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Persistent organic pollutants (PCB, DDT, HCH, HCB & BDE) in eels (*Anguilla anguilla*) in Scotland: current levels and temporal trends

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Abstract

Eels are an ideal biomonitor for persistent organic pollutants (POPs) because of their high lipid content, longevity and tendency to remain within a defined range during their freshwater life phase. This study investigated concentrations of POPs in eels (*Anguilla anguilla*) from 30 sites across Scotland, including polychlorinated biphenyls (PCBs), polybrominated diphenylethers (BDEs), DDT (and metabolites), hexachlorocyclohexanes (α,β,γ-HCH), hexachlorobenzene (HCB), hexachlorobutadiene (HCBD) and pentachlorobenzene. Despite its EU-wide ban ~30 years ago, DDT and its derivatives were detected in almost all samples. PCB 153 and 138 were the most widely detected PCB congeners, while BDE 47 was the dominant BDE. Pentachlorobenzene was not detected, while HCBD was detected once only. α-HCH, β-HCH and HCB concentrations were very low (generally <3 μg/kg or below detection). When compared with 1986 and 1995 data, the results revealed considerable decreases in p,p'-DDE concentrations. More drastic reductions were evident for γ-HCH, reflecting the tightening restrictions on pesticide use imposed over the previous decades.

Capsule

Persistent Organic Pollutants (POPs) are detectable in eels in many parts of Scotland. The concentrations are generally low, with evidence that they are decreasing for DDE and HCH.

Key words

Persistent organic pollutants; BDE; PCB; European eel; biota monitoring

Introduction

Persistent organic pollutants (POPs) are of global concern because of their toxicity, resistance to degradation, potential for long-range transport and their tendency to accumulate in fatty tissues (lipophilicity), the latter of which renders them likely to bioaccumulate through food-webs (Jones and de Voogt, 1999). The serious potential risks they pose to the environment and to human health are such that international treaties, e.g. the United Nations' Aarhus Protocol (UN, 1998) and Stockholm Convention (UNEP, 2005), which aim to eliminate or restrict their production and use, have been established. Dichlorodiphenyltrichloroethane (DDT), the infamous

organochlorine insecticide used extensively in the 1940s and 1950s, and polychlorinated biphenyls (PCBs), used in plastics, lubricants and dielectric fluids, were each targeted by both international treaties. Hexachlorocyclohexane (including forms α-HCH, β-HCH and γ-HCH), primarily used in pesticides, was covered by the Aarhus Protocol. Both legislations have mechanisms for incorporating additional POPs and it is understood that brominated or poly-brominated diphenyl ethers (BDEs) are to be included or are being considered (Scheringer, 2009). BDEs are also on the Oslo and Paris Commission (OSPAR) list of chemicals for priority action and their use as flame retardants, once widespread in a host of consumer and industrial goods, is now severely restricted within the European Union (EU) under the recast Restriction of Hazardous Substances (RoHS) Directive (European Commission, 2008). They are also named as priority substances under the EU Water Framework Directive (European Commission, 2000). Such recognition of these BDEs as potential environmental threats reflects the growing evidence of their ready accumulation in aquatic animal species and the resultant risks (Covaci et al., 2005; Darnerud, 2003; Law et al., 2006; Webster et al., 2007; Weijs et al., 2009).

In order to assess the extent and patterns of POPs contamination across Scotland, this study assessed the presence and concentrations of DDT (and its metabolites), PCBs, BDEs, HCHs, hexachlorobenzene (HCB), hexachlorobutadiene (HCBD) and pentachlorobenzene in tissues of freshwater eels (*Anguilla anguilla*) collected from rivers and streams from various parts of the country between 2004 and 2008. Scotland, having several population and heavy industry centres as well as large expanses of relatively pristine wilderness, is well-suited for studying dispersion of these substances and the range of environmental contamination that can develop. Moreover, eels are an ideal biomonitor species for POPs because of their morphology, geographic distribution, lifestyle and behaviour: eels are long-lived and relatively sedentary, spending up to 20 years in freshwater before migrating back to the sea to spawn (Larsson et al., 1991; FRS, 2009); they feed on fish and benthic organisms such as worms and snails; and they

have a high proportion of lipids that facilitates the accumulation of lipophilic contaminants such as POPs (Roose et al., 2003). Their extensive geographical distribution also allows for wider comparisons with studies from other parts of Europe (Bressa et al., 1997; Covaci et al., 2005; Oliveira Ribeiro et al., 2008; Roose et al., 2003; Weatherley et al., 1997). In this study, eels were collected from 30 sites across Scotland that encompass spatial, land use and population pressure variations. Nineteen of these sites were selected because they were previously surveyed for p,p'-DDE and γ-HCH concentrations in 1986 (Wells et al., 1987), while a further two of the sites were surveyed for p,p'-DDE, γ-HCH and selected PCBs in 1995 (Walmsley and Ridgway, 1995). This study therefore aimed to investigate the contamination extent of selected POPs in freshwater eels in Scotland, incorporating assessments of regional variation as well as temporal differences.

Methods & Materials

Sample Collection Sites

Eel samples were collected over a four-year period (2004 - 2008) from 30 sites across Scotland, representing various geographic regions and catchments of differing land use pressures (Figure 1, Table 1). Generally, sites in the Central Belt (between Glasgow and Edinburgh) and around Aberdeen are subject to industrial and population pressures, whereas sites in the south and in the north east (between Aberdeen and Inverness) are predominantly influenced by agricultural activities.

Eel collection and Processing

Eels were caught and collected by electrofishing. Ideally, five eels of length ≥30 cm were collected from each site. However, as it was not always possible to obtain sufficient eels of this size, 19% of eels analysed were between 23 and 30cm (Table 2). Once caught, eels were

transported alive to the laboratory, where they were humanely killed by freezing at -20°C. Samples were stored at -20°C until analysed. Eel physical parameters were measured and then tissue samples were prepared by removing the skin and flesh of each eel from the region behind the anus (i.e. distally from the anus). Each tissue sample was individually homogenised (Fisherbrand Powergen 125 Homogeniser), weighed and freeze dried. One gram of freeze-dried tissue from each eel was then extracted using Accelerated Solvent Extraction (ASE) with a mobile phase of acetone:hexane (1:2 v/v). Diatamaceous earth (DE, Aldrich) was used as a packing material (eel tissue was homogenised with DE, with an additional layer of pure DE above and below). The ASE conditions comprised: system pressure 1500 psi, oven temperature 100°C, oven heat up time 5 min, static time 5 min, flush volume 60%, 1 min nitrogen purge and 5 cycles per sample. ASE cells of 11 ml capacity were used. After extraction, samples were filtered, concentrated and exchanged to iso-octane using turbovaps (Zymark). Extracts were then adsorbed onto a 6 g acid silica column (column id 9 mm, with acid silica pre-conditioned by shaking vigorously 60 g silica gel with 4 ml concentrated sulfuric acid, 18M) and eluted with 180 ml hexane into a Zymark® tube. Once eluted, samples were again concentrated and exchanged to iso-octane using turbovaps. Extracts were then put through a 6 g deactivated alumina oxide column (id 9mm, with alumina prepared by mixing 100 g alumina oxide with 10 ml deionised water, 18MΏ) and eluted with 60 ml hexane. Eluents were concentrated and exchanged as above by turbovaps and capped for instrument analysis.

Eel lipid content was estimated by extracting a second portion of freeze dried eel tissue using the ASE conditions listed above but omitting column chromatography clean up. Once extracted, samples were quantitatively transferred to a 5 ml volumetric flask and made to volume using hexane. One ml was dispensed into a small pre-weighed beaker, placed on a hotplate at 90 ºC and periodically weighed until a constant weight was attained (50 min). The final weight was used to calculate the lipid content (Walmsley and Ridgway, 1995; Weatherley et al., 1997).

Chemical Analysis

Samples collected and processed during 2004-2006 were analysed for p,p'-DDT, o,p'-DDT, p,p'-DDD, p,p'-DDE, α,β,γ-HCH, HCB and HCBD as well as PCB congeners (IUPAC no.) 28, 52, 101, 105, 118, 138, 153, 156 and 180. Importantly, these PCB congeners included the 7 'indicator PCBs' (28, 52, 101, 118, 138, 153, 180) suggested by the International Committee for the Exploration of the Sea (ICES), which are recognised by the wider scientific community as a representative index of PCB contamination. Samples collected between 2006 and 2008 were analysed for the substances listed above but were additionally analysed for pentachlorobenzene; PCB congeners 13, 44, 47, 49, 66, 77, 81, 110, 114, 123, 126, 128, 141, 151, 157, 167, 169, 170, 183, 187, 189 and 194; and BDE congeners 28, 47, 66, 71, 75, 85, 99, 100, 138, 153, 154, 183 and 209.

