

BROMINATED FLAME RETARDANTS IN TROUT AND EELS FROM THE SKERNE-TEES RIVER SYSTEM AND TOTAL DIET STUDY SAMPLES

Key Facts

- This work was carried out following reports of elevated concentrations of polybrominated flame retardants (BFRs) in fish and sediment in the River Skerne.
- Polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs) were analysed in samples of trout and eels caught at various locations in the Skerne-Tees River system.
- Most of the individual PBDE and HBCD congeners were detected in the majority of samples of trout and eels analysed.
- The concentrations of these chemicals were elevated in fish caught downstream of a site where these flame retardants were being manufactured, concentrations decreasing with increasing distance from the site.
- The independent expert Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment (COT) concluded that the estimated dietary intakes of PBDEs and HBCDs from the consumption of a weekly single portion of eels or trout from the Skerne-Tees were unlikely to represent a risk to health. However, this opinion was reached against a background of uncertainties surrounding the toxicological database and exposure assessments.
- To provide supporting information for this study, the chemicals referred to above, together with tetrabromobisphenol A (TBBP-A) and polybrominated biphenyls (PBBs), were tested for in food group samples from the 2001 Total Diet Study (TDS). No BFRs were detected in any of the TDS food groups.
- Dietary intakes were estimated from the limits of determination for the BFRs in the different food groups. The estimated *upper bound* adult dietary intakes, from the whole diet, of total PBDEs and total HBCDs respectively would be less than 41 and 11 ng/kg bodyweight/day for average consumers, and 74 and 18 ng/kg bodyweight/day for

high level consumers. The COT recommend continued monitoring of deca-BDE and HBCD, particularly in fatty foods.

Background

Purpose and Nature of Brominated flame retardants

Brominated Flame Retardants (BFRs) are used to reduce the risk of fire in a wide variety of applications including plastics, electronic equipment and textiles. The term BFRs, as used in this report, refers to three groups of chemicals - polybrominated diphenyl ethers (PBDEs), hexabromocyclododecanes (HBCDs) and polybrominated biphenyls (PBBs), and one individual chemical, tetrabromobisphenol A (TBBP-A). There are 209 closely related PBDEs, referred to as congeners. Three formulations are available commercially - penta-, octa- and decabromodiphenyl or bis(pentabromophenyl) ethers (PeBDEs, OBDEs and DeBDE, respectively). Of these formulations, DeBDE (consisting predominantly of only a single PBDE congener) is used in the greatest quantities, whilst PeBDE is used the least. Production of PeBDEs and OBDEs will cease in the EU 2004, although use will continue at least until 2006, while alternative products are developed.

HBCDs consist of three closely related chemicals, known as diastereoisomers (alpha-, beta- and gamma-HBCD). These and TBBP-A are usually added to polymers as flame retardants. PBBs, of which there are also 209 congeners, are also used as flame retardants, mainly in synthetic fibres and plastics for electrical equipment, although their production ceased in 2000.

BFRs can be released to the environment during their production and use in manufacturing other products, and during disposal of products containing these chemicals. PBDEs do not degrade easily and, consequently, they are becoming widespread in the environment. Additionally, being lipophilic, they are able to bioaccumulate in the food chain. Less is known about the environmental distribution of HBCD and TBBP-A.

There does not appear to be a clear trend in concentrations of PBDEs in the environment against time. However, most studies that have been carried out seem to indicate that concentrations of PBDEs in fish and in the environment have been rising until recently, but

may now be stabilising.^{1,2} This would coincide with the fact that production peaked in 2000 and is now beginning to drop off.

There is less information about HBCDs, PBBs and TBBP-A. However, as the usage of HBCDs and TBBP-A began more recently than that of PBDEs and PBBs, it is possible that concentrations of HBCDs and TBBP-A are still rising.

Controls on the usage of BFRs

An EC ban on the marketing and use of penta- and octa-BDE has recently been agreed^{3,4} and is due to come into effect from 1 July 2004. However, since these substances are persistent, they are still likely to be present in the environment for some time after the start of the ban. There are no plans to regulate the use of DeBDE. In-depth risk assessments for HBCD and TBBP-A are both in progress under the Existing Substances Regulation (EC, 1993).⁵ Under the Oslo and Paris Convention for the Protection of the Marine Environment,⁶ the Oslo and Paris Commission (OSPAR) has identified PBDEs and HBCDs for priority action, and certain PBBs as 'substances of potential concern'. The OSPAR objective, which the UK has accepted, is to 'make every endeavour' to move towards the target of cessation of discharges, emissions and losses of hazardous substances by 2020. A monitoring strategy is being developed to monitor progress in achieving the target. PBDEs (but not HBCDs) are on the Water Framework Directive (WFD) Priority List that came into force on 16 December 2001, and PeBDE is further classified as a Priority Hazardous Substance (PHS) (EC, 2001).^{7,8} Under the WFD, member states must implement measures in accordance with Article 16 with the objective of progressively reducing discharges, emissions and losses of Priority Substances. In the case of Priority Hazardous Substances, the aim is to cease or phase out discharges, emissions and losses within 20 years of adoption of these proposals by the European Parliament and the Council. It is now anticipated that emission controls and environmental quality standards will be proposed during 2004.

