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Citation for final published version:

O'Rourke, Emily, Schumacher, Melanie, Charlesworth, Mark, Andrade, Thomaz, Pereira, M. Gloria, Hailer, Frank and Chadwick, Elizabeth 2026. Increasing concentrations of polychlorinated biphenyls (PCBs) in Eurasian otters (*Lutra lutra*) from Wales suggest remobilisation from sediment sinks. *Environmental Pollution* 388 , 127354. 10.1016/j.envpol.2025.127354

Publishers page: <https://doi.org/10.1016/j.envpol.2025.127354>

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1 Increasing concentrations of polychlorinated biphenyls
2 (PCBs) in Eurasian otters (*Lutra lutra*) from Wales
3 suggest remobilisation from sediment sinks
4

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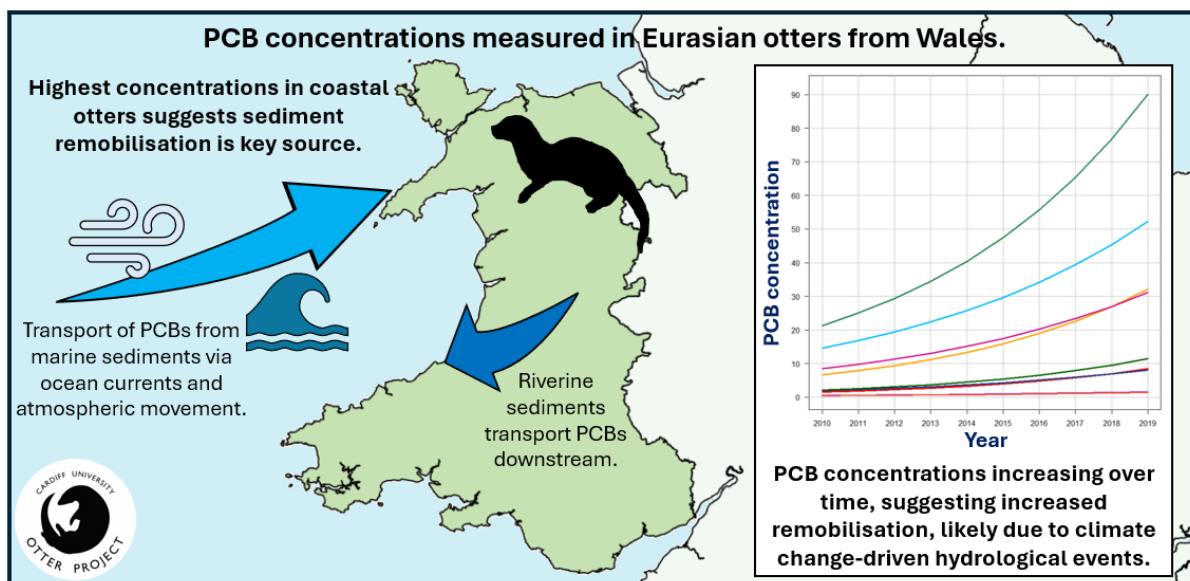
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16

Abstract

Despite ongoing regulatory efforts to mitigate PCB pollution, their presence remains pervasive in the environment, with concentrations in top predators still reaching toxicologically significant levels. To assess temporal and spatial variation of PCB concentrations in Wales between 2010 and 2019, we analysed liver samples of a sentinel predator, the Eurasian otter (*Lutra lutra*), for 23 PCB congeners. PCBs were detected in all livers analysed, with PCB 153 recording the highest individual concentration (545.8 µg/kg wet weight); sixteen percent of otters had concentrations above a toxic threshold for dioxin-like PCBs. Statistical modelling revealed a negative correlation between concentrations of dioxin-like PCBs and otter body condition. While previous studies on otters from the UK showed declining PCB concentrations between 1983 and 2009, our model predictions indicated a significant increase between 2010-2019, mirroring trends seen in marine mammals. Higher concentrations were observed in otters from coastal, low-altitude areas. A multi-model inference approach was used to identify the best groups of predictors for each congener, and suggest that remobilisation of PCBs from riverine and coastal sediment sinks is now a major driver of PCB concentrations, with the impacts of climate change likely exacerbating remobilisation. While PCB concentrations were often below limits of detection in water, and below the current Water Framework Directive Environmental Quality Standard in fish, they remained at toxicologically relevant levels in otters from Wales. These findings underscore the importance of biomonitoring across trophic levels, and suggest that current environmental quality standards for water and fish are not protective of top predators. Addressing environmental PCB contamination will require strengthened international efforts both to manage the significant sinks of legacy pollutants, as well as to meet climate change mitigation targets.

Graphical abstract



Keywords

Polychlorinated biphenyls (PCBs)

Biomonitoring

Eurasian otter

Freshwater ecosystems

Remobilisation

Sediment sinks

Secondary sources

Highlights

- PCBs were detected in all otters analysed from Wales.
- Otters from coastal, low-altitude areas had the highest PCB concentrations.
- PCBs in otters increased from 2010–2019, reversing previous PCB declines.
- We infer that remobilisation of PCBs from sediment sinks is the likely driver of increases.
- We suggest that safety standards designed to protect top predators are not effective.

Introduction

The extensive use of polychlorinated biphenyls (PCBs) in industrial and commercial applications has led to pervasive contamination of the environment (Hoffman et al. 2001; Desforges et al. 2018).

Their lipophilicity and persistence means PCBs partition into adipose tissues, and can biomagnify up trophic levels, reaching concentrations high enough to have toxic effects in both wildlife and humans (Jepson et al. 2016; Kean et al. 2021; Montano et al. 2022). PCBs have been found in abiotic and biotic samples from the Arctic to deep ocean trenches, far away from PCB production sources (Rigét et al. 2019; Vorkamp et al. 2022; Sobek et al. 2023), and have been linked to population declines in freshwater and marine top predators (Harding and Härkönen 1999; Roos et al. 2012; Jepson and Law 2016; Kean et al. 2021).

The commercial production of PCBs started in 1929 and peaked in the 1970s (Othman et al. 2022). Once evidence of toxicity became clear, most countries banned PCB production in the late 1970s or early 1980s. This curtailed the main sources of emissions, and concentrations in biota declined as a result. However, this initial decline was not sustained, and a number of recent studies have shown a plateauing, and even increases, in PCB concentrations over time (Shaw et al. 2005; Shore et al. 2005; Law et al. 2012; Jepson et al. 2016; Rigét et al. 2016; Rigét et al. 2019). Studies showing increases in PCB concentrations have been primarily conducted using marine biota, leaving uncertainty about whether similar upward trends are occurring in freshwater ecosystems.

PCBs are still entering the environment via the continued use of products (such as transformers, capacitors and some building materials) which were manufactured before PCB production was banned (Othman et al. 2022). Disposal of PCB containing products via landfill can give rise to leachate (Weber et al. 2011; Brand and Spencer 2023) while waste incineration can cause emissions (Montano et al. 2022). In accordance with the Stockholm Convention, parties have until 2025 to eliminate the use of legacy equipment containing PCBs and until 2028 to ensure environmentally sound waste management (UNEP 2016), but many countries are falling behind these targets (Melymuk et al. 2022). Additionally, the resistance of PCBs to chemical and thermal degradation means that environmental reservoirs, especially sediment sinks, have become important secondary sources of PCBs (Lohmann et al. 2007; Ma et al. 2016). In the aquatic environment, the hydrophobic nature of PCBs and their high tendency to sorb to organic carbon, causes them to partition out of water and into sediment (Jönsson et al. 2003; Wernicke et al. 2022; Sobek et al. 2023). During the period of high use, a cumulative loading of river and marine sediment sinks occurred, and now they have become important sources in the continued cycling of PCBs (Ma et al. 2016). Cycling is driven by the physicochemical properties of PCBs, such as volatility and partitioning, by environmental

factors, such as precipitation, flooding, winds, temperature, currents and wave action (Ma et al. 2016; Pizzini et al. 2022), and by human activity, such as dredging and trawling (Nicolaus et al. 2015). There is concern that the effects of climate change, such as increasing temperatures, rainfall and storm events, may be remobilising PCBs and making them more bioavailable to wildlife. In the freshwater environment, increased rainfall and flash flooding cause increased runoff and water turbulence, which can disturb sediments and expose contaminated sediment layers, thus potentially remobilising PCBs (Whitehead et al. 2009). Subsequently, PCBs can be transported downstream via suspended sediments to lowland rivers and estuaries (Dawson and Macklin 1998). In the marine environment, deep ocean sediments are regarded as a final sink for PCBs (Lohmann et al. 2007), whereas PCBs held in continental shelves can be remobilised (Jönsson et al. 2003). Storm events, especially those with strong winds and waves, can disturb coastal sediments, remobilising PCBs into both the water and atmosphere (Dachs and Méjanelle 2010), from where they can be transported vast distances to land via oceanic currents and prevailing winds (Vorkamp et al. 2022).