Gas Chromatography *(samples collected 2004-2006)*

The prepared eel samples were analysed for organochlorines and PCBs using a Hewlett-Packard 5890 gas chromatograph with electron capture detector (GC-ECD) fitted with a fused silica capillary column (J & W Scientific, HP-5 60 m length x 0.25 mm id x 0.25 µm film thickness). Samples in hexane were injected (1 µL) on-column. The oven temperature program was 100 ºC to 210 ºC at 25 ºC/min held for 5 min, ramped to 240 ºC at 4 ºC/min, then to 260 ºC at 10 ºC/min and held for 5 min. Hydrogen was used as the carrier gas (30 psi head pressure) at a flow rate of 2-4 ml/min. The injection port tracked the oven temperature. The ECD was set to 300 ºC with a make up gas of 45 ml/min nitrogen.

Gas Chromatography/Triple Quadrupole Mass Spectrometry *(2006-2008 samples)*

Prepared samples were analysed for organochlorines and PCBs using a Varian 1200 L gas chromatograph with a triple quadrupole mass spectrometer (GC-MS/MS) fitted with a fused silica

capillary column (J & W Scientific, HP-5 60 m length x 0.25 mm id x 0.25 µm film thickness). Using 1079 injector, 1 µl of extract was injected at a temperature of 280 ºC. The injector program was: split off at 0.01 min, on at 1.5 min and then off at 5.0 min, with a split ratio of 100. The oven temperature program was 50 ºC held for 1 min ramped to 100 ºC at 12 ºC/min, then to 220 ºC at 7 ºC/min and then ramped to 325 ºC at 2.5 ºC/min and held for 2 min. The GC-MS/MS was used in MRM (multiple reaction monitoring) mode with the following conditions; transfer line temperature 325 ºC, ion source 250 ºC, electron impact mode with an electron energy of 70 eV, Argon CID gas set at 1.80 mTorr. The carrier gas was Helium with a flow rate of 1.5 ml/min. Quantitative analysis for BDEs was performed using a rapid MS fused silica capillary column (Varian, VF-5MS 10 m length x 0.53 mm i.d. x 0.25 µm film thickness). Using 1177 injector, 1 µl of extract was injected at a temperature of 250 ºC. The injector program was: split off at 0.01 min, on at 1.5 min and then off at 5.0 min, with a split ratio of 100. The oven temperature program was 80 ºC held for 1.5 min ramped to 250 ºC at 12 ºC/min and then to 300 ºC at 25 ºC/min and then held for 5.33 min. The GC-MS/MS was again used in MRM mode with the same settings as above, and tuned to include higher mass 614 for BDE 209. The carrier gas was Helium with a constant column flow 2 ml/min, pulse pressure 45 ml/min and pulse duration 1.60 min.

Quality Control

Certified standards, used for calibration, purchased from Ultra Scientific, included; 31 PCB congeners, HCH (α, β, γ-), HCB, p,p'-DDE, p,p'-DDD and p,p'-DDT. QMx Laboratories supplied HCBD, pentachlorobenzene and o,p'-DDT, while Cambridge Isotope Laboratory supplied 13 BDE congeners. Independent standards were prepared using QMx-supplied PCBs 28, 52, 101, 105, 118, 138, 153, 156, 180, 194, HCBD, pentachlorobenzene and o,p'-DDT. Independent standards were also prepared from the Ultra Scientific products α, β, γ-HCH, HCB, p,p'-DDE,

p,p'-DDD and p,p'-DDT, plus products from Greyhound (BDE 28, 47, 99, 153, 183 and 209). Internal standards were made using PCB congeners 112 and 198, supplied by QMx.

Extractions were conducted in small batches (5 samples), with each batch also including a process blank, eel matrix blank, spiked eel matrix and a certified reference material (Cambridge Isotopes EDF-2525). For analysis, the samples were batched with independent standards and also with calibration drift standards to assess concentration drift throughout the run. Calibration curves used for quantification were all better than r^2>0.995. Percentage recoveries (100× measured/certified values) for certified standards were 101% for BDE 153, 91% for p,p'-DDE and 92% for PCB 138.

All solvents used were glass distilled grade. Sodium sulphate (Fisher) and aluminium oxide (standard 90) were purified at 300 °C in a furnace for 2 h and cooled before use. Silica gel 60 (Merck) 0.063-0.200 mesh was purified at 500 °C for 4 h, then allowed to cool before use. Whatman filter papers (1PS) were cleaned in an ultrasonic bath for 1 h using hexane:acetone 1:1(v/v) and dried overnight. Glasswool was cleaned similarly to the filter papers but using hexane instead of hexane:acetone. All glassware was cleaned using an ultrasonic bath with 10% Decon for 1 h. Glassware was then rinsed thoroughly with water and placed in an oven for 3 hrs at 90 °C before it was cooled and used.

Although numerous advances in analytical methodology have been achieved since 1986, the methods used here for DDE and HCH quantification were comparable to those employed in the earlier studies (Walmsley and Ridgway, 1995; Wells et al., 1987) and thus comparisons between the data sets can be made. However, it should be noted that while the current study (and that from 1995) performed individual analyses on separate replicate eel samples for each location (i.e. n=5), the 1986 investigation analysed composite samples for each location that were pooled

from six eels (Wells et al., 1987). Importantly, the size range of eels in the current study matched those of the two previous studies.

Data treatment and statistical analysis

Following the convention adopted elsewhere (e.g. Webster et al., 2009; Zhao et al., 2009), when calculating means and summed values (e.g. Σ7PCB and total BDE burden) any measured concentrations falling below the method detection limit (MDL) were treated as equalling half the MDL. Statistical analyses for temporal comparisons were performed via Mann-Whitney U tests, using the statistical package Analyse-It. This non-parametric test was selected because the data, as commonly encountered in studies of contaminant concentrations in eels (e.g. Oliveira Ribeiro et al., 2008), did not consistently adhere to a normal distribution. For the same reason, data were also $\log_{10}$-transformed prior to the calculation of means for graphical presentations.

Results

Extent of contaminant detection

Considering all geographic regions, p,p'-DDT and its derivative products p,p'-DDE and p,p'-DDD were detected in almost all samples (Figure 2). β-HCH and γ-HCH were detected in a high proportion of samples (51% and 29% of all samples, respectively), while α-HCH was only detected in 2% of eels tested. HCB was detected in 45% of eels. HCBD (formerly used as an algaecide and also present in some industrial solvents) was detected in one eel only, while pentachlorobenzene was not detected in any sample. For PCBs, congeners 153, 138, 118 and 180 were most prevalent (Figure 2), with PCB 153 detected in 90% of samples. PCB 47, 49, 157, 169, 183, 189 and 194 were not detected in any sample. Of the BDEs, congeners 47 and 100 were the most frequently detected, while BDE 85, 138 and 183 were not detected in any of the eels examined (Figure 2).

Percentage detection of the various contaminants is presented on a regional basis in Figure 2. Very little regional difference was evident among the organochlorine pesticides (OC-Ps). There was similarly little difference between the regions for the most frequently detected PCBs (153, 138, 118, 101, 180, 105, 52). For the less prevalent PCBs, however, there was a general pattern in percentage detection of SE>SW>N, with the exception of PCB 151 that was only detected in eels from the northern region (at marginally quantifiable levels). Excluding BDE 47 and 100, i.e. the two most commonly detected congeners, this general SE>SW>N regional pattern was also observed in the percentage detections for BDEs (Figure 2).

Contaminant Concentrations (all values presented as wet weight)

PCB congener concentrations varied widely among sites (Table 3a and 3b), with sums of the 7 indicator congeners (Σ7PCBs) ranging from less than 10 μg/kg in eels from the Rivers Cree, Nith and Ythan to values of 2000 μg/kg or more in those from the River Clyde. The PCB results reflect the expected pattern of observing the greatest Σ7PCB concentrations in eels from industrial-influenced areas (e.g. the River Clyde in Glasgow and the River Don in Aberdeen, Table 3a). While it was rarely detected in eels from other regions, PCB 81 was detected in 60% of eels tested for it in the SE (Figure 2) and the highest concentrations for this congener were also observed in eels from the SE (site 22 and 23, Table 3b).

Concentrations of most BDE congeners were low, with many consistently below the respective MDLs (Table 4). However, as might be expected, sites in urbanised catchments tended to have greater eel total BDE burden than those in rural areas. Regarding OC-Ps (Table 5, Figure 3), concentrations of HCBD and α-HCH were almost all below detection, while HCB and β-HCH

were also very low (<3 μg/kg or below quantifiable levels). Concentrations of DDT derivatives varied, with the highest concentrations observed in eels from the Lunan Burn (eg 251 μg/kg median p,p'-DDT).