Health effects of BFRs

The usage of PBDEs, HBCDs and TBBP-A as fire retardants contributes to the saving of many lives each year by prevention and reduction in the incidence and severity of fires.

Toxicology data on PBDEs and HBCDs are limited. The liver is a target organ. There is evidence for neurodevelopmental effects and OBDE only has been found to exhibit reproductive toxicity. The PBDEs have also shown weak dioxin-like and endocrine disruptor activity

The International Agency for Research on Cancer (IARC) has classified PBBs as possible human carcinogens and, under the US National Toxicology Program, some PBBs have been classified as reasonably anticipated to be human carcinogens.¹¹

Background to current work

The Centre for Environment, Fisheries and Aquaculture Science (CEFAS) has investigated PBDEs and HBCDs in fish and sediment from UK rivers and the North Sea.^{12,13} The study was carried out to assess the effects of BFRs on the fish. CEFAS found elevated concentrations of the BFRs in sediment from the River Skerne (a tributary of the River Tees), with levels much higher in sediment downstream than upstream of the Great Lakes Newton Aycliffe site where BFRs were being manufactured. Other investigators have found elevated concentrations of the chemicals in marine fish from the North Sea.¹⁴ Similar work has been carried out in other countries.¹⁵⁻¹⁹ Two Japanese surveys of fish and shellfish did not detect TBBP-A in any samples.^{19,20} One survey also targeted HBCD.²¹

The Food Standards Agency's surveys were carried out to assess the food safety implications of the presence of BFRs in fish and other foods.

Levels of BFRs in other foods

Very little work has been published on BFRs in foodstuffs other than fish. Low levels of PBDEs have been reported in vegetables, meat and human breast milk in Japan.²² BFRs have also been detected in human milk.²³⁻²⁵ A market basket study has been carried out in Sweden in which PBDE intake was investigated in samples of fish, meat, milk products, eggs, fats and oils and pastry food groups, which were the food groups assumed to contribute most to total intake.²⁶ Almost half the total intake originated from fish products whilst approximately 15 per cent was accounted for by meat, milk products and fats and oils. Low concentrations of PBDEs and PBBs have also been reported in cows' milk in Germany.²⁷

Methodology

Fish sampling

Brown trout and eels were caught at eight locations along the Skerne-Tees river system, including at Haughton Road (downstream of the Newton Aycliffe plant) and at the Tees barrage where the River Tees becomes tidal. In addition, control samples were obtained from the River Skerne upstream of the plant and from the River Tees well upstream of the confluence of the two rivers. As the project examined fish caught directly from the river system, brand names are not available.

The fish sampling locations were based on their position relative to the factory, ease of access and the sufficient availability of fish. A total of 42 samples (26 trout and 16 eel) were obtained for this survey from the River Skerne at Ricknall Grange (control site), Haughton Road and Oxenfield Bridge, and from the River Tees at Middleton in Teesdale (control site), Low Coniscliffe, Croft-on-Tees, Low Moor and the Tees Barrage. A map showing the sampling locations is included in Annex I. Only at Oxenfield Bridge and Croft-on-Tees were both trout and eels available.

Total Diet Samples

The Total Diet Study is a model of the average domestic diet in the UK.^{28,29} A total of 121 categories of food and drink are specified for inclusion in the Total Diet. These are assigned to one of twenty broad food groups. Foods are grouped so that commodities known to be susceptible to a build-up of environmental contaminants (e.g. offals and fish) are kept separate, as are foods that are consumed in large quantities, e.g. bread, potatoes, milk. Food samples are purchased fortnightly from 24 randomly selected locations representative of the UK as a whole. The food samples are prepared and cooked according to normal consumer practice. Further information on the Total Diet Study can be found in Food Survey Information Sheet No **38/03**.³⁰

Analysis

Samples were analysed by CEFAS, and full details of the analytical methodology can be found in the contractor's final reports for these surveys.^{31,32} Sub-samples of fish and TDS food groups were ground with anhydrous sodium sulphate, left for 24 hours and then extracted using a Soxhlet apparatus with a mixture of n-hexane and acetone for four hours. Aliquots of the crude extracts were purified and fractionated using alumina and

silica adsorption chromatography for PBDE analysis. For HBCD analysis, additional aliquots of crude extracts were mixed with concentrated sulphuric acid in order to remove lipid and other co-extractants.

The purified aliquots were analysed by CEFAS for PBDEs 28, 47, 99, 100, 153, 154 and, for the TDS food groups only, PBDE 183 and PBBs (the latter as a commercial formulation), using recently published methods developed by CEFAS in association with The Netherlands Institute for Fisheries Research (RIVO) and The Royal Netherlands Institute for Sea Research (NIOZ).³³ Aliquots were analysed by gas chromatography-negative ion chemical ionisation mass spectroscopy (GC-NICI-MS) using an Agilent 5973 mass selective detector. Quantities of PBDEs in the extracts were determined by comparing signal responses in the extracts to appropriate PBDE standards, which were supplied by Cambridge Isotope Laboratories. To improve the measurement accuracy of PBDE levels in an extract and to ensure correct PBDE identification, a known amount of the polychlorinated biphenyl compound CB198 was added as an internal standard prior to PBDE analysis.

