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The occurrence of persistent chlorinated and brominated organic contaminants in the European eel (*Anguilla anguilla*) in Irish waters

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ABSTRACT

The European eel (*Anguilla anguilla*) is a relatively high lipid, long lived species capable of living in a variety of brackish, fresh and marine habitats. As such, eels can accumulate organic pollutants and have been incorporated into environmental monitoring programs as a suitable "bioindicator" species for the determination of the levels of organic contaminants within different water bodies. The global eel stock is now in decline and while the cause of the collapse remains unidentified, it is likely to include a combination of anthropogenic mortality in addition to environmental degradation. This study provides valuable data on a range of contaminants (PCDD/Fs, PCBs, OCPs, PBDEs, HBCD, TBBPA and PBBs) and extractable lipid levels in eel muscle tissue collected from five Irish catchments. Extractable lipid levels were lower in the yellow eels compared to those in the silver eels. These levels were similar to those reported elsewhere and it has been posited that a decline in the lipid content in yellow eels may have consequences for the future viability of the stock. With the exception of higher substituted dioxins (especially OCDD), in three samples collected from one catchment (Burrishoole) in the West of Ireland, POP levels in general were determined to be low in eels from Irish waters compared to those in other countries.

1. Introduction

Throughout Europe the numbers of glass eels (*Anguilla anguilla*) returning from sea has declined since the early 1980s and the stocks have declined in most of the distributional area and are considered below safe biological limits (Bertin, 1956; Feunteun, 2002; Dekker, 2003; Tesch, 2003). The International Council for the Exploration of the Sea (ICES, 2006) suggested that spawner quality issues (i.e. parasites, disease, contaminants) are highly likely to impact on migration and spawning success, however a lack of reliable data over the wide geographical range of the eel makes it difficult to assess the impact of these issues on the global spawning stock of European Eel. While the cause of the eel stock collapse remains unidentified, it is likely to include a combination of anthropogenic mortality (e.g. fishing and turbines) in addition to environmental degradation. It is additionally suggested that accumulation of contaminants and/or a reduction in energy reserves (lipid levels) may be impairing the quality of potential spawners (Belpaire et al., 2009).

It is well documented that the eel progresses through five principal life cycle stages namely, the leptocephalus, glass eel, elver, yellow and silver eel stages. The leptocephali metamorphose into glass eels and a proportion migrate upstream as elvers. At the latter stages elvers develop into the yellow eel stage, which continue to feed and grow in a wide range of habitats in marine and freshwater, before completing their life cycle and metamorphosing to the silver eel stage for migration to the spawning grounds of the Sargasso Sea (Bertin, 1956; Tesch, 2003). This ability for eels to inhabit such a diverse range of marine and freshwater environments, along with a wide variety of dietary influences, may subject eels to multiple routes (sources) of exposure to environmental contaminants Eels spend a large proportion of their lives in estuarine and/or freshwater systems which when combined with their long lifecycle and diverse feeding patterns can result in the eel accumulating substantial body burdens of environmental pollutants (de Boer et al., 1994; Versonnen et al., 2004).

Dioxins (PCDDs), furans (PCDFs) and polychlorinated biphenyls (PCBs) are reported to be toxic and bioaccumulative, and thus potentially pose a major health risk to the consumer of seafood and possibly to the eels themselves (Ryan et al., 1990; Giesy and

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Kannan, 1998; van den Berg et al., 2006). The most toxic dioxin congener is 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD). Particular concern is expressed about the 12 so-called *non-ortho* (PCBs 77, 81, 126, 169) and *mono-ortho* (PCBs 105, 114, 118, 123, 156, 157, 167, 189) dioxin-like (DL) PCBs, for which essentially the toxic effects are based on the same principle as that of PCDDs and PCDFs, since they also bind to the aryl hydroxyl (Ah) receptor. Other PCBs are known as non-dioxin-like (NDL) PCBs and they do not exert their toxicological effects via binding to the Ah receptor but nonetheless are associated with a wide spectrum of toxic responses (Giesy and Kannan, 1998; Legare et al., 2000).

Brominated flame retardants (BFRs) comprise a group of chemicals, which are added to many household products for the purpose of fire prevention. Limited toxicity (and often environmental prevalence) data are available for polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD), polybrominated biphenyl (PBB) and tetrabromobisphenol-A (TBBPA). The potential of these chemicals to cause toxic effects though has been documented (Meironyte et al., 1999; de Boer et al., 2000; Spiegelstein, 2000; Kitamura et al., 2002; Alaee et al., 2003; Vos et al., 2003; Birnbaum and Staskal, 2004; European Commission, 2006). Based on their lipophilic nature, environmental persistence and bioaccumulation potential, chlorinated pesticides (OCPs) represent a threat to aquatic organisms a number having been linked to many health problems in marine mammals (Reijnders, 1986, 1994; de Swart et al., 1994).