Temporal Changes

When combined with the previous studies, the current investigation provides a means to examine any temporal changes in the level of contaminants in eels in Scottish rivers. For p,p'-DDE, the highly environmentally persistent daughter product of DDT, a substantial decrease in concentration was observed at almost all locations when compared with 1986 values (Figure 3). Exceptions were one location on the River Cree (site 8), which had current concentrations equivalent to those in 1986, and the River Don (site 30), which had current concentrations above those recorded in 1986 (Figure 3). A more dramatic reduction was observed in γ-HCH levels, as all sites exhibited large decreases with many in the current investigation having concentrations below detection (Figure 4).

A comparison of the concentrations at the two River Devon sites (sites 14 and 15) with the levels recorded in 1995 (Walmsley and Ridgway, 1995) also revealed substantial reductions in γ-HCH concentrations at both locations (Figure 5). However, p,p'-DDE and Σ7PCB concentrations showed no significant temporal differences at either site ($p>0.05$, Mann-Whitney U test, Figure 5), indicating that the levels of these contaminants have not decreased in eels of the River Devon since 1995.

Discussion

To assess the relative environmental impact of the contaminant levels revealed by this study, the results need to be viewed in the context of the wider European situation and beyond. In terms of

γ-HCH concentration, the results observed here were similar to those reported for eels collected from rivers in Wales (Weatherley et al., 1997), as the concentrations there were also frequently below detection. The range of p,p'-DDE concentrations determined here for eels from Scottish rivers (<1 to 227 μg/kg, Figure 4) was greater than that observed in a Flemish study (Covaci et al., 2005) which examined eels from three ponds and one canal (and reported concentrations from 6 to 24 μg/kg), though this may reflect a greater historical agricultural influence on the Scottish sites than those examined in Flanders. However, a comparison of the PCB concentrations reported in that study (138 to 494 μg/kg, Covaci et al., 2005) with those of the current investigation (Tables 3a and 3b) revealed that PCB levels in the Scottish eels were generally much lower. The same was true when PCB levels from the present study were compared with those reported for eels from the Po Delta in NE Italy (Bressa et al., 1997) and for an analogous eel species (*Anguilla rostrata*) from the Delaware and Hudson River basins in the USA (Ashley et al., 2003). The Scottish eels examined here can therefore be considered to have comparatively low PCB levels, with the notable exception of eels taken from the River Clyde (*c.f.* Tables 3a and 3b) which has a history of heavy industrial inputs. Indeed, the range of PCB concentrations determined for the River Clyde eels were in keeping with those reported for eels resident in contaminated inland waters of the Netherlands (de Boer and Hagel, 1994). A further exception may be for PCB 81 in eels from sites 21-23 which, after conversion to dry weight equivalents for comparison (data in Tables 1 and 3b), were above the levels recorded in the Camargue Nature Reserve in France (Oliveira Ribeiro et al., 2008). The underlying cause of the higher concentrations of PCB 81 (a dioxin-like PCB) at sites 21-23 (SE region) is not known, but may reflect a different PCB input source or different balance of sources.

The relative abundances of the PCBs analysed in the eels of this study were similar to those reported elsewhere: in a study of eels taken from the Lesina Lagoon in SE Italy PCB 138 and 153 each accounted for ~20% of the total PCB load (Storelli et al., 2007), which matches the median proportions observed here of 20% for PCB 138 (median contribution to PCB load considering all locations, with min 2.5% and max 25%) and 20% for PCB 153 (min 2.7%, max 31%; see data in Tables 3a and 3b). Fromme et al. (1999) also reported very similar proportions for PCB 138 and 153 (23% and 21% of total PCB load, respectively) in eels from German rivers. This proportional dominance of PCB 138 and 153, as well as their widespread detection recorded here along with PCB 118 and 180 (Figure 2), may reflect historic use and subsequent environmental dispersion of commercial PCB mixtures Aroclor 1254 and Aroclor 1260, for which these congeners were important constituents. It may also reflect lower chemical degradation rates of these congeners and thus a higher retention in aquatic systems.

Regarding BDEs, the dominance of congener 47 observed here (Table 4) matches that observed by Covaci and co-workers (2005) who found that BDE 47 accounted for ~60 to 70% of the total BDE burden in the eels they examined. Moreover, the concentrations of BDE 47 recorded here for eels from Scottish rivers (Table 4) span a similar range to that reported in a study of river and lake sites across 10 European countries (Santillo et al., 2005). The concentrations of other BDE congeners measured in that study were also similar to those observed here, with BDE 66, 85, 99, 138, 153, 154 and 183 all being extremely low and/or below detection (Table 4).

The decreases in p,p'-DDE and γ-HCH concentrations observed at most sites in this study relative to values from 1986 reflects the various bans and tightening restrictions placed on the use and release of organochlorine pesticides which have come into force over the last few decades. These include the requirements under the Aarhus Protocol (UN, 1998), the EU Water

Framework Directive and UK legislation such as the Control of Pesticides Regulations Act. Similar, corresponding decreases in concentrations of POPs in biota have been reported elsewhere, such as for the brain and liver of glaucous gulls (*Larus hyperboreus*) from the Barents Sea which have shown notable decreases in concentration of these contaminants since 1989 (Sagerup et al., 2009).

Although this study was not intentionally designed to examine aspects of human health, and although the sampling sites involved were not necessarily in recognised fishing areas, it is informative to compare the results obtained with recommended tolerance limits. Considering PCBs, all but two individual eels from the most contaminated site (i.e. two eels from the River Clyde, Table 3a) were below the maximum tolerance limit for PCBs (2 mg/kg) stipulated by the US Food and Drug Administration (USFDA, 2009). With regard to DDT and its metabolites, including p,p'-DDE (Figure 3), none of the eels examined here would exceed the USFDA limit set for all fish types (5 mg/kg).

Conclusions

This investigation revealed that while DDT and its derivatives are still readily detectable in eels from Scottish rivers ~30 years after its EU ban, concentrations are lower compared to those observed in the 1980s. Reductions in γ-HCH have been even more distinct, to the point that eels from many sites now do not have detectable concentrations. PCB 153 was detected in 90% of samples, regardless of region or land use. Concentrations of Σ7PCB congeners and total BDE were higher in urbanised catchments. Nevertheless, in general the PCB levels observed were lower than reported in many other studies, while the BDE levels were in line with those reported elsewhere.

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Figures and Tables are presented in the following pages.

