 eggs from the coast of the Baltic Proper. DecaBDE was not detected in any sample. The detection limit of the analytical method used was not given.

Jiménez et al. (2005) [ABST] reported that decaBDE and nonaBDEs (BDE-206 and BDE-207 along with a further unidentified nonaBDE, presumably BDE-208) were detectable in peregrine falcon (*Falco peregrinus*) eggs collected from Spain during 2000 and 2001. No further details of the concentrations found were given.

The levels of decaBDE in livers of Glaucous Gulls from Berentsberg (West Spitsbergen) have been determined by Savinov et al. (2005) [ABST]. A total of 20 birds were sampled in 2001. DecaBDE was found at an average concentration of 0.95 µg/kg wet weight but the levels in some individuals were much higher (7.5 and 86.5 µg/kg wet weight). Few other details are given.

Verreault et al. (2007) found that decaBDE was "virtually non-detectable" and nonaBDEs were found "sporadically" in samples of plasma and egg yolk from Glaucous Gulls (*Larus hyperboreus*) collected from the Norwegian Arctic (Bear Island) in May and June 2006. The survey included blood plasma samples from nineteen males and thirty female gulls and thirty one unincubated, third-laid eggs from the same colonies. The quantification limit for decaBDE was not given in the paper (the quantification limit for nonaBDEs was given as 0.16 µg/kg wet weight in plasma and 0.36 µg/kg wet weight in egg yolk and it is likely that the quantification limit for decaBDE was similar). The quality assurance/quality control procedures used included analysis of procedural blanks and standard reference material. The blank samples were found to show negligible background contamination.

The level of decaBDE was below the quantification limit of the method in all samples. There were some indications that nonaBDEs may have been present in some samples, but only sporadically and at concentrations below the quantification limit. Several octaBDEs, including BDE-196, BDE-197, BDE-201, BDE-202, BDE-203 and BDE-205) were positively identified in the samples, with BDE-197, BDE-201 and BDE-202 being detected in 29 per cent of the egg yolk samples at a maximum concentration of 1.75 µg/kg wet weight (these three congeners were detectable in only one plasma sample). The finding of BDE-202 in these samples is of interest as this has been suggested as a possible breakdown product of decaBDE (see Section 4.2.2).

Table 4.14 Levels of decaBDE and nonaBDE in birds and birds' eggs.

Species	Sample	Congener	Concentration	Reference
Europe				
White-tailed Sea Eagle (*Haliaeetus albicilla*)	44 eggs from Sweden collected in 1991-2004.	DecaBDE	not detected	Nordlöf et al. (2007)
Peregrine Falcon (*Falco peregrinus*)	Eggs collected from Spain during 2000 and 2001.	NonaBDE and decaBDE	not detectable	Jiménez et al. (2005) [ABST]
Glaucous Gull (*Larus hyperboreus*)	20 liver samples from Barentsburg, Spitsbergen, collected in 2001.	DecaBDE	86.5 µg/kg wet weight (maximum) 0.95 µg/kg wet weight (mean)	Savinov et al. (2005) [ABST]
Glaucous Gull (*Larus hyperboreus*)	61 blood plasma samples and 31 egg yolk samples from Bear Island, Norwegian Arctic, 2006.	NonaBDE and decaBDE	<0.16 µg/kg wet weight (plasma) <0.36 µg/kg wet weight (egg yolk)	Verreault et al. (2007)
Ivory Gull (*Pagophila eburnea*)	35 eggs from four colonies in Svalbard (Skenskøya), Franz Josef Land (Nagurskoe and Klyuv Cape) and Severnaya Zemlya (Domashny).	DecaBDE	not detected to 0.075 µg/kg wet weight	Miljeteig et al. (2007)
Tawny Owl (*Strix aluco*)	A total of 137 eggs from central Norway covering the period between 1986 and 2004. DecaBDE was detectable in 63.5% of the samples.	DecaBDE	2.11 µg/kg lipid (overall mean) <0.03 µg/kg wet weight to 90.6 µg/kg lipid (overall range) 1.25 µg/kg lipid (mean of 12 eggs, 1986-1989) 1.28 µg/kg lipid (mean of 42 eggs, 2001-2004)	Bustnes et al. (2007)
Rest of the world				
Dabchick (*Tachybaptus ruficollis*)	1 egg from Koppies (Ground Dam), South Africa, 2004-2005.	DecaBDE	<0.028 µg/kg lipid (not detected)	Polder et al. (2007) [ABST]

Species	Sample	Congener	Concentration	Reference
African Darter (*Anhinga rufa*)	14 eggs from the Vaal River, South Africa, 2004-2005.	DecaBDE	0.32 µg/kg lipid (mean)	Polder et al. (2007) [ABST]
Common Cormorant (*Phalacrocorax carbo*)	4 muscle samples from Lake Biwa, Shiga, 2001.	BDE-206	0.75 µg/kg lipid (mean) <0.2-2.5 µg/kg lipid (range)	Kunisue et al. (2008a)
		BDE-207	0.6 µg/kg lipid (mean) <0.2-2.0 µg/kg lipid (range)	
		DecaBDE	1.4 µg/kg lipid (mean) <0.5-3.5 µg/kg lipid (range)	
Reed Cormorant (*Phalacrocorax africanus*)	3 eggs from the Vaal River, South Africa, 2004-2005.	DecaBDE	0.43 µg/kg lipid (mean)	Polder et al. (2007) [ABST]
Sacred Ibis (*Threskiornis aethiopicus*)	2 eggs from the Vaal River, South Africa, 2004-2005.	DecaBDE	18.0 µg/kg lipid (mean)	Polder et al. (2007) [ABST]
		BDE-207	0.93 µg/kg lipid	
		DecaBDE	<0.5 µg/kg lipid (not detected)	
Cattle Egret (*Bubulcus ibis*)	20 egg samples from the Vaal River and Barberspan Bird Sanctuary, South Africa, 2004-2005.	DecaBDE	0.44 ug/kg lipid (mean)	Polder et al. (2007) [ABST]
	5 egg samples from Xiamen, China, 2004.	BDE-206	0.86 µg/kg lipid (mean) 0.030-2.4 µg/kg lipid (range)	Lam et al. (2007)
		BDE-207	9.6 µg/kg lipid (mean) 0.21-24 µg/kg lipid (range)	
		DecaBDE	28 µg/kg lipid (mean) <0.5-75 µg/kg lipid (range)	
Chinese Pond Heron (*Ardeola bacchus*)	5 egg samples from Xiamen, China, 2004.	BDE-206	1.8 µg/kg lipid (mean) 0.25-4.7 µg/kg lipid (range)	Lam et al. (2007)
		BDE-207	12 µg/kg lipid (mean) 0.90-33 µg/kg lipid (range)	
		DecaBDE	99 µg/kg lipid (mean) <3.1-290 µg/kg lipid (range)	

Species	Sample	Congener	Concentration	Reference
Little Egret (*Egretta garzetta*)	5 eggs from Hong Kong, China, 2004.	BDE-206	0.11 µg/kg lipid (mean) <0.02-0.25 µg/kg lipid (range)	Lam et al. (2007)
		BDE-207	0.60 µg/kg lipid (mean) 0.21-1.1 µg/kg lipid (range)	
		DecaBDE	2.6 µg/kg lipid (mean) <0.5-3.8 µg/kg lipid (range)	
	5 eggs from Xiamen, China, 2004.	BDE-206	0.046 µg/kg lipid (mean) 0.029-0.085 µg/kg lipid (range)	
		BDE-207	0.17 µg/kg lipid (mean) 0.087-0.29 µg/kg lipid (range)	
		DecaBDE	0.89 µg/kg lipid (mean) 0.42-2.0 µg/kg lipid (range)	
	5 eggs from Quanzhou, China, 2004.	BDE-206	0.088 µg/kg lipid (mean) 0.046-0.13 µg/kg lipid (range)	
		BDE-207	0.49 µg/kg lipid (mean) 0.31-0.65 µg/kg lipid (range)	
		DecaBDE	1.6 µg/kg lipid (mean) 0.88-2.3 µg/kg lipid (range)	
Black-crowned Night Heron (*Nycticorax nycticorax*)	5 eggs from Hong Kong, China, 2004.	BDE-206	1.1 µg/kg lipid (mean) <0.02-5.2 µg/kg lipid (range)	Lam et al. (2007)
		BDE-207	8.2 µg/kg lipid (mean) 0.32-35 µg/kg lipid (range)	
		DecaBDE	14 µg/kg lipid (mean) 0.62-59 µg/kg lipid (range)	

Species	Sample	Congener	Concentration	Reference
	5 eggs from Xiamen, China, 2004.	BDE-206	0.11 µg/kg lipid (mean) 0.054-0.25 µg/kg lipid (range)	
		BDE-207	0.49 µg/kg lipid (mean) 0.11-0.92 µg/kg lipid (range)	
		DecaBDE	1.8 µg/kg lipid (mean) <0.5-5.2 µg/kg lipid (range)	
	5 eggs from Quanzhou, China, 2004.	BDE-206	<0.02 µg/kg lipid (not detected)	
		BDE-207	0.34 µg/kg lipid (mean) <0.02-0.64 µg/kg lipid (range)	
		DecaBDE	<0.5 µg/kg lipid (not detected)	
Northern Fulmar (*Fulmarus glacialis*)	5 muscle samples from birds caught on the open sea, 1999.	BDE-206	<0.2 µg/kg lipid (not detected)	Kunisue et al. (2008a)
		BDE-207	<0.2 µg/kg lipid (not detected)	
		DecaBDE	1.9 µg/kg lipid (mean) <0.5-4.8 (range)	
Black-footed Albatross (*Diomedea nigripes*)	5 muscle samples from birds caught on the open sea and off the coast of Ogasawara Island, 1998-1999.	BDE-206	<0.2 µg/kg lipid (not detected)	Kunisue et al. (2008a)
		BDE-207	<0.2 µg/kg lipid (not detected)	
		DecaBDE	<0.5 µg/kg lipid (not detected)	
Laysan Albatross (*Diomedea immutabilis*)	5 muscle samples from birds caught on the open sea, 1998.	BDE-206	<0.2 µg/kg lipid (not detected)	Kunisue et al. (2008a)
		BDE-207	<0.2 µg/kg lipid (not detected)	
		DecaBDE	<0.5 µg/kg lipid (not detected)	

Species	Sample	Congener	Concentration	Reference
Golden Eagle (*Aquila chrysaetos*)	2 muscle samples from Iwate, 1994 and 1995.	BDE-206	<0.2-0.45 µg/kg lipid (range)	Kunisue et al. (2008a)
		BDE-207	2.3-3.7 µg/kg lipid (range)	
		DecaBDE	<0.5-10 µg/kg lipid (range)	
Steller's Sea-eagle (*Haliaeetus pelagicus*)	1 sample from Hokkaido, 1994.	BDE-206	<0.2 µg/kg lipid (not detected)	Kunisue et al. (2008a)
Goshawk (*Accipiter gentilis*)	1 muscle sample from Tochigi, 1997.	BDE-206	370 µg/kg lipid	Kunisue et al. (2008a)
		BDE-207	290 µg/kg lipid	
		DecaBDE	120 µg/kg lipid	
Peregrine Falcon (*Falco peregrinus*)	20 eggs from California collected in 1986-2007.	DecaBDE	3,400 µg/kg lipid (maximum)	Hooper et al. (2007) [ABST]
Peregrine Falcon (*Falco peregrinus*)	14 eggs from Connecticut, Massachusetts, Maine, New Hampshire, Rhode Island and Vermont collected between 1996 and 2006	BDE-206	not detected to 19 µg/kg wet weight	Chen et al. (2008b)
		BDE-207	0.4 to 260 µg/kg wet weight	
		BDE-208	not detected to 120 µg/kg wet weight	
		DecaBDE	1.4 to 420 µg/kg wet weight (range)	
Herring Gull (*Larus argentatus*)	Pooled egg samples from seven locations in the Great Lakes. Collected in 2006 (one sample was from 2005).	BDE-206	<0.10 µg/kg wet weight	Gauthier et al. (2008)
		BDE-207	1.5 to 6.4 µg/kg wet weight	
		BDE-208	0.40 to 2.1 µg/kg wet weight	
		DecaBDE	4.5 to 20 µg/kg wet weight	
Herring Gull (*Larus argentatus*)	Pooled egg samples from six locations in the Great Lakes. Collected in 2004.	NonaBDEs	0.1 to 5.6 µg/kg wet weight	Gauthier et al. (2007a) [ABST] and Gauthier et al. (2007b)
		DecaBDE	0.14 to 0.21 µg/kg wet weight (two colonies)	
			Detected in a further two colonies and not detected in two colonies.	
Kelp Gull (*Larus dominicanus*)	1 egg from Velddrif, South Africa, 2004-2005.	DecaBDE	<0.028 µg/kg lipid (not detected)	Polder et al. (2007) [ABST]
Black-tailed Gull (*Larus	5 muscle samples from Rishiro Island,	BDE-206	<0.2 µg/kg lipid (not detected)	Kunisue et al.

Species	Sample	Congener	Concentration	Reference
crassirostris)	Hokkaido, 1999-2001.	BDE-207	0.36 µg/kg lipid (mean) <0.2-1.5 µg/kg lipid (range)	(2008a)
		DecaBDE	2.0 µg/kg lipid (mean) <0.5-6.8 µg/kg lipid (range)	
Ivory Gull (*Pagophila eburnea*)	Pooled egg samples from 1976 (3 samples each of 3 eggs), 1987 (3 samples each of 3 eggs and 2004 (2 samples each of 3 eggs). The eggs were collected from Seymour Island, Nunavut, Canada.	DecaBDE	<0.01 µg/kg wet weight (not detected)	Braune et al. (2007)
Sea birds (Black-legged Kittiwake (*Rissa tridactyla*), Razorbill (*Alca torda*), Atlantic Puffin (*Fratercula arctica*), Common Guillemot (*Uria aalge*) and Brünnich's Guillemot (*Uria lomvia*))	Liver samples from the Seven Islands Archipelago, Barents Sea, Russia, 2002.	BDE-206	<10 ng/kg wet weight (not detected)	Savinova et al. (2007) [ABST]
		BDE-207	<10 ng/kg wet weight (not detected)	
		BDE-208	<10 ng/kg wet weight (not detected)	
		DecaBDE	<10 ng/kg wet weight (not detected)	
White-fronted Plover (*Charadrius marginatus*)	1 egg from Velddrif, South Africa, 2004-2005.	DecaBDE	<0.028 µg/kg lipid (not detected)	Polder et al. (2007) [ABST]
Crowned Plover (*Vanellus coronatus*)	1 egg from Koppies (Ground Dam), South Africa, 2004-2005.	DecaBDE	2.79 µg/kg lipid	Polder et al. (2007) [ABST]
Jungle Crow (*Corvus macrorhynchos*)	5 muscle samples from Osaka, 1998.	BDE-206	20 µg/kg lipid (mean) 1.8-83 µg/kg lipid (range)	Kunisue et al. (2008a)
		BDE-207	220 µg/kg lipid (mean) 12-980 µg/kg lipid (range)	
		DecaBDE	440 µg/kg lipid (mean) 36-1,800 µg/kg lipid (range)	

A study of the levels of decaBDE in eggs of Ivory Gulls (*Pagophila eburnea*) has been carried out by Miljeteig et al. (2007). A total of 35 eggs from four colonies in Svalbard (Svenskøya), Franz Josef Land (Nagurskoe and Klyuv Cape) and Severnaya Zemlya (Domashny) were analysed for the presence of decaBDE. The analysis was carried out by an accredited laboratory and the quality control/quality assurance procedures included the routine analysis of procedural blanks and reference samples. The detection limit of the analytical method was not given. DecaBDE was not detectable in ten samples from Svalbard. It was detected in one out of six samples from Nagurskoe at a concentration of 0.031 µg/kg wet weight, in four out of seven samples from Klyuv Cape at a concentration between 0.021 and 0.075 µg/kg wet weight and in two out of twelve samples from Domashny at a concentration of between 0.03 and 0.04 µg/kg wet weight. The levels are very low, but these findings are interesting because this species spends its whole life cycle in the Arctic (though it sometimes nests close to human habitation).

DecaBDE was not detected in samples of liver from seabirds (including ten samples from Black-legged Kittiwake (*Rissa tridactyla*), five samples from Razorbill (*Alca torda*), six samples from Atlantic Puffin (*Fratercula arctica*), ten samples from Common Guillemot (*Uria aalge*) and ten samples from Brünnich’s Guillemot (*Uria lomvia*)) collected in July 2002 from the Seven Islands Archipelago, Barents Sea, Russia (Savinova et al. 2007) [ABST]. The detection limit of the analytical method used was 10 ng/kg wet weight. The paper indicates that other components of the commercial decabromodiphenyl ether products (i.e. BDE-206, BDE-207 and BDE-208) were also not detected (detection limit again 10 ng/kg wet weight).

Bustnes et al. (2007) analysed a total of 137 eggs of Tawny Owl (*Strix aluco*) for the presence of decaBDE. The eggs were collected from the area surrounding Tondheim in central Norway and covered the period between 1986 and 2004. The analysis was carried out by a laboratory accredited for the analysis of polybrominated diphenyl ethers and the quality control/quality assurance procedures included the routine analysis of procedural blanks and reference samples. DecaBDE was detected at low levels in the procedural blanks and the detection limit was set at 0.03 µg/kg wet weight to take account of this. DecaBDE was detectable in 63.5 per cent of the eggs analysed at a mean concentration of 2.11 µg/kg lipid (range 0.13 to 90.6 µg/kg lipid). A time trend analysis was also undertaken but no significant trends were identified for decaBDE. The mean level of decaBDE in eggs collected between 1986 and 1989 (a total of twelve eggs) was 1.25 µg/kg lipid which was comparable with the mean level of 1.28 µg/kg lipid in eggs collected between 2001 and 2004 (a total of forty two eggs).

Rest of the world

The levels of decaBDE and nonaBDE found in birds from other parts of the world are summarised in Table 4.14.

Hooper et al. (2007) [ABST] investigated the levels of decaBDE in eggs of Peregrine Falcons (*Falco peregrinus*) from California. The eggs were collected between 1986 and 2007 and a total of 20 eggs were analysed for the presence of decaBDE. The highest concentration measured was 3,400 µg/kg lipid (the levels in the other eggs were presented graphically only and appeared to be in the approximate range 100-1,500 µg/kg lipid). Higher levels were generally found in eggs from urban areas compared with eggs from rural areas.

Another study of the levels of decaBDE in eggs of Peregrine Falcons (*Falco peregrinus*) from the United States has been carried out by Chen et al. (2008b). In this study a total of 114 eggs were collected from nests in Connecticut (4 eggs), Massachusetts (26 eggs), Maine (1 egg), New Hampshire (58 eggs), Rhode Island (8 eggs) and Vermont (17 eggs) covering each year from 1996 to 2006 (excluding 1997 and 1998). The analytical methodology used incorporated several quality control procedures to minimise and evaluate the potential for analyte loss, sample contamination, and PBDE degradation during the analysis. DecaBDE was found to be detectable in one analytical blank sample but only at a non-quantifiable level. DecaBDE was detectable in all of the eggs at concentrations between 1.4 and 420 μg/kg wet weight. The study also found that nonaBDEs were present in at least 70 per cent of the samples, with levels in the range 'not detectable' to 19 μg/kg wet weight for BDE-206, 0.4 to 260 μg/kg wet weight for BDE-207 and 'not detected' to 120 μg/kg wet weight for BDE-208. In addition, the study found that BDE-202 was also present in 107 of the samples (concentration range 'not detectable' to 16 μg/kg wet weight). In terms of the total PBDEs present in the eggs, Chen et al. (2008b) found that the congener pattern was dominated by BDE-153, followed by BDE-199, BDE-183, BDE-209, BDE-197, BDE-207, BDE-154, BDE-100 and BDE-196, with lesser contributions from BDE-47, BDE-208, BDE-203, BDE-201, BDE-206, BDE-202, BDE-138 and BDE-119. Chen et al. (2008b) noted that this congener pattern differed markedly for that typically reported for aquatic birds (where BDE-47 is usually dominant) and commented that the pattern here suggests that terrestrial birds may experience greater exposure to the more highly brominated congeners compared with aquatic birds.

Chen et al. (2008b) carried out a comparison of the concentrations found in eggs from urban environments (for example nests located on high rise buildings or bridges) with those from rural areas (most nests were located on natural cliffs). This analysis found the concentration of decaBDE was statistically significantly higher ($P<0.005$) in eggs from urban areas than in eggs from rural areas, and that a significant correlation existed between the decaBDE concentration and the population of the area.

A time-trend analysis was also carried out on the data. Chen et al. (2008b) noted that although the majority of the nests had been sampled in each year, some of the nests were sampled only once and this may bias the results. The concentration of decaBDE was, however, found to show a significant increase with time ($r=0.384$, $p<0.005$) over the period of the study with an estimated doubling time of five years.

Gauthier et al. (2008) investigated the temporal trends in the levels of polybrominated diphenyl ethers in Herring Gull (*Larus argentatus*) eggs from the Laurentian Great Lakes. Egg samples were collected from seven colonies for each of the years 1982, 1987, 1992 and 1995 to 2006 inclusive. Three of the colonies were considered to be in remote areas away from likely sources of polybrominated diphenyl ethers, although the relevance of this is unclear given that the population winters elsewhere, and that there could be exchange of birds between colonies. The eggs were collected in April to May and each sample represented a pooled sample from ten to thirteen individual eggs from each site. The eggs were analysed for the presence of decaBDE, all three nonaBDE congeners and various other lower brominated congeners. The levels found were reported only for the 2006 samples (for one site the levels were for 2005 samples). Here the level of decaBDE ranged from 4.5 μg/kg wet weight to 20 μg/kg wet weight. The levels of BDE-206, BDE-207 and BDE-208 were <0.10 μg/kg wet weight, between 1.5 and 6.4 μg/kg wet weight and between 0.40 and 2.1 μg/kg wet weight respectively.

The time trend analysis was carried out using data for all fifteen years. The doubling time for each congener was estimated for six sites using the rate of change (i.e. slope) of a curve from a non-linear, exponential growth regression model using nontransformed concentration data. Few other details of the method or data used are given (for example the concentrations found in years other than 2006 are not given). Gauthier et al. (2008) also indicates that data points for one site were not included in the time trend analysis as they covered only the period between 1995 and 2006. The doubling time was estimated as between 2.1 to 3.0 years for the six sites analysed for decaBDE and between 2.6 and 5.3 years for the sum of nonaBDEs and the regression results were reported to be statistically significant at the p=0.05 level. The lack of raw data from the earlier years in this study makes it difficult to assess the reliability of these estimated doubling times. The usefulness of 'doubling time' as an indicator of accumulation is also questionable, since in some cases concentrations in 2004/5 are only slightly higher than levels in the mid-1990s.

Gauthier et al. (2008) also considered the changes in the congener profiles present in each year in relation to possible evidence for debromination of decaBDE to lower brominated congeners. These data are discussed in Section 4.3.3.

Gauthier et al. (2007a [ABST] and 2007b) also report levels of decaBDE in eggs of Herring Gulls (*Larus argentatus*) from the Great Lakes. These papers indicate that eggs were collected from six colonies in April-May 2004[12] with a total of ten to thirteen eggs being collected at each site (the eggs were pooled for analysis). DecaBDE was detected in the pooled samples from four of the six colonies from 2006, and was quantifiable in two of these four colonies at a concentration of 0.14 µg/kg wet weight (colony from Channel-Shelter Island) and 0.21 µg/kg wet weight (colony from Niagara River). For the other four colonies (Agawa Rock, Gull Island, Fighting Island and Toronto Harbour) decaBDE was below the limit of quantification (<0.1 µg/kg wet weight) at all sites but was detectable but not quantifiable at two of these colonies. NonaBDE, of which BDE-207 was most prominent at all locations except Channel-Shelter Island (where BDE-206 was most prominent), and several octaBDEs (of which BDE-197/199 was most prominent at most locations except for Channel-Shelter Island where BDE-195 and BDE-196 were also prominent) were also found to be present in the samples. The levels of total nonaBDEs (BDE-206, BDE-207 and BDE-208) were in the range 0.10 to 5.6 µg/kg wet weight. These samples appear to be part of the same study as Gauthier et al. (2008).

DecaBDE was not detectable (concentration <0.01 µg/kg wet weight) in pooled samples of Ivory Gull (*Pagophila eburnean*) eggs from Seymour Island, Nunavut, Canada. A total of nine eggs were collected in 1976, nine eggs in 1987 and a further six eggs in 2004 (Braune et al. 2007). The eggs were pooled (each pooled sample consisting of three eggs) prior to analysis. The quality assurance/quality control procedures used during the analysis included the routine analysis of procedural blanks and in-house reference tissues.

The levels of decaBDE in eggs of several species of birds from South Africa have been reported by Polder et al. (2007) [ABST]. The eggs were collected in the summer period from November 2004 to March 2005. The samples included fourteen African Darter (*Anhinga rufa*), three Reed Cormorant (*Phalacrocorax africanus*) and two Sacred Ibis (*Threskiornis aethiopicus*) eggs from the Vaal River, twenty Cattle Egret (*Bubulcus ibis*) eggs from the Vaal River and Barberspan Bird Sanctuary, one Crowned Plover (*Vanellus coronatus*) and one Dabchick (*Tachybaptus ruficollis*) egg from Koppies (Ground Dam) and one White-fronted Plover (*Charadrius marginatus*) and one Kelp Gull (*Larus dominicanus*) egg from Velddrif, Western Cape Province Salt pan. The

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[12] They are given as 2006 in the Gauthier et al. (2007a) paper but this appears to be an error.

eggs from the Vaal River were collected downstream of a heavily industrialised area. The Barberspan Bird Sanctuary is located approximately 300 km to the west of Johannesburg. Koppies is an agricultural area close to a side river of the River Vaal, about 150 km south of Johannesburg and Velddrif is a coastal area about 1,500 km from Johannesburg.

The mean levels of decaBDE found were 0.32 µg/kg lipid in African Darter eggs, 0.44 µg/kg lipid in Cattle Egret eggs, 0.43 µg/kg lipid in Reed Cormorant eggs, 18.0 µg/kg lipid in Sacred Ibis eggs, 2.79 µg/kg lipid in Crowned Plover eggs and not detected (<0.028 µg/kg lipid) in Dabchick, White-fronted Plover and Kelp Gull eggs. The lipid contents of the eggs ranged from 4.84 per cent to 10.8 per cent. The highest levels were found in Sacred Ibis. This species is known to forage on waste dumps and this was thought to be a possible explanation for the elevated levels. It is interesting to note that some of the species in which decaBDE was found are almost exclusively piscivorous.

A study of the levels of decaBDE in eggs of water birds from South China has been carried out by Lam et al. (2007). The egg samples included Black-crowned Night Herons (*Nycticorax nycticorax*) and Little Egret (*Egretta garzetta*) from Hong Kong, Xiamen and Quanzhou and Chinese Pond Heron (*Ardeola bacchus*) and Cattle Egret (*Bubulcus ibis*) from Xiamen. The eggs were collected during the breeding season (March to May) in 2004 and a total of five eggs per species were collected from each location. Individual eggs were analysed (it is not entirely clear from the paper whether the shells were removed or not prior to the analysis). The detection limit of the analytical method was 0.02 µg/kg lipid for nonaBDEs and 0.5 µg/kg lipid for decaBDE (these values were three times the levels found in the procedural blank samples). DecaBDE was detectable in egg samples from all three locations. The mean[13] (and range) of the concentrations found was 2.6 µg/kg lipid (<0.5 to 3.8 µg/kg lipid), 0.89 µg/kg lipid (0.42 to 2.0 µg/kg/lipid) and 1.6 µg/kg lipid (0.88 to 2.3 µg/kg lipid) in Little Egret eggs from Hong Kong, Xiamen and Quanzhou respectively, 14 µg/kg lipid (0.62 to 59 µg/kg lipid), 1.8 µg/kg lipid (<0.5 to 5.2 µg/kg lipid and <0.5 µg/kg lipid in Black-crowned Night Heron eggs from the three locations respectively, 99 µg/kg lipid (3.1 to 290 µg/kg lipid) in Chinese Pond Heron eggs from Xiamen and 28 µg/kg lipid (<0.5-75 µg/kg lipid) in Cattle Egret eggs from Xiamen. NonaBDEs (BDE-206 and BDE-207) were also detectable in the samples, with concentrations in the range <0.02 µg/kg lipid to 5.2 µg/kg lipid for BDE-206 and <0.02 to 35 µg/kg lipid for BDE-207 (the levels of nonaBDEs in each species are summarised in Table 4.14).

Kunisue et al. (2008a) determined the levels of decaBDE and nonaBDEs (BDE-206 and BDE-207) in samples of pectoral muscle in birds from in and around Japan. The samples were collected between 1994 and 2001 and the species included open sea birds (five samples each of Black-footed Albatross (*Diomedea nigripes*), Laysan Albatross (*Diomedea immutabilis*) and Northern Fulmar (*Fulmarus glacialis*)), coastal birds (five samples of Black-tailed Gull (*Larus crassirostris*) and one sample of Steller's Sea-eagle (*Haliaeetus pelagicus*)) and inland birds (four samples of Common Cormorant (*Phalacrocorax carbo*), five samples of Jungle Crow (*Corvus macrorhynchos*), two samples of Golden Eagle (*Aquila chrysaetos*) and one sample of Goshawk *(Accipiter gentilis*)). The analytical methodology used included the routine analysis of laboratory blanks and the laboratory had previously taken part in an inter-laboratory comparison exercise in which reliable results were obtained. DecaBDE and nonaBDEs were not detectable or present only at low levels in the samples of

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[13] Samples below the detection limit were assigned a value of ½ the detection limit when calculating the mean.

open sea birds (not detectable in both species of albatross (detection limit was 0.5 µg/kg lipid for decaBDE and 0.2 µg/kg lipid for the two nonaBDEs) and decaBDE was found at a mean concentration of 1.9 µg/kg lipid (range <0.5 to 4.8 µg/kg lipid) in Northern Fulmar; BDE-206 and BDE-207 were not detected in this species). Similarly only low levels were measured in the coastal birds (BDE-206 was not detectable in either Black-tailed Gull or Steller's Sea-eagle samples; the levels of decaBDE in these two species were a mean of 2.0 µg/kg lipid (range <0.5 to 6.8 µg/kg lipid) and <0.5 µg/kg lipid respectively; the levels of BDE-207 in these two species were a mean of 0.36 µg/kg lipid (range <0.2 to 1.5 µg/kg lipid) and 0.93 µg/kg lipid respectively).

The levels present in the inland birds were higher, particularly in the Jungle Crow and Goshawk. Here the levels of decaBDE found were a mean of 1.4 µg/kg lipid (range <0.5 to 3.5 µg/kg lipid) in Common Cormorant, a mean of 440 µg/kg lipid (range 36-1,800 µg/kg lipid) in Jungle Crow, <0.5 and 10 µg/kg lipid in the two Golden Eagle samples and 120 µg/kg lipid in the Goshawk sample. The levels of BDE-206 were a mean of 0.75 µg/kg lipid (range <0.2 to 2.5 µg/kg lipid) in Common Cormorant, a mean of 20 µg/kg lipid (range 1.8 to 83 µg/kg lipid) in Jungle Crow, <0.2 and 0.45 µg/kg lipid in Golden Eagle and 370 µg/kg lipid in Goshawk; the levels of BDE-207 were a mean of 0.6 µg/kg lipid (range <0.2-2.0 µg/kg lipid) in Common Cormorant, a mean of 220 µg/kg lipid (range 12 to 980 µg/kg lipid) in Jungle Crow, 2.3 and 3.7 µg/kg lipid in Golden Eagle and 290 µg/kg lipid in Goshawk.

These results should be interpreted cautiously given the small number of samples and large variability in some of the concentrations measured in individual species. However, there are two main points of note from this study:

- DecaBDE and nonaBDE appear to occur at higher concentrations in terrestrial birds, particularly those feeding on small birds and mammals (e.g. Goshawk) or crops, fruits, insects, carrion and garbage (e.g. Jungle Crow). This implies that exposure via the terrestrial food chain is important for this substance.
- The levels of nonaBDEs (sum of BDE-206 and BDE-207) in both the Jungle Crow and Goshawk appear to have been around 50 to >100 per cent of those of decaBDE.

There are a number of possible explanations for this second finding. One could be that these birds were exposed to nonaBDE as a result of the use of both commercial octabromodiphenyl ether and commercial decabromodiphenyl ether products (nonaBDEs are present in both). However, the study also investigated the levels of two octaBDEs (BDE-197 and BDE-196) and a heptaBDE (BDE-183; this is a known (major) component of the commercial octabromodiphenyl ether product) and the levels of these substances were around ten to seventy times lower in the Jungle Crow than in the Goshawk (the levels of these congeners in the Jungle Crow were of a similar order to, but lower than those of the nonaBDEs and decaBDE (the mean levels in the Jungle Crow were 130 µg/kg lipid for BDE-183, 72 and 99 µg/kg lipid for the two octaBDEs compared with 44 µg/kg lipid for decaBDE and 220 µg/kg lipid for BDE-207)[14]). For comparison, commercial octabromodiphenyl ether products generally consisted of around 42-58 per cent heptaBDEs, 26-35 per cent octaBDEs and 8-14 per cent nonaBDEs (EC 2003), and commercial decabromodiphenyl ether typically contains ≤3 per cent nonaBDEs and ≥97 per cent decaBDE[15] (EC, 2002). Thus the fact that the

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[14] The data are not given for individual birds and so it is only possible to do this comparison based on the mean concentrations.

[15] Some older products may have contained higher amounts of nonaBDEs, for example a composition of 21.8 per cent nonaBDEs and 77.4 per cent decaBDE is given in EC (2002). It should be noted that the bird samples analysed were collected between 1994 and 2001 and so

concentration of BDE-207 found in the Jungle Crow was higher than both decaBDE and BDE-183 (a marker for exposure to the commercial octaBDE product) suggest that it is unlikely that the pattern of congeners seen in the birds, particularly in the Jungle Crow, can be fully explained in terms of the combined exposure from exposure to octabromodiphenyl ether and decabromodiphenyl ether commercial products alone. Two other alternative explanations are that either nonaBDEs are much more accumulative than decaBDE in these species or that they are formed by metabolic breakdown of decaBDE in these species. It is not currently possible to distinguish between these two possibilities but the net result is effectively the same, i.e. the concentration of the nonaBDEs in the birds appears to be higher than would be expected based on the amount of nonaBDE present in the commercial decabromodiphenyl ether product.

Terrestrial mammals

The levels of decaBDE and nonaBDEs in terrestrial mammals are summarised in Table 4.15.

Europe

The concentrations of decaBDE in liver tissues of otter (*Lutra lutra*) from the United Kingdom have been determined to be up to 6,808 µg/kg lipid with a mean 172 µg/kg lipid (Pountney et al., 2007) [ABST]. A total of 117 liver samples were analysed in the study and the concentration of decaBDE present in the sample varied with location. Otters from the north and east of England were found to have significantly higher levels than those from the southwest of England and Wales. There were also strong indications (although the correlations were not statistically significant) that sex and life stage/age of the otters also influenced the concentration found, with higher liver concentrations being evident in older animals compared with younger animals and in females compared with males. The samples appear to have been collected over a number of years, ranging from 1997 to 2006.

Rest of the world

The levels of decaBDE and nonaBDEs in liver samples from brown rats (*Rattus norvegicus*) from Japan have been reported by Kurunthachalam et al. (2005) [ABST]. The rats were collected from urban (some of the rats may have been collected from the subway/underground system), rural and remote areas and also from waste dumps and landfill sites. Laboratory Wistar rats were used as a control for comparison. The levels found for decaBDE were 6 µg/kg lipid in the laboratory rats and between 3.6 and 74 µg/kg lipid in the wild rats. For the nonaBDEs, the level of BDE-207 in the laboratory rats was 0.47 µg/kg lipid and between 0.31 and 8.4 µg/kg lipid in the wild rats and the level of BDE-208 was <0.001 µg/kg in the laboratory rats and 0.11 to 3.6 µg/kg in the wild rats. The highest levels were associated with a rats from a landfill site. Kurunthachalam et al. (2005) suggested that the rats could be exposed through feeding on cables, electrical appliances and plastics, etc.

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the composition of the products in use at that time may not reflect the composition of current products.

Table 4.15 Levels of decaBDE and nonaBDE in terrestrial mammals.