Since gas chromatography was unable to separate the three HBCD diastereoisomers (alpha-, beta- and gamma-HBCD), these and, for the TDS food group samples only, TBBP-A, were analysed by liquid chromatography-mass spectroscopy (LC-MS) using an electrospray technique developed by CEFAS in collaboration with NIOZ and RIVO.³⁴ Further details of the analytical methodology for measuring HBCDs and TBBP-A are presented in the contractor's final reports.^{31,32}

Results

Trout and eels from the Skerne-Tees river system

This report presents the *upper bound* results for selected individual PBDEs and for HBCDs in the fish samples, expressed as parts per billion fresh weight (Tables 1a and 2a) and on a fat basis (Tables 1b and 2b). *Upper bound* concentrations assume that the compound being measured, when present at concentrations below the limit of detection (LOD), are present at the limit of detection. Consequently, they may be an overestimate of the true concentrations. The ranges of results, including all congeners analysed, for individual samples taken for this survey are presented in Tables 1a and 1b (PBDEs) and Tables 2a and 2b (HBCDs) respectively. All results refer to the edible portions of the fish. Full details can be found in the contractor's final report for the work.³¹

In the majority of samples, all of the individual congeners were present at concentrations above the limit of detection (LOD). The highest fresh weight concentrations both of total PBDEs and total HBCDs were found in trout from Haughton Road and eels from Oxenfield Bridge. PBDE 47 was always the most abundant PBDE, generally followed by PBDE 99 in trout and PBDE 100 in eels. Alpha-HBCD was generally the most abundant HBCD congener.

Dietary intakes of PBDEs and HBCDs from trout and eels were estimated from the concentrations found in the fish from Haughton Road, Oxenfield Bridge and Croft-on-Tees and, for comparison purposes, from Ricknall Grange. Intakes were estimated from consumption of one portion per week of either trout or eels. This was based on the Food Standards Agency's recommendation to all consumers that they should eat at least two portions of fish per week, one of which should be oily, as part of a balanced diet.³⁵ Typical portion sizes of 120 g trout or 70 g eel were assumed.³⁶ Estimated dietary intakes are presented in Table 4 for PBDEs, and Table 5 for HBCDs. For total PBDEs by adults, the maximum intakes would be 56 ng/kg bodyweight per day from trout and 48 ng/kg bodyweight per day from eel, based on the most contaminated fish tested. The corresponding intakes of total HBCDs were 1,931 and 1,572 ng/kg bodyweight per day from trout and eel, respectively. Estimated intakes from fish caught upstream or further downstream from the Newton Aycliffe Great Lakes site were lower.

Total Diet Study food group samples

Concentrations of BFRs in the food groups from the 2001 TDS are presented in Table 3. No BFRs were detected at concentrations exceeding the LOD in any of the food group samples. Further details can be found in the contractor's final report for the survey.³² As a result, planned analysis of these samples additionally for decaPBDE (PBDE 209) and for individual PBB congeners was abandoned.

Interpretation

For both trout and eels, the highest concentrations were found in the samples caught closest to the Newton Aycliffe plant (Haughton Road) and the concentrations fell with distance downstream from these locations. However, no fishing for eels takes place at the Haughton Road site, leaving Oxenfield Bridge as the site where the most contaminated eels are caught by anglers. There are no commercial fisheries in the River Skerne.

Concentrations in fish at locations in the River Skerne upstream of the site and in the River Tees upstream of its confluence with the River Skerne were low. These findings are consistent with release of BFRs into the River Skerne between Haughton Road and Ricknall Grange. Manufacture of BFRs in the UK by Great Lakes ceased on 23 December 2003, so with no further inputs from this source it is reasonable to expect that concentrations of BFRs in fish from the Skerne-Tees river system will fall.

The estimated dietary intakes of PBDEs and HBCDs from consuming one portion of trout or eel per week are shown in Tables 4 and 5. The independent expert Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment (COT) has concluded that, because only limited toxicological data are available, no Tolerable Daily Intakes (TDIs) can be set for any of the BFRs.³⁷ However, the Committee considered that a risk to human health was not expected if the Margin of Exposure between the doses found to cause effects in experimental animals and the human exposure exceeded 1000 for the PBDEs and 10,000 for the HBCDs. The Margin of Exposure for the estimated dietary intake was about 10,000 for total PBDEs and about 50,000 for the total HBCDs.

COT concluded that even the highest concentrations of PBDEs and HBCDs found in the trout and eels were unlikely to pose a health risk. However, this opinion was reached against a background of uncertainties surrounding the toxicological database and exposure assessments. COT also recommended that concentrations of deca-BDE and HBCD should continue to be monitored, especially in fatty foods.³⁷

Future work

Samples of a number of farmed and wild fish and shellfish species, currently being collected for dioxins and PCBs analysis, are also to be analysed for BFRs, including deca-BDE and TBBP-A.

Summary of Units

ppb	parts per billion, equivalent to one microgram per kilogram (kg)
kg	a kilogram (kg) is one thousand grams (g)
µg	microgram, one millionth of a gram
ng	a nanogram is one thousand millionth of a gram
ng/kg bw/day	nanograms per kilogram of bodyweight per day; equivalent to parts per million million (parts per trillion) by weight.

