



# Polybrominated diphenyl ethers (PBDEs): sources, pathways and environmental data

October 2019

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# Executive summary

Polybrominated diphenyl ethers (PBDEs) are a group of man-made organobromine compounds. They have been used as flame retardants in a wide range of products including electrical and electronic equipment, textiles and foams.

The persistent and bioaccumulative properties of PBDEs, along with their potential adverse effects on aquatic life and humans have led to commercially supplied penta-, octa- and decaBDE being classified as persistent organic pollutants under the Stockholm Convention. In addition, the specific compounds tetra-, penta-, hexa-, and heptaBDE, also components of commercial penta- and octaBDE, are designated priority hazardous substances and ubiquitous persistent, bioaccumulative and toxic compounds under the Water Framework Directive (WFD) in the related Environmental Quality Standards Directive (EQSD) ([2008/105/EC](#) amended by [2013/39/EU](#)).

This narrative considers those PBDEs listed in Annex I of the amended EQSD and includes the 6 structurally similar compounds (congeners) most commonly used in commercial penta- and octaBDE products. The term PBDEs, as used in this narrative, refers specifically to these compounds. Where relevant, decaBDE is identified separately.

There are currently national and international measures in place to prevent the use of PBDEs in products. However, there are a wide range of old consumer products used in homes and businesses which contain PBDEs. Releases occur during use and disposal of these products and they continue to enter waste water treatment works (WwTWs). Much of the PBDEs in WwTWs partition to the sewage sludge, however there are continuing widespread low level emissions of PBDEs to surface waters via WwTWs' effluent. Evidence shows that concentrations of PBDEs in effluent have reduced compared with those measured earlier this decade, with over 30% reduction in average concentrations in effluents observed within a few years for 2 of the key congeners.

PBDEs may be present in soil as a result of the spreading of sludge to land. Based on the properties of PBDEs, they are unlikely to leach from soil, but may be present on soil particles entering the water environment as a result of rainfall.

PBDEs may also be released into the water column by the re-suspension of contaminated sediment or the transformation of decaBDE, which is still in use in industrial products, to smaller congeners such as octa- and pentaBDE. This may mean that it will take a long time for the environment to respond to reductions in emissions. The marketing and use of decaBDE has been restricted under the REACH regulation since March 2019. The only remaining permitted uses are in aircraft, and spare parts for vehicles and machinery manufactured prior to the restriction.

All surface water sites assessed are above the biota environmental quality standard (EQS) set in 2013 ([2013/39/EU](#)) of 0.0085 micrograms per kilogram ( $\mu\text{g}/\text{kg}$ ) wet weight for PBDEs in fish. While our monitoring is limited, our sampling locations are geographically spread across river basin districts. The presence of PBDEs is widespread and at levels significantly above the biota EQS.

The biota EQS for PBDEs was set by the EU based on protecting humans as the most sensitive receptor. There are currently no regulatory food limits, following discussion on an opinion from the European Food Safety Authority (EFSA) published in 2011. The need for regulatory limits will be reviewed on the basis of any new EFSA opinions. The levels observed are not a risk to aquatic life or wildlife eating the fish based on corresponding standards developed for these protection goals in water and biota, respectively.

It will be some time before we have enough data to be able to say anything definitive about trends in environmental concentrations of PBDEs. Available evidence indicates that at some sampled locations in marine waters, concentrations in fish are declining.

There is little scope to increase source control measures on these substances and if the current trends in reducing emissions from waste water treatment works continue, they are estimated to be an order of magnitude lower in the next 10 years. Advanced treatment could be considered at waste water treatment plants to further reduce emissions of PBDEs, but even should this prove technically feasible this would be very costly and energy intensive.

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# 1. Introduction

Polybrominated diphenyl ethers (PBDEs) are a group of man-made organobromine compounds. They have been used as flame retardants in polyurethane foams in upholstery and in polymer resins and plastics used as components in electrical equipment.

PBDEs are persistent in the environment, with degradation half-lives estimated in the range of months to years. They also have the potential for long-range transport and have been detected in remote regions including the Arctic.

They adsorb to sediment and suspended particles and are present in sewage sludge following sewage treatment.

PBDEs bioaccumulate and bio magnify in food chains. They have been detected in top predatory birds and mammals, such as gulls and seals. They also have potential adverse effects on aquatic life and humans, including possible effects on the thyroid system, potential endocrine disrupting effects, reproductive toxicity, and neurodevelopment toxicity.

The above hazardous properties of PBDEs have led to commercially supplied penta-, octa- and decaBDE, which contain a range of PBDEs as well as the title substances, being classified as persistent organic pollutants (POPs) under the Stockholm Convention on Persistent Organic Pollutants.

The specific compounds tetra-, penta-, hexa-, and heptaBDE, also components of commercial penta- and octaBDE, are designated priority hazardous substances and ubiquitous persistent, bioaccumulative and toxic (uPBT) compounds under the Water Framework Directive (WFD) ([2000/60/EC](#)) in the related Environmental Quality Standards Directive (EQSD) ([2008/105/EC](#)) amended by the Priority Substances Directive ([2013/39/EU](#)).

This narrative considers those PBDEs listed in Annex I of the Priority Substances Directive ([2013/39/EU](#)) and includes the 6 structurally similar compounds (congeners) most commonly used in commercial penta- and octaBDE products. The term PBDEs, as used in this narrative, refers specifically to the penta- and octaBDE congeners and not to decaBDE. Where relevant, decaBDE is identified separately.

The requirements of the 2013 Priority Substances Directive ([2013/39/EU](#)) took effect in December 2015 through national legislation (UK Government, 2015). Previous statutory requirements for PBDEs under the EQSD ([2008/105/EC](#)) covered the same six congeners and classified pentaBDE as a priority hazardous substance.

Directive [2013/39/EU](#) introduced a biota standard of 0.0085 micrograms per kilogram ( $\mu\text{g}/\text{kg}$ ) wet weight for PBDEs in fish and removed the annual average environmental quality standard (EQS) for concentrations in water. The setting of a biota EQS reflects the persistent, bioaccumulative and toxic (PBT) characteristics of this group of substances. The biota EQS is driven by risks to human health based on the assessment of human health toxicity data (see Section 4).

The focus of this narrative is on contributing effects to and assessment of the condition of surface waters in relation to PBDEs. However, the contribution that groundwater makes to the surface water status failures is also considered as part of the WFD and the Groundwater Directive ([2006/118/EC](#) as amended by [2014/80/EU](#)). If groundwater contributes more than 50% of the PBDE load in any surface water status failure then the groundwater body will also go to poor chemical status. We will be assessing this further as we develop water body classifications for the draft river basin plans.

## 2. Use pattern

The family of PBDEs consists of 209 structurally similar compounds with degrees of bromination ranging between 2 to 10 bromine atoms on the phenyl rings. Three products, comprising different mixtures of congeners have been available commercially: a 'pentaBDE product', an 'octaBDE product' and a 'decaBDE product'. The penta- and octaBDE products are mainly discussed in this narrative and are collectively referred to as PBDEs.