Here we investigate the presence of PCBs in the freshwater environment of Wales, a country of historic PCB production (Harrad et al. 1994) and high PCB usage (Melymuk et al. 2022). Due to its geographic location and prevailing weather conditions, Wales is also vulnerable to pollutant recirculation from sediments in freshwater and marine environments. Prevailing west-southwesterly winds mean that Wales is one of the first parts of Britain to receive frontal rainfall from Atlantic weather systems, and orographic rainfall due to Wales' hilly terrain is also an important feature (Mayes 2013; Met Office 2013). Consequently, Wales has much higher annual rainfall than other parts of Britain (Met Office 2013), and, due to climate change, average annual rainfall volumes and extreme storm events have increased (Blöschl et al. 2019; Cotterill et al. 2021; Madge 2021). The predominant ocean current influencing Wales is the Gulf Stream and its eastern extension, the North Atlantic Drift, which moves in a north-easterly direction across the North Atlantic Ocean from the Gulf of Mexico to north-western Europe. The basin of the North Atlantic Ocean is particularly highly contaminated with PCBs, compared to other ocean basins, due to its location adjacent to the greatest historic PCB producers and emitters in eastern USA and western Europe (Jönsson et al. 2003; Breivik et al. 2007).

Regulatory monitoring in Wales and England does not currently give rise to concern regarding PCBs; concentrations in water samples are typically at or near to the limit of detection (Lu et al. 2017), and concentrations of dioxin like PCBs in fish are below the Water Framework Directive Environmental Quality Standard (EQS_{biota}, 0.0065 µg/kg Toxic Equivalency [TEQ]) (Jürgens et al. 2015; Natural Resources Wales 2022). However, concentrations in liver tissue from the Eurasian otter *Lutra lutra* (the top predator of freshwaters in Great Britain) have previously revealed concentrations above

thresholds indicative of harm, in otters collected between 2000 and 2009 (Kean et al. 2021). As a long-lived apex predator, with a primarily piscivorous diet (Moorhouse-Gann et al. 2020) and non-migratory nature, the Eurasian otter can integrate high concentrations of bioaccumulating contaminants over its lifetime. Therefore, Eurasian otters are an excellent sentinel for bioaccumulating contaminants, and have been shown to be vulnerable to their toxic effects, with PCBs likely playing a major role in population declines between the 1950s-70s (Mason 1989; Mason and Wren 2001; Roos et al. 2001; Sainsbury et al. 2019). Analysis of otter tissues can therefore provide a valuable benchmark to evaluate whether current environmental quality standards for water and fish are protective of top predators.

The overall aim of this study was to evaluate the drivers and potential impacts of PCB pollution in Eurasian otters in Wales. To achieve this, we analysed the livers of 100 Eurasian otters that died between 2010 and 2019 for 23 PCB congeners. We hypothesised that (1) higher PCB concentrations would be associated with historic production and waste disposal sites, (2) otters from coastal locations would have higher PCB concentrations, due to exposure from freshwater and marine sediment sinks, (3) previously identified declines in concentrations (Mason 1998; Kean et al. 2021) are likely to have ceased due to increased remobilisation of PCBs as a result of climate change, and (4) some individuals would have PCB concentrations above toxic thresholds, indicative of potential harmful effects.

143

Materials and methods

Otter sample selection and associated biotic data

Since 1994, Cardiff University Otter Project has collected otters found dead (primarily as roadkill) for the purpose of ecological research. Finders record the location (as National Grid Reference) and date when each otter was found, and arrange delivery to Cardiff University. A standardised post-mortem examination is carried out, during which a range of biometric data (including sex, age-class, length, weight and reproductive status) are recorded, and tissue samples are retained (adapted from Simpson, 2001). Liver samples were stored wrapped in aluminium foil, in individual grip seal bags at minus 20°C.

One hundred liver samples were selected for analysis from the Cardiff University Otter Project archive, from otters which died between 2010 and 2019. To facilitate analysis of temporal change, 9-11 otters were selected from each of the ten years. To enable comparison between otters found in the western Atlantic fringe populations, versus those in the east, an even split was selected from eastern and western river basin districts (RBDs) (n=50 from Dee and Severn RBDs representing the east, and n=50 from Western Wales RBD representing the west), with between 4-6 individuals from each year, in both areas. Although sample availability meant a fully balanced sample design was not possible, we aimed to balance selection across six catchments, chosen to ensure a range of geographical, land use and ecological status across Wales (Carmarthen Bay and Gower, Llŷn and Eryri, and Ynys Môn in the west, and Dee, Usk, and Wye in the east); samples from adjacent catchments were included where necessary to ensure an east-west balance and even selection across sampling years (Figure 1, Table S1). Juvenile otters (males <3 kg, females <2.1 kg), otters which showed signs of decomposition, and very emaciated otters (determined at post mortem) were removed from sample selection. Too few juveniles were available to robustly test associations across the full age range, so analysis was focused on sub-adult and adult individuals which together (based on previous analysis of the age-distribution of road-killed otters in the UK (Sherrard-Smith and Chadwick 2010) are likely to result in samples from otters predominantly 1-3 years old. Setting these selection criteria allowed us to focus analysis on spatial and temporal comparisons while restricting variation in other potentially confounding variables. An even split by sex was selected so that sex could be included in statistical modelling to control for maternal transfer of PCBs, which has been evidenced in other mammals (Mauritsson et al. 2022) and is believed to occur in otters (Kean et al. 2021). We also ensured normal distributions of body length and body condition score (derived from length and weight using scaled mass index [SMI]) (Peig and Green 2009), thus providing a representative selection of individuals by size and condition (Figure S1).