References

1. Kierkegaard, A. *et al.* (1997). Temporal trends of a polybrominated diphenyl ether (PBDE), a methoxylated PBDE, and hexabromocyclododecane (HBCD) in Swedish biota. *Organohalogen Compounds*. **40**, 367-370.
2. de Boer, J. and Allchin, C. (2001). An indication of temporal trends in environmental PBDE levels in Europe. *Organohalogen Compounds*.
3. The European Commission (EC). (2002). European Parliament and EU ministers agree to ban potentially toxic flame retardants. Enterprise Europe News Update, 18 December 2002, available at <http://europa.eu.int/comm/enterprise/library/enterprise-europe/news-updates/industry/20021218.htm>.
4. The European Commission (EC). (2001). Proposal for a Directive of the European Parliament and of the Council amending for the 24th time Council Directive 76/769/EEC relating to restrictions on the marketing and use of certain dangerous substances and preparations (pentabromodiphenyl ether), COM (2001) 12, available at http://europa.eu.int/eur-lex/en/com/pdf/2001/en_501PC0012.pdf.
5. European Commission. (2001). Proposal to amend Directive 76/769/EEC on restrictions on the marketing and use of dangerous substances and preparations. COM (2001) 12, available at <http://europa.eu.int/eur-lex/en/com/index1.html>.
6. Convention for the Protection of the Marine Environment of the North-East Atlantic. (2001). Certain brominated flame retardants – polybrominated diphenylethers, polybrominated biphenyls, hexabromocyclododecane, available at www.ospar.org/eng/html/welcome.html.
7. European Commission. (2001). Priority substances under the Water Framework Directive, available at http://europa.eu.int/comm/environment/water/water-dangersub/wd_env_191000_01_final.pdf
8. European Commission. (2000). Directive 2000/60/EC of the European Parliament and of the Council establishing a framework for the Community action in the field of water policy. *Official Journal of the European Communities*, **L 327**, 22 December 2000, available at http://europa.eu.int/comm/environment/water/water-framework/index_en.html.
9. Cordle, F. *et al.* (1978). Human exposures to polychlorinated biphenyls and polybrominated biphenyls. *Environmental Health Perspectives*. **24**,157-172.
10. Kay, K. (1977). Polybrominated biphenyls (PBB) environmental contamination in Michigan 1973-1976. *Environmental Research*. **13**,74-93.

11. International Agency for Research on Cancer. (1978). IARC monographs on the evaluation of carcinogenic risk of chemicals to humans. Vol. 18. Polychlorinated biphenyls and polybrominated biphenyls. World Health Organization, Lyon, France, 107-125.
12. Allchin, C.R., Law, R.J. and Morris, S. (1999). Polybrominated diphenylethers in sediments and biota downstream of potential sources in the UK. *Environmental Pollution* **105**, 197-207.
13. Allchin, C. and de Boer, J. (2001). Results of a comprehensive survey for PBDEs in the River Tees, UK. *Organohalogen Compounds*.
14. Zegers, B.N. *et al.* (2001). Levels of some polybrominated diphenyl ether (PBDE) flame-retardants in animals of different trophic levels of the North Sea food web. *Organohalogen Compounds*.
15. Christensen, J.H. *et al.* (2002). Polybrominated diphenyl ethers (PBDEs) in marine fish and blue mussels from southern Greenland. *Chemosphere*. **47**, 631-638.
16. Luross, J.M. *et al.* (2002). Spatial distribution of polybrominated diphenyl ethers and polybrominatedbiphenyls in lake trout from the Laurentian Great Lakes. *Chemosphere*. **46**, 665-672.
17. Ikonomou, M.G. *et al.* (2002). Occurrence and congener profiles of polybrominated diphenyl ethers (PBDEs) in environmental samples from Coastal British Columbia, Canada. *Chemosphere*. **46**, 649-663.
18. Rice, C.P. *et al.* (2002). Comparisons of PBDE composition and concentration in fish collected from the Detroit River, MI and Dews Plaines River, IL. *Chemosphere*. **49**, 731-737.
19. Tatsukawa, R. and Watanabe, I. (1990). [Environmental problems from brominated organic flame retardants.] *Kogai Taisaku*. **26**, 658-668.
20. Environment Agency Japan (1991) Chemicals in the environment. Report on environmental survey and wildlife monitoring of chemicals in F.Y. 1988 and 1989. Environment Agency Japan, Department of Environmental Health, Office of Health Studies. Tokyo.
21. Sellström, U. *et al.* (1998). Polybrominated diphenyl ethers and hexabromocyclododecane in sediment and fish from a Swedish river. *Environmental Toxicology and Chemistry*. **17**, 1065-1072.
22. Ohta, S. *et al.* (2002). Comparison of polybrominated diphenyl ethers in fish, vegetables, and meats and levels in human milk of nursing women in Japan. *Chemosphere*. **46**,689-696.