PBDEs were used as flame retardants in rigid and flexible polyurethane foams, mostly used in upholstery and furnishing, and in a variety of polymer resins and plastics, particularly for components in electrical and electronic equipment ([Environment Agency, 2013a](#)). Typically, the flame retardants were added at concentrations between 5 and 30% by weight, that is 1kg of polymer would contain 50–300g of flame retardant ([European Chemicals Bureau, 2001](#)).

Historically there have been significant differences between EU countries on fire safety requirements for furniture and fittings, with the most stringent in the UK and Ireland ([Guillame et al., 2000](#)). This resulted in a greater demand for flame retardants in the UK from the late 1980s onwards. The UK also saw a drop in furniture and furnishing related deaths of 64% in 2002–2007 compared with 1981–1985 ([BIS, 2009](#)).

Estimates of the historic annual usage of total PBDEs are around 10,000–11,000 tonnes/year in the EU and 40,000 tonnes/year globally (European Chemicals Bureau, 2001). Production of PBDEs ceased in the UK in 1996. Imports into the UK have also reduced to zero. The UK use of PBDEs peaked in the early 1990s followed by a steady decline until 2004 when restrictions on marketing and use were imposed under Commission Regulation ([EC No 552/2009](#)) ([Oakdene Hollis, 2010](#)).

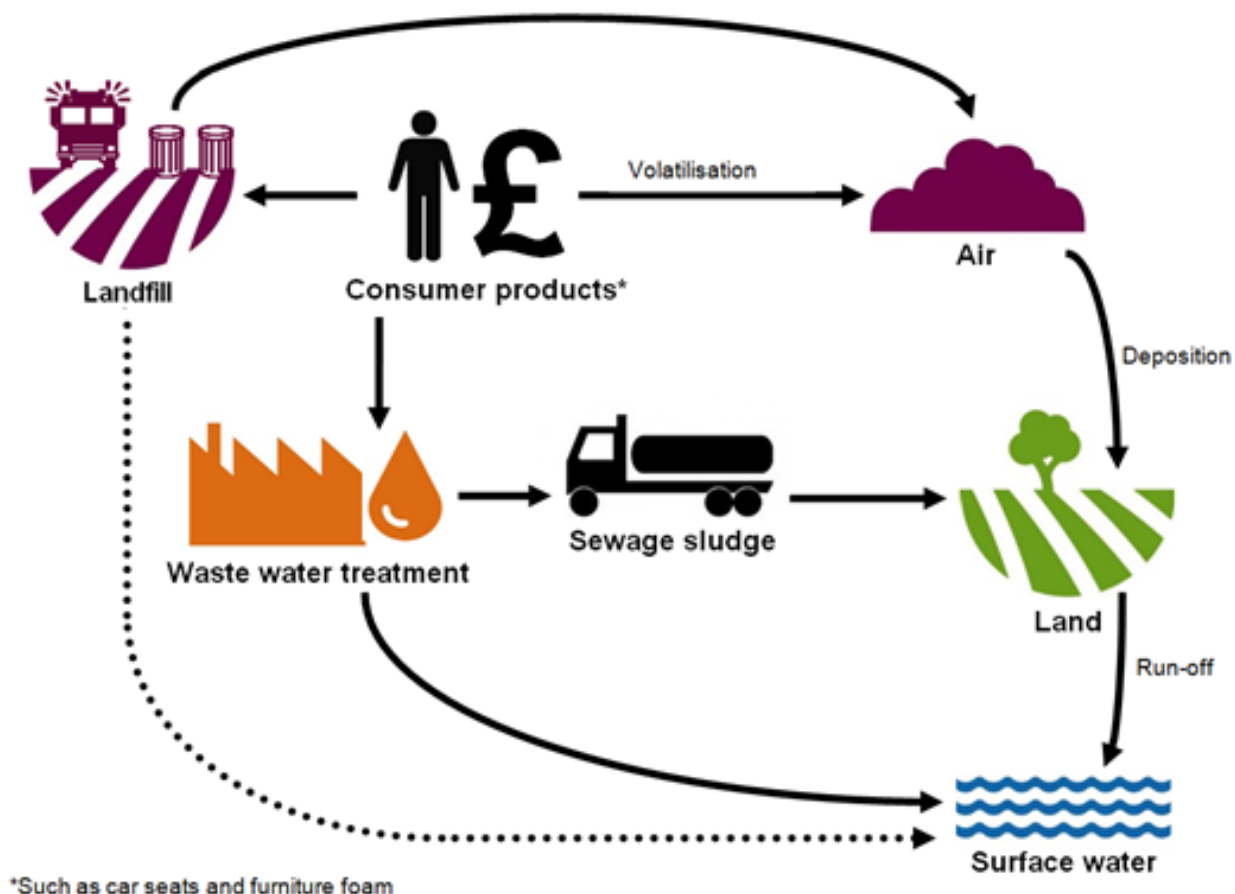
DecaBDE was used in electrical and electronic equipment and as a flame retardant in textiles and various types of polymers. Most uses of decaBDE have been restricted since March 2019 (see Section 5). The only remaining permitted uses are in the production of aircraft and spare parts for motor and agricultural vehicles or machinery manufactured prior to the restriction ([Commission Regulation \(EU\) 2017/227](#)).

PBDEs have also been detected in samples taken from stack emissions from UK iron ore sinter plants. These were already present in the raw materials, such as iron ores and coke breeze, and a significant proportion of these PBDEs were estimated to be destroyed during the sinter process ([Drage et al., 2014](#)).

### 3. Sources, emissions and pathways

No known natural sources of PBDEs have been identified, although related substances are known ([European Chemicals Bureau, 2001](#)). Figure 1 shows the main sources and pathways of PBDEs into surface water. Atmospheric releases of PBDEs are proportionally greatest, with indoor concentrations of particle-bound PBDEs being significantly larger than outdoor air concentrations. Emissions to soil and water are lower ([Earnshaw et al., 2013](#)).

Figure 1: Sources and pathways of polybrominated diphenyl ethers into the environment



The production and use of penta- and octaBDEs are banned in the EU. Releases, assuming no illegal imports, will be from historic use in items such as car seats and foam furniture for the pentaBDE product, or electrical equipment, such as televisions and computers, and other plastics for the octaBDE product.

Decreasing UK ambient air concentrations of PBDEs between 1999 and 2012 suggest that these regulatory controls have been effective in controlling emissions to the environment ([Graf et al., 2016](#)). A Swiss study on e-waste showed a significant decline in the concentrations of penta- (-93%), octa- (-77%) and decaBDE (-24%) between 2003 and 2011, also proving the restrictions effective ([BAFU, 2017](#)).