Species	Sample	Congener	Concentration	Reference
Europe				
Otter (*Lutra lutra*)	117 liver samples from the United Kingdom.	DecaBDE	6,808 µg/kg lipid (maximum) 172 µg/kg lipid (mean)	Pountney et al. (2007) [ABST]
Rest of the world				
Brown rat (*Rattus norvegicus*)	Liver samples from rats from urban, rural and remote areas and a waste dump and landfill site in Japan. The highest levels were found in rats from the landfill site.	BDE-207 BDE-208 DecaBDE	3.6 to 74 µg/kg lipid 0.31 to 8.4 µg/kg lipid 0.11 to 3.6 µg/kg lipid	Kurunthachalam et al. (2005) [ABST]
Cat	23 serum samples from domestic cats in the United States.	DecaBDE	detected	Venier et al. (2007) [ABST] and Dye et al. (2007a and 2007b)
Cat and dog	Pooled serum sample from 17 domestic cats in the United States.	BDE-206 BDE-207 BDE-208 DecaBDE	11.7 µg/kg lipid 52.1 µg/kg lipid 20.6 µg/kg lipid 360 µg/kg lipid	EWG (2008)
	Pooled serum sampled from 10 domestic dogs in the United States.	BDE-206 BDE-207 BDE-208 DecaBDE	3.4 µg/kg lipid 3.3 µg/kg lipid 2.3 µg/kg lipid 98.8 µg/kg lipid	

Species	Sample	Congener	Concentration	Reference
Giant panda (*Ailuropoda melanoleuca*)	Samples (5 liver, 2 kidney, 1 brain and 1 gonad) from 8 individuals from Beijing Zoo, 2006.	BDE-206	9.22-141.2 μg/kg lipid (liver; mean 42 μg/kg lipid) 12-14 μg/kg lipid (kidney) 5.4 μg/kg lipid (brain) 434 μg/kg lipid (gonad)	Guo-Cheng et al. (2008)
		BDE-207	0.8-31 μg/kg lipid (liver; mean 10 μg/kg lipid) 3.4-4.2 μg/kg lipid (kidney) 2.1 μg/kg lipid (brain) 69 μg/kg lipid (gonad)	
		BDE-208	3.8-122 μg/kg lipid (liver; mean 33 μg/kg lipid) 7.7-7.8 μg/kg lipid (kidney) 1.0 μg/kg lipid (brain) 148 μg/kg lipid (gonad)	
		DecaBDE	6.8-390 μg/kg lipid (liver; mean 113 μg/kg lipid) 36-39 μg/kg lipid (kidney) 23 μg/kg lipid (brain) 1,479 μg/kg lipid (gonad)	

Species	Sample	Congener	Concentration	Reference
Raccoon dog (*Nyctereutes procyonoides*)	21 liver samples from Ehime, Japan, collected in 2001-2003.	BDE-206	0.47 µg/kg lipid (mean) <0.02-1.8 µg/kg (range)	Isobe et al. (2007) [ABST] and Kunisue et al. (2008b)
		BDE-207	0.90 µg/kg lipid (mean) <0.02-2.8 µg/kg (range)	
		DecaBDE	16 µg/kg lipid (mean) 1.1-67 µg/kg (range)	
	8 liver samples from Osaka, Japan, collected in 2004-2006.	BDE-206	0.87 µg/kg lipid (mean) <0.16-1.9 µg/kg (range)	
		BDE-207	1.5 µg/kg lipid (mean) <0.48-3.0 µg/kg (range)	
		DecaBDE	28 µg/kg lipid (mean) 5.8-76 µg/kg (range)	
	8 adipose samples from Osaka, Japan, collected in 2004-2006.	BDE-206	0.59 µg/kg lipid (mean) 0.069-1.4 µg/kg lipid (range)	
		BDE-207	0.95 µg/kg lipid (mean) 0.20-1.7 µg/kg lipid (range)	
		DecaBDE	5.9 µg/kg lipid (mean) <0.1-11 µg/kg lipid (range)	
	10 liver samples from Kanagawa, Japan, collected in 2001-2003.	BDE-206	2.4 µg/kg lipid (mean) <0.059-9.4 µg/kg (range)	
		BDE-207	9.6 µg/kg lipid (mean) <0.02-32 µg/kg (range)	
		DecaBDE	39 µg/kg lipid (mean) <0.1-160 µg/kg (range)	

Species	Sample	Congener	Concentration	Reference
Red panda (*Ailurus fulgens*)	Samples (4 liver, 3 kidney, 4 adipose and 6 muscle) from 8 individuals from Chongqing Zoo, 2006.	BDE-206	10-79 µg/kg lipid (liver; mean 29 µg/kg lipid) 14-42 µg/kg lipid (kidney; mean 23 µg/kg lipid) 8.9-366 µg/kg lipid (muscle; mean 79 µg/kg lipid) 2.6-26 µg/kg lipid (adipose; mean 9.3 µg/kg lipid)	Guo-Cheng et al. (2008)
		BDE-207	4.2-39 µg/kg lipid (liver; mean 14 µg/kg lipid) 7.1-14µg/kg lipid (kidney; mean 9.5 µg/kg lipid) not detected-297 µg/kg lipid (muscle; mean 85 µg/kg lipid) 1.1-7.7 µg/kg lipid (adipose; mean 3.4 µg/kg lipid)	
		BDE-208	15-183 µg/kg lipid (liver; mean 59 µg/kg lipid) 15-20 µg/kg lipid (kidney; mean 16 µg/kg lipid) not detected-502 µg/kg lipid (muscle; mean 112 µg/kg lipid) 2.7-24 µg/kg lipid (adipose; mean 9.3 µg/kg lipid)	
		DecaBDE	13-154 µg/kg lipid (liver; mean 67 µg/kg lipid) 17-143 µg/kg lipid (kidney; mean 70 µg/kg lipid) not detected-730 µg/kg lipid (muscle; mean 17 µg/kg lipid) 5.1-200 µg/kg lipid (adipose; mean 59 µg/kg lipid)	

Another study on the levels of polybrominated diphenyl ethers in terrestrial mammals from Japan has found decaBDE and nonaBDEs (BDE-206 and BDE-207) to be present in the liver of raccoon dogs (*Nyctereutes procyonoides*) (Isobe et al. (2007) [ABST] and Kunisue et al. (2008b)). The samples were collected during 2001-2003 from Ehime (21 samples) and Kanagawa (ten samples) and during 2004-2006 at Osaka (eight samples). The quality assurance/quality control procedures included the routine analysis of procedural blanks and the laboratory had participated previously in an inter-laboratory comparison exercise. Very low levels of decaBDE were evident in some of the blank samples and the concentrations reported in the raccoon samples were corrected for this. DecaBDE was found to be present in 100 per cent of the samples from Ehime and Osaka, and 90 per cent of the samples from Kanagawa, and the mean concentration found was 16 µg/kg lipid in Ehime, 28 µg/kg lipid in Osaka and 39 µg/kg from Kanagawa. BDE-206 and BDE-207 were also found to be present in the majority of the samples (found in between 76 per cent and 100 per cent of the samples) with mean concentrations in the range 0.47 µg/kg lipid to 2.4 µg/kg lipid for BDE-206 and 0.9 µg/kg lipid to 9.6 µg/kg lipid for BDE-207. The highest concentrations were again found in the samples from Kanagawa.

As well as liver samples, samples of adipose tissue were also collected from the raccoon dogs collected in Osaka (Kunisue et al. 2008b). The mean decaBDE concentration found in these samples was 5.9 µg/kg lipid. BDE-206 and BDE-207 were also present at a mean concentration of 0.59 µg/kg lipid and 0.95 µg/kg lipid respectively. The ratio of the lipid normalised concentration in liver to the lipid normalised concentration in adipose was determined to be 4.9 for decaBDE, 2.7 for BDE-206 and 1.8 for BDE-207 suggesting preferential accumulation of these substances in the liver (for comparison the liver to adipose ratio for lower brominated congeners (tri- to heptaBDEs) were all <1 and the ratios for two octaBDEs (BDE-196 and BDE-197) were 1.5 and 1.3 respectively).

An investigation of the levels of polybrominated diphenyl ethers in serum of pet cats and cat food has been undertaken (Venier et al. (2007) [ABST] and Dye et al. (2007a) [ABST]). The serum samples (23 samples) were obtained from veterinary teaching hospitals in North Carolina, Massachusetts and Georgia, United States. The cats were grouped into three categories: hyperthyroid cats, sick (non-hyperthyroid) cats and young cats. The food samples (12 samples of dry cat food and 24 samples of canned cat food) were obtained from stores in Indiana. The mean concentration of total polybrominated diphenyl ethers found was 9.8 µg/kg wet weight in the cat serum samples, 1.8 µg/kg for the dry cat food samples and 0.66 µg/kg in the canned food samples. The levels of decaBDE alone were not given but the relative proportion of decaBDE was reported to be highest in dry food samples and the serum of cats that ate predominately dry cat food. The same study is also reported by Dye et al. (2007b) and this gives further details of the levels of decaBDE found. The quality assurance and quality control procedures included the routine analysis of procedural blanks and the levels of polybrominated diphenyl ethers in these samples were low and so it was not necessary to correct from this in the samples. The mean concentrations of total polybrominated diphenyl ethers in cat serum samples were 4.3 µg/l in young cats, 10.5 µg/l in sick (non-hyperthyroid) cats and 12.7 µg/l in hyperthyroid cats. The contribution of decaBDE to these levels was given graphically only and appeared to be up to around 1 µg/l. The levels of decaBDE found in the canned food were in the range 0.003 to 0.04 µg/kg and the level of decaBDE in the dry food was between 0.42 and 2.28 µg/kg. A nonaBDE (BDE-207) was also found in the cat food samples at a concentration between not detectable to 0.06 µg/kg in canned food and between 0.01 and 0.07 µg/kg in dry food. Although the amount of BDE-207 present in dry food accounted for only 1-3% of the total polybrominated diphenyl ethers in the food, the serum levels of BDE-207 accounted for around 17% of the total polybrominated diphenyl ethers in dry-food-eating cats (again the actual serum levels were displayed graphically only). Similarly the ratio of BDE-207/decaBDE in dry food was ~0.03 but the ratio found in serum was fairly constant across all cats (0.51, 0.54 and 0.63 in young, sick (non-hyperthyroid) and hyperthyroid cats respectively). These findings suggest that BDE-207 is either much more accumulative than decaBDE or that decaBDE is metabolised to BDE-207 in cats.

EWG (2008) carried out a study of the levels of decaBDE and nonaBDE (BDE-206, BDE-207 and BDE-208) in blood serum of pet dogs and cats from the United States. Blood samples were collected from ten dogs and 17 cats and the individual samples were pooled into a single dog and cat sample for analysis. The quality control/quality assurance procedures used during the analysis included the routine analysis of procedural blanks and a spiked laboratory control sample. The levels found in pets were higher than in the human population, for example the serum levels of decaBDE in dogs and cats were 98.8 and 360 µg/kg lipid compared with 5.9 µg/kg lipid in humans. The levels of the nonaBDEs found were 3.4 µg/kg lipid for BDE-206, 3.3 µg/kg lipid for BDE-207 and 2.3 µg/kg lipid for BDE-208 in dogs and 11.7 µg/kg lipid for BDE-206, 52.1 µg/kg lipid for BDE-207 and 20.6 µg/kg lipid for BDE-208 in cats. For comparison the levels in human blood serum were reported to be 0.25 µg/kg lipid for BDE-206, 0.51 µg/kg lipid for BDE-207 and 0.33 µg/kg lipid for BDE-208. EWG (2008) suggested that one source of exposure of cats and dogs could be through dust ingested from feeding on floors or through grooming. (This also has some relevance for human babies.)

Guo-Cheng et al. (2008) reports the levels of polybrominated diphenyl ethers in captive giant panda (*Ailuropoda melanoleuca*) and red panda (*Ailurus fulgens*) from China. Nine tissue samples (including liver (five samples) kidney (two samples), brain (one sample) and gonad (one sample)) were collected from eight individual giant pandas from Beijing Zoo in December 2006 and a further seventeen tissue samples (including liver (four samples), adipose (four samples), muscle (six samples) and kidney (three samples)) were collected from eight individual red pandas from Chongqing Zoo in November 2006. The detection limit for decaBDE was ≤1.45 µg/kg lipid and precautions were taken during the sample collection and analysis to prevent sample contamination, cross-contamination and degradation. For the giant panda samples, decaBDE was detected at a concentration between 6.8 and 390 µg/kg lipid (mean 113 µg/kg lipid) in liver samples, 36 to 39 µg/kg lipid in kidney samples, 23 µg/kg lipid in the brain sample, and 1,479 µg/kg lipid in the gonad sample. The levels of decaBDE found in the red panda samples were 13 to 154 µg/kg lipid in liver (mean 67 µg/kg lipid), 17 to 143 µg/kg lipid in kidney (mean 70 µg/kg lipid), 5.1 to 200 µg/kg lipid in adipose (mean 59 µg/kg lipid) and not detected to 730 µg/kg lipid (mean 17 µg/kg lipid) in muscle.

The survey also determined the levels of nonaBDEs (BDE-206, BDE-207 and BDE-208). The levels found in individual tissues are summarised in Table 4.14 and the concentrations were in the general range 5 to 434 µg/kg lipid, 0.8 to 69 µg/kg lipid and 1 to 148 µg/kg lipid for BDE-206, BDE-207 and BDE-208 respectively in giant panda tissues and 2.6 to 366 µg/kg lipid, not detected to 297 µg/kg lipid and not detected to 502 µg/kg lipid respectively in red panda tissues.

Plants

The levels of decaBDE (and nonaBDE) in plants are summarised in Table 4.16.

Europe

DecaBDE has been found in moss (*Hylocomoum splendens*) from twelve locations in Norway at a concentration of 105-1,593 ng/kg dry weight (Mariussen et al. (2005) [ABST]). DecaBDE was also found in the procedural blanks but the amount found in most, but not all, of the moss samples was at least two times higher than found in the procedural blank samples. The occurrence in moss was thought to result from atmospheric transport of decaBDE. The study also investigated the levels of a nonaBDE (BDE-206) but this was only "detectable in one or two samples".

Mariussen et al. (2008) reports levels of decaBDE of between 452 and 1,198 ng/kg dry weight in samples of moss (*Hylocomoum splendens*) from five locations in Norway (Nannestad, Hurdal, Minnesund, Kise and Vingrom). It is possible that some of these samples are the same are reported above by Mariussen et al. (2005).

DecaBDE has also been determined to be present in tree bark from Italy and Germany (Qiu and Hites 2008; see below).

Rest of the world

Jin et al. (2007) [ABST] reported that decaBDE and nonaBDEs (BDE-206, BDE-207 and BDE-208) were detected in samples of plants from Bohai Bay in China. The samples included four grass samples from a brominated flame retardant production plant, and two samples of haricot beans and one sample of saline seepweed from a local market. DecaBDE was detected in all samples at a concentration of 291 to 3,086 µg/kg dry weight in grass, 90 µg/kg dry weight in haricot bean and 52 µg/kg dry weight in saline seepweed. NonaBDEs (BDE-206, BDE-207 and BDE-208) were also detected. The concentrations of these three congeners found were respectively 14 to 407 µg/kg dry weight, 8.9 to 47 µg/kg dry weight and 8.9 to 47 µg/kg dry weight in grass, 8.1 µg/kg dry weight, 4.1 µg/kg dry weight and 2.5 µg/kg dry weight in haricot bean and 12 µg/kg dry weight, 4.5 µg/kg dry weight and 2.2 µg/kg dry weight in saline seepweed. The samples do not appear to have been washed prior to analysis and so the levels reported may represent substance deposited on the surface of the plants as well as substance absorbed by the plants. Few other details are currently available.

DecaBDE has been found to be present in tree bark (pine trees) from the northeastern United States (Qiu and Hites 2008). Samples of tree bark were collected from 26 locations (three samples at each location) and decaBDE was found to be present in all samples at mean concentrations between 0.12 and 5.8 µg/kg. NonaBDEs were also found in the samples with mean concentrations in the range not detected to 0.12 µg/kg for BDE-206, not detected to 0.06 µg/kg for BDE-207 and not detected to 0.04 µg/kg for BDE-208. Samples of tree bark from other countries were also analysed and the mean decaBDE concentrations in these samples were 0.007 µg/kg in southern Norwest Territories, Canada, 1.6 µg/kg in Halle, Germany, 0.12 µg/kg in Trieste, Italy, 6.5 µg/kg in Hanam, South Korea and between 14 and 43 µg/kg in three locations in China (the levels of nonaBDEs in these samples were not given).

The levels of polybrominated diphenyl ethers in pine needles from a rural area of Ansung City, South Korea, have been determined by Yeo et al. (2006) [ABST]. The levels of total polybrominated diphenyl ethers were between 0.2 and 3,104 ng/kg dry weight. The most abundant congener present in both needles and soil was decaBDE. Soil samples from the same area were also analysed (see Section 4.2.3).

Table 4.16 Levels of decaBDE and nonaBDE in plants.

Sample	Comments	Congener	Concentration	Reference
Europe				
Moss (*Hylocomium splendens*)	12 locations in Norway collected in summer 2004.	DecaBDE	0.11 to 1.6 µg/kg dry weight	Mariussen et al. (2005) [ABST]
Moss (*Hylocomium splendens*)	5 locations in Norway collected in summer 2004.	DecaBDE	0.45 to 1.2 µg/kg dry weight	Mariussen et al. (2008)
Tree bark (pine trees)	Samples from Halle, Germany, 2005	DecaBDE	1.6 µg/kg (mean)	Qiu and Hites (2008)
	Samples from Trieste, Italy, 2005	DecaBDE	0.12 µg/kg (mean)	
Rest of the world				
Grass	Four samples from close to a brominated flame retardant production plant, Bohai Bay, China, 2006	BDE-206	14-407 µg/kg dry weight	Jin et al. (2007) [ABST]
		BDE-207	8.9-47 µg/kg dry weight	
		BDE-208	8.9-47 µg/kg dry weight	
		DecaBDE	291-3,086 µg/kg dry weight	
Haricot bean	Two samples from a local market in Bohai Bay, China, 2006	BDE-206	8.1 µg/kg dry weight	Jin et al. (2007) [ABST]
		BDE-207	4.1 µg/kg dry weight	
		BDE-208	2.5 µg/kg dry weight	
		DecaBDE	90 µg/kg dry weight	
Pine needles	Samples from a rural area of Ansung City, South Korea. The levels are given as total polybrominated diphenyl ethers but decaBDE was reported to be the most abundant congener present.	Total BDEs	0.0002-3.1 µg/kg dry weight	Yeo et al. (2006) [ABST]
Saline seepweed	One sample from a local market in Bohai Bay, China, 2006	BDE-206	12 µg/kg dry weight	Jin et al. (2007) [ABST]
		BDE-207	4.5 µg/kg dry weight	
		BDE-208	2.2 µg/kg dry weight	
		DecaBDE	52 µg/kg dry weight	

Sample	Comments	Congener	Concentration	Reference
Tree bark (pine trees)	26 locations in the northeastern United States, autumn, 2006.	BDE-206	not detected to 0.12 µg/kg (range of mean values)	Qiu and Hites (2008)
		BDE-207	not detected to 0.06 µg/kg (range of mean values	
		BDE-208	not detected to 0.04 µg/kg (range of mean values	
		DecaBDE	0.12 to 5.8 µg/kg (range of mean values)	
	Samples from southern Northwest Territories, Canada, 2000	DecaBDE	0.007 µg/kg (mean)	
	Samples from Hanam, South Korea, 2005	DecaBDE	6.5 µg/kg (mean)	
	Samples from Shenzheng, China, 2007	DecaBDE	43 µg/kg (mean)	
	Samples form Hangzhou, China, 2007	DecaBDE	19 µg/kg (mean)	
	Samples from Tianjin, China, 2007	DecaBDE	14 µg/kg (mean)	

Foodstuffs

The levels of decaBDE and nonaBDEs found in foodstuffs are summarised in Table 4.17. As these data are most relevant for exposure to humans they have not been reviewed in detail in this report. Other sources of information on the levels of polybrominated diphenyl ethers in food include a recent review of the levels in food from the United States by Schecter et al. (2008).

Summary of new data

Many new studies are available on the levels of decaBDE (and nonaBDE) in biota. The analytical determination of decaBDE in biota samples in particular is problematic (e.g. see ECB (2005) for a discussion of the issues) and the occurrence of decaBDE in laboratory blank samples can lead to “false positive” results. However, most of the new studies appear to have been carried out using appropriate quality control procedures to minimise these problems (although full details of the procedures used are often not given).

As reported in the previous ESR reports, decaBDE is detectable in a wide range of species including fish and shellfish, aquatic mammals, birds and birds’ eggs, terrestrial mammals and plants. In general terms, the concentration of decaBDE in fish is very low (<<1 µg/kg) but higher levels up to a few hundred µg/kg lipid have been measured in fish close to point sources (for example in the River Vero, Spain). Similarly decaBDE is frequently not detectable or present at concentrations up to a few µg/kg lipid in marine mammals.

Taking into account the new information reported here, and the data reported previously in EC (2002) and ECB (2004 and 2007), decaBDE appears to be detected more frequently in birds, particularly terrestrial birds and birds eggs, and terrestrial mammals. For example Chen et al. (2008b) suggested that terrestrial birds may be exposed to more of the higher brominated congeners than aquatic birds and that proximity to urban sources may also be important. The results of the conclusion (i) monitoring programme do not permit any conclusions about trends in decaBDE concentrations in Glaucous Gull eggs and Eurasian Sparrowhawk eggs over the time period of the study (2005 to 2007), although the levels in Eurasian Sparrowhawks in particular may be somewhat higher in 2007 than in 2005. An increasing time trend has been suggested by Gauthier et al. (2008) based on Herring Gull eggs from the Great Lakes (although the reliability of these findings are difficult to assess at present), and by Chen et al. (2008b) for Peregrine Falcon eggs from several locations in the United States (although again there are some uncertainties in the data). Whether concentrations continue to rise will depend on several factors, and presumably a plateau could be reached depending on the kinetics and the sources of accumulation.

Some of the available monitoring data for birds and terrestrial mammals are suggestive that either debromination of decaBDE to nonaBDEs may be occurring or that some nonaBDEs are much more accumulative than decaBDE itself. This is considered further in Section 4.3.3.

Table 4.17 Levels of decaBDE and nonaBDE in food.

Sample	Comments	Congener	Concentration	Reference
Palm oil	Sample of palm oil and distillate from deodorizing of palm oil, 2004.	DecaBDE	390 ng/kg fresh weight	Haug et al. (2005) [ABST]
Seafood	212 samples (oysters, mussels, ark shell, clam, razor clam and other molluscs) from fishery markets in south China, 2005.	DecaBDE	Detected in “some samples”	Guo et al. (2007a) [ABST]
Seafood	Seafood products (six species of shrimp, two species of crab and 13 species of shellfish) collected from fishery markets at 11 coastal sites in south China between June and October 2005.	DecaBDE	<100 to 962 ng/kg wet weight (detected in 44.4% of the samples)	Guo et al. (2007b)
Seafood	Study of dietary intake of decaBDE from seafood (fish, bivalves, shrimp, crab and cephalopods) from China.	Total PBDEs (decaBDE was one of the predominant congeners found)	46.3 µg/kg lipid (maximum)	Miyake et al. (2008)
Fish and shellfish	Samples from various locations in the North Sea and the Netherlands, 2003. DecaBDE found only in two samples of mussels.	DecaBDE	<0.1-0.8 µg/kg wet weight	van Leeuwen and de Boer (2008)
Fish	59 samples from 2003-2005. Origin unknown. DecaBDE found to dominate in one sample only.	DecaBDE	4.8 mg/kg lipid	Herrmann et al. (2005) [ABST]
Fish	45 samples from markets in three areas of Japan, 2004-2005.	DecaBDE	Detected in some samples (data shown graphically in the paper)	Ashizuka et al. (2008)
Trout	From Lake Mjøsa, Norway, 2004	DecaBDE	63 ng/kg fresh weight	Haug et al. (2005) [ABST]
Chicken	Sample of chicken meat from the “Belgium Crisis”, 2004.	DecaBDE	64 ng//kg fresh weight	Haug et al. (2005) [ABST]

Sample	Comments	Congener	Concentration	Reference
Food (commercial)	Large range of commercial foodstuffs from markets in Spain.	Total PBDEs	<0.01 to 2,282 ng/kg fresh weight (range) – decaBDE made a large contribution to the levels except in fish and shellfish	Gómara et al. (2006)
Diet sample	Composite diet samples (of all meals, snacks and drinks consumed over 3 consecutive days). 12 Samples collected for 2003 and 2004 and 9 samples collected for 2005.	NonaBDEs (sum of BDE-206 and BDE-207)	2.5 ng/kg (median) – 2003 2 ng/kg (median) – 2004 3 ng/kg (median) - 2005	Nomura et al. (2005) [ABST]
		DecaBDE	18.5 ng/kg (median) – 2003 10 ng/kg (median) – 2004 12 ng/kg (median) - 2005	

4.3 Environmental fate and distribution

4.3.1 Degradation

Atmospheric degradation

Stapleton and Dodder (2008) investigated the photodegradation of decaBDE in household dust by natural sunlight. Details of this study were included in ECB (2007) but the Stapleton and Dodder (2008) paper considers further the possible degradation products. The study was carried out using pre-cleaned house dust (to remove all polybrominated diphenyl ethers present) that was spiked with decaBDE at a concentration of around 2.2 mg/kg. The spiking was carried out by adding a solution of decaBDE in toluene to the dust and allowing the solvent to evaporate. In addition a standard reference material sample of indoor dust was also used (this contained several polybrominated diphenyl ether congeners including decaBDE at a concentration of 2.7 mg/kg; no further decaBDE was spiked on to this sample). The dust samples (0.5 g aliquots in UV cuvettes) were exposed to sunlight outdoors (39º08'13'' latitude and 77º13'00'' longitude) between 9 am and 4 pm Monday to Friday on days on which no precipitation was forecast for a total of up to 200 hours exposure. Control samples were wrapped with foil and maintained at room temperature in the laboratory. The average incident solar radiation during the experiment was 545 W/m^2 (range 61 to 929 W/m^2).

At the start of the experiment, as well as decaBDE, the spiked dust sample contained detectable amounts of nonaBDEs (BDE-206, BDE-207 and BDE-208; accounting for around 3.8 per cent of the total polybrominated diphenyl ether concentration) and a trace of an unidentified heptaBDE. The reference dust contained a range of polybrominated diphenyl ether congeners from tri- to decaBDE. In both samples the concentration of decaBDE was found to decrease with time and a half-life of around 301 hours in the spiked dust and 408 hours in the standard reference material was determined for decaBDE. Stapleton and Dodder (2008) considered that the half-life of decaBDE in dust samples in actual houses will be considerably longer than these values owing to the facts that house dust would not be expected to receive full sunlight exposure for the majority of the day and that windows filter out a substantial fraction of light in the UV-A region. They estimated a more realistic half-life of around 200 days based on two hours exposure to sunlight per day.

In the experiments with spiked dust, increasing concentrations of several hepta-, octa- and nonaBDEs were evident as the concentration of decaBDE decreased with exposure. The degradation products formed included all three nonaBDEs (BDE-206, BDE-207 and BDE-208), at least six octaBDEs (BDE-196, BDE-197, BDE-200/203, BDE-201, BDE-202 and one unknown congener) and three heptaBDEs (BDE-183 and two unknown congeners). Stapleton and Dodder (2008) indicated that the BDE-202 has not been previously identified to be present in a commercial polybrominated diphenyl ether product. As noted in Section 4.2, this congener has been recently determined to be present in the environment. However, no information on the source, synthesis or confirmation of identity for BDE-202 was given in the paper. In the experiment with the standard reference material there was some evidence for increasing concentrations of BDE-208, BDE-201 and BDE-202 during the test but the results here are more difficult to interpret owing to the larger number of congeners initially present in the starting material.

Stapleton and Dodder (2008) also investigated the ratio of the BDE-197 to BDE-201 in the samples as a further indication that debromination was occurring. These two octaBDEs are present in commercial octabromodiphenyl ether products and the relative proportion has been found to be relatively constant in these products (ranging from 28 to 35 in two different octabromodiphenyl ether products). The ratio of these two congeners in the spiked dust samples were found to be 0.9 after 20 hours exposure to light and 0.6 by the end of the exposure period (the average ratio over the entire exposure period was 0.8). In the standard reference material used in the study the ratio of these two congeners decreased from a value of 4.8 to a value of 1.2 over the 200 hour exposure period, with the trend in the ratio appearing to be towards that seen in the spiked dust samples with increasing exposure. This trend was thought to result from either a faster formation of BDE-201 relative to BDE-197 or the slower degradation of BDE-197 relative to BDE-201.

Stapleton and Dodder (2008) also carried out a mass balance analysis during the study. At the start of the study the total amount of polybrominated diphenyl ethers present in the standard reference material was 2,300 pmol (primarily decaBDE) and at the end of the exposure period approximately 850 pmol (or 38 per cent) of the decaBDE mass had been lost or degraded. Of this 850 pmol, 300 pmol was accounted for by the formation of lower brominated congeners (around 240 pmol (~28 per cent of the mass loss) as nonaBDEs, around 56 pmol (~6.5 per cent of the mass loss) as octaBDEs and around 4 pmol (~0.5 per cent of the mass loss) as heptaBDEs) but the fate of the remaining 550 pmol (or 65 per cent of the mass that was lost) is currently unknown (possibilities include loss by volatilisation of decaBDE or degradation products, adsorption of decaBDE or degradation products to the walls of the test vessels, non-extraction of decaBDE or degradation products from the dust samples or formation of unknown degradation products). The mass balance also showed that there was an overall reduction in the total amount of polybrominated diphenyl ethers present in the dust samples. For example in the spiked dust the total concentration of all polybrominated diphenyl ethers detected was 2,310 pmol/g dry weight and this had fallen to 1,750 pmol/g dry weight (around 76% of the initial amount present) after 200 hours (a similar reduction in the total amount of polybrominated diphenyl ethers was also evident in the experiments with the standard reference material).

The photolysis of decaBDE in plastics has been studied by Kajiwara et al. (2008a). Some earlier results from the study are also given in Kajiwara et al. (2007) [ABST]. The experiments were carried out using samples of high impact polystyrene (HIPS) to which decaBDE was added, and composite samples of used TV cases (which already contained decaBDE). The HIPS samples were prepared by adding 50 g of HIPS to 500 ml of toluene containing 10 µg/ml of decaBDE. The mixture was shaken overnight to facilitate complete dissolution of the HIPS, the solvent evaporated in the dark and the HIPS sample was pulverized and screened (a fine powder of diameter between 106 and 300 µm was used in the experiments). The TV cases were obtained from a recycling plant in Japan. The casings from fifty TVs were crushed (<2 mm) and homogenised in a large volume mixer and a sub-sample was further pulverized and screened to give a fine powder (again with a diameter between 106 and 300 µm). The specific surface areas were 0.222 m^2/g for the HIPS sample and 0.275 m^2/g for the TV sample. The photolysis experiments were carried out using 0.30 g of the powdered plastics. The powder was placed in quartz tubes, sealed and exposed to natural sunlight (September 2006 to May 2007 in Tsukuba, Japan (36°02'N, 140°07'E)) for up to 224 days. During the exposure the tubes were kept in a temperature controlled glass room at 22°C and the tubes were constantly rotated (twelve revolutions per minute). Dark control experiments were also carried out. In addition, some samples of the HIPS containing decaBDE were hydrated with water (0.5 ml per tube) before the start of the experiment in order to investigate the effect of moisture on the photodegradation.

At various times during the exposure, one dark control and duplicate exposure samples were analysed for the presence of polybrominated diphenyl ethers (a total of twenty-five congeners were analysed covering di- to decaBDE), polybrominated dibenzo-*p*-dioxins (five congeners were analysed covering tri-, tetra-, penta-, hexa- and octa-congeners) and polybrominated dibenzofurans (seven congeners were analysed covering di- to octa- congeners). For these substances positive identification was made by comparison with authentic standards. In addition, several unknown peaks were also found to be present and these were assigned to the appropriate homologue group.

In the experiments with HIPS to which decaBDE was added, the initial concentration of decaBDE was 1,300 mg/kg (i.e. 0.13 per cent by weight), with smaller amounts of nonaBDEs (140 mg/kg), octaBDEs (4.4 mg/kg) and heptaBDEs (0.93 mg/kg) and no detectable di- to hexaBDEs. DecaBDE was found to disappear from the HIPS sample on exposure to sunlight, with around 50 per cent loss after around seven days. No loss of decaBDE was evident in the dark controls throughout the experiment and so the loss seen from the HIPS samples exposed to light represents photodegradation. The degradation seen over the whole experiment is shown in Figure 4.7.

Assuming a first order decay, the half-life of decaBDE under these conditions was estimated to be 51 days. The degradation rate in the experiments with added water was reported to be faster than seen in HIPS alone although data were available after 112 days exposure only and few other details of the experiments with water were given. Kajiwara et al. (2008a) also indicated that the rate of photodegradation of decaBDE in actual plastic articles would be much longer than suggested by the experiments with pulverised plastics as the surface area exposed to light had been maximised in these experiments.

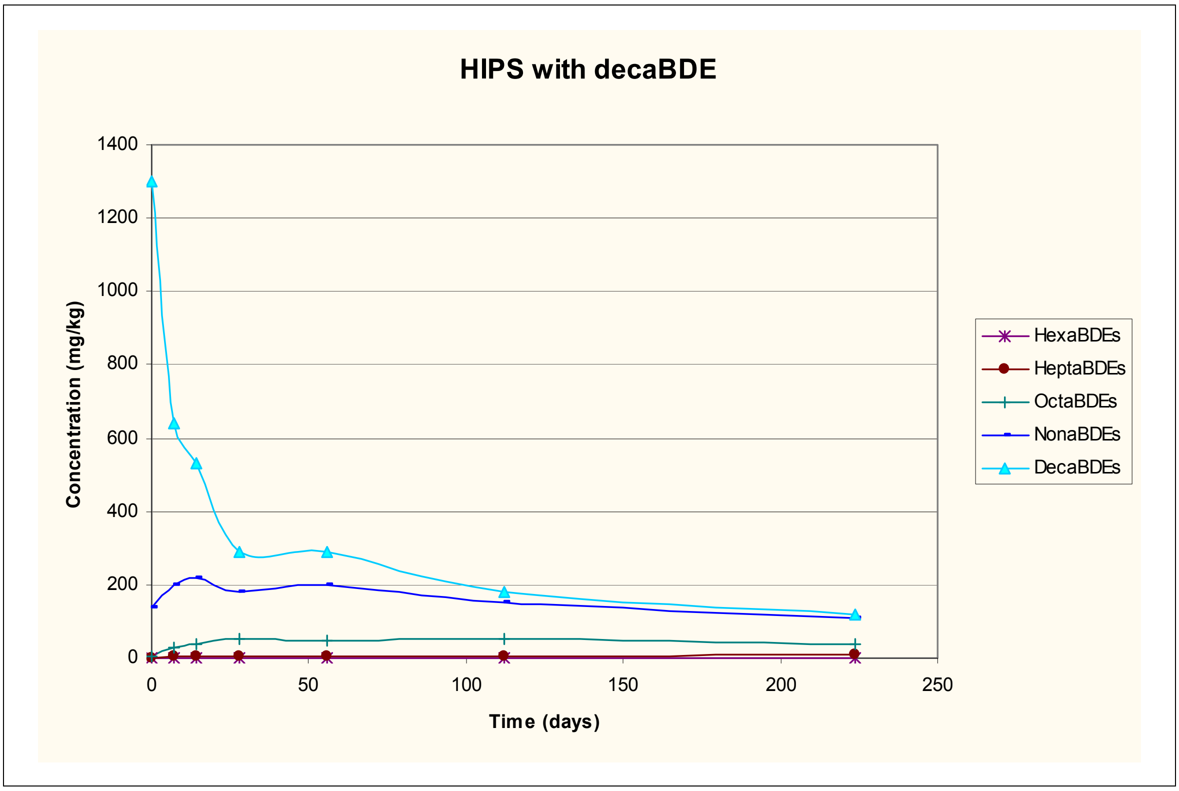

Figure 4.7 Photolysis of decaBDE in HIPS (after Kajiwara et al, 2008a).

The study with HIPS containing decaBDE showed that the concentration of several lower brominated congeners (hexa- to nonaBDEs) increased after one week of exposure (see Figure 4.7) indicating that they were products of the photodegradation of decaBDE. However, from one week onwards the concentrations of these lower brominated congeners remained relatively constant (or decreased slightly) while the concentration of decaBDE continuously declined. By the end of the study the concentration of total polybrominated diphenyl ethers found in the sample was less than 20 per cent of the initial concentration and the proportion of the total polybrominated diphenyl ethers that was attributable to decaBDE had changed from around 90 per cent at the start of the study to around 44 per cent by the end. No di- to pentabrominated congeners were seen at any time point.

For the other possible degradation products, no polybrominated dibenzo-*p*-dioxins were detected during the experiment with HIPS and decaBDE. However the concentration of total polybrominated dibenzofurans was found to show a marked increase (greater than 40 times) over the first seven days of the study (see Figure 4.8). At the start of the study, only traces of octabromodibenzofuran (0.15 mg/kg) and heptabromodibenzofuran (0.099 mg/kg) were determined in the samples but after one weeks irradiation tri- to hexabromodibenzofurans were also found to be present at concentrations ranging from 0.036 to 3.1 mg/kg. The concentrations of the brominated dibenzofurans then decreased with increasing irradiation, indicating that these substances were themselves subject to photodegradation.

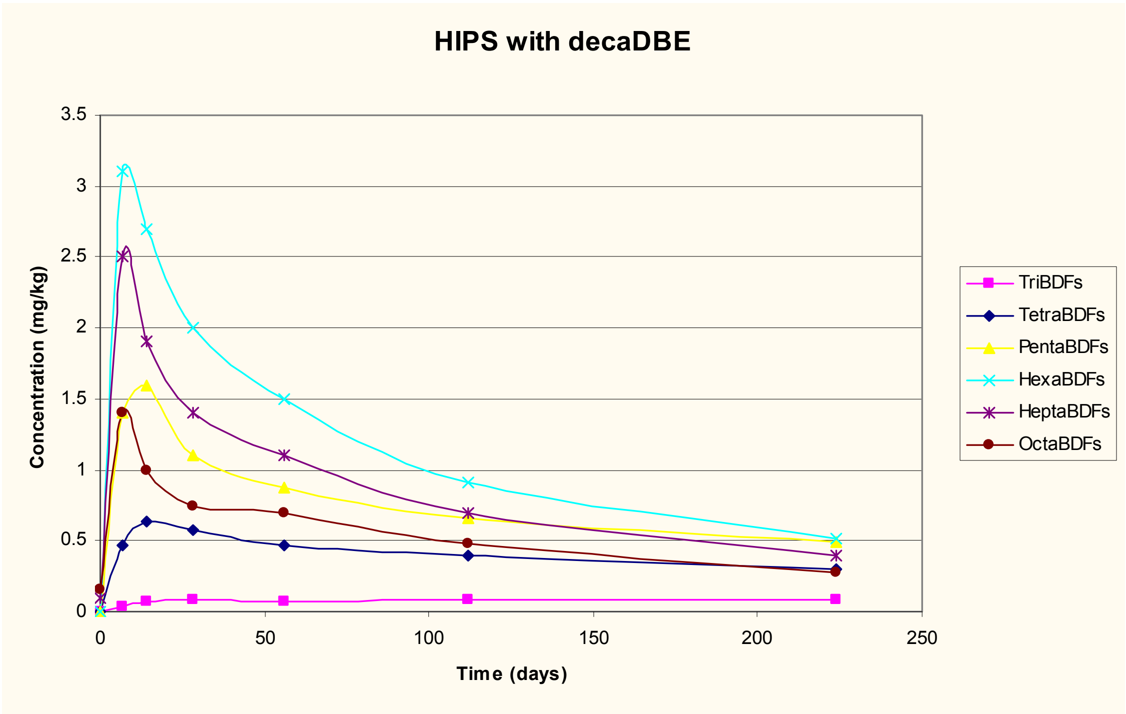

Figure 4.8 Formation of dibenzofurans during photolysis of decaBDE in HIPS (after Kajiwara et al, 2008a).

The experiments with the TV casing samples showed no clear degradation of decaBDE. The initial concentration of decaBDE in the samples was 96,000 mg/kg (i.e. 9.6 per cent) and the concentrations measured between day seven and day 224 were in the range 96,000 to 110,000 mg/kg. Although no significant loss of decaBDE was seen in these studies, the concentrations of di- to octabrominated dibenzofurans were found to show a continuous increase over the course of the study, with the level of total brominated dibenzofurans increasing approximately twenty times over the initial level present by the end of the study. A probable explanation for the different behaviour of decaBDE in the TV casing samples compared with the HIPS samples put forward by Kajiwara et al. (2008a) was that the TV casing samples would have contained additives other than decaBDE, for example colouring agents, UV absorbers and stabilizers, and these substances could have had a marked effect on the amount of light to which the decaBDE present was exposed (for example the TV casing samples were black but the HIPS samples were creamy white in colour). In addition the high concentration of decaBDE present in these samples made it difficult to detect any small amount of degradation of decaBDE that may have occurred. However the increase in the levels of brominated dibenzofurans during the experiment was thought to result from the photodegradation of decaBDE.

Mass balance calculations showed that in the experiments with decaBDE in HIPS, over 90 per cent of the decaBDE initially present was degraded in 224 days. However the identified products (lower brominated polybrominated diphenyl ethers and brominated dibenzofurans) only accounted for around 1.1 and 0.24 per cent respectively of the initial decaBDE at the end of the study. This implies that other, as yet unidentified products, may be major photodegradation products of decaBDE.

Although the Kajiwara et al. (2008a) study was carried out using ground samples of HIPS containing decaBDE, it is relevant to this assessment since dust containing decaBDE could be generated from plastic articles in use. However, the results are not appropriate for determining the fate of the bulk of decaBDE incorporated within articles manufactured from HIPS (or other plastics) as the surface area exposed to light would be much reduced compared to the experiments using ground samples.