23. Norén, K. and Meironyté, D. (2000). Certain organochlorine and organobromine contaminants in Swedish human milk in perspective of past 20-30 years. *Chemosphere*. **40**, 1111-1123.
24. Meironyté, D., Norén, K. and Bergman, Å. (1999). Analysis of polybrominated diphenyl ethers in Swedish human milk. A time-related study, 1972-1997. *Journal of Toxicology and Environmental Health A*. **58**, 329-341.
25. Hooper, K. and McDonald, T.A. (2000). The PBDEs: an emerging environmental challenge and another reason for breast-milk monitoring programs. *Environmental Health Perspectives*, **108**,387-392.
26. Darnerud, P.O. *et al.* (2001). Polybrominated diphenyl ethers: occurrence, dietary exposure, and toxicology. *Environmental Health Perspectives*, **109**, 49-67.
27. Krüger, C. (1988). Polybrominated biphenyls and polybrominated diphenyl ethers – detection and quantitation in selected foods. PhD Thesis, University of Münster, Germany.
28. Ministry of Agriculture, Fisheries and Food. (1994). The British Diet: finding the facts. *Food Surveillance Paper*, **40**. HMSO.
29. Peattie, M.E. *et al.* (1983). Reorganisation of the British Total Diet Study for monitoring food constituents from 1981. *Food and Chemical Toxicology*. **21**, 503-507.
30. Food Standards Agency. (2003). Dioxins and dioxin-like PCBs in the UK diet: 2001 Total Diet Study samples *Food Surveillance Information Sheet*, **38/03**. Food Standards Agency, available at http://www.food.gov.uk/multimedia/pdfs/fsis38_2003.pdf .
31. Centre for Environment, Fisheries and Aquaculture Science. (2003). Survey of brominated flame retardant residues in eel and trout from R.Skerne and R.Tees Centre for Environment, Fisheries and Aquaculture Science.
32. Centre for Environment, Fisheries and Aquaculture Science. (2003). Congener specific analysis of brominated fire retardants in samples from the 2001 Total Diet study. Centre for Environment, Fisheries and Aquaculture Science.
33. de Boer, J., Allchin, C., Law, R., Zegers, B. and Boon, J.P. (2001). Method for the analysis of polybrominated diphenyl ethers in sediments and biota. *Trends in Analytical Chemistry*, **20**, 591-599.
34. Morris, S *et al.* In preparation, to be submitted to *Trends in Analytical Chemistry*.
35. Food Standards Agency. (2003). Eat more fish. Food Standards Agency, available at www.food.gov.uk/multimedia/webpage/resolutions/eatmorefish.

36. Ministry of Agriculture, Fisheries and Food/Department of Health. (1993). Food portion sizes. 2nd Edition. HMSO.
37. Food Standards Agency. (2003). Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment. Statement on brominated flame retardants in fish from the Skerne-Tees river system. Food Standards Agency.
<http://www.food.gov.uk/science/ouradvisors/toxicity/statements/cotstatements2003/>

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Table 1a: Concentrations of PBDEs in trout and eels from the Skerne-Tees River system

Food	PBDEs analysed	Concentrations ($\mu\text{g}/\text{kg}$ fresh weight)	Location
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.02-0.1 5.5-7.8 3.9-4.5 1.0-1.6 0.2-0.3 0.3-0.5 12-14	Ricknall Grange (Control for Skerne)
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.5-1.3 30-107 17-52 6.9-25 2.2-9.1 2.9-20 59 -197	Haughton Road
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.02-0.7 14-61 5.0-42 3.3-11 0.8-3.6 1.7-4.9 27-123	Oxenfield Bridge
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.02-0.1 0.6-9.1 0.5-6.0 1.0-1.6 0.02-0.3 0.04-0.5 1.3-18	Middleton in Teesdale (Control for Tees)

Table 1a (continued): Concentrations of PBDEs in trout and eels from the Skerne-Tees River system

Food	PBDEs analysed	Concentrations (µg/kg fresh weight)	Location
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.02-0.02 1.5-4.1 1.0-2.2 0.3-0.7 0.09-0.3 0.1-0.3 3.0-7.6	Low Coniscliffe
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.02-0.6 0.5-39 0.4-14 0.07-4.4 0.05-1.5 0.02-2.3 1.0-57	Croft-on-Tees
Eels (single sample)	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	1.3 34 2.0 12 1.8 1.9 53	Ricknall Grange (Control for Skerne)
Eels	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.02-2.5 84-185 10-14 59-117 6.8-13 9.8-27 164-288	Oxenfield Bridge

Table 1a (continued): Concentrations of PBDEs in trout and eels from the Skerne-Tees River system

Food	PBDEs analysed	Concentrations (µg/kg fresh weight)	Location
Eels	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	1.3-2.5 30-153 2.1-3.8 14-50 2.0-7.7 3.1-11 52 –221	Croft on Tees
Eels	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.1-2.5 17-189 0.1-22 14-49 1.5-13 3.7-9.3 37-266	Low Moor
Eels	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.8-2.1 25-145 0.8-2.1 18-58 2-6.4 3.9-22 52-234	Tees Barrage

Table 1b: Concentrations of PBDEs in trout and eels from the Skerne-Tees River system

Food	PBDEs analysed	Concentrations (µg/kg fat basis)	Location
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	2.0-5.7 263-550 163-450 58-160 13-31 19-51 521-1244	Ricknall Grange (Control for Skerne)
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	13-36 705-3286 393-2368 164-814 52-300 69-629 1408-6627	Houghton Road
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	5.0-91 1713-7650 625-5200 484-1413 138-450 350-613 3331-15416	Oxenfield Bridge
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	1.7-8.3 49-758 41-500 8.2-133 2.5-28 3.1-44 109-1473	Middleton in Teesdale (Control for Tees)

Table 1b (continued): Concentrations of PBDEs in trout and eels from the Skerne-Tees River system