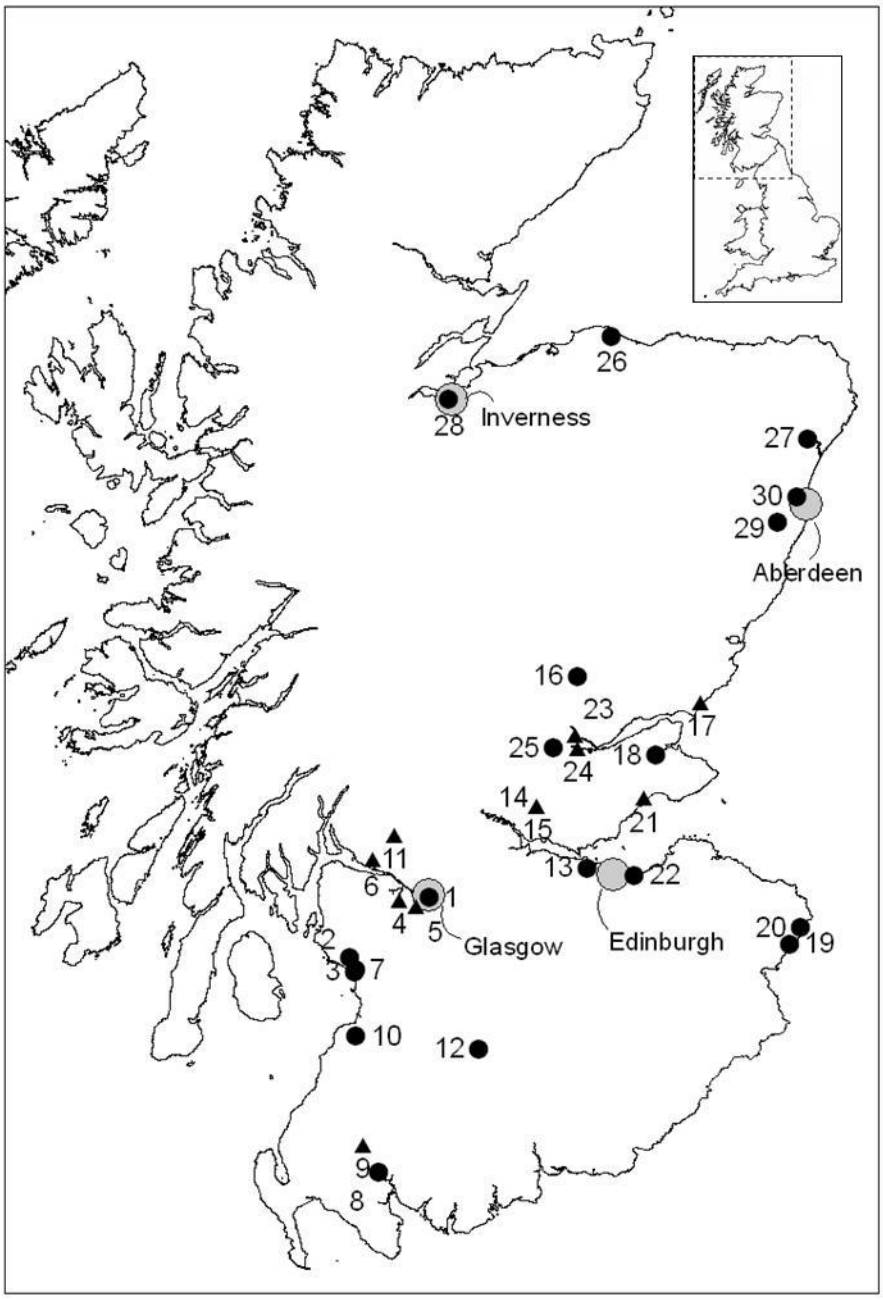

Figure 1. Map of Scotland (mainland and near-shore islands, with inset of Great Britain) showing the 30 eel collection sites (see Table 1 for location names). Black circles, as opposed to triangles, indicate sites that were also sampled in the 1986 study. Locations of the cities Aberdeen, Edinburgh, Glasgow and Inverness are indicated by grey circles.

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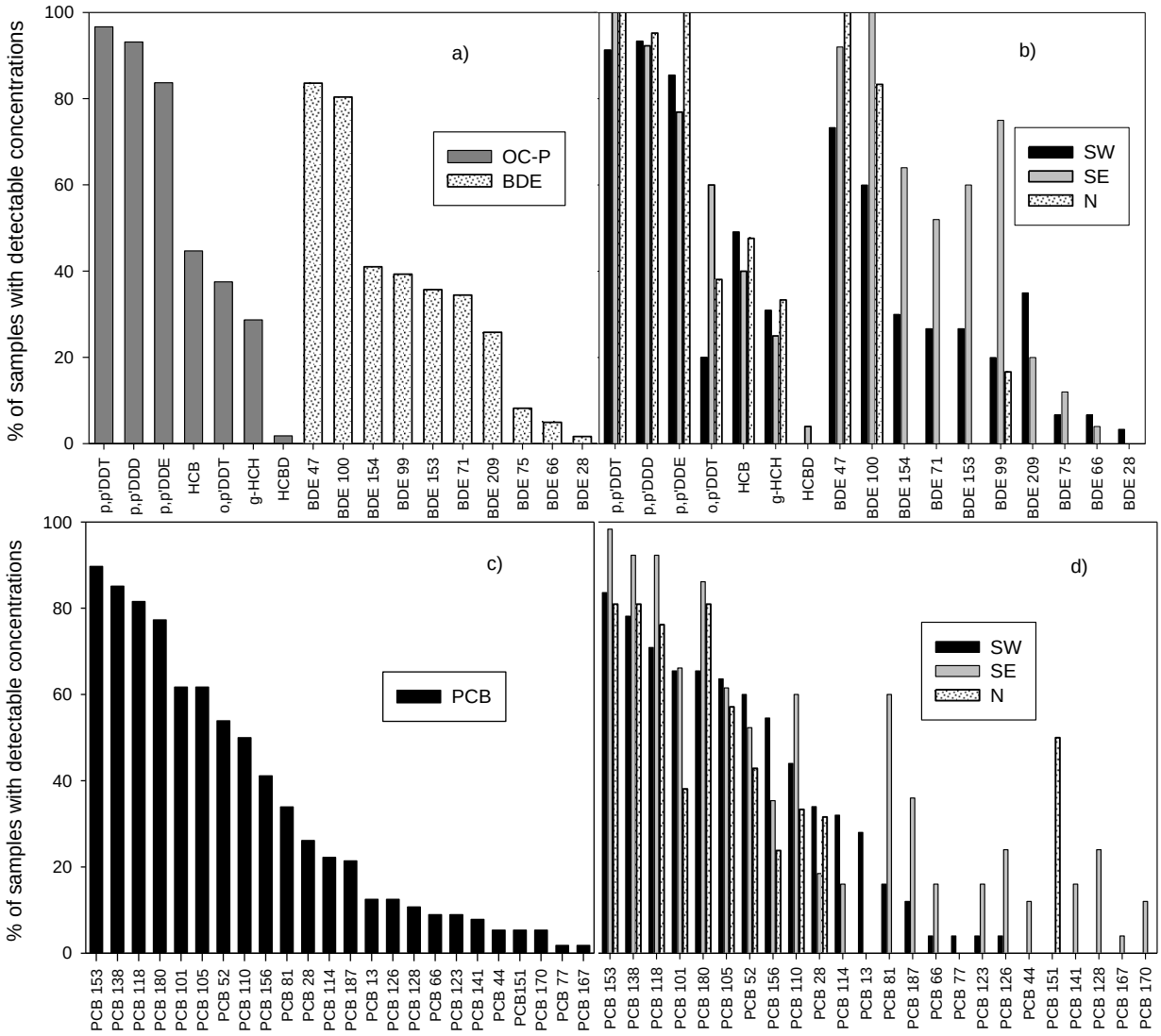

Figure 2. Percentage of samples in which the measured analyte was detected [i.e. 100x number of samples with detectable concentrations / number of samples analysed for a given POP]; a) percentages for organochlorine pesticides and BDEs considering all sites; b) percentages for organochlorine pesticides and BDEs by region; c) percentages for PCBs considering all sites; d) percentages for PCBs by region. BDE 85, 138 and 183 and PCB 47, 49, 157, 169, 183, 189 and 194 were not detected in any sample.

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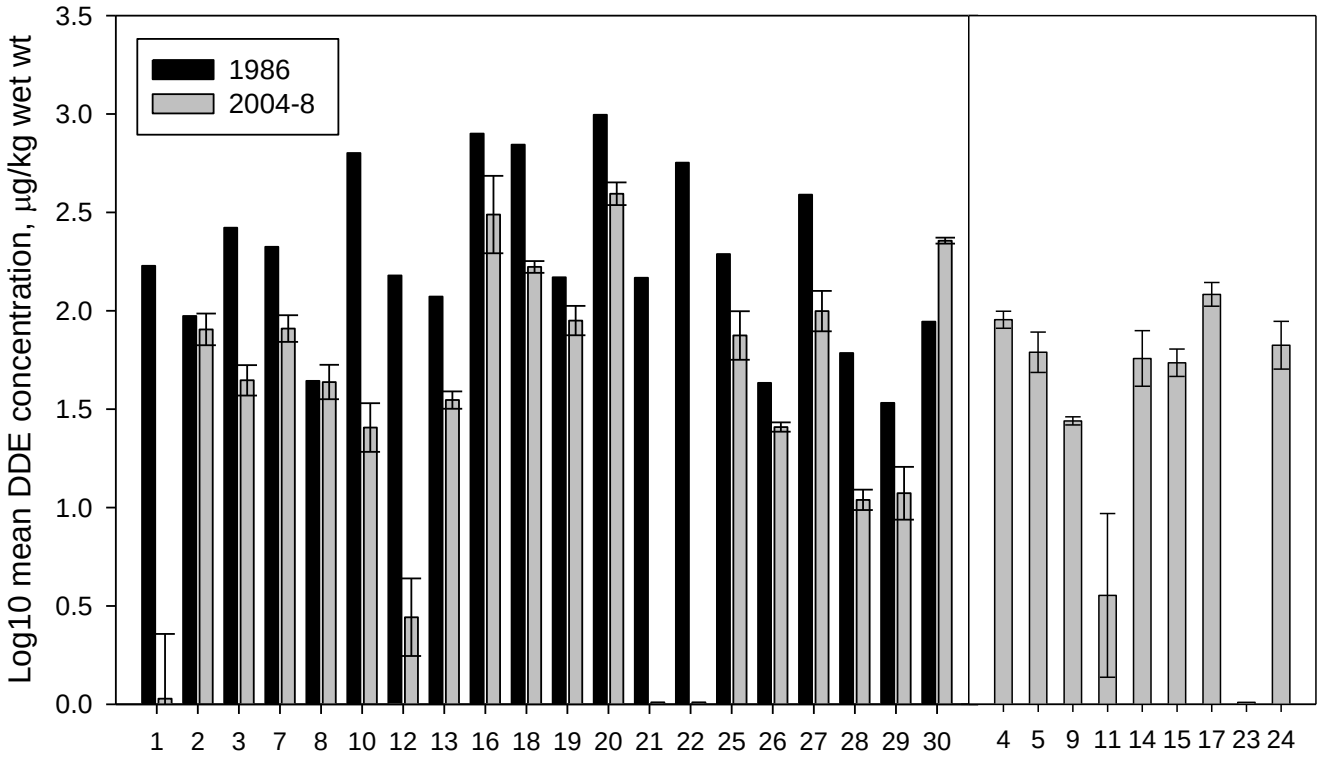

Figure 3. Log10 transformed DDE concentrations in eel muscle tissue sampled in 1986 and 2004-2008 (location names corresponding to site numbers, x-axis, are given in Table 1). Sites from the current investigation that were not sampled in the 1986 survey are presented in the right-most panel. Error bars indicate standard errors.