Raff and Hites (2007a [ABST] and 2007b) carried out an investigation into the relative importance of various atmospheric removal processes for polybrominated diphenyl ethers. Firstly a series of experiments were carried out to determine the rate constant for reaction with atmospheric hydroxyl radicals, so that they could be compared with those predicted from a structure-activity relationship developed by Zetzsch (1982) and Kwok and Atkinson (1995) (the latter structure-activity relationship was used to estimate the rate constant for decaBDE in EC (2002)). The experiments were carried out using a series of mono- and dibrominated diphenyl ethers (BDE-1, BDE-2, BDE-3, BDE-4, BDE-7, BDE-11 and BDE-15). The results showed reasonable agreement between the measured rate constants and those estimated from the structure activity relationships (this comparison is only shown graphically in the paper), although it was noted that the structure-activity relationships were not able to predict changes in the rate constant resulting from differences in the bromine substitution pattern.

The second series of experiments examined the direct photolysis rate for biphenyl ether, 4-bromodiphenyl ether (BDE-3) and 2,4-dibromodiphenyl ether (BDE-7) in the gas phase. The substances were irradiated at 325 K over six to eight minutes using light of wavelength 310 nm. The degradation followed first order kinetics and the photolysis rates were found to increase with the degree of bromination (the results are displayed graphically only in the paper). A direct correlation between the photolysis rate and the absorption cross-section of the substance at 310 nm was found and the quantum yield for the photolysis of BDE-7 was determined to be 0.5 at 325 K.

The final part of the study was to compare the atmospheric residence times expected for reaction with hydroxyl radicals and direct photolysis for polybrominated diphenyl ethers. Estimated lifetimes were calculated for 20 congeners with between one and ten bromine atoms. The lifetimes for reaction with hydroxyl radicals were calculated using the reaction rate constant estimated from the structure-activity relationships and assuming an average global hydroxyl radical concentration of 9.7×10^5 molecules/cm^3. The lifetimes for direct photolysis were calculated using the absorption cross-section (determined from the UV-visible spectra of the polybrominated diphenyl ethers in iso-octane solution), assuming a quantum yield of 0.5 for all congeners and the actinic flux was estimated from the Tropospheric Ultra-violet-Visible (TUV 4.2) software package. The results of this comparison (again the data were only shown graphically) found that direct photolysis is likely to be the dominant loss process for polybrominated diphenyl ethers in the atmosphere. However, it was noted that this is only likely to be true for molecules present in the gas phase as a number of physical and chemical effects may hinder the direct photolysis of polybrominated diphenyl ethers adsorbed onto particles. Examples of such effects include shielding of UV-light by particulates, photophysical quenching by neighbouring molecules in the condensed phase and enhanced recombination in a 'solvent cage' within the aerosol. When the effect of partitioning to atmospheric particulates was taken into account (assuming that polybrominated diphenyl ethers in the particulate phase do not undergo photolysis) the analysis suggested that the congeners present mainly in the gas phase will be removed from the atmosphere primarily by direct photolysis but those that are bound mainly to atmospheric particulates, as is the case with decaBDE, will not be significantly degraded by sunlight but rather are likely to be removed from the atmosphere mainly by wet and dry deposition.

Evidence for this hypothesis was obtained by comparison of the polybrominated diphenyl ether congener profiles found in surface sediment from Siskiwit Lake located on Isle Royale in Lake Superior with the congener pattern in air (particulates plus vapour phase) from Eagle Harbour located near the shore of Lake Superior. Lake Siskiwit is a remote lake with few visitors that receives no water from Lake Superior and hence atmospheric deposition is the main source of polybrominated diphenyl ethers into the lake. The most abundant polybrominated diphenyl ether present in the lake sediment was decaBDE, accounting for 95 per cent of the total PBDEs. When the congener profile in sediment was compared to that in air it was seen that there was a higher proportion of BDE-209 in the sediment compared to that in the air. Raff and Hites (2007a and 2007b) suggested that if atmospheric deposition was the only loss process for polybrominated diphenyl ethers from air then the congener profile in the sediment would be expected to be similar to that in air but this comparison suggested that there was some depletion of the lower brominated congeners by photolysis (and/or reaction with hydroxyl radicals) compared to decaBDE in the air sample before deposition.

Schenker et al. (2008a and 2008b [ABST]) considered the importance of including photolysis of decaBDE in modelling the environmental fate of polybrominated diphenyl ethers. The model used was the global model CliMoChem (Climate Zone Model for Chemicals), which had previously been validated for other substances such as DDT (Schenker et al. 2008c). The rate constants for direct photodegradation in the atmosphere (both in the vapour phase and sorbed to aerosols) used in the study were extrapolated from experiments carried out in isooctane and cyclohexane by Raff and Hites (2007b) combined with data from other studies using organic solvents and silica gel. On the basis of these data it was assumed that the rate constants for photolysis in the gas phase were around two times larger than the values obtained in organic solvents and the rate constants for photolysis for substances adsorbed to aerosols were around sixty times smaller than the values obtained in organic solvents. Indirect photooxidation by reaction with hydroxyl radicals was also taken into account in the gas phase (it was assumed that this reaction did not occur when the substance was adsorbed to particulates). Photolysis on the surface of soil was not found to be important in the model (and so was not included in the main modelling exercise). However microbial degradation was found to be important and estimates for the rate of degradation (obtained from the EPIWIN Software package and an estimation method by Arnot et al. (2005)) were included in the model. The overall half-lives assumed in the model for decaBDE were 3,165 days in soil, 1,583 days in water, 318 days for atmospheric reaction with hydroxyl radicals and 0.42 days for direct photolysis in the gas phase. It was assumed that the yield of lower brominated congeners from the photolysis reaction was 80 per cent for each degradation step from decaBDE to hexaBDEs and 50 per cent for each subsequent step and that microbial degradation and reaction with hydroxyl radicals did not result in the formation of lower brominated congeners.

The model was run using estimates of the emissions into the various compartments of the model (based on previous published emission estimates). The overall persistence of decaBDE in the model was estimated to be 1,552 days assuming that no direct photolysis occurs; this was reduced to 183 days assuming that direct photolysis occurred. Similarly the spatial range (a measure of long-range transport potential given as a percentage of the pole-to-pole distance) was found to decrease from 30 percent (assuming no direct photolysis) to 10 per cent (assuming direct photolysis). The results obtained including direct photolysis were found to be the best predictor for the pattern of long-range transport for polybrominated diphenyl ethers as a whole.

Schenker et al. (2008a) carried out a comparison of the predicted concentrations in air obtained by the model with field data (no comparison was made for the other environmental compartments). It was assumed that the average background concentration of decaBDE measured in air in field studies was around 3.7 pg/m^3 in Europe and Asia and 0.88 pg/m^3 in North America. The predicted concentration of decaBDE in air in the northern temporate zone of the model was around 0.15 pg/m^3 which was an order of magnitude below the field data. Similarly, the predictions obtained for the model for tetraBDEs and pentaBDEs (predicted concentrations of 0.18 pg/m^3 and 0.08 pg/m^3 respectively) were again around one order of magnitude lower than the levels found in field studies (average levels around 5.3 pg/m^3 for tetraBDEs and 2.9 pg/m^3 for pentaBDEs). Possible explanations for the discrepancy between the predicted concentrations and the field data include underestimation of the actual emissions used as input to the model or assumption of a faster rate of degradation than actually occurs in the environment. Schenker et al. (2008a) noted, however, that despite these discrepancies in the predicted concentrations, the model correctly predicted the *relative* order of the concentrations of PBDEs found in the environment (i.e. the concentration of tetraBDEs> decaBDE>pentaBDE).

Schenker et al. (2008a) also used the model to investigate the contribution of decaBDE to the formation of lower brominated diphenyl ether congeners. It was estimated that around 50 per cent of the hepta- and hexaBDE congeners present in the environment could originate from degradation of decaBDE (with decaBDE also contributing around 11 per cent and 2.5 per cent to the penta- and tetraBDE congeners respectively). It should be noted that, owing to the nature of this study, there are considerable uncertainties in the input data used in the model, and hence the modelled results might not be realistic. Areas of high uncertainty include the emission estimates, degradation rates and yields of debromination products assumed in the model and these would all affect the outcomes of the modelling. Considering some of these uncertainties, Schenker et al. (2008a) put a lower and upper limit to the contribution decaBDE could make to the amounts of pentaBDE congeners in the environment as 3-20 per cent. However it should be noted that the actual rate of atmospheric photodegradation of decaBDE (and the other PBDEs included in the study), along with the yield of lower brominated congeners from the reaction in the atmosphere, is not known and this is a key parameter in the model.

Aquatic degradation

The photochemical degradation of decabromodiphenyl ether dissolved in isooctane solvent has been studied (Kuivikko et al. 2007[16]). The substance tested had a purity of >98.3 per cent. The photochemical degradation was carried out by exposing the solution of decabromodiphenyl ether (initial concentration 250 ng/ml) to natural sunlight on the roof of a building in Helsinki, Finland (60°20'N 24°97'E) for 60 minutes on 5 October. During the exposure the solutions were placed in glass vials and the glass vials were placed in a tank of water of depth 5-6 cm. The degradation of decabromodiphenyl ether was found to follow first order kinetics with a half-life of around 0.6 hours, and the quantum yield (Φ) was determined to be 0.28±0.06.

Based on this quantum yield, Kuivikko et al. (2007) also carried out model simulations of the predicted degradation half-life in the Baltic Sea (59.9°N, 23.3°E) and north Atlantic Ocean (60°N, 30°W). The model simulations assumed environmentally relevant concentrations of 30-40 pg/l in the water phase and the simulations were carried out assuming typical summer solar radiation at 60°N, accounting for the absorption by the natural dissolved and particulate matter. Close to the surface, the predicted photolysis half-life for decabromodiphenyl ether was around 0.02 days for the conditions in the Baltic Sea. Simulations were also carried out for the Baltic Sea and Atlantic Ocean for a mixing stratum of 10 m in the Baltic Sea and 30 m in the Atlantic Ocean. Under these conditions, the estimated photolytic half-lives were 1.8 days in the Baltic Sea and 0.4 days in the Atlantic Ocean.

Simulations were also carried out investigating the effects of latitude (between 0°N and 60°N along the 30°W meridian for a concentration of 3-4 pg/l) and season on the estimated photolytic half-life of decaBDE in the Atlantic Ocean. The half-life values calculated ranged from 0.1 days in the summer at 20 to 40°N to 33 days in the Winter at 60°N. Overall Kuivikko et al. (2007) concluded that decaBDE has a high potential to undergo photodegradation in the ocean at all latitudes and in all seasons.

The photolytic degradation of decaBDE in hexane solution has been studied by Shih and Wang (2009). In the study, solutions of decaBDE (concentration of 12.5 mg/l) were exposed to artificial light in wavelength ranges of 300-330 nm, 350-400 nm, 300-400 nm or natural sunlight (Taiwan, 24°07'14.9"N 120°40'29.0"E). Photodegradation was found to occur rapidly with a reaction rate constant of 0.11 min^{-1} using artificial light in the 300-330 nm range, 0.051 min^{-1} using artificial light in the 350-400 nm range, 0.050 min^{-1} using artificial light in the 300-400 nm range and 0.06-0.08 min^{-1} using natural sunlight. The rate was found to increase linearly with light intensitiy in the 714-896 W/m^2 range. Lower brominated diphenyl ethers (tri- to nonaBDEs) were identified as the main degradation products. The reaction was thought to procede via a step-wise loss of bromine, with the rate of loss of bromine from decaBDE in the first step being para- > meta- > ortho-. As this experiment was carried out using hexane solvent the environmental relevance of the reaction rates is unclear.

A further photodegradation study with decaBDE in organic solvents has been carried out by Zeng et al. (2008). In this study, decaBDE was dissolved in isooctane (concentration 196 µmol/l) and irradiated with artificial light (254 nm) at 35°C. Under these conditions, reductive debromination to form lower brominated congeners was found to occur and these results were used to test and validate a photodegradation model developed by Zeng et al. (2008) based on calculated enthalpies of formation of

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[16] The results from this study were available in abstract form in 2006 and were considered in ECB (2007). However there are some small differences in the data reported in ECB (2007) and that in the Kuivikko et al. (2007) paper and so the new results are considered here.

the PBDE congeners. However the conditions used in this experiment (organic solvent and light with a wavelength of 254 nm) mean that it is difficult to extrapolate the results to the environment.

Mas et al. (2008a) also studied the photodegradation of decaBDE using light with a wavelength of 254 nm. In this study solutions of decaBDE (concentration 120 mg/l) in tetrahydrofuran or in tetrahydrofuran:water mixtures (ratios of tetrahydrofuran to water of 90:10, 80:20, 70:30 and 60:40 volume/volume were used). The degradation reaction was monitored by determining the UV-visible spectrum in the 250-400 nm range at intervals during the exposure to the 254 nm light. This showed the degradation of decaBDE occurred under the conditions used and the UV-visible spectra suggested that as well as lower brominated diphenyl ether congeners, brominated dibenzofurans may have been formed. However it should be noted that this was based on the appearance of peaks in the UV-visible spectrum and no definitive analysis specific for brominated dibenzofurans was carried out. The rate of the photodegradation reaction was found to decrease as the amount of water present increased (first order reaction rate constants were determined as $2.66\text{-}3.32\times10^{-4}$ s^{-1} in the experiment with a 90:10 ratio of tetrahydrofuran:water and $1.64\text{-}2.4\times10^{-4}$ s^{-1} in the experiment with a 60:40 ratio of tetrahydrofuran:water. Again, the conditions used in this experiment (organic solvent and light with a wavelength of 254 nm), and the relatively high concentration of decaBDE used (it is doubtful that decaBDE would be soluble at this concentration in some of the solvent:water mixtures) mean that it is difficult to extrapolate the results to the environment.

A detailed investigation of the products formed from photolysis of decaBDE in tetrahydrofuran, methanol and methanol-water mixtures has been carried out by Christiansson et al. (2009). The photolysis was carried out by exposing solutions of decaBDE (concentration 1 µmol/l) for either 100 minutes (for experiments carried out in tetrahydrofuran, methanol, a 90:10 mixture of methanol and water and a 80:20 mixture of methanol and water) or 200 minutes (for experiments carried out using a 90:10 mixture of methanol and water and a 80:20 mixture of methanol and water) to an artificial UV light source (a 20 Watt fluorescent lamp). The irridation time of 100 minutes corresponded to around seven half-lives for decaBDE in tetrahydrofuran and the irradiation time of 200 minutes corresponded to around seven half-lives for decaBDE in an 80:20 mixture of methanol and water. The wavelengths emitted by the lamp were not given in the paper but it was reported that the system was the same as used in the Eriksson et al. (2004) study that is summarised in ECB (2007). This former system used artificial UV light in the sunlight region and so it is assumed that this was also the case in the Christiansson et al. (2009) study.

At the end of the irridation phase a detailed analysis of the products formed was undertaken. The results of this analysis are displayed graphically in Figure 4.9.

The yield of identified products (effectively a mass balance) was found to be 104 per cent in the experiment with tetrahydrofuran, 105 per cent in the experiment with methanol, 65 per cent (after 100 minutes irridation) and 33 per cent (after 200 minutes irridation) in the experiments with a 90:10 mixture of methanol and water and 53 per cent (after 100 minutes irradiation) and 42 per cent (after 200 minutes irridation) in the experiments with a 80:20 mixture of methanol and water. The data presented in Figure 4.9 represent the actual yield of the various products taking into account the overall yield (the data are presented in Christiansson et al. (2009) in terms of the percentage of the identified products). In all experiments, photolysis of decaBDE was found to lead to the formation of lower brominated diphenyl ether congeners (mainly hexa- to nonabrominated congeners), and also polybrominated dibenzofurans (PBDFs) (mainly mono- to pentabrominated congeners). The brominated diphenyl ethers accounted for approximately 90 per cent of the total amount of substances identified and the brominated dibenzofurans accounted for around 10 per cent of the total amount of substance identified in each case. In addition, the experiments in methanol led to the formation of methoxylated-brominated dibenzofurans (MeO-BDFs). As noted in Christiansson et al. (2009) it is unlikely that methoxylated-brominated dibenzofurans would be formed in the environment by this route as their formation in these experiments is dependent on the presence of the methanol solvent. Other products formed included pentabromophenol, dihydroxytetrabromobenzene, di-hydroxydibromodibenzofuran, dihydroxytribromodibenzofuran, and dihydroxytetrabromodibenzofuran.

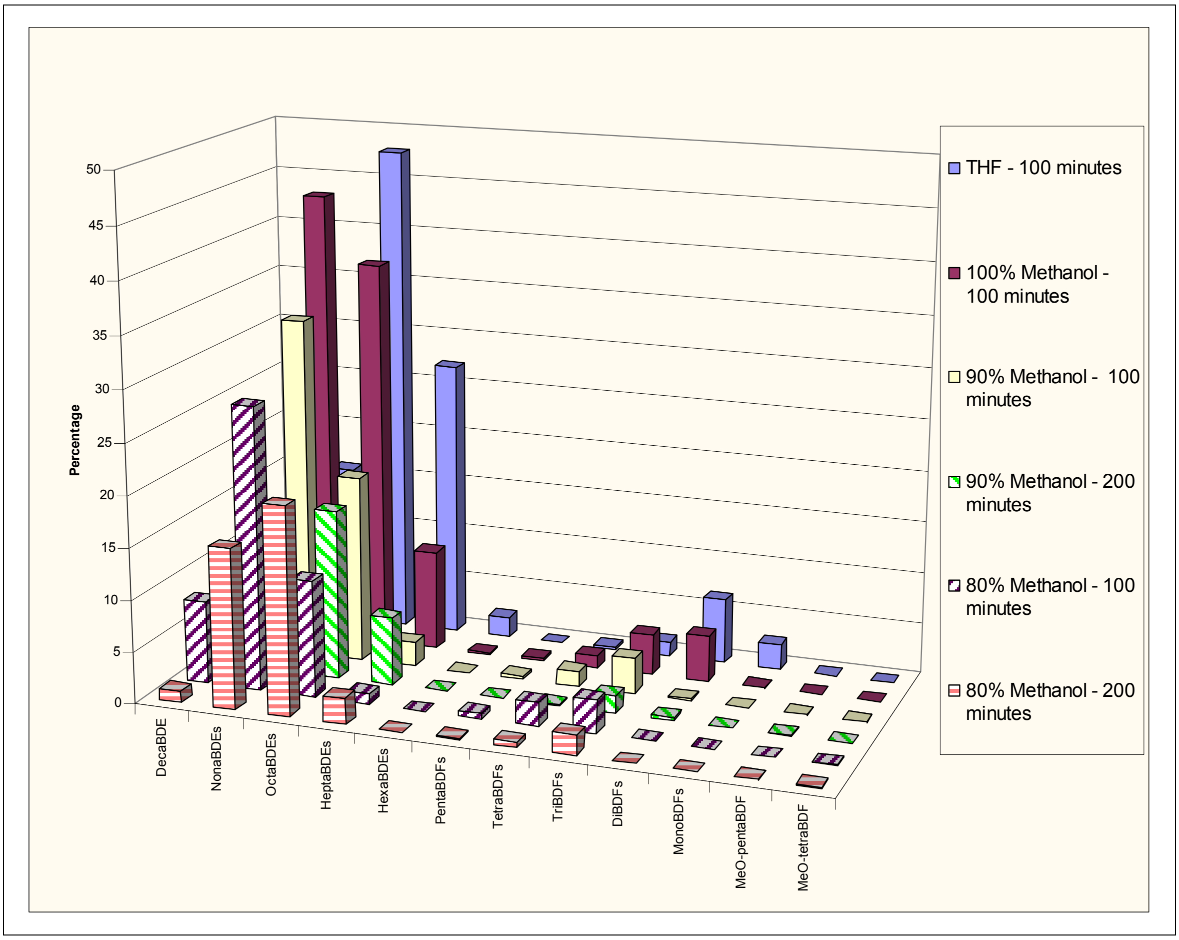

Figure 4.9 Summary of products from photodegradation of decaBDE in various solvents.

It should be noted that, as the debromination reaction is a sequential reaction, the product distributions found are a function of the irridation time. Longer or shorter irridation times would lead to different product distributions.

The degradation of decaBDE has been studied by Tokarz et al. (2008). The study was carried out using a co-solvent enhanced biomimetic system with vitamin B_{12} as an electron-transfer mediator. Vitamin B_{12} was chosen as it occurs widely in anaerobic microorganisms. In addition, experiments were carried out using sediment microcosms for comparison with the results from the biomimetic system.

The decaBDE used in the study contained small amounts of nonaBDE (2.0 per cent BDE-206, 1.9 per cent BDE-207 and 0.9 per cent BDE-208 on a mole fraction basis).

The biomimetic experiments were carried out with a mixture of decaBDE (0.03 mM), titanium citrate (5.0 mM; acted as electron-donating reducing agent) and vitamin B_{12} (0.2 mM) in a buffer solution (pH 7.4) containing tetrahydrofuran (33 per cent by volume). The final volume was 31 ml and the calculated redox potential of the solution was -400 mV (this was thought to be similar to the reducing conditions in methanogenic sediments). Control experiments were carried out without the addition of titanium citrate. The experiments were carried out in triplicate and the solutions were incubated in the dark in an anaerobic chamber for up to 24 hours. At intervals during the test, sub-samples were collected from each chamber and analysed for the presence of polybrominated diphenyl ethers.

Rapid debromination of decaBDE was evident in the biomimetic experiments (the half-life was ~18 seconds; complete debromination occurred within five minutes). At least twelve degradation products were observed but the dominant products were hexabromodiphenyl ethers after five minutes. At longer time periods (24 hours) the dominant products present were tetra- (e.g. BDE-47 and BDE-66) and pentabromodiphenyl ethers (e.g. BDE-99 and BDE-119). These are the PBDE congeners that are most frequently detected in the environment (contrary to other studies, no selective meta- or para-debromination was observed).

The sediment microcosms were constructed using a natural loam sediment (which contained no detectable polybrominated diphenyl ethers) with a pH of 6.3 and an organic carbon content of 16.4 per cent. DecaBDE (3.5 mg) was firstly added to 10 g of air-dried, sieved (2 mm) sediment as a solution in toluene and the toluene allowed to evaporate. The spiked dried sediment was then mixed into 70 g of sieved wet sediment (water content approximately 50 per cent) giving a final decaBDE concentration of 5.0 mg/kg [Note: the paper indicates that the final concentration was 5.0 mg/kg but this does not appear to be correct as a total of 3.5 mg of decaBDE was added to 80 g of sediment – thus the initial concentration would appear to be approximately 3.5/0.080 = 44 mg/kg]. The spiked sediment was added to 125 ml serum bottles containing 50 ml of phosphate buffer and the sediments were fed with 50 µl of methanol and 25 mg of dextrose in order to provide an organic electron donor and to ensure anaerobic conditions (the combination of dextrose and methanol promotes the rapid onset of anaerobic conditions without the need for adding exogenous reducing chemicals). The microcosms were sealed, shaken, and then incubated in the dark at 22°C. Three replicate microcosms appear to have been prepared (although this is not altogether clear from the paper). Control microcosms were prepared in a similar manner and were autoclaved three times prior to use. In addition, a second set of sediment microcosms that had been constructed three years earlier were also used in the study (the concentration of decaBDE in these microcosms was 0.3 mg/kg).

The methane gas produced by the sediment microcosms was measured by inserting a glass-barrelled syringe through the stopper. It is not clear from the paper how many replicate microcosms were used. At intervals during the test, sub-samples of the sediment were collected and analysed for the presence of polybrominated diphenyl ethers.

DecaBDE was found to degrade only very slowly in the sediment microcosms. For example, it was found that only a very slight decrease in the amount of decaBDE was evident after 10 months' incubation, with a concurrent increase in all three nonaBDEs. However, more extensive degradation was evident in the older microcosms after 3.5 years of incubation (the amount of decaBDE present was around 55 per cent of that initially added in one replicate, around 80 per cent of that initially added in two other replicates and around 85 per cent of that initially added in the control; values read from a graph). In addition, a statistically significant increase in the mole fraction of all three nonaBDEs was evident, along with the appearance of a number of lower brominated congeners (mainly hepta- and octaBDEs, but small amounts of hexaBDEs (e.g. BDE-128 and BDE-138) was also apparent in one of the replicates). The concentrations are low (the mole fraction of the hexaBDE congeners appears to be less than 1%), and the influence of analytical variability on these results is unknown. The half-life of decaBDE was estimated to be between 6 and 50 years, with an average of around 14 years.

In order to compare the rates of degradation seen in the biomimetic system with those obtained in the sediment system, Tokarz et al. (2008) estimated an “effective” debromination rate constant by dividing the rate constant obtained in the biomimetic system by a term that accounted for adsorption of decaBDE onto organic matter[17]. Using this method, Tokarz et al. (2008) estimated an “effective” rate constant of 0.284 year^{-1} and hence an “effective” half-life of 2.4 years. This half-life compares reasonably well with the lower limit of those obtained in the sediment microcosms. When considering these data it is relevant to take into account the fact that the sediment used in this study had a higher organic carbon content (16.4 per cent) than is usually considered in the REACH Guidance Document (typically 5 per cent organic carbon content is assumed). Assuming an organic carbon content of 5 per cent in the methodology used by Tokarz et al. (2008) results in an “effective” half-life of 0.93 year^{-1} and an “effective” half-life of 0.75 years.

Overall, the results obtained in the biomimetic system suggest that decaBDE has the potential to debrominate in the environment under anaerobic conditions to form more toxic and accumulative lower brominated congeners. However, the conditions used in the biomimetic system mean that it is difficult to relate these results directly to the degradation behaviour of decaBDE in the environment. The experiments with sediment microcosms are less conclusive. Only one out of the three replicates appeared to show significant degradation – there is little information about how this microcosm was treated during the 3.5 year period (e.g. whether feeding with methanol and dextrose was carried out at intervals over this time period, etc.), so the viability of the microcosm is unclear. The study was carried out at 22°C and so at more environmentally-relevant temperatures the degradation would be expected to be even slower. Conversely, the sediment used in this study had a relatively high organic carbon content (16.4 per cent) and it is possible that bioavailability, and hence degradation rate, could be higher in sediments with lower organic carbon contents.

The main implication of this study is that there is a long time lag between disappearance of decaBDE and the subsequent formation of the PBDE congeners of concern (probably owing to the number of sequential steps that need to be gone through). The timeframe for this degradation is therefore very long (half-lives of the order of tens of years). In other words, the degradation of decaBDE to lower congeners of concern is possible, but is likely to be so slow as to be practically immeasurable using normal laboratory systems.

Nose et al. (2005 [ABST] and 2007) investigated the degradation of decaBDE using hydrothermal treatment. Hydrothermal treatment is one of several method used in Japan for treating waste containing polychlorinated biphenyls. Experiments were carried out with decaBDE using a stainless steel micro autoclave (50 ml capacity) filled with 40 ml of hexane-washed distilled water. DecaBDE was added to the water phase as a solution in toluene (100 µl of a 100 mg/l solution in toluene was added; the

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[17] This term was based on the fraction of organic carbon in the sediment and the log K_{ow} of decaBDE and took into account the soil mass to solution volume ratio in the sediment microcosms.

resulting nominal decaBDE concentration in the chamber would therefore be 0.25 mg/l). The vessel was then sealed and held at 300°C and a pressure of 8 MPa for 120 or 240 minutes. After this time, the water phase was extracted with hexane and the extracts were analysed for the presence of polybrominated diphenyl ethers and polybrominated dibenzo-*p*-dioxins and furans.

Under these conditions, decaBDE was found to be degraded to lower brominated congeners. More than 99.999 per cent degradation of decaBDE was seen after 120 minutes. The products formed during the reaction are summarised in Table 4.18.

Table 4.18 Degradation of decaBDE by hydrothermal treatment.

Substance	**Concentration (µmol/l)**		
	Initial	**120 minutes**	**240 minutes**
Brominated diphenyl ethers			
Decabromodiphenyl ether	990	0.10	0.095
Nonabromodiphenyl ethers	50	0.67	0.057
Octabromodiphenyl ethers	3.1	1.7	0.17
Heptabromodiphenyl ethers	1.9	0.55	0.29
Hexabromodiphenyl ethers		0.022	0.071
Pentabromodiphenyl ethers		0.013	0.064
Tetrabromodiphenyl ethers		0.014	0.049
Tribromodiphenyl ethers		0.019	0.079
Dibromodiphenyl ethers		0.020	0.091
Monobromodiphenyl ethers		0.037	0.11
Brominated dibenzo-*p*-dioxins			
Octabromodibenzo-*p*-dioxin		ND	ND
Heptabromodibenzo-*p*-dioxin		0.024	ND
Hexabromodibenzo-*p*-dioxin		0.52	0.030
Pentabromodibenzo-*p*-dioxin		0.55	0.040
Tetrabromodibenzo-*p*-dioxin		0.012	0.036
Brominated dibenzofurans			
Octabromodibenzofuran		ND	ND
Heptabromodibenzofuran		0.042	0.0015
Hexabromodibenzofuran		0.37	0.044
Pentabromodibenzofuran		0.094	0.11
Tetrabromodibenzofuran		0.054	0.27

Notes: ND = not detectable.

As can be seen, lower brominated congeners (including tetra- and pentaBDEs) were formed, along with polybrominated dibenzo-*p*-dioxins and furans, and the yield of these products depended on the reaction time. The main nonaBDEs found were BDE-207 and BDE-208 in approximately equal amounts, indicating that the reactivities of para- and meta-substituted bromines were similar and higher than the reactivity of ortho-substituted bromines.

The degradation of decaBDE in solution by zero-valent iron has been demonstrated previously (see the study by Keum and Li (2005) reported in ECB (2007)). Further analysis of the products from this reaction has been carried out Wang et al. (2008) using a new analytical methodology designed to optimise the simultaneous analysis of both lower brominated congeners (in this case mono- to heptaBDEs) and the higher brominated congeners (e.g. octa- to decaBDEs). The analysis was carried out on a solution of decaBDE dissolved in ethyl acetate (concentration of 50 µg/ml) that had been exposed to zerovalent iron powder for 40 days. A total of 43 BDE-congeners were detected in this solution, and twenty four of these could be identified and quantified (using authentic PBDE standards). These are shown in Table 4.19.

For comparison Keum and Li (2005) identified BDE-17, BDE-28/33, BDE-47, BDE-66, BDE-99, BDE-100, BDE-138. BDE-153, BDE-154, BDE-183 and BDE-190 from the same solution. The identified products accounted for around 74 per cent of the initial amount of BDE-209 added to the solution with the nineteen unidentified BDE congeners accounting for the remaining 26 per cent.

Li et al. (2007) have carried out further investigations of the degradation of decaBDE by zerovalent iron. The study used nanoparticles of zerovalent iron embedded in an ion-exchange resin to effect the debromination. The tests were carried out using a 1 mg/l solution of decaBDE (purity 97 per cent) in a 1:1 mixture of acetone:water (total volume 8 ml) to which 2 g of the resin (containing 0.11 g of the nanoparticulate zerovalent iron) was added. The test vessels were capped and maintained at 25 °C with shaking for up to ten days. Under these conditions, debromination of the decaBDE was found to occur rapidly with a half-life of around 2.4 hours. No debromination of decaBDE was seen in control solutions. Nona- to tribrominated diphenyl ether congeners were found to be formed in a sequential manner. All three possible nonaBDE congeners were found to be formed in significant amounts within one hour of the start of the experiment, and their concentration increased over the first eight hours of the experiment. After this period the concentrations of the nonaBDE decreased to not detectable levels after 24 hours. OctaBDEs (five main congeners; two of which were identified as BDE-196 and BDE-197)) were also evident over this time period. After two days exposure, heptaBDEs (three major congeners, including BDE-183, and one minor congener) were the predominant congeners present and after longer exposure periods, hexaBDEs were evident (at least six congeners including BDE-153 and BDE-154). At the end of the experiment (ten days total exposure) pentaBDEs (including BDE-99) were present in significant amounts and tetraBDEs (including BDE-47) had started to appear.

Sun et al. (2009) investigated the photocatalytic degradation of decaBDE using titanium dioxide. The study found that dispersions of decaBDE (2×10^{-5} mol/l) and titanium dioxide (1 g/l) in acetonitrile were rapidly degraded by a reductive debromination mechanisms on exposure to light at wavelengths >360 nm in the absence of air. However, the degradation reaction was found to be markedly inhibited by the presence of air and traces of water (1 per cent water was found to stop the reaction), and was also found to depend markedly on the solvent used (for example little or no degradation was found using hexane, toluene, tetrahydrofuran, acetone or dimethyl sulphoxide and the degradation rate was reduced in methanol compared with that seen in acetonitrile). The dependence of this reaction on the solvent and, in particular, the need for a lack of water or air, means that the results from this study have little relevance to the fate of decaBDE in the environment.

Table 4.19 Products identified from the treatment of decaBDE with zerovalent iron.

Congener	Concentration (µg/ml)
Monobromodiphenyl ethers	
BDE-3	Not detected
Dibromodiphenyl ethers	
BDE-7	0.020
Tribromodiphenyl ethers	
BDE-15	0.046
BDE-17	1.7
BDE-28	1.3
Tetrabromodiphenyl ethers	
BDE-47	3.4
BDE-49	11
BDE-66	1.3
BDE-71	Not detected
BDE-77	0.14
Pentabromodiphenyl ethers	
BDE-85	Not detected
BDE-99	0.64
BDE-100	0.043
BDE-119	0.51
BDE-126	0.020
Hexabromodiphenyl ethers	
BDE-138	0.15
BDE-153	1.5
BDE-154	2.2
BDE-156	0.067
Heptabromodiphenyl ethers	
BDE-183	0.65
BDE-184	0.19
BDE-191	0.35
Octabromodiphenyl ethers	
BDE-197	0.60
BDE-196	1.8
Nonabromodiphenyl ethers	
BDE-206	2.2
BDE-207	1.32
Decabromodiphenyl ether	
BDE-209	1.6

Olsman et al. (2007) carried out a study to investigate whether the degradation of decaBDE under anaerobic conditions led to the formation of Ah Receptor agonists. Batch experiments were carried out using around 49 and 46 g of digested household waste from semi-continuous laboratory scale (four litre) mesophilic and thermophilic digesters[18] amended with 2.4 g and 4 g respectively of household waste. DecaBDE was added as a solution in dimethyl sulphoxide to give a final concentration of 0.24 g/l. Sterile controls and solvent controls were also run. The bottles were capped and incubated for up to 60 days at 37°C (mesophilic conditions) or up to 40 days at 55°C (thermophilic conditions). At various times during the incubations the Ah Receptor agonist activity was determined using both the CELCAD assay (chick embryo liver cell assay for dioxins) and the DR-CALUX assay (using a transfected rat hepatoma cell line). No significant changes in the response of these assays were seen over time in any of the treatments. The tetrachlorodibenzo-*p*-dioxin (TCDD) equivalents (TEQs) determined in the assays were all close to the detection limit and were thought to reflect the variability in the background bioactivity of the household waste rather than any biotransformation of decaBDE.

It is understood that a long-term study to investigate the fate of decaBDE in aquatic mesocosms in a boreal lake is being undertaken (Orihel et al. 2008). No results of this study are available at the current time.

Degradation in soil and sewage sludge

No new data have been located on the degradation of decaBDE in soil. However, there is one study (Zhou et al. 2007) that has investigated the degradation of decaBDE by white rot fungi under aerobic conditions. As fungi are present in soils the results are presented here.

The decaBDE used in the study had a purity of 98 per cent (Zhou et al. 2007). The cultures used for the degradation experiments were prepared by adding one millilitre of a solution of decaBDE in dichloromethane (concentration 160 mg/l) to a 250 ml flask and allowing the solvent to evaporate (this produced a coating of 160 µg of decaBDE on the bottom of the flask). Aqueous culture medium (100 ml of a medium containing yeast, peptone and glucose) was added and the flasks were inoculated with white rot fungi GIM3.383 (the amount added was not given) and the flasks were incubated in the dark for up to ten days at 30°C on an orbital shaker. Experiments were also carried out to investigate the effects of a non-ionic surfactant (Tween 80) and ß-cyclodextrin on the bioavailability of decaBDE in this system. The culture flasks for these experiments were prepared in the same way as above except that either Tween 80 or ß-cyclodextrin was added to the aqueous culture medium at concentrations up to 900 mg/l. All experiments were carried out in triplicate and sterile samples were also prepared as controls.

Degradation of decaBDE was evident in the experiments without Tween 80 or ß-cyclodextrin. The amount of decaBDE present in the flask was found to decrease from an initial (nominal) value of 160 µg to 92.5 µg by day ten (42.2 per cent degradation). No significant change in the amount of decaBDE in the control cultures occurred over the same timeframe. The extent of degradation was found to be increased by both Tween 80 and ß-cyclodextrin. With Tween 80, the maximum degradation was seen at a concentration of 500 mg/l where 96.5 per cent degradation

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[18] The digesters had been operated at 37°C (mesophilic conditions) or 55°C (thermophilic conditions) using a feed of organic household waste (the organic loads used were typical of those used in large-scale biogas processes) and the digesters had hydraulic retention times of 30 days (mesophilic digester) and 20 days (thermophilic digester).

of decaBDE occurred in ten days (and no decaBDE could be detected after 12 days), however the degradation appeared to be inhibited at higher concentrations (for example 31.5 per cent degradation of decaBDE was evident after ten days at a Tween 80 concentration of 900 mg/l). ß-Cyclodextrin showed a similar, but less marked, enhancement on the degradation of decaBDE as Tween 80 with the highest degradation (78.4 per cent degradation after ten days) of decaBDE occurring at a ß-cyclodextrin concentration of 700 mg/l, and a slightly lower degradation (76.4 per cent degradation after ten days) occurring at a ß-cyclodextrin of 900 mg/l. The degradation products were not determined in this study. It should be noted that the mass of mycelium present in the cultures increased during the course of the experiment, and the increase in the mass of the mycelium in the presence of both Tween 80 and ß-cyclodextrin followed a broadly similar trend to the degradation rate. Although the analytical method included a step for the extraction of decaBDE from the mycelium no information is given in the paper as to the effectiveness of the extraction method used (sonication for fifteen minutes with dichloromethane). Therefore it is possible that some of the loss of decaBDE seen in this study could result from adsorption onto the mycelium (and incomplete extraction during the analytical method).

The degradation of polybrominated diphenyl ethers (including decaBDE) in sewage sludge has been investigated by Stiborová et al. (2008) [ABST]. The sludge used in the study was taken from two waste water treatment plants from industrialised areas in the Czech Republic. The polybrominated diphenyl ethers initially present in the sediment included BDE-28, -47, -49, -66, -85, -99, -100, -154, -183)[19] at a total concentration of 920.9 and 220.4 µg/kg dry weight in the two sludge samples, along with decaBDE at a concentration of 685.3 and 1,402.6 µg/kg dry weight respectively. The sewage sludge samples were stored at 4°C for up to four weeks before use.

The degradation of the polybrominated diphenyl ethers was investigated under both aerobic and anaerobic conditions. For the aerobic conditions, slurries of the sewage sludge were used (consisting of 15 g wet sewage sludge and 35 ml of mineral medium). Three series of flasks were prepared (three parallel flasks for each series). The first series contained the sewage sludge slurry alone, the second series contained the sewage sludge slurry amended with yeast extract (50 mg/l) and the third series contained the sewage sludge amended with both yeast extract (50 mg/l) and 4-bromobiphenyl[20] (0.6 mg/l). Control flasks were prepared by heat-sterilisation of the sludge. The flasks were then incubated in the dark at 28°C with constant shaking (150 rpm) for three months.