Food	PBDEs analysed	Concentrations ($\mu\text{g}/\text{kg}$ fat basis)	Location
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	2.0-2.5 150-513 95-275 30-88 9.0-35 11-34 297-946	Low Coniscliffe
Trout	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	3.3-42 87-2779 60-1100 12-325 8.3-250 3.3-183 173-4078	Croft-on-Tees
Eels (single sample)	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	5.6 152 8.9 52 8.0 8.5 235	Ricknall Grange (Control for Skerne)
Eels	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.1-8.9 264-571 30-51 218-344 25-40 30-80 604-890	Oxenfield Bridge

Table 1b (continued): Concentrations of PBDEs in trout and eels from the Skerne-Tees River system

Food	PBDEs analysed	Concentrations ($\mu\text{g}/\text{kg}$ fat basis)	Location
Eels	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	6.1-6.6 157-743 11-18 72-222 11-37 16-49 274-1075	Croft on Tees
Eels	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	0.5-9.0 244-782 5.0-65 121-636 36-68 22-168 471-1665	Low Moor
Eels	BDE28 BDE47 BDE99 BDE100 BDE153 BDE154 Total	6.1-20 180-620 4.8-15 133-248 15-27 28-122 377-1003	Tees Barrage

Table 2a: Concentrations of HBCDs in trout from the Skerne-Tees River system

Food	HBCDs analysed	Concentrations (µg/kg fresh weight)	Location
Trout	α-HBCD β- HBCD γ- HBCD Total	12-87 5.9-18 3.3-17 21-119	Ricknall Grange (Control for Skerne)
Trout	α-HBCD β- HBCD γ- HBCD Total	84-5597 34-398 42-763 159-6758	Haughton Road
Trout	α-HBCD β- HBCD γ- HBCD Total	47-259 20-92 40-63 106-414	Oxenfield Bridge
Trout	α-HBCD β- HBCD γ- HBCD Total	1.2-31 1.2-18 1.2-2.3 3.6-51	Middleton in Teesdale (Control for Tees)
Trout	α-HBCD β-HBCD γ-HBCD Total HBCD	1.2-4.9 1.2-12 1.2-9.5 3.6-26	Low Coniscliffe
Trout	α-HBCD β-HBCD γ-HBCD Total HBCD	22-145 3.8-32 1.2-21 27-198	Croft-on-Tees

Table 2a (continued): Concentrations of HBCDs in eels from the Skerne-Tees River system

Food	HBCDs analysed	Concentrations ($\mu\text{g}/\text{kg}$ fresh weight)	Location
Eels (single sample)	α -HBCD β -HBCD γ -HBCD Total HBCD	110 15 34 159	Ricknall Grange (Control for Skerne)
Eels	α -HBCD β -HBCD γ -HBCD Total HBCD	535-8895 29-273 6.9-263 570-9432	Oxenfield Bridge
Eels	α -HBCD β -HBCD γ -HBCD Total HBCD	156-610 6.9-207 10-88 173-720	Croft on Tees
Eels	α -HBCD β -HBCD γ -HBCD Total HBCD	53-814 2.1-30 1.2-54 68-862	Low Moor
Eels	α -HBCD β -HBCD γ -HBCD Total HBCD	34-881 1.8-24 4.4-53 40-951	Tees Barrage

Table 2b: Concentrations of HBCDs in trout from the Skerne-Tees River system

Food	HBCDs analysed	Concentrations ($\mu\text{g}/\text{kg}$ fat basis)	Location
Trout	α -HBCD β - HBCD γ - HBCD Total	504-6710 244-1640 137-1240 885-9530	Ricknall Grange (Control for Skerne)
Trout	α -HBCD β - HBCD γ - HBCD Total	3011-133262 1196-9476 1486-18167 5693-160905	Haughton Road
Trout	α -HBCD β - HBCD γ - HBCD Total	8250-32375 3388-11475 6288-9900 17925-51763	Oxenfield Bridge
Trout	α -HBCD β - HBCD γ - HBCD Total	103-2567 103-1483 103-194 309-4244	Middleton in Teesdale (Control for Tees)
Trout	α -HBCD β -HBCD γ -HBCD Total HBCD	120-492 120-1150 120-948 360-2590	Low Coniscliffe
Trout	α -HBCD β -HBCD γ -HBCD Total HBCD	2905-31000 573-3570 200-5075 4145-38125	Croft-on-Tees

Table 2b (continued): Concentrations of HBCDs in eels from the Skerne-Tees River system

Food	PBDEs analysed	Concentrations ($\mu\text{g}/\text{kg}$ fat basis)	Location
Eels (single sample)	α -HBCD β -HBCD γ -HBCD Total HBCD	491 67 153 711	Ricknall Grange (Control for Skerne)
Eels	α -HBCD β -HBCD γ -HBCD Total HBCD	1651-26127 88-804 21-774 1760-27705	Oxenfield Bridge
Eels	α -HBCD β -HBCD γ -HBCD Total HBCD	586-2961 36-525 53-426 919-3496	Croft on Tees
Eels	α -HBCD β -HBCD γ -HBCD Total HBCD	158-37000 6.4-1368 3.6-800 204-39168	Low Moor
Eels	α -HBCD β -HBCD γ -HBCD Total HBCD	247-3781 13-231 32-323 292-4082	Tees Barrage