1.1. Summary contaminant related effects and legislation

There are currently no EU maximum limits for BFRs in food and tolerable daily intakes (TDIs) have not been derived, primarily due to limited toxicological data and the associated uncertainties with such studies. EFSA currently recommends the monitoring of BDE 28, 47, 99, 100, 153, 154, 183 and 209, in addition to PBB153 and HBCD in foods and feeds (European Food Safety Authority, 2005). With the exception of PBDE209 all of these compounds were analysed in this study.

No legislation exists restricting the production of TBBPA or its derivatives and it is placed on the fourth list of priority chemicals under European Council (EC) Regulation No. 793 regarding the evaluation and control of the risks of existing substances (European Commission, 2000). Due to its high production volumes TBBPA will however be one of the first substances to go through the EU REACH (registration, evaluation, authorisation and restriction of chemicals) registration process (European Commission, 2006a) and as such monitoring data are of importance.

There are currently no maximum levels for NDL–PCBs set by the EC, however, a number of Member-States have set national levels for individual or sum of seven indicator/marker-PCBs. A stringent level has been set in Belgium for the sum of seven indicator PCBs with a maximum level of 75 $\mu g\,kg^{-1}$ product for "Fish, including shellfish, crustaceans and foodstuffs derived thereof" (European Commission, 2005). The European Commission is currently considering the regulation of NDL–PCBs, possibly via setting maximum levels for six indicator PCBs (28, 52, 101, 138, 153 and 180).

EFSA report that no health based guidance value for humans can be established for NDL–PCBs because simultaneous exposure to NDL–PCB and DL-compounds hampers the interpretation of the results of the toxicological and epidemiological studies, and the database on effects of individual NDL–PCB congeners is rather limited (European Food Safety Association, 2003). There are, however, indications that subtle developmental effects, being caused by NDL–PCB, DL-PCB, or PCDD/Fs alone, or in combination, may occur at maternal body burdens that are only slightly higher than those expected from the average daily intake in European countries (EFSA, 2005). Because some individuals and some European (sub)-populations may be exposed to considerably higher average intakes, EFSA report that a continued effort to lower the levels of NDL–PCB in food is warranted.

As part of its exposure reduction strategy the EC has also introduced maximum levels for PCDDs, PCDFs and DL-PCBs in foodstuffs, via Council Regulation (EC) No. 1881/2006; this sets maximum levels for certain contaminants in foodstuffs (European Commission, 2006b). The maximum level established for the sum of PCDD/Fs in eel muscle is currently 4 $pg\,g^{-1}$ WHO-PCDD/F-TEQ (toxic equivalents) whole weight and 12 $pg\,g^{-1}$ WHO-PCDD/F-PCB-TEQ whole weight for the sum of PCDD/F and DL-PCBs. Toxic equivalency factors (TEFs) are additionally available for fish and wildlife (Table 2) however their application to address the significance of potential and/or existing exposure to DL-compounds can be subject to a number of limitations (Tillett, 1999). As TEFs (van den Berg et al., 2006) for PCDD/Fs and DL-PCBs have not been adopted in EC legislation to date they have not been utilised in this report.

Relatively few data documenting contaminant levels in eels from Irish waters are available, as such, levels of extractable lipid, PCDD/Fs, PCBs, OCPs, PBDEs, HBCD, TBBPA and PBBs in eel muscle tissue are reported.

2. Experimental

Between October and November 2005 eels were obtained at an initial five separate river systems throughout Ireland from commercial fishermen or from fish monitoring traps in the case of those from the Burrishoole catchment (Fig. 1). Primarily on the basis of dioxin profiling in these initial samples, a second sampling event was completed in the Burrishoole (silver eel) and L. Feeagh (from where the majority of Burrishoole silver eel originate) area in 2007 where two additional samples were collected. Sampling details are presented in Table 1 and Fig. 1.

2.1. Sampling methodology – eel biology

At each site at least 210 eels were randomly selected from each commercial catch or from the traps at Burrishoole (Poole et al., 1990). Commercial eels were captured either by coghill/fyke net (mesh size 8–10 mm bar) or with the use of eel-pots in tidal waters. 100 eels were immediately anaesthetised, with chlorobutanol, measured (±0.1 cm) and frozen (<−18 °C) for further biological examination and for subsequent removal of otoliths for ageing purposes. The remaining eels were also anaesthetised, measured for the purposes of estimating eel size distributions, then revived in freshwater before being returned alive to the water. 10 random eels, which were not subjected to anaesthetic, were individually bagged and frozen for contaminant analysis. Eels were sexed macroscopically by dissection and ageing analysis was carried out utilising otoliths prepared by burning and cracking (Moriarty, 1983; Poole and Reynolds, 1996), followed by reading under 100× magnification by two independent readers. Where discrepancies were reported, a third reading was taken and all three readings were then averaged.

2007 confirmatory sampling was completed in Burrishoole with the capture of 15 yellow eels from a fyke net survey catch of 106 eels on L. Feeagh and 12 silver eels from the traps. These samples were treated in a similar fashion to those taken in 2005.

2.2. Sampling methodology – sub-sampling

Pooled mixed sex samples of eel muscle were removed from between the pectoral fin and tail. Subcutaneous lipid was removed from skin and returned to the sample muscle tissue; 9–15 individual eels (Table 1) were then pooled, samples