At the end of the three months period the amount of polybrominated diphenyl ethers present in the sludge slurry had decreased by around 30 per cent (total of BDE-29 to BDE-183) and 20 per cent (decaBDE). Yeast extract and 4-bromobiphenyl was found to have no significant effect on the loss seen.

The anaerobic experiments were carried out by suspending the sewage sludge samples in a mineral medium (ratio 40 sludge:60 medium; total volume 50 ml) in capped serum bottles. Starch (20 mg) and yeast extract (50 mg) were also added to the test system. A second series of experiments were carried out with the addition of starch, yeast extract and 4-bromobiphenyl[20] (amount not given). Sterile control bottles were also prepared. The bottles were incubated in the dark at 28 °C (and 150 rpm) for six months. Shortly after incubation had started, gas production was noted in the bottles indicating that methanogenic conditions had been attained. Significant loss of decaBDE was evident in experiments with only one of the sewage sludge samples (the concentration of decaBDE was found to remain constant in the other sludge sample).

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[19] These were the congeners analysed for. It is possible that other congeners may have been present that were not detected by the analytical method used.

[20] At one place in the paper this is referred to as 4-bromobiphenyl ether and so it is not clear exactly what was added.

Loss of the lower brominated congeners initially present was evident in both sludges (up to 50 per cent in one sludge and up to 30 per cent in the other sludge) and the loss was generally higher in the experiments where 4-bromobiphenyl had been added.

It should be noted that few other details of this study are currently available. In particular no information is given on the loss of polybrominated diphenyl ethers in the sterile controls at the end of the incubation period. Therefore it is difficult to assess the significance of the loss of polybrominated diphenyl ethers seen at present

Summary and evaluation of environmental degradation data

A number of important new studies have been carried out investigating the possible degradation of decaBDE in the environment.

The studies by Stapleton and Dodder (2008) and Kajiwara et al. (2008a) both demonstrate that photodegradation of decaBDE adsorbed on to the surface of particulates can occur leading to the formation of nonaBDEs, octaBDEs and heptaBDEs (and possibly hexaBDEs in the case of the Kajiwara et al. (2008a) study). In addition Kajiwara et al. (2008a) found that polybrominated dibenzofurans (tri- to octabromo) were also formed. Although the rate of degradation appears to be slow (half-lives estimated to be in the range 50 to 200 days) and the yield of the lower brominated congeners also appears to be low, these are significant findings as the available monitoring data in Section 4.2.4 demonstrates that decaBDE is a common contaminant of household dust and is present in some samples at milligram per kilogram levels. It should also be noted that these reactions may also lead to the formation of as yet unidentified products.

Photodegradation of decaBDE in solution has also been studied further. Although the study by Kuivikko et al. (2007), as is the case with many of the previous studies (see EC (2002), ECB (2004) and ECB (2007)), was carried out using organic solvents (and so the results are not directly applicable to the environment), the fact that photolytic debromination has been shown to occur with decaBDE adsorbed to particles in the atmosphere suggests that similar debromination reactions involving decaBDE adsorbed onto the surface of particulates in the aquatic compartment could be expected to occur if exposed to sufficient amounts of light.

Christiansson et al. (2009) carried out a detailed analysis of the products formed during photodegradation of decaBDE in organic solvents and organic solvent/water mixtures. The main products identified were lower brominated PBDE congeners but the study also showed that brominated dibenzofurans could potentially be formed.

It is still not possible to reliably estimate the amounts of lower brominated congeners (and other compounds) that may be formed in the environment from photodegradation of decaBDE, for a number of reasons. For example, decaBDE will adsorb to particulates in both water and the atmosphere and so some shielding from light would be expected. In addition, other removal mechanisms may operate before significant degradation could occur, for example wet and dry deposition of atmospheric particulates. The monitoring studies reported in Section 4.2.4 are also inconclusive on this point – the study of Thomas and Jones (2007) suggests that atmospheric degradation of decaBDE is not the primary source of the octaBDE congeners found in the air samples that were measured. Conversely, the congener profile found in the Wilford et al. (2008) study is suggestive of debromination of decaBDE to nonaBDEs in indoor air samples (although other explanations are also possible).

A recent global modelling study by Schenker et al. (2008a) suggests that if photodegradation of decaBDE in the atmosphere does occur then the reaction could represent a significant source of the hepta- and hexaBDEs (and a small source of penta- and tetraBDEs) present in the environment. However, the model used includes a number of assumptions and approximations and so the results do not prove the significance of such reactions.

Degradation (debromination) of decaBDE using reductants (zero valent iron and vitamin B_{12}) has been studied further. The experiments suggest that debromination of decaBDE can be carried out using certain reductants that are found in natural systems (and add to those considered in ECB (2007)). In particular, Tokarz et al. (2008) have shown that hexaBDE congeners may be formed in sediment microcosm experiments given sufficient time (half-lives of the order of tens of years). The sediment used in this study had a relatively high organic carbon content (16.4 per cent) and it is possible that bioavailability, and hence degradation rate, could be higher in sediments with lower organic carbon contents. Although the conditions used in many of the experiments are not representative of those in the environment (making it impossible to estimate the likely rate or yield of any such reactions), it cannot be ruled out that such reactions could occur in the environment.

Degradation of decaBDE by white rot fungus has also been demonstrated but no information on the products from the degradation was provided.

Overall the new data provide evidence that decaBDE can undergo photolytic debromination when adsorbed onto particulates forming nona- to heptaBDEs (and possibly hexaBDEs) as products. The formation of polybrominated dibenzofurans (tri- to octabromo) also appears to be possible. It is also likely that debromination could occur in other systems where natural reductants are present (e.g. sediments). It is not, however, possible to reliably quantify the rate of degradation in the environment or the yield of debrominated products in the environment.

4.3.2 Environmental partitioning

A study of the partitioning behaviour of polybrominated diphenyl ethers between air, water, sediment and fish has been carried out in Lake Thun, Switzerland (Bogdal et al. 2007) [ABST]. Lake Thun is an oligo-mesotrophic lake in an alpine region and has a surface area of 47.69 km^2, a mean depth of 136 m, a volume of 6.42 km^3 and a hydraulic residence time of 684 days. The samples analysed included a single sample of surface sediment from the deepest part of the lake (depth 217 m) collected in July 2005, 57 samples of whitefish (*Coregonus* sp.) muscle collected in September 2005, a water sample collected in March 2007 from the same location as the sediment sample and a sample of bulk atmospheric deposition collected over a six-week period from the southern shore of the lake during July and August 2006. The levels found are summarised in Table 4.20. The data showed that decaBDE partitioned mainly to the sediment and particulate phase. Based on the known sedimentation rate in the lake and the concentration of decaBDE found in sediment, Bogdal et al. (2007) estimated that the flux of decaBDE to sediment is around 1.7 $\mu g/m^2$ per year. This compares well with the blank-corrected atmospheric deposition data (5.0 ng/m^2 per day or 1.8 $\mu g/m^2$ per year) and suggests that the sedimentation flux of decaBDE in the lake can be largely explained by atmospheric deposition. The octa- and nonaBDEs included in the analysis were BDE-194, BDE-196/200, BDE-197/204, BDE-198/203, BDE-201 and BDE-202 (octaBDEs) and BDE-206, BDE-207 and BDE-208 (nonaBDEs). The concentrations found for the individual congeners were not given.

The data showed that decaBDE partitioned mainly to the sediment and particulate phase. Based on the known sedimentation rate in the lake and the concentration of decaBDE found in sediment, Bogdal et al. (2007) estimated that the flux of decaBDE to sediment is around 1.7 $\mu g/m^2$ per year. This compares well with the blank-corrected atmospheric deposition data (5.0 ng/m^2 per day or 1.8 $\mu g/m^2$ per year) and suggests

that the sedimentation flux of decaBDE in the lake can be largely explained by atmospheric deposition.

Table 4.20 Partitioning behaviour in Lake Thun.

Compartment	Number of samples	Concentration[a]	
		DecaBDE	Total octa- and nonaBDEs
Surface sediment	1	0.9 µg/kg dry weight {0.05 µg/kg dry weight}	0.06 µg/kg dry weight {<0.01 µg/kg dry weight}
Fish muscle	57	7.2 µg/kg lipid (mean) 3-10 µg/kg lipid (range) {1.9 µg/kg lipid}	<0.1 µg/kg lipid {<0.1 lipid}
Water – suspended particulates	1	1.0 pg/l {<0.1 pg/l}	<0.01 pg/l {<0.01 pg/l}
Water – dissolved phase	1	<0.1 pg/l {<0.1 pg/l}	<0.01 pg/l {<0.01 pg/l}
Atmospheric deposition	1	6.1 ng/m^2 per day {1.1 ng/m^2 per day}	0.2 ng/m^2 per day {0.02 ng/m^2 per day}

Notes: a - Values in parentheses represent the concentrations found in laboratory blanks.

Although Bogdal et al. (2007) concluded that the mean concentration of decaBDE in the fish is low (mean concentration in muscle of 7.2 µg/kg lipid) indicating limited potential for bioaccumulation, it is interesting to note that the levels of decaBDE in the water phase were also low (<0.1 pg/l in the dissolved phase and around 1 pg/l in the particulate phase). Although not included in the Bogdal et al. (2007) paper, it is possible to estimate a tentative bioaccumulation factor for decaBDE from these data. The lipid concentration of the fish was not given, but assuming a lipid content of around 5 per cent, a concentration of 7.2 µg/kg lipid would be equivalent to a whole fish concentration of 0.36 µg/kg fresh weight. Allowing for the concentration of decaBDE found in laboratory blanks, this would imply that the overall bioaccumulation factor for decaBDE is of the order of at least 3×10^5 l/kg. This is a relatively simplistic and worst case interpretation of the data - exposure of the fish to other sources of decaBDE (e.g. in their food) would modify this value.

Vives et al. (2007) investigated the distribution of polybrominated diphenyl ethers in air, bulk deposition, aquatic settling material and sediments from Lake Maggiore (Italy and Switzerland). The concentrations of total polybrominated diphenyl ethers, along with the percentage contribution from decaBDE in the samples, are summarised in Table 4.21. For the air samples, decaBDE was detectable only in the particulate phase and was the predominant congener present in the bulk deposition samples. For the surface sediment samples, two distinct congener distribution patterns were evident. DecaBDE was the predominant congener present (accounting for around 70 per cent of the total) in the samples where the higher concentration of total polybrominated diphenyl ethers was found, but made a much smaller contribution (around 17 per cent of the total) in the samples with lower concentrations of total polybrominated diphenyl ethers. This was thought to reflect local input sources of decaBDE into the lake.

Table 4.21 Distribution of decaBDE in air, atmospheric deposition and sediments from Lake Maggiore.

Sample	Sampling locations	Concentration of total PBDE	Percentage contribution from decaBDE (%)
Air sample – gas phase	Collected from Ispra, 22-30 March 2005.	77 pg/m^3	0%
Air sample – particulate phase	Collected from Ispra, 22-30 March 2005.	29 pg/m^3	16%
Bulk deposition	Collected from Ispra, 22-30 March 2005.	17 ng/m^2 per day	48%
Aquatic settling material	Collected from Ispra Bay, 17 December 2004-19 April 2005. Sample collected 3 m above the sediment at a depth of 27 m.	5.7 µg/kg dry weight	9%
Surface sediments	48 samples collected at depths between 5 and 60 m in 2005. The samples consisted of the top 10 cm of sediment.	5.1 µg/kg dry weight	17% or 70%

Cetin and Odabasi (2007a) investigated the air-water exchange and dry deposition of decaBDE at a coastal site in Izmir Bay, Turkey. The study analysed the levels of polybrominated diphenyl ethers in paired air and water samples collected in July and December 2005. Based on the concentrations found in these samples, the net air-water exchange flux for decaBDE was estimated to be 11.1 ng/m^2 per day indicating that the main net transfer between air (gaseous phase) and water was deposition from air rather than volatilisation from water. Dry deposition was also found to be significant for decaBDE with an estimated dry particulate deposition flux of 116 ng/m^2 per day being estimated for decaBDE.

A further study by Cetin and Odabasi (2007b) investigated the particle-phase dry deposition to soil and air-soil gas exchange at Izmir, Turkey. Concurrent ambient air and particle dry deposition samples were collected at a suburban, urban and industrial area (with a nearby steel plant, power plant, fertilizer factory with other industrial plants within a few kilometres). The samples were collected from the suburban and urban sites in summer 2004 and winter 2005 and the samples from the industrial site were collected in winter 2005 and summer 2005. Soil samples (0-5 cm depth) were also collected from the same areas at the time of air sampling. The mean concentration of decaBDE found in the air samples (gas plus particulate phase) was 16 pg/m^3 at the suburban site, 26 pg/m^3 at the urban site and 47 pg/m^3 at the industrial site. DecaBDE was the dominant polybrominated diphenyl ether congener present in these samples and around 27 per cent of the decaBDE was associated with the gaseous phase and 73 per cent associated with the particulate phase. The particle phase dry deposition flux for decaBDE was measured at the urban and suburban areas and was determined to be 49 ng/m^2 per day at the suburban site and 108 ng/m^2 per day at the urban site (the deposition flux was not measured at the industrial site). The mean levels of total polybrominated diphenyl ethers in the soils were in the range 0.50 to 2,840 µg/kg dry weight (the highest concentration was found in soil at an electronics industry). Using these data, Cetin and Odabasi (2007b) modelled the gas-soil exchange using a fugacity approach. Based on this analysis, it was concluded that the soil concentration was not in equilibrium with the air concentration (the fugacity of the air was higher than that of the soil) and concluded that the atmosphere is a major source of polybrominated diphenyl ethers in the surface soil. It was estimated that dry particle, wet and gaseous deposition contributed around 60 per cent, 32 per cent and 8 per cent respectively to the annual flux of total polybrominated diphenyl ethers to soil.

Overall, the available new data show that decaBDE is expected to be associated mainly with the particulate phases in air and water. Atmospheric deposition (wet and dry deposition of particulates) appears to be an important distribution process for decaBDE. These conclusions are consistent with the previous ESR reports.

A review of methods for estimating the long-range transport of organic chemicals in the environment has been carried out by Scheringer (2008). The review is general in nature but it also considers the implications for modelling the long-range transport of specific substances, including decaBDE. Scheringer (2008) notes that the long-range transport potential of decaBDE is “particularly difficult to understand” as it adsorbs strongly to aerosol particles, undergoes a slow reaction with atmospheric hydroxyl radicals but is potentally degraded rapidly by direct photolysis, and this latter process has rarely been taken into account in modelling work carried out so far for decaBDE. Although a limited transport potential is often predicted because of a predicted fast deposition with aerosol particles, Scheringer (2008) considered that decaBDE could potentially be transported over longer distances (1,000 km or more) during episodic transport events taking place during dry periods.

It should be noted that a global modelling study with decaBDE specifically investigating the influence of direct photolysis on the modelled results has recently been carried out by Schenker et al. (2008a) (see Section 4.3.1).

4.3.3 Bioaccumulation and metabolism

Aquatic organisms

Nyholm et al. (2008a) investigated the maternal transfer of decaBDE in zebra fish (*Danio rerio*). Feed contaminated with various brominated flame retardants (including decaBDE, a heptaBDE (BDE-183), a triBDE (BDE-28) and eight other non-PBDE brominated flame retardants) was prepared by adding a mixture of the brominated flame retardants in ethanol to freeze-dried chironomid feed and allowing the ethanol to evaporate. Two nominal concentrations were prepared, a high dose where the concentration of each brominated flame retardant was 100 nmol/g dry food (equivalent to a decaBDE concentration of 96 mg/kg dry food) and a low dose where the concentration of each brominated flame retardant was 10 nmol/g dry food (equivalent to a decaBDE concentration of 9.6 mg/kg dry food). Groups of 23 males and 23 females were used for each exposure concentration and were fed at a rate of 2 per cent of their body weight per day (to ensure the feed was completely consumed, half the feed was given in the morning and half in the afternoon) for up to 42 days. Fish were sampled on days 0, 3, 7, 14, 28, 35 and 42 of the experiment and eggs were collected on days 0, 2-3, 6-7, 13-14, 27-28, 34-36 and 41-42. The average lipid contents of the fish and eggs during the study were 3.36 per cent and 0.47 per cent respectively. The lipid content of the feed was not given.

DecaBDE was detected in the egg samples during the study and by day 34-36 had reached around 10 nmol/g lipid (~10 mg/kg lipid) in the eggs in the high dose treatment and 1 nmol/g lipid (~1 mg/kg lipid) in the eggs in the low dose treatment (values read from a graph). DecaBDE was also found to be present in the fish (whole body). The actual levels found in the fish were not given in the paper but the ratios of the concentration in eggs (nmol/g lipid) to the concentration in fish (nmol/g lipid) were calculated for each sampling time in the study. The median egg to fish ratio for decaBDE was around two for the high dose group and around five for the low dose treatment (values read from a graph[21]). These ratios were shown to be statistically significantly higher than one (a ratio of one would be expected from fugacity considerations) implying enhanced accumulation in the eggs compared with the adults. A possible reason for this considered by Nyholm et al. (2008a) was that decaBDE may have bound to, and been transported by, lipoproteins that are needed for egg production, or that the eggs have a lower capacity for transformation (metabolism) than the fish. The results for BDE-183 showed that this substance was also taken up in the fish and transferred to eggs. The concentration in the eggs was around 100 nmol/g lipid (~70 mg/kg lipid) in the high dose group and 10 nmol/g lipid (~7 mg/kg lipid) in the low dose group and the ratio of the concentration in eggs to fish (on a lipid normalised basis) was around 1.5 in the high dose group and 2.3 in the low dose group.

It should be noted that, as well as maternal transfer, it is also possible that uptake of decaBDE (and other PBDEs) into the eggs could have occurred via the water phase (or in contact with faeces or uneaten food). Although precautions were taken to limit this possibility (for example it was reported that the bottom of each aquarium was rinsed before eggs were sampled in order to minimize possible contamination, and the eggs were collected directly after spawning and were rinsed with water prior to analysis) such uptake could still have occurred.

Nyholm et al. (2008a) also found that three hexabromodiphenyl ethers accumulated in the eggs and fish during the study (the actual congeners were not identified). For two of these the cumulative exposure via impurities in the spiked food was less than one per cent of the level measured in the fish after 42 days exposure implying that they were metabolically derived in the fish from debromination of higher congeners. However, for the third relatively high amounts were present in the spiked food and this could have accounted for the amount accumulated in the fish. As the food was spiked with both BDE-183 and BDE-209 it cannot be distinguished which of these congeners contributed to the metabolic formation of the hexabromodiphenyl ethers seen.

It is possible to estimate an approximate biomagnification factor (BMF) from the data reported in the Nyholm et al. (2008a) study based on the concentration in the fish (mg/kg wet weight)/concentration in food (mg/kg dry weight). For example in the low exposure group the concentration of decaBDE in the eggs was determined to be around 1 nmol/g lipid (~1 mg/kg lipid) and so, as the ratio of the concentration in eggs to fish was around five, the concentration in fish would have been around 0.2 mg/kg lipid. The mean lipid content of the fish was 3.36 per cent and so the concentration in fish on a wet weight basis would have been around 7×10^{-3} mg/kg wet weight. Thus the

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[21] The error bars on the graph indicated that there was considerable variability in the ratios obtained. A possible reason for this is that the eggs and fish analysed each day were not paired as female and offspring (one tank was used to house the fish in each exposure level). Also, it was reported that the average recovery of a ^{13}C-labelled decaBDE internal standard was low and variable in the fish and egg samples (mean±standard deviation was 47± 24 per cent) which could have added to the variability seen in the ratios.

BMF for the fish in the low dose group can be estimated as approximately $7\times10^{-3}/9.6 = 7\times10^{-4}$. The equivalent calculation for the high dose group for decaBDE would lead to a BMF of around 2×10^{-3}. For comparison the BMF that would be calculated from these data for BDE-183 would be around 0.01 for the low dose group and 0.02 for the high dose group. There is no indication in the paper as to whether steady state was reached over the 42 day exposure period (data are shown for BDE-28 that shows that this substance was approaching steady state).

A second study by Nyholm et al. (2008b) using a similar exposure method has looked further at the uptake and biotransformation of decaBDE in male zebrafish (*Danio rerio*). In this study groups of fish were exposed to the same mixture of brominated flame retardants as before via diet at nominal concentrations of either 1 or 100 nmol/g food for each component (this is equivalent to a decaBDE concentration of either 0.96 mg/kg dry weight or 96 mg/kg dry weight in the food). The fish were fed the contaminated diet at a rate of 2 per cent of the body weight (the fish had an initial weight of 500 mg) for a total of 42 days, which was followed by a 14-day elimination period where the fish were fed an uncontaminated diet. Fish were analysed for the presence of decaBDE and possible metabolites on days 0, 3, 7, 14, 28, 35 and 42 of the uptake period and days 7 and 14 of the elimination period (the fish were sampled 24 hours after feeding). A composite sample of two whole fish was analysed on each occasion. DecaBDE was found to be present in fish exposed to the high-dose feed, but the uptake efficiency was found to be very low at around 0.62 per cent. Detectable amounts of decaBDE were still present in the fish at the end of the elimination period and the elimination half-life was estimated to be 6.5 days. The actual concentration reached in the fish is shown graphically in the paper. In the high dose group the concentration of decaBDE reached around 0.08 nmol/g wet weight in the fish after 42 days exposure and the plot indicated that steady state was being approached after this time. The measured concentration of decaBDE in the food for this group was 72 nmol/g dry weight. Using these data it is possible to estimate the BMF for decaBDE (as the ratio of the concentration in fish to the concentration in food) as 0.0011.

Several lower brominated congeners, for example hexaBDEs, were also detectable in the exposed fish. However, as the fish were exposed to a mixture of brominated flame retardants, including a heptaBDE, it is not possible to ascribe the presence of these lower congeners solely to exposure to decaBDE.

A study investigating the uptake and accumulation of decaBDE in zebra mussels (*Dreissena polymorpha*) has been undertaken by Riva et al. (2007) as part of an evaluation of the potential for decaBDE to induce DNA damage. The toxicological results from this study are summarised in Section 5. The mussels for the study were collected from Lake Maggiore, Lake Garda, Lake Como, Lake Iseo and Lake Lugano and were acclimated to laboratory conditions by maintaining in dechlorinated tap water at 20°C for 21 days (with daily renewal of the water to facilitate depuration). One week prior to the start of the accumulation study, the mussels were placed on sheets of glass suspended in fifteen litre glass tanks and only animals that were able to re-attach themselves by the byssus were kept for the study (around 150 mussels per treatment).

The decaBDE used in the test had a purity of 98% (the major impurities were two nonaBDE congeners; the identities were not given). The substance was firstly dissolved in a 9:1 volume/volume mixture of isooctane/toluene to give a concentration of 10 mg/l. This solution was then dissolved in dimethylsulfoxide to give a stock solution with a decaBDE concentration of 1 mg/l. This was added to the tanks containing the mussels to give nominal concentrations of 0.1, 2 and 10 µg/l. A control and solvent control tank were also run. The water in the exposure tanks was renewed daily and the mussels were fed with green alga two hours prior to each renewal. During the exposure period (up to 168 hours) the tanks were screened against direct sunlight in order to avoid possible photodegradation of decaBDE.

The levels of decaBDE present in the mussel soft tissues were measured after 48, 96 and 168 hours exposure. The mussels were collected approximately 24 hours after the last addition of food in order to allow sufficient time for depuration of any particulate matter present in the gastro-intestinal tract prior to analysis. The levels of decaBDE found in the mussels were displayed graphically in the paper but the levels appeared to be reasonably constant from 48 hours exposure onwards. The approximate levels found in the mussels were ~32,000 µg/kg lipid in the 10 µg/l treatment group, ~1,000 µg/kg lipid to ~1,500 µg/kg lipid in the 2 µg/l treatment group and ~100 to ~1,000 µg/kg lipid in the 0.1 µg/l treatment group (values read from graph). Based on these data, the mussel bioconcentration factor (BCF)/bioaccumulation factor (BAF) can be estimated at approximately 3,200 l/kg lipid for the 10 µg/l treatment group, 500 to 750 l/kg lipid for the 2 µg/l treatment group and 1,000 to 10,000 l/kg lipid for the 0.1 µg/l treatment group. A further study was carried out exposing mussels to a nominal concentration of 2 µg/l for eleven days. Here the concentration in the mussels reached 20,000 µg/kg lipid, which is equivalent to a BCF/BAF of 10,000 l/kg lipid. The lipid contents of the mussels were not given and so it is not possible to estimate the BCF/BAF on a mussel wet weight basis but these would be lower than the lipid based BCF/BAF values (for example if the lipid content was 5-10 per cent the BCF of 10,000 l/kg lipid would be equivalent to a wet weight BCF/BAF of 500-1,000 l/kg).

It should be noted that the concentrations in water were not determined analytically and may have exceeded the actual water solubility of decaBDE in the test medium (the exact water solubility of decaBDE is unclear but is likely to be <0.1 µg/l (EC, 2002)). This has some important implications for the interpretation of the results for this study. For example, if undissolved test material was present it is possible that the mussels could have directly ingested this material constantly during the study or alternatively if it is considered that the mussels were exposed solely via the dissolved phase then the actual exposure concentrations could have been much lower than indicated by the nominal concentrations, and hence the actual BCF/BAF could be higher than estimated above[22]. Overall these data are suggestive that the BCF/BAF for decaBDE in mussels may be of the order of 1,000 l/kg or above.

As well as decaBDE, Riva et al. (2007) indicate that there was also evidence for the presence of lower brominated congeners in the mussels after 168 hours exposure. These were not determined quantitatively but they were thought to be three heptaBDEs, three octaBDEs and three nonaBDEs.

Jenssen et al. (2007) studied decaBDE bioaccumulation in north-east Atlantic marine ecosystems. The study uses some of the monitoring data obtained by Sørmo et al. (2006) (discussed in detail in ECB (2007)). Sampling occurred along a latitudinal gradient from southern Norway to the Arctic and included Oslofjord (59°N), Froan (64°N), Bear Island (74°N) and Spitsbergen (>78°N). The samples analysed at each location included calanoid species, Atlantic cod (*Gadus morhua*), polar cod (Borreogadus), harbour seal (*Phoca vitulina*) blubber, ringed seal (*Phoca hispida*) blubber, Common Tern (*Sterna hirundo*) eggs and Arctic Tern (*Sterna paradisaea*) eggs. Not all species were collected at all locations. The samples were collected in 2003 except for blubber samples from harbour seals from Oslofjord (collected in 1998). The levels of decaBDE found in the various species are summarised in Table 4.22.

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[22] BSEF (2009) also indicated that no control mussels appear to have been evaluated at each time point. In addition, procedural blanks included in the analytical procedure also appeared to show the presence of decaBDE. This adds further uncertainty to the values estimated.

Table 4.22 Accumulation of decaBDE in marine ecosystems.

Location	Species	Number of samples	Mean concentration of decaBDE (± standard deviation) (µg/kg lipid)
Oslofjord	*Calanas* sp.	3	0.58±0.29
	Atlantic cod	21	0.64±0.66
	Harbour seal blubber		No data
	Common tern eggs	10	0.26±0.29
Froan	*Calanus finmarchicus*		No data
	Atlantic cod	9	0.64±0.92
	Harbour seal blubber	6	0.11±0.07
	Arctic tern eggs	10	0.22±0.13
Bear Island	Polar cod		No data
Spitsbergen	*Calanus glacialis*		No data
	Polar cod[a]	5	0.27±0.08
	Harbour seal	5	0.59±0.54
	Ringed seal		No data
	Arctic tern	10	0.84±0.89

Notes: a - The data are taken from Sørmo et al. (2006) but the mean has been recalculated based on specimens with levels above the detection limit.

Based on these data, Jenssen et al. (2007) considered the biomagnification potential for decaBDE in a simple cod-harbour seal food chain. The biomagnification factor (BMF) was defined as the mean concentration in seal (µg/kg lipid)/mean concentration in cod (µg/kg lipid). The following BMFs were determined for decaBDE.

Froan	BMF = 0.2
Spitsbergen	BMF = 2.2

There are a number of factors that need to be considered when considering the results of this study. Firstly, the number of samples analysed for some species is relatively low and so the representivity of the data is unclear. Secondly, for a number of species the paper indicates that there are “no data” on the concentration of decaBDE but it is not entirely clear whether this means that decaBDE was not detected in the samples or whether the samples were not analysed for decaBDE. Overall, given that the analysis resulted in one BMF >1 and one BMF <1, the study is inconclusive as to the biomagnification potential of decaBDE

Tomy et al. (2008) investigated the bioaccumulation of decaBDE in an Arctic marine food web in Eastern Canada. The organisms included in the study and the levels of decaBDE found are summarised in Table 4.23. The quality assurance and quality control procedures used in analytical method included routine analysis of both procedural blanks and instrument blanks, along with analysis of a standard reference material (beluga blubber sample). DecaBDE was found to be present in the procedural blanks in low amounts and the concentrations found in the samples were all corrected for this.

Table 4.23 Accumulation of decaBDE in an Arctic marine food web in Eastern Canada.

Sample	Number of samples	Sampling location and date	Mean lipid content (%)	$\delta^{15}N$	Trophic level[a]	Analytical detection limit (µg/kg lipid)	Concentration of decaBDE (µg/kg lipid)
Zooplankton (predominantly copepods)	5	Frobisher Bay, 2002.	3.69	10.06	2.5	0.0350	18.7 (mean) 8.9-36.9 (range)
Shrimp (*Pandalus borealis* and *Hymenodora glacialis*)	8	Davis Strait, 2000 and 2001	7.50	12.24	3.0	0.0876	4.5 (mean) 1.1-17.6 (range)
Clams (*Mya truncate* and *Serripes groenlandica*)	5	Frobisher Bay, 2002.	1.01	8.12	2.0	0.0381	5.9 (mean) 0.9-18.9 (range)
Arctic cod (*Boreogadus saida*) - whole body	8	Davis Strait, 2000 and 2001.	2.09	14.19	3.6	0.0735	13.0 (mean) 1.1-84.2 (range)
Deepwater redfish (*Sebastes mentella*)	5	Davis Strait, 2000 and 2001	1.81	12.39	3.1	0.0514	3.6 (mean) 2.9-3.7 (range)
Beluga whale (*Delphinapterus leucas*) - blubber	5	Frobisher Bay near Iqaluit, 1996.	92.32	15.32	3.8	0.470	<0.47 (not detected)
Narwhal (*Monodon monoceros*) -blubber	5	Broughton Island, 2000.	82.96	19.03	4.8	0.397	<0.40 (not detected)
Walrus (*Odebenus rosmarus*) - blubber	5	Cape Dorset, 1998.	88.79	11.72	2.9	0.430	<0.43 (not detected)

Note: a - The trophic levels were shown graphically in Tomy et al. (2008). The values given here are approximate (read from the graphs).

The trophic level of the organisms were determined based on $\delta^{15}N$ as defined in an earlier paper by Tomy et al. (2004b). The actual values for the trophic level of each species are only given graphically in the Tomy et al. (2008) paper. The highest concentrations of decaBDE (on a lipid normalised basis) were found in zooplankton and the concentrations were found to generally decrease with increasing trophic level. A plot of ln [lipid normalised concentration] against trophic level gave the following statistically significant ($P = 0.002$; correlation coefficient $R^2 = 0.25$) relationship and a trophic magnification factor of 0.3 was estimated for decaBDE from the data.

$$\ln[\text{concentration of decaBDE}] = -1.21 \times \text{Trophic level} + 4.87$$

Overall Tomy et al. (2008) concluded that the concentrations of decaBDE were decreasing with increasing trophic level in this food chain (i.e. trophic dilution was occurring). A possible explanation for this put forward by Tomy et al. (2008) was that the capacity for metabolism of decaBDE by organisms increases with increasing trophic level. However, other alternative explanations exist. For example, the dietary assimilation efficiencies could decrease (see discussion of bioaccumulation data below). It should also be noted that the organisms included in the study were collected from different locations at different times, and this adds some uncertainty to the conclusions that can be drawn.

In a further study, Tomy et al. (2009) investigated the bioaccumulation of decaBDE in an Arctic marine food web in Western Canada. The results from the study are summarised in Table 4.24. The quality assurance and quality control procedures were the same as for the Tomy et al. (2008) study.

Using these data Tomy et al. (2009) found no positive correlation between the concentration of decaBDE and trophic level for the Western Canadian Arctic marine food web and concluded that the trophic magnification factor (TMF) was <1 for decaBDE in this food chain. Tomy et al. (2009) also estimate BMF values (on a lipid weight basis) for various combinations of predator:prey. The BMF values determined for decaBDE were 0.3 for ring seal blubber:cod liver, 0.3 for beluga blubber:cod liver, 0.9 for beluga blubber:herring liver, 0.03 for beluga blubber: Arctic cisco liver, 12.7 for cod liver:*Calanus* sp. whole body and 4.8 for cod liver:*Thermisto* sp. whole body. The significance of these BMF values, given that comparison is made between different combinations of liver, blubber and whole body, is not clear.

Table 4.24 Accumulation of decaBDE in an Arctic marine food web in Western Canada.

Sample	Number of samples	Sampling location and date	Mean lipid content (%)	δ^{15}N	Concentration of decaBDE (µg/kg lipid)[a]
Arctic copepod (*Calanus hyperboreus*)	2	Amundsen Gulf, 2004	0.31	9.3	0.04 (composite sample analysed)
Pelagic amphipod (*Themisto libellula*)	1	Eastern Beafort Sea, 2005	0.07	10.2	0.11
Arctic cisco (*Coregonus autumnalis*)	9	Mackenzie Estuary, 2005	1.94	12.3	7.23 (median) 2.05-21.67 (range) (detected in 100% of samples)
Pacific herring (*Clupea pallasi*)	10	Mackenzie Shelf, 2005	6.42	12.7	0.25 (median) 0.04-0.70 (range) (detected in 70% of samples)
Arctic cod	9	Amundsen Glulf, 2004/5	1.62	14.9	1.01 (median) 0.23-1.77 (range) (detected in 100% of samples)
Ringed seal	8	Holman Island, 2004	86.11	15.6	not detected
Beluga	10	Hendrickson Island, 2007	81.75	16.5	0.23 (median) 0.23-1.13 (range) (detected in 20% of samples)

Note: a - Where not detected, the median was calculated using a value of ½ of the detection limit.

A study on the bioaccumulation of decaBDE in organisms downstream of a waste water treatment plant on Gaobeidian Lake, Beijing, China has been carried out by Wang et al. (2007). The waste water treatment plant treated around one million tonnes of waste water per day (eighty per cent was from municipal sources) and discharged thirty per cent of the effluent directly into the lake and the remaining effluent was used as cooling water for a nearby power plant before being discharged to the lake (resulting in temperature of the lake water being above 30°C from May to October). For the study, samples of effluent and lake water were collected in December 2006 and organisms from the aquatic food web were sampled in September 2006. The organisms included spirogyra and March brown, coccid, zooplankton (*Monia rectirostris*, *Monia micrura* and *Monia macrocopa*), fish (common carp (*Cyprinus carpio*), crucian carp (*Carassius auratus*), leather catfish (*Silurus meridionalis*) and java tilapia (*Tilapia nilotica*)) and Chinese soft-shell turtle (*Chinemys reevesii*). In addition a sediment core was collected both upstream and downstream of the outfall from the waste water treatment plant.

The samples collected were analysed for decaBDE. The paper indicates that strict quality controls were used to ensure correct identification and accurate quantification in the analysis and included the analysis of method blanks, etc. However the paper only gives details of the response in the method blanks for lower brominated congeners and not decaBDE. The method limit of detection for decaBDE was 1 µg/kg. The levels of decaBDE found in the various samples are summarised in Table 4.25.

Table 4.25 Distribution of decaBDE in a Gaobeidian Lake ecosystem.

Sample	Concentration of decaBDE
Effluent from the waste water treatment plant (particulates plus dissolved)	<1 µg/l[a]
Lake water (particulates plus dissolved)	<1 µg/l[a]
Sediment cores	237 µg/kg dry weight (mean)[d]
	<1 to 743 µg/kg dry weight (range)[d]
Lichen/spirogyra[e]	1,572 µg/kg dry weight
March brown[f]	11.4 µg/kg dry weight
Coccid[g]	114 µg/kg dry weight
Zooplankton	152 µg/kg dry weight
Common carp (four samples)	5.0 µg/kg[b]
	[135 µg/kg lipid[c]]
Java Tilapia (three samples)	<1 µg/kg (not detected)
Leather catfish (four samples)	19.3 µg/kg (mean)
	<1-61 µg/kg (range)
	[90 µg/kg lipid (mean)]
Crucian carp (five samples)	1.8 µg/kg (mean)
	<1-29 µg/kg (range)
	[12 µg/kg lipid (mean)]
Chinese soft-shell turtle (three samples)	2.6 µg/kg (mean)
	<1-6.7 µg/kg (range)
	[84 µg/kg lipid (mean)]

Notes: a - The paper indicates that this was less than the limit of detection. The limit of detection was given in the paper as 1 µg/kg. For the effluent and water samples it is assumed here that the limit of detection was 1 µg/l.

b - The paper indicates that decaBDE was detected in only one sample; the other samples were less than the limit of detection.

c - The paper gives the levels as µg/kg OC. The term OC is usually used for “organic carbon”. In this case it is assumed that it relates to the lipid content of the fish.

d - The values given appear to relate to the concentrations found over all depths in both sediment cores (the sediment cores were collected to a total depth of 36 cm).

e - In one table in the paper this is referred to as lichen but elsewhere it is indicated to be spirogyra.

f - The species identity is not entirely clear from the paper. The paper appears to give a Latin name of *Limnodrilus hoffmeisteri* which is an oligochaete worm. However the name “March Brown” appears to relate to the fly *Rithrogena germanica*.

g - The actual species is unclear. The term Coccid is generally used for various insects of the superfamily *Coccoidea*, including the scale insects and mealybugs

DecaBDE was not detectable in either the effluent samples or the lake water samples but was present in the sediment cores at a mean concentration of 237 µg/kg dry weight (this appears to relate to the mean concentration across all layers of both the sediment cores (down to a depth of thirty six centimetres)). The upper layers (topmost twelve centimetres) had the highest concentrations. Based on the sediment core data, the inventory of decaBDE in the lake was estimated at around 1,150 ng/cm^2.

For the aquatic species, the highest concentrations of decaBDE were found in spirogyra, March brown, coccid and zooplankton with lower levels being found in the fish and turtles. In order to determine whether biomagnification was occurring in the food chain, Wang et al. (2007) determined the trophic level of each species based on nitrogen isotope ratios. Based on this analysis it was determined that java tilapia were at the highest trophic level and no indication for biomagnification was found in the available data.

The study also determined the levels of lower brominated polybrominated diphenyl ether congeners (tri- to heptaBDEs). For these congeners a linear relationship was found between the bioaccumulation factor in fish and the number of bromine atoms in the molecule as follows.

$$\log BAF = 0.65 - 0.38 \times N_{Br} \qquad R^2 = 0.439, P < 0.05$$

where BAF = bioaccumulation factor (l/kg); it is not entirely clear from the paper whether these are estimated on the lipid normalised data or the whole body weight data.

N_{Br} = number of bromine atoms per molecule.

R = correlation coefficient.

P = level of statistical significance.

Using this relationship, the following BAFs can be estimated for the more highly brominated congeners (assuming that the relationship still holds at higher bromine contents).