Table 3: Concentrations (µg/kg fat basis) of BFRs in food groups from the 2001 TDS

Chemical	Concentration (ug/kg fat basis)										
	Food group:	Carcase meat	Offals	Meat products	Poultry	Fish	Fats & oils	Potatoes	Milk	Milk products	Eggs
PBDE28		<5.5	<3.6	<6.1	<5.8	<5.1	<6.5	<5.3	<2	<6.1	<5.3
PBDE47		<5.5	<3.6	<6.1	<5.8	<5.1	<6.5	<5.3	<2	<6.1	<5.3
PBDE85		<5.5	<3.6	<6.1	<5.8	<5.1	<6.5	<5.3	<2	<6.1	<5.3
PBDE99		<5.5	<3.6	<6.1	<5.8	<5.1	<6.5	<5.3	<2	<6.1	<5.3
PBDE100		<5.5	<3.6		<5.8	<5.1	<6.5	<5.3	<2	<6.1	<5.3
PBDE153		<5.5	<3.6	<6.1	<5.8	<5.1	<6.5	<5.3	<2	<6.1	<5.3
PBDE154		<5.5	<3.6	<6.1	<5.8	<5.1	<6.5	<5.3	<2	<6.1	<5.3
PBDE183		<5.5	<3.6	<6.1	<5.8	<5.1	<6.5	<5.3	<2	<6.1	<5.3
Total PBDEs		<44	<28.8	<48.8	<46.4	<40.8	<52	<42.4	<16	<48.8	<42.4
PBB formulation		<5.5	<3.6	<6.1	<5.8	<5.1	<6.5	<5.3	<2	<6.1	<5.3
Alpha-HBCD		<2	<4.4	<1.5	<3.5	<3.6	<0.78	<9.4	<15	<1.4	<3.2
Beta-HBCD		<2	<4.4	<1.5	<3.5	<3.6	<0.78	<9.4	<15	<1.4	<3.2
Gamma -HBCD		<2	<4.4	<1.5	<3.5	<3.6	<0.78	<9.4	<15	<1.4	<3.2
Total HBCD		<6	<13.2	<4.5	<10.5	<10.8	<2.3	<28.2	<45	<4.2	<9.6
TBBP-A		<2	<4.4	<1.5	<3.5	<3.6	<0.78	<9.4	<15	<1.4	<3.2

Notes: Concentrations of total PBDEs and total HBCDs may not equal the sums of the concentrations of the individual congeners due to rounding.

Total PBDE values take no account of congeners that were not measured.

* These samples had very low fat contents. The fresh weight concentrations were low (see text.)

Table 3b (continued): Concentrations (µg/kg fat basis) of BFRs in food groups from the 2001 TDS

Chemical	Concentration (ug/kg fat basis)								
	Food group:	Misc. cereals	Bread	Nuts	Sugar & preserves	Fruit products	Green veg	Other veg	Canned veg
PBDE28	<5.4	<5	<5.2	<5.3	<4	<10	<2.5	<3.3	<3.3
PBDE47	<5.4	<5	<5.2	<5.3	<4	<10	<2.5	<3.3	<3.3
PBDE85	<5.4	<5	<5.2	<5.3	<4	<10	<2.5	<3.3	<3.3
PBDE99	<5.4	<5	<5.2	<5.3	<4	<10	<2.5	<3.3	<3.3
PBDE100	<5.4	<5	<5.2	<5.3	<4	<10	<2.5	<3.3	<3.3
PBDE153	<5.4	<5	<5.2	<5.3	<4	<10	<2.5	<3.3	<3.3
PBDE154	<5.4	<5	<5.2	<5.3	<4	<10	<2.5	<3.3	<3.3
PBDE183	<5.4	<5	<5.2	<5.3	<4	<10	<2.5	<3.3	<3.3
Total PBDEs	<43.2	<40	<41.6	<42.4	<32	<80	<20	<26.4	<26.4
PBB formulation	<5.4	<5	<5.2	<5.3	<4	<10	<2.5	<3.3	<3.3
Alpha-HBCD	<5.7	<13.7	<1.4	<2.5	<30	<30	<15	<30	<30
Beta-HBCD	<5.7	<13.7	<1.4	<2.5	<30	<30	<15	<30	<30
Gamma-HBCD	<5.7	<13.7	<1.4	<2.5	<30	<30	<15	<30	<30
Total HBCD	<17.1	<41.1	<4.2	<7.5	<90	<90	<45	<90	<90
TBBP-A	<5.7	<13.7	<1.4	<2.5	<30	<30	<15	<30	<30

Notes: Concentrations of total PBDEs and total HBCDs may not equal the sums of the concentrations of the individual congeners due to rounding.

Total PBDE values take no account of congeners that were not measured.

* These samples had very low fat contents. The fresh weight concentrations were low (see text.)