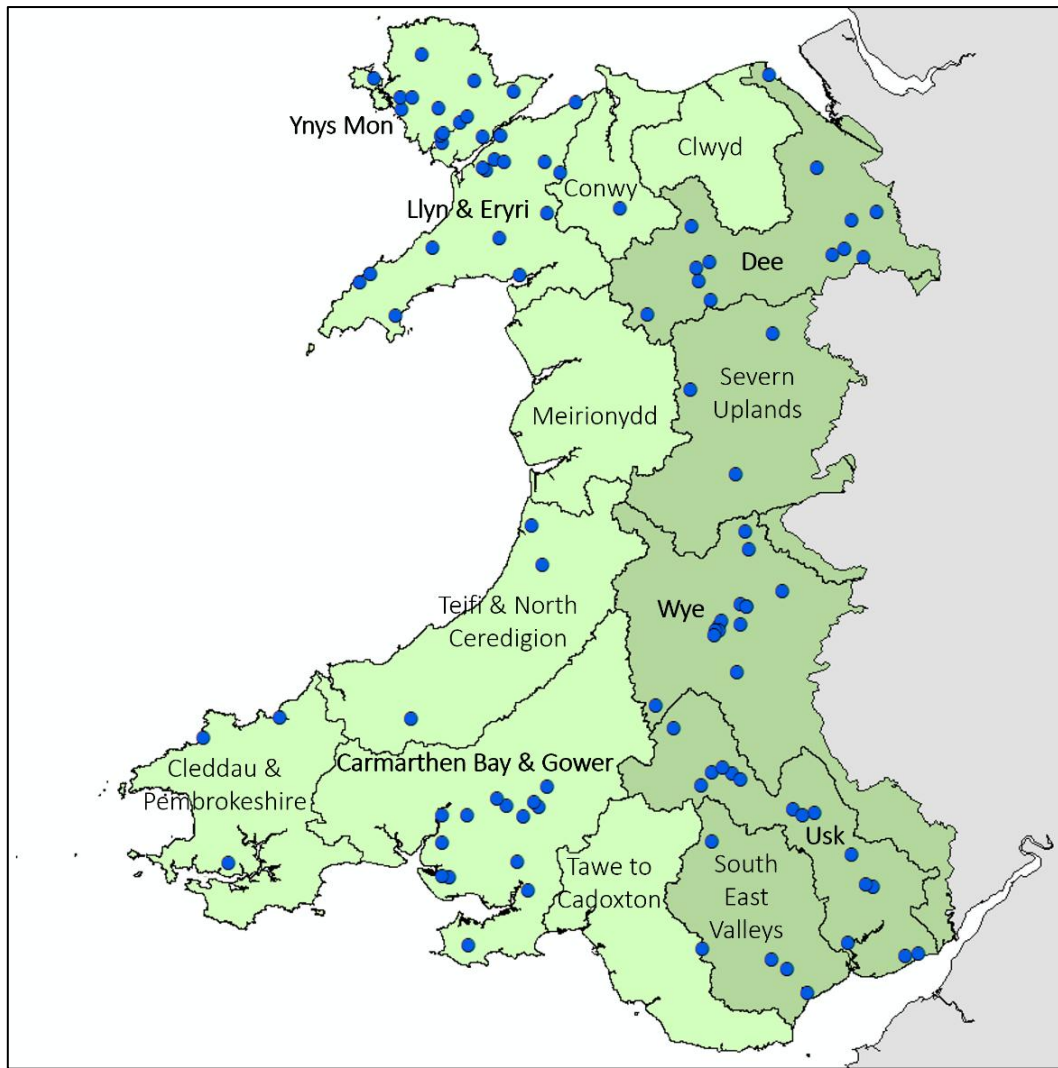


Figure 1: Geographical locations of 100 otters selected for analysis from Wales.

Locations of otter deaths (carcass sampling locations) are shown as blue dots. River catchments are outlined in black and coloured green. The differing shades of green show the split between catchments used for the western sample (light green – from the Western Wales river basin district [RBD]) and those used for the eastern sample (dark green – from the Severn and Dee RBDs). Individual river catchments are labelled in black. The six main river catchments compared in our models were Ynys Mon, Llyn & Eryri, Carmarthen Bay and Gower, Dee, Wye, and Usk. Adjacent areas of England are shown in grey. Data mapped using ArcMap GIS 10.7.1.

188 Spatial data sources and extraction

189 We collated anthropogenic and environmental data identified from the literature as pertinent to
190 describing spatial variation of PCBs in the freshwater environment, including both primary sources,
191 and environmental variables likely to influence recirculation and transport from secondary sources.
192 These data are outlined in Table S2, and the spatial distributions of these data are shown in Figure
193 S2. Spatial data and otter locations were mapped as shapefiles in ArcMap GIS 10.7.1. To collate data
194 from the likely area of exposure to contaminants during the otters life (rather than just at the
195 location of death), the location where the otter was found was used as a centre point for a buffer,
196 10km in radius, to create circular polygons (based on estimates of otters home range between 5 and
197 40km) (Erlinge 1967; Kruuk 1995; Ó Néill et al. 2009), within which environmental data were
198 summarised. The ArcMap tabulate intersection tool was used to calculate the percentage coverage
199 of landfill site area and arable area in each buffer. The sum of PCB emissions and the mean values
200 for annual rainfall, altitude and soil organic carbon were extracted for each buffer, using the join tool
201 in ArcMap. The linear distance from the location of death for each otter and the coast was calculated
202 with the join tool, and the influence of historic PCB production or disposal sites was quantified as a
203 binary variable, where otters were identified either as within a 10 km radius of any identified site, or
204 not (production and disposal sites were clustered in the same areas, and there was insufficient data
205 to treat them separately for the purposes of analysis). The range of values for spatial variables used
206 in statistical modelling is shown in Figure S3.

207

208 Analytical determination

209 Liver samples were sent for analysis to the Natural Resources Wales Analytical Services (NRWAS),
210 Swansea, which is accredited to international industry standard ISO/IEC 17025:2017 by UKAS (the
211 national accreditation body for the United Kingdom). Preliminary analysis was undertaken to explore
212 variation in deposition within the liver: three whole livers were selected, and PCB concentrations
213 compared between six subsamples taken randomly from different lobes. Results showed that
214 variation between lobes was minimal compared to variation between livers (Figure S4) and
215 subsequent analysis used a single subsample of approximately 5 grams from each liver.

216 The concentrations of 23 PCB congeners (listed with limits of quantification in Table S3) were
217 quantified in the otter liver samples, and reported as $\mu\text{g/kg}$ wet weight (ww). Lipid corrected
218 concentrations are used in some studies to facilitate comparison across different tissue types or

species with differing lipid content. In the current study, preliminary exploration of the data showed that wet weight concentrations and lipid corrected concentrations were highly correlated (for the sum of the top 9 most frequently detected PCBs, Spearman's rank correlation coefficient $\rho = 0.942$, $p < 0.001$). Because small errors in measurement of lipid content would significantly impact estimates of PCB concentration, we have chosen to report by wet weight. For conversion, the median value of 3.4% lipid content can be used to estimate values as $\mu\text{g/kg}$ lipid weight.

A detailed description of the method used is given in supporting information. In brief, the target compounds were extracted into 95:5% Dichloromethane:Toluene (DCM:Tol) mix using a Pressurised Solvent Extraction (PSE) system, where 0.5 g of sample was extracted in 45 mL solvent. Prior to extraction the sample was ground with sodium sulphate to dehydrate the sample and was spiked with internal standard. After PSE the samples were speed evaporated to 0.5 mL and exchanged into hexane prior to silica SPE clean up. The samples were eluted with 40% DCM in hexane solution, evaporated to 0.5 mL and then further cleaned using Gel Permeated Chromatography (GPC). Post GPC samples were again evaporated to 0.5 mL and exchanged into hexane prior to Florisil SPE clean up. The samples were eluted with 5% DCM in hexane solution and the eluent was evaporated and solvent exchanged into hexane with a final volume of 0.5 mL and vialled. For analysis, a 5 mL injection was made into a gas chromatograph (GC) using a PTV injector operating in solvent vent mode. The GC was interfaced with a mass spectrometer operating in atmospheric pressure chemical ionisation mode, acquiring accurate mass high resolution MS data. The analytes were quantitated against a standard curve using an internal standard calibration approach. Limits of quantification (LOQs) were determined as 10 times the within-batch standard deviation (SD) of the blank samples.

The method was validated by running duplicate blank matrix samples alongside duplicate test matrix samples (pig liver) that had been spiked with a known concentration of target analytes at two concentration levels. Six batches of validation samples were run prior to sample analysis. To pass for quantitative analysis, accuracy and precision targets were set as 20% bias and 15% relative standard deviation (RSD) and any failures were tested for statistical significance. Precision and/or accuracy of the results for four of the 23 PCB congeners (35, 105, 149 and 180) did not meet method validation thresholds (see Table S4), and were therefore assigned semi-quantitative values only. These values are considered indicative of the approximate concentration, enabling the measurements to be used to compare relative concentrations, and to explore spatial and temporal variation, but the reported absolute concentrations should be treated with caution.