Congener	**BAF (l/kg)**
HeptaBDE	6,900
OctaBDE	2,900
NonaBDE	1,200
DecaBDE	500

Biota-sediment accumulation factors (BSAFs) have been determined for decaBDE based on the levels found in samples from the Pearl River estuary in South China (Xiang et al. 2007). The biota and sediment samples were collected in April and August 2004 and included three benthic invertebrate species (jinga shrimp (*Metapenaeus affinis*), greasy-back shrimp (*Metapenaeus ensis*) and mantis shrimp (*Oratosquilla oratoria*)), five fish species (large yellow croaker (*Pseudosiaena crocea*), silvery pomfret (*Pampus argenteus*), flathead fish (*Platycephalus indicus*), robust tonguefish (*Cynoglossus robustus*) and Bombay duck (*Harpodon nehereus*)) along with surface sediment samples (top 5 cm). The muscle fillets from the fish and the soft part (edible part) of the shrimp were used for the analysis. The fish samples analyzed included both individual fish and pooled samples (of four or five similar sized fish) and the shrimp were pooled samples of five individuals. In all a total of 86 biota samples were analysed. The quality control/quality assurance procedures used during the analysis included routine analysis of procedural blanks and spiked blank and matrix samples. The detection limit for decaBDE was 0.016 µg/kg lipid and decaBDE was found to be present in only one procedural blank sample at a non-quantifiable level. The levels of decaBDE found in the various samples of biota and sediment are summarised in Table 4.26.

For the fish included in the study, decaBDE was generally detectable more frequently, and at a higher median concentration, in the large yellow croaker and silvery pomfret. The trophic levels of the fish species were determined using $\delta^{15}N$ measurements (Xiang et al. (2007) considered that $\delta^{15}N$ values increase by approximately 3.4‰ for each unit increase in trophic level). These showed that the large yellow croaker, flathead fish and Bombay duck occupied approximately the same trophic level. The $\delta^{15}N$ values for silvery pomfret were found to be variable (possibly as a result of differences in the sizes of fish used) but the mean $\delta^{15}N$ put this species at a higher trophic level than the other species. Based on this analysis, no evidence for increasing levels of decaBDE in increasing trophic level was apparent.

Xiang et al. (2007) also determined the BSAF for decaBDE in the species (defined as the lipid normalised concentration in biota/organic carbon normalised concentration in sediment). The median BSAF for decaBDE was given as zero (presumably some of the values were too low to be determined reliably), with a maximum value of around 0.4.

Further BSAF values for decaBDE have been determined from field data by Viganò et al. (2009). In this study samples of gammarids (mainly *Echinogammarus stammeri*), caddisflies (mainly of the family Hydropsychidae) and sediments were collected from the River Po, Italy and analysed for the presence of decaBDE. The samples were collected both upstream and downstream of a polluted tributary in both 2003 and 2005 The sediments analysed were the <63 µm fraction. The analytical method used included routine analysis of procedural blanks (no decaBDE was detected in these blank samples) and the detection limit for decaBDE was 0.5 µg/kg dry weight. The highest levels of decaBDE found in the sediments were up to 7,353 µg/kg dry weight in 2003 and up to 64,042 µg/kg dry weight in 2005. DecaBDE was also detectable in the samples of gammarids (up to around 180 µg/kg lipid) and caddisflies (up to around 340 µg/kg lipid) and BSAFs of the order of 0.01-0.02 on a lipid/organic carbon normalised basis were determined for decaBDE. It should be noted that the organisms were not allowed to purge their gut contents prior to analysis and so the gut contents may have contributed to the levels found in the organisms. However the levels in the organisms were found to correlate with the lipid content of the organisms.

Very low BSAF values for decaBDE have been determined for mud snails (*Cipangopaludina cahayensis*) in a study by Yang et al. (2009). Samples of snails and sediments were collected from an electronic waste recycling area in South China in November 2006. The analytical method used included regular analysis of procedural blanks, solvent blanks and standards and the detection limit of decaBDE was set at 0.1 µg/kg dry weight (based on the mean procedural blank level plus three times the standard deviation). The levels of decaBDE in sediment at the four sites sampled were 14.7, 2,934, 896 and 3,510 µg/kg dry weight and the levels of decaBDE found in the snails at the same four sites were not detectable, 1.2, 2.5 and 9.5 µg/kg dry weight respectively. The BSAF determined was in the range 0.0002 to 0.0019 (on a lipid normalised/organic carbon normalised basis) at the three sites where calculation was possible. It is not clear from the paper whether the gut contents of the snails were purged prior to analysis.

A modelling study of the fate of polybrominated diphenyl ethers in juvenile lake trout (*Salvelinus namaycush*) has been undertaken by Bhavsar et al. (2008a). The model used in the study was a non-steady-state (time dependent) multi-chemical fugacity-based fish model and was parameterised to correspond to the feeding rates, fish sizes, lipid contents, etc., in the bioaccumulation study carried out by Tomy et al. (2004a) that is discussed in detail in ECB (2004). The model was then used to back-calculate the concentrations of the various polybrominated diphenyl ethers measured in the Tomy et al. (2004a) study by optimizing the values for metabolic half-life and gut absorption efficiency for each congener that led to the best agreement between the modelled and experimental data for both the uptake and depuration phase. The model assumed that certain lower brominated congeners could be formed from higher brominated congeners by debromination (for example it was assumed that decaBDE could be biotransformed to nonaBDEs). A total of thirteen polybrominated diphenyl ethers were considered in the study. The results of the exercise found that a metabolic half-life and a gut absorption efficiency (or assimilation efficiency) of 1,000 hours and 35 per cent respectively gave the best fit of the data for decaBDE. It should be noted that the model assumes that biotransformation occurred mainly in the fish body and that biotransformation in the gut was assumed to be insignificant (this assumption would affect the concentrations predicted by the model).

It should be noted that in the Tomy et al. (2004a) study the measured assimilation efficiency for decaBDE was 5.2 per cent (although a lower value of 0.3 per cent was also given in this paper). Bhavsar et al. (2008a) indicates that the value determined by Tomy et al. (2004a) does not take into account biotransformation of decaBDE into lower brominated congeners and hence a higher value of 35 per cent is estimated by the model here. The relevance of the model estimates is not clear.

Bhavsar et al. (2008b) [ABST] have used the model described above to investigate the potential biotransformation of decaBDE into lower brominated congeners over extended periods (for example over 10 to 15 years). The model was run assuming two dietary concentrations of decaBDE (3.4 and 27.5 µg/kg wet weight respectively). Two possible food consumption rates were considered: the first assumed that the food consumption rate varied with the size of the fish (based on published information) and the second assumed a constant feeding rate of 3 per cent of the body weight over the entire period.

Table 4.26 Levels of decaBDE in sediment and biota from the Pearl River estuary, China.

Sample	Number of samples	Lipid content (%)	$\delta^{15}N$ (‰)	Feeding/living habitat	Detection rate[a] (%)	Concentration of decaBDE (µg/kg lipid (biota) or µg/kg organic carbon (sediment))
Sediment	9					1,372 (median) 792-4,173 (range)
Mantis shrimp	9	0.98-3.59		Benthic invertebrate	66.7	42.5 (median) <0.016-88.5 (range)
Greasy-back shrimp	10	0.47-5.15		Benthic invertebrate	60.0	30.0 (median) <0.016-405 (range)
Jinga shrimp	10	0.99-2.81		Benthic invertebrate	30.0	0 (median) <0.016-556 (range)
Large yellow croaker	13	0.77-1.83	15.8 (mean) 14.6-17.7 (range)	Omnivorous	69.2	117 (median) <0.016-532 (range)
Silvery pomfret	10	1.46-11.04	21.8 (mean) 13.2-24.8 (range)	Omnivorous	60.0	24.4 (median) <0.016-624 (range)
Flathead fish	17	0.28-1.19	16.0 (mean) 13.6-18.7 (range)	Benthivorous	17.6	0 (median) <0.016-38.4 (range)
Robust tongue fish	8	0.51-1.23	13.5 (mean) 12.6-15.0 (range)	Benthivorous	25.0	0 (median) <0.016-273 (range)
Bombay duck	9	0.77-3.03	16.8 (mean) 14.3-18.4 (range)	Piscivorous	44.4	0 (median) <0.016-150 (range)

Notes: a - Percentage of samples in which decaBDE was detectable.

Assuming a variable food consumption rate the model predicted that the decaBDE concentration in the fish would reach around 30 µg/kg lipid (for the low dose) or 240 µg/kg lipid (for the high dose) after around 300 days. The model predicted that the concentrations would then decrease by around 65 per cent over fifteen years mainly due to changes in the consumption rate relative to the fish growth rate. Using a constant feeding rate the model predicted that a steady state concentration in the fish of around 425 µg/kg lipid would result for the high dose after around three to four years.

The modelling results also suggested that debromination of decaBDE into nona-, octa-, hepta- and hexaBDEs (but not penta- or tetraBDEs) may occur in the juvenile fish. However over longer periods (up to fifteen years), the model predicted that small amounts of pentaBDEs (e.g. BDE-99) and tetraBDEs (e.g. BDE-47) may be formed, reaching steady state concentrations of 3 and 23 µg/kg lipid at the low and high dose respectively for BDE-99 (similar concentrations were predicted for BDE-100) and 1 and 8 µg/kg lipid at the low and high dose respectively for BDE-47. Again the relevance of the model estimates to real scenarios is not clear as the model has effectively been used to predict what might happen in a prolonged feeding study (up to fifteen years). In particular, the concentrations predicted to occur in the fish will depend on the dietary concentration of decaBDE assumed in the model and so it is not possible to compare these concentrations directly with concentrations actually found in wildlife.

La Guardia et al. (2007) provided evidence that metabolic debromination of decaBDE was occurring in fish in the field. The study analysed the concentrations of polybrominated diphenyl ethers (triBDEs to decaBDE) in samples of sewage sludge (activated sludge) from an industrial waste water treatment plant (plastics manufacturer in the United States) along with samples of surface sediment and biota from the receiving water system. Samples were collected in both November 2002 and November 2005. The biota samples included chub (*Semolilus atromaculatus*; six samples in 2002), crayfish (*Cambarus puncticambarus*; five samples in 2002) and sunfish (*Lepomis gibbosus*; thirteen samples in 2002 and twenty two samples in 2005) and a single composite sample of each species was analysed. The fish and crayfish were kept in holding tanks for 72 hours prior to analysis to allow depuration of gut contents to occur. The levels of polybrominated diphenyl ethers found in the samples are summarised in Table 4.27 (the study also included several tri- to hexabromodiphenyl ether congeners but these are not shown in Table 4.27).

The pattern of lower brominated congeners found in the sludge samples resembled closely that in the commercial pentabromodiphenyl ether product and the authors concluded that this suggested that debromination of decaBDE during the waste water treatment process was unlikely to be a major process contributing to the levels of lower brominated congeners found in the sludge. The congener profile in the sediment downstream of the waste water treatment plant was broadly similar to that in the sludge and it was concluded that minimal debromination of decaBDE was also occurring in the sediment.

For the biota samples, a total of twenty three polybrominated diphenyl ethers were detected, with the congeners BDE-47 (tetraBDE), BDE-153 (hexaBDE), BDE-196, BDE-201, BDE-202, BDE-203 (octaBDEs) and BDE-206, BDE-207 and BDE-208 (nonaBDEs) being detected in all of the samples. DecaBDE was detectable in crayfish and one of the sunfish samples but was not detectable in chub.

Table 4.27 Distribution of polybrominated diphenyl ethers in sediment and biota downstream of a waste water treatment plant.

Sample		Concentrations							
		DecaBDE	NonaBDEs			OctaBDEs			HeptaBDEs
		BDE-209	BDE-208	BDE-207	BDE-206	BDE-203	BDE-197	BDE-196	BDE-183
November 2002									
Sewage sludge (µg/kg dry weight)		58,800	726	1,340	27,400	1,190	993	1,600	310
Sediment (µg/kg organic carbon)	0.2 km upstream	36,800	not detected	not detected	not detected	not detected	not detected	not detected	not detected
	0 km (15 m) downstream	1,630,000	3,530	5,810	67,700	434	not detected	380	not detected
	1.3 km downstream	3,150,000	not detected	6,660	84,000	386	not detected	771	not detected
	5.6 km downstream	642,000	577	2,630	24,300	322	156	620	249
	10.8 km downstream	300,000	not detected	945	10,900	166	73	210	125
Chub (µg/kg lipid)		not detected	103	79	94	117	not detected	45	not detected
Crayfish (µg/kg lipid)		21,600	143	1,920	2,650	132	43	200	not detected
Sunfish (µg/kg lipid)		2,880	201	276	411	74	193	65	83
November 2005									
Sewage sludge (µg/kg dry weight)		37,400	295	276	1,490	220	171	202	89

Sample		**Concentrations**							
		DecaBDE	**NonaBDEs**			**OctaBDEs**			**HeptaBDEs**
		BDE-209	**BDE-208**	**BDE-207**	**BDE-206**	**BDE-203**	**BDE-197**	**BDE-196**	**BDE-183**
Sediment (µg/kg organic carbon)	0.2 km upstream	33,300	not detected	not detected	not detected	not detected	not detected	not detected	not detected
	0 km (15 m) downstream	181,000	not detected	not detected	11,200	not detected	not detected	not detected	not detected
	1.3 km downstream	2,310,000	not detected	not detected	31,700	not detected	not detected	not detected	not detected
	5.6 km downstream	2,390,000	1,690	6,520	35,500	553	388	1,120	not detected
	10.8 km downstream	247,000	375	544	3,120	not detected	not detected	not detected	not detected
Sunfish (µg/kg lipid)		not detected	67	73	133	20	86	28	77

The congener profile in the chub was of interest as it showed some differences with the profiles found in the other species. For example no BDE-99 (a pentaBDE) or BDE-183 (a heptaBDE) were found in chub even though these were present in both the sediments and other species in the same area. La Guardia et al. (2007) indicated that a similar congener profile had been reported previously for common carp (*Cyprinus carpio*) in a study by Hale et al. (2001). Stapleton et al. (2004) had previously carried out a study exposing common carp to BDE-99 and BDE-183 via the diet and found that carp could metabolise these substances to BDE-47 (a tetraBDE) and BDE-154 (a hexaBDE) respectively. As chub and common carp belong to the same family (*Cyprinidae*) La Guardia et al. (2007) speculated that a similar metabolic route as found for carp may be occurring in chub, and so species-specific differences in metabolic capacity may explain the differences in the congener patters between the various species.

La Guardia et al. (2007) also concluded that the congener profiles found provide some evidence for metabolic debromination of decaBDE. For example, although only the sunfish from 2002 contained detectable amounts of decaBDE, both the chub and sunfish samples contained detectable amounts of two octaBDEs (BDE-201 and BDE-202) and three heptaBDEs (BDE-179, BDE-184 and BDE-188) that were not detectable in either the sludge or sediment samples. It is therefore possible that the presence of these congeners is the result of metabolic debromination of decaBDE (or nonaBDEs).

This hypothesis is further strengthened by the results of the dietary study exposing common carp and rainbow trout to decaBDE that was carried out by Stapleton et al. (2006) (this study is considered in detail in ECB (2007)). In these studies, no decaBDE was detectable in carp after 60 days exposure but there were indications of the presence of BDE-202, BDE-188 and BDE-179 (amongst other congeners) suggesting that carp can metabolise decaBDE to these congeners. In the trout study, decaBDE was detectable in the fish after 112 days exposure, along with several heptaBDEs (including BDE-179, BDE-183, BDE-184 and BDE-188) and octaBDEs (including BDE-196, BDE-201, BDE-202, BDE-203 and BDE-204/197). The metabolic debromination of decaBDE was confirmed by Stapleton et al. (2006) using both trout and carp liver microsomes. La Guardia et al. (2007) noted that the congener patterns seen in trout in the dietary exposure study was similar to that seen in sunfish and the pattern seen in carp in the dietary exposure study was similar to that seen in chub.

Another study of the congener distribution pattern in fish has been carried out by Labandeira et al. (2007). In this study, samples of sediment and carp (*Cyrpinus carpio*) were collected from the Anoia and Cardener Rivers (tributaries of the Llobregat River) in Spain. The samples were collected upstream (1 km) and downstream (23 and 27 km) of an industrial town (with textile and tannery industries) on the Anoia River and upstream (5 km) and downstream (4 and 8 km) of an industrial town (with a rubber industry) on the Cardener River. A total of four sediment samples (one sample from all three sites on the Anoia River and one downstream site on the Cardener River) and 43 carp (26 from the sites on the Anoia River and 17 from the sites on the Cardener River) were collected during 2000. The samples were analysed for the presence of decaBDE along with several lower brominated congeners (mono- to heptaBDEs). The quality assurance/quality control procedures included the routine analysis of procedural blanks (no polybrominated diphenyl ethers were reported to be detected in these).

Of the four sediment samples, only three samples (from the Anoia River) were analysed for the presence of decaBDE. In these samples, decaBDE was not detectable (<0.05 μg/kg dry weight) in the upstream samples and was present at concentrations of 4.6 and 4.0 µg/kg dry weight in the two downstream samples. DecaBDE was not detectable in any of the fish muscle samples (<0.74 µg/kg lipid). This indicates a low accumulation potential from decaBDE from sediment in carp.

Labandeira et al. (2007) also investigated the pattern of lower brominated congeners (particularly penta- to heptaBDEs) in the fish in comparison to the pattern in the sediments. One hexaBDE (BDE-154) was detectable in only one of the four sediment samples but was found to be present in 91 per cent of the carp muscle samples (and contributed between five and 22 per cent of the total polybrominated diphenyl ether burden in the muscle samples). It was speculated that the presence of this congener could be due, at least in part, to debromination of decaBDE. It should be noted that there are several limitations with this study; in particular, relatively few sediment samples were analysed and no information was available on octaBDE and nonaBDE (other possible (intermediate) debromination products of decaBDE), so it is not possible to conclude definitively that the findings indicate debromination of decaBDE.

The uptake of decaBDE from sediment by the marine polychaete worm *Neries virens* has been studied by Klosterhaus et al. (2007) [ABST]. The worms were exposed to a sediment spiked with decaBDE, a sediment spiked with a mixture of decaBDE, pentaBDE and decachlorobiphenyl, a sediment spiked with decachlorobiphenyl alone (positive control) and a field sediment that was contaminated with polybrominated diphenyl ethers. The concentration of decaBDE in the sediments was approximately 2,500 µg/kg dry sediment. A solvent control sediment was also used. The exposures were carried out for 28 days and the worms were not fed over this time period. In addition to the sediment experiments, experiments were also carried out using contaminated food where the worms were fed a diet of clams spiked with either decaBDE or decachlorobiphenyl (the concentration in the clams was 2,500 µg/kg wet weight) for 28 days. A control experiment using uncontaminated clams was also carried out. The worms used in the feeding experiments were maintained in water only (no sediment present).

The worms were analysed for the presence of polybrominated diphenyl ethers at various times during the study. The worms were allowed to purge their gut contents in clean sediment and water for 24 hours prior to analysis.

The results from the sediment experiments indicated that decaBDE was more bioavailable from the spiked sediments than the field sediments. No decaBDE was detected (concentration <0.3 µg/kg wet weight) in the worms exposed to the field sediments. The concentration of decaBDE found in the worms after 28 days exposure to the sediment spiked with decaBDE alone reached around 17 µg/kg wet weight (giving an approximate bioaccumulation factor (concentration in worms/concentration in dry sediment) of 0.007), whereas the concentration of decaBDE in worms after 28 days in the sediment spiked with a mixture of decaBDE, pentaBDE and decachlorobiphenyl was <1 µg/kg wet weight (the concentrations in worms in the spiked sediments were given graphically and the values quoted here have been read from the graphs; the paper indicates that the concentration in worms in the sediment spiked with the mixture of substances was around thirty times lower than in worms exposed to the sediment spiked with decaBDE alone). The levels found in the worms exposed via diet were not given but the paper indicates that the bioaccumulation factors (concentration in worm/concentration in sediment or concentration in diet) for decaBDE for exposure via both sediment and feed were <0.05, with the bioaccumulation factor via diet being around three to four times higher than the spiked sediment exposure.

The study also determined the levels of nonaBDEs (BDE-206, BDE-207 and BDE-208) and octaBDEs (BDE-196 and BDE-197) in both the sediments and worms exposed to the sediments. The concentrations of these components in the field-contaminated sediment were between 0.5 and 40 µg/kg dry weight, and the concentrations found in the worms exposed to the sediment for 28 days were generally very low (BDE-197, BDE-207 and BDE-208 in the range 0.02 to 0.08 µg/kg wet weight) or not detectable (BDE-196 and BDE-206 <0.06 µg/kg wet weight). Similar to the case with decaBDE, a higher bioavailability for these components was evident in the experiments with the sediment freshly spiked with decaBDE. In this case the concentrations of these components in the sediment were between 1 and 190 µg/kg dry weight and the concentrations found in the worms after 28 days exposure were between 0.1 and 3 µg/kg wet weight (the concentrations for individual components are shown graphically in the paper and are difficult to determine but it is clear that BDE-206, BDE-207 and BDE-208 were present in the worms in this case)

Overall these experiments show that decaBDE is bioavailable from sediment but the actual bioaccumulation factor is low. The study also suggests that the bioavailability of decaBDE may be reduced by the presence of other substances that are more bioaccumulative than decaBDE, and that the bioaccumulation from field sediments is much lower than found in freshly spiked sediments. Few other details of this study are available; in particular, important information such as the organic carbon contents of the sediments and information on the levels of decaBDE in the worms at time points prior to 28 days are not given (and so it is not possible to assess if steady state was reached within 28 days). The paper suggests that the concentrations in the worms increased with time.

Guo et al. (2008a) investigated the accumulation of polybrominated diphenyl ethers (including decaBDE and a nonaBDE (BDE-207)) in fish, shrimps and crab from the Pearl River Estuary and Daya Bay, South China. DecaBDE was reported to account for <3% of the total polybrominated diphenyl ethers in these samples (the actual levels of decaBDE were not given) but BDE-207 appeared to account for around 10% of the total polybrominated diphenyl ethers in some samples (again the actual levels of BDE-207 were not given). The bioaccumulation of polybrominated diphenyl ethers was considered in terms of total polybrominated diphenyl ethers and no analysis was presented for decaBDE or BDE-207 alone.

Another study by Guo et al. (2008b) determined the tissue distribution of decaBDE in fish from the Pearl River Delta. The fish sampled included three species of freshwater farmed fish (big head carp (*Aristichthys nobilis*), mandarin fish (*Siniperca chuatsi*) and northern snakehead (*Nemipterus virgatus*)), one species of marine farmed fish (crimson snapper (*Lutjanus erythropterus*)) and one species of wild marine fish (golden thread (*Nemipterus virgatus*)). A total of eight individuals of each species were included in the study. The levels of decaBDE in the gastrointestinal tract (food, digest fluid and fat were removed prior to analysis), muscle, gills, liver and skin was determined. The analytical method used had a detection limit of 10 µg/kg dry weight and no decaBDE was detected in procedural blanks run during the analysis. In all, a total of 187 samples were analysed and decaBDE was detected in 70 of these samples in the range 0.39 to 59.9 µg/kg dry weight[23]. The 70 samples in which decaBDE was detected included 22 skin samples, 18 gill samples, 11 gastrointestinal tract samples, 11 muscle samples and eight liver samples. The study also investigated the ratio of decaBDE concentration to the total polybrominated diphenyl ether concentration (a total of 11 congeners were analysed) and this ratio was found to be higher in the skin

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[23] Some of the concentrations reported in this paper appear to be below the stated analytical detection limit. In addition, the raw data given in the paper give mean levels of decaBDE in individual species/tissues in the range 0.22 to 15.54 µg/kg dry weight but it is not clear how ‘not detected’ values were treated in the estimation of these mean values.

samples (mean 48 per cent) than in the liver samples (mean 8.2 per cent) suggesting that metabolism of decaBDE in the liver may be occurring. However it should be noted that this analysis is limited by the relatively high detection limit of the analytical method used.

Birds

Gauthier et al. (2008) investigated the presence of possible metabolites of decaBDE in eggs of Herring Gulls (*Larus argentatus*) from the Great Lakes. The study was part of a temporal trend study on the concentrations of polybrominated diphenyl ethers (see Section 4.2.5 for further details). The samples were collected in 2006 (and in one case 2005) and the concentrations of nonaBDEs found in the samples were in the order BDE-207 > BDE-208 >> BDE-206. BDE-207 and BDE-208 have been suggested previously as possible metabolites of decaBDE in experiments using subcutaneous silastic implants with European Starlings (*Sturnus vulgaris*) (see ECB, 2007). In addition, Gauthier et al. (2008) found that the profile of octaBDE in the gull eggs was similar to that found in starlings and that there was a strong covariance between the levels of decaBDE and the levels of both octaBDEs and nonaBDEs found in the gull eggs. On this basis Gauthier et al. (2008) concluded that this study provided evidence that decaBDE was debrominated to lower brominated congeners (nona- and octaBDEs). However, the relative metabolic capacities of these two species are unknown, and it should be noted that other explanations could exist. For example, the gulls could be exposed to nona- and octaBDE congeners arising from the commercial octabromodiphenyl ether product. Some support for this explanation comes from the fact that the proportion of octaBDE congeners in relation to decaBDE seems much higher across several years at some sites than others. In addition, some sites have quite different ratios between years. Therefore the reported conclusions from this study, in terms of metabolism of decaBDE, are not necessarily correct.

An investigation of the tissue distribution of decaBDE in foraging hens has been carried out by Liang et al. (2008). The hens were collected from near a site in Taizhou, China, where electronic waste (mainly television sets) had been dismantled approximately six years previously (the workers separated out the useable material, crushed/powdered the waste, stacked it on a riverbed and covered with soil). The area has since been covered with vegetation, although some parts were bared by weathering. The hens were dissected into twelve tissues and analysed for the presence of decaBDE (the quality control/quality assurance procedures used included the routine analysis of procedural blanks and field blanks and the levels of decaBDE in these were well below ten per cent of the levels found in the tissue samples). The levels found in the various tissues are summarised in Table 4.28.

DecaBDE was the predominant polybrominated diphenyl ether found in the samples and the following trend in tissue levels (based on lipid weight concentrations) was evident: muscle > fat > intestine > heart > liver > oviduct > gizzard > ovum > skin > blood.

Table 4.28 Tissue distribution of decaBDE in foraging hens.

Tissue	Lipid (% of wet weight)[a]	Concentration[b]	
		µg/kg lipid	µg/kg wet weight
Liver	NA	495	
Heart	0.51	1,135	5.8
Skin	30.6	71	22
Fat	88.7	2,308	2,047
Intestine	2.4	1,474	35
Muscle	0.80	17,796	142
Oviduct	NA	274	
Gizzard	NA	229	
Ovum	NA	72	
Blood	0.30	33	0.10
Spleen			154
Kidney			46

Notes: a - NA = not available. The values given have been estimated here based on the wet weight of the samples and the amount of lipid determined in the samples based on the raw data presented by Liang et al. (2008).

b - Data are given in the paper on a lipid weight basis for all tissues except for spleen and kidney. The remaining wet weight data here have been estimated using the lipid contents in this table.

Polybrominated diphenyl ethers were also detected in air, water, soil, animal and vegetation samples from the area. These were stated to have a similar congener profile to that found in the hen tissues (with decaBDE again being the predominant congener present) and the levels of total polybrominated diphenyl ethers in these samples were 967 pg/m^3 in air, 201 ng/l in field water, 28 mg/kg dry soil, between 16 and 687 µg/kg wet weight in eight species of animal and between 195 and 9,502 µg/kg dry weight in eighteen species of vegetation. Few other details of these samples are given.

Herbert *et al.* (2009) studied the accumulation of decaBDE in Herring Gulls (*Larus argentatus*) breeding along the shores of the St Lawrence River in Canada. Eggs were collected from 13 nests at Strachan Island (one egg per nest) each year between 1986 and 2005 (except for 1987). The collection site was in a relatively polluted area of Lake St Lawrence. The study used changes in the ratios of stable nitrogen isotopes ($\delta^{15}N$) and fatty acid methyl esters in the eggs as tracers to provide information on the trophic position and food webs utilized by the birds ($\delta^{15}N$ values were determined in ten individual eggs each year between 1986 and 2002, and on pooled samples of eggs for 2003 to 2005; fatty acid methyl ester values were detemined in pooled egg samples from 1986 to 2005, and additionally ten individual eggs from 2001 and 2002). In addition, ten individual eggs from both 2001 and 2002[24] were analysed for the presence of PBDEs (including decaBDE). The analytical methodology for PBDEs used in this study was reported to be the same as in the Gauthier et al. (2008) study (no further details were given).

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[24] These years were chosen as they represented the most recent years for which individual values of $\delta^{15}N$ and fatty acid methyl esters were available.

The study found no correlation between the total concentration of PBDEs (a total of 29 congeners ranging from tri- to decabrominated congeners were included in the study; only the concentration for total PBDEs was given in the paper and this was in the range 0.206-0.902 mg/kg wet weight) and the $\delta^{15}N$ value. However, principal component analysis indicated that eggs with higher $\delta^{15}N$ values were proportionally more contaminated with the lower brominated congeners than eggs with lower $\delta^{15}N$, and that the eggs with the lower $\delta^{15}N$ values generally had higher levels of the higher brominated congeners (hepta- to decaBDE). This difference in the congener pattern was discussed by Herbert et al. (2009) in terms of potential differences in the diet of the birds. The $\delta^{15}N$ and fatty acid methy ester values suggested that eggs with higher $\delta^{15}N$ were from birds which incorporated a greater proportion of aquatic prey into their diets and, as the PBDE congener profile of aquatic organisms is generally dominated by the lower brominated congeners (particularly BDE-47, BDE-99 and BDE-100) this may explain the congener pattern seen in the eggs with high $\delta^{15}N$ values in this study. Conversely, Herbert et al. (2009) suggested that the congener profile in terrestrial prey may be dominated by the more highly brominated congeners and that terrestrial prey will likely occupy a lower trophic position than aquatic prey (lower $\delta^{15}N$ value) and so feeding on terrestrial prey could at least partly explain the congener pattern seen in the eggs with the lower $\delta^{15}N$ values. Overall, Herbert et al. (2009) concluded that for PBDEs, trophic position may not be the most important factor in relation to PBDE concentrations in food webs but rather the type of food (aquatic versus terrestrial prey) may be more important.

Holden et al. (2008 [ABST] and 2009) studied the polybrominated diphenyl ether congener pattern in Peregrine Falcon (*Falco peregrinus*) eggs from California, USA. The study used 95 eggs collected between 1986 and 2007 and the eggs were analysed for a range of polybrominated diphenyl ether congeners. The paper indicates that most of the egg samples were analysed without the shell but some of the samples also included the shell. No distinction is made between the samples with and without shell in the rest of the paper. The analytical method involved high resolution gas chromatography coupled with high resolution mass spectrometry and the quality assurance/quality control measures included minimising exposure to UV light and routine analysis of blank and quality control samples. The recovery for decaBDE was reported to be 59.6 per cent and it is not clear if the concentrations of decaBDE found were corrected for this recovery.

The median level of decaBDE in the eggs was 0.49 mg/kg lipid. NonaBDEs (sum of BDE-206, BDE-207 and BDE-208) were also present at a median level of 0.18 mg/kg lipid. Also present were octaBDEs (median concentration 0.80 mg/kg lipid), heptaBDEs (median concentration 0.95 mg/kg lipid), hexaBDEs (median concentration 1.95 mg/kg lipid), pentaBDEs (median concentration 1.90 mg/kg lipid) and tetraBDEs (median concentration 0.54 mg/kg lipid). Holden et al. (2009) found that the total PBDE congener profile in the eggs differed markedly from that found in aquatic biota, where the lower brominated congeners (particularly tetraBDEs and pentaBDEs) tended to dominate.

Holden et al. (2009) also investigated the profiles of the major heptaBDEs (BDE-179 and BDE-183), octaBDEs (BDE-196, BDE-197, BDE-201, BDE-202 and BDE-203) and nonaBDEs in comparison with the profiles reported for commercial penta-, octa- and decabromodiphenyl ether products. It was found that two congeners (BDE-202 and an unknown heptaBDE[25]) were present in the eggs that were not reported to occur in commercial products and the authors suggested that these provide evidence for biological debromination of decaBDE. In addition, two nonaBDEs (BDE-207 and BDE-

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[25] The paper is not entirely clear about this congener. For example, in one place it is mentioned that it has been present in trace amounts in a commercial octabromodiphenyl ether product.

208) were present in a higher proportion in the eggs than found in penta- and decabromodiphenyl ether commercial products. Holden et al. (2009) discounted that these elevated nonaBDE congener proportions could arise from exposure to commercial octabromodiphenyl ether products (which contain a substantial amount of nonaBDEs) based on a simple consideration of the relatively small usage of the commercial octabromodiphenyl ether, and considered that they provided further evidence for biological debromination of decaBDE. However this latter argument appears to be flawed as the eggs were found to also contain substantial amounts of octaBDEs and heptaBDEs indicating that exposure to the components of the commercial octaBDE products may have been significant in this case. Therefore it is considered here that at least part of the explanation for the pattern of nonaBDEs seen in these samples could result from exposure to the commercial octabromodiphenyl ether products. Overall, although Holden et al. (2009) concluded that there were several strands of evidence for metabolic debromination of decaBDE in these data, there are many uncertainties in making this interpretation. The most important finding in this respect is that BDE-202 was present in the samples (as this congener has been suggested as a potential marker for debromination of decaBDE).

Mammals

Huwe (2005) [ABST] and Huwe and Smith (2007) studied the uptake and accumulation of decaBDE in rats exposed via their diet. Sprague-Dawley rats (80-d old) were trained to eat a diet consisting of 12 g ground rat chow topped with 200 µl of corn oil over a one hour period each morning (this method of dosing a restricted diet ensured that the dose was completely consumed by the rats and also minimised the impact of body mass changes during the study). No traces of hepta- to decaBDE were evident in the food or corn oil used in the study. The decaBDE used in the experiment had a purity of 98.5 per cent and was dissolved in corn oil and a small amount of toluene. Other polybrominated diphenyl ethers that were found to be present in the decaBDE used included nonaBDEs, octaBDEs and a trace of a heptaBDE (BDE-183). The solution was stirred overnight under a stream of nitrogen to allow the toluene to evaporate, giving a final concentration of 18.9 µg decaBDE/ml oil. At the start of the test the decaBDE solution was added to the diet of eighteen rats each day for 21 days at a concentration of 0.3 mg/kg food (each rat received a daily dose of 3.8 µg). Eight control rats received the same diet without the addition of decaBDE.

At various times after the last dose was administered (starting 24 hours after the last feeding up to 21 days after last feeding) groups of three dosed rats were killed and the amounts of polybrominated diphenyl ethers in the various tissues and organs were determined. The levels found 24 hours after dosing for 21 days are summarised in Table 4.29. Low amounts of decaBDE and nonaBDE were also found to be present in the control rats but the tissue levels were 10 to 20 times lower than in the exposed rats. In addition, some PBDEs were also found to be present in laboratory blanks. The data reported in Table 4.29 have been corrected for the levels found in the control rats.

The concentration of decaBDE in dosed rats was found to be around two to three times higher in the liver than the carcass (the lipid contents were similar at 4.13 per cent for the liver and 4.32 per cent for the carcass). This is consistent with the uptake of decaBDE being associated with blood proteins rather than lipid fractions. Biomagnification factors (BMFs; based on the concentration in rat (µg/kg fresh weight)/concentration in food (µg/kg fresh weight)) were estimated by Huwe (2005) for decaBDE as 0.16 for liver and 0.05 for carcass. The value for liver is similar to those obtained previously for several lower brominated congeners (e.g. Huwe (2005) gives biomagnification factors for liver of 0.17 for BDE-17, 0.24 for BDE-99, 0.31 for BDE-100, 0.33 for BDE-153, 0.11 for BDE-154 and 0.28 for BDE-183) but much lower than those found for carcass (biomagnification factors for carcass were given by Huwe

(2005) as 0.41 for BDE-17, 0.59 for BDE-99, 0.49 for BDE-100, 0.46 for BDE-153, 0.34 for BDE-154 and 0.33 for BDE-183). This suggests that decaBDE has a different deposition pattern to the tetra- to hexabrominated congeners in rats.

The estimated percentage of the dose retained within the animals and excreted after 21 days is summarised in Table 4.30. A potentially important finding from these data is that the amounts of several lower brominated congeners present in the rat after 21-days exposure were higher than could be accounted for by the dose given. This includes two octaBDEs (BDE-197 and BDE-201) and one nonaBDE (BDE-207)[26]. For these congeners the amounts present in the tissues were around 155% (BDE-207), 845% (BDE-201) and 1,170% (BDE-197) of the administered dose and suggest that these congeners were formed by the debromination (meta-debromination) of higher brominated congeners in the rat (and as a result of this, the apparent BMF values for these congeners were >1 (see Table 4.29)).

However, another possible explanation for the elevated levels of these congeners is as a result of breakdown of decaBDE during the tissue handling, extraction and analytical procedure. Although precautions were taken to minimise exposure to light during the sample preparation and analysis during the study, this possibility was investigated further by Huwe and Smith (2007) using a control carcass that had been spiked with decaBDE at similar concentrations to those found in the dosed animals. These spiked control samples (experiment was carried out in triplicate) were processed using the same analytical methodology as the dosed animals and this found that the concentrations of BDE-197, BDE-201 and BDE-207 were approximately ten times lower than found in the dosed animals and so formation during the analytical procedure could not, on its own, account for the levels of these congeners found in the dosed animals. Furthermore, the pattern of octaBDEs and nonaBDE found in the spiked samples was different from that found in the dosed animals. Overall, Huwe and Smith (2007) concluded that reductive debromination of decaBDE was occurring in the rats by meta- debromination, but that such metabolism accounted for a relatively small fraction of the total decaBDE dose (<3 per cent).

Evidence for more extensive metabolism of decaBDE came from mass balance considerations. The amount of unchanged decaBDE retained in the tissue and plasma after 21 days exposure was around 5 per cent, with the amount in faeces being around 50% of the dose (see Table 4.30). As the debromination to lower brominated congeners was estimated to be <3 per cent this left around 42 per cent of the dose unaccounted for, possibly as non-extractable bound residues or as unknown metabolites. No further analysis for possible bound or hydroxylated metabolites was carried out however.

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[26] The absence of analytical standards for some congeners at the time of the analysis does not affect the findings, because the same analytical uncertainties would apply equally to the analysis of the amounts present in the food samples and the rats (i.e. although the *absolute* concentrations may be uncertain, the *relative* concentrations between the food and rats would not be affected by the lack of standards).

Table 4.29 Uptake and accumulation in rats via the diet.