PBDEs are not chemically bound to the plastic or foam in which they are incorporated. This means that at the production stage they were physically combined with the material being treated rather than chemically combined (as in reactive flame retardants). Therefore, there is potential for them to leach out of products. The most probable route for this is transfer from furniture or gradual breakdown of foam or plastic containing the chemicals



contributing to household dust particles. Transfer of PBDEs, particularly decaBDE, to dust can also occur via direct contact between the treated products and dust ([Rauert and Harrad, 2015](#)). High levels of PBDEs have been found in the UK in dust sampled from homes, offices, classrooms and, in particular, car cabins ([Harrad et al., 2008](#); [Harrad and Abdallah, 2011](#); [Muenhor, 2011](#); [Muenhor and Harrad, 2012](#); [Kuang et al., 2016](#)). Dust particles then adhere to clothes and are released during washing, reaching waste water treatment works (WwTWs) via the sewerage system ([Prevedouros et al., 2004](#); [Earnshaw et al. 2013](#); [2015](#); [Environment Agency, 2013a](#); [Saini et al., 2016](#)).

Furniture typically has a long lifetime in use and even when disposed of may be recycled by another user, meaning that there are likely to be releases of PBDEs for years to come ([Bjorklund, 2011](#)).

Volatilisation from PBDE-containing products and partitioning to dust particles will result in emissions to air and subsequent deposition onto land, reaching surface waters during rainfall as urban run-off ([European Chemicals Bureau, 2001](#)).

Monitoring of waste water in an urban catchment identified domestic waste water as the main source of PBDEs to WwTWs ([Rule et al., 2006](#); [Comber et al., 2015](#)). Even though PBDE concentrations in effluents from WwTWs are relatively low, the large volumes of effluent treated leads to these being a significant source of PBDEs to receiving waters.

PBDEs have very low water solubility and a significant proportion of the load in a WwTW will partition to sewage sludge. Application of sludge to agricultural land may be a significant exposure route. Elevated concentrations of PBDEs are reported in soils ([Eljerrat et al., 2008](#); [Matscheko et al., 2002](#)). PBDEs are very persistent, and evidence of a cumulative effect of sludge application has been reported by [Eljerrat et al. \(2008\)](#). Based on the properties of PBDEs, they are likely to be immobile in soil ([ATSDR 2017](#); [European Chemicals Bureau, 2001](#)), but may be present on soil particles entering the water environment as a result of rainfall.

In a West Midlands study on both soil and air, concentrations of PBDEs decrease with increasing distance from the city centre, suggesting that urban conurbations are sources of PBDEs to the wider environment ([Harrad and Hunter, 2006](#); [Drage et al., 2016](#)). In a rural study of airborne particles in north-west England, highest PBDE concentrations, including decaBDE, were found when air masses passed over urban and industrial areas, although small local influences from towns may also have contributed ([Wilford et al. 2008](#)).

As older furniture and other items containing PBDEs are disposed of, PBDEs enter the waste stream. The final mode of disposal for polyurethane foam containing PBDEs is likely to be ultimately to landfill or incineration ([European Chemicals Bureau, 2001](#)). Landfills that are designed and operated with engineering requirements in accordance with the EU Landfill Directive ([Council Directive 1999/31/EC](#)) include measures to minimise the breakout of leachate, and hence minimise the potential for release of PBDEs present in the leachate to the environment. However, leachate may be discharged off-site via a treatment system with the potential for PBDEs to be discharged to the environment, if the treatment technology does not remove PBDEs.

A number of studies are underway concerning the amount of POPs, including PBDEs, in waste streams, and additionally in landfill leachate itself. Information from these studies will become available progressively over the next 5 years and form part of the UK Government's endeavours to better understand the pathways and impacts of POPs in the wider environment.

Further details of this work are periodically made available through the website of the [Stockholm Convention](#) as part of the UK POPs National Implementation Plan.

Based on the highly sorptive properties of PBDEs, the chemicals are unlikely to be very mobile. A screening study of leachate from a range of landfill sites did not find any PBDEs in either landfill leachate or groundwater samples ([Environment Agency, 2013b](#)). Particle-bound emissions of PBDEs generated during the unloading of waste and from vehicular disturbances are thought to be the dominant pathways from landfill to the atmosphere ([Earnshaw et al., 2013](#)).

The magnitude of loss of PBDEs following disposal from landfill, incineration or accidental burning is considered to be negligible compared with losses during product use ([Prevedouros et al., 2004](#)). PBDEs can form brominated dibenzofurans and brominated dibenzo-p-dioxins on combustion and the total bromine load in waste has an effect on their formation during the waste incineration process ([European Chemicals Bureau, 2001](#)).

Remobilisation of PBDEs buried at depth in sediment may also lead to re-release in the future, as will transformation in sediment of decaBDE by loss of bromine atoms (debromination) to lower brominated congeners such as octaBDE and pentaBDE ([ECHA, 2012](#)). This could be a significant source of PBDEs to surface waters, given the large volumes of decaBDE produced in the last few decades ([Earnshaw et al., 2013](#)).

The inventory of emissions, discharges and losses of priority and priority hazardous substances is a formal requirement of the WFD under the EQSD 2008 (2008/105/EC). The inventory for England can be found under the [Emissions Inventory and Chemical Trends information](#) for the cycle 2 River Basin Management Plans. It provides estimates of emissions, discharges and losses of PBDEs from both point and diffuse sources, by river basin district, for the baseline year of reporting (2010).

Estimates have been determined using SAGIS (Source Apportionment Geographic Information System, v2010), a GIS-based source apportionment tool and surface water model (Comber et al., 2012), combined with estimates of releases to sea from long sewer outfalls from the Pollution Inventory (PI).