Table 4: Estimated upper bound adult dietary intakes of PBDEs via one portion per week of brown trout or eels from the Skerne-Tees River system

Fish type	Trout (7)			Trout (3)			Eels (5)			Trout (5)			Eels (3)			Trout (5)	Eels (1)
Location	Houghton Road			Oxenfield Bridge						Croft-on-Tees						Ricknall Grange	
Notes	Skerne, immediately downstream of Great Lakes			Skerne, downstream of Houghton Road and close to confluence						Tees, close to and downstream of confluence						Skerne, well upstream of Great Lakes, control	
Portion size (g)	120	120	120	120	120	120	70	70	70	120	120	120	70	70	70	120	70
Concentrations (µg/kg fresh weight)																	
	Mean	Min*	Max*	Mean	Min*	Max*	Mean	Min*	Max*	Mean	Min*	Max*	Mean	Min*	Max*	Mean	Mean
PBDE 28	0.8	0.5	1.3	0.3	0.0	0.7	1.5	0.0	2.5	0.2	0.0	0.6	1.7	1.3	2.5	0.1	1.3
PBDE 47	58	30	107	31	14	61	120	84	185	17	0.5	39	92	30	153	6.3	34
PBDE 99	30	17	34	18	5.0	42	11	10	14	6.2	0.4	13.6	3.0	2.1	4	4.2	2.0
PBDE 100	14	6.9	25	6.2	3.3	11	79	59	117	2.2	0.1	4.4	37	14	50	1.4	12
PBDE 153	5.1	2.2	9.1	1.8	0.8	3.6	11	6.8	13	0.8	0.1	1.5	5.5	2.0	8	0.3	1.8
PBDE 154	9.2	2.9	20	3.1	1.7	4.9	14	-	27	1.1	0.02	2.3	8.0	3.1	11	0.4	1.9
Total PBDEs	118	59	197	60	27	123	233	164	288	28	1.0	57	146	52	221	13	53
Estimated dietary intake (ng/kg bw/day) from one portion per week																	
PBDE 28	0.2	0.2	0.4	0.09	0.006	0.2	0.26	0.003	0.4	0.07	0.01	0.2	0.28	0.21	0.4	0.02	0.21
PBDE 47	17	8.5	31	8.7	3.9	17	19.9	14.0	30.8	4.9	0.1	11	15.4	4.9	25.5	1.8	5.7
PBDE 99	8.5	4.7	9.7	5.1	1.4	12	1.9	1.7	2.3	1.8	0.1	3.9	0.5	0.4	0.6	1.2	0.3
PBDE 100	4.1	2.0	7.2	1.8	0.9	3.2	13.2	9.9	19.5	0.6	0.02	1.3	6.1	2.3	8.4	0.4	2.0
PBDE 153	1.4	0.6	2.6	0.5	0.2	1.0	1.8	1.1	2.2	0.2	0.01	0.4	0.9	0.3	1.3	0.08	0.30
PBDE 154	2.6	0.8	5.7	0.9	0.5	1.4	2.4	1.6	4.5	0.3	0.01	0.7	1.3	0.5	1.8	0.1	0.32
Total PBDEs	34	17	56	17	7.0	35	39	27.3	48	7.9	0.3	18	24.3	8.6	37	3.6	8.8

Notes: * Samples with the minimum and maximum total PBDE concentrations. The minimum and maximum concentrations of the individual congeners were not always found in the same two samples. The figures in parentheses are the numbers of samples analysed.

Table 5: Estimated upper bound adult dietary intakes of HBCDs via one portion per week of brown trout or eels from the Skerne-Tees River system

Fish type	Trout (7)			Trout (3)			Eels (5)			Trout (5)			Eels (3)			Trout (5)	Eels (1)
Location	Houghton Road			Oxenfield Bridge						Croft-on-Tees						Ricknall Grange	
Notes	Skerne, immediately downstream of Great Lakes			Skerne, downstream of Houghton Road and close to confluence						Tees, close to and downstream of confluence						Skerne, well upstream of Great Lakes, control	
Portion size (g)	120	120	120	120	120	120	70	70	70	120	120	120	70	70	70	120	70
	Concentrations (µg/kg fresh weight)																
	Mean	Min*	Max*	Mean	Min*	Max*	Mean	Min*	Max*	Mean	Min*	Max*	Mean	Min*	Max*	Mean	Mean
Alpha-HBCD	1842	84	5597	91	47	144	2798	535	8895	41	22	67	291	106	610	54	110
Beta-HBCD	198	34	398	28	20	42	125	29	274	6.7	3.6	15	41	6.9	22	13	15.0
Gamma-HBCD	302	42	763	28	17	40	124	6.9	253	6.0	1.2	11	40	10	88	10	34
Total HBCDs	2341	159	6758	146	106	213	3047	570	9432	54	27	92	372	173	720	78	159
	Estimated dietary intake (ng/kg bw/day) from one portion per week																
Alpha-HBCD	526	24	1599	26	13	41	466	89	1483	12	6.3	19.1	55	26.0	102	15	18.3
Beta-HBCD	57	9.6	114	7.9	5.6	12	20.8	4.8	46	1.9	1.0	4.3	13.1	1.1	34.5	3.8	2.5
Gamma-HBCD	86	12	218	8.0	4.7	11	20.6	1.1	42.2	1.7	0.3	3.0	9.1	1.7	14.6	3.0	5.7
Total HBCDs	669	45	1931	42	24	64	508	95	1572	15	7.7	26	62	28.8	120	22	26.5

Notes: * Samples with the minimum and maximum total HBCD concentrations. The minimum and maximum concentrations of the individual congeners were not always found in the same two samples. The figures in parentheses are the numbers of samples analysed.

Sampling locations for the project