Congener	Total dose administered via food (ng per rat)	Total amount present in rat after 21 days exposure[a]									Carcass BMF[c]
		Liver		Carcass		GI tract		Plasma		Faeces	
		Conc. (µg/kg fresh weight)	Amount per rat (ng)	Conc. (µg/kg fresh weight)	Amount per rat (ng)	Conc. (µg/kg fresh weight)	Amount per rat (ng)	Conc. (µg/kg fresh weight)	Amount per rat (ng)	Amount per rat (ng)	
DecaBDE	71,770	48.2	320	14.0	3,020	35.9	190	3.6	11.6	35,940	0.05
BDE-208[b]	90	1.0	6	0.3	70	0.7	4	0.05	<0.15	270	0.9
BDE-207[b]	450	7.6	50	2.9	620	5.8	30	0.5	1.7	830	1.6
BDE-206	1,060	0.9	6	0.4	80	0.7	4	0.04	0.1	830	0.09
BDE-203	12	0.03	0.2	0.02	4	0.02	0.1	0.002	<0.01	6.4	0.4
BDE-201[b]	<2.4	0.2	1	0.08	20	0.1	0.5	0.004	0.01	7.6	8.8
BDE-197[b]	8	0.8	5	0.4	80	0.8	4	0.02	0.07	11.1	12.1
BDE-196[b]	24	0.2	1	0.1	20	0.2	1	0.004	0.01	8.8	1.1
BDE-183	4	0.02	0.2	0.03	6	0.04	0.2	0.002	0.01	<0.13	1.6

Notes:
a - Amounts are corrected for the amounts found in the control rats.
b - The values given for these congeners are estimates as standards were not available at the time of the analysis. However, the identities were confirmed later once standards became available.
c - The biomagnification factor (BMF) was defined as the concentration in carcass/concentration in feed on a wet weight basis. As the lipid contents of the carcass (lipid content 4.32%) and feed (the lipid contents of the feed were not given) were said to be similar, the BMF values would be similar on a lipid weight basis.

Table 4.30 Percentage retained and excreted in rats exposed for 21 days to decaBDE via diet.

Congener	Percentage of dose retained (%)[a]	Percentage of dose excreted (%)[b]
DecaBDE	5	50
BDE-208	85	290
BDE-207	155	183
BDE-206	9	79
BDE-203	35	56
BDE-201	845	217
BDE-197	1,170	146
BDE-196	108	37
BDE-183	150	3

Notes: a - Based on amount of substance present in liver, carcass, GI tract and faeces after 21 days exposure/total amount administered.

b - Based on amount of substance in faeces after 21 days exposure/total amount administered.

Huwe and Smith (2007) also investigated the depuration kinetics of decaBDE in the rats for up to 21 days following the last dosing. The concentrations of decaBDE in carcass, liver and plasma were analysed using either simple first order kinetics or by fitting the data to a biphasic model. For the carcass, the data best fitted simple first order kinetics, and a half-life of 8.6 days was obtained. For the liver and plasma the data were best fitted to a biphasic model. For the liver, the half-lives of the two phases were estimated to be 0.7 days and 20.2 days. For plasma, the half-lives of the two phases were estimated to be 1.2 and 75.9 days. The plasma levels were also fitted to a simple first order kinetic model (half-life 3.9 days) but the fit was not as good as with the biphasic model. The relatively long half-life of 75.9 days was thought to possibly reflect slow elimination from body stores following chronic dosing, however, Huwe and Smith (2007) cautioned that the estimate contained significant error owing to the relatively short depuration phase (21 days) used in the study.

Huwe and Smith (2007) also found that the concentration of two of the octaBDE congeners (BDE-197 and BDE-201) continued to increase during the 21-day depuration phase of the study. This provided further support for their formation in the rats by metabolism of higher brominated congeners.

Riu et al. (2008) investigated the disposition and metabolism of ^{14}C-labelled decaBDE in pregnant Wistar rats. The substance used had a radiochemical purity of >99.8 per cent. Three rats were used in the study (mean bodyweight 284 g). The animals were force fed with daily dosed of the ^{14}C-decaBDE dissolved in peanut oil from gestational days 16 to 19 (the dose rate was 2.0 mg/kg bodyweight/day). During the study the rats were allowed free access to water and a standard diet, and urine and faeces were collected daily. The animals were killed on day 20 of gestation (24 hours after the last dose of ^{14}C-decaBDE) and the amounts of ^{14}C-label, parent compound and metabolites were determined in various organs and tissues. The distribution of radiolabel in the animals on gestational day 20, and the concentration of radiolabel (as ^{14}C-decaBDE equivalents) are shown in Table 4.31.

The total radioactivity recovered in the experiment was 91.1 per cent of that dosed. The main route of elimination of radiolabel from the animals was via the faeces (accounting for around two thirds of the administered dose). In concentration terms, the highest concentrations of radiolabel were present in the adrenals, ovaries and liver, where concentrations above 10 mg/kg were present. The concentration of radiolabel calculated for whole blood was around 1 mg/l with around 75 per cent of this being associated with the plasma fraction. The concentrations of radiolabel in adipose tissue and skeletal muscle were <0.8 mg/kg. The radiolabel present in carcass (remaining muscle and fat, bones, skin and eyes) was mainly located in the skin and adipose tissue.

A more detailed analysis of the ^{14}C-metabolites present was also carried out using HPLC with a radioactivity detector. Around 97 per cent of the radiolabel recovered from the organic fractions of the faeces was found to be unchanged decaBDE. However, three ^{14}C-labelled metabolites that were more polar than decaBDE were also evident. Similarly, unchanged decaBDE accounted for 96.4 per cent, 86.4 per cent and 91.9 per cent of the radiolabel in the organic fractions of the stomach, small intestine and large intestine respectively. Metabolites with a higher polarity than decaBDE were also evident in the aqueous fractions from the stomach, small intestine, large intestine and faeces accounting for around 34.4 per cent, 70.5 per cent, 50.6 per cent and 17.5 per cent respectively of the radiolabel present in the aqueous fractions. For urine, the majority of the radiolabel present was found to be as more polar metabolites and no unchanged decaBDE was evident.

For plasma and other tissues, the proportion of metabolites present ranged from around 9 per cent in adrenals, ovaries and liver to around 30 per cent in the carcass. The remaining radiolabel was generally attributable to unchanged decaBDE although a small amount of non-extractable radiolabel was evident in some tissues.

Several main metabolites were evident in the various tissues. These were isolated from faeces and tissues and identified using mass spectrometric methods. These were identified as nonaBDEs (BDE-206, BDE-207 and BDE-208), an unidentified octaBDE and a hydroxylated octaBDE derivative. Checks were carried out to determine whether degradation of decaBDE occurred during the preparation and administration of the dosage solution or during the analytical procedure. This showed that no degradation occurred meaning that the metabolic products were formed in the animals.

Overall, the metabolites were estimated to account for around 7 per cent of the total radioactivity administered. The result showed that at least 19 per cent of the administered dose was absorbed and recovered in the body tissues plus carcass. Excretion via faeces was the main elimination route. Metabolism of decaBDE was likely to be occurring in the rats, with the initial formation of nonaBDEs (BDE-206, BDE-207 and BDE-208), although it is possible that such reductive debromination could be occurring in the gut by the microflora present. The liver was found to be a target tissue for decaBDE (6.5 per cent of the total radioactivity and a concentration of 11 mg/kg) but the highest concentrations were found to be present in certain endocrine glands (e.g. adrenals and ovaries). The amount of radiolabel found to cross the blood-brain barrier was relatively small but 0.5 per cent of the dose was found in fetuses indicating that decaBDE or metabolites can cross the placental barrier.

Table 4.31 Distribution of ^{14}C-labelled decaBDE in pregnant rats.

Organ/tissue	Mean percentage of administered radioactivity (%)	Mean concentration (mg/kg of mg/l) as ^{14}C-decaBDE equivalents[a]
Faeces	66.29	
Digestive tract content	5.33	
Urine	0.11	0.06
Adrenals	0.16	33.06
Ovaries	0.13	16.05
Liver	6.48	11.16
Kidneys	0.29	3.90
Stomach	0.20	3.10
Small intestine	0.64	2.81
Heart	0.08	2.47
Lung	0.13	1.93
Spleen	0.06	1.82
Large intestine	0.16	1.61
Plasma	0	1.53
Uterus	0.24	1.39
Adipose tissue		0.79
Muscle		0.41
Brain	0.01	0.11
Placentas	0.47	2.48
Fetuses (whole litter)	0.43	0.46
Amniotic fluid	0.02	0.11
Carcass	9.23	1.11
Cages	<0.0002	

Notes: a - Assuming all the radiolabel is unchanged decaBDE.

The bioavailability of polybrominated diphenyl ethers in household dust to rats has been investigated by Richardson et al. (2007) [ABST] and Huwe et al. (2008a). In the study male Sprague-Dawley rats were fed a diet supplemented with either a standard reference dust[27] containing known amounts of polybrominated diphenyl ethers or oil containing a mixture of commercial penta-, octa- and decabromodiphenyl ether products for 21 days. Two exposure groups were used for both the dust- and oil-amended diets. The daily doses of total polybrominated diphenyl ethers received by the rats were approximately 1 µg/kg bw per day (low dose – oil and low dose – dust groups) and 6 µg/kg bw per day (high dose – oil and high dose – dust groups). A control group fed diet without any added polybrominated diphenyl ethers was also included (the daily dose of total polybrominated diphenyl ethers received by this group

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[27] The dust used was National Institute of Standards and Technology (NIST) SRM-2585 dust. Kucklick et al. (2007) [ABST] gives the decaBDE content of this dust as 2,510 µg/kg dry weight and the concentration of BDE-206 (a nonaBDE) as 271 µg/kg dry weight.

was around 0.24 µg/kg bw per day resulting from the levels present in feed only). The amounts of the individual polybrominated diphenyl ethers in each diet are shown in Table 4.32 for the hepta- to decaBDE congeners (tri- to hexaBDEs were also present).

The animals were sacrificed after 21 days' exposure (the animals were killed 24 hours after the last feeding) and the amounts of polybrominated diphenyl ethers present in various organs and tissues were determined. DecaBDE was found to be equally as bioavailable when administered in dust compared with oil with around 4.0 and 4.8 per cent of the dose being retained respectively after 21 days exposure (see Table 4.32).

Richardson et al. (2007) [ABST] reported that the lower brominated congeners (tri- to hexaBDEs) were found to account for the majority of the polybrominated diphenyl ethers present in adipose tissue, brain and carcass (nonaBDEs and decaBDE were not consistently detected in adipose tissue at concentrations above the control), but higher brominated congeners (hepta- to decaBDE) accounted for over 50% of the total polybrominated diphenyl ethers present in livers, with decaBDE being the predominant congener found in liver (the amount of nonaBDE and decaBDE in liver in the high dose - dust group was found to be lower than that in the high dose – oil group) and it was suggested by Richardson et al. (2007) that this may result from alterations in metabolism (for example the study found that hepatic Cyp1a1 mRNA expression increased significantly with dust exposure, but not with oil exposure, suggesting that components of the dust other than polybrominated diphenyl ethers may have been causing Cyp1a1 induction). This possibility was not discussed in the Huwe et al. (2008a) paper.

Huwe et al. (2008a) also calculated biomagnification factors (concentration in tissue/concentration in diet) for the polybrominated diphenyl ethers included in the study. The BMFs were determined separately for epididymal fat/adipose tissue and liver. The BMFs for decaBDE were in the range 0-0.6 for adipose tissue (in this case the BMF was determined based on the lipid normalised concentration in adipose tissue/the concentration in feed (not lipid normalised)). For liver, the BMFs for decaBDE were between 0.07 and 0.41 (in this case the BMF was determined based on the fresh weight concentrations in liver and food). Higher BMFs were determined for two nonaBDE congeners (BDE-206 and BDE-207). The BMFs for BDE-206 were between 0 and 2.1 in adipose tissue and 0.05 and 2.4 in liver and the BMFs for BDE-207 were between 0.75 and 2.29 in adipose tissue and 0.40 and 1.09 in liver.

Table 4.32 Uptake and accumulation of polybrominated diphenyl ethers by rats from dust and oil.

Congener	Concentration in feed (µg/kg)					Percentage of dose retained and excreted via faeces after 21 days (%)			
	Dust – low dose	Dust – high dose	Oil – low dose	Oil – high dose	Control	Dust – high dose		Oil – high dose	
						Retained	Excreted	Retained	Excreted
Heptabromodiphenyl ether									
BDE-183	0.17	0.98	0.21	1.20	0.016	47.9	49.3	45.7	21.7
Octabromodiphenyl ethers									
BDE-196	0.13	0.76	0.035	0.21	0.0092	14.6	48.7	21.0	54.3
BDE-197	0.52	0.090	0.088	0.53	0.024	48.7	53.0	49.6	29.1
BDE-203	0.14	0.81	0.025	0.15	0.0050	23.7	47.8	6.6	44.9
Nonabromodiphenyl ethers									
BDE-206	0.66	3.8	0.23	1.4	0.18	10.1	85.0	11.6	67.6
BDE-207	0.33	2.0	0.19	1.1	0.078	22.6	91.7	20.0	55.5
Decabromodiphenyl ether									
DecaBDE	16.8	97.2	11.4	68.1	2.8	4.0	68.0	4.8	63.3

A further study of the tissue distribution of decaBDE in rats has been carried out by Huwe et al. (2008b). In this study, male rats were exposed using a diet supplemented with either the same reference dust as the Richardson et al. (2007) and Huwe et al. (2008a) study above (24 mg of dust g^{-1} diet) or corn oil containing a mixture of commercial penta-, octa- and decabromodiphenyl ether products (the ratio of the products was 16:1:25 in order to mimic the congener profile found in the dust) for 21 days. The doses of total PBDEs given to the rats were approximately 8 µg/kg body weight/day for the dust experiments and 6 µg/kg body weight/day for the corn oil experiments. Control animals received uncontaminated diet only. Twenty-four hours after the last feeding, the rats were killed and various tissues (including epididymal fat, brain, kidney, liver, lung, gastrointestinal tract and residual carcass) were analysed for the presence of polybrominated diphenyl ethers (PBDEs). Faeces were also collected daily during the course of the study. The study found that decaBDE was not readily distributed to most of the tissues and was extensively excreted in the faeces (decaBDE was found to account for over 70 per cent of the total PBDEs present in the faeces and based on Huwe et al. (2008a) over 60 per cent of the total dose of decaBDE administered to the rats was excreted unchanged via the faeces). The two body compartments in which decaBDE was detected most often were the liver and plasma (the amount of decaBDE found in liver was around 0.2 per cent of the total dose in the dust experiments and around 1.2 per cent of the total dose in the oil experiments after 21 days).

A study of the hibernation-associated changes in contaminant profiles in grizzly bears (*Ursus arctos horribilis*) has been carried out by Christensen et al. (2007). Samples of subcutaneous fat and hair samples were collected from eleven bears in the autumn of 2003 (the decaBDE levels in these bears were published previously by Christensen et al. (2005) and considered in ECB (2007)) and fourteen bears in early spring of 2004. The hair samples were used to obtain dietary information on the bears included in the study. The hairs were subdivided into one cm segments (each segment reflected approximately twenty days growth with the root end reflecting the most recent diet) and each segment was analysed for stable isotopes of carbon and nitrogen (i.e. ^{13}C and ^{15}N). The ^{15}N-measurements were used to determine the dietary index of the bears (a high dietary index implies that the bears consumed a diet consisting of a relatively high trophic level (e.g. salmon) and a low dietary index implies that the bears consumed a diet from a lower trophic level within a terrestrial food chain).

The fat samples were analysed for the presence of decaBDE and nonaBDEs (BDE-206, BDE-207 and BDE-208). The analytical method used included the routine analysis of procedural blanks and certified reference material. The levels of polybrominated diphenyl ethers in the blank samples were generally < 5 ng/kg and the detection limit of polybrominated diphenyl ethers was <1 ng/kg. The levels of decaBDE found in the fat samples were in the range 0.17 to 29.9 µg/kg wet weight and the levels of nonaBDEs were in the range 0.016 to 2.7 µg/kg wet weight for BDE-206, 0.021 to 1.9 µg/kg wet weight for BDE-207 and 0.015 to 1.6 µg/kg wet weight for BDE-208.

Christensen et al. (2007) carried out a detailed analysis of the lipid normalised concentrations of polybrominated diphenyl ethers in the bears before and after hibernation. The analysis was based broadly on a comparison of the concentrations of polybrominated diphenyl ethers before and after hibernation with the pattern seen for 2,2',4,4',5,5'-hexachlorobenzene, taking into account differences in dietary intake. The concentration of 2,2',4,4',5,5'-hexachlorobenzene was found to increase by a factor of 2.25 in spring (after hibernation) compared to autumn (before hibernation). However the analysis that could be carried out for nonaBDE and decaBDE was limited as the variation of the concentration with dietary index followed a different trend to that seen for 2,2',4,4',5,5'-hexachlorobenzene (and lower brominated PBDE congeners up to hexaBDE). Although not discussed in the paper, this difference could result from the fact that decaBDE exposure may be mainly through the terrestrial food chain rather than the aquatic food chain.

As a result of this, for the higher brominated polybrominated diphenyl ether congeners, Christensen et al. (2007) carried out a comparison for each congener by comparing the changes in concentrations during hibernation with those seen for an octaBDE congener (BDE-203; the concentrations of each congener before and after hibernation are not given separately in the paper). Based on this comparison, the relative order of "persistence" of the higher polybrominated diphenyl ethers (hepta- to decaBDEs) within the bears was BDE-183 > BDE-203 > BDE-208 > BDE-207 > BDE-206 > BDE-209 with relative persistence values of 3.13, 1.00, 0.48, 0.45, 0.40 and 0.26 respectively.

Christensen et al. (2007) indicates that hibernating bears provide a good system for investigating the persistence of chemicals. In general terms the main processes that lead to differences in the persistence of substances in fat in mammalian systems include substance-related differences in excretion, placental and lactational transfer, contaminant mobilization and redistribution, binding to cellular receptors and metabolism. However, a number of these possible processes are not operating in the hibernating bears in the current study. For example, hibernating bears do not urinate or defecate during hibernation and the bears used in the study were either adult males or females below reproductive age. A detailed principal component analysis of the whole data set (comprising a large number of polybrominated diphenyl ethers and polychlorinated biphenyls) indicated strongly that substance-specific differences in metabolism were key in explaining the patterns of substances seen.

An *in vitro* metabolism study with decaBDE using rat microsomes has been carried out by Mas et al. (2008) [ABST]. The tests were carried out using phenobarbital-, β-naphthoflavone- and clofibrate-treated rat liver microsomes. The decaBDE used in the study had a purity of 98 per cent, and stock solutions of the test substance were prepared in dimethyl sulphoxide (concentration 140 mg/l) for use in the test. DecaBDE was thought to be soluble in dimethyl sulphoxide at this concentration.

The metabolism tests were carried out by pre-incubating 1.5 µM of decaBDE (in dimethyl sulphoxide) with 1 mg/ml of the hepatic microsomes in a 0.1 M buffer (pH 7.5) for five minutes with shaking at 37°C. After preincubation the reaction was initated by the addition of a NADPH regenerating system and metabolism was stopped after 30, 60 and 120 minutes incubation (the total volume of the system was 1 ml). Control incubations were carried out in the same way, except sodium bicarbonate was added instead of the NADPH regenerating system.

The control incubations did not show any decrease in the concentration of decaBDE present, showing that there was no significant abiotic degradation of decaBDE under the conditions used. Loss (or metabolism) of decaBDE was evident in the hepatic microsomes, with around a 31 per cent to 58 per cent decrease in concentration being evident after 30 minutes in the different systems used. The highest loss was found with the phenobarbital-treated system. No further decrease in the decaBDE concentration was evident in any of the systems between 30 minutes and 120 minutes incubation time.

Mas et al. (2008) [ABST] attempted to determine the decomposition/metabolic products from decaBDE in this study. This analysis did not find evidence of any lower PBDE congeners as products. The study also investigated the formation of hydroxy derivatives (with between two and four bromine atoms per molecule) but again there was no evidence for the formation of hydroxy derivatives with this level of bromination.

Plants

Zlámalíková et al. (2008) [ABST] has investigated the uptake of decaBDE by plants from sewage sludge. The sewage sludge used in the study was from a waste water treatment plant in the Czech Republic and it contained a range of polybrominated diphenyl ethers including decaBDE (the decaBDE concentration was 2,973 µg/kg). Plants (tobacco plant (*Nicotiana tabacum*) and nightshade (*Solanum nigrum*)) were grown in aluminium foil-lined plastic pots containing the sewage sludge. A total of five replicates was used for each plant species. Control plants were grown using a proprietary growing medium. The plants were maintained at laboratory temperature with regular watering for three months. At the end of the growth period the amount of decaBDE present in various parts of the plant was analysed. For the tobacco plants, the concentration of decaBDE found was 0 µg/kg dry weight in roots, 16.1 µg/kg dry weight in stalk and 0 µg/kg dry weight in roots. For the nightshade plants, the concentration of decaBDE found was 28.2 µg/kg dry weight in roots, 0.8 µg/kg dry weight in stalks, 0 µg/kg dry weight in leaves and 1.2 µg/kg dry weight in fruit. It should be noted that no information is reported in the paper on the levels of decaBDE found in the control plants and therefore it is currently difficult to assess the significance of these results.

Discussion of bioaccumulation and metabolism data

A significant body of new information is available on the bioaccumulation and metabolism of decaBDE.

Uptake of decaBDE by aquatic organisms, birds and mammals has been shown in laboratory studies, confirming that decaBDE is bioavailable. In addition, decaBDE adsorbed onto dust has also been shown to be bioavailable.

No new data are available on the bioconcentration factor for decaBDE in fish. The BCF value for decaBDE in the previous risk assessment reports was assumed to be low (a BCF of around 4 l/kg was assumed in EC (2002)) However a new study with mussels is suggestive of a significantly higher BCF/BAF in mussels, possibly of the order of 1,000 l/kg or above. Studies determining the uptake from sediment generally show very low BSAF values.

The results of several new feeding studies are also available. These generally show that decaBDE is absorbed from the diet but the BMF values obtained are generally <1 for decaBDE. In addition, several studies have investigated the biomagnification of decaBDE in aquatic food chains. With one exception, these data generally show no evidence of increasing concentrations with increasing trophic level, demonstrating that decaBDE is not biomagnifying.

Further evidence for metabolism of decaBDE to lower brominated congeners has become available for both aquatic and terrestrial species. For example experiments with rats have shown higher levels of accumulation of nonaBDEs and octaBDEs than can be accounted for by the dose given (leading to apparent BMFs >1). In addition, some environmental monitoring studies (see Section 4.2.5) have shown that the levels of nonaBDEs in some species of biota are higher relative to decaBDE than would be expected based on their known occurrence in the commercial decabromodiphenyl ether product (nonaBDE content typically ≤3 per cent in the products currently supplied in the EU). There are several possible explanations for this. These include metabolic breakdown of decaBDE in the organism to nonaBDE, a higher accumulation potential for nonaBDE compared with decaBDE, degradation of decaBDE in the environment to nonaBDE with subsequent accumulation of the formed nonaBDE, or a higher release of nonaBDE to the environment than would be expected based on its content in the commercial decabromodiphenyl ether product (for example from use of older decabromodiphenyl ether products with higher nonaBDE contents than the current product or as a component of the commercial octabromodiphenyl ether products).

A potential marker for debromination of decaBDE (BDE-202) has been found in some organisms (e.g. Holden et al. 2009). However fields studies are not generally able to distinguish between metabolic formation of BDE-202 or degradation of decaBDE to BDE-202 followed by subsequent uptake into the organism.

Of interest to this discussion is a recent study by Geller et al. (2008) that has investigated the congener distribution in a commercial octabromodiphenyl ether product (DE-79^{TM}). The study found that this commercial product consisted of 36% BDE-183, 19% BDE-197, 13.1% BDE-207, 9.1% BDE-196, 7.3% BDE-153, 6.2% BDE-203, 2.2% BDE-180, 1.6% BDE-171, 1.2% BDE-154, 0.7% BDE-206 and 1.3% decaBDE.

Although there is undoubtedly some contribution to the total environmental burden of nonaBDE from older products and the commercial octabromodiphenyl ether products, these sources can be more or less ruled out when considering the congener pattern seen in biota as other environmental media (for example sludge and sediment) would also be expected to show a similar enhancement of nonaBDEs in comparison with decaBDE if this source were significant. No such enhancement is generally seen in the levels determined in sediments and soils. Similarly the degradation of decaBDE in the environment to nonaBDE can be ruled out as the main explanation for the patterns seen, particularly in fish, as such degradation would also be expected to alter the congener pattern seen in sediments, etc., which again generally show little or no enhancement of nonaBDE in comparison with decaBDE over that which would be expected based on the nonaBDE content of the current products. A possible exception to this could be if dust exposure was significant, as there is some evidence for enhancement of the levels of nonaBDE compared with decaBDE in dusts as a result of photodegradation (see Section 4.3.1).

This therefore leaves the most likely explanation(s) for the congener pattern seen in biota in a number of studies as either metabolic debromination of decaBDE to nonaBDE or a higher accumulation potential for nonaBDE compared to decaBDE. It is not always possible to distinguish between these two possibilities and, based on the available evidence, it is possible (or perhaps even likely) that both processes contribute to the pattern seen. In terms of the risks from decaBDE, the net effect of the two processes is the same; the levels of nonaBDE in the organism are higher than would be expected based on the amount present in the commercial decabromodiphenyl ether product alone.

There is also growing evidence that the metabolism of decaBDE may be species-specific. For example, studies carried out with rainbow trout and carp indicate metabolism may be occurring in these speces; a study by Lebeuf et al. (2006) reported in EC (2007) found that Atlantic tomcod (*Microgadus tomcod*) exhibited a very limited capacity to metabolise decabromodiphenyl ether to the more toxic lower brominated congeners. Based on the findings of Holden et al. (2009) dietary factors may also be an important factor in the uptake of decaBDE by organisms. This may explain why decaBDE is found in some organisms and not others. Although enhanced metabolism of decaBDE in some species may explain the very low (or not detectable) levels of decaBDE in the organism, it is also possible that the formation of metabolites which are more toxic and accumulative than decaBDE may be enhanced in such species. There is currently very limited evidence that this is the case but it is a logical consequence if it is accepted that metabolism of decaBDE leads to the formation of lower brominated congeners (or other more toxic and accumulative products) and that metabolism is species-specific.

Overall there is now strong evidence from both the available laboratory and field data (see Section 4.3.1) that metabolism of decaBDE to lower brominated congeners (down to nonaBDEs, octaBDEs and probably heptaBDEs) occurs in a number of species. The yield of these metabolites is generally low (typically <5 per cent of the dosed decaBDE in the various studies), but several studies indicate that other, as yet unidentified metabolites, may also be formed.

The final point of note from the new studies is that there is further evidence that the uptake of decaBDE by several organisms appears to be associated more with the blood proteins than the lipid content of the organisms. Some, but not all, studies appear to show preferential accumulation of decaBDE in organs like the liver rather than body fat. This may have implications for whether it is relevant to lipid normalise environmental monitoring (or BCF/BMF) data for decaBDE.

5 Ecotoxicity

The potential for disruption of thyroid hormone in fish by decaBDE and a nonaBDE (BDE-206) has been studied in a competitive binding assay (Morgado et al. 2007). The study used an assay developed using sea bream recombinant transthyretin (TTR) and 50 ng of TTR was incubated in 200 µl of buffer containing 0.1 nM [^{125}-I]-3,5,3'-L-triisdothyronine (T_3) in the presence of increasing amounts of unlabelled T_3 or polybrominated diphenyl ethers (concentration 0-10 µM) for 2 hours on ice and the binding of [^{125}I]-T_3 was determined. Neither decaBDE or BDE-206 (or indeed several hexa- to octaBDEs) showed any binding to TTR as shown by their inability to displace [^{125}I]-T_3.

The potential for decaBDE to induce DNA damage in zebra mussels (*Dreissena polymorpha*) has been studied by Riva et al. (2007). The mussels used were the same as those used for the bioconcentration study reported in Section 4.3.3 and were exposed to nominal concentrations of decaBDE of 0.1, 2 and 10 µg/l under semi-static conditions for up to 168 hours. Two assays were used, an alkaline Comet assay using haemolymph collected from the posterior adductor muscle sinus of the mussels after 48, 96 and 168 hours exposure and a micronucleus assay using haemolymph collected after 48 and 96 hours exposure. A control and solvent control (dimethyl sulphoxide) and positive control (benzo[*a*]pyrene) were also run.

No mussels died during the study. Cell viability tests showed that neither decaBDE nor the positive control had any significant effect on the viability of haemocytes indicating that the concentrations used were not cytotoxic to the mussels and that any DNA damage seen in the test was not the result of cell death.

The Comet assay showed a significant increase ($P < 0.01$) in the length diameter ratio (LDR) in the exposed groups compared with the control groups and this increase was dose dependent at the decaBDE concentrations of 0.1 and 2 µg/l. The response at a decaBDE concentration of 10 µg/l, although still significantly increased over the control groups, was lower at 168 hours than at 96 hours, and was lower than the response seen in the 2 µg/l group after 168 hours. A possible explanation for the response seen in the 10 µg/l treatment group given by Riva et al. (2007) could be due to a large amount of apoptotic cells that were not included in the score. Other possible explanations put forward by Riva et al. (2007) to explain the lack of dose-response seen included the possible formation of lower brominated congeners in the various treatment groups.

In contrast to the Comet assay, no treatment related effects were seen to result from exposure to decaBDE in the micronucleus assay showing that decaBDE exposure did not induce irreversible genotoxic effects on haemocyte.

Overall, Riva et al. (2007) concluded that decaBDE has the potential to induce DNA damage but not irreversible effects on DNA haemocytes. This significance of this finding, in terms of population survival, is not clear and so the results are not considered further here[28].

The toxicity of decaBDE to the harpacticoid copepod *Nitocra spinipes* has been investigated by Breitholtz et al. (2008). The test was carried out using silica gel as a carrier for decaBDE. The decaBDE was firstly dissolved in toluene, loaded onto 100 mg of silica gel and then the solvent was allowed to evaporate over 24 hours. The test solution was then prepared by adding 5 ml of brackish water (salinity 6.5‰) to the silica gel and allowing the system to equilibrate for one week. A total of eight concentrations were tested, corresponding to a nominal concentration of between 0.033 and 100 mg decaBDE/l, and six replicates were used in each test. The toxicity tests were carried out according to Swedish standard procedures (SIS, 1991) and under these conditions decaBDE was found to have no effect on the survival of the test species over four days (the 96-h LC_{50} was >100 mg/l).

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[28] BSEF (2009) has indicated that there may be a number of short-comings with the Comet assay carried out by Riva et al. (2007). For example, the data were evaluated using a parametric statistic but as Riva et al. (2007) used 'scored' data for the percentage of DNA in the tail to indicate DNA damage, a non-parametric statistic would have been more appropriate. In addition BSEF (2009) indicates that a possible source of error when evaluating DNA damage using the Comet assay is the formation of DNA strand breaks through apoptosis and/or necrosis and therefore it is preferable to carry out certain confirmatory tests. According to BSEF (2009) such confirmatory tests do not appear to have been carried out in this study.

6 Information related to human health

6.1 Mammalian toxicity/effects on human health

A review of all of the new data available for decaBDE and nonaBDEs on mammalian toxicity and effects on humans is outside the scope of this report. Table 6.1 outlines studies that have been located that could contain significant new information. Other studies related to human health that are of limited relevance to decaBDE (for example, those containing information on the toxicity of lower brominated congeners only) are listed in the other references of limited relevance at the end of this report.

Table 6.1 Summary of new studies related to mammalian toxicity.

Reference	Brief summary of study/findings
Akutsu et al*.* (2008a and 2008b)	Pilot study on the relationship between polybrominated diphenyl ethers in serum and sperm quality. It is not clear from the abstract if decaBDE was included in the study.
Barber et al. (2006)	Investigated the micronucleus-forming activities of decaBDE in MCF-7 cells. The findings for decaBDE are not clear from the abstract.
Beck (2009)	OECD 426 Test Guideline developmental neurotoxicity study with decaBDE in rats. The summary report indicates that no effects on any of the neurobehavioural endpoints were seen in the study.
Brown et al. (2004)	Studied Ah receptor pathway activation by brominated flame retardants. DecaBDE was tested.
Cantón et al. (2004)	*In vivo* effects of decaBDE on the adreno cortical enzyme (CYP17).
Cantón et al. (2006)	*In vivo* effects of decaBDE on CYP17 catalytic activity.
Cantón et al. (2007)	*In vivo* steroidogenic effects in rats. An inhibiting effect on adrenal CYP17 activity was seen with decaBDE.
Darnerud, (2008)	Review of the effects of polybrominated diphenyl ethers on the endocrine system.
Eriksson et al. (2006)	Interaction of decaBDE with perfluorinated chemicals during neonatal brain development and enhancement of developmental neurobehavioural defects.
Eriksson et al. (2008)	Interaction of decaBDE with perfluorinated chemicals during neonatal brain development and enhancement of developmental neurobehavioural defects.
Fonnum et al. (2006)	Considers the molecular mechanisms that may be involved in the toxicity of polybrominated diphenyl ethers.
Goodman (2009)	Review of the neurodevelopmental effects of decaBDE. Concludes that Viberg *et al.* (2003) study is not suitable for establishing a reference dose for decaBDE.

Reference	Brief summary of study/findings
Gregoraszczuk et al. (2008)	Abstract not yet available. Study investigated steroid secretion following exposure of ovarian follicular cells to polybrominated diphenyl ethers. Not clear of decaBDE was tested.
Hamers et al. (2004a, 2004b and 2006)	*In vitro* screening of the endocrine disrupting potency of decaBDE.
Harju et al. (2007) (see also Harju et al. (2004))	Quantitative structure-activity modelling of *in vitro* endocrine effects of 26 selected brominated flame retardants. It is not clear from the abstract if decaBDE was included in the study.
Hu et al. (2007)	Investigated the anti-proliferative, apoptotic properties of decaBDE using a human hepatoma HEP G2 line as a model system. The abstract indicates that decaBDE had the toxicity activity of anti-proliferation and induction of apoptosis in tumour cells *in vitro*.
Jiang et al. (2008)	Investigated the effect of maternal exposure of rats to decaBDE on the expression of growth associated protein-43 and brain-derived neurotrophic factors in the hippocampus of offspring.
Johansson et al. (2008)	Further investigation of the developmental neurotoxicity of decaBDE.
Kawashiro et al. (2007)	Studied the effects of decaBDE on human umbilical vein endothelial cells.
Kirkland et al. (2005a, 2005b and 2006)	May discus the *in vitro* genotoxicity data for polybrominated diphenyl ethers.
Mühlebach et al. (2006)	Investigated the cellular kinetics of decaBDE in cell culture systems (human skin fibroblasts, rat astrocytoma C6 cells and non-differentiated and differentiated mouse 3T3 cells).
Onos et al. (2007)	Investigated effects of decaBDE following developmental exposure (may be related to the Rice et al. (2007) study).
Pacyniak et al. (2007)	Investigated the potential for decaBDE to induce gene expression of cyp3a11, 2b10 and cypl1a1/2 in C57BL6 mice. Induced gene expression of cyp3a11 and 2b10 but not cypl1a1/2 was seen on exposure to decaBDE. Both cyp3a11 and 2b10 are known target genes of pregnane X receptor (PXR) and so it was hypothesised that decaBDE is a PXR activator. This was confirmed using reporter gene luciferase assays.
Peters et al. (2004)	Effects on EROD activity and cytochrome P450-1A1 expression.
Rice et al. (2007)	Investigated the effect of neonatal exposure to decaBDE in inbred C57BL6/J. Male and female mice were exposed to daily oral doses of 0, 6 or 20 mg/kg bw from postnatal day 2 to 15. The dose of 20 mg/kg bw/day was found to lead to developmental delays in the acquisition of the palpebral reflex in immature animals. Exposure to decaBDE at 20 mg/kg bw/day over this period also lead to alteration of the normal sex- and age-specific characteristics of spontaneous locomotor activity.

Reference	Brief summary of study/findings
Schricks et al. (2006)	Used the T-screen (GH3 rat pituitary cell line) to identify potentiating, antagonistic and thyroid hormone-like activities of polybrominated diphenyl ethers. Effects may have been seen with nonaBDE.
Serafimova et al. (2007)	Structure-activity model for mutagenicity. The findings for decaBDE are not clear from the abstract.
Stapleton et al. (2009)	Study of metabolism by human hepatocytes *in vitro*.
Tseng et al. (2007) [ABST]	Male mice pups were exposed daily to decaBDE (10,100, 500 or 1,500 mg/kg bw/day) from postnatal day 21 to 70. The results suggested that exposure to decaBDE impaired morphology of hepatocytes but did not affect hepatic enzyme activities and thyroid hormone homeostasis in the adult mouse.
Tseng et al. (2008a)	Investigated the effects on thyroid hormone and hepatic enzyme in male offspring following oral exposure of pregnant mice to decaBDE (10, 500 and 1,500 mg/kg bw/day over gestation days 0 to 17). Effects seen on several endpoints at 1,500 mg/kg bw/day and for one endpoint (decrease in serum triiodothyronine (T3)) at 10 mg/kg bw/day.
Tseng et al. (2008b) [ABST]	Assessed whether prenatal exposure to decaBDE affects the development and hepatic enzyme levels in dams and female offspring mice. Little or no effects appear to have been seen on in the female offspring but the results suggested that exposure of pregnant mice to decaBDE might induce hepatic enzyme activities in the dams.
Van der Ven et al. (2008) (also Van der Ven et al. (2006))	28-Day oral toxicity study in rats designed to detect endocrine and immune modulating effects of decaBDE. Effects seen on weight of seminal vesicles/coagulation gland in males (benchmark dose lower confidence limit) (BMDL) = 0.2 mg/kg bw/day) and decrease enzyme activity in adrenals in females (BMDL = 0.18 mg/kg bw/day).
Viberg et al. (2008)	Neonatal NMRI male mice were orally exposed to 20.1 mg/kg bw of decaBDE on day 3 (? this is not entirely clear from the abstract). The levels of CAMKII, GAP-43 and BDNF in different brain regions were analysed 7 days after exposure. These showed that exposure to decaBDE resulted in significantly increased CaMKII in hippocampus, but not in cortex, a significant increase in GAP-43 in hippocampus, a significant decrease in GAP-43 in cortex and a significant decrease of BDNF in hippocampus, but not in cortex. The results show that exposure to decaBDE affects important brain proteins that are involved in normal maturation of the brain.
Viberg (2009)	Study on the effects on the developmental expression of synaptophysin and tau following exposure of neonatal male mice to decaBDE. Effects on the level of synaptophysin in the developing brain were reported.
Vinggaard et al. (2008)	Screening of 397 chemicals, including brominated flame retardants, for androgen receptor antagonism. It is not clear from the abstract whether decaBDE was included in the study.

Reference	Brief summary of study/findings
Wang et al. (2005a and 2005b)	Quantitative structure-activity relationship for prediction of the toxicity of polybrominated diphenyl ethers.
Wang et al. (2006)	Quantitative structure-activity relationship for prediction of the toxicity of polybrominated diphenyl ethers.
Weiss and Bergman (2006)	Considered the risk from human exposure to PBDEs.
Wu et al. (2008)	Investiged the effect of maternal exposure of rats to decaBDE on the learning and memory ability of offspring rats.
Yang et al. (2008)	Studied the effect of decaBDE on cultured rat hippocampal neurons. The study found that development inhibition in neurons occurred at a concentration of 15 µmol/l, suggesting that decaBDE is a neurotoxicant.

Other reviews and articles on the health effects of polybrominated diphenyl ethers include VCCEP (2008), TERA (2003) and Kucher and Purvis (2004).

Developmental neurotoxicity potential of decaBDE

As a result of EU Member State concerns about a potential neurotoxic effect of decaBDE, a study has recently been carried out by Industry following the OECD 426 Test Guideline (Beck, 2009). The study was carried out using rats exposed to doses of 1, 10, 100 and 1,000 mg/kg body weight/day from gestation day 6 to lactation day 21. The study summary indicates that exposure to decaBDE caused no maternal toxicity, no effects on offspring survival and growth, and no effects on any of the neurobehavioural endpoints studied compared with the control groups at all dosage levels. The study is currently being reviewed by the French competent authority to decide what, if any, further action is necessary.