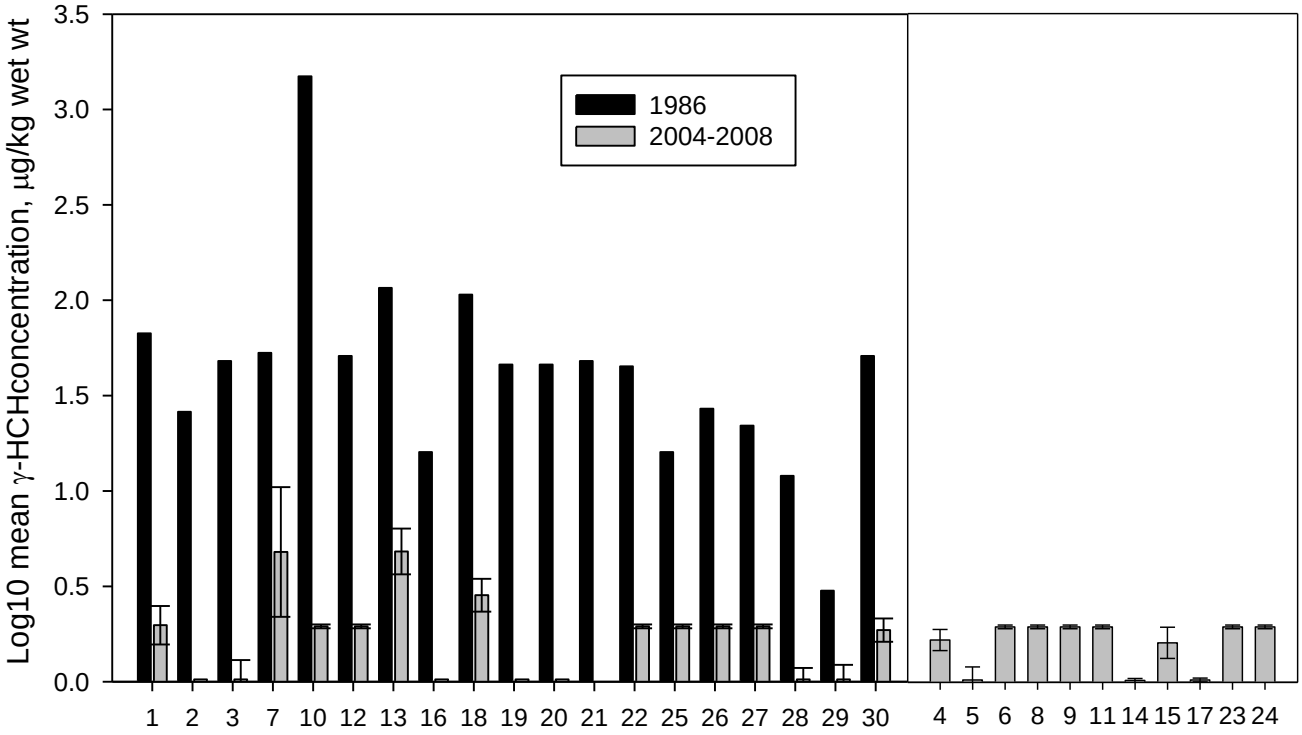

Figure 4. Log10 transformed γ-HCH concentrations in eel muscle tissue sampled in 1986 and 2004-2008 (location names corresponding to site numbers, x-axis, are given in Table 1). Sites from the current investigation that were not sampled in the 1986 survey are presented in the right-most panel. Error bars indicate standard errors. Note that the MDL varied from 1.0 to 3.9 μg/kg for the current data because of a methodology change. Also note that γ-HCH was not determined for site 21 in the current investigation.

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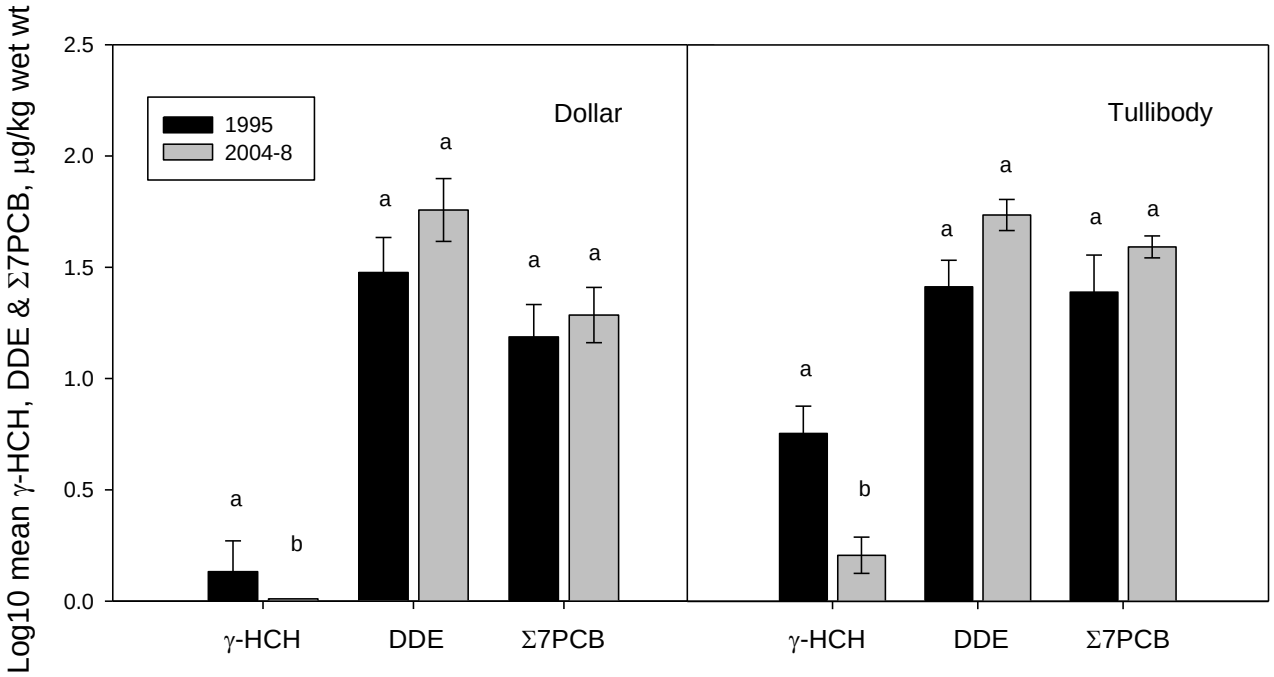

Figure 5. γ-HCH, DDE and Σ7PCB concentrations in eels from the River Devon sites of Dollar (site 14) and Tullibody (site 15) plotted against corresponding concentrations from 1995 (Walmsley and Ridgway, 1995). Error bars indicate standard error. For each contaminant and site, concentration values with different letters (*a* or *b*) above are significantly different according to a non-parametric Mann-Whitney U test ($p<0.05$).

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Table 1. Eel collection site locations and analytical suites measured

Site #	Location	NGR	Region	Urban/Rural	Analytical Suite
1*	River Clyde	NS 595645	SW	U	OC-P, PCB[a]
2*	River Garnock	NS 308427	SW	U	OC-P, PCB
3*	River Irvine	NS 325375	SW	U	OC-P, PCB
4	White Cart Water – Hammils	NS 486638	SW	U	OC-P, PCB
5	White Cart Water – Pollock	NS 548617	SW	U	OC-P, PCB
6	River Leven – Renton	NS 390785	SW	U	BDE
7*	Annick Water	NS 331383	SW	U	OC-P, PCB, BDE
8*	River Cree- Newton Stewart	NX 412653	SW	U	OC-P, PCB, BDE
9	River Cree – Bargrennan	NX 355753	SW	R	OC-P, PCB, BDE
10*	Culroy Burn	NS 331143	SW	R	OC-P, PCB, BDE
11	Endrick Water	NS 471873	SW	R	OC-P, PCB, BDE
12*	River Nith	NS 775096	SW	R	OC-P, PCB, BDE
13*	River Almond	NT 165752	SE	U+R	OC-P, PCB
14	River Devon – Dollar	NS 985979	SE	U+R	OC-P, PCB
15	River Devon – Tullibody	NS 985979	SE	U	OC-P, PCB
16*	Lunan Burn	NO 133445	SE	R	OC-P, PCB
17	Monikie Burn	NO 579353	SE	R	OC-P, PCB
18*	River Eden	NO 415158	SE	R	OC-P, PCB
19*	River Tweed	NT 898477	SE	R	OC-P, PCB
20*	Whiteadder Water	NT 939536	SE	R	OC-P, PCB
21	River Leven	NO 373006	SE	U	OC-P, PCB, BDE
22*	River Esk	NT 339724	SE	U	OC-P, PCB, BDE
23	River Tay	NO 122232	SE	U	OC-P, PCB, BDE
24	River Earn - Bridge of Earn	NO 132186	SE	R	OC-P, PCB, BDE
25*	River Earn	NO 043184	SE	R	OC-P, PCB, BDE
26*	River Lossie	NJ 254673	N	R	OC-P, PCB, BDE
27*	River Ythan	NJ 966304	N	R	OC-P, PCB, BDE
28*	River Ness	NH 665445	N	U+R	OC-P, PCB
29*	River Dee	NJ 858003	N	R	OC-P, PCB
30*	River Don	NJ 924093	N	U	OC-P, PCB

* Sites marked with an asterisk were also included in the 1986 survey by Wells *et al. (*1987), while sites 14 and 15 were surveyed in 1995 by Walmsley and Ridgway.