Data analysis

Principal component analysis

Prior to carrying out multivariate analysis to explore associations of PCB concentrations with spatial variables, we checked all spatial variables for collinearity. Pearson's correlation showed that some of the continuous spatial variables were correlated with one another (e.g. sum PCB emissions with percentage landfill area [correlation coefficient of 0.80], and distance to coast with altitude [correlation coefficient of 0.85], Figure S5). Principal component analysis (PCA) was used to explore the environmental variation between river catchments, and to derive principal components (PCs) which can be used as aggregate variables to describe the nature of that variation. All environmental variables were log transformed prior to analysis to avoid undue influence of outliers, and data were scaled (to means of zero and variance of one), to avoid emphasis on variables with greater variance. The Singular Value Decomposition method ("prcomp") in R version 4.1.2 (R Core Team 2021) was used for calculating the components. The first three principal components (PCs) explained 88.2% of the spatial variation with PC1 explaining 56.3%, PC2 18.7%, and PC3 13.3%. PC1 was driven by PCB emissions, arable land and soil organic carbon, PC2 was primarily driven by distance to the coast and altitude and PC3 by rainfall (Table S5). Overall, the PCs showed that environmental characteristics were highly variable within, and showed a great deal of overlap between, river catchments, but in combination, the PCs do allow distinction between some catchment pairs, for example the Wye and Ynys Môn catchments are separated on PC1 (Figure 2)

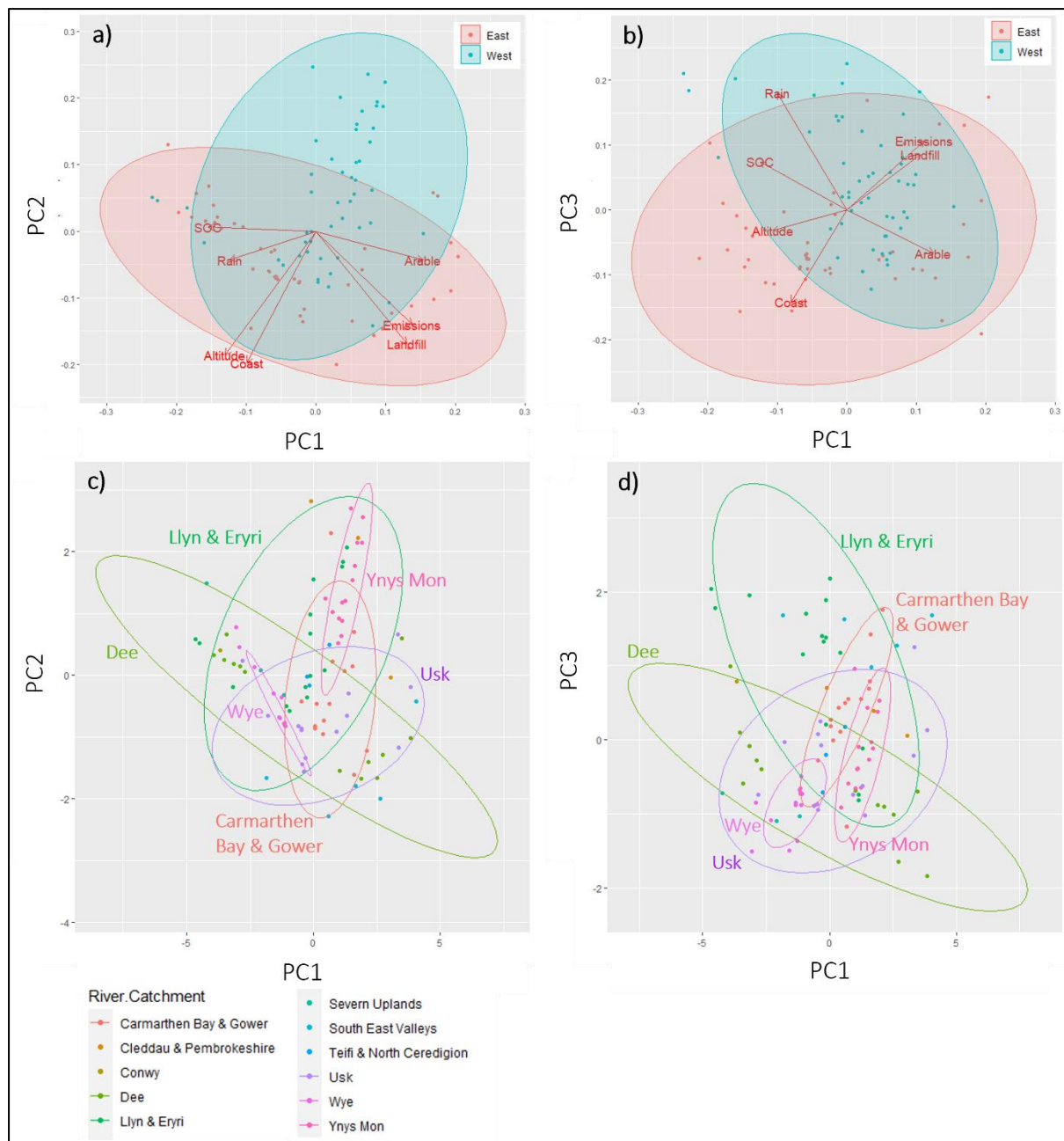


Figure 2: Differences between spatial areas in environmental characteristics, illustrated via results from principal component analysis (PCA).

Plots (a) and (b) show the loadings of the spatial variables for PCs 1 and 2, and PCs 1 and 3, respectively. Ellipses show the separation of values in east and west Wales. Plots (c) and (d) show the separation of values for the river catchments, ellipses for the six main river catchments are shown (others not shown due to low sample size) for PCs 1 and 2, and PCs 1 and 3, respectively.

General linear models

To explore spatial, temporal and biotic variation in PCB concentrations, we used general linear models (GLMs) with contaminant concentrations as the dependent term. Each of the nine congeners with detection >70% (PCBs 105, 118, 128, 153, 156, 157, 167, 170 and 180) were modelled, and the sum of all nine (Σ_9 PCB) was also modelled separately. For statistical modelling, samples with PCB concentrations below the limit of quantification (LOQ) were assigned $0.5 \times \text{LOQ}$. Measured concentrations (dependent variables) were highly skewed (Figure S6), comparison of model residuals across alternate error families and link functions indicated that log-transformed dependent data with a Gaussian error family and identity link function provided the best fit for all models.

To test for associations with the putative spatial drivers of concentration, the first three principal components derived from PCA of environmental characteristics (as described above), and the presence / absence of a PCB site within a 10km radius, were all included as independent variables in the model. To test for change over time, year was also included as an independent term in all models. Sex, length and SMI (body condition) were included as independent terms in all models, to explore and control for biotic variation between individuals; the interaction between sex and length was also included in order to explore and control for potential differences in the sex effect for otters of different ages (Table 1, model group 1).

To allow further exploration of potential spatial variation in time trends, we additionally modelled concentration (of each congener listed above, and with Σ_9 PCB) with year, while also including a location term: *either* east / west, or river catchment, as well as the interaction of the location term with Year, in order to test whether putative temporal trends might differ between the east and west of Wales or between catchments. Biotic variables (sex, length, SMI, sex:length) were controlled in all models (Table 1, model groups 2 and 3).

Independent variables in each model were standardised using the standardise function in the Arm package (Gelman 2011). To determine the most important variables in each model we used multi-model inference. The MuMIn package's dredge function (Barton 2009) was used to rank models by AICc, and model averaging was applied to models where delta AICc was <2 (Symonds and Moussalli 2011). Model estimates were obtained using the full average method, whereby values are averaged across all top models for all retained parameters, and set to zero for parameters not retained. The importance of each independent variable was determined based on its effect size, relative importance in the model and statistical significance. For each congener/ Σ_9 PCB, the average model was used to derive model predictions (using the "predict" function in R), while controlling for other

311 retained variables to their mean value for continuous data, or most frequently occurring value for
312 categorical data. Statistical analyses were conducted in R version 4.1.2 (R Core Team 2021).

313

314

Table 1: Starting variables used in each model group. The 9 individual congeners used in the models are PCBs 105, 118, 128, 153, 156, 157, 167, 170 and 180. n=100 for model groups 1 and 2. Data from the 6 main river catchments (Carmarthen Bay and Gower, Dee, Llŷn and Eryri, Usk, Wye and Ynys Môn) are included in model group 3, the 15 otters falling outside these catchments were excluded, therefore n=85 for model group 3. SMI stands for scaled mass index, an estimation of body condition calculated from Peig and Green 2009. PC stands for principal component, produced from the principal component analysis (PCA) conducted on the spatial variables, for details of which spatial variables each PC represents see Table S5.