It should be noted that there are a number of other recently published studies that suggest that decaBDE does have the potential to cause developmental neurotoxic effects. Several important concerns about some of these studies have been identified in a critical review submitted to the Environment Agency by industry. An initial review has also been prepared by the Institute of Environment and Health (Cranfield University) for the Environment Agency as an unpublished internal report. They concluded that many of industry's reservations are valid, highlighting the need to take due consideration of study limitations when considering the various reported findings. *Therefore it is appropriate to wait until the whole data set has been fully evaluated by appropriate experts before the implications for setting a no-observed adverse effect level (NOAEL) for decaBDE are considered.*

6.2 Human biomonitoring and human exposure

A number of new studies have been published that either do or may contain further information on the levels of decaBDE in human samples (e.g. skin, blood serum/plasma, breast milk, etc.), and estimates of human exposure. These are listed below with full details in the section of human health-related references. The information in these papers has not been considered in this evaluation as the focus of this report is on the environmental fate and effects of decaBDE (and it is possible that not all of these papers contain information on decaBDE). In addition, the interpretation of human concentration data is complicated by occupational and consumer exposure.

Akutsu et al. (2007) - dietary intake
Antignac et al. (2008) - maternal and umbilical serum, adipose tissue and breast milk
Antignac et al. (2009) - maternal and umbilical serum, adipose tissue and breast milk
Anderson et al. (2007) - serum
Anderson et al. (2008) - serum
Ashizuka et al. (2007) - dietary intake
Athanasiadou et al. (2008) - serum
Bethune et al. (2006) - plasma levels following consumption of salmon
Bi et al. (2007a) - serum
Bi et al. (2007b) - serum
Brouwers et al. (2009) - occupational exposure
Chao et al. (2007a) - breast milk
Chao et al. (2007b) - breast milk
Christiansson et al. (2008) - serum
Covaci et al. (2008) - liver and adipose tissue
Doucet et al. (2009) - fetal liver and placenta
EWG (2008) - serum (see alsoTable 4.15)
Fäldt et al. (2005) - serum
Fängström et al. (2008) - breast milk
Fernandes et al. (2008) - dietary exposure (see also Table 4.11)
Frederiksen et al. (2008) - placenta
Frederiksen et al. (2009) - review of human exposure
Fukata et al. (2005) - maternal serum, umbilical cord and cord serum
Furst, 2006 - breast milk
Gómara et al. (2007a) - serum and breast milk
Gómara et al. (2007b) - serum, placenta, breast milk
Harrad and Porter (2007) - serum
Jakobsson et al. (2005) - serum
Jareczewska et al. (2006) - serum and breast milk
Johnson-Restrepo et al. (2007) - breast milk

Johnson-Restrepo and Kannan (2009) - estimated human exposure

Jones et al. (2006) - serum

Jones-Otazo et al. (2005) - estimated human exposure

Karlsson et al. (2007) - plasma (see also Table 4.9 and Table 4.10)

Kawashiro et al. (2008) - maternal blood, breast milk and umbilical cord/blood

Knutsen et al. (2008) - serum

Kunisue et al. (2007) - adipose tissue

Leijs et al. (2008) - serum

Li et al. (2005) - adipose tissue

Lorber (2008) - estimated human exposure/body burden, breast milk and serum

Medina et al. (2008) - breast tissue

Meng et al. (2007) - estimated human exposure including via breast milk

Miyata et al. (2005) - breast milk

Murata et al. (2007) - dietary intake

Nomura et al. (2007) - dietary intake (see also Table 4.17)

Ohta et al. (2005) - breast milk and dairy milk

Pérez-Maldonado et al. (2009) - plasma

Petterson et al. (2004) - occupational exposure

Petterson-Julander et al. (2004) - occupational exposure

Polder et al. (2008a) - breast milk

Polder et al. (2008b) - breast milk

Pulkrabová et al. (2009) - adipose tissue

Qu et al. (2007) - serum

Raab et al. (2008) - breast milk

Schecter et al. (2006a) - serum

Schecter et al. (2006b) - serum and breast milk

Schecter et al. (2007a) - liver (fetus/newborns)

Schecter et al. (2007b) - liver (fetus) and dietary intake

Schecter et al. (2008) - breast milk and dietary intake

Schecter et al. (2009) - occupational exposure

Schuhmacher et al. (2009) - breast milk

SFT (2007) - plasma

She et al. (2005a) - breast milk

She et al. (2005b) - breast milk

She et al. (2007) - breast milk

Sjödin et al. (2006) - breast milk

Sjödin et al. (2008) - serum

Stapleton et al. (2008a) - hand to mouth contact

Stapleton et al. (2008b) - serum

Sudaryanto et al. (2008a) - breast milk

Sudaryanto et al. (2008b) - breast milk

Sudaryanto et al. (2008c) - human exposure assessment

Takasuga et al. (2006) - serum

Thomsen et al. (2005) - serum and breast milk

Thomsen et al. (2007a) - serum

Thomsen et al. (2007b) - serum and dietary intake

Thomsen et al. (2008) - serum

Thuresson et al. (2004) - serum

Thuresson et al. (2006) - serum

Toms et al. (2007a) - breast milk

Toms et al. (2007b) - breast milk

Toms et al. (2008) - serum

Toms et al. (2009) - breast milk

Wang et al. (2008) - breast milk and dietary exposure

Wei-Yu et al. (2007) - serum

Wu et al. (2007) - breast milk (linked to dust (see Table 4.10) and diet)

Yuan et al. (2008) - serum

Zalko et al. (2007) - adipose tissue, breast milk, maternal serum and cord serum

Zhang et al. (2008) - placenta tissue

Zhao et al. (2008) - human hair

Zhu et al. (2009) - breast milk

Other studies related to human biomonitoring that are of limited relevance to decaBDE (for example where it is clear from the abstract that the paper contains information on lower brominated congeners only) are listed in the other references of limited relevance at the end of this report.

6.3 Implications for secondary poisoning assessment

A predicted no effect concentration (PNEC) for secondary poisoning for decaBDE of 833 mg/kg food has been derived based on a long-term NOAEL of 1,120 mg/kg bw/day and an assessment factor of 30 (EC, 2002 and ECB, 2007). New mammalian toxicity data are available for decaBDE but the implications of these data for the NOAEL for decaBDE are currently unknown. Therefore it is appropriate to wait until the whole data set has been fully evaluated by appropriate experts before a revised PNEC for secondary poisoning for decaBDE is considered.

7 Assessment against the REACH Article 57 criteria

The following sections summarise the most relevant studies in the context of the REACH Article 57 and Annex XIII criteria, in accordance with the guidance provided by ECHA (2008).

7.1 Persistence

Based on the information in the original risk assessment reports (EC 2002, ECB 2004 and ECB 2007) decaBDE is already considered to be very persistent in the environment, meeting the vP criterion of REACH Annex XIII. The half-life is substantially longer than 180 days in freshwater sediment, as indicated by a complete lack of degradation in a simulation test with anaerobic sediment after 32 weeks (8 months or 224 days).

The new information supports this conclusion. For example, a sediment microcosm study by Tokarz et al. (2008) showed only very limited degradation over ten months in the laboratory. Further monitoring data show that the substance is widely distributed in the environment, and sewage sludge and estuarine sediment concentrations in Europe have not yet declined significantly following the implementation of voluntary industry emission reduction measures. In addition, one study shows increasing trends in sediment levels in Switzerland over the period 1974-2001 (Kohler et al., 2008), although it is acknowledged that much of the time trend data is inconclusive as to whether the levels in Europe are still currently increasing or not.

There is also further evidence of long-range transport via the atmosphere (e.g. Su et al., 2007a & 2007b; Cheng et al., 2007). However, a study at the McMurdo research base in Antarctica by Hale et al. (2008) shows that there can be significant local sources even in supposedly pristine environments and, given the widespread use of decaBDE in articles, local sources cannot totally be ruled out in most of these studies. DecaBDE in the atmosphere is likely to be associated mainly with atmospheric particulates and so would be removed by atmospheric deposition processes, limiting the potential for long-range transport. However, although the study by Wilford et al. (2008) found that the majority of of decaBDE in the indoor air was associated with particulates with diameters > 3 µm, a small fraction was associated with either the fine particulate fraction (diameters <1 µm) or the vapour phase. DecaBDE in these latter two phases could be subject to transport over longer distances than that associated with the larger particulates. Scheringer (2008) noted that modelling the long-range transport potential of decaBDE is difficult but suggested that decaBDE could potentially be transported over long distances during episodic transport events taking place during dry periods, where the potential for rapid deposition of particle-bound or aerosol-bound decaBDE is reduced. In addition Schenker et al. (2008a) have indicated that photodegradation in the atmosphere is an important consideration in determining the long-range transport potential for decaBDE. Overall, although there is evidence that decaBDE is present in the environment in remote regions there is still some uncertainty over the long-range transport potential for decaBDE.

The slow rate of degradation could nevertheless lead to the formation of other substances that are of concern (including some that are considered to meet the PBT/vPvB criteria) and this is considered in Section 7.4.

7.2 Bioaccumulation

The bioaccumulation criteria are based on a measured BCF for an aquatic organism being above 2,000 l/kg (B) or above 5,000 l/kg (vB). A BCF of around 4 l/kg was measured for decaBDE but this result is invalid (as discussed in ECB, 2007). No other fish BCF data are available, so alternative evidence needs to be considered.

DecaBDE is detected in a wide range of wildlife species from all parts of the world, particularly in terrestrial organisms, including top predators and sensitive stages of the life cycle (e.g. birds' eggs). Whilst many of the measured levels are in the low µg/kg (wet weight) range, the ultimate level of accumulation that is possible for such a persistent substance is unknown – there is evidence that tissue levels in some birds are continuing to increase (e.g. Gauthier et al., 2008 and Chen et al., 2008b; although the reliability of these findings is difficult to assess at present). ECB (2007) reported findings of levels up to 122 µg/kg wet weight in birds collected in China and levels of up to 420 µg/kg wet weight were detected in birds' eggs in the United States by Chen et al. 2008b.

The BCF value measures the uptake into an organism (fish) through water (dissolved phase) exposure only. As decaBDE strongly adsorbs to suspended matter, it is likely that aquatic organisms will be exposed through both the dissolved phase and through ingestion of particulates, including prey items (e.g. algae, zooplankton, invertebrates, etc.). Therefore a better measure of the accumulation potential for decaBDE would be the overall bioaccumulation factor (BAF) that incorporates all possible routes of exposure.

The tentative conclusion of ECB (2007) was that a BAF of the order of at least 1,000 l/kg is appropriate for decaBDE for aquatic species. This was based on evidence from monitoring data - dissolved concentrations of decaBDE in the environment are usually at the ng/l level or less (e.g. see Section 4.2.1 of this report, EC (2002) and ECB (2004 and 2007)), whilst fish concentrations tend to be in the µg/kg wet weight range (e.g. see Section 4.2.5, EC (2002) and ECB (2004 and 2007)). The new data provide additional evidence that the overall BAF for decaBDE in aquatic organisms is relatively high. For example, the study by Bogdal et al. (2007) discussed in Section 4.3.2 allows a BAF of at least 3×10^5 to be estimated for fish (although this interpretation is somewhat simplistic and a worst case approach). Similarly, the study by Riva et al. (2007) reported in Section 4.3.3 would suggest a BCF or BAF of at least 1,000 l/kg. Finally, the Wang et al. (2007) study in Section 4.3.3 allows a BAF of around 500 l/kg to be predicted.

Although none of these data are considered sufficiently robust to determine the actual BCF/BAF for decaBDE (and indeed the Annex XIII criteria are currently based on BCF rather than BAF values), they do suggest that the accumulation of decaBDE in wildlife is significant. In addition, the determination of BCF or BAF data based on decaBDE concentrations alone could *underestimate* its actual uptake potential, since any initial metabolic debromination to form nonaBDE congeners would be considered to be a 'removal' process. Whilst the importance of this metabolic pathway appears to vary across species, given the likely similarity in toxicity between nonaBDEs and decaBDE, a more realistic approach would be to consider the total concentration of nonaBDEs plus decaBDE in each medium. This *could* indicate higher levels of accumulation than consideration of decaBDE alone.

Several new studies also investigate the biomagnification of decaBDE. The majority of these new data suggest that decaBDE has a BMF <1 (although there is one exception to this). Overall, taking these new data together with the information available previously in EC (2002) and ECB (2004 and 2007), the overall weight of evidence is that decaBDE does not biomagnify. However, it should be noted that there is evidence that the metabolism of decaBDE is species specific, and this may at least partly explain some of the variability in the results obtained in the various studies.

On the basis of this analysis, decaBDE does not meet the Annex XIII bioaccumulation criteria based on BCF considerations. Whilst it does not appear to biomagnify, a BAF in aquatic organisms somewhere in the region of 1,000 l/kg or above seems to be appropriate.

7.3 Toxicity

DecaBDE is not classified for environmental or human health hazards, and ECB (2007) concluded that the toxicity criteria are not triggered by the available data from standard studies. The limited new data available for aquatic organisms do not change this conclusion. Further studies have been carried out to investigate the developmental neurotoxicity of decaBDE. These will need to be reviewed in detail by appropriate experts to clarify the level of concern for this endpoint. It is not yet known whether the human health classification will be affected and so it is not possible to currently conclude on the Annex XIII toxicity criterion for decaBDE.

7.4 Degradation products of concern

Whilst decaBDE is highly persistent, a large number of uncertainties remain about its environmental fate and behaviour. In particular, questions about the significance of the formation of hazardous breakdown products has been an ongoing issue for many years. These possible hazardous breakdown products include substances that meet the PBT/vPvB criteria (such as hexaBDE, which is persistent and has a BCF of 5,640 l/kg (EC, 2000) and potentially some brominated dibenzofuran congeners) or have other properties of concern[29]. Previous European risk assessments (e.g. ECB, 2007) concluded that the significance of such degradation has not been established, and an environmental monitoring programme is being performed over a 10 year time frame to investigate this further.

This report considers the latest results of this conclusion (i) monitoring programme, as well as new published data. The main findings are summarised below.

1) There is strong evidence from both laboratory and field data (see Section 4.3.1) that metabolism of decaBDE to nonaBDEs, octaBDEs and probably heptaBDEs occurs in a number of species (e.g. Stapleton et al., 2006; La Guardia et al., 2007; ECB (2007) also reports a study with European Starlings (*Sturnus vulgaris*) that provides evidence for metabolism to nona- and octaBDE congeners). HexaBDEs may also be formed in some fish species (e.g. Nyholm et al., 2008a). The yield of these metabolites is generally low (typically <5 per cent of the dosed decaBDE in the various studies), but several studies indicate that other, as yet unidentified metabolites, may also be formed. For example, Huwe and Smith (2007) found that around 42 per cent of the decaBDE dose in rats after 21 days exposure had been converted to unknown metabolites. Hydroxylated derivatives of polybrominated diphenyl ethers

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[29] For example, the commercial octabromodiphenyl ether product has been classified as toxic to reproduction Category 2 (R61 May cause harm to the unborn child) and toxic to reproduction Category 3 (R62 Possible risk of impaired fertility) and so would meet the Annex XIII criteria for a substance of very high concern. The main components of the octabromodiphenyl ether product tested were hepta- and octaBDEs (see Appendix 1) and heptaBDE has recently been added to the Stockholm Convention on persistent organic pollutants (POPs).

have been found in some metabolism studies (for example see EC (2002) and ECB (2007)) and other as yet unidentified degradation/metabolism products could also exist. The fate and behaviour of these derivatives have not been investigated fully in either the current or previous risk assessment reports. They would be expected to be more soluble than the corresponding polybrominated diphenyl ether and hence potentially more available for uptake in the environment. However, it could also be expected that the hydroxyl group would make them more prone to biodegradation and excretion/elimination from organisms (the latter would reduce their potential for bioaccumulation). This adds further uncertainty to the evaluation of the environmental effects of decaBDE as, for example, if the metabolite burden were increasing with time, toxicity studies of shorter duration may be less relevant than longer-term studies. However, it is recognised that it is difficult to make firm statements about the likely consequences of the formation of these other metabolites.

2) The available new data show that decaBDE occurs widely in indoor and outdoor air and dust. Levels up to 520 mg/kg have been measured in indoor dust from the United Kingdom. The study of Wilford et al. (2008) is particularly relevant, since it provides evidence that the decaBDE found in indoor air is predominantly associated with particles formed by abrasion of articles, indicating the importance of this emission source. Whilst the congener profile found in this study is suggestive of debromination of decaBDE to nonaBDEs in dust samples, Stapleton and Dodder (2008) provide further evidence that heptaBDEs can be formed by photodegradation of decaBDE adsorbed onto dust. This appears to be an environmentally relevant degradation mechanism, and although it is difficult to determine the extent of exposure of such dust to light in the environment, such exposure is likely to occur in the atmosphere and on windows, etc. It is therefore possible that decaBDE on indoor dust is a source of heptaBDE congeners in the wider environment.

3) A large number of studies show that a range of hazardous PBDEs can be formed via numerous mechanisms in the laboratory (see EC (2002) and ECB (2004 and 2007) for examples). Evidence of extensive debromination of decaBDE in the presence of natural reductants (e.g. vitamin B_{12}) can now be added to this list (Tokarz et al., 2008). Most laboratory experiments are of relatively short duration and use conditions that are not representative of those in the environment, which makes it difficult to estimate the likely rate or yield of any such reactions. Given the very long periods that may be needed before significant amounts of lower PBDE congeners (and other hazardous substances) can be formed under natural conditions, it may prove very difficult to measure reliable rates in the laboratory in any case. At the same time, such reactions *appear to be occurring* in the environment, as demonstrated by the results of field monitoring.

4) Of particular interest is the finding by Tokarz et al. (2008) that hexaBDE could be formed from the degradation of decaBDE over a three year time frame using sediment microcosms (although it is recognised that there are some limitations with this part of the study). The implication is that the formation of lower PBDE congeners with PBT/vPvB properties is so slow in the environment that it might only become apparent after decades rather than years. Given the small amounts likely to be formed, sediment degradation might not be very significant. This appears to be substantiated by sediment core studies that have investigated changing

congener patterns over long time periods (generally starting from around 1975), which provide little or no evidence of degradation (e.g. Kohler et al. (2008) and Moon et al. (2007c and 2007d); other studies are summarised in ECB (2004 and 2007) and EC (2002)).

5) Nevertheless, it should also be noted that a specific pentabromodiphenyl ether congener (BDE-126) has been recently found in sediment in the conclusion (i) monitoring programme, at levels up to about 0.3 µg/kg dry weight (the actual concentrations could not be quantified). This congener has also been found in sewage sludge samples from 2007, at ~0.1 µg/kg dry weight at both sites investigated. This is an important finding because this congener was chosen as a marker of abiotic degradation. These results therefore suggest that pentaBDE congeners *are* being formed in the environment (though not necessarily in sediment), presumably as a result of degradation of decaBDE. In addition, although the sediment core study by Kohler et al. (2008) found that the congener profile present in the sediment core did not change significantly from year to year, there was some evidence for another possible degradation product (BDE-202, an octabromodiphenyl ether congener that is not usually found in commercial products) in some of the samples (although, given that only a limited set of commercial products was analysed, it cannot totally be ruled out that BDE-202 arises from the use of octaBDE products (either current or historic)). BDE-202 has been found to be present in some organisms in recent studies (e.g. Holden et al. 2009).

5) Further information is also available on the formation and release of brominated dibenzofurans and brominated dibenzo-*p*-dioxins, including from photodegradation of decaBDE adsorbed to plastic particles (e.g. Kajiwara et al., 2008a). Some of these substances have PBT or vPvB properties. However, for some of these studies few details of the analytical methodologies used are available and the identities of the polybrominated dibenzofurans and dibenzo-*p*-dioxins found are not always clear. Much of the earlier available information on the formation of polybrominated dibenzofurans and dibenzo-*p*-dioxins from the use and pyrolysis of decaBDE has been summarised in the previous risk assessment reports, particularly EC (2002). The new data are generally consistent with the previous information. Although it is clear that polybrominated dibenzofurans and dibenzo-*p*-dioxins are present at low levels in some samples of plastic shredder waste, and have the potential to be formed during combustion or pyrolysis processes involving plastic waste, it is difficult to ascribe their presence or formation solely to the use of decaBDE. Therefore it is not possible to quantify the risks from decaBDE for these sources. It should also be noted that the requirements of the RoHS Directive effectively mean that decaBDE can no longer be used in new polymers for electrical and electronic equipment in the EU. Therefore the amount of decaBDE present in the waste polymer streams in the UK and EU should gradually reduce with time in the future.

The persistence of decaBDE is such that the formation of even small quantities of hazardous substances in percentage terms is undesirable, since the actual amounts may become significant over long time frames. Whilst the strongest evidence is for the formation of nona- to octaBDE congeners (some of which appear to be both very persistent and toxic – see Appendix 1), there is now fresh evidence that hexa- and even pentaBDEs (which clearly have PBT/vPvB properties) may be formed (for example the Tokarz et al. (2008) study and the finding of BDE-126 in the conclusion (i) monitoring programme).

Finally, it should also be noted that new manufacturers may be producing products that are not as pure as those made by the main European suppliers. For example, Chen et al. (2007) reported that the octaBDE and nonaBDE content of two commercial decabromodiphenyl ether products from China was in the range 8.2 to 10.4 per cent. This may also need to be taken into account in any further assessment.

8 Summary

DecaBDE is very persistent and widely dispersed in the environment, including sediment, soil and many types of biota. This means that its properties need careful consideration. Whilst a PEC/PNEC risk has not been established, and decaBDE does not itself meet the PBT/vPvB criteria of Annex XIII of REACH, there continue to be concerns related to its presence in food chains (including top predators) and the unknown significance of degradation to more hazardous substances. It is possible that some of the uncertainties surrounding this substance may be addressed by the results of on-going investigations (or new studies). However, new evidence suggests that detection of hazardous degradation products (e.g. hexaBDE congeners which themselves have vPvB properties, and heptaBDE which has recently been added to the Stockholm Convention on persistent organic pollutants (POPs)) in sediment, sewage sludge and biota may be linked to emissions of decaBDE, although it is still difficult to estimate the rates and amounts of formation.

Within the context of environment levels, it is worth considering the impact of the Voluntary Emissions Control and reduction Action Programme (VECAP). This was set up five years ago by the manufacturing industry, with approval of the EU Member State Competent Authorities, to promote new codes of good practice for the use of decaBDE in the plastics and textile industries. The aim was to ensure improved emission control for industrial point sources. Although data are only available from the conclusion (i) monitoring programme for a couple of years, there is little or no evidence of a decline in levels in any of the media sampled. This suggests that the VECAP has not made any substantial immediate impact on the general levels of decaBDE in the environment, although it may have prevented further increases (it is recognised that it has been effective at reducing the emissions from certain point sources). In addition, substantial stocks of the substance are present in treated articles that are still in use (which may be increasing year on year), and the VECAP was not designed to address emissions from this source (e.g. dust arising from textile wear).

It is also worth considering the industry monitoring programme under Commission Regulation (EC) No. 565/2006, which still has several years to run before it is complete. The original purpose of the programme was to (a) measure the levels of PBDE congeners being found in the environment (to try to identify patterns that might suggest degradation or not) and (b) assess any trends which could give rise to a risk in the future. This was always going to be difficult because of ongoing contributions from emissions of the commercial penta- and octabromodiphenyl ether products, despite their removal from the market. Given the low levels of some of the congeners found and the high level of variability in some sample types, it is in any case rather difficult to detect any patterns or trends other than very striking ones. The change in use pattern might also be a confounding factor now that one of the major uses of decabromodiphenyl ether has been restricted (in electrical and electronic equipment). The possibility of lower purity material being imported from developing countries cannot be ruled out, and this would also obscure any relationship between one congener and another. Therefore the continued utility of the monitoring programme as a scientific and policy support tool might need to be re-evaluated.

These factors should be considered in any further prioritisation of decaBDE for regulation under REACH.

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WILFORD, B.H., THOMAS, G.O., JONES, K.C., DAVISON, B., AND HURST, D.K., 2008. Decabromodiphenyl ether (deca-BDE) commercial mixture components, and other PBDEs, in airborne particulates at a UK site. Environmental International, 34 (3), 412-419.

WONG, M.H., WU, S.C., DENG, W.J., YU, X.Z., LUO, Q., LEUNG, A.O., WONG, C.S., LUKSEMBURG, W.J. AND WONG, A.S., 2007. Export of toxic chemicals – a review of the case of uncontrolled electronic-waste recycling. Environmental Pollution, 149 (2), 131-140.

WU, N., HERRMANN, T., PAEPKE, O., TICKNER, J., HALE, R., HARVEY, E., LA GUARDIA, M., MCCLEAN, M.D. AND WEBSTER, T.F., 2007. Human exposure to PBDEs: Associations of PBDE body burdens with food consumption and house dust concentrations. Environmental Science and Technology, 41 (5), 1584-1589.

WURL, O., LAM, P.K.S. AND OBBARD, J.P., 2006. Occurrence and distribution of polybrominated diphenyl ethers (PBDEs) in the dissolved and suspended phases of the sea-surface microlayer and seawater in Hong Kong, China. Chemosphere, 65, 1660-1666.

XIANG, C-H., LUO, X-J., CHEN, S-J., YU, M., MAI, B-X. AND ZENG, E.Y., 2007. Polybrominated diphenyl ethers in biota and sediments of the Pearl River Estuary, South China. Environmental Toxicology and Chemistry, 26 (4), 616-623.

XU, H-Y., ZOU, J-W., YU, Q-S., WANG, Y-H., ZHANG, J-Y. and JIN, H-X., 2007. QSPR/QSAR models for prediction of the physicochemical properties and biological activity of polybrominated diphenyl ethers. Chemosphere, 66 (10), 1998-2010.

YANG, Z.Z., ZHAO, X.R., QIN, Z.F., FU, S., LI, X.H., QIN, X.F., XU, X.B. AND JIN, Z.X., 2009. Polybrominated diphenyl ethers in mudsnails (*Cipangopaludina cahayensis*) and sediments from an electronic waste recycling region in South China. Bulletin of Environmental Contamination and Toxicology, 82 (2), 206-210.

YEO, H-Y., CHO, K-C. AND CHUN, M-Y., 2006. Concentration and characteristic of PBDEs in pine needle and soil of Ansung-City. English abstract only available from the Korean Medical Database.

YUN, S.H., ADDINK, R., MCCABE, J.M., OSTASZEWSKI, A., MACKENZIE-TAYLOR, D., TAYLOR, A.B. AND KANNAN, K., 2008. Polybrominated diphenyl ethers and polybrominated biphenyls in sediment and floodplain soils of the Saginaw River Watershed, Michigan, USA. Archives of Environmental Contamination and Toxicology, 55 (1), 1-10.

ZENG, X., MASSEY SIMONICH, S.L., ROBROCK, K.R., KORYTÁR, P., ALVAREZ-COHEN, L. AND BAROFSKY, D.F., 2008. Development and validation of a congener-specific photodegradation model for polybrominated diphenyl ethers. Environmental Toxicology and Chemistry, 27 (12), 2427-2435.

ZENNEGG, M., BRÄNDII, R.C., KUPPER, T., BURCHELI, T.D., GUJER, E., SCHMID, P., STADELMANN, F.X. AND TARRADELLAS, J., 2005. PCDD/Fs, PCBs, PBDEs, TBBPA and HBCD in compost and digestate. Organohalogen Compounds, 67, 1040-1043.

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ZHOU, J., JIANG, W., DING, J., ZHANG, X. AND GAO, S., 2007. Effect of Tween 80 and beta-cyclodextrin on degradation of decabromodiphenyl ether (BDE-209) by white rot fungi. Chemosphere, 70 (2), 172-177.

ZLÁMALÍKOVÁ, J., DEMNEROVÁ, K., MACKOVÁ, M., HAJŠLOVÁ, J., PULKRABOVÁ, J., HRÁDKOVÁ, P., NÁPRAVNÍKOVÁ, M., MACEK, T. AND STIBOROVÁ, H., 2008. Plant uptake of polybrominated diphenyl ethers from contaminated sludge. Organohalogen Compounds, 70, 2232-2235.

ZOU, M-Y., RAN., Y., GONG, J., MAI, B-X. AND ZENG, E.Y., 2007. Polybrominated diphenyl ethers in watershed soils of the Pearl River Delta, China: Occurrence, inventory, and fate. Environmental Science and Technology, 41 (24), 8262-8267.

Other papers that have not been reviewed

There are a number of conferences on brominated flame retardants held each year. For these, short abstracts of the presentations are often available. The papers below are papers for which the abstract is not yet available or could not be located. In addition to this a large number of papers containing information on polybrominated diphenyl ethers are published in the literature each week and it is not possible, for a review of this type, to be fully up to date. As noted in Section 1.1, an initial literature search for this report was undertaken in June 2008 and a further targeted literature search was undertaken in May 2009. Only the most relevant papers from the 2009 targeted literature search have been included in this review. The remaining papers from the 2009 literature search, and any other papers we are aware of at the time of writing that may contain information on decaBDE, are given below.

ACKERMAN, L.K., SCHWINDT, A.R., SIMONICH, S.L., KOCH, D.C., BLETT, T.F., SCHRECK, C.B., KENT, M.L. AND LANDERS, D.H., 2008. Atmospherically deposited PBDEs, pesticides, PCBs, and PAHs in wester U.S. National Park fish: concentrations and consumption guidelines. Environmental Science and Technology, 42, 2334-2341. [further information on levels in the environment]

ALLEN, J.G., MCCLEAN, M.D., STAPLETON, H.M. AND WEBSTER, T.F., 2008. Critical factors in assessing exposure to PBDEs via house dust. Environment International, 34 (8), 1085-1091. [further information on levels in dust]

ALSHURAFA, M., KHAN, J., KIM, M., ALAEE, M., STEER, H., PACEPAVICIUS, G. AND WAINMAN, B., 2008. Polybrominated diphenyl ethers in rats at McMaster University animal quarters and exposure assessment. Organohalogen Compounds, 70, 1849-1852. [background levels in laboratory animals]

ALVAREZ, D.A., CRANOR, W.L., PERKINS, S.D., SCHROEDER, V.L., IWANOWICZ, L.R., CLARK, R.C., GUY, C.P., PINKNEY, A.E., BLAZER, V.S. AND MULLICAN, J.E., 2008. Reproductive health of bass in the Potomac, USA drainage: Part 2. Seasonal occurrence of persistent and emerging organic contaminants. Environmental Toxicology and Chemistry, in press. [further information on levels in the environment; it is not clear from the abstract which flame retardants are included]

ARNOLD, R.G., TESKE, S., TOMANEK, M., ENGSTROM, J., LEUNG, C., ZHANG, J., BANIHANI, Q., QUANDRUD, D., ELA, W.P. AND SÁEZ, A.E., 2008. Fate of polybrominated diphenyl ethers during wastewater treatment/polishing and sludge stabilization/disposal. Annals of the New York Academy of Sciences, 1140, 394-411. [further information on levels in the environment and fate during waste water treatment]

ATHANASIADOU, M. AND BERGMAN, Å, 2008. Insamling av bröstmjölksprover från Stockholm, Göteborg, Lund och Umeå samt analyser av insamlade bröstmjölksprover. Resultat från 2007 års arbete. Rapport till Naturvårdsverket. [in Swedish; reported in Kemi (2009)]

BENGTSON NASH, S.M., POULSEN, A.H., KAWAGUCHI, S., VETTER, W. AND SHLABACH, M., 2008. Persistent organohalogen contaminant burdens in Antarctic krill (*Euphausia superba*) from the eastern Antarctic sector: a baseline study. Science of the Total Environment, 407 (1), 304-314. [further information on levels in the environment]

BETTERMAN, S., CHEMYAK, L., JIA, C., Godwin, C. AND CHARLES, S., 2008. Mass balance of PBDEs in air, floor dust, and ventilation filters in a new office building. BFR 2008 Workshop, Institute of Ocean Sciences, Sidney, BC, June 3-4, 2008. [further information on levels in the environment]

BETTS, K., 2009. New data suggest PBDE byproducts are ubiquitous in U.S. waters. Environmental Science and Technology, in press. [news article discussing the possible occurrence of hydroxylated PBDEs and brominated dibenzo-*p*-dioxins in water]

BOGDAL, C., SCHMID, P., KOHLER, M., MÜLLER, C.E., IOZZA, S, BUCHELI, T.D., SCHERINGER, M. AND HUNGERBÜHLER, K, 2008. Sediment record and atmospheric deposition of brominated flame retardants and organochlorine compounds in Lake Thun, Switzerland: lessons from the past and evaluation of the present. Environmental Science and Technology, 42 (18), 6817-6822. [further information on levels in the environment and atmospheric deposition]

BORGHESI, N., CORSOLINI, S. AND FOCARDI, S., 2008. Levels of polybrominated diphenyl ethers (PBDEs) and organochlorine pollutants in two species of Antarctic fish (*Chinodraco hamatus* and *Trematomus bernacchii*). Chemosphere, 73 (2), 155-160. [further information on levels in the environment]

CANUEL, E.A., LERBERG, E.J., DICKHUT, R.M., KUEHL, S.A., BIANCHI, T.S. AND WAKEHAM, S.G., 2009. Changes in sediment and organic carbon accumulation in a highly-disturbed ecosystem: The Sacramento-San Joaquin River Delta (California, USA). Marine Pollution Bulletin, in press. [further information on levels in the environment]

CETIN, B. AND ODABASI, M. , 2008. Atmospheric concentrations and phase partitioning of polybrominated diphenyl ethers (PBDEs) in Izmir, Turkey. Chemosphere, 71, 1067-1078. [further information on levels in the environment; similar to Cetin and Odabasi (2007a and 2007b)]

CHEAIB, Z., GRANDJEAN, D., KUPPER, T., AND DE ALENCASTRO, L.F., 2009. Brominated flame retardants in fish of Lake Geneva (Switzerland). Bulletin of Environmental Contamination and Toxicology, 82 (4), 522-527. [further information on levels in the environment]

CHEN. D.H., LI. L.P., BI, X.H., ZHAO, J.P., SHENG, G.Y. AND FU, J.M., 2008. PBDEs pollution in the atmosphere of a typical E-waste dismantling region. Huan Jing Ke Xue, 29 (8), 2105-2110. [further information on levels in the environment; article in Chinese]

CHEN, D., BI, X., ZHAO, J., CHEN, L., TAN, J., MAI, B., SHENG, G., FU, J. AND WONG, M., 2009. Pollution characterization and diurnal variation of PBDEs in the atmosphere of an E-waste dismantling region. Environmental Pollution, 157 (3), 1051-1057. [further information on levels in the environment]

CHEN, L. HUANG, Y., PENG, X., XU, Z., ZHANG, S., REN, M., YE, Z. AND WANG, X., 2009. PBDEs in sediments of the Beijiang River, China: Levels, distribution, and influence of total organic carbon. Chemosphere, 76 (2), 226-231. [further information on levels in the environment]

CHEN, S-J., MA, Y-J., WANG, J., CHEN, D., LUO, X-J. AND MAI, B-X., 2009. Brominated flame retardants in children's toys: concentration, composition, and children's exposure and risk assessment. Environmental Science and Technology, 43 (11), 4200-4206. [further information on potential sources of exposure; human health related]

CHERNYAK, S. AND BATTERMAN, S., 2008. Trends and mass balance of flame retardant chemicals in a new building. Organohalogen Compounds, 70, 693-696. [further information on levels in the indoor environment]

CHERNYAK, S., KONOPLEV, A., BATTERMAN, S., KOCHETKOV, A., PASYNKOVA, E., SAMSONOV, D. AND JIA, C., 2008. PBDEs in ambient and indoor air in four locations in the Russian Federation. Organohalogen Compounds, 70, 2163-2166. [further information on levels in the environment]

CHEUNG, K.C., ZHENG, J.S., LEUNG, H.M. AND WONG, M.H., 2008. Exposure to polybrominated diphenyl ethers associated with consumption of marine and freshwater fish in Hong Kong. Chemosphere, 70 (9), 1707-1720. [further information on levels in the environment and dietary exposure]

CHOI, K.I., LEE, S.H. AND OSAKO, M., 2009. Leaching of brominated flame retardants from TV housing plastics in the presence of dissolved humic matter. Chemosphere, 74 (3), 460-466. [further information on leaching from polymers]

CHRISTIANSSON, A., HOVANDER, L., ATHANASSIADIS, I., JAKOBSSON, K. AND BERGMAN, A., 2008. Polybrominated diphenyl ethers in aircraft cabins – a source of human exposure? Chemosphere, 73 (10), 1654-1660. [further information on levels in indoor air]

CIESIELSKI, T., JENSSEN, B.M., PEDERSEN, H.C., BAEK, K., SKAARE, J.U. AND RINGSBY, T.H., 2008. Decabrominated diphenyl ether in ptarmigans and sparrows in Norway. Organohalogen Compounds, 70, 1181-1184. [further information on levels in the environment]

CINCINELLI, A., MARTELLINI, T., DEL BUBBA, M., MISURI, L., LASCHI, S., GIANNONI, M. AND LEPRI, L., 2008. Distribution and levels of brominated diphenyl ethers in sewage sludge in Italy. Organohalogen Compounds, 70, 2276-2279. [further information on levels in the environment]

CLARKE, B., PORTER, N., SYMONS, R., MARRIOTT, P., ADES, P., SEVENSON, G. AND BLACKBEARD, J., 2008. Polybrominated diphenyl ethers and polybrominated biphenyls in Australian sewage sludge. Chemosphere, 73 (6), 980-989. [further information on levels in sewage sludge]

COVACI, A., ROOSENS, L., DIRTU, A.C., WAEGENEERS, N., VAN OVERMEIRE, I., NEELS, H. AND GOEYENS, L., 2008. Brominated flame retardants in Belgium home-produced eggs: Levels and contamination sources. Science of the Total Environment, in press. [further information on levels in the environment]

DANON-SCHAFFER, M.N., GRACE, J.R. AND IKONOMOU, M.G., 2008. PBDE concentrations in landfill leachates from across Canada. BFR 2008 Workshop, Institute of Ocean Sciences, Sidney, BC, June 3-4, 2008. [further information on levels in the environment]

DANON-SCHAFFER, M.N., GRACE, J.R. AND IKONOMOU, M.G., 2008. PBDEs in waste disposal sites from northern Canada. Organohalogen Compounds, 70, 365-368. [further information on levels in the environment]

DE LA TORRE, A., CONCEJERO MORENO, M.A., SANZ, P. AND MARINÉZ, M.A., 2008. Waste characterization: Occurrence and concentration of PCDD/Fs, PCBs, PBDEs and HCB. Organohalogen Compounds, 70, 1776-1779. [further information on levels in the environment]

DE LA TORRE, A., SANZ, P. AND MARINEZ, M.Á., 2008. Presence of PBDEs and DBDPE in leachate and compost from different Spanish landfills. Organohalogen Compounds, 70, 1958-1961. [further information on levels in the environment]

DE WIT, C., THURESSON, K. AND BJORKLUND, J., 2008. Tri- to decabrominated diphenyl ethers and HBCD in indoor air and dust from Stockholm microenvironments. Organohalogen Compounds, 70, 846-849. [further information on levels in the environment]

DEL VENTO, S., CODLING, G., JONES, K.C. AND HALSALL, C.J., 2008. The role and significance of the Arctic for the cycling and fate of persistent organic pollutants (ARCPOP). Organohalogen Compounds, 70, 1228-1231. [decaBDE appears to be included in the study but no specific results are given]