[a] OC-P= Organochlorine pesticides; PCB= polychlorinated biphenyls; BDE= Brominated diphenylethers. Note that capacity to measure BDEs dates from 2006, thus samples collected and analysed between 2004 and 2006 were not assessed for BDE concentrations.

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Table 2. Physical parameters of eels (mean±standard error) examined in the study

Site #	*n*	Length, cm	Fresh Weight, g	Lipid, g/g
1	5	36.8±0.7	84.4±4.2	0.33±0.09
2	5	39.4±3.6	116±30	0.40±0.02
3	5	34.9±1.1	80.6±8.0	0.44±0.02
4	5	35.0±2.6	90.2±20.2	0.45±0.02
5	5	54.8±2.1	366±50	0.49±0.03
6	5	41.4±4.7	133±38	0.43±0.05
7	7	34.0±1.0	72.9±7.7	0.43±0.05
8	4	31.2±1.2	48.4±7.5	0.52±0.05
9	5	27.9±2.4	36.6±9.7	0.55[a]
10	4	30.2±2.7	48.5±13.0	0.40±0.07
11	5	30.3±2.3	45.4±13.1	0.14±0.03
12	5	32.8±1.4	48.9±8.2	0.29±0.11
13	5	40.9±1.1	106±9.4	0.35±0.06
14	5	34.4±1.8	76.8±9.9	0.32±0.10
15	5	30.1±1.8	47.7±9.9	0.36±0.11
16	5	44.3±2.8	153±28	0.40±0.08
17	5	40.0±2.5	123±21	0.31±0.05
18	5	28.2±1.9	39.2±8.9	*na*
19	5	26.1±1.3	31.6±8.8	0.23±0.11
20	5	44.4±2.9	191±47	0.41±0.05
21	5	38.9±2.1	107±17	0.43±0.09
22	5	29.7±1.5	45.5±7.7	0.46±0.07
23	5	33.1±0.5	58.6±7.6	0.44±0.03
24	5	30.1±2.4	49.0±12.3	0.31±0.08
25	5	49.7±4.9	238±68	0.37±0.07
26	2	29.2±1.9	44.2±11.9	0.47[a]
27	4	28.3±0.9	37.2±4.6	0.22±0.01
28	5	33.8±1.2	71.2±4.8	0.30±0.13
29	5	32.5±1.1	52.9±3.7	0.21±0.06
30	5	33.8±0.6	80.2±9.4	0.51±0.02

[a] single lipid value determined; *na* not applicable (no value determined).

Table 3a: Concentrations of PCB congeners measured in eels from all sites: median (min-max[a]), expressed as μg/kg wet weight

Site #	PCB 28	PCB 52	PCB 101	PCB 105	PCB 118	PCB 138	PCB 153	PCB 156	PCB 180	Σ7PCB [d]
1	2.2 (1.4-18)	140 (12-1038)	315 (13-3197)	230 (6.2-1137)	501 (<1.0-2445)	454 (20-2171)	306 (22-1343)	65 (2.6-310)	64 (9.8-274)	1878 (79-10487)
2	<1.0[b]	9.9 (4.4-18)	10 (5.0-12)	5.7 (3.7-7.8)	16 (10-30)	24 (14-40)	28 (14-43)	2.8 (1.4-3.3)	12 (4.9-14)	103 (72-146)
3	<1.0 (1.3)	3.6 (2.1-4.8)	5.8 (3.7-11)	7.9 (2.7-8.3)	18 (6.4-22)	32 (10-53)	30 (11-44)	3.5 (1.3-5.1)	10 (3.8-13)	107 (38-137)
4	1.6 (1.2-2.0)	17 (13-37)	39 (15-46)	14 (12-17)	55 (38-56)	45 (37-57)	43 (33-59)	3.3 (2.7-3.5)	6.1 (4.0-6.7)	218 (147-239)
5	1.2 (1.0-1.2)	6.4 (5.1-8.2)	10 (5.2-12)	7.2 (6.9-8.8)	9.5 (7.9-12)	20 (19-49)	17 (16-21)	2.6 (2.5-3.2)	7.9 (7.0-9.4)	69 (63-114)
7	<1.0	3.3 (1.0-10)	5.0 (1.7-21)	2.4 (<1.8-15)	10 (5.5-38)	9.7 (6.0-44)	7.9 (5.3-38)	1.2 (<1.0-5.2)	3.0 (2.2-8.6)	41 (25-161)
8	<1.7	<2.1	<1.3	<1.8	<1.8	<1.8	<1.9 (2.3)	<1.8	<1.3	7.1 (6.7-8.1)
9	<1.7	<2.1	<1.3	<1.8	<1.8	<1.8 (2.19)	<1.9 (3.1)	<1.8	<1.3	7.5 (6.5-8.9)
10	<1.7	<2.1	<1.3	<1.8	<1.8	<1.8 (2.8)	<1.9 (3.4)	<1.8	<1.3	7.0 (6.5-11)
11	<1.7	<2.1	1.76 (<1.3-3.7)	1.4 (1.4-2.6)	4.5 (4.3-7.2)	5.9 (5.8-9.4)	5.1 (4.0-7.7)	<1.8	1.8 (<1.3-3.1)	21 (18-34)
12	*na*	<2.1	<1.3	<1.8	<1.8 (2.1)	<1.8	<1.9 (2.8)	<1.8	<1.3	5.9 (5.6-9.4) [e]
13	1.3 (<1.0-2.0)	15 (8.0-21)	21 (19.7-35)	16 (14-16)	42 (40-69)	45 (42-79)	37 (5.2-69)	4.2 (3.9-56)	7.7 (6.8-11.8)	172 (155-270)
14	<1.0	<1.0	1.3 (<1.0-1.6)	1.3 (<1.0-1.8)	3.0 (1.3-5.1)	6.1 (1.9-31)	4.1 (2.5-9.7)	<1.0	1.3 (<1.0-2.6)	22 (8-41)
15	<1.0	1.9 (1.1-4.6)	3.2 (1.4-5.6)	5.3 (3.4-6.7)	10 (5.7-13)	11 (7.5-14.3)	9.7 (7.8-14)	1.6 (1.0-2.4)	2.3 (1.7-3.9)	42 (26-51)
16	<1.0	<1.0	<1.0	<1.0	<1.0 (2.2)	2.0 (1.5-4.8)	2.6 (1.7-6.3)	<1.0	1.1 (<1.0-2.0)	7.9 (5.7-17)
17	<1.0	4.4 (4.0-5.1)	2.9 (<1.0-3.7)	1.3 (<1.0-1.8)	2.9 (2.2-5.0)	4.6 (3.7-8.9)	5.7 (5.0-12)	<1.0 (1.1)	2.2 (1.8-3.8)	24 (20-38)
18	<1.0 (1.8)	2.1 (1.9-3.9)	2.3 (2.0-5.5)	2.1 (1.9-2.6)	4.6 (4.0-5.4)	7.0 (6.4-7.5)	8.2 (7.8-8.6)	1.2 (1.0-1.6)	2.7 (2.6-2.9)	29 (26-34)
19	<1.0	2.1 (1.4-2.7)	1.8 (1.5-2.1)	1.2 (1.1-1.6)	2.2 (1.7-3.7)	3.5 (2.4-5.4)	4.2 (3.1-7.0)	<1.0 (1.6)	1.4 (1.1-2.7)	17 (13-23)
20	1.4 (<1.0-3.3)	1.0 (<1.0-2.8)	2.6 (<1.0-4.1)	2.0 (1.6-4.7)	5.5 (4.2-13)	6.4 (5.8-15)	8.7 (7.9-18)	1.3 (1.0-1.8)	3.2 (2.8-5.4)	29 (22-61)
21	<1.7 (2.0)	<2.1 (6.4)	2.8 (<1.3-13)	<1.8 (8.9)	5.0 (1.1-26)	5.7 (3.9-27)	7.14 (1.7-25)	<1.8 (3.4)	4.0 (<1.3-8.1)	27 (7.8-107)
22	<1.7 (2.1)	2.7 (<2.1-5.8)	2.0 (<1.3-4.7)	3.5 (<1.8-5.8)	8.6 (3.3-16)	15 (6.7-23)	12 (6.0-18)	<1.8	4.5 (3.0-6.6)	43 (25-75)
23	<1.7	<2.1	<1.3	<1.8	1.9 (<1.8-2.6)	2.6 (1.1-3.8)	*na*	<1.8	1.8 (<1.3-3.3)	[f]
24	<1.7	<2.1	<1.3 (1.6)	<1.8	1.8 (1.2-3.9)	<1.8 (6.3)	<1.9 (9.9)	<1.8	<1.3 (5.2)	7.3 (6.9-29)
25	<1.7	<2.1	3.2 (<1.3-5.0)	<1.8	5.9 (<1.8-7.8)	13 (<1.8-30)	24 (4.4-25.2)	<1.8	4.3 (<1.3-5.0)	54 (9.9-62)
26	*na*	<2.1	(1.6-2.2) [c]	(1.8-2.0)[c]	(3.6-5.9)[c]	(6.0-7.6)[c]	(7.1-9.4) [c]	<1.8	(2.1-2.6)[c]	(29-22) [c e]
27	<1.7	<2.1	<1.3	<1.8	<1.8	<1.8	<1.9	<1.8	<1.3	6.5 (6.5-6.6)
28	<1.0	1.6 (<1.0-4.0)	<1.0 (1.6)	<1.0 (1.5)	1.8 (1.3-3.2)	4.1 (2.9-4.2)	4.8 (3.7-5.4)	<1.0	2.2 (1.4-2.7)	15 (13-22)
29	<1.0	<1.0 (1.0)	<1.0	1.2 (<1.0-2.9)	1.7 (<1.0-2.6)	3.0 (1.0-5.2)	5.0 (2.1-6.2)	<1.0	2.2 (1.0-2.6)	13 (6.1-19)
30	2.4 (2.1-2.8)	18 (4.7-101)	18 (8.7-73)	21 (15-269)	63 (45-482)	84 (53-437)	100 (55-370)	9.8 (6.7-76)	34 (13-259)	327 (197-1723)