Model group	Dependent variable	Biotic independent variables	Temporal independent variable	Spatial independent variables	Interaction terms	n
1	10 separate models: Σ_9 PCB, and 9 individual congeners	Sex Length SMI	Year	PC1 PC2 PC3 PCB site in 10km radius of otter (Y/N)	Sex:Length	100
2	10 separate models: Σ_9 PCB, and 9 individual congeners	Sex Length SMI	Year	Categorical east/west location (E/W)	EW:Year Sex:Length	100
3	10 separate models: Σ_9 PCB, and 9 individual congeners	Sex Length SMI	Year	River catchment (RC) n=6	RC:Year Sex:Length	85

Calculation of Σ PCB-TEQ

To analyse potential health impacts, PCB concentrations in the otters were compared to a known toxic threshold. Due to their protected species status, PCB toxicity to otters has not been tested experimentally. Values for American mink (*Neovison vison*), a closely related mustelid, can be used instead due to their similar anatomy and physiology (Kruuk and Conroy 1996). A Σ PCB-TEQ toxic threshold of 0.077 μ g TEQs/kg liver wet weight, which is a threshold based on mink kit survivability in three maternal feeding experiments (Zwiernik et al. 2011), was used by Kean et al (2021) to assess the health of otters found dead in England and Wales between 2000 and 2009. We repeated the same analysis for the current dataset. Σ PCB-TEQ was calculated using the toxic equivalency factors (TEFs) of six dioxin-like PCB congeners; these were 77, 105, 118, 126, 156 and 169, with TEFs of 0.0001, 0.00003, 0.00003, 0.1, 0.00003 and 0.03 respectively (Van den Berg et al. 2006). The mono-ortho congeners (PCBs 105, 118 and 156) had high detection of $\geq 98\%$, whereas the non-ortho congeners, PCB 77, 126 and 169, were detected in 10%, 48% and 7% of samples respectively. In this part of the analysis non-detected concentrations were assigned a value of zero (rather than assignment of half detection limit) to avoid falsely inflated TEQ which occurred particularly in samples when PCB 126 was detected (following Kean et al 2021). It should be noted that PCB 105 is

included in the calculation for Σ PCB-TEQ, concentrations for this congener are semi-quantitative and therefore this may introduce a degree of inaccuracy into the results.

Results and discussion

Detection and concentrations of PCBs

Detectable concentrations of PCBs were found in all 100 otter livers, showing that PCBs are still present in the environment almost four decades after PCB production was banned in the UK, in 1982.

The data were highly skewed (Figure S6), with Σ_{23} PCB (the sum of the concentrations of the 23 PCB congeners determined) of <200 $\mu\text{g}/\text{kg}$ ww in 67 otters, and >1000 $\mu\text{g}/\text{kg}$ ww in three otters (from the Ynys Môn, Wye and Dee river catchments). Nine of the twenty-three congeners analysed were found in >70% of livers; PCB congeners 118, 153, 170 and 180 were detected in 100% of livers, and PCB congeners 105, 156, 167, 128 and 157 were found in 98%, 98%, 95%, 75% and 72% of livers respectively (Table S6). These nine frequently detected congeners were also found at the highest concentrations in the livers, collectively accounting for 96.5% of the Σ_{23} PCB profile (Figure 3). PCB 153 had the highest maximum and median concentrations (545.82 $\mu\text{g}/\text{kg}$ ww and 39.51 $\mu\text{g}/\text{kg}$ ww respectively), individually accounting for 37% of the Σ_{23} PCB profile (Figure 3). PCB 153 is extremely stable (Fiolet et al. 2021) and has previously been found to be the most abundant congener in Eurasian otters (Kean et al. 2021), American river otters (*Lontra canadensis*) (Wainstein et al. 2022), harbour porpoises (*Phocoena Phocoena*) (Williams et al. 2020), predatory birds (Pereira et al. 2009; Walker et al. 2011) and humans (Fiolet et al. 2021). The rank order, by median concentration, for the frequently detected congeners was PCB 153 > 180 > 170 > 118 > 156 > 105 > 167 > 128 > 157 (Figure 3). These nine congeners are all penta-, hexa- or hepta-chlorinated congeners (congeners containing 5, 6, and 7 chlorine atoms respectively); it is expected that these high-chlorine PCBs would be seen at the highest concentrations in the otters due to their greater persistence and biomagnification potential, compared to low-chlorine PCBs (Smit et al. 1996; Montano et al. 2022). All low-chlorine PCBs (containing ≤ 4 chlorine atoms) in this study were detected in $\leq 21\%$ of otters. Low-chlorine PCBs have shorter half-lives and are more rapidly metabolised, and therefore will be eliminated from the body faster than high-chlorine homologues (Gabryszewska and Gworek 2021; Montano et al. 2022). Low-chlorine PCBs also have lower octanol-water partition coefficients and are more volatile, meaning they are more readily transported through freshwater systems to marine, and are more prone to atmospheric degradation (Harrad et al. 1994). The properties of PCBs, especially high-chlorine congeners, mean they will remain in the environment for decades; environmental residence

times are potentially as long as 110 years for PCB 153 and 70 years for PCB 180, the two most abundant congeners in our sample (Jönsson et al. 2003). This highlights the need to continue biomonitoring programmes for very persistent and bioaccumulative pollutants many years after regulation.

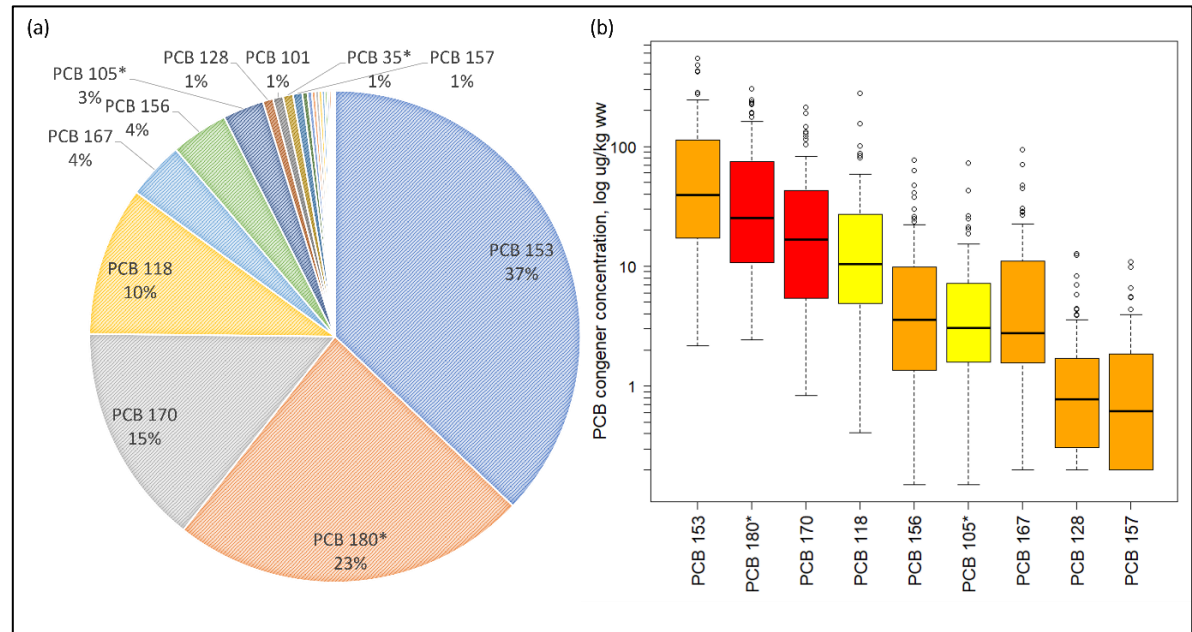


Figure 3: _PCB profile: 3a) Proportion of individual congeners in relation to the total of all PCBs. PCB congeners 8, 20, 28, 52, 77, 81, 114, 123, 126, 149, 169 and 189 represented <0.5% of the profile and are not labelled. 3b) Concentrations of PCB congeners with detection frequency >70%. Boxes are colour coded by degree of chlorination: yellow: penta-chlorinated, orange: hexa-chlorinated, red: hepta-chlorinated. Concentrations are recorded in $\mu\text{g/kg}$ wet weight and plotted on a log scale. * Denotes concentrations that were measured with less precision, and should be treated as semi-quantitative (see Supporting Information Analytical Methods section). Concentrations are presented as a boxplot; the thick black line indicates the median concentration, the lower and upper extents of the box indicate the 25th (Q1) and 75th (Q3) percentiles of the data distribution, whiskers show the lowest and highest values excluding outliers, and circles indicate outliers (1.5x the interquartile range).