DING, J., ZHOU, J., JIANG, W.Y. AND GAO, S.X., 2008. Aerobic microbial degradation of polybrominated diphenyl ethers. Huan Jing Ke Xue, 29 (11), 3179-3184. [article in Chinese]

DIRTU, A. AND COVACI, A., 2008. Daily intake of OCPs, PCBs and PBDEs from food consumption and indoor dust ingestion in Romania. Organohalogen Compounds, 70, 562-565. [further information on levels in the environment]

Environment Canada, 2009. Draft "State of Science Report on the Bioaccumulation and Biotransformation of decabromodiphenyl ether. Environment Canada, March 2009. [review; this has been checked for additional references]

ERDOĞRUL, O., 2008. Pesticide residues in liquid pekmez (grape molasses). Environmental Monitoring and Assessment, 144 (1-3), 323-328. [further information on levels in the environment/food]

ERICSON, I., VAN BAVEL, B. AND LINDSTRÖM, G., 2008. Resultatrapport för projecktet: Screening av humanvävnad. Rapport till Naturvårdsverket, 2008-03-31. [in Swedish; reported in Kemi (2009)]

FERNANDES, A., MORTIMER, D., GEM, M., DICKS, P., SMITH, F., WHITE, S. AND ROSE, M., 2008. Brominated contaminants (PBDD/FS and PBDEs) in shellfish. Organohalogen Compounds, 70, 1024-1027. [further information on levels in the environment]

FU, S, YANG, Z.Z., ZHANG, L., LI, K. AND XU, X.B., 2009. Composition, distribution, and characterization of polybrominated diphenyl ethers in sandstorm depositions in Beijing, China. Bulletin of Environmental Contamination and Toxicology, in press. [further information on levels in the environment]

GEARHART, J. AND POSSELT, H., 2006. BFRs in vehicles. 8th Annual Workshop on BFRs in the Environment, Ontario Ministry of the Environment, Toronto, ON, June 27-29, 2006. [further information on levels in the environment]

GEBBINK, W., SONNE, C., DIETZ, R., KIRKEGAARD, M., RIGET, F.F., BORN, E.W., MUIR, D.C.G. AND LETCHER, R.J., 2006. Polybrominated diphenyl ethers and degradation products: Bioaccumulation and biotransformation in polar bears and ringed seals from East Greenland. 8th Annual Workshop on BFRs in the Environment, Ontario Ministry of the Environment, Toronto, ON, June 27-29, 2006. [further information on levels in the environment]

GEBBINK, W.A., SONNE, C., DIETZ, R., KIRKEGAARD, M., RIGET, F.F., BORN, E.W., MUIR, D.C. AND LETCHER, R.J., 2008. Tissue-specific congener composition of organohalogen and metabolite contaminants in East Greenland polar bears (*Ursus maritimus*). Environmental Pollution, 152 (3), 621-629. [further information on levels in the environment]

GOODBRED, S.L., BRYANT, W.L., ROSEN, M.R., ALVAREZ, D. AND SPENCER, T., 2009. How useful are the "other" semipermeable membrane devices (SPMDs); the mini-unit (15.2 cm long)?. Science of the Total Environment, 407 (13), 4149-4156. [further information on levels in the environment; not clear from the abstract which flame retardants were included]

GRÜMPING, R., PETERSEN, M., NEUGEBAUER, F. AND OPEL, M., 2008. Levels of dioxins, PCBs, BFRs, PFCs and organotins in fishery products from Latvia. Organohalogen Compounds, 70, 582-585. [further information on levels in the environment]

GUAN, Y.F., SOJINU, O.S., LI, S.M., AND ZENG, E.Y., 2009. Fate of polybrominated diphenyl ethers in the environment of the Pearl River Estuary, South China. Environmental Pollution, 157 (7), 2166-2172. [further infomraiton on levels in the environment]

GUO, Y., YU, H.Y., ZHANG, B.Z. AND ZENG. E.Y., 2009. Persistent halogenated hydrocarbons in fish feeds manufactured in South China. Journal of Agriculture and Food Chemistry, 57 (9), 3674-3680. [further information on levels in the environment]

HAARSTAD, K. AND BORCH, H., 2008. Halogenated compounds, PCBs and pesticides in landfill leachate, downstream lake sediments and fish. Journal of Environmental Science and Health Part A, 43 (12), 1346-1352. [further information on levels in the environment]

HAMILTON, M.C. AND FISHER, T., 2006. Brominated diphenyl ethers in selected foods. 8th Annual Workshop on BFRs in the Environment, Ontario Ministry of the Environment, Toronto, ON, June 27-29, 2006. [further information on levels in food]

HARRAD, S., IBARRA, C., ABDALLAH, M.A.-E., BOON, R., NEELS, H. AND COVACI, A., 2008. Concentrations of brominated flame retardants in dust from United Kingdom cars, homes, and offices: Causes of variability and implications for human exposure. Environment International, **34**, 1170-1175. [further information on levels in dust]

HELGASON, L.B., POLDER, A., FØREID, S., BAEK, K., LIE, E., GABRIELSEN, G.W., BARRETT, R.T. AND SKAARE, J.U., 2008. Levels and temporal trends (1983-2003) of polybrominated diphenyl ethers and hexabromocyclododecanes in seabird eggs from North Norway. Environmental Toxicology and Chemistry, in press. [further information on levels in the environment; high variability in the procedural blanks was noted for decaBE]

HERMANUSSEN, S., PAEPKE, O., MATTHEWS, V., EBSEN, P., FOEDE, D., LIMPUS, C.J., AND GAUS, C., 2006. Flame retardants (PBDEs) in seafood and turtles from Australia. 8th Annual Workshop on BFRs in the Environment, Ontario Ministry of the Environment, Toronto, ON, June 27-29, 2006. [further information on levels in the environment]

HIRAI, Y., SATO, S. AND SAKAI, S., 2008. Impact of PBDD/DFs in lifecycle assessments on recycling of TV cabinet back covers. Organohalogen Compounds, 70, 1418-1421. [further information on emissions of decaBDE (and PBDD/DFs)]

HITES, R. AND VENIER, M., 2008. Flame retardants in the atmosphere of cities near the North American Great Lakes. Organohalogen Compounds, 70, 672-675. [further information on levels in the environment]

HOOPER, K., HOLDEN, A., CHOI, G., CHU, V., KIM, M., SHI, Y., CHIN, T., LINTHICUM, J. AND WALTON, B.J., 2008. PBDE and PCB congener patterns in California peregrine falcons: Evidence for BDE-209 debromination. BFR 2008 Workshop, Institute of Ocean Sciences, Sidney, BC, June 3-4, 2008. [probably related to the Holden et al. (2009) papers included in the evaluation]

HOOPER, K., HOLDEN, A., CHOI, G., CHU, V., KIM, M., SHI, Y., CHIN, T., LINTHICUM, J. AND WALTON, B.J., 2008. PBDE and PCB congener patterns in California peregrine falcons: Evidence for BDE-209 debromination? Paper presented at DIOXIN 2008. [probably related to the and Holden et al. (2009) papers included in the evaluation]

IBARRA, C., HARRAD, S, ABDALLAH, M., NEELS, H. AND COVACI, A., 2008. Brominated flame retardants in dust from the United Kingdom: within-room spatial and temporal variability. Organohalogen Compounds, 70, 1149-1152. [further information on levels in the indoor environment]

IKONOMOU, M.G. AND ADDISON, R.F., 2008. Polybrominated diphenyl ethers (PBDEs) in seal populations from eastern and western Canada: an assessment of the processes and factors controlling PBDE distribution in seals. Marine Envrionmental Research, 66 (2), 225-230. [further information on levels in the environment; it is not clear from the abstract whether decaBDE was included]

JIN, J., LIU, W. , WANG, Y. AND YAN TANG, X., 2008. Levels and distribution of polybrominated diphenyl ethers in plant, shellfish and sediment samples from Laizhou Bay in China. Chemosphere, 71, 1043-1050. [further information on levels in the environment]

JOHANSSON, A.K., SELLSTRÖM, U., LINDBERG, P., BIGNERT, A. AND DE WITT, C.A., 2009. Polybrominated diphenyl ether congener patterns, hexabromocyclododecane, and brominated biphenyl 153 in eggs of peregrine falcons (*Falco peregrinus*) breeding in Sweden. Environmental Toxicology and Chemistry, 28 (1), 9-17. [further information on levels in the environment]

JOHANNESSEN, S.C., MACDONALD, R.W., WRIGHT, C.A., BURD, B., SHAW, D.P. AND ROODSELAAR, A., 2008. Joined by geochemistry, divided by history: PCBs and PBDEs in Strait of Georgia sediments. Marine Environmental Research, 66, Supplement, 112-120. [further information on levels in the environment]

JÖRUNDSDÓTTIR, H., BIGNERT, A., SVAVARSSON, J. NYGÅRD, T., WEIHE, P. AND BERGMAN, A, 2009. Assessment of emerging and traditional halogenated contaminants in guillemot (*Uria aalge*) egg from north-western europe and the Baltic Sea. Science of the Total Environment, 407 (13), 4174-4183. [further information on levels in the environment]

KANNAN, K., MOON, H.B., YUN, S.H., AGUSA, T., THOMAS, N.J., AND TANABE, S. Chlorinated, brominated, and perfluorinated compounds, polycyclic aromatic hydrocarbons and trace elements in livers of sea otters from California, Washington, and Alaska (USA), and Kamchatka (Russia). Journal of Environmental Monitoring, 10 (4), 552-558. [further information on levels in the environment]

KEMI, 2009. DekaBDE – rapport från ett regeringsuppdrag. Rapport Ne 1/09, Kemikalieinspectionen. [review in Swedish]

KENNEDY, K., HEARN, L., TOMS, L-M.L., THOMPSON, J., BARTKOW, M. AND MUELLER, J., 2008. Ambient atmospheric levels of PBDEs across urban and regional centres in Australia determined using polyurethane foam (PUF) passive air sampling. Organohalogen Compounds, 70, 1621-1624. [further information on levels in the environment]

KIERKEGAARD, A., 2007. PBDEs in the environment. Time trends, bioaccumulation and the identification of their successor, decabromodiphenyl ethane. Doctoral dissertation, Department of Applied and Environmental Sciences, Stockholm University, Sweden, 68 pp. [further information on levels in the environment; as quoted in Environment Canada, 2009]

KIMBROUGH, K.L., JOHNSON, W.E., LAUENSTEIN, G.G., CHRISTENSEN, J.D. AND APETI, D.A., 2009. An assessment of polybrominated diphenyl ethers (PBDEs) in sediments and bivalves of the U.S. coastal zone. NOAA Technical Memorandum, NOS NCCOS 94, Silver Spring, MD, 87 pp. [further information on levels in the environment]

KIVIRANTA, H., RUOKOJÄRVI, P., LAAKSONEN, S., SUUTARI, A., RAJAKANGAS, L. AND HALLIKAINEN, A., 2008. PCDD/Fs, PCBs, AND PBDEs in Finnish game and semi-domesticated animals in 2006-7. Organohalogen Compounds, 70, 1422-1425.

KLOSTERHAUS, S., FISHER, D., ZIEGLER, G. AND BAKER, J., 2006. Bioavailability of brominated biphenyl ethers to deposit-feeding invertebrates exposed to 'deca-BDE'-dominated estuarine sediments. 8th Annual Workshop on BFRs in the Environment, Ontario Ministry of the Environment, Toronto, ON, June 27-29, 2006. [further information on levels in the environment]

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ST-AMAND, A.D., MAYER, P.M. AND BLAIS, J.M., 2007. Modeling atmospheric vegetation uptake of PBDEs using field measurements. Environmental Science and Technology, 41 (12), 4234-4239. [study did include decaBDE but little or no information was reported specifically for this congener]

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List of abbreviations

Abbreviations

ABS	Acrylonitrile-butadiene-styrene
[ABST]	Information available in abstract form only
BAF	Bioaccumulation factor
BCF	Bioconcentration factor
BDE	Brominated diphenyl ether
BMDL	Benchmark dose level
BMF	Biomagnification factor
CAS number	Chemical Abstracts registry number
CELAD assay	Chick embryo liver cell assay for dioxins
DecaBDE	Decabromodiphenyl ether
DiBDE	Dibromodiphenyl ether
Dmax aver	Average maximum diameter
DR-CALUX assay	An assay using a transfected rat hepatoma cell line
ESR	Existing Substances Regulation
GC	Gas chromatography
GC-MS	Gas chromatography – mass spectrometry
HeptaBDE	Heptabromodiphenyl ether
HexaBDE	Hexabromodiphenyl ether
HIPS	High impact polystyrene
HxBDD	Hexabromodibenzo-*p*-dioxin
log Koa	Logarithm (base 10) of the n-octanol-air partition coefficient
log Kow	Logarithm (base 10) of the n-octanol-water partition coefficient
MML	Maximum molecular length
NM^3	Cubic metre at normal temperature and pressure
NOAEL	No adverse effect level
NonaBDE	Nonabromodiphenyl ether
OctaBDE	Octabromodiphenyl ether
PBDE	Polybrominated diphenyl ether
PeBDD	Pentabromodibenzo-*p*-dioxin
PeBDF	Pentabromodibenzofuran
PEC	Predicted environmental concentration
PentaBDE	Pentabromodiphenyl ether
PNEC	Predicted no effect concentration
PVC	Polyvinylchloride
REACH	Regulation (EC) No 1907/2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals

RoHS Directive	Directive 2002/95/EC on the restriction of the use of certain hazardous substances in electrical and electronic equipment
TeBDD	Tetrabromodibenzo-*p*-dioxin
TeBDF	Tetrabromodibenzofuran
TetraBDE	Tetrabromodiphenyl ether
TEQ	Toxicity equivalent
TriBDE	Tribromodiphenyl ether
VECAP	Voluntary Emissions Control Action Programme
WEEE	Waste electrical and electronic equipment
WEEE Directive	Directive 2002/96/EC on waste electrical and electronic equipment

Congener numbers and names

Congener	Name
BDE-1	2-Bromodiphenyl ether
BDE-2	3-Bromodiphenyl ether
BDE-3	4-Bromodiphenyl ether
BDE-4	2,2'-Dibromodiphenyl ether
BDE-5	2,3-Dibromodiphenyl ether
BDE-6	2,3'-Dibromodiphenyl ether
BDE-7	2,4-Dibromodiphenyl ether
BDE-8	2,4'-Dibromodiphenyl ether
BDE-9	2,5-Dibromodiphenyl ether
BDE-10	2,6-Dibromodiphenyl ether
BDE-11	3,3'-Dibromodiphenyl ether
BDE-12	3,4-Dibromodiphenyl ether
BDE-13	3,4'-Dibromodiphenyl ether
BDE-14	3,5-Dibromodiphenyl ether
BDE-15	4,4'-Dibromodiphenyl ether
BDE-16	2,2',3-Tribromodiphenyl ether
BDE-17	2,2',4-Tribromodiphenyl ether
BDE-18	2,2',5-Tribromodiphenyl ether
BDE-19	2,2',6-Tribromodiphenyl ether
BDE-20	2,3,3'-Tribromodiphenyl ether
BDE-21	2,3,4-Tribromodiphenyl ether
BDE-22	2,3,4'-Tribromodiphenyl ether
BDE-23	2,3,5-Tribromodiphenyl ether
BDE-24	2,3,6-Tribromodiphenyl ether
BDE-25	2,3',4-Tribromodiphenyl ether
BDE-26	2,3',5-Tribromodiphenyl ether
BDE-27	2,3',6-Tribromodiphenyl ether
BDE-28	2,4,4'-Tribromodiphenyl ether
BDE-29	2,4,5-Tribromodiphenyl ether
BDE-30	2,4,6-Tribromodiphenyl ether
BDE-31	2,4',5-Tribromodiphenyl ether
BDE-32	2,4',6-Tribromodiphenyl ether
BDE-33	2,3',4'-Tribromodiphenyl ether
BDE-34	2,3',5'-Tribromodiphenyl ether
BDE-35	3,3',4-Tribromodiphenyl ether
BDE-36	3,3',5-Tribromodiphenyl ether
BDE-37	3,4,4'-Tribromodiphenyl ether
BDE-38	3,4,5-Tricholodiphenyl ether

Congener	**Name**
BDE-39	3,4',5-Tribromodiphenyl ether
BDE-40	2,2',3,3'-Tetrabromodiphenyl ether
BDE-41	2,2',3,4-Tetrabromodiphenyl ether
BDE-42	2,2',3,4'-Tetrabromodiphenyl ether
BDE-43	2,2',3,5-Tetrabromodiphenyl ether
BDE-44	2,2',3,5'-Tetrabromodiphenyl ether
BDE-45	2,2',3,6-Tetrabromodiphenyl ether
BDE-46	2,2',3,6'-Tetrabromodiphenyl ether
BDE-47	2,2',4,4'-Tetrabromodiphenyl ether
BDE-48	2,2',4,5-Tetrabromodiphenyl ether
BDE-49	2,2',4,5'-Tetrabromodiphenyl ether
BDE-50	2,2',4,6-Tetrabromodiphenyl ether
BDE-51	2,2',4,6'-Tetrabromodiphenyl ether
BDE-52	2,2',5,5'-Tetrabromodiphenyl ether
BDE-53	2,2',5,6'-Tetrabromodiphenyl ether
BDE-54	2,2',6,6'-Tetrabromodiphenyl ether
BDE-55	2,3,3',4-Tetrabromodiphenyl ether
BDE-56	2,3,3',4'-Tetrabromodiphenyl ether
BDE-57	2,3,3',5-Tetrabromodiphenyl ether
BDE-58	2,3,3',5'-Tetrabromodiphenyl ether
BDE-59	2,3,3',6-Tetrabromodiphenyl ether
BDE-60	2,3,4,4'-Tetrabromodiphenyl ether
BDE-61	2,3,4,5-Tetrabromodiphenyl ether
BDE-62	2,3,4,6-Tetrabromodiphenyl ether
BDE-63	2,3,4',5-Tetrabromodiphenyl ether
BDE-64	2,3,4',6-Tetrabromodiphenyl ether
BDE-65	2,3,5,6-Tetrabromodiphenyl ether
BDE-66	2,3',4,4'-Tetrabromodiphenyl ether
BDE-67	2,3',4,5-Tetrabromodiphenyl ether
BDE-68	2,3',4,5'-Tetrabromodiphenyl ether
BDE-69	2,3',4,6-Tetrabromodiphenyl ether
BDE-70	2,3',4',5-Tetrabromodiphenyl ether
BDE-71	2,3',4',6-Tetrabromodiphenyl ether
BDE-72	2,3',5,5'-Tetrabromodiphenyl ether
BDE-73	2,3',5',6-Tetrabromodiphenyl ether
BDE-74	2,4,4',5-Tetrabromodiphenyl ether
BDE-75	2,4,4',6-Tetrabromodiphenyl ether
BDE-76	2,3',4',5'-Tetrabromodiphenyl ether
BDE-77	3,3',4,4'-Tetrabromodiphenyl ether

Congener	Name
BDE-78	3,3',4,5-Tetrabromodiphenyl ether
BDE-79	3,3',4,5'-Tetrabromodiphenyl ether
BDE-80	3,3',5,5'-Tetrabromodiphenyl ether
BDE-81	3,4,4',5-Tetrabromodiphenyl ether
BDE-82	2,2',3,3',4-Pentabromodiphenyl ether
BDE-83	2,2',3,3',5-Pentabromodiphenyl ether
BDE-84	2,2',3,3',6-Pentabromodiphenyl ether
BDE-85	2,2',3,4,4'-Pentabromodiphenyl ether
BDE-86	2,2',3,4,5-Pentabromodiphenyl ether
BDE-87	2,2',3,4,5'-Pentabromodiphenyl ether
BDE-88	2,2',3,4,6-Pentabromodiphenyl ether
BDE-89	2,2',3,4,6'-Pentabromodiphenyl ether
BDE-90	2,2',3,4',5-Pentabromodiphenyl ether
BDE-91	2,2',3,4',6-Pentabromodiphenyl ether
BDE-92	2,2',3,5,5'-Pentabromodiphenyl ether
BDE-93	2,2',3,5,6-Pentabromodiphenyl ether
BDE-94	2,2',3,5,6'-Pentabromodiphenyl ether
BDE-95	2,2',3,5',6-Pentabromodiphenyl ether
BDE-96	2,2',3,6,6'-Pentabromodiphenyl ether
BDE-97	2,2',3,4',5'-Pentabromodiphenyl ether
BDE-98	2,2',3,4',6'-Pentabromodiphenyl ether
BDE-99	2,2',4,4',5-Pentabromodiphenyl ether
BDE-100	2,2',4,4',6-Pentabromodiphenyl ether
BDE-101	2,2',4,5,5'-Pentabromodiphenyl ether
BDE-102	2,2',4,5,6'-Pentabromodiphenyl ether
BDE-103	2,2',4,5',6-Pentabromodiphenyl ether
BDE-104	2,2',4,6,6'-Pentabromodiphenyl ether
BDE-105	2,3,3',4,4'-Pentabromodiphenyl ether
BDE-106	2,3,3',4,5-Pentabromodiphenyl ether
BDE-107	2,3,3',4',5-Pentabromodiphenyl ether
BDE-108	2,3,3',4,5'-Pentabromodiphenyl ether
BDE-109	2,3,3',4,6-Pentabromodiphenyl ether
BDE-110	2,3,3',4',6-Pentabromodiphenyl ether
BDE-111	2,3,3',5,5'-Pentabromodiphenyl ether
BDE-112	2,3,3',5,6-Pentabromodiphenyl ether
BDE-113	2,3,3',5',6-Pentabromodiphenyl ether
BDE-114	2,3,4,4',5-Pentabromodiphenyl ether
BDE-115	2,3,4,4',6-Pentabromodiphenyl ether
BDE-116	2,3,4,5,6-Pentabromodiphenyl ether

Congener	**Name**
BDE-117	2,3,4',5,6-Pentabromodiphenyl ether
BDE-118	2,3',4,4',5-Pentabromodiphenyl ether
BDE-119	2,3',4,4',6-Pentabromodiphenyl ether
BDE-120	2,3',4,5,5'-Pentabromodiphenyl ether
BDE-121	2,3',4,5',6-Pentabromodiphenyl ether
BDE-122	2,3,3',4',5'-Pentabromodiphenyl ether
BDE-123	2,3',4,4',5'-Pentabromodiphenyl ether
BDE-124	2,3',4',5,5'-Pentabromodiphenyl ether
BDE-125	2,3',4',5',6-Pentabromodiphenyl ether
BDE-126	3,3',4,4',5-Pentabromodiphenyl ether
BDE-127	3,3',4,5,5'-Pentabromodiphenyl ether
BDE-128	2,2',3,3',4,4'-Hexabromodiphenyl ether
BDE-129	2,2',3,3',4,5-Hexabromodiphenyl ether
BDE-130	2,2',3,3',4,5'-Hexabromodiphenyl ether
BDE-131	2,2',3,3',4,6-Hexabromodiphenyl ether
BDE-132	2,2',3,3',4,6'-Hexabromodiphenyl ether
BDE-133	2,2',3,3',5,5'-Hexabromodiphenyl ether
BDE-134	2,2',3,3',5,6-Hexabromodiphenyl ether
BDE-135	2,2',3,3',5,6'-Hexabromodiphenyl ether
BDE-136	2,2',3,3',6,6'-Hexabromodiphenyl ether
BDE-137	2,2',3,4,4',5-Hexabromodiphenyl ether
BDE-138	2,2',3,4,4',5'-Hexabromodiphenyl ether
BDE-139	2,2',3,4,4',6-Hexabromodiphenyl ether
BDE-140	2,2',3,4,4',6'-Hexabromodiphenyl ether
BDE-141	2,2',3,4,5,5'-Hexabromodiphenyl ether
BDE-142	2,2',3,4,5,6-Hexabromodiphenyl ether
BDE-143	2,2',3,4,5,6'-Hexabromodiphenyl ether
BDE-144	2,2',3,4,5',6-Hexabromodiphenyl ether
BDE-145	2,2',3,4,6,6'-Hexabromodiphenyl ether
BDE-146	2,2',3,4',5,5'-Hexabromodiphenyl ether
BDE-147	2,2',3,4',5,6-Hexabromodiphenyl ether
BDE-148	2,2',3,4',5,6'-Hexabromodiphenyl ether
BDE-149	2,2',3,4',5',6-Hexabromodiphenyl ether
BDE-150	2,2',3,4',6,6'-Hexabromodiphenyl ether
BDE-151	2,2',3,5,5',6-Hexabromodiphenyl ether
BDE-152	2,2',3,5,6,6'-Hexabromodiphenyl ether
BDE-153	2,2',4,4',5,5'-Hexabromodiphenyl ether
BDE-154	2,2',4,4',5,6'-Hexabromodiphenyl ether
BDE-155	2,2',4,4',6,6'-Hexabromodiphenyl ether

Congener	Name
BDE-156	2,3,3',4,4',5-Hexabromodiphenyl ether
BDE-157	2,3,3',4,4',5'-Hexabromodiphenyl ether
BDE-158	2,3,3',4,4',6-Hexabromodiphenyl ether
BDE-159	2,3,3',4,5,5'-Hexabromodiphenyl ether
BDE-160	2,3,3',4,5,6-Hexabromodiphenyl ether
BDE-161	2,3,3',4,5',6-Hexabromodiphenyl ether
BDE-162	2,3,3',4',5,5'-Hexabromodiphenyl ether
BDE-163	2,3,3',4',5,6-Hexabromodiphenyl ether
BDE-164	2,3,3',4',5',6-Hexabromodiphenyl ether
BDE-165	2,3,3',5,5',6-Hexabromodiphenyl ether
BDE-166	2,3,4,4',5,6-Hexabromodiphenyl ether
BDE-167	2,3',4,4',5,5'-Hexabromodiphenyl ether
BDE-168	2,3',4,4',5',6-Hexabromodiphenyl ether
BDE-169	3,3',4,4',5,5'-Hexabromodiphenyl ether
BDE-170	2,2',3,3',4,4',5-Heptabromodiphenyl ether
BDE-171	2,2',3,3',4,4',6-Heptabromodiphenyl ether
BDE-172	2,2',3,3',4,5,5'-Heptabromodiphenyl ether
BDE-173	2,2',3,3',4,5,6-Heptabromodiphenyl ether
BDE-174	2,2',3,3',4,5,6'-Heptabromodiphenyl ether
BDE-175	2,2',3,3',4,5',6-Heptabromodiphenyl ether
BDE-176	2,2',3,3',4,6,6'-Heptabromodiphenyl ether
BDE-177	2,2',3,3',4,5',6'-Heptabromodiphenyl ether
BDE-178	2,2',3,3',5,5',6-Heptabromodiphenyl ether
BDE-179	2,2',3,3',5,6,6'-Heptabromodiphenyl ether
BDE-180	2,2',3,4,4',5,5'-Heptabromodiphenyl ether
BDE-181	2,2',3,4,4',5,6-Heptabromodiphenyl ether
BDE-182	2,2',3,4,4',5,6'-Heptabromodiphenyl ether
BDE-183	2,2',3,4,4',5',6-Heptabromodiphenyl ether
BDE-184	2,2',3,4,4',6,6'-Heptabromodiphenyl ether
BDE-185	2,2',3,4,5,5',6-Heptabromodiphenyl ether
BDE-186	2,2',3,4,5,6,6'-Heptabromodiphenyl ether
BDE-187	2,2',3,4',5,5',6-Heptabromodiphenyl ether
BDE-188	2,2',3,4',5,6,6'-Heptabromodiphenyl ether
BDE-189	2,3,3',4,4',5,5'-Heptabromodiphenyl ether
BDE-190	2,3,3',4,4',5,6-Heptabromodiphenyl ether
BDE-191	2,3,3',4,4',5',6-Heptabromodiphenyl ether
BDE-192	2,3,3',4,5,5',6-Heptabromodiphenyl ether
BDE-193	2,3,3',4',5,5',6-Heptabromodiphenyl ether
BDE-194	2,2',3,3',4,4',5,5'-Octabromodiphenyl ether

Congener	Name
BDE-195	2,2',3,3',4,4',5,6-Octabromodiphenyl ether
BDE-196	2,2',3,3',4,4',5,6'-Octabromodiphenyl ether
BDE-197	2,2',3,3',4,4',6,6'-Octabromodiphenyl ether
BDE-198	2,2',3,3',4,5,5',6-Octabromodiphenyl ether
BDE-199	2,2',3,3',4,5,5',6'-Octabromodiphenyl ether
BDE-200	2,2',3,3',4,5,6,6'-Octabromodiphenyl ether
BDE-201	2,2',3,3',4,5',6,6'-Octabromodiphenyl ether
BDE-202	2,2',3,3',5,5',6,6'-Octabromodiphenyl ether
BDE-203	2,2',3,4,4',5,5',6-Octabromodiphenyl ether
BDE-204	2,2',3,4,4',5,6,6'-Octabromodiphenyl ether
BDE-205	2,3,3',4,4',5,5',6-Octabromodiphenyl ether
BDE-206	2,2',3,3',4,4',5,5',6-Nonabromodiphenyl ether
BDE-207	2,2',3,3',4,4',5,6,6'-Nonabromodiphenyl ether
BDE-208	2,2',3,3',4,5,5',6,6'-Nonabromodiphenyl ether
BDE-209	Decabromodiphenyl ether

Appendix 1 The PBT status of nona- to heptaBDE congeners

Persistence: Since pentaBDEs and decaBDE are considered to be very persistent, hepta- to nonaBDE congeners can also be assumed to be very persistent. There is also the potential for debromination.

Bioaccumulation: Whilst heptaBDE congeners have been considered to have Persistent Organic Pollutant (POP) characteristics under ongoing global discussions (and have recently been added to Annex A of the Stockholm Convention[30]), there is no reliable information about the bioaccumulative behaviour of the nona-, octa- or heptaBDEs. This is considered below.

From the available evidence it is likely that the BCF for nonaBDEs is <<2,000 l/kg (EC, 2003) and so these congeners are unlikely to meet the B-criterion themselves based on a BCF. However, based on the study of Wang et al. (2007), the BAF for nonaBDE would be expected to be around 1,200 l/kg, although again it should be noted that this is a prediction with an inherent uncertainty. For hexaBDEs, an experimental fish BCF of around 5,640 l/kg has been determined (EC, 2000) and so these congeners clearly meet the criteria for vB.

No BCF data appear to be available for heptaBDEs. In order to estimate the BCF an analysis has been carried out using the available BCF data for tetra-, penta- and hexaBDEs, along with the log K_{ow} values for these substances. The data used in this analysis are summarised below: the BCF values were taken from EC (2002), whilst the log K_{ow} values were taken from a number of sources (given the difficulty in their measurement) including the following:

- Estimates obtained using the USEPA EPIWIN v3.12 program.
- Mean log K_{ow} values estimated using the ALOGPS v2.18 program.
- Values estimated by Ellinger et al. (2003) based on total surface area (TSA) correlations with the known log K_{ow} values for tetra- to hexaBDEs.
- Values determined by Ellinger et al. (2003) by a GC-MS method using the selected polychlorinated biphenyls with known log K_{ow} values as reference substances.

A plot of measured log BCF against log K_{ow} is shown in Figure A2.1 for each of the data sets.

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[30] See http://chm.pops.int/Convention/Media/Pressreleases/COP4Geneva9May2009/tabid/542/language/en-US/Default.aspx

Table A2.1 Data used to estimate the BCF for heptaBDE.

Congener	Predicted log K_{ow}				Measured fish BCF (l/kg)	Log BCF
	EPIWIN	**ALOGPS**	**Ellinger et al. (2003) TSA**	**Ellinger et al. (2003) GC-MS**		
TetraBDE	6.77	6.32	6.8	7.4	66,700	4.82
PentaBDE	7.66	7.03	7.3	7.7	17,700	4.24
HexaBDE	8.55	7.60	7.8	8.0	5,640	3.75
HeptaBDE	9.44	8.31	9.3	8.4	-	-

These data sets are used in this analysis as they allow log K_{ow} values to be predicted (or measured) for each substance of interest on the same basis. It should be noted, however, that the experimentally derived log K_{ow} is often lower than predicted. For example, measured values of the log K_{ow} of the *commercial* products have been determined as 6.57 for pentabromodiphenyl ether (EC, 2000), 6.29 for octabromodiphenyl ether (EC, 2003). In addition EC (2003) gives a log K_{ow} value of 7.14 for a heptaBDE (BDE-183) obtained using a slow-stirring method. (For comparison, values of between 6.27 and 9.97 have been measured for decabromodiphenyl ether (EC, 2002).) Although the agreement between these measured values and the predictions used in the analysis is poor, this does not necessarily impact adversely on the BCF values obtained as the log K_{ow} is effectively used only as a scaler in Figure A2.1 – it is the relative change in log K_{ow} with increasing bromination that is most important in this approach rather than the absolute log K_{ow} value.

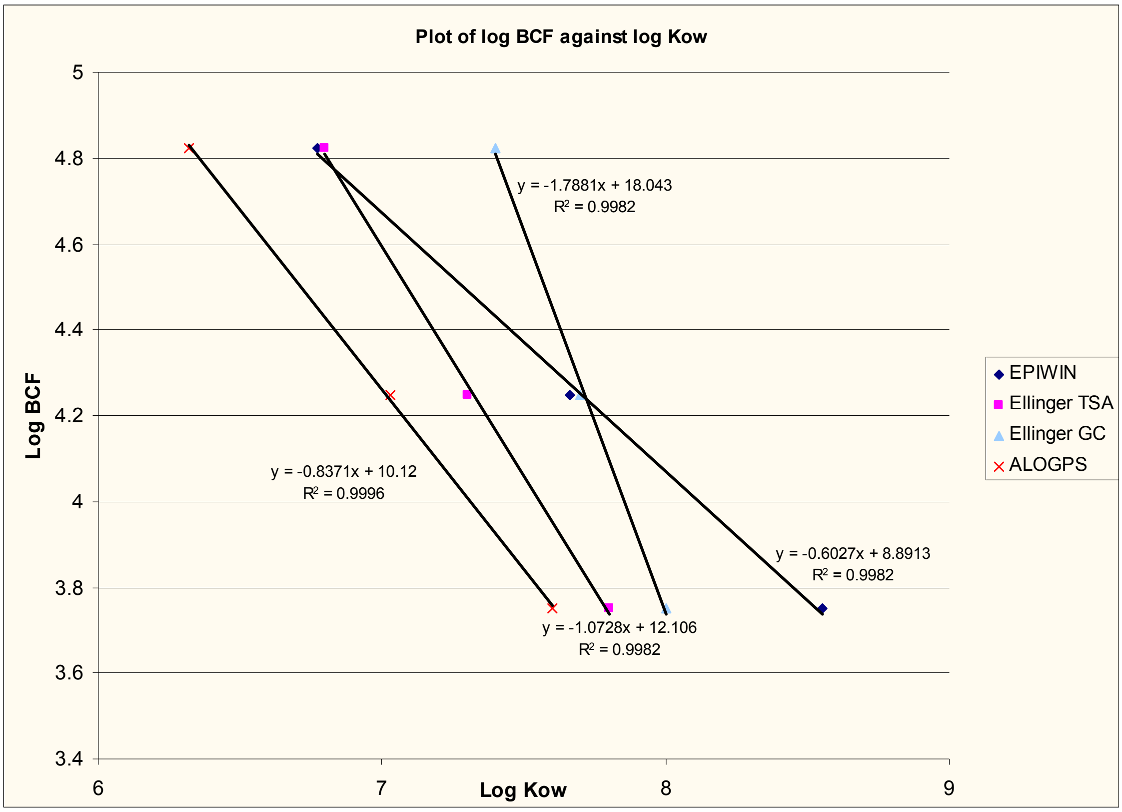

Figure A2.1 Plot of measured log BCF against predicted log Kow.

The regression line fitted to these data points gives the following relationships:

$\log BCF = -0.603 \times \log K_{ow} + 8.89$ for the EPIWIN values

$\log BCF = -1.79 \times \log K_{ow} + 18.04$ for the Ellinger et al. (2003) GC values

$\log BCF = -1.07 \times \log K_{ow} + 12.11$ for the Ellinger et al. (2003) TSA values

$\log BCF = -0.837 \times \log K_{ow} + 10.12$ for the ALOGPS values

Using these equations, a BCF of around 1,580 l/kg (EPIWIN values), 1,010 l/kg (Ellinger et al. (2003) GC values), 144 l/kg (Ellinger et al. (2003) TSA values) and 1,460 l/kg (ALOGPS values) can be estimated for heptaBDE. Thus it would appear that heptaBDE itself does not meet the B criterion in relation to the (predicted) BCF value.

However, it should be noted that, as is the case with decaBDE, the actual accumulation potential for nona-, octa- and heptaBDEs in aquatic organisms may be better expressed in terms of a BAF rather than a BCF. As is the case with the BCF, there are no conclusive data available for the actual BAFs for decaBDE or the lower brominated congeners. As discussed in Section 7.2, there are data available that suggest that the BAF for decaBDE could be of the order of 1,000 l/kg or above. Similarly, based on the results of the study by Wang et al. (2007) given in Section 4.3.3, BAFs of around 1,200, 2,900 and 6,900 l/kg can be estimated for nonaBDE, octaBDE and heptaBDE respectively (although it is not clear if these are lipid normalised or wet weight values). This does suggest that the estimated BCF values for heptaBDEs in the range 144-1,580 l/kg could be misleading with respect to bioaccumulation potential (in that it could be underestimated).

Toxicity: With regard to octa- and heptaBDE toxicity, it is relevant to consider that commercial octabromodiphenyl ether products typically consisted of ≤45% by weight heptaBDEs and ≤33% by weight octaBDEs. Annex I of Directive 67/548/EEC indicates that the commercial octabromodiphenyl ether product is classified as toxic to reproduction Category 2 (R61 May cause harm to the unborn child) and toxic to reproduction Category 3 (R62 Possible risk of impaired fertility). Consequently, the commercial octaBDE product would be considered to be a substance of very high concern (and so be an Annex XIV candidate) under REACH had it not been banned already.

According to EC (2003) the composition of the commercial product used in the toxicity test leading to the toxic to reproduction Category 2 (R 61) classification was 0.2 per cent pentaBDE, 8.6 per cent hexaBDE, 45 per cent heptaBDE, 33.5 per cent octaBDE, 11.2 per cent nonaBDE and 1.4 per cent decaBDE (the composition of the commercial product tested that led to the R62 classification is not reported in EC (2003)). It is not known which components of the commercial product contribute to the toxicity that led to this classification but the two main components to which the animals would have been exposed would be the heptaBDEs and octaBDEs. There is also new evidence that neurotoxicity may be expressed by many lower brominated congeners (for example see Costa and Giordano (2007)).

Thus, based on the existing hazard classification for the commercial octaBDE it cannot be ruled out that the heptaBDEs and octaBDE components would meet the toxicity criterion within Annex XIII of REACH.

Summary: OctaBDE and heptaBDE congeners are highly persistent in the environment, and whilst their actual bioaccumulation potential is unclear, it could be significant (e.g. BAFs tentatively in the region of 2,000 – 7,000 l/kg). These congeners are considered to be toxic, since the commercial octaBDE product is a Category 2 reprotoxin.

In summary, heptaBDEs in particular could be considered to be of equivalent concern to a PBT substance. This would be consistent with the POP discussions.

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