[a] In cases where the median is also the minimum, the max value only is presented inside parentheses; [b] <*x* indicates value was below the method detection limit (MDL), with the MDL equalling x; [c] Two samples only, thus min and max are presented; [d] Sum of 7 indicator PCBs (28, 52, 101, 118, 138, 153, 180), as per the International Committee for the Exploration of the Sea (ICES), for which any congener with a concentration below the MDL was counted as having a concentration equal to half the MDL. [e] Value calculated excluding missing minor constituent (PCB 28); [f] Σ7PCB not calculated for River Tay (site 23) because a major potential constituent (PCB 153) was not determined; *na* not applicable as concentration was not determined.

Table 3b: Additional PCB congeners[a] measured only in eels collected from sites sampled during 2006-2008: median (min-max[b]) concentration, expressed as μg/kg wet weight

Site	PCB 13	PCB 44	PCB 66	PCB 81	PCB 110	PCB 114	PCB 123	PCB 126	PCB 128	PCB 141	PCB 187
7	<2.4	<1.3	<1.9	<2.4	(4.2-8.1)[c]	<1.6	(<2.2-10)[c]	(<3.1-4.48)[c]	<2.2	<2.6	<3.0
8	<2.4	<1.3	<1.9	<2.4	<2.2	<1.6	<2.2	<3.1	<2.2	<2.6	<3.0
9	<2.4	<1.3	<1.9	<2.4	<2.2	<1.6	<2.2	<3.1	<2.2	<2.6	<3.0 (3.1)
10	<2.4	<1.3	<1.9	<2.4	<2.2	<1.6	<2.2	<3.1	<2.2	<2.6	<3.0 (4.69)
11	2.9 (<2.4-3.5)	<1.3	<1.9	22 (<2.4-42)	2.9 (<2.2-6.0)	2.9 (2.7-5.1)	<2.2	<3.1	<2.2	*na*	<3.0 (3.3)
12	4.3 (<2.4-6.4)	<1.3	<1.9	<2.4 (13)	<2.2 (3.0)	1.8 (<1.6-2.6)	<2.2	<3.1	<2.2	<2.6	<3.0
21	<2.4	<1.3 (2.3)	<1.9 (3.5)	26 (12-33)	3.7 (1.4-18)	<1.6	<2.2	<3.1	<2.2	<2.6 (3.9)	3.21 (5.9)
22	<2.4	<1.3 (2.18)	<1.9 (2.12)	33 (13-61)	6.3 (<2.2-11)	<1.6	<2.2 (9.6)	4.2 (<3.1-5.7)	<2.2 (3.0)	2.8 (<2.6-4.0)	<3.0 (4.0)
23	<2.4	<1.3	<1.9	50 (17-73)	<2.2	<1.6	<2.2	<3.1	<2.2	<2.6	<3.0 (4.7)
24	<2.4	<1.3	<1.9	<2.4	<2.2 (2.5)	<1.6 (17)	<2.2	<3.1	<2.2	<2.6	<3.0 (3.6)
25	<2.4	<1.3	<1.9	<2.4	2.8 (<2.2-5.1)	3.3 (<1.6-23)	<2.2 (8.3)	7.4 (<3.1-8.0)	4.3 (<2.2-6.2)	<2.6	<3.0 (7.2)
26	<2.4	<1.3	<1.9	<2.4	(3.0-3.7)*	*nd*	<2.2	<3.1	<2.2	<2.6	<3.0
27	<2.4	<1.3	<1.9	<2.4	<2.2	<1.6	<2.2	<3.1	<2.2	<2.6	<3.0

[a] Additional PCBs also included congeners 47 (MDL 2.1μg/kg), 49 (MDL 2.1μg/kg), 169 (MDL 2.0μg/kg), 157 (MDL 1.7μg/kg), PCB 183 (MDL 3.7μg/kg), PCB 189 (MDL 3.2μg/kg) and PCB 194 (MDL 2.8μg/kg), but these were not detected in any sample. Further congeners analysed but not displayed above were 77 (MDL 1.9μg/kg, observed in one eel only at site 11, concentration 3.4μg/kg), 151 (MDL 2.1μg/kg, detected only at site 27, <2.1-2.9μg/kg), 167 (MDL 1.9μg/kg, detected in one eel only at site 21, 4.1μg/kg) and 170 (MDL 3.8μg/kg, detected at site 24 only, <3.8-4.4μg/kg); [b] In cases where the median is also the minimum, the max value only is presented inside parentheses; [c] Two samples only, thus min and max are presented; *na* not applicable as no value obtained.

Table 4: BDE concentrations in eel tissue: median (min-max[a]), expressed as μg/kg wet weight

Site #	BDE 28	BDE 47	BDE 66	BDE 71	BDE 75	BDE 85	BDE 99	BDE 100
6	<0.36	9.2 (2.9-21)	<0.42	<0.52 (1.18)	<0.32 (0.52)	<0.48	0.99 (<0.52-5.4)	3.5 (1.9-7.9)
7	<0.36	(48-96) [c]	(0.43-0.52) [c]	(1.7-3.0) [c]	(<0.32-0.35) [c]	<0.48	(2.4-3.1) [c]	(19-37) [c]
8	<0.36	<0.58	<0.42	<0.52	<0.32	<0.48	<0.52	<0.44
9	<0.36	0.62 (<0.58-1.26)	<0.42	<0.52	<0.32	<0.48	<0.52	<0.44 (0.54)
10	<0.36	1.2 (<0.58-2.0)	<0.42	<0.52	<0.32	<0.48	<0.52	0.58 (<0.44-1.4)
11	<0.36 (0.38)	16.4 (9.3-31)	<0.42	<0.52 (0.98)	<0.32	<0.48	<0.52 (0.90)	*na*
12	<0.36	2.58 (<0.58-16)	<0.42	<0.52 (0.65)	<0.32	<0.48	<0.52	3.13 (0.73-5.7)
21	<0.36	12.0 (6.6-20)	<0.42	<0.52 (0.87)	<0.32	<0.48	1.07 (<0.52-1.7)	4.20 (2.4-6.9)
22	<0.36	16.2 (7.6-49)	<0.42 (0.46)	0.61 (<0.52-1.5)	<0.32 (0.48)	<0.48	1.9 (0.4-4.6)	17 (7.4-31)
23	<0.36	18.9 (4.6-22)	<0.42	0.71 (<0.52-1.2)	<0.32	<0.48	1.10 (<0.52-2.6)	6.75 (3.2-9.6)
24	<0.36	1.71 (<0.58-13)	<0.42	<0.52 (1.1)	<0.32	<0.48	<0.52 (0.84)	1.24 (0.48-7.1)
25	<0.36	10.9 (3.3-16)	<0.42	0.54 (<0.52-1.1)	<0.32 (0.32)	<0.48	*na*	2.85 (0.94-4.30)
26	<0.36	(0.94-3.7) [c]	<0.42	<0.52	<0.32	<0.48	<0.52	(<0.44-2.0) [c]
27	<0.36	2.5 (1.4-5.2)	<0.42	<0.52	<0.32	<0.48	<0.52 (0.6)	1.22 (0.9-2.1)