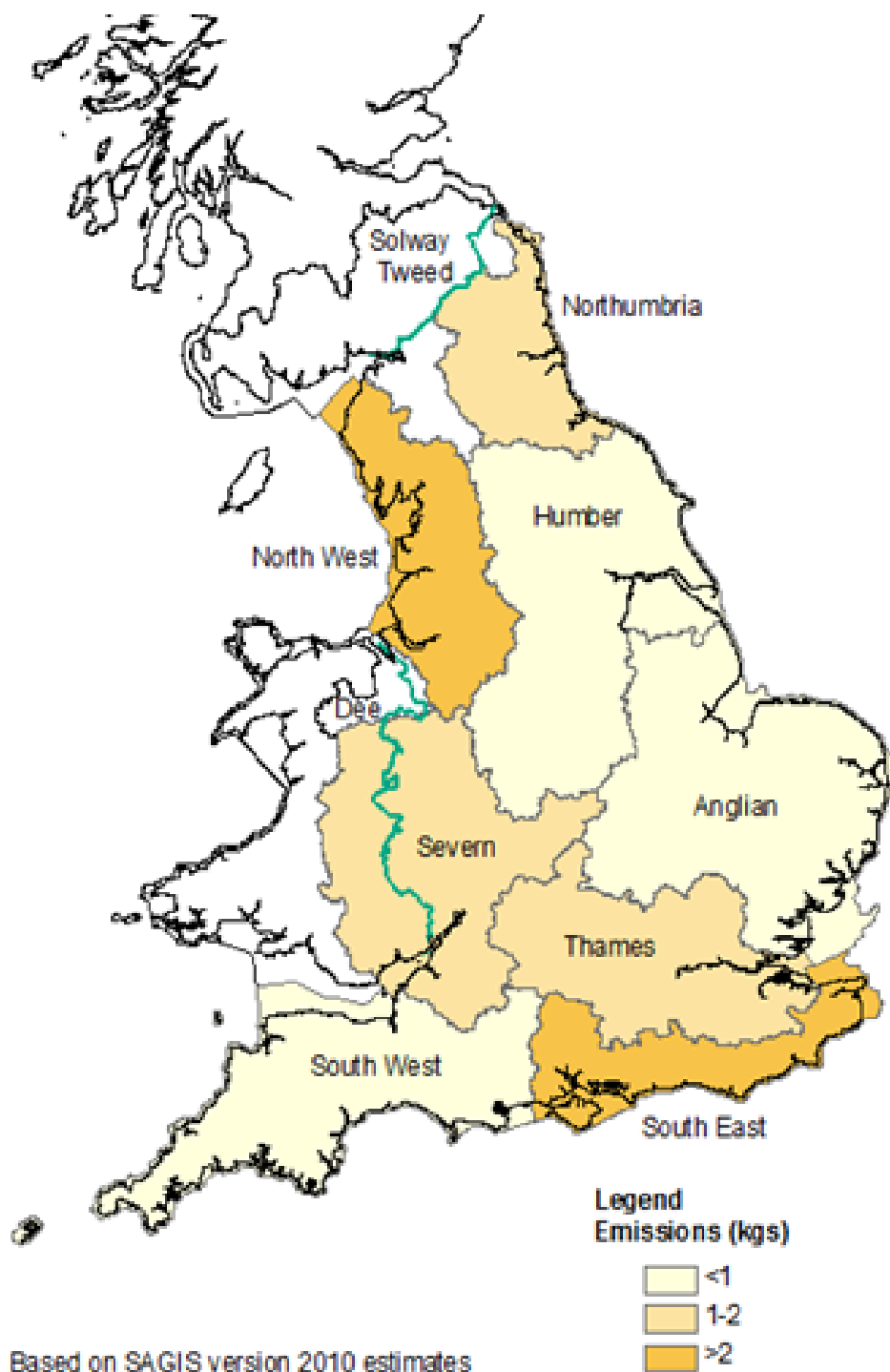
We estimate that, despite restrictions on the use of PBDEs, approximately 11kg of PBDEs were released to surface waters in England in 2010. These were estimated to be predominantly owing to point source releases. Total emissions, discharges and losses of PBDEs to water per river basin district are shown in Figure 2 (results for Solway Tweed are reported by the Scottish Environmental Protection Agency and the Dee by Natural Resources Wales).

Emissions of PBDEs are considered to have peaked around 1997 and, based on our knowledge of use, are now considered to be declining ([Prevedouros et al., 2004](#); [Earnshaw et al., 2013](#); [2015](#)).

Estimates of emissions of PBDEs to water are broadly similar across all river basin districts, with emissions ranging from 0.58kg/year in Anglian to 2.69kg/year in South East.

With only one year of data in the inventory we cannot yet describe any temporal trends in emissions. The next update of the inventory will be published as part of the 3rd cycle river basin management plans in 2021.

Figure 2. Baseline year (2010) total emissions, discharges and losses to water for polybrominated diphenyl ethers (PBDEs) (as listed in Annex I of the Priority Substances Directive [2013/39/EU](#)) for each river basin district



We have reviewed PI data up until the most recent year reported (2017). Highest levels of emissions in 2017 were reported for the Thames river basin district, followed by the Humber and North West.

Reported emissions of PBDEs to water dropped from 51kg in 2012 to 3kg in 2013. Emissions have remained at similar level since suggesting controls have influenced inputs into the water environment from large regulated industrial sites to some extent.

Given the data are generated in a different manner to the baseline inventory and focus solely on point sources, it is not possible to draw any further conclusions.

SAGIS (v2010) estimates of emissions of PBDEs by industry sector show that WwTWs are the main source of PBDEs to surface waters. There are also minor emissions of PBDEs from urban runoff and other water company discharges such as those from combined sewer overflows and storm tanks.

Monitoring data collected from 2010 to 2013 through the Chemicals Investigation Programme (CIP) undertaken by water companies (UKWIR, 2014a; 2014b) has been used as input data in the SAGIS tool for estimating chemical loads per river basin district. The CIP data suggest that sewage treatment can remove over 85% of PBDEs entering the works.

Monitoring data collected as part of the second phase of the Chemicals Investigation Programme (CIP2) ([UKWIR, 2019](#)) suggest that restrictions on use are having an impact on emissions, with declines observed in concentrations of PBDEs in effluents. At 29 sites common to both CIP, which finished in 2013, and CIP2 monitoring conducted from 2015 to 2017, the average concentrations of two of the main congeners of PBDEs – BDE-47 and BDE-99 – have decreased in effluent by over 30%. More-recent CIP2 monitoring at common sites to CIP suggests further significant declines, although the sites studied differ from those above.

Analysis of measured concentrations of BDE-47 in WwTWs' effluent ([UKWIR, 2019](#)) suggests that levels in effluent will have declined by roughly an order of magnitude by 2027, posing lower environmental risk. This assumes a continued decline owing to control measures in place.

## 4. Environmental monitoring data

We have recent monitoring data for PBDEs in surface waters and biota – fish and blue mussels – from both English freshwaters and estuarine and coastal waters. In addition, CIP2 measures concentrations of PBDEs in effluent and in water around WwTWs across England ([UKWIR, 2019](#)); data collection and analysis are ongoing and the current results are summarised in Section 4.2. Marine monitoring programmes under the Oslo Paris Convention (OSPAR) have also included the analysis of levels of PBDEs in biota and sediment in UK coastal waters over time (Section 4.4). We use saline waters in this narrative to cover estuaries and coastal waters out to 12 nautical miles (territorial waters).

We can compare our data against EQSs to get an understanding of risk. The 2013 Priority Substances Directive ([2013/39/EU](#)) lists the PBDEs given in Section 1 of this narrative as WFD priority hazardous substances. It specifies a biota-based EQS of 0.0085µg/kg for the sum of the concentrations of 6 PBDE congeners – BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, and BDE-154. This EQS refers to the presence of this group of substances in fish and is derived from human health toxicity data. It represents a level that is considered to be protective against risks to the environment and human health from the consumption of contaminated fish.

The data used to derive this value are summarised in the EQS dossier ([European Commission, 2012](#)).

The Directive removed previous annual average EQSs in surface waters for PBDEs.

### 4.1. Biota monitoring data

As part of the programme of work to assess risks from PBT substances in England, we have gathered data on PBDE residues in freshwater and saline biota from a limited number of locations around England.

Our sampling began in 2015. Between 2015 and 2018, we have sampled at 36 freshwater sites for fish (brown trout, chubb or roach) and at 23 saline sites for blue mussels 'Mytilus edulis'. The number of sites and their locations have varied year on year, although a subset of sites have been consistently sampled and can be used to observe trends in concentrations over time (see Section 4.4).

In 2018, we extended our freshwater sampling programme to generate biota data from more widespread locations and by the end of this year should have data from about 40% of WFD management catchments.

Similarly our saline monitoring was extended to include fish (flounder and dab) from 15 sites in 2018. Originally sampling focused on blue mussels for trend analysis and water body risk assessment. However, questions regarding the trophic level correction factors necessary to compare mussel tissue (trophic level 2) with EQSs set for fish (trophic level 4) has resulted in the inclusion of whole fish sampling in recent years to ensure equivalency. We are currently considering our future approach to biological sampling based on our experiences to date.

A summary of the concentrations of PBDEs we have measured in freshwater and saline fish, as well as in blue mussels is presented below. We have looked at mean measured concentrations based on the most recent 3 years of data where available. The results for fish are shown in Figure 3.

Because PBDEs are hydrophobic and tend to partition to lipids, fish with different lipid content may have different concentrations of the substances. Normalisation to a default lipid content reduces the variability in the data and allows comparison between different

fish. Therefore, when we consider fish results for PBDEs, the concentrations we use are lipid normalised using a default of 5% lipid content ([European Commission, 2014](#)).

In freshwater fish, mean concentrations of PBDEs range from 1.4 to 36.1 µg/kg wet weight (lipid normalised). All sites (100%) exceed the EQS and 42% of sites are over 1000 times greater than this value. The highest mean concentrations of PBDEs are recorded for the River Seven and River Trent in the Midlands at values over 4000 times the EQS.

Fish from all saline waters sampled have mean lipid-normalised concentrations above the EQS threshold. Mean concentrations of PBDEs in saline fish range from 0.59 to 11.3 µg/kg wet weight (lipid normalised).

Some of the site results for saline waters are based on only one replicate. However, the results for those sites have a broadly similar range to those generated at sites with 3 or more replicates (0.59 to 8.1 µg/kg wet weight (lipid normalised)). Highest mean concentrations in saline waters were recorded in the Thames and Mersey estuaries.

The results in both types of water suggest the EQS is exceeded in widespread locations across the country. While the measured values are relatively high, they do not appear to be confined to extremely populated areas. However, there are relatively few data in heavily populated areas for comparison.

Mean measured concentrations in blue mussels in saline waters range from 0.0000006 – the reported value when the individual PBDE congeners are below the limit of quantification (LOQ) – to 1.3 µg/kg wet weight. Similar to freshwaters, most sites (96%) exceeded the EQS with only 1 site in Cornwall with mean concentrations below the EQS.

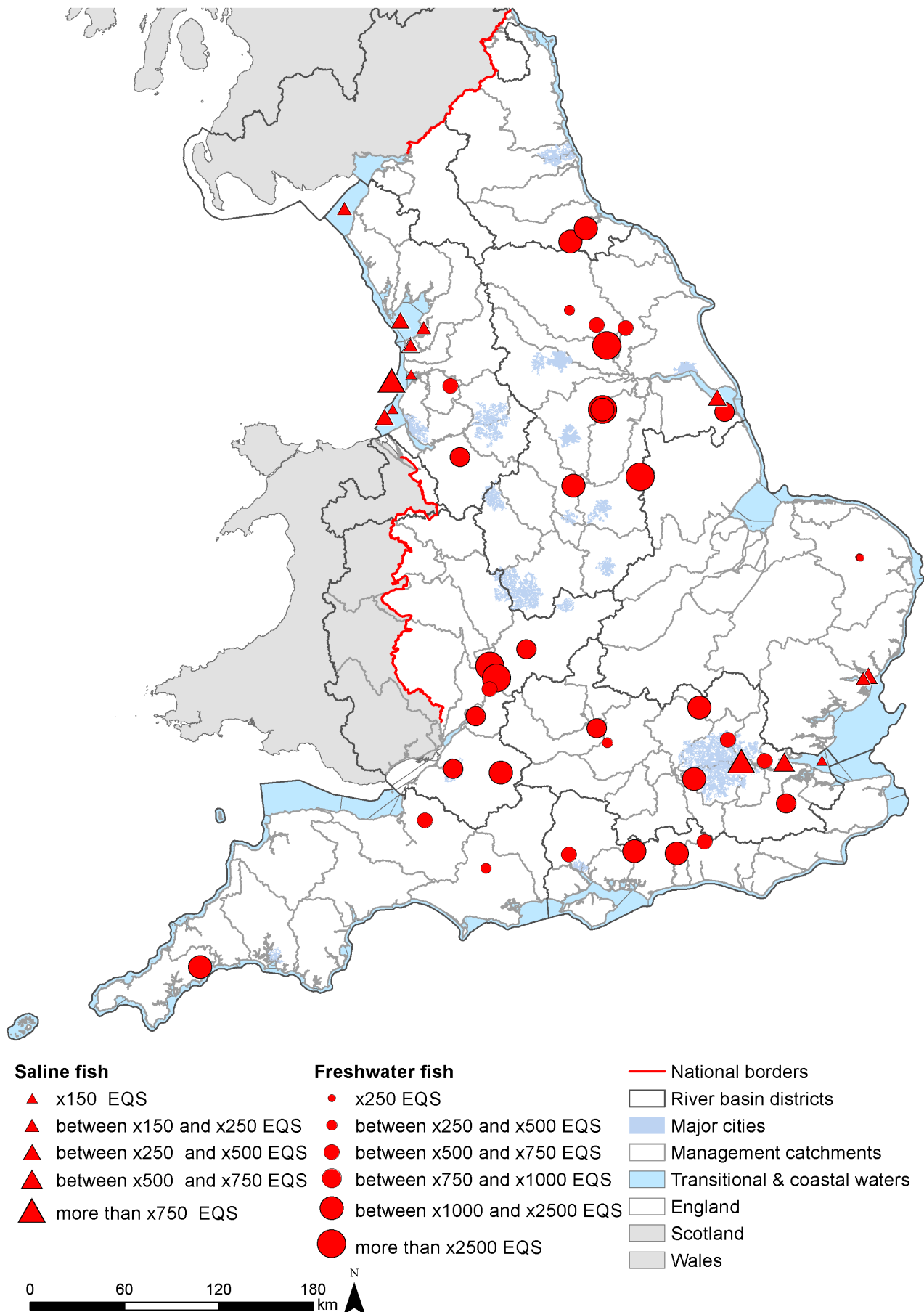
While levels in blue mussels are lower than those in saline fish, it should be noted these values have not been normalised for lipid content and represent a lower trophic level. In addition, the recorded measured concentrations are still high. The highest measured concentration is at a site near Southampton.

There is no obvious association between the estimated levels of current emissions, discharges and losses to water for each river basin district (see Figure 2) and the location of high levels of PBDEs from our biota monitoring data (Figure 3). This suggests atmospheric contributions also play a significant role in environmental concentrations in the water column.

The EQS dossier for PBDEs ([European Commission, 2012](#)) calculated a biota standard of 44 µg/kg wet weight for the protection of predators from secondary poisoning. This was not proposed as a statutory value as the protection goal of human health was the driver for the final EQS. However, based on a comparison against the secondary poisoning standard, our monitoring data in biota suggest there is no risk to wildlife eating fish.



**Figure 3. Site results showing three-year mean concentrations of PBDEs in freshwater and saline fish from 2015 to 2018 compared against the biota EQS**



Recent data and literature from other organisations support the widespread presence of PBDEs in freshwater and marine biota.

Studies on the levels of PBDEs in freshwater fish, including roach and trout, sampled from locations across the UK have shown widespread occurrence of PBDEs, frequently at levels above the biota EQS ([Jurgens et al., 2013](#); [Rose and Fernandes, 2010](#)). This was also the finding from a number of studies of marine fish, such as grey mullet and mackerel, sampled from waters around the UK ([The Food and Environment Research Agency, 2015](#); [Aznar-Aleman et al., 2017](#)).

Studying 3 rivers in South Wales, [Windsor et al. \(2019\)](#) found that contaminants such as PBDEs were significantly higher in invertebrates in areas with greater urban land cover and wastewater discharge. The sites with high contamination also showed macroinvertebrate communities with reduced taxonomic and functional diversity, simplified food webs and less abundant prey for apex predators. The data support the idea that contaminant mixtures containing substances such as PBDEs, may inhibit the complete recovery of macroinvertebrate food webs from the past effects of gross pollution.

## 4.2. Water column monitoring

The water company CIP2 is ongoing and includes contaminants in rivers upstream and downstream of over 600 WwTWs, as well as in sewage effluent from those works. The sites were selected because the receiving waters for their effluent offer low dilution and, therefore, they represent sites at greatest risk of contamination.

The findings from the CIP2 to date covers results for sites monitored for the 6 PBDE congeners of interest between 2015 and 2019 ([UKWIR, 2019](#)). Mean results for samples of individual congeners taken at up to 448 sites upstream and downstream of WwTWs' discharge locations were 0.22 to 0.27ng/l and 0.23 to 0.27ng/l, respectively. The similarity in these values is in part a reflection of the fact that many samples were below the chosen limit of detection for the investigations.

We took samples at our routine water monitoring sites between 2016 and 2018; 272 freshwater and 51 saline sites were assessed. The majority of sites were monitored for two years with 41 sites having only 1 year's data and 55 sites having 3 years' worth. Between 3 and 37 samples were taken at the sites. The mean concentrations of PBDEs in freshwaters ranged from 0.0006 to 1.6ng/l. For saline waters, the concentrations of PBDEs ranged from 0.0006 to 0.77ng/l.

Assessment of risk using water column monitoring data is difficult because there is no statutory value to compare against. The EQS dossier for PBDEs (European Commission, 2012) calculated water concentrations equivalent to the biota EQS of  $4.9 \times 10^{-8}$  and  $2.4 \times 10^{-9}$ µg/l for fresh and saline waters, respectively, but recommended caution over their use. These values, therefore, were not included in the Priority Substances Directive ([2013/39/EU](#)).

The dossier ([European Commission, 2012](#)) also calculated standards to reflect long-term risks to aquatic life owing to direct toxic effects. These values are far less stringent than the values that were considered equivalent to the biota EQS at 0.049 and 0.0049µg/l for fresh and saline waters, respectively. These are not statutory values as the protection goal of human health was the driver for the final EQS.

None of our monitoring data or that undertaken by water companies upstream and downstream of their WwTWs exceeds these values suggesting that the levels in water are below those likely to result in direct toxic effects to aquatic organisms.



The Priority Substances Directive ([2013/39/EU](#)) also sets out maximum allowable concentration EQSs (MAC EQSs) for PBDEs of 0.14 and 0.014µg/l for inland surface waters and other surface waters, respectively. The MAC EQS is a threshold intended to protect the aquatic environment from short-term toxic effects. The MAC EQSs should be applied where the biota EQS is applied or where emissions or environmental concentrations indicate potential short-term risk.

All our measured concentrations in fresh and saline waters are below the MAC EQSs.

[Cristale et al. \(2013\)](#) analysed water samples from sites along the River Aire for PBDEs. Whilst the main congener of decaBDE, BDE-209, was detected in the majority of water samples, none of the congeners from either penta- or octaBDE was detected. This is broadly consistent with other studies, which also report a dominance of decaBDE and lower concentrations of both penta- and octaBDE in water, sediment and biota samples at most locations ([Law et al., 2014](#), [OSPAR, 2010](#); [Law, 2014](#)).

### 4.3. Trends in freshwaters

As described in Section 4.1, we have been measuring PBDE residues in biota since 2015. Sampling locations have changed over time and we only have one freshwater site where residues of PBDEs in fish have been sampled for consecutive years from 2015 to 2018. We have visually examined the data for trends rather than applying any statistical analysis to the data because we do not yet have sufficient data. Overall, there is no consistent temporal trend observed at the site. We will continue to collect data on trends to enable future analysis.

There are only limited environmental monitoring data reported for PBDEs in the wider scientific literature. There is a general consensus that although elevated levels of penta- and octaBDE in biota and sediments are still present and there is substantial spatial variation, concentrations have probably peaked and, at some UK sites are now declining ([Law et al., 2014](#), [OSPAR, 2010](#); [Law, 2014](#); [Yang et al., 2016](#)). A downward trend is not however observed at all sites, and in some locations sediment concentrations of PBDEs, including BDE-209, are still increasing (Yang et al., 2016).

A study by Walker et al. (2012) measuring levels of PBDEs in the livers of the Eurasian otter 'Lutra lutra' reported PBDE concentrations of 3 to 718ng/g wet weight. These levels were within the range previously reported between 1995 to 2005; implying little decline in concentrations since restrictions were introduced.

Birds have been widely used as monitors or sentinels of environmental contaminants including PBDEs. Trends in residues of PBDEs in archive sparrowhawk 'Accipiter nisus' eggs collected between 1985 and 2007 in the UK as part of the Predatory Bird Monitoring Scheme show little decline in concentrations following a peak in the 1990s. [Crosse et al. \(2012a\)](#) suggest that these data may indicate some formation of pentaBDE congeners as a result of debromination of decaBDE.

A further study of PBDE concentrations in sparrowhawk livers has revealed sparrowhawks in Britain accumulate some of the highest PBDE residues reported for terrestrial feeding birds ([Crosse et al., 2013](#)).

In a study on decaBDE in wild birds from the UK, Sweden and the Netherlands, there was no significant change in sparrowhawk concentrations between 1973 and 2001, although in later years concentrations were found more frequently above the limit of detection. In peregrine falcons, peak concentrations were measured in 1995, followed by a decline until 2001 ([Leslie et al., 2011](#)).

## 4.4. Trends in transitional and coastal waters

Under OSPAR, the Clean Seas Environment Monitoring Programme (CSEMP) has collected data on concentrations of PBDEs in UK marine waters over several years. Data generated under this programme is collated for the International Council for the Exploration of the Sea (ICES) and can be viewed using the Database on Oceanography and Marine Ecosystems ([DOME](#)) web portal. Data are available for the 6 congeners of interest.

An overview of the trend monitoring data collected through the CSEMP for BDE-47 within English territorial waters (12 nautical miles), the saline area to which the Priority Substance Directive (2013/39/EU) applies, is given in Figure 4.

Trend information has been reported for residues of BDE-47 in the livers of common dab 'Limanda limanda' sampled at 8 sites around England between 2003 and 2017. At 7 of these sites, downward trends were observed. Only one site in Northumberland showed no trend.

For the other congeners, the majority of sites that can be assessed for trends also show declines in PBDE residues over time.

Law et al. (2014) compiled and reviewed recent reports of PBDEs in the marine environment between 2010 and 2012. They concluded that despite some evidence of declining trends in some fish species and locations, significant concentrations of PBDEs were still being reported.

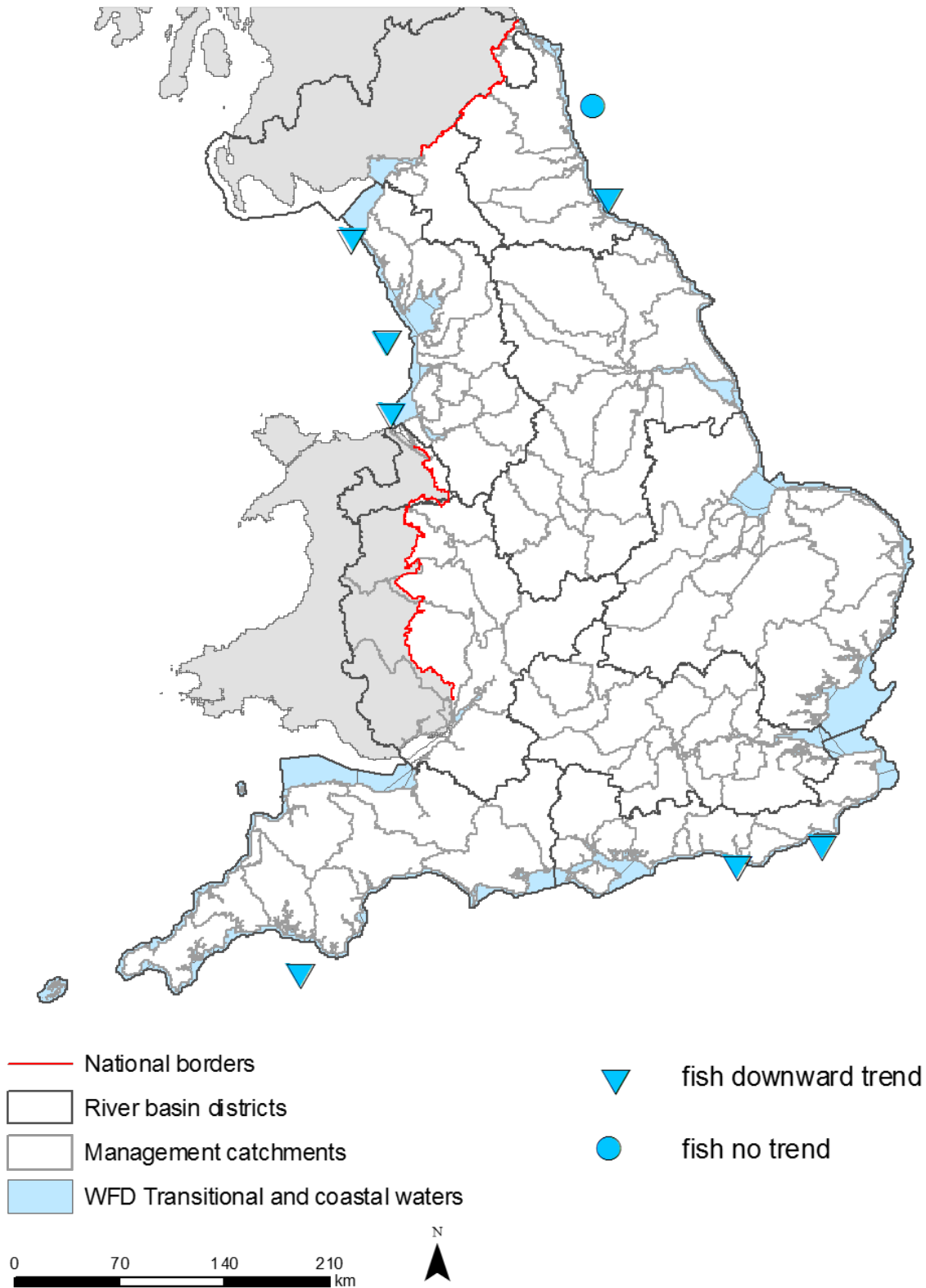
In contrast to the high residues of PBDEs reported in terrestrial feeding birds ([Crosse et al., 2012a](#); [2013](#)), PBDE concentrations in the eggs of the Northern gannet (*Morus bassanus*) from two UK colonies showed a rise in the late 1980s, peaking in 1994, and a steady decline until 2002, where levels fell back to pre-1980 levels ([Crosse et al., 2012b](#)).

A study of PBDEs in ringed seals from the Baltic Sea between 1974 and 2015 also reported an increasing trend until the end of the 1990s, followed by a decreasing trend until the end of the period ([Bjurlid et al., 2018](#)).

Since 2009, [CEFAS \(2012\)](#) have monitored PBDEs in coastal sediments around the UK. Whilst there are insufficient data to report a temporal trend yet, results show that tetraBDE and pentaBDE were present. Samples were dominated by decaBDE. Similar results have been reported by [Vane et al. \(2010\)](#).

A study by [Sühring et al. \(2015\)](#) investigated brominated flame retardants, including PBDEs, in sediments around the UK east coast. High concentrations of BDE-209 were detected in North Sea sediments, which were expected owing to high use in Europe. Samples with the highest overall PBDE concentrations were found near the UK coast with BDE-100 being the main congener followed by BDE-154, BDE-47 and BDE-99, components of technical penta-BDE.

Figure 4. Trends in concentrations of BDE-47, as proxy for PBDEs, in fish liver (dab) in English coastal waters over the period from 2003 to 2017. Data accessed from the ICES DOME portal, extraction 15 February, 2019; downloaded May 2019. Chemical data for the OSPAR Coordinated Environmental Monitoring Programme, reporting laboratories via the British Oceanographic Data Centre (UK). ICES, Copenhagen



## 5. Control measures

### 5.1. Restrictions

Commercial penta- and octaBDE were originally restricted in 2003 in Europe under the Marketing and Use Directive (2003/11/EC) and then in 2006 by the REACH Regulation (EC 1907/2006). These restrict the placing on the market and use of penta- and octaBDE above 0.1% by weight.

In 2009, penta- and octaBDE were designated as Annex A POPs at the 4th Conference of the Parties to the Stockholm Convention. Parties to the Stockholm Convention must take measures to eliminate production and use of these substances.

This international agreement was implemented into EU legislation through a Council Decision in 2006 (2006/507/EC) as an amendment to Regulation (EC) No 850/2004 on POPs which prohibits the production and placing on the market of the substances listed in Annex I of the legislation and regulates the management of waste containing these substances.

DecaBDE was designated as an Annex A POP at the 8th meeting of the parties to the Stockholm Convention in spring 2017.

DecaBDE, along with other PBDEs, has been banned for use in electrical and electronic equipment since 2008 under the Directive on the Restriction of the Use of Certain Hazardous Substances in Electrical and Electronic Equipment ([2011/65/EU](#)). Most other uses of decaBDE (at or above 0.1%) have now been restricted under the REACH regulation since March 2019 ([Commission Regulation \(EU\) 2017/227](#)). The only remaining permitted uses are in aircraft, and spare parts for vehicles and machinery manufactured prior to the restriction.

The EU Regulation on POPs was recast in June 2019 ([Regulation \(EU\) 2019/1021](#)) to include decaBDE and specifies a concentration limit for PBDEs, including decaBDE, in waste of 1000mg/kg. This limit is to be reviewed by the European Commission by July 2021 with a view to lowering it to 500mg/kg.

Land spreading of sewage sludge is regulated by the Sludge (Use in Agriculture) (Amendment) Regulations 1990 ([SI 1990/880](#)) which set limits for the amounts of certain trace metals. There are currently no limits set on the levels of PBDEs in sewage sludge spread to agricultural land.

We have continued to develop and improve the way we assess the risks from waste applications to agricultural land under the Environmental Permitting Regulations ([SI 2016/1154](#)).

The biota EQS for PBDEs was set by the EC based on protecting humans as the most sensitive receptor. There are currently no regulatory food limits, following discussion on an opinion from the European Food Safety Authority (EFSA) published in 2011 ([EFSA, 2011](#)). The need for regulatory limits will be reviewed on the basis of any new EFSA opinions.

## 6. Discussion

PBDEs are stable and very persistent compounds which have potential adverse effects on aquatic life and humans.

Data, such as that generated through the CIP and CIP2 and in the Pollution Inventory, show declines in emissions to water suggesting that restrictions on use have influenced inputs to some extent.

The evidence we have from monitoring data in biota suggests that the presence of PBDEs is widespread across England. The biota monitoring reports concentrations of PBDEs several orders of magnitude greater than the biota EQS. As such, widespread failure of this EQS in the future seems likely.

It will be some time before we have enough data to be able to say anything definitive about trends in environmental concentrations of PBDEs. The available evidence indicates that at some sampled locations, particularly in marine waters, concentrations are declining. However, this is not true for all sites. The continued release of PBDEs from historic sources and potentially from the breakdown of decaBDE, as well as their persistence and potential to bioaccumulate means that in some instances, little or no decline in environmental concentrations is observed.

PBDEs are already heavily restricted, and there is little scope to increase source control measures on these substances. We continue to monitor compliance with these restrictions. Even if complete compliance with these restrictions is achieved, it is unclear how long it will be before environmental concentrations of PBDEs are no longer problematic because of the persistence of these substances in the environment and the amounts still in use by society.

The limited evidence available suggests that environmental concentrations will take decades to decline significantly ([Earnshaw et al., 2015](#)). DecaBDE may remain a source of lower level congeners in the environment for some time. DecaBDE has been used in high volumes until recently, but rigorous enforcement of recent restrictions could help bring down emissions and reduce the potential for them to form PBDEs as they de-brominate.

The stability of these substances also means that they are not amenable to destruction by traditional waste water treatment technology, making 'end of pipe' control difficult. Most of the PBDE load will be removed to sludge. If the current trends in reducing emissions from WwTWs continue, PBDE concentrations in effluents are estimated to be an order of magnitude lower in the next 10 years. Advanced treatment could be considered at WwTWs to further reduce emissions of PBDEs, but even should this prove technically feasible this would be very costly and energy intensive and it is uncertain what this would deliver in terms of improved compliance with biota standards.

Further work is needed to understand what environmental improvements could be achieved. We will continue to develop our understanding of this and will engage with stakeholders on the implications as our knowledge improves.

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## 8. List of abbreviations and acronyms

### **BDE-47 pentaBDE**

2,2',4,4'-tetrabromodiphenyl ether; a congener present in commercial

### **BDE-99 pentaBDE**

2,2',4,4',5-pentabromodiphenyl ether; a congener present in commercial

### **BDE-209**

decabromodiphenyl ether (decaBDE)

### **BDE**

bromodiphenyl ether

### **BIS**

Department for Business, Innovation and Skills

### **CEFAS**

Centre for Environment, Fisheries and Aquaculture Science

### **CIP**

Chemicals Investigation Programme

### **CSEMP**

Clean Seas Environment Monitoring Programme

### **DOME**

Database on Oceanography and Marine Ecosystems

### **ECHA**

European Chemicals Agency

### **EFSA**

European Food Safety Authority

### **EQS**

Environmental quality standard

### **EQSD**

Environmental Quality Standards Directive

### **ICES**

International Council for the Exploration of the Sea

### **LOQ**

Limit of quantification

### **MAC**

Maximum allowable concentration

### **OSPAR**

Oslo Paris Convention

### **PBDEs**

Polybrominated diphenyl ethers

### **POPs**



Persistent organic pollutants

**QA/QC**

Quality assurance/quality control

**REACH**

Registration, Evaluation, Authorisation and Restriction of Chemicals

**SAGIS**

Source Apportionment Geographic Information System

**TraC**

Transitional and coastal

**UKWIR**

UK Water Industry Research

**WFD**

Water Framework Directive

**WwTWs**

Waste water treatment works

## 9. Glossary

### **Adsorb**

A sorption process in which one substance becomes attached to another via adhesion to the surface

### **Bioaccumulate**

The accumulation of a substance, such as a toxic chemical, in various tissues of a living organism

### **Biomagnify**

When the concentration of a substance in an organism exceeds the background concentration of the substance in its diet

### **Congeners**

Chemical substances related to each other by origin, structure or function

### **Debromination**

Loss of bromine atoms from compound

### **Limit of qualification**

The lowest concentration of a substance in a sample that can be reliably quantified with an acceptable level of precision and accuracy

### **Lipid normalisation**

Normalisation of measured concentrations of PBDEs by taking into account actual lipid content in a fish and adjusting the concentrations relative to a default lipid content of 5%. PBDEs are hydrophobic and tend to partition into the lipids of organisms. This approach allows for comparability across results for different fish.

### **Organobromine**

Organic molecule containing one or more bromine atoms

### **OSPAR**

The mechanism by which 15 governments and the EU cooperate to protect the marine environment of the North-East Atlantic

### **Passive samplers**

Sampling device left in situ in the water column

### **Pollution inventory**

The inventory collates data from large regulated industrial sites on emissions of specified substances to air and controlled waters and sewers, as well as quantities of waste transferred off site.

### **Stockholm Convention**

The Stockholm Convention is a global treaty to protect human health and the environment from persistent organic pollutants (POPs) and came into force in May 2004.

### **Sorption**

A physical or chemical process in which one substance takes up or holds another

### **Wet weight**

Whole weight, fresh weight and wet weight refer to the sample as it is received whole or wet, regardless of whether it is a whole organism or parts of the organism.



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