Temporal trends

PCB concentrations increased between 2010 and 2019 (Figure 4). Year was significant, and had a relative variable importance (RVI) of 1, in all models for $\Sigma_9\text{PCB}$ and for all 9 individually modelled congeners which make up this sum (PCBs 105, 118, 128, 153, 156, 157, 167, 170 and 180; Tables S7-9). $\Sigma_9\text{PCB}$ increased from a model predicted concentration of 56.88 $\mu\text{g/kg}$ ww in 2010 to 230.16 $\mu\text{g/kg}$ ww in 2019. Of the individual congeners, PCB 153 had the greatest model predicted increase, a rise of 68.77 $\mu\text{g/kg}$ ww, from 21.29 $\mu\text{g/kg}$ ww in 2010 to 90.06 $\mu\text{g/kg}$ ww in 2019 (Figure 4).

Temporal trends were consistent across east and west Wales (Table S8) and the six main river catchments (Table S9) (i.e. the interaction terms of location with year were not significant in any model); however, the small sample size within each of the individual catchments means that our power to detect spatial differences in trend was limited.

Previous studies on otters have shown declines in PCBs since regulation (Mason 1998; Kean et al. 2021), which were mirrored in marine mammals. However, more recent studies on marine mammals such as polar bears (*Ursus maritimus*) (Rigét et al. 2016) and harbour seals (*Phoca vitulina concolor*) (Shaw et al. 2005), have shown increasing concentrations of PCBs over time. Although differences in tissue type and species mean that direct comparison of concentrations is not informative, the detection of similarly rising temporal trends suggests that the recent increase in environmental PCB exposure is widespread, spanning both marine and freshwater ecosystems.

The increase in PCB concentrations since 2010, following declines between 1983 and 2009 (Mason 1998; Kean et al. 2021), supports a hypothesis of increasing remobilisation from sediments. This trend is consistent with evidence that climate change is increasing the remobilisation of legacy pollutants via rising temperatures, increased rainfall, and sediment disturbance (Ma et al. 2016; Pizzini et al. 2022).

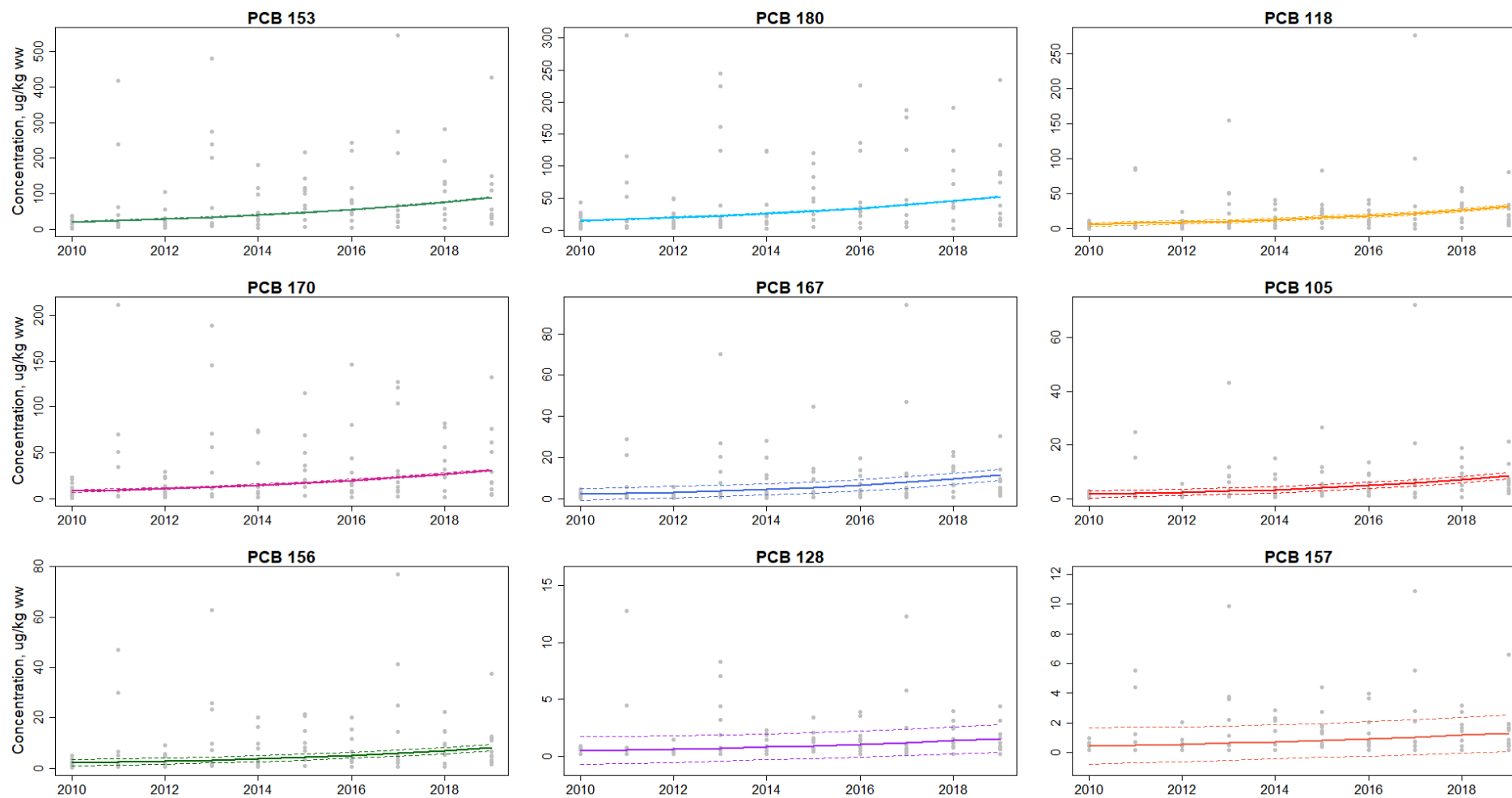


Figure 4: Temporal change in PCB concentrations. Plots show model-predicted PCB concentrations with year of otter death (coloured lines, $\pm$ SE), created using model group 1 (see Table 1)

418 Table 1). Other variables in the averaged models are controlled to their mean values, otter sex is controlled to 'male' and presence of historic PCB site is
419 controlled to 'no', see Table S7 for details. Grey dots show measured concentrations (raw data). Note the differing scales on the y axis, PCB congeners have
420 been ordered by decreasing median concentration.

Spatial variation

Otters from the west of Wales had slightly higher concentrations of Σ_9 PCB and individual congeners than those from the east (Figure S7). The east/west categorical variable was retained in all group 2 models, but was not significant and had a low relative importance (≤ 0.38 in all models, Table S8). On average, otters from the Llŷn and Eryri, and Ynys Môn river catchments (both in the west) had the highest PCB concentrations (Figure S7), but river catchment was not retained in any group 3 top models (Table S9).

Results of group 1 modelling with principal components (PCs), showed that spatial variation in concentration was driven primarily by distance to the coast and altitude, with higher concentrations in low lying, coastal areas. Results from multi-model inference showed that PC1 (which described 56.3% of the total environmental variation and was positively associated with atmospheric PCB emissions and percentage arable area, and negatively with soil organic carbon, Table S5); was retained in only 3/10 models (PCBs 105, 118 and 156), in which it had very low relative variable importance (all ≤ 0.14). PC2 described less environmental variation (18.7%) and was primarily negatively associated with distance to the coast and altitude, Table S5) but was retained with a positive association in all models, with a high relative variable importance, and was significant in five of the ten models (Table S7). Rainfall made a small contribution to spatial variation, with higher concentrations in areas of higher rainfall. PC3 (which described 13.3% of the variation and on which rainfall loaded most heavily, Table S5) was retained with a positive association in most models (exceptions were PCB 128 and 157) but with a low relative importance (all ≤ 0.21 , Table S7).

Atmospheric PCB emissions and landfill site area were negatively associated with PC2 (although with lower loadings than distance to coast and altitude), so the positive association between PC2 and PCB concentrations suggests, counterintuitively, higher PCB concentrations in areas of lower emissions or landfill site area. Similarly, the presence / absence of a historic PCB site within a 10km radius of each otter was retained in all models except PCBs 105 and 118 (Table S7) and suggested lower concentrations in otters near to historic sources. Areas of highest emissions, landfill sites and historic PCB sites are, however, predominantly in the east of Wales (Figure S2); it seems likely that the strong coastal effect outweighs any remaining input from legacy sites. This interpretation is supported by the lack of retention of PC1 (which was positively associated with atmospheric PCB emissions) in most models. Combined, these results suggest anthropogenic sources via the release of PCBs from legacy products, thermal processes, leachate from landfill, sewage sludge amended soils, and historic production and waste disposal sites are of lesser importance when explaining spatial variation of PCB concentrations in Wales than distance to the coast, altitude and rainfall.

It is likely that the higher PCB concentrations found in Wales's low lying coastal areas reflect the remobilisation and recirculation of PCBs from secondary sources, delivered via ocean currents and atmospheric movement from the north-east Atlantic Ocean (Ma et al. 2016; Vorkamp et al. 2022), as well as via the erosion and downstream transport of contaminated river sediments (Dawson and Macklin 1998). Elevated PCB concentrations at coastal locations have previously been reported in British otters (Kruuk and Conroy 1996; Kean et al. 2021) and other wildlife (Herceg Romanić et al. 2012; Jürgens et al. 2015), and similar mechanisms proposed. In Shetland, the high PCB content of the North-East Atlantic, delivered via oceanic and atmospheric deposition, was proposed as a key driver of elevated PCB concentrations in Eurasian otters compared to mainland Scotland (Kruuk and Conroy 1996). Likewise, North Atlantic currents have been linked to higher PCB burdens in European Arctic biota relative to the Canadian Arctic (Wang-Andersen et al. 1993; Wolkers et al. 1998).

Remobilisation from both marine and freshwater sediments is likely to be exacerbated by climate change. In the oceans, climate change is driving altered currents and increased upwelling, which can bring PCBs from deeper waters to the surface (Noyes et al. 2025), where rising air and sea temperatures can increase volatilisation to air (Ma et al. 2016; Othman et al. 2022). Climatic patterns such as the North Atlantic Oscillation (NAO) also influence these processes by changing wind patterns and ocean circulation. During positive NAO phases, stronger westerly winds and shifts in ocean currents can increase mixing and water movement, which may enhance the vertical and horizontal transport of contaminants (Outten and Davy 2024). Coastal waters have been identified as a significant secondary source of PCBs to the atmosphere (García-Flor et al. 2009; Nicolaus et al. 2015), with subsequent deposition in coastal areas via frontal rainfall, as well as in upland regions via orographic precipitation. In freshwater systems, heavy rainfall and flooding events (which are becoming more frequent in Wales as elsewhere in Northwestern Europe (Blöschl et al. 2019; Cotterill et al. 2021; Madge 2021)) are associated with increased runoff and water turbulence, which can destabilise riverbanks, disrupt sediments and re-expose historically contaminated layers, potentially releasing stored PCBs back into the water column (Whitehead et al. 2009). These remobilised contaminants can then be transported downstream on suspended particles, contributing to elevated PCB levels in lowland rivers and estuarine environments (Dawson and Macklin 1998).

It is difficult to disentangle the relative contributions made by oceanic versus freshwater sources; further research quantifying PCB concentrations in river and marine sediments is needed to help identify dominant sources and inform mitigation strategies. However, differences in model results between more- and less-chlorinated congeners suggests that marine sediments may make a greater contribution. The observed association between PCB concentration and PC2 (describing proximity to

coast and altitude) was stronger for the less chlorinated congeners, such as penta- and hexa-chlorinated PCBs, than more chlorinated hepta-chlorinated congeners (Table S7). The less chlorinated congeners are more soluble, volatile and mobile (Zhou et al 2014) making them more prone to long-range atmospheric transport (Sari et al. 2023). These patterns are consistent with coastal exposure via atmospheric deposition and prevailing Atlantic winds. However, this does not rule out the contribution of downstream transport of sediments, particularly for more chlorinated congeners; overall, both coastal exposure and downstream transport of sediments are likely to contribute to PCB concentrations in coastal waters. Dietary variation (between areas, and change over time) could potentially confound comparisons of PCB exposure. If, for example, prey taken at the coast were higher in lipid content, age or trophic level, their consumption would be associated with higher exposure to PCBs. However, even where otters are found near the coast, they rely on access to freshwater (Chanin 2003). Dietary analysis suggests that otters living close to the coast in England and Wales consume marine species (predominantly flatfish) only infrequently, with freshwater prey predominating (Drake et al. 2023). Changes in dietary composition over time reflect changing prey communities, with declines in species such as European eels (*Anguilla anguilla*) and salmonids (e.g. *Salmo salar*) reflecting population declines in these species, and increased consumption of smaller, lower-trophic-level fish, such as European bullheads (*Cottus gobio*) and sticklebacks in compensation (Moorhouse-Gann et al. 2020). This dietary shift would be expected to result in a decline in PCB exposure for otters as long-lived, fat rich, high trophic level species are replaced with shorter lived, less fatty, lower trophic level fish; despite this, PCB concentrations have increased. Temporal change in diet potentially means that the measured increases in PCB exposure are underestimated.

Biotic variation and potential health effects

Twelve PCBs share similar toxicological properties to 2,3,7,8-tetra-chloro-dibenzo-dioxin (TCDD) (Alcock et al. 1998). These dioxin-like PCBs are the non-ortho and mono-ortho substituted PCBs and have been assigned toxicity equivalency factors (TEFs) (Van den Berg et al. 2006). Sixteen percent of otters in this study exceeded a Σ PCB-TEQ toxic threshold which was calculated from the TEFs of six dioxin-like PCB congeners (77, 105, 118, 126, 156 and 169, see methods). Kean et al. (2021) reported concentrations above the same threshold in 38% of otters across England and Wales; reanalysis of their data for Wales only showed exceedance in 28% of otters in 2000-2009. In both Kean et al. 2021 and the current study, threshold exceedance occurred in otters from all years, and from a spatially widespread distribution (Figure 5). The higher percentage exceedance described by Kean et al. 2021

reflects much higher PCB concentrations in the earlier years of their study, which have not been reached during 2010-2019, despite the increasing trend during the latter time period. While environmental PCB concentrations do not appear to be having a strong detrimental impact at a population level, as seen during the 1950s to 1970s, in a minority of otters, PCB concentrations may still be having an impact on health.

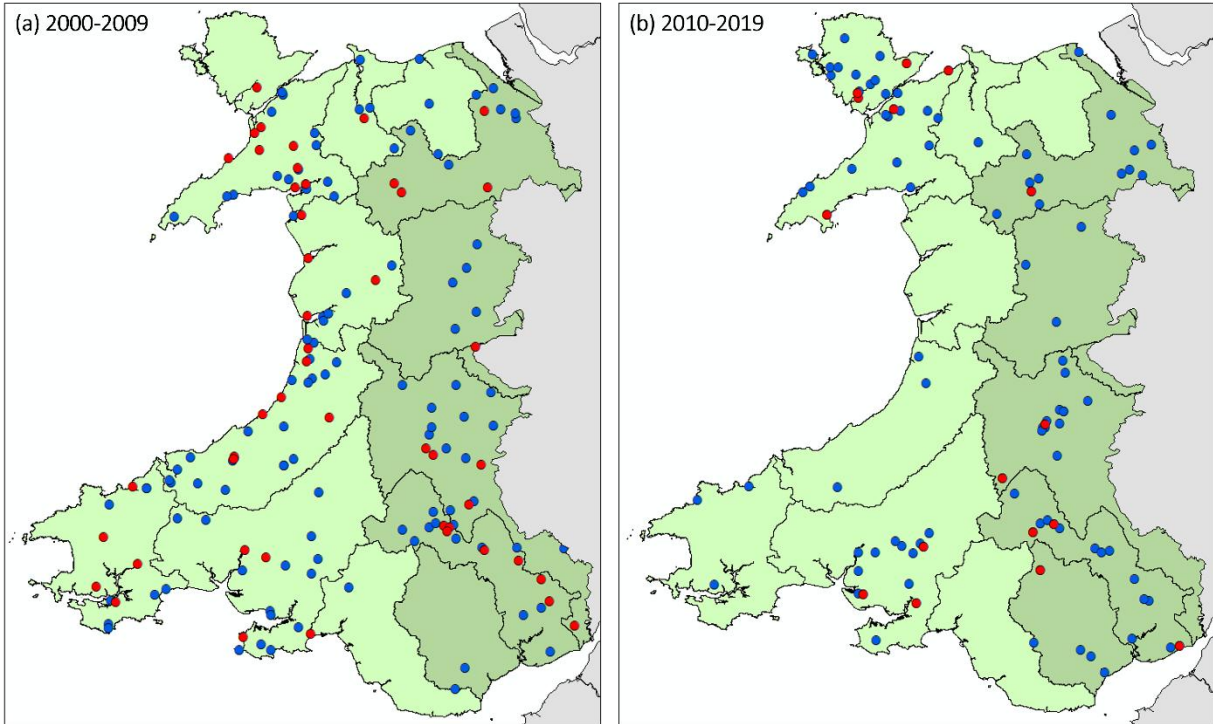


Figure 5: Distribution of otters in which a toxicity threshold for dioxin-like PCB congeners was exceeded for otters found dead between (a) 2000-2009 and (b) 2010-2019. The total TEQ value for PCB congeners 77, 105, 118, 126, 156 and 169 was summed. Individuals in which the sum was greater than published toxicity threshold of 0.077 ug TEQs/kg liver wet weight (Zwiernik et al., 2011) are shown in red, those below threshold in blue. Western Wales RBD is coloured light green, eastern RBDs (Dee and Severn) are coloured dark green. River Catchments are outlined in black. Data for 2000-2009 is taken from Kean et al. 2021. Data mapped using ArcMap GIS 10.7.1.

In the current study, PCB concentrations were negatively correlated with an index of body condition (SMI) for all five of the mono-ortho dioxin-like congeners that were modelled (PCBs 105, 118, 156, 157 and 167). SMI was retained for 5/5 in model group 2, for 3/5 (PCBs 105, 118, and 167) in model group 3, and for 1/5 (PCB 105) in model group 1, although relative importance of SMI was less than 0.42 in all cases (Tables S7-9). In comparison, of the four non dioxin-like congeners modelled (PCBs 128, 153, 170, 180), SMI was only retained once (for PCB 128 in model group 3; none in model groups 1 or 2, see Tables S7-9). Correlations between PCB congener concentrations and body

condition might occur either because PCBs are harmful to health, or because remobilisation of fat reserves (e.g. during starvation, associated with low body condition scores) results in increased concentrations of PCBs in the liver (Wania 1999), thus making it difficult to conclusively identify cause / effect. However, if association with SMI was driven by remobilisation of fats, the expectation would be for similar results for both non- and dioxin-like congeners. The clear difference between groups supports the interpretation that the dioxin-like congeners, which are the most toxic PCBs (suggested to interfere with the metabolism of vitamin A and linked to increased frequency of disease in mink and otters) (Håkansson et al. 1992; Leonards et al. 1996) are still impacting otter health. These findings highlight the importance of congener specific analysis of health effects, concentrations, toxicity, and persistence.

Male otters had slightly higher concentrations of penta- and hexa-chlorinated PCBs than females (sex was retained), whereas for the hepta-chlorinated congeners (PCBs 170 and 180) there was no difference between the sexes (sex was not retained, with a single exception of congener 170 for model group 3; Tables S7-9). A sex difference has been observed before for Σ PCB in Eurasian otters (Kean et al. 2021) and harbour porpoises (Law et al. 2012) and is suggested to reflect a differing diet between the sexes or maternal transfer by females (as evidenced in other mammals and birds, Bargar et al. 2001; Mauritsson et al. 2022). The smaller, more mobile, lower-chlorinated congeners have greater potential to be transferred and therefore maternal transfer has been found to be inversely related to congener chlorination (Bargar et al. 2001). This would explain why a sex difference was not seen in the hepta-chlorinated congeners in the present study.

We found no correlation between PCB concentrations and otter length (a proxy for age) for almost all the congeners (exceptions of PCB 167 in model group 1, PCB 105 in model group 2, and PCB 180 in model group 3; Tables S7-9). The unimportance of length in the models was consistent between the sexes (the interaction term of length with sex was not retained in any model). Some other studies on otters have also found no relationship (Kruuk and Conroy 1996; Roos et al. 2001), while Kean et al. (2021) found a non-linear relationship with age, with juveniles having higher concentrations than sub-adults and adults. It is important to note that because juveniles were excluded from this study, the absence of evidence for an association with length in our data does not rule out accumulation with age.

Conclusions

This study highlights the ongoing challenges posed by legacy contaminants, emphasising that their ecological impacts remain unresolved. PCB concentrations remain at concentrations that are toxicologically relevant in otters from Wales, as in other top predator species (Jepson et al. 2016; Williams et al. 2020). Exceedance of thresholds for otters suggests that the current EQS_{biota} set for fish may not be adequately protective of higher trophic-level species from PCB exposure as intended, which has important implications for the implementation of the Water Framework Directive. Fish monitoring under the WFD relies on destructive sampling and regulatory authorities in some areas now struggle to source sufficient fish due to population declines. Otters, which can be monitored non-invasively through post-mortem analysis of carcasses found dead, offer a valuable alternative for assessing PCB contamination in freshwater ecosystems. This highlights the potential role of otters as sentinel species to complement existing monitoring efforts and bridge critical data gaps in contaminant surveillance.

Our findings contribute to a growing body of evidence indicating that, with primary anthropogenic PCB emissions now regulated, secondary sources have become more important to the occurrence, fate and exposure of PCBs in the environment (Ma et al. 2016). Furthermore, climate change may be influencing their global cycling, further exacerbating the challenges associated with these persistent pollutants (Lohmann et al. 2007; Ma et al. 2016; Merhaby et al. 2019), as evidenced by the increasing PCB concentrations observed in Welsh otters between 2010 and 2019. The remobilisation of PCBs highlights the complex interconnection between oceanic, atmospheric, and hydrological pathways in driving PCB distribution in Welsh otter populations, reflecting broader global patterns of contaminant transport and deposition in aquatic ecosystems. To more fully evaluate future risks to wildlife and human health, further research is needed to quantify the concentrations of persistent chemicals stored in sediments, paired with hydrological modelling to explore remobilisation under future management and climate scenarios.

Due to their persistence, PCBs will continue to cycle between environmental compartments for decades, possibly centuries (Herceg Romanić et al. 2012) with increasing remobilisation posing a significant global challenge given their capacity for long-range atmospheric transport (Lohmann et al. 2007). Addressing this issue requires strengthened international cooperation and a concerted effort to eliminate PCB stocks by the Stockholm Convention's deadline, alongside efforts to meet climate change targets and mitigate the resurgence of legacy pollutants in the environment.

Acknowledgments

EO's PhD studentship was co-funded by the Waterloo Foundation and Cardiff University. Natural Resources Wales funded and carried out the chemical analyses of otter livers. Otter carcasses were collected by members of the public, Natural Resources Wales, Trunk Roads Agencies, Wildlife Trusts and other organisations. Cardiff University Otter Project Research Assistants assisted with postmortem procedures. We thank the editor and anonymous reviewers for helpful feedback on a previous version of the manuscript.

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