Table 4: *continued*

Site #	BDE 138	BDE 153	BDE 154	BDE 183	BDE 209	Total BDE [d] (ex. BDE 209)	%BDE47 [e]
6	<0.32	0.81 (<0.54-1.8)	0.63 (<0.52-1.3)	<0.64	<3.0 (10)	16.9 (7.1-44)	55 (40-61)
7	<0.32	(3.5-4.4) [c]	(2.9-4.3) [c]	<0.64	*na*	(98-132) [c]	(49-73)[c]
8	<0.32	<0.54	<0.52	<0.64	*na*	2.83 (2.8-3.0)	10 (10-11)
9	<0.32	<0.54	<0.52	<0.64	<3.0	3.16 (3.15-3.8)	20 (9.2-33)
10	<0.32	<0.54	<0.52	<0.64	*na*	3.0 (1.3-3.3)	27 (10-44)
11	<0.32	<0.54 (0.77)	0.57 (<0.52-0.87)	<0.64	1.63 (1.1-6.2)	20.4 (11-40) [f]	86 (81-87) [f]
12	<0.32	<0.54 (0.72)	<0.52 (0.96)	*na*	<3.0	8.03 (3.0-26) [f]	32 (8.8-64) [f]
21	<0.32	*na*	1.05 (<0.52-1.6)	<0.64	*na*	21.0 (12-32) [f]	57 (49-63) [f]
22	<0.32	1.66 (<0.54-3.0)	1.65 (<0.52-2.4)	<0.64	<3.0 (3.8)	34 (18-84)	48 (32-59)
23	<0.32	0.93 (0.3-1.2)	0.89 (<0.52-1.4)	<0.64	*na*	33.2 (10-38)	59 (44-65)
24	<0.32	<0.54 (1.07)	<0.52 (1.18)	<0.64	*na*	3.60 (1.6-24)	34 (9.4-51)
25	<0.32	<0.54 (0.87)	<0.52 (1.04)	*na*	*na*	15.0 (4.4-23) [f]	66 (55-68) [f]
26	<0.32	<0.54	<0.52	<0.64	<3.0	3.47-7.56	(27-46) [c]
27	<0.32	<0.54	<0.52	<0.64	<3.0	6.0 (4.6-10)	40 (30-52)

[a] In cases where the median is also the minimum, the max value only is presented inside parentheses; [b] <*x* indicates value was below the method detection limit (MDL), with the MDL equalling x; [c] Two samples only, thus min and max are presented; [d] Total BDE concentration, excluding BDE 209, calculated following the principal that when individual congener concentrations are <MDL they are treated as equalling half the MDL (Webster et al. 2009; Zhao et al 2009); [e] %BDE 47 equals the percentage of total BDE load, excluding BDE 209, represented by BDE 47 (calculated with <MDLs set to half MDL). [f] Value calculated excluding missing minor constituent. *na* not applicable as concentration was not determined.

Table 5: Concentrations of organo-chlorine contaminants determined in eels: median (min-max[a]), expressed as μg/kg wet weight

Site #	HCBD	HCB	α-HCH	β-HCH	p,p'-DDT	o,p'-DDT	p,p'-DDD
1	<1.0 [b]	1.0 (<1.0-2.3)	<1.0	1.5 (<1.0-2.0)	<1.0 (7.1)	<1.0 (1.3)	34.8 (<1.0-181)
2	<1.0	1.5 (1.0-2.1)	<1.0	1.9 (1.2-2.6)	13.9 (7.6-15)	2.4 (1.4-4.2)	12.3 (8.7-38)
3	<1.0	1.2 (1.1-1.6)	<1.0	1.4 (<1.0-2.3)	7.5 (5.8-10)	<1.0 (1.9)	10.4 (8.1-17)
4	<1.0	2.1 (1.9-3.0)	<1.0	2.9 (1.5-3.1)	21.3 (15.0-44.3)	<1.0	48.8 (35-61)
5	<1.0	1.5 (1.3-1.9)	<1.0	<1.0 (1.2)	5.5 (3.4-8.9)	<1.0	10.2 (8.3-13)
7	<1.0	1.7 (<1.0-2.8)	<1.0	1.2 (<1.0-2.2)	9.7 (6.4-19)	<1.0 (2.0)	18.7 (6.5-34)
8	<3.3	<3.0	<2.0	<1.9 (2.3)	3.0 (1.7-6.1)	<1.1	5.5 (3.5-6.9)
9	<3.3	<3.0	<2.0	*na*	*na*	<1.1	11.0 (8.2-12)
10	<3.3	<3.0	<2.0	<1.9	*na*	<1.1	3.4 (<2.0-6.3)
11	<3.3	<3.0	<2.0 (3.0)	<1.9	5.1 (3.4-8.2)	*na*	*na*
12	<3.3	<3.0	<2.0	5.2 (4.5-8.0)	5.6 (3.2-9.2)	<1.1	*na*
13	<1.0	1.8 (<1.0-2.8)	<1.0	1.8 (1.0-4.5)	15.4 (12.6-25)	3.2 (<1.0-3.4)	8.1 (5.6-15)
14	<1.0	<1.0	<1.0	1.3 (1.2-2.7)	17.7 (8.8-41)	1.0 (<1.0-1.4)	9.7 (2.8-16)
15	<1.0	1.5 (<1.0-3.6)	<1.0	2.6 (1.8-3.6)	20.7 (13.1-26.4)	1.3 (<1.0-4.5)	12.5 (6.7-19)
16	<1.0	1.4 (1.2-1.7)	<1.0	2.3 (4.3)	251 (11.3-992)	10.5 (<1.0-42.6)	53.4 (16-191)
17	<1.0	1.9 (<1.0-2.4)	<1.0	<1.0 (1.2)	71.9 (40.4-94)	5.6 (2.7-8.6)	24.9 (14.8-33.3)
18	<1.0	1.2 (<1.0-2.7)	<1.0	3.2 (2.3-5.6)	97.7 (73.4-105)	5.0 (4.4-8.4)	76.6 (59-89)
19	<1.0	<1.0 (1.5)	<1.0	3.4 (2.6-6.0)	20.1 (14.1-26)	<1.0 (2.5)	22.3 (13-28)
20	<1.0	1.3 (<1.0-1.8)	<1.0	<1.0 (8.9)	36.6 (20.1-52)	3.1 (1.6-5.5)	35.9 (32-58)
21	<3.3	<3.0	<2.0	<1.9	*na*	<1.1	6.4 (4.2-6.8)
22	<3.3	<3.0	<2.0	<1.9	14.2 (5.3-28)	<1.1	7.2 (2.7-17)
23	<3.3 (3.9)	<3.0	<2.0	<1.9	*na*	<1.1 (1.18)	12.9 (4.0-15.5)
24	<3.3	<3.0	<2.0	<1.9	12.3 (<1.0-40)	<1.1	<2.0
25	<3.3	<3.0 (7.2)	<2.0	<1.9	12.2 (4.2-19)	<1.1	18.6 (5.3-19)
26	<3.3	<3.0	<2.0	<1.9	*na*	(1.49-3.83)	(5.9-6.3)
27	<3.3	<3.0	<2.0	<1.9	45.5 (35-69)	<1.1	19.9 (17-58)
28	<1.0	<1.0 (2.4)	<1.0 (1.2)	<1.0 (1.0)	3.5 (2.7-8.6)	<1.0 (1.6)	1.8 (1.3-5.1)
29	<1.0	1.1 (<1.0-1.2)	<1.0	2.1 (1.2-24)	10.2 (2.5-10)	<1.0	2.9 (<1.0-13)
30	<1.0	2.3 (2.0-2.6)	<1.0	<1.0	74.0 (69-87)	9.7 (6.4-23.5)	46.4 (43-58)

[a] In cases where the median is also the minimum, the max value only is presented inside parentheses; [b] <*x* indicates value was below the method detection limit (MDL), with the MDL equalling x; *na* not applicable as concentration was